Fluctuating Topological Defects in 2D Liquids: Heterogeneous Motion and Noise

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We measure the defect density as a function of time at different temperatures in simulations of a two-dimensional system of interacting particles. Just above the solid to liquid transition temperature, the power spectrum of the defect fluctuations shows a $1/f$ signature, which crosses over to a white noise signature at higher temperatures. When $1/f$ noise is present, the 5–7 defects predominantly form stringlike structures, and the particle trajectories show a 1D correlated motion that follows the defect strings. At higher temperatures this heterogeneous motion is lost.

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In liquids and glassy systems there has been considerable interest in dynamical heterogeneities which occur when certain regions of the sample have a higher mobility than the rest of the sample. Particle motion in these systems often occurs by means of correlated motion of a group of particles along a stringlike structure [1]. Additionally, studies of 2D liquids have found that the equilibrium structure of 5–7 dislocation pairs in the lattice form highly inhomogeneous distributions [2]. In this case, percolating networks of defects form strings or grain boundaries throughout the system, suggesting strong interactions between defects.

Most of the studies of heterogeneous motion in 2D and 3D have been performed on systems supercooled near the glass transition in simulations [3] and experiments [4–6]. In a recent study, a relatively simple system of a 2D dense monodisperse colloidal assembly showed an ordered to disordered transition as a function of density [5]. Here the colloids form a defect-free triangular lattice at high densities and disorder for lower densities. Near the disordering transition at the close packing density, collective heterogeneous motion of the particles appears, and the system consists of domains of sixfold coordinated particles, $n_c = 6$, surrounded by grain boundaries composed of strings of $n_c = 7$ and $n_c = 5$ dislocations. The formation of grain boundaries or defect condensation in 2D monodisperse liquids has also been observed in colloidal assemblies [6,7] and in dusty plasmas [8–10]. Additionally, numerical simulations of the 2D liquid phase [2] found that percolating grain boundaries of dislocations are an inherent equilibrium structure of 2D liquids.

Since the dislocations have a complex interaction with one another, one could expect complicated dislocation dynamics for systems with even a low density of dislocations. In the liquid state, creation and annihilation of dislocation pairs also occurs due to the thermally induced motions of the underlying particles. Although extensive numerical studies have shown that the defects form stringlike grain boundaries in 2D liquids [2] associated with heterogeneous particle motion, the fluctuations in the *defect density,* defined as the number of particles with $n_c \neq 6$, as a function of time has not been investigated. This measure would be easy to access in experiments on colloids and dusty plasmas where the individual particles can be directly imaged. It is not known how the density of $n_c \neq 6$ particles would fluctuate with time as the ordering transition is approached from the disordered side. Since in the liquid phase defects are strongly correlated, forming clumps or grain boundaries [2,8], it is likely that the creation or annihilation of defects will also be highly correlated, producing $1/f^{\alpha}$ fluctuation spectra. Conversely, if the defects appear and disappear in an uncorrelated manner, such as may occur at high temperatures, a white noise spectra would arise. We also wish to connect the formation of strings of defects with the appearance of correlated particle motion along 1D strings.

In this work we show that for 2D systems which form triangular lattices at low temperatures, for increasing temperature there is a transition from a nondefected regime to a defected regime where there is a proliferation of defects. Here we do not attempt to examine the nature of the disordering transition, such as whether there is an intermediate hexatic phase [11]. Instead, we concentrate on the motions of the particles and the defect fluctuations in the liquid phase. Close to the disordering transition, in the defected regime, the system consists of regions of particles with sixfold order surrounded by strings or grain boundaries of fivefold to sevenfold defects, and there are few free dislocations [2,8]. In this region, collective particle motion occurs in 1D strings associated with creation and annihilation of the defects. For a series of fixed temperatures we monitor the time dependence of the defect fluctuations. Near the disordering transition, the defect density shows large fluctuations with $1/f$ spectra. For increasing temperatures, the defect density increases and the stringlike structures of the defects and the 1D stringlike motion of the particles are lost. At the same time, the low frequency noise power increases and the spectrum crosses over to a white noise signature, indicating the loss of correlations in the creation or annihilation of defects with the concurrent loss of correlation in the particle motion.

We numerically study 2D systems at finite temperature using Langevin dynamics. We consider monodisperse particles in a sample with periodic boundary conditions and have investigated two types of interactions. Most of the results presented here are for particles interacting via a Yukawa or screened Coulomb interaction potential, $V(r_{ij}) = (Q^2/|\mathbf{r}_i - \mathbf{r}_j|)\exp(-\kappa|\mathbf{r}_i - \mathbf{r}_j|)$. Here $\mathbf{r}_{i(j)}$ is the position of particle $i(j)$, Q is the particle charge, $1/\kappa$ is the screening length, and we use $\kappa = 2/a_0$, where a_0 is the lattice constant. We choose to study this interaction since the quantities we study can be accessed in systems such as 2D colloidal assemblies and dusty plasmas, in which the particles interact via a screened Coulomb interaction. We also consider particles interacting via a logarithmic potential, $V(r) = \ln(r)$, which we treat with the summation method of Ref. [12]. This interaction can represent vortices in thin-film type-II superconductors.

The equation of motion for particle i is

$$\frac{d\mathbf{r}_i}{dt} = -\sum_{j\neq i}^{N_c} \nabla_i V(r_{ij}) + \mathbf{f}_T, \tag{1}$$

where $\mathbf{f}_T$ is a randomly fluctuating force due to thermal kicks with $\langle \mathbf{f}^T(t)\rangle = 0$ and $\langle \mathbf{f}^T(t)\mathbf{f}^T(t')\rangle = 2k_B T\delta(t - t')$. We initially start from an ordered triangular configuration, and fix the temperature $\mathbf{f}_T$ for 10^6 time steps before we begin to take data. We note that previous simulations have shown that extremely long time transients can arise near the order to disorder transition of up to 2×10^6 time steps for system sizes of 36864 particles [2]. In this work we limit ourselves to system sizes of $N_c = 2000$ or less, so that 10^6 time steps is adequate for equilibration. In addition, histograms of the time series of the defect density are Gaussian, which is further evidence that our systems are equilibrated. We measure temperature in units of T/T_d where T_d is the temperature at which the first free dislocations appear.

In Fig. 1(a) we show the Voronoi (or Wigner-Seitz) construction of a system of particles with a screened Coulomb interaction for $T/T_d = 1.04$. An individual particle is located in the center of each cell. If a particle has six nearest neighbors, $n_c = 6$, then the polygon has six sides. In Fig. 1 particles with $n_c = 6$ are white, $n_c = 5$ are dark gray, and $n_c = 7$ are black. We analyze a series of such images at fixed $T/T_d = 1.04$. We find that 0.229 of the particles are defected and that most (94.5%) of the defects are part of a cluster or condensate rather than free. In some regions, the defects form strings or grain boundaries. There are also a small number of free disclinations present, indicating that we are in the liquid phase rather than a hexatic phase, according to 2D continuous melting theories. This agrees with previous observations of clustering of defects into string structures and small numbers of free disclinations in the dense liquid regime close to the ordering transition in simulations [2] of 2D liquids

(a)

(b)

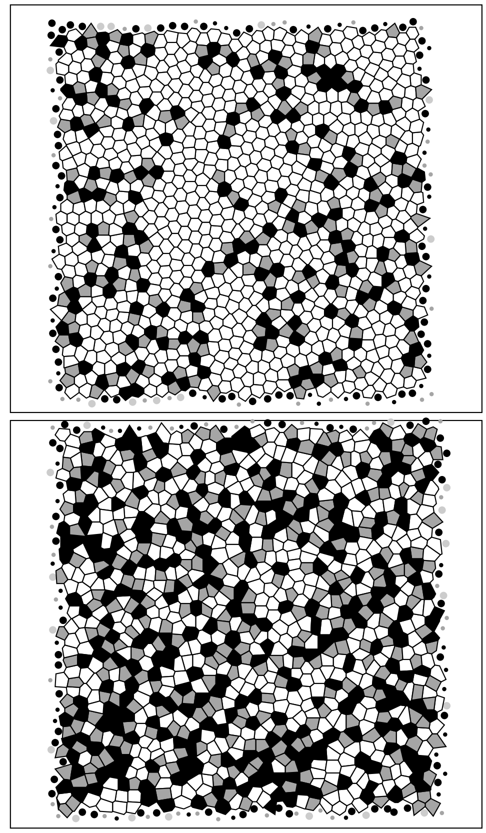

FIG. 1. The Voronoi constructions for a 2D system of particles interacting with a screened Coulomb potential. A particle is centered at each polygon and the polygon color is as follows: $n_c = 6$, white; $n_c = 5$, dark gray; $n_c = 7$, black. (a) $T/T_d = 1.04$; (b) $T/T_d = 7.0$.

and experiments on dusty plasmas [8]. In Fig. 1(b) we show the same system at a higher temperature, $T/T_d = 9$. Here the number of defects is higher with most of the defects again predominantly appearing in clusters; however, there is no stringlike characteristic to the clusters. Instead, the defects form clumps.

In Fig. 2(a) we show the positions of the particles (black dots) and trajectories (lines) over 10 000 time steps for the same system in Fig. 1(a). The motion is heterogeneous, and in the defect-free regions the particles do not move. In the moving areas the particles translate by a, and there is some tendency for the motion to occur along stringlike paths. We also often find motion occurring in a circular path with an immobile particle in the center. Similar 1D stringlike motion and rotations were observed in experiments on dense colloidal liquids [5]. If we take trajectories for longer times, eventually all the particles take part in some motion. In Fig. 2(b) we show the positions and the trajectories for the high temperature phase in Fig. 1(b) for 2000 time steps. Here the motion occurs everywhere with no evidence for correlated stationary regions. We find that even for very short times, there are no correlations in the motion.

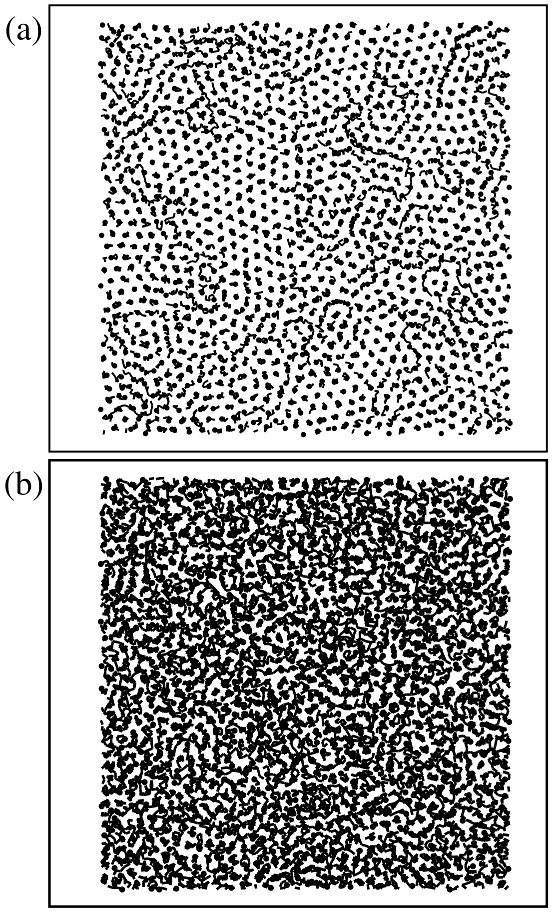

FIG. 2. Particle positions (black dots) and trajectories (black lines) for a fixed period of time for (a) $T/T_d = 1.04$ and (b) $T/T_d = 7.0$.

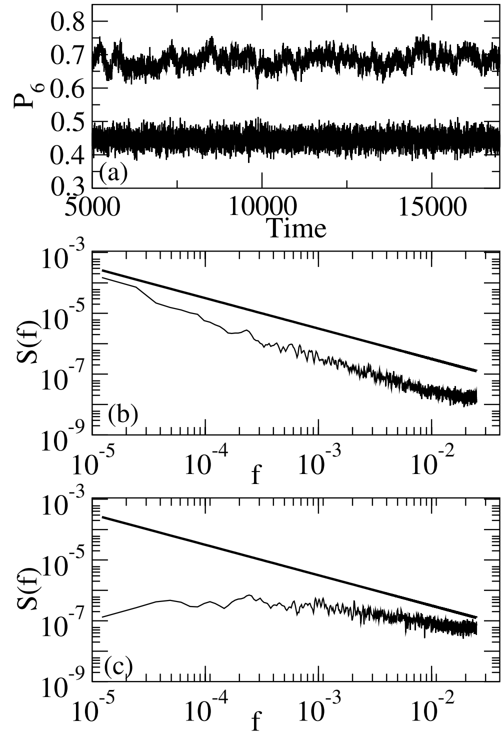

FIG. 3. (a) A portion of the time series of the fraction of $n_c = 6$ particles, $P_6(t)$, for (upper curve) $T/T_d = 1.04$ and (lower curve) $T/T_d = 7.0$. (b) The power spectrum for the time series at $T/T_d = 1.04$. The solid line indicates a slope of $1/f$. (c) The power spectrum for the time series at $T/T_d = 7.0$ along with a $1/f$ line.

We next consider a way to characterize the behaviors of the system at the different temperatures by measuring the fluctuations of the defect density as a function of time. We compute the defect configuration and density every 20 time steps for 20 000 frames and obtain a time series of the defect density for several temperatures from $T/T_d = 1.04$ to 10. We have considered a variety of sampling rates and find consistent results. In Fig. 3(a) we show a portion of the time series of the fraction of $n_c = 6$ particles, $P_6(t)$, for $T/T_d = 1.04$ (upper curve) and $T/T_d = 7.0$ (lower curve). For $T/T_d = 1.04$ the fluctuations show long time variations, while for the higher temperature the fluctuations are very rapid. In Fig. 3(b) we plot the power spectrum $S(f)$ of $P_6(t)$ for $T/T_d = 1.04$, which fits well to a $1/f^\alpha$ scaling over more than three decades with the best fit $\alpha = 1.04$, close to $1/f$ noise. As the temperature increases, the spectrum changes from $1/f$ to white noise ($\alpha = 0$) for low frequencies. The small frequencies correspond to the long time correlations in the system; thus, the defect creation/annihilation correlation times decrease at higher temperatures. In Fig. 3(c), the power spectrum for $T/T_d = 7.0$ is white with $\alpha = 0$. We have also considered $P_6(t)$ for $T/T_d < 1.0$ where there are a small number of bound dislocation pairs. Here we find white noise with small amplitude fluctuations.

The magnitude of the noise power, obtained by integrating $S(f)$ over the first octave of frequencies, is related to the defect density and the disordering transition. In Fig. 4 we show P_6 as a function of T and the corresponding noise power S_0. The peak in S_0 coincides with the onset of the defect proliferation. For increasing T, the noise power falls and saturates when the spectrum becomes white. There is a finite S_0 for $T/T_d < 1.0$ since pairs of bound dislocations can still be thermally created. We have repeated the simulations for particles interacting with a logarithmic potential and again find a noise power peak coinciding with the defect proliferation (insets of Fig. 4) and a $1/f$ power spectrum just above T_d.

We have also considered a sudden quench from $T/T_d > 1.0$ to $T/T_d < 1.0$. Here the system is out of equilibrium, and approaches equilibrium through defect annihilation. In Fig. 5 we show the particle motions after a quench from $T/T_d = 1.04$ to $T/T_d = 0.2$. The number of defects decreases rapidly to a saturation point where a few defects or grain boundaries remain, in agreement with previous quench simulations [2]. The motion of the particles corresponds to the defect annihilations. As seen in Fig. 5, the motion during the defect annihilation process has the same stringlike nature as the equilibrium $T > T_d$ motion

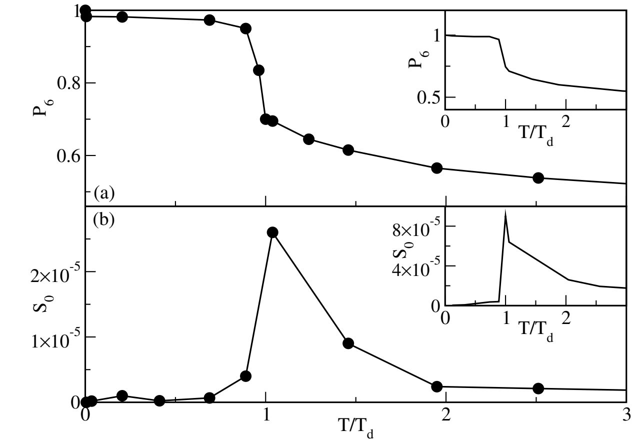

FIG. 4. (a) The fraction of sixfold particles P_6 vs T for a system of particles with a screened Coulomb interaction. (b) The integrated noise power S_0, obtained from $S(f)$, vs T for the same system in (a). Insets: P_6 and S_0 vs T for a system with logarithmic interactions.

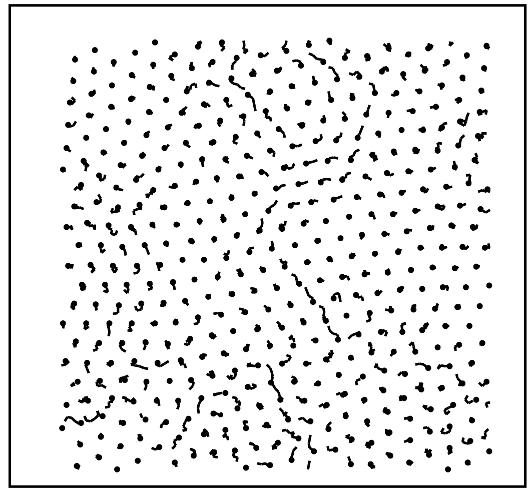

FIG. 5. The particle positions (black dots) and trajectories (black lines) for a system quenched from $T/T_d = 1.04$ to $T/T_d = 0.3$.

near the defect proliferation transition. This suggests that the heterogeneous particle motion is produced by the motion of defects, particularly the correlated creation or annihilation of these defects.

We briefly compare our results to recent experiments. Experiments performed in the dense liquid phase of a 2D dusty plasma show intermittent heterogeneous motion, which occurs in avalanchelike pulses with a power law distribution in the number of fast events [13]. There is also numerical evidence that certain models with avalanche-type dynamics produce $1/f$ noise signatures in the power spectra [14]. In our system, since the particle motion occurs near the defect clusters, we expect the fluctuations in the particle motion to be correlated with the fluctuations of the clusters. Thus, if the clusters show power law fluctuations in the spectra, as observed in Ref. [13], the particles should as well. Also, in recent experiments measuring the local structural ordering in colloidal systems exhibiting heterogeneous motion, particle motion occurs in the most disordered regions [15], which agrees well with our picture that the heterogeneous particle motion occurs where there are defects present.

In summary, we have proposed a new method for examining dynamical heterogeneities in a liquid by measuring the fluctuations in the topological defect density. We find that near the onset of defect proliferation, the particle motion is heterogeneous and the defects cluster together into grain boundaries or strings in agreement with previous work. The defect fluctuations in this phase have a $1/f$ character and a large noise power. For higher temperatures the heterogeneities are lost and the fluctuation spectrum is white. We have also examined the defect annihilation after a quench from the disordered phase to the ordered phase and find that the particle motion also shows heterogeneities as the defects are annihilated. We have considered systems with screened Coulomb and logarithmic interactions and find similar behavior. Our predictions can be easily tested in 2D dense colloidal liquids. It would also be interesting to study the fluctuations of defects in 3D. This would be experimentally possible in 3D colloidal assemblies using confocal microscopy.

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