Chapter 9

CONCLUSION AND OUTLOOK

This thesis presented a comprehensive investigation on various aspects related to the constant stress and pressure rheology of polydisperse colloidal suspensions. Using bidisperse suspensions as a model system, we first investigated the effects of particle sizes on the suspension short-time transport properties. Our results showed that introducing a second species of different particle size leads to qualitative differences in the suspension transport properties such as the sedimentation velocity. However, in many cases, the influences of particle sizes are only quantitative. We found that the pairwise additive approximation with proper structural input is valid in most cases up to a volume fraction of $\phi = 0.1$, demonstrating the strong influences of many-body effects in transport properties. In contrast, the semi-analytical approximation scheme with partial resummation of the many-body hydrodynamic interactions is valid up to $\phi = 0.4$.

Our work on the short-time transport properties serves as a critical reference for future experiments and simulations on bidisperse systems, and provides insight and guidance on using particle sizes to tune the suspension transport properties. The approximation scheme on the hydrodynamic functions signficantly simplifies the interpretation of scattering experiments for bidisperse suspensions. The computational scheme can be straightforwardly extended to other systems such as charge-stabilized colloidal suspensions. In this case, in addition to the size differences, the charge differences also affect the transport properties by changing the suspension structures. With proper structure predictions, we can significantly extend our capability to predict transport properties in other colloidal systems.

We also developed the Spectral Ewald Accelerated Stokesian Dynamics (SEASD) for computing hydrodynamic interactions in polydisperse colloidal suspensions. The main advantages of the SEASD include the polydisperse capability, the flexible error control via the Spectral Ewald (SE) method, and the GPU acceleration. We extensively validated the SEASD, and found that SEASD-nf, the near-field Brownian approximation of the algorithm, can quantitatively capture the hydrodynamic interactions in the rheology of bidisperse suspensions.

Comparing to other Particle-Mesh-Ewald (PME) techniques, the SE method is

simple and allows full error controls. The SEASD method can be conveniently extended to compute the hydrodynamic interactions of active particles with surface velocities. By casting the swimming model with surface velocity [1] into a form independent of the reference frame [2], we can adapt the SEASD to study dynamics of swimming particles. Furthermore, an area for improvement in the current SEASD is the computation of Brownian force, which is the most time-consuming step in dynamic simulations of colloidal systems. A possibility is to exploit the symmetry of the computation kernel, similarly to the approach in fluctuating hydrodynamics [3].

Our investigations on the film drying process of colloidal suspensions revealed the critical role of the confining boundary on the stress and structures of the colloidal suspensions. Interestingly, both fast and slow boundary motion contribute to structural formation, and the most amorphous structures occurs at a moderate boundary velocity. The simple continuum model is surprisingly effective for predicting the structural and stress evolution, but cannot capture the boundary-layer-like stress distributions and the volume fraction distribution at fast boundary motion. The model can be improved by incorporating non-local effects in way similarly to Kamrin and Koval [4].

We exhaustively investigated the behaviors of dense colloidal suspensions including the suspension mechanics, structures, and diffusive dynamics with constant imposed stress and pressure, and evaluated the role of hydrodynamic interactions through non-hydrodynamic and hydrodynamic simulations. We found that hydrodynamic interactions profoundly affect the suspension structures, but only quantitatively changes the behaviors of amorphous and glassy suspensions. We devised a simple model to describe the rheology and diffusion of glassy suspensions, highlighting the role of jamming physics on the suspension dynamics. We also discovered an universal collapse of simulation data representing an effective Stokes-Einstein-Sutherland (SES) relation with an effective temperature proportional to the osmotic pressure. This revelation suggests that a mean-field description for the rheology and dynamics of dense suspensions is possible.

We can use the rheology model as a constitutive relation to predict macroscopic behaviors of glassy suspensions using the approach of Jop et al. [5]. An example is microrheology in glass: i.e., the structure and stress of the glassy suspensions in response to a moving probe. The model prediction can be compared with the simulations and Mode Coupling Theory predictions [6]. Another point of interest is the validity of the effective SES relation beyond the hard-sphere systems.

References

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