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The Influence of Gravity on Nucleation, Growth, Stability and Structure in Crystallizing Colloidal Suspensions
NAG3-1652

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TASK OBJECTIVE:

Our goal is to understand the dynamics of particles within colloidal crystals. In particular, we focus on the influence of the cell walls and gravity on the particle dynamics. In this study, we will use a novel light scattering experiment, known as diffusing wave spectroscopy, to probe particle motions in turbid suspensions. This is a noninvasive experimental probe of interparticle dynamics.

TASK DESCRIPTION:

Aqueous suspensions of charged colloidal particles provide attractive model systems for the study of crystal dynamics due to the long range Coulombic repulsion that diminishes the influence of hydrodynamic interactions in the suspension. In this system of charged polystyrene latex synthesized in our laboratory with ionic comonomers, we can observe and provoke homogeneous nucleation to two crystal lattices. We can also study twinning and a crystallization growth instability analogous to those observed in molecular systems. These phenomena motivate and provide a means for us to investigate the role of interfacial tension, surface interactions and gravity on the crystallization process. A key feature of this study is the use of density matching experiments to study crystal growth without the influence of gravity. We suspend our particles in mixtures of deuterated and normal water to render them neutrally buoyant. We focus our investigation on the dynamics of particles in colloidal crystals using light scattering as our measurement tool. Future studies on more dense colloidal particles would require low gravity experiments.

TASK SIGNIFICANCE:

An important fundamental feature of this work is the application of diffusing wave spectroscopy to ordered suspensions. This has potential implications for the study of other interesting systems such as liquid crystals or protein arrays. Our combination of experimental and theoretical treatments provides a means to interpret the dynamics of particles in a lattice through the analysis of the multiply scattered light interacting with the array. This study will provide a test of our understanding of crystalline dynamics, the influence of external fields such as produced by walls or gravity and the nature of crystal defect formation.

PROGRESS DURING FY 1996:

In this program we aim to investigate the influence of gravity on particle dynamics in colloidal crystals. Our system is a electrostatically stabilized suspension of polystyrene latices in water. We control the interparticle interactions and hence the suspension structure and dynamics through the ionic strength of the suspending aqueous fluid. Unfortunately, such suspensions are quite opaque and require the multiple scattering technique known as diffusing wave spectroscopy (DWS) for their interrogation. We have extended this approach by developing a spatially resolved (SR-DWS) method capable of handling ordered colloidal suspensions. This novel scattering geometry with its variable incident beam and detector positions allows us to characterize the propagation of the diffusing light over different length scales.

We developed a mathematical analysis of the problem of light diffusing from a point source into a semi-infinite solid. The resulting autocorrelation function depends on the distance of our point detector from the point source and thus allows us to perform a series of experiments to test the diffusion model for ordered arrays. We illuminate the front face of our suspension with a focused beam of an Argon ion laser. The detection occurs through a 0.2 mm fiber optic cable located at a variable distance from the point source. The source to detection distance ranges from 400 to 3500 micrometers; ordered samples require the longer distance measurements to ensure that the light is fully diffusing. The autocorrelation function is calculated on a high-speed correlator.

We have shown that the autocorrelation function obtained from the diffusing photon model gives excellent agreement with the measured autocorrelation functions. This confirms the diffusive photon transport in both disordered and ordered samples; however, the transport mean free path is much larger for the ordered samples than for the disordered ones. This suggests that the optical transport in these suspensions is primarily affected by structures on length scales smaller than the size of the individual crystallites but much larger than the crystal lattice spacing.

Our fitting procedure also yields a measurement of the characteristic dynamic processes in the suspension through the relaxation time. We observe a dramatic enhancement in the dynamics for ordered suspensions. In contrast to the results for the transport mean free path, we see a dependence of the ordered dynamics on the suspension ionic strength. This suggests that the dynamic processes are primarily governed by structures and interactions on the length scale of the lattice.

We have also observed for the first time underdamped lattice modes at long times in the autocorrelation functions. These modes could not be observed in other experiments due to the thin sample cells used. Our new DWS geometry allowed us to measure these modes without the influence of confining walls. The analysis of these modes serves as a test of the harmonic lattice model.

Our new SR-DWS scattering experiment has given us a tool for properly interrogating ordered colloidal suspensions. The spatial resolution that it provides allows us to more fully characterize the suspension to obtain accurate measurements of its optical and dynamic properties. We have shown its applicability to ordered colloidal suspensions, but it should be equally useful for interrogating any number of other anisotropic systems such as those composed of anisotropic particles. This new modification to the DWS formalism will hopefully make it more applicable to a wider variety of experimental systems with the Laser Light Scattering facility being developed at NASA Lewis Research Center.

Students funded under research: PhD 1.3

Bibliographic Citations for FY 1996:

S. J. Nilsen and A. P. Gast , "Photon Transport in Disordered and Ordered Suspensions of Colloidal Particles," submitted J. Optical Society of America, April 1996. (sorry held up in review)

S. J. Nilsen "Structure and Dynamics in Ordered and Disordered Colloidal Suspensions," Dissertation, Stanford University, March 1996.

American Institute of Chemical Engineers, Miami Beach, Florida November 1995
"Dynamics of Ordered and Disordered Colloidal Suspensions" paper presented by A. Gast.