MAR07-2006-006318
Abstract for an Invited Paper
for the MAR07 Meeting of
the American Physical Society

Imaging the Dynamics of Freezing and Sublimation of Colloidal Crystals[1]

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We study the kinetics of freezing and sublimating colloidal crystals with single-particle resolution. In experiments, a short-ranged depletion attraction between spheres leads to crystallites that are one to three layers thick. The spheres are tracked with optical microscopy and the sizes and bond-orientational order parameters of the crystallites are measured. The inter-particle attraction is reduced or increased by modest changes in temperature, which lead either to sublimation of crystallites or to formation of crystallites from a gas phase. The sublimation process is also investigated using Brownian Dynamics simulations. In both experiments and simulations of sublimation, we find a two-stage process: at first, large crystallites sublimate by escape of particles from the perimeter. The rate of crystallite shrinkage is then greatly enhanced as the size falls below a cross-over value that ranges between 20 and 50 in different regions of the phase diagram. Simultaneous with the enhanced sublimation rate, the crystallites transform to a dense amorphous structure, which then rapidly vaporizes. The two-step kinetics are also seen in freezing at sphere area fractions near 0.3, but not at substantially higher or lower area fractions. The two-step kinetics are attributed to a thermodynamically meta- or unstable amorphous phase (ten Wolde and Frenkel, Science **277**, 1975 (1997).). The results should be relevant in diverse systems including colloids, proteins, and atoms such as Argon. We gratefully acknowledge support from Research Corporation and from the NSF through grant DMR-0605839.

[1]In collaboration with J. R. Savage, D. W. Blair, R. A. Guyer, and A. J. Levine.