Dynamic simulations of colloidal dispersions: sticky and polarizable particles as building blocks for novel materials

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Advanced materials are ubiquitous in almost every aspect of modern life. Continued development of these materials is crucial to maintain and improve societal well-being in areas such as healthcare, food, clean water, and environmentally responsible energy. The importance of materials discovery and development in society has been recognized in many ways, including by the U.S. Government with the launch of the Materials Genome Initiative(MGI) in 2011. More than $1 billion in resources has been devoted to this effort since it's start, and we have seen a great deal of scientific progress as a result. Clearly, research that allows us to more quickly discover, create, and scale up the manufacture of advanced materials is important to society.

Once class of promising materials is soft matter composed of colloidal particles. They offer a large range of material properties that can be accessed simply by changing the components and processing parameters. The interactions are often complex and occur at different length scales, thus making them challenging to control and model. Due to these complexities it can be costly to discover new soft materials, but the integration of computation and theory with experiment can help make this search more efficient. Motivated by the need to accelerate new materials design, this thesis aims to be a small piece of the puzzle in the endeavor of many past, current, and future scientists to improve computational tools for the study of soft materials, and to explore novel materials that show promise for future applications.

Within the realm of colloidal soft materials, there are a variety of interparticle interactions one can employ to achieve design goals. Colloidal particles that interact in the presence of an external applied field are appealing for many applications due to the change in material properties upon application of the field. While there has been a number of experimental studies of polarizable colloidal particle systems, there have been less computational studies until recent years due to a lack of computational tools to accurately model their behavior. Throughout this thesis we study polarizable colloidal particles via dynamic simulations in order to gain insight into their use in novel materials. In the first part of this thesis, we describe the derivation and numerical calculation of the stress tensor in dispersions of polarizable colloids based on a mutual dipole model for polarization. The stress tensor we derive builds upon previous work to more accurately model these dispersions in particle simulations.

In the remainder of this thesis we describe two Brownian dynamics (BD) simulation studies of a composite colloidal system that leverages short-range colloidal attractions and dipole-dipole interactions between polarizable particles in an applied field. The focus of this work is on two novel processing schemes that lead to different microstructures. Simulations of this scale and level of accuracy for this composite colloidal system have not been performed before. While this work gives insight into a specific composite material and processing scheme, it is also a part of a larger emerging area in colloidal gels: leveraging processing to unlock novel structures. These studies demonstrate the use of sophisticated BD simulations to explore processing conditions to achieve structures and properties of interest that can help guide experimental efforts.

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