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Slightly perturbed two-dimensional solid: crystal, hexatic or glass ?⁽¹⁾

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Abstract. -- Melting properties of a two-dimensional system comprised of $\sim 1 \mu\text{m}$ diameter, monodispersed, polystyrene spheres are compared with those of a system containing a one-percent concentration of larger spheres which have a larger interparticle interaction. The spheres interact via a dipolar interaction which is controlled externally. The translational and bond-orientational correlation functions are examined as a function of the dipole strength. The monodispersed system exhibits two-stage melting. In the system with a one-percent concentration of larger spheres the correlation functions are characteristic of a hexatic phase at the largest interaction strengths. Dislocations are pinned to the larger particles.

1. Introduction.

The nature of order in two-dimensional systems is an interesting problem. Perfect translational symmetry cannot exist in a two-dimensional solid due to the large fluctuations from long wavelength phonons. In the crystal phase the translational correlation function decays algebraically with distance while long-range order exists in the bond orientation, the direction parallel to the crystallographic axis [1]. Both of these correlations are destroyed by melting in three dimensions. In two dimensions the possibility exists that only translational order is destroyed by melting. In the KTHNY theory by Kosterlitz, Thouless, Halperin, Nelson and Young [2-4] dislocation pairs, which are thermally excited defects in the crystal, break up into free dislocations, at the melting point. Quasi-long-range translational order is destroyed by free dislocations but bond-orientational order persists. This leads to the hexatic phase in which translational correlations decay exponentially while bond-orientational correlations decay algebraically. At a higher temperature dislocations break up into free disclinations and destroy the hexatic phase. The high temperature phase is a fluid.

Another question of interest is whether translational correlation will be retained in a slightly perturbed two-dimensional solid. That is, does the crystal phase exist in the presence of interaction-potential perturbations. This question is closely related to whether a pinned electron solid exists in the crystal phase. We attempt to shed light on this latter question by comparing the correlation functions of a system with uniform interparticle interactions with one in which one-percent of the particles have a larger interaction potential.

Monolayers of polystyrene spheres have been used by a number of coworkers as a model system to study melting in two dimensions. Murray and Van Winkle [5], Armstrong et al. [6] and Tang et al. [7] have studied melting in monolayers of charged, polystyrene spheres interacting via a screened Coulomb interaction. Skjeltorp examined spheres in a ferrofluid. He observed the formation of a crystal phase when a magnetic field was applied normal to the monolayer. This was a result of dipole interactions induced

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when the ferrofluid was polarized.

Our system is the electrical analog of spheres in the ferrofluid. A monolayer of $\sim 1 \mu\text{m}$ diameter polystyrene spheres in water is confined between two, parallel, glass plates. A 3.5 MHz electric field is applied normal to the layer. The spheres can be treated as voids in the highly polarizable water layer. They have a dipole moment which is antiparallel to the applied field. The strength of the dipole moment is proportional to the product of the sphere volume and the applied field. Thus, the interaction strength is proportional to the square of the applied field.

2. Correlation functions.

The translational correlation function for the crystal phase is [3]

$$C_G(\mathbf{r} - \mathbf{r}') \propto |\mathbf{r} - \mathbf{r}'|^{-\eta_G}$$

where η_G is a function of the reciprocal lattice vector $\mathbf{G}$, the temperature and the Lamé coefficients of the lattice. Translational correlations decay exponentially in the hexatic and fluid phases. The bond-orientational correlation function approaches a constant at large r in the crystal phase. In the hexatic phase it decays algebraically:

$$C_6 \propto |\mathbf{r}|^{-\eta_6}$$

where η_6 depends on the Frank constant and the temperature. It decays exponentially in the fluid phase. The KTHNY theory predicts that the hexagonal crystal melts when $\eta_6 = 1/3$ and the hexatic phase is destroyed when $\eta_6 = 1/4$.

3. Experimental Cell.

A cross section of our cell is shown in Fig. 1. Semi-transparent electrodes for the application of an electric field are evaporated onto the outer sides of glass cover slides. These slides are sealed onto sapphire windows for optical access. The cell is held in an aluminum jig. Three differential micrometers are used to align the glass plates. Configurations of the particles are video-taped and digitized with a frame grabber for analysis.

Fig. 1. A schematic cross section of the colloidal monolayer cell

4. Results.

Below, data from a monodispersed, $2.0 \mu\text{m}$ sample and an impure $2.2 \mu\text{m}$ sample are presented. The thermodynamic parameter used to characterize the data is Γ , the ratio of the interparticle potential energy to the thermal energy. The raw data, i.e., the particle positions, are presented as triangulations. The triangulation employed is the dual of the Wigner-Seitz or Voronoi construction. The vertices of the triangulation mark particle positions. The interparticle bonds in the triangulation identify nearest neighbors.

Fig. 2. Triangulations of particle positions for a monodispersed sample (a.) and impure sample (b.): black circles - large impurity spheres. Dislocations can be discerned by looking at the images at a glancing angle.

Non-sixfold coordinated particles are identified as defects in the topological crystal. The correlation functions are displayed on log-log scales on which an algebraic dependence exhibits a straight line and exponentially decaying behavior exhibits a negative second derivative.

Fig. 2a. and 2b. show the triangulations for a monodispersed sample and the impure sample contaminated by a one-percent concentration of large spheres. The interparticle potential is high; for Fig. 2a. $\Gamma = 1.6 \Gamma_i$ and for 2b. $\Gamma = 1.7 \Gamma_i$, where Γ_i is the ratio of the interparticle potential energy to the temperature at the isotropic transition, i.e., where bond-orientational order is lost. Translational order is lost at $1.3 \Gamma_i$ in the monodispersed sample. This sample is well ordered showing large patches of sixfold coordinated particles; however, a few dislocations pairs and a triplet can be observed. The impure sample shows a high concentration of dislocations, each of which is pinned at a large impurity particle.

Fig. 3a. and 3b. show the correlations for the monodispersed sample in the crystalline and the hexatic phase, respectively. The two phases are distinguished by the distinct difference in the behavior of the translational correlation function. Such a change from algebraic to exponential behavior is not observed in the translational correlation functions of the impure sample, Fig. 4a. and 4b.

Fig. 3. Correlation functions for monodispersed sample on log-log scale: dashed line - translational correlation function; solid line - bond-orientational correlation function; upper and lower dotted lines - theoretical limits of algebraic decay for bond orientation and translation, respectively.

Fig. 4. Correlation functions on log-log scale for sample contaminated with one-percent concentration of large spheres: dashed line - translational correlation function; solid line - bond-orientational correlation function; upper and lower dotted lines - theoretical limits of algebraic decay for bond orientation and translation, respectively.

5. Discussion.

These results and others lead us to conclude that monodispersed samples of spheres confined to two dimensions, interacting via a dipole potential melt by a two-stage process consistent with KTHNY. The impure sample with a one-percent concentration large spheres does not show this behavior. No translationally ordered phase is detected, even at interaction strengths well above that needed to order a monodispersed sample. We find that this disorder is caused by the pinning of dislocations to the large particles, for which we estimate the interparticle potential to be approximately four times greater than that between the plentiful, smaller particles in the sample. While this cannot be considered a weak perturbation, the fact that such a low concentration of impurities, i.e., one-percent, can destroy order is interesting and possibly significant to the pinned electron solid.

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