THEORY OF GAS BUBBLE DYNAMICS IN OSCILLATING PRESSURE FIELDS

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ABSTRACT

The behavior of a permanent gas bubble in liquids under oscillating pressure fields is studied by a linearized theory. The derived thermodynamic relation tends to indicate average isothermality for high frequency limit, contrary to the usual intuitive reasonings. The growth of the gas bubble under the oscillating pressure fields due to the effect of rectification of mass is also investigated. The effect is small, being of second order, but accumulating. The absence of resulting large bubbles is explained briefly by the considerations on the stability of spherical shape of the bubbles.

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INTRODUCTION

In the absence of an external force field, a spherical gas bubble in a liquid of infinite extent will stay at rest if the internal pressure is in equilibrium with the ambient pressure. Any disturbance from the equilibrium conditions will result in a complicated motion of the gas bubble. It may undergo oscillating expansion and contraction, and cause diffusion of gas to and from the surrounding medium due to the inbalance of the pressure; or it may undergo translational or rotational motion and distortion of the shape due to the asymmetry of the disturbance. Moreover, as the bubble usually contains some vapor of the surrounding fluid, evaporation and condensation will occur due to the disturbance.

A complete, quantitative analysis of the problem is very difficult. However, it is hoped that partial understanding of a simplified problem would eventually lead to the full understanding of the underlying mechanism of the complete problem. In this sense, the simplification is not so much based on approximation to the physical situation as on the simplicity of the model for theoretical considerations.

Now let us take the case in which the disturbance is spherically symmetric, and the temperature is low enough so that practically no vapor of the surrounding liquid is present inside the gas bubble. As the initial geometrical conditions and the disturbances are all spherically symmetric, it is to be expected that the subsequent changes would show the same symmetry. In general, the resulting motion will be different according to the duration of the disturbance.

If the duration of the disturbance is a finite interval of time, we expect that the system will be in a new equilibrium state after some kind of oscillations damped out by the effects of viscosity, heat conduction, surface tension and perhaps also the non-linearity of the problem. On the other hand, if the external disturbance persists for a long time, its effects would also vary according to whether the disturbance is oscillatory or monotonic. For the latter case, the bubble may collapse or grow indefinitely until the stability of the system is of significance. For the former case, we may expect that a final steady-state oscillatory motion will be attained after the transients die out.

For the case of a disturbance of finite duration, an underwater explosion seems to be a good example. This phenomenon, however, is not only violent but has important asymmetries so that it hardly fits into the category of our simplified models. It is not likely that some time after the explosion we can actually find the "stationary gas bubble" in equilibrium with the water. In one of the earliest papers dealing with gas bubble dynamics, Rayleigh [1] has deduced how the gas bubble would perform free oscillations. This free oscillation would not persist in a model permitting heat conduction and viscous effects. On the other hand, the treatment of the growth of a bubble in super-heated [2] or super-saturated [3] medium could be taken as the case of the disturbance of long duration with monotonic nature at least before the very final equilibrium state is reached.

For the case of an oscillatory disturbance, the existence of a steady-state motion is actually only a conjecture even for this simplified model. In the first place, it may not be true for certain non-linear cases. Secondly, even if the dynamical problem has a steady-state solution, other factors such as mass diffusion and heat conduction may produce some one-sided effects which could cause a steady growth of the bubble. However if the disturbance is small enough so that the problem can be linearized, we shall see that the steady-state is actually attained, and the one-sided effects are of the second order.

The problem with oscillating disturbance is closely related to the problem of wave propagation in a medium containing gas bubbles. Both problems of scattering of sound waves by a gas bubble and the propagation of shock waves through a gassy medium have been treated before [4, 5]. The thermodynamic behavior of the bubble under the influence of the wave, which was not considered in those investigations, will be examined here. Furthermore, in the limit of very long wave-lengths, this spherically symmetric problem is a kind of "dipole approximation" for the study of the behavior of the gas bubble when the sound waves pass by.

The present paper contains two main parts. The first part deals with the dynamics of the gas bubble under oscillating pressure fields. The second part deals with the above mentioned one-sided effect, which is sometimes called "rectification".

The thermodynamic behavior of the bubble which is of main interest in the first part of this analysis may also be considered by the following qualitative arguments.

Let D_1 be the thermal diffusivity of the liquid; then we may define the diffusion length as $R_{D_1} = \sqrt{\frac{D_1}{\omega}}$, which should characterize the situation in conduction of heat for the system under oscillatory disturbance with frequency ω . Now for any increment of temperature ΔT in the bubble, the increase in internal energy is

$$\Delta h_2 = \frac{4}{3} \pi R_o^3 \rho_2 C_2 \Delta T \quad ,$$

where R_o is the radius of the bubble, ρ_2 is the density of the gas and C_2 is the specific heat at constant volume of the gas. On the other hand, the flow of heat from the bubble during a half-cycle is approximately

$$\Delta h_1 = K_1 \frac{\Delta T}{R_{D_1}} 4\pi R_o^2 \times \frac{\pi}{\omega} \quad ,$$

where k_1 is the coefficient of thermal conductivity for the liquid. Since $D_1 = \frac{k_1}{\rho_1 C_1}$, where ρ_1 is the density of the liquid and C_1 is the specific heat of the liquid, thus

$$\Delta h_1 = 4\pi^2 R_o^2 \rho_1 C_1 D_1 \frac{\Delta T}{\omega \sqrt{\frac{D_1}{\omega}}}$$

$$= 4\pi^2 R_o^2 \rho_1 C_1 \sqrt{\frac{D_1}{\omega}} \Delta T \ .$$

Hence

$$\frac{\Delta h_1}{\Delta h_2} = 3\pi \frac{\rho_1 C_1 \sqrt{\frac{D_1}{\omega}}}{\rho_2 C_2 R_o} \ .$$

If $\Delta h_2 << \Delta h_1$, i.e. $3\pi \frac{\rho_1 C_1}{\rho_2 C_2 R_o} \sqrt{\frac{D_1}{\omega}} >> 1$, then only an insignificant part of the transferred energy is available for the increase of internal energy, or equivalently, for raising the temperature of the bubble. Hence, it is legitimate to say that as far as the gas bubble is concerned, the entire thermodynamic process is essentially isothermal. On the other hand, if $\Delta h_1 << \Delta h_2$, adiabatic behavior may be expected from corresponding arguments.

It will be shown below that quite similar results can be derived from a detailed analysis if the condition of uniformity inside the bubble is imposed on our properly chosen model. The condition of uniform interior is equivalent to the assumption of an infinite value for the coefficient of heat conductivity of the gas, which is far from the case. After taking care of the finiteness of this coefficient of heat conductivity, we no longer get the adiabatic limit. In a way, this result is quite puzzling because it is contrary to usual belief that adiabaticity should be a natural result in the high frequency limit, based on the traditional physical argument made by Laplace to account for the correct velocity of sound in air. But no matter how successful Laplace's theory is, we still do not know the details of the thermodynamic process involved in the propagation of sound waves.

In the following analysis, we shall consider the model that consists of a spherical bubble of perfect gas in an incompressible, inviscid liquid of infinite extent. In the first part, we shall first deal with the case of uniform interior; then the more realistic case without this requirement will be considered. With this model it can be seen that potential flow may be assumed as far as the liquid is concerned. It may be remarked that for this spherically symmetric problem, the irrotationality will not be destroyed by the introduction of viscous effects. Moreover, in the linearized analysis, the inclusion of the viscous term will not change the essential features of the problem, except for a slight modification if the liquid is not very viscous. This is partly due to the fact that the effect of heat conduction has already played the role of a damping factor in the system.

For the second part on the phenomenon of rectification, the underlying physical picture may be seen as follows. When the gas bubble is compressed, due to the increase in internal pressure, the gas concentration at the bubble wall will rise above the equilibrium value, and thus results in the outflow of gas. On the other hand, gas will flow into the bubble during expansion. But owing to the difference in surface area of the bubble wall between the half-cycles of compression and expansion, there is a net inflow of gas over a complete cycle. A quasi-static approach has been adopted by Blake [6]to account for this phenomenon. The present paper gives a more complete analysis based on a linearization procedure.

As already remarked, the net inflow obtained in rectification is a small quantity of the second order if we consider the disturbance introduced in the analysis of part I as a first order small quantity. Thus, there is no inconsistency in neglecting this effect when we deal with the problem of part I. The effect of convection, which is neglected by Blake, being also of second order, is properly included in our treatment to give a complete consistent analysis.

An analogous phenomenon of rectification of heat may be expected to occur also. After knowing the exact, detailed thermodynamic behavior of the oscillating gas bubble, the analysis can be carried out in a similar fashion.

I. LINEARIZED THEORY OF PERMANENT GAS BUBBLE IN LIQUIDS UNDER OSCILLATING PRESSURE FIELDS.

A. Bubble with Uniform Interior

1. Linearized Formulation

The Bernoulli equation for the potential flow of an incompressible, inviscid fluid with spherical symmetry is

$$\frac{P_1}{\rho_1} + \frac{1}{2}(\nabla\phi)^2 - \frac{\partial\phi}{\partial t} = K(t) \quad ,$$

where ρ_1 is the density of the fluid, P_1 denotes the pressure, ϕ is the velocity potential such that the velocity of the fluid particle at any point $\vec{q}_1 = -\nabla\phi$, and K(t) is some function of time only. For the spherically symmetric case, ϕ can be expressed up to some additive function of time only, as

$$\phi = \frac{R^2\dot{R}}{r} \quad ,$$

where $\dot{R}$ is dR/dt, and where $r = R$ describes the location of the bubble wall. Thus, with $P_\infty(t)$ as the pressure at infinity, the Bernoulli equation can be re-expressed as

$$\frac{P_1}{\rho_1} + \frac{1}{2}\left(\frac{R^4\dot{R}^2}{r^4}\right) - \frac{1}{r}\left(2R\dot{R}^2 + R^2\ddot{R}\right) = \frac{P_\infty(t)}{\rho_1} \quad . \tag{1}$$

In particular, at the bubble wall where $r = R$, we have

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{P_e}{\rho_1} - \frac{P_\infty}{\rho_1} \quad , \tag{2}$$

where P_e is the pressure of the liquid, or external pressure at $r = R$ (cf. [7]). Let P_2 be the pressure of the gas inside the bubble, which is assumed here to be uniform, and let σ be the surface tension constant. Then, due to the effect of surface tension, the balance of external and internal pressure is

$$P_2 = P_e + \frac{2\sigma}{R} \quad .$$

Hence the dynamic equation (2) becomes

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{1}{\rho_1}\left[P_2 - \frac{2\sigma}{R} - P_\infty(t) \right] . \qquad (2')$$

Let T_1 be the temperature at any point in the liquid; then the heat equation, which expresses the conservation of energy, assumes the following form:

$$\nabla^2 T_1 = \frac{1}{D_1}\left(\frac{\partial T_1}{\partial t} + \vec{q}_1 \cdot \nabla T_1 \right) , \qquad (3)$$

where $D_1 = \frac{k_1}{\rho_1 C_1}$ is the thermal diffusivity of the liquid, k_1 and C_1 are coefficients of thermal conductivity and specific heat of the liquid respectively. The gas inside the bubble is assumed to be perfect and the temperature is uniform throughout the gas bubble. Thus we have

$$P_2(R) R^3 = N T_2 \quad , \qquad (4)$$

where N is a constant and is actually equal to $\frac{3}{4\pi}$ nR, R is the universal gas constant, and n is the number of mols of gas inside the bubble. Also we note that by the requirement of continuity of temperature, we have $T_2 = T_1$ (r=R).

Because of the assumed uniformity inside the gas bubble, the problem of the entire gas-liquid system is equivalent to the problem of liquid only, with the state of the gas serving as boundary condition at the bubble wall the equation of motion of which is defined by equation(2). In this sense, the boundary condition of the heat equation (3) at the bubble wall can be formulated on the basis of energy consideration for the gas bubble, as follows:

$$M C_2 \frac{dT_2}{dt} = -4\pi R^2 P_2(R) \frac{dR}{dt} + k_1 4\pi R^2 \left(\frac{\partial T_1}{\partial r}\right)_{r=R} , \quad (5)$$

where M is the total mass of the gas which is assumed to be constant, and C_2 is the specific heat at constant volume of the gas. The left hand side of equation(5) represents the increase of internal energy of the gas bubble, while the terms on the right hand side represent the work done on the bubble and the heat flow into the bubble. Since the temperature at infinity is assumed to be constant at all times, we have

$$T_1(\infty, t) = T_\infty , \quad (6)$$

where T_∞ is constant.

We also assume that the disturbance begins at a certain instant which we call t = 0. Thus everything is in equilibrium for $t \leqslant 0$. In other words, we have, for $t \leqslant 0$:

$$R(t) = R_0 , \quad (7)$$

$$\dot{R}(t) = 0 , \quad (8)$$

$$\ddot{R}(t) = 0 , \quad (8')$$

$$P_\infty(t) = P_0 , \quad (9)$$

and

$$T_1(r,t) = T_2(t) = T_\infty , \quad (10)$$

where R_o is the equilibrium radius of the bubble and P_o is the equilibrium pressure in the liquid.

The disturbance is introduced by a perturbing oscillating pressure field at infinity which can be expressed as

$$P_\infty(t) = P_o\left[1+\epsilon(t)\right] = P_o\left[1+\epsilon_o e^{i\omega t}\right], \quad \text{for } t>0. \tag{11}$$

It should be remarked that the complex quantity is introduced solely for simplification of computations, and only the real part has physical significance. Since all the subsequent operations are linear, the physically significant solution is just the real part of the solution of the same problem with complex quantities.

The linearization procedure is carried out with respect to the equilibrium configuration and is based on the smallness of ϵ_o in comparison with unity. It is conceivable that as ϵ_o gets smaller and smaller, the system will approach the equilibrium configuration unless the equilibrium is an instable one. That such instability does not appear is confirmed by our results.

Since the system is in equilibrium for $t \leqslant 0$, we immediately have the following relations, namely,

$$P_2(R_o) - P_o = \frac{2\sigma}{R_o}; \tag{12}$$

$$T_2(R_o) = T_\infty; \tag{13}$$

and

$$P_2(R_o)\,R_o^3 = N\,T_\infty. \tag{14}$$

Now let

$$R = R_0 (1 + x) \quad ; \tag{15}$$

$$P_2 = P_0 (a + p) = P_{2(R_0)} \left(1 + \frac{p}{a}\right) \quad ; \tag{16}$$

$$T_1 = T_\infty (1 + \theta_1) \quad ; \qquad T_2 = T_\infty (1 + \theta_2) \quad ; \tag{17}$$

where x, p, θ_1, θ_2 as well as ϵ are small quantities in comparison with unity. The linearized equations are obtained by inserting (15), (16), and (17) into the governing equations and boundary conditions and neglecting those terms of second order.

Thus equation (2) becomes

$$R_0 \ddot{x} = \frac{1}{\rho_1} \left[P_0 (a + p) - \frac{2\sigma}{R_0} (1 - x) - P_0 (1 + \epsilon) \right] . \tag{18}$$

Let us now write

$$\alpha = \frac{P_0}{\rho_1 R_0^2} \quad ;$$

and

$$W_r = \frac{2\sigma}{P_0 R_0} \quad . \tag{19}$$

W_r is sometimes called the Weber's number; it measures the relative importance of the surface tension effect relative to the inertia effects. Using equations (16) and (12) we obtain

$$a = 1 + W_r \quad , \tag{20}$$

and by equation (12), equation (18) becomes

$$\ddot{x} - \alpha W_r x = \alpha (p - \epsilon) \quad . \tag{21}$$

Inserting equations (15) - (17) into (4), using (14) and linearizing, we have

$$p = a\theta_2 - 3ax \quad . \tag{22}$$

The heat equation (3), after linearization, becomes the ordinary diffusion equation without convection:

$$\nabla^2 \theta_1 = \frac{1}{D_1} \frac{\partial \theta_1}{\partial t} \quad , \tag{23}$$

while the boundary condition becomes

$$\beta \frac{d\theta_2}{dt} = -aP_0 R_0 \frac{dx}{dt} + k_1 T_\infty \left(\frac{\partial \theta_1}{\partial r}\right)_{r=R_0} \quad , \tag{24}$$

where

$$\beta = \frac{M C_2 T_\infty}{4\pi R_0^2} \quad .$$

Similarly:

for $t \leqslant 0$: $$x = \dot{x} = \ddot{x} = \theta_1 = \theta_2 = 0 \tag{25}$$

while

$$\theta_1(\infty, t) = 0 \qquad \text{for all } t \quad . \tag{26}$$

2. Formal Solutions.

The problem is now reduced to the solution of the following equation

$$\nabla^2 \theta_1 = \frac{1}{D_1} \frac{\partial \theta_1}{\partial t} \quad , \tag{27}$$

with the boundary conditions:

$$\theta_1(\infty, t) = 0 \quad , \tag{28}$$

and

$$\beta \frac{d\theta_2}{dt} = -P_o R_o (1+W_r) \frac{dx}{dt} + k_1 T_\infty \left(\frac{\partial \theta_1}{\partial r}\right)_{r=R_o} , \quad (29)$$

where $\theta_2(t) = \theta_1(R_o, t)$. The motion of the bubble wall is determined by

$$\ddot{x} + \alpha(3+2W_r)x = \alpha\left[(1+W_r)\theta_2 - \epsilon\right]. \quad (30)$$

The initial conditions are

$$x = \dot{x} = \ddot{x} = \theta_1 = \theta_2 = 0 \quad , \quad \text{for} \quad t \leq 0 . \quad (31)$$

With spherical symmetry, we can rewrite equation (27) as

$$\frac{\partial^2}{\partial r^2}(r\theta_1) = \frac{1}{D_1}\frac{\partial}{\partial t}(r\theta_1) \quad . \quad (32)$$

Now let us define the Laplace transform of θ_1 as

$$w_1 \equiv \mathcal{L}\{\theta_1\} \equiv \int_0^\infty \theta_1 e^{-st}\, dt \quad ,$$

and, similarly, $w_2 \equiv \mathcal{L}\{\theta_2\}$, $z \equiv \mathcal{L}\{x\}$, $\mu \equiv \mathcal{L}\{\epsilon\}$.

With initial conditions (31), equations (27) - (30) now become

$$\frac{d^2}{dr^2}(r w_1) = \frac{s}{D_1}(r w_1) \quad , \quad (33)$$

$$w_1(\infty) = 0 \quad , \quad (34)$$

$$\beta s w_2 = -P_o R_o (1+W_r) s z + k_1 T_\infty \left(\frac{dw_1}{dr}\right)_{r=R_o} , \quad \text{with} \quad w_2 = (w_1)_{r=R_o} , \quad (35)$$

and

$$\left[s^2 + \alpha(3+2W_r)\right] z = \alpha\left[(1+W_r) w_2 - \mu\right] . \quad (36)$$

Solving equations (33) - (36) we obtain

$$w_1 = \frac{\alpha E S \mu}{P(\sqrt{s})} \frac{R_o}{r} e^{-(r-R_o)\sqrt{\frac{s}{D_1}}} , \tag{37}$$

where P is a polynomial defined by

$$P(u) \equiv (\beta u^2 + Au + B)(u^4 + \omega_1^2) + \alpha E(1+W_r)u^2 , \tag{38}$$

and where

$$w_1^2 = \alpha(3 + 2W_r) \quad ; \qquad A = \frac{k_1 T_\infty}{\sqrt{D_1}} \quad ;$$

$$B = \frac{k_1 T_\infty}{R_o} \quad ; \qquad E = P_o R_o (1 + W_r) \quad . \tag{39}$$

When $\epsilon = \epsilon_o e^{i\omega t}$, then $\mu = \mathcal{L}\{\epsilon\} = \frac{\epsilon_o}{s - i\omega}$.

Thus

$$w_1 = \frac{\alpha E \epsilon_o S}{(s - i\omega) P(\sqrt{s})} \frac{R_o}{r} e^{-(r-R_o)\sqrt{\frac{s}{D_1}}} . \tag{40}$$

From equations (36), (35) and (40), we have

$$Z = - \frac{\alpha \epsilon_o (\beta s + A s^{1/2} + B)}{(s - i\omega) \; P(s^{1/2})} . \tag{41}$$

The formal solutions are obtained by the inversion of (40) and (41). Let the roots of P(u) = 0 be $-a_1$, $-a_2$, $-a_3$, $-a_4$, $-a_5$ and $-a_6$. Also let $a_7 = \sqrt{\omega}\, e^{i\pi/4}$, $a_8 = -\sqrt{\omega}\, e^{i\pi/4} = \sqrt{\omega}\, e^{-i3\pi/4}$. Then by partial fractions we can, in principle, express (40) and (41) as:

$$w_1(r; s) = \frac{R_o}{r} e^{-(r-R_o)\sqrt{\frac{s}{D_1}}} \sum_{i=1}^{8} \frac{b_i}{s^{1/2} + a_i} , \tag{42}$$

and

$$Z(s) = \sum_{i=1}^{8} \frac{c_i}{s^{1/2} + a_i} . \tag{43}$$

Since $\sum_{i=1}^{8} b_i = 0$, $\sum_{i=1}^{8} c_i = 0$, we thus obtain [8]

$$\theta_1(r,t) = -\frac{R_0}{r}\sum_{i=1}^{8} a_i b_i \, e^{\frac{a_i(r-R_0)}{\sqrt{D_1}} + a_i^2 t} \, Erfc\left(\frac{r-R_0}{2\sqrt{D_1 t}} + a_i t^{\frac{1}{2}}\right), \quad (44)$$

consequently

$$\theta_2(t) = -\sum_{i=1}^{8} a_i b_i \, e^{a_i^2 t} \, Erfc\,(a_i t^{\frac{1}{2}}) \;, \quad (45)$$

and

$$x(t) = -\sum_{i=1}^{8} a_i c_i \, e^{a_i^2 t} \, Erfc\,(a_i t^{\frac{1}{2}}) \;. \quad (46)$$

Hence

$$\begin{aligned} p(t) &= (1+W_r)(\theta_2 - 3x) \\ &= (1+W_r)\sum_{i=1}^{8} a_i(3c_i - b_i)\, e^{a_i^2 t}\, Erfc\,(a_i t^{\frac{1}{2}}) \,. \end{aligned} \quad (47)$$

Here we have used the notation:

$$Erfc\,(x) = 2\pi^{-\frac{1}{2}} \int_x^{\infty} e^{-t^2} dt \;.$$

3. Asymptotic Behavior of the Solutions for Large t and Thermodynamic Relations

The above formal solutions have little practical significance since first of all it is hard to find the roots a_i except by numerical methods when the proper physical constants are furnished. Secondly, even if the roots are found, it is not a very simple matter to visualize the behavior of the error functions with complex arguments. However, the search for asymptotic expressions for the solutions is not only dictated by practical considerations, but also because it yields the steady-state solution which is actually of most physical significance.

As a preliminary step, we have to investigate the location of the roots of P(u) = 0. Since all the coefficients in the polynomial P(u) are real and positive, it is possible to show that for this special form of P(u), all the roots $-a_1$, $-a_2$, $-a_3$, $-a_4$, $-a_5$ and $-a_6$ lie in the sector:

$$|\arg(-a_i)| > \frac{\pi}{4} \qquad \text{or} \qquad |\arg(a_i)| < \frac{3\pi}{4} , \tag{48}$$

by principle of argument in theory of functions, (cf. Appendix 1). This result is essential for the boundedness of the asymptotic expressions.

To obtain the asymptotic expressions we may proceed by two different approaches. Either we may start from the formal solutions, using the equivalent relation between error functions and confluent hypergeometric functions, and then from the known asymptotic expansions of the confluent hypergeometric functions obtain the asymptotic expressions of our solutions (cf. Appendix 2) or we may work directly with the inversion integral of our transformed solutions, change the contour of integration by use of Cauchy's residue theorem, and then obtain the asymptotic expressions of our solutions by applying the method of steepest descent. (cf. Appendix 3).

Both approaches, after making use of the relation 48, arrive at the same result which is as $t \to \infty$:

$$\theta_1(r,t) = \frac{\alpha \epsilon_0 E \omega}{P(\sqrt{\omega}\, e^{i\pi/4})} \frac{R_0}{r} e^{-(r-R_0)\sqrt{\frac{i\omega}{D_1}}} e^{i(\omega t + \frac{\pi}{2})} + O(t^{-5/2}); \tag{49}$$

$$\theta_2(t) = \frac{\alpha \epsilon_0 E \omega}{P(\sqrt{\omega}\, e^{i\pi/4})} e^{i(\omega t + \pi/2)} + O(t^{-5/2}) ; \tag{50}$$

$$\chi(t) = -\frac{\alpha \epsilon_0 (\beta\omega + A\sqrt{\omega}\, e^{-i\pi/4} - iB)}{P(\sqrt{\omega}\, e^{i\pi/4})} e^{i(\omega t + \pi/2)} + O(t^{-3/2}) ; \tag{51}$$

and

$$p(t) = \frac{\alpha \epsilon_0 (1+W_r)\left[(E+3\beta)\omega + 3A\sqrt{\omega}\, e^{-i\pi/4} - 3iB\right]}{P(\sqrt{\omega}\, e^{i\pi/4})} e^{i(\omega t + \frac{\pi}{2})} + O(t^{-3/2}). \quad (52)$$

The forms of these steady-state solutions are typical for the problem of linear forced oscillation with damping. In this case the damping is indirectly introduced by the effect of heat conduction through the energy equation (29). In considering the thermodynamic behavior of the bubble, we notice that the energy equation (29):

$$K_1 T_\infty \left(\frac{\partial \theta_1}{\partial r}\right)_{r=R_0} = \beta \frac{d\theta_2}{dt} + P_0 R_0 (1+W_r) \frac{dx}{dt} \quad (53)$$

can be recognized as expressing the first law of thermodynamics, namely,

$$dQ = C_2\, dT_2 + P\, dV \quad , \quad (54)$$

and the relative magnitudes of $C_2 dT$ and dQ will indicate the tendency of the system to be isothermal or adiabatic.

Now, using the asymptotic expressions, we have

$$\beta \frac{d\theta_2}{dt} = \frac{g}{3} \frac{\rho_2 C_2}{\rho_1 C_1} \left[R_0^2 \frac{i\omega}{D_1}\right] e^{i(\omega t + \frac{\pi}{2})} \quad ; \quad (55)$$

while

$$K_1 T_\infty \left(\frac{\partial \theta_1}{\partial r}\right)_{r=R_0} = -g\left[1 + R_0 \sqrt{\frac{i\omega}{D_1}}\right] e^{i(\omega t + \frac{\pi}{2})} \quad ; \quad (56)$$

where

$$g = \frac{\alpha E \epsilon_0 K_1 T_\infty \omega}{R_0 P(\sqrt{\omega}\, e^{i\pi/4})} \quad ;$$

and we have also used the relation

$$\beta = \frac{1}{3} \rho_2 C_2 R_0 T_\infty \quad .$$

Comparing the expressions (55) and (56), we are led to the following conclusion regarding the average behavior of the thermodynamic process of the oscillating bubble, namely: The thermodynamic process may be said to be isothermal if $\frac{\rho_2 C_2}{\rho_1 C_1} R_0^2 \frac{\omega}{D_1} \ll 1$, and adiabatic if $\frac{\rho_2 C_2}{\rho_1 C_1} R_0 \sqrt{\frac{\omega}{D_1}} \gg 1$.

We have intentionally stated that the above conclusion can only be applied to "average" behavior, since nothing can be said about instantaneous behaviors for any finite non-vanishing frequencies, due to the phase differences between θ_2 and x. From the basic equation of state satisfied by any perfect gas, it seems certain that the phase difference between these state variables is inevitable for any general thermodynamic processes.

B. Bubble with Non-uniform Interior

1. Linearized Formulation

When we relax the requirement that conditions inside the bubble are uniform throughout the bubble, the situation is much more complex than the previous case although nothing is changed in the formulation of the problem in the surrounding liquid. Thus we have the same set of equations for the motion of the bubble wall and heat transfer in the liquid as before, namely,

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{1}{\rho_1}\left[P_2(R,t) - \frac{2\sigma}{R} - P_\infty(t) \right], \tag{1}$$

and

$$\nabla^2 T_1 = \frac{1}{D_1}\left(\frac{\partial T_1}{\partial t} + \vec{q}_1 \cdot \nabla T_1 \right) \quad \text{for } r \geq R . \tag{2}$$

The gas inside the bubble is assumed to be a perfect gas. Now due to the non-uniformity of the interior of the bubble, the equation of state can only be satisfied locally and we have therefore

$$P_2(r,t) = B\,\rho_2(r,t)\,T_2(r,t) \quad , \quad \text{for} \quad r \leq R \, , \qquad (3)$$

where B is a constant which is equal to $\frac{R}{m}$, where R is the universal gas constant and m is the molecular weight of the gas. The conservation of mass is expressed by the following equation of continuity:

$$\frac{\partial \rho_2}{\partial t} + \rho_2\, \nabla \cdot \vec{q}_2 + \vec{q}_2 \cdot \nabla \rho_2 = 0 \, , \qquad r \leq R \, . \qquad (4)$$

If we neglect viscous effects and assume that there is no external force field, then the equation of motion is

$$\rho_2 \left[\frac{\partial \vec{q}_2}{\partial t} + (\vec{q}_2 \cdot \nabla)\, \vec{q}_2 \right] = - \nabla P_2 \quad , \quad r \leq R \, , \qquad (5)$$

while the energy equation takes the following form:

$$\nabla^2 T_2 = \frac{1}{D_2} \left[\frac{\partial T_2}{\partial t} + \vec{q}_2 \cdot \nabla T_2 \right] + \frac{1}{K_2}\, P_2 (\nabla \cdot \vec{q}_2) , \quad r \leq R \, , \qquad (6)$$

where the last term represents the increment in internal energy due to compressibility; k_2 is the coefficient of heat conduction in the gas, and $D_2 = \frac{k_2}{\rho_2 c_2}$ is the coefficient of thermal diffusivity for the gas.

With $P_\infty(t)$ prescribed at a great distance from the bubble, the boundary conditions are such that all the physical quantities must remain finite at $r = 0$ and as $r \to \infty$; further, the pressure, particle velocity, temperature and flux of heat must be continuous at the bubble wall, $r = R$. The continuity of pressure is actually a restatement of Newton's Third law, while the continuity of fluid particle velocity is a consequence of the requirement that we allow no gap to arise in this continuum system. The continuity of temperature and heat flux follows from the arguments that the heat flux across the boundary is finite and that no heat can be accumulated on the boundary. These continuity conditions are expressed mathematically as follows:

$$P_1(R,t) = P_2(R,t) - \frac{2\sigma}{R} , \tag{7}$$

$$q_1(R,t) = \dot{R}(t) = q_2(R,t) , \tag{8}$$

$$T_1(R,t) = T_2(R,t) , \tag{9}$$

and

$$K_1 \frac{\partial T_1}{\partial r}(R,t) = K_2 \frac{\partial T_2}{\partial r}(R,t) . \tag{10}$$

The initial conditions are the same as before in that equilibrium is not disturbed until t = 0 at which time some perturbation in $P_\infty(t)$ starts the entire system in motion.

We shall linearize the problem in the same way as before. Let

$$R = R_0(1 + x) , \tag{11}$$

$$T_1 = T_\infty(1 + \theta_1) , \tag{12}$$

$$T_2 = T_\infty(1 + \theta_2), \tag{13}$$

$$P_\infty = P_0(1 + \epsilon), \tag{14}$$

$$P_2 = P_0(a + p), \tag{15}$$

$$\rho = \rho_0(1 + \eta), \tag{16}$$

where x, θ_1, θ_2, p, η are all small quantities in comparison with unity and are in the same order as the term in the perturbing pressure, ϵ.

Neglecting all the second order effects, we obtain the linearized equations by inserting the expressions (11) - (16) in equations (1) - (6). Thus, equation (1) becomes

$$\ddot{x} - \alpha W_r x = \alpha [p(R_o,t) - \epsilon] , \tag{17}$$

where as before $\alpha = \frac{P_o}{\rho_1 R_o^2}$ and $W_r = \frac{2\sigma}{P_o R_o}$.

Using the equilibrium condition $aP_o = P_2(R_o, t) = \frac{2\sigma}{R_o} + P_o$, we get

$$a = 1 + W_r . \tag{18}$$

Equation (2) now becomes

$$\frac{1}{D_1} \frac{\partial \theta_1}{\partial t} = \nabla^2 \theta_1 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial \theta_1}{\partial r} \right) . \tag{19}$$

Here we have used the spherical symmetry of the problem. With the equilibrium condition $P_o a = B \rho_o T_\infty$, the linearized local equation of state is now

$$\frac{p}{a} = \eta + \theta_2 . \tag{20}$$

Since for this problem with spherical symmetry

$$\vec{q}_2 = \vec{e}_r \, q_2(r,t) ,$$

the equation of continuity becomes, after linearization:

$$\frac{\partial \eta}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} (r^2 q_2) = 0 , \tag{21}$$

while the equation of motion becomes

$$\rho_0 \frac{\partial q_2}{\partial t} = -P_0 \frac{\partial p}{\partial r} \quad . \tag{22}$$

In a similar manner we obtain the linearized energy equation, namely,

$$\frac{1}{r^2} \frac{\partial}{\partial r}\left[r^2 \left(\frac{\partial \theta_2}{\partial r} - \frac{P_0 a}{K_2 T_\infty} q_2 \right) \right] = \frac{1}{D_2} \frac{\partial \theta_2}{\partial t} \quad . \tag{23}$$

For boundary conditions at the bubble wall, we have now

$$q_1(R_0,t) = R_0 \dot{x} = q_2(R_0,t) \ , \tag{24}$$

$$\theta_1(R_0,t) = \theta_2(R_0,t) \ , \tag{25}$$

and

$$K_1 \frac{\partial \theta_1}{\partial r}(R_0,t) = k_2 \frac{\partial \theta_2}{\partial r}(R_0,t) \ . \tag{26}$$

It should be remarked here that, in carrying out the linearization process, we have already treated the quantities q_1, q_2 as being of the same order of magnitude as $R_o x$. There may be some questions raised concerning the validity of this linearization process for high frequency oscillations. However, there should be no difficulty in this respect so long as we are free to set the amplitude of perturbation as small as we please. As the frequency gets higher and higher, deviations may appear for other reasons, such as departures from instantaneous validity of the equation of state, or a lack of constancy of the physical coefficients.

2. The Asymptotic Solutions

As before, we define the Laplace transform of u(t) as

$\mathcal{L}\{u\} = \int_0^\infty e^{-st} u \, dt$, and let

$$\begin{aligned} &\mathcal{L}\{x\} = z \ , && \mathcal{L}\{\theta_1\} = w_1 \ , && \mathcal{L}\{\theta_2\} = w_2 \ , \\ &\mathcal{L}\{p\} = f \ , && \mathcal{L}\{\eta\} = g \ , && \mathcal{L}\{q_2\} = v \ , \end{aligned} \tag{27}$$

and $\mathcal{L}\{\epsilon\} = \mu$.

Also we note that $\mathcal{L}\{R_o \dot{x}\} = R_o s Z = v(R_o) = \mathcal{L}\{q_2(R_o, t)\}$.

Thus using the initial conditions, we have

$$\chi(r,0) = \dot{\chi}(r,0) = \theta_1(r,0) = \theta_2(r,0) = q_2(r,0) = \eta(r,0) = p(r,0) = 0 \ . \qquad (28)$$

We obtain the following transformed equations:

$$(s^2 - \alpha W_r) Z = \alpha [f(R_o) - \mu] \ ; \qquad (29)$$

$$\frac{d^2}{dr^2}(r w_1) = \frac{s}{D_1}(r w_1) \ , \qquad \text{for} \quad r \geq R_o \ ; \qquad (30)$$

$$f = a(g + w_2) \ , \qquad \text{for} \quad r \leq R_o \ ; \qquad (31)$$

$$s g + \frac{1}{r^2}\frac{d}{dr}(r^2 v) = 0 \ , \qquad \text{for} \quad r \leq R_o \ ; \qquad (32)$$

$$\rho_o s v = - P_o \frac{df}{dr} \ ; \qquad (33)$$

and

$$\frac{1}{r}\frac{d^2}{dr^2}(r w_2) - \frac{P_o a}{K_2 T_\infty}\frac{1}{r^2}\frac{d}{dr}(r^2 v) = \frac{s}{D_2} w_2 \ . \qquad (34)$$

From equations (33) and (32) we have

$$g = \frac{P_o}{\rho_o s^2}\frac{1}{r}\frac{d^2}{dr^2}(r f) \ . \qquad (35)$$

Then from (31), we obtain

$$w_2 = \frac{f}{a} - g = \frac{f}{a} - \frac{P_o}{\rho_o s^2}\frac{1}{r}\frac{d^2}{dr^2}(r f) \ . \qquad (36)$$

Application of equations (33) and (36) gives for (34)

$$\frac{d^4}{dr^4}(rf) - \left[\frac{\rho_o s^2}{P_o a} + \left(\frac{P_o a}{K_2 T_\infty} + \frac{1}{D_2}\right)s\right]\frac{d^2}{dr^2}(rf) + \frac{\rho_o s^3}{P_o a D_2}(rf) = 0, \quad (37)$$

or

$$\left(\frac{d^2}{dr^2} - \alpha_1^2\right)\left(\frac{d^2}{dr^2} - \alpha_2^2\right)(rf) = 0 , \quad (38)$$

where

$$\alpha_{1,2}^2(s) = \frac{1}{2}\left\{\left(\frac{P_o a}{K_2 T_\infty} + \frac{1}{D_2}\right)s + \frac{\rho_o s^2}{P_o a} \pm \sqrt{\left[\left(\frac{P_o a}{K_2 T_\infty} + \frac{1}{D_2}\right)s + \frac{\rho_o s^2}{P_o a}\right]^2 - \frac{4\rho_o s^3}{P_o a D_2}}\right\}. \quad (39)$$

Thus the general solution of f can be written as

$$f = \frac{1}{r}\left[A_1 \cosh\alpha_1 r + A_2 \sinh\alpha_1 r + A_3 \cosh\alpha_2 r + A_4 \sinh\alpha_2 r\right]. \quad (40)$$

At this stage, it is obvious that it is immaterial whether we take the positive or the negative part of the square root in the expressions of α_1 and α_2. What is essential is that we have to adopt a consistent definition for further calculations.

From equation (35) we then obtain

$$g = \frac{P_o}{\rho_o s^2}\frac{1}{r}\left[\alpha_1^2 A_1 \cosh\alpha_1 r + \alpha_1^2 A_2 \sinh\alpha_1 r + \alpha_2^2 A_3 \cosh\alpha_2 r + \alpha_2^2 A_4 \sinh\alpha_2 r\right]. \quad (41)$$

As f is finite at $r = 0$, $A_1 = -A_3$; as g is also finite at $r = 0$, and $\alpha_1 \neq \alpha_2$ in general, we find that $A_1 = A_3 = 0$. Thus

$$f = \frac{1}{r}\left[A_2 \sinh\alpha_1 r + A_4 \sinh\alpha_2 r\right]; \quad (42)$$

$$g = \frac{P_o}{\rho_o s^2}\frac{1}{r}\left[A_2 \alpha_1^2 \sinh\alpha_1 r + A_4 \alpha_2^2 \sinh\alpha_2 r\right]. \quad (43)$$

Now from equation (33), we have

$$v = -\frac{P_o}{\rho_o s}\left[A_2\left(\frac{\alpha_1 \cosh\alpha_1 r}{r} - \frac{\sinh\alpha_1 r}{r^2}\right) + A_4\left(\frac{\alpha_2 \cosh\alpha_2 r}{r} - \frac{\sinh\alpha_2 r}{r^2}\right)\right]. \quad (44)$$

It is easily verified that as $r \to 0$, $v \to 0$, which is a satisfactory result. Also from (36) we have

$$w_2 = \frac{1}{r}\left[A_2\left(\frac{1}{a} - \frac{P_o \alpha_1^2}{\rho_o s^2}\right)\sinh\alpha_1 r + A_4\left(\frac{1}{a} - \frac{P_o \alpha_2^2}{\rho_o s^2}\right)\sinh\alpha_2 r\right] . \quad (45)$$

For the problem in the liquid, the solution of equation (27), after requiring that w_1 be finite as $r \to \infty$, is

$$w_1 = \frac{A_o}{r} e^{-\sqrt{\frac{s}{D_1}}\, r} . \quad (46)$$

The constants A_o, A_2 and A_4 which are all functions of the parameter s can be determined by making use of the boundary conditions at the bubble wall r = R , namely,

$$w_1(R_o) = w_2(R_o) , \quad (47)$$

$$K_1 \frac{dw_1}{dr}(R_o) = K_2 \frac{dw_2}{dr}(R_o) \quad (48)$$

and

$$v(R_o) = s Z R_o , \quad (49)$$

together with the equation of motion of the bubble wall (equation 29). Thus:

$$A_2 = \frac{1}{\Delta(s)} \frac{\alpha \mu s R_o}{s^2 - \alpha W_r}\left(\frac{1}{a} - \frac{P_o \alpha_2^2}{\rho_o s^2}\right) \times$$

$$\left[K_2\left(\alpha_2 \cosh\alpha_2 R_o - \frac{\sinh\alpha_2 R_o}{R_o}\right) + K_1\left(\sqrt{\frac{s}{D_1}} + \frac{1}{R_o}\right)\sinh\alpha_2 R_o\right] ; \quad (50)$$

and

$$A_4 = -\frac{1}{\Delta(s)} \frac{\alpha\mu S R_o}{s^2 - \alpha W_r}\left(\frac{1}{a} - \frac{P_o\alpha_1^2}{\rho_o s^2}\right) \times$$
$$\left[K_2\left(\alpha_1 \cosh\alpha_1 R_0 - \frac{\sinh\alpha_1 R_0}{R_0}\right) + K_1\left(\sqrt{\frac{s}{D_1}} + \frac{1}{R_0}\right)\sinh\alpha_1 R_0\right], \tag{51}$$

where

$$\Delta(s) = \Delta_1(\alpha_1,\alpha_2) - \Delta_1(\alpha_2,\alpha_1) \quad ,$$

and

$$\Delta_1(\alpha_1,\alpha_2) = \left[\frac{P_o\alpha_1}{\rho_o R_o s}\cosh\alpha_1 R_o - \sinh\alpha_1 R_o\left(\frac{P_o}{\rho_o s R_o^2} - \frac{\alpha s}{s^2 - \alpha W_r}\right)\right]\left[\frac{1}{a} - \frac{P_o\alpha_2^2}{\rho_o s^2}\right]$$
$$\times\left[K_2\left(\alpha_2\cosh\alpha_2 R_0 - \frac{\sinh\alpha_2 R_0}{R_0}\right) + K_1\left(\sqrt{\frac{s}{D_1}} + \frac{1}{R_0}\right)\sinh\alpha_2 R_0\right] . \tag{52}$$

Having found f, w_1, w_2, z, v, g, we may in principle obtain p, θ_1, θ_2, q_2, η by inverse transform. However, we shall be content with the asymptotic expressions for large t.

Let us try to find the asymptotic expression of p. Now

$$p(r,t) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} f(r;s)\, e^{st} ds \quad ,$$
$$= \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} \frac{1}{r}\left[A_2(s)\sinh\alpha_1 r + A_4(s)\sinh\alpha_2 r\right] e^{st} ds \,, \tag{53}$$

where the path of integration is to the right of all the singularities of the integrand.

From the expressions of A_2 and A_4 in equations (50) and (51), we see that the expression in the square bracket in (53) is of the form

$$\frac{1}{\Delta_1(\alpha_1,\alpha_2) - \Delta_1(\alpha_2,\alpha_1)}\left[\psi(\alpha_1,\alpha_2) - \psi(\alpha_2,\alpha_1)\right]$$

which is equivalent to the form

$$\chi(\alpha_1,\alpha_2) + \chi(\alpha_2,\alpha_1) \quad .$$

In other words the integrand is not to be changed with the interchange of its dependence on α_1 and α_2.

Now, from the expressions for α_1, α_2, A_2 and A_4, it appears that there are three branch points in the integrand of equation (53), namely $s = 0$, and the two roots $s_{1,2}$ given by

$$\left[\left(\frac{P_o}{K_2 T_\infty} + \frac{1}{D_2} \right)^2 + \frac{\rho_o s}{P_o a} \right]^2 - \frac{4 \rho_o s}{P_o a D_2} = 0 \quad ,$$

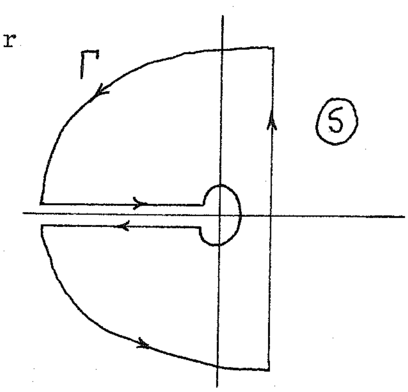
as seen from the expression for $\alpha_{1,2}^2$ in equation (39).

Now, due to the symmetric form of the integrand with respect to the interchange of α_1 and α_2, and also the interchange of values of α_1 and α_2 when we complete a circle surrounding any of the branch points $\alpha_{1,2}$, it is not difficult to see that $s_{1,2}$ are just apparent branch points. Now let us specify $\epsilon = \epsilon_o e^{i\omega t}$. (55)

Then

$$\mu = \mathcal{L}\{\epsilon\} = \frac{\epsilon_o}{s - i\omega} \quad . \tag{56}$$

It is obvious that $i\omega$ is a pole inside the contour Γ shown in the figure. It is not easy to see whether there are any other poles of the integrand inside Γ. Nevertheless, from the physical argument that for the steady state case the only mode of oscillation that can possibly persist is one with the forcing frequency, we may expect that the only other poles, if there are any, must lie in the left half plane so as to yield exponentially decaying terms.



5

As the branch integral from the branch point $s = 0$ is of the order $(\frac{1}{t})$ as $t \to \infty$, by Cauchy's Theorem, we obtain

$$p(r,t) = \frac{1}{r}\left[B_2(i\omega)\sinh\alpha_1(i\omega)r + B_4(i\omega)\sinh\alpha_2(i\omega)r\right]e^{i\omega t} + O\left(\frac{1}{t}\right), \qquad \text{as } t \to \infty, \tag{57}$$

where

$$B_2(i\omega) = -\frac{R_0}{\Delta(i\omega)}\,\frac{\alpha\epsilon_0 i\omega}{\omega^2+\alpha W_r}\left(\frac{1}{a} + \frac{P_0\alpha_2^2(i\omega)}{\rho_0\omega^2}\right) \times \left\{K_2\left[\alpha_2(i\omega)\cosh\alpha_2(i\omega)R_0 - \frac{\sinh\alpha_2(i\omega)R_0}{R_0}\right] + k_1\left(\sqrt{\frac{i\omega}{D_1}} + \frac{1}{R_0}\right)\sinh\alpha_2(i\omega)R_0\right\}; \tag{58}$$

and

$$B_4(i\omega) = \frac{R_0}{\Delta(i\omega)}\,\frac{\alpha\epsilon_0 i\omega}{\omega^2+\alpha W_r}\left(\frac{1}{a} + \frac{P_0\alpha_1^2(i\omega)}{\rho_0\omega^2}\right) \times \left\{K_2\left[\alpha_1(i\omega)\cosh\alpha_1(i\omega)R_0 - \frac{\sinh\alpha_1(i\omega)R_0}{R_0}\right] + K_1\left(\sqrt{\frac{i\omega}{D_1}} + \frac{1}{R_0}\right)\sinh\alpha_1(i\omega)R_0\right\}; \tag{59}$$

and $\Delta(i\omega)$, $\alpha_1(i\omega)$ and $\alpha_2(i\omega)$ are obtained from equations (52) and (39) upon replacing s by $i\omega$. Similarly we have for large t

$$\Theta_2(r,t) \sim \frac{1}{r}\left\{B_2(i\omega)\left[\frac{1}{a} + \frac{P_0\alpha_1^2(i\omega)}{\rho_0\omega^2}\right]\sinh\alpha_1(i\omega)r + B_4(i\omega)\left[\frac{1}{a} + \frac{P_0\alpha_2^2(i\omega)}{\rho_0\omega^2}\right]\sinh\alpha_2(i\omega)r\right\}e^{i\omega t}, \tag{60}$$

and

$$x(t) \sim \frac{P_0}{\rho_0 R_0\omega^2}\left\{B_2(i\omega)\left[\frac{\alpha_1(i\omega)\cosh\alpha_1(i\omega)R_0}{R_0} - \frac{\sinh\alpha_1(i\omega)R_0}{R_0^2}\right] + B_4(i\omega)\left[\frac{\alpha_2(i\omega)\cosh\alpha_2(i\omega)R_0}{R_0} - \frac{\sinh\alpha_2(i\omega)R_0}{R_0^2}\right]\right\}e^{i\omega t}. \tag{61}$$

Thus even for these asymptotic expressions, the results are of such complexity that very little can be seen from them. Even numerical expressions would require laborious computations.

In consideration of thermodynamic behavior, we see the equivalent form of the first law of thermodynamics

$$dQ - P\,dV = C_v\,dT$$

is

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left[r^2\left(\frac{\partial \theta_2}{\partial r} - \frac{P_0 a}{K_2 T_\infty} q_2\right)\right] = \frac{1}{D_2}\frac{\partial \theta_2}{\partial t} \; .$$

In order to be able to compare the results with what is concluded from the previous problem, the average behavior rather than the instantaneous, local behavior is to be considered. Thus the above relation is to be integrated over the entire volume occupied by the gas. And for the limiting cases $\omega \to 0$ and $\omega \to \infty$, the order of magnitude of dQ and C_vdT can be compared. The results obtained after lengthy algebraic manipulation can be stated briefly as follows:

$$\frac{C_v\,dT}{dQ} \sim O(\omega) \qquad \text{as} \qquad \omega \longrightarrow 0 \; ,$$

and

$$\frac{C_v\,dT}{dQ} \sim \sqrt{\frac{K_2 \rho_2 C_2}{K_1 \rho_1 C_1}} \qquad \text{as} \qquad \omega \longrightarrow \infty \; .$$

Thus for very low frequency it appears that the relation is isothermal in agreement with the result of the previous analysis. For very high frequency, no adiabatic limit is indicated. Adiabatic behavior would have to be found for $\frac{K_2}{K_1} \to \infty$, which is actually implied in the assumption of uniformity of the interior of the gas bubble. However, for actual physical situations k_2 is of the same order as k_1, or even smaller. (For instance, for air $k_2 = 2.5 \times 10^3$ erg/cm sec°C;

while for water $k_1 = 6 \times 10^4$ erg/cm. sec. $^{\circ}$C). Hence $\sqrt{\frac{K_2 \rho_2 c_2}{K_1 \rho_1 c_1}}$ is usually a small number. This result tends to indicate that even at very high frequency isothermality rather than adiabaticity is to be expected for the average thermodynamic behavior of a gas bubble in an oscillating pressure field.

II. RECTIFICATION OF MASS

A. Formulation of the Problem

When a gas bubble is subject to an oscillating pressure field in a liquid saturated with the dissolved gas, it is to be expected that there is net flow of gas into the bubble over any complete cycle of oscillation. An intuitive physical explanation is as follows: When the gas bubble is compressed, the increase of pressure at bubble wall will cause the rise of gas concentration above the equilibrium value, and results in the outflow of gas. On the other hand, by similar arguments, gas will flow into the bubble during the expansion half-cycle of the gas bubble. Owing to the difference in surface areas of the bubble walls between these two half-cycles, there will be net flow gas into the bubble over a complete cycle. Obvious as it appears from this intuitive argument, the quantitative analysis is not quite simple. First of all, it is closely associated with the complicated dynamic problem which determines the motion of bubble wall in terms of the applied pressure field and other relevant parameters. Secondly, even after knowing the motion of the bubble wall, we are still left with a non-linear diffusion problem which involves conditions specified on some moving boundary. In general, the dynamic problem and the diffusion problem are unseparably coupled; and this leads to even greater complexity. The purpose of the present analysis is mainly to study the mechanism involved in the above mentioned phenomenon of one-sided diffusion, which is often termed as "rectification of mass". Therefore some simplifying assumptions are to be made on other aspects of the entire problem, and a linearization procedure is to be applied to the study of the diffusion problem itself.

Actually the dynamic problem can be by-passed if we prescribe the oscillating pressure inside the gas bubble rather than with the pressure at infinity. This procedure indeed is not so arbitrary as might appear, for when the steady state is attained, except for a definite phase difference and some modification of amplitude, the pressure inside the bubble behaves essentially in the same manner as the applied pressure field so far as the linearized theory is concerned.

We shall denote the internal pressure of the gas bubble, which is assumed to be uniform throughout the bubble, as P(t). P(t) is prescribed in the following manner:

$$P(t) = P_0 \left(1 + \epsilon \sin \omega t \right) . \tag{1}$$

We assume ϵ to be much smaller than unity in order to be able to carry out the linearization procedure.

Now we again assume that the gas inside the bubble behaves isothermally while it undergoes expansion and compression. Then this will lead to the following result:

$$R(t) = R_0 \left(1 + \delta \sin \omega t \right) + O(\delta^2) , \tag{2}$$

where $3\delta = -\epsilon$, R is radius of the bubble, and R_o is the equilibrium radius corresponding to P_o.

For a more general thermodynamic behavior, there will be a phase difference between $P - P_o$ and $R - R_o$. This is ignored since no essentially new feature can be learned from those complications.

Now the amount of gas flowing into the bubble during a time interval Δ T is just

$$\int_{T_0}^{T_0+\Delta T} dt \int_S D\,\nabla C \cdot \vec{ds} \quad ,$$

where C is the gas concentration in the liquid, D is the coefficient of diffusion, and the integration is taken over the surface S of the entire bubble wall.

When the problem possesses spherical symmetry, as in the present case, the above expression can be simplified to

$$\int_{T_0}^{T_0+\Delta T} dt \cdot 4\pi D R^2 \left(\frac{\partial C}{\partial r}\right)_{r=R} \quad .$$

Since C is determined by the diffusion equation, the problem is equivalent to finding C from the equation

$$\frac{\partial C}{\partial t} + \vec{q} \cdot \nabla C = D \nabla^2 C \ , \qquad (3)$$

with suitable initial and boundary conditions. In the above expression $\vec{q}$ is the particle velocity of the fluid. When the flow field in the liquid is irrotational, it is known that

$$\vec{q} = \frac{R^2 \dot{R}}{r^2} \vec{e}_r \quad . \qquad (4)$$

The boundary conditions will be as follows: $C = C_\infty$, which is a constant as $r \rightarrow \infty$; while at $r = R$, $C = aP(R)$ in accordance with Henry's Law. As $C = C_\infty$ everywhere when there is no disturbance, we have $aP_0 = C_\infty$. We may now formulate the boundary conditions for the solution of Eq. (1) as follows:

$$C = C_\infty \qquad \text{as} \qquad r \longrightarrow \infty \ , \qquad (5)$$

$$C = C_\infty (1 + \epsilon \sin \omega t) , \qquad \text{at} \qquad r = R . \qquad (6)$$

The initial condition is set as follows:

$$C = C_\infty \,, \qquad \text{for} \quad t \leq 0, \quad \text{and all} \quad r \,. \tag{7}$$

As we are only interested in the steady state solution, only the asymptotic solution for large t is sought.

B. Solution of the Problem

Let $\theta(r, t) = C(r, t) - C_\infty$. Then the problem is reduced to the solution of the equation

$$L(\theta) = \frac{\partial^2(r\theta)}{\partial r^2} - \frac{1}{D}\frac{\partial(r\theta)}{\partial t} = \frac{R^2\dot{R}}{rD}\frac{\partial\theta}{\partial r} \equiv g(r,t) = g(\theta) \,, \tag{8}$$

with the conditions

$$\theta(r, 0) = \theta(\infty, t) = 0 \,, \tag{9}$$

and

$$\begin{aligned} \theta(R(t), t) &= C_\infty \epsilon \sin \omega t \,, \\ &= -3\delta C_\infty \sin \omega t \,. \end{aligned} \tag{10}$$

A scheme of successive approximations in powers of the small parameter ϵ can be developed, and we shall evaluate the leading term that contributes to the rectification of mass. This leading term is of the order of ϵ^2. The successive approximations, in general, will be carried out in two steps. First we solve the following problem, namely,

$$L(\theta_1) = 0 \,, \tag{11}$$

with

$$\theta_1(r, 0) = \theta_1(\infty, t) = 0 \,, \tag{12}$$

and

$$\theta_1(R(t), t) = -3\delta C_\infty \sin \omega t \,. \tag{13}$$

Since $$R(t) = R_0 (1 + \delta \sin \omega t) + O(\delta^2) ,$$

successive approximations have to be carried out to the desired order of accuracy due to the boundary condition (13).

Using this solution as the first approximation in $g(\theta)$, we then carry out the next step of successive approximations by solving the equation

$$L(\theta_2) = g(\theta_1 + \theta_2) , \tag{14}$$

with

$$\theta_2(r, 0) = \theta_2(\infty, t) = 0 , \tag{15}$$

and

$$\theta_2(R(t), t) = 0 . \tag{16}$$

After both θ_1 and θ_2 have been obtained in this way, then $\theta = \theta_1 + \theta_2$ will be the final solution to the desired order of accuracy. It may be pointed out that to obtain solutions to the orders higher than ϵ^2, the expression for R(t) should also be corrected to the appropriate degree of accuracy. It is easy to see that this scheme, although workable in principle to obtain higher order solutions, becomes quite complicated.

The asymptotic solution of Eqs. (11), (12) and (13) for large t, to the order of ϵ^2, is (cf. Appendix 4)

$$\begin{aligned} \theta_1(r,t) = -\frac{3R_0 C_\infty \delta}{r} \Big\{ & e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \sin\left[\omega t - (r-R_0)\sqrt{\frac{\omega}{2D}}\right] \\ & + \frac{\delta}{2}\Big[\left(1+R_0\sqrt{\frac{\omega}{2D}}\right) \mathrm{Erfc}\left(\frac{r-R_0}{\sqrt{4Dt}}\right) - \left(1+R_0\sqrt{\frac{\omega}{2D}}\right) e^{-(r-R_0)\sqrt{\frac{\omega}{D}}} \cos\left(2\omega t - (r-R_0)\sqrt{\frac{\omega}{D}}\right) \\ & + R_0\sqrt{\frac{\omega}{2D}}\, e^{-(r-R_0)\sqrt{\frac{\omega}{D}}} \sin\left(2\omega t - (r-R_0)\sqrt{\frac{\omega}{D}}\right)\Big]\Big\} + O(t^{-3/2}) . \end{aligned} \tag{17}$$

For the purpose of studying the effect of rectification of mass, it is not necessary to evaluate θ_2 explicitly. The relevant quantity is just $\left(\frac{\partial\theta_2}{\partial r}\right)_{r=R}$, since the amount of gas flowing into the bubble during any time interval ΔT is $\int_{T_0}^{T_0+\Delta T} dt\, 4\pi D R^2 \left(\frac{\partial\theta}{\partial r}\right)_{r=R}$.

Thus using (17), we obtain, for large t, up to the order of ϵ^2, the following result (cf. Appendix 5):

$$\left(\frac{\partial\theta_2}{\partial r}\right)_{r=R} = -\frac{1}{R_0}\int_{R_0}^{\infty} g_1(r)\,dr + O\left(\frac{1}{t^{1/2}}\right) + S\,, \tag{18}$$

where S is some sinusoidal terms which would not contribute to the net flow of gas into the bubble over a complete cycle up to the order of ϵ^2, and

$$g_1(r) = 3R_0^4 C_\infty\left(\frac{\omega}{2D}\right)\delta^2 e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}}\left\{-\left(\frac{1}{r^3}+\frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\right)\sin\left[(r-R_0)\sqrt{\frac{\omega}{2D}}\right]\right.$$
$$\left. + \frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\cos\left[(r-R_0)\sqrt{\frac{\omega}{2D}}\right]\right\}. \tag{19}$$

The integral in Eq. (18) may not be easy to evaluate. However, for the case that $R_0\sqrt{\frac{\omega}{2D}} \gg 1$, i.e. when the diffusion length $\sqrt{\frac{D}{\omega}}$ is small in comparison with the radius of the bubble, we can obtain an asymptotic expression. In this way we find (cf. Appendices 5, 6)

$$\left(\frac{\partial\theta_2}{\partial r}\right)_{r=R} = C_\infty\delta^2\left[\left(-\frac{3}{2}\sqrt{\frac{\omega}{2D}}+\frac{9}{2R_0}\right) + O\left(\frac{1}{R_0^2\sqrt{\frac{\omega}{2D}}}\right) + O\left(\frac{1}{t^{1/2}}\right) + S\right] + O(\delta^3). \tag{20}$$

Now from Eq. (17), we obtain (cf. Appendix 4)

$$\left(\frac{\partial\theta_1}{\partial r}\right)_{r=R} = 3R_0C_\infty\delta\left[\left(\frac{1}{R_0^2}+\frac{1}{R_0}\sqrt{\frac{\omega}{2D}}\right)\sin\omega t + \frac{1}{R_0}\sqrt{\frac{\omega}{2D}}\cos\omega t\right]$$
$$+ 3R_0C_\infty\delta^2\left[\frac{1}{2R_0^2}\left(1+R_0\sqrt{\frac{\omega}{2D}}\right) - \left(\frac{2}{R_0^2}+\frac{2}{R_0}\sqrt{\frac{\omega}{2D}}\right)\sin^2\omega t\right.$$
$$\left. + O\left(\frac{1}{t^{1/2}}\right) + S'\right] + O(\delta^3)\,, \tag{21}$$

when S' also contains only sinusoidal terms that will not contribute to rectification up to the order of ϵ^2.

Now the rate of flow of gas into the bubble is

$$J = 4\pi D R^2 \left(\frac{\partial\theta}{\partial r}\right)_{r=R} = 4\pi D R_0^2 \left(1+2\delta\sin\omega t + O(\delta^2)\right)\left(\frac{\partial(\theta_1+\theta_2)}{\partial r}\right)_{r=R} .$$

Using (20) and (21), we obtain, up to order of ϵ^2,

$$J = 24\pi D C_\infty R_0 \delta^2 \left[1 + O\left(\frac{1}{R_0\sqrt{\frac{\omega}{2D}}}\right) + O\left(\frac{1}{t^{1/2}}\right) \right] + S + O(\delta^3) . \quad (22)$$

Thus, taking only the leading term, we have the average rate of flow of gas into the bubble

$$\bar{J} = 24\pi D C_\infty R_0 \delta^2 . \quad (23)$$

Or, since

$$\delta = -\frac{\epsilon}{3} = -\frac{1}{3}\,\frac{P_{max.} - P_0}{P_0} = -\frac{1}{3}\,\frac{\Delta P}{P_0} ,$$

we have alternatively,

$$\bar{J} = \frac{8}{3}\pi D C_\infty R_0 \left(\frac{\Delta P}{P_0}\right)^2 . \quad (24)$$

When $\left(\frac{\Delta P}{P_0}\right)$ is sufficiently small, the effect on the growth of the bubble due to rectification comes from this leading term only. The rate of growth due to the effect of rectification is indeed slow.

The mass inside the gas bubble is

$$m = \frac{4}{3}\pi \rho_g R^3 . \quad (25)$$

The density of gas ρ_g, remains practically unchanged during this slow growth, so that

$$\frac{dm}{dt} = 4\pi \rho_g R^2 \frac{dR}{dt} . \quad (26)$$

On the other hand, the rate of increase of mass in the bubble due to rectification is essentially

$$\frac{dm}{dt} = \bar{J} = \frac{8}{3}\pi D C_\infty \epsilon^2 R \;. \tag{27}$$

Equating (26) and (27), we obtain

$$\frac{dR}{dt} = \frac{2}{3}\frac{DC_\infty}{\rho_g}\epsilon^2 \frac{1}{R} \;. \tag{28}$$

Therefore

$$R^2 = R_0^2 + \frac{4}{3}\frac{DC_\infty}{\rho_g}\epsilon^2 t \;, \tag{29}$$

if we set $R = R_o$ when $t = 0$. Or

$$R = \sqrt{\frac{DC_\infty}{3\rho_g}}\, 2\epsilon\,(t + T_0)^{1/2} \;, \tag{30}$$

when

$$T_0 = \frac{3\rho_g}{DC_\infty}\left(\frac{R_0}{\epsilon}\right)^2 \;.$$

Summing up the above results, we conclude that within the range of small oscillations, the greater the relative amplitude of oscillation, the greater the parameter $\left(\frac{C_\infty}{\rho_g}\right)$, and the greater the coefficient of diffusivity of gas in the liquid D, the faster will be the growth of bubble; and also the growth behaves like $R \sim t^{1/2}$ for large enough t.

Let us denote T as the time required for the bubble to double its size. From Eq. (29), we obtain

$$T = \frac{9R_0^2\rho_g}{4C_\infty D\epsilon^2} \;. \tag{31}$$

Take the case of air in water, at 20°C, and 1 atm of pressure, we obtain the results as shown in Table I.

TABLE I

TIME REQUIRED TO DOUBLE THE SIZE OF GAS BUBBLES BY THE EFFECT OF MASS RECTIFICATION

R_o (cm)	$\frac{P_{max} - P_o}{P_o}$	T (sec)
10^{-1}	10^{-1}	6.7×10^{6}
10^{-1}	10^{-2}	6.7×10^{8}
10^{-3}	10^{-1}	6.7×10^{2}
10^{-3}	10^{-2}	6.7×10^{4}

III. THE STABILITY OF A SPHERICAL GAS BUBBLE IN OSCILLATING PRESSURE FIELDS

It is known that a spherical gas bubble in liquid under oscillating pressure fields would grow indefinitely due to the effect of rectification of mass (cf. Part II). The fact that we do not observe large bubbles under these conditions might be explained by the consideration of stability of the spherical shape of the bubble under oscillating pressure fields.

The stability of the flow with spherical symmetry has been discussed by Plesset [9] and applied to the cases of growth and collapse of vapor bubble by Plesset and Mitchell [10]. The following investigation will make use of the basic relations derived in the papers just mentioned.

So far as the effect of rectification of mass is concerned, the growth of gas bubble is indeed very slow. Therefore, the stability considerations may just be applied to the case where the mean radius of the gas bubble remains essentially constant all the time.

Let the bubble wall be distorted from the surface of a sphere of radius R to a surface with radius vector r_s; then one may write

$$r_s = R + \sum a_n Y_n \quad , \tag{1}$$

where Y_n is a spherical harmonic of degree n and the a_n's are functions of time to be determined. The growth or decay of $a_n(t)$ from a small initial value would determine whether the spherical shape is stable or not. When a linearized perturbation procedure under the assumption that

$$|a_n(t)| << R(t)$$

is applied to the case of two immiscible, incompressible, inviscid fluids separated by a spherical interface, it is found [8] that the a_n's are independent of each other, and they satisfy the following differential equation:

$$\frac{d^2a_n}{dt^2} + \frac{3}{R}\frac{dR}{dt}\frac{da_n}{dt} - Aa_n = 0 . \tag{2}$$

The function A of Eq. (2) is given by

$$A = \frac{[n(n-1)\rho_2 - (n+1)(n+2)\rho_1]\frac{d^2R}{dt^2} - (n-1)n(n+1)(n+2)\frac{\sigma}{R^2}}{[n\rho_2 + (n+1)\rho_1]R} , \tag{3}$$

where ρ_1 is the density of the fluid inside the sphere, ρ_2 is the density of the fluid occupying the region exterior to the sphere, and σ is the surface tension constant. Although the stability of spherical shape from small distortion may be inferred from the decay of $a_n(t)$ with time, strictly speaking because of the linearization process the instability as derived from the growth of $a_n(t)$ with time is rather a reasonable conjecture than a necessary consequence.

For the case of gas bubble, the gas density ρ_1 may be neglected in comparison with the liquid density ρ_2. Then A becomes

$$A = \frac{(n-1)}{R}\frac{d^2R}{dt^2} - (n-1)(n+1)(n+2)\frac{\sigma}{\rho R^3} , \tag{4}$$

where $\rho = \rho_2$ is the liquid density.

Let

$$b_n = R^{3/2} a_n , \tag{5}$$

Then equation (2) is transformed into

$$\frac{d^2 b_n}{dt^2} + G(t) b_n = 0 \,, \tag{6}$$

where

$$G(t) = (n-1)(n+1)(n+2)\frac{\sigma}{\rho R^3} - \frac{3}{4}\frac{1}{R^2}\left(\frac{dR}{dt}\right)^2 - \frac{(n+\frac{1}{2})}{R}\frac{d^2R}{dt^2} \,. \tag{7}$$

Now the radius of the undisturbed bubble R is governed by the equation

$$R\frac{d^2R}{dt^2} + \frac{3}{2}\left(\frac{dR}{dt}\right)^2 = \frac{1}{\rho}\left(P_i - P_\infty - \frac{2\sigma}{R}\right) \,, \tag{8}$$

where P_i is the pressure inside the bubble, while P_∞ is the pressure at distance from the bubble which in the present case may be represented by

$$P_\infty = P_0 (1 + \epsilon \sin \omega t) \,. \tag{9}$$

When ϵ is small in comparison with unity, it may be shown from a linearized theory that R can be represented in the form

$$R = R_0 [1 + \delta \sin(\omega t + \phi)] \,, \tag{10}$$

where δ is of the same order of magnitude as ϵ and ϕ denotes a constant phase shift, which for convenience may be put equal to zero. Then G(t) may be expressed as

$$G(t) = (n-1)(n+1)(n+2)\frac{\sigma}{\rho R_0^3}\left[1 - 3\delta \sin \omega t + O(\delta^2)\right] - \frac{3}{4}\delta^2\omega^2\left[1 - 2\delta \sin \omega t + O(\delta^2)\right]$$

$$+ \left(n + \frac{1}{2}\right)\delta\omega^2 \sin \omega t \left[1 - \delta \sin \omega t + O(\delta^2)\right] \,. \tag{11}$$

With G(t) so expressed, the differential equation (6) can be recognized as belonging to the kind of equations known as Hill's equation.

When $\delta \ll 1$, by retaining only those leading terms in the expression of G(t), we may write (11) as:

$$G(t) = \alpha + \beta \sin \omega t \,, \tag{12}$$

where

$$\alpha = (n-1)(n+1)(n+2)\frac{\sigma}{\rho R_0^3} - \frac{1}{2}\left(n+\frac{5}{4}\right)\delta^2\omega^2 \,, \tag{13}$$

and

$$\beta = \delta\left[\left(n+\frac{1}{2}\right)\omega^2 - (n-1)(n+1)(n+2)\frac{3\sigma}{\rho R_0^3}\right] \,. \tag{14}$$

Equation (6) is then just the Mathieu Equation.

The stability theory of solutions of Mathieu equation is well known [11]. Relations between parameters n, σ, ρ, δ, ω and R_o may be obtained to determine the region of stability or instability of the solutions. Or to be more specific, with known values of σ, ρ, δ and ω, we may determine the critical value of R_o which is the transition value between stability and instability.

Without going into details of determining the exact stability conditions, the critical radius may be roughly evaluated by the following considerations, with the aid of the stability chart for the Mathieu equation [11]. First of all, the solution is essentially unstable if $\alpha < 0$. From (13), it may be concluded that the solution is always unstable for the case $n = 1$. But this case corresponds to the displacement of the entire bubble in the fluid, and therefore is not significant for the consideration of the stability of the spherical shape of the bubble.

For $n \geq 2$, the condition $\alpha < 0$ requires that

$$R_0 > \left(\frac{2(n-1)(n+1)(n+2)\,\sigma}{\left(n+\frac{5}{4}\right)\rho\,\omega^2\delta^2}\right)^{1/3} \,.$$

Or, in an equivalent way of speaking, it is necessary, to insure stability, that

$$R_0 < \left(\frac{2(n-1)(n+1)(n+2)\sigma}{(n+\frac{5}{4})\rho\omega^2\delta^2}\right)^{1/3} . \tag{15}$$

Now, either from the preceding discussion or by comparing the coefficients α and β in (12), it is evident from the stability chart that for $n \geqq 2$, the higher the mode of distortion, the more stable is the solution. Therefore, for the determination of the critical radius, it is sufficient to consider the case $n = 2$ only.

For $n = 2$, equation (12) becomes

$$G(t) = \left(\frac{12\sigma}{\rho R_0^3} - \frac{13}{8}\omega^2\delta^2\right) + \delta\left(\frac{5}{2}\omega^2 - \frac{36\sigma}{\rho R_0^3}\right)\sin\omega t . \tag{16}$$

An order of magnitude criterion of stability is thus

$$\frac{12\sigma}{\rho R_0^3} \gtrsim \frac{5}{2}\delta\omega^2 . \tag{17}$$

Notice that the condition (15) is implied in the above condition. From equation (17), we obtain the critical radius of stability as

$$R_{cr.} \sim \left(\frac{24\sigma}{5\rho\delta\omega^2}\right)^{1/3} . \tag{18}$$

The solution is stable only if the mean radius R_0 is less than this critical radius R_{cr}.

To illustrate this general result, let us consider the case of an air bubble in water. For this case we have

$$\sigma = 73.5 \ dyne/cm , \qquad \text{and} \qquad \rho = 1 \ gm/cm^3 .$$

Now take $\delta = \frac{1}{100}$ and $\omega = 10^4$ rad/sec. Then

$$R_{cr.} \sim 10^{-1} \text{ cm.}$$

This value is not inconsistent with the experimental observations with sonic and ultrasonic pressure oscillations in water. However, whether the absence of large air bubbles is due solely to the instability just discussed, still awaits the exact experimental verifications.

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Appendix 1

Consider the polynomial

$$P(u) = (\beta u^2 + Au + B)(u^4 + \omega_1^2) + \alpha E(1+W_r)u^2 ,$$

where β, A, B, ω , α, E, W_r are all positive real constants. Let us denote $G = \alpha E(1 + W_r) > 0$, then

$$P(u) = (\beta u^2 + Au + B)(u^4 + \omega_1^2) + Gu^2 .$$

Since β, A, B, ω and G are all positive real constants, it is clear that $P(u) = 0$ has no positive real root. Let

$$u = v e^{i\pi/4} = \frac{1}{\sqrt{2}}(1+i)v$$

Thus

$$P(ve^{i\pi/4}) = [i\beta v^2 + \frac{A}{\sqrt{2}}(1+i)v + B](-v^4 + \omega_1^2) + iGv^2 ,$$

$$= M(v) + iN(v) ,$$

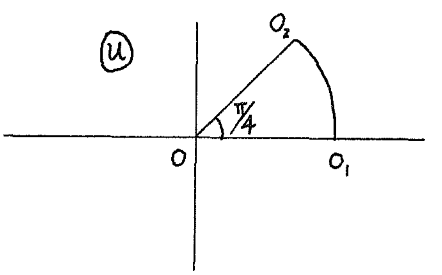
where

$$M(v) = -(\frac{A}{\sqrt{2}}v + B)(v^4 - \omega_1^2) ,$$

and

$$N(v) = (\beta v^2 + \frac{A}{\sqrt{2}}v)(-v^4 + \omega_1^2) + Gv^2 .$$

Now if $P(Ve^{i\pi/4}) = 0$ has any real root, for this real V, M(V) and N(V) must vanish separately. Now the only real roots of $M(V) = 0$ are $V = +\sqrt{\omega_1}$, $-\sqrt{\omega_1}$ and $-\frac{\sqrt{2}B}{A}$. Since $N(+\sqrt{\omega_1}) \neq 0$. Therefore we may conclude in particular that $P(Ve^{i\pi/4}) = 0$ has no positive real root. In other words, P(u) has no zero along the line OO_2 shown in the figure.



Now the principle of the argument [12] says that, if f(Z) is regular inside a closed contour and is not zero at any point on the contour, then

$$N = \frac{1}{2\pi} \Delta_c \arg f(z) ,$$

where N is the number of zeros of f(Z) inside C and $\Delta_c \arg f(Z)$ is the variation of the argument of f(Z) round the contour C.

Now consider the contour OO_1O_2O which consists of the positive real axis OO_1, the infinite arc O_1O_2 and the line OO_2 which is obtained from rotating OO_1 by an angle $\pi/4$.

Now since P(u) is real and positive along OO_1, then

$$\Delta_{OO_1} \arg P(u) = 0 .$$

We express u by its absolute value and argument, i.e. $u = Re^{i\theta}$.

Then

$$P(u) = \beta R^6 e^{i6\theta} [1 + O(\tfrac{1}{R})] , \quad \text{as } R \to \infty .$$

Hence

$$\Delta_{O_1O_2} \arg P(u) = 6 \times \frac{\pi}{4} = \frac{3\pi}{2} .$$

Along OO_2

$$\arg P(u) = \tan^{-1} \frac{N(v)}{M(v)} .$$

For V = 0:

$$M(0) = \beta \omega_1^2 ;$$

$$N(0) = 0 ;$$

and let us take arg P(0) = 0.

For $V \to \infty$:

$$M(v) \sim - \frac{A}{\sqrt{2}} V^5 ;$$

$$N(v) \sim - \beta V^6 .$$

Therefore

$$\arg P(ve^{i\pi/4}) = \tan^{-1} V \;, \qquad as \quad V \longrightarrow +\infty \;;$$

and

$$\arg P(ve^{i\pi/4}) = \frac{n}{2}\pi \;, \qquad as \quad V \longrightarrow +\infty \;;$$

where n may take any positive or negative integral value, and n is determined by the number of infinities of $\frac{N(V)}{M(V)}$, and the way it jumps.

In our case, along OO_2, there is only one infinity of $\frac{N(V)}{M(V)}$, namely, $V = +\sqrt{\omega_1}$, and $\frac{N(V)}{M(V)}$ changes from $+\infty$ to $-\infty$, as V passes $V = +\sqrt{\omega_1}$, in the direction of increasing V. Therefore we conclude that

$$\arg P(ve^{i\pi/4}) = \frac{3}{2}\pi \;, \qquad as \quad V \longrightarrow +\infty \;.$$

Hence

$$\Delta_{O_2O} \arg P(u) = 0 - \frac{3}{2}\pi = -\frac{3}{2}\pi \;.$$

Thus

$$\Delta_c \arg P(u) = \Delta_{OO_1O_2O} \arg P(u)$$
$$= 0 \;.$$

Therefore there is no zero of P(u) for $0 \leq \arg u \leq \frac{\pi}{4}$. Since the complex roots of a polynomial equation with real coefficients appear in pairs of complex conjugates, it thus follows that there is no zero of P(u) for $-\frac{\pi}{4} \leq \arg u \leq 0$.

Thus $P(u) = 0$ has no root for which $|\arg u| \leq \frac{\pi}{4}$.

Appendix 2

From the correlation between the error function and the confluent hypergeometric functions (cf. [13]) we obtain

$$\mathrm{Erfc}(z) = \frac{e^{-z^2}}{z\sqrt{\pi}}\left(1 - \frac{2!}{1!(2z)^2} + \frac{4!}{2!(2z)^4} - \frac{6!}{3!(2z)^6} + \cdots\right),$$

for large $|z|$ and $|\arg z| < \frac{3\pi}{4}$. Thus $e^{z^2}\mathrm{Erfc}(z) = O(\frac{1}{z})$, for large $|z|$, $|\arg z| < \frac{3\pi}{4}$; we have

$$1 - \mathrm{Erfc}\left(\sqrt{\tfrac{\pi}{2}}\, x\, e^{i\pi/4}\right) = (1+i)\left[C(x) - i\,S(x)\right],$$

where x is real, and also

$$C(x) = \int_0^x \cos\left(\tfrac{\pi}{2}t^2\right)dt \quad \text{and} \quad S(x) = \int_0^x \sin\left(\tfrac{\pi}{2}t^2\right)dt,$$

so that

$$\begin{aligned} 1 - \mathrm{Erfc}\left(-\sqrt{\tfrac{\pi}{2}}\, x\, e^{i\pi/4}\right) &= (1+i)\left[C(-x) - i\,S(-x)\right], \\ &= -(1+i)\left[C(x) - i\,S(x)\right]. \end{aligned}$$

Now for large x, (cf. [14])

$$C(x) = \frac{1}{2} + \frac{1}{\pi x}\sin\frac{\pi}{2}x^2 + O\left(\frac{1}{x^2}\right);$$

$$S(x) = \frac{1}{2} - \frac{1}{\pi x}\cos\frac{\pi}{2}x^2 + O\left(\frac{1}{x^2}\right).$$

Thus

$$\mathrm{Erfc}\left(-\sqrt{\tfrac{\pi}{2}}\, x\, e^{i\pi/4}\right) = 2 + O\left(\frac{1}{x}\right), \quad \text{as } x \to \infty,$$

while

$$\mathrm{Erfc}\left(\sqrt{\tfrac{\pi}{2}}\, x\, e^{i\pi/4}\right) = O\left(\frac{1}{x}\right), \quad \text{as } x \to \infty.$$

Thus Eq. (45) Part IA and Eq. (46) Part IA yield for large t:

$$\theta_2(t) = 2\sqrt{\omega}\, b_8\, e^{i(\omega t + \pi/4)} + O\left(\frac{1}{t}\right);$$

and

$$x(t) = 2\sqrt{\omega}\, C_8\, e^{i(\omega t + \pi/4)} + O\left(\frac{1}{t}\right).$$

Now from (I, A-40, 42) we have

$$\sum_{i=1}^{8} \frac{b_i}{s^{1/2} + a_i} = \frac{\alpha E \epsilon_0 s}{(s - i\omega) P(s^{1/2})} .$$

Thus

$$b_8 = \left(\frac{(s^{1/2} + a_8) \alpha E \epsilon_0 s}{(s - i\omega) P(s^{1/2})} \right)_{s^{1/2} = -a_8} = \frac{\alpha E \epsilon_0 (-a_8)^2}{(-a_8 + a_7) P(-a_8)} .$$

Or

$$b_8 = \frac{\alpha E \epsilon_0 i\omega}{2\sqrt{\omega} e^{i\pi/4} P(\sqrt{\omega} e^{i\pi/4})} .$$

Again from (I, A-41, 43) we have

$$\sum_{i=1}^{8} \frac{c_i}{s^{1/2} + a_i} = - \frac{\alpha \epsilon_0 (\beta s + A s^{1/2} + B)}{(s - i\omega) P(s^{1/2})} .$$

Thus

$$c_8 = - \frac{\alpha \epsilon_0 (i\beta\omega + A\sqrt{\omega} e^{i\pi/4} + B)}{2\sqrt{\omega} e^{i\pi/4} P(\sqrt{\omega} e^{i\pi/4})} .$$

Therefore, for large t, we have

$$\theta_2(t) \sim \frac{\alpha E \epsilon_0 \omega}{P(\sqrt{\omega} e^{i\pi/4})} e^{i(\omega t + \frac{\pi}{2})} ;$$

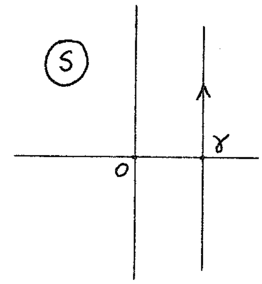
$$x(t) \sim - \frac{\alpha \epsilon_0 (\beta\omega + A\sqrt{\omega} e^{-i\pi/4} - iB)}{P(\sqrt{\omega} e^{i\pi/4})} e^{i(\omega t + \frac{\pi}{2})} ;$$

$$p(t) \sim \frac{\alpha \epsilon_0 (1 + W_r) [(E + 3\beta)\omega + 3A e^{-i\pi/4} \sqrt{\omega} - 3iB]}{P(\sqrt{\omega} e^{i\pi/4})} e^{i(\omega t + \frac{\pi}{2})} .$$

Appendix 3

From (I, A, 40) and (I, A, 41), we have

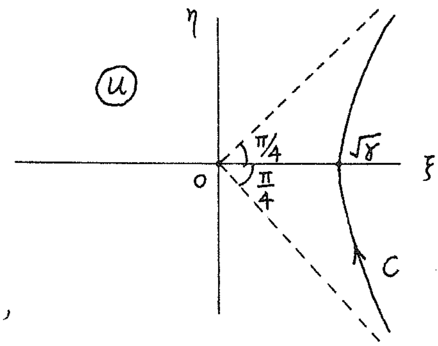
$$\theta_2(t) = \frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty} \frac{\alpha \epsilon_0 E s\, e^{st}}{(s-i\omega)P(s^{1/2})}\, ds \, , \qquad (1)$$



and

$$\chi(t) = \frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty} - \frac{\alpha \epsilon_0 (\beta s + A s^{1/2} + B)\, e^{st}}{(s-i\omega)\, P(s^{1/2})}\, ds \, , \qquad (2)$$

where the path of integration is to the right of all the singularities of the integrands concerned.



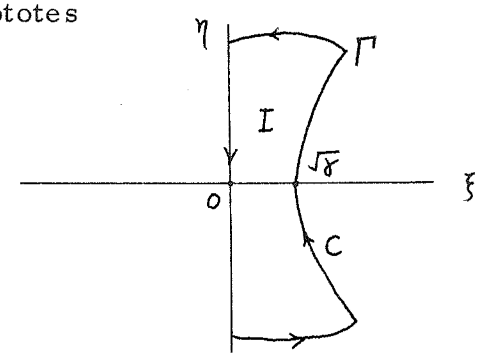
Let $s = u^2$ and $u = \xi + i\eta$.

Then in the u-plane, the integral (1) becomes

$$\theta_2(t) = \frac{1}{2\pi i}\int_c \frac{2\alpha \epsilon_0 E u^3 e^{u^2 t}\, du}{(u^2 - i\omega)\, P(u)} \, ,$$

where c is the branch of the hyperbola $\xi^2 - \eta^2 = \gamma$ that lies in the right half plane. This hyperbola has asymptotes

$$\xi = \pm \eta \, .$$



Consider the closed contour Γ ; due to the factor $e^{u^2 t}$, the previous integral becomes, after applying Cauchy's theorem:

$$\theta_2(t) = \frac{1}{2\pi i}\int_{-i\infty}^{i\infty} \frac{2\alpha \epsilon_0 E u^3 e^{u^2 t}\, du}{(u^2 - i\omega)\, P(u)} + \sum R \, ,$$

where $\sum R$ denotes the sum of residues of the integrand at its poles in I. As mentioned before, all the roots of P(u), i.e., $-a_i$, have arguments of absolute magnitude greater than $\pi/4$; thus it is easy to see that, due to the factor $e^{u^2 t}$, no significant contribution can result from them for large t. The residue due to the pole $u = \sqrt{\omega}\, e^{i\pi/4}$, however, is equal to

$$\frac{\alpha \epsilon_0 E i \omega e^{i\omega t}}{P(\sqrt{\omega}\, e^{i\pi/4})} .$$

Finally

$$\frac{1}{2\pi i}\int_{-i\infty}^{i\infty} \frac{2\alpha \epsilon_0 E u^3 e^{u^2 t}\, du}{(u^2 - i\omega)\, P(u)} \sim O(t^{-5/2}) ,$$

for large t, either by the method of steepest descent or Watson's Lemma [10]. Thus

$$\theta_2(t) = \frac{\alpha \epsilon_0 E \omega\, e^{i(\omega t + \pi/2)}}{P(\sqrt{\omega}\, e^{i\pi/4})} + O(t^{-5/2}), \quad as\ t \to \infty .$$

Similarly

$$x(t) = -\frac{\alpha \epsilon_0 (\beta \omega + A\sqrt{\omega}\, e^{-i\pi/4} - iB)}{P(\sqrt{\omega}\, e^{i\pi/4})}\, e^{i(\omega t + \pi/2)} + O(t^{-3/2}) ,$$

$$as \quad t \to \infty .$$

Appendix 4

To solve

$$\frac{\partial}{\partial t}(r\theta_1) = D\frac{\partial^2}{\partial r^2}(r\theta_1), \tag{1}$$

with

$$\theta_1(r,0) = \theta_1(\infty,t) = 0, \tag{2}$$

and

$$\theta_1(R(t),t) = -3C_\infty\delta\sin\omega t, \tag{3}$$

up to $O(\delta^2)$, we note that

$$\theta_1(R(t),t) = \theta_1(R_o,t) + (R-R_o)\left(\frac{\partial\theta_1}{\partial r}\right)_{r=R_o} + \cdots\cdots,$$

or

$$\theta_1(R_o,t) = -3C_\infty\delta\sin\omega t - R_o\delta\sin\omega t\left(\frac{\partial\theta_1}{\partial r}\right)_{r=R_o} - \cdots\cdots \tag{4}$$

We may ignore the remaining terms, since they are of the order of δ^3. Now let us solve first the equation

$$\frac{\partial}{\partial t}(r\theta_o) = D\frac{\partial^2}{\partial r^2}(r\theta_o), \tag{5}$$

with

$$\theta_o(r,0) = \theta_o(\infty,t) = 0, \tag{6}$$

and

$$\theta_o(R_o,t) = -3C_\infty\delta\sin\omega t. \tag{7}$$

Denote

$$v_o(r;s) = \mathcal{L}\{r\theta_o\} = \int_0^\infty r\theta_o(r,t)e^{-st}dt. \tag{8}$$

Then the transformed equation and conditions become

$$\frac{d^2 v_0}{d r^2} - \frac{s}{D} v_0 = 0 \ , \tag{9}$$

with

$$\lim_{r \to \infty} \frac{v_0}{r} = 0 \ ,$$

and

$$v_0 (R_0 ; s) = - 3 R_0 C_\infty \delta \frac{\omega}{s^2 + \omega^2} \ . \tag{10}$$

Thus

$$v(r; s) = - \frac{3 R_0 C_\infty \omega \delta}{s^2 + \omega^2} e^{-(r - R_0)\sqrt{\frac{s}{D}}} \ . \tag{11}$$

Using the inversion formula, we obtain

$$r \theta_0 (r,t) = - \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{3 R_0 C_\infty \omega \delta \, e^{-(r-R_0)\sqrt{\frac{s}{D}}}}{s^2 + \omega^2} e^{st} ds \ . \tag{12}$$

Thus for large t, we have

$$\theta_0 (r,t) = - \frac{3 R_0 C_\infty \delta}{r} e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \sin\left[\omega t - (r-R_0)\sqrt{\frac{\omega}{2D}}\right] + O(t^{-3/2}) ; \tag{13}$$

and

$$\frac{\partial \theta_0}{\partial r} = 3 R_0 C_\infty \delta \, e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \left\{ \left(\frac{1}{r^2} + \frac{1}{r}\sqrt{\frac{\omega}{2D}}\right) \sin\left[\omega t - (r-R_0)\sqrt{\frac{\omega}{2D}}\right] + \frac{1}{r}\sqrt{\frac{\omega}{2D}} \cos\left[\omega t - (r-R_0)\sqrt{\frac{\omega}{2D}}\right] \right\} + O(t^{-3/2}) \ . \tag{14}$$

Neglecting terms of the order of $t^{-3/2}$, we thus have

$$\left(\frac{\partial \theta_0}{\partial r}\right)_{r=R_0} = \frac{3 C_\infty \delta}{R_0} \left[\left(1 + R_0 \sqrt{\frac{\omega}{2D}}\right) \sin\omega t + R_0 \sqrt{\frac{\omega}{2D}} \cos \omega t \right] \ . \tag{15}$$

Now θ_1 will be solved by putting in (4) $\left(\frac{\partial \theta_0}{\partial r}\right)_{r=R_0}$ in place of $\left(\frac{\partial \theta_1}{\partial r}\right)_{r=R_0}$.

Then

$$R_0\theta_1(R_0,t) = -3R_0 C_\infty \delta \left\{ \sin\omega t + \delta\left[\left(1+R_0\sqrt{\tfrac{\omega}{2D}}\right)\sin^2\omega t + \frac{R_0}{2}\sqrt{\frac{\omega}{2D}}\,\sin 2\omega t \right]\right\}. \quad (16)$$

Now let $v_1(r;s) = \mathcal{L}\{r\theta_1\}$; it is easy to see that

$$v_1(r;s) = v_1(R_0;s)\, e^{-(r-R_0)\sqrt{\frac{s}{D}}}, \quad (17)$$

where

$$v_1(R_0;s) = -3R_0 C_\infty \delta \left\{ \frac{\omega^2}{s^2+\omega^2} + \frac{\delta}{2}\left[\left(1+R_0\sqrt{\tfrac{\omega}{2D}}\right)\left(\frac{1}{s} - \frac{s}{s^2+4\omega^2}\right) + R_0\sqrt{\frac{\omega}{2D}}\,\frac{2\omega}{s^2+4\omega^2} \right]\right\}. \quad (18)$$

Thus we obtain from the inversion formula asymptotically for large t:

$$\theta_1(r,t) = -\frac{3R_0 C_\infty \delta}{r}\left\{ e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}}\sin\left[\omega t - (r-R_0)\sqrt{\tfrac{\omega}{2D}}\right] + \frac{\delta}{2}\left[\left(1+R_0\sqrt{\tfrac{\omega}{2D}}\right)\mathrm{Erfc}\left(\frac{r-R_0}{\sqrt{4Dt}}\right) - \left(1+R_0\sqrt{\tfrac{\omega}{2D}}\right)e^{-(r-R_0)\sqrt{\frac{\omega}{D}}}\cos\left(2\omega t-(r-R_0)\sqrt{\tfrac{\omega}{D}}\right) + R_0\sqrt{\frac{\omega}{2D}}\,e^{-(r-R_0)\sqrt{\frac{\omega}{D}}}\sin\left(2\omega t-(r-R_0)\sqrt{\tfrac{\omega}{D}}\right)\right]\right\} + O(t^{-3/2}). \quad (19)$$

Now

$$\left(\frac{\partial\theta_1}{\partial r}\right)_{r=R} = \left(\frac{\partial\theta_1}{\partial r}\right)_{r=R_0} + (R-R_0)\left(\frac{\partial^2\theta_1}{\partial r^2}\right)_{r=R_0} + O(\delta^3). \quad (20)$$

From (19), we have

$$\frac{\partial\theta_1}{\partial r} = 3R_0C_\infty\delta\left\{\left(\frac{1}{r^2}+\frac{1}{r}\sqrt{\frac{\omega}{2D}}\right)\sin\left[\omega t-(r-R_0)\sqrt{\tfrac{\omega}{2D}}\right] + \frac{1}{r}\sqrt{\frac{\omega}{2D}}\cos\left[\omega t-(r-R_0)\sqrt{\tfrac{\omega}{2D}}\right]\right\}e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}}$$

$$+\frac{3}{2}R_0C_\infty\delta^2\left\{\left(1+R_0\sqrt{\tfrac{\omega}{2D}}\right)\left[\frac{1}{r^2}\mathrm{Erfc}\left(\frac{r-R_0}{\sqrt{4Dt}}\right) + \frac{1}{r\sqrt{\pi Dt}}\,e^{-\frac{(r-R_0)^2}{4Dt}}\right] + S\right\}, \quad (21)$$

where S denotes those sinusoidal terms which will not contribute to the rectification up to the second order.

Also

$$\frac{\partial^2\theta_1}{\partial r^2} = -3R_0C_\infty\delta\left\{\left(\frac{2}{r^3}+\frac{2}{r^2}\sqrt{\frac{\omega}{2D}}\right)\sin\left[\omega t-(r-R_0)\sqrt{\frac{\omega}{2D}}\right]\right.$$

$$\left.+\left(\frac{2}{r^2}\sqrt{\frac{\omega}{2D}}+\frac{2}{r}\frac{\omega}{2D}\right)\cos\left[\omega t-(r-R_0)\sqrt{\frac{\omega}{2D}}\right]\right\}e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} + O(\delta^2). \quad (22)$$

Thus from (20), since $R - R_o = R_o\delta\sin\omega t$, we have

$$\left(\frac{\partial\theta_1}{\partial r}\right)_{r=R} = 3R_0C_\infty\delta\left\{\left(\frac{1}{R_0^2}+\frac{1}{R_0}\sqrt{\frac{\omega}{2D}}\right)\sin\omega t+\frac{1}{R_0}\sqrt{\frac{\omega}{2D}}\cos\omega t\right\}$$

$$+3R_0C_\infty\delta^2\left\{\left(1+R_0\sqrt{\frac{\omega}{2D}}\right)\left(\frac{1}{2R_0^2}+\frac{1}{2R_0\sqrt{\pi Dt}}+\frac{2}{R_0^2}\sin^2\omega t\right)+S\right\}$$

$$+O(t^{-3/2})+O(\delta^3). \quad (23)$$

Appendix 5

We want to solve the following equation:

$$\frac{\partial^2}{\partial r^2}(r\theta_2) - \frac{1}{D}\frac{\partial}{\partial t}(r\theta_2) = g(r,t) \,, \tag{1}$$

where $g(r,t) = \frac{R^2\dot{R}}{Dr}\frac{\partial\theta_1}{\partial r}$ to the order of δ^2. From the result in Appendix 4, since $\dot{R} = \delta R_0 \omega \cos\omega t$, we have

$$g(r,t) = \frac{3R_0^4 C_\infty \omega \delta^2}{Dr} e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \left\{ \left(\frac{1}{r^2} + \frac{1}{r}\sqrt{\frac{\omega}{2D}}\right) \sin\left[\omega t - (r-R_0)\sqrt{\frac{\omega}{2D}}\right] \right.$$

$$\left. + \frac{1}{r}\sqrt{\frac{\omega}{2D}} \cos\left[\omega t - (r-R_0)\sqrt{\frac{\omega}{2D}}\right] \right\} \cos\omega t + O(\delta^3) \,. \tag{2}$$

The initial and boundary conditions, up to the same order, are

$$\theta_2(r,0) = \theta_2(\infty,t) = 0 \,, \tag{3}$$

and

$$\theta_2(R_0,t) = 0 \,. \tag{4}$$

Apply Laplace Transformation, and let $\phi = \mathcal{L}\{\theta_2\}$, also put

$$h(r;s) = \mathcal{L}\{g(r,t)\} \;; \tag{5}$$

then it may be verified that:

$$\phi(r;s) = -\frac{1}{2r}\sqrt{\frac{D}{s}} \left[e^{-(r-R_0)\sqrt{\frac{s}{D}}} \int_{R_0}^{r} h(x;s) e^{(x-R_0)\sqrt{\frac{s}{D}}} dx + e^{(r-R_0)\sqrt{\frac{s}{D}}} \int_{r}^{\infty} h(x;s) e^{-(x-R_0)\sqrt{\frac{s}{D}}} dx \right.$$

$$\left. - e^{-(r-R_0)\sqrt{\frac{s}{D}}} \int_{R_0}^{\infty} e^{-(x-R_0)\sqrt{\frac{s}{D}}} h(x;s)\, dx \right] . \tag{6}$$

Hence

$$\frac{d\phi}{dr} = -\frac{1}{2}\sqrt{\frac{D}{s}}\Big[\Big(-\frac{1}{r^2}-\frac{1}{r}\sqrt{\frac{s}{D}}\Big)\int_{R_0}^{r} e^{-(r-R_0)\sqrt{\frac{s}{D}}}\, e^{(x-R_0)\sqrt{\frac{s}{D}}}\, h(x;s)\,dx$$

$$+\Big(-\frac{1}{r^2}+\frac{1}{r}\sqrt{\frac{s}{D}}\Big)\, e^{(r-R_0)\sqrt{\frac{s}{D}}}\int_{r}^{\infty} e^{-(x-R_0)\sqrt{\frac{s}{D}}}\, h(x;s)\,dx$$

$$+\Big(\frac{1}{r^2}+\frac{1}{r}\sqrt{\frac{s}{D}}\Big)\, e^{-(r-R_0)\sqrt{\frac{s}{D}}}\int_{R_0}^{\infty} e^{-(x-R_0)\sqrt{\frac{s}{D}}}\, h(x;s)\,dx\Big] \quad . \tag{7}$$

Thus

$$\left(\frac{d\phi}{dr}\right)_{r=R_0} = -\frac{1}{R_0}\int_{R_0}^{\infty} e^{-(x-R_0)\sqrt{\frac{s}{D}}}\, h(x;s)\,dx \quad . \tag{8}$$

Now let us rewrite the expression of $g(r,t)$ in (2). Then we have

$$g(r,t) = 3R_0^4 C_\infty\Big(\frac{\omega}{2D}\Big)\delta^2\Big\{\Big[\Big(\frac{1}{r^3}+\frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\Big)\cos(r-R_0)\sqrt{\frac{\omega}{2D}} + \frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\, \sin(r-R_0)\sqrt{\frac{\omega}{2D}}\Big]\sin 2\omega t$$

$$+\Big[\Big(-\frac{1}{r^3}-\frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\Big)\sin(r-R_0)\sqrt{\frac{\omega}{2D}} + \frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\cos(r-R_0)\sqrt{\frac{\omega}{2D}}\Big]$$

$$\times\Big[1+\cos 2\omega t\Big]\Big\}\, e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \qquad + \quad O(\delta^3) \quad . \tag{9}$$

From (8) it is fairly obvious that excluding those terms which at most contribute to the rectification of the order of $O(\delta^3)$ and $O(t^{-3/2})$ the relevant term in $g(r,t)$ is just

$$g_1(r) = 3R_0^4 C_\infty\Big(\frac{\omega}{2D}\Big)\delta^2 e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}}\Big\{\Big(-\frac{1}{r^3}-\frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\Big)\sin\Big[(r-R_0)\sqrt{\frac{\omega}{2D}}\Big]$$

$$+\frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\cos\Big[(r-R_0)\sqrt{\frac{\omega}{2D}}\Big]\Big\} \quad . \tag{10}$$

As $\quad h_1(r;s) = \mathcal{L}\{g_1(r)\} = \frac{1}{s}\, g_1(r)$, it follows that

$$\left(\frac{\partial\theta_2}{\partial r}\right)_{r=R_0} = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} ds\Big[-\frac{1}{R_0 s}\, e^{st}\int_{R_0}^{\infty} e^{-(x-R_0)\sqrt{\frac{s}{D}}}\, g_1(x)\,dx\Big] \quad + \quad T \, . \tag{11}$$

where T denotes irrelevant terms. As we are only interested in the behavior of the solution for large t, we thus expand $e^{-(x-R_0)\sqrt{\frac{s}{D}}}$ in ascending powers of $s^{1/2}$, and obtain:

$$\left(\frac{\partial\theta_2}{\partial r}\right)_{r=R_0} = -\frac{1}{R_0}\left[\int_{R_0}^{\infty} g_1(x)\,dx - \frac{1}{\sqrt{\pi D t}}\int_{R_0}^{\infty}(x-R_0)\,g_1(x)\,dx\right] + O(t^{-3/2}) . \tag{12}$$

Since

$$r\,g_1(r) = 3R_0^4 C_\infty\left(\frac{\omega}{2D}\right)\delta^2 \frac{d}{dr}\left[\frac{1}{r}\, e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}}\sin (r-R_0)\sqrt{\frac{\omega}{2D}}\right],$$

therefore

$$\int_{R_0}^{\infty} x\, g_1(x)\,dx = 0 \quad ;$$

and this leads to the result that

$$\left(\frac{\partial\theta_2}{\partial r}\right)_{r=R_0} = -\frac{1}{R_0}\left(1+\frac{R_0}{\sqrt{\pi D t}}\right)\int_{R_0}^{\infty} g_1(x)\,dx + O(t^{-3/2}) . \tag{13}$$

We may observe that $\left(\frac{\partial\theta_2}{\partial r}\right)_{r=R_0} = O(\delta^2)$. Hence, up to this order we have

$$\left(\frac{\partial\theta_2}{\partial r}\right)_{r=R} \cong \left(\frac{\partial\theta_2}{\partial r}\right)_{r=R_0} = -\frac{1}{R_0}\left(1+\frac{R_0}{\sqrt{\pi D t}}\right)\int_{R_0}^{\infty} g_1(x)\,dx + O(t^{-3/2}) ,$$

$$= -3R_0^3 C_\infty\left(\frac{\omega}{2D}\right)\delta^2\left(1+\frac{R_0}{\sqrt{\pi D t}}\right) I_1 + O(t^{-3/2}) , \tag{14}$$

where

$$I_1 = \int_{R_0}^{\infty} e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}}\left[\left(-\frac{1}{r^3}-\frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\right)\sin (r-R_0)\sqrt{\frac{\omega}{2D}}\right.$$

$$\left. + \frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\cos (r-R_0)\sqrt{\frac{\omega}{2D}}\right] dr . \tag{15}$$

Appendix 6

To evaluate the integral

$$I_1 = \int_{R_0}^{\infty} \Big[\Big(-\frac{1}{r^3} - \frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\Big) \sin (r-R_0)\sqrt{\frac{\omega}{2D}} + \frac{1}{r^2}\sqrt{\frac{\omega}{2D}} \cos (r-R_0)\sqrt{\frac{\omega}{2D}} \Big] e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \, dr \quad , \tag{1}$$

let us note that

$$\frac{d}{dr}\Big[\frac{1}{r^2} e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \sin (r-R_0)\sqrt{\frac{\omega}{2D}}\Big]$$

$$= \Big[\Big(-\frac{2}{r^3} - \frac{1}{r^2}\sqrt{\frac{\omega}{2D}}\Big)\sin(r-R_0)\sqrt{\frac{\omega}{2D}} + \frac{1}{r^2}\sqrt{\frac{\omega}{2D}} \cos (r-R_0)\sqrt{\frac{\omega}{2D}}\Big] e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} ,$$

$$= -\frac{1}{r^3} e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \sin(r-R_0)\sqrt{\frac{\omega}{2D}} + i_1 \quad , \tag{2}$$

where i_1 is the integrand in I_1. From (2), we may thus write

$$I_1 = \int_{R_0}^{\infty} \frac{1}{r^3} e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \sin(r-R_0)\sqrt{\frac{\omega}{2D}} \, dr \; . \tag{3}$$

After changing variables, we have

$$I_1 = \int_0^{\infty} \frac{1}{(x+R_0)^3} e^{-x\sqrt{\frac{\omega}{2D}}} \sin \sqrt{\frac{\omega}{2D}}\, x \, dx \quad ,$$

$$= \mathrm{Im.} \Big[\int_0^{\infty} \frac{1}{(x+R_0)^3} e^{-\sqrt{\frac{\omega}{2D}}\,(x-ix)} dx \Big] \; . \tag{4}$$

Now let $y = \sqrt{2}\, e^{-i\pi/4} x = (1-i)x$, then apply Cauchy's Theorem to transform the integral along the real axis of the new coordinate system, and we then obtain

$$I_1 = \mathrm{Im.} \Big[\frac{e^{i\pi/4}}{\sqrt{2}} \int_0^{\infty} \frac{e^{-\sqrt{\frac{\omega}{2D}}\,y}}{(R_0 + \frac{e^{i\pi/4}}{\sqrt{2}} y)^3} dy \Big] \; . \tag{15}$$

For the case that $R_0\sqrt{\frac{\omega}{2D}} \gg 1$, we may apply Watson's Lemma, and get

$$I_1 = \text{Im.}\left[\frac{e^{i\pi/4}}{\sqrt{2}\, R_0^3} \left(\frac{1}{\sqrt{\frac{\omega}{2D}}} - \frac{3}{\sqrt{2}}\, e^{i\pi/4} \frac{1}{R_0 \frac{\omega}{2D}} \right) \right] + O\left(\frac{1}{R_0^5 \left(\frac{\omega}{2D}\right)^{3/2}} \right),$$

$$= \frac{1}{2R_0^2} \left[\left(\frac{1}{R_0\sqrt{\frac{\omega}{2D}}} - \frac{3}{R_0^2 \frac{\omega}{2D}} \right) + O\left(\frac{1}{\left(R_0\sqrt{\frac{\omega}{2D}}\right)^3} \right) \right] . \qquad (6)$$