I. On a New Constitutive Equation for Non-Newtonian Fluids

II. Brownian Motion with Fluid-Fluid Interfaces

Thesis by

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to Alain

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INTRODUCTION

This thesis is a small contribution to the ambitious goal of understanding some of the more complex flows that are found in nature, namely, flows of fluids with a microstructure. There is a great diversity of such flows: suspensions, emulsions, polymeric solutions, ..., each exhibitting phenomena not found in the flow of homogeneous Newtonian fluids. A bit of this diversity has been incorporated in this thesis: The first part of it is on some aspects of non-Newtonian fluid flow, and in the second part Brownian motion involving interfaces between Newtonian fluids is studied.

Given the large amount of effort devoted recently to thenumerical simulation of non-Newtonian fluid flow, the absence of mathematical proofs that any of the standard computational methods for solving the equations will converge except for nearly Newtonian flows seems somewhat disturbing. While there is evidence that investigators may have overcome the so-called "high Weissenberg-number problem," at least in specific cases, confidence in the numerical solutions would undoubtedly be increased by a rigorous mathematical foundation for the numerical algorithm. The first, and in many cases nontrivial, step towards this is to prove that a solution actually exists. In the first part of this thesis, a proof of existence without restriction on the parameters is given for a particular modified finitely extendible nonlinear elastic dumbbell model. A physical basis for the modifications is given.

For numerical computation, the issue of stability of a flow is also an important one, as the small errors introduced by discretization are essentially perturbations in the flow, which, if they grow too fast, can make convergence impossible. An energy method calculation is given here for the same FENE dumbbell model considered in the existence proof (except for the modifications) to show that for any flow in a bounded domain, at small enough Reynolds number and high enough Deborah number, all disturbances will remain bounded. While the estimate found for the highest Reynolds number and lowest Deborah number for guaranteed stability may be very conservative, the result is nevertheless useful in that it shows that if there is an instability, it must occur at a critical Reynolds or Deborah number.

While the Brownian motion of a rigid particle has received much attention in the literature, and the Stokes-Einstein diffusivity of a rigid particle is a result almost as well-known as the Stokes drag law, the Brownian motion of systems that are more complex hydrodynamically has only recently begun to be investigated. Most recent work on such systems has been for systems with rigid boundaries, e.g., suspensions of rigid spheres. In this thesis, the case of deformable fluid-fluid interfaces is considered. Since the understanding of the behavior of clusters or suspensions of particles can only follow an understanding of the behavior of a single particle, the two cases considered here are a drop in an infinite fluid, and an isolated particle in the presence of an approximately planar interface. Expressions for statistical quantities, such as the velocity autocorrelation, of the particle and drop motion are derived. In the case of the interface, the nature of its effect on the particle's behavior, beyond the obvious fact that it changes the particle's mobility, is explored. Similarly, the surface-tension dependence of the drop's motion is investigated.

Finally, in a slight digression, the problem of high-frequency oscillatory Stokes flow around two spheres, with specified velocity at their surfaces, is reduced to an infinite system of algebraic equations for the (frequency-dependent) coefficients in a spherical harmonic expansion of the solution. This is expected to be useful for computations of such flows where better accuracy than an approximate solution obtained by the method of reflections is desired.

CHAPTER I

Existence of Solutions for all Deborah Numbers for a Non-Newtonian Model Modified to Include Diffusion

Chapter I consists of the text of a paper
submitted to *Journal of Non-Newtonian Fluid Mechanics*

Existence of Solutions for all Deborah Numbers for a Non-Newtonian Model Modified to Include Diffusion

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INTRODUCTION

The question of existence of solutions and convergence of numerical methods for models proposed to describe non-Newtonian fluids has become particularly relevant on account of significant difficulties that have been encountered in computational studies. There is evidence that at least part of the problem may be due to intrinsic flaws in the model, namely, that they allow as mathematical possibilities phenomena that are physically unreasonable. In this paper, it will be shown by examining the conditions needed for a mathematical proof of existence, that attention can be drawn to the specific terms in the constitutive equation that could give rise to unphysical behavior. This should not be surprising, since the definition of "existence" generally has built into it a restriction to classes of functions that are physically reasonable.

Only constitutive equations of the so-called "molecular" type that come from simple "dumbbell" models will be considered here. Bird *et al.* [1] discuss these models and their derivation in detail. In particular, the following (nondimensionalized) model (used by Chilcott and Rallison [2] in their numerical calculations of flow past a cylinder and sphere) will be used:

$$\begin{aligned} Re\ \mathbf{u}\cdot\nabla\mathbf{u} &= -\nabla p + \nabla^2\mathbf{u} + \frac{c}{D}\nabla\cdot(f(R)\mathbf{A}); \\ \mathbf{u}\cdot\nabla\mathbf{A} &= \mathbf{A}\cdot\nabla\mathbf{u} + \nabla\mathbf{u}^T\cdot\mathbf{A} - \frac{f(R)}{D}(\mathbf{A}-\mathbf{I}); \end{aligned} \qquad (1.1a,b)$$

where

$$f(R) = \frac{1}{1-\frac{R^2}{L^2}}; \qquad (1.2)$$

and R^2 is the trace of $\mathbf{A}$. Here, $\mathbf{u}$ is the velocity, Re is the Reynolds number, c is the dumbbell concentration, D is the Deborah number, and $\mathbf{A} = <\mathbf{rr}>$, where $\mathbf{r}$ is the end-to-end vector for a dumbbell and the brackets denote an ensemble average. The Deborah number $D = \tau/T$, where τ is the polymer relaxation time, and T is the characteristic time scale of the flow. This is a FENE (finitely extendable nonlinear elastic) dumbbell, meaning that the two beads are connected by a spring that can be stretched by the flow, and which exerts a restoring force tending to bring the dumbbell back to its equilibrium extension. The words "finitely extendable" refer to the fact that the spring coefficient $f(R)$ becomes infinite as R approaches L, (L being an adjustable model parameter) so that the (average) dumbbell extension can never exceed L. It has already been pointed out by Rallison and Hinch [3] that without this feature of finite extension, there are certain flows in which the dumbbell would extend to infinite length, a clearly nonphysical phenomenon. If it was attempted to prove existence of solutions for the equations of motion based upon such a model, (or derivatives such as the Oldroyd model, which can be derived from a dumbbell model with a *constant* spring coefficient), it would immediately be clear that the possibility of infinite extension is an obstacle, since all currently available methods of proof require *a priori* estimates guaranteeing that all possible solutions lie in a compact subset of some reasonable function space, so that sequences of approximate solutions will converge to an element in that space.

While the need for a bound on the extension of the dumbbell has been recognized for some time by non-Newtonian fluid mechanicians, there are two other potential flaws in models such as the one above that seem not to have received much notice yet. It is the purpose of this paper to draw attention to them and to show that, once they are corrected, the existence of a solution can be guaranteed *independent* of the Deborah number. While no *proof* is provided here that solutions in some Sobolev space fail to exist without the modifications to the model suggested here, it is certainly true that none of the currently available methods to prove existence can be applied successfully. Our point of view is that this is an indication of problems with the model rather than any inadequacy of available mathematical theory. Undoubtedly it is possible to come up with a sufficiently generalized definition for a solution (i.e., the solution would be sought in a function space with sufficiently relaxed continuity and boundedness properties) that existence could be proved regardless of physically unrealistic aspects of the model. Since this would mean that the solution might be physically unrealistic, however, this seems to be a much less useful approach than modifying the model to disallow any undesirable behavior, and *then* proving existence.

One of the problems with the model in (1.1) is brought to light in one of the essential steps of an existence proof, namely, getting an *a priori* estimate on $\mathbf{A}$ in the Sobolev space in which a solution is sought, which means that the components of $\mathbf{A}$, as well as all their first-order partial derivatives, should be Lebesgue-square-integrable (which implies a certain degree of smoothness and boundedness). However, a constitutive equation of the type given in (1.1) is really just an ordinary differential equation giving the derivative of $\mathbf{A}$ along a streamline. If a physically reasonable value of $\mathbf{A}$ is prescribed as an upstream boundary condition for a streamline, and the velocity field is not too pathological (this will be made more precise later by requiring that $\mathbf{u}$ lie in a Sobolev function space so that it and its first- order derivatives are square-integrable), then the terms on the right- hand side of (1.1b), which determine $\mathbf{u} \cdot \nabla \mathbf{A}$ (the derivative of $\mathbf{A}$ along the streamline), will integrate to give bounded and continuous values of $\mathbf{A}$ along the streamline. If the streamline is closed, the constitutive equation is like an eigenvalue equation for $\mathbf{u}$, since the value for $\mathbf{A}$ obtained by integrating along the streamline must equal the value at the starting point of the integration. In this case it is less clear what conditions on $\mathbf{u}$ are necessary to ensure that $\nabla \mathbf{A}$ will be bounded. In any case, it is natural to ask the following question: Is there any limitation on the derivatives of $\mathbf{A}$ normal to a streamline? Is it possible to construct solutions in which $\mathbf{A}$ is discontinuous across streamlines?

DISCONTINUOUS SOLUTIONS

We begin by showing that discontinuous solutions can definitely be constructed for the case where the inlet conditions are allowed to be discontinuous. A very simple example is provided by uniform flow with a constant velocity U in the x direction. In this case the equations reduce to

$$U\frac{\partial \mathbf{A}}{\partial x} = -\frac{f(R)}{D}(\mathbf{A} - \mathbf{I}); \tag{2.1}$$

$$0 = -\nabla p + \nabla \cdot \mathbf{A}. \tag{2.2}$$

It is evident upon examination of (2.1) that an asymptotic solution for $\mathbf{A}$ as $x \to \infty$ for *any* initial condition at $x = 0$ is simply $\mathbf{A} \to \mathbf{I}$. Thus, sufficiently far downstream from any initial condition, the distribution for $\mathbf{A}$ will approach arbitrarily closely to being continuous. Nevertheless, if a discontinuity is specified for $\mathbf{A}$ at $x = 0$, a discontinuity of decreasing magnitude, at which $\nabla \mathbf{A} = \infty$, will exist for all finite x. To demonstrate this behavior, (2.1) can be solved directly. Any solution of (2.1) for $\mathbf{A}$ that is independent of y will satisfy (2.2). It is convenient to solve for $\zeta = R^2 = Tr\mathbf{A}$ rather than for $\mathbf{A}$ itself. For this purpose, (2.1) can be rewritten in the form

$$U\frac{d\zeta}{dx} = -\frac{1}{D}\frac{1}{1-\dfrac{\zeta}{L^2}}(\zeta - 3), \tag{2.3}$$

which, if ζ_0 denotes the value of ζ at $x = 0$, may be integrated to give

$$\exp\Big(-\frac{1}{L^2}(\zeta - \zeta_0)\Big)\left(\frac{\zeta - 3}{\zeta_0 - 3}\right)^{1-\frac{3}{L^2}} = \exp\Big[-\frac{x}{DU}\Big]. \tag{2.4}$$

Since the solution depends only on x, it is clear that two solutions with different inlet conditions may be pieced together at some value of y, at which there will be a discontinuity in $\mathbf{A}$. Equation (2.2) indicates that the pressure must also be discontinuous at this value of y. It may be seen from (2.4) that for any choice of inlet condition other than the equilibrium $\mathbf{A} = \mathbf{I}, R^2 = 3$, the discontinuity persists *infinitely far* downstream, though its magnitude decreases with increasing x. Although viscosity tends to smooth velocity gradients, it does not necessarily smooth gradients in $\mathbf{A}$, since these can exist without a velocity gradient.

The question remains as to whether it is possible to construct a solution with a discontinuity in $\mathbf{A}$ for *smooth* inlet conditions. Consider Equations (1.1), this time including time dependence, in the sense of generalized functions:

$$Re\left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{u}\right) = -\nabla p + \nabla^2\mathbf{u} + \frac{c}{D}\nabla\cdot(f(R)\mathbf{A})$$
$$\frac{\partial \mathbf{A}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{A} = \mathbf{A}\cdot\nabla\mathbf{u} + \nabla\mathbf{u}^T\cdot\mathbf{A} - \frac{f(R)}{D}(\mathbf{A} - \mathbf{I}). \tag{2.5a, b}$$

First, for a steady solution to have a jump in $\mathbf{A}$, so that $\nabla\mathbf{A}$ will behave like a delta function, there must be a jump in $\nabla\mathbf{u}$ and/or p, since the delta function must be matched by another delta function in (2.5a). To show that the jump in $\mathbf{A}$ must *necessarily* be *across* a streamline, note that if the term $\mathbf{u}\cdot\nabla\mathbf{A}$ had delta-function behavior, it could not be balanced by any other term in (2.5b) (since only first derivatives of $\mathbf{u}$ appear in (2.5b)). However, for an *unsteady* solution, if the position of the discontinuity were allowed to move (analogous to a travelling shock wave), the $\mathbf{u}\cdot\nabla\mathbf{A}$ term in (2.5b) could possibly be balanced by the time-derivative term, so that a jump would *not* necessarily have to be in a direction normal to a streamline.

To derive conditions, analogous to the Hugoniot relations for shock waves, that a discontinuous solution must satisfy, the problem must be reformulated in "weak form." These conditions will actually be derived here only for the steady case, although how to extend this derivation to the case of a moving discontinuity will be obvious. As shown above, in the steady case, only the momentum equation has discontinuous terms, so it is not necessary to deal with the constitutive equation to derive jump conditions. The weak form of the momentum equation is, in index notation,

$$\int_V \left[-Re\; u_j \frac{\partial u_i}{\partial x_j}\phi_i + p\frac{\partial \phi_i}{\partial x_i} - \frac{\partial u_i}{\partial x_j}\frac{\partial \phi_i}{\partial x_j} - \frac{c}{D} f(R) A_{ji} \frac{\partial \phi_i}{\partial x_j} \right] d\mathbf{x} = 0, \qquad (2.6)$$

where this is to hold for all divergence-free test function vectors ϕ with compact support. Here, V denotes the flow domain, and S will be used to denote the discontinuity surface, V^+ and V^- will designate the two regions on either side of the discontinuity, and "+" or "-" will denote values of quantities in the limit of approaching the discontinuity surface from the V^+ or V^- sides, respectively. Within regions V^+ and V^-, this weak form can be rewritten as

$$\left[\int_{V^+} + \int_{V^-}\right] \left[-Re\; u_j \frac{\partial u_i}{\partial x_j}\phi_i + \frac{\partial}{\partial x_i}(\phi_i p) - \frac{\partial}{\partial x_j}\left(\phi_i \frac{\partial u_i}{\partial x_j}\right) - \frac{c}{D}\frac{\partial}{\partial x_j}\left(f(R) A_{ji}\phi_i\right) \right] dx$$

$$-\left[\int_{V^+} + \int_{V^-}\right]\left[-\phi_i \frac{\partial p}{\partial x_i} + \frac{\partial^2 u_i}{\partial x_j^2}\phi_i + \frac{c}{D}\frac{\partial f(R)A_{ji}}{\partial x_j}\phi_i\right] d\mathbf{x}. \tag{2.7}$$

If the differential form of the equation (i.e. Equation (2.5a)) holds within V^+ and V^-, then the second two integrals are zero. The divergence theorem may be applied to the first two integrals, so that only boundary terms from along the discontinuity surface result, giving

$$\int_{S^+}\left[p\delta_{ij} - \frac{\partial u_i}{\partial x_j} - \frac{c}{D}f(R)A_{ji}\right]\phi_i n_j^+ \cdot dS$$
$$+\int_{S^-}\left[p\delta_{ij} - \frac{\partial u_i}{\partial x_j} - \frac{c}{D}f(R)A_{ji}\right]\phi_i n_j^- \cdot dS \quad . \tag{2.8}$$

Here, n_j denote components of the outward-pointing normal to the region, so that clearly $n_j^+ = -n_j^-$. Since this equation is to hold for all test functions ϕ, it must be that at all points on the surface of discontinuity

$$\left\{-\left[-p\delta_{ij} + \frac{\partial u_i}{\partial x_j} + \frac{c}{D}f(R)A_{ji}\right]^+ + \left[-p\delta_{ij} + \frac{\partial u_i}{\partial x_j} + \frac{c}{D}f(R)A_{ji}\right]^-\right\} n_j^+ = 0. \tag{2.9}$$

These are the jump conditions for steady flow. How to extend this derivation to the unsteady case by integrating by parts in the time t should also be clear. The velocity of the surface of discontinuity will appear when the time derivatives of integrals over the domain are taken. Again, this is analogous to the derivation of the Hugoniot relations for inviscid shock waves.

It remains a challenge to prove or disprove the existence of discontinuous solutions satisfying these jump conditions without having discontinuous initial or boundary data. There certainly seems not to be any *obvious* reason that such a flow field could not exist. The condition (2.9) demonstrates that even velocity-gradient jumps are not disallowed for a fluid of type (1.1), although viscosity is present. By comparison, jump conditions allowing for discontinuous velocity gradients can *not* be derived for the steady Navier-Stokes equations, because there is no deviatoric stress term in the momentum equation that can balance the delta-function behavior of the viscous term.

The uniform flow example given above at least demonstrates that a solution with discontinuous $\mathbf{A}$ can exist if the boundary data are allowed to be discontinuous. The question that now arises is what such discontinuous solutions mean (if anything) physically. The flow variables that jump across the discontinuity in the particular example above are the components of $\mathbf{A}$. However, $\mathbf{A}$ is the second moment of the distribution of extension and orientation of the dumbbells, so that a jump in $\mathbf{A}$ means a sudden change in this distribution. This is not physically reasonable: clearly, dumbbells (if they are to model, and therefore behave like, polymer molecules) on either side of a streamline are not completely independent of each other; *Brownian motion will continually move them across streamlines*, and thus smooth any discontinuities in $\mathbf{A}$. This is an important effect that is missing in all differential constitutive equations so far proposed.

MODEL MODIFICATION

It turns out that this effect fails to be included in most models because of the "local homogeneity" approximation made in their derivation. To see how this assumption eliminates Brownian motion of dumbbells across streamlines from the model, one can consider Phan-Thien's [4] derivation for the dumbbell model as an example. Phan-Thien's analysis starts with the stochastic differential equations governing the positions $\mathbf{R}_1$ and $\mathbf{R}_2$ of the two dumbbell beads. The equations are stochastic because a Brownian (random) force on each bead is included. After changing variables to $\mathbf{R}^{(c)}$, the center-of-mass of the dumbbell, and $\mathbf{R}$, the end-to-end vector for the dumbbell, the stochastic differential equations become (neglecting inertia associated with the mass of the beads):

$$\xi\dot{\mathbf{R}}^{(c)} - \xi\mathbf{u}(\mathbf{R}^{(c)}) = \mathbf{f}^{(c)}(t); \tag{3.1a}$$

$$\xi\dot{\mathbf{R}} + \frac{6k\theta}{Na^2}\kappa\mathbf{R} - \xi\mathbf{L}(\mathbf{R}^{(c)})\cdot\mathbf{R} = \mathbf{f}(t). \tag{3.1b}$$

(Phan-Thien's "internal viscosity" term has been omitted here.) Here, ξ is the hydrodynamic resistance of one dumbbell bead (assumed constant); $\mathbf{u}$ is once again the velocity; $\mathbf{L}$ is the velocity gradient (a function of $\mathbf{R}^{(c)}$ in general); k is Boltzmann's constant; θ is the absolute temperature; N is the number of sublinks in a dumbbell; a is the length of a "sublink"; and $\kappa(R)$ is the spring law. The important thing to note here is that there are two Brownian forces: $\mathbf{f}^{(c)}(t)$ acts on the center of mass, and $\mathbf{f}(t)$ acts on the end-to-end vector; the two forces are uncorrelated. Clearly, the first of the two gives rise to motion of dumbbells across streamlines (without changing their orientation or extension!). It should be noted that one approximation has been made in writing (3.1b), namely that the difference in the velocity field experienced by the two beads of a single dumbbell has been assumed to be $\mathbf{L}(\mathbf{R}^{(c)}) \cdot \mathbf{R}$ rather than $\mathbf{u}(\mathbf{R}_2) - \mathbf{u}(\mathbf{R}_1)$. In other words, while the Equations (3.1) allow for the fact that the velocity field may change in a nonlinear way over distances comparable to the size of an ensemble of dumbbells, it is assumed that it can be linearized on the length scale of a single dumbbell. The Fokker-Planck equation corresponding to the stochastic differential equations can be derived, following Phan-Thien [4], as

$$\frac{\partial \phi}{\partial t} = \frac{\partial}{\partial R_i^{(c)}} \left\{ \frac{k\theta}{2\xi} \frac{\partial \phi}{\partial R_i^{(c)}} - u_i(\mathbf{R}^{(c)})\phi \right\} + \frac{\partial}{\partial R_i} \left\{ \frac{2k\theta}{\xi} \frac{\partial \phi}{\partial R_i} + \left[\frac{6k\theta}{\xi N a^2} \, \kappa(R) \; R_i - L_{ik} R_k \right] \phi \right\} \quad . \tag{3.2}$$

Here, "$\phi(\mathbf{R}^{(c)}, \mathbf{R}, t)$ is the probability density function of the vector Markovian process $(\mathbf{R}^{(c)}, \mathbf{R})$." The constitutive equation is the second moment of this equation, and is thus obtained by multiplying by $\mathbf{RR}$, and integrating over $\mathbf{R}$, the integration corresponding to an average over an ensemble of dumbbells. A few integrations by parts are necessary to make all the terms look like expectations; consequently, some surface terms are obtained, since the integration is over a finite range of possible values of $\mathbf{R}$, but they are omitted because the probability density is assumed to go to zero as the two extremes of a maximally extended dumbbell ($R \to L$) and a completely collapsed dumbbell ($R \to 0$) are approached. After these manipulations, the result is

$$\frac{\partial \mathbf{A}}{\partial t} = \frac{k\theta}{2\xi} \nabla^2 \mathbf{A} - \mathbf{u} \cdot \nabla \mathbf{A} + \frac{8k\theta}{\xi} \mathbf{I} - \frac{12k\theta}{\xi N a^2} \kappa(R) \mathbf{A} + \mathbf{L} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{L}^T, \tag{3.3}$$

where the terms have been written in the same order as their antecedents in the Fokker-Planck Equation (3.2). Here, gradients with respect to the center of mass position $\mathbf{R}^{(c)}$ have been written as ∇, since the variable $\mathbf{R}^{(c)}$ in (3.1), (3.2) corresponds to the position variable $\mathbf{x}$ in (1.1b). The only approximation made in obtaining this moment equation is the usual "preaveraging" assumption made in replacing the expectation of the nonlinear spring force by its value at the expectation of R. If the above-mentioned assumption of linearization of the velocity on the length scale of a *single* dumbbell had not been made, some preaveraging assumption would be needed for the last term in (3.2) also, and there would be some correction to the last two terms in (3.3). Apart from this, *no assumption* of constancy of the velocity gradient over the averaging region has been made. If the Deborah number is defined as

$$D = \frac{\xi N a^2}{12k\theta}, \tag{3.4}$$

and the usual assumption is made that the isotropic term

$$\frac{8k\theta}{\xi}\mathbf{I} \tag{3.5}$$

should be replaced by

$$\frac{\kappa(R)\mathbf{I}}{D}, \tag{3.6}$$

so that the quiescent state will correspond to an equilibrium value of $\mathbf{A} = \mathbf{I}$, then it can be seen that equation (3.3) is almost identical to (1.1b) (in the steady case). The one difference is the presence of the term

$$\frac{k\theta}{2\xi}\nabla^2\mathbf{A}, \tag{3.7}$$

which originates in the Brownian motion of the center of mass of the dumbbell. In other words, Brownian motion should give rise to a *diffusion* term in $\mathbf{A}$, in addition to its other effect (namely, the Brownian force along the connector which turns out to prevent the dumbbell from collapsing to zero length by giving rise to an isotropic term). Since both these effects arise from the same cause, it is inconsistent to include one and not the other, which, in fact, is what is done. This diffusion effect is lost in derivations such as Phan-Thien's because the local homogeneity assumption is taken to imply that the probability density ϕ can be factored into two parts, one a function of $\mathbf{R}$ only, and the other a function of $\mathbf{R}^{(c)}$ only. This gives rise to two separate Fokker-Planck equations [4], one having gradients in $\mathbf{R}$ only, and the other having gradients in $\mathbf{R}^{(c)}$ only. It is from the former that the constitutive equation is derived; thus all dependence on position is lost except the dependence implicitly left in the velocity gradient in the terms $\mathbf{L}\cdot\mathbf{A}$ and $\mathbf{A}\cdot\mathbf{L}^T$. There is no need to make this factoring assumption, however, as has been shown above.

A relevant question at this point is the magnitude of the spatial diffusion term relative to other terms in the equation for $\mathbf{A}$. Let U and d denote typical velocity and length scales for a flow. Then, the magnitude of the diffusion term, the coefficient of which is essentially a Stokes-Einstein diffusivity, can be estimated as

$$\frac{k\theta}{\mu a}\frac{L}{d^2} \tag{3.8}$$

(where the hydrodynamic resistance has been estimated by μa, with μ being the solvent viscosity, and the magnitude of $\mathbf{A}$ has been estimated by the upper bound for its trace, namely L, the maximum extension). On the other hand, the convective derivative term $\mathbf{u} \cdot \nabla \mathbf{A}$ in the constitutive equation can be estimated as UL/d. The ratio is then

$$\frac{k\theta}{\mu a d^2 U}. \tag{3.9}$$

If typical values of $\theta = 10^2$K, $\mu = 10^{-2}$ dyne/cm · s, $a = 10^{-4}$ cm, $U = 1$ cm/s, $d = 10$ cm are taken, and it is recalled that $k \sim 10^{-16}$erg/K, the dimensionless ratio is then on the order of 10^{-11}, because Boltzmann's constant is so small (in other words, because the Brownian diffusivity of a dumbbell is small). Thus, the diffusion term will be negligible except in regions of extremely high gradients of $\mathbf{A}$. However, in such a region, it plays an essential role in tending to smoothen the distribution for $\mathbf{A}$.

It is significant that such a diffusion term should be included in the constitutive equation, because this actually results in a change of the apparent type of the equation from hyperbolic to parabolic. Much attention has been given recently [5] to the implications of apparent hyperbolicity of the constitutive equation, such as possible loss of evolution, and difficulties in both proving the existence of a solution and proving convergence of numerical approximate solutions. However, the present analysis suggests that the constitutive equation should really be unambiguously parabolic (this is ensured when the highest-order derivative term is a diffusion term).

The presence of the diffusion term obviates the issue of whether the absence of "inlet" conditions in regions of closed streamlines will necessarily cause the flow to be nonunique in those regions. (The flow may of course be nonunique for other reasons, depending on the flow parameters; this would be expected since the Navier-Stokes equations do not have unique solutions at higher Reynolds numbers.) As mentioned above, the constitutive equation without the diffusion term is like an ordinary differential equation for $\mathbf{A}$ along a streamline, indicating that it is necessary to specify the value of $\mathbf{A}$ at one point on each streamline. This is straightforward only if there is a clearly defined surface on which the desired value of $\mathbf{A}$ is known (like a boundary condition) and through which *all* streamlines pass exactly once. Not all flows will have this property. Some may have closed eddies; the exact location of these and the conditions in them would be unknown until the entire flow is computed. There may be nonuniqueness of $\mathbf{A}$ in these eddies. However, with the diffusion term, boundary conditions for $\mathbf{A}$ are evidently necessary on the entire boundary of the domain (just as $\mathbf{u}$ must be specified on the entire boundary of the domain when the Navier-Stokes equations are to be solved), and it should not be necessary to know anything about the geometry of the streamlines to specify correct boundary conditions for a well-posed problem. On the other hand, specifying appropriate values of $\mathbf{A}$ (or possibly its normal derivative) on all the boundaries, including solid ones, rather than just at the "inlet" boundary, may itself be a problem, at least temporarily, since very little work has been done to determine anything about the distribution of dumbbell (macromolecular) configurations near solid walls.

The absence of this important diffusion term is not the only problem that arises in proving the existence of solutions for the FENE model, Equations (1a,b). Another problem is that it seems to be impossible to prove mathematically that terms involving the spring-law $f(R)$ remain bounded in all flow conditions (because $f(R)$ becomes unbounded as $R \rightarrow L$), since it cannot be assured that R is somehow prevented from getting arbitrarily close to L. (Examination of the existence proof given later in this paper will show that bounding these terms is essential for proving existence.) From a qualitative physical point of view, the existence of an unbounded force (i.e. $R \rightarrow L$ and $f(R) \rightarrow \infty$) exerted by the fluid on the spring (and vice versa) in some region seems implausible. In order to balance the spring force, an extremely large velocity gradient would be required. Moreover, it is generally believed that the kinematics of a flow will be modified in regions of high stress, so this type of singularity, in principle allowed by the constitutive model, never actually occurs. Unfortunately, this implausibility argument does not constitute a mathematical proof, and it is necessary for the existence proof to modify the model in such a way as to explicitly limit the maximum magnitude of the spring force. There is no question that the spring force should be bounded, since realistically a dumbbell, which corresponds in some sense to a polymer molecule, should not be able to withstand an infinite force trying to pull it apart. The question is whether the bound on the spring force is already implicit in the model or whether it must be explicitly built in as a modification. Since this question cannot be answered with complete certainty, the (perhaps redundant) modification *will* be made.

The modification proposed here corresponds to the introduction of a "breaking function" $g(R)$ that simply "turns off" the dumbbell contribution to the bulk stress in the momentum equation, and the stretching and rotating of the dumbbell in the constitutive (moment) equation, whenever R exceeds some finite threshold value R_1 that may be placed as close to L as desired. One possibility, which is chosen here, is for the function $g(R)$ to equal 1 for all R less than R_1, and to ramp rapidly down to zero in the range $R_1 \leq R \leq R_2$, where R_2 is still less than L. It should be noted that this is to a large extent an arbitrary choice; however, the existence proof given below will not work for a discontinuous $g(R)$ (for example, if $g(R)$ suddenly dropped to zero at $R = R_1$). The model becomes

$$Re\, \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla p + \nabla^2 \mathbf{u} + \frac{1}{D} \nabla \cdot (g(R) c f(R) \mathbb{A});$$

$$\mathbf{u} \cdot \nabla \mathbf{A} = g(R) \left[\mathbf{A} \cdot \nabla \mathbf{u} + \nabla \mathbf{u}^T \cdot \mathbf{A} - \frac{f(R)}{D} (\mathbf{A} - \mathbf{I}) \right] + \epsilon \nabla^2 \mathbf{A} \quad (3.10a, b)$$

(where ϵ has been used to denote the nondimensionalized coefficient of the diffusion term). By this somewhat artificial mechanism, it can be guaranteed that the spring force does not exceed some finite threshold value. The function $g(R)$ is denoted here as the "breaking function" because one interpretation of the resulting model is that $g(R)c$ is the dumbbell concentration, which goes to zero in regions where R exceeds its threshold value at which the spring force $f(R)$ starts to become greater than the maximum force that can be exerted on the dumbbell before it breaks. To continue with this interpretation, once dumbbells break in a certain region, they should cease to affect the flow (which is ensured because the deviatoric stress term is "turned off" in the momentum equation), and they should also cease to be affected by the flow (hence the factor $g(R)$ in the constitutive equation). In any region where $g(R)$ is less than 1 (i.e., dumbbells are breaking), there will still be convection and diffusion of (unbroken) dumbbells into the region, so the convective and diffusive terms are retained without modification in the constitutive equation. It is not claimed here that the above interpretation is rigorous; it should merely be viewed as providing an intuitive justification. Of course, the model (3.10) may seem somewhat simplistic, since, strictly speaking, if dumbbells are allowed to break, a conservation equation for the dumbbell concentration should also be included. In addition, it would perhaps be more realistic to allow a dumbbell to break into two smaller dumbbells rather than to consider it to essentially vanish when it breaks. However, *the goal here is to find the* simplest *model that can be expected to give reasonable results*, rather than to try to include every possible effect. In this spirit, it is conjectured that since the regions where the spring force will tend to be very large will be only a very small fraction of the entire flow domain (and perhaps nonexistent, since the threshold force can be chosen at any arbitrarily large, but finite value), the change in dumbbell concentration that is due to breaking will not be very significant, so that no conservation equation will be included here. This conjecture could be verified *a posteriori* by looking at a numerical solution to this model and seeing how large the regions are where $g(R) \neq 1$ (if, in fact, there are any) relative to the size of the whole domain. (In a numerical study, it will probably be desirable to choose the arbitrary upper bound on the spring force based on how large a number can be reasonably handled on the computer - a consideration that has no bearing on proving that the solution exists.) As a final comment we would only reiterate the following point: If the interaction between dumbbell stretching and local modifications in the flow is such that an unbounded spring force can never be obtained in a "real" flow, then the introduction of a *mathematical* bound on the spring force is superfluous, and the function $g(R)$ will play no role in the behavior of the model (3.10). If, on the other hand, an unbounded spring force is possible with the original FENE model (1.1) (and this would be unexpected and interesting), then $g(R)$ is essential to any proof of existence of solutions for arbitrary Deborah number D.

It will now be shown that a solution to the modified model (3.10) with appropriate boundary conditions can be guaranteed for a bounded domain, *independent* of the Deborah number.

FUNCTION SPACES

Before giving the existence proof, the function spaces in which the problem is to be formulated must be introduced. This is necessary to make precise the notion of "existence." It is also necessary to know exactly what Sobolev spaces the functions lie in, since Sobolev embedding theorems will be used later to get estimates on the norms of certain functions. The notation and definitions of these spaces are taken in large part from Ladyzhenskaya [6], and are just reviewed briefly here. The domain Ω on which a solution is to exist will be assumed to have a Lipschitz-continuous boundary. To begin with, the space $L_2(\Omega)$ will be needed. This is the space of all scalar functions on Ω that are Lebesgue-square-integrable. The inner product on this space is defined by

$$(u,v)_{L_2} = \int_\Omega u \cdot v d\mathbf{x} \tag{4.1}$$

and the norm is given by

$$\|u\|_{L_2} = \sqrt{(u,u)_{L_2}} \ . \tag{4.2}$$

Once this space has been defined, the Sobolev space $W_2^1(\Omega)$ can be defined by

$$W_2^1(\Omega) = \left\{ u : u \in L_2(\Omega) \text{ and } \frac{\partial u}{\partial x_i} \in L_2(\Omega) \right\} \tag{4.3}$$

with the inner product

$$(u,v)_{W_2^1} = \left(\frac{\partial u}{\partial x_i}, \frac{\partial v}{\partial x_i} \right)_{L_2} \tag{4.4}$$

and the norm

$$\|u\|_{W_2^1} = (u,u)_{W_2^1}^{\frac{1}{2}} \quad , \tag{4.5}$$

which make it a Hilbert space. Note that for a bounded domain, the other obvious choice of inner product,

$$(u,v)_{W_2^1} = (u,v)_{L_2} + \left(\frac{\partial u}{\partial x_i}, \frac{\partial v}{\partial x_i}\right)_{L_2} , \tag{4.6}$$

is equivalent to this one by Poincare's inequality. Now define the space $\dot{W}_2^1$ to be the closure of the set of all infinitely differentiable functions with compact support in Ω. It can be shown that this is a subspace of $W_2^1(\Omega)$.

Since only incompressible flows will be considered here, the velocity field will be divergence-free. It is therefore convenient to work with the space

$$\dot{J}(\Omega) = \{\mathbf{u} \in [\dot{W}_2^1(\Omega)]^3 : \nabla \cdot \mathbf{u} = 0 \ ,\} \tag{4.7}$$

where derivatives are interpreted in the generalized sense. The superscript "3" refers, of course, to the fact that $\mathbf{u}$ is a vector function with 3 scalar components. This space will be given the inner product

$$[\mathbf{u},\mathbf{v}]_{\dot{J}} = \int_\Omega \nabla\mathbf{u} : \nabla\mathbf{v}^T d\mathbf{x} \tag{4.8}$$

and the norm

$$\|\mathbf{u}\|_{\dot{J}} = [\mathbf{u},\mathbf{u}]_{\dot{J}}^{\frac{1}{2}} \quad . \tag{4.9}$$

Let $H(\Omega)$ denote the completion of $\dot{J}(\Omega)$ in the metric generated by this norm. Its inner product and norm will be the same as for $\dot{J}$, and will be indicated by a subscript H instead of $\dot{J}$. (In fact, for the *bounded* domain Ω to be considered here, the spaces $H(\Omega)$ and $\dot{J}(\Omega)$ are the same.)

The variable $\mathbf{A}$ that appears in the constitutive equation will lie in the space

$$[\dot{W}_2^1]^9 \quad , \tag{4.10}$$

which will hereafter be denoted by H'. (Of course, only 6 of the 9 components of $\mathbf{A}$ are actually independent, so the space $[\dot{W}_2^1]^6$ could be used instead; however, this would not affect the results in any significant way.) It is also convenient to define

$$H'' = H \times H' \quad , \tag{4.11}$$

which will be the space where the full solution vector

$$\begin{pmatrix} \mathbf{u} \\ \mathbf{A} \end{pmatrix} \tag{4.12}$$

(the 3 velocity components and the 9 components of $\mathbf{A}$) will lie. The inner product on these compound spaces will be defined as the sum of the inner products on the component spaces:

$$[\mathbf{A}, \chi]_{H'} = \sum_{i,j} [A_{ij}, \chi_{ij}]_{\dot{W}_2^1} \quad , \tag{4.13}$$

$$\left[\begin{pmatrix} \mathbf{u} \\ \mathbf{A} \end{pmatrix}, \begin{pmatrix} \phi \\ \chi \end{pmatrix} \right]_{H''} = [\mathbf{u}, \phi]_H + [\mathbf{A}, \chi]_{H'} \quad , \tag{4.14}$$

and as usual the norms will be defined by

$$\|\mathbf{A}\|_{H'} = [\mathbf{A}, \mathbf{A}]_{H'}^{\frac{1}{2}} \quad , \tag{4.15}$$

$$\left\|\begin{pmatrix}\mathbf{u}\\ \mathbf{A}\end{pmatrix}\right\|_{H''} = \left[\begin{pmatrix}\mathbf{u}\\ \mathbf{A}\end{pmatrix}, \begin{pmatrix}\mathbf{u}\\ \mathbf{A}\end{pmatrix}\right]_{H''}^{\frac{1}{2}} \quad . \tag{4.16}$$

Finally, the space $L_4(\Omega)$ of all scalar functions whose fourth powers are Lebesgue integrable will be needed; its norm is

$$|v|_{L_4} = \left(\int_\Omega v^4 d\mathbf{x}\right)^{\frac{1}{4}}. \tag{4.17}$$

At times it will be convenient to consider the velocity $\mathbf{u}$ as a member of

$$[L_4(\Omega)]^3 \tag{4.18}$$

and the tensor $\mathbf{A}$ as a member of

$$[L_4(\Omega)]^9. \tag{4.19}$$

FORMULATION

The existence proof given here is similar in many ways to Ladyzhenskaya's [6] presentation for the Navier-Stokes equations. As in her version of the proof (due originally to Leray), it is first necessary to reformulate the problem to allow for weak (i.e., generalized) solutions, rather than to leave the problem as a boundary-value problem for a system of differential equations. This is the purpose of this section. Continue to consider the stationary case, and for simplicity, a bounded domain with the flow driven by an external forcing function $\mathbf{F}(\mathbf{x})$, with no slip at the boundary ($\mathbf{u} = \mathbf{0}$) and the dumbbells at equilibrium at the boundary ($\mathbf{A} = \mathbf{I}$). Of course, for most engineering purposes, flows driven by specified (nonhomogeneous) boundary conditions are of interest; it should be noted that the proof given below can be *easily modified* to include such a case (as Ladyzhenskaya does for the Navier-Stokes equations).

The model formulated as a system of differential equations thus becomes

$$
\begin{aligned}
Re\, \mathbf{u} \cdot \nabla \mathbf{u} &= -\nabla p + \nabla^2 \mathbf{u} + \frac{1}{D} \nabla \cdot (g(R) c f(R)(\mathbf{B} + \mathbf{I})) + \mathbf{F}(\mathbf{x}), \\
\mathbf{u} \cdot \nabla \mathbf{B} &= g(R) \left[\mathbf{B} \cdot \nabla \mathbf{u} + \nabla \mathbf{u}^T \cdot \mathbf{B} + \nabla \mathbf{u} + \nabla \mathbf{u}^T - \frac{f(R)}{D} \mathbf{B} \right] + \epsilon \nabla^2 \mathbf{B},
\end{aligned}
\tag{5.1a, b}
$$

$$
\nabla \cdot \mathbf{u} = 0, \tag{5.2}
$$

which are to hold in the domain Ω, with boundary conditions

$$
\mathbf{u} = \mathbf{0} \text{ and } \mathbf{B} = \mathbf{0} \text{ on } \partial\Omega \quad , \tag{5.3}
$$

where a slight change of variables to $\mathbf{B} = \mathbf{A} - \mathbf{I}$ was made in order to make the boundary conditions formally homogeneous. As above,

$$
R^2 = Tr\mathbf{A} = Tr\mathbf{B} + 3 \quad , \tag{5.4}
$$

and

$$
f(R) = \frac{1}{1 - \frac{R^2}{L^2}} \quad , \tag{5.5}
$$

and the particular choice of the polymer-breaking function g(R) will be (with $R_2 < L$):

$$g(R) = 1 \quad \text{if } R \leq R_1$$
$$= 1 - \frac{R - R_1}{R_2 - R_1} \text{ if } R_1 \leq R \leq R_2$$
$$= 0 \quad \text{if } R_2 \leq R. \tag{5.6}$$

To use functional-analytic methods, the model must be reformulated in a generalized sense. The idea is to look for solutions

$$\begin{pmatrix} \mathbf{u} \\ \mathbf{B} \end{pmatrix} \tag{5.7}$$

in the space H'' (defined above), but such solutions may not have derivatives defined at every point. So, in standard fashion, the inner product of the equations is taken with an arbitrary element

$$\begin{pmatrix} \phi \\ \chi \end{pmatrix} \tag{5.8}$$

of H'', and this is integrated over the domain Ω. This results in:

$$\int_\Omega \Big\{ \big[Re\, \mathbf{u} \cdot \nabla \mathbf{u} + \nabla p - \nabla^2 \mathbf{u} - \frac{1}{D} \nabla \cdot (g(R) c f(R)(\mathbf{B} + \mathbf{I}))\big] \cdot \phi$$
$$+ \big[\mathbf{u} \cdot \nabla \mathbf{B} - g(R)[\mathbf{B} \cdot \nabla \mathbf{u} + \nabla \mathbf{u}^T \cdot \mathbf{B} + \nabla \mathbf{u} + \nabla \mathbf{u}^T - \frac{f(R)}{D} \mathbf{B}] - \epsilon \nabla^2 \mathbf{B}\big] : \chi \Big\} d\mathbf{x}$$
$$- \int_\Omega \mathbf{F}(\mathbf{x}) \cdot \phi d\mathbf{x} = 0 \quad , \tag{5.9}$$

which should hold for all

$$\begin{pmatrix} \phi \\ \chi \end{pmatrix} \in H''. \tag{5.10}$$

As written, it is not obvious that these integrals exist except for certain $\binom{\mathbf{u}}{\mathbf{B}}$ in H'' that are smooth enough. This is fixed by a few integrations by parts (and using the fact that $\mathbf{u}$ and ϕ are divergence-free):

$$\int_\Omega \Big\{ -Re\,\mathbf{uu} : \nabla\phi + \nabla\mathbf{u} : \nabla\phi^T + \frac{c}{D} g(R) f(R) \mathbf{B} : \nabla\phi - \mathbf{B} : (\mathbf{u}\cdot\nabla\chi)$$
$$+ g(R)\big[-B\cdot\nabla\mathbf{u} - \nabla\mathbf{u}^T\cdot\mathbf{B} - \nabla\mathbf{u} - \nabla\mathbf{u}^T + \frac{f(R)}{D}\mathbf{B}\big] : \chi + \epsilon\nabla\mathbf{A} \vdots \nabla\chi^T \Big\} dx$$
$$- \int_\Omega \mathbf{F}(\mathbf{x})\cdot\phi d\mathbf{x} = 0 \quad , \tag{5.11}$$

where the notation convention

$$\nabla\mathbf{B} \vdots \nabla\chi^T = \sum_{i,j,k} \frac{\partial B_{ij}}{\partial x_k}\frac{\partial \chi_{ij}}{\partial x_k} \tag{5.12}$$

is used. Now recall that

$$\int_\Omega \nabla\mathbf{B} \vdots \nabla\chi^T = \sum_{i,j} [B_{ij}, \chi_{ij}]_{\dot{W}_2^1} = [\mathbf{B}, \chi]_{H'} \,. \tag{5.13}$$

and

$$\int_\Omega \nabla\mathbf{u} : \nabla\phi^T = \sum_{i,k} \frac{\partial u_i}{\partial x_k}\frac{\partial \phi_i}{\partial x_k} = [\mathbf{u}, \phi]_H \quad . \tag{5.14}$$

EXISTENCE PROOF

Now that the problem has been properly formulated, the actual proof of existence of solutions can be given. The proof will be in several steps; to assist in following the proof, the steps will first be outlined briefly here. A familiarity with Ladyzhenskaya's presentation for the Navier-Stokes equations is helpful here, since this proof is similar in outline. Step 1 involves viewing the weak formulation of the problem obtained in the previous section as a linear functional on the test functions. It is shown that this linear functional is bounded by deriving an estimate for its norm. Step 2 invokes the Riesz Representation Theorem to rewrite the action of the linear functional on a vector function as the inner product of another (unique) vector function with it. The unique vector function thus defined by the linear functional must be shown to be completely continuous. This makes Step 3 possible, which involves rewriting the problem again, this time as an operator equation, with a solution corresponding to a fixed point of the operator. To assure existence of a fixed point (the desired solution!) the Leray-Schauder theorem is used; this is Step 5. Before this, however, some *a priori* estimates satisfied by all possible solutions must be derived; this is Step 4.

So to begin with Step 1, note that for a fixed $\binom{\mathbf{u}}{\mathbf{B}} \in H''$, the integral

$$\int_\Omega \Big\{ - Re\, \mathbf{uu} : \nabla\phi + \frac{c}{D} g(R) f(R) \mathbf{B} : \nabla\phi \quad - \mathbf{B} : (\mathbf{u}\cdot\nabla\chi) + g(R)\left[-\mathbf{B}\cdot\nabla\mathbf{u} - \nabla\mathbf{u}^T\cdot\mathbf{B} - \nabla\mathbf{u} - \nabla\mathbf{u}^T + \frac{f(R)}{D}\mathbf{B}\right] : \chi \Big\} d\mathbf{x} \qquad (6.1)$$

defines a linear functional of $\begin{pmatrix}\phi\\ \chi\end{pmatrix} \in H''$. It will now be shown that this is a bounded linear functional, by estimating each of the terms. In deriving the estimates, frequent use will be made of the following two inequalities, valid for any space in which

$\dot{W}_2^1(\Omega)$ or a subspace of $\dot{W}_2^1(\Omega)$ is dense:

$$\int_\Omega v^4 d\mathbf{x} \leq 4 \left(\int_\Omega v^2 d\mathbf{x} \right)^{\frac{1}{2}} \left(\int_\Omega \nabla v \cdot \nabla v^T d\mathbf{x} \right)^{\frac{3}{2}} \quad , \tag{6.2}$$

$$\int_\Omega v^2 d\mathbf{x} \leq \frac{1}{\mu_1} \int_\Omega \nabla v \cdot \nabla v^T d\mathbf{x} \quad . \tag{6.3}$$

Both these inequalities are proved by Ladyzhenskaya [6]; the latter is generally known as Poincaré's inequality, and μ_1 is a constant depending on the domain. In all that follows, K will be a generic designation for a positive constant; i.e., *it will not always be the same constant.*

$$\begin{aligned} \left| \int_\Omega Re\, \mathbf{uu} : \nabla \phi d\mathbf{x} \right| &\leq Re \sum_{i,j} \left[\int_\Omega (u_i u_j)^2 d\mathbf{x} \right]^{\frac{1}{2}} \left[\int_\Omega \frac{\partial \phi_j}{\partial x_i}^2 d\mathbf{x} \right]^{\frac{1}{2}} \\ &\leq Re \sum_{i,j} \left[\int_\Omega u_1^4 d\mathbf{x} \right]^{\frac{1}{4}} \left[\int_\Omega u_j^4 d\mathbf{x} \right]^{\frac{1}{4}} K \|\phi\|_H \\ &\leq K \|\mathbf{u}\|_H^2 \|\phi\|_H \quad . \end{aligned} \tag{6.4}$$

The fact that $g(R) \leq 1$ and that $g(R) = 0$ if $R \geq R_2$ will be used in some of the following estimates.

$$\begin{aligned} \left| \int_\Omega \frac{c}{D} g(R) f(R) \mathbf{B} : \nabla \phi d\mathbf{x} \right| &\leq \frac{c}{D} \sum_{i,j} \int_\Omega \left| g(R) f(R) B_{ij} \frac{\partial \phi_j}{\partial x_i} \right| d\mathbf{x} \\ &\leq \frac{c}{D} \sum_{i,j} f(R_2) \left[\int_\Omega B_{ij}^2 d\mathbf{x} \right]^{\frac{1}{2}} \left[\int_\Omega \frac{\partial \phi_j}{\partial x_i}^2 d\mathbf{x} \right]^{\frac{1}{2}} \\ &\leq K \|\mathbf{B}\|_{H'} \|\phi\|_H \quad . \end{aligned} \tag{6.5}$$

$$\left|\int_\Omega \mathbf{B} : (\mathbf{u}\cdot\nabla\chi) d\mathbf{x}\right| \le \sum_{i,j,k}\int_\Omega \left|B_{ij}u_k \frac{\partial \chi_{ij}}{\partial x_k}\right| d\mathbf{x}$$
$$\le \sum_{i,j,k}\left[\int_\Omega |B_{ij}u_k|^2 d\mathbf{x}\right]^{\frac{1}{2}}\left[\int_\Omega \left|\frac{\partial \chi_{ji}}{\partial x_k}\right|^2 d\mathbf{x}\right]^{\frac{1}{2}}$$
$$\le \sum_{i,j,k}\left[\int_\Omega |B_{ij}|^4 d\mathbf{x}\right]^{\frac{1}{4}}\left[\int_\Omega |u_k|^4 d\mathbf{x}\right]^{\frac{1}{4}}\|\chi\|_{H'}$$
$$\le \sum_{i,j,k} 16\left[\int_\Omega |B_{ij}|^2 d\mathbf{x}\right]^{\frac{1}{8}}\left[\int_\Omega \nabla B_{ij}\cdot\nabla B_{ij} d\mathbf{x}\right]^{\frac{3}{8}}\left[\int_\Omega |u_k|^2 d\mathbf{x}\right]^{\frac{1}{8}}$$
$$\times\left[\int_\Omega \nabla u_k\cdot\nabla u_k d\mathbf{x}\right]^{\frac{3}{8}} K\|\chi\|_{H'}$$
$$\le \sum_{i,j,k} 16K\|\mathbf{B}\|_{H'}\|\mathbf{u}\|_H\|\chi\|_{H'}$$
$$\le K\|\mathbf{B}\|_{H'}\|\mathbf{u}\|_H\|\chi\|_{H'} \quad . \tag{6.6}$$

$$\left|\int_\Omega g(R)\mathbf{B}\cdot\nabla\mathbf{u} : \chi d\mathbf{x}\right| \le \sum_{i,j,k}\int_\Omega \left|B_{ij}\frac{\partial u_k}{\partial x_j}\chi_{ki}\right| d\mathbf{x}$$
$$\le \sum_{i,j,k}\left[\int_\Omega |B_{ij}|^4 d\mathbf{x}\right]^{\frac{1}{4}}\left[\int_\Omega |\chi_{ki}|^4 d\mathbf{x}^{\frac{1}{4}}\right]\left[\int_\Omega \left(\frac{\partial u_k}{\partial x_j}\right)^2 d\mathbf{x}\right]^{\frac{1}{4}}$$
$$\le \sum_{i,j,k} 16K\|\mathbf{B}\|_{H'}\|\mathbf{u}\|_H\|\chi\|_{H'}$$
$$\le K\|\mathbf{B}\|_{H'}\|\mathbf{u}\|_H\|\chi\|_{H'} \quad . \tag{6.7a}$$

The estimate

$$\left|\int_\Omega g(R)\nabla\mathbf{u}^T\cdot\mathbf{B}\cdot\chi d\mathbf{x}\right| \le K\|\mathbf{u}\|_H\|\mathbf{B}\|_{H'}\|\chi\|_{H'} \tag{6.7b}$$

follows in a very similar way to the previous estimate. Analogously to (6.7a,b), the estimates

$$\left|\int_\Omega g(R)\nabla\mathbf{u} : \chi d\mathbf{x}\right| \leq K\|\mathbf{u}\|_H\|\chi\|_{H'} \tag{6.8a}$$

and

$$\left|\int_\Omega g(R)\nabla\mathbf{u}^T : \chi d\mathbf{x}\right| \leq K\|\mathbf{u}\|_H\|\chi\|_{H'} \tag{6.8b}$$

can be obtained. Finally,

$$\begin{aligned}\left|\int_\Omega \frac{g(R)f(R)}{D}\mathbf{B} : \chi d\mathbf{x}\right| &\leq \frac{f(R_2)}{D}\sum_{i,j}\int_\Omega |B_{ij}\chi_{ij}|d\mathbf{x} \\ &\leq \frac{f(R_2)}{D}\sum_{i,j}\left[\int_\Omega B_{ij}^2 d\mathbf{x}\right]^{\frac{1}{2}}\left[\int_\Omega \chi_{ji}^2 d\mathbf{x}\right]^{\frac{1}{2}} \\ &\leq \frac{f(R_2)}{D}K\|\mathbf{B}\|_{H'}\|\chi\|_{H'} \quad .\end{aligned} \tag{6.9}$$

So the entire functional is estimated by

$$\begin{aligned}&\int_\Omega\Big\{-Re\,\mathbf{uu} : \nabla\phi + \frac{c}{D}g(R)f(R)\mathbf{B} : \nabla\phi - \mathbf{B} : (\mathbf{u}\cdot\nabla\chi) \\ &\qquad + g(R)\big[-B\cdot\nabla\mathbf{u} - \nabla\mathbf{u}^T\cdot\mathbf{B} + \frac{f(R)}{D}\mathbf{B}\big] : \chi\Big\}d\mathbf{x} \\ &\leq K\|\mathbf{u}\|_H^2\|\phi\|_H + K\|\mathbf{B}\|_{H'}\|\phi\|_H \\ &\quad + K\|\mathbf{B}\|_{H'}\|\mathbf{u}\|_H\|\chi\|_{H'} + K\|\mathbf{B}\|_{H'}\|\mathbf{u}\|_H\|\chi\|_{H'} \\ &\quad + K\|\mathbf{u}\|_H\|\mathbf{B}\|_{H'}\|\chi\|_{H'} + K\|\mathbf{u}\|_H\|\chi\|_{H'} \\ &\quad + K\|\mathbf{u}\|_H\|\chi\|_{H'} + K\|\mathbf{B}\|_{H'}\|\chi\|_{H'} \\ &\leq K\left\|\binom{\phi}{\chi}\right\|_{H''}\end{aligned} \tag{6.10}$$

for a fixed $\binom{\mathbf{u}}{\mathbf{B}} \in \{H''\}$.

By the Riesz representation theorem, the functional can therefore be written as

$$\left[\mathbf{M}\begin{pmatrix}\mathbf{u}\\\mathbf{B}\end{pmatrix}, \begin{pmatrix}\phi\\\chi\end{pmatrix}\right]_{H''} \quad , \tag{6.11}$$

where $\mathbf{M}$ is a unique (9-component) element of H'' depending nonlinearly on $\binom{\mathbf{u}}{\mathbf{B}} \in H''$. This completes Step 1.

The next step, (Step 2), is to show that the operator taking $\binom{\mathbf{u}}{\mathbf{B}}$ to $\mathbf{M}\binom{\mathbf{u}}{\mathbf{B}}$ is completely continuous on H''. By definition, the operator is completely continuous if for any sequence

$$\left\{\begin{pmatrix}\mathbf{u}^{(m)}\\\mathbf{B}^{(m)}\end{pmatrix}\right\} \tag{6.12}$$

in H'' such that

$$\begin{pmatrix}\mathbf{u}^{(m)}\\\mathbf{B}^{(m)}\end{pmatrix} \rightarrow \begin{pmatrix}\mathbf{u}\\\mathbf{B}\end{pmatrix} \tag{6.13}$$

weakly in H'', it follows that

$$\left\|M\begin{pmatrix}\mathbf{u}^{(m)}\\\mathbf{B}^{(m)}\end{pmatrix} - M\begin{pmatrix}\mathbf{u}\\\mathbf{B}\end{pmatrix}\right\|_{H''} \rightarrow 0. \tag{6.14}$$

So consider a weakly convergent sequence and estimate the inner product

$$\left[M\begin{pmatrix}\mathbf{u}^{(m)}\\\mathbf{B}^{(m)}\end{pmatrix} - M\begin{pmatrix}\mathbf{u}^{(n)}\\\mathbf{B}^{(n)}\end{pmatrix}, \begin{pmatrix}\phi\\\chi\end{pmatrix}\right]_{H''}$$

$$= -(I) + (II) - (III) - (IV) - (V) - (VI) - (VII) - (VIII), \tag{6.15}$$

where

$$(I) = \int_\Omega Re\, \mathbf{u}^{(m)}\mathbf{u}^{(m)} : \nabla\phi d\mathbf{x} - \int_\Omega Re\, \mathbf{u}^{(n)}\mathbf{u}^{(n)} : \nabla\phi d\mathbf{x}\ , \qquad (6.16a)$$

$$(II) = \int_\Omega \frac{c}{D} g(R^{(m)}) f(R^{(m)}) \mathbf{B}^{(m)} : \nabla\phi d\mathbf{x} - \int_\Omega \frac{c}{D} g(R^{(n)}) f(R^{(n)}) \mathbf{B}^{(n)} : \nabla\phi d\mathbf{x}\ , \qquad (6.16b)$$

$$(III) = \int_\Omega \mathbf{B}^{(m)} : (\mathbf{u}^{(m)} \cdot \nabla\chi) d\mathbf{x} - \int_\Omega \mathbf{B}^{(n)} : (\mathbf{u}^{(n)} \cdot \nabla\chi) d\mathbf{x}\ , \qquad (6.16c)$$

$$(IV) = \int_\Omega g(R^{(m)}) \mathbf{B}^{(m)} \cdot \nabla\mathbf{u}^{(m)} : \chi d\mathbf{x} - \int_\Omega g(R^{(n)}) \mathbf{B}^{(n)} \cdot \nabla\mathbf{u}^{(n)} : \chi d\mathbf{x}\ , \qquad (6.16d)$$

$$(V) = \int_\Omega g(R^{(m)}) \nabla\mathbf{u}^{(m)^T} \cdot \mathbf{B}^{(m)} : \chi d\mathbf{x} - \int_\Omega g(R^{(n)}) \nabla\mathbf{u}^{(n)^T} \cdot \mathbf{B}^{(n)} : \chi d\mathbf{x}\ , \qquad (6.16e)$$

$$(VI) = \int_\Omega g(R^{(m)}) \nabla\mathbf{u}^{(m)} : \chi d\mathbf{x} - \int_\Omega g(R^{(n)}) \nabla\mathbf{u}^{(n)} : \chi d\mathbf{x}\ , \qquad (6.16f)$$

$$(VII) = \int_\Omega g(R^{(m)}) \nabla\mathbf{u}^{(m)^T} : \chi d\mathbf{x} - \int_\Omega g(R^{(n)}) \nabla\mathbf{u}^{(n)^T} : \chi d\mathbf{x}\ , \qquad (6.16g)$$

$$(VIII) = \int_\Omega \frac{g(R^{(m)}) f(R^{(m)})}{D} \mathbf{B}^{(m)} : \chi d\mathbf{x} - \int_\Omega \frac{g(R^{(n)}) f(R^{(n)})}{D} \mathbf{B}^{(n)} : \chi d\mathbf{x}. \qquad (6.16g)$$

These eight terms are estimated using Hölder's inequality, the two inequalities (6.2) and (6.3), and integration by parts. For example, the necessary steps for term (I) are shown in detail below; first it is rewritten as

$$
\begin{aligned}
(I) = &\int_\Omega Re\, \mathbf{u}^{(m)}(\mathbf{u}^{(m)} - \mathbf{u}^{(n)}) : \nabla\phi d\mathbf{x} \\
&+ \int_\Omega Re\, \mathbf{u}^{(n)}(\mathbf{u}^{(m)} - \mathbf{u}^{(n)}) : \nabla\phi d\mathbf{x} \quad ,
\end{aligned} \tag{6.17}
$$

and then it is estimated by

$$
\begin{aligned}
|(I)| \le\ & Re \sum_{i,j} \int_\Omega \left| u_i^{(m)} (u_j^{(m)} - u_j^{(n)}) \frac{\partial \phi_i}{\partial x_j} \right| d\mathbf{x} \\
& + Re \sum_{i,j} \int_\Omega \left| u_i^{(m)} (u_j^{(m)} - u_j^{(n)}) \frac{\partial \phi_i}{\partial x_j} \right| d\mathbf{x} \\
\le\ & Re \sum_{i,j} \left[\int_\Omega \left| u_i^{(m)} (u_j^{(m)} - u_j^{(n)}) \right|^2 d\mathbf{x} \right]^{\frac{1}{2}} \left[\int_\Omega \left(\frac{\partial \phi_i}{\partial x_j}\right)^2 d\mathbf{x} \right]^{\frac{1}{2}} \\
& + Re \sum_{i,j} \left[\int_\Omega \left| u_i^{(m)} (u_j^{(m)} - u_j^{(n)}) \right|^2 d\mathbf{x} \right]^{\frac{1}{2}} \left[\int_\Omega \left(\frac{\partial \phi_i}{\partial x_j}\right)^2 d\mathbf{x} \right]^{\frac{1}{2}} \\
\le\ & Re \sum_{i,j} \left[\int_\Omega (u_i^{(m)})^4 d\mathbf{x} \right]^{\frac{1}{4}} \left[\int_\Omega (u_j^{(m)} - u_j^{(n)})^4 d\mathbf{x} \right]^{\frac{1}{4}} \left[\int_\Omega \left(\frac{\partial \phi_i}{\partial x_j}\right)^2 d\mathbf{x} \right]^{\frac{1}{2}} \\
& + \left[\int_\Omega (u_i^{(n)})^4 d\mathbf{x} \right]^{\frac{1}{4}} \left[\int_\Omega (u_j^{(m)} - u_j^{(n)})^4 d\mathbf{x} \right]^{\frac{1}{4}} \left[\int_\Omega \left(\frac{\partial \phi_i}{\partial x_j}\right)^2 d\mathbf{x} \right]^{\frac{1}{2}} \\
\le\ & Re\ K \left\|\mathbf{u}^{(m)}\right\|_{[L_4(\Omega)]^3} \left\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3} \|\phi\|_H \\
& + Re\ K \left\|\mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3} \left\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3} \|\phi\|_H \quad .
\end{aligned} \tag{6.18}
$$

Similarly, term (II) is first rewritten as two integrals:

$$
(II) = \frac{c}{D} \int_\Omega g(R^{(m)}) f(R^{(m)}) (\mathbf{B}^{(m)} - \mathbf{B}^{(n)}) \nabla\phi d\mathbf{x}
$$

$$+\frac{c}{D}\int_\Omega \left(g(R^{(m)})f(R^{(m)}) - g(R^{(n)})f(R^{(n)})\right)\mathbf{B}^{(n)} : \nabla\phi d\mathbf{x} \quad . \tag{6.19}$$

In estimating (II), use is made of the fact that $g(R)f(R)$ has an upper bound of $f(R_2)$ (recall that for $R \geq R_2$ the dumbbells have all broken, so that $g(R) = 0$).

$$\begin{aligned}|(II)| &\leq \frac{c}{D}f(R_2)\int_\Omega \left|(\mathbf{B}^{(m)} - \mathbf{B}^{(n)}) : \nabla\phi\right| d\mathbf{x} \\ &\quad + \frac{c}{D}\int_\Omega \left|(g(R^{(m)})f(R^{(m)}) - g(R^{(n)})f(R^{(n)}))\mathbf{B}^{(n)} : \nabla\phi\right| d\mathbf{x} \\ &\leq K\|\mathbf{B}^{(m)} - \mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9}\|\phi\|_H \\ &\quad + K\|g(R^{(m)})f(R^{(m)}) - g(R^{(n)})f(R^{(n)})\|_{L_4(\Omega)}\|\mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9}\|\phi\|_H \quad .\end{aligned} \tag{6.20}$$

By rewriting term (III) as

$$\begin{aligned}(III) &= \int_\Omega (\mathbf{B}^{(m)} - \mathbf{B}^{(n)}) : (\mathbf{u}^{(m)} \cdot \nabla\chi) d\mathbf{x} \\ &\quad + \int_\Omega \mathbf{B}^{(n)} : \left((\mathbf{u}^{(m)} - \mathbf{u}^{(n)}) \cdot \chi\right) d\mathbf{x} \quad ,\end{aligned} \tag{6.21}$$

it is clear that

$$\begin{aligned}|(III)| &\leq K\|\mathbf{B}^{(m)} - \mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9}\|\mathbf{u}^{(m)}\|_{[L_4(\Omega)]^3}\|\chi\|_{H'} \\ &\quad + K\ \|\mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9}\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\|_{[L_4(\Omega)]^3}\|\chi\|_{H'} \quad .\end{aligned} \tag{6.22}$$

Now rewrite Term (IV) as

$$(IV) = \int_\Omega [g(R^{(m)}) - g(R^{(n)})]\mathbf{B}^{(m)} \cdot \nabla\mathbf{u}^{(m)} : \chi d\mathbf{x}$$

$$
\begin{aligned}
&+\int_\Omega g(R^{(n)})[\mathbf{B}^{(m)}-\mathbf{B}^{(n)}]\cdot\nabla\mathbf{u}^{(m)}:\chi dx \\
&+\int_\Omega g(R^{(n)})\mathbf{B}^{(n)}\cdot(\nabla\mathbf{u}^{(m)}-\nabla\mathbf{u}^{(n)}):\chi dx \\
&=(IVa)+(IVb)+(IVc).
\end{aligned}
\tag{6.23}
$$

Now the finite extensibility of the dumbbells will again be used; this means that R is bounded by R_2 (and certainly by L !). Since $R^2 = Tr\mathbf{A} = Tr\mathbf{B}+3$, the diagonal components of $\mathbf{B}$ are therefore bounded. Since the equations are frame-invariant (this concept is discussed in detail in [7]), the off-diagonal components of $\mathbf{B}$ are also bounded. (This fact could have been used earlier in any of the estimates involving $\mathbf{B}$, but it may be useful to make it clear exactly where the finite extensibility is really needed.) Consequently,

$$
|(IVa)|\le K\|g(R^{(m)})-g(R^{(n)})\|_{L_4(\Omega)}\|\mathbf{u}^{(m)}\|_H\|\chi\|_{[L_4(\Omega)]^9} \quad . \tag{6.24}
$$

To estimate (IVb), use the fact that $g(R)\le 1$ always:

$$
|(IVb)|\le K\|\mathbf{B}^{(m)}-\mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9}\|\mathbf{u}^{(m)}\|_H\|\chi\|_{[L_4(\Omega)]^9} \quad . \tag{6.25}
$$

Term (IVc) is somewhat more involved, since an integration by parts is necessary:

$$
\begin{aligned}
(IVc) &= \int_\Omega g(R^{(n)})B_{ij}^{(n)}\Big(\frac{\partial u_k^{(m)}}{\partial x_j}-\frac{\partial u_k^{(n)}}{\partial x_j}\Big)\chi_{ki}d\mathbf{x} \\
&= \int_\Omega h(R^{(n)})\frac{\partial Tr(\mathbf{B}^{(n)})}{\partial x_j}B_{ij}^{(n)}(u_k^{(m)}-u_k^{(n)})\chi_{ki}d\mathbf{x} \\
&+\int_\Omega g(R^{(n)})\frac{\partial B_{ij}^{(n)}}{\partial x_j}(u_k^{(m)}-u_k^{(n)})\chi_{ki}d\mathbf{x} \\
&+\int_\Omega g(R^{(n)})B_{ij}^{(n)}(u_k^{(m)}-u_k^{(n)})\frac{\partial \chi_{ki}}{\partial x_j}d\mathbf{x} \quad ,
\end{aligned}
\tag{6.26}
$$

where for convenience $h(R)$ denotes the derivative of $g(R)$ with respect to R^2 rather than R. Note that $h(R)$ is bounded except at $R = 0$ and $R = R_2$ (where it is discontinuous), but since it is defined everywhere except on a set of measure zero, it exists as a generalized derivative, and its points of discontinuity can be ignored as far as the integrals are concerned. Again, by using the fact that all components of **B** are bounded, it should be clear that

$$\begin{aligned} |(IVc)| \le\ & K\|\mathbf{B}\|_{H'}\left\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3}\|\chi\|_{[L_4(\Omega)]^9} \\ &+ K\|\mathbf{B}\|_{H'}\left\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3}\|\chi\|_{[L_4(\Omega)]^9} \\ &+ K\left\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3}\|\chi\|_{H'} \quad . \end{aligned} \tag{6.27}$$

Term (V) can be handled in a very similar way to term (IV), and the result is the same as for (IV). Terms (VI) and (VII) take fewer (analogous) steps to estimate than (IV) and (V), because their integrands do not contain the **B** factors; thus

$$\begin{aligned} (VI),(VII) \le\ & K\left\|g(R^{(m)}) - g(R^{(n)})\right\|_{L_4(\Omega)}\left\|\mathbf{u}^{(m)}\right\|_{H}\left\|\chi\right\|_{[L_4(\Omega)]^9} \\ &+K\left\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3}\|\chi\|_{H'} \quad . \end{aligned} \tag{6.28}$$

Finally, term (VIII) is rewritten as

$$\begin{aligned} (VIII) =\ & \int_\Omega \frac{g(R^{(m)})f(R^{(m)})}{D}(\mathbf{B}^{(m)} - \mathbf{B}^{(n)}) : \chi d\mathbf{x} \\ &+ \int_\Omega (\frac{g(R^{(m)})f(R^{(m)})}{D} - \frac{g(R^{(n)})f(R^{(n)})}{D})\mathbf{B}^{(n)} : \chi d\mathbf{x} \ , \end{aligned} \tag{6.29}$$

from which the estimate

$$|(VIII)| \leq K \; \|g(R^{(m)})f(R^{(m)}) - g(R^{(n)})f(R^{(n)})\|_{L_2(\Omega)} \|\mathbf{B}^{(m)}\|_{[L_4(\Omega)]^9} \|\chi\|_{[L_4(\Omega)]^9}$$
$$+ K \; \|\mathbf{B}^{(m)} - \mathbf{B}^{(n)}\|_{[L_2(\Omega)]^9} \|\chi\|_{[L_2(\Omega)]^9} \tag{6.30}$$

is easily obtained, where again the fact that $g(R)f(R)$ is a bounded function has been used.

All the above estimates of the eight integral terms can be put together to obtain:

$$\left| \left[M \begin{pmatrix} \mathbf{u}^{(m)} \\ \mathbf{B}^{(m)} \end{pmatrix} - M \begin{pmatrix} \mathbf{u}^{(n)} \\ \mathbf{B}^{(n)} \end{pmatrix}, \begin{pmatrix} \phi \\ \chi \end{pmatrix} \right]_{H''} \right|$$

$$\leq K \; \left\|\mathbf{u}^{(m)}\right\|_{[L_4(\Omega)]^3} \left\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3} \|\phi\|_H$$
$$+ K \; \left\|\mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3} \left\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\right\|_{[L_4(\Omega)]^3} \|\phi\|_H$$
$$+ K \; \|\mathbf{B}^{(m)} - \mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9} \|\phi\|_H$$
$$+ K \; \|g(R^{(m)})f(R^{(m)}) - g(R^{(n)})f(R^{(n)})\|_{L_4(\Omega)} \|\mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9} \|\phi\|_H$$
$$+ K \; \|\mathbf{B}^{(m)} - \mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9} \|\mathbf{u}^{(m)}\|_{[L_4(\Omega)]^3} \|\chi\|_{H'}$$
$$+ K \; \|\mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9} \|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\|_{[L_4(\Omega)]^3} \|\chi\|_{H'}$$
$$+ K \; \|g(R^{(m)}) - g(R^{(n)})\|_{L_4(\Omega)} \|\mathbf{u}^{(m)}\|_H \|\chi\|_{[L_4(\Omega)]^9}$$
$$+ K \; \|\mathbf{B}^{(m)} - \mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9} \|\mathbf{u}^{(m)}\|_H \|\chi\|_{[L_4(\Omega)]^9}$$
$$+ K \; \|\mathbf{B}\|_{H'} \|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\|_{[L_4(\Omega)]^3} \|\chi\|_{[L_4(\Omega)]^9}$$
$$+ K \; \|\mathbf{B}\|_{H'} \|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\|_{[L_4(\Omega)]^3} \|\chi\|_{[L_4(\Omega)]^9}$$
$$+ K \; \|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\|_{[L_4(\Omega)]^3} \|\chi\|_{H'}$$
$$+ K \; \|g(R^{(m)}) - g(R^{(n)})\|_{L_4(\Omega)} \|\mathbf{u}^{(m)}\|_H \|\chi\|_{[L_4(\Omega)]^9}$$
$$+ K \; \|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\|_{[L_4(\Omega)]^3} \|\chi\|_{H'}$$

$$+ K \ \|g(R^{(m)})f(R^{(m)}) - g(R^{(n)})f(R^{(n)})\|_{L_2(\Omega)} \|\mathbf{B}^{(m)}\|_{[L_4(\Omega)]^9} \|\chi\|_{[L_4(\Omega)]^9}$$

$$+ K \ \|\mathbf{B}^{(m)} - \mathbf{B}^{(n)}\|_{[L_2(\Omega)]^9} \|\chi\|_{[L_2(\Omega)]^9} \quad . \tag{6.31}$$

To simplify this, note first that $\mathbf{u}^{(m)}$ and $\mathbf{u}^{(n)}$ are bounded in $[L_4(\Omega)]^3$ and in H, and that $\mathbf{B}^{(m)}$ and $\mathbf{B}^{(n)}$ are bounded in $[L_4(\Omega)]^9$ and in H'. Then note that

$$\|\chi\|_{[L_2(\Omega)]^9} \ , \ \|\chi\|_{[L_4(\Omega)]^9} \ , \ \|\chi\|_{H'} \qquad \text{and} \qquad \|\phi\|_H \ \leq \ \left\| \begin{pmatrix} \phi \\ \chi \end{pmatrix} \right\|_{H''} \quad . \tag{6.32}$$

This allows the inequality to be rewritten as

$$\begin{aligned}
&\left| \left[\mathbf{M} \begin{pmatrix} \mathbf{u}^{(m)} \\ \mathbf{B}^{(m)} \end{pmatrix} - \mathbf{M} \begin{pmatrix} \mathbf{u}^{(n)} \\ \mathbf{B}^{(n)} \end{pmatrix} , \begin{pmatrix} \phi \\ \chi \end{pmatrix} \right]_{H''} \right| \\
&\leq K \ \Big[\|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\|_{[L_4(\Omega)]^3} \\
&+ \|\mathbf{B}^{(m)} - \mathbf{B}^{(n)}\|_{[L_4(\Omega)]^9} \\
&+ \|g(R^{(m)})f(R^{(m)}) - g(R^{(n)})f(R^{(n)})\|_{L_4(\Omega)} \\
&+ \|g(R^{(m)}) - g(R^{(n)})\|_{L_4(\Omega)} \Big] \\
&\times \left\| \begin{matrix} \phi \\ \chi \end{matrix} \right\|_{H''} \quad .
\end{aligned} \tag{6.33}$$

Now by choosing

$$\begin{pmatrix} \phi \\ \chi \end{pmatrix} = \mathbf{M} \begin{pmatrix} \mathbf{u}^{(m)} \\ \mathbf{B}^{(m)} \end{pmatrix} - \mathbf{M} \begin{pmatrix} \mathbf{u}^{(n)} \\ \mathbf{B}^{(n)} \end{pmatrix} , \tag{6.34}$$

it is clear that

$$\left\| \mathbf{M} \begin{pmatrix} \mathbf{u}^{(m)} \\ \mathbf{B}^{(m)} \end{pmatrix} - \mathbf{M} \begin{pmatrix} \mathbf{u}^{(n)} \\ \mathbf{B}^{(n)} \end{pmatrix} \right\|_{H''} \leq K \ \|\mathbf{u}^{(m)} - \mathbf{u}^{(n)}\|_{[L_4(\Omega)]^3}$$

$$+K\left\|\mathbf{B}^{(m)}-\mathbf{B}^{(n)}\right\|_{[L_4(\Omega)]^9}$$
$$+K\left\|g(R^{(m)})f(R^{(m)})-g(R^{(n)})f(R^{(n)})\right\|_{L_4(\Omega)}$$
$$+K\left\|g(R^{(m)})-g(R^{(n)})\right\|_{L_4(\Omega)} \quad . \tag{6.35}$$

At this point, recall the fact (a proof of which is given by Ladyzhenskaya [6]) that if a sequence is weakly convergent in $\dot{W}_2^1$, H, H', or H'', then it is strongly convergent in $[L_4(\Omega)]^q$, where $q = 1, 3, 9, 12$, whichever is the appropriate dimension of the space. In considering the behavior of this expression as n and $m \to \infty$ the fact that $g(R)f(R)$ is a bounded and continuous function of R should be noted; this implies that if

$$\left\{R^{(m)}\right\} \tag{6.36a}$$

is a strongly convergent sequence, then

$$\left\{g(R^{(m)})f(R^{(m)})\right\} \tag{6.36b}$$

is also strongly convergent. From these remarks and the above inequality, it is clear that

$$\left\|\mathbf{M}\binom{\mathbf{u}^{(m)}}{\mathbf{B}^{(m)}}-\mathbf{M}\binom{\mathbf{u}^{(n)}}{\mathbf{B}^{(n)}}\right\|_{H''} \to 0 \tag{6.37}$$

as $m, n \to \infty$. This concludes the proof that the operator $\mathbf{M}$ is completely continuous, and thus completes Step 2.

At this point, the statement of the problem is:

Find $\binom{\mathbf{u}}{\mathbf{B}} \in H''$ such that

$$\left[\mathbf{M}\binom{\mathbf{u}}{\mathbf{B}},\binom{\phi}{\chi}\right]_{H''} + [\mathbf{u},\phi]_H + \epsilon[\mathbf{B},\chi]_{H'}$$
$$-\int_\Omega \mathbf{F}(\mathbf{x})\cdot\phi d\mathbf{x} = 0 \tag{6.38}$$

holds for all $\binom{\phi}{\chi} \in H''$, where $\mathbf{M}$ is completely continuous as a function of $\binom{\mathbf{u}}{\mathbf{B}}$. Now comes Step 3, the reformulation of the problem as that of guaranteeing existence of at least one fixed point of a certain operator. For a reasonable forcing function $\mathbf{F}(\mathbf{x})$, the Riesz representation theorem can again be applied to assure existence of an element $\tilde{\mathbf{F}}$ in H' such that

$$\int_\Omega \mathbf{F}(\mathbf{x})\cdot\phi d\mathbf{x} = \left[\binom{\tilde{\mathbf{F}}}{\mathbf{0}},\binom{\phi}{\chi}\right]_{H''} \quad . \tag{6.39}$$

Note that

$$[\mathbf{u},\phi]_H + \epsilon[\mathbf{B},\chi]_{H'} = \left[\binom{\mathbf{u}}{\mathbf{B}},\binom{\phi}{\epsilon\chi}\right]_{H''} \quad . \tag{6.40}$$

Clearly, it is possible to define a new operator $\tilde{\mathbf{M}}\binom{\mathbf{u}}{\mathbf{B}}$, which is also completely continuous, such that

$$\left[\mathbf{M}\binom{\mathbf{u}}{\mathbf{B}},\binom{\phi}{\chi}\right]_{H''} = \left[\tilde{\mathbf{M}}\binom{\mathbf{u}}{\mathbf{B}},\binom{\phi}{\epsilon\chi}\right]_{H''} \quad . \tag{6.41}$$

Thus the problem becomes:

Find $\binom{\mathbf{u}}{\mathbf{B}} \in H''$ such that

$$\left[\tilde{\mathbf{M}}\binom{\mathbf{u}}{\mathbf{B}},\binom{\phi}{\epsilon\chi}\right]_{H''} + \left[\binom{\mathbf{u}}{\mathbf{B}},\binom{\phi}{\epsilon\chi}\right]_{H''}$$
$$-\left[\binom{\tilde{\mathbf{F}}}{\mathbf{0}},\binom{\phi}{\epsilon\chi}\right]_{H''} = 0 \tag{6.42}$$

holds for all $\binom{\phi}{\chi} \in H''$, which can be restated as:

Find $\binom{\mathbf{u}}{\mathbf{B}} \in H''$ such that

$$\tilde{\mathrm{M}}\binom{\mathbf{u}}{\mathbf{B}} + \binom{\mathbf{u}}{\mathbf{B}} - \binom{\tilde{\mathbf{F}}}{\mathbf{0}} = 0, \tag{6.43}$$

which is equivalent to looking for a fixed point of the completely continuous operator

$$\binom{\tilde{\mathbf{F}}}{\mathbf{0}} - \tilde{\mathrm{M}}\binom{\mathbf{u}}{\mathbf{B}}. \tag{6.44}$$

This completes Step 3. By the Leray-Schauder theorem, at least one fixed point will exist if it is true that all possible solutions of

$$\binom{\mathbf{u}}{\mathbf{B}} = \lambda\Big[\binom{\tilde{\mathbf{F}}}{\mathbf{0}} - \tilde{\mathrm{M}}\binom{\mathbf{u}}{\mathbf{B}}\Big] \tag{6.45}$$

for $\lambda \in [0,1]$ lie in a bounded subset of H''. This equation can be rewritten as

$$\lambda\tilde{\mathrm{M}}\binom{\mathbf{u}}{\mathbf{B}} + \binom{\mathbf{u}}{\mathbf{B}} - \lambda\binom{\tilde{\mathbf{F}}}{\mathbf{0}} = 0. \tag{6.46}$$

That all possible solutions are bounded in this space will now be shown (Step 4, the derivation of *a priori* estimates).

First, take the inner product of (6.46) with

$$\binom{\mathbf{0}}{f(R)\mathbf{I}}. \tag{6.47}$$

The first term of this is

$$\lambda\Big[\tilde{\mathrm{M}}\binom{\mathbf{u}}{\mathbf{B}}, \binom{\mathbf{0}}{f(R)\mathbf{I}}\Big]_{H''} = \frac{\lambda}{\epsilon}\int_\Omega \Big\{-\mathbf{B} : (\mathbf{u}\cdot\nabla(f(R)\mathbf{I}))$$
$$+ g(R)\big[-\mathbf{B}\cdot\nabla\mathbf{u} - \nabla\mathbf{u}^T\cdot\mathbf{B} + \frac{f(R)}{D}\mathbf{B}\big] : f(R)\mathbf{I}\Big\}\,d\mathrm{x}$$

$$
\begin{aligned}
&= \frac{\lambda}{\epsilon}\int_\Omega \Big\{-(R^2+3)[f(R)]^2\mathbf{u}\cdot\nabla R^2 \\
&\quad - 2g(R)f(R)\mathbf{B}:\nabla\mathbf{u} \\
&\quad + \frac{g(R)[f(R)]^2}{D}(R^2+3)\Big\}d\mathbf{x} \ . \qquad (6.48)
\end{aligned}
$$

By noting that $(R^2+3)[f(R)]^2\nabla R^2 = \nabla q(R)$, where $q(R)$ is some function, and by then integrating by parts, recalling that $\mathbf{u} = \mathbf{0}$ on the boundary, and that $\nabla\cdot\mathbf{u} = 0$, it can be shown that

$$
\int_\Omega \Big\{-(R^2+3)[f(R)]^2\mathbf{u}\cdot\nabla R^2\Big\}d\mathbf{x} = 0. \qquad (6.49)
$$

The next term in the inner product is

$$
\begin{aligned}
\left[\begin{pmatrix}\mathbf{u}\\ \mathbf{B}\end{pmatrix}, \begin{pmatrix}\mathbf{0}\\ f(R)\mathbf{I}\end{pmatrix}\right]_{H''} &= \int_\Omega \nabla\mathbf{B} \vdots \nabla(f(R)\mathbf{I})d\mathbf{x} \\
&= \int_\Omega \nabla R^2\cdot\nabla f(R)d\mathbf{x} \\
&= \int_\Omega [f(R)]^2\nabla R^2\cdot\nabla R^2 d\mathbf{x}, \qquad (6.50)
\end{aligned}
$$

and the last term in the inner product of (6.46) with (6.47) is zero by orthogonality. The result is the following equation, which must hold for all possible solutions:

$$
\begin{aligned}
&\frac{\lambda}{\epsilon}\int_\Omega \Big[-2g(R)f(R)\mathbf{B}:\nabla\mathbf{u} + \frac{g(R)[f(R)]^2}{D}(R^2+3)\Big]d\mathbf{x} \\
&\qquad + \int_\Omega [f(R)]^2\nabla R^2\cdot\nabla R^2 d\mathbf{x} = 0 \ . \qquad (6.51)
\end{aligned}
$$

Equation (6.51) will be used soon. Now take the inner product of (6.46) with $\binom{\mathbf{u}}{\mathbf{0}}$. The first term is:

$$\lambda\left[\tilde{\mathbf{M}}\begin{pmatrix}\mathbf{u}\\ \mathbf{B}\end{pmatrix}, \begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}\right]_{H''} = \lambda\int_\Omega\Big\{-Re\ \mathbf{u}\mathbf{u}:\nabla\mathbf{u} + \frac{c}{D}g(R)f(R)\mathbf{B}:\nabla\mathbf{u}\Big\}d\mathbf{x}\ , \tag{6.52}$$

where once again the first term in the integrand integrates to zero on account of continuity and the fact that $\mathbf{u} = \mathbf{0}$ on the boundary. The next term is

$$\left[\begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}, \begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}\right]_{H''} \quad \Big(= \int_\Omega \nabla\mathbf{u}:\nabla\mathbf{u}d\mathbf{x} = [\mathbf{u},\mathbf{u}]_H\Big), \tag{6.53}$$

and the third term is, of course, $\lambda[\mathbf{F},\mathbf{u}]_H$, all resulting in the equation

$$\lambda\int_\Omega \frac{c}{D}g(R)f(R)\mathbf{B}:\nabla\mathbf{u}d\mathbf{x} + \left[\begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}, \begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}\right]_{H''} - \lambda[\mathbf{F},\mathbf{u}]_H = 0. \tag{6.54}$$

Substitute (6.51) into this to get

$$\left[\begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}, \begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}\right]_{H''} + \lambda\frac{1}{2}\frac{c}{D}\int_\Omega \frac{g(R)[f(R)]^2}{D}(R^2+3)d\mathbf{x} + \frac{\epsilon}{2}\frac{c}{D}\int_\Omega [f(R)]^2\nabla R^2\cdot\nabla R^2 d\mathbf{x} = \lambda[\mathbf{F},\mathbf{u}]_H\quad , \tag{6.55}$$

which, recalling that $g(R) = 0$ if $R > R_2$, and that the domain Ω was assumed bounded (with some finite volume V_Ω), gives

$$\left[\begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}, \begin{pmatrix}\mathbf{u}\\ \mathbf{0}\end{pmatrix}\right]_{H''} + \frac{\epsilon}{2}\frac{c}{D}\int_\Omega [f(R)]^2\nabla R^2\cdot\nabla R^2 d\mathbf{x} \leq -\frac{1}{2}\frac{c}{D}\frac{[f(R_2)]^2}{D}(R_2^2+3)V_\Omega + \lambda K\|\mathbf{F}\|_H\|\mathbf{u}\|_H\quad . \tag{6.56}$$

By definition, $[\mathbf{u},\mathbf{u}]_H = \|\mathbf{u}\|_H^2$ and, since $f(R) \geq 1$,

$$\frac{c}{2D}\int_\Omega [f(R)]^2 \nabla R^2 \cdot \nabla R^2 dx \geq \frac{c}{2D}[R^2, R^2]_{\dot{W}_2^1}$$
$$= \frac{c}{2D}\|R^2\|_{\dot{W}_2^1} \quad , \tag{6.57}$$

from which it is clear that $\|\mathbf{u}\|_H$ and $\|R^2\|_{\dot{W}_2^1}$ are bounded. As mentioned above, the pointwise bound on $R^2 = Tr\mathbf{B}+3$ implies that *all* components of $\mathbf{B}$ are bounded pointwise. To get a bound on $\mathbf{B}$ in H' from the bounds on $\mathbf{u}$ in H and R^2 in $\dot{W}_2^1$ which have just been obtained, take the inner product of (5.1b) with $\mathbf{B}$ to get

$$0 = \int_\Omega g(R)\mathbf{B}\cdot\nabla\mathbf{u} : \mathbf{B}dx + \int_\Omega g(R)\nabla\mathbf{u}^T\cdot\mathbf{B} : \mathbf{B}dx$$
$$- \int_\Omega \frac{g(R)f(R)}{D}\mathbf{B} : \mathbf{B}dx + \epsilon\int_\Omega \nabla^2\mathbf{B} : \mathbf{B}dx \ , \tag{6.58}$$

which gives

$$\epsilon\|\mathbf{B}\|_{H'}^2 \leq K\|u\|_H + \frac{g(R_2)f(R_2)}{D}(9L^2)V_\Omega \leq K\|u\|_\Omega + const., \tag{6.59}$$

the desired bound. Thus, the Leray-Schauder theorem can be applied (Step 5) to assure existence of at least one fixed point. Since it was shown earlier that a fixed point corresponds to a weak solution of the original problem, this completes the existence proof. It should be possible to extend the methods that Ladyzhenskaya [6] uses for the Navier Stokes equations on unbounded domains to this problem.

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CHAPTER II

Criteria for General Stability of Bounded Flows of a Non-Newtonian Fluid

Serrin [1] has given stability criteria for general solutions to the Navier-Stokes equations. He was able to obtain these because the presence of the viscosity term makes it possible to place an upper negative bound on the rate of growth of the energy integral for sufficiently low Reynolds number. While many flows of interest are at a higher Reynolds number than his results apply to, it is nevertheless useful to know that a large class of flows are stable if they are sufficiently slow. One of the implications of this result is that it implies the existence of a critical Reynolds number associated with the onset of instability for a quite general class of flows.

Given the interest in non-Newtonian flows and their stability, it would clearly be useful to know if a similar stability result can be obtained for low enough Reynolds number and (possibly) some range of Deborah number. It will be shown here that criteria similar to Serrin's criteria for universal stability can be derived for a FENE dumbbell model used by Chilcott and Rallison [2] to describe non-Newtonian fluids. The equations for this model are

$$Re\left(\frac{\partial \mathbf{U}}{\partial t} + \mathbf{U}\cdot\nabla\mathbf{U}\right) = -\nabla P + \nabla^2\mathbf{U} + \frac{c}{D}\nabla\cdot[f(R)\mathbf{A}], \tag{1}$$

$$\nabla\cdot\mathbf{u} = 0, \tag{2}$$

$$\frac{\partial \mathbf{A}}{\partial t} + \mathbf{U}\cdot\nabla\mathbf{A} = \mathbf{A}\cdot\nabla\mathbf{U} + (\nabla\mathbf{U})^T\cdot\mathbf{A} - \frac{f(R)}{D}(\mathbf{A}-\mathbf{I}) + \epsilon\nabla^2\mathbf{A}, \tag{3}$$

where the spring force $f(R)$ is given by

$$f(R) = \frac{1}{1 - R^2/L^2}, \tag{4}$$

with

$$R^2 = Tr\mathbf{A}, \tag{5}$$

so that the force becomes infinite at the "maximum extension" L. The use of a *finitely* extendible dumbbell model will be shown to be critical to the derivation of these criteria, as without the bound on the moment of the end-to-end displacement vector, certain integrals cannot be bounded.

The assumption will thus be made that the presence of the nonlinear spring term, which tends to contract the dumbbells more and more strongly as they approach their maximum elongation, will keep the dumbbell "lengths" $R < L$. This assumption should in no way restrict the physical meaning of the results here. For indeed, if it was possible to achieve a solution to the equations that violated this inequality, starting with initial conditions that did not, it would mean that the equations allowed completely meaningless behavior (from a physical standpoint). If R exceeds L, the spring force changes sign, corresponding to a force tending to push the two ends of the dumbbells apart!

Since the goal here is to present a mathematical proof of stability, it is desirable to justify this assumption from a mathematical standpoint also, especially since the constitutive equation does not correspond *exactly* to the original model of a dumbbell in a flow with certain stochastic forces acting on it. The assumption of preaveraging is made in deriving the constitutive equation, so that while it is clear that an actual dumbbell cannot overcome an infinite spring force with the result that its beads fly apart, it is not entirely clear that all possible solutions of the equations of motion will satisfy $R < L$. Therefore, some comments will now be made to provide some (mathematical) justification for the assumption that the average length of the end-to-end vector R in the FENE dumbbell model (1) cannot exceed L, the value of R at which the spring force becomes infinite.

It is convenient to speak in terms of generalized function solutions. If R exceeds L anywhere in the flow domain, there will be some streamline or streamsurface on one side of which $f(R) > 0$ and on the other side of which $f(R) < 0$, and on which $f(R) = \infty$. Thus, around this surface, $f(R)$ will behave like the function $\delta'(\mathbf{x}-\mathbf{x}_s)$, where δ is the Dirac delta function, and $\mathbf{x}_s$ is any point on the surface. This implies that the term

$$\frac{1}{D}\nabla \cdot [f(R)\mathbf{A}]$$

in the momentum Equation (1) will behave like $\delta''(\mathbf{x}-\mathbf{x}_s)$. The only term that could possibly balance this is $\nabla^2\mathbf{u}$, and this would mean that the velocity $\mathbf{u}$ itself would behave like $\delta(\mathbf{x}-\mathbf{x}_s)$. However, this would leave the term $\mathbf{u}\cdot\nabla\mathbf{u}$ as an even stronger singularity (actually not definable in terms of generalized functions) that could not be balanced by anything. From this argument it appears that the model equations do not admit solutions with $R < L$ in part of the domain (there will always be such a region if physical boundary or initial conditions are imposed), and $R > L$ in the rest of the domain.

Following Serrin, consider an undisturbed flow with velocity $\mathbf{v}$, pressure q, and moment tensor $\mathbf{B}$ (with trace S^2), occupying a region of space $\mathcal{V}$, with a prescribed velocity on its boundary $\mathcal{S}$. The flow perturbation will be denoted by $\mathbf{u}$, p, $\mathbf{A}$, R^2 for the velocity, pressure, moment tensor, and its trace, respectively. The equations for the total flow are

$$Re\frac{\partial \mathbf{u}}{\partial t} + Re\frac{\partial \mathbf{v}}{\partial t} + Re(\mathbf{u}+\mathbf{v})\cdot\nabla(\mathbf{u}+\mathbf{v})$$

$$= -\nabla(p+q) + \nabla^2\mathbf{u} + \nabla^2\mathbf{v}\frac{1}{D}\nabla\cdot[f(R^2+S^2)(\mathbf{A}+\mathbf{B})]; \quad (6)$$

$$\frac{\partial\mathbf{A}}{\partial t} + \frac{\partial\mathbf{B}}{\partial t} + (\mathbf{u}+\mathbf{v})\cdot\nabla(\mathbf{A}+\mathbf{B})$$

$$= (\mathbf{A}+\mathbf{B})\cdot\nabla(\mathbf{u}+\mathbf{v}) + (\nabla[(\mathbf{u})^T+(\mathbf{v})^T]\cdot(\mathbf{A}+\mathbf{B}) - \frac{f(R^2+S^2)}{D}(\mathbf{A}+\mathbf{B}-\mathbf{I}); \quad (7)$$

$$Re\frac{\partial\mathbf{v}}{\partial t} + Re\mathbf{v}\cdot\nabla\mathbf{v} = -\nabla q + \nabla^2\mathbf{v} + \frac{1}{D}\nabla\cdot(f(S^2)\mathbf{B}); \quad (8)$$

$$\frac{\partial\mathbf{B}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{B} = \mathbf{B}\cdot\nabla\mathbf{u} + (\nabla\mathbf{u})^T\cdot\mathbf{B} - \frac{f(S^2)}{D}(\mathbf{B}-\mathbf{I})\ . \quad (9)$$

Subtracting Equation (8) from (6) gives

$$Re\frac{\partial\mathbf{u}}{\partial t} + Re\mathbf{u}\cdot\nabla\mathbf{u} + Re\mathbf{u}\cdot\nabla\mathbf{v} + Re\mathbf{v}\cdot\nabla\mathbf{u}$$

$$= -\nabla(p) + \nabla^2\mathbf{u} + \frac{1}{D}\nabla\cdot[f(R^2+S^2)(\mathbf{A}+\mathbf{B})] - \frac{1}{D}\nabla\cdot(f(S^2)\mathbf{B})\ . \quad (10)$$

In a slight deviation from the usual approach to stability analyses, where Equation (9) would be subtracted from (7), and the resulting difference used, here only equation (7) will be used. In fact, only the trace of (7), given by the following equation, will be used.

$$\frac{\partial(R^2+S^2)}{\partial t} + (\mathbf{u}+\mathbf{v})\cdot\nabla(R^2+S^2) =$$

$$2\mathbf{A}:\nabla\mathbf{u} + 2\mathbf{B}:\nabla\mathbf{u} + 2\mathbf{A}:\nabla\mathbf{v} + 2\mathbf{B}:\nabla\mathbf{v} - \frac{f(R^2+S^2)}{D}(R^2+S^2-3)\ . \quad (11)$$

If the inner product of $\mathbf{u}$ with Equation (10) is taken, and the result is integrated over the entire flow domain, the following is obtained:

$$\frac{Re}{2}\frac{\partial}{\partial t}\int_{\mathcal{V}} \mathbf{u}\cdot\mathbf{u}d\mathcal{V}$$

$$+Re\int_{\mathcal{V}}\mathbf{u}\cdot(\nabla\mathbf{u})\cdot\mathbf{u}d\mathcal{V}+Re\int_{\mathcal{V}}\mathbf{u}\cdot(\nabla\mathbf{v})\cdot\mathbf{u}d\mathcal{V}+Re\int_{\mathcal{V}}\mathbf{v}\cdot(\nabla\mathbf{u})\cdot\mathbf{u}d\mathcal{V}$$

$$=-\int_{\mathcal{V}}\mathbf{u}\cdot\nabla(p)d\mathcal{V}+\int_{\mathcal{V}}\mathbf{u}\cdot\nabla^2\mathbf{u}d\mathcal{V}$$

$$+\frac{1}{D}\int_{\mathcal{V}}\mathbf{u}\cdot(\nabla\cdot[f(R^2+S^2)(\mathbf{A}+\mathbf{B})])d\mathcal{V}-\frac{1}{D}\int_{\mathcal{V}}\mathbf{u}\cdot\nabla\cdot(f(S^2)\mathbf{B})d\mathcal{V}\quad . \tag{12}$$

Regardless of what boundary condition the flow must satisfy at the boundary $\mathcal{S}$, the disturbance flow must satisfy $\mathbf{u}=0$ on the boundary. This, and the fact that $\nabla\cdot\mathbf{u}$ (continuity) can be used when some of the terms in (12) are integrated by parts to give

$$\frac{Re}{2}\frac{\partial}{\partial t}\int_{\mathcal{V}}\mathbf{u}\cdot\mathbf{u}d\mathcal{V}+Re\int_{\mathcal{V}}\mathbf{u}\cdot(\nabla\mathbf{v})\cdot\mathbf{u}d\mathcal{V}$$

$$=-\int_{\mathcal{V}}\nabla\mathbf{u}:\nabla\mathbf{u}d\mathcal{V}$$

$$-\frac{1}{D}\int_{\mathcal{V}}f(R^2+S^2)(\mathbf{A}+\mathbf{B}):(\nabla\mathbf{u})d\mathcal{V}+\frac{1}{D}\int_{\mathcal{V}}\mathbf{u}\cdot\nabla[f(S^2)\mathbf{B}]d\mathcal{V}\quad . \tag{13}$$

Now multiply (11) by

$$\frac{1}{2D}f(R^2+S^2), \tag{14}$$

and integrate over the entire fluid domain, to get

$$\frac{1}{2D}\int_{\mathcal{V}}f(R^2+S^2)\frac{\partial(R^2+S^2)}{\partial t}d\mathcal{V}+\frac{1}{2D}\int_{\mathcal{V}}f(R^2+S^2)(\mathbf{u}+\mathbf{v})\cdot\nabla(R^2+S^2)d\mathcal{V}$$

$$=\frac{1}{2D}\int_{\mathcal{V}}f(R^2+S^2)2\mathbf{A}:\nabla\mathbf{u}d\mathcal{V}+\frac{1}{2D}\int_{\mathcal{V}}f(R^2+S^2)2\mathbf{B}:\nabla\mathbf{u}d\mathcal{V}$$

$$+\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)2\mathbf{A}:\nabla\mathbf{v}d\mathcal{V}+\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)2\mathbf{B}:\nabla\mathbf{v}d\mathcal{V}$$

$$-\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)\frac{f(R^2+S^2)}{D}(R^2+S^2-3) \quad . \tag{15}$$

If the second integral on the left side of (15) is integrated by parts, the result can be seen to be zero by first noting that

$$f(R^2+S^2)\nabla(R^2+S^2)=\nabla g(R^2+S^2), \tag{16}$$

where

$$g(R^2+S^2)=-\frac{1}{L^2}ln[1-(R^2+S^2)], \tag{17}$$

and then using continuity again. This leaves

$$\frac{1}{2D}\int_{\mathcal{V}}\frac{\partial g(R^2+S^2)}{\partial t}d\mathcal{V}$$

$$=\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)2\mathbf{A}:\nabla\mathbf{u}d\mathcal{V}+\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)2\mathbf{B}:\nabla\mathbf{u}d\mathcal{V}$$

$$+\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)2\mathbf{A}:\nabla\mathbf{v}d\mathcal{V}+\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)2\mathbf{B}:\nabla\mathbf{v}d\mathcal{V}$$

$$-\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)\frac{f(R^2+S^2)}{D}(R^2+S^2-3) \quad . \tag{18}$$

Now, Equation (18) and Equation (13) are added together to give

$$\frac{Re}{2}\frac{\partial}{\partial t}\int_{\mathcal{V}}\mathbf{u}\cdot\mathbf{u}d\mathcal{V}+\frac{1}{2D}\frac{\partial}{\partial t}\int_{\mathcal{V}} g(R^2+S^2)d\mathcal{V}$$

$$=-Re\int_{\mathcal{V}}\mathbf{u}\cdot(\nabla\mathbf{v})\cdot\mathbf{u}d\mathcal{V}-\int_{\mathcal{V}}\nabla\mathbf{u}:\nabla\mathbf{u}d\mathcal{V}$$

$$+\frac{1}{D}\int_{\mathcal{V}}\mathbf{u}\cdot\nabla[f(S^2)\mathbf{B}]d\mathcal{V}$$

$$+\frac{1}{D}\int_{\mathcal{V}} f(R^2+S^2)\mathbf{A}:\nabla\mathbf{v}d\mathcal{V}+\frac{1}{D}\int_{\mathcal{V}} f(R^2+S^2)\mathbf{B}:\nabla\mathbf{v}d\mathcal{V}$$

$$-\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)\frac{f(R^2+S^2)}{D}(R^2+S^2-3)\ . \tag{19}$$

The various terms in (19) will now be considered. First, as shown by Serrin [1], if $-m$ is a lower bound for the eigenvalues of the matrix $\nabla\mathbf{v}$ (the velocity gradient of the undisturbed flow), then

$$\mathbf{u}\cdot(\nabla\mathbf{v})\cdot\mathbf{u}\geq -mu^2. \tag{20}$$

This allows the term

$$-\int_{\mathcal{V}}\nabla\mathbf{u}:\nabla\mathbf{u}d\mathcal{V} \tag{21}$$

to be bounded *above* by

$$m\int_{\mathcal{V}} u^2 d\mathcal{V}. \tag{22}$$

The term

$$\int_{\mathcal{V}}\nabla\mathbf{u}:\nabla\mathbf{u}d\mathcal{V} \tag{23}$$

can be bounded *below* by a Poincare inequality if it is assumed that the domain $\mathcal{V}$ is bounded; as shown by Serrin,

$$\int_{\mathcal{V}}\nabla\mathbf{u}:\nabla\mathbf{u}d\mathcal{V}\geq \alpha d^{-2}\int_{\mathcal{V}} u^2 d\mathcal{V}, \tag{24}$$

where

$$\alpha = \frac{3+\sqrt{13}}{2}\pi^2, \tag{25}$$

and d is the maximum distance between any two points in the flow domain.

The next term to be bounded is

$$\left|\int_{\mathcal{V}} f(R^2+S^2)\mathbf{A}:\nabla\mathbf{v}d\mathcal{V}\right| \le \left|\int_{\mathcal{V}_1} f(R^2+S^2)\mathbf{A}:\nabla\mathbf{v}d\mathcal{V}\right| + \left|\int_{\mathcal{V}_2} f(R^2+S^2)\mathbf{A}:\nabla\mathbf{v}d\mathcal{V}\right|$$

$$\le f(3+q)L^2(9|\nabla\mathbf{v}|_{max}) + L^2(9|\nabla\mathbf{v}|_{max})\left|\int_{\mathcal{V}_2} |f(R^2+S^2)|d\mathcal{V}\right|, \tag{26}$$

where $|\nabla\mathbf{v}|_{max}$ is an upper bound on the components of the base-flow velocity gradient over the entire domain. Similarly,

$$\left|\int_{\mathcal{V}} f(R^2+S^2)\mathbf{B}:\nabla\mathbf{v}d\mathcal{V}\right| \le \left|\int_{\mathcal{V}_1} f(R^2+S^2)\mathbf{B}:\nabla\mathbf{v}d\mathcal{V}\right| + \left|\int_{\mathcal{V}_2} f(R^2+S^2)\mathbf{B}:\nabla\mathbf{v}d\mathcal{V}\right|$$

$$\le f(3+q)L^2(9|\nabla\mathbf{v}|_{max}) + L^2(9|\nabla\mathbf{v}|_{max})\left|\int_{\mathcal{V}_2} |f(R^2+S^2)|d\mathcal{V}\right|. \tag{27}$$

Here, the assumption that the components of $\mathbf{A}$ and $\mathbf{B}$ are bounded by L^2 has been used.

Next, let q be an arbitrary number greater than 0, and divide the flow into two regions: $\mathcal{V}_1$, where $R^2+S^2 \le 3+q$, and $\mathcal{V}_2$, where $R^2+S^2 > 3+q$. Then note that

$$\frac{1}{2D}\int_{\mathcal{V}} f(R^2+S^2)\frac{f(R^2+S^2)}{D}(R^2+S^2-3)$$

$$\ge \frac{q}{2D^2}\int_{\mathcal{V}_2}\left[f(R^2+S^2)\right]^2 d\mathcal{V} - \frac{1}{2D^2}\int_{\mathcal{V}_1}(3+q)f(3+q)d\mathcal{V}. \tag{28}$$

Finally, observe that

$$-\frac{1}{D}\left|\int_{\mathcal{V}} f(S^2)\mathbf{B} : \nabla \mathbf{u} d\mathcal{V}\right| \le \frac{9L^2}{D}|f(S^2)|_{max} \int_{\mathcal{V}} |\nabla \mathbf{u}| d\mathcal{V}. \tag{29}$$

With all the above estimates, Equation (19) gives

$$\begin{aligned}
&\frac{Re}{2}\frac{\partial}{\partial t}\int_{\mathcal{V}} \mathbf{u}\cdot\mathbf{u} d\mathcal{V} + \frac{1}{2D}\frac{\partial}{\partial t}\int_{\mathcal{V}} g(R^2+S^2) d\mathcal{V} \\
&\le Re\alpha d^{-2}\int_{\mathcal{V}} u^2 d\mathcal{V} - \alpha d^{-2}\int_{\mathcal{V}} u^2 d\mathcal{V} \\
&+\frac{1}{D}|\nabla\cdot[f(S^2)\mathbf{B}]|_{max}\int_{\mathcal{V}} |\mathbf{u}| d\mathcal{V} \\
&+2L^2(9|\nabla\mathbf{v}|_{max})\left|\int_{\mathcal{V}_2} |f(R^2+S^2)| d\mathcal{V}\right| \\
&-\frac{q}{2D^2}\int_{\mathcal{V}_2}\left[f(R^2+S^2)\right]^2 d\mathcal{V} + \frac{1}{2D^2}\int_{\mathcal{V}_1}(3+q)f(3+q) d\mathcal{V} \quad .
\end{aligned} \tag{30}$$

Now the Schwarz inequality will be used:

$$\int_{\mathcal{V}} p d\mathcal{V} \le \mathcal{V}\left(\int_{\mathcal{V}} p^2 d\mathcal{V}\right)^{\frac{1}{2}} \tag{31}$$

for any function p, where $\mathcal{V}$ is now also used to denote the volume of the domain $\mathcal{V}$. Let $p = |\mathbf{u}| = u$; the result is

$$\int_{\mathcal{V}} |u| d\mathcal{V} \le \mathcal{V}\left(\int_{\mathcal{V}} u^2 d\mathcal{V}\right)^{\frac{1}{2}} \quad , \tag{32}$$

which, when substituted into (30) gives

$$\begin{aligned}
&\frac{Re}{2}\frac{\partial}{\partial t}\int_{\mathcal{V}} \mathbf{u}\cdot\mathbf{u} d\mathcal{V} + \frac{1}{2D}\frac{\partial}{\partial t}\int_{\mathcal{V}} g(R^2+S^2) d\mathcal{V} \\
&\le Re\alpha d^{-2}\int_{\mathcal{V}} u^2 d\mathcal{V} - \alpha d^{-2}\int_{\mathcal{V}} u^2 d\mathcal{V}
\end{aligned}$$

$$+\frac{1}{D}|\nabla\cdot[f(S^2)\mathbf{B}]|_{max}\mathcal{V}\left(\int_{\mathcal{V}}u^2d\mathcal{V}\right)^{\frac{1}{2}}$$

$$+2L^2(9|\nabla\mathbf{v}|_{max})\mathcal{V}\left[\int_{\mathcal{V}_2}[f(R^2+S^2)]^2d\mathcal{V}\right]^{\frac{1}{2}}$$

$$-\frac{q}{2D^2}\int_{\mathcal{V}_2}\left[f(R^2+S^2)\right]^2d\mathcal{V}+\frac{1}{2D^2}\int_{\mathcal{V}_1}(3+q)f(3+q)d\mathcal{V}\quad. \tag{33}$$

The left-hand side of this equation is the time derivative of an "energy" for the system. This "energy" will be denoted by $E = E_1 + E_2$, where

$$E_1 = \frac{Re}{2}\int_{\mathcal{V}}\mathbf{u}\cdot\mathbf{u}d\mathcal{V}; \tag{34}$$

$$E_2 = \frac{1}{2D}\int_{\mathcal{V}}g(R^2+S^2)d\mathcal{V}. \tag{35}$$

Then (33) can be written as

$$\frac{\partial E}{\partial t}\le\left[\frac{2\alpha}{d^2}-\frac{2\alpha}{Re\ d^2}+\frac{2}{Re\ D}|\nabla\cdot[f(S^2)\mathbf{B}]|_{max}\mathcal{V}\right]E_1$$

$$+2L^2(9|\nabla\mathbf{v}|_{max})\left[\int_{\mathcal{V}_2}[f(R^2+S^2)]^2d\mathcal{V}\right]^{\frac{1}{2}}$$

$$-\frac{q}{2D^2}\int_{\mathcal{V}_2}\left[f(R^2+S^2)\right]^2d\mathcal{V}$$

$$+\frac{1}{2D^2}(3+q)f(3+q)\mathcal{V}+2f(3+q)L^2(9|\nabla\mathbf{v}|_{max}). \tag{36}$$

From this equation it should be clear that if the Reynolds and Deborah numbers satisfy

$$\frac{2\alpha}{d^2}-\frac{2\alpha}{Re\ d^2}+\frac{2}{Re\ D}|\nabla\cdot[f(S^2)\mathbf{B}]|_{max}\mathcal{V}<0, \tag{37}$$

the flow will remain bounded, even though it cannot be proved from this that it actually goes to zero. This is because with this condition the terms on the right-hand side of (36) that dominate at large values of E_1 and E_2 are negative. Growth of the disturbance can therefore not continue unbounded. For small enough Reynolds number and large enough Deborah number, this condition can be satisfied for any bounded base flow in a bounded domain.

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CHAPTER III

Brownian Motion of a Slightly Deformable Drop

The Brownian motion of a drop in another fluid is of interest in such applications as emulsions or liquid-liquid extraction. There is reason to believe that its motion when subjected to small random forces that are due to fluctuations in the surrounding fluid may be somewhat different from a solid particle. A solid particle in an infinite fluid will undergo translational and rotational diffusion because of its Brownian motion. The particle, being solid, has a finite number of degrees of freedom (three for translational, and from zero to two for orientation depending on its degree of rotational symmetry). In the case of a sphere, this diffusivity is given by the Stokes-Einstein result. For a particle of arbitrary shape, there will be both translational and rotational diffusivities. Essentially, the diffusivity will still be the amount of thermal energy associated with a degree of freedom divided by the mobility of the particle associated with that degree of freedom. However, translation and rotation become coupled for a particle of arbitrary shape.

The case of a deformable drop is yet more complicated because there are an infinite number of degrees of freedom. "Rotation" does not have a well-defined meaning in this case. While it is possible to define an "angular velocity" of a drop by integrating the vorticity over its volume, this will not be done here, since it does not relate directly to the motion of the surface. Instead, the motion of the surface will be divided into center-of-mass translation and deformations of the surface that leave the center of mass fixed. While for a rigid particle the Stokes-Einstein diffusivity can be derived using the simple equilibrium argument first presented by Einstein [1], it is not clear whether this method can be generalized to the case of a deformable body.

To investigate how the differences between drops and rigid particles affect Brownian motion, a single drop in an infinite fluid will be considered here. Its diffusivity and velocity-autocorrelation function will be calculated, as well as the correlation functions associated with its deformation, in terms of the density, viscosity and surface tension. Gravity will be neglected here even though the drop density will not necessarily be assumed to be the same as the surrounding fluid density. As in other physical problems (such as suspensions) where the geometrical configuration changes with time, a distinction must be made between "short-time" and "long-time" diffusivities. In this analysis, the diffusivity to be calculated will be short-time in the sense that the drop will be assumed not to have moved far from its initial position over the averaging time interval, but long-time insofar as the diffusivity will not be a function of the drop configuration, but rather will be the result of an average over all possible configurations of the drop weighted in some appropriate way according to their probabilities.

A key step in the understanding of Brownian motion is the determination of a fluctuation-dissipation theorem, which relates the fluctuation variables to macroscopic properties. The fluctuation-dissipation theorem for the drop will show how the random thermal energy of the fluid molecules gets partitioned between translation of the center of mass of the drop and deformation that is centrally symmetric. Its derivation is a two-step process: first, correlations of the fluctuating variables at equilibrium must be determined using thermodynamics; then, the time-dependent hydrodynamic problem must be solved to obtain correlations of the fluctuating variables at different times, up to a constant that must be determined by comparison with the equilibrium results.

EQUILIBRIUM RESULTS FROM THERMODYNAMICS

As stated above, the first step in deriving the fluctuation-dissipation theorem is to determine how the energy is partitioned among the various spherical harmonic modes at equilibrium. In choosing the fluctuating variables, it is convenient to decompose the deformation into spherical harmonic modes. The drop shape will be described in spherical coordinates (r, θ, ϕ) by

$$r = a + \eta(\theta, \phi, t) \ , \tag{1.1}$$

where

$$\eta = \sum_{n=0}^{\infty} \eta_n(\theta, \phi, t) = \sum_{n=0}^{\infty} \sum_{m=-n}^{n} a_n^m(t) P_n^{|m|}(cos\theta) e^{im\phi} \ . \tag{1.2}$$

It will be assumed here that η is small relative to the drop radius; in other words, when nondimensionalized with respect to a, η will be of order ϵ, with $\epsilon << 1$. Since η includes not only deformations of the drop shape relative to a sphere, but also center-of-mass translations, it is important to justify the assumption that enough Brownian events occur before the drop has moved even a small fraction of its radius to allow meaningful statistical analysis. Einstein [1] estimates that a 0.001 mm rigid Brownian particle in water will move 0.8 microns in a second. Chandrasekhar [2] states that a Brownian particle will experience about 10^{21} collisions in a second. Clearly, the drop will not have to move far before such quantities as a diffusivity will be well-defined.

Since the objective here is to study Brownian motion of a drop (with no mass transfer) at constant temperature, a canonical ensemble is the natural choice from the statistical mechanical viewpoint. (The canonical ensemble consists of a large number of systems, each with a fixed volume, temperature, and number of molecules.) According to standard statistical thermodynamic theory, the choice of ensemble determines which form of energy (e.g., internal, Gibbs, Helmholtz) appears in the probability distribution for the states of the system. As shown in Hill [3] for a canonical ensemble at equilibrium, the probability of a certain interface configuration is related exponentially to the energy associated with it:

$$P[\eta] \propto exp[-\frac{E(\eta)}{\kappa T}] \, , \tag{1.3}$$

where κ is Boltzmann's constant, and T is the absolute temperature, and the reference state with respect to which the energy will be measured will be taken as the spherical (undeformed) state. The energy can be considered to be a function(al) of the a_n^m, since the set of values of these coefficients completely describes η:

$$E[\eta(\theta, \phi)] = E[\{a_n^m\}]. \tag{1.4}$$

The energy associated with a certain state (i.e., a certain shape and center-of-mass velocity) of the drop consists of two parts: potential energy associated with surface tension, and kinetic energy if the center of mass of the drop has a nonzero velocity. As mentioned earlier, gravitational potential energy will be ignored. Moreover, it will be assumed that the kinetic energy associated with *deformation rates* is negligible; this is consistent with the assumption to be made later for the time-dependent hydrodynamic problem that the Reynolds number of the flow is very small. By thermodynamics, the potential energy associated with a certain deformation is equal in magnitude to the work done against surface tension in isothermally and reversibly deforming the drop from the undeformed state (which is chosen as the reference state) to that particular deformed state . (When the drop is deformed reversibly, work is done only against surface tension; there is no viscous dissipation, because the process occurs "infinitely" slowly.) By the definition of surface tension, the work done in isothermally increasing the drop surface area by δA is just

$$W = \gamma \delta A, \tag{1.5}$$

where γ is the surface tension. So it is necessary to calculate δA for a given deformation η. For convenience, define a function Π by

$$\Pi(r, \theta, \phi) = r - a - \eta(\theta, \phi) \ , \tag{1.6}$$

where a is the radius of the drop in its spherical state. Then the deformed drop surface is the solution to $\Pi = 0$, and the unit normal to the surface is given by

$$\mathbf{n}(r, \theta, \phi) = \frac{\nabla \Pi}{|\nabla \Pi|}, \tag{1.7}$$

so that

$$\mathbf{n}(r, \theta, \phi) = \frac{\mathbf{e}_r - \frac{1}{r}\mathbf{e}_\theta \frac{\partial \eta}{\partial \theta} - \mathbf{e}_\phi \frac{1}{r sin\theta} \frac{\partial \eta}{\partial \phi}}{\left[1 + \frac{1}{r^2}\left(\frac{\partial \eta}{\partial \theta}\right)^2 + \frac{1}{r^2 sin^2\theta}\left(\frac{\partial \eta}{\partial \phi}\right)^2\right]^{\frac{1}{2}}}. \tag{1.8}$$

Now consider a point $(a + \eta(\theta, \phi), \theta, \phi)$ on the surface, and the element of area dA within $d\theta$ and $d\phi$ of this point. If the surface element were on a sphere of radius $a + \eta$, the area element would be

$$dA' = (a + \eta)^2 sin\theta d\theta d\phi. \tag{1.9}$$

However, since the surface element is in general tilted (i.e., $\mathbf{n} \neq \mathbf{e}_r$), this is just the projected area; the actual area is

$$dA = \frac{dA'}{cos\alpha} = \frac{dA'}{\mathbf{n} \cdot \mathbf{e}_r}, \tag{1.10}$$

where α is the angle between the surface normal and $\mathbf{e}_r$. Thus,

$$dA = (a+\eta)^2 sin\theta d\theta d\phi \big[1 + \frac{1}{r^2}\big(\frac{\partial\eta}{\partial\theta}\big)^2 + \frac{1}{r^2 sin^2\theta}\big(\frac{\partial\eta}{\partial\phi}\big)^2\big]^{\frac{1}{2}}, \tag{1.11}$$

with $r = a + \eta$. The deformation will be assumed to be small, say of $O(\epsilon)$, (large amplitude or rapidly varying deformations are very improbable since the probability goes exponentially like the energy). Therefore, this expression will be approximated for small η by the terms up to $O(\epsilon^2)$ (the reason for carrying the approximation to this order will become clear below):

$$dA = \Big[a^2 + 2a\eta + \eta^2 + \frac{1}{2}\big(\frac{\partial\eta}{\partial\theta}\big)^2 + \frac{1}{2sin^2\theta}\big(\frac{\partial\eta}{\partial\phi}\big)^2\Big] sin\theta d\theta d\phi. \tag{1.12}$$

The change in area ΔA is, of course, the integral of this over the ranges of ϕ and θ, minus the area of the undeformed spherical drop, $4\pi a^2$. However, the fluid inside the drop is assumed to be incompressible, leading to the condition

$$V = \frac{4}{3}\pi a^3 \ , \tag{1.13}$$

where V is the drop volume, which is also given by

$$V = \int_0^{\pi}\int_0^{2\pi}\int_0^{a+\eta} r^2 sin\theta dr d\phi d\theta \ , \tag{1.14}$$

which, if the r-integral is done, results in

$$V = \int_0^{\pi}\int_0^{2\pi} \frac{(a+\eta)^3}{3} sin\theta d\phi d\theta. \tag{1.15}$$

This expression can also be approximated for small η as

$$V = \frac{4}{3}\pi a^3 + \int_0^{\pi}\int_0^{2\pi} a^2 \eta sin\theta d\theta d\phi, \tag{1.16}$$

from which the incompressibility condition

$$\int_0^{\pi}\int_0^{2\pi} \eta sin\theta d\theta d\phi = 0 \tag{1.17}$$

is obtained. This simplifies the expression for the area change to

$$\Delta A = \int_0^{\pi}\int_0^{2\pi}\Big[\eta^2 + \frac{1}{2}\big(\frac{\partial\eta}{\partial\theta}\big)^2 + \frac{1}{2sin^2\theta}\big(\frac{\partial\eta}{\partial\phi}\big)^2\Big] sin\theta d\theta d\phi. \tag{1.18}$$

To evaluate the change in surface area, the expression for η in terms of spherical harmonics is substituted into this formula for the area change:

$$\begin{aligned}\Delta A = \int_0^{\pi}\int_0^{2\pi} \sum_{n=0}^{\infty}\sum_{m=-n}^{n}\sum_{j=0}^{\infty}\sum_{k=-j}^{j} a_n^m a_j^k \Big[& P_n^{|m|}(cos\theta)P_j^{|k|}(cos\theta) \\ & + \frac{1}{2}\frac{dP_n^{|m|}(cos\theta)}{d\theta}\frac{dP_j^{|k|}(cos\theta)}{d\theta} \\ & - \frac{mk}{2sin^2\theta}P_n^{|m|}(cos\theta)P_j^{|k|}(cos\theta)\Big] \cdot e^{i(m+k)\phi} sin\theta d\theta d\phi. \end{aligned} \tag{1.19}$$

The integration with respect to ϕ can be easily done since

$$\int_0^{2\pi} e^{i(m+k)\phi} d\phi = 2\pi\delta_{m,-k} \tag{1.20}$$

(where δ is, of course, the Kronecker delta), so that

$$\begin{aligned}\Delta A = 2\pi \int_0^{\pi} \sum_{n=0}^{\infty}\sum_{m=-n}^{n}\sum_{j=0}^{\infty} a_n^m a_j^{-m} \Big[& P_n^{|m|}(cos\theta)P_j^{|m|}(cos\theta) \\ & + \frac{1}{2}\frac{dP_n^{|m|}(cos\theta)}{d\theta}\frac{dP_j^{|m|}(cos\theta)}{d\theta} \\ & + \frac{m^2}{2sin^2\theta}P_n^{|m|}(cos\theta)P_j^{|m|}(cos\theta)\Big]\ sin\theta\ d\theta. \end{aligned} \tag{1.21}$$

In evaluating the portion of this integral associated with the derivatives of the associated Legendre polynomials, it is helpful to make the change of variables $x = cos\theta$ and then integrate by parts (for $m \geq 0$):

$$
\begin{aligned}
I &\equiv \int_0^\pi \frac{dP_n^m(cos\theta)}{d\theta}\frac{dP_j^m(cos\theta)}{d\theta}\ sin\theta\ d\theta \\
&= \int_{-1}^1 \frac{dP_n^m}{dx}\frac{dP_j^m}{dx}\ (1-x^2)\ dx \\
&= P_n^m\left[(1-x^2)\ \frac{dP_j^m}{dx}\right]_{-1}^1 \ - \ \int_{-1}^1 P_n^m \frac{d}{dx}\left((1-x^2)\frac{dP_j^m}{dx}\right)dx. \qquad (1.22)
\end{aligned}
$$

Since the P_n^m are associated Legendre polynomials, and satisfy the differential equation

$$\frac{d}{dx}\left[(1-x^2)\frac{dP_n^m}{dx}\right] = -\left[n(n+1) - \frac{m^2}{1-x^2}\right]P_n^m(x), \qquad (1.23)$$

the remaining integral in (1.22) can be expressed in the form

$$I = \int_{-1}^1 P_n^m\left[j(j+1) - \frac{m^2}{1-x^2}\right]P_j^m\, dx. \qquad (1.24)$$

Now by changing variables from x back to $cos\theta$ and substituting the result into the expression for ΔA,

$$
\begin{aligned}
\Delta A = 2\pi \ &\sum_{n=0}^{\infty}\ \sum_{m=-n}^{n}\ \sum_{j=0}^{\infty} a_n^m a_j^{-m}\left[\int_0^\pi P_n^{|m|}(\cos\theta)P_j^{|m|}(\cos\theta)\ \sin\theta\ d\theta\right. \\
+ \quad &\frac{1}{2}j(j+1)\int_0^\pi P_n^{|m|}(\cos\theta)P_j^{|m|}(\cos\theta)\ \sin\theta\ d\theta \qquad (1.25)
\end{aligned}
$$

is obtained. Finally, using the orthogonality property of the associated Legendre polynomials

$$\int_0^\pi P_n^{|m|}(cos\theta)P_j^{|m|}(cos\theta)\ sin\theta\ d\theta \quad = \quad \frac{2}{2n+1}\frac{(n+m)!}{(n-m)!}\delta_{nj}, \qquad (1.27)$$

the expression (1.25) can be expressed in the form

$$\Delta A \quad = \quad 2\pi\sum_{n=0}^{\infty}\sum_{m=-n}^{n} a_n^m a_n^{-m}\left[1+\frac{n(n+1)}{2}\right]\frac{2}{2n+1}\frac{(n+|m|)!}{(n-|m|)!}. \qquad (1.28)$$

Since η must be real, it follows that

$$a_n^m = \overline{a_n^{-m}} \qquad (1.29)$$

(with the overbar denoting complex conjugation), and consequently,

$$a_n^m a_n^{-m} = |a_n^m|^2. \qquad (1.30)$$

To get the constraint on the a_n^m imposed by imcompressibility, the decomposition for η in spherical harmonics is substituted into the incompressibility condition:

$$\int_0^\pi\int_0^{2\pi}\eta sin\theta d\theta d\phi = \sum_{n=0}^{\infty}\sum_{m=-n}^{n} a_n^m \int_0^\pi\int_0^{2\pi} P_n^{|m|}(cos\theta)sin\theta e^{im\phi}\,d\theta d\phi\ ,$$
$$= 4\pi a_0^0, \qquad (1.31)$$

which implies that $a_0^0 = 0$. Henceforth, the $n = 0$ harmonic will not even be included in the complete decompositions, since this dilitational mode is disallowed by incompressibility. This completes the calculation of the component of the energy associated with surface tension.

Now the other component of the energy, the kinetic part, will also be expressed in terms of the a_n^m. It will be assumed here that the kinetic energy is due entirely to the purely translational motion of the drop. This is actually only a good approximation for a high-density, high-viscosity drop. Otherwise, the kinetic energy associated with the motion of the surrounding fluid and the circulation inside the drop must be included. If a more generally valid solution is desired, so that these other components of the kinetic energy must be included, it is useful to note that whereas in principle this kinetic energy could be obtained by integrating $\frac{1}{2}\rho u^2$ over the fluid volume, it is much easier to deduce it by looking at the unsteady force on the drop needed to "accelerate" any of its modes $a_{n,m}(t)$ from the stationary state to a quasi-steady motion. In this unsteady force, there will be a term analogous to the "added mass" term in the unsteady force on a rigid particle, which will represent the force needed to overcome the fluid inertia, rather than the viscous dissipation. From this force term, it should be possible to deduce the kinetic energy of the fluid associated with quasi-steady motion of a mode, just as it can be deduced from the added mass term in the unsteady drag on a rigid particle that the kinetic energy of the fluid in which a particle is translating with steady velocity U is just $\frac{1}{2}m_A U^2$, where m_A is the added mass. Since this requires knowledge of the time-dependent response of the system, it will not be considered in this section.

Since the translational kinetic energy is $\frac{1}{2}m_D \mathbf{U}\cdot\mathbf{U}$, where m_D is the mass of the drop, and $\mathbf{U}$ is the velocity of the center of mass of the drop, first the center of mass $\mathbf{X}$ must be expressed in terms of the a_n^m. By definition, the center of mass $\mathbf{X}$ is

$$\mathbf{X} = \frac{3}{4\pi a^3}\int_D \mathbf{x}\,dV, \tag{1.32}$$

where the integral is over the drop volume. This can be rewritten (using spherical coordinates) as

$$\mathbf{X} = \frac{3}{4\pi a^3} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \int_{r=0}^{a+\eta(\theta,\phi)} r\left[cos\theta \mathbf{e}_z + sin\theta cos\phi \mathbf{e}_x + sin\theta sin\phi \mathbf{e}_y\right]$$

$$\times \; r^2 sin\theta \; dr \; d\theta \; d\phi. \tag{1.33}$$

Since the deformations are assumed to be very small, and only the $O(\epsilon)$ approximation is sought here, this expression can be linearized in η (after the trivial r-integration is done) to give

$$\mathbf{X} = \frac{3}{4\pi a^3} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \left[\frac{a^4}{4} + a^3\eta\right] \left[cos\theta \mathbf{e}_z + sin\theta cos\phi \mathbf{e}_x + sin\theta sin\phi \mathbf{e}_z\right]$$

$$\times \; sin\theta d\theta d\phi. \tag{1.34}$$

Clearly, the $\frac{a^4}{4}$ term integrates to zero, since if the sphere were undeformed its center of mass would be at the origin. Thus,

$$\mathbf{X} = \frac{3}{4\pi} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \eta(\theta,\phi) \left[cos\theta \mathbf{e}_z + sin\theta cos\phi \mathbf{e}_x + sin\theta sin\phi \mathbf{e}_z\right] sin\theta d\theta d\phi. \tag{1.35}$$

Since orthogonality relations are known for the spherical harmonics $P_n^m(\cos\theta)e^{im\phi}$, it is convenient to express $\cos\theta$, $\sin\theta\cos\phi$, and $\sin\theta\sin\phi$ in terms of surface spherical harmonics:

$$\cos\theta = P_1^0(\cos\theta); \tag{1.36}$$

$$\sin\theta\cos\phi = -\frac{1}{2} P_1^1(cos\theta)\left[e^{i\phi} + e^{-i\phi}\right]; \tag{1.37}$$

$$\sin\theta sin\phi = -\frac{1}{2i}P_1^1(\cos\theta)\big[e^{i\phi} - e^{-i\phi}\big] \ , \tag{1.38}$$

so that the integral (1.35) can be rewritten as

$$\mathbf{X} = \frac{3}{4\pi}\int_{\theta=0}^{\pi}\int_{\phi=0}^{2\pi}\sum_{n=0}^{\infty}\sum_{m=-n}^{n} a_n^m P_n^{|m|}(\cos\theta)e^{im\phi}$$
$$\Big[P_1^0(\cos\theta)\mathbf{e}_z - \frac{1}{2}P_1^1(\cos\theta)\big[e^{i\phi}+e^{-i\phi}\big]\mathbf{e}_x - \frac{1}{2i}P_1^1(\cos\theta)\big[e^{i\phi}-e^{-i\phi}\big]\mathbf{e}_y\Big]$$
$$\times \sin\theta d\theta d\phi. \tag{1.39}$$

Doing the ϕ-integration (using (1.20)) gives

$$\mathbf{X} = \frac{3}{4\pi}\int_{\theta=0}^{\pi}\sum_{n=0}^{\infty}2\pi\Big[a_n^0 P_n^0(\cos\theta)P_1^0(\cos\theta)\mathbf{e}_z$$
$$-\frac{1}{2}(a_n^{-1}+a_n^1)P_n^1(cos\theta)P_1^1(cos\theta)\mathbf{e}_x$$
$$-\frac{1}{2i}(a_n^{-1}-a_n^1)P_n^1(cos\theta)P_1^1(cos\theta)\mathbf{e}_y\Big]\cdot sin\theta d\theta, \tag{1.40}$$

and then doing the θ-integration (using (1.27)) gives

$$\mathbf{X} = \frac{3}{4\pi}2\pi\ \Big[\frac{2}{3}a_1^0\mathbf{e}_z - (\frac{4}{3})\frac{1}{2}(a_1^{-1}+a_1^1)\mathbf{e}_x - (\frac{4}{3})\frac{1}{2i}(a_1^{-1}-a_1^1)\mathbf{e}_y\Big]. \tag{1.41}$$

It is convenient now to write the number a_n^m, which in general is complex, as

$$a_n^m = b_n^m + i\ c_n^m, \tag{1.42}$$

where b_n^m and c_n^m are real, so that

$$|a_n^m|^2 = (b_n^m)^2 + (c_n^m)^2 \quad , \tag{1.43}$$

where both b_n^m and c_n^m are *real* numbers. Then (1.41) simplifies to

$$\mathbf{X} = b_1^0 \mathbf{e}_z - 2b_1^1 \mathbf{e}_x + 2c_1^1 \mathbf{e}_y. \qquad (1.44)$$

Thus, the velocity of the center of mass of the drop is

$$\mathbf{U} = \frac{d\mathbf{X}}{dt} = \frac{db_1^0}{dt}\mathbf{e}_z - 2\frac{db_1^1}{dt}\mathbf{e}_x + 2\frac{dc_1^1}{dt}\mathbf{e}_y, \qquad (1.45)$$

and the kinetic energy of the drop motion is

$$E_{kin} = \frac{1}{2} m_D \Big[\Big(\frac{db_1^0}{dt}\Big)^2 + 4\Big(\frac{db_1^1}{dt}\Big)^2 + 4\Big(\frac{dc_1^1}{dt}\Big)^2 \Big]. \qquad (1.46)$$

At this point the probability distribution for interface deformations (described by $\{a_n^m(t)\}$) can be written (using (1.28)) as

$$P[\{a_n^m\}] = N exp\Bigg\{ -\frac{1}{\kappa T}\Bigg[\frac{m_D}{2}\Big[\Big(\frac{db_1^0}{dt}\Big)^2 + 4\Big(\frac{db_1^1}{dt}\Big)^2 + 4\Big(\frac{dc_1^1}{dt}\Big)^2 \Big] + 2\pi\gamma \sum_{n=0}^{\infty} \sum_{m=-n}^{n} d_n^m [(b_n^m)^2 + (c_n^m)^2] \Bigg]\Bigg\} , \qquad (1.48)$$

where

$$d_n^m = \Big[1 + \frac{n(n+1)}{2}\Big] \frac{2}{2n+1} \frac{(n+|m|)!}{(n-|m|)!} \qquad (1.49)$$

and N is a normalization constant, chosen so that if P is integrated over all possible values of $\{a_n^m\}$, the result is 1. Note that the independent variables

$$\{b_m^m\}, \{c_m^m\}, \qquad n = 0,1,2,...;\ m = -n,...,0,...,n \qquad (1.50)$$

$$\frac{db_1^0}{dt}, \frac{db_1^1}{dt}, \frac{dc_1^1}{dt}, \qquad (1.51)$$

can be thought of as coordinates in phase space, with (1.50) being the position coordinates (configuration space) and (1.51) being equivalent to coordinates in momemtum space. It can be seen from Equation (1.48) for the probability distribution function that it can be factored into functions each of which depends on only *one* of the b_n^m or c_n^m or their time derivatives. In fact, the probability distribution is just the composite of independent Gaussian distributions; i.e., the spherical harmonic modes (and their time derivatives) of different order and degree are *independent.*

Hence, if particular values of m and n are chosen, the probability distribution for b_n^m *alone* is

$$P(b_n^m) = N_{n,m}\ exp\big[-\frac{2\pi\gamma}{\kappa T} d_n^m\ (b_n^m)^2\big], \tag{1.52}$$

where $N_{n,m}$ is now a normalization constant for this distribution only. (This normalization constant is chosen so that this probability integrated over all possible values of b_n^m (i.e., all real numbers), gives 1.) The same equation holds for c_n^m by just replacing b_n^m with c_n^m in (1.52), from which it follows that b_n^m and c_n^m will have the same autocorrelations. In the following section, a fluctuation-dissipation theorem for the drop will be derived; for this, the equilibrium autocorrelations of the b_n^m and c_n^m will be needed. They are computed by noting that the probability distribution has the general form

$$P(x) = N exp[-qx^2], \tag{1.53}$$

where q is some constant, N is the normalization constant, and x stands for one of the b_n^m or c_n^m. Thus, the expectation of x^2 is simply

$$< x^2 >= \frac{\int_{-\infty}^{\infty} x^2 exp[-qx^2]\ dx}{\int_{-\infty}^{\infty} exp[-qx^2]\ dx} = \frac{1}{2q}. \tag{1.54}$$

This shows that the expectations of $b_n^m b_n^m$ and $c_n^m c_n^m$ are given by

$$\begin{aligned} \langle b_n^m b_n^m \rangle = \langle c_n^m c_n^m \rangle &= \Big(\frac{4\pi\gamma}{\kappa T} d_n^m\Big)^{-1} \\ &= \frac{\kappa T}{4\pi\gamma}\Big[1 + \frac{n(n+1)}{2}\Big]^{-1}\Big(n + \frac{1}{2}\Big)\frac{(n-|m|)!}{(n+|m|)!}, \end{aligned} \tag{1.55}$$

with d_n^m as given above. This relation holds for all m, n except $m = 0, n = 0$, since incompressibility necessitates $b_0^0 = c_0^0 = 0$. Note that this shows the qualitatively correct dependence on T, γ, and n, m: as the temperature goes up, the mean-square fluctuations should increase; as the surface tension goes up, the fluctuations should decrease, and they should also decrease as m and n increase, since larger n and/or m correspond(s) to smaller wavelength, i.e., higher energy, deformations. For zero surface tension, the fluctuations become infinite, as expected, since the drop loses its integrity in this limit.

The equilibrium autocorrelations for the center-of-mass velocity components will also be needed, and these are calculated similarly:

$$P(\dot{b}_1^0) = N exp\Big\{-\frac{m_D}{2\kappa T}(\dot{b}_1^0)^2\Big\}; \tag{1.56}$$

$$< \dot{b}_1^0 \dot{b}_1^0 >= \frac{\kappa T}{m_D}, \tag{1.57}$$

and similarly for b_1^1 and c_1^1:

$$< \dot{b}_1^1 \dot{b}_1^1 >=< \dot{c}_1^1 \dot{c}_1^1 >= \frac{\kappa T}{4 m_D}\ . \tag{1.58}$$

This is, of course, just the familiar form of equipartition that holds for the velocity of a rigid particle, too.

THE TIME-DEPENDENT PROBLEM

The fluctuation-dissipation theorem gives the autocorrelations of the parameters specifying the interface configuration (b_n^m and c_n^m) at *different* times. The equilibrium results calculated in the previous section are needed to get the absolute magnitudes of the correlations, but to obtain the time-dependent part, it is necessary to solve the time-dependent hydrodynamic problem. This consists of determining the flow field due to a (stochastic) forcing that is assumed to model the Brownian fluctuations. The Brownian (thermal) energy, which is of the magnitude of κT for each degree of freedom, will be assumed to cause only small fluctuations in the drop shape. With the assumption that the interface deformations due to fluctuations are small in amplitude, it is possible to expand the boundary conditions about the spherical state, and still work in spherical coordinates (i.e., a domain perturbation). It will also be assumed that the Reynolds number for the fluctuating velocity field is small, so that the equations for fluid motion inside and outside the drop are the unsteady Stokes equations. Thus, for the interior flow

$$\rho_1 \frac{\partial \mathbf{u}^i}{\partial t} = -\nabla p^i + \mu_1 \nabla^2 \mathbf{u}^i \ , \tag{2.1}$$

$$\nabla \cdot \mathbf{u}^i = 0 \ , \tag{2.2}$$

where p_i is the pressure, $\mathbf{u}^i$ is the fluid velocity, and ρ_1,μ_1 are, respectively, the density and the viscosity of the fluid inside the drop; similarly, for the exterior flow,

$$\rho_2 \frac{\partial \mathbf{u}^o}{\partial t} = -\nabla p^o + \mu_2 \nabla^2 \mathbf{u}^o \ , \tag{2.3}$$

$$\nabla \cdot \mathbf{u}^o = 0 \ . \tag{2.4}$$

(The subscripts "i" and "o" refer to *inside* and *outside*, respectively.) For a time-*independent* flow with boundary conditions to be applied at a sphere, Lamb's [4] general solution, involving spherical harmonics, is a natural choice. Lamb discusses briefly the general solution for the time-*dependent* case, and Yang [5] has expressed this solution explicitly in vector notation. If the velocities (and pressures) are Fourier-transformed according to

$$\hat{\mathbf{u}}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathbf{u}(t)\ e^{i\omega t} dt \ , \tag{2.5}$$

$$\mathbf{u}(t) = \int_{-\infty}^{\infty} \hat{\mathbf{u}}(\omega)\ e^{-i\omega t} d\omega \ , \tag{2.6}$$

(the same formulas hold for the pressure by replacing $\mathbf{u}$ with p), then Yang's form for the general solution in spherical coordinates (r, θ, ϕ) is:

$$\hat{\mathbf{u}}^i = \sum_{n=1}^{\infty} \Bigg[\frac{1}{h_1^2 \mu_1} \nabla p_n^i - \psi_n(h_1 r) \nabla \times (\mathbf{r} \chi_n^i)$$
$$+ [(n+1)\psi_{n-1}(h_1 r) - n\psi_{n+1}(h_1 r) h_1^2 r^2] \nabla \varphi_n^i$$
$$+ n(2n+1)\psi_{n+1}(h_1 r) h_1^2 \varphi_n^i \mathbf{r} \Bigg] \ , \tag{2.7a}$$

$$\hat{p}^i = \sum_{n=1}^{\infty} p_n^i \ , \tag{2.7b}$$

for the inside flow, and

$$\hat{\mathbf{u}}^o = \sum_{n=1}^{\infty}\left[\frac{1}{h_2^2\mu_2}\nabla p^o_{-(n+1)} - \psi_n(h_2 r)\nabla\times(\mathbf{r}\chi^o_n)\right.$$
$$+[(n+1)\psi_{n-1}(h_2 r) - n\psi_{n+1}(h_2 r)h_2^2 r^2]\nabla\varphi^o_n$$
$$\left. + n(2n+1)\psi_{n+1}(h_2 r)h_2^2\varphi^o_n\mathbf{r}\right] , \qquad (2.8a)$$

$$\hat{p}^o = \sum_{n=1}^{\infty}\hat{p}^o_{-(n+1)} , \qquad (2.8b)$$

for the outside flow, where

$$p^i_n,\ \varphi^i_n,\ \chi^i_n,\ \varphi^o_n,\ \chi^o_n$$

are *general* spherical harmonics of order n (the specific choice is determined from the boundary conditions), and the

$$p^o_{-(n+1)}$$

are spherical harmonics of order $-(n+1)$. The functions ψ_n and f_n are defined by

$$f_n(x) = i\cdot\sqrt{\frac{\pi}{2}}\, x^{-\left(n+\frac{1}{2}\right)}\, H^{(2)}_{n+\frac{1}{2}}(x) , \qquad (2.9)$$

$$\psi_n(x) = \sqrt{\frac{\pi}{2}} x^{-\left(n+\frac{1}{2}\right)} J_{\left(n+\frac{1}{2}\right)}(x) , \qquad (2.10)$$

where

$$H^{(2)}_{n+\frac{1}{2}}(x)$$

are Hankel functions of the second kind of order $n + \frac{1}{2}$, and

$$J_{\left(n+\frac{1}{2}\right)}(x)$$

are Bessel functions of the first kind of order $n + \frac{1}{2}$. The symbols h_1 and h_2 are an abbreviated notation for

$$h_1 = \sqrt{\frac{i\omega\rho_1}{\mu_1}} \quad , \tag{2.11}$$

$$h_2 = \sqrt{\frac{i\omega\rho_2}{\mu_2}} \quad , \tag{2.12}$$

with the *positive* branch of the square root function being taken.

While Yang (5) presented this solution in dimensional form, it will be found convenient later to work with dimensionless variables here. To avoid introducing any additional complications in the notation, the *same* symbols will be used below for the *nondimensional* velocity, pressure, and spherical harmonics as were used in Yang's solution above. Since henceforth everything will be dimensionless, there should be no confusion. The characteristic velocity used to make $\mathbf{u}$ dimensionless is $kT/\mu_2 a^2$; the characteristic pressure is kT/a^3; the interface deformation η and the position variables are nondimensionalized by a; the characteristic time is $\mu_2 a^3/kT$; the spherical harmonics φ_n are nondimensionalized by $kT/\mu_2 a$, and the spherical harmonics χ_n by $kT/\mu_2 a^2$. The outside (i.e., subscript 2) fluid parameters are used in the nondimensionalization, so that later on, when limits of the inside viscosity going to zero and to infinity are taken (to compare with the cases of a bubble and a rigid particle), none of the dimensionless variables will become infinite just because of the way they were nondimensionalized. With this, the general solution in dimensionless form becomes

$$\hat{\mathbf{u}}^i = \sum_{n=1}^{\infty} \Bigg[\frac{1}{i\Omega\alpha} \nabla p_n^i - \psi_n\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big) \nabla \times (\mathbf{r}\chi_n^i)$$

$$+ \Big[(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big) - n\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)\Big(\frac{i\Omega\alpha}{\lambda}\Big)r^2\Big] \nabla \varphi_n^i$$

$$+ n(2n+1)\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)\Big(\frac{i\Omega\alpha}{\lambda}\Big)\varphi_n^i \mathbf{r} \Bigg] , \tag{2.13a}$$

$$\hat{p}^i = \sum_{n=1}^{\infty} p_n^i , \tag{2.13b}$$

for the inside flow, and

$$\hat{\mathbf{u}}^o = \sum_{n=1}^{\infty} \Bigg[\frac{1}{i\Omega} \nabla p_{-(n+1)}^o - f_n(\sqrt{i\Omega}r) \nabla \times (\mathbf{r}\chi_n^o)$$

$$+ \Big[(n+1) f_{n-1}(\sqrt{i\Omega}r) - n f_{n+1}(\sqrt{i\Omega}r)(i\Omega)r^2\Big] \nabla \varphi_n^o$$

$$+ n(2n+1) f_{n+1}(\sqrt{i\Omega}r)(i\Omega)\varphi_n^o \mathbf{r} \Bigg] , \tag{2.14a}$$

$$\hat{p}^o = \sum_{n=1}^{\infty} \hat{p}_{-(n+1)}^o , \tag{2.14b}$$

for the outside flow. Here, the dimensionless parameters that appear are

$$\alpha = \frac{\rho_1}{\rho_2}, \tag{2.15a}$$

$$\lambda = \frac{\mu_1}{\mu_2}, \tag{2.15b}$$

$$\beta = \frac{kT\rho_2}{\mu_2^2 a}, \tag{2.15c}$$

$$\Gamma = \frac{\gamma a^2}{kT}, \tag{2.15d}$$

and the dimensionless variable

$$\Omega = \frac{\omega \rho_2 a^2}{\mu_2}. \tag{2.16}$$

To determine the particular choice of the spherical harmonics p_n, φ_n, χ_n appearing in the general solution (2.13),(2.14), boundary conditions must be invoked. The boundary conditions to be applied at the interface ($r = 1 + \eta(\theta, \phi, t)$) for this time-dependent problem are continuity of tangential velocity,

$$\mathbf{t} \cdot \mathbf{u}^i = \mathbf{t} \cdot \mathbf{u}^o \quad , \tag{2.17a}$$

where $\mathbf{t}$ denotes either of two independent unit vectors tangent to the surface; the kinematic condition

$$\mathbf{n} \cdot \mathbf{u}^i \; = \; \mathbf{n} \cdot \mathbf{u}^o \; = \; \frac{\partial \eta}{\partial t} \quad , \tag{2.17b}$$

where $\mathbf{n}$ is the outward normal to the interface; continuity of tangential stress,

$$\mathbf{t} \cdot \mathbf{n} \cdot \sigma^i = \mathbf{t} \cdot \mathbf{n} \cdot \sigma^o; \tag{2.17c}$$

and finally, continuity of normal stress,

$$\mathbf{n} \cdot \mathbf{n} \cdot \sigma^i \; - \; \mathbf{n} \cdot \mathbf{n} \cdot \sigma^o \; = \; \Gamma \nabla \cdot \mathbf{n} + y(\theta, \phi, t). \tag{2.17d}$$

The function $y(\theta, \phi, t)$ which appears in (2.17d) represents the random normal stress on the interface caused by collisions of fluid molecules with it (nondimensionalized by kT/a^3). It is convenient to decompose y also in spherical harmonics,

$$y(\theta, \phi, t) = \sum_{n=0}^{\infty} y_n(t) = \sum_{n=0}^{\infty} \sum_{m=-n}^{n} y_n^m(t) P_n^{|m|}(cos\theta) e^{im\phi}. \tag{2.18}$$

Such a decomposition is always possible for a continuous function of θ and ϕ; it is being assumed here that an individual realization of the stochastic function y is the average, on some length-scale intermediate between the drop size and the mean free path of the fluid molecules, of the force per area caused by fluid molecules impinging on the surface, so that this assumption of continuity of y is reasonable.

Since the flow is driven only by this random normal stress, the velocity and interface deformation, and also the pressure (apart from the term that is independent of θ and ϕ) are of the same order as y, namely, $O(\epsilon)$. (The θ- and ϕ-independent part of the pressure would be present even if there were no flow and the drop were perfectly spherical; it balances the surface tension term for the constant curvature of a sphere.) Hence, if the boundary conditions (which are actually to be evaluated at $r = 1 + \eta$) are expanded about $r = 1$, the corrections (due to the displacement of the interface by η) are of $O(\epsilon^2)$ and are thus negligible for the $O(\epsilon)$ problem being solved here. In other words, the boundary conditions can simply be applied at $r = 1$ with no changes, the only exception being the surface tension term $\Gamma\nabla \cdot \mathbf{n}$. For this term, it is necessary to determine $\nabla \cdot \mathbf{n}$ to first order in η. The result is

$$\nabla \cdot \mathbf{n} \;=\; 2 - \frac{1}{2}\Big[2\eta + \frac{cos\theta}{sin\theta}\frac{\partial\eta}{\partial\theta} + \frac{\partial^2\eta}{\partial\theta^2} + \frac{1}{sin^2\theta}\frac{\partial^2\eta}{\partial\phi^2}\Big] \;+ H.O.T. \tag{2.19}$$

The first member in the right-hand side of (2.19) is the θ- and ϕ-independent term mentioned above, and it can be ignored, since it is part of the trivial $O(1)$ solution (corresponding to a completely quiescent fluid and a perfectly spherical drop). Since the boundary conditions will be imposed by requiring all terms of each degree of surface harmonic to balance, it is desirable to simplify the surface tension term

$$S.T.Term = \frac{\Gamma}{2}\sum_{n=1}^{\infty}\Big[2\hat{\eta}_n + \frac{cos\theta}{sin\theta}\frac{\partial\hat{\eta}_n}{\partial\theta} + \frac{\partial^2\hat{\eta}_n}{\partial\theta^2} + \frac{1}{sin^2\theta}\frac{\partial^2\hat{\eta}_n}{\partial\phi^2}\Big] \tag{2.20}$$

appearing in the normal stress condition so that it appears as a sum of surface spherical harmonics, rather than a sum of various derivatives of spherical harmonics. The associated Legendre polynomials P_n^m in the surface harmonic expansion (1.2) for η satisfy (1.23), which, if the change of variables $x = \cos\theta$ is made, becomes

$$\frac{d^2P_n^m(\cos\theta)}{d\theta^2} + \frac{\cos\theta}{\sin\theta}\frac{dP_n^m(\cos\theta)}{d\theta} + \Big[n(n+1) - \frac{m^2}{\sin^2\theta}\Big]P_n^m(\cos\theta) = 0. \tag{2.21}$$

Since by (1.2)

$$\eta_n = \sum_{m=-n}^{n} a_n^m P_n^{|m|}(cos\theta)e^{im\phi}, \tag{2.22}$$

it follows that

$$\sum_{m=-n}^{n}\hat{a}_n^m\Bigg[\frac{d^2P_n^{|m|}(cos\theta)}{d\theta^2} + \frac{cos\theta}{sin\theta}\frac{dP_n^{|m|}(cos\theta)}{d\theta}$$

$$+\Big(n(n+1) - \frac{m^2}{sin^2\theta}\Big)P_n^{|m|}(cos\theta)\Bigg]e^{im\theta} = 0, \tag{2.23}$$

which can be rewritten as

$$\frac{\partial^2\hat{\eta}_n}{\partial\theta^2} + \frac{cos\theta}{sin\theta}\frac{\partial\hat{\eta}_n}{\partial\theta} + n(n+1)\hat{\eta}_n + \frac{1}{sin^2\theta}\frac{\partial^2\hat{\eta}_n}{\partial\phi^2} = 0. \tag{2.24}$$

From (2.24) it is clear that the surface tension term (2.20) can be rewritten as

$$S.T.Term = \frac{\Gamma}{2}\sum_{n=1}^{\infty}[2 - n(n+1)]\hat{\eta}_n. \tag{2.25}$$

Since only the $O(\epsilon)$ approximation of the flow field is being computed here, it is consistent to ignore the higher-order corrections to the unit vectors, which change the boundary conditions only at $O(\epsilon^2)$:

$$\mathbf{n} = \mathbf{e}_r + H.O.T. \tag{2.26a}$$

$$\mathbf{t} = \begin{cases} \mathbf{e}_\theta + H.O.T. \\ \mathbf{e}_\phi + H.O.T. \end{cases} \tag{2.26b}$$

For application of the stress boundary conditions, it is convenient to use Brenner's [6] result that (after nondimensionalization),

$$\mathbf{e}_r \cdot \sigma^o = -\frac{\mathbf{r}}{r}p^o + \left[\frac{\partial \mathbf{u}^o}{\partial r} - \frac{\mathbf{u}^o}{r}\right] + \frac{1}{r}\nabla(\mathbf{r}\cdot\mathbf{u}^o), \tag{2.27a}$$

$$\mathbf{e}_r \cdot \sigma^i = -\frac{\mathbf{r}}{r}p^i + \lambda\left[\frac{\partial \mathbf{u}^i}{\partial r} - \frac{\mathbf{u}^i}{r}\right] + \frac{\lambda}{r}\nabla(\mathbf{r}\cdot\mathbf{u}^i). \tag{2.27b}$$

It can be shown (as done by Yang in dimensional form) that

$$\mathbf{e}_r \cdot \sigma^o = \frac{1}{r}\sum_{n=1}^{\infty}\Big[-\frac{2(n+2)}{(i\Omega)}\nabla p^o_{-(n+1)}$$
$$-Q^o_n(\sqrt{i\Omega}r)\nabla\times(\mathbf{r}\chi^o_n) + R^o_n(\sqrt{i\Omega}r)\nabla\varphi^o_n$$
$$-\frac{(2n+1)}{r^2}S^o_n(\sqrt{i\Omega}r)\varphi^o_n\mathbf{r} - p^o_{-(n+1)}\mathbf{r}\Big] \tag{2.28a}$$

for the exterior flow, and

$$\mathbf{e}_r \cdot \sigma^i = \frac{1}{r}\sum_{n=1}^{\infty}\Big\{\lambda\Big[\frac{2(n-1)}{(i\Omega)\alpha}\nabla p^i_n$$
$$-Q^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\nabla\times(\mathbf{r}\chi^i_n) + R^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\nabla\varphi^i_n$$

$$-\frac{(2n+1)}{r^2} S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r)\varphi_n^i \mathbf{r}\Big] - p_n^i \mathbf{r}\Big\} \tag{2.28b}$$

for the interior flow, where

$$Q_n^o(z) = -z^2 f_{n+1}(z) + (n-1) f_n(z), \tag{2.29a}$$

$$R_n^o(z) = nz^4 f_{n+2}(z) + nz^2 f_{n+1}(z) - (n+1) z^2 f_n(z) + 2(n+1)(n-1) f_{n-1}(z), \tag{2.29b}$$

$$S_n^o(z) = nz^4 f_{n+2}(z) + nz^2 f_{n+1}(z), \tag{2.29c}$$

$$Q_n^i(z) = -z^2 \psi_{n+1}(z) + (n-1)\psi_n(z), \tag{2.30a}$$

$$R_n^i(z) = nz^4 \psi_{n+2}(z) + nz^2 \psi_{n+1}(z) - (n+1) z^2 \psi_n(z) + 2(n+1)(n-1)\psi_{n-1}(z), \tag{2.30b}$$

$$S_n^i(z) = nz^4 \psi_{n+2}(z) + nz^2 \psi_{n+1}(z). \tag{2.30c}$$

Since the general solution is in terms of Fourier-transformed variables, the next step is to Fourier-transform the boundary conditions. The only change from the boundary conditions in time given above is that the velocity, pressure and stress are replaced by their Fourier transforms, and

$$\frac{\partial \eta}{\partial t}$$

is replaced by

$$-\frac{i\Omega}{\beta}\hat{\eta}.$$

Now, the general solution will be substituted into the boundary conditions in order to obtain algebraic equations for the arbitrary spherical harmonics that appear in it. First, the kinematic condition gives

$$-\frac{i\Omega}{\beta}\sum_{n=0}^{\infty}\hat{\eta}_n = \sum_{n=1}^{\infty}\Bigg\{\frac{1}{i\alpha\Omega}\frac{\partial p_n^i}{\partial r} + \left[(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big) - n\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)\frac{i\Omega\alpha}{\lambda}r^2\right]\frac{\partial\varphi_n^i}{\partial r} + n(2n+1)\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)\frac{i\Omega\alpha}{\lambda}\varphi_n^i r\Bigg\}, \qquad (2.31a)$$

and

$$-\frac{i\Omega}{\beta}\sum_{n=0}^{\infty}\hat{\eta}_n = \sum_{n=1}^{\infty}\Bigg\{\frac{1}{i\Omega}\frac{\partial p^o_{-(n+1)}}{\partial r} + \left[(n+1)f_{n-1}\Big(\sqrt{i\Omega}r\Big) - nf_{n+1}\Big(\sqrt{i\Omega}r\Big)(i\Omega)r^2\right]\frac{\partial\varphi_n^o}{\partial r} + n(2n+1)f_{n+1}\Big(\sqrt{i\Omega}r\Big)(i\Omega)\varphi_n^o r\Bigg\}, \qquad (2.31b)$$

which gives the two equations (to be satisfied for $n = 1, 2, 3, ...$)

$$-\frac{i\Omega}{\beta}\hat{\eta}_n = \frac{n}{i\Omega\alpha}p_n^i + \left[n(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big) + n(n+1)\frac{i\Omega\alpha}{\lambda}\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)\right]\varphi_n^i\Bigg|_{r=1}, \qquad (2.32a)$$

and

$$-\frac{i\Omega}{\beta}\hat{\eta}_n = -\frac{(n+1)}{i\Omega}p^o_{-(n+1)}$$

$$+\left[n(n+1)f_{n-1}(\sqrt{i\Omega}) + n(n+1)i\Omega f_{n+1}(\sqrt{i\Omega})\right]\varphi^o_n\Big|_{r=1} \quad . \tag{2.32b}$$

By the methodology developed by Brenner [6], the condition of continuity of tangential velocity at the drop surface will be satisfied by requiring that

$$r\frac{\partial u^i_r}{\partial r}\Big|_{r=1} = r\frac{\partial u^o_r}{\partial r}\Big|_{r=1}, \tag{2.33}$$

and

$$\mathbf{r}\cdot\nabla\times\mathbf{u}^i\Big|_{r=1} = \mathbf{r}\cdot\nabla\times\mathbf{u}^o\Big|_{r=1} \quad . \tag{2.34}$$

Similarly, the condition of continuity of tangential stress at the drop surface will be satisfied by requiring that

$$r\frac{\partial(\mathbf{e}_r\cdot\mathbf{e}_r\cdot\sigma^i)}{\partial r} - r\nabla\cdot(\mathbf{e}_r\cdot\sigma^i)\Big|_{r=1} = r\frac{\partial(\mathbf{e}_r\cdot\mathbf{e}_r\cdot\sigma^o)}{\partial r} - r\nabla\cdot(\mathbf{e}_r\cdot\sigma^o)\Big|_{r=1}, \tag{2.35}$$

and

$$\mathbf{r}\cdot\nabla\times(\mathbf{e}_r\cdot\sigma^i_r)\Big|_{r=1} = \mathbf{r}\cdot\nabla\times(\mathbf{e}_r\cdot\sigma^o_r)\Big|_{r=1} \quad . \tag{2.36}$$

It can be shown that Equations (2.34) and (2.36), which imply that there is no torque on the drop, necessitate that

$$\chi^i_n = 0; \tag{2.37a}$$

$$\chi_n^o = 0. \tag{2.37b}$$

Equation (2.33) leads to the conditions (for $n = 1, 2, 3, ...$):

$$\frac{n(n-1)}{i\Omega\alpha} p_n^i$$
$$+\left[n(n-1)(n+1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right) - n(n+1)\frac{i\Omega\alpha}{\lambda}\psi_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right)\right.$$
$$\left. +n(n+1)^2\frac{i\Omega\alpha}{\lambda}\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right) - n(n+1)\left(\frac{i\Omega\alpha}{\lambda}\right)^2\psi_{n+2}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right)\right]\varphi_n^i\Bigg|_{r=1}$$
$$= \frac{(n+1)(n+2)}{i\Omega} p^o_{-(n+1)}$$
$$+\left[n(n-1)(n+1)f_{n-1}(\sqrt{i\Omega}) - n(n+1)(i\Omega)f_n(\sqrt{i\Omega})\right.$$
$$\left. +n(n+1)^2(i\Omega)f_{n+1}(\sqrt{i\Omega}) - n(n+1)(i\Omega)^2 f_{n+2}(\sqrt{i\Omega})\right]\varphi_n^o\Bigg|_{r=1}. \tag{2.38}$$

Before applying the tangential stress condition, it is useful to note that

$$-\nabla\cdot(\mathbf{e}_r\cdot\sigma^i) + \nabla\cdot(\mathbf{e}_r\cdot\sigma^o)\Big|_{r=1} = \mathbf{e}_r\cdot[\nabla\cdot(\sigma^o - \sigma^i)] + (\nabla\mathbf{e}_r):(\sigma^o - \sigma^i)\Big|_{r=1}$$
$$= \frac{-i\Omega}{\beta}(u_r^o - u_r^i) - \frac{3}{r}(p^o - p^i) + \frac{1}{r}\mathbf{e}_r\mathbf{e}_r:(\sigma^o - \sigma^i)\Big|_{r=1}. \tag{2.39}$$

It is also useful to note that

$$\mathbf{e}_r\cdot[\mathbf{e}_r\cdot(\sigma^o - \sigma^i)] = \sum_{n=1}^{\infty} -\Big[\frac{2(n+1)(n+2)}{(i\Omega)r} p^o_{-(n+1)} + R_n^o(\sqrt{i\Omega}r)\frac{n\varphi_n^o}{r}$$

$$-\frac{(2n+1)}{r^2}S^o_n(\sqrt{i\Omega}r)\varphi^o_n r - p^o_{-(n+1)}r\Big]$$

$$+\Bigg\{\lambda\Big[\frac{2(n-1)n}{(i\Omega)\alpha r}p^i_n + R^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n\varphi^i_n}{r}$$

$$-\frac{(2n+1)}{r^2}S^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\varphi^i_n r\Big] - p^i_n r\Bigg\}, \tag{2.40}$$

since this expression appears in both the tangential and the normal stress conditions. Equation (2.35) leads to the conditions (again for $n = 1, 2, 3, ...$):

$$\frac{2\lambda n(n-1)(n-2)}{i\Omega\alpha}p^i_n - np^i_n - 3p^i_n$$

$$+\lambda\Bigg[n(n-2)R^i_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right) + n\frac{dR^i_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)}{dr}$$

$$-(2n+1)(n-2)S^i_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - (2n+1)\frac{dS^i_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)}{dr}\Bigg]\Bigg|_{r=1}$$

$$-\Big[\frac{2(n+1)(n+2)}{(i\Omega)r}p^o_{-(n+1)} + R^o_n(\sqrt{i\Omega}r)\frac{n\varphi^o_n}{r}$$

$$-\frac{(2n+1)}{r^2}S^o_n(\sqrt{i\Omega}r)\varphi^o_n r - p^o_{-(n+1)}r\Big]$$

$$= -\frac{2(n+1)(n+2)(n+3)}{i\Omega}p^o_{-(n+1)} + (n+1)p^o_{-(n+1)} - 3p^o_{-(n+1)}$$

$$+\lambda\Bigg[n(n-2)R^o_n(\sqrt{i\Omega}) + n\frac{dR^o_n(\sqrt{i\Omega}r)}{dr}$$

$$-(2n+1)(n-2)S^o_n(\sqrt{i\Omega}r) - (2n+1)\frac{dS^o_n(\sqrt{i\Omega}r)}{dr}\Bigg]$$

$$-\Bigg\{\lambda\Big[\frac{2(n-1)n}{(i\Omega)\alpha r}p^i_n + R^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n\varphi^i_n}{r}$$

$$-\frac{(2n+1)}{r^2}S^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\varphi^i_n r\Big] - p^i_n r\Big\}\Big|_{r=1}. \qquad (2.41)$$

Here,

$$\frac{dR^o_1(z)}{dz} = -2n(n+2)(2n+1)zf_{n+1}(z) + (2n+1)^2 zf_n(z) - (2n+1)zf_{n-1}(z), \quad (2.42)$$

and

$$\frac{dS^o_1(z)}{dz} = -2n(n+2)(2n+1)zf_{n+1}(z) + n(4n+3)zf_n(z) - nzf_{n-1}(z), \quad (2.43)$$

and similarly for the inside flow with the superscript "o" replaced by "i," and the functions $f(z)$ replaced by $\psi(z)$. In deriving these expressions from (2.29) and (2.30), the recursion relation

$$z^2 f_{n+1}(z) = (2n+1)f_n(z) - f_{n-1}(z), \qquad (2.44)$$

which also holds for $\psi_n(z)$, was used. Also, extensive use has been made of the fact that the derivatives of the functions f_n and ψ_n satisfy the relations

$$f'_n(z) = -zf_{n+1}(z); \qquad (2.45a)$$

$$\psi'_n(z) = -z\psi_{n+1}(z). \qquad (2.45b)$$

Finally, the normal stress condition gives

$$-\mathbf{e}_r \cdot \frac{1}{r}\sum_{n=1}^{\infty}\Big[-\frac{2(n+2)}{(i\Omega)}\nabla p^o_{-(n+1)}$$

$$+R^o_n(\sqrt{i\Omega}r)\nabla\varphi^o_n$$

$$-\frac{(2n+1)}{r^2}S_n^o(\sqrt{i\Omega}r)\varphi_n^o\mathbf{r} - p_{-(n+1)}^o\mathbf{r}\Big]$$

$$+\mathbf{e}_r\cdot\frac{1}{r}\sum_{n=1}^{\infty}\Big\{\lambda\Big[\frac{2(n-1)}{(i\Omega)\alpha}\nabla p_n^i$$

$$+R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\nabla\varphi_n^i$$

$$-\frac{(2n+1)}{r^2}S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\varphi_n^i\mathbf{r}\Big] - p_n^i\mathbf{r}\Big\}$$

$$= -\frac{\Gamma}{2}\sum_{n=1}^{\infty}[2-n(n+1)]\hat{\eta}_n + \sum_{n=1}^{\infty}\hat{y}_n, \tag{2.46}$$

which gives the equation

$$-\Big[\frac{2(n+1)(n+2)}{(i\Omega)r}p_{-(n+1)}^o + R_n^o(\sqrt{i\Omega}r)\frac{n\varphi_n^o}{r}$$

$$-\frac{(2n+1)}{r^2}S_n^o(\sqrt{i\Omega}r)\varphi_n^o r - p_{-(n+1)}^o r\Big]$$

$$+\Big\{\lambda\Big[\frac{2(n-1)n}{(i\Omega)\alpha r}p_n^i + R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n\varphi_n^i}{r}$$

$$-\frac{(2n+1)}{r^2}S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\varphi_n^i r\Big] - p_n^i r\Big\}$$

$$= -\frac{\Gamma}{2}[2-n(n+1)]\hat{\eta}_n + \hat{y}_n. \tag{2.47}$$

All of the above conditions are to be applied at $r = 1$. The above equations were simplified by making repeated use of the property of any spherical harmonic function S_n of order n that

$$\frac{\partial S_n}{\partial r} = \frac{nS_n}{r}. \tag{2.48}$$

The Equations (2.32a), (2.32b), (2.38), (2.41), and (2.47) constitute 5 equations from which the desired relation between the interface deformation (or other flow variables) and the random normal stress is to be obtained. (There are two more equations, that determine χ_n, but these are independent of the five, and thus are not needed here.) The 5 equations are

$$-\frac{i\Omega}{\beta}\hat{\eta}_n = \tilde{A}p^i_n + \tilde{B}\varphi^i_n, \tag{2.49a}$$

$$-\frac{i\Omega}{\beta}\hat{\eta}_n = \tilde{C}p^o_{-(n+1)} + \tilde{D}\varphi^o_n, \tag{2.49b}$$

$$\tilde{E}\varphi^i_n + \tilde{F}p^i_n = \tilde{G}\varphi^o_n + \tilde{H}p^o_{-(n+1)}, \tag{2.49c}$$

$$\tilde{I}p^i_n + \tilde{J}\varphi^i_n = \tilde{K}p^o_{-(n+1)} + \tilde{L}\varphi^o_n, \tag{2.49d}$$

and

$$\tilde{M}p^i_n + \tilde{N}\varphi^i_n + \tilde{O}p^o_{-(n+1)} + \tilde{P}\varphi^o_n = \tilde{Q}\hat{\eta}_n + \hat{y}_n. \tag{2.49e}$$

In these equations, $\tilde{A}, \tilde{B}, ...$ are given by

$$\tilde{A} = \frac{n}{i\Omega\alpha}, \tag{2.50a}$$

$$\tilde{B} = n(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big) + n(n+1)\Big(\frac{i\Omega\alpha}{\lambda}\Big)\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big), \tag{2.50b}$$

$$\tilde{C} = -\frac{(n+1)}{i\Omega}, \tag{2.50c}$$

$$\tilde{D} = n(n+1)f_{n-1}(\sqrt{i\Omega}) + n(n+1)i\Omega f_{n+1}(\sqrt{i\Omega}), \tag{2.50d}$$

$$\tilde{E} = n(n-1)(n+1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right) - n(n+1)(n+2)\frac{i\Omega\alpha}{\lambda}\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right) \quad (2.50e)$$

$$\tilde{F} = \frac{n(n-1)}{i\Omega\alpha}, \quad (2.50f)$$

$$\tilde{G} = n(n-1)(n+1)f_{n-1}(\sqrt{i\Omega}) - n(n+1)(n+2)(i\Omega)f_{n+1}(\sqrt{i\Omega}) \quad (2.50g)$$

$$\tilde{H} = \frac{(n+1)(n+2)}{i\Omega}, \quad (2.50h)$$

$$\tilde{I} = \frac{2n(n-1)^2\lambda}{i\Omega\alpha} - n + 2, \quad (2.50i)$$

$$\tilde{J} = \lambda\Bigg[2n(n+1)(n+2)^2\left(\frac{i\Omega\alpha}{\lambda}\right)\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right) - 2n(2n+1)(n+1)\left(\frac{i\Omega\alpha}{\lambda}\right)\psi_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right)$$

$$+2n(n+1)(n-1)^2\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right)\Bigg] \quad (2.50j)$$

$$\tilde{K} = \frac{-2(n+1)(n+2)^2}{i\Omega} + n + 3, \quad (2.50k)$$

$$\tilde{L} = \Bigg[2n(n+1)(n+2)^2 i\Omega f_{n+1}(\sqrt{i\Omega}) - 2n(2n+1)(n+1)(i\Omega)f_n(\sqrt{i\Omega})$$

$$+2n(n+1)(n-1)^2 f_{n-1}(\sqrt{i\Omega})\Bigg] \quad (2.50l)$$

$$\tilde{M} = \frac{2\lambda n(n-1)}{i\Omega\alpha} - 1, \quad (2.50m)$$

$$\tilde{N} = -2\lambda n(n+1)\left[(n+2)\left(\frac{i\Omega\alpha}{\lambda}\right)\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right) - (n-1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}\right)\right] \quad (2.50n)$$

$$\tilde{O} = -\frac{2(n+1)(n+2)}{i\Omega} + 1, \quad (2.50o)$$

$$\tilde{P} = 2n(n+1)\left[(n+2)(i\Omega)f_{n+1}(\sqrt{i\Omega}) - (n-1)f_{n-1}(\sqrt{i\Omega})\right], \quad (2.50p)$$

and finally,

$$\tilde{Q} = -\frac{\Gamma}{2}[2 - n(n+1)]. \tag{2.50q}$$

This system of equations can be rewritten in matrix form:

$$\begin{pmatrix} \tilde{A} & 0 & \tilde{B} & 0 & \frac{-i\Omega}{\beta} \\ 0 & \tilde{C} & 0 & \tilde{D} & \frac{-i\Omega}{\beta} \\ \tilde{F} & -\tilde{H} & \tilde{E} & -\tilde{G} & 0 \\ \tilde{I} & -\tilde{K} & \tilde{J} & -\tilde{L} & 0 \\ \tilde{M} & \tilde{O} & \tilde{N} & \tilde{P} & -\tilde{Q} \end{pmatrix} \begin{pmatrix} p_n^i \\ p^o_{-(n+1)} \\ \varphi_n^i \\ \varphi_n^o \\ \hat{\eta}_n \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ \hat{y}_n \end{pmatrix}. \tag{2.51}$$

The desired result is the solution to this linear system of equations for the flow variables in terms of the imposed random normal stress y. The solution for the deformation can be written as

$$\hat{\eta}_n = \hat{G}_n^1(\Omega)\ \hat{y}_n, \tag{2.52}$$

where $\hat{G}_n^1(\Omega)$ is the "transfer function" which, when multiplied by the input, gives the output. It is given by:

$$\hat{G}_n^1(\Omega) = \frac{\beta}{i\Omega}\frac{\tilde{T}}{(\tilde{S} - \tilde{R} - \frac{i\Omega}{\beta}\tilde{T}\tilde{Q})}, \tag{2.53}$$

where

$$\begin{aligned} \tilde{R} = -\tilde{C}\begin{vmatrix} \tilde{F} & \tilde{E} & -\tilde{G} \\ \tilde{I} & \tilde{J} & -\tilde{L} \\ \tilde{M} & \tilde{N} & \tilde{P} \end{vmatrix} - \tilde{D}\begin{vmatrix} \tilde{F} & -\tilde{H} & \tilde{E} \\ \tilde{I} & -\tilde{K} & \tilde{J} \\ \tilde{M} & \tilde{O} & \tilde{N} \end{vmatrix} \\ = -\tilde{C}\Big[\tilde{F}(\tilde{J}\tilde{P} + \tilde{L}\tilde{N}) - \tilde{E}(\tilde{I}\tilde{P} + \tilde{L}\tilde{M}) - \tilde{G}(\tilde{I}\tilde{N} - \tilde{J}\tilde{M})\Big] \\ -\tilde{D}\Big[- \tilde{F}(\tilde{K}\tilde{N} + \tilde{J}\tilde{O}) + \tilde{H}(\tilde{I}\tilde{N} - \tilde{J}\tilde{M}) + \tilde{E}(\tilde{I}\tilde{O} + \tilde{K}\tilde{M})\Big], \end{aligned} \tag{2.54}$$

$$\tilde{S} = \tilde{A}\begin{vmatrix} -\tilde{H} & \tilde{E} & -\tilde{G} \\ -\tilde{K} & \tilde{J} & -\tilde{L} \\ \tilde{M} & \tilde{N} & \tilde{P} \end{vmatrix} - \tilde{D}\begin{vmatrix} \tilde{F} & -\tilde{H} & -\tilde{G} \\ \tilde{I} & -\tilde{K} & -\tilde{L} \\ \tilde{M} & \tilde{O} & \tilde{P} \end{vmatrix}$$

$$= -\tilde{A}\Big[-\tilde{H}(\tilde{J}\tilde{P} + \tilde{L}\tilde{N}) - \tilde{E}(-\tilde{K}\tilde{P} + \tilde{L}\tilde{O}) - \tilde{G}(-\tilde{K}\tilde{N} - \tilde{J}\tilde{O})\Big]$$

$$-\tilde{B}\Big[\tilde{F}(-\tilde{K}\tilde{P} + \tilde{L}\tilde{O}) + \tilde{H}(\tilde{I}\tilde{P} + \tilde{L}\tilde{M}) - \tilde{G}(\tilde{I}\tilde{O} + \tilde{K}\tilde{M})\Big] \,, \qquad (2.55)$$

and

$$\tilde{T} = \tilde{A}\begin{vmatrix} \tilde{C} & 0 & \tilde{D} \\ -\tilde{H} & -\tilde{E} & -\tilde{G} \\ -\tilde{K} & \tilde{J} & -\tilde{L} \end{vmatrix} + \tilde{B}\begin{vmatrix} 0 & \tilde{C} & \tilde{D} \\ \tilde{F} & -\tilde{H} & -\tilde{G} \\ \tilde{I} & -\tilde{K} & -\tilde{L} \end{vmatrix}$$

$$= \tilde{A}\Big[\tilde{C}(-\tilde{E}\tilde{L} + \tilde{G}\tilde{J}) + \tilde{D}(-\tilde{H}\tilde{J} + \tilde{E}\tilde{K})\Big]$$

$$+\tilde{B}\Big[-\tilde{C}(-\tilde{F}\tilde{L} + \tilde{G}\tilde{I}) + \tilde{D}(-\tilde{F}\tilde{K} + \tilde{H}\tilde{I})\Big]. \qquad (2.56)$$

The solutions for the other flow variables, which are needed in the calculation given below for the $O(\epsilon)^2$ correction to the diffusivity, can also be written in terms of transfer functions:

$$p_n^i = \hat{G}^{i,2}(\Omega)\hat{y}_n; \qquad (2.57)$$

$$\varphi_n^i = \hat{G}^{i,3}(\Omega)\hat{y}_n; \qquad (2.58)$$

$$p_{-(n+1)}^o = \hat{G}^{o,2}(\Omega)\hat{y}_n; \qquad (2.59)$$

$$\varphi_n^o = \hat{G}^{o,3}(\Omega)\hat{y}_n. \qquad (2.60)$$

The transfer functions appearing here are given in Appendix 1.

With this solution to the time-dependent problem, it is now possible to obtain the fluctuation-dissipation theorems for the fluctuating variables η_n, or more precisely, for the time-dependent coefficients $b_{n,m}$ and $c_{n,m}$, and to compute the diffusivity and velocity autocorrelation of the drop. Up to this point, the Fourier transform has been used, primarily because Lamb's and Yang's general solutions are given in Fourier-transformed variables. However, it should be noted that the variables such as particle velocity involved in the Brownian motion of the drop do *not* go to zero as $t \to \infty$. This is of course why the particle has a diffusivity. In such a case, the Fourier transforms will exist in the usual sense only for Ω having a nonzero positive imaginary component. This raises subtle points in inverting the transforms if the Fourier inversion formula (2.6) is to be used. The integrals will not exist in the usual sense and will have to be interpreted as generalized functions, which do not follow the usual rules of multiplication and differentiation. For this reason it is convenient to convert to Laplace transforms at this point, which can be done very simply. (Hauge and Martin-Löf [7], in their treatment of a solid Brownian particle, use Fourier transforms, and they also point out that it is more straightforward to convert to Laplace transforms before inverting.) To switch to Laplace transforms, consider everything to be motionless before $t = 0$ (as pointed out by Hinch [8] in his treatment of a solid particle, this does not result in any loss of generality), and define the Laplace transform

$$\check{\mathbf{X}}(s) = \frac{1}{2\pi}\int_0^\infty e^{-st}\mathbf{X}(t)dt = \hat{\mathbf{X}}(\Omega)\Big|_{\Omega=is}, \tag{2.61}$$

(where $\hat{\mathbf{X}}(\Omega)$ is of course the same Fourier transform as defined in (2.5)) for which the inversion formula is

$$\mathbf{X}(t) = \frac{1}{i}\int_{\gamma-i\infty}^{\gamma+i\infty} e^{st}\check{\mathbf{X}}(s)\; ds. \tag{2.62}$$

In this inversion formula, γ is chosen so that the path of integration is to the right of all the poles of $\check{\mathbf{X}}(s)$. For a system initially at zero (i.e. the Brownian drop at the origin with zero velocity and acceleration, and motionless surrounding fluid), the transfer function for Laplace-transformed "input" and "output" variables is related to that for Fourier-transformed variables by

$$\check{G}(s) = \hat{G}(\Omega)\Big|_{\Omega=is}, \tag{2.63}$$

with

$$\hat{\eta}(\Omega) = \hat{G}(\Omega)\ \hat{y}(\Omega); \tag{2.64}$$

$$\check{\eta}(s) = \check{G}(s)\ \check{y}(s). \tag{2.65}$$

Now the diffusivity of the drop will be computed. The diffusivity is the 3×3 tensor defined by

$$\mathbf{D} = \lim_{t \to \infty} \frac{d}{dt} < \mathbf{X}(t)\mathbf{X}(t) > . \tag{2.66}$$

Since, as observed above, the spherical harmonic modes decouple to first order, and only the first-order problem is being done here, the diffusivity will be diagonal. Furthermore, by symmetry, it must be that

$$D_{xx} = D_{yy} = D_{zz}. \tag{2.67}$$

Therefore, only the calculation for D_{zz} need be done here. From (1.44),

$$D_{zz} = \lim_{t \to \infty} \frac{d}{dt} < b_1^0(t)b_1^0(t) >, \tag{2.68}$$

and from (2.44), $\hat{\eta}_1 = \hat{G}_1^1(\Omega)\hat{y}_1$. From (1.2),

$$\hat{\eta}_1 = \sum_{m=-1}^{m=1} \hat{a}_1^m P_1^{|m|}(cos\theta)e^{im\phi}, \tag{2.69}$$

and from (2.18),

$$\hat{y}_1 = \sum_{m=-1}^{m=1} \hat{y}_1^m P_1^{|m|}(cos\theta)e^{im\phi}. \tag{2.70}$$

Because of (1.29), $a_1^0 = b_1^0$; all this may be combined to give

$$\hat{b}_1^0(\Omega) = \hat{G}_1^1(\Omega)\hat{y}_1^0. \tag{2.71}$$

In terms of Laplace transforms this is

$$\check{b}_1^0(s) = \hat{G}_1^1(is)\check{y}_1^0. \tag{2.72}$$

In the expression (2.55) for D_{zz}, b_1^0 rather than $\check{b}_1^0$ appears, so the inverse Laplace transform must be taken according to (2.62):

$$b_1^0(t) = \frac{1}{i}\int_{\gamma-i\infty}^{\gamma+i\infty} e^{st}\check{b}_1^0(s)\ ds, \tag{2.73}$$

where γ is any positive real number; substitution of this into (2.68) gives

$$D_{zz} = \lim_{t\to\infty}\frac{d}{dt}\left\langle -\int_{\gamma-i\infty}^{\gamma+i\infty}\int_{\gamma-i\infty}^{\gamma+i\infty} e^{s_1t}e^{s_2t}\hat{G}_1^1(is_1)\hat{G}_1^1(is_2)\check{y}_1^0(s_1)\check{y}_1^0(s_2)ds_1ds_2\right\rangle. \tag{2.74}$$

The assumption will be made that the random normal stress that the drop surface experiences changes on a much faster time scale than the macroscopic drop deformation, since it is caused by a large number of microscopic events (namely, collisions of fluid molecules). Thus, for the calculation here, the random normal stress will be assumed to have essentially zero autocorrelation time:

$$< y(\theta,\phi,t)y(\theta,\phi,t+\tau) >= C_y\delta(\tau)\ , \tag{2.75}$$

where C_y is some constant (which will be determined below from the equilibrium autocorrelations.) Thus, specifically,

$$< y_1^0(\theta,\phi,t)y_1^0(\theta,\phi,t+\tau) >= C_y^{1,0}\delta(\tau). \tag{2.76}$$

Note that in (2.74), the term

$$R_y =< \check{y}_1^0(s_1)\check{y}_1^0(s_2) > \tag{2.77}$$

appears (since the expectation operator $<>$ can be commuted with the integrals). Substituting the formula for the Laplace transform into this gives

$$\begin{aligned} R_y &= \left\langle \frac{1}{(2\pi)^2}\int_0^\infty \int_0^\infty e^{-s_1\tau_1}e^{-s_2\tau_2}y_1^0(\tau_1)y_1^0(\tau_2)\ d\tau_1\ d\tau_2 \right\rangle \\ &= \frac{1}{(2\pi)^2}\int_0^\infty \int_0^\infty e^{-s_1\tau_1}e^{-s_2\tau_2}\left\langle y_1^0(\tau_1)y_1^0(\tau_2)\right\rangle\ d\tau_1\ d\tau_2, \end{aligned} \tag{2.78}$$

which, using (2.76), becomes

$$\begin{aligned} R_y &= \frac{1}{(2\pi)^2}\int_0^\infty \int_0^\infty e^{-s_1\tau_1}e^{-s_2\tau_2}C_y^{1,0}\delta(\tau_1-\tau_2)\ d\tau_1\ d\tau_2 \\ &= \frac{1}{(2\pi)^2}\int_0^\infty e^{-s_1\tau}e^{-s_2\tau}C_y^{1,0}\ d\tau \\ &= \frac{C_y^{1,0}}{(2\pi)^2(s_1+s_2)}. \end{aligned} \tag{2.79}$$

Substituting this into (2.74) gives

$$D_{zz} = \lim_{t \to \infty} \frac{d}{dt}\Big[-\int_{\gamma-i\infty}^{\gamma+i\infty}\int_{\gamma-i\infty}^{\gamma+i\infty} e^{s_1 t}e^{s_2 t}\hat{G}_1^1(is_1)\hat{G}_1^1(is_2)\frac{C_y^{1,0}}{(2\pi)^2(s_1+s_2)}ds_1 ds_2\Big]. \tag{2.80}$$

The time derivative may be commuted with the integrals, resulting in the simpler expression

$$D_{zz} = \lim_{t \to \infty}\Big[-\int_{\gamma-i\infty}^{\gamma+i\infty}\int_{\gamma-i\infty}^{\gamma+i\infty} e^{s_1 t}e^{s_2 t}\hat{G}_1^1(is_1)\hat{G}_1^1(is_2)\frac{C_y^{1,0}}{(2\pi)^2}ds_1 ds_2\Big]. \tag{2.81}$$

These integrals can be evaluated by residue calculus. Here γ is any real positive number; i.e., the transfer functions $\hat{G}_1^1$ should have poles only in the left half-plane for physical reasons. Since when $n = 1$ the coefficient $\tilde{Q} = 0$ by (2.50q), it is clear from (2.52) that $\hat{G}_1^1$ has a simple pole at the origin. Assume that the other poles of $\hat{G}_1^1$ are simple, and denote them by r_i, with $r_0 = 0$. Then doing the s_1 integral gives

$$D_{zz} = \lim_{t \to \infty}\Big[-(2\pi i)\int_{\gamma-i\infty}^{\gamma+i\infty}\sum_i e^{r_i t}e^{s_2 t}R_i\hat{G}_1^1(is_2)\frac{C_y^{1,0}}{(2\pi)^2}ds_2\Big], \tag{2.82}$$

where R_i is the residue from the i-th pole (except for the exponential term); i.e.,

$$R_i = \lim_{s_1 \to r_i}(s_1 - r_i)\hat{G}_1^1(is_1). \tag{2.83}$$

Similarly, doing the s_2 integral gives

$$D_{zz} = \lim_{t \to \infty}\Big[-(2\pi i)^2\sum_i\sum_j e^{r_i t}e^{r_j t}R_iR_j\frac{C_y^{1,0}}{(2\pi)^2}\Big]. \tag{2.84}$$

Now clearly, in the long-time limit, all the terms from poles r_i with negative real part will go to zero. Thus, only the terms for $i = 0$ and $j = 0$ will remain, giving

$$D_{zz} = \left[-(2\pi i)^2 (R_0)^2 \frac{C_y^{1,0}}{(2\pi)^2} \right]. \tag{2.85}$$

The constant $C_y^{1,0}$ in (2.85) is as yet unknown and will now be determined from the equilibrium correlation (1.57). From (2.72) it is clear that

$$\check{\dot{b}}_1^0 = s\ \check{b}_1^0 = s\ \hat{G}_1^1(is)\ \check{y}_1^0(s), \tag{2.86}$$

where the dot over a variable indicates its time derivative. By again using the inversion formula (2.62), this implies

$$< \dot{b}_1^0(t)\dot{b}_1^0(t) >= \left\langle - \int_{\gamma-i\infty}^{\gamma+i\infty} \int_{\gamma-i\infty}^{\gamma+i\infty} e^{s_1 t} e^{s_2 t} s_1 s_2 \hat{G}_1^1(is_1)\hat{G}_1^1(is_2)\check{y}_1^0(s_1)\check{y}_1^0(s_2) ds_1 ds_2 \right\rangle, \tag{2.87}$$

which by manipulations very similar to those done in calculating the diffusivity, and in particular by using (2.79), can be reduced to

$$< \dot{b}_1^0(t)\dot{b}_1^0(t) >= - \int_{\gamma-i\infty}^{\gamma+i\infty} \int_{\gamma-i\infty}^{\gamma+i\infty} e^{s_1 t} e^{s_2 t} s_1 s_2 \hat{G}_1^1(is_1)\hat{G}_1^1(is_2) \frac{C_y^{1,0}}{(2\pi)^2 (s_1 + s_2)} ds_1 ds_2. \tag{2.88}$$

What is needed, for comparison with the equilibrium correlation derived in Section 1, is the long-time limit of this. If the s_1 integral is considered first, there will be residues from the poles of $\hat{G}_1^1$ and from the simple pole at $s_1 = -s_2$. It can be shown, however, that only the residue from $s_1 = -s_2$ survives as $t \to \infty$. Thus,

$$< \dot{b}_1^0(t)\dot{b}_1^0(t) >= (2\pi i) \int_{\gamma-i\infty}^{\gamma+i\infty} s^2 \hat{G}_1^1(is)\hat{G}_1^1(-is) \frac{C_y^{1,0}}{(2\pi)^2} ds. \tag{2.89}$$

Combining (1.57) and (2.89) gives

$$C_y^{1,0} = -\frac{\kappa_B T}{m_D}(2\pi i)\left[\int_{\gamma-i\infty}^{\gamma+i\infty} s^2 \hat{G}_1^1(is)\hat{G}_1^1(-is)ds\right]^{-1} \tag{2.90}$$

for the coefficient in the autocorrelation of the component of the random stress, y_1^0. From (2.85), the diffusivity is then

$$D_{zz} = -\frac{\kappa_B T}{m_D}(2\pi i)\left[\int_{\gamma-i\infty}^{\gamma+i\infty} s^2 \hat{G}_1^1(is)\hat{G}_1^1(-is)ds\right]^{-1}(R_0)^2\Big], \tag{2.91}$$

where

$$R_0 = \lim_{s \to 0} s\hat{G}_1^1(is). \tag{2.92}$$

The transfer function $\hat{G}_1^1$ appearing in (2.91) can be simplified to

$$\hat{G}_1^1 = \frac{\beta}{i\Omega}\frac{Num}{Denom}, \tag{2.93}$$

where

$$Num = 24\frac{i\Omega}{\lambda}f_2\psi_2 - \frac{72}{\lambda}f_1\psi_2 + \left[216\frac{1}{i\Omega}\left(1 - \frac{1}{\lambda} + \frac{24}{\lambda} - \frac{12\alpha}{\lambda}\right)\right]f_0\psi_2$$
$$-\frac{72}{i\Omega}f_0\psi_1 - \frac{12}{i\Omega}f_0\psi_0; \tag{2.94}$$

$$Denom = 36i\Omega\frac{\lambda-1}{\lambda}f_2\psi_2 + 36i\Omega f_2\psi_1 - 18i\Omega f_2\psi_0$$
$$-\left[36\frac{i\Omega}{\lambda} + 432\frac{\lambda-1}{\lambda}\right]f_1\psi_0 + \left[144\alpha\frac{\lambda-1}{\lambda} + 18\frac{i\Omega\alpha}{\lambda}\right]f_0\psi_2 - 72\alpha f_0\psi_1. \tag{2.95}$$

In the above, the arguments of all the functions f_n are $\sqrt{i\Omega}$, and the arguments of all the functions ψ_n are $\sqrt{i\Omega\alpha/\lambda}$. The functions involved in these expressions are

$$f_0(x) = \frac{e^{-ix}}{x}; \tag{2.96a}$$

$$f_1(x) = e^{-ix}\Big[\frac{1}{x^3} + \frac{i}{x^2}\Big]; \tag{2.96b}$$

$$f_2(x) = e^{-ix}\Big[\frac{3i}{x^4} - \frac{1}{x^3} + \frac{3}{x^5}\Big]; \tag{2.96c}$$

$$f_3(x) = e^{-ix}\Big[-\frac{i}{x^4} - \frac{6}{x^5} + \frac{15i}{x^6} + \frac{15}{x^7}\Big]; \tag{2.96d}$$

and

$$\psi_0(x) = \frac{sinx}{x}; \tag{2.97a}$$

$$\psi_1(x) = \frac{-xcosx + sinx}{x^3}; \tag{2.97b}$$

$$\psi_2(x) = \frac{-3xcosx + 3sinx - x^2 sinx}{x^5}; \tag{2.97c}$$

$$\psi_3(x) = \frac{x^3 cosx - 6x^2 sinx - 15xcosx + 15sinx}{x^7}. \tag{2.97d}$$

The velocity-autocorrelation function for the drop is also of interest. In the long-time limit, this approaches the function

$$< \dot{b}_1^0(t)\dot{b}_1^0(t+\tau) >= \frac{iC_y^{1,0}}{(2\pi)} \int_{\gamma-i\infty}^{\gamma+i\infty} e^{s\tau} s^2 \hat{G}_1^1(is)\hat{G}_1^1(-is)ds \tag{2.98}$$

(with $C_y^{1,0}$ given in (2.90)), which depends only on the time difference τ.

For the higher-degree spherical harmonic modes, $n = 2, 3, ...$, the equilibrium correlation gives $< b_n^m b_n^m >$ rather than $< \dot{b}_n^m \dot{b}_n^m >$, so the calculation of $C_y^{n,m}$ is slightly different. Thus,

$$< b_n^m(t)b_n^m(t+\tau) >= -\int_{\gamma-i\infty}^{\gamma+i\infty}\int_{\gamma-i\infty}^{\gamma+i\infty} e^{s_1 t}e^{s_2(t+\tau)}$$

$$\hat{G}_n^1(is_1)\hat{G}_n^1(is_2)\frac{C_y^{n,m}}{(2\pi)^2(s_1+s_2)}ds_1 ds_2, \tag{2.99}$$

which for $t \to \infty$ and $\tau = 0$ gives the following expression,

$$\lim_{t\to\infty} < b_n^m(t) b_n^m(t) >= -i \int_{\gamma-i\infty}^{\gamma+i\infty}\int_{\gamma-i\infty}^{\gamma+i\infty} \hat{G}_n^1(is)\hat{G}_n^1(-is)\frac{C_y^{n,m}}{(2\pi)}ds, \tag{2.100}$$

which, by comparison with the equilibrium correlations (1.55), gives

$$C_y^{n,m} = \frac{\kappa T}{4\pi\gamma}\left[1+\frac{n(n+1)}{2}\right]^{-1}\left(n+\frac{1}{2}\right)\frac{(n-|m|)!}{(n+|m|)!}$$
$$\cdot i\left[\int_{\gamma-i\infty}^{\gamma+i\infty}\int_{\gamma-i\infty}^{\gamma+i\infty} \hat{G}_n^1(is)\hat{G}_n^1(-is)\frac{1}{(2\pi)}ds\right]^{-1}. \tag{2.101}$$

The time autocorrelation of the amplitude of the n, m mode is then (for long times)

$$< b_n^m(t) b_n^m(t+\tau) >= -i \int_{\gamma-i\infty}^{\gamma+i\infty}\int_{\gamma-i\infty}^{\gamma+i\infty} e^{s\tau}\hat{G}_n^1(is)\hat{G}_n^1(-is)\frac{C_y^{n,m}}{(2\pi)}ds, \tag{2.102}$$

with $C_y^{n,m}$ given by (2.89), and the transfer function $\hat{G}_n^1$ given by (2.45). This is the fluctuation-dissipation theorem for the deformations; it relates the autocorrelation of the deformation amplitudes to the macroscopic properties of surface tension, viscosity and density. It is interesting to note that as the surface tension becomes very large,

$$\hat{G}_n(\Omega) \to -\frac{1}{\tilde{Q}} = \frac{2}{\Gamma[2-n(n+1)]}, \tag{2.103}$$

which is independent of Ω. Thus, as the surface tension becomes large, the time dependence of the autocorrelations of the amplitudes of the higher-order harmonics approaches a delta function.

FIRST CORRECTION TO CENTER-OF-MASS MOTION DUE TO FINITE SURFACE-TENSION

Since the spherical harmonic modes decouple to $O(\epsilon)$ (where ϵ denotes some measure of the magnitude of the deformation), to get any dependence of the mean-square displacement on surface tension, the $O(\epsilon^2)$ problem must be considered. In the $O(\epsilon^2)$ boundary conditions, products of the $O(\epsilon)$ quantities appear. These products must be expanded in spherical harmonics, and in general the product of an n-th order harmonic with a j-th order harmonic will have a first-order term, no matter how large n and j get. *Every* order of harmonics in the $O(\epsilon)$ solution will thus contribute to the $O(\epsilon^2)$ first-order harmonics; i.e., every term in the $O(\epsilon)$ solution will contribute to the center-of-mass motion. While the complete second-order solution could be written out in terms of the Clebsch-Gordon coefficients for the expansion of products of associated Legendre polynomials, this is hardly worth doing in a treatment that is asymptotic and therefore approximate anyway. What is of more interest here is just an approximation of the effect of surface tension on the drop center-of-mass motion. The probability of a deformation corresponding to a spherical harmonic of order n decreases with increasing n, and the rate of decrease becomes rapid as the surface tension increases (corresponding to a very "stiff" drop.) Thus, to get an approximation to the effect of surface tension for the case of large surface tension, all the harmonics beyond the N-th one in the $O(\epsilon)$ solution could be assumed to be zero for purposes of calculating the $O(\epsilon^2)$ solution. The minimum value of N to get any dependence on surface tension is 2.

Let the various flow variables have the expansions

$$\epsilon \mathbf{u}^i + \epsilon^2 \mathbf{v}^i + \ldots \tag{3.1a}$$

$$\epsilon p^i + \epsilon^2 q^i + ... \tag{3.1b}$$

$$\epsilon \eta + \epsilon^2 \xi + ... \tag{3.1c}$$

$$\epsilon \chi^i + \epsilon^2 \Xi^i + ... \tag{3.1d}$$

$$\epsilon \varphi^i + \epsilon^2 \Phi^i + ... \tag{3.1e}$$

$$\epsilon \sigma^i + \epsilon^2 \Sigma^i + ... \tag{3.1f}$$

and similarly for the outside solution, with the superscript "i" replaced by "o." The time-dependent hydrodynamic problem solved in Section 2 was of course the $O(\epsilon)$ problem, involving $\mathbf{u}^i$,$\mathbf{u}^o$, η, etc. The procedure that will be used here to calculate $\mathbf{v}^i$,$\mathbf{v}^o$, etc., is that outlined in Brenner's 1964 paper [6].

First, it is useful to note that the unit vectors normal and tangent to the surface have expansions

$$\mathbf{t}_\theta = \mathbf{e}_\theta + \epsilon \frac{\partial \eta}{\partial \theta} \mathbf{e}_r = \mathbf{e}_\theta + \mathbf{t}'_\theta; \tag{3.2a}$$

$$\mathbf{t}_\phi = \mathbf{e}_\phi + \epsilon \frac{1}{\sin\theta} \frac{\partial \eta}{\partial \phi} \mathbf{e}_r = \mathbf{e}_\phi + \mathbf{t}'_\phi; \tag{3.2b}$$

$$\mathbf{n} = \mathbf{e}_r - \epsilon \frac{\partial \eta}{\partial \theta} \mathbf{e}_\theta - \epsilon \frac{1}{\sin\theta} \frac{\partial \eta}{\partial \phi} \mathbf{e}_\phi = \mathbf{e}_r + \mathbf{n}'. \tag{3.2c}$$

Next, the expansion of the surface-tension term is needed. It was shown above (1.8) that

$$\mathbf{n}(\theta,\phi) = \left. \frac{\mathbf{e}_r - \frac{1}{r}\mathbf{e}_\theta \frac{\partial(\epsilon\eta + \epsilon^2\xi)}{\partial\theta} - \mathbf{e}_\phi \frac{1}{r sin\theta} \frac{\partial(\epsilon\eta + \epsilon^2\xi)}{\partial\phi}}{\left[1 + \frac{1}{r^2}\left(\frac{\partial(\epsilon\eta + \epsilon^2\xi)}{\partial\theta}\right)^2 + \frac{1}{r^2 sin^2\theta}\left(\frac{\partial(\epsilon\eta + \epsilon^2\xi)}{\partial\phi}\right)^2\right]^{\frac{1}{2}}} \right|_{r=1}. \tag{3.3}$$

From this it follows that

$$\nabla \cdot \mathbf{n} = 2 + \epsilon \left[-2\eta - \frac{\partial^2 \eta}{\partial \theta^2} - \frac{\cos\theta}{\sin\theta} \frac{\partial \eta}{\partial \theta} - \frac{1}{\sin\theta} \frac{\partial^2 \eta}{\partial \phi^2} \right]$$

$$+\epsilon^2 \left[-2\xi - \frac{\partial^2 \xi}{\partial \theta^2} - \frac{\cos\theta}{\sin\theta} \frac{\partial \xi}{\partial \theta} - \frac{1}{\sin\theta} \frac{\partial^2 \xi}{\partial \phi^2} \right.$$

$$\left. +2\eta \left(\eta + \frac{\partial^2 \eta}{\partial \theta^2} + \frac{\cos\theta}{\sin\theta} \frac{\partial \eta}{\partial \theta} + \frac{1}{\sin\theta} \frac{\partial^2 \eta}{\partial \phi^2} \right) \right] \quad . \tag{3.4}$$

The $O(\epsilon)^2$ deformation ξ has a surface harmonic expansion, just like η:

$$\xi = \sum_{n=1}^{\infty} \xi_n. \tag{3.5}$$

It was shown earlier that each surface harmonic satisfies

$$\left[2\hat{\eta}_n + \frac{cos\theta}{sin\theta} \frac{\partial \hat{\eta}_n}{\partial \theta} + \frac{\partial^2 \hat{\eta}_n}{\partial \theta^2} + \frac{1}{sin^2\theta} \frac{\partial^2 \hat{\eta}_n}{\partial \phi^2} \right] = \left[2 - n(n+1) \right] \hat{\eta}_n, \tag{3.6}$$

which is just a consequence of Legendre's equation. Of course, the same equation holds with ξ substituted for η. With this, the expression for $\nabla \cdot \mathbf{n}$ can be simplified to

$$\nabla \cdot \mathbf{n} = 2 + \epsilon \sum_{n=1}^{\infty} (-)[2 - n(n+1)]\eta_n$$

$$+\epsilon^2 \sum_{n=1}^{\infty} \left[-[2 - n(n+1)]\xi_n + \sum_{m=0}^{\infty} 2\eta_n([2 - n(n+1)] - 1)\eta_m \right] + \ldots \quad . \tag{3.7}$$

The kinematic velocity boundary conditions are

$$[\mathbf{e}_r + \epsilon \mathbf{n}'] \cdot \left[\epsilon \mathbf{u}^i + \epsilon^2 \left(\eta \frac{\partial \mathbf{u}^i}{\partial r} + \mathbf{v}^i \right) \right]$$

$$= [\mathbf{e}_r + \epsilon \mathbf{n}'] \cdot \left[\epsilon \mathbf{u}^o + \epsilon^2 \left(\eta \frac{\partial \mathbf{u}^o}{\partial r} + \mathbf{v}^o \right) \right]$$

$$= -\frac{i\Omega}{\beta}[\epsilon\hat{\eta} + \epsilon^2\hat{\xi}] \quad , \tag{3.8}$$

where all functions of r in this condition (as well as the conditions to be given below) are to be evaluated at $r = 1$, since the correction for the interface being at $r = 1 + \epsilon\eta + \epsilon^2\xi + ...$ has already been made to $O(\epsilon^2)$. Continuity of velocity gives the conditions

$$\epsilon\mathbf{u}^i + \epsilon^2\Big(\eta\frac{\partial\mathbf{u}^i}{\partial r} + \mathbf{v}^i\Big) = \epsilon\mathbf{u}^o + \epsilon^2\Big(\eta\frac{\partial\mathbf{u}^o}{\partial r} + \mathbf{v}^o\Big). \tag{3.9}$$

Collecting the $O(\epsilon^2)$ terms in these equations gives for the $O(\epsilon^2)$ kinematic conditions:

$$\begin{aligned}&\mathbf{e}_r\cdot\Big(\eta\frac{\partial\mathbf{u}^i}{\partial r} + \mathbf{v}^i\Big) + \mathbf{n}'\cdot\mathbf{u}^i\\ &= \mathbf{e}_r\cdot\Big(\eta\frac{\partial\mathbf{u}^o}{\partial r} + \mathbf{v}^o\Big) + \mathbf{n}'\cdot\mathbf{u}^o = -\frac{i\Omega}{\beta}\hat{\xi};\end{aligned} \tag{3.10}$$

and for the $O(\epsilon^2)$ continuity of velocity conditions:

$$\eta\frac{\partial\mathbf{u}^i}{\partial r} + \mathbf{v}^i = \eta\frac{\partial\mathbf{u}^o}{\partial r} + \mathbf{v}^o. \tag{3.11}$$

The stress boundary conditions are given by

$$\begin{aligned}&(\mathbf{e}_r + \epsilon\mathbf{n}')\cdot\Big[-\epsilon\sigma^i - \epsilon^2\Sigma^i - \epsilon^2\eta\frac{\partial\sigma^i}{\partial r} + \epsilon\sigma^o + \epsilon^2\Sigma^o + \epsilon^2\eta\frac{\partial\sigma^o}{\partial r}\Big]\\ &= (\mathbf{e}_r + \epsilon\mathbf{n}')\Big\{\epsilon y + \Gamma\Big[-\epsilon\sum_{n=1}^{\infty}[2 - n(n+1)]\eta_n\\ &+\epsilon^2\sum_{n=1}^{\infty}\Big(-[2 - n(n+1)]\xi_n + \sum_{m=1}^{\infty}2\eta_n([2 - n(n+1)] - 1)\eta_m\Big)\Big]\Big\}.\end{aligned} \tag{3.12}$$

Collecting the terms of $O(\epsilon)^2$ in this gives

$$\mathbf{e}_r \cdot \left[-\Sigma^i - \eta \frac{\partial \sigma^i}{\partial r} + \Sigma^o + \eta \frac{\partial \sigma^o}{\partial r} \right]$$

$$+\mathbf{n}' \cdot \left[-\sigma^i + \sigma^o \right]$$

$$= \mathbf{e}_r \Gamma \left[\sum_{n=1}^{\infty} \left(-[2 - n(n+1)]\xi_n + \sum_{m=1}^{\infty} 2\eta_n([2 - n(n+1)] - 1)\eta_m \right) \right]$$

$$+\mathbf{n}' \left\{ y + \Gamma \left[-\sum_{n=1}^{\infty} [2 - n(n+1)]\eta_n \right] \right\}. \tag{3.13}$$

These conditions may be rewritten as

$$\mathbf{e}_r \cdot \mathbf{v}^i + \frac{i\Omega}{\beta} \hat{\xi} = -\mathbf{e}_r \cdot \eta \frac{\partial \mathbf{u}^i}{\partial r} - \mathbf{n}' \cdot \mathbf{u}^i; \tag{3.14}$$

$$\mathbf{e}_r \cdot \mathbf{v}^o + \frac{i\Omega}{\beta} \hat{\xi} = -\mathbf{e}_r \cdot \eta \frac{\partial \mathbf{u}^o}{\partial r} - \mathbf{n}' \cdot \mathbf{u}^o; \tag{3.15}$$

$$\mathbf{v}^i - \mathbf{v}^o = \eta \frac{\partial \mathbf{u}^o}{\partial r} - \eta \frac{\partial \mathbf{u}^i}{\partial r}; \tag{3.16}$$

$$- \mathbf{e}_r \cdot \Sigma^i + \mathbf{e}_r \cdot \Sigma^o + \mathbf{e}_r \Gamma \sum_{n=1}^{\infty} \left([2 - n(n+1)]\xi_n \right)$$

$$= \epsilon \mathbf{n}' \left\{ y + \Gamma \left[-\sum_{n=1}^{\infty} [2 - n(n+1)]\eta_n \right] \right\}$$

$$+ \mathbf{e}_r \cdot \eta \frac{\partial \sigma^i}{\partial r} - \mathbf{e}_r \cdot \eta \frac{\partial \sigma^o}{\partial r} + \mathbf{e}_r \Gamma \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} 2\eta_n([2 - n(n+1)] - 1)\eta_m$$

$$- \mathbf{n}' \cdot \left[-\sigma^i + \sigma^o \right]; \tag{3.17}$$

to emphasize the fact that the right-hand terms are known, and the left-hand terms, which involve the unknown $O(\epsilon)^2$ flow field, have the same structure as the $O(\epsilon)$

boundary conditions. For convenience, the right-hand sides of (3.14) and (3.15) will be denoted by the scalars V^i and V^o, respectively; the right-hand-side of (3.16) will be denoted by the vector $\mathbf{W}$; and the right-hand side of (3.17) will be denoted by the vector $\mathbf{T}$. Thus, the boundary conditions at $r = 1$ become

$$\mathbf{e}_r \cdot \mathbf{v}^i + \frac{i\Omega}{\beta}\hat{\xi} = V^i(\theta, \phi) \; ; \tag{3.18}$$

$$\mathbf{e}_r \cdot \mathbf{v}^o + \frac{i\Omega}{\beta}\hat{\xi} = V^o(\theta, \phi) \; ; \tag{3.19}$$

$$\mathbf{v}^i - \mathbf{v}^o = \mathbf{W} \; ; \tag{3.20}$$

$$-\mathbf{e}_r \cdot \Sigma^i + \mathbf{e}_r \cdot \Sigma^o + \mathbf{e}_r \Gamma \sum_{n=1}^{\infty} \left([2 - n(n+1)]\xi_n \right) = \mathbf{T} \; , \tag{3.21}$$

where

$$V^i(\theta, \phi) = \sum_{n=1}^{\infty} V_n^i = -\mathbf{e}_r \cdot \eta \frac{\partial \mathbf{u}^i}{\partial r} - \mathbf{n}' \cdot \mathbf{u}^i \Bigg|_{r=1} \; ; \tag{3.22}$$

$$V^o(\theta, \phi) = \sum_{n=1}^{\infty} V_n^o = -\mathbf{e}_r \cdot \eta \frac{\partial \mathbf{u}^o}{\partial r} - \mathbf{n}' \cdot \mathbf{u}^o \Bigg|_{r=1} \; ; \tag{3.23}$$

$$\mathbf{W}(\theta, \phi) = \eta \frac{\partial \mathbf{u}^o}{\partial r} - \eta \frac{\partial \mathbf{u}^i}{\partial r} \Bigg|_{r=1} \; ; \tag{3.24}$$

$$\mathbf{T}(\theta, \phi) = \epsilon \mathbf{n}' \left\{ y + \Gamma \left[- \sum_{n=1}^{\infty} [2 - n(n+1)]\eta_n \right] \right\}$$

$$+ \mathbf{e}_r \cdot \eta \frac{\partial \sigma^i}{\partial r} - \mathbf{e}_r \cdot \eta \frac{\partial \sigma^o}{\partial r} + \mathbf{e}_r \Gamma \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} 2\eta_n([2 - n(n+1)] - 1)\eta_m$$

$$- \mathbf{n}' \cdot \Big[-\sigma^i + \sigma^o \Big] \Bigg|_{r=1} . \qquad (3.25)$$

The $O(\epsilon)^2$ flow field has the same form of expansion as the $O(\epsilon)$ flow field:

$$\hat{\mathbf{v}}^i = \sum_{n=1}^{\infty} \Bigg[\frac{1}{i\Omega\alpha} \nabla q_n^i - \psi_n\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}} r\Big) \nabla \times (\mathbf{r}\Xi_n^i)$$

$$+ \Big[(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}} r\Big) - n\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}} r\Big) \Big(\frac{i\Omega\alpha}{\lambda}\Big) r^2 \Big] \nabla \Phi_n^i$$

$$+ n(2n+1)\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}} r\Big) \Big(\frac{i\Omega\alpha}{\lambda}\Big) \Phi_n^i \mathbf{r} \Bigg] ; \qquad (3.26a)$$

$$\hat{q}^i = \sum_{n=1}^{\infty} q_n^i , \qquad (3.26b)$$

for the inside flow, and

$$\hat{\mathbf{v}}^o = \sum_{n=1}^{\infty} \Bigg[\frac{1}{i\Omega} \nabla q_{-(n+1)}^o - f_n(\sqrt{i\Omega} r) \nabla \times (\mathbf{r}\Xi_n^o)$$

$$+ \Big[(n+1) f_{n-1}(\sqrt{i\Omega} r) - n f_{n+1}(\sqrt{i\Omega} r)(i\Omega) r^2 \Big] \nabla \Phi_n^o$$

$$+ n(2n+1) f_{n+1}(\sqrt{i\Omega} r)(i\Omega) \Phi_n^o \mathbf{r} \Bigg] ; \qquad (3.27b)$$

$$\hat{q}^o = \sum_{n=1}^{\infty} \hat{q}_{-(n+1)}^o . \qquad (3.27b)$$

Similarly, the $O(\epsilon)^2$ stress on the (undeformed) sphere surface has the same form of expansion as the $O(\epsilon)$ stress does:

$$\mathbf{e}_r \cdot \mathbf{\Sigma}^o = \frac{1}{r}\sum_{n=1}^{\infty}\Big[-\frac{2(n+2)}{(i\Omega)}\nabla q^o_{-(n+1)}$$

$$-Q^o_n(\sqrt{i\Omega}r)\nabla \times (\mathbf{r}\Xi^o_n) + R^o_n(\sqrt{i\Omega}r)\nabla\Phi^o_n$$

$$-\frac{(2n+1)}{r^2}S^o_n(\sqrt{i\Omega}r)\Phi^o_n\mathbf{r} - q^o_{-(n+1)}\mathbf{r}\Big] \tag{3.28}$$

for the exterior flow, and

$$\mathbf{e}_r \cdot \mathbf{\Sigma}^i = \frac{1}{r}\sum_{n=1}^{\infty}\Big\{\lambda\Big[\frac{2(n-1)}{(i\Omega)\alpha}\nabla q^i_n$$

$$-Q^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\nabla \times (\mathbf{r}\Xi^i_n) + R^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\nabla\Phi^i_n$$

$$-\frac{(2n+1)}{r^2}S^i_n(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\Phi^i_n\mathbf{r}\Big] - q^i_n\mathbf{r}\Big\} \tag{3.29}$$

for the interior flow.

Now these general solution expansions will be substituted into the boundary conditions. This step is very similar to what was done above for the $O(\epsilon)$ solution, so only the resulting equations will be given here. The kinematic conditions become (for $n = 1, 2, 3, ...$):

$$\frac{i\Omega}{\beta}\hat{\xi}_n + \frac{n}{i\Omega\alpha}q^i_n + \Big[n(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big) + n(n+1)\frac{i\Omega\alpha}{\lambda}\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)\Big]\Phi^i_n = V^i_n, \tag{3.30}$$

and

$$\frac{i\Omega}{\beta}\hat{\xi}_n + -\frac{(n+1)}{i\Omega}q^o_{-(n+1)} + \big[n(n+1)f_{n-1}(\sqrt{i\Omega}) + n(n+1)i\Omega f_{n+1}(\sqrt{i\Omega})\big]\Phi^o_n = V^o_n. \tag{3.31}$$

Continuity of tangential velocity gives

$$\frac{n(n-1)}{i\Omega\alpha}q_n^i$$

$$+\Bigg[n(n-1)(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)-n(n+1)\frac{i\Omega\alpha}{\lambda}\psi_n\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)$$

$$+n(n+1)^2\frac{i\Omega\alpha}{\lambda}\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)-n(n+1)\Big(\frac{i\Omega\alpha}{\lambda}\Big)^2\psi_{n+2}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)\Bigg]\Phi_n^i\Bigg|_{r=1}$$

$$-\frac{(n+1)(n+2)}{i\Omega}q^o_{-(n+1)}$$

$$-\Bigg[n(n-1)(n+1)f_{n-1}(\sqrt{i\Omega})-n(n+1)(i\Omega)f_n(\sqrt{i\Omega})$$

$$+n(n+1)^2(i\Omega)f_{n+1}(\sqrt{i\Omega})-n(n+1)(i\Omega)^2f_{n+2}(\sqrt{i\Omega})\Bigg]\Phi_n^o\Bigg|_{r=1}=-r\nabla\cdot\mathbf{W},\quad(3.32)$$

and

$$-n(n+1)\psi_n\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)\Xi_n^i+n(n+1)f_n(\sqrt{i\Omega}r)\Xi_n^i=\mathbf{r}\cdot\nabla\mathbf{W}.\qquad(3.33)$$

Continuity of tangential stress leads to the conditions (again for $n=1,2,3,...$):

$$\frac{2\lambda n(n-1)(n-2)}{i\Omega\alpha}q_n^i-nq_n^i$$

$$+\lambda\Bigg[n(n-2)R_n^i\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)+n\frac{dR_n^i\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)}{dr}$$

$$-(2n+1)(n-2)S_n^i\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)-(2n+1)\frac{dS_n^i\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)}{dr}\Bigg]\Phi_n^i\Bigg|_{r=1}$$

$$+\frac{2(n+1)(n+2)(n+3)}{i\Omega}q^o_{-(n+1)}-(n+1)q^o_{-(n+1)}$$

$$-\lambda\Bigg[n(n-2)R_n^o(\sqrt{i\Omega})+n\frac{dR_n^o(\sqrt{i\Omega}r)}{dr}$$

$$-(2n+1)(n-2)S_n^o(\sqrt{i\Omega}r)-(2n+1)\frac{dS_n^o(\sqrt{i\Omega}r)}{dr}\Bigg]\Phi_n^o\Bigg|_{r=1}$$

$$=-r\nabla\cdot\mathbf{T}; \tag{3.34}$$

and

$$-n(n+1)Q_n^i\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)\Xi_n^i+n(n+1)Q_n^o(\sqrt{i\Omega}r)\Xi_n^i=\mathbf{r}\cdot\nabla\mathbf{T}. \tag{3.35}$$

Finally, the normal stress condition gives

$$\Big[\frac{2(n+1)(n+2)}{(i\Omega)r}q_{-(n+1)}^o-R_n^o(\sqrt{i\Omega}r)\frac{n\Phi_n^o}{r}$$

$$+\frac{(2n+1)}{r^2}S_n^o(\sqrt{i\Omega}r)\Phi_n^o r+q_{-(n+1)}^o r\Big]$$

$$-\Bigg\{\lambda\Big[\frac{2(n-1)n}{(i\Omega)\alpha r}q_n^i-R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n\Phi_n^i}{r}$$

$$+\frac{(2n+1)}{r^2}S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\Phi_n^i r\Big]+q_n^i r\Bigg\}$$

$$+\frac{\Gamma}{2}[2-n(n+1)]\hat{\xi}_n=\mathbf{e}_r\cdot\mathbf{T}. \tag{3.36}$$

Clearly, it is necessary to have the surface harmonic expansions of V^i, V^o, $-r\nabla\cdot\mathbf{W}$, $\mathbf{r}\cdot\nabla\times\mathbf{W}$, $-r\nabla\cdot\mathbf{T}$, $\mathbf{r}\cdot\nabla\times\mathbf{T}$, and $\mathbf{e}_r\cdot\mathbf{T}$, which involve the $O(\epsilon)$ solution, to be able to solve these equations for the $O(\epsilon)$ flow variables. However, since what is of interest here is only the contribution of the $O(\epsilon)$ solution to the diffusivity, only ξ (the $O(\epsilon^2)$ deformation) has to be solved for. Since the two equations involving Ξ_n^i and Ξ_n^o decouple from the five equations involving q_n^i,$q_{-(n+1)}^o$, Φ_n^i,Φ_n^o, and ξ_n, exactly as happened in the $O(\epsilon)$ problem, these two equations can be dispensed with. It is therefore not actually necessary here to expand $\mathbf{r}\cdot\nabla\times\mathbf{W}$ and $\mathbf{r}\cdot\nabla\times\mathbf{T}$ in surface harmonics .

These surface harmonic expansions may be written as

$$V^i = \sum_{n=1}^{\infty} V_n^i, \tag{3.37a}$$

$$V^o = \sum_{n=1}^{\infty} V_n^o, \tag{3.37b}$$

$$-r\nabla\cdot\mathbf{W} = 0, \tag{3.37c}$$

$$-r\nabla\cdot\mathbf{T} = \sum_{n=1}^{\infty} T_n^a, \tag{3.37d}$$

$$\mathbf{e}_r\cdot\mathbf{T} = \sum_{n=1}^{\infty} T_n^b, \tag{3.37e}$$

where the expansion terms are calculated in Appendix 2. The five equations may then be written in a way completely analogous to the $O(\epsilon)$ calculation, as

$$V_n^i - \frac{i\Omega}{\beta}\hat{\xi}_n = \tilde{A}q_n^i + \tilde{B}\Phi_n^i, \tag{3.38a}$$

$$V_n^o - \frac{i\Omega}{\beta}\hat{\xi}_n = \tilde{C}q_{-(n+1)}^o + \tilde{D}\Phi_n^o, \tag{3.38b}$$

$$\tilde{E}\Phi_n^i + \tilde{F}q_n^i = \tilde{G}\Phi_n^o + \tilde{H}q_{-(n+1)}^o, \tag{3.38c}$$

$$\tilde{I}q_n^i + \tilde{J}\Phi_n^i - \tilde{K}q_{-(n+1)}^o - \tilde{L}\Phi_n^o = T_n^a, \tag{3.38d}$$

and

$$\tilde{M}q^i_n + \tilde{N}\Phi^i_n + \tilde{O}q^o_{-(n+1)} + \tilde{P}\Phi^o_n = \tilde{Q}\hat{\xi}_n + T^b_n, \tag{3.38e}$$

which in matrix form is

$$\begin{pmatrix} \tilde{A} & 0 & \tilde{B} & 0 & \frac{-i\Omega}{\beta} \\ 0 & \tilde{C} & 0 & \tilde{D} & \frac{-i\Omega}{\beta} \\ \tilde{F} & -\tilde{H} & \tilde{E} & -\tilde{G} & 0 \\ \tilde{I} & -\tilde{K} & \tilde{J} & -\tilde{L} & 0 \\ \tilde{M} & \tilde{O} & \tilde{N} & \tilde{P} & -\tilde{Q} \end{pmatrix} \begin{pmatrix} q^i_n \\ q^o_{-(n+1)} \\ \Phi^i_n \\ \Phi^o_n \\ \hat{\xi}_n \end{pmatrix} = \begin{pmatrix} V^i_n \\ V^o_n \\ 0 \\ T^a_n \\ T^b_n \end{pmatrix}. \tag{3.39}$$

The solution to this system is

$$\xi_n = \frac{1}{\Delta_0}\Big[\Delta_1 V^i_n - \Delta_2 V^o_n + \Delta_3 T^a_n - \Delta_4 T^b_n\Big], \tag{3.40}$$

where $\Delta_0, \ldots$ are the determinants

$$\Delta_0 = \begin{pmatrix} \tilde{A} & 0 & \tilde{B} & 0 & \frac{-i\Omega}{\beta} \\ 0 & \tilde{C} & 0 & \tilde{D} & \frac{-i\Omega}{\beta} \\ \tilde{F} & -\tilde{H} & \tilde{E} & -\tilde{G} & 0 \\ \tilde{I} & -\tilde{K} & \tilde{J} & -\tilde{L} & 0 \\ \tilde{M} & \tilde{O} & \tilde{N} & \tilde{P} & -\tilde{Q} \end{pmatrix}; \tag{3.41a}$$

$$\Delta_1 = \begin{vmatrix} 0 & \tilde{C} & 0 & \tilde{D} \\ \tilde{F} & -\tilde{H} & \tilde{E} & -\tilde{G} \\ \tilde{I} & -\tilde{K} & \tilde{J} & -\tilde{L} \\ \tilde{M} & \tilde{O} & \tilde{N} & \tilde{P} \end{vmatrix}; \tag{3.41b}$$

$$\Delta_2 = \begin{vmatrix} \tilde{A} & 0 & \tilde{B} & 0 \\ \tilde{F} & -\tilde{H} & \tilde{E} & -\tilde{G} \\ \tilde{I} & -\tilde{K} & \tilde{J} & -\tilde{L} \\ \tilde{M} & \tilde{O} & \tilde{N} & \tilde{P} \end{vmatrix}; \tag{3.41c}$$

$$\Delta_3 = \begin{vmatrix} \tilde{A} & 0 & \tilde{B} & 0 \\ 0 & \tilde{C} & 0 & \tilde{D} \\ \tilde{F} & -\tilde{H} & \tilde{E} & -\tilde{G} \\ \tilde{M} & \tilde{O} & \tilde{N} & \tilde{P} \end{vmatrix}; \tag{3.41d}$$

$$\Delta_4 = \begin{vmatrix} \tilde{A} & 0 & \tilde{B} & 0 \\ 0 & \tilde{C} & 0 & \tilde{D} \\ \tilde{F} & -\tilde{H} & \tilde{E} & -\tilde{G} \\ \tilde{I} & -\tilde{K} & \tilde{J} & -\tilde{L} \end{vmatrix}. \tag{3.41e}$$

It is helpful to note that the determinant Δ_0 was already computed for the $O(\epsilon)$ problem.

From Appendix 2,

$$V^i_{1,0}(\theta,\phi) =$$

$$\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j} \hat{H}^{i,1}(n,m,j,k;\Omega)\eta_{j,k}p^i_{n,m} + \hat{H}^{i,2}(n,m,j,k;\Omega)\eta_{j,k}\varphi^i_{n,m}; \tag{3.42}$$

$$V^o_{1,0}(\theta,\phi) =$$

$$\sum_{n=1}^{2}\sum_{j=1}^{2}\sum_{m=-n}^{n}\sum_{k=-j}^{j} \hat{H}^{o,1}(n,m,j,k;\Omega)\eta_{j,k}p^o_{n,m} + \hat{H}^{o,2}(n,m,j,k;\Omega)\eta_{j,k}\varphi^o_{n,m}; \tag{3.43}$$

$$T^a_{1,0} =$$

$$\begin{aligned}\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j} \Big\{ &\hat{H}^3(n,m,j,k;\Omega)\eta_{j,k}\eta_{n,m} + \hat{H}^4(n,m,j,k;\Omega)\eta_{j,k}\hat{y}_{n,m} \\ &+\hat{H}^{i,5}\eta_{j,k}p^i_{n,m} + \hat{H}^{i,6}\eta_{j,k}\varphi^i_{n,m} \\ &+\hat{H}^{o,5}\eta_{j,k}p^o_{-(n+1),m} + \hat{H}^{o,6}\eta_{j,k}\varphi^o_{n,m}\Big\};\end{aligned} \tag{3.44}$$

$$T^b_{1,0} = \mathcal{P}\mathbf{e}_r \cdot \mathbf{T}\Big|_{r=1} =$$

$$\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j} \hat{H}^{o,7}\eta_{j,k}p^o_{-(n+1),m} + \hat{H}^{o,8}\eta_{j,k}\varphi^o_{n,m}$$

$$+\hat{H}^{i,7}\eta_{j,k}p^i_{n,m} + \hat{H}^{i,8}\eta_{j,k}\varphi^i_{n,m} + \hat{H}^9\eta_{j,k}\eta_{n,m} \ , \tag{3.45}$$

where the transfer functions $\hat{H}$ (which are functions of $n, m, j, k, \Omega,$) are given explicitly in Appendix 2. Consider now only translation in the z-direction, which corresponds to the degree 1, order 0 surface harmonic. The $O(\epsilon)$ displacement $\xi_{1,0}$ in the z-direction is:

$$\xi_{1,0} = \frac{1}{\Delta_0} \sum_{n=1}^{\infty} \sum_{j=1}^{\infty} \sum_{m=-n}^{n} \sum_{k=-j}^{j}$$

$$\Big\{ \Delta_1 \Big[\hat{H}^{i,1}(n,m,j,k;\Omega)\eta_{j,k}p^i_{n,m} + \hat{H}^{i,2}(n,m,j,k;\Omega)\eta_{j,k}\varphi^i_{n,m} \Big]$$

$$-\Delta_2 \Big[\hat{H}^{o,1}(n,m,j,k;\Omega)\eta_{j,k}p^o_{n,m} + \hat{H}^{o,2}(n,m,j,k;\Omega)\eta_{j,k}\varphi^o_{n,m} \Big]$$

$$+\Delta_3 \Big[\hat{H}^3(n,m,j,k;\Omega)\eta_{j,k}\eta_{n,m} + \hat{H}^4(n,m,j,k;\Omega)\eta_{j,k}\hat{y}_{n,m}$$

$$+\hat{H}^{i,5}\eta_{j,k}p^i_{n,m} + \hat{H}^{i,6}\eta_{j,k}\varphi^i_{n,m}$$

$$+\hat{H}^{o,5}\eta_{j,k}p^o_{-(n+1),m} + \hat{H}^{o,6}\eta_{j,k}\varphi^o_{n,m} \Big]$$

$$-\Delta_4 \Big[\hat{H}^{o,7}\eta_{j,k}p^o_{-(n+1),m} + \hat{H}^{o,8}\eta_{j,k}\varphi^o_{n,m}$$

$$+\hat{H}^{i,7}\eta_{j,k}p^i_{n,m} + \hat{H}^{i,8}\eta_{j,k}\varphi^i_{n,m} + \hat{H}^9\eta_{j,k}\eta_{n,m} \Big] \Big\}. \tag{3.46}$$

From Section 2, the $O(\epsilon)$ flow variables are given by

$$\eta_{n,m} = \hat{G}^1_{n,m}\hat{y}_{n,m}; \tag{3.47a}$$

$$p^i_{n,m} = \hat{G}^{i,2}_{n,m}\hat{y}_{n,m}; \tag{3.47b}$$

$$p^o_{-(n+1),m} = \hat{G}^{o,2}_{n,m}\hat{y}_{n,m}; \tag{3.47c}$$

$$\varphi^i_n = \hat{G}^{i,3}_{n,m}\hat{y}_{n,m}; \tag{3.47d}$$

$$\varphi^o_n = \hat{G}^{o,3}_{n,m}\hat{y}_{n,m}; \tag{3.47e}$$

where the transfer functions $\hat{G}$ are given there. By using this, the $O(\epsilon)^2$ displacement can be expressed as

$$\xi_{1,0} = \frac{1}{\Delta_0}\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j}\hat{y}_{n,m}(\Omega)\hat{y}_{j,k}(\Omega)$$

$$\Bigg\{\hat{G}^{i,2}_{n,m}(\Omega)\hat{G}^{1}_{j,k}(\Omega)\Bigg[\Delta_1(n,\Omega)\hat{H}^{i,1}(n,m,j,k;\Omega) + \Delta_3(n,\Omega)\hat{H}^{i,5}(n,m,j,k;\Omega)$$

$$-\Delta_4(n,\Omega)\hat{H}^{i,7}(n,m,j,k;\Omega)\Bigg]$$

$$+\hat{G}^{i,3}_{n,m}(\Omega)\hat{G}^{1}_{j,k}(\Omega)\Bigg[\Delta_1(n,\Omega)\hat{H}^{i,2}(n,m,j,k;\Omega) + \Delta_3(n,\Omega)\hat{H}^{i,6}(n,m,j,k;\Omega)$$

$$-\Delta_4(n,\Omega)\hat{H}^{i,8}(n,m,j,k;\Omega)\Bigg]$$

$$+\hat{G}^{o,2}_{n,m}(\Omega)\hat{G}^{1}_{j,k}(\Omega)\Bigg[-\Delta_2(n,\Omega)\hat{H}^{o,1}(n,m,j,k;\Omega) + \Delta_3(n,\Omega)\hat{H}^{o,5}(n,m,j,k;\Omega)$$

$$-\Delta_4(n,\Omega)\hat{H}^{o,7}(n,m,j,k;\Omega)\Bigg]$$

$$+\hat{G}^{o,3}_{n,m}(\Omega)\hat{G}^{1}_{j,k}(\Omega)\Bigg[-\Delta_2(n,\Omega)\hat{H}^{o,2}(n,m,j,k;\Omega) + \Delta_3(n,\Omega)\hat{H}^{o,6}(n,m,j,k;\Omega)$$

$$-\Delta_4(n,\Omega)\hat{H}^{o,8}(n,m,j,k;\Omega)\Big]$$

$$+\hat{G}^1_{n,m}(\Omega)\hat{G}^1_{j,k}(\Omega)\Big[\Delta_3(n,\Omega)\hat{H}^3(n,m,j,k;\Omega)-\Delta_4(n,\Omega)\hat{H}^9(n,m,j,k;\Omega)\Big]$$

$$+\hat{G}^1_{j,k}(\Omega)\Delta_3(n,\Omega)\hat{H}^4(n,m,j,k;\Omega)\Big\}. \tag{3.48}$$

It is convenient to define a new transfer function $\hat{G}_\xi(n,m;\Omega)$ so that

$$\xi_{1,0}(\Omega)=\frac{1}{\Delta_0}\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j}\hat{G}_\xi(n,m;\Omega)\hat{y}_{n,m}(\Omega)\hat{y}_{j,k}(\Omega). \tag{3.49}$$

The mean-square displacement due to the deformation $\xi_{1,0}(\Omega)$ is

$$<\xi_{1,0}\xi_{1,0}>=\lim_{t\to\infty}\frac{1}{(2\pi i)^2}\int_{\gamma-i\infty}^{\gamma+i\infty}\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{1}{\Delta_0}\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j}$$

$$\hat{G}_\xi(n,m;is_1)\hat{G}_\xi(n,m;is_2)<\hat{y}_{n,m}(is_1)\hat{y}_{j,k}(is_1)\hat{y}_{n,m}(is_1)\hat{y}_{j,k}(is_2)>e^{(s_1+s_2)t}ds_1ds_2. \tag{3.50}$$

(Note that since the expectation of odd powers of $\hat{y}$ is zero, because $\hat{y}$ is assumed Gaussian, there is no $O(\epsilon)^3$ contribution to the mean-square displacement consisting of the expectation of products of $O(\epsilon)$ flow variables with $O(\epsilon)^2$ flow variables.) This expression includes the contributions from harmonics of all degrees. As pointed out earlier, it is not really necessary to include contributions from all the higher-degree modes, since the probability weighting of a mode $\eta_{n,m}$ decreases significantly as n and m increase. Thus, to obtain a good approximation, the summation could be truncated to include only the contributions from the first- and second-order harmonics.

It can be seen from this expression that this first correction to the mean-square displacement will include surface-tension dependence, since it includes autocorrelations of $y_{n,m}$ for n greater than 1, and these were shown earlier (in the equilibrium calculation, as well as in the transfer function) to depend on surface tension. Since it is assumed that the random stress y can be modelled by Gaussian white noise, the autocorrelations of a product like

$$< \hat{y}_{n,m}(is_1)\hat{y}_{j,k}(is_1)\hat{y}_{n,m}(is_1)\hat{y}_{j,k}(is_2) > \tag{3.51}$$

can be obtained by adding all possible pairwise correlations:

$$\begin{aligned}
&< \hat{y}_{n,m}(is_1)\hat{y}_{j,k}(is_1) >< \hat{y}_{n,m}(is_1)\hat{y}_{j,k}(is_2) > + \\
&< \hat{y}_{n,m}(is_1)\hat{y}_{n,m}(is_1) >< \hat{y}_{j,k}(is_1)\hat{y}_{j,k}(is_2) > + \\
&< \hat{y}_{n,m}(is_1)\hat{y}_{j,k}(is_2) >< \hat{y}_{n,m}(is_1)\hat{y}_{j,k}(is_1) > .
\end{aligned} \tag{3.52}$$

Since, just as in the $O(\epsilon)$ calculation, the determinant Δ_0 appears in the numerator, evidently the poles of the transfer function $\hat{G}_\xi(n, m; is)$ will be the same as those for the transfer function $\hat{G}$ of the $O(\epsilon)$ calculation in Section 2.

DISCUSSION

The above analysis has given general expressions for a number of statistical quantities related to the Brownian motion of the drop. Expressions for the mean-square displacement and deformations have been found for small times, when the drop has not moved or deformed much relative to its initial position and shape, but there have nevertheless been enough collisions to make averaging meaningful. Fluctuation-dissipation theorems for the spherical harmonic coefficients describing the drop motion have been given.

It is also interesting to consider what has been learned in a more qualitative sense. Before doing any calculations, a number of arguments could have been put forth as to how the deformability of the drop might affect its mean-square displacement. The molecules of the surrounding fluid impinge upon the drop and thereby transfer energy to it; it might be reasoned that some of this Brownian energy causes center-of-mass translations, whereas the rest of it gets "consumed" in deforming the drop in a centrally symmetric way. This would suggest that the diffusivity of a drop would decrease as surface tension decreased, all other things being equal.

Another viewpoint involves a consideration of the effect of mobility. The asymptotic analysis of Taylor and Acrivos [9] for the steady sedimentation of a slightly deformed near-spherical drop gives a positive first correction in Weber number for the drag force on the drop; given that the diffusivity of a solid particle is the ratio of kT (Brownian energy) over the *steady* drag, it seems reasonable to conjecture that the diffusivity of a drop will also depend on the steady drag and will therefore be less for a deformable drop, since the drag force on it will be greater.

On the other hand, statistical mechanical theory predicts that a certain amount of thermal energy will be associated with each degree of freedom, regardless of how many degrees of freedom there are. Each translational degree of freedom gets energy $kT/2$, whether there be one, two or three; in other words, the viewpoint that there is a fixed amount of thermal energy which must somehow be distributed among all the modes of motion seems not to be appropriate. In light of this, it might be reasoned that the addition of deformational modes simply gives more ways in which energy can be transmitted from the surrounding fluid molecules to the drop, and that therefore there will be more motion, i.e., higher diffusivity. This argument can be summed up as: "more modes means more energy means more motion."

Examination of the quantitative results obtained in the previous sections indicates that this last line of reasoning is the most appropriate. If the surface tension becomes infinite, the quantity $\tilde{Q}$ also becomes infinite, from which it can be seen from Equation (2.53) for the transfer functions that $\hat{G}_n^1 \to 0$ unless $n = 1$. This, of course, just reflects the obvious fact that a drop with infinite surface tension will not have the higher-order harmonic components of the flow field associated with deformation, and therefore will not have any $O(\epsilon)$ correction to its mean-square displacement. Changing the surface tension does not affect the $O(\epsilon)$ mean-square displacement at all (this can be seen from the fact that $\tilde{Q} = 0$ for $n = 1$). Thus, all other things being equal, a drop with finite surface tension compared to a drop with infinite surface tension will have a larger mean-square displacement. (The $O(\epsilon)$ correction to the mean-square displacement can, of course, take on only nonnegative values.)

It is not unreasonable that the change in mobility due to deformation should not be the factor determining the trend in the surface-tension dependence of mean-square displacement, since it has been shown by Happel and Brenner [10] that the average mobility of a slightly deformed sphere with deformation of $O(\epsilon)$ is, at least to $O(\epsilon)$, exactly the same as the mobility of the sphere with the same volume as the deformed particle. Since the net random force on the drop should take on all directions with equal probability, it should be the *average* mobility that affects the mean-square displacement, and this average mobility is at least approximately independent of surface tension.

The above comments have been with regard to the mean-square displacement, rather than the diffusivity, because if the classical definition of diffusivity is used, namely, that it is one-half the *infinite* time limit of the rate of growth of the mean-square displacement, the $O(\epsilon)$ correction will be zero in all cases, because the transfer functions go to zero as $\Omega \to 0$. However, it is important to note that this infinite-time limit may be "too long," first in the sense that the particle displacement may become too large for the small-displacement analysis used in deriving the transfer functions to be still valid, and moreover, that the particle may move significantly due to the relaxation of its deformations (represented by the $O(\epsilon)$ correction) before the surface-tension independent diffusivity "wins out", as it evidently will, eventually.

Appendix 1 of Chapter 3

In this appendix, the linear system of equations

$$\begin{pmatrix} \tilde{A} & 0 & \tilde{B} & 0 & \frac{-i\Omega}{\beta} \\ 0 & \tilde{C} & 0 & \tilde{D} & \frac{-i\Omega}{\beta} \\ \tilde{F} & -\tilde{H} & \tilde{E} & -\tilde{G} & 0 \\ \tilde{I} & -\tilde{K} & \tilde{J} & -\tilde{L} & 0 \\ \tilde{M} & \tilde{O} & \tilde{N} & \tilde{P} & -\tilde{Q} \end{pmatrix} \begin{pmatrix} p_n^i \\ p_{-(n+1)}^o \\ \varphi_n^i \\ \varphi_n^o \\ \hat{\eta}_n \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ \hat{y}_n \end{pmatrix} \qquad (A1.1)$$

is solved to give

$$p_n^i = \hat{G}^{i,2}(\Omega)\hat{y}_n; \qquad (A1.2a)$$

$$\varphi_n^i = \hat{G}^{i,3}(\Omega)\hat{y}_n; \qquad (A1.2b)$$

$$p_{-(n+1)}^o = \hat{G}^{o,2}(\Omega)\hat{y}_n; \qquad (A1.2c)$$

$$\varphi_n^o = \hat{G}^{o,3}(\Omega)\hat{y}_n, \qquad (A1.2d)$$

where the transfer functions $\hat{G}$ are given by

$$\hat{G}^{i,2}(\Omega) = \hat{y}_n \begin{vmatrix} 0 & \tilde{B} & 0 & -\frac{i\Omega}{\beta} \\ \tilde{C} & 0 & \tilde{D} & -\frac{i\Omega}{\beta} \\ -\tilde{H} & \tilde{E} & -\tilde{G} & 0 \\ -\tilde{K} & \tilde{J} & -\tilde{L} & 0 \end{vmatrix}; \qquad (A1.3a)$$

$$\hat{G}^{i,3}(\Omega) = -\hat{y}_n \begin{vmatrix} \tilde{A} & \tilde{B} & 0 & -\frac{i\Omega}{\beta} \\ 0 & 0 & \tilde{D} & -\frac{i\Omega}{\beta} \\ \tilde{F} & \tilde{E} & -\tilde{G} & 0 \\ \tilde{I} & \tilde{J} & -\tilde{L} & 0 \end{vmatrix}; \qquad (A1.3b)$$

$$\hat{G}^{o,2}(\Omega) = \hat{y}_n \begin{vmatrix} \tilde{A} & 0 & 0 & -\frac{i\Omega}{\beta} \\ 0 & \tilde{C} & \tilde{D} & -\frac{i\Omega}{\beta} \\ \tilde{F} & -\tilde{H} & -\tilde{G} & 0 \\ \tilde{I} & -\tilde{K} & -\tilde{L} & 0 \end{vmatrix}; \qquad (A1.3c)$$

$$\hat{G}^{o,3}(\Omega) = -\hat{y}_n \begin{vmatrix} \tilde{A} & 0 & \tilde{B} & -\frac{i\Omega}{\beta} \\ 0 & \tilde{C} & 0 & -\frac{i\Omega}{\beta} \\ \tilde{F} & -\tilde{H} & \tilde{E} & 0 \\ \tilde{I} & -\tilde{K} & \tilde{J} & 0 \end{vmatrix} . \qquad (A1.3d)$$

Appendix 2 of Chapter III

In this appendix, the $P_1^0(\cos\theta) = \cos\theta$ component of the surface harmonic expansions of the following quantities will be determined: V^i, V^o, $-r\nabla\cdot\mathbf{W}$, $\mathbf{r}\cdot\nabla\times\mathbf{W}$, $-r\nabla\cdot\mathbf{T}$, $\mathbf{r}\cdot\nabla\times\mathbf{T}$, and $\mathbf{e}_r\cdot\mathbf{T}$, where

$$V^i(\theta,\phi) = \sum_{n=1}^{\infty} V_n^i = -\mathbf{e}_r\cdot\eta\frac{\partial\mathbf{u}^i}{\partial r} - \mathbf{n}'\cdot\mathbf{u}^i \Bigg|_{r=1}; \qquad (A2.1)$$

$$V^o(\theta,\phi) = \sum_{n=1}^{\infty} V_n^o = -\mathbf{e}_r\cdot\eta\frac{\partial\mathbf{u}^o}{\partial r} - \mathbf{n}'\cdot\mathbf{u}^o \Bigg|_{r=1}; \qquad (A2.2)$$

$$\mathbf{W}(\theta,\phi) = \eta\frac{\partial\mathbf{u}^o}{\partial r} - \eta\frac{\partial\mathbf{u}^i}{\partial r} \Bigg|_{r=1}; \qquad (A2.3)$$

$$\begin{aligned}\mathbf{T}(\theta,\phi) = \mathbf{n}'\left\{y + \Gamma\left[-\sum_{n=1}^{\infty}[2-n(n+1)]\eta_n\right]\right\} \\ + \mathbf{e}_r\cdot\eta\frac{\partial\sigma^i}{\partial r} - \mathbf{e}_r\cdot\eta\frac{\partial\sigma^o}{\partial r} + \mathbf{e}_r\Gamma\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}2\eta_n([2-n(n+1)]-1)\eta_m \\ - \mathbf{n}'\cdot\left[-\sigma^i+\sigma^o\right] \Bigg|_{r=1}. \qquad (A2.4)\end{aligned}$$

(Throughout this appendix, all quantities are to be evaluated at $r = 1$, even if this is not explicitly indicated, unless they appear inside an r-derivative, in which case they are first to be differentiated and *then* evaluated at $r = 1$.) These expressions all involve *products* of spherical harmonics.

Expansion of V^i and V^o

Recall that

$$\hat{u}_r^i = \sum_{n=1}^{\infty}\left\{\frac{1}{i\alpha\Omega}\frac{\partial p_n^i}{\partial r} + \left[(n+1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\frac{i\Omega\alpha}{\lambda}r^2\right]\frac{\partial \varphi_n^i}{\partial r}\right.$$
$$\left. + \; n(2n+1)\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\frac{i\Omega\alpha}{\lambda}\varphi_n^i r\right\} \qquad (A2.5)$$

and that $\chi_n^i = 0$. From this it follows that

$$\mathbf{e}_r \cdot \frac{\partial \mathbf{u}^i}{\partial r}\bigg|_{r=1} = \frac{\partial u_r^i}{\partial r} = \sum_{n=1}^{\infty}\left\{\frac{n(n-1)}{i\Omega\alpha}p_n^i\right.$$
$$+\left[n(n+1)(n-1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n(n+1)\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\psi_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\right.$$
$$\left.\left.+n(n+1)^2\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n(n+1)\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)^2\psi_{n+2}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\right]\varphi_n^i\right\}. \qquad (A2.6)$$

Next, note that

$$-\mathbf{n}' \cdot \mathbf{u}^i = \frac{\partial \eta}{\partial \theta}u_\theta^i + \frac{1}{\sin\theta}\frac{\partial \eta}{\partial \phi}u_\phi^i. \qquad (A2.7)$$

By recalling that

$$\hat{u}_\theta^i = \sum_{n=1}^{\infty}\left\{\frac{1}{i\alpha\Omega r}\frac{\partial p_n^i}{\partial \theta}\right.$$
$$\left. + \left[(n+1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\frac{i\Omega\alpha}{\lambda}r^2\right]\frac{1}{r}\frac{\partial \varphi_n^i}{\partial \theta}\right\} \qquad (A2.8)$$

$$\hat{u}_\phi^i = \sum_{n=1}^{\infty}\left\{\frac{1}{i\alpha\Omega r sin\theta}\frac{\partial p_n^i}{\partial \phi}\right.$$

$$+ \left[(n+1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\frac{i\Omega\alpha}{\lambda}r^2\right]\frac{1}{rsin\theta}\frac{\partial\varphi_n^i}{\partial\theta}\Bigg\} \qquad (A2.9)$$

(where again the fact that $\chi_n^i = 0$ has been used), it follows that

$$V^i(\theta,\phi) = \sum_{n=1}^{\infty} V_n^i = -\mathbf{e}_r \cdot \eta\frac{\partial \mathbf{u}^i}{\partial r} - \mathbf{n}' \cdot \mathbf{u}^i \Bigg|_{r=1};$$

$$= \left[\sum_{j=1}^{\infty}\eta_j\right] \cdot \sum_{n=1}^{\infty}\Bigg\{\frac{n(n-1)}{i\Omega\alpha}p_n^i$$

$$+\Bigg[n(n+1)(n-1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n(n+1)\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\psi_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)$$

$$+n(n+1)^2\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n(n+1)\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)^2\psi_{n+2}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\Bigg]\varphi_n^i\Bigg\}$$

$$+\left[\sum_{j=1}^{\infty}\frac{\partial\eta_j}{\partial\theta}\right]\sum_{n=1}^{\infty}\Bigg\{\frac{1}{i\alpha\Omega r}\frac{\partial p_n^i}{\partial\theta}$$

$$\left[(n+1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\frac{i\Omega\alpha}{\lambda}r^2\right]\frac{1}{r}\frac{\partial\varphi_n^i}{\partial\theta}\Bigg\}$$

$$+\frac{1}{\sin\theta}\left[\sum_{j=1}^{\infty}\frac{\partial\eta_j}{\partial\phi}\right]\sum_{n=1}^{\infty}\Bigg\{\frac{1}{i\alpha\Omega rsin\theta}\frac{\partial p_n^i}{\partial\phi}$$

$$\left[(n+1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\frac{i\Omega\alpha}{\lambda}r^2\right]\frac{1}{rsin\theta}\frac{\partial\varphi_n^i}{\partial\theta}\Bigg\}. \qquad (A2.10)$$

Consider the various products appearing in this expression:

$$\eta_j p_n^i, \quad \eta_n\varphi_n^i, \quad \frac{\partial\eta_j}{\partial\theta}\frac{\partial p_n^i}{\partial\theta}, \quad \frac{\partial\eta_j}{\partial\theta}\frac{\partial\varphi_n^i}{\partial\theta}, \quad \frac{1}{\sin^2\theta}\frac{\partial\eta_j}{\partial\phi}\frac{\partial p_n^i}{\partial\phi}, \quad \frac{1}{\sin^2\theta}\frac{\partial\eta_j}{\partial\phi}\frac{\partial\varphi_n^i}{\partial\phi}.$$

Only the order 1, degree 0 (i.e., $P_1^0 = \cos\theta$) terms in their surface harmonic expansions are desired here. These are given by:

$$\mathcal{P}\eta_j p_n^i = \sum_{k=-j}^{j} \sum_{m=-n}^{n} \eta_{j,k} p_{n,m}^i I_1(n,m,j,k)$$

$$= \sum_{k=-j}^{j} \sum_{m=-n}^{n} \eta_{j,k} p_{n,m}^i [\delta_{m,-k} \zeta_{n,m}^a \delta_{j,n+1} + \delta_{m,-k} \zeta_{n,m}^b \delta_{j,n-1}], \qquad (A2.11)$$

$$\mathcal{P} \frac{\partial \eta_j}{\partial \theta} \frac{\partial p_n^i}{\partial \theta} = \sum_{k=-j}^{j} \sum_{m=-n}^{n} \eta_{j,k} p_{n,m}^i I_2(n,m,j,k), \qquad (A2.12)$$

$$\mathcal{P} \frac{1}{\sin^2\theta} \frac{\partial \eta_j}{\partial \phi} \frac{\partial p_n^i}{\partial \phi} = \sum_{k=-j}^{j} \sum_{m=-n}^{n} \eta_{j,k} p_{n,m}^i I_3(n,m,j,k), \qquad (A2.13)$$

where

$$I_1(n,m,j,k) = \int_0^{\pi} \int_0^{2\pi} P_j^k(\cos\theta) e^{ik\phi} P_n^m(\cos\theta) e^{im\phi} \cos\theta \sin\theta d\phi d\theta; \qquad (A2.14)$$

$$I_2(n,m,j,k) = \int_0^{\pi} \int_0^{2\pi} \frac{\partial P_j^k(\cos\theta)}{\partial\theta} e^{ik\phi} \frac{\partial P_n^m(\cos\theta)}{\partial\theta} e^{im\phi} \cos\theta \sin\theta d\phi d\theta; \qquad (A2.15)$$

$$I_3(n,m,j,k) = \int_0^{\pi} \int_0^{2\pi} \frac{1}{\sin^2\theta} P_j^k(\cos\theta)(ik) e^{ik\phi} P_n^m(\cos\theta)(im) e^{im\phi} \cos\theta \sin\theta d\phi d\theta, \qquad (A2.16)$$

and where, for convenience, $\mathcal{P}$ denotes the operator that projects a function of θ, ϕ onto its P_1^0 component. By taking advantage of the following properties of associated Legendre polynomials:

$$\cos\theta P_n^m(\cos\theta) = \frac{(n-m+1)}{(2n+1)} P_{n+1}^m(\cos\theta) + \frac{(n+m)}{(2n+1)} P_{n-1}^m(\cos\theta); \qquad (A2.17)$$

$$\int_0^\pi P_n^m(\cos\theta) P_j^m(\cos\theta)\sin\theta d\theta = \delta_{n,j}\frac{2}{2n+1}\frac{(n+m)!}{(n-m)!}; \qquad (A2.18)$$

it can be seen that

$$I_1(n,m,j,k) = \int_0^{2\pi}\int_0^\pi P_n^{|m|}(\cos\theta)e^{im\phi} P_j^{|k|}(\cos\theta)e^{ik\phi}\cos\theta\sin\theta d\theta$$

$$= 2\pi\delta_{m,-k}\left[\frac{2(n-m+1)}{(2n+1)(2n+3)}\frac{(n+m+1)!}{(n-m+1)!}\delta_{j,n+1}\right.$$

$$\left. + \frac{2(n+m)}{(2n+1)(2n-1)}\frac{(n+m-1)!}{(n-m-1)!}\delta_{j,n-1}\right.$$

$$= \delta_{m,-k}\zeta_{n,m}^a\delta_{j,n+1} + \delta_{m,-k}\zeta_{n,m}^b\delta_{j,n-1}. \qquad (A2.19)$$

Similar general expressions can be derived for I_2 and I_3. Thus,

$$V_{1,0}^i(\theta,\phi) = \sum_{n=1}^\infty\sum_{j=1}^\infty\sum_{m=-n}^n\sum_{k=-j}^j\left\{\eta_{j,k}I_1(n,m,j,k)\left[\frac{n(n-1)}{i\Omega\alpha}p_{n,m}^i\right.\right.$$

$$+\left(n(n+1)(n-1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n(n+1)\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\psi_n\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\right.$$

$$\left.\left. +n(n+1)^2\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n(n+1)\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)^2\psi_{n+2}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\right)\varphi_{n,m}^i\right]$$

$$+\eta_{j,k}I_2(n,m,j,k)\left[\frac{1}{i\alpha\Omega r}p_{n,m}^i\right.$$

$$\left.+\left[(n+1)\psi_{n-1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right) - n\psi_{n+1}\left(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\right)\frac{i\Omega\alpha}{\lambda}r^2\right]\frac{1}{r}\varphi_{n,m}^i\right]$$

$$+\eta_{j,k}I_3(n,m,j,k)\Bigg[\frac{1}{i\alpha\Omega r}p^i_{n,m}$$

$$+\Big[(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)-n\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}r\Big)\frac{i\Omega\alpha}{\lambda}r^2\Big]\frac{1}{r}\varphi^i_{n,m}\Bigg]\Bigg\}. \qquad (A2.20)$$

By a similar procedure, it can be shown that

$$V^o_{1,0}(\theta,\phi)=\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j}\Bigg\{\eta_{j,k}I_1(n,m,j,k)\Bigg[\frac{(n+1)(n+2)}{i\Omega\alpha}p^o_{-(n+1),m}$$

$$+\Bigg(n(n+1)(n-1)f_{n-1}(\sqrt{i\Omega})-n(n+1)(\sqrt{i\Omega})f_n(\sqrt{i\Omega})$$

$$+n(n+1)^2(\sqrt{i\Omega})f_{n+1}(\sqrt{i\Omega})-n(n+1)(\sqrt{i\Omega})^2f_{n+2}(\sqrt{i\Omega})\Bigg)\varphi^o_{n,m}\Bigg]$$

$$+\eta_{j,k}I_2(n,m,j,k)\Bigg[\frac{1}{i\alpha\Omega}p^o_{-(n+1),m}$$

$$+\Big[(n+1)f_{n-1}(\sqrt{i\Omega})-nf_{n+1}(\sqrt{i\Omega})i\Omega\Big]\varphi^o_{n,m}\Bigg]$$

$$+\eta_{j,k}I_3(n,m,j,k)\Bigg[\frac{1}{i\alpha\Omega}p^o_{-(n+1),m}$$

$$+\Big[(n+1)f_{n-1}(\sqrt{i\Omega})-nf_{n+1}(\sqrt{i\Omega})(i\Omega)\Big]\varphi^o_{-(n+1),m}\Bigg]\Bigg\}. \qquad (A2.21)$$

As discussed earlier, what will be determined here is only the $O(\epsilon^2)$ contribution to the diffusivity due to interaction of the $n=1$ and $n=2$ modes of the $O(\epsilon)$ solution. Therefore, the above (complete) sums giving $V^i_{1,0}$ and $V^o_{1,0}$ will be truncated so that n and j take on only the values 1 and 2. It is convenient to rewrite these truncated expressions as

$$V^i_{1,0}(\theta,\phi) =$$
$$\sum_{n=1}^{2}\sum_{j=1}^{2}\sum_{m=-n}^{n}\sum_{k=-j}^{j} \hat{H}^{i,1}(n,m,j,k;\Omega)\eta_{j,k}p^i_{n,m} + \hat{H}^{i,2}(n,m,j,k;\Omega)\eta_{j,k}\varphi^i_{n,m}; \quad (A2.22)$$

$$V^o_{1,0}(\theta,\phi) =$$
$$\sum_{n=1}^{2}\sum_{j=1}^{2}\sum_{m=-n}^{n}\sum_{k=-j}^{j} \hat{H}^{o,1}(n,m,j,k;\Omega)\eta_{j,k}p^o_{n,m} + \hat{H}^{o,2}(n,m,j,k;\Omega)\eta_{j,k}\varphi^o_{n,m}, \quad (A2.23)$$

where the transfer functions $\hat{H}$ are given by

$$\hat{H}^{i,1}(n,m,j,k;\Omega) = \frac{1}{i\alpha\Omega}\Big[n(n-1)I_1(n,m,j,k) + I_2(n,m,j,k) + I_3(n,m,j,k)\Big]; \quad (A2.24)$$

$$\hat{H}^{i,2}(n,m,j,k;\Omega) =$$
$$I_1(n,m,j,k)\Bigg(n(n+1)(n-1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big) - n(n+1)\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)\psi_n\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)$$
$$+n(n+1)^2\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big) - n(n+1)\Big(\frac{i\Omega\alpha}{\lambda}\Big)\psi_{n+2}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)\Bigg)$$
$$+I_2(n,m,j,k)\Big[(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big) - n\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)\frac{i\Omega\alpha}{\lambda}\Big]$$
$$+I_3(n,m,j,k)\Big[(n+1)\psi_{n-1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big) - n\psi_{n+1}\Big(\sqrt{\frac{i\Omega\alpha}{\lambda}}\Big)\frac{i\Omega\alpha}{\lambda}\Big]; \quad (A2.25)$$

$$\hat{H}^{o,1}(n,m,j,k;\Omega) = I_1(n,m,j,k)\frac{(n+1)(n+2)}{i\Omega\alpha}p^o_{-(n+1),m}$$
$$+I_2(n,m,j,k)\frac{1}{i\alpha\Omega} + I_3(n,m,j,k)\frac{1}{i\alpha\Omega}; \quad (A2.26)$$

$$\hat{H}^{o,2}(n,m,j,k;\Omega) =$$

$$I_1(n,m,j,k)\Bigg(n(n+1)(n-1)f_{n-1}(\sqrt{i\Omega}) - n(n+1)(\sqrt{i\Omega})f_n(\sqrt{i\Omega})$$

$$+n(n+1)^2(\sqrt{i\Omega})f_{n+1}(\sqrt{i\Omega}) - n(n+1)(i\Omega)f_{n+2}(\sqrt{i\Omega})\Bigg)$$

$$+I_2(n,m,j,k)\Big[(n+1)f_{n-1}(\sqrt{i\Omega}) - nf_{n+1}(\sqrt{i\Omega})i\Omega\Big]$$

$$+I_3(n,m,j,k)\Big[(n+1)f_{n-1}(\sqrt{i\Omega}) - nf_{n+1}(\sqrt{i\Omega})(i\Omega)\Big]. \tag{A2.27}$$

Expansion of $-r\nabla \cdot \mathbf{W}$

Recall that

$$\mathbf{W}(\theta,\phi) = \eta\frac{\partial \mathbf{u}^o}{\partial r} - \eta\frac{\partial \mathbf{u}^i}{\partial r}\Bigg|_{r=1}. \tag{A2.28}$$

After some manipulations, it can be shown that

$$\nabla \cdot \Big(\frac{\partial \mathbf{u}^i}{\partial r}\Big) = \frac{\partial}{\partial r}(\nabla \cdot \mathbf{u}^i) + \frac{1}{r}\nabla \cdot \mathbf{u}^i + \frac{1}{r}\frac{\partial u_r^i}{\partial r}, \tag{A2.29}$$

and similarly for $\mathbf{u}^o$. Since by continuity $\nabla \cdot \mathbf{u}^i = \nabla \cdot \mathbf{u}^o = 0$,

$$-\nabla \cdot \mathbf{W} = -\eta\Big(\frac{1}{r}\frac{\partial u_r^o}{\partial r} - \frac{1}{r}\frac{\partial u_r^i}{\partial r}\Big) + \nabla\eta \cdot \Big(\frac{\partial \mathbf{u}^o}{\partial r} - \frac{\partial \mathbf{u}^i}{\partial r}\Big). \tag{A2.30}$$

The $O(\epsilon)$ tangential velocity boundary condition was that

$$\frac{\partial u_r^i}{\partial r}\Bigg|_{r=1} = \frac{\partial u_r^o}{\partial r}\Bigg|_{r=1}; \tag{A2.31}$$

this can be used to simplify to

$$-\nabla \cdot \mathbf{W} = +\nabla\eta \cdot \left(\frac{\partial \mathbf{u}^o}{\partial r} - \frac{\partial \mathbf{u}^i}{\partial r} \right) \bigg|_{r=1} . \qquad (A2.32)$$

Since the unit vectors in spherical coordinates do not depend on r, this can be rewritten as

$$-\nabla \cdot \mathbf{W} = +\frac{\partial \tilde{\eta}}{\partial \theta} \left[\frac{\partial u^o_\theta}{\partial r} - \frac{\partial u^i_\theta}{\partial r} \right] + \frac{1}{\sin\theta} \frac{\partial \tilde{\eta}}{\partial \theta} \left[\frac{\partial u^o_\phi}{\partial r} - \frac{\partial u^i_\phi}{\partial r} \right]. \qquad (A2.33)$$

Since the condition of continuity of tangential stress at $r = 1$ was imposed on the $O(\epsilon)$ solution,

$$\sigma^i_{r\theta} \Big|_{r=1} = \sigma^o_{r\theta} \Big|_{r=1}, \qquad (A2.34)$$

which is equivalent to

$$\frac{\partial u^i_\theta}{\partial r} - \frac{u^i_\theta}{r} + \frac{1}{r} \frac{\partial u^i_r}{\partial \theta} \bigg|_{r=1} = \frac{\partial u^o_\theta}{\partial r} - \frac{u^o_\theta}{r} + \frac{1}{r} \frac{\partial u^o_r}{\partial \theta} \bigg|_{r=1} . \qquad (A2.35)$$

By the continuity of the $O(\epsilon)$ velocity field on the sphere surface $r = 1$,

$$u^i_\theta \Big|_{r=1} = u^o_\theta \Big|_{r=1}, \qquad (A2.36)$$

which implies

$$\frac{\partial u^i_r}{\partial \theta} \bigg|_{r=1} = \frac{\partial u^o_r}{\partial \theta} \bigg|_{r=1} . \qquad (A2.37)$$

From all this it can be deduced that

$$\frac{\partial u^i_\theta}{\partial r} = \frac{\partial u^o_\theta}{\partial r}, \qquad (A2.38)$$

and similarly, using the continuity of stress in the ϕ direction ($\sigma_{r\phi}$), that

$$\frac{\partial u^i_\phi}{\partial r} = \frac{\partial u^o_\phi}{\partial r}, \qquad (A2.39)$$

so a further simplification can be made to give

$$-\nabla \cdot \mathbf{W} = 0. \qquad (A2.40)$$

Expansion of $-r\nabla \cdot \mathbf{T}$

$$\begin{aligned} -r\nabla \cdot \mathbf{T}|_{r=1} = -\nabla \cdot \Bigg\{ \mathbf{n}' \Bigg[y + \Gamma\Bigg(-\sum_{n=1}^{\infty} [2 - n(n+1)]\eta_n \Bigg) \Bigg] \\ + \mathbf{e}_r \cdot \eta \frac{\partial \sigma^i}{\partial r} - \mathbf{e}_r \cdot \eta \frac{\partial \sigma^o}{\partial r} + \mathbf{e}_r \Gamma \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} 2\eta_n([2 - n(n+1)] - 1)\eta_m \\ - \mathbf{n}' \cdot \Big[-\sigma^i + \sigma^o \Big] \Bigg\} \Bigg|_{r=1} \end{aligned} \qquad (A2.41)$$

On account of the length of this expression, it is convenient to rewrite it as the sum of several separate terms:

$$-r\nabla \cdot \mathbf{T}|_{r=1} = \tau_1 + \tau_2 + \tau_3 + \tau_4 + \tau_5, \qquad (A2.42)$$

where

$$\tau_1 = -\nabla \cdot \Bigg[\mathbf{n}' \Big[y + \Gamma\Big(-\sum_{n=1}^{\infty} [2 - n(n+1)]\eta_n \Big) \Big] \Bigg]; \qquad (A2.43a)$$

$$\tau_2 = -\nabla \cdot \Big[\mathbf{e}_r \cdot \eta \frac{\partial \sigma^i}{\partial r} \Big]; \qquad (A2.43b)$$

$$\tau_3 = -\nabla \cdot \Big[-\mathbf{e}_r \cdot \eta \frac{\partial \sigma^o}{\partial r} \Big]; \qquad (A2.43c)$$

$$\tau_4 = -\nabla \cdot \Big[\mathbf{e}_r \Gamma \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} 2\eta_n [1 - n(n+1)]\eta_m \Big] \qquad (A2.43d)$$

$$\tau_5 = -\nabla \cdot \left[\mathbf{n}' \cdot \sigma^i\right] + \nabla \cdot \left[\mathbf{n}' \cdot \sigma^o\right]. \qquad (A2.43e)$$

Consider first τ_1:

$$\tau_1 = -\left[y + \Gamma\left(-\sum_{n=1}^{\infty}[2 - n(n+1)]\eta_n\right)\right]\nabla \cdot \mathbf{n}'$$

$$-\nabla\left[y + \Gamma\left(-\sum_{n=1}^{\infty}[2 - n(n+1)]\eta_n\right)\right] \cdot \mathbf{n}'. \qquad (A2.44)$$

The first correction to the surface unit normal vector, $\mathbf{n}'$, was given above as $\mathbf{n}' = \frac{\partial \eta}{\partial \theta}\mathbf{e}_\theta + \frac{1}{\sin\theta}\frac{\partial \eta}{\partial \phi}\mathbf{e}_\phi$, so that

$$\nabla \cdot \mathbf{n}'|_{r=1} = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\eta}{\partial\theta}\right) + \frac{1}{\sin\theta}\frac{\partial}{\partial\phi}\left(\frac{1}{\sin\theta}\frac{\partial\eta}{\partial\phi}\right). \qquad (A2.45)$$

By using Legendre's equation again, this can be rewritten as

$$\nabla \cdot \mathbf{n}'|_{r=1} = -\sum_{j=1}^{\infty} j(j+1)\eta_j, \qquad (A2.46)$$

so that

$$\mathcal{P}\tau_1 = \sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j}\Bigg\{\left[y_{n,m} - \Gamma[2 - n(n+1)]\eta_{n,m}\right]j(j+1)\eta_{j,k}I_1(n,m,j,k)$$

$$-\left[y_{n,m} - \Gamma\left([2 - n(n+1)]\eta_{n,m}\right)\right]\eta_{j,k}I_2(n,m,j,k)$$

$$-\left[y_{n,m} - \Gamma\left([2 - n(n+1)]\eta_{n,m}\right)\right]\eta_{j,k}I_3(n,m,j,k)\Bigg\}, \qquad (A2.47)$$

where as before $\mathcal{P}$ denotes the operator that projects a function onto its $\cos\theta$ component. Next, expand τ_2:

$$\tau_2 = -\nabla \cdot \left[\eta \frac{\partial \mathbf{e}_r \cdot \sigma^i}{\partial r}\right] = -\eta \nabla \cdot \frac{\partial \mathbf{e}_r \cdot \sigma^i}{\partial r} - (\nabla \eta) \cdot \frac{\partial \mathbf{e}_r \cdot \sigma^i}{\partial r} . \qquad (A2.48)$$

Recall that

$$\mathbf{e}_r \cdot \sigma^i = \frac{1}{r} \sum_{n=1}^{\infty} \Bigg\{ \lambda \Big[\frac{2(n-1)}{(i\Omega)\alpha} \nabla p_n^i + R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r) \nabla \varphi_n^i - \frac{(2n+1)}{r^2} S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r) \varphi_n^i \mathbf{r} \Big] - p_n^i \mathbf{r} \Bigg\}, \qquad (A2.49)$$

from which it can be shown that

$$\frac{\partial(\mathbf{e}_r \cdot \sigma^i)}{\partial r} = \sum_{n=1}^{\infty} \Bigg\{ \lambda \Big[\frac{2(n-1)(n-2)}{(i\Omega)\alpha r^2} \nabla p_n^i + R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r) \frac{n-2}{r^2} \nabla \varphi_n^i + \frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r)}{dr} \frac{1}{r} \nabla \varphi_n^i - \frac{(2n+1)(n-2)}{r^4} S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r) \varphi_n^i \mathbf{r} - \frac{(2n+1)}{r^3} \frac{dS_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r)}{dr} \varphi_n^i \mathbf{r} \Big] - \frac{n}{r} p_n^i \mathbf{e}_r \Bigg\}, \qquad (A2.50))$$

which in turn gives

$$\nabla \cdot \frac{\partial(\mathbf{e}_r \cdot \sigma^i)}{\partial r}\Bigg|_{r=1} = \sum_{n=1}^{\infty} \Bigg\{ \lambda \Bigg[-\frac{4(n-1)(n-2)}{i\Omega\alpha} p_n^i + \left(-2n(n-2) R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}) + n(n-3) \frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r)}{dr} + n \frac{d^2 R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r)}{dr^2} \right) \varphi_n^i - \Bigg([n-(2n+1)(n-2)] S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r) + [n+(2n+1)(n-2)] \frac{dS_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}} r)}{dr}$$

$$+(2n+1)\frac{d^2 S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr^2}\Bigg)\varphi_n^i\Bigg] - n(n+1)p_n^i\Bigg\}. \qquad (A2.51)$$

Putting this together gives

$$\mathcal{P}_{\mathcal{T}_2} = -\sum_{j=1}^{\infty}\sum_{n=1}^{\infty}\sum_{k=-j}^{j}\sum_{m=-n}^{n}$$

$$\Bigg\{\eta_{j,k}p_{n,m}^i\Bigg[\Bigg(-\frac{4\lambda(n-1)(n-2)}{i\Omega\alpha} - n(n+1)\Bigg)I_1(n,m,j,k)$$

$$+\lambda\frac{2(n-1)(n-2)}{(i\Omega)\alpha r^2}I_2(n,m,j,k) + \lambda\frac{2(n-1)(n-2)}{(i\Omega)\alpha r^2}I_3(n,m,j,k)\Bigg]$$

$$+\eta_{j,k}\varphi_{n,m}^i\lambda\Bigg[\Bigg(-2n(n-2)R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}) + n(n-3)\frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr} + n\frac{d^2R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr^2}$$

$$-[n-(2n+1)(n-2)]S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r) + [n+(2n+1)(n-2)]\frac{dS_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr}$$

$$+(2n+1)\frac{d^2 S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr^2}\Bigg)I_1(n,m,j,k)$$

$$+\Bigg(R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n-2}{r^2} + \frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr}\Bigg)I_2(n,m,j,k)$$

$$+\Bigg(R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n-2}{r^2} + \frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr}\Bigg)I_3(n,m,j,k)\Bigg]\Bigg\}. \qquad (A2.52)$$

Similarly,

$$\mathcal{P}_{\mathcal{T}_3} = -\sum_{j=1}^{\infty}\sum_{n=1}^{\infty}\sum_{k=-j}^{j}\sum_{m=-n}^{n}$$

$$\Bigg\{\eta_{j,k}p_{-(n+1),m}^o\Bigg[\Bigg(-\frac{4(n+2)(n+3)}{i\Omega} - n(n+1)\Bigg)I_1(n,m,j,k)$$

$$+\frac{2(n+2)(n+3)}{i\Omega}I_2(n,m,j,k)+\frac{2(n+2)(n+3)}{i\Omega}I_3(n,m,j,k)\Bigg]$$

$$+\eta_{j,k}\varphi^o_{n,m}\Bigg[\Bigg(-2n(n-2)R^o_n(\sqrt{i\Omega})+n(n-3)\frac{dR^o_n(\sqrt{i\Omega}r)}{dr}+n\frac{d^2R^o_n(\sqrt{i\Omega}r)}{dr^2}$$

$$-[n-(2n+1)(n-2)]S^o_n(\sqrt{i\Omega})+[n+(2n+1)(n-2)]\frac{dS^o_n(\sqrt{i\Omega}r)}{dr}$$

$$+(2n+1)\frac{d^2S^o_n(\sqrt{i\Omega}r)}{dr^2}\Bigg)I_1(n,m,j,k)+\Bigg((n-2)R^o_n(\sqrt{i\Omega})+\frac{dR^o_n(\sqrt{i\Omega}r)}{dr}\Bigg)I_2(n,m,j,k)$$

$$+\Bigg((n-2)R^o_n(\sqrt{i\Omega})+\frac{dR^o_n(\sqrt{i\Omega}r)}{dr}\Bigg)I_3(n,m,j,k)\Bigg]\Bigg\}. \qquad (A2.53)$$

Next, expand τ_4:

$$\mathcal{P}\tau_4=-\mathcal{P}\nabla\cdot\left[\mathrm{e}_r\Gamma\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}2\eta_n[1-n(n+1)]\eta_j\right]$$

$$=-\mathcal{P}\frac{\partial}{\partial r}\left[r^2\Gamma\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}2\eta_n[1-n(n+1)]\eta_j\right]$$

$$=-\mathcal{P}\Gamma\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}2[1-n(n+1)](n+j+2)\eta_n\eta_j$$

$$=-\Gamma\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j}2[1-n(n+1)](n+j+2)\eta_{n,m}\eta_{j,k}I_1(n,m,j,k). \qquad (A2.54)$$

Next, expand τ_5:

$$\tau_5=-\nabla\cdot\left[\mathrm{n}'\cdot\sigma^i\right]+\nabla\cdot\left[\mathrm{n}'\cdot\sigma^o\right]$$

$$=\mathrm{n}'\cdot\left[\nabla\cdot(\sigma^o-\sigma^i)\right]+(\nabla\mathrm{n}'):(\sigma^o-\sigma^i)\ . \qquad (A2.55)$$

By the condition of continuity of stress that was imposed on the $O(\epsilon)$ solution, $\sigma^o-\sigma^i=0$ at $r=1$. Now by the equations of motion,

$$\nabla \cdot \sigma^o - \nabla \cdot \sigma^i = -\frac{i\Omega}{\beta}[\hat{\mathbf{u}}^o - \hat{\mathbf{u}}^i], \qquad (A2.56)$$

so that

$$\tau_5 = \mathbf{n}' \cdot \frac{i\Omega}{\beta}[\hat{\mathbf{u}}^i - \hat{\mathbf{u}}^o] \ . \qquad (A2.57)$$

By continuity of velocity at $r = 1$, which was imposed on the $O(\epsilon)$ solution, $[\hat{\mathbf{u}}^i - \hat{\mathbf{u}}^o] = 0$. Moreover, all θ or ϕ derivatives of this difference will also be zero at $r = 1$. Since $\mathbf{n}'$ has only components in the θ and ϕ directions, it follows that $\tau_5 = 0$. So, finally,

$$T^a_{1,0} = \sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j}\Big\{\hat{H}^3(n,m,j,k;\Omega)\eta_{j,k}\eta_{n,m} + \hat{H}^4(n,m,j,k;\Omega)\eta_{j,k}\hat{y}_{n,m}$$

$$+\hat{H}^{i,5}\eta_{j,k}p^i_{n,m} + \hat{H}^{i,6}\eta_{j,k}\varphi^i_{n,m}$$

$$+\hat{H}^{o,5}\eta_{j,k}p^o_{-(n+1),m} + \hat{H}^{o,6}\eta_{j,k}\varphi^o_{n,m}\Big\}, \qquad (A2.58)$$

where

$$\hat{H}^3(n,m,j,k;\Omega)$$

$$= -\Gamma[2 - n(n+1)][j(j+1)I_1(n,m,j,k) - I_2(n,m,j,k) - I_3(n,m,j,k)]$$

$$-2\Gamma[1 - n(n+1)](n+j+2)I_1(n,m,j,k); \qquad (A2.59a)$$

$$\hat{H}^4(n,m,j,k;\Omega) = j(j+1)I_1(n,m,j,k) - I_2(n,m,j,k) - I_3(n,m,j,k); \quad (A2.59b)$$

$$\hat{H}^{i,5} = \Bigg[\Bigg(-\frac{4\lambda(n-1)(n-2)}{i\Omega\alpha} - n(n+1)\Bigg) I_1(n,m,j,k)$$

$$+\lambda\frac{2(n-1)(n-2)}{(i\Omega)\alpha r^2} I_2(n,m,j,k) + \lambda\frac{2(n-1)(n-2)}{(i\Omega)\alpha r^2} I_3(n,m,j,k)\Bigg]; \qquad (A2.59c)$$

$$\hat{H}^{i,6} = \Bigg[\Bigg(-2n(n-2)R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}) + n(n-3)\frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr} + n\frac{d^2R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr^2}$$

$$-[n-(2n+1)(n-2)]S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r) + [n+(2n+1)(n-2)]\frac{dS_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr}$$

$$+(2n+1)\frac{d^2S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr^2}\Bigg) I_1(n,m,j,k)$$

$$+\Bigg(R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n-2}{r^2} + \frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr}\Bigg) I_2(n,m,j,k)$$

$$+\Bigg(R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n-2}{r^2} + \frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr}\Bigg) I_3(n,m,j,k)\Bigg] \qquad (A2.59d)$$

$$\hat{H}^{o,5} = \Bigg[\Bigg(-\frac{4(n+2)(n+3)}{i\Omega} - n(n+1)\Bigg) I_1(n,m,j,k)$$

$$+\frac{2(n+2)(n+3)}{i\Omega} I_2(n,m,j,k) + \frac{2(n+2)(n+3)}{i\Omega} I_3(n,m,j,k)\Bigg]; \qquad (A2.59e)$$

$$\hat{H}^{o,6} = \Bigg[\Bigg(-2n(n-2)R_n^o(\sqrt{i\Omega}) + n(n-3)\frac{dR_n^o(\sqrt{i\Omega}r)}{dr} + n\frac{d^2R_n^o(\sqrt{i\Omega}r)}{dr^2}$$

$$-[n-(2n+1)(n-2)]S_n^o(\sqrt{i\Omega}) + [n+(2n+1)(n-2)]\frac{dS_n^o(\sqrt{i\Omega}r)}{dr}$$

$$+(2n+1)\frac{d^2 S_n^o(\sqrt{i\Omega}r)}{dr^2}\Bigg) I_1(n,m,j,k) + \left((n-2)R_n^o(\sqrt{i\Omega}) + \frac{dR_n^o(\sqrt{i\Omega}r)}{dr}\right) I_2(n,m,j,k)$$

$$+\left((n-2)R_n^o(\sqrt{i\Omega}) + \frac{dR_n^o(\sqrt{i\Omega}r)}{dr}\right) I_3(n,m,j,k)\Bigg] \quad . \qquad (A2.59f)$$

Expansion of $\mathbf{e}_r \cdot \mathbf{T}$

$$\mathbf{e}_r \cdot \mathbf{T} = \eta[\mathbf{e}_r \cdot \mathbf{e}_r \cdot \frac{\partial \sigma^i}{\partial r} - \mathbf{e}_r \cdot \mathbf{e}_r \cdot \frac{\partial \sigma^o}{\partial r}] + \Gamma \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} 2\eta_n([2 - n(n+1)] - 1)\eta_m$$

$$-\mathbf{e}_r \cdot \mathbf{n}' \cdot \left[-\sigma^i + \sigma^o\right] \Bigg|_{r=1} \qquad (A2.60)$$

All components of the stress tensor σ are continuous across the boundary except the (r,r) component. Since $\mathbf{n}'$ has only θ and ϕ components,

$$\mathbf{n}' \cdot \left[-\sigma^i + \sigma^o\right] = 0. \qquad (A2.61)$$

With this, and the fact that the unit vector $\mathbf{e}_r$ does not depend on r, (A2.60) may be rewritten as

$$\mathbf{e}_r \cdot \mathbf{T} = \eta[\frac{\partial \mathbf{e}_r \cdot \mathbf{e}_r \cdot \sigma^i}{\partial r} - \frac{\partial \mathbf{e}_r \cdot \mathbf{e}_r \cdot \sigma^o}{\partial r}]$$

$$+\Gamma \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} 2\eta_n([2 - n(n+1)] - 1)\eta_m \Bigg|_{r=1} \quad , \qquad (A2.62)$$

which, using (A2.49) and the companion equation for the outside region, becomes

$$\mathbf{e}_r \cdot \mathbf{T}\Big|_{r=1} =$$

$$\eta \frac{\partial}{\partial r}\left\{\frac{1}{r}\sum_{n=1}^{\infty} -\left[\frac{2(n+1)(n+2)}{(i\Omega)r} p^o_{-(n+1)} + R_n^o(\sqrt{i\Omega}r)\frac{n\varphi_n^o}{r}\right.\right.$$

$$-\frac{(2n+1)}{r^2}S_n^o(\sqrt{i\Omega}r)\varphi_n^o r - p_{-(n+1)}^o r\Big]$$

$$+\Bigg[\lambda\Big[\frac{2(n-1)n}{(i\Omega)\alpha r}p_n^i + R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\frac{n\varphi_n^i}{r}$$

$$-\frac{(2n+1)}{r^2}S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)\varphi_n^i r\Big] - p_n^i r\Bigg]\Bigg\}$$

$$+\Gamma\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}2\eta_n([2-n(n+1)]-1)\eta_m \Bigg|_{r=1}, \qquad (A2.63)$$

so that

$$T_{1,0}^b = \mathcal{P}\mathbf{e}_r\cdot\mathbf{T}\Big|_{r=1} =$$

$$\sum_{n=1}^{\infty}\sum_{j=1}^{\infty}\sum_{m=-n}^{n}\sum_{k=-j}^{j}\hat{H}^{o,7}\eta_{j,k}p_{-(n+1),m}^o + \hat{H}^{o,8}\eta_{j,k}\varphi_{n,m}^o$$

$$+\hat{H}^{i,7}\eta_{j,k}p_{n,m}^i + \hat{H}^{i,8}\eta_{j,k}\varphi_{n,m}^i + \hat{H}^9\eta_{j,k}\eta_{n,m}, \qquad (A2.64)$$

where

$$\hat{H}^{o,7} = \Big[\frac{2(n+1)(n+2)(n+3)}{i\Omega} + n + 1\Big]I_1(n,m,j,k); \qquad (A2.65a)$$

$$\hat{H}^{o,8} = \Bigg[-n^2(n-2)R_n^o(\sqrt{i\Omega}) + n\frac{dR_n^o(\sqrt{i\Omega}r)}{dr}$$

$$+(2n+1)(n-2)S_n^o(\sqrt{i\Omega}) + (2n+1)\frac{dS_n^o(\sqrt{i\Omega}r)}{dr}\Bigg]I_1(n,m,j,k); \qquad (A2.65b)$$

$$\hat{H}^{i,7} = -\Big[\frac{2n(n-1)(n-2)}{i\Omega} - n\Big]I_1(n,m,j,k); \qquad (A2.65c)$$

$$\hat{H}^{i,8} = \left[-n^2(n-2)R_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r) + n\frac{dR_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr} \right.$$

$$\left. +(2n+1)(n-2)S_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r) + (2n+1)\frac{dS_n^i(\sqrt{\frac{i\Omega\alpha}{\lambda}}r)}{dr} \right] I_1(n,m,j,k); \quad (A2.65d)$$

$$\hat{H}^9 = \Gamma 2([2-n(n+1)]-1)I_1(n,m,j,k). \quad (A2.65e)$$

CHAPTER IV

On the Effect of a Fluid-Fluid Interface on the Brownian Motion of a Solid Particle

While the Brownian motion of a rigid particle in an infinite fluid has been studied initially by Einstein [1], and subsequently via a dynamic approach by Hinch [2], Hauge and Martin-Löf [3], Zwanzig and Bixon [4] and others, more general situations have begun to be considered only more recently. The case of clusters or suspensions of rigid particles has been investigated by numerous researchers, (Russell [5], Brady and Bossis [6], Felderhof and Jones [7], Brenner, Nadim and Haber [8],) and wall effects have also been considered (e.g., Gotoh and Kaneda [9].) However, Brownian motion in systems with free fluid interfaces seems to have received comparatively less attention.

Each additional rigid particle introduced into a multiparticle system adds a finite number of degrees of freedom (the coordinates of the particle). Rigid particles have only translational and rotational degrees of freedom, and since the kinetic energy associated with each is well-known (depending only on the mass and moments of inertia), it is clear how Brownian energy is distributed to each of the modes. When more than one particle is present, the time-dependent system response will depend on the configuration, so the challenging problem is how to average properly over the different configurations to obtain the mean motion of a particle.

The case of a rigid particle near a fluid-fluid interface is somewhat different, in that the interface introduces an infinite number of degrees of freedom, since it can take on an infinite number of different configurations. While this may also be true of an unbounded suspension, the behavior of one particle in a suspension is influenced by the detailed configuration of only a finite number of particles in its vicinity, beyond which the remaining particles affect it only in some averaged way, with their detailed configuration not being important. In contrast, even the portion of the interface that is near enough to the particle to influence it significantly can take on an infinite number of configurations. Also, the interface can "store" energy in potential form because it has surface tension. It is natural to ask how the effect of this interface on the Brownian motion of a particle differs from the effect of other rigid Brownian particles placed in the proximity of the particle.

In his thesis, Yang [10] studied this problem of the Brownian motion of a rigid particle in the presence of a fluid-fluid interface. His results seemed to indicate that the interface affects the motion of the particle only insofar as it changes its mobility. Since his work forms a good starting point for consideration of this problem, it will be summarized briefly here, along with some discussion that will attempt to elucidate the physical meaning of his results.

Yang assumed that the complete fluid fluctuations could be modelled by considering the flow to be driven by a random force on the particle and a random normal stress on the interface. The governing equations then became

$$\rho_j \frac{\partial \mathbf{u}^j}{\partial t} = -\nabla p^j + \mu_j \nabla^2 \mathbf{u}^j; \tag{1.1}$$

$$\nabla \cdot \mathbf{u}^j = 0 \tag{1.2}$$

(where j=1,2, the particle is in fluid 2, μ_i, $i = 1,2$ is the viscosity of fluid i, and ρ_i, $i = 1,2$ is the density of fluid i) with boundary conditions of continuity of velocity at the interface

$$\mathbf{u}^1 = \mathbf{u}^2, \tag{1.3}$$

the kinematic condition

$$\mathbf{u}^i = \frac{\partial \eta}{\partial t}, \tag{1.4}$$

(where η is the displacement of the interface from the plane $x_3 = 0$), the two conditions of continuity of tangential stress,

$$\mathbf{t} \cdot \mu_1[\nabla \mathbf{u}^1 + (\nabla \mathbf{u}^1)^T] = \mathbf{t} \cdot \mu_2[\nabla \mathbf{u}^2 + (\nabla \mathbf{u}^2)^T] \tag{1.5}$$

(where $\mathbf{t}$ is either of two independent vectors tangent to the interface), the normal stress condition

$$-p^1 + \mathbf{n} \cdot \mu_1[\nabla \mathbf{u}^1 + (\nabla \mathbf{u}^1)^T] + p^2 - \mathbf{n} \cdot \mu_2[\nabla \mathbf{u}^2 + (\nabla \mathbf{u}^2)^T] = \gamma \nabla \cdot \mathbf{n} + (\Delta \rho) g \eta + y \tag{1.6}$$

(where $\mathbf{n}$ is the normal to the interface, γ is the surface tension, $\Delta\rho$ is the density difference between the two fluids, g is of course the acceleration of gravity, and y is the random normal stress on the interface, assumed by Yang to model the fluctuations); and finally, the no-slip condition at the surface of the particle, $|\mathbf{x} - \mathbf{X}| = a$,

$$\mathbf{u}^2 = \mathbf{U}. \tag{1.7}$$

Here, $\mathbf{X}$ denotes the particle position, $\mathbf{U}$ denotes its velocity, and a is its radius. The particle motion is described by the "Langevin equation"

$$m_P \frac{d\mathbf{U}}{dt} + \mathbf{B}(t)[\mathbf{U}] = \mathbf{F}_{fluc} \qquad \Big(= \mathbf{F}(t) + \mathbf{A}(t)\Big) \ , \tag{1.8}$$

where the right-hand side is the force on the particle due to the random fluctuations. As discussed below, Yang assumed that this can be divided into the "indirect" random force $\mathbf{F}(t)$ due to the flow created by the random normal stress y acting on the fluid-fluid interface, and the "direct" random force $\mathbf{A}(t)$ due to the random thermal motion of the fluid molecules in the immediate vicinity of the particle. This separation is of course artificial, since in fact there are random driving forces throughout the entire fluid. Nevertheless, it seems reasonable to focus on the Brownian forces acting in the immediate vicinity of the interface if what is desired is to determine whether the deformability of the interface (which introduces extra macroscopic degrees of freedom) affects the Brownian motion of the particle in some way beyond just the obvious fact that it changes the particle's mobility.

Since the position of the particle boundary varies with time, the complete problem is nonlinear in the unknown particle position. Yang's investigation was a first approximation in the case that the motion of the particle is negligible on a length scale characterizing the flow field caused by interface fluctuations. The problem becomes linear in the particle position in this case, since the hydrodynamic force on the particle is calculated with the assumption that the particle remains at a fixed position. This will be referred to below as "**Assumption 1.**" His analysis also assumed that the fluctuations could be modelled by having only a random *normal* stress at the boundaries. This will be referred to henceforth as "**Assumption 2.**" In later sections, the effect of weakening these assumptions will be considered.

With these two assumptions, Yang could consider the particle diffusivity to consist of two *separate* contributions: one from the "direct" random force $\mathbf{A}(t)$, and the other from the "indirect" force $\mathbf{F}(\mathbf{x}, t)$ created by the motion of the interface. This is possible because these two forces are uncorrelated, and the problem is now linear. (That is, the force on the particle as a function of time is a linear functional of the random normal stress. If the variation of the force with the particle's change of position *were* taken into account, then this relation would be nonlinear.) In the actual problem (without Assumptions 1 and 2,) the diffusivity cannot be separated into two parts in this simple way, because of (a) the hydrodynamic interaction between the particle and the interface, and (b) the fact that the force created by the interface motion is a function of position as well as of time, and the particle's position is changing. With Assumption 1, the Langevin equation becomes

$$m_P \frac{d\mathbf{U}}{dt} + \mathbf{B}(t)[\mathbf{U}] = \mathbf{F}(\mathbf{x}, t)\Big|_{\mathbf{x}=\mathbf{X}(t)} + \mathbf{A}(t), \tag{1.9}$$

and with Assumption 2, it becomes

$$m_P \frac{d\mathbf{U}}{dt} + \mathbf{B}(t)[\mathbf{U}] = \mathbf{F}(\mathbf{x}, t)\Big|_{\mathbf{x}=\mathbf{X}_0} + \mathbf{A}(t), \tag{1.10}$$

where $\mathbf{X}_0$ is the initial position of the particle (assumed for simplicity to be $(0, 0, d)$.)

The force $\mathbf{F}$ in (1.10) is the hydrodynamic force on the particle due to the flow field created by interface fluctuations. The interface fluctuations arise, in turn, from the random normal stress y (appearing in (1.6)) that is assumed to model the thermal forces. The autocorrelation of $\mathbf{F}$ is needed to get information about the particle motion from the Langevin equation, and the autocorrelation of y is needed to get the autocorrelation of $\mathbf{F}$. The important assumption that is made about y is that it has an essentially zero correlation time:

$$< y(t)y(t+\tau) > \ \propto \delta(t-\tau). \tag{1.11}$$

This is the assumption that the time scale on which the random stress varies is much smaller than the time scale on which the interface fluctuates. Information about the spatial (equilibrium) correlations of y must come from statistical thermodynamic considerations, via the usual assumption that the probability of a certain configuration is related to the energy of the configuration by

$$P[\eta(\mathbf{x}_s] = \exp\left[-\frac{E[\eta(\mathbf{x}_s)]}{\kappa_B T}\right], \tag{1.12}$$

where κ_B is Boltzmann's constant, and T is absolute temperature. Since this is an equilibrium calculation, the energy here is due to surface tension and gravity. Yang found the equilibrium correlations of η (Fourier-transformed in the two spatial directions of the interface) to be

$$< \eta(\mathbf{k},t)\eta(\mathbf{k}',t) >= \frac{\kappa_B T}{2\pi}\frac{1}{\left[(\Delta\rho)g - \gamma\mathbf{k}\cdot\mathbf{k}'\right]}\delta(\mathbf{k}+\mathbf{k}'). \tag{1.13}$$

Here, $\mathbf{k}$ and $\mathbf{k}'$ are wavevectors, $\Delta\rho$ is the density difference across the interface, and γ is the surface tension. It can be seen from this that the deformations with the highest amplitude on average are the ones with the longest wavelength. This makes Assumption 1 more plausible, since the wavelength of the deformation is also the length scale characterizing spatial variations in the flow.

In calculating this force, Yang neglected the hydrodynamic interaction between the particle and the interface. In other words, he solved the hydrodynamic problem of Equations (1.1) and (1.2), subject to the boundary conditions (1.3), (1.4), (1.5) and (1.6), with no particle present. By assuming that the interface deformation is small, it is possible to linearize the nonlinear surface tension term in (1.6), and also to apply the boundary conditions at the undeformed interface to get a first-order approximation to the flow field. Since the problem is linear, with boundary conditions on a plane, it is convenient to Fourier transform in the two space directions tangent to the plane of the undeformed interface, as well as in time. The wavevector is denoted by $\mathbf{k}$, and ω is the frequency. Yang's calculations lead to the following velocity field in fluid 1 (which occupies $x_3 > 0$):

$$\mathbf{w}^{(1)}(\mathbf{k}, x_3, \omega) = \Big[\Phi_1(\mathbf{k}, \omega) e^{-k x_3} + \Psi_1(\mathbf{k}, \omega) e^{-\alpha_1 x_3}\Big] \hat{\eta}(\mathbf{k}, \omega); \tag{1.14}$$

and in fluid 2 (which occupies the region $x_3 < 0$; the particle is in fluid 2):

$$\mathbf{w}^{(2)}(\mathbf{k}, x_3, \omega) = \Big[\Phi_2(\mathbf{k}, \omega) e^{-k x_3} + \Psi_2(\mathbf{k}, \omega) e^{-\alpha_2 x_3}\Big] \hat{\eta}(\mathbf{k}, \omega). \tag{1.15}$$

Here,

$$\alpha_1 = \left(k^2 - \frac{i\omega}{\nu_1}\right)^{1/2}; \tag{1.16a}$$

$$\alpha_2 = \left(k^2 - \frac{i\omega}{\nu_2}\right)^{1/2}; \tag{1.16b}$$

$$\Phi_1(k, \omega) = \frac{i\omega\nu_1(k + \alpha_1) + \nu_2(\alpha_2 - k)[-2k^2\nu_1(\lambda - 1) + i\omega\lambda]}{\nu_1(k - \alpha_1) + \lambda(k - \alpha_2)}; \tag{1.17a}$$

$$\Phi_2(k, \omega) = \frac{i\omega\lambda\nu_1(k + \alpha_2) + \nu_1(\alpha_1 - k)[2k^2\nu_2(\lambda - 1) + i\omega]}{\nu_1(k - \alpha_1) + \lambda(k - \alpha_2)}; \tag{1.17b}$$

$$\Psi_1(k, \omega) = \frac{-2\nu_1\nu_2 k^2(\lambda - 1)(\alpha_1 - k) - 21\omega k\nu_2}{\nu_1(k - \alpha_1) + \lambda(k - \alpha_2)}; \tag{1.18a}$$

$$\Psi_2(k, \omega) = \frac{2\nu_1\nu_2 k^2(\lambda - 1)(\alpha_1 - k) - 21\omega k\lambda\nu_2}{\nu_1(k - \alpha_1) + \lambda(k - \alpha_2)}. \tag{1.18b}$$

The solution for the interface deformation is

$$\hat{\eta}(\mathbf{k},\omega) = \frac{\hat{y}(\mathbf{k},\omega)}{\hat{H}_I(\mathbf{k},\omega)}, \tag{1.19}$$

where

$$\begin{aligned}\hat{H}_I(k,\omega) = \frac{i\omega}{k}\Big[\rho_2\Phi_2(k,\omega) + \rho_1\Phi_1(k,\omega)\Big] \\ - 2(\mu_2 - \mu_1)\Big[k\Phi_2(k,\omega) + \alpha_2\Psi_2(k,\omega)\Big] \\ - \Big[(\Delta\rho)g + \gamma k^2\Big].\end{aligned} \tag{1.20}$$

By comparison with his equilibrium correlations (1.13) for the deformation, Yang finds that the spatial autocorrelation of the random normal stress is

$$< y(\mathbf{k},\omega)y(\mathbf{k}',\omega') >= \frac{\kappa_B T\delta(\mathbf{k}+\mathbf{k}')\delta(\omega+\omega')}{(2\pi)^2\Big[(\Delta\rho)g - \gamma\mathbf{k}\cdot\mathbf{k}'\Big]\int_{-\infty}^{\infty}\frac{ds}{\hat{H}_I(\mathbf{k},s)\hat{H}_I(\mathbf{k}',-s)}}. \tag{1.21}$$

It is noteworthy that the **k**-dependent factors in (1.21) outside the delta function will cause a "spreading out" upon taking inverse Fourier transforms, so that in physical space, the random stress will *not* be delta-correlated.

The force on the particle is then determined from the flow field by using the unsteady Faxen's Law for a rigid particle in Fourier-transformed form. These hydrodynamical calculations for the force give $\mathbf{F}(\mathbf{k}, x_3, \omega)$, i.e., the force Fourier- transformed in the two space directions x_1, x_2. By Assumption 1, which says that the particle does not move far before many Brownian events have occurred, the force on the particle at all times can be approximated by the force at the particle's initial position. Thus, it is necessary to express **F** at the fixed point $\mathbf{X}_0 = (0, 0, d)$, which can be taken without loss of generality to be the initial position of the particle, in terms of this Fourier-transformed solution. This is done using the inverse Fourier transform:

$$\mathbf{F}(\mathbf{X}_0,\omega) = \int_{\mathbf{k}} \mathbf{F}(\mathbf{k},d,\omega)e^{i\mathbf{k}\cdot\mathbf{X}_0}d\mathbf{k} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbf{F}(\mathbf{k},d,\omega)dk_1dk_2. \tag{1.22}$$

Thus the Fourier-transformed Langevin equation for particle motion due only to interface fluctuations is

$$-i\omega m_P\mathbf{U} + \hat{\mathbf{B}}(\omega)[\mathbf{U}] = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbf{F}(\mathbf{k},d,\omega)dk_1dk_1. \tag{1.23}$$

By the usual methods of linear response theory, this Langevin equation can be used to get the particle velocity autocorrelation in terms of the force autocorrelation. The force autocorrelation has been related to the random stress autocorrelation via the Faxen's Law and the hydrodynamical results given above. Thus, Yang was able to compute the particle velocity autocorrelation. The particle diffusivity, if defined as the infinite-time limit of the rate of growth of the mean-square particle displacement, can be obtained from

$$\mathbf{D} = \lim_{\omega \to 0} \pi\mathbf{R}_U(d;\omega,-\omega), \tag{1.24}$$

where $\mathbf{R}_U$ is the particle velocity autocorrelation. Yang found this limit to be zero.

A number of questions arise on consideration of Yang's analysis. If some hydrodynamic interaction between the particle and the interface were to be included, would this change the result that the diffusivity is zero? Even if the diffusivity (in the classical sense) is zero, does the mean-square particle displacement continue to grow with time in some sort of anomalous diffusion, or does the particle remain in some finite region for all time? Is the time limit $t \to \infty$ "too long" in the sense that for some time interval, the mean-square displacement will grow diffusively, but that after a very long time it will level off? What happens if Assumption 1 is weakened, i.e., if the particle moves significantly relative to the length scale characterizing the flow field created by interface fluctuations before a meaningful diffusivity can be defined? What happens if Assumption 2 is dropped? For instance, what if there are random tangential stresses at the interface in addition to normal stresses? Finally, why do the normal stresses on the surface have a *finite* length scale for autocorrelation? Their physical origin is the collisions of fluid molecules with the surface, so that no correlation would be expected between the stress on nearby points on the interface. Each of these questions will be addressed below.

The Spring Analogy to Surface Tension

First, consider the question of whether using a better approximation to the hydrodynamic interaction between the interface and the particle could change the result that the interface fluctuations ("independent" of particle motion) do not give rise to a particle diffusivity. (For example, when the force on the particle due to the random flow field is computed, instead of using a Faxen's Law for an infinite fluid, a Faxen's Law that takes into account the presence of the interface could be used.) It will be shown here that the answer is definitely "No" as long as Assumption 1 is retained. With Assumption 1, it is possible to use the technique of linear-response theory which is the standard method for dealing with hydrodynamic fluctuation problems. Without Assumption 1, the problem cannot be linearized, so it becomes a nonlinear stochastic differential system, for which very few techniques of solution are available. (The nonlinearity arises because the force depends on the particle position in a nonlinear way, and it is the particle position that is to be solved for.) The Langevin equation without Assumption 1 takes the form of a differential equation driven by a stochastic field. This is equivalent to the problem that arises in stochastic quantization of field theory, for which perturbation methods (i.e., expansion about the initial position) have been developed and used extensively by physicists. It is unfortunately still an open problem as to how to deal with the case where the particle position changes by more than just a small amount relative to the length scale of the field it is placed in (the field in this case being the flow field created by the interface motion). Thus, only the perturbation method will be discussed below, and it is hoped that it will at least help in the qualitative understanding of how Assumption 1 affects the result.

The Fourier-transformed particle Langevin equation will be considered again:

$$m_P \frac{d\mathbf{U}}{dt} + \mathbf{B}(t)[\mathbf{U}] = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbf{F}(\mathbf{k}, d, \omega) dk_1 dk_1 + \mathbf{A}(t), \tag{1.25}$$

and the way it is used to relate the particle velocity to the random stress will be reviewed. First, the particle velocity must be related to the random normal stress on the interface, for which the autocorrelations in space and time are known. Yang obtains this relation through several hydrodynamic calculations. In terms of Fourier-transformed variables, these relations take the form of multiplication by "transfer functions." First, the particle velocity is related to the force on the particle: (this comes from the Fourier- transformed Langevin equation)

$$\hat{H}_U(\omega)\hat{\mathbf{U}} = \hat{\mathbf{F}}, \tag{1.26}$$

where $\hat{H}_U$ includes the particle inertia and the unsteady particle mobility. Then the force on the particle is related to the flow field generated by the interface motion, via the unsteady Faxen's Law:

$$\hat{\mathbf{F}} = \hat{H}_{Fax}(\omega)\hat{\mathbf{u}}(0,0,d,\omega) = \hat{H}_{Fax}(\omega)\int_{\mathbf{k}} \hat{\mathbf{u}}(\mathbf{k},d,\omega)d\mathbf{k}, \tag{1.27}$$

where the fact that the particle position is approximated as $(0,0,d)$ for all time has been used. The undisturbed fluid velocity $\hat{\mathbf{u}}(\mathbf{k},d,\omega)$, which, unlike the particle velocity $\hat{\mathbf{U}}(\omega)$ (but like the force $\hat{\mathbf{F}}(\mathbf{k},x_3,\omega)$) is Fourier-transformed in x_1, x_2 as well as in t, (it being a function of *space* as well as of time,) is in turn expressed in terms of the interface deformation (the relation coming from solving the Stokes equations in the two fluids for small deformations):

$$\hat{\mathbf{u}}(\mathbf{k},d,\omega) = \hat{H}_{flow}(\mathbf{k},d,\omega)\hat{\eta}(\mathbf{k},\omega); \tag{1.28}$$

and finally, the interface deformation is related to the random normal stress on the interface by the so-called "interface susceptibility":

$$\hat{H}_I(\mathbf{k},\omega)\hat{\eta}(\mathbf{k},\omega) = \hat{y}(\mathbf{k},\omega). \tag{1.29}$$

Combining all these relations gives:

$$\hat{\mathbf{U}} = \frac{1}{\hat{H}_U(\omega)}\hat{H}_{Fax}(\omega)\int_{\mathbf{k}} \hat{H}_{flow}(\mathbf{k},d,\omega)\frac{1}{\hat{H}_I(\mathbf{k},\omega)}\hat{y}(\mathbf{k},\omega)d\mathbf{k}. \tag{1.30}$$

Yang assumed that the diffusivity of the particle is the limit as $\omega \to 0$ of the Fourier transform of its velocity-autocorrelation function. Consequently, it is clear that if any of the factors (or "transfer functions") in the above equation go to zero as $\omega \to 0$, the diffusivity will also be zero. Now consider the factors, one by one. First, the function $\hat{H}_U$ clearly tends to the finite limit $6\pi\mu a$, the steady drag on a solid sphere, as $\omega \to 0$. Similarly, the factor $\hat{H}_{Fax}$ goes to $6\pi\mu a$, by the steady Faxen's Law for a rigid particle. These two statements can in fact be combined into one: At very low frequency, a neutrally buoyant particle placed in a zero-Reynolds-number flow field will move with the undisturbed fluid velocity. Consider next the function $\hat{H}_I$, which determines how much interface deformation there is for a given imposed normal stress. Clearly, this is finite even in the zero- frequency limit. A constant normal stress will cause the interface to deform until the surface tension force balances the imposed force. Finally, consider the function $\hat{H}_{flow}$. Here is the key to the matter. As the rate at which the interface is deformed goes to zero, the flow field in the fluid caused by the interface motion goes to zero, at least in the case of an infinite planar interface considered by Yang. In the limit as the interface just remains fixed in some deformed state, there is no flow at all. In other words, the function $\hat{H}_{flow} \to 0$ as $\omega \to 0$. *This is why the diffusivity is zero.* This also makes it clear that including more sophisticated hydrodynamics will not change this result. Hydrodynamical interaction between the interface and the particle *will not change the fact that* $\hat{H}_{flow} \to 0$.

Why is forcing a particle via a fluid medium different from forcing it directly? Clearly, the direct random force on the particle $\mathbf{A}(t)$ gives rise to a diffusivity similar to the Stokes-Einstein result but with the mobility modified from $6\pi\mu a$ to something else because of the presence of the interface. In contrast, $\mathbf{F}$, the "indirect" force, will not. It turns out that the essential difference is due to the fact that the interface, when idealized as infinite, "can't go anywhere." Surface tension (and gravity) act as a spring, pulling it back towards its initial position. This analogy will now be pursued to get a better understanding of the situation.

Instead of an interface, consider another large solid particle, free to wander around, that is acted on by a random force. As above, consider only the particle's motion due to the motion of this larger particle, and not the motion due to the force

$\mathbf{A}(t)$ that acts directly on the particle. Landau and Lifshitz [11] have determined the unsteady mobility of a particle in a viscous fluid (at zero Reynolds number):

$$\hat{\mathbf{F}}_{hyd} = \left[6\pi\mu R\left(1 + R\sqrt{\frac{\rho\omega}{2\mu}}\right) - i\omega(3\pi R^2)\sqrt{\frac{2\mu\rho}{\omega}}\left(1 + \frac{2R}{9}\sqrt{\frac{\rho\omega}{2\mu}}\right)\right]\hat{\mathbf{U}}, \tag{1.31}$$

where $\mathbf{F}_{hyd}$ is the hydrodynamic force on the particle, $\mathbf{U}$ is the particle velocity, R is the particle radius, μ is the fluid viscosity, ρ is the fluid density, and the circumflex indicates that the variables have been Fourier-transformed according to

$$\mathbf{U}(t) = \int_{-\infty}^{\infty} \hat{\mathbf{U}}(\omega)e^{-i\omega t}d\omega, \tag{1.32}$$

and similarly for $\mathbf{F}_{hyd}$. For convenience, rewrite this relation as

$$\hat{\mathbf{F}}_{hyd} = \hat{G}_1(\omega)\hat{\mathbf{U}}. \tag{1.33}$$

The large particle's motion is thus described by the Langevin equation

$$-i\omega m_L\hat{\mathbf{U}} = \hat{G}_1(\omega)\hat{\mathbf{U}} + \hat{\mathbf{F}}, \tag{1.34}$$

where $\hat{\mathbf{F}}$ is the ("direct") random force on the particle, and m_L is the mass of the large particle. This can be rewritten as

$$\hat{\mathbf{U}} = \frac{\hat{\mathbf{F}}}{-i\omega m_L - \hat{G}_1(\omega)}. \tag{1.35}$$

Landau and Lifshitz also give the velocity field in the fluid generated by the sphere's motion as (with r being the spherical coordinate of distance from the particle center),

$$\hat{\mathbf{u}} = \hat{U}_r \mathbf{e}_r \left[-\frac{2}{r^3} \left[A e^{ikr} (r - \frac{1}{ik}) + B \right] + \frac{1}{r^2} aike^{ikr} (r - \frac{1}{ik}) + \frac{ae^{ikr}}{r^2} \right]$$

$$-\frac{1}{r} ikAe^{ikr} \hat{\mathbf{U}}, \tag{1.36}$$

where

$$k = (1+i)\sqrt{\frac{2\mu}{\rho\omega}}, \tag{1.37}$$

$$A = -\frac{3R}{2ik} e^{-ikR}, \tag{1.38}$$

$$B = -\frac{R^3}{2}(1 - \frac{3}{ikR} - \frac{3}{k^2R^2}). \tag{1.39}$$

Write this relation as

$$\hat{\mathbf{u}} = \hat{\mathbf{G}}_2(r,\omega) \cdot \hat{\mathbf{U}}. \tag{1.40}$$

Finally, there is an unsteady Faxen's Law that gives the force on the smaller sphere in terms of its velocity and the flow field that it is placed in. (This flow field will be taken as the field generated by the motion of the bigger sphere. Hydrodynamic interaction between the two spheres is being neglected here, as it will not qualitatively affect the result.) Clearly, if the sphere is neutrally buoyant and sufficiently small, this relation can be approximated by just having the small sphere move affinely with the fluid (i.e., with the undisturbed fluid velocity at its center). This is certainly true in the limit as $\omega \to 0$. Combining all this gives

$$\hat{\mathbf{U}}_s = \hat{\mathbf{u}}\Big|_{r=r_s} = \hat{\mathbf{G}}_2(r_s,\omega)\cdot\mathbf{U} = \hat{\mathbf{G}}_2(r_s,\omega)\cdot\frac{\hat{\mathbf{F}}(\omega)}{-[i\omega m_L + \hat{G}_1(\omega)]}. \qquad (1.41)$$

Here, $\mathbf{U}_s$ denotes the velocity of the smaller sphere, and r_s denotes its position in the spherical coordinate system centered at the larger sphere. It follows from this and linear response theory that

$$\hat{R}_{\mathbf{U}_s} = \left|\frac{\hat{\mathbf{G}}_2(r_s,\omega)}{i\omega + \hat{G}_1(\omega)}\right|\hat{R}_{\mathbf{F}}, \qquad (1.42)$$

where $\hat{R}_{\mathbf{U}_s}$ denotes the Fourier transform of the autocorrelation of the small sphere's velocity, and $\hat{R}_{\mathbf{F}}$ denotes the Fourier transform of the autocorrelation of the (random) force on the large sphere. Now within the framework of this calculation, $\hat{R}_{\mathbf{F}}$ should be given by the fluctuation-dissipation theorem given in, for example, Hinch [2]:

$$\hat{R}_{\mathbf{F}} = \frac{1}{2\pi}\frac{kT}{6\pi\mu R}. \qquad (1.43)$$

(The $1/2\pi$ factor is the Fourier transform of the Dirac delta function.) Also, the diffusivity of the small sphere should be given by

$$\mathbf{D} = \lim_{\omega\to 0}\hat{R}_{\mathbf{U}_s} \qquad (1.44)$$

(this is shown in Hinch [2] among other sources.) Thus,

$$\mathbf{D} = \lim_{\omega\to 0}\left|\frac{\hat{\mathbf{G}}_2(r_s,\omega)}{[i\omega m_L + \hat{G}_1(\omega)]}\right|\frac{1}{2\pi}\frac{kT}{6\pi\mu R}. \qquad (1.45)$$

To see that the limit in (1.45) is nonzero, note that by (1.31) and (1.33),

$$\hat{G}_1(\omega) \rightarrow 6\pi\mu R \tag{1.46}$$

as $\omega \rightarrow 0$; also, it is clear that the flow field generated by the large sphere's motion will tend to the Stokes solution for steady motion of a sphere in this limit, so that

$$\hat{\mathbf{G}}_2(r_s,\omega) \rightarrow \left[\frac{3R}{4} + \frac{R^3}{4r_s^2}\right]\mathbf{I} + \left[\frac{3R}{4r_s} - \frac{3R^3}{4r_s^3}\right]\frac{\mathbf{x}_s\mathbf{x}_s}{r_s^2}. \tag{1.47}$$

The main point that is illustrated by all this is that the small sphere has a *nonzero* diffusivity due only to the motion of the large sphere driven by a random force directly on the large sphere.

It was pointed out above that while the effect of the interface on the small particle's diffusivity should be qualitatively similar to that of many particles (or even just one) undergoing Brownian motion and thereby creating flow fields, the interface is not free to move an indefinite distance from its initial position. To model this feature, to the simplified case of a single large particle considered above, a spring tending to bring the large particle back to its initial position will be added. The spring force will be given by

$$\mathbf{F}_{spr} = k\mathbf{X}, \tag{1.48}$$

where k is a spring constant, and $\mathbf{X}$ is the position of the large particle. Thus, the Langevin equation of the large particle is changed to

$$-i\omega m_L\hat{\mathbf{U}} = -\frac{k}{i\omega}\hat{\mathbf{U}} + \hat{G}_1(\omega)\hat{\mathbf{U}} + \hat{\mathbf{F}}, \tag{1.49}$$

so that the expression for the diffusivity is changed to

$$\mathbf{D} = \lim_{\omega \to 0} \left| \frac{\hat{\mathrm{G}}_2(r_s, \omega)}{\left[i\omega m_L + \hat{G}_1(\omega) + \frac{ik}{\omega}\right]} \right| \frac{1}{2\pi} \frac{kT}{6\pi\mu R}. \tag{1.50}$$

If this is rewritten as

$$\mathbf{D} = \lim_{\omega \to 0} \left| \frac{\omega\hat{\mathrm{G}}_2(r_s, \omega)}{[i\omega^2 m_L + \omega\hat{G}_1(\omega) + ik]} \right| \frac{1}{2\pi} \frac{kT}{6\pi\mu R}, \tag{1.51}$$

and the fact that $\hat{G}_1$ and $\hat{G}_2$ go to a finite nonzero limit as $\omega \to 0$ is recalled, it becomes clear that adding the spring makes a significant difference: The diffusivity of the small particle is now zero! In fact, the diffusivity of the large particle is also zero for the same reason, namely, that the transfer function giving the large particle velocity in terms of the random force on it goes to zero as $\omega \to 0$, because of the spring force. This is entirely analogous to the fact that $H_{flow} \to 0$ in the same limit, as observed above in the discussion of Yang's analysis with a fluid-fluid interface instead of a large particle on a spring. Like the interface, the large particle cannot roam away farther and farther as time passes; instead, it approaches a finite mean-square displacement from its initial position.

Is the particle "trapped" or does it diffuse anomalously?

The fact that the diffusivity in the classical sense is zero does not necessarily mean that the mean-square particle displacement stops growing at long times. It means only that the rate of growth is slower than linear in time. The limiting behavior of the particle velocity autocorrelation at long times will now be considered in more detail, to determine whether the particle diffuses anomalously, or whether its mean-square displacement actually reaches a finite limit at large times.

There is another reason for considering the long-time $\omega \to \infty$ limit more closely. The arguments made above that the particle is "trapped" because surface-tension acts like a spring pulling the interface back do not hold if the interface simply undergoes rigid-body translation and if gravity effects are not present ($\Delta\rho = 0$.) Gravity is clearly also analogous to a spring force, because any displacement from the equilibrium flat state will give rise to a gravitational force tending to restore the interface to its flat state, and the larger the displacement, the larger the force (it is actually a linear relationship). Surface tension only exerts a restoring force if the displacement is not uniform, i.e., if the curvature becomes nonzero. If the entire interface were displaced by a constant distance, there would be no restoring surface tension force. Thus, if gravity were considered to be absent from the problem, it would seem that infinite wavelength disturbances ($k \to 0$) might give rise to a nonzero diffusivity. In fact, when there is no density difference between the two fluids, the interface becomes unstable, and the equilibrium average amplitude of zero-wavevector (infinite-wavelength) disturbances becomes infinite.

When the limit as $\omega \to 0$ of the velocity-autocorrelation function (which is an integral over the wavenumber vector $\mathbf{k}$), is taken in Yang's analysis, there seems to be an implicit assumption that the limit commutes with the integral, i.e., that the integrand is uniformly continuous. A close examination of the integral reveals that taking the limit is not such a simple process. In fact, it can be shown that the major contribution to the integral comes from the neighborhood of $\mathbf{k} = 0$, where the integrand is not uniformly continuous. This makes sense physically, since the limit $\omega \to 0$ corresponds to taking the limit of long time, and in this limit only the infinite wavelength disturbances should survive. This may be compared with the calculation of transport coefficients via linear-response theory in Kreuzer [12], where both the limits $\mathbf{k} \to 0$ and $\omega \to 0$ must be taken, and it is found that the ω limit must be taken *after* the $\mathbf{k}$ limit to insure getting a nonzero result. In the case of diffusion, this corresponds to the fact that if **k** is not allowed to go to zero first, the particle is confined to a finite region, and thus in infinite time, since its mean-square displacement can't grow beyond this finite region, it will not diffuse. This suggests that the limit is singular, and should be examined more carefully.

So first, a change of variables must be made to make the integrand uniformly continuous. Second, the integrand itself contains another integral over a dummy variable which is a function of k. After the change of variables is made to insure uniform convergence, it also becomes a function of ω. Thus, its asymptotic expansion as $\omega \to 0$ must also be determined. At this point, it is important to note that it is necessary to go through these steps for two reasons: First, it must be determined whether the limit of the integral giving the diffusivity is really zero. It will be shown below that there is a factor in the integrand that tends to a delta function at $k = 0$ in the limit $\omega \to 0$. This factor when integrated would give a *finite* result even though it goes to zero for all *nonzero* k. Whether or not the limit of the integral is still zero will thus depend on the asymptotic behavior of the other factors in the integrand. Secondly, even if the asymptotic behavior is still $\propto \omega^p$, where p is positive, so that the diffusivity really is zero, it is of interest to know the value of p, so that it can be determined whether the mean-square displacement of the particle continues to grow in time even if slower than a diffusive rate (i.e., *anomalous diffusion*), or whether it asymptotes to a finite limit, so that the particle essentially never leaves a bounded region around its initial position.

If the mean-square particle displacement asymptotes to Dt^q, where q is a positive number, and D is a diffusion coefficient (in a general sense), then the case $q = 1$ corresponds to the usual definition of diffusion, in the sense that Yang uses it. However, all other strictly positive values of q correspond to so-called anomalous diffusion, in which the mean-square particle displacement *still grows in time*, but not at a linear rate. In particular, if $0 < q < 1$, then the diffusion coefficient as Yang calculates it, meaning the time derivative of the mean-square particle displacement (in the long-time limit), will be *zero even though the mean-square particle displacement is growing in time.* Therefore, to obtain a more complete understanding of the particle motion, the exponent p, discussed above (from which the exponent q can be determined), will be calculated here, since it is not given in Yang's thesis.

Yang used the following expression for the diffusivity,

$$\mathbf{D} = \lim_{\omega \to 0} \pi \langle \mathbf{U}(\omega)\mathbf{U}(-\omega) \rangle, \tag{1.52}$$

and if the velocity autocorrelation function he obtained (i.e., with Assumptions 1 and 2) is substituted into this, the result is

$$\begin{aligned}
\mathbf{D} = \lim_{\omega \to 0} \int_{k=0}^{k=\infty} \Bigg\{ & \frac{6\pi\mu_2 a}{m}\left[1 + a\sqrt{\frac{\omega}{2\nu_2}}(1-i)\right] \\
& \times \left[\Phi_2(k,\omega)e^{kx_3}(-i\mathbf{e}_1 + \mathbf{e}_3) + W_s(\omega)\Psi_2(k,\omega)e^{\alpha_2 x_3}\left(\frac{-i\alpha_2}{k}\mathbf{e}_1 + \mathbf{e}_3\right)\right] \\
& - \frac{2\pi a^3 \rho_2 \omega i}{m}\left[\Phi_2(k,\omega)e^{(}kx_3)(-i\mathbf{e}_1 + \mathbf{e}_3) + W_v(\omega)\Psi_2(k,\omega)e^{\alpha_2 x_3}\left(-\frac{i\alpha_2}{k}\mathbf{e}_1 + \mathbf{e}_3\right)\right]\Bigg\} \\
& \times \Bigg\{ \frac{6\pi\mu_2 a}{m}\left[1 + a\sqrt{\frac{-\omega}{2\nu_2}}(1-i)\right] \\
& \times \left[\Phi_2(k,-\omega)e^{kx_3}(-i\mathbf{e}_1 + \mathbf{e}_3) + W_s(-\omega)\Psi_2(k,-\omega)e^{\tilde{\alpha}_2 x_3}\left(\frac{-i\tilde{\alpha}_2}{k}\mathbf{e}_1 + \mathbf{e}_3\right)\right] \\
& + \frac{2\pi a^3 \rho_2 \omega i}{m}\Big[\Phi_2(k,-\omega)e^{(}kx_3)(-i\mathbf{e}_1 + \mathbf{e}_3) \\
& \quad + W_v(-\omega)\Psi_2(k,-\omega)e^{\tilde{\alpha}_2 x_3}\left(-\frac{i\tilde{\alpha}_2}{k}\mathbf{e}_1 + \mathbf{e}_3\right)\Big]\Bigg\}
\end{aligned}$$

$$\times \frac{\kappa_B T}{(2\pi)^2[(\Delta\rho)g + \gamma k^2]\hat{H}_I(k,\omega)\hat{H}_I(k,-\omega)\int_{-\infty}^{\infty}\frac{d\tilde{\omega}}{\hat{H}_I(k,\tilde{\omega})\hat{H}_I(k,-\tilde{\omega})}} k\,dk, \tag{1.53}$$

where some of the quantities appearing here have been defined earlier in (1.16), (1.17), (1.18), and the rest are given by

$$\tilde{\alpha}_1 = \left(k^2 + \frac{i\omega}{\nu_1}\right)^{1/2}; \tag{1.54a}$$

$$\tilde{\alpha}_2 = \left(k^2 + \frac{i\omega}{\nu_2}\right)^{1/2}; \tag{1.54b}$$

$$W_s(\omega) = \frac{\sin\left(a\sqrt{\frac{i\omega}{\nu_2}}\right)}{\left(a\sqrt{\frac{i\omega}{\nu_2}}\right)}; \tag{1.55}$$

$$W_v(\omega) = \frac{\sin\left(a\sqrt{\frac{i\omega}{\nu_2}}\right) - \left(a\sqrt{\frac{i\omega}{\nu_2}}\right)\cos\left(a\sqrt{\frac{i\omega}{\nu_2}}\right)}{\left(a\sqrt{\frac{i\omega}{\nu_2}}\right)^3}. \tag{1.56}$$

Since the derivation of this expression (which is just Equation (1.24) with all the transfer functions explicitly written) is given in Yang's thesis, it will not be reproduced here. Since this integral is cumbersome, it is convenient to consider only the special case where the two fluids, though immiscible, have the same density and viscosity. The integrand simplifies considerably in this case, and there is no reason to suspect that the qualitative behavior in this case is any different from the case of general density and viscosity ratios (except perhaps when they take on extreme values).

In this special case, the expression for the diffusivity reduces to (with only the (1,1) component shown here)

$$D_{11} = \frac{6\pi\mu a}{m}\frac{\kappa_B T}{\gamma^2} \begin{matrix} lim \\ \omega \to 0 \end{matrix} \int_{k=0}^{\infty} \left[e^{kx_3} \frac{\omega\left(k^2 - \frac{i\omega}{\nu}\right)^{1/2}}{k - \left(k^2 - \frac{i\omega}{\nu}\right)^{1/2}} \right.$$

$$+ \frac{\sin\left(a\sqrt{\frac{i\omega}{\nu}}\right)}{\left(a\sqrt{\frac{i\omega}{\nu}}\right)} \frac{\omega\left(k^2 - \frac{i\omega}{\nu}\right)^{1/2}}{\left(k^2 - \frac{i\omega}{\nu}\right)^{1/2} - k} \exp\left[\left(k^2 - \frac{i\omega}{\nu}\right)^{1/2} x_3\right] \Bigg]$$

$$\left[-e^{kx_3} \frac{\omega\left(k^2 + \frac{i\omega}{\nu}\right)^{1/2}}{k - \left(k^2 + \frac{i\omega}{\nu}\right)^{1/2}} \right.$$

$$- \frac{sin\left(a\sqrt{\frac{-i\omega}{\nu}}\right)}{\left(a\sqrt{\frac{-i\omega}{\nu}}\right)} \frac{\omega\left(k^2 + \frac{i\omega}{\nu}\right)^{1/2}}{\left(k^2 + \frac{i\omega}{\nu}\right)^{1/2} - k} \exp\left[\left(k^2 + \frac{i\omega}{\nu}\right)^{1/2} x_3\right] \Bigg]$$

$$\times \left[\frac{2\omega^2\rho\left(k^2 - \frac{i\omega}{\nu}\right)^{1/2}}{k\left(k - \left(k^2 - \frac{i\omega}{\nu}\right)^{1/2}\right)} - \gamma k^2 \right]^{-1}$$

$$\times \left[\frac{2\omega^2\rho\left(k^2 + \frac{i\omega}{\nu}\right)^{1/2}}{k\left(k - \left(k^2 + \frac{i\omega}{\nu}\right)^{1/2}\right)} - \gamma k^2 \right]^{-1} \frac{kdk}{k^2 F(k)}, \tag{1.57}$$

where

$$F(k) = \int_{-\infty}^{\infty} \frac{ds}{\hat{H}_I(k,s)\hat{H}_I(k,-s)}, \tag{1.58}$$

and $\hat{H}_I$ is now given by just

$$\hat{H}_I(k,\omega) = -\frac{2\omega^2\rho\alpha}{k(k-\alpha)} - \gamma k^2. \tag{1.59}$$

Now it should be clear that the terms

$$k - \left(k^2 - \frac{i\omega}{\nu}\right)^{1/2} \tag{1.60}$$

are the reason why the integral is not uniformly convergent in this form. When k and ω both go to zero, these terms vanish, but the order to which they vanish is unknown until what is generally called a "distinguished limit" is specified, i.e., the relative rate at which k and ω go to zero. Thus, taking the ω limit inside the integral in this form is clearly not legitimate. This difficulty can be removed by making the change of variables suggested by the form of the terms, namely, letting

$$k = \sqrt{\omega} u, \tag{1.61}$$

so that the diffusivity is now expressed as

$$\begin{aligned} D_{11} = \frac{6\pi\mu a}{m}\frac{\kappa_B T}{\gamma^2} \lim_{\omega \to 0} \int_{k=0}^{\infty} \Bigg[& e^{\sqrt{\omega} u x_3} \frac{\omega\left(u^2 - \frac{i}{\nu}\right)^{1/2}}{u - \left(u^2 - \frac{i}{\nu}\right)^{1/2}} \\ & + \frac{sin\left(a\sqrt{\frac{i\omega}{\nu}}\right)}{\left(a\sqrt{\frac{i\omega}{\nu}}\right)} \frac{\omega\left(u^2 - \frac{i}{\nu}\right)^{1/2}}{\left(u^2 - \frac{i}{\nu}\right)^{1/2} - u} \exp\left[\sqrt{\omega}\left(u^2 - \frac{i}{\nu}\right)^{1/2} x_3\right]\Bigg] \\ & \Bigg[-e^{\sqrt{\omega} k x_3} \frac{\omega\left(u^2 + \frac{i}{\nu}\right)^{1/2}}{u - \left(u^2 + \frac{i}{\nu}\right)^{1/2}} \\ & - \frac{sin\left(a\sqrt{\frac{-i\omega}{\nu}}\right)}{\left(a\sqrt{\frac{-i\omega}{\nu}}\right)} \frac{\omega\left(u^2 + \frac{i}{\nu}\right)^{1/2}}{\left(u^2 + \frac{i}{\nu}\right)^{1/2} - u} \exp\left[\sqrt{\omega}\left(u^2 + \frac{i}{\nu}\right)^{1/2} x_3\right]\Bigg] \\ & \times \left[\frac{2\omega^2\rho\left(u^2 - \frac{i}{\nu}\right)^{1/2}}{\sqrt{\omega}\left(u - \left(u^2 - \frac{i}{\nu}\right)^{1/2}\right)} - \gamma\omega u^3\right]^{-1} \\ & \times \left[\frac{2\omega^2\rho\left(u^2 + \frac{i}{\nu}\right)^{1/2}}{\left(u - \left(u^2 + \frac{i}{\nu}\right)^{1/2}\right)} - \gamma\omega u^3\right]^{-1} \frac{\omega u du}{\omega F(\sqrt{\omega} u)}. \end{aligned} \tag{1.62}$$

Now the limit *may* be commuted with the integral. As mentioned earlier, it is necessary to find the asymptotic expansion of the inside integral $F(\sqrt{\omega}u)$ as $\omega \to 0$. This is done in Appendix 1, where it is shown that

$$F(k) \sim k^{-5/2}, \qquad k \to 0, \tag{1.63}$$

which implies that

$$F(\sqrt{\omega}u) \sim \omega^{-5/4}, \qquad \omega \to 0. \tag{1.64}$$

If the other terms in the integrand are carefully expanded, it is then found that the entire integral goes like $\omega^{9/4}$. Thus, the particle is not undergoing anomalous diffusion; its mean-square displacement asymptotes to a finite limit as $t \to \infty$. It remains essentially in a finite region. It is worthwhile to recall that this result is for the (unstable) case where the density of the two fluids are the same. Curiously, the instability does not affect this calculation; even the fact that infinite-wavelength deformations have infinite average amplitude when $\Delta\rho = 0$ is not enough to keep the particle moving outwards. If there was a density difference, the mean-square displacement would grow even more slowly at large times.

The reason why infinite wavelength disturbances are not giving rise to a nonzero diffusivity can be traced back to the equilibrium calculations. The interface displacement $\eta(\mathbf{x}_s)$ has a mean- square equilbrium value, because the potential energy due to surface tension (or gravity) has, after linearization, quadratic terms in the interface displacement. Loosely speaking, this means that the interface will continue indefinitely to wiggle a small amount around its initial plane. For the interface actually to *diffuse*, the (absolute) interface displacement (i.e., the average value of

$\eta(\mathbf{x}_s)$ over the surface, a measure of how far the interface has left its initial plane), which may be denoted by h, would have to keep growing in time. This would be the case if there were a term quadratic in dh/dt (rather than quadratic in h, as the gravitational energy is), in the potential energy expression. That this is so is clear by comparison with a particle, which diffuses because equipartition gives it a mean square energy of $kT/2$ for each translational degree of freedom and it has three degrees of freedom corresponding to its kinetic energy $mU^2/2$. *No such kinetic energy can be assigned to an interface if it is idealized as infinite, because it is not possible to associate a mass (or inertia) to its rigid-body motion.* Alternatively, if the kinetic energy associated with its rigid-body motion is taken to be the kinetic energy of all the fluid that it sets in motion, this energy would be infinite, and therefore the equilibrium correlation of dh/dt would be zero.

These considerations make it clear that the case of an infinite interface is different from the case of a very large drop. It might be thought that to a very small particle near the large drop, the drop interface might cause the same effects as an infinite interface. This is not so, because the drop has translational degrees of freedom, and thus will diffuse, causing the small particle to diffuse, as shown above.

It is also necessary to take into account the fact that in any real physical situation to which this analysis would be applied, the interface would not be infinite, but rather, the fluid would have boundaries. Suppose the two fluids were in a container. If the contained had a fixed volume and were entirely filled with the fluids, then the interface could not shift over by a constant amount because that would violate incompressibility. Even if the container were open and the top surface were free (at atmospheric pressure) since the volume of the lower fluid is constant, the average absolute interface position would have to remain *constant.* This makes it clear that in such a situation the interface has no translational degrees of freedom. The only way that the interface could nevertheless give rise to a diffusivity of the small particle is if the transfer function referred to above as H_{flow} did not have to go to zero as $\omega \to 0$, i.e., in the steady limit. Above, it was stated that this function must always go to zero, but that was for an idealized infinite interface, where there are no boundary conditions, so there is always a solution to the problem of finding the interface shape to support any applied steady normal stress. For a finite interface in a container, there are boundary conditions, namely, that the interface must meet the container walls at the contact angle, which is a property of the fluids and wall material. If there were no *steady* solution to the problem of finding the interface shape to balance a given applied normal stress with these boundary conditions, then H_{flow} would not have to go to zero as ω does. Now it is known [13] that certain container shapes do not allow such a steady solution, for instance, containers having triangular cross sections. These nonexistence results are for the full nonlinear problem, however. It should be noted that in the above discussion it has been assumed that all displacements were small enough so that the equations (and in particular the curvature term in the surface tension force) could be linearized, so that linear response theory could be used. The nonexistence results may not carry over to the linearized case. This would mean either that solutions would always exist for the magnitude of normal stress associated with random fluctuations, or that solutions actually would not exist in some containers, but it could not be proved that this leads to a nonzero diffusivity using linear response theory. A nonlinear stochastic system would have to be solved. This could be very difficult.

None of these considerations change the essential observation that care must be taken in idealizing an interface as infinite, as the results may not even be meaningful qualitatively. The actual nature of the boundaries, even if they are far away, may need to be known. It was pointed out earlier that taking into account fluid inertia (the part involving the square of the velocity, not just the part with acceleration) might change the above results, since then there would be kinetic energy associated with steady interface motion. This could be done analytically by finding an asymptotic solution for small Reynolds number. As a final comment, it would also be interesting to see whether allowing the fluids to be slightly compressible would change the results qualitatively.

SECTION 2: AN ALTERNATIVE CALCULATION WITH A WEAKENED ASSUMPTION 1

It has been shown above that with Assumptions 1 and 2, the mean-square displacement of the particle asymptotes to a finite value as $t \to \infty$, rather than continues to grow at some rate slower than $\sqrt{t}$ (since its motion is not diffusive). If the force field the particle is placed in were white noise, the particle would definitely diffuse. Thus, the nature of the autocorelation of the force field must be such as to systematically tend to move the particle back right after it has pushed it out. Yang explains this in terms of the interface creating wavelike disturbances, with the particle essentially going around in circles. However, with Assumption 1, the fact is being neglected, that once the force field has pushed the particle away from its initial position, the particle now experiences the force at its *new* position. Even though an infinite time limit is being taken, so that loosely speaking the particle has plenty of opportunity to stray from its initial position, the force in the Langevin equation is still being evaluated at its initial position! It certainly seems reasonable to question whether the fact that the probability distribution for the particle position stops spreading after a very long time is perhaps just an artifact of this assumption.

It was mentioned earlier that there is an available technique for dealing with the Langevin equation with a weaker assumption than Assumption 1. If the force can be expanded in a convergent Taylor series about the particle's initial position, then a type of approximate solution can be found, by a perturbation method that is very popular with physicists working with quantum field theory. The method is explained in Bern's thesis [14], and by Abrikosov *et al.* [15] among undoubtedly many other references. The perturbation scheme can be carried out efficiently to a large number of terms by using the "diagram technique," in which lines stand for Green's functions; dots, crosses or other symbols for functions; and loops for autocorrelations (see Appendix 2 or [14]). While it may be necessary to use many terms to get a good approximation, what is mostly of interest here is the issue of whether using this method will qualitatively change the long-time behavior of the particle-position probability distribution. Will its mean square position now tend to infinity? Will it grow at a diffusive rate?

The scheme is set up as follows. First, expand the force in the Langevin equation about the particle's initial position $\mathbf{X}_0$:

$$\begin{aligned} m_P\frac{d\mathbf{U}}{dt} + \mathbf{B}(t)[\mathbf{U}] &= \mathbf{F}(\mathbf{x},t)\Big|_{\mathbf{x}=\mathbf{X}_0} \\ &= \mathbf{F}(\mathbf{X}_0,t) + \epsilon\nabla\mathbf{F}(\mathbf{X}_0,t)\cdot(\mathbf{X}-\mathbf{X}_0) \\ &\quad + \epsilon^2\nabla\nabla\mathbf{F}(\mathbf{X}_0,t) : (\mathbf{X}-\mathbf{X}_0)(\mathbf{X}-\mathbf{X}_0) + \dots \quad . \end{aligned} \tag{2.1}$$

Then look for a solution of the form

$$\mathbf{X}(t) = \mathbf{X}^{(1)} + \epsilon\mathbf{X}^{(2)} + \epsilon^2\mathbf{X}^{(3)} + \dots \quad . \tag{2.2}$$

Then by the usual procedure of collecting terms of each order in ϵ, the following equations are obtained:

$$\begin{aligned} m_P\frac{d^2\mathbf{X}^{(1)}}{dt^2} + B\left\{\frac{d\mathbf{X}^{(1)}(t)}{dt}\right\} &= \mathbf{F}(\mathbf{X}_0,t) \\ m_P\frac{d^2\mathbf{X}^{(2)}}{dt^2} + B\left\{\frac{d\mathbf{X}^{(2)}(t)}{dt}\right\} &= \nabla\mathbf{F}(\mathbf{X}_0,t)\cdot(\mathbf{X}^{(1)}-\mathbf{X}_0) \\ m_P\frac{d^2\mathbf{X}^{(3)}}{dt^2} + B\left\{\frac{d\mathbf{X}^{(3)}(t)}{dt}\right\} &= \nabla\mathbf{F}(\mathbf{X}_0,t)\cdot\mathbf{X}^{(2)} \\ &\quad + \nabla\nabla\mathbf{F}(\mathbf{X}_0,t) : (\mathbf{X}^{(1)}-\mathbf{X}_0)(\mathbf{X}^{(1)}-\mathbf{X}_0) \\ &\dots\dots \end{aligned} \tag{2.3}$$

These equations have to be solved for the $\mathbf{X}^{(i)}$ in terms of the force $\mathbf{F}$ at $\mathbf{X}_0$ (and its derivatives). Of course, what is actually *known* about $\mathbf{F}$ is the Fourier transform of its autocorrelation function. Thus, the solutions of these equations can only be used to get the autocorrelation of $\mathbf{X}$, which is what is desired to determine the diffusivity. The fact that all these equations are of the form

$$m_P \frac{d^2\mathbf{X}^{(i)}}{dt^2} + B\left\{\frac{d\mathbf{X}^{(i)}(t)}{dt}\right\} = \mathbf{f}^{(i)}(t), \tag{2.4}$$

where $\mathbf{f}^{(i)}$ is the forcing function on the right-hand side, will be exploited. Take the Fourier transform of this "generic" equation:

$$\left[-\omega^2 m_P - i\omega\hat{B}\right]\hat{\mathbf{X}}^{(i)} = \hat{\mathbf{f}}^{(i)}, \tag{2.5}$$

where $\hat{B}$ is the unsteady mobility operator in Fourier space. It is just a multiplicative operator in this space, whereas it is a functional in time. From this it is easy to get

$$\hat{\mathbf{X}}^{(i)} = \frac{\hat{\mathbf{f}}^{(i)}}{[-\omega^2 m_P - i\omega\hat{B}]}, \tag{2.6}$$

and the Fourier inversion formula now gives

$$\mathbf{X}^{(i)}(t) = \int_{-\infty}^{\infty} \frac{\hat{\mathbf{f}}^{(i)}}{[-\omega^2 m_P - i\omega\hat{B}]} e^{-i\omega t}\, d\omega. \tag{2.7}$$

These are the formulae to be used for solving the equations. From them, it is evident that

$$\hat{X}_j^{(2)} = \frac{1}{[-\omega^2 m_P - i\omega\hat{B}]}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\Big[ik_1\hat{F}_j(k_1,k_2,x_3,\omega)\hat{X}_1^{(1)}(\omega) + ik_2\hat{F}_j(k_1,k_2,x_3,\omega) + \frac{d}{dx_3}\hat{F}_j(k_1,k_2,x_3,\omega)[\hat{X}_3^{(1)}(\omega)-d]dk_1dk_2\Big|_{x_3=d}. \tag{2.8}$$

Now by noting that

$$\hat{U}_j^{(2)} = -i\omega\hat{X}_j^{(2)}, \tag{2.9}$$

it is clear that

$$< \hat{U}_j^{(2)}(\omega)\hat{U}_k^{(2)}(\omega') >$$
$$= \frac{1}{[-\omega^2 m_P - i\omega\hat{B}(\omega)]}\frac{1}{[-\omega'^2 m_P - i\omega'\hat{B}(\omega')]}$$
$$\times\Bigg\langle\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{1}{[-\omega^2 m_P - i\omega\hat{B}(\omega)]}$$
$$\cdot\Bigg\{ik_1\hat{F}_j(k_1,k_2,d,\omega)\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\hat{F}_1(k_1'',k_2'',d,\omega)dk_1''dk_2''$$
$$+ ik_2\hat{F}_j(k_1,k_2,d,\omega)\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\hat{F}_2(k_1'',k_2'',d,\omega)dk_1''dk_2''$$
$$+ \frac{d\hat{F}_j(k_1,k_2,d,\omega)}{dx_3}\Big|_{x_3=d}\Big[\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\hat{F}_1(k_1'',k_2'',d,\omega)dk_1''dk_2'' - d\Big]\Bigg\}dk_1dk_2$$
$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{1}{[-\omega'^2 m_P - i\omega'\hat{B}(\omega')]}$$
$$\cdot\Bigg\{ik_1'\hat{F}_j(k_1',k_2',d,\omega')\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\hat{F}_1(k_1''',k_2''',d,\omega')dk_1'''dk_2'''$$
$$+ ik_2\hat{F}_j(k_1',k_2',d,\omega')\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\hat{F}_2(k_1''',k_2''',d,\omega')dk_1'''dk_2'''$$
$$+ \frac{d\hat{F}_j(k_1,k_2,d,\omega')}{dx_3}\Big|_{x_3=d}\Big[\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\hat{F}_1(k_1''',k_2''',d,\omega)dk_1'''dk_2''' - d\Big]\Bigg\}$$

$$dk_1' dk_2' \Big\rangle \, . \tag{2.10}$$

This expression is expanded into 27 terms in Appendix 3. It will not be necessary to deal with all these terms to answer the question of whether including these terms as first corrections to the velocity autocorrelation will allow the particle to "escape" from the bounded region it is essentially confined in to first-order. It is only necessary to observe that the terms are all of the general form

$$\frac{1}{(i\omega)^2} Q(\omega) \int\int k_{i_1} < F_{i_2} F_{i_3} > \times \int\int k_{i_4} < F_{i_5} F_{i_6} >, \tag{2.11}$$

with the $i_1, i_2, \ldots$ being general indices. Here, $Q(\omega) \rightarrow$ constant as $\omega \rightarrow 0$. This is to be compared with the first-order approximation, which is of the form

$$\tilde{Q}(\omega) \int\int < F_{i_1} F_{i_2} > . \tag{2.12}$$

It will be recalled that this latter expression behaved like $\omega^{9/4}$ as $\omega \rightarrow 0$. If the asymptotic expansion of this integral for the first-order approximation, discussed at length earlier, is reexamined, and, in particular, if it is recalled that the variable k was changed to $\sqrt{\omega} u$ to make it possible to approximate the integrand uniformly, it is found that the behavior of the above generic correction term can be found by inspection to be

$$\frac{1}{\omega^2} \left(\omega^{9/4}\right)^2 \left(\sqrt{\omega}\right)^2 = \omega^{7/2}. \tag{2.13}$$

Here, the factor

$$\left(\omega^{9/4}\right)^2$$

comes from the fact that by the first-order calculation, integrals of $< FF >$ are known to behave like $\omega^{9/4}$, and the second-order terms look like $< FF >< FF >$ (hence the squaring). The factor $\left(\sqrt{\omega}\right)^2$ is present because there are now the two additional factors kk in the integrand, and the change of variables gives a factor $\sqrt{\omega}$ for each factor k. The asymptotic behavior $\omega^{7/2}$ of the Fourier-transformed velocity-autocorrelation function means that correcting for the fact that the random forcing changes with position does not cause the particle to undergo any sort of diffusion, normal *or* anomalous, at least under the assumption that the forcing is only a weak function of position.

SECTION 3: AN ALTERNATIVE CALCULATION WITHOUT ASSUMPTION 2

While it may seem reasonable at first glance to model the fluctuations using Assumption 2, namely, by putting only a random normal stress at the interface and a random force directly on the particle, in actuality there are fluctuations throughout the entire fluid. Moreover, there could be tangential as well as normal stresses at the interface. The discussion earlier about how motion of the particle due to interface fluctuations is limited by the fact that surface tension acts like a spring pulling the interface back was valid for fluctuations driven by normal stresses only. A fluid element on the interface is free to move infinitely far in a tangential direction; there is no "spring analogy" for motion driven by tangential stresses. Thus, it is interesting to consider how alternatives to Assumption 2 could change the particle behavior.

A classical finding of Landau and Lifshitz [11] is that the fluctuations in an infinite fluid are driven by a random stress tensor $\mathbf{s}$ with strength

$$< s_{ij}(\mathbf{x},t_1)s_{km}(\mathbf{x}',t_2) >= 2kT\mu\delta(\mathbf{x}-\mathbf{x}')\delta(t_1-t_2)\left[\delta_{ik}\delta_{jm}+\delta_{im}\delta_{jk}-\frac{2}{3}\delta_{ij}\delta_{km}\right]. \quad (3.1)$$

Hinch [2] has shown by applying a generalized Langevin analysis that for a rigid particle in an infinite fluid, the "correct" (i.e., Stokes-Einstein) diffusivity, *and* the "correct" (i.e., in agreement with simulations) velocity autocorrelation long-time tail can be retrieved if the fluctuations are modelled as driven by a random stress throughout the fluid, with the same correlation as (3.1). Therefore, in the present work, the calculation of the diffusivity will be redone using this random stress instead of only the normal stress at the interface. In comparing the calculation to be done below with Yang's approach of putting the driving random forcing only on the interface and the particle surface, it should be remembered that even though the particle will not be introduced in the calculation of the fluctuating velocity field here, since the random forcing is distributed *throughout* the fluid, some of that forcing corresponds to what was assigned to the particle surface by Yang. (This is just a consequence of the artificiality of Assumption 2, which puts all the forcing at the boundaries.)

The strategy will be first to calculate the random flow field assuming there is a planar interface, but no particle. Then the particle will be placed in this flow field, and its motion will be calculated as if there is no interface (except insofar as it generates the flow field). In this respect the calculation will be done just as in Yang's thesis, except, of course, that the random flow field will be different as discussed above.

Thus, for the first step of the calculation, the equations of motion will be

$$S\alpha \frac{\partial \mathbf{u}^{(1)}}{\partial t} = -\nabla p^{(1)} + \lambda \nabla^2 \mathbf{u}^{(1)} + \nabla \cdot \mathbf{s}^{(1)}, \tag{3.2a}$$

$$\nabla \cdot \mathbf{u}^{(1)} = 0, \tag{3.2b}$$

for fluid 1 (occupying $x_3 \geq 0$); and for fluid 2 (occupying $x_3 \leq 0$),

$$S \frac{\partial \mathbf{u}^{(2)}}{\partial t} = -\nabla p^{(2)} + \nabla^2 \mathbf{u}^{(2)} + \nabla \cdot \mathbf{s}^{(2)}, \tag{3.3a}$$

$$\nabla \cdot \mathbf{u}^{(2)} = 0. \tag{3.3b}$$

Here $\alpha = \rho_1/\rho_2$ is the density ratio of the two fluids, and $\lambda = \mu_1/\mu_2$ is the viscosity ratio. The equations have been nondimensionalized, with the characteristic velocity taken to be $kT/L^2\mu$, the characteristic stress and pressure kT/L^3, and the characteristic length L (this is arbitrary; there is no length scale inherent in this problem until the wavelength of the interface disturbances is specified). $S = \rho_2 kT/L\mu_2^2$ is a Strouhal number. The tensor $\mathbf{s}$ is the random stress of (3.1). The boundary conditions at the planar interface $x_3 = \eta(x_1, x_2)$ are the kinematic conditions

$$\mathbf{u}^{(1)} \cdot \mathbf{n} = \mathbf{u}^{(2)} \cdot \mathbf{n} = \frac{\partial \eta}{\partial t}; \qquad (3.4a)$$

continuity of tangential velocity

$$\mathbf{u}^{(1)} \cdot \mathbf{t} = \mathbf{u}^{(2)} \cdot \mathbf{t}; \qquad (3.4b)$$

continuity of tangential stress

$$\lambda \mathbf{t}\cdot\mathbf{n}\cdot\left[\nabla\mathbf{u}^{(1)} + (\nabla\mathbf{u}^{(1)})^T\right] + \mathbf{t}\cdot\mathbf{n}\cdot\mathbf{s}^{(1)} = \mathbf{t}\cdot\mathbf{n}\cdot\left[\nabla\mathbf{u}^{(2)} + (\nabla\mathbf{u}^{(2)})^T\right] + \mathbf{t}\cdot\mathbf{n}\cdot\mathbf{s}^{(2)}, \qquad (3.4c)$$

where $\mathbf{t}$ is either of two independent unit vectors tangent to the interface; and finally, the normal stress condition

$$-p^{(1)} + \lambda \mathbf{n}\cdot\mathbf{n}\cdot\left[\nabla\mathbf{u}^{(1)} + (\nabla\mathbf{u}^{(1)})^T + \mathbf{s}^{(1)}\right] + p^{(2)} - \mathbf{n}\cdot\mathbf{n}\cdot\left[\nabla\mathbf{u}^{(2)} + (\nabla\mathbf{u}^{(2)})^T + \mathbf{s}^{(2)}\right]$$
$$= \Gamma\nabla\cdot\mathbf{n} + \beta\eta. \qquad (3.4d)$$

Here, $\Gamma = \gamma L^2/kT$ is a form of the reciprocal Capillary number, with γ being the surface tension, and $\beta = (\rho_2 - \rho_1)gL^4/kT$ is a dimensionless measure of the gravitational force tending to restore the interface to its flat state.

Now Fourier transform these equations and boundary conditions according to

$$\hat{v} = \frac{1}{(2\pi)^3}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} v(\mathrm{x},t)\exp[i(-k_1x_1 - k_2x_2 + i\omega t)]dx_1dx_2dt; \qquad (3.5a)$$

$$v = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \hat{v}(k_1,k_2,x_3,\omega)\exp[i(k_1x_1 + k_2x_2 - \omega t)]dk_1dk_2d\omega, \qquad (3.5b)$$

where v stands for any of the variables $\mathrm{u}^{(i)}, p^{(i)}, \mathrm{s}^{(i)}$, with $i = 1,2$. The equations (3.2) become, for fluid 1,

$$\begin{aligned} -i\omega S\alpha\hat{u}_1^{(1)} &= -ik_1\hat{p}^{(1)} + \lambda\Big(\frac{d^2}{dx_3^2} - k_1^2 - k_2^2\Big)\hat{u}_1^{(1)} \\ &\quad + ik_1\hat{s}_{11}^{(1)} + ik_2\hat{s}_{21}^{(1)} + \frac{d}{dx_3}\hat{s}_{31}^{(1)}; \end{aligned} \qquad (3.6a)$$

$$\begin{aligned} -i\omega S\alpha\hat{u}_2^{(1)} &= -ik_2\hat{p}^{(1)} + \lambda\Big(\frac{d^2}{dx_3^2} - k_1^2 - k_2^2\Big)\hat{u}_2^{(1)} \\ &\quad + ik_1\hat{s}_{12}^{(1)} + ik_2\hat{s}_{22}^{(1)} + \frac{d}{dx_3}\hat{s}_{32}^{(1)}; \end{aligned} \qquad (3.6b)$$

$$\begin{aligned} -i\omega S\alpha\hat{u}_3^{(1)} &= -\frac{d}{dx_3}\hat{p}^{(1)} + \lambda\Big(\frac{d^2}{dx_3^2} - k_1^2 - k_2^2\Big)\hat{u}_3^{(1)} \\ &\quad + ik_1\hat{s}_{13}^{(1)} + ik_2\hat{s}_{23}^{(1)} + \frac{d}{dx_3}\hat{s}_{33}^{(1)}; \end{aligned} \qquad (3.6c)$$

$$ik_1\hat{u}_1^{(1)} + ik_2\hat{u}_2^{(1)} + \frac{d}{dx_3}\hat{u}_3^{(1)}, \qquad (3.6d)$$

with the same equations holding for Fluid 2, if α and λ are set equal to 1, and the superscript "(1)" is replaced by "(2)."

Equations for the pressures $p^{(1)}$ and $p^{(2)}$ can be obtained by using continuity to eliminate the velocity. Taking ik_1 times Equation (3.6a), plus ik_2 times (3.6b), plus $\frac{d}{dx_3}$ of (3.6c) gives

$$\begin{aligned} &-(ik_1)^2\hat{p}^{(1)} - (ik_2)^2\hat{p}^{(1)} - \frac{d^2}{dx_3^2}\hat{p}^{(1)} \\ &+ik_1\left[ik_1\hat{s}_{11}^{(1)} + ik_2\hat{s}_{21}^{(1)} + \frac{d}{dx_3}\hat{s}_{31}^{(1)}\right] \\ &+ik_2\left[ik_1\hat{s}_{12}^{(1)} + ik_2\hat{s}_{22}^{(1)} + \frac{d}{dx_3}\hat{s}_{32}^{(1)}\right] \\ &+\frac{d}{dx_3}\left[ik_1\hat{s}_{13}^{(1)} + ik_2\hat{s}_{23}^{(1)} + \frac{d}{dx_3}\hat{s}_{33}^{(1)}\right] = 0, \end{aligned} \tag{3.7}$$

which can be rewritten as

$$\frac{d^2\hat{p}^{(1)}}{dx_3^2} - c_p\hat{p}^{(1)} = q^{(1)}, \tag{3.8}$$

where $c_p = k_1^2 + k_2^2$, and

$$\begin{aligned} q^{(1)} = &\; ik_1\left[ik_1\hat{s}_{11}^{(1)} + ik_2\hat{s}_{21}^{(1)} + \frac{d}{dx_3}\hat{s}_{31}^{(1)}\right] \\ &+ik_2\left[ik_1\hat{s}_{12}^{(1)} + ik_2\hat{s}_{22}^{(1)} + \frac{d}{dx_3}\hat{s}_{32}^{(1)}\right] \\ &+\frac{d}{dx_3}\left[ik_1\hat{s}_{13}^{(1)} + ik_2\hat{s}_{23}^{(1)} + \frac{d}{dx_3}\hat{s}_{33}^{(1)}\right]. \end{aligned} \tag{3.9}$$

Similarly, the equations for the velocity can be rewritten as

$$\frac{d^2\hat{u}_1^{(1)}}{dx_3^2} - c_1\hat{u}_1^{(1)} = r_1^{(1)}, \tag{3.10a}$$

$$\frac{d^2\hat{u}_2^{(1)}}{dx_3^2} - c_1\hat{u}_2^{(1)} = r_2^{(1)}, \tag{3.10b}$$

$$\frac{d^2\hat{u}_3^{(1)}}{dx_3^2} - c_1\hat{u}_3^{(1)} = r_3^{(1)}, \tag{3.10c}$$

with

$$c_1 = k_1^2 + k_2^2 - \frac{i\omega S\alpha}{\lambda}, \tag{3.11a}$$

and the same equations hold for the components of $\hat{u}^{(2)}$, with the superscript (1) replaced by (2), λ and α set equal to one, and c_1 replaced by

$$c_2 = k_1^2 + k_2^2 - i\omega S. \tag{3.11b}$$

All these equations have the form

$$\frac{d^2u}{dx^2} - cu = r, \tag{3.12}$$

which has the general solution

$$\begin{aligned} u(x) = e^{-\sqrt{c}x} \int_0^x e^{2\sqrt{c}y} \int_0^y r(z)e^{-\sqrt{c}z}dzdy \\ + ae^{\sqrt{c}x} + be^{-\sqrt{c}x}, \end{aligned} \tag{3.13}$$

where a and b, which appear in the homogeneous solution, are arbitrary constants (determined by boundary conditions). The convention will be taken in the following that $\sqrt{\cdot}$ always stands for the *positive* branch. From this it is clear that the solutions are

$$\hat{p}^{(1)} = e^{-\sqrt{c_p}x_3}\int_0^{x_3} e^{2\sqrt{c_p}y}\int_0^{y} q^{(1)}(z)e^{-\sqrt{c_p}z}dzdy$$
$$A_1' e^{\sqrt{c_p}x_3} + \quad A_1 e^{-\sqrt{c_p}x_3}; \tag{3.14a}$$

$$\hat{u}_1^{(1)} = e^{-\sqrt{c_1}x_3}\int_0^{x_3} e^{2\sqrt{c_1}y}\int_0^{y} r_1^{(1)}(z)e^{-\sqrt{c_1}z}dzdy$$
$$+ \quad A_2' e^{\sqrt{c_1}x_3} \quad + \quad A_2 e^{-\sqrt{c_1}x_3}; \tag{3.14b}$$

$$\hat{u}_2^{(1)} = e^{-\sqrt{c_1}x_3}\int_0^{x_3} e^{2\sqrt{c_1}y}\int_0^{y} r_2^{(1)}(z)e^{-\sqrt{c_1}z}dzdy$$
$$+ \quad A_3' e^{\sqrt{c_1}x_3} \quad + \quad A_3 e^{-\sqrt{c_1}x_3}; \tag{3.14c}$$

$$\hat{u}_3^{(1)} = e^{-\sqrt{c_1}x_3}\int_0^{x_3} e^{2\sqrt{c_1}y}\int_0^{y} r_3^{(1)}(z)e^{-\sqrt{c_1}z}dzdy$$
$$+ \quad A_4' e^{\sqrt{c_1}x_3} \quad + \quad A_4 e^{-\sqrt{c_1}x_3}, \tag{3.14d}$$

and similarly for the pressure and velocity in fluid 2, if the superscript (1) is replaced by (2), and c_1 is replaced by c_2, and the constants A_i, A_i' are replaced by B_i, B_i' for $i = 1, 2, 3, 4$. Here, the quantities r have been defined for convenience by

$$r_1^{(1)} = \frac{ik_1}{\lambda}\hat{p}^{(1)} - \frac{1}{\lambda}\Big[ik_1\hat{s}_{11}^{(1)} + ik_2\hat{s}_{21}^{(1)} + \frac{d}{dx_3}\hat{s}_{31}^{(1)}\Big]; \tag{3.15a}$$

$$r_2^{(1)} = \frac{ik_2}{\lambda}\hat{p}^{(1)} - \frac{1}{\lambda}\Big[ik_1\hat{s}_{12}^{(1)} + ik_2\hat{s}_{22}^{(1)} + \frac{d}{dx_3}\hat{s}_{32}^{(1)}\Big]; \tag{3.15b}$$

$$r_3^{(1)} = \frac{d\hat{p}^{(1)}}{dx_3} - \frac{1}{\lambda}\Big[ik_1\hat{s}_{13}^{(1)} + ik_2\hat{s}_{23}^{(1)} + \frac{1}{\lambda}\frac{d}{dx_3}\hat{s}_{33}^{(1)}\Big]. \tag{3.15c}$$

The constants A_i, A_i', B_i, B_i' are to be determined from the boundary conditions (3.4). Although the boundary conditions are in general nonlinear, only the first linear approximation for small flow and small deformation is sought here. Thus, the terms appearing in the boundary conditions, which are evaluated at $x_3 = \eta$, are written as Taylor-series expansions about $x_3 = 0$. Since only the first approximation is desired, it is easily verified that only the first term in the Taylor series must be retained; i.e., the terms are evaluated at $x_3 = 0$. The interface deformation appears only in the linearized surface tension and gravity terms of the normal stress condition, and in the right-hand-side of the kinematic condition. Thus, the kinematic conditions (3.4a) become

$$u_3^{(1)}\Big|_{x_3=0} = u_3^{(2)}\Big|_{x_3=0} = \frac{\partial \eta}{\partial t}. \tag{3.16}$$

After Fourier-transforming, this becomes

$$\hat{u}_3^{(1)}\Big|_{x_3=0} = \hat{u}_3^{(2)}\Big|_{x_3=0} = -i\omega\hat{\eta}. \tag{3.17}$$

Substituting (3.14d) into this gives

$$A_4 + A_4' = B_4 + B_4' = -i\omega\eta. \tag{3.18}$$

Now consider the conditions of continuity of tangential velocity (3.4b). To first order, they are

$$u_1^{(1)}\Big|_{x_3=0} = u_1^{(2)}\Big|_{x_3=0}; \tag{3.19a}$$

$$u_2^{(1)}\Big|_{x_3=0} = u_2^{(2)}\Big|_{x_3=0}; \tag{3.19b}$$

and after Fourier-transforming they become

$$\hat{u}_1^{(1)}\Big|_{x_3=0} = \hat{u}_1^{(2)}\Big|_{x_3=0}; \tag{3.20a}$$

$$\hat{u}_2^{(1)}\Big|_{x_3=0} = \hat{u}_2^{(2)}\Big|_{x_3=0}. \tag{3.20b}$$

Substituting (3.14b,c) into these gives

$$A_2 + A_2' = B_2 + B_2'; \tag{3.21a}$$

$$A_3 + A_3' = B_3 + B_3'. \tag{3.21b}$$

Continuity of tangential stress (3.4c) implies

$$\lambda \frac{\partial u_3^{(1)}}{\partial x_1}\Big|_{x_3=0} + \lambda \frac{\partial u_1^{(1)}}{\partial x_3}\Big|_{x_3=0} + s_{13}^{(1)}\Big|_{x_3=0} = \frac{\partial u_3^{(2)}}{\partial x_1}\Big|_{x_3=0} + \frac{\partial u_1^{(2)}}{\partial x_3}\Big|_{x_3=0} + s_{13}^{(2)}\Big|_{x_3=0}; \tag{3.22a}$$

and

$$\lambda \frac{\partial u_3^{(1)}}{\partial x_2}\Big|_{x_3=0} + \lambda \frac{\partial u_2^{(1)}}{\partial x_3}\Big|_{x_3=0} + s_{23}^{(1)}\Big|_{x_3=0} = \frac{\partial u_3^{(2)}}{\partial x_2}\Big|_{x_3=0} + \frac{\partial u_2^{(2)}}{\partial x_3}\Big|_{x_3=0} + s_{23}^{(2)}\Big|_{x_3=0}. \tag{3.22b}$$

These Fourier-transform to

$$ik_1\lambda\hat{u}_3^{(1)}\Big|_{x_3=0} + \lambda\frac{d\hat{u}_1^{(1)}}{dx_3}\Big|_{x_3=0} + \hat{s}_{13}^{(1)}(0) = \hat{u}_3^{(2)}\Big|_{x_3=0} + \frac{d\hat{u}_1^{(2)}}{dx_3}\Big|_{x_3=0} + \hat{s}_{13}^{(2)}(0); \tag{3.23a}$$

and

$$ik_2\lambda\hat{u}_3^{(1)}\Big|_{x_3=0} + \lambda\frac{d\hat{u}_2^{(1)}}{dx_3}\Big|_{x_3=0} + \hat{s}_{23}^{(1)} = ik_2\hat{u}_3^{(2)}\Big|_{x_3=0} + \frac{d\hat{u}_2^{(2)}}{dx_3}\Big|_{x_3=0} + \hat{s}_{23}^{(2)}, \qquad (3.23b)$$

and substitution of (3.14b,c,d) into them gives

$$ik_1\lambda A_4' + ik_1\lambda A_4 + \sqrt{c_1}\lambda A_2' - \sqrt{c_1}\lambda A_2 + \hat{s}_{13}^{(1)}(0)$$
$$= ik_1 B_4' + ik_1 B_4 - \sqrt{c_2}B_2' + \sqrt{c_2}B_2 + \hat{s}_{13}^{(2)}(0), \qquad (3.24a)$$

and

$$ik_2\lambda A_4 + ik_2\lambda A_4' + \sqrt{c_1}\lambda A_3' - \sqrt{c_1}\lambda A_3 + \hat{s}_{23}^{(1)}(0)$$
$$= ik_2 B_4 + ik_2 B_4' - \sqrt{c_2}B_3' + \sqrt{c_2}B_3 + \hat{s}_{23}^{(2)}(0). \qquad (3.24b)$$

The last boundary condition is the normal stress condition (3.4d), which to first order is

$$-p^{(1)}\Big|_{x_3=0} + \lambda\frac{\partial u_3^{(1)}}{\partial x_3}\Big|_{x_3=0} + p^{(2)}\Big|_{x_3=0} - \frac{\partial u_3^{(2)}}{\partial x_3}\Big|_{x_3=0} + s_{33}^{(1)}\Big|_{x_3=0} - s_{33}^{(2)}\Big|_{x_3=0}$$
$$= -\Gamma\left[\frac{\partial^2\eta}{\partial x_1^2} + \frac{\partial^2\eta}{\partial x_2^2}\right] + \beta\eta. \qquad (3.25)$$

This Fourier-transforms to

$$-\hat{p}^{(1)}\Big|_{x_3=0} + \lambda\frac{d\hat{u}_3^{(1)}}{dx_3}\Big|_{x_3=0} + \hat{p}^{(2)}\Big|_{x_3=0} - \frac{d\hat{u}_3^{(2)}}{dx_3}\Big|_{x_3=0} + \hat{s}_{33}^{(1)}(0) - \hat{s}_{33}^{(2)}(0)$$
$$= -\Gamma\Big[(ik_1)^2 + (ik_2)^2\Big]\hat{\eta} + \beta\hat{\eta}. \qquad (3.26)$$

Substitution of (3.14d) into this gives

$$-A_1+\sqrt{c_1}\lambda A_4'-\sqrt{c_1}\lambda A_4+B_1+\sqrt{c_2}B_4'-\sqrt{c_2}B_4+\hat{s}_{33}^{(1)}(0)-\hat{s}_{33}^{(2)}(0)=\Gamma(k_1^2+k_2^2)\hat{\eta}+\beta\hat{\eta}. \tag{3.27}$$

Thus, the boundary conditions provide 7 equations, namely, (3.18), (3.21), (3.24), (3.27), for determining the 17 unknowns $A_i,\ \tilde{A}_i,\ B_i,\ \tilde{B}_i,\ i=1,2,3,4$ and $\hat{\eta}$, all of which are, of course, functions of the transformed variables $\omega,\ k_1, k_2$. Ten additional conditions come from the continuity equation and from the requirement that the flow should not grow exponentially in fluid 1 as $x_3 \to \infty$, and in fluid 2 as $x_3 \to -\infty$. Two of these conditions are immediate: Clearly, $A_1' = 0$ and $B_1' = 0$, so that the homogeneous pressure field doesn't increase exponentially. To derive the remaining conditions, it will be convenient first to rewrite the general solutions for the velocity field. First, by substituting the solution (3.14a) for the pressure into Equations (3.15), the following is obtained:

$$r_1^{(1)} = \frac{ik_1}{\lambda}\left[e^{-\sqrt{c_p}x_3}\int_0^{x_3} e^{2\sqrt{c_p}y}\int_0^{y} q^{(1)}(z)e^{-\sqrt{c_p}z}\,dz\,dy + A_1 e^{-\sqrt{c_p}x_3}\right] - \frac{1}{\lambda}\left[ik_1\hat{s}_{11}^{(1)} + ik_2\hat{s}_{21}^{(1)} + \frac{d}{dx_3}\hat{s}_{31}^{(1)}\right]; \tag{3.28a}$$

$$r_2^{(1)} = \frac{ik_2}{\lambda}\left[e^{-\sqrt{c_p}x_3}\int_0^{x_3} e^{2\sqrt{c_p}y}\int_0^{y} q^{(1)}(z)e^{-\sqrt{c_p}z}\,dz\,dy + A_1 e^{-\sqrt{c_p}x_3}\right] - \frac{1}{\lambda}\left[ik_1\hat{s}_{12}^{(1)} + ik_2\hat{s}_{22}^{(1)} + \frac{d}{dx_3}\hat{s}_{32}^{(1)}\right]; \tag{3.28b}$$

$$r_3^{(1)} = \frac{d}{dx_3}\left[e^{-\sqrt{c_p}x_3}\int_0^{x_3} e^{2\sqrt{c_p}y}\int_0^{y} q^{(1)}(z)e^{-\sqrt{c_p}z}\,dz\,dy + A_1 e^{-\sqrt{c_p}x_3}\right]$$

$$-\frac{1}{\lambda}\left[ik_1\hat{s}_{13}^{(1)} + ik_2\hat{s}_{23}^{(1)} + \frac{d}{dx_3}\hat{s}_{33}^{(1)}\right]. \tag{3.28c}$$

The continuity conditions will involve only the homogeneous parts of the solutions (i.e., the parts involving the unknown "constants" A_i, B_i, A_i', B_i', as opposed to the parts involving the stress field s). Use the letter w to denote this homogeneous part of the velocity field. Then it is clear from (3.14b,c,d) and (3.28) that

$$\begin{aligned}\hat{w}_1^{(1)} &= e^{-\sqrt{c_1}x_3}\int_0^{x_3} e^{2\sqrt{c_1}y}\int_0^{y}\frac{ik_1}{\lambda}\left[A_1e^{-\sqrt{c_p}z}\right]e^{-\sqrt{c_1}z}dzdy \\ &+ \quad A_2'e^{\sqrt{c_1}x_3} + A_2e^{-\sqrt{c_1}x_3};\end{aligned} \tag{3.29a}$$

$$\begin{aligned}\hat{w}_2^{(1)} &= e^{-\sqrt{c_1}x_3}\int_0^{x_3} e^{2\sqrt{c_1}y}\int_0^{y}\frac{ik_1}{\lambda}\left[A_1e^{-\sqrt{c_p}z}\right]e^{-\sqrt{c_1}z}dzdy \\ &+ \quad A_3'e^{\sqrt{c_1}x_3} + A_3e^{-\sqrt{c_1}x_3};\end{aligned} \tag{3.29b}$$

$$\begin{aligned}\hat{w}_3^{(1)} &= e^{-\sqrt{c_1}x_3}\int_0^{x_3} e^{2\sqrt{c_1}y}\int_0^{y}\frac{1}{\lambda}\frac{d}{dz}\left[A_1e^{-\sqrt{c_p}z}\right]e^{-\sqrt{c_1}z}dzdy \\ &+ \quad A_4'e^{\sqrt{c_1}x_3} + A_4e^{-\sqrt{c_1}x_3}.\end{aligned} \tag{3.29c}$$

The integrals in these equations can be evaluated to give

$$\begin{aligned}\hat{w}_1^{(1)} = &-\frac{ik_1A_1}{\lambda(c_1-c_p)}\left(e^{-\sqrt{c_p}x_3} - e^{-\sqrt{c_1}x_3}\right) \\ &+ \frac{ik_1A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_p})}\left(e^{\sqrt{c_1}x_3} - e^{-\sqrt{c_1}x_3}\right) \\ &+ A_2'e^{\sqrt{c_1}x_3} + A_2e^{-\sqrt{c_1}x_3};\end{aligned} \tag{3.30a}$$

$$\hat{w}_2^{(1)} = -\frac{ik_2 A_1}{\lambda(c_1 - c_p)}\left(e^{-\sqrt{c_p}x_3} - e^{-\sqrt{c_1}x_3}\right) + \frac{ik_2 A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_p})}\left(e^{\sqrt{c_1}x_3} - e^{-\sqrt{c_1}x_3}\right) + A_3' e^{\sqrt{c_1}x_3} + A_3 e^{-\sqrt{c_1}x_3}; \tag{3.30b}$$

$$\hat{w}_3^{(1)} = \frac{\sqrt{c_p}A_1}{\lambda(c_1 - c_p)}\left(e^{-\sqrt{c_p}x_3} - e^{-\sqrt{c_1}x_3}\right) - \frac{\sqrt{c_p}A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_p})}\left(e^{\sqrt{c_1}x_3} - e^{-\sqrt{c_1}x_3}\right) + A_4' e^{\sqrt{c_1}x_3} + A_4 e^{-\sqrt{c_1}x_3}. \tag{3.30c}$$

This time, the corresponding equations for the flow in fluid 2 *will* be given explicitly, since they differ from those in fluid 1 in a less obvious way:

$$\hat{w}_1^{(2)} = e^{-\sqrt{c_2}x_3}\int_0^{x_3} e^{2\sqrt{c_2}y}\int_0^{y} ik_1\left[B_1 e^{\sqrt{c_p}z}\right]e^{-\sqrt{c_2}z}\,dz\,dy + B_2 e^{\sqrt{c_2}x_3} + B_2' e^{-\sqrt{c_2}x_3}; \tag{3.31a}$$

$$\hat{w}_2^{(2)} = e^{-\sqrt{c_2}x_3}\int_0^{x_3} e^{2\sqrt{c_2}y}\int_0^{y} ik_1\left[B_1 e^{\sqrt{c_p}z}\right]e^{-\sqrt{c_2}z}\,dz\,dy + B_3 e^{\sqrt{c_2}x_3} + B_3' e^{-\sqrt{c_2}x_3}; \tag{3.31b}$$

$$\hat{w}_3^{(2)} = e^{-\sqrt{c_2}x_3}\int_0^{x_3} e^{2\sqrt{c_2}y}\int_0^{y} \frac{d}{dz}\left[B_1 e^{\sqrt{c_p}z}\right]e^{-\sqrt{c_2}z}\,dz\,dy + B_4 e^{\sqrt{c_2}x_3} + B_4' e^{-\sqrt{c_2}x_3}. \tag{3.31c}$$

Integration gives

$$\begin{aligned}\hat{w}_1^{(2)} = & \frac{ik_1 B_1}{(c_p - c_2)}\left(e^{\sqrt{c_p}x_3} - e^{-\sqrt{c_2}x_3}\right) \\ & - \frac{ik_1 B_1}{2\sqrt{c_2}(\sqrt{c_p} - \sqrt{c_2})}\left(e^{\sqrt{c_2}x_3} - e^{-\sqrt{c_2}x_3}\right) \\ & + B_2 e^{\sqrt{c_2}x_3} + B_2' e^{-\sqrt{c_2}x_3};\end{aligned} \tag{3.32a}$$

$$\begin{aligned}\hat{w}_2^{(2)} = & -\frac{ik_2 B_1}{(c_p - c_2)}\left(e^{\sqrt{c_p}x_3} - e^{-\sqrt{c_2}x_3}\right) \\ & - \frac{ik_2 B_1}{2\sqrt{c_2}(\sqrt{c_p} - \sqrt{c_2})}\left(e^{\sqrt{c_2}x_3} - e^{-\sqrt{c_2}x_3}\right) \\ & + B_3 e^{\sqrt{c_2}x_3} + B_3' e^{-\sqrt{c_2}x_3};\end{aligned} \tag{3.32b}$$

$$\begin{aligned}\hat{w}_3^{(2)} = & \frac{\sqrt{c_p} B_1}{(c_p - c_2)}\left(e^{\sqrt{c_p}x_3} - e^{-\sqrt{c_2}x_3}\right) \\ & - \frac{\sqrt{c_p} B_1}{2\sqrt{c_2}(\sqrt{c_p} - \sqrt{c_2})}\left(e^{\sqrt{c_2}x_3} - e^{-\sqrt{c_2}x_3}\right) \\ + \quad & B_4 e^{\sqrt{c_2}x_3} + B_4' e^{-\sqrt{c_2}x_3}.\end{aligned} \tag{3.32c}$$

From the fact that the homogeneous velocity solution should not grow exponentially at infinity, it is now immediately clear that

$$A_2' = -\frac{ik_1 A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_p})}; \tag{3.33a}$$

$$A_3' = -\frac{ik_2 A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_p})}; \tag{3.33b}$$

$$A_4' = \frac{\sqrt{c_p}A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_p})}; \tag{3.33c}$$

and similarly,

$$B_2' = \frac{ik_1B_1}{2\sqrt{c_2}(\sqrt{c_2}+\sqrt{c_p})}; \tag{3.34a}$$

$$B_3' = \frac{ik_2B_1}{2\sqrt{c_2}(\sqrt{c_2}+\sqrt{c_p})}; \tag{3.34b}$$

$$B_4' = \frac{\sqrt{c_p}B_1}{2\sqrt{c_2}(\sqrt{c_2}+\sqrt{c_p})}. \tag{3.34c}$$

Now impose the continuity requirement on the homogeneous velocity solution:

$$ik_1\hat{w}_1^{(1)} + ik_2\hat{w}_2^{(1)} + \frac{d\hat{w}_3^{(1)}}{dx_3} = 0, \tag{3.35}$$

and similarly for fluid 2, which gives

$$ik_1A_2 + ik_2A_3 - \sqrt{c_1}A_4 \tag{3.36a}$$

$$+\Big[-\frac{c_P}{\lambda(c_1-c_P)} + \frac{c_P}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_P})} + \frac{\sqrt{c_P}}{2\lambda(\sqrt{c_1}+\sqrt{c_P})}\Big]A_1 = 0; \tag{3.36b}$$

$$ik_1B_2 + ik_2B_3 + \sqrt{c_2}B_4; \tag{3.36c}$$

$$+\Big[\frac{c_P - \sqrt{c_p}\sqrt{c_2}}{2\sqrt{c_2}(\sqrt{c_p}-\sqrt{c_2})}\Big]B_1 = 0. \tag{3.36d}$$

Now all the 17 equations determining A_i, A_i', B_i, B_i' $(i = 1,2,3,4)$ and $\hat{\eta}$ have been derived. Since they are scattered throughout the above text, they are collected for convenience in Appendix 4.

The solution to this system of 17 algebraic equations is given in Appendix 5, since it is quite cumbersome. At this point the solution for the fluid velocity field is fully known in terms of the imposed random stress field. However, what is actually of interest is the velocity autocorrelation. From Appendix 5 it is evident that the velocity autocorrelation will involve not only terms like

$$< s_{ij}(k_1, k_2, x_3, t) s_{km}(k_1', k_2', x_3', t') > \tag{3.37}$$

appearing inside three-dimensional integrals, for Equation (3.1) can be used, but also terms like

$$< s_{ij}(k_1, k_2, 0, t) s_{km}(k_1', k_2', 0, t') >, \tag{3.38}$$

(which arise because of the boundary conditions at $x_3 = 0$) appearing inside *two* dimensional integrals. If (3.1), which has a *three*-dimensional delta function, were substituted for these, the integral would formally give infinity, the value of the delta function when its argument is zero, because the range of integration would include the point where $k_1 = k_1'$ and $k_2 = k_2'$. Of course, in actuality the stress field has a finite but macroscopically very small length scale, say L_a, over which it has a nonzero autocorrelation. Equation (3.1) holds on *macroscopic* length scales over which L_a can be approximated by zero. Thus, the delta function is only an approximation, and the real function has a finite height at zero. This height cannot be obtained without some further manipulations, however, since the coefficient in (3.1) gives only the area under the function. The way to evaluate terms like (3.38) will now be described.

The desired autocorrelations, of the general form of Equation (3.38), are all of stress components *at the interface*, so instead of working with all components of s, only components of $\mathbf{n}\cdot\mathbf{s}$ will be considered. To do this, it is first helpful to rewrite the desired autocorrelations, which are of Fourier-transformed variables, to show explicitly the transform integrals:

$$< \mathbf{n}\cdot\mathbf{s}(k_1,k_2,0,t)\mathbf{n}\cdot\mathbf{s}(k_1',k_2',0,t') >$$

$$= \left\langle \int_{x_1=-\infty}^{\infty}\int_{x_2=-\infty}^{\infty} \mathbf{n}\cdot\mathbf{s}(x_1,x_2,0,t)e^{i(k_1x_1+k_2x_2)}dx_1dx_2 \right.$$

$$\left. \int_{x_1'=-\infty}^{\infty}\int_{x_2'=-\infty}^{\infty} \mathbf{n}\cdot\mathbf{s}(x_1',x_2',0,t')e^{i(k_1'x_1'+k_2'x_2')}dx_1'dx_2' \right\rangle. \quad (3.39)$$

To the order of approximation here, the interface is the plane $x_3 = 0$, so that the integrals here are over the surface that has $\mathbf{e}_3$ as unit normal. Thus, the divergence theorem can be applied, to give

$$< \mathbf{n}\cdot\mathbf{s}^{(1)}(k_1,k_2,0,t)\mathbf{n}\cdot\mathbf{s}^{(1)}(k_1',k_2',0,t') >$$

$$= \left\langle \int_{x_1=-\infty}^{\infty}\int_{x_2=-\infty}^{\infty}\int_{x_3=0}^{\infty} \nabla\cdot[\mathbf{s}^{(1)}(x_1,x_2,x_3,t)e^{i(k_1x_1+k_2x_2)}]dx_1dx_2dx_3 \right.$$

$$\left. \int_{x_1'=-\infty}^{\infty}\int_{x_2'=-\infty}^{\infty}\int_{x_3'=0}^{\infty} \nabla\cdot[\mathbf{s}^{(1)}(x_1',x_2',x_3',t')e^{i(k_1'x_1'+k_2'x_2')}]dx_1'dx_2'dx_3' \right\rangle, \quad (3.40a)$$

for Fluid 1, and

$$< \mathbf{n}\cdot\mathbf{s}^{(2)}(k_1,k_2,0,t)\mathbf{n}\cdot\mathbf{s}^{(2)}(k_1',k_2',0,t') >$$

$$= \left\langle \int_{x_1=-\infty}^{\infty}\int_{x_2=-\infty}^{\infty}\int_{x_3=-\infty}^{0} \nabla\cdot[\mathbf{s}^{(2)}(x_1,x_2,x_3,t)e^{i(k_1x_1+k_2x_2)}]dx_1dx_2dx_3 \right.$$

$$\left. \int_{x_1'=-\infty}^{\infty}\int_{x_2'=-\infty}^{\infty}\int_{x_3'=-\infty}^{0} \nabla\cdot[\mathbf{s}^{(2)}(x_1',x_2',x_3',t')e^{i(k_1'x_1'+k_2'x_2')}]dx_1'dx_2'dx_3' \right\rangle, \quad (3.40b)$$

for Fluid 2. In these expressions, even though the stress field $\mathbf{s}^{(1)}$ in Fluid 1 exists only for $0 \le x_3 \le \infty$, and the stress field for Fluid 2 exists only for $-\infty \le x_3 \le 0$, in writing these stress fields as the inverse Fourier transform of their Fourier transforms, for mathematical convenience, they have been imagined as extending to the entire space, so that complications of half-space Fourier transforms would not have to be introduced. For this purpose this extension is legitimate; however, the physically correct domains had to be used in applying the divergence theorem.

By using the linearity of the expectation operator, (3.40a) can be rewritten as

$$
\begin{aligned}
&< \mathbf{n} \cdot \mathbf{s}^{(1)}(k_1, k_2, 0, t)\mathbf{n} \cdot \mathbf{s}^{(1)}(k_1', k_2', 0, t') > \\
&= \int_{x_1=-\infty}^{\infty} \int_{x_2=-\infty}^{\infty} \int_{x_3=0}^{\infty} \int_{x_1'=-\infty}^{\infty} \int_{x_2'=-\infty}^{\infty} \int_{x_3'=0}^{\infty} \\
&\Bigg[e^{i(k_1x_1+k_2x_2)} e^{i(k_1'x_1'+k_2'x_2')} \Big\langle \nabla \cdot \mathbf{s}^{(1)}(x_1, x_2, x_3, t) \nabla \cdot \mathbf{s}^{(1)}(x_1', x_2', x_3', t') \Big\rangle \\
&+ \Big\langle (ik_1\mathbf{e}_1 + ik_2\mathbf{e}_2) \cdot \mathbf{s}^{(1)}(x_1, x_2, x_3, t)(ik_1'\mathbf{e}_1 + ik_2'\mathbf{e}_2) \cdot \mathbf{s}^{(1)}(x_1', x_2', x_3', t') \Big\rangle \Bigg] \\
&\qquad dx_1 dx_2 dx_3 dx_1' dx_2' dx_3', \qquad (3.41)
\end{aligned}
$$

and similarly for Fluid 2. By the usual methods of linear response theory, the autocorrelation

$$
\Big\langle \nabla \cdot \mathbf{s}^{(l)}(x_1, x_2, x_3, t) \nabla \cdot \mathbf{s}^{(l)}(x_1', x_2', x_3', t') \Big\rangle \qquad (3.42)
$$

can be determined in terms of the known autocorrelation (3.1), which does not involve any derivatives. For convenience, rewrite (3.1) as

$$
< s_{ij}^{(l)}(\mathbf{x}, t) s_{km}^{(l)}(\mathbf{x}', t') >
$$

$$= C^{(l)}_{ijmn}\delta(\mathbf{x}-\mathbf{x}')\delta(t-t'), \tag{3.43}$$

where $l = 1, 2$. Then,

$$\left\langle \frac{\partial s^{(l)}_{ij}(\mathbf{x},t)}{\partial x_1}\frac{\partial s^{(l)}_{mn}(\mathbf{x}',t')}{\partial x_1}\right\rangle = -C^{(l)}_{ijmn}\delta''(d_1)\delta(d_2)\delta(d_3) \tag{3.44}$$

(where, for brevity, $\mathbf{d} = (d_1, d_2, d_3) = \mathbf{x}' - \mathbf{x}$ is introduced), with similar expressions where the partial derivatives with respect to x_1 are replaced by derivatives with respect to x_2, or x_3, and

$$\left\langle \frac{\partial s^{(l)}_{ij}(\mathbf{x},t)}{\partial x_1}\frac{\partial s^{(l)}_{mn}(\mathbf{x}+\mathbf{d},t')}{\partial x_2}\right\rangle = -C^{(l)}_{ijmn}\delta'(d_1)\delta'(d_2)\delta(d_3), \tag{3.45}$$

with similar expressions where the partial derivatives with respect to (x_1,x_2) are replaced by derivatives with respect to (x_1,x_3), or (x_2,x_3). From this it follows that the (j,n) component of the tensor (3.42) is

$$\left\langle \nabla\cdot\mathbf{s}^{(l)}(x_1,x_2,x_3,t)\nabla\cdot\mathbf{s}^{(l)}(x_1',x_2',x_3',t')\right\rangle_{jn} =$$
$$-\left[C^{(l)}_{1j1n}\delta''(d_1)\delta(d_2)\delta(d_3) + C^{(l)}_{2j2n}\delta(d_1)\delta''(d_2)\delta(d_3) + C^{(l)}_{3j3n}\delta(d_1)\delta(d_2)\delta''(d_3)\right]$$
$$-C^{(l)}_{1j2n}\delta'(d_1)\delta'(d_2)\delta(d_3) - C^{(l)}_{2j1n}\delta'(d_1)\delta(d_2)\delta'(d_3) - C^{(l)}_{1j3n}\delta(d_1)\delta'(d_2)\delta'(d_3)$$
$$-C^{(l)}_{3j1n}\delta'(d_1)\delta'(d_2)\delta(d_3) - C^{(l)}_{1j2n}\delta'(d_1)\delta(d_2)\delta'(d_3) - C^{(l)}_{2j1n}\delta(d_1)\delta'(d_2)\delta'(d_3). \tag{3.46}$$

Substituting (3.46) and (3.43) into (3.41) gives

$$< \mathbf{n}\cdot\mathbf{s}^{(l)}(k_1,k_2,0,t)\mathbf{n}\cdot\mathbf{s}^{(l)}(k_1',k_2',0,t') >_{jn}$$

$$= \int_{x_1=-\infty}^{\infty}\int_{x_2=-\infty}^{\infty}\int_{x_3=0}^{\infty}\int_{x_1'=-\infty}^{\infty}\int_{x_2'=-\infty}^{\infty}\int_{x_3'=0}^{\infty} e^{i(k_1x_1+k_2x_2)}e^{i(k_1'x_1'+k_2'x_2')}$$

$$\Big[-C^{(l)}_{1j1n}\delta''(d_1)\delta(d_2)\delta(d_3) - C^{(l)}_{2j2n}\delta(d_1)\delta''(d_2)\delta(d_3) - C^{(l)}_{3j3n}\delta(d_1)\delta(d_2)\delta''(d_3)$$

$$-C^{(l)}_{1j2n}\delta'(d_1)\delta'(d_2)\delta(d_3) - C^{(l)}_{2j1n}\delta'(d_1)\delta'(d_2)\delta(d_3) - C^{(l)}_{1j3n}\delta(d_1)'\delta(d_2)\delta'(d_3)$$

$$-C^{(l)}_{3j1n}\delta'(d_1)\delta(d_2)\delta'(d_3) - C^{(l)}_{2j3n}\delta(d_1)\delta'(d_2)\delta'(d_3) - C^{(l)}_{3j2n}\delta(d_1)\delta'(d_2)\delta'(d_3)$$

$$+(ik_1ik'_1C^{(l)}_{1j1n} + ik_1ik'_2C^{(l)}_{1j2n} + ik_2ik'_1C^{(l)}_{2j1n} + ik_2ik'_2C^{(l)}_{2j2n})\delta(\mathbf{x}-\mathbf{x}')\Big]$$

$$\delta(t-t')dx_1dx_2dx_3dx'_1dx'_2dx'_3. \tag{3.47}$$

If it is assumed that the fluid is homogeneous, then the correlation coefficients, $C^{(l)}_{ijmn}$, which depend on temperature and viscosity, are *independent* of x_3. In this case, all terms in the integrals involving $\delta'(d_3)$ or $\delta''(d_3)$, when integrated by parts, will give zero contribution. The rest of the integrals cancel, showing that

$$< \mathbf{n}\cdot\mathbf{s}^{(l)}(k_1,k_2,0,t)\mathbf{n}\cdot\mathbf{s}^{(l)}(k'_1,k'_2,0,t') >_{jn} = 0. \tag{3.48}$$

On the other hand, if the fluid is not homogeneous, and the $C^{(l)}_{ijmn}$ depend on x_3, the distance from the interface (corresponding to either the viscosity, or the temperature, or both, varying in the x_3 direction), then

$$< \mathbf{n}\cdot\mathbf{s}^{(l)}(k_1,k_2,0,t)\mathbf{n}\cdot\mathbf{s}^{(l)}(k'_1,k'_2,0,t') >_{jn}$$

$$= \int_{x_3=0}^{\infty}\Big[-\frac{dC^{(l)}_{3j3n}}{dx_3}\delta(k_1+k'_1)\delta(k_2+k'_2)$$

$$+\frac{dC^{(l)}_{1j3n}}{dx_3}\delta'(k_1+k'_1)\delta(k_2+k'_2) + \frac{dC^{(l)}_{3j1n}}{dx_3}\delta'(k_1+k'_1)\delta(k_2+k'_2)$$

$$+\frac{dC^{(l)}_{2j3n}}{dx_3}\delta(k_1+k'_1)\delta'(k_2+k'_2) + \frac{dC^{(l)}_{3j2n}}{dx_3}\delta(k_1+k'_1)\delta'(k_2+k'_2)\Big]$$

$$\delta(t-t')dx_1dx_2dx_3dx_1'dx_2'dx_3'. \tag{3.49}$$

An interesting thing that has been recovered here is the observation made by Brenner [17] that an interface between two homogeneous fluids can be modelled as a finite region of rapid change in fluid properties rather than a (zero-volume) plane of discontinuity. The nonzero contributions to the integral in (3.49) will then come entirely from the small region of rapidly varying viscosity between the two bulk fluids. For the normal stress ($j = 3, n = 3$), the integral in (3.49), which depends on how rapidly the viscosity varies in this finite region, is accounted for in some lumped way by the surface tension and is therefore definitely nonzero. It corresponds to the autocorrelation (1.21) for the random normal stress on the interface, which was derived by modelling the interface as a discontinuity, with its own inherent properties distinct from the bulk (namely, surface tension), and having a random Brownian stress in addition to the fluctuating stresses in the bulk fluid. Whether or not the tangential stresses have a nonzero correlation, in the case where the two bulk fluids are assumed homogeneous, will depend on other fluid properties not included in Yang's analysis. These properties would presumably enter into the equilibrium correlations because there would be some component of the energy of the system associated with them. Thus, if the two fluids had "interfacial viscosity," the autocorrelation of the tangential stresses would be nonzero.

It is, of course, also possible that the correlations will be nonzero because the bulk fluid is not homogeneous; i.e., the properties will not just vary rapidly in a small region around the interface, but will vary on a macroscopic length scale. This could occur if, for example, there was a temperature gradient in the fluid, or if surfactants were present on the interface. (Note that (3.48) also shows that some of the correlations would be nonzero if the fluid properties varied *along* the interface.) In such a case there might well be a number of other effects not included in the analysis here, such as convective currents, or diffusion of surfactants. Also, if thermodynamic quantities such as temperature and concentration vary in the system, the equilibrium correlations may change. The internal energy appears in the probability distribution (1.12) for a canonical ensemble; this is appropriate only for an isothermal system. For a nonisothermal system, some other free energy must be used. Aside from these detailed considerations, it is interesting that it can at least be seen qualitatively from (3.49) how the tangential stresses at the interface could have a nonzero autocorrelation.

Now all the above can be pieced together to get the fluid-velocity autocorrelation. While the particle velocity should really be obtained from the fluid velocity via the unsteady Faxen's Law, for simplicity, it will be assumed initially that the particle moves affinely with the fluid. Since the limit of low frequency will be taken to get the diffusivity, this assumption is valid for a small enough particle. (Of course, the unsteady Faxen's Law must be used to get the correct *particle* velocity autocorrelation, but that is another issue.) The full expression for the Fourier-transformed velocity at a distance x_3 from the interface is

$$\hat{u}_1^{(2)}(k_1,k_2,x_3,\omega) =$$

$$- e^{-\sqrt{c_1}x_3}\int_0^{x_3} e^{2\sqrt{c_1}y}\int_0^{y}\left[ik_1\hat{s}_{11}^{(2)}(k_1,k_2,z,\omega)+ik_2\hat{s}_{21}^{(2)}(k_1,k_2,z,\omega)\right]e^{-\sqrt{c_1}z}dzdy$$

$$- e^{-\sqrt{c_1}x_3}\int_0^{x_3} e^{\sqrt{c_1}y}\hat{s}_{31}^{(2)}(k_1,k_2,y,\omega)dy$$

$$+ e^{-\sqrt{c_1}x_3}\frac{1}{2\sqrt{c_1}}\left[e^{2\sqrt{c_1}x_3}-1\right]\hat{s}_{31}^{(2)}(k_1,k_2,0,\omega)$$

$$+\left[\frac{-ik_1a_{17}}{2\sqrt{c_2}(\sqrt{c_p}+\sqrt{c_2})}+a_{23}e^{\sqrt{c_2}x_3}\right]\left[\hat{s}_{13}^{(1)}(k_1,k_2,0,\omega)-\hat{s}_{13}^{(2)}(k_1,k_2,0,\omega)\right]$$

$$+ \left[\frac{-ik_1 a_{18}}{2\sqrt{c_2}(\sqrt{c_p}+\sqrt{c_2})} + a_{24} e^{\sqrt{c_2} x_3} \right] \left[\hat{s}_{23}^{(1)}(k_1,k_2,0,\omega) - \hat{s}_{23}^{(2)}(k_1,k_2,0,\omega) \right]$$

$$+ \left[\frac{-ik_1 a_{19}}{2\sqrt{c_2}(\sqrt{c_p}+\sqrt{c_2})} + a_{25} e^{\sqrt{c_2} x_3} \right] \left[\hat{s}_{33}^{(1)}(k_1,k_2,0,\omega) - \hat{s}_{33}^{(2)}(k_1,k_2,0,\omega) \right] . \quad (3.47)$$

Again, the constants a_{ij} appearing here are given in Appendix 5. The autocorrelation function is by definition

$$\mathbf{R}_u(k_1,k_2,x_3,\omega,k_1',k_2',x_3,\omega') \equiv < \mathbf{u}(k_1,k_2,x_3,\omega)\mathbf{u}(k_1',k_2',x_3,\omega') >, \quad (3.48)$$

and it has the form

$$\mathbf{R}_u(k_1,k_2,x_3,\omega,k_1',k_2',x_3,\omega') = \mathbf{C}_u(k_1,k_2,x_3,\omega,k_1',k_2',x_3,\omega')\delta(\omega+\omega'), \quad (3.49)$$

where the tensor $\mathbf{C}_u$ can be determined from (3.47) in the obvious way (i.e., by taking the expectation of the product of the expression for the velocity evaluated at the unprimed argument times the expression for the velocity evaluated at the primed argument). If Assumption 1 (the "frozen particle") is made again (for simplicity), and the assumption of affine motion is made, then the particle velocity is related to the fluid velocity by

$$\hat{\mathbf{U}}(x_3,\omega) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \hat{\mathbf{u}}(k_1,k_2,x_3,\omega)dk_1dk_2. \quad (3.50)$$

Instead of being regarded as a variable, here x_3 is viewed as a parameter, namely, the distance of the particle from the interface. Now the particle diffusivity is given by

$$\mathbf{D} = \lim_{\omega \to 0} \hat{\mathbf{T}}(\omega), \tag{3.51}$$

where $\hat{\mathbf{T}}(\omega)$ is the time-Fourier-transform of the particle autocorrelation defined in a different way, as for a stationary process:

$$\mathbf{T}(\omega)(\tau) \equiv < \mathbf{U}(t)\mathbf{U}(t+\tau) >, \tag{3.52}$$

where it depends only on the time difference. By some manipulations (transforming and inverse-transforming), it can be shown that

$$\hat{\mathbf{T}}(\omega) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbf{C}_u(k_1, k_2, x_3, \omega, k_1', k_2', x_3, -\omega) dk_1 dk_2 dk_1' dk_2'; \tag{3.53}$$

so that finally, the particle diffusivity is given by

$$\mathbf{D} = \lim_{\omega \to 0} \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbf{C}_u(k_1, k_2, x_3, \omega, k_1', k_2', x_3, -\omega) dk_1 dk_2 dk_1' dk_2'. \tag{3.54}$$

In the expression for the velocity (3.45), some of the stress terms appear inside integrals over x_3 (these come from the particular solution for the velocity field driven by the random stress in the bulk), whereas other stress terms are not inside integrals, but are evaluated at $x_3 = 0$ (since they come from imposing the boundary conditions). Since the purpose of the analysis is to determine whether the interface has any effect on the particle's diffusivity beyond the obvious change in mobility, it is the second type of stress term that should be considered, the type that is evaluated at the interface, rather than is integrated over the bulk, and that thus depends on interface properties. Consider then the following two members of the expression for the velocity (3.45):

$$\left[\frac{-ik_1 a_{18}}{2\sqrt{c_2}(\sqrt{c_p}+\sqrt{c_2})} + a_{24} e^{\sqrt{c_2} x_3}\right] \hat{s}_{23}^{(1)}(k_1, k_2, 0, \omega); \tag{3.55}$$

and

$$\left[\frac{-ik_1 a_{19}}{2\sqrt{c_2}(\sqrt{c_p}+\sqrt{c_2})} + a_{25} e^{\sqrt{c_2} x_3}\right] \hat{s}_{33}^{(1)}(k_1, k_2, 0, \omega). \tag{3.56}$$

The term in (3.55), which involves the tangential stress in the "2" direction on the interface, will lead to the following term in the velocity autocorrelation:

$$\left[\frac{-ik_1 a_{18}}{2\sqrt{c_2}(\sqrt{c_p}+\sqrt{c_2})} + a_{24} e^{\sqrt{c_2} x_3}\right] \left[\frac{-ik_1' a_{18}'}{2\sqrt{c_2'}(\sqrt{c_p'}+\sqrt{c_2'})} + a_{24}' e^{\sqrt{c_2'} x_3}\right]$$
$$\left\langle \hat{s}_{23}^{(1)}(k_1, k_2, 0, \omega) \hat{s}_{23}^{(1)}(k_1', k_2', 0, \omega') \right\rangle$$
$$\equiv (C_u)_{11}^a(k_1, k_2, x_3, \omega, k_1', k_2', x_3, -\omega) \left\langle \hat{s}_{23}^{(1)}(k_1, k_2, 0, \omega) \hat{s}_{23}^{(1)}(k_1', k_2', 0, \omega') \right\rangle, \tag{3.57}$$

where a prime denotes evaluation at k_1', k_2', ω' instead of k_1, k_2, ω. The term in (3.56), which involves the normal stress on the interface, gives rise to the following term in the velocity autocorrelation:

$$\left[\frac{-ik_1 a_{19}}{2\sqrt{c_2}(\sqrt{c_p}+\sqrt{c_2})} + a_{25} e^{\sqrt{c_2} x_3}\right] \left[\frac{-ik_1' a_{19}'}{2\sqrt{c_2'}(\sqrt{c_p'}+\sqrt{c_2'})} + a_{25}' e^{\sqrt{c_2'} x_3}\right]$$
$$\left\langle \hat{s}_{33}^{(1)}(k_1, k_2, 0, \omega) \hat{s}_{33}^{(1)}(k_1', k_2', 0, \omega') \right\rangle$$
$$\equiv (C_u)_{11}^b(k_1, k_2, x_3, \omega, k_1', k_2', x_3, -\omega) \left\langle \hat{s}_{33}^{(1)}(k_1, k_2, 0, \omega) \hat{s}_{33}^{(1)}(k_1', k_2', 0, \omega') \right\rangle. \tag{3.58}$$

The superscripts "a" and "b" on $(C_u)^a_{11}$ and $(C_u)^b_{11}$ are merely to indicate that these each represent only one of a number of terms in the (1,1) component of the total velocity autocorrelation. Now by the calculation of the stresses at the interface given earlier, the stress autocorrelations appearing here can be written in the form of (3.49) (to show the delta correlation in time) as

$$\left\langle \hat{s}^{(1)}_{23}(k_1,k_2,0,\omega)\hat{s}^{(1)}_{23}(k'_1,k'_2,0,\omega')\right\rangle = S^a(k_1,k_2,k'_1,k'_2)\delta(\omega+\omega'); \tag{3.59}$$

and

$$\left\langle \hat{s}^{(1)}_{33}(k_1,k_2,0,\omega)\hat{s}^{(1)}_{33}(k'_1,k'_2,0,\omega')\right\rangle = S^b(k_1,k_2,k'_1,k'_2)\delta(\omega+\omega'), \tag{3.60}$$

where S_a and S_b are going to depend on the interface properties. Now by examination of Equation (3.54), the contribution to the (1,1) diffusivity component from these terms will be

$$D^a_{11} = \lim_{\omega\to 0}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} S^a(C_u)^a_{11}(k_1,k_2,x_3,\omega,k'_1,k'_2,x_3,-\omega)dk_1dk_2dk'_1dk'_2 \tag{3.61}$$

from (3.55), and

$$D^b_{11} = \lim_{\omega\to 0}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} S^b(C_u)^b_{11}(k_1,k_2,x_3,\omega,k'_1,k'_2,x_3,-\omega)dk_1dk_2dk'_1dk'_2 \tag{3.62}$$

from (3.56). The integrands here turn out to be uniformly continuous, so that the limits can be commuted with the integrals. If it is assumed that the fluid properties are such that the $(C_u)^a_{11}$ and $(C_u)^b_{11}$ are nonzero, then whether or not these contributions to the diffusivity are nonzero depends on the limits of the (transfer) functions S^a and S^b. It is here that the difference between the tangential and the normal stresses at the interface is made evident. As $\omega \to 0$, the transfer function S^a, which relates fluid velocity to tangential stresses at the interface, approaches a finite limit. On the other hand, the transfer function S^b goes to zero in this limit. This latter result is consistent with the observation made in previous sections that the normal stresses at the interface cannot cause the particle mean-square displacement to grow indefinitely, because surface tension acts like a spring, preventing the interface deformations from growing indefinitely, and the surrounding fluid cannot be displaced with an amplitude growing faster than the interface displacements. There is, on the other hand, nothing to prevent an arbitrarily large displacement of a fluid element *along* the interface, and this is reflected in the fact that tangential stresses at the interface can give a contribution to the diffusivity. Whether or not real interfaces actually *have* such tangential stresses is, of course, open to question.

SECTION 4: A DIFFERENT DEFINITION OF PARTICLE DIFFUSIVITY

From the discussions in Sections 2 and 3 it is clear that as long as the "direct" forcing on the particle is ignored, if the mean-square displacement of the particle is plotted as a function of time, it will asymptote to a horizontal line. The definition of diffusivity that has been used in the above sections was the slope of this plot for very long times. It is of interest to see if the particle undergoes motion that can be regarded as diffusive at shorter times, specifically, short enough that its mean-square displacement has not approached the flat part, but long enough that there is still time for the particle to sample different interface configurations. The usual definition of the short-time diffusivity, which is the initial slope, will include the effect of the interface only insofar as it affects the particle *mobility*. Since the goal here is to determine whether the interface gives rise to other less obvious effects, the short-time diffusivity will not be calculated here (its calculation is essentially a purely hydrodynamic one). There is, however, yet another definition of diffusivity that can be used: the coefficient of any diffusion-type term that appears in the particle Fokker-Planck equation once the interface configuration variables have been "averaged out." This definition will now be made more precise.

If the variables describing the entire system configuration are listed, namely the particle position $\mathbf{X}(t)$ and velocity $\mathbf{U}(t)$, and the interface deformation $\eta(t)$, it is clear that there is a Fokker-Planck equation for the entire system, i.e., an equation for the probability density P for finding the system in a particular configuration at a particular time, given some initial state. This equation will be linear in the probability density, although it will probably have nonconstant coefficients. One way to define a diffusivity might be to integrate this equation with respect to all the coordinates except the particle position, and to try to rewrite the resulting equation in terms of a new probability density P' for just the particle coordinates (where, of course, P' is just P integrated with respect to all the coordinates except the particle position). Were this possible, the diffusivity could then be defined as the coefficient of any "diffusion-type" term that appears in the resulting equation (i.e., any term involving $\nabla^2 P'$). However, since the coefficients of the Fokker-Planck equation for P are not constant, some assumptions about certain moments of P would have to be made before the integrated equation could be expressed solely in terms of P'. These assumptions could not be justified without knowing something about the actual solution. It should also be noted that since the interface configuration requires an infinite number of coordinates to describe it, "integrating" with respect to all the interface coordinates is integration over a function space rather than integration in the usual sense.

A simpler approach in which the assumptions to be made are at least more evident, even if not more correct, is to start with the particle Langevin equation:

$$m_P \frac{d\mathbf{U}}{dt} + \mathbf{B}(t)[\mathbf{U}] = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbb{F}[\eta(\mathbf{x}_s, t); (\mathbf{k}, d, \omega)]dk_1 dk_1 + \mathbb{A}(t). \tag{4.1}$$

Here, the functional dependence of the indirect random forcing F on the interface deformation η has been explicitly shown. What is desired is to average out η. The only known probability distribution for η is the time-independent equilibrium distribution. The time-dependent (second-order) autocorrelations are known, but that is far less information than knowing the actual time-dependent distribution. In principle, higher-order autocorrelations could be calculated by making some assumption about the random forcing being Gaussian (so that higher-order moments would be given by products of second-order ones), and the probability distribution could be retrieved from all the autocorrelations of all orders (or approximated by using a finite number of the autocorrelations). This would be a very complicated calculation, however. It would be simpler just to average over the equilibrium distribution. The first problem that arises in doing this is that the forcing depends on time derivatives of η as well as on η, if it is computed from the full unsteady hydrodynamic problem. The equilibrium distribution, coming from thermodynamic considerations, gives no information about time derivatives. This can be bypassed by assuming that the interface starts out with the initial configuration but that the fluid is initially quiescent. The relaxation of the system can then be calculated from the unsteady hydrodynamic equations with these initial conditions, and the force on the particle as a function of time due to the flow field can be calculated. The second problem, mentioned earlier, is that this averaging involves an integration over function space, which is essentially an infinite number of integrals. To put this integration in more concrete terms, choose a basis:

$$\{\eta_0, \eta_1, \eta_2, \ldots\}, \tag{4.2}$$

so that any η can be written as

$$\eta = \sum_{m=0}^{\infty} a_m \eta_m. \tag{4.3}$$

To get an approximate result, truncate this expansion after N terms:

$$\eta = \sum_{m=0}^{N} a_m \eta_m. \tag{4.4}$$

Then the interface configuration is approximately described by the *finite* set of coordinates $\{a_0, a_1, a_2, ..., a_N\}$. The average mean-square force then becomes

$$\int_{a_0}\int_{a_1}\cdots\int_{a_N} \mathbf{F}[\eta(\mathbf{x}_s,t);(\mathbf{k},d,\omega)]\mathbf{F}[\eta(\mathbf{x}_s,t);(\mathbf{k},d,\omega)]P(a_0,a_1,a_2,...,a_N). \quad (4.5)$$

The probability distribution for the truncated set of coefficients can be determined from the full probability distribution by renormalizing it. Yang [10] found the equilibrium probability distribution to be (cf. (1.12))

$$P[\eta(\mathbf{x}_s,t)] = N\exp\left[-\frac{1}{2\kappa T}\int_{\mathbf{x}_s}\left((\Delta\rho)\eta^2(\mathbf{x}_s,t)+\gamma|\nabla_{\mathbf{x}_s}(\mathbf{x}_s,t)|^2\right)d\mathbf{x}_s\right]. \quad (4.6)$$

Here, N is a normalization constant, which cannot really be determined without considering integration over some infinite-dimensional space. However, this equation gives the *relative* magnitudes of probabilities for different configurations η, which is all that is needed to determine the normalized distribution for the projection onto a finite-dimensional space.

A natural choice of a basis is the set of functions

$$\eta_m = \exp\left[\frac{i\mathbf{y}\cdot\mathbf{x}}{m}\right], \quad (4.7)$$

which are spatially periodic, with a decreasing wavelength as m increases. The maximum wavelength this set allows is $\mathbf{y}$, which should therefore be chosen on the order of the maximum dimension of the system of interest. In fact, it will be far more convenient to consider these functions over only a bounded range (i.e., have $\eta_m = 0$ outside this range), since they give infinity when integrated over the whole space. The choice of N, the number of terms in the expansion, is determined by how far towards the other extreme of short wavelengths it is necessary to go; the shorter the wavelength, the more improbable the configuration, since it will have a higher energy associated with it because of surface tension.

With this choice of basis, the probability distribution becomes

$$P(a_0, a_1, a_2, ..., a_N) = N \exp\left[-\frac{1}{2\kappa T}\int_{\mathbf{x}_s}\left\{(\Delta\rho)\left[\sum_{m=1}^{N} a_m \exp\left(\frac{i\mathbf{y}\cdot\mathbf{x}}{m}\right)\right]^2 + \gamma\left|\sum_{m=1}^{N} a_m \frac{i\mathbf{y}}{m}\exp\left(\frac{i\mathbf{y}\cdot\mathbf{x}}{m}\right)\right|^2\right\} d\mathbf{x}_s\right]. \tag{4.8}$$

It is necessary now to have a solution to the hydrodynamic problem for flow with an interface initially stationary with a specified deformation away from the flat state. This is similar enough to the hydrodynamic problem solved in Section 3 that the equations can be taken from there with a few modifications. In Section 3 the problem of flow driven by an imposed stress throughout the fluid was solved. Only the homogeneous part of this solution is needed here, since the flow is driven by the initial interface condition rather than by a forcing in the equations of motion. From Section 3, the *homogeneous* velocity field in fluid 2 is:

$$\hat{w}_1^{(2)} = e^{-\sqrt{c_2}x_3}\int_0^{x_3} e^{2\sqrt{c_2}y}\int_0^{y} ik_1\left[B_1 e^{\sqrt{c_p}z}\right]e^{-\sqrt{c_2}z}dzdy + B_2 e^{\sqrt{c_2}x_3} + B_2' e^{-\sqrt{c_2}x_3}; \tag{4.9a}$$

$$\hat{w}_2^{(2)} = e^{-\sqrt{c_2}x_3}\int_0^{x_3} e^{2\sqrt{c_2}y}\int_0^{y} ik_1\left[B_1 e^{\sqrt{c_p}z}\right]e^{-\sqrt{c_2}z}dzdy + B_3 e^{\sqrt{c_2}x_3} + B_3' e^{-\sqrt{c_2}x_3}; \tag{4.9b}$$

$$\hat{w}_3^{(2)} = e^{-\sqrt{c_2}x_3} \int_0^{x_3} e^{2\sqrt{c_2}y} \int_0^y \frac{d}{dz}\left[B_1 e^{\sqrt{c_P}z}\right] e^{-\sqrt{c_2}z} dz dy$$
$$+ \qquad B_4 e^{\sqrt{c_2}x_3} + B_4' e^{-\sqrt{c_2}x_3}, \tag{4.9c}$$

where the constants were determined from the boundary conditions, the application of which resulted in the equations in Appendix 4. Here, the quasi-steady problem is to be solved, so that the frequency ω will be set to zero. This means that $c_1 = c_2 = c_P$. The two kinematic conditions (A4.1) and (A4.2) must be replaced by the one condition

$$A_4 + A_4' = B_4 + B_4'. \tag{4.10}$$

Also, there is no random stress field throughout the fluid in this problem, so that all the terms $\hat{s}_{i3}^{(l)}(0)$ that appear in Appendix 4 will not appear in the boundary conditions here. Apart from these changes, the same equations as in Appendix 4 can be used. The solution to these equations is given in detail in Appendix 6, but will be written here as

$$\hat{w}_i^{(2)}(k_1, k_2, x_3) = G_i(k_1, k_2, x_3)\eta(k_1, k_2), \tag{4.11}$$

so that if affine motion is assumed (i.e., very small particle), the mean-square velocity is

$$< \hat{w}_i^{(2)}(k_1, k_2, x_3)\hat{w}_i^{(2)}(k_1, k_2, x_3) >=$$
$$\int G_i(k_1, k_2, x_3)\hat{\eta}(k_1, k_2)G_i(k_1, k_2, x_3)\hat{\eta}(k_1, k_2)P[\hat{\eta}(k_1, k_2, t)]d\eta, \tag{4.12}$$

where now $P[\hat{\eta}(k_1, k_2, t)]$ is the probability density in Fourier space. This can be related to the density in physical space by the Plancherel theorem, which states that the Fourier transform is an isometry; i.e., if $f(\mathbf{x})$ and $\hat{f}(\mathbf{k})$ are a Fourier transform pair, then

$$\int_{\mathbf{x}} [f(\mathbf{x})]^2 d\mathbf{x} = \int_{\mathbf{k}} [\hat{f}(\mathbf{k})]^2 d\mathbf{k}. \tag{4.13}$$

From this it is evident that

$$P[\eta(\mathbf{x}_s, t)] = N \exp\left[-\frac{1}{2\kappa T}\int_{\mathbf{x}_s}\left((\Delta\rho)\eta^2(\mathbf{x}_s, t) + \gamma|\nabla_{\mathbf{x}_s}(\mathbf{x}_s, t)|^2\right) d\mathbf{x}_s\right]$$

$$= N \exp\left[-\frac{1}{2\kappa T}\int_{\mathbf{k}}\left((\Delta\rho)\hat{\eta}^2(\mathbf{k}, t) + \gamma|(-k^2)\hat{\eta}(\mathbf{k}, t)|^2\right) d\mathbf{k}\right]. \tag{4.14}$$

Thus, the fluid-velocity autocorrelation becomes

$$< \hat{w}_i^{(2)}(k_1, k_2, x_3)\hat{w}_i^{(2)}(k_1, k_2, x_3) >=$$

$$\frac{\int G_i(k_1, k_2, x_3)\eta(k_1, k_2)G_i(k_1, k_2, x_3)\eta(k_1, k_2)P[\hat{\eta}(k_1, k_2, t)]}{\int P[\hat{\eta}(k_1, k_2, t)]d\eta}. \tag{4.15}$$

Once again the assumption will be made that the particle velocity can be approximated by the fluid velocity at the particle's initial position $x_1 = 0, x_2 = 0$. (To get a better approximation, the perturbative method discussed in Section 2 could be used.) With this simplest assumption, the mean-square particle velocity becomes

$$< U_i U_j >= \frac{1}{2\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}$$

$$\left[\frac{\int G_i(k_1, k_2, x_3)\eta(k_1, k_2)G_j(k_1, k_2, x_3)\eta(k_1, k_2)P[\hat{\eta}(k_1, k_2)]d\eta}{\int P[\hat{\eta}(k_1, k_2)]d\eta}\right] dk_1 dk_2. \tag{4.16}$$

If the basis of functions suggested earlier is used, this becomes

$$< U_i U_j >= \frac{1}{2\pi} \int_{k_1=-\infty}^{\infty} \int_{k_2=-\infty}^{\infty}$$

$$\left[\int G_i(k_1, k_2, x_3) \left(\sum_{j=0}^{N} a_j \hat{\eta}_j(k_1, k_2) \right) G_j(k_1, k_2, x_3) \right.$$

$$\left. \left(\sum_{m=0}^{N} a_m \hat{\eta}_m(k_1, k_2) \right) P[\hat{\eta}(k_1, k_2)] da_1 da_2 \cdots da_N \right]$$

$$\left[\int P[\hat{\eta}(k_1, k_2)] da_1 da_2 \cdots da_N \right]^{-1} dk_1 dk_2. \tag{4.17}$$

Another way of approaching this is to regard the η_j's as "trial functions," and the mean-square particle velocity as a functional. As more and more linearly independent trial functions are used, the estimate for the mean-square velocity improves.

By analogy with the Stokes-Einstein result, where the particle diffusivity is twice the mean-square velocity divided by the steady mobility, the (dimensionless) particle diffusivity will be assumed, for simplicity, to be given by

$$D_{ij} = \frac{2 < U_i U_j >}{(6\pi)}, \tag{4.18}$$

with $< U_i U_j >$ as given above in (4.17). More detailed calculations than the above could be carried out, for instance by using the Faxen's Law for the force on a finite-sized particle placed in a flow field rather than assuming affine motion. However, the above illustrates that a nonzero diffusivity can be calculated by this method. The assumption made here is that the equilibrium distribution of interface configurations gives a representative sample of the configurations that the particle sees as it undergoes random motion. While it has been shown earlier (subject to certain assumptions) that the particle will eventually asymptote to a finite mean-square displacement and will remain within a finite region, it is conjectured that the diffusivity derived above gives the rate at which the particle's mean-square displacement grows for shorter times when it has not yet neared its limiting value.

Appendix 1 to Chapter IV

In this appendix, the asymptotic expansion for $k \to 0$ of the integral

$$F(k) = \int_{-\infty}^{\infty} \frac{ds}{\hat{H}_I(k,s)\hat{H}_I(k,-s)},$$

where $\hat{H}_I$ is given by

$$\hat{H}_I(k,\omega) = -\frac{2\omega^2 \rho\alpha}{k(k-\alpha)} - \gamma k^2,$$

and

$$\alpha = \left(k^2 - \frac{is}{\nu}\right)^{1/2},$$

will be determined. Here the square-root sign will always refer to the *positive* branch of the square-root function. The difficulty lies in the fact that the integrand cannot be uniformly approximated, because the nature of the expansion of terms like

$$k - \sqrt{\left(k^2 - \frac{is}{\nu}\right)}$$

will depend on the relative magnitudes of k and s. This motivates the following change of variables. Let

$$\tau = \frac{s}{\nu k^2};$$

$$\zeta = \frac{\rho\nu^2 k}{\gamma};$$

(these have also been chosen to be dimensionless). A few manipulations then give that

$$F(k) = \frac{\nu}{k^2\gamma}\int_{-\infty}^{\infty}\frac{d\tau}{\left[-2\zeta\tau^2\frac{\sqrt{1-i\tau}}{1-\sqrt{1-i\tau}}-1\right]\left[-2\zeta\tau^2\frac{\sqrt{1+i\tau}}{1-\sqrt{1+i\tau}}-1\right]}.$$

To avoid dealing with the factor 2, let $2\zeta = \epsilon$, and define for convenience

$$J(\epsilon) = \frac{k^2\gamma}{\nu}F(k),$$

to get rid of some more factors temporarily. It also proves to be convenient to make one further change of variables,

$$\theta = \sqrt{\epsilon}\tau.$$

So, finally, the integral to be dealt with is

$$J(\epsilon) \equiv \frac{1}{\sqrt{\epsilon}}I(\epsilon),$$

where

$$I(\epsilon) = \int_{-\infty}^{\infty}\frac{d\theta}{\left[-\theta^2\frac{\sqrt{1-i\theta/\epsilon}}{1-\sqrt{1-i\theta/\epsilon}}-1\right]\left[-\theta^2\frac{\sqrt{1+i\theta/\epsilon}}{1-\sqrt{1+i\theta/\epsilon}}-1\right]}.$$

The only evident way to deal with this integral analytically is to use contour integration. The poles can be found only approximately. If the following definitions,

$$f(\theta) = -\theta^2\frac{\sqrt{1-i\theta/\epsilon}}{1-\sqrt{1-i\theta/\epsilon}}-1,$$

$$g(\theta) = -\theta^2\frac{\sqrt{1+i\theta/\epsilon}}{1-\sqrt{1+i\theta/\epsilon}}-1,$$

are made, then it is clear that if θ_a is a zero of $f(\theta)$, then $-\theta_a$ is a zero of $g(\theta)$. To solve the equation $f(\theta) = 0$, the square-root terms are collected on one side of the equation, and both sides of the equation are then squared, resulting in a quartic equation for θ. Two of its four roots are spurious solutions that arise from squaring. The other two are actually solutions to $f(\theta) = 0$. They can be shown to be given approximately by

$$\theta_a = 1 + \frac{1+i}{2\sqrt{2}}\epsilon^{1/4},$$

and

$$\tilde{\theta}_a = -1 + \frac{1-i}{2\sqrt{2}}\epsilon^{1/4},$$

so that $g(\theta)$ must have zeros approximately at

$$\theta_b = 1 - \frac{1-i}{2\sqrt{2}}\epsilon^{1/4},$$

and

$$\tilde{\theta}_b = -1 - \frac{1+i}{2\sqrt{2}}\epsilon^{1/4}.$$

Figure 1 shows the relative location of these roots, as well as the branch cuts for the functions $\sqrt{1 + i\theta/\sqrt{\epsilon}}$ and $\sqrt{1 - i\theta/\sqrt{\epsilon}}$, and the keyhole contour (which avoids crossing the branch cuts so as to remain in a domain of analyticity), chosen for integration. This contour is closed in the upper half-plane, so that there will be two residues, from θ_a and θ_b. The residue from θ_a is

$$\lim_{\theta \to \theta_a} \frac{\theta - \theta_a}{f(\theta)g(\theta)},$$

which by L'Hospital's rule is

$$\lim_{\theta \to \theta_a} \frac{1}{f\frac{dg}{d\theta} + g\frac{df}{d\theta}} = \frac{1}{g(\theta_a)\frac{df(\theta_a)}{d\theta}}.$$

Similarly, the residue at θ_b is given by

$$\lim_{\theta \to \theta_b} \frac{1}{f\frac{dg}{d\theta} + g\frac{df}{d\theta}} = \frac{1}{f(\theta_b)\frac{dg(\theta_b)}{d\theta}}.$$

After some tedious calculations it is found that (approximately)

$$f(\theta_b) = -\sqrt{2}\epsilon^{1/4};$$

$$g(\theta_a) = \sqrt{2}\epsilon^{1/4};$$

$$\frac{df(\theta_a)}{d\theta} = 2 - \frac{1+i}{2\sqrt{2}}\epsilon^{\frac{1}{4}};$$

$$\frac{dg(\theta_b)}{d\theta} = 2 + \frac{1-i}{2\sqrt{2}}\epsilon^{\frac{1}{4}},$$

so that the sum of the residues is

$$\frac{1}{\sqrt{2}\epsilon^{1/4}\left[2 - \frac{1+i}{2\sqrt{2}}\epsilon^{\frac{1}{4}}\right]} - \frac{1}{\sqrt{2}\epsilon^{1/4}\left[2 + \frac{1-i}{2\sqrt{2}}\epsilon^{\frac{1}{4}}\right]} \sim \frac{1}{8}.$$

Therefore, by the Cauchy integral theorem,

$$I(\epsilon) = \frac{2\pi i}{8} - I_B^{down} - I_{BP} - I_B^{up},$$

where I_B^{down} is the integral going down on the left side of the branch cut, I_{BP} is the circular integral around the branch point, and I_B^{down} is the integral going up on the right side of the branch cut. These integrals, which are easier to handle because their integrands can be uniformly approximated for small ϵ, will now be considered. First of all, since the integrand does not have a pole at the branch point, I_{BP} will obviously go to zero as the keyhole contour is moved closer and closer to the branch cut. So it can be ignored. Now let the path along the branch cut (i.e., along the imaginary axis) be parameterized by the real number t, so that $\theta = it$. The angles α and $\tilde{\alpha}$ shown in Figure 1 were chosen so that the desired positive branch of the square-root function can be obtained by choosing

$$\left(1 + \frac{i\theta}{\sqrt{\epsilon}}\right) = \left|1 + \frac{i\theta}{\sqrt{\epsilon}}\right| e^{i\alpha},$$

and

$$\left(1 - \frac{i\theta}{\sqrt{\epsilon}}\right) = \left|1 - \frac{i\theta}{\sqrt{\epsilon}}\right| e^{i\tilde{\alpha}},$$

so that

$$\left(1 + \frac{i\theta}{\sqrt{\epsilon}}\right)^{1/2} = \left|1 + \frac{i\theta}{\sqrt{\epsilon}}\right|^{1/2} e^{i\alpha/2},$$

and similarly,

$$\left(1 - \frac{i\theta}{\sqrt{\epsilon}}\right)^{1/2} = \left|1 - \frac{i\theta}{\sqrt{\epsilon}}\right|^{1/2} e^{i\tilde{\alpha}/2}.$$

Along the branch cut for $\sqrt{1 + i\theta/\sqrt{\epsilon}}$, it is clear that $\tilde{\alpha} = 0$, and that $\alpha = -\pi$ on the "left side" of the branch cut, whereas $\alpha = \pi$ on the "right side" of the branch cut. From all this it is clear that on the left side,

$$\left(1 + \frac{i\theta}{\sqrt{\epsilon}}\right)^{1/2} = -i\left|1 - \frac{t}{\sqrt{\epsilon}}\right|^{1/2};$$

$$\left(1 - \frac{i\theta}{\sqrt{\epsilon}}\right)^{1/2} = \left|1 + \frac{t}{\sqrt{\epsilon}}\right|^{1/2},$$

whereas on the right side,

$$\left(1 + \frac{i\theta}{\sqrt{\epsilon}}\right)^{1/2} = i\left|1 - \frac{t}{\sqrt{\epsilon}}\right|^{1/2};$$

$$\left(1 - \frac{i\theta}{\sqrt{\epsilon}}\right)^{1/2} = \left|1 + \frac{t}{\sqrt{\epsilon}}\right|^{1/2}.$$

Substituting this into the integrands gives

$$I_B^{down} + I_B^{up} = \int_{\infty}^{\sqrt{\epsilon}} \frac{idt}{\left[-(it)^2 \frac{(-i)\sqrt{t/\epsilon - 1}}{1-(-i)\sqrt{t/\epsilon-1}} - 1\right]\left[-(it)^2 \frac{\sqrt{1+t/\epsilon}}{1-\sqrt{1+t/\epsilon}} - 1\right]}$$

$$+ \int_{\sqrt{\epsilon}}^{\infty} \frac{idt}{\left[-(it)^2 \frac{i\sqrt{t/\epsilon-1}}{1-i\sqrt{t/\epsilon-1}} - 1\right]\left[-(it)^2 \frac{\sqrt{1+t/\epsilon}}{1-\sqrt{1+t/\epsilon}} - 1\right]}$$

$$= 0 + o(1).$$

In other words, the integrals along the branch cut contribute only at higher order. Thus, finally,

$$\lim_{\epsilon \to 0} I(\epsilon) = \frac{\pi i}{4},$$

a somewhat surprising result since it would be guessed by inspection that the integral would be real (since it contains two factors that look like complex conjugates) and that it would go to infinity as $\epsilon \to 0$, since naively bringing the limit inside the integral (an illegitimate operation, of course) gives a divergent result.

Thus,

$$J(\epsilon) \sim \frac{1}{\sqrt{\epsilon}} \frac{\pi i}{4},$$

as $\epsilon \to 0$, so that

$$F(k) \sim \frac{\nu}{k^2 \gamma} \frac{1}{\sqrt{2 \frac{\rho \nu^2 k}{\gamma}}} \frac{\pi i}{4},$$

as $k \to 0$, the desired result.

Appendix 2 to Chapter IV

In this appendix, the rules for constructing diagrams to represent the perturbation solution of the stochastic differential equation

$$L\{\mathbf{X}\} = \mathbf{F}(\mathbf{X}(t), t),$$

where L is a linear operator with a known Green's function G, will be given. This shorthand notation is useful since these perturbation calculations result in many terms, even for just the first correction (and higher-order corrections are extremely complicated). It gives a convenient and simple overview of the perturbation scheme.

The symbols will stand for factors in the integrand. A thin line with an arrow (pointing forward in time) will stand for the Green's function. A thin line terminating in the symbol $F^{(i)}$ will stand for a factor of $\nabla ... \nabla \mathbf{F}$ (with i ∇ operators) (all evaluated at the initial particle position $\mathbf{X}_0$). A thick line terminating in the symbol $\mathbf{X}$ will stand for a factor of $\mathbf{X} - \mathbf{X}_0$ (the particle displacement), which, of course, is the variable being solved for.

It is assumed that the function $\mathbf{F}(\mathbf{X}(t), t)$ can be expanded in a Taylor series about $\mathbf{X}_0$ so that

$$\begin{aligned} L\{\mathbf{X}\} = {} & \mathbf{F}(\mathbf{X}_0, t) \\ & + \nabla \mathbf{F}(\mathbf{X}_0, t) \cdot (\mathbf{X}(t) - \mathbf{X}_0) \\ & + \nabla\nabla \mathbf{F}(\mathbf{X}_0, t) \cdot (\mathbf{X}(t) - \mathbf{X}_0)(\mathbf{X}(t) - \mathbf{X}_0) \\ & + \nabla\nabla\nabla \mathbf{F}(\mathbf{X}_0, t) \cdot (\mathbf{X}(t) - \mathbf{X}_0)(\mathbf{X}(t) - \mathbf{X}_0)(\mathbf{X}(t) - \mathbf{X}_0) \\ & + \ldots\ldots \quad . \end{aligned}$$

Thus, the solution $X(t)$ is the linear superposition of the solutions to the following series of equations, where some approximation for $\mathbf{X}$ must be substituted for $\mathbf{X}$ wherever it appears in the right-hand side:

$$L\{\mathbf{X}\} = \mathbf{F}(\mathbf{X}_0, t);$$

$$L\{\mathbf{X}\} = \nabla\mathbf{F}(\mathbf{X}_0, t) \cdot (\mathbf{X}(t) - \mathbf{X}_0);$$

$$L\{\mathbf{X}\} = \nabla\nabla\mathbf{F}(\mathbf{X}_0, t) \cdot (\mathbf{X}(t) - \mathbf{X}_0)(\mathbf{X}(t) - \mathbf{X}_0);$$

$$L\{\mathbf{X}\} = \nabla\nabla\nabla\mathbf{F}(\mathbf{X}_0, t) \cdot (\mathbf{X}(t) - \mathbf{X}_0)(\mathbf{X}(t) - \mathbf{X}_0)(\mathbf{X}(t) - \mathbf{X}_0);$$

$$\vdots$$

If it is recalled that the solution to $L\{\mathbf{X}\} = R$, where R stands for a general right-hand-side term, is $\mathbf{X} = \int GR$, where G is the Green's function (and the arguments of the functions are suppressed here for simplicity), then it is evident that the diagram representation of the solution is given by Figure 2, and then, by replacing the unknown solution by its approximations to various orders, the diagram representation of Figure 3 is obtained. To get the autocorrelations, these diagrams are "glued" together in all possible combinations. Any diagram with a "dangling" end, i.e., an odd number of random terms, will represent a zero correction. The diagrams for the autocorrelations are shown in Figure 4.

Appendix 3 to Chapter IV

In this appendix, the first correction to the velocity autocorrelation of the Brownian particle considered in Section 2 of Chapter IV, which is due to the fact that the forcing depends on the particle's position, is given:

$$< \hat{U}_j^{(2)}(\omega)\hat{U}_m^{(2)}(\omega') >= \frac{1}{(-i\omega)(-i\omega')[-i\omega m_P + \hat{B}(\omega)]^2[-i\omega' m_P + \hat{B}(\omega')]^2}$$
$$\times \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} I(k_1,k_2,k_1',k_2',k_1'',k_2'',k_1''',k_2''',\omega,\omega')$$
$$dk_1\ dk_2\ dk_1'\ dk_2'\ dk_1''\ dk_2''\ dk_1'''\ dk_2''',$$

where

$$\begin{aligned}
I(k_1,&k_2,k_1',k_2',k_1'',k_2'',k_1''',k_2''',\omega,\omega')\\
=&\Big\langle ik_1\hat{F}_j(k_1,k_2,d,\omega)\hat{F}_1(k_1'',k_2'',d,\omega)ik_1'\hat{F}_m(k_1',k_2',d,\omega')\hat{F}_1(k_1''',k_2''',d,\omega')\Big\rangle\\
+&\Big\langle ik_1\hat{F}_j(k_1,k_2,d,\omega)\hat{F}_1(k_1'',k_2'',d,\omega)ik_2'\hat{F}_m(k_1',k_2',d,\omega')\hat{F}_2(k_1''',k_2''',d,\omega')\Big\rangle\\
+&\Big\langle ik_1\hat{F}_j(k_1,k_2,d,\omega)\hat{F}_1(k_1'',k_2'',d,\omega)\frac{\partial\hat{F}_m(k_1',k_2',d,\omega')}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1''',k_2''',d,\omega')\Big\rangle\\
+&\Big\langle ik_2\hat{F}_j(k_1,k_2,d,\omega)\hat{F}_2(k_1'',k_2'',d,\omega)ik_1'\hat{F}_m(k_1',k_2',d,\omega')\hat{F}_1(k_1''',k_2''',d,\omega')\Big\rangle\\
+&\Big\langle ik_2\hat{F}_j(k_1,k_2,d,\omega)\hat{F}_2(k_1'',k_2'',d,\omega)ik_2'\hat{F}_m(k_1',k_2',d,\omega')\hat{F}_2(k_1''',k_2''',d,\omega')\Big\rangle\\
+&\Big\langle ik_2\hat{F}_j(k_1,k_2,d,\omega)\hat{F}_2(k_1'',k_2'',d,\omega)\frac{\partial\hat{F}_m(k_1',k_2',d,\omega')}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1''',k_2''',d,\omega')\Big\rangle\\
+&\Big\langle\frac{\partial\hat{F}_j(k_1,k_2,d,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1'',k_2'',d,\omega)ik_1'\hat{F}_m(k_1',k_2',d,\omega')\hat{F}_1(k_1''',k_2''',d,\omega')\Big\rangle\\
+&\Big\langle\frac{\partial\hat{F}_j(k_1,k_2,d,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1'',k_2'',d,\omega)ik_2'\hat{F}_m(k_1',k_2',d,\omega')\hat{F}_2(k_1''',k_2''',d,\omega')\Big\rangle\\
+&\Big\langle\frac{\partial\hat{F}_j(k_1,k_2,d,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1'',k_2'',d,\omega)\frac{\partial\hat{F}_m(k_1',k_2',d,\omega')}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1''',k_2''',d,\omega')\Big\rangle.
\end{aligned}$$

It has been assumed that the forcing function F is Gaussian, since it is a linear functional of the random normal stress, which is assumed to have a Gaussian distribution. It is well-known [17] that if $x_1,\ x_2,\ x_3,\ x_4$ are arbitrary Gaussian random variables with zero mean, then

$$< x_1 x_2 x_3 >= 0$$

and

$$< x_1, x_2, x_3, x_4 >=< x_1 x_2 >< x_3 x_4 > + < x_1 x_3 >< x_2 x_4 > + < x_1 x_4 >< x_2 x_3 >,$$

with $< \cdot >$ as usual denoting the expectation operator, or ensemble average. Therefore, the above 9-term expression for the velocity autocorrelation can be expanded into the 27-term expression,

$$\begin{aligned}
I(k_1,&k_2,k_1',k_2',k_1'',k_2'',k_1''',k_2''',\omega,\omega') \\
=&ik_1 ik_1' \left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_1(k_1'',k_2'',d,\omega)\right\rangle \left\langle \hat{F}_m(k_1',k_2',d,\omega')\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle \\
+&ik_1 ik_1' \left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_m(k_1',k_2',d,\omega')\right\rangle \left\langle \hat{F}_1(k_1'',k_2'',d,\omega)\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle \\
+&ik_1 ik_1' \left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle \left\langle \hat{F}_1(k_1'',k_2'',d,\omega)\hat{F}_m(k_1',k_2',d,\omega')\right\rangle \\
+&ik_1 ik_2' \left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_1(k_1'',k_2'',d,\omega)\right\rangle \left\langle \hat{F}_m(k_1',k_2',d,\omega')\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle \\
+&ik_1 ik_2' \left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_m(k_1',k_2',d,\omega')\right\rangle \left\langle \hat{F}_1(k_1'',k_2'',d,\omega)\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle \\
+&ik_1 ik_2' \left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle \left\langle \hat{F}_1(k_1'',k_2'',d,\omega)\hat{F}_m(k_1',k_2',d,\omega')\right\rangle\Big\rangle \\
+&ik_1 \left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_1(k_1'',k_2'',d,\omega)\right\rangle \left\langle \frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\bigg|_{x_3=d} \hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle \\
+&ik_1 \left\langle \hat{F}_j(k_1,k_2,d,\omega)\frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\bigg|_{x_3=d}\right\rangle \left\langle \hat{F}_1(k_1'',k_2'',d,\omega)\hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle
\end{aligned}$$

$$+ik_1\left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle\left\langle \hat{F}_1(k_1'',k_2'',d,\omega)\frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\Big|_{x_3=d}\right\rangle$$

$$+ik_2ik_1'\left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_2(k_1'',k_2'',d,\omega)\right\rangle\left\langle \hat{F}_m(k_1',k_2',d,\omega')\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_2ik_1'\left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_m(k_1',k_2',d,\omega')\right\rangle\left\langle \hat{F}_2(k_1'',k_2'',d,\omega)\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_2ik_1'\left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle\left\langle \hat{F}_2(k_1'',k_2'',d,\omega)\hat{F}_m(k_1',k_2',d,\omega')\right\rangle$$

$$+\left\langle ik_2\hat{F}_j(k_1,k_2,d,\omega)\hat{F}_2(k_1'',k_2'',d,\omega)\right\rangle\left\langle ik_2'\hat{F}_m(k_1',k_2',d,\omega')\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_2\left\langle \hat{F}_j(k_1,k_2,d,\omega)ik_2'\hat{F}_m(k_1',k_2',d,\omega')\right\rangle\left\langle \hat{F}_2(k_1'',k_2'',d,\omega)\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle$$

$$+\left\langle ik_2\hat{F}_j(k_1,k_2,d,\omega)\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle\left\langle ik_2'\hat{F}_m(k_1',k_2',d,\omega')\hat{F}_2(k_1'',k_2'',d,\omega)\right\rangle$$

$$+ik_2\left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_2(k_1'',k_2'',d,\omega)\right\rangle\left\langle \frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_2\left\langle \hat{F}_j(k_1,k_2,d,\omega)\frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\Big|_{x_3=d}\right\rangle\left\langle \hat{F}_2(k_1'',k_2'',d,\omega)\hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_2\left\langle \hat{F}_j(k_1,k_2,d,\omega)\hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle\left\langle \hat{F}_2(k_1'',k_2'',d,\omega)\frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\Big|_{x_3=d}\right\rangle$$

$$+ik_1'\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1'',k_2'',d,\omega)\right\rangle\left\langle \hat{F}_m(k_1',k_2',d,\omega')\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_1'\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_m(k_1',k_2',d,\omega')\right\rangle\left\langle \hat{F}_3(k_1'',k_2'',d,\omega)\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_1'\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_1(k_1''',k_2''',d,\omega')\right\rangle\left\langle \hat{F}_3(k_1'',k_2'',d,\omega)\hat{F}_m(k_1',k_2',d,\omega')\right\rangle$$

$$+ik_2'\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1'',k_2'',d,\omega)\right\rangle\left\langle \hat{F}_m(k_1',k_2',d,\omega')\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_2'\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_m(k_1',k_2',d,\omega')\right\rangle\left\langle \hat{F}_3(k_1'',k_2'',d,\omega)\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle$$

$$+ik_2'\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_2(k_1''',k_2''',d,\omega')\right\rangle\left\langle \hat{F}_3(k_1'',k_2'',d,\omega)\hat{F}_m(k_1',k_2',d,\omega')\right\rangle$$

$$+\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1'',k_2'',d,\omega)\right\rangle\left\langle \frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\Big|_{x_3=d}\hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle$$

$$+\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\Big|_{x_3=d}\frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\Big|_{x_3=d}\right\rangle\left\langle \hat{F}_3(k_1'',k_2'',d,\omega)\hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle$$

$$+\left\langle \frac{\partial \hat{F}_j(k_1,k_2,x_3,\omega)}{\partial x_3}\bigg|_{x_3=d} \hat{F}_3(k_1''',k_2''',d,\omega')\right\rangle \left\langle \hat{F}_3(k_1'',k_2'',d,\omega)\frac{\partial \hat{F}_m(k_1',k_2',x_3,\omega')}{\partial x_3}\bigg|_{x_3=d}\right\rangle .$$

Appendix 4 to Chapter IV

This appendix lists the 17 equations determining A_i, A'_i, B_i, B'_i $(i = 1,2,3,4)$ and $\hat{\eta}$. These are the "constants" (actually functions of Fourier-transformed variables) appearing in the general solution for the velocity field (driven by the random stress) that must be determined:

$$A_4 + A'_4 = -i\omega\hat{\eta}; \tag{A4.1}$$

$$B_4 + B'_4 = -i\omega\hat{\eta}; \tag{A4.2}$$

$$A_2 + A'_2 = B_2 + B'_2; \tag{A4.3}$$

$$A_3 + A'_3 = B_3 + B'_3; \tag{A4.4}$$

$$\begin{aligned} &ik_1\lambda A'_4 + ik_1\lambda A_4 + \sqrt{c_1}\lambda A'_2 - \sqrt{c_1}\lambda A_2 + \hat{s}^{(1)}_{13}(0) \\ &= ik_1 B'_4 + ik_1 B_4 - \sqrt{c_2}B'_2 + \sqrt{c_2}B_2 + \hat{s}^{(2)}_{13}(0); \end{aligned} \tag{A4.5}$$

and

$$\begin{aligned} &ik_2\lambda A_4 + ik_2\lambda A'_4 + \sqrt{c_1}\lambda A'_3 - \sqrt{c_1}\lambda A_3 + \hat{s}^{(1)}_{23}(0) \\ &= ik_2 B_4 + ik_2 B'_4 - \sqrt{c_2}B'_3 + \sqrt{c_2}B_3 + \hat{s}^{(2)}_{23}(0); \end{aligned} \tag{A4.6}$$

$$\begin{aligned} &-A_1 + \sqrt{c_1}\lambda A'_4 - \sqrt{c_1}\lambda A_4 + B_1 + \sqrt{c_2}B'_4 - \sqrt{c_2}B_4 + \hat{s}^{(1)}_{33}(0) - \hat{s}^{(2)}_{33}(0) \\ &= \Gamma(k_1^2 + k_2^2)\hat{\eta} + \beta\hat{\eta}; \end{aligned} \tag{A4.7}$$

$$A_1' = 0; \tag{A4.8}$$

$$B_1' = 0; \tag{A4.9}$$

$$A_2' = -\frac{ik_1 A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_p})}; \tag{A4.10}$$

$$A_3' = -\frac{ik_2 A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_p})}; \tag{A4.11}$$

$$A_4' = \frac{\sqrt{c_p} A_1}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_p})}; \tag{A4.12}$$

$$B_2' = \frac{ik_1 B_1}{2\sqrt{c_2}(\sqrt{c_2}+\sqrt{c_p})}; \tag{A4.13}$$

$$B_3' = \frac{ik_2 B_1}{2\sqrt{c_2}(\sqrt{c_2}+\sqrt{c_p})}; \tag{A4.14}$$

$$B_4' = -\frac{\sqrt{c_p} B_1}{2\sqrt{c_2}(\sqrt{c_2}+\sqrt{c_p})}. \tag{A4.15}$$

$$ik_1 A_2 + ik_2 A_3 - \sqrt{c_1} A_4 + \frac{\sqrt{c_P}}{2\lambda\sqrt{c_1}} A_1 = 0; \tag{A4.16}$$

$$\begin{aligned} &ik_1 B_2 + ik_2 B_3 + \sqrt{c_2} B_4 \\ &+ \frac{c_P}{2\sqrt{c_2}} B_1 = 0. \end{aligned} \tag{A4.17}$$

Appendix 5 to Chapter IV

This appendix gives the solution to the system of 17 algebraic equations listed in Appendix 4:

$$\hat{\eta} = \frac{a_{12}}{a_{11}}\Delta_1 + \frac{a_{13}}{a_{11}}\Delta_2 - \frac{1}{a_{11}}\Delta_3; \qquad (A5.1)$$

$$A_1 = a_{14}\Delta_1 + a_{15}\Delta_2 + a_{16}\Delta_3; \qquad (A5.2)$$

$$B_1 = a_{17}\Delta_1 + a_{18}\Delta_2 + a_{19}\Delta_3; \qquad (A5.3)$$

$$A_2 = a_{20}\Delta_1 + a_{21}\Delta_2 + a_{22}\Delta_3; \qquad (A5.4)$$

$$B_2 = a_{23}\Delta_1 + a_{24}\Delta_2 + a_{25}\Delta_3; \qquad (A5.5)$$

$$A_3 = a_{26}\Delta_1 + a_{27}\Delta_2 + a_{28}\Delta_3; \qquad (A5.6)$$

$$B_3 = a_{29}\Delta_1 + a_{30}\Delta_2 + a_{31}\Delta_3; \qquad (A5.7)$$

$$A_4 = -i\omega\hat{\eta} - \frac{\sqrt{c_P}}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_P})}[a_{14}\Delta_1 + a_{15}\Delta_2 + a_{16}\Delta_3]; \qquad (A5.8)$$

$$B_4 = -i\omega\hat{\eta} + \frac{\sqrt{c_P}}{2\sqrt{c_2}(\sqrt{c_1} + \sqrt{c_P})}[a_{17}\Delta_1 + a_{18}\Delta_2 + a_{19}\Delta_3]; \qquad (A5.9)$$

$$A_1' = 0; \qquad (A5.10)$$

$$B_1' = 0; \qquad (A5.11)$$

$$A_2' = -\frac{ik_1}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_P})}[a_{14}\Delta_1 + a_{15}\Delta_2 + a_{16}\Delta_3]; \qquad (A5.12)$$

$$B_2' = \frac{ik_1}{2\lambda\sqrt{c_2}(\sqrt{c_1}+\sqrt{c_P})}[a_{17}\Delta_1 + a_{18}\Delta_2 + a_{19}\Delta_3]; \qquad (A5.13)$$

$$A_3' = -\frac{ik_2}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_P})}[a_{14}\Delta_1 + a_{15}\Delta_2 + a_{16}\Delta_3]; \qquad (A5.14)$$

$$B_3' = \frac{ik_2}{2\lambda\sqrt{c_2}(\sqrt{c_1}+\sqrt{c_P})}[a_{17}\Delta_1 + a_{18}\Delta_2 + a_{19}\Delta_3]; \qquad (A5.15)$$

$$A_4' = \frac{\sqrt{c_P}}{2\lambda\sqrt{c_1}(\sqrt{c_1}+\sqrt{c_P})}[a_{14}\Delta_1 + a_{15}\Delta_2 + a_{16}\Delta_3]; \qquad (A5.16)$$

$$B_4' = -\frac{\sqrt{c_p}}{2\lambda\sqrt{c_2}(\sqrt{c_1}+\sqrt{c_P})}[a_{17}\Delta_1 + a_{18}\Delta_2 + a_{19}\Delta_3]; \qquad (A5.17)$$

where

$$a_1 = \frac{c_P}{(\sqrt{c_1}+\sqrt{c_P})(\sqrt{c_2}+\lambda\sqrt{c_1})} + \frac{\sqrt{c_P}}{\lambda(\sqrt{c_1}+\sqrt{c_P})}; \qquad (A5.18)$$

$$a_2 = -\frac{c_P}{(\sqrt{c_2}+\sqrt{c_P})(\sqrt{c_2}+\lambda\sqrt{c_1})}; \qquad (A5.19)$$

$$a_3 = -\frac{i\omega(\lambda - 1)c_P}{\sqrt{c_2} + \lambda\sqrt{c_1}} + i\omega\sqrt{c_1}; \tag{A5.20}$$

$$a_4 = -\frac{ik_1}{\sqrt{c_2} + \lambda\sqrt{c_1}}; \tag{A5.21}$$

$$a_5 = -\frac{ik_2}{\sqrt{c_2} + \lambda\sqrt{c_1}}; \tag{A5.22}$$

$$a_6 = \frac{c_P}{(\sqrt{c_2} + \lambda\sqrt{c_1})(\sqrt{c_1} + \sqrt{c_P})}; \tag{A5.23}$$

$$a_7 = \frac{\sqrt{c_P}}{2\sqrt{c_2}} + \frac{c_P(\sqrt{c_1}\lambda - \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})(\sqrt{c_2} + \lambda\sqrt{c_1})} + \frac{\sqrt{c_2}\sqrt{c_P}}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})}; \tag{A5.24}$$

$$a_8 = -\frac{ic_P\omega(\lambda - 1)}{\sqrt{c_2} + \lambda\sqrt{c_1}} + i\omega\sqrt{c_2}; \tag{A5.25}$$

$$a_9 \ (= a_4) \ = -\frac{ik_1}{\sqrt{c_2} + \lambda\sqrt{c_1}}; \tag{A5.26}$$

$$a_{10} \ (= a_5) \ = -\frac{ik_2}{\sqrt{c_2} + \lambda\sqrt{c_1}}; \tag{A5.27}$$

$$a_{11} = -\frac{\sqrt{c_1}}{\sqrt{c_1} + \sqrt{c_P}} \frac{a_7 a_3 - a_2 a_8}{a_1 a_7 - a_2 a_6} + \frac{\sqrt{c_2}}{\sqrt{c_2} + \sqrt{c_P}} \frac{a_1 a_8 - a_6 a_3}{a_1 a_7 - a_2 a_6}$$
$$+ i\omega(\sqrt{c_2} + \lambda\sqrt{c_1}) - \Gamma c_P - \beta; \tag{A5.28}$$

$$a_{12} = \frac{\sqrt{c_1}}{\sqrt{c_1} + \sqrt{c_P}} \frac{a_7 a_4 - a_2 a_9}{a_1 a_7 - a_2 a_6} - \frac{\sqrt{c_2}}{\sqrt{c_2} + \sqrt{c_P}} \frac{a_1 a_9 - a_6 a_4}{a_1 a_7 - a_2 a_6}; \tag{A5.29}$$

$$a_{13} = \frac{\sqrt{c_1}}{\sqrt{c_1} + \sqrt{c_P}} \frac{a_7 a_5 - a_2 a_{10}}{a_1 a_7 - a_2 a_6} - \frac{\sqrt{c_2}}{\sqrt{c_2} + \sqrt{c_P}} \frac{a_1 a_{10} - a_6 a_5}{a_1 a_7 - a_2 a_6}; \qquad (A5.30)$$

$$a_{14} = \frac{a_7 a_4 - a_2 a_9 + \frac{a_{12}}{a_{11}}(a_7 a_3 - a_2 a_8)}{a_1 a_7 - a_2 a_6}; \qquad (A5.31)$$

$$a_{15} = \frac{a_7 a_5 - a_2 a_{10} + \frac{a_{13}}{a_{11}}(a_7 a_3 - a_2 a_8)}{a_1 a_7 - a_2 a_6}; \qquad (A5.32)$$

$$a_{16} = \frac{a_2 a_8 - a_7 a_3}{a_{11}(a_1 a_7 - a_2 a_6)}; \qquad (A5.33)$$

$$a_{17} = \frac{a_1 a_9 - a_6 a_4 + \frac{a_{12}}{a_{11}}(a_1 a_8 - a_6 a_3)}{a_1 a_7 - a_2 a_6}; \qquad (A5.34)$$

$$a_{18} = \frac{a_1 a_{10} - a_6 a_5 + \frac{a_{13}}{a_{11}}(a_1 a_8 - a_6 a_3)}{a_1 a_7 - a_2 a_6}; \qquad (A5.35)$$

$$a_{19} = \frac{a_6 a_3 - a_1 a_8}{a_{11}(a_1 a_7 - a_2 a_6)}; \qquad (A5.36)$$

$$a_{20} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[1 + \frac{a_{12}}{a_{11}} k_1 \omega(\lambda - 1) + \frac{ik_1(1 - 2\lambda\sqrt{c_1})}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_P})} a_{14} \right.$$
$$\left. + \frac{ik_1(1 - \sqrt{c_1}\lambda + \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})} a_{17} \right]; \qquad (A5.37)$$

$$a_{21} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[\frac{a_{13}}{a_{11}} k_1 \omega(\lambda - 1) + \frac{ik_1(1 - 2\lambda\sqrt{c_1})}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_P})} a_{15} \right.$$
$$+ \frac{ik_1(1 - \sqrt{c_1}\lambda + \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P}} a_{18}; \qquad (A5.38)$$

$$a_{22} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[-\frac{1}{a_{11}} k_1 \omega(\lambda - 1) + \frac{ik_1(1 - 2\lambda\sqrt{c_1})}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_P})} a_{16} \right.$$

$$\left. + \frac{ik_1(1 - \sqrt{c_1}\lambda + \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P}} a_{19}; \right. \qquad (A5.39)$$

$$a_{23} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[1 + \frac{a_{12}}{a_{11}} k_1 \omega(\lambda - 1) - \frac{ik_1}{(\sqrt{c_1} + \sqrt{c_P})} a_{14} \right.$$

$$\left. - \frac{ik_1(\sqrt{c_1}\lambda - \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})} a_{17} \right]; \qquad (A5.40)$$

$$a_{24} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[\frac{a_{13}}{a_{11}} k_1 \omega(\lambda - 1) - \frac{ik_1}{(\sqrt{c_1} + \sqrt{c_P})} a_{15} \right.$$

$$\left. - \frac{ik_1(\sqrt{c_1}\lambda - \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})} a_{18} \right]; \qquad (A5.41)$$

$$a_{25} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[-\frac{1}{a_{11}} k_1 \omega(\lambda - 1) - \frac{ik_1}{(\sqrt{c_1} + \sqrt{c_P})} a_{16} \right.$$

$$\left. - \frac{ik_1(\sqrt{c_1}\lambda - \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})} a_{19} \right]; \qquad (A5.42)$$

$$a_{26} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[\frac{a_{12}}{a_{11}} k_2 \omega(\lambda - 1) + \frac{ik_2(1 - 2\lambda\sqrt{c_1})}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_P})} a_{14} \right.$$

$$\left. + \frac{ik_2(1 - \sqrt{c_1}\lambda + \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})} a_{17} \right]; \qquad (A5.43)$$

$$a_{27} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[1 + \frac{a_{13}}{a_{11}} k_2 \omega(\lambda - 1) + \frac{ik_2(1 - 2\lambda\sqrt{c_1})}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_P})} a_{15} \right.$$

$$\left. + \frac{ik_2(1 - \sqrt{c_1}\lambda + \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P}} a_{18}; \right. \qquad (A5.44)$$

$$a_{28} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[-\frac{1}{a_{11}} k_2 \omega(\lambda - 1) + \frac{ik_2(1 - 2\lambda\sqrt{c_1})}{2\lambda\sqrt{c_1}(\sqrt{c_1} + \sqrt{c_P})} a_{16} \right.$$

$$\left. + \frac{ik_2(1 - \sqrt{c_1}\lambda + \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P}} a_{19}; \right. \tag{A5.45}$$

$$a_{29} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[\frac{a_{12}}{a_{11}} k_2 \omega(\lambda - 1) - \frac{ik_2}{(\sqrt{c_1} + \sqrt{c_P})} a_{14} \right.$$

$$\left. - \frac{ik_2(\sqrt{c_1}\lambda - \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})} a_{17} \right]; \tag{A5.46}$$

$$a_{30} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[\frac{1 + a_{13}}{a_{11}} k_2 \omega(\lambda - 1) - \frac{ik_2}{(\sqrt{c_1} + \sqrt{c_P})} a_{15} \right.$$

$$\left. - \frac{ik_2(\sqrt{c_1}\lambda - \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})} a_{18} \right]; \tag{A5.47}$$

$$a_{31} = \frac{1}{\sqrt{c_2} + \lambda\sqrt{c_1}} \left[-\frac{1}{a_{11}} k_2 \omega(\lambda - 1) - \frac{ik_2}{(\sqrt{c_1} + \sqrt{c_P})} a_{16} \right.$$

$$\left. - \frac{ik_2(\sqrt{c_1}\lambda - \sqrt{c_2})}{2\sqrt{c_2}(\sqrt{c_2} + \sqrt{c_P})} a_{19} \right]. \tag{A5.48}$$

Appendix 6 to Chapter IV

This appendix gives the solution for the constants appearing in the expression for the quasi-steady flow field that results when the interface initially has a specified deformation $\eta(\mathbf{x})$.

$$A_1 = -\frac{8\lambda(\lambda+1)}{8\lambda^2+\lambda+3}(\Gamma c_P + \beta)\eta; \tag{A6.1}$$

$$A_2 = -\frac{2ik_1(3+\lambda)}{8\lambda^2+\lambda+3}\frac{\Gamma c_P + \beta}{c_P}\eta; \tag{A6.2}$$

$$A_3 = -\frac{2ik_2(3+\lambda)}{(8\lambda^2+\lambda+3)c_P}\eta; \tag{A6.3}$$

$$A_4 = \frac{2(1-\lambda)}{(8\lambda^2+\lambda+3)}\frac{(\Gamma c_P + \beta)}{\sqrt{c_P}}\eta; \tag{A6.4}$$

$$B_1 = \frac{16\lambda}{(8\lambda^2+\lambda+3)}(\Gamma c_P + \beta)\eta; \tag{A6.5}$$

$$B_2 = -(ik_1)(7\lambda-3)(\Gamma c_P + \beta)(8\lambda^2+\lambda+3)c_P\eta; \tag{A6.6}$$

$$B_3 = -(ik_2)(7\lambda-3)(\Gamma c_P + \beta)(8\lambda^2+\lambda+3)c_P\eta; \tag{A6.7}$$

$$B_4 = \frac{3(5\lambda-1)}{(8\lambda^2+\lambda+3)}\frac{\Gamma c_P + \beta}{\sqrt{c_P}}\eta; \tag{A6.8}$$

The constants A_1', A_2', A_3', A_4', B_1', B_2', B_3', and B_4' are given by equations (A4.8) through (A4.15) of Appendix 4, combined with the above equations (A6.1) through (A6.8). (For brevity, they will not be written explicitly here).

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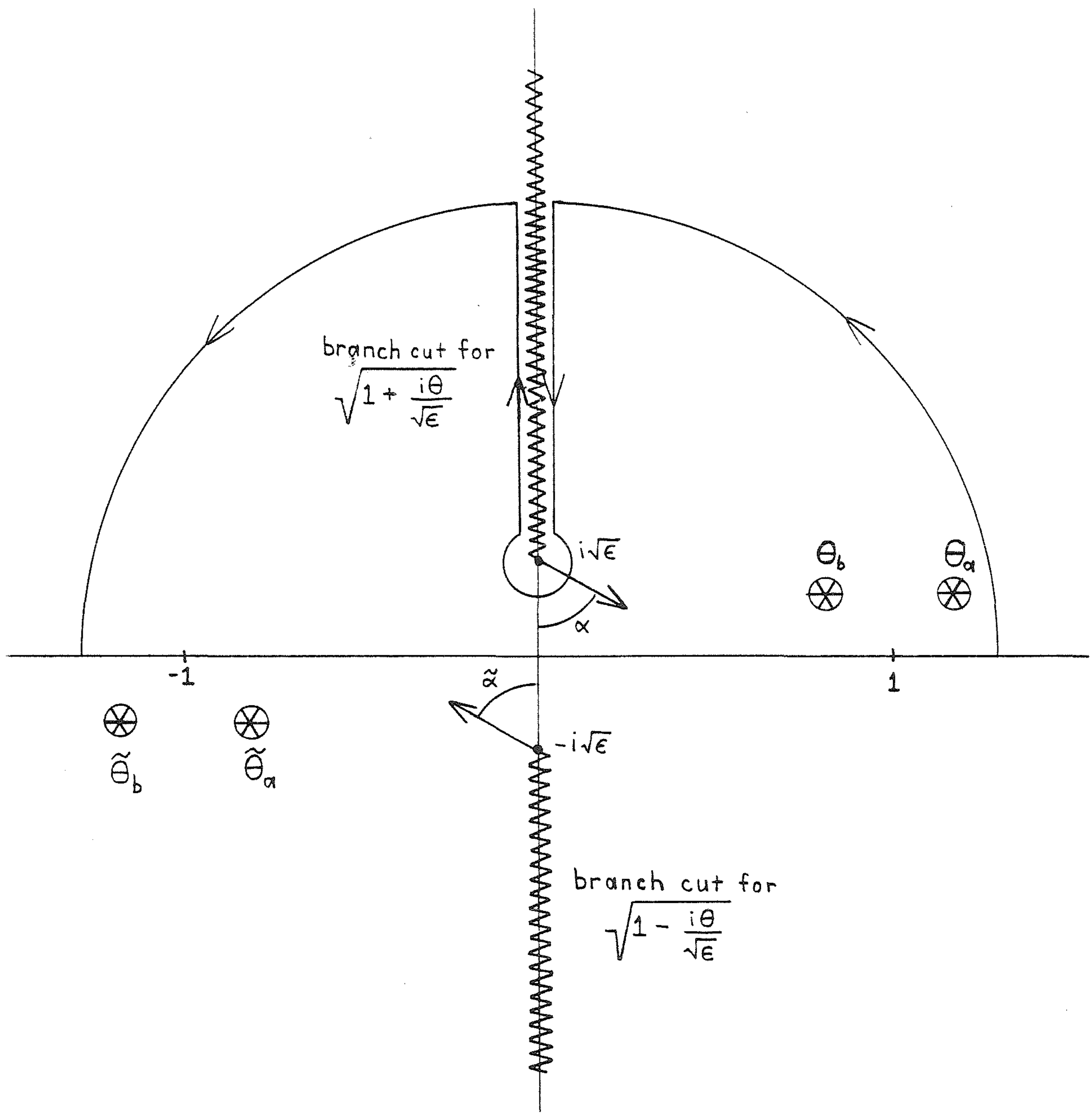
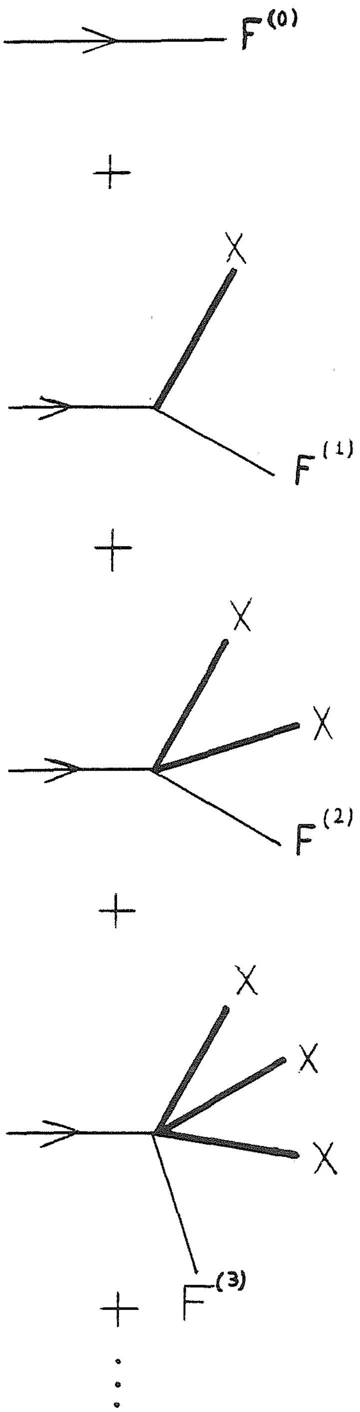


FIGURE 1

The θ-plane showing the contour of integration and the poles.



$$\int G \underline{F}$$

$$+$$

$$\int G \nabla \underline{F} \cdot (\underline{X} - \underline{X}_0)$$

$$+$$

$$\int G \nabla \nabla \underline{F} : (\underline{X} - \underline{X}_0)^2$$

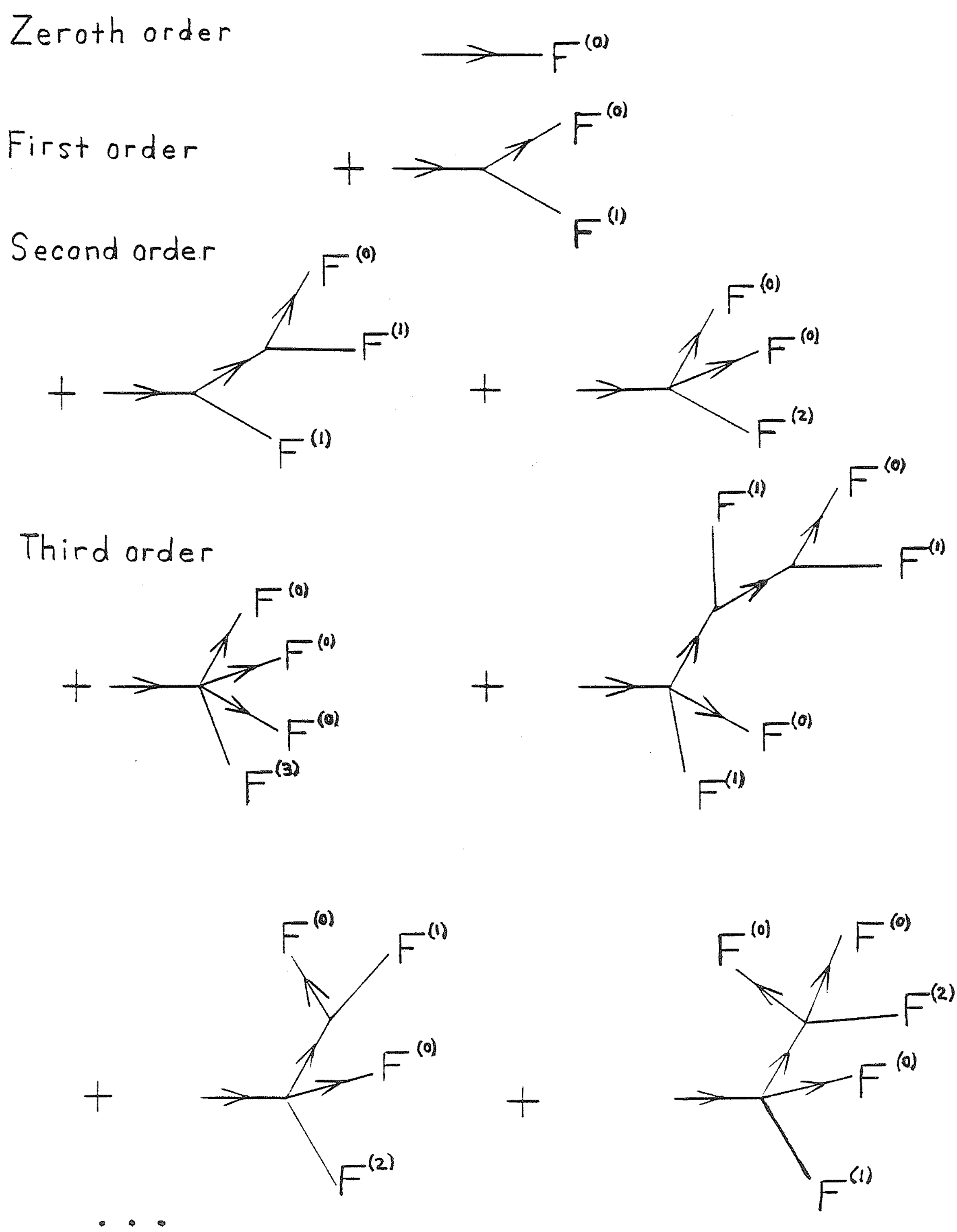
$$+$$

$$\int G \nabla \nabla \nabla \underline{F} \,\vdots\, (\underline{X} - \underline{X}_0)^3$$

$$+$$

$$\vdots$$

FIGURE 2


Zeroth order
F(0)
First order
F(0)
F(1)
Second order
F(0)
F(1)
F(1)
F(0)
F(0)
F(2)
Third order
F(0)
F(0)
F(0)
F(3)
F(1)
F(0)
F(1)
F(0)
F(1)
F(0)
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F(1)
. . .

FIGURE 3

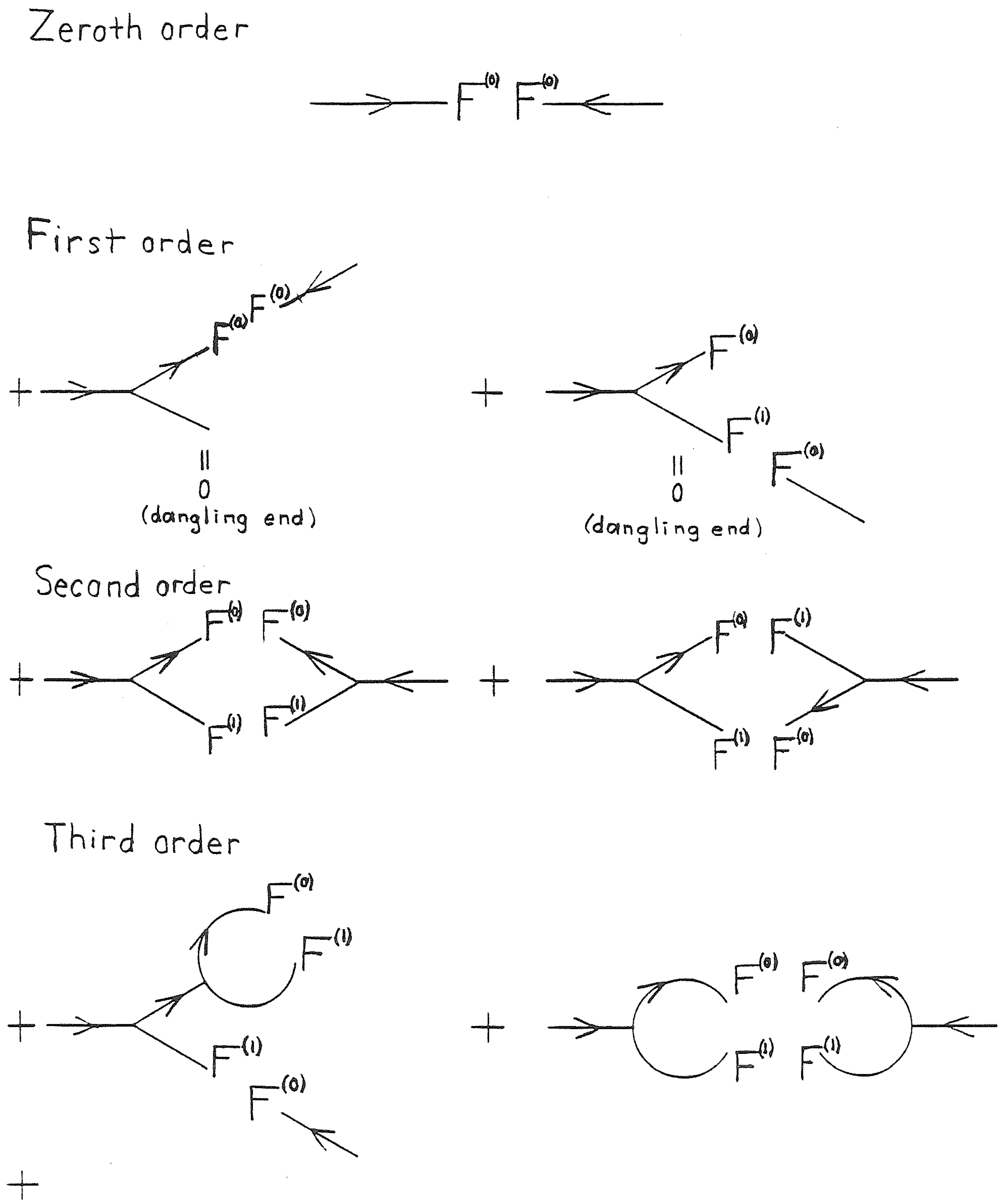
Zeroth order
First order
= 0
(dangling end)
= 0
(dangling end)
Second order
Third order

FIGURE 4

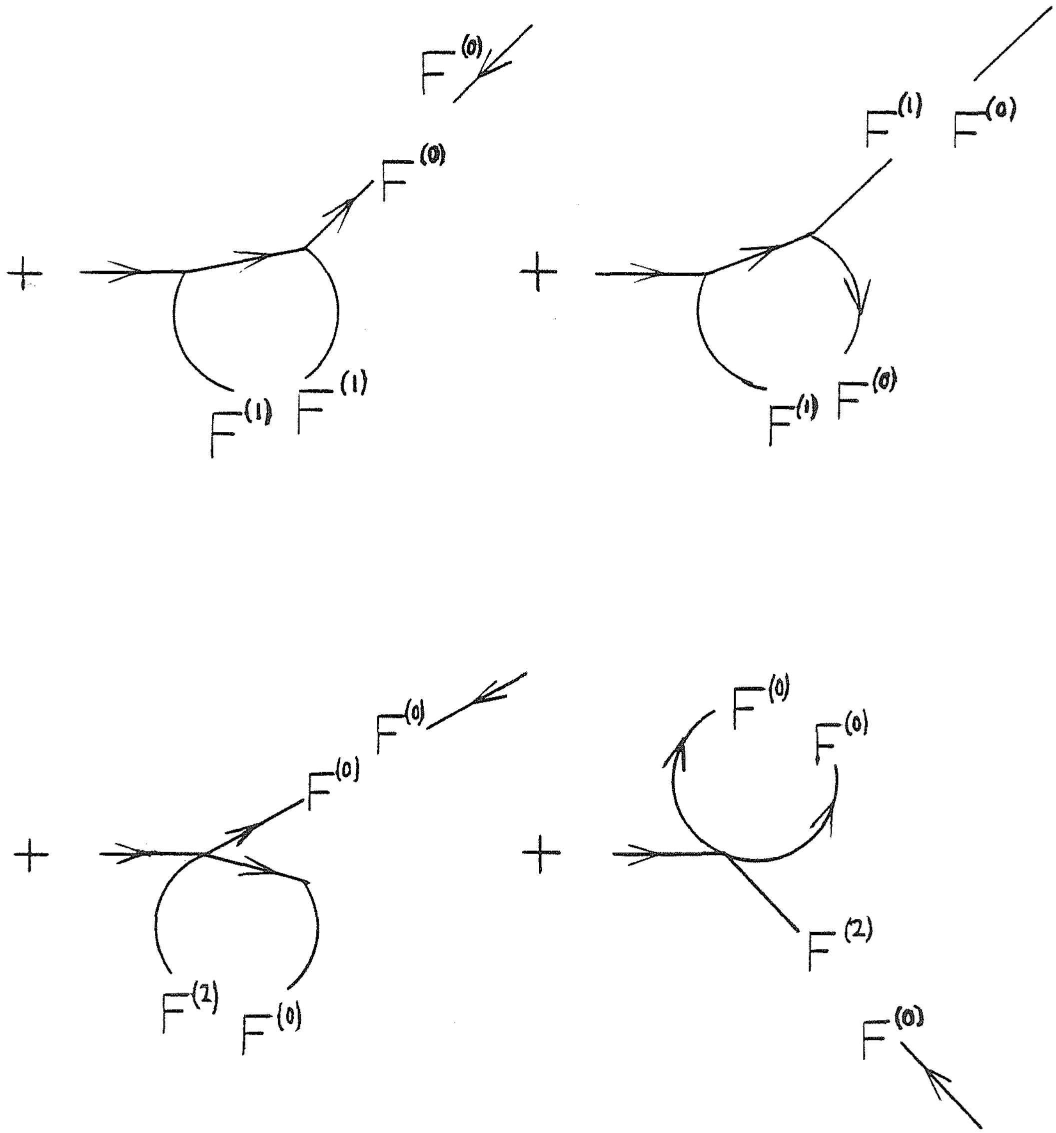

FIGURE 4 CTD.

CHAPTER V

On Unsteady Heat Conduction and Unsteady Stokes Flow with Two Spheres

While steady heat conduction and zero-Reynolds-number fluid flow around two spheres have been investigated by exploiting the separability of Laplace's equation in bipolar coordinates, the corresponding unsteady problems seem only to have been investigated by approximating the spheres as point forces, or by using the method of reflections, on account of the failure of the Helmholtz equation to separate in bipolar coordinates. There is reason to believe that more accurate hydrodynamics will show some effects missing in these solutions. For example, Hassonjee, Ganatos and Pfeffer [1] have solved for steady Stokes flow around clusters of more than two spherical particles by using addition theorems for spherical harmonics to expand the eigenfunctions in one coordinate system in terms of the other, with each particle having a spherical coordinate system centered on it. (This general idea was probably first used in electromagnetic theory for problems with two spheres, and it appeared in Jeffery's [2] paper on heat conduction). Hassonjee, et al. [1] found "multiparticle interaction effects ... which would not be present if only pair interactions of the particles were considered." While their solution method is still approximate in a sense, namely, in that only a finite number of eigenfunctions can be used in the solution expansion, it seems to approach the exact solution within a reasonable amount of computational time.

This general approach of using more than one coordinate system can also be used for unsteady Stokes flow, since it is possible to expand the eigenfunctions of the Helmholtz equation in one spherical coordinate system in terms of the eigenfunctions in the other spherical coordinate system. This should have application to suspensions of spherical particles moving with small amplitude but at high frequencies.

In general, these expansions of eigenfunctions in one coordinate system in terms of eigenfunctions in another coordinate system lead to an infinite *nontriangular* system of algebraic equations for the coefficients, so that it is not possible to solve for the first n coefficients without making the approximation that all the remaining ones are zero. It is in this sense that the solution is not quite "exact," although as the number of coefficients solved for is increased, any desired accuracy can be achieved. It should also be noted that for any given configuration, the linear system has only to be solved *once*, and the result is an essentially analytic solution for the entire flow field with some constants appearing in it that were determined numerically. Such a solution has the advantage that it gives complete information about the flow, and can be differentiated any number of times without losing accuracy.

The fact that the infinite linear system is nontriangular is arguably an artifact of the eigenfunction basis chosen, since it is possible to choose a particular basis for the eigenfunctions to get a triangular system. This change of basis is actually just another way of viewing the "LDU" decomposition that is possible for any matrix. For suppose that the vector $\mathbf{b}$ is the vector of unknown coefficients in the general solution, to be determined from the boundary conditions. The solution $\mathbf{u}$ is then given by

$$\mathbf{u} = \mathbf{b} \cdot \mathbf{x}, \tag{1.1}$$

where $\mathbf{x}$ is the function basis chosen for the expansion of the solution. (For example, $\mathbf{x}$ might be a vector of spherical harmonics). Then application of the boundary conditions yields a system of equations

$$\mathbf{M} \cdot \mathbf{b} = \mathbf{f}, \tag{1.2}$$

where $\mathbf{M}$ is a known matrix, and $\mathbf{f}$ is a vector of boundary data. By decomposing $\mathbf{M}$ into the product of a lower and upper diagonal matrix, it can be seen that

$$\mathbf{L} \cdot [\mathbf{U} \cdot \mathbf{b}] = \mathbf{f}. \tag{1.3}$$

Now if the new basis

$$\mathbf{x}' = [\mathbf{U}^{-1}]^T \cdot bfx, \tag{1.4}$$

is chosen, then the solution can be rewritten as

$$\mathbf{u} = \mathbf{b}' \cdot \mathbf{x}', \tag{1.5}$$

where the new coefficient matrix $\mathbf{b}'$ satisfies

$$\mathbf{L} \cdot \mathbf{b} = \mathbf{f}, \tag{1.6}$$

a lower triangular system. Since in general these vectors and matrices will be infinite-dimensional, it is important to note several things. First, the decomposition of a matrix into the product $\mathbf{L} \cdot \mathbf{U}$ can be done recursively, starting with the first entries, so that it is possible to do the decomposition up to the first $n \times n$ block by knowing only the first $n \times n$ block of the matrix $\mathbf{M}$. Secondly, the new basis is related to the old basis by the lower triangular matrix $[\mathbf{U}^{-1}]^T$, which again can be determined up to the first $n \times n$ block by knowing only the first $n \times n$ block of $\mathbf{U}$ so that the first n elements of the basis can be determined by knowing only the first $n \times n$ block of $\mathbf{M}$.

Infinite-dimensional systems arising from expanding solutions in one coordinate system in terms of another coordinate system have been encountered by other researchers. For example, Haberman and Sayre [3] obtain an "exact analytical solution" for the motion of a spherical particle in a cylindrical wall, in the form of a series for the stream function where the coefficients are the solutions of an infinite (square) system of algebraic equations. Like the solution of Hassonjee et al. for several spheres, it is not quite exact in the sense that it cannot be written explicitly in terms of known functions. However, they are able to obtain an excellent approximation by solving for the first N coefficients, where $N = O(10)$. Systems like this are inherently ill-posed, however, in the sense that the solution vector elements must get progressively smaller in moving down the column (a necessary condition for the series to converge). As more and more coefficients are simultaneously solved for, this imbalance in the magnitude of the unknowns (and the coefficient matrix) will create more and more problems. While this difficulty could be overcome by rescaling (i.e., renormalizing the basis functions), the above approach of changing the basis to make the system triangular seems more natural.

UNSTEADY HEAT CONDUCTION WITH TWO SPHERES

The problem of unsteady heat conduction with two spheres is interesting in its own right and is simpler than the unsteady Stokes flow problem, so it will be considered first here, to illustrate the general idea of change of basis. Rather than first derive the infinite system of algebraic equations, and then decompose it, the basis will be chosen "along the way." For further simplicity, only the axisymmetric case will be considered here, although it will be clear how to extend this analysis to the nonaxisymmetric case. Two spherical coordinate systems (r_1, θ_1) and (r_2, θ_2), with centers separated by a distance R, will be used (Figure 1). Since it is consistent to use the unsteady Stokes equations only for flows at high frequency where the particle displacement is small, R will be assumed constant in the following. Sphere

1 (2), of radius L_1 (L_2), will be at the origin of system 1 (2). The heat equation can be Fourier-transformed to yield the Helmholtz equation

$$\nabla^2 u + h^2 u = 0, \tag{1.7}$$

and the eigenfunction basis for this problem obtained by separation of variables (with no ϕ dependence) in spherical coordinate system 1 is $\{u_n^{(1)}\}$ and $\{\tilde{u}_n^{(1)}\}$, where

$$u_n^{(1)} = r_1^{-1/2} J_{n+\frac{1}{2}}(hr_1) P_n(\cos\theta_1); \tag{1.8a}$$

$$\tilde{u}_n^{(1)} = r_1^{-1/2} H_{n+\frac{1}{2}}^{(2)}(hr_1) P_n(\cos\theta_1). \tag{1.8b}$$

Similarly, the eigenfunction basis obtained by separation of variables in spherical coordinate system 2 is $\{u_n^{(2)}\}$ and $\{\tilde{u}_n^{(2)}\}$, where

$$u_n^{(2)} = r_2^{-1/2} J_{n+\frac{1}{2}}(hr_2) P_n(\cos\theta_2); \tag{1.8c}$$

$$\tilde{u}_n^{(2)} = r_2^{-1/2} H_{n+\frac{1}{2}}^{(2)}(hr_2) P_n(\cos\theta_2). \tag{1.8d}$$

The Bessel functions of the first kind, $J_{n+\frac{1}{2}}(hr)$, are bounded at $r = 0$, and go to zero as $r \to \infty$. The Hankel functions of the second kind, $H_{n+\frac{1}{2}}^{(2)}(hr)$, diverge as $r \to 0$, but go to zero as $r \to \infty$. Since the eigenfunctions form a complete set for solutions of the equation (subject to the appropriate boundedness and continuity restrictions), the eigenfunctions of coordinate system 1 must possess an expansion in terms of the eigenfunctions of coordinate system 2 and vice versa. However, the expansion for the Hankel functions of coordinate system 1, which have a singularity at $r_1 = 0$, must involve *only* Bessel functions in coordinate system 2, since they do *not* have singularities at $r_2 = 0$. In other words,

$$r_1^{-1/2} H^{(2)}_{n+\frac{1}{2}}(hr_1) P_n(\cos\theta_1) = \sum_{m=0}^{\infty} c_{n,m} r_2^{-1/2} J_{m+\frac{1}{2}}(hr_2) P_m(\cos\theta_2). \tag{1.9a}$$

Likewise, the Bessel functions of coordinate system 1, which do not have any singularities, must have an expansion in coordinate system 2 involving only Bessel functions:

$$r_1^{-1/2} J_{n+\frac{1}{2}}(hr_1) P_n(\cos\theta_1) = \sum_{m=0}^{\infty} \tilde{c}_{n,m} r_2^{-1/2} J_{m+\frac{1}{2}}(hr_2) P_m(\cos\theta_2). \tag{1.9b}$$

These two expansions can be written as

$$u_n^{(1)} = \sum_{m=0}^{\infty} c_{n,m} u_m^{(2)}, \tag{1.10a}$$

$$\tilde{u}_n^{(1)} = \sum_{m=0}^{\infty} \tilde{c}_{n,m} u_m^{(2)}. \tag{1.10b}$$

Instead of expanding in the basis $\{u_n^{(1)}\}$ and $\{\tilde{u}_n^{(1)}\}$, the solution will be expanded in the special basis $\{q_n^{(1)}\}$ and $\{\tilde{q}_n^{(1)}\}$, where

$$q_n^{(1)} = \sum_{m=0}^{n} [a_{n,m} u_n^{(1)} + \tilde{a}_{n,m} \tilde{u}_n^{(1)}], \tag{1.11a}$$

$$\tilde{q}_n^{(1)} = \sum_{m=0}^{n} [d_{n,m} u_n^{(1)} + \tilde{d}_{n,m} \tilde{u}_n^{(1)}]. \tag{1.11b}$$

The constants $a_{n,m}$, $\tilde{a}_{n,m}$, $d_{n,m}$ and $\tilde{d}_{n,m}$, which can be considered as elements of lower triangular matrices $\mathbf{A}$, $\tilde{\mathbf{A}}$, $\mathbf{D}$, and $\tilde{\mathbf{D}}$, will be chosen below to satisfy the desirable transformation property. Thus, the solution will be expressed as

$$u = \sum_{n=0}^{\infty} b_n q_n^{(1)} + \sum_{n=0}^{\infty} \tilde{b}_n \tilde{q}_n^{(1)}$$

$$= \sum_{n=0}^{\infty} b_n \sum_{m=0}^{n} [a_{n,m} u_n^{(1)} + \tilde{a}_{n,m} \tilde{u}_n^{(1)}] + \sum_{n=0}^{\infty} \tilde{b}_n \sum_{m=0}^{n} [d_{n,m} u_n^{(1)} + \tilde{d}_{n,m} \tilde{u}_n^{(1)}]. \qquad (1.12)$$

This form can be used to apply boundary conditions on sphere 1. For applying boundary conditions on sphere 2, the expansions (1.10) can be used, to give

$$u = \sum_{n=0}^{\infty} b_n q_n^{(1)} + \sum_{n=0}^{\infty} \tilde{b}_n \tilde{q}_n^{(1)}$$

$$= \sum_{n=0}^{\infty} b_n \sum_{m=0}^{n} [a_{n,m} \sum_{j=0}^{\infty} c_{m,j} u_j^{(2)} + \tilde{a}_{n,m} \sum_{j=0}^{\infty} \tilde{c}_{m,j} u_j^{(2)}]$$

$$+ \sum_{n=0}^{\infty} \tilde{b}_n \sum_{m=0}^{n} [d_{n,m} \sum_{j=0}^{\infty} c_{m,j} u_j^{(2)} + \tilde{d}_{n,m} \sum_{j=0}^{\infty} \tilde{c}_{m,j} u_j^{(2)}]. \qquad (1.13)$$

Thus, the solution in coordinate system 1 is

$$u = \sum_{n=0}^{\infty} b_n \sum_{m=0}^{n} [a_{n,m} r_1^{-1/2} J_{m+\frac{1}{2}}(hr_1) P_m(\cos\theta_1) + \tilde{a}_{n,m} r_1^{-1/2} H_{m+\frac{1}{2}}^{(2)}(hr_1) P_m(\cos\theta_1)]$$

$$+ \sum_{n=0}^{\infty} \tilde{b}_n \sum_{m=0}^{n} [d_{n,m} r_1^{-1/2} J_{m+\frac{1}{2}}(hr_1) P_m(\cos\theta_1) + \tilde{d}_{n,m} r_1^{-1/2} H_{m+\frac{1}{2}}^{(2)}(hr_1) P_m(\cos\theta_1)], \qquad (1.14a)$$

and the solution in coordinate system 2 is

$$u = \sum_{n=0}^{\infty} b_n \sum_{m=0}^{n} [a_{n,m} \sum_{j=0}^{\infty} c_{m,j} + \tilde{a}_{n,m} \sum_{j=0}^{\infty} \tilde{c}_{m,j}] r_2^{-1/2} J_{j+\frac{1}{2}}(hr_2) P_j(\cos\theta_2)$$

$$+\sum_{n=0}^{\infty}\tilde{b}_n\sum_{m=0}^{n}[d_{n,m}\sum_{j=0}^{\infty}c_{m,j}+\tilde{d}_{n,m}\sum_{j=0}^{\infty}\tilde{c}_{m,j}]r_2^{-1/2}J_{j+\frac{1}{2}}(hr_2)P_j(\cos\theta_2). \qquad (1.14b)$$

Now the constants $a_{n,m}$, $\tilde{a}_{n,m}$, $d_{n,m}$, $\tilde{d}_{n,m}$, will be chosen to make the boundary conditions take a convenient form. On sphere 1, let the boundary condition be $u = f(\theta_1)$; then

$$f(\theta_1)=\sum_{n=0}^{\infty}b_n\sum_{m=0}^{n}[a_{n,m}L_1^{-1/2}J_{m+\frac{1}{2}}(hL_1)+\tilde{a}_{n,m}L_1^{-1/2}H^{(2)}_{m+\frac{1}{2}}(hL_1)]P_m(\cos\theta_1)$$

$$+\sum_{n=0}^{\infty}\tilde{b}_n\sum_{m=0}^{n}[d_{n,m}L_1^{-1/2}J_{m+\frac{1}{2}}(hL_1)+\tilde{d}_{n,m}L_1^{-1/2}H^{(2)}_{m+\frac{1}{2}}(hL_1)]P_m(\cos\theta_1). \qquad (1.15)$$

The first thing that will be required of the matrices $\mathbf{A}$, $\tilde{\mathbf{A}}$, $\mathbf{D}$, and $\tilde{\mathbf{D}}$, is that

$$[a_{n,m}L_1^{-1/2}J_{m+\frac{1}{2}}(hL_1)+\tilde{a}_{n,m}L_1^{-1/2}H^{(2)}_{m+\frac{1}{2}}(hL_1)]$$

$$=e_n[d_{n,m}L_1^{-1/2}J_{m+\frac{1}{2}}(hL_1)+\tilde{d}_{n,m}L_1^{-1/2}H^{(2)}_{m+\frac{1}{2}}(hL_1)] \qquad (1.16)$$

for all possible n and m (no summation over m in this equation), where e_n is any constant depending only on n. If this is satisfied, then the basis for functions of θ_1 in the boundary condition can be taken to be

$$t_n(\theta_1)=\sum_{m=0}^{n}[a_{n,m}L_1^{-1/2}J_{m+\frac{1}{2}}(hL_1)+\tilde{a}_{n,m}L_1^{-1/2}H^{(2)}_{m+\frac{1}{2}}(hL_1)]P_m(\cos\theta_1), \qquad (1.17)$$

and (1.15) can be rewritten as

$$f(\theta_1)=\sum_{n=0}^{\infty}f_n t_n(\theta_1)=\sum_{n=0}^{\infty}b_n t_n(\theta_1)$$

$$+\sum_{n=0}^{\infty} \tilde{b}_n e_n t_n(\theta_1). \tag{1.18}$$

Thus, the boundary condition at sphere 1 will give the algebraic equations

$$f_n = b_n + e_n \tilde{b}_n, \tag{1.19}$$

for $n = 0, 1, 2, \dots$.

On sphere 2, let the boundary condition be $u = g(\theta_2)$; then

$$g(\theta_2) = \sum_{j=0}^{\infty} g_j P_j(\cos\theta_2)$$

$$= \sum_{n=0}^{\infty} b_n \sum_{m=0}^{n} [a_{n,m} \sum_{j=0}^{\infty} c_{m,j} + \tilde{a}_{n,m} \sum_{j=0}^{\infty} \tilde{c}_{m,j}] L_2^{-1/2} J_{j+\frac{1}{2}}(hL_2) P_j(\cos\theta_2)$$

$$+\sum_{n=0}^{\infty} \tilde{b}_n \sum_{m=0}^{n} [d_{n,m} \sum_{j=0}^{\infty} c_{m,j} + \tilde{d}_{n,m} \sum_{j=0}^{\infty} \tilde{c}_{m,j}] L_2^{-1/2} J_{j+\frac{1}{2}}(hL_2) P_j(\cos\theta_2). \tag{1.20}$$

To have this equation take a convenient form, it will be required that

$$\sum_{m=0}^{n} [a_{n,m} c_{m,j} + \tilde{a}_{n,m} \tilde{c}_{m,j}] = 0 \tag{1.21a}$$

for $j < n$, and that

$$\sum_{m=0}^{n} [d_{n,m} c_{m,j} + \tilde{d}_{n,m} \tilde{c}_{m,j}] = 0 \tag{1.21b}$$

for $j < n$. Then, for $j = 0, 1, 2, ..$

$$g_j = \sum_{n=0}^{\infty} b_n \sum_{m=0}^{n} [a_{n,m} c_{m,j} + \tilde{a}_{n,m} \tilde{c}_{m,j}] L_2^{-1/2} J_{j+\frac{1}{2}}(hL_2)$$

$$+\sum_{n=0}^{\infty} \tilde{b}_n \sum_{m=0}^{n} [d_{n,m} c_{m,j} + \tilde{d}_{n,m} \tilde{c}_{m,j}] L_2^{-1/2} J_{j+\frac{1}{2}}(hL_2), \tag{1.22}$$

which, by the requirements (1.21) is

$$g_j = \sum_{n=0}^{j} b_n \sum_{m=0}^{n} [a_{n,m}c_{m,j} + \tilde{a}_{n,m}\tilde{c}_{m,j}] L_2^{-1/2} J_{j+\frac{1}{2}}(hL_2)$$

$$+\sum_{n=0}^{j} \tilde{b}_n \sum_{m=0}^{n} [d_{n,m}c_{m,j} + \tilde{d}_{n,m}\tilde{c}_{m,j}] L_2^{-1/2} J_{j+\frac{1}{2}}(hL_2). \tag{1.23}$$

Now it must be shown how lower triangular matrices $a_{n,m}$, $\tilde{a}_{n,m}$, $d_{n,m}$, $\tilde{d}_{n,m}$, can be found to satisfy all these conditions. Apparently, it is possible to set $\tilde{\mathbf{D}} = 0$, $e_n = 1$ for all n, and to make the diagonal elements of $\mathbf{A}$ and $\mathbf{D}$ arbitrary. Equation (1.16) can be solved to give the matrix $\tilde{\mathbf{A}}$ in terms of $\mathbf{A}$ and $\mathbf{D}$:

$$\tilde{a}_{n,m} = [L_1^{-1/2} H^{(2)}_{m+\frac{1}{2}}(hL_1)]^{-1} [d_{n,m} L_1^{-1/2} J_{m+\frac{1}{2}}(hL_1) - a_{n,m} L_1^{-1/2} J_{m+\frac{1}{2}}(hL_1)]. \tag{1.24}$$

If this is substituted into (1.21a) , the result is

$$\sum_{m=0}^{n} \frac{[d_{n,m} L_1^{-1/2} J_{m+\frac{1}{2}}(hL_1) - a_{n,m} L_1^{-1/2} J_{m+\frac{1}{2}}(hL_1)]\tilde{c}_{m,j}]}{[a_{n,m}c_{m,j} + [L_1^{-1/2} H^{(2)}_{m+\frac{1}{2}}(hL_1)]} = 0. \tag{1.25}$$

Now it will be shown that Equations (1.25) and (1.21b) can be solved to give the off-diagonal components of $\mathbf{A}$ and $\mathbf{D}$. This can be done by induction. Assume the rows 1,2,...,$k-1$ of these matrices have been determined, and consider the diagonal components (which are arbitrary) to be known. Then if Equations (1.25) and (1.21b) are written out for $n = k$, $j = 1, 2, ..., k-1$, there result $2(k-1)$ simultaneous equations for the $2(k-1)$ unknowns $a_{k,k-1}, a_{k,k-2}, ..., a_{k,0}, d_{k,k-1}, d_{k,k-2}, ..., d_{k,0}$. For example, the choice $n = 1, j = 0$ gives the equations

$$[a_{1,0}c_{0,0}+a_{1,1}c_{1,0}+[L_1^{-1/2}H_{\frac{1}{2}}^{(2)}(hL_1)]^{-1}[d_{1,0}L_1^{-1/2}J_{\frac{1}{2}}(hL_1)-a_{1,0}L_1^{-1/2}J_{\frac{1}{2}}(hL_1)]\tilde{c}_{0,0}]$$

$$+[L_1^{-1/2}H_{1+\frac{1}{2}}^{(2)}(hL_1)]^{-1}[d_{1,1}L_1^{-1/2}J_{\frac{3}{2}}(hL_1)-a_{1,1}L_1^{-1/2}J_{\frac{3}{2}}(hL_1)]\tilde{c}_{1,0}]=0, \quad (1.26a)$$

and

$$[d_{1,0}c_{0,0}+\tilde{d}_{1,0}\tilde{c}_{0,0}][d_{1,1}c_{1,0}+\tilde{d}_{1,1}\tilde{c}_{1,0}]=0, \quad (1.26b)$$

which can be solved for $d_{1,0}$ and $a_{1,0}$.

It will now be shown how to compute the coefficients $c_{n,m}$. First, an "addition formula" for Bessel functions given in Lebedev [4] will be used:

$$\frac{J_{n+\frac{1}{2}}(hr_1)}{(hr_1)^{\frac{n+1}{2}}}=2^{n+\frac{1}{2}}\Gamma(n+\frac{1}{2})\sum_{m=0}^{\infty}(n+\frac{1}{2}+m)\frac{J_{n+\frac{1}{2}+m}(hr_2)J_{n+\frac{1}{2}+m}(hR)}{(hr_2)^{\frac{n+1}{2}}(hR)^{\frac{n+1}{2}}}C_m^{n+\frac{1}{2}}(\cos\theta_2), \quad (1.27)$$

as well as another formula for spherical harmonics given in Hobson [5] :

$$r_1^n P_n^m(\cos\theta_1)=\sum_{s=0}^{n}\binom{n}{s}(-1)^s r_2^s R^s P_s(\cos\theta_2). \quad (1.28)$$

If (1.27) and (1.28) are substituted into the left member of (1.9a), the result is

$$h^{n+\frac{1}{2}}2^{n+\frac{1}{2}}\Gamma(n+\frac{1}{2})\left[\sum_{m=0}^{\infty}(n+\frac{1}{2}+m)\frac{J_{n+\frac{1}{2}+m}(hr_2)J_{n+\frac{1}{2}+m}(hR)}{(hr_2)^{\frac{n+1}{2}}(hR)^{\frac{n+1}{2}}}C_m^{n+\frac{1}{2}}(\cos\theta_2)\right]$$

$$\times\left[\sum_{s=0}^{n}\binom{n}{s}(-1)^s r_2^s R^s P_s(\cos\theta_2)\right]$$

$$= \sum_{m=0}^{\infty} c_{n,m} r_2^{-1/2} J_{m+\frac{1}{2}}(hr_2) P_m(\cos\theta_2). \tag{1.29}$$

The orthogonality of the Legendre polynomials can now be exploited. If the equation is multiplied by $P_k(\cos\theta_2)\sin\theta_2$ and integrated with respect to θ_2 from 0 to π, the result is

$$\frac{2}{2m+1} c_{n,k} r_2^{-1/2} J_{k+\frac{1}{2}}(hr_2) P_k$$
$$= h^{n+\frac{1}{2}} 2^{n+\frac{1}{2}} \Gamma(n+\frac{1}{2}) \Bigg[\sum_{m=0}^{\infty} \sum_{s=0}^{n} (n+\frac{1}{2}+m) \frac{J_{n+\frac{1}{2}+m}(hr_2) J_{n+\frac{1}{2}+m}(hR)}{(hr_2)^{\frac{n+1}{2}} (hR)^{\frac{n+1}{2}}}$$
$$\binom{n}{s} (-1)^s r_2^s R^s \int_0^{2\pi} C_m^{n+\frac{1}{2}}(\cos\theta_2) P_s(\cos\theta_2) P_k(\cos\theta_2) \sin\theta_2 d\theta_2 \Bigg]. \tag{1.30}$$

The integral

$$\int_0^{2\pi} C_m^{n+\frac{1}{2}}(\cos\theta_2) P_s(\cos\theta_2) P_k(\cos\theta_2) \sin\theta_2 d\theta_2 \tag{1.31}$$

can be evaluated as follows. First, use the Clebsch-Gordon series as given in Vilenkin [6]:

$$P_s(\cos\theta) P_k(\cos\theta)$$
$$= \sum_{\substack{j=|s-k| \\ j+s+k \ \text{even}}}^{s+k} \frac{(2j+1)(j+s-k)!(s+k-j)!(j-s+k)!(g!)^2}{(s+k+j+1)![(g-s)!(g-k)!(g-j)!]^2} P_j(\cos\theta), \tag{1.32}$$

where $g = (s+k+j)/2$. It follows that

$$\int_0^{2\pi} C_m^{n+\frac{1}{2}}(\cos\theta_2) P_s(\cos\theta_2) P_k(\cos\theta_2) \sin\theta_2 d\theta_2$$
$$= \sum_{\substack{j=|s-k| \\ j+s+k \ \text{even}}}^{s+k} \frac{(2j+1)(j+s-k)!(s+k-j)!(j-s+k)!(g!)^2}{(s+k+j+1)![(g-s)!(g-k)!(g-j)!]^2}$$

$$\times \int_0^{2\pi} C_m^{n+\frac{1}{2}}(\cos\theta_2)P_j(\cos\theta_2)\sin\theta_2 d\theta_2. \tag{1.33}$$

For this it is necessary to determine

$$\int_0^{2\pi} C_m^{n+\frac{1}{2}}(\cos\theta_2)P_j(\cos\theta_2)\sin\theta_2 d\theta_2. \tag{1.34}$$

Recall that

$$P_j(\cos\theta) = C_j^{\frac{1}{2}}(\cos\theta), \tag{1.35}$$

and use the result from Gradshteyn and Ryzhik [7] that

$$\int_0^{2\pi} C_m^{n+\frac{1}{2}}(\cos\theta)C_j^{\frac{1}{2}}(\cos\theta)\sin\theta d\theta = \frac{2^{n+1}\Gamma(1)\Gamma(n+1)\Gamma(2n)}{j!m!\Gamma(n)\Gamma(n+2)}$$

$$\times \frac{\Gamma(j+1)\Gamma(m+2n+1)}{\Gamma(1)\Gamma(2n+1)} {}_4F_3(-j, j+1, 1, -n+1; 1, 2n+2, -2n+1; 1). \tag{1.36}$$

Thus,

$$\xi(m,n,s,k) \equiv \int_0^{2\pi} C_m^{n+\frac{1}{2}}(\cos\theta_2)P_s(\cos\theta_2)P_k(\cos\theta_2)\sin\theta_2 d\theta_2$$

$$= \sum_{\substack{j=|s-k| \\ j+s+k \quad \text{even}}}^{s+k} \frac{(2j+1)(j+s-k)!(s+k-j)!(j-s+k)!(g!)^2}{(s+k+j+1)![(g-s)!(g-k)!(g-j)!]^2}$$

$$\times \frac{2^{n+1}\Gamma(1)\Gamma(n+1)\Gamma(2n)}{j!m!\Gamma(n)\Gamma(n+2)} \frac{\Gamma(j+1)\Gamma(m+2n+1)}{\Gamma(1)\Gamma(2n+1)}$$

$${}_4F_3(-j, j+1, 1, -n+1; 1, 2n+2, -2n+1; 1). \tag{1.37}$$

Here the quantity $\xi(m,n,s,k)$ has been introduced for brevity. It is explicitly given by (1.37). It is useful to note a certain property of the function ξ. By recalling another expression for Gegenbauer polynomials, namely,

$$C_m^{n+\frac{1}{2}}(\cos\theta) = \frac{1}{(2n-1)!!}\frac{d^n P_{m+n}(\cos\theta)}{d(\cos\theta)^n}, \tag{1.38}$$

it can be seen by integrating the integral

$$\int_0^{2\pi} C_m^{n+\frac{1}{2}}(\cos\theta)P_s(\cos\theta)P_k(\cos\theta)\sin\theta d\theta \tag{1.39}$$

by parts $(s+k)$ times, and recalling that $P_s(\cos\theta)P_k(\cos\theta)$ is an $s+k$-th order polynomial in $\cos\theta$, that

$$\xi(m,n,s,k) = 0 \qquad \text{for } n > s+k+1\ . \tag{1.40}$$

In summary, the matrix $\mathbf{C} = [c_{n,k}]$ is given by

$$c_{n,k} = \frac{2m+1}{2} r_2^{1/2} \frac{1}{J_{k+\frac{1}{2}}(hr_2)} h^{n+\frac{1}{2}} 2^{n+\frac{1}{2}} \Gamma(n+\frac{1}{2})$$
$$\times \left[\sum_{m=0}^{\infty}\sum_{s=0}^{n}(n+\frac{1}{2}+m)\frac{J_{n+\frac{1}{2}+m}(hr_2)J_{n+\frac{1}{2}+m}(hR)}{(hr_2)^{\frac{n+1}{2}}(hR)^{\frac{n+1}{2}}}\right.$$
$$\left.\binom{n}{s}(-1)^s r_2^s R^s \xi(m,n,s,k)\right]. \tag{1.41}$$

This identity must hold for all $r_2 < R$, and in particular, the value $r_2 = l_2$ can be substituted in:

$$c_{n,k} = \frac{2m+1}{2} L_2^{1/2} \frac{1}{J_{k+\frac{1}{2}}(hL_2)} h^{n+\frac{1}{2}} 2^{n+\frac{1}{2}} \Gamma(n+\frac{1}{2})$$
$$\times \left[\sum_{m=0}^{\infty}\sum_{s=0}^{n}(n+\frac{1}{2}+m)\frac{J_{n+\frac{1}{2}+m}(hL_2)J_{n+\frac{1}{2}+m}(hR)}{(hL_2)^{\frac{n+1}{2}}(hR)^{n+1/over2}}\right.$$
$$\left.\binom{n}{s}(-1)^s L_2^s R^s \xi(m,n,s,k)\right], \tag{1.42}$$

where $\xi(m,n,s,k)$ is given explicitly by (1.37).

UNSTEADY STOKES FLOW WITH TWO SPHERES

The problem of unsteady Stokes flow with two spheres will now be considered. The two spheres, A and B, are separated by a distance R. This time the infinite system of algebraic systems for the eigenfunction coefficients will be derived without making any change of basis to force the system to be triangular.

The general solution for unsteady flow with boundary conditions at two spheres can be written as

$$\mathbf{u} = \mathbf{u}_A + \mathbf{u}_B, \tag{2.1}$$

where $\mathbf{u}_A$ is expressed in the spherical coordinate system centered on sphere A (r_1, θ_1, ϕ):

$$\begin{aligned}\hat{\mathbf{u}}_A = \sum_{n=1}^{\infty}\Bigg[& \frac{1}{i\Omega}\nabla p^A_{-(n+1)} - f_n(\sqrt{i\Omega}r_1)\nabla\times(\mathbf{r}_1\chi^A_n) \\ & +[(n+1)f_{n-1}(\sqrt{i\Omega}r_1) - nf_{n+1}(\sqrt{i\Omega}r_1)(i\Omega)r_1^2]\nabla\varphi^A_n \\ & +n(2n+1)f_{n+1}(\sqrt{i\Omega}r_1)(i\Omega)\varphi^A_n\mathbf{r}_1\Bigg];\end{aligned} \tag{2.2}$$

$$\hat{p}^A = \sum_{n=1}^{\infty} p^A_{-(n+1)}. \tag{2.3}$$

Here,

$$p^A_{-(n+1)} = \sum_{m=-n}^{m=n} p^A_{-(n+1),m} r_1^{-(n+1)} P^m_n(\cos\theta_1)e^{im\phi}; \tag{2.4a}$$

$$\chi_n^A = \sum_{m=-n}^{m=n} \chi_{n,m}^A r_1^n P_n^m(\cos\theta_1) e^{im\phi}; \tag{2.4b}$$

$$\varphi_n^A = \sum_{m=-n}^{m=n} \varphi_{n,m}^A r_1^n P_n^m(\cos\theta_1) e^{im\phi}. \tag{2.4c}$$

The exact same expression holds for $\mathbf{u}_B$, with the superscript "A" replaced everywhere by "B," and the coordinates (r_1, θ_1, ϕ) replaced by (r_2, θ_2, ϕ). The summation is from $n = 1$ to $n = \infty$ because it is required that the flow field go to zero far from the spheres, and that the spheres are not a source of fluid (this rules out the dilatational $n = 0$ harmonics).

To get the solution of interest here, it is necessary to apply the boundary conditions at the surfaces of both the spheres. To apply the boundary conditions at the surface of sphere B, it is necessary to express $\mathbf{u}_A$ in the (r_2, θ_2, ϕ) coordinate system. This will be done as follows.

First, note that the following addition theorems hold for spherical harmonics. For negative harmonics,

$$\frac{1}{r_i^{n+1}} P_n^m(\cos\theta_i) = \frac{1}{R}^{n+1} \sum_{s=m}^{\infty} \binom{s+m}{n+s} \left(\frac{R}{r_{3-i}}\right)^s P_s^m(\cos\theta_{3-i}), \tag{2.5}$$

where $i = 1, 2$. This result is from Jeffrey [2], and allows the spherical harmonics $p_{-(n+1)}$ of one coordinate system to be expressed in the other coordinate system. For positive harmonics, there is a result from Hobson [5]

$$r_1^n P_n^m(\cos\theta_1) = \sum_{s=0}^{n-m} r_2^{n-s} P_{n-s}^m(\cos\theta_2) \frac{R^s}{s!} \frac{(n+m)!}{(n+m-s)!}. \tag{2.6}$$

This allows the spherical harmonics χ_n and φ_n of one coordinate system to be expressed in the other coordinate system.

The functions f_n are defined by

$$f_n(z) = i\sqrt{\frac{\pi}{2}} z^{-(n+\frac{1}{2})} H^{(2)}_{n+\frac{1}{2}}(z), \tag{2.7}$$

where $H^{(2)}_{n+\frac{1}{2}}$ are Hankel functions. These functions satisfy the addition theorem (see Lebedev [4])

$$\frac{H^{(2)}_{n+\frac{1}{2}}(\sqrt{i\Omega}r_1)}{(\sqrt{i\Omega}r_1)^{n+\frac{1}{2}}}$$
$$= 2^{n+\frac{1}{2}}\Gamma(n+\frac{1}{2})\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)\left[\frac{H^{(2)}_{n+p+\frac{1}{2}}(\sqrt{i\Omega}R)J_{n+p+\frac{1}{2}}(\sqrt{i\Omega}r_2)}{(\sqrt{i\Omega}R)^{n+\frac{1}{2}}(\sqrt{i\Omega}r_2)^{n+\frac{1}{2}}}\right]C_m^{n+\frac{1}{2}}(\cos\theta_2). \tag{2.8}$$

Thus,

$$f_n(\sqrt{i\Omega}r_1) = \frac{(2n-1)!}{2^{n-2}(n-1)!}\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_n(\sqrt{i\Omega}R)\psi_n(\sqrt{i\Omega}r_2)C_p^{\frac{n+1}{2}}(cos\theta_2). \tag{2.9}$$

Finally, the vector $\mathbf{r}_1$, which appears in the general solution (2.2), must be expressed in the coordinate system centered on sphere B:

$$\mathbf{e}_{r_1} = \mathbf{R} + \mathbf{r}_2$$
$$= R\cos\theta_2\mathbf{e}_{r_2} - R\sin\theta_2\mathbf{e}_{\theta_2} + r_2\mathbf{e}_{r_2}\ . \tag{2.10}$$

By noting that

$$r_1\sin\theta_1 = r_2\sin\theta_2; \tag{2.11}$$

$$r_1\cos\theta_1 = R - r_2\cos\theta_2, \tag{2.12}$$

and that the law of cosines gives

$$r_1^2 = R^2 + r_2^2 - 2Rr_2 \cos\theta_2, \tag{2.13}$$

all the above may be combined to give

$$\begin{aligned} \mathbf{r}_1 &= r_1 \mathbf{e}_{r_1} \\ &= \mathbf{e}_{r_2} r_2 \sin\theta_2 \sin\theta_2 + (R - r_2\cos\theta_2)\cos\theta_2] \\ &+ \mathbf{e}_{\theta_2}[r_2 \sin\theta_2 \cos\theta_2 - (R - r_2\cos\theta_2)\sin\theta_2] \\ &= \mathbf{e}_{r_2}[R\cos\theta_2 - r_2\cos(2\theta_2)] - \mathbf{e}_{\theta_2}[R\sin\theta_2], \end{aligned} \tag{2.14}$$

the desired expression. Since the vector operator ∇ can be easily expressed in either coordinate system, at this point, everything in the solution (2.2) has been expressed in the (r_2, θ_2, ϕ) coordinate system. Putting this altogether gives

$$\begin{aligned} \hat{\mathbf{u}}_A = \sum_{n=1}^{\infty}\Bigg\{ & \frac{1}{i\Omega}\nabla\left[\sum_{m=-n}^{m=n} p^A_{-(n+1),m}\left(\frac{1}{R}\right)^{n+1}\sum_{s=m}^{\infty}\binom{n+s}{s+m}\left(\frac{r_2}{R}\right)^s P_s^m(\cos\theta_2)e^{im\phi}\right] \\ & -\frac{(2n-1)!}{2^{n-2}(n-1)!}\left[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}r_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\right] \\ & \cdot\nabla\times\left[\left(\mathbf{R}+\mathbf{r}_2\right)\left(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m} r_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\right)\right] \\ & +\left[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\left(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}r_2)C_p^{n-\frac{1}{2}}(cos\theta_2)\right)\right. \\ & -n\frac{(2n+1)!}{2^{n-1}n!}\left(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\right) \end{aligned}$$

$$\cdot (i\Omega)(R^2 + r_2^2 - 2Rr_2\cos\theta_2)\Big]$$

$$\cdot \nabla\Big[\sum_{m=-n}^{m=n} \varphi_{n,m}^A \sum_{s=0}^{n-m} r_2^{n-s} P_{n-s}^m(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$(i\Omega)\Big[\sum_{m=-n}^{m=n} \varphi_{n,m}^A \sum_{s=0}^{n-m} \frac{(n-s)(n-s-1)}{r_2^2} r_2^{n-s} P_{n-s}^m(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$\Big(\mathbf{e}_{r_2}[R\cos\theta_2 - r_2\cos(2\theta_2)] - \mathbf{e}_{\theta_2}[R\sin\theta_2]\Big)\Big\}. \tag{2.15}$$

Now the the boundary conditions will be applied. Let the prescribed boundary condition on sphere 2 be

$$\mathbf{u} = \mathbf{V}_2(\theta, \phi, \omega), \tag{2.16}$$

and, in keeping with Brenner's method for solving flow problems with spheres, use the spherical harmonic expansions

$$\frac{\mathbf{r}}{r}\cdot\mathbf{V}_2 = \sum_{n=1}^{\infty} X_n^2; \tag{2.17a}$$

$$-r\nabla\cdot\mathbf{V}_2 = \sum_{n=1}^{\infty} Y_n^2; \tag{2.17b}$$

$$\mathbf{r}\cdot\nabla\times\mathbf{V}_2 = \sum_{n=1}^{\infty} Z_n^2. \tag{2.17c}$$

As shown by Yang [8] for the unsteady case,

$$\frac{\mathbf{r}_2}{r_2}\cdot\mathbf{u}^B = \sum_{n=1}^{\infty}\Big[\frac{(n+1)}{r_2(i\Omega)}p_{-(n+1)}^B$$

$$+n\Big(\frac{(n+1)}{r_2}f_{n-1}(\sqrt{i\Omega}r_2)+(n+1)f_{n+1}(\sqrt{i\Omega}r_2)(i\Omega)r_2\Big)\varphi_n^B, \tag{2.18a}$$

$$-r_2\nabla\cdot\mathbf{u}^B = \sum_{n=1}^{\infty}\Bigg[\frac{(n+1)(n+2)}{i\Omega r_2}p_{-(n+1)}^B + n\Big(\frac{(n^2-1)}{r_2}f_{n-1}(\sqrt{i\Omega}r_2)$$

$$+(n+1)^2(i\Omega)r_2 f_{n+1}(\sqrt{i\Omega}r_2)+(n+1)(i\Omega)r_2 f_n(\sqrt{i\Omega}r_2)$$

$$+(n+1)(i\Omega)^2 r_2^3 f_{n+2}(\sqrt{i\Omega}r_2)\Big)\varphi_n^B\Bigg], \tag{2.18b}$$

$$\mathbf{r}_2\cdot\nabla\times\mathbf{u}^B = -\sum_{n=1}^{\infty}n(n+1)f_n(\sqrt{i\Omega}r_2)\chi_n^B. \tag{2.18c}$$

Thus, to apply the boundary conditions, it is first necessary to determine what

$$\mathbf{e}_{r_2}\cdot\mathbf{u}_A(a_2,\theta_2,\phi,\omega) = \mathbf{e}_{r_2}\cdot\mathbf{u}_A(r_2,\theta_2,\phi,\omega)\Big|_{r=a_2}, \tag{2.19}$$

$$-r_2\nabla\cdot\mathbf{u}^A(a_2,\theta_2,\phi,\omega) = \frac{\partial}{\partial r_2}\Big[\mathbf{e}_{r_2}\cdot\mathbf{u}_A(r_2,\theta_2,\phi,\omega)\Big]\Big|_{r=a_2}, \tag{2.20}$$

and

$$\mathbf{r}_2\cdot\nabla\times\mathbf{u}^A(a_2,\theta_2,\phi,\omega) = \mathbf{r}_2\cdot\nabla\times\mathbf{u}^A(r_2,\theta_2,\phi,\omega)\Big|_{r_2=a_2}, \tag{2.21}$$

are in the coordinate system 2. This is done by substituting (2.14) for $\mathbf{u}^A$ into (2.19), (2.20), (2.21), and applying the operators. The resulting three expressions are given in Appendix 1, since they are lengthy. In obtaining (A1.4), (A1.5), (A1.6) of Appendix 1, use was made of the recursion relations

$$f_n'(z) = -zf_{n+1}(z); \tag{2.22a}$$

$$\psi_n'(z) = -z\psi_{n+1}(z). \tag{2.22b}$$

In calculating

$$r_2\mathbf{e}_{r_2} \cdot \nabla \times \mathbf{u}^A, \tag{2.23}$$

it is helpful to first note that $\mathbf{u}^A$ has the generic form

$$\mathbf{u}^A = \nabla P + F(r_2, \theta_2)\nabla \times [(\mathbf{R} + \mathbf{r}_2)\Xi]$$

$$+G(r_2, \theta_2)\nabla\Phi + H(r_2, \theta_2)\Phi(\mathbf{R} + \mathbf{r}_2), \tag{2.24}$$

where P, Ξ and Φ are harmonic, F, G and H are scalar functions, and $\mathbf{R}$, as before, is the constant vector $R\cos\theta_2\mathbf{e}_{r_2} - R\sin\theta_2\mathbf{e}_{\theta_2}$. It can be shown from this that

$$\mathbf{r}_2 \cdot \nabla \times \mathbf{u}^A = \frac{1}{r_2\sin\theta_2}\frac{\partial G}{\partial\theta_2}\frac{\partial\Phi}{\partial\phi}$$

$$+r_2F\Big[\frac{\partial}{\partial r_2}(\mathbf{R}\cdot\nabla\Xi) + 2\frac{\partial\Xi}{\partial r_2} + r_2\frac{\partial^2\Xi}{\partial r_2^2}\Big]$$

$$+\frac{\partial F}{\partial\theta_2}\Big[\frac{R\cos\theta_2 + r_2}{r_2}\frac{\partial\Xi}{\partial\theta_2} + R\sin\theta_2\frac{\partial\Xi}{\partial r_2}\Big] - R\sin\theta_2\frac{\partial(H\Phi)}{\partial\theta_2}. \tag{2.25}$$

In the above,

$$F(r_2, \theta_2) = -\frac{(2n-1)!}{2^{n-2}(n-1)!}\left[\sum_{p=0}^{\infty}(n + \frac{1}{2} + p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}r_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\right]; \tag{2.26a}$$

$$G(r_2,\theta_2)$$

$$= \Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\Big(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}r_2)C_p^{n-\frac{1}{2}}(cos\theta_2)\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(R^2+r_2^2-2Rr_2\cos\theta_2)\Bigg] \qquad (2.26b)$$

$$H(r_2,\theta_2) = n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}(i\Omega)$$

$$\times \Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big); \qquad (2.26c)$$

$$\Xi = \Big(\sum_{m=-n}^{m=n}\chi_{n,m}^A\sum_{s=0}^{n-m}r_2^{n-s}P_{n-s}^m(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big); \qquad (2.27a)$$

$$\Phi = \Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^A\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{r_2^2}r_2^{n-s}P_{n-s}^m(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]. \qquad (2.27b)$$

The boundary conditions at the surface of sphere 2 ($r_2 = a_2$), in which the expressions in Appendix 1 appear (with r_2 set equal to a_2), are given in Appendix 2.

To obtain algebraic equations from the boundary conditions (A2.1), (A2.2), (A2.3), the usual procedure will be taken of multiplying by

$$P_j^k(\cos\theta_2)e^{-ik\phi}, \qquad (2.28)$$

and integrating over θ_2 and ϕ, to exploit the orthogonality of the surface spherical harmonics. The first step is to rewrite the sums

$$\sum_{n=1}^{\infty} \sum_{m=-n}^{m=n} \tag{2.29}$$

as

$$\sum_{m=-\infty}^{\infty} \sum_{n=|m|}^{\infty} \tag{2.30}$$

(where it is understood that $n \neq 0$), and to do the ϕ integrals using

$$\int_0^{2\pi} e^{im\phi} e^{-ik\phi} d\phi = \delta_{m,k}. \tag{2.31}$$

For convenience, after doing this operation, replace the index k by m (a mere change in notation at that point). The resulting three algebraic equations are given in Appendix 3 as Equations (A3.1), (A3.2), (A3.3). The various theta integrals appearing in these three equations are evaluated in Appendix 4. They are abbreviated by $I_1, I_2, ...$, as defined in this appendix. These values for the integrals can be substituted into Equations (A3.1), (A3.2), and (A3.3) to give, finally, the algebraic equations for the coefficients $p^A_{-(n+1),m}$, $p^B_{-(n+1),m}$, $\chi^A_{n,m}$, $\chi^B_{n,m}$, $\varphi^A_{n,m}$, $\varphi^B_{n,m}$. By the symmetry of the geometry of the two spherical coordinate systems, the other 3 equations can be obtained from the following three equations by exchanging subscripts "1" and "2," and exchanging superscripts "A" and "B." The three algebraic equations resulting from applying the no-slip condition at sphere B are

$$a^{(1)}_{j,m} p^B_{-(j+1),m} + a^{(2)}_{j,m} \varphi^B_{j,m} + \sum_{n=|m|}^{\infty} \left[a^{(3)}_{n,j,m} p^A_{n,m} a^{(4)}_{n,j,m} \varphi^A_{n,m} a^{(5)}_{n,j,m} \chi^A_{n,m} \right]$$

$$= X^2_{j,m} \frac{2}{(2j+1)} \frac{(j+m)!}{(j-m)!} \quad ; \tag{2.32a}$$

$$b^{(1)}_{j,m} p^B_{-(j+1),m} + b^{(2)}_{j,m} \varphi^B_{j,m} + \sum_{n=|m|}^{\infty} \left[b^{(3)}_{n,j,m} p^A_{n,m} b^{(4)}_{n,j,m} \varphi^A_{n,m} \right]$$

$$= Y^2_{j,m} \frac{2}{(2j+1)} \frac{(j+m)!}{(j-m)!} \quad ; \tag{2.32b}$$

$$c^{(1)}_{n,m} \chi^B_{n,m} + \sum_{n=|m|}^{\infty} \left[c^{(3)}_{n,j,m} p^A_{n,m} + c^{(4)}_{n,j,m} \varphi^A_{n,m} + c^{(5)}_{n,j,m} \chi^A_{n,m} \right]$$

$$= Z^2_{j,m} \frac{2}{(2j+1)} \frac{(j+m)!}{(j-m)!} \quad , \tag{2.32c}$$

where the coefficients are given in Appendix 5.

In summary, the problem of unsteady Stokes flow around two spheres with specified velocities at their surfaces (and vanishing flow at infinity) has been reduced to the solution of an infinite system of algebraic equations. To obtain actual numerical values, the system can be truncated to N equations, where N can be chosen as large as necessary to get the desired accuracy. For cases where the two spheres are separated by a distance on the order of their size, this method should give more accurate results than the method of reflections.

Appendix 1 to Chapter V

In this appendix, the three quantities

$$\mathbf{e}_{r_2} \cdot \mathbf{u}_A, \tag{A1.1}$$

$$r_2 \frac{\partial}{\partial r_2}(\mathbf{e}_{r_2} \cdot \mathbf{u}_A), \tag{A1.2}$$

$$r_2 \mathbf{e}_{r_2} \cdot \nabla \times \mathbf{u}^A, \tag{A1.3}$$

are given, where $\mathbf{u}_A$ is expressed in coordinate system 2. The first of these quantities appears in the normal velocity condition on sphere 2; the other two appear in the two tangential velocity conditions.

TERM IN NORMAL VELOCITY CONDITION.

$$\mathbf{e}_{r_2} \cdot \mathbf{u}_A$$

$$= \sum_{n=1}^{\infty} \Bigg\{ \frac{1}{i\Omega} \Bigg[\sum_{m=-n}^{m=n} p^A_{-(n+1),m} \left(\frac{1}{R}\right)^{n+1} \sum_{s=m}^{\infty} \binom{n+s}{s+m} \frac{s}{r_2} \left(\frac{r_2}{R}\right)^s P_s^m(\cos\theta_2) e^{im\phi} \Bigg]$$

$$- \frac{(2n-1)!}{2^{n-2}(n-1)!} \Bigg[\sum_{p=0}^{\infty} (n + \frac{1}{2} + p) f_{n+p}(\sqrt{i\Omega}R) \psi_{n+p}(\sqrt{i\Omega}r_2) C_p^{n+\frac{1}{2}}(cos\theta_2) \Bigg]$$

$$\cdot \frac{R}{r_2} \Bigg[\sum_{m=-n}^{m=n} \chi^A_{n,m} \Big(\sum_{s=0}^{n-m} r_2^{n-s} P^m_{n-s}(cos\theta_2) \frac{R^s}{s!} \frac{(n+m)!}{(n+m-s)!} (im) e^{im\phi} \Big) \Bigg]$$

$$+ \Bigg[(n+1) \frac{(2n-3)!}{2^{n-3}(n-2)!} \Big(\sum_{p=0}^{\infty} (n - \frac{1}{2} + p) f_{n+p-1}(\sqrt{i\Omega}R) \psi_{n+p-1}(\sqrt{i\Omega}r_2) C_p^{n-\frac{1}{2}}(cos\theta_2) \Big)$$

$$- n \frac{(2n+1)!}{2^{n-1}n!} \Big(\sum_{p=0}^{\infty} (n + \frac{3}{2} + p) f_{n+p+1}(\sqrt{i\Omega}R) \psi_{n+p+1}(\sqrt{i\Omega}r_2) C_p^{n+\frac{3}{2}}(cos\theta_2) \Big)$$

$$\cdot(i\Omega)(R^2 + r_2^2 - 2Rr_2\cos\theta_2)\Big]$$

$$\cdot\Big[\sum_{m=-n}^{m=n}\varphi^A_{n,m}\sum_{s=0}^{n-m}\frac{(s-n)}{r_2}r_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$(i\Omega)\Big[\sum_{m=-n}^{m=n}\varphi^A_{n,m}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{r_2^2}r_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$\Big(R\cos\theta_2 + r_2\Big)\Big\} \qquad (A1.4)$$

TERM IN FIRST TANGENTIAL VELOCITY CONDITION

$$r_2\frac{\partial}{\partial r_2}(\mathbf{e}_{r_2}\cdot\mathbf{u}_A) =$$

$$r_2\sum_{n=1}^{\infty}\Big\{\frac{1}{i\Omega}\Big[\sum_{m=-n}^{m=n}p^A_{-(n+1),m}\Big(\frac{1}{R}\Big)^{n+1}\sum_{s=m}^{\infty}\binom{n+s}{s+m}\frac{s(s-1)}{r_2^2}\Big(\frac{r_2}{R}\Big)^s P^m_s(\cos\theta_2)e^{im\phi}\Big]$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Big[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}r_2)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\Big]$$

$$\cdot\frac{R}{r_2}\Big[\sum_{m=-n}^{m=n}\chi^A_{n,m}\Big(\sum_{s=0}^{n-m}r_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)e^{im\phi}\Big)\Big]$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Big[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}r_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\Big]$$

$$\cdot\Big[R\Big(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m}(s-n-1)r_2^{n-s+2}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)e^{im\phi}\Big)\Big]$$

$$+\Big[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}$$

$$\Big(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}r_2)\psi_{n+p}(\sqrt{i\Omega}r_2)C_p^{n-\frac{1}{2}}(cos\theta_2)\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}r_2)\psi_{n+p+2}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(R^2+r_2^2-2Rr_2\cos\theta_2)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(2r_2-2R\cos\theta_2)\Big]$$

$$\cdot\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(s-n)}{r_2}r_2^{n-s}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$+\Big[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\Big(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}r_2)C_p^{n-\frac{1}{2}}(cos\theta_2)\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(R^2+r_2^2-2Rr_2\cos\theta_2)\Big]$$

$$\cdot\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{r_2^2}r_2^{n-s-2}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}$$

$$\cdot\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}r_2)\psi_{n+p+2}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$(i\Omega)\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{r_2^2}r_2^{n-s-2}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$\Big([R\cos\theta_2 + r_2]\Big)$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$(i\Omega)\Big\{\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(s-n)(n-s-1)(n-s-3)}{r_2^3}$$

$$\cdot r_2^{n-s}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]\Big([R\cos\theta_2 + r_2]\Big)$$

$$+\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{r_2^2}r_2^{n-s}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]\Big\}\Bigg\} \qquad (A1.5)$$

TERM IN SECOND TANGENTIAL VELOCITY CONDITION

$$\mathbf{r}_2\cdot\nabla\times\mathbf{u}^A$$

$$=\frac{1}{r_2\sin\theta_2}\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}$$

$$\Big(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}r_2)\frac{\partial C_p^{n-\frac{1}{2}}(cos\theta_2)}{\partial\theta_2}\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)\frac{\partial C_p^{n+\frac{3}{2}}(cos\theta_2)}{\theta_2}\Big)$$

$$\cdot(i\Omega)(R^2+r_2^2-2Rr_2\cos\theta_2)\Bigg]$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(2Rr_2\sin\theta_2)\Bigg]$$

$$\cdot\left[\sum_{m=-n}^{m=n}\varphi^A_{n,m}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{r_2^2}r_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)e^{im\phi}\right]$$

$$+r_2\left\{-\frac{(2n-1)!}{2^{n-2}(n-1)!}\left[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}r_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\right]\right\}$$

$$\Bigg[R\cos\theta_2\Big(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m}(n-s)(n-s-1)r_2^{n-s-2}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)$$

$$-R\sin\theta_2\frac{\partial}{\partial\theta_2}\Big(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m}(n-s-1)r_2^{n-s-2}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)$$

$$+\Big(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m}[(n-s)(n-s+1)]r_2^{n-s-1}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)\Bigg]$$

$$+\left\{-\frac{(2n-1)!}{2^{n-2}(n-1)!}\left[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}r_2)\frac{\partial C_p^{n+\frac{1}{2}}(cos\theta_2)}{\partial\theta_2}\right]\right\}$$

$$\Bigg[\frac{R\cos\theta_2+r_2}{r_2}\Big(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m}r_2^{n-s}\frac{\partial P^m_{n-s}(cos\theta_2)}{\partial\theta_2}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)$$

$$+R\sin\theta_2\Big(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m}(n-s)r_2^{n-s-1}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)\Bigg]$$

$$-R\sin\theta_2$$

$$\Bigg[n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)\frac{\partial C_p^{n+\frac{3}{2}}}{\partial\theta_2}(cos\theta_2)\Big)$$

$$(i\Omega)\Big[\sum_{m=-n}^{m=n}\varphi^A_{n,m}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{r_2^2}r_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$(i\Omega)\Big[\sum_{m=-n}^{m=n}\varphi^A_{n,m}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{r_2^2}r_2^{n-s}\frac{\partial P^m_{n-s}(cos\theta_2)}{\theta_2}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]\Bigg]$$

$$(A1.6)$$

Appendix 2 of Chapter V

In this Appendix, the three velocity conditions for sphere 2 are given.

FIRST CONDITION

$$\sum_{n=1}^{\infty}\left[\frac{(n+1)}{a_2(i\Omega)}[\sum_{m=-n}^{n} p^{B}_{-(n+1),m} P_n^m(\cos\theta_2)e^{im\phi}]\right.$$

$$\left.+n\Big(\frac{(n+1)}{a_2} f_{n-1}(\sqrt{i\Omega}a_2)+(n+1)f_{n+1}(\sqrt{i\Omega}a_2)(i\Omega)a_2\Big)\sum_{m=-n}^{n}\varphi^{B}_{n,m}P_n^m(\cos\theta_2)e^{im\phi}\right]$$

$$+\sum_{n=1}^{\infty}\left\{\frac{1}{i\Omega}\left[\sum_{m=-n}^{m=n} p^{A}_{-(n+1),m}\left(\frac{1}{R}\right)^{n+1}\sum_{s=m}^{\infty}\binom{n+s}{s+m}\frac{s}{a_2}\left(\frac{a_2}{R}\right)^{s} P_s^m(\cos\theta_2)e^{im\phi}\right]\right.$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\left[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\right]$$

$$\cdot\frac{R}{a_2}\left[\sum_{m=-n}^{m=n}\chi^{A}_{n,m}\sum_{s=0}^{n-m}a_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)e^{im\phi}\right]$$

$$+\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\Big(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}a_2)C_p^{n-\frac{1}{2}}(cos\theta_2)\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(R^2+a_2^2-2Ra_2\cos\theta_2)\Bigg]$$

$$\cdot\left[\sum_{m=-n}^{m=n}\varphi^{A}_{n,m}\sum_{s=0}^{n-m}\frac{(s-n)}{a_2}a_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\right]$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\left(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\right)$$

$$(i\Omega)\left[\sum_{m=-n}^{m=n}\varphi^{A}_{n,m}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\right]$$

$$\cdot\Big(R\cos\theta_2 + a_2\Big)\Big\} = \sum_{n=1}^{\infty} X_n^2 \qquad (A2.1)$$

SECOND CONDITION

$$\sum_{n=1}^{\infty}\Bigg[\frac{(n+1)(n+2)}{i\Omega a_2}\Big[\sum_{m=-n}^{n} p^B_{-(n+1),m}P_n^m(\cos\theta_2)e^{im\phi}\Big] + n\Big(\frac{(n^2-1)}{a_2}f_{n-1}(\sqrt{i\Omega}a_2)$$

$$+(n+1)^2(i\Omega)a_2 f_{n+1}(\sqrt{i\Omega}a_2) + (n+1)(i\Omega)a_2 f_n(\sqrt{i\Omega}a_2)$$

$$+(n+1)(i\Omega)^2 a_2^3 f_{n+2}(\sqrt{i\Omega}a_2)\Big)\Big[\sum_{m=-n}^{m}\varphi^B_{n,m}P_n^m(\cos\theta_2)e^{im\phi}\Big]\Bigg]$$

$$+a_2\sum_{n=1}^{\infty}\Bigg\{\frac{1}{i\Omega}\Bigg[\sum_{m=-n}^{m=n} p^A_{-(n+1),m}\Big(\frac{1}{R}\Big)^{n+1}$$

$$\cdot\sum_{s=m}^{\infty}\binom{n+s}{s+m}\frac{s(s-1)}{a_2^2}\Big(\frac{a_2}{R}\Big)^s P_s^m(\cos\theta_2)e^{im\phi}\Bigg]$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Bigg[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}a_2)\psi_{n+p+1}(\sqrt{i\Omega}a_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\Bigg]$$

$$\cdot\frac{1}{a_2\sin\theta_2}\Bigg[\Big(R\sin\theta_2\Big)$$

$$\Big(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m}a_2^{n-s}P_{n-s}^m(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)e^{im\phi}\Big)\Bigg]$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Bigg[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\Bigg]$$

$$\cdot\frac{1}{\sin\theta_2}\Bigg[\Big(R\sin\theta_2\Big)$$

$$\Big(\sum_{m=-n}^{m=n}\chi^A_{n,m}\sum_{s=0}^{n-m}(s-n-1)a_2^{n-s+2}P_{n-s}^m(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)e^{im\phi}\Big)\Bigg]$$

$$+\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}$$

$$\cdot\Big(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}a_2)\psi_{n+p}(\sqrt{i\Omega}r_2)C_p^{n-\frac{1}{2}}(cos\theta_2)\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}a_2)\psi_{n+p+2}(\sqrt{i\Omega}a_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(R^2+a_2^2-2Ra_2\cos\theta_2)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(2a_2-2R\cos\theta_2)\Bigg]$$

$$\cdot\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(s-n)}{a_2}a_2^{n-s}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$+\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\Big(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}a_2)C_p^{n-\frac{1}{2}}(cos\theta_2)\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(R^2+a_2^2-2Ra_2\cos\theta_2)\Bigg]$$

$$\cdot\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s-2}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}$$

$$\cdot\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}s_2)\psi_{n+p+2}(\sqrt{i\Omega}a_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$(i\Omega)\Big[\sum_{m=-n}^{m=n}\varphi^A_{n,m}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s-2}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$\cdot\Big([R\cos\theta_2+a_2]\Big)$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$(i\Omega)\Big\{\Big[\sum_{m=-n}^{m=n}\varphi^A_{n,m}\sum_{s=0}^{n-m}\frac{(s-n)(n-s-1)(n-s-3)}{a_2^3}$$

$$\cdot a_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]\Big([R\cos\theta_2+a_2]\Big)$$

$$+\Big[\sum_{m=-n}^{m=n}\varphi^A_{n,m}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}P^m_{n-s}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]\Big\}$$

$$=\sum_{n=1}^{\infty}Y_n^2 \qquad (A2.2)$$

THIRD CONDITION

$$-\Big(\sum_{n=1}^{\infty}n(n+1)f_n(\sqrt{i\Omega}r_a)\Big)\Big(\sum_{m=-n}^{m=n}\chi^B_{n,m}P^m_n(\cos\theta_2)e^{im\phi}\Big)$$

$$+\frac{1}{a_2\sin\theta_2}\Big[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}$$

$$\cdot\Big(\sum_{p=0}^{\infty}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}a_2)\frac{\partial C_p^{n-\frac{1}{2}}(cos\theta_2)}{\partial\theta_2}\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)\frac{\partial C_p^{n+\frac{3}{2}}(cos\theta_2)}{\partial\theta_2}\Big)$$

$$\cdot(i\Omega)(R^2+a_2^2-2Ra_2\cos\theta_2)\Big]$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$\cdot(i\Omega)(2Ra_2\sin\theta_2)\Big]$$

$$\cdot\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)e^{im\phi}\Big]$$

$$+a_2\Bigg\{-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Bigg[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\Bigg]\Bigg\}$$

$$\Bigg[R\cos\theta_2\Big(\sum_{m=-n}^{m=n}\chi_{n,m}^{A}\sum_{s=0}^{n-m}(n-s)(n-s-1)a_2^{n-s-2}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)$$

$$--R\sin\theta_2\frac{\partial}{\partial\theta_2}\Big(\sum_{m=-n}^{m=n}\chi_{n,m}^{A}\sum_{s=0}^{n-m}(n-s-1)a_2^{n-s-2}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)$$

$$+\Big(\sum_{m=-n}^{m=n}\chi_{n,m}^{A}\sum_{s=0}^{n-m}(n-s)(n-s+1)a_2^{n-s-1}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)\Bigg]$$

$$+\Bigg\{-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Bigg[\sum_{p=0}^{\infty}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)\frac{\partial C_p^{n+\frac{1}{2}}(cos\theta_2)}{\partial\theta_2}\Bigg]\Bigg\}$$

$$\Bigg[\frac{R\cos\theta_2+a_2}{a_2}\Big(\sum_{m=-n}^{m=n}\chi_{n,m}^{A}\sum_{s=0}^{n-m}a_2^{n-s}\frac{\partial P_{n-s}^{m}(cos\theta_2)}{\partial\theta_2}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)$$

$$+R\sin\theta_2\Big(\sum_{m=-n}^{m=n}\chi_{n,m}^{A}\sum_{s=0}^{n-m}(n-s)a_2^{n-s-1}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big)\Bigg]$$

$$-R\sin\theta_2$$

$$\Bigg[n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)\frac{\partial C_p^{n+\frac{3}{2}}}{\partial\theta_2}(cos\theta_2)\Big)$$

$$(i\Omega)\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}P_{n-s}^{m}(cos\theta_2)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big(\sum_{p=0}^{\infty}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\Big)$$

$$(i\Omega)\Big[\sum_{m=-n}^{m=n}\varphi_{n,m}^{A}\sum_{s=0}^{n-m}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}\frac{\partial P_{n-s}^{m}(cos\theta_2)}{\partial\theta_2}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}e^{im\phi}\Big]\Big]$$

$$=\sum_{n=1}^{\infty}Z_n^2 \qquad (A2.3)$$

Appendix 3 of Chapter V

FIRST EQUATION

$$\sum_{n=|m|}^{\infty}\left[\frac{(n+1)}{a_2(i\Omega)}[p^B_{-(n+1),m}\int_0^{\pi}P_n^m(\cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2\,d\theta_2]\right.$$

$$+n\Big(\frac{(n+1)}{a_2}f_{n-1}(\sqrt{i\Omega}a_2)+(n+1)f_{n+1}(\sqrt{i\Omega}a_2)(i\Omega)a_2\Big)$$

$$\left.\varphi^B_{n,m}\int_0^{\pi}P_n^m(\cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2\,d\theta_2\right]$$

$$+\sum_{n=|m|}^{\infty}\left\{\frac{1}{i\Omega}\left[p^A_{-(n+1),m}\left(\frac{1}{R}\right)^{n+1}\sum_{s=m}^{\infty}\binom{n+s}{s+m}\frac{s}{a_2}\left(\frac{a_2}{R}\right)^s\right.\right.$$

$$\left.\int_0^{\pi}P_s^m(\cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2\,d\theta_2\right]$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\left[\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)\right.$$

$$\cdot\frac{R}{a_2}\chi^A_{n,m}\int_0^{\pi}P^m_{n-s}(\cos\theta_2)P_j^m(\cos\theta_2)C_p^{n+\frac{1}{2}}(cos\theta_2)\sin\theta_2\,d\theta_2$$

$$\left.\cdot\,a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)\right]$$

$$+\left[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}a_2)\right.$$

$$\left.\cdot\,\varphi^A_{n,m}\frac{(s-n)}{a_2}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\right]$$

$$\int_0^{\pi}P^m_{n-s}(cos\theta_2)C_p^{n-\frac{1}{2}}(cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2\,d\theta_2$$

$$+\left[-n\frac{(2n+1)!}{2^{n-1}n!}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)\cdot(i\Omega)\right.$$

$$\cdot \varphi^A_{n,m} \frac{(s-n)}{a_2} a_2^{n-s} \Big((R^2 + a_2^2) \int_0^\pi C_p^{n+\frac{3}{2}}(cos\theta_2) P^m_{n-s}(cos\theta_2) P^m_j(\cos\theta_2) \sin\theta_2 d\theta_2$$

$$-2Ra_2 \int_0^\pi \cos\theta_2 C_p^{n+\frac{3}{2}}(cos\theta_2) P^m_{n-s}(cos\theta_2) P^m_j(\cos\theta_2) \sin\theta_2 d\theta_2 \Big) \frac{R^s}{s!} \frac{(n+m)!}{(n+m-s)!} \Bigg]$$

$$+n(2n+1) \frac{(2n+1)!}{2^{n-1} n!} \Bigg[\sum_{p=0}^{\infty} \sum_{s=0}^{n-m} (n + \frac{3}{2} + p) f_{n+p+1}(\sqrt{i\Omega}R) \psi_{n+p+1}(\sqrt{i\Omega} a_2)$$

$$\cdot (i\Omega) \varphi^A_{n,m} \frac{(n-s)(n-s-1)}{a_2^2} a_2^{n-s} \frac{R^s}{s!} \frac{(n+m)!}{(n+m-s)!} \Bigg]$$

$$\cdot \Big(R \int_0^\pi \cos\theta_2 C_p^{n+\frac{3}{2}}(cos\theta_2) P^m_{n-s}(cos\theta_2) P^m_j(\cos\theta_2) \sin\theta_2 d\theta_2$$

$$+a_2 \int_0^\pi C_p^{n+\frac{3}{2}}(cos\theta_2) P^m_{n-s}(cos\theta_2) P^m_k(\cos\theta_2) \sin\theta_2 d\theta_2 \Big) \Bigg\}$$

$$= \sum_{n=|m|}^{\infty} X^2_{n,m} \int_0^\pi P^m_n(\cos\theta_2) P^m_j(\cos\theta_2) \sin\theta_2 d\theta_2 \qquad (A3.1)$$

SECOND EQUATION

$$\sum_{n=|m|}^{\infty} \Bigg[\frac{(n+1)(n+2)}{i\Omega a_2} \Big[p^B_{-(n+1),m} \int_0^\pi P^m_n(\cos\theta_2) P^m_j(\cos\theta_2) \sin\theta_2 \Big]$$

$$+n \Big(\frac{(n^2-1)}{a_2} f_{n-1}(\sqrt{i\Omega} a_2) + (n+1)^2 (i\Omega) a_2 f_{n+1}(\sqrt{i\Omega} a_2)$$

$$+(n+1)(i\Omega) a_2 f_n(\sqrt{i\Omega} a_2) + (n+1)(i\Omega)^2 a_2^3 f_{n+2}(\sqrt{i\Omega} a_2) \Big)$$

$$\cdot [\varphi^B_{n,m} \int_0^\pi P^m_n(\cos\theta_2) P^m_j(\cos\theta_2) \sin\theta_2 d\theta_2] \Bigg]$$

$$+a_2 \sum_{n=|m|}^{\infty} \Bigg\{ \frac{1}{i\Omega} \Bigg[p^A_{-(n+1),m} \Big(\frac{1}{R} \Big)^{n+1}$$

$$\sum_{s=m}^{\infty}\binom{n+s}{s+m}\frac{s(s-1)}{a_2^2}\left(\frac{a_2}{R}\right)^s\int_0^{\pi}P_s^m(\cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2\Bigg]$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\left[\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}a_2)\psi_{n+p+1}(\sqrt{i\Omega}a_2)\right.$$

$$\cdot\frac{R}{a_2}\Bigg(\chi_{n,m}^{A}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)$$

$$\left.\int_0^{\pi}C_p^{n+\frac{1}{2}}(cos\theta_2)P_{n-s}^m(cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2\Bigg)\right]$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\left[\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)\right.$$

$$\cdot\Bigg(\chi_{n,m}^{A}(s-n-1)a_2^{n-s+2}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)$$

$$\left.\int_0^{\pi}C_p^{n+\frac{1}{2}}(cos\theta_2)P_{n-s}^m(cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2\Bigg)\right]$$

$$+\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Bigg((n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}a_2)$$

$$\psi_{n+p}(\sqrt{i\Omega}r_2)\int_0^{\pi}C_p^{n-\frac{1}{2}}(cos\theta_2)P_{n-s}^m(cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2$$

$$+n\frac{(2n+1)!}{2^{n-1}n!}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)(\sqrt{i\Omega}a_2)(i\Omega)\Big[\big(\psi_{n+p+2}(\sqrt{i\Omega}a_2)(R^2+a_2^2)$$

$$-2a_2\psi_{n+p+1}(\sqrt{i\Omega}a_2)\big)\int_0^{\pi}C_p^{n+\frac{3}{2}}(cos\theta_2)P_{n-s}^m(cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2$$

$$+\big(2R\psi_{n+p+1}(\sqrt{i\Omega}a_2)-\psi_{n+p+2}(\sqrt{i\Omega}a_2)(2Ra_2)\big)$$

$$\int_0^{\pi}C_p^{n+\frac{3}{2}}(cos\theta_2)P_{n-s}^m(cos\theta_2)P_j^m(\cos\theta_2)\cos\theta_2\sin\theta_2 d\theta_2\Big]$$

$$\cdot\varphi_{n,m}^{A}\frac{(s-n)}{a_2}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Bigg)$$

$$+\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\Big((n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}a_2)$$

$$\int_0^{\pi} C_p^{n-\frac{1}{2}}(cos\theta_2)P_{n-s}^{m}(cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2\, d\theta_2\Big)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}\Bigg((n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)$$

$$\Big[(R^2+a_2^2)\int_0^{\pi} C_p^{n+\frac{3}{2}}(cos\theta_2)P_{n-s}^{m}(cos\theta_2)P_j^m(\cos\theta_2)\cos\theta_2\, d\theta_2$$

$$-2Ra_2\int_0^{\pi}\cos\theta_2 C_p^{n+\frac{3}{2}}(cos\theta_2)P_{n-s}^{m}(cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2\, d\theta_2\Big]\Bigg)\cdot(i\Omega)\Bigg]$$

$$\cdot\Big[\varphi_{n,m}^{A}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s-2}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big]$$

$$+n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Big((n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}s_2)\psi_{n+p+2}(\sqrt{i\Omega}a_2)\Big)$$

$$(i\Omega)\Big[\varphi_{n,m}^{A}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s-2}$$

$$\Big(R\int_0^{\pi} P_{n-s}^{m}(cos\theta_2)C_p^{n+\frac{3}{2}}(cos\theta_2)\cos\theta_2 P_j^m(\cos\theta_2)\sin\theta_2\, d\theta_2$$

$$+a_2\int_0^{\pi} P_{n-s}^{m}(cos\theta_2)C_p^{n+\frac{3}{2}}(cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2\, d\theta_2\Big)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big]\Bigg]$$

$$+\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big((n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)\Big)$$

$$(i\Omega)\Big\{\Big[\varphi_{n,m}^{A}\frac{(s-n)(n-s-1)(n-s-3)}{a_2^3}$$

$$\cdot a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big]$$

$$\Big([R\int_0^{\pi} C_p^{n+\frac{3}{2}}(cos\theta_2)P_{n-s}^{m}(cos\theta_2)\cos\theta_2 P_j^m(\cos\theta_2)\sin\theta_2\, d\theta_2$$

$$+a_2 \int_0^{\pi} C_p^{n+\frac{3}{2}}(cos\theta_2) P_{n-s}^m(cos\theta_2) P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2]\Big)$$

$$+\Big[\varphi_{n,m}^A \frac{(n-s)(n-s-1)}{a_2^2} a_2^{n-s} \frac{R^s}{s!} \frac{(n+m)!}{(n+m-s)!}$$

$$\int_0^{\pi} C_p^{n+\frac{3}{2}}(cos\theta_2) P_{n-s}^m(cos\theta_2) P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2\Big]\Big\}\Bigg\}$$

$$= \sum_{n=|m|}^{\infty} Y_{n,m}^2 \int_0^{\pi} P_n^m(\cos\theta_2) P_j^k(\cos\theta_2) d\theta_2 \qquad (A3.2)$$

THIRD EQUATION

$$-\sum_{n=|m|}^{\infty} n(n+1) f_n(\sqrt{i\Omega} r_a) \chi_{n,m}^B \int_0^{\pi} P_n^m(\cos\theta_2) P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2$$

$$+\sum_{p=0}^{\infty}\sum_{s=0}^{n-m} \frac{1}{a_2}\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}(n-\frac{1}{2}+p) f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}a_2)$$

$$\int_0^{\pi} \frac{\partial C_p^{n-\frac{1}{2}}(cos\theta_2)}{\partial\theta_2} P_{n-s}^m(cos\theta_2) P_j^m(\cos\theta_2) d\theta_2$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}(n+\frac{3}{2}+p)(i\Omega) f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)$$

$$\Bigg((R^2+a_2^2)\int_0^{\pi} \frac{\partial C_p^{n+\frac{3}{2}}(cos\theta_2)}{\partial\theta_2} P_{n-s}^m(cos\theta_2) P_j^m(\cos\theta_2) d\theta_2$$

$$-(2Ra_2)\int_0^{\pi} \cos\theta_2 \frac{\partial C_p^{n+\frac{3}{2}}(cos\theta_2)}{\partial\theta_2} P_{n-s}^m(cos\theta_2) P_j^m(\cos\theta_2) d\theta_2\Bigg)$$

$$-n\frac{(2n+1)!}{2^{n-1}n!}(n+\frac{3}{2}+p) f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)$$

$$\int_0^{\pi} C_p^{n+\frac{3}{2}}(cos\theta_2) P_{n-s}^m(cos\theta_2) P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2 \cdot (i\Omega)(2Ra_2)\Bigg]$$

$$\cdot\left[\varphi^A_{n,m}\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)\right]$$

$$+a_2\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\left[-\frac{(2n-1)!}{2^{n-2}(n-1)!}\left((n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)\right)\right]$$

$$\left[R\chi^A_{n,m}(n-s)(n-s-1)a_2^{n-s-2}\right.$$

$$\int_0^\pi \cos\theta_2 C_p^{n+\frac{1}{2}}(\cos\theta_2)P^m_{n-s}(cos\theta_2)P^m_j(\cos\theta_2)\sin\theta_2 d\theta_2\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}$$

$$-R\left(\chi^A_{n,m}(n-s-1)a_2^{n-s-2}\right.$$

$$\int_0^\pi \sin\theta_2 C_p^{n+\frac{1}{2}}(\cos\theta_2)\frac{\partial P^m_{n-s}(cos\theta_2)}{\partial\theta_2}P^m_j(\cos\theta_2)\sin\theta_2 d\theta_2$$

$$\left.\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\right)$$

$$+\left(\chi^A_{n,m}[2(n-s)+(n-s)(n-s-1)]a_2^{n-s-1}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\right.$$

$$\left.\left.\int_0^\pi C_p^{n+\frac{1}{2}}(\cos\theta_2)P^m_{n-s}(cos\theta_2)P^m_j(\cos\theta_2)\sin\theta_2 d\theta_2\right)\right]$$

$$+\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\left[-\frac{(2n-1)!}{2^{n-2}(n-1)!}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)\right]$$

$$\left[\left(\chi^A_{n,m}a_2^{n-s-1}\left(R\int_0^\pi \cos\theta_2\frac{\partial P^m_{n-s}(cos\theta_2)}{\partial\theta_2}\frac{\partial C_p^{n+\frac{1}{2}}(cos\theta_2)}{\partial\theta_2}P^m_j(\cos\theta_2)\sin\theta_2 d\theta_2\right.\right.\right.$$

$$\left.\left.+a_2\int_0^\pi\frac{\partial P^m_{n-s}(cos\theta_2)}{\partial\theta_2}\frac{\partial C_p^{n+\frac{1}{2}}(cos\theta_2)}{\partial\theta_2}P^m_j(\cos\theta_2)\sin\theta_2 d\theta_2\right)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\right)$$

$$+R\left(\chi^A_{n,m}(n-s)a_2^{n-s-1}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\right.$$

$$\left(R \int_0^\pi \cos\theta_2 \sin^2\theta_2 P_{n-s}^m(cos\theta_2) \frac{\partial C_p^{n+\frac{1}{2}}(cos\theta_2)}{\partial\theta_2} P_j^m(\cos\theta_2) d\theta_2 \right.$$

$$\left. \left. \left. + a_2 \int_0^\pi \sin^2\theta_2 P_{n-s}^m(cos\theta_2) \frac{\partial C_p^{n+\frac{1}{2}}(cos\theta_2)}{\partial\theta_2} P_j^m(\cos\theta_2) d\theta_2 \right) \right) \right]$$

$$-R \sum_{p=0}^{\infty} \sum_{s=0}^{n-m} \left[n(2n+1) \frac{(2n+1)!}{2^{n-1} n!} \Big((n + \frac{3}{2} + p) f_{n+p+1}(\sqrt{i\Omega} R) \psi_{n+p+1}(\sqrt{i\Omega} a_2) \Big) \right.$$

$$(i\Omega) \Big[\varphi_{n,m}^A \frac{(n-s)(n-s-1)}{a_2^2} a_2^{n-s} \frac{R^s}{s!} \frac{(n+m)!}{(n+m-s)!}$$

$$\int_0^\pi \frac{\partial C_p^{n+\frac{3}{2}}(\cos\theta_2)}{\partial\theta_2} \sin^2\theta_2 P_{n-s}^m(cos\theta_2) P_j^m(\cos\theta_2) d\theta_2 \Big]$$

$$+ n(2n+1) \frac{(2n+1)!}{2^{n-1} n!} \Big((n + \frac{3}{2} + p) f_{n+p+1}(\sqrt{i\Omega} R) \psi_{n+p+1}(\sqrt{i\Omega} a_2) \Big)$$

$$(i\Omega) \Big[\varphi_{n,m}^A \frac{(n-s)(n-s-1)}{a_2^2} a_2^{n-s} \frac{R^s}{s!} \frac{(n+m)!}{(n+m-s)!}$$

$$\left. \int_0^\pi C_p^{n+\frac{3}{2}}(cos\theta_2) \sin^2\theta_2 \frac{\partial P_{n-s}^m(cos\theta_2)}{\partial\theta_2} d\theta_2 \Big] \right]$$

$$= \sum_{n=|m|}^{\infty} Z_{n,m}^2 \int_0^\pi P_n^m(\cos\theta_2) P_j^m(\cos\theta_2) \sin\theta_2 \, d\theta_2 \qquad (A3.3)$$

Appendix 4 to Chapter V

The various theta integrals appearing in the algebraic conditions obtained from the boundary conditions using orthogonality are evaluated here. First, there is the well-known fact that

$$\int_0^\pi P_n^m(\cos\theta_2)P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2 = \frac{2}{2n+1}\frac{(n+m)!}{(n-m)!}\delta_{nj}. \qquad (A4.1)$$

Next, define functions

$$I_1(p,n,m,s,j) = \int_0^\pi C_p^{n+\frac{1}{2}}(\cos\theta_2)P_{n-s}^m(\cos\theta_2)P_j^m(\cos\theta_2)d\theta_2; \qquad (A4.2)$$

$$I_2(p,n,m,s,j) = \int_0^\pi C_p^{n+\frac{1}{2}}(\cos\theta_2)P_{n-s}^m(\cos\theta_2)P_j^m(\cos\theta_2)sin\theta_2 d\theta_2; \qquad (A4.3)$$

$$I_3(p,n,m,s,j) = \int_0^\pi C_p^{n+\frac{1}{2}}(\cos\theta_2)P_{n-s}^m(\cos\theta_2)P_j^m(\cos\theta_2)\cos\theta_2 sin\theta_2 d\theta_2; \quad (A4.4)$$

$$I_4(p,n,m,s,j) = \int_0^\pi \cos\theta_2 C_p^{n+\frac{1}{2}}(\cos\theta_2)P_{n-s}^m(\cos\theta_2)P_j^m(\cos\theta_2)d\theta_2; \qquad (A4.5)$$

$$I_5(p,n,m,s,j) = \int_0^\pi \frac{dC_p^{n+\frac{1}{2}}(\cos\theta_2)}{d\theta_2}P_{n-s}^m(\cos\theta_2)P_j^m(\cos\theta_2)d\theta_2; \qquad (A4.6)$$

$$I_6(p,n,m,s,j) = \int_0^\pi \frac{dC_p^{n+\frac{1}{2}}(\cos\theta_2)}{d\theta_2}P_{n-s}^m(\cos\theta_2)P_j^m(\cos\theta_2)\cos\theta_2 d\theta_2; \qquad (A4.7)$$

$$I_7(p,n,m,s,j) = \int_0^\pi C_p^{n+\frac{1}{2}}(\cos\theta_2)\frac{dP_{n-s}^m(\cos\theta_2)}{d\theta_2}P_j^m(\cos\theta_2)\sin^2\theta_2 d\theta_2; \qquad (A4.8)$$

$$I_8(p,n,m,s,j) = \int_0^\pi \cos\theta_2\frac{dC_p^{n+\frac{1}{2}}(\cos\theta_2)}{d\theta_2}\frac{dP_{n-s}^m(\cos\theta_2)}{d\theta_2}P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2; \qquad (A4.9)$$

$$I_9(p,n,m,s,j) = \int_0^\pi \frac{\partial C_p^{n+\frac{1}{2}}(\cos\theta_2)}{\partial\theta_2}P_{n-s}^m(\cos\theta_2)P_j^m(\cos\theta_2)\sin^2\theta_2 d\theta_2; \qquad (A4.10)$$

$$I_{10}(p,n,m,s,j) = \int_0^\pi \frac{\partial P_{n-s}^m(cos\theta_2)}{\partial\theta_2}\frac{\partial C_p^{n+\frac{1}{2}}(cos\theta_2)}{\partial\theta_2}P_j^m(\cos\theta_2)\sin\theta_2 d\theta_2; \quad (A4.11)$$

$$I_{11}(p,n,m,s,j) = \int_0^\pi P_{n-s}^m(cos\theta_2)\frac{\partial C_p^{n+\frac{1}{2}}(cos\theta_2)}{\partial\theta_2}P_j^m(\cos\theta_2)\cos\theta_2\sin^2\theta_2 d\theta_2. \quad (A4.12)$$

By use of identities like

$$\cos\theta_2 P_n^m(\cos\theta_2) = \frac{(n-m+1)P_{n+1}^m(\cos\theta_2)+(n+m)P_{n-1}^m(\cos\theta_2)}{(2n+1)}, \quad (A4.13))$$

$$\sin^2\theta_2\frac{dP_n^m(\cos\theta_2)}{d\theta_2} = (n+m)P_{n-1}^m(\cos\theta_2) - n\cos\theta_2 P_n^m(\cos\theta_2), \quad (A4.14)$$

$$\frac{1}{\sin\theta_2}\frac{dC_n^\lambda(\cos\theta_2)}{d\theta_2} = -2\lambda C_{n-1}^{\lambda+1}(\cos\theta_2), \quad (A4.15)$$

and

$$\sin\theta_2 P_n^m(\cos\theta_2) = P_{n+1}^{m+1}(\cos\theta_2) - P_{n-1}^{m+1}(\cos\theta_2), \quad (A4.16)$$

it can be shown that

$$I_2(p,n,m,s,j) = I_1(p,n,m+1,s,j+1) - I_1(p,n,m+1,s,j-1); \quad (A4.17)$$

$$I_3(p,n,m,s,j) = \frac{j-m+1}{2n+1}I_2(p,n,m,s,j+1) + \frac{j+m}{2n+1}I_2(p,n.m,s,j-1); \quad (A4.18)$$

$$I_4(p,n,m,s,j) = \frac{(j-m+1)}{(2j+1)}I_1(p,n,m,s,j+1) + \frac{(j+m)}{(2j+1)}I_1(p,n,m,s,j-1); \quad (A4.19)$$

$$I_5(p,n,m,s,j) = -2I(p-1,n+1,m,s+1,j); \tag{A4.20}$$

$$\begin{aligned} I_6(p,n,m,s,j) =&(-2)(n+\frac{3}{2})[\frac{j-m+1}{2n+3}I(p-1,n+1,m,s+1,j+1) \\ &+\frac{j+m}{2n+3}I(p-1,n+1,m,s+1,j-1)]; \end{aligned} \tag{A4.21}$$

$$\begin{aligned} I_7(p,n,m,s,j) =&I_1(p,n,m,s+1,j) - \frac{(n-s)(j+m)}{(2j+1)}I_1(p,n,m,s,j+1) \\ &-\frac{(n-s)(j+m)}{(2j+1)}I_1(p,n,m,s,j-1); \end{aligned} \tag{A4.22}$$

$$\begin{aligned} I_8(p,n,m,s,j) =& -(2n+1)(n-s+m)I_3(p-1,n,m,s+1,j) \\ &+\frac{(2n+1)(n-s)(j-m+1)}{(2j+1)}I_4(p-1,n+1,m,s+1,j+1) \\ &+\frac{(2n+1)(n-s)(j+m)}{(2j+1)}I_4(p-1,n+1,m,s+1,j-1); \end{aligned} \tag{A4.23}$$

$$\begin{aligned} I_9(p,n,m,s,j) =& -(2n+1)I_1(p-1,n+1,m,s+1,j) \\ &+\frac{(2n+1)(j-m+1)}{(2j+1)}I_3(p-1,n+1,m,s+1,j+1) \\ &+\frac{(2n+1)(j+m)}{(2j+1)}I_3(p-1,n+1,m,s+1,j-1); \end{aligned} \tag{A4.24}$$

$$\begin{aligned} I_{10}(p,n,m,s,j) =& -(2n+1)(n-s+m)I_1(p-1,n+1,m,s,j) \\ &-(2n+1)(n-s)I_1(p-1,n+1,m,s+1,j); \end{aligned} \tag{A4.25}$$

$$\begin{aligned} I_{11}(p,n,m,s,j) =& -(2n+1)\frac{(j-m+1)(j-m+2)}{(2j+1)(2j+3)}I_3(p-1,n+1,m,s+1,j+2) \\ &-(2n+1)\frac{[(j+1)^2+j^2-2m^2]}{(2j+3)(2j+1)^2(2j-1)}I_3(p-1,n+1,m,s+1,j) \\ &-(2n+1)\frac{(j+m)(j+m-1)}{(2j+1)(2j-1)}I_3(p-1,n+1,m,s+1,j-2). \end{aligned} \tag{A4.26}$$

Appendix 5 to Chapter V

This appendix gives the constants appearing in the algebraic equations for the coefficients $p^A_{-(n+1),m}, p^B_{-(n+1),m}$, $\chi^A_{n,m}, \chi^B_{n,m}$, $\varphi^A_{n,m}$, $\varphi^B_{n,m}$ that result from applying the boundary conditions at sphere B. Recall that the equations are (for $j = 1, 2, 3, ...$ and $m = 1, 2, ..., j$):

$$a^{(1)}_{j,m} p^B_{-(j+1),m} + a^{(2)}_{j,m}\varphi^B_{j,m} + \sum_{n=|m|}^{\infty}\left[a^{(3)}_{n,j,m}p^A_{n,m} a^{(4)}_{n,j,m}\varphi^A_{n,m} a^{(5)}_{n,j,m}\chi^A_{n,m}\right]$$

$$= X^2_{j,m}\frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}; \qquad (A5.1)$$

$$b^{(1)}_{j,m} p^B_{-(j+1),m} + b^{(2)}_{j,m}\varphi^B_{j,m} + \sum_{n=|m|}^{\infty}\left[b^{(3)}_{n,j,m}p^A_{n,m} b^{(4)}_{n,j,m}\varphi^A_{n,m}\right]$$

$$= Y^2_{j,m}\frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}; \qquad (A5.2)$$

$$c^{(1)}_{n,m}\chi^B_{n,m} + \sum_{n=|m|}^{\infty}\left[c^{(3)}_{n,j,m}p^A_{n,m} + c^{(4)}_{n,j,m}\varphi^A_{n,m} + c^{(5)}_{n,j,m}\chi^A_{n,m}\right]$$

$$= Z^2_{j,m}\frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}. \qquad (A5.3)$$

In these equations,

$$a^{(1)}_{j,m} = \frac{(j+1)}{a_2(i\Omega)}\frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}; \qquad (A5.4)$$

$$a^{(2)}_{j,m} = j\Big(\frac{(j+1)}{a_2}f_{j-1}(\sqrt{i\Omega}a_2) + (j+1)f_{j+1}(\sqrt{i\Omega}a_2)(i\Omega)a_2\Big)\frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}; \qquad (A5.5)$$

$$a^{(3)}_{n,j,m} = \frac{1}{i\Omega}\left[\left(\frac{1}{R}\right)^{n+1} \sum_{s=m}^{\infty} \binom{n+s}{s+m} \frac{s}{a_2}\left(\frac{a_2}{R}\right)^s \frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}\right]; \qquad (A5.6)$$

$$a^{(4)}_{n,j,m} = \sum_{n=|m|}^{\infty}\Bigg\{\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)$$

$$\cdot\ \psi_{n+p-1}(\sqrt{i\Omega}a_2)\frac{(s-n)}{a_2}a_2^{n-s}I_2(p,n,m,s,j)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Bigg]$$

$$- n\frac{(2n+1)!}{2^{n-1}n!}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Bigg[(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}r_2)\cdot(i\Omega)$$

$$\cdot\ \frac{(s-n)}{a_2}a_2^{n-s}\Big((R^2+a_2^2)I_2(p,n+1,m,s+1,j)$$

$$- 2Ra_2I_3(p,n+1,m,s+1,j)\Big)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Bigg]$$

$$+ n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Bigg[\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)$$

$$\cdot(i\Omega)\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}$$

$$\cdot\Big(RI_3(p,n+1,m,s+1,j)+a_2I_2(p,n+1,m,s+1,j)\Big)\Bigg]\Bigg\}; \qquad (A5.7)$$

$$a^{(5)}_{n,j,m} = \sum_{n=|m|}^{\infty}\Bigg\{-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Bigg[\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)$$

$$\cdot\ \frac{R}{a_2}I_2(p,n,m,s,j)a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)\Bigg]\Bigg\}; \qquad (A5.8)$$

$$b^{(1)}_{j,m} = \left[\frac{(j+1)(j+2)}{i\Omega a_2}\left[\frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}\right]\right.; \qquad (A5.9)$$

$$b^{(2)}_{j,m} = j\Big(\frac{(j^2-1)}{a_2}f_{j-1}(\sqrt{i\Omega}a_2)+(j+1)^2(i\Omega)a_2f_{j+1}(\sqrt{i\Omega}a_2)$$

$$+ (j+1)(i\Omega)a_2 f_j(\sqrt{i\Omega}a_2) + (j+1)(i\Omega)^2 a_2^3 f_{j+2}(\sqrt{i\Omega}a_2)\Big) \cdot \frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}; \tag{A5.10}$$

$$b^{(3)}_{n,j,m} = a_2 \sum_{n=|m|}^{\infty} \Bigg\{ \frac{1}{i\Omega}\Bigg[\Big(\frac{1}{R}\Big)^{n+1}$$

$$\sum_{s=m}^{\infty} \binom{n+s}{s+m} \frac{s(s-1)}{a_2^2}\left(\frac{a_2}{R}\right)^s \frac{2}{(2s+1)}\frac{(s+m)!}{(s-m)!}\delta_{sj}\Bigg]; \tag{A5.11}$$

$$b^{(4)}_{n,j,m} = a_2 \sum_{n=|m|}^{\infty} \Bigg\{ -\frac{(2n-1)!}{2^{n-2}(n-1)!}\Bigg[\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n+\frac{1}{2}+p) f_{n+p}(\sqrt{i\Omega}R)(-\sqrt{i\Omega}a_2)$$

$$\psi_{n+p+1}(\sqrt{i\Omega}a_2) \cdot \frac{R}{a_2}\Big(a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im) I_2(p,n,m,s,j)\Big)\Bigg]$$

$$-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Bigg[\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}(n+\frac{1}{2}+p) f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)$$

$$\cdot \Big((s-n-1)a_2^{n-s+2}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im) I_2(p,n,m,s,j)\Big)\Bigg]\Bigg\}; \tag{A5.12}$$

$$b^{(5)}_{n,j,m} = a_2 \sum_{n=|m|}^{\infty} \Bigg\{ (n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Bigg((\frac{1}{2}-n-p) f_{n+p-1}(\sqrt{i\Omega}R)$$

$$(\sqrt{i\Omega}a_2)\psi_{n+p}(\sqrt{i\Omega}a_2) I_2(p,n-1,m,s-1,j)$$

$$+ n\frac{(2n+1)!}{2^{n-1}n!}(n+\frac{3}{2}+p) f_{n+p+1}(\sqrt{i\Omega}R)(\sqrt{i\Omega}a_2)(i\Omega)\Big[\big(\psi_{n+p+2}(\sqrt{i\Omega}a_2)(R^2+a_2^2)$$

$$-2a_2\psi_{n+p+1}(\sqrt{i\Omega}a_2)\big) I_2(p,n+1,m,s+1,j)$$

$$+\big(2R\psi_{n+p+1}(\sqrt{i\Omega}a_2) - \psi_{n+p+2}(\sqrt{i\Omega}a_2)(2Ra_2)\big) I_3(p,n+1,m,s+1,j)\Big]$$

$$\cdot \frac{(s-n)}{a_2} a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Bigg)$$

$$+\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}\Big((n-\frac{1}{2}+p) f_{n+p-1}(\sqrt{i\Omega}R)\psi_{n+p-1}(\sqrt{i\Omega}a_2)$$

$$I_2(p,n-1,m,s-1,j)\Big)$$

$$- n\frac{(2n+1)!}{2^{n-1}n!}\Bigg((n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)$$

$$\Big[(R^2+a_2^2)I_4(p,n,m,s,j) - 2Ra_2I_3(p,n+1,m,s+1,j)\Big]\Bigg)\cdot(i\Omega)\Bigg]$$

$$\cdot\ \Big[\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s-2}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big]$$

$$+ n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Big((\frac{3}{2}-n-p)f_{n+p+1}(\sqrt{i\Omega}R)(\sqrt{i\Omega}s_2)$$

$$\psi_{n+p+2}(\sqrt{i\Omega}a_2)\Big)(i\Omega)\Big[\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s-2}$$

$$\Big(RI_3(p,n+1,m,s+1,j)+a_2I_2(p,n+1,m,s+1,j)\Big)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big]$$

$$+\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big((n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)\Big)$$

$$(i\Omega)\Big\{\Big[\frac{(s-n)(n-s-1)(n-s-3)}{a_2^3}\cdot a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big]$$

$$\Big([RI_3(p,n+1,m,s+1,j)+a_2I_2(p,n+1,m,s+1,j)]\Big)$$

$$+\Big[\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}$$

$$I_2(p,n+1,m,s+1,j)\Big]\Big\}\Bigg\}; \qquad (A5.13)$$

$$c_{n,m}^{(1)} = -j(j+1)f_j(\sqrt{i\Omega}r_a)\frac{2}{(2j+1)}\frac{(j+m)!}{(j-m)!}; \qquad (A5.14)$$

$$c_{n,j,m}^{(4)} = -\sum_{n=|m|}^{\infty}\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\frac{1}{a_2}\Bigg[(n+1)\frac{(2n-3)!}{2^{n-3}(n-2)!}(n-\frac{1}{2}+p)f_{n+p-1}(\sqrt{i\Omega}R)$$

$$\psi_{n+p-1}(\sqrt{i\Omega}a_2)I_5(p,n-1,m,s-1,j)$$

$$- n\frac{(2n+1)!}{2^{n-1}n!}(n+\frac{3}{2}+p)(i\Omega)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)$$

$$\Bigg((R^2+a_2^2)I_5(p,n+1,m,s+1,j)-(2Ra_2)I_6(p,n+1,m,s+1,j)\Bigg)$$

$$- n\frac{(2n+1)!}{2^{n-1}n!}(n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)$$

$$I_2(p,n+1,m,s+1,j)\cdot(i\Omega)(2Ra_2)\Bigg]$$

$$\cdot\Big[\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}(im)\Big]$$

$$- R\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Bigg[n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big((n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)\Big)$$

$$(i\Omega)\Big[\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}I_{12}(p,n+1,m,s+1,j)\Big]$$

$$+ n(2n+1)\frac{(2n+1)!}{2^{n-1}n!}\Big((n+\frac{3}{2}+p)f_{n+p+1}(\sqrt{i\Omega}R)\psi_{n+p+1}(\sqrt{i\Omega}a_2)\Big)$$

$$(i\Omega)\Big[\frac{(n-s)(n-s-1)}{a_2^2}a_2^{n-s}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}I_7(p,n+1,m,s+1,j)\Big]\Bigg] \quad (A5.15)$$

$$c_{n,j,m}^{(5)} = -\sum_{n=|m|}^{\infty}a_2\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Bigg[-\frac{(2n-1)!}{2^{n-2}(n-1)!}(n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)\Bigg]$$

$$\Bigg[\Bigg(R\Big((n-s)(n-s-1)a_2^{n-s-2}I_3(p,n,m,s,j)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big)\Bigg)$$

$$+\Bigg(\Big(-R\Big((n-s-1)a_2^{n-s-2}I_7(p,n,m,s,j)\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big)\Bigg)$$

$$+\Big(\big(2(n-s)+(n-s)(n-s-1)\big]a_2^{n-s-1}\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}I_2(p,n,m,s,j)\Big)\Big)\Bigg]$$

$$+\sum_{p=0}^{\infty}\sum_{s=0}^{n-m}\Bigg[-\frac{(2n-1)!}{2^{n-2}(n-1)!}\Bigg((n+\frac{1}{2}+p)f_{n+p}(\sqrt{i\Omega}R)\psi_{n+p}(\sqrt{i\Omega}a_2)\Bigg)\Bigg]$$

$$\Bigg[\frac{1}{a_2}\Big(a_2^{n-s}\Bigg[RI_8(p,n,m,s,j)+a_2I_{13}(p,n,m,s,j)\Bigg]\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\Big)$$

$$+ R\left((n-s)a_2^{n-s-1}\left[RI_{14}(p,n,m,s,j)+a_2 I_{12}(p,n,m,s,j)\right]\frac{R^s}{s!}\frac{(n+m)!}{(n+m-s)!}\right)\Bigg]. \tag{A5.16}$$

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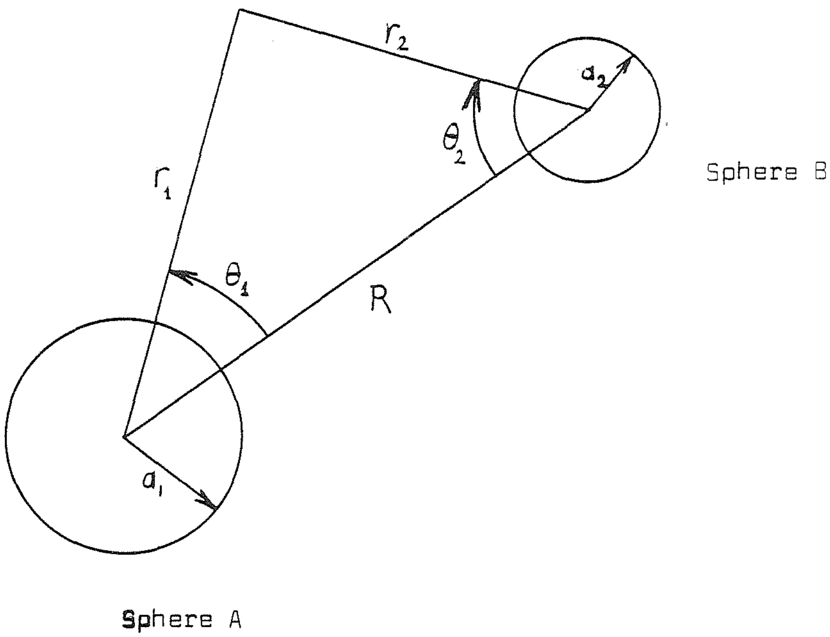

FIGURE 1

The two spherical coordinate systems.
The plane $\phi = 0$ is shown.