Coexistence of hexatic and isotropic phases in two-dimensional Yukawa systems

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Abstract  
We performed Brownian-dynamics simulations on the melting of two-dimensional colloidal crystals in which particles interact via a Yukawa potential. A stable hexatic phase was found in the Yukawa systems, but we also found that the melting of Yukawa systems is a two-stage melting, which is inconsistent with the Kosterlitz–Thouless–Halperin–Nelson–Young (KTHNY) theory. A two-phase coexistence region between the stable hexatic phase and the isotropic liquid phase was found. The behavior of point defects in the coexistence region is very complicated. The emergence of some unstable free disclinations and grain boundaries was a characteristic representative of the isotropic liquid phase, and a large number of free dislocations indicated the existence of a hexatic phase. This indicates the existence of a phase of hexatic–isotropic liquid phase coexistence. The big picture in the melting of a two-dimensional Yukawa system is that first the system undergoes a transition induced by the formation of free dislocations, then it goes through a phase coexistence, and finally, it comes into an isotropic fluid phase. This melting process is consistent with experiments and simulations.  
(Some figures in this article are in colour only in the electronic version)

1. Introduction  
In contrast to the case of melting in three-dimensional (3D) systems, it has by now been well established that in two-dimensional (2D) crystals, long-range positional order does not exist due to long-wavelength fluctuations [1]. Despite this, in a 2D crystal, there exists a special kind of long-range bond-orientational order. A microscopic scenario of 2D melting has been posited in the form of the Kosterlitz–Thouless–Halperin–Nelson–Young (KTHNY) theory [2–5]. The KTHNY theory predicts a new phase, the so-called hexatic phase, that exists between the solid and liquid phases in 2D melting [6].  
According to the KTHNY theory, the melting of a 2D system is a two-stage transition. In the first stage we start with the 2D system in a solid phase, which has both quasi-long-range positional order and long-range bond orientation order; the system then undergoes a continuous transition and becomes a hexatic phase with short-range positional order and quasi-long-range orientational order. In the second stage, another continuous transition drives the hexatic phase to an isotropic liquid phase in which both positional and bond-orientational order have short ranges.  
The KTHNY theory predicts the unbinding of topological defects to break the symmetry in the two-stage transitions. The physical driving force behind the two-stage transitions is the dissociation of bound defect pairs, specifically pairs of dislocation (solid \(\rightarrow\) hexatic) and pairs of disclinations (hexatic \(\rightarrow\) liquid). Two-dimensional systems are characterized by two different order parameters, namely, the orientational and translational order, corresponding to the two types of topological defects. Dissociation of the dislocation pairs causes the translational symmetry to be broken, and dissociation of free dislocations melts, causing the orientational symmetry to be broken.  
In recent years, a large number of experiments and computer simulations have indicated that there is, indeed, a two-stage melting scenario in 2D systems as prescribed by the KTHNY theory. The hexatic phase has been observed in...
colloidal crystals [7, 8], magnetic bubble arrays [9], and the freestanding liquid-crystal films [10]. The hexatic phase not only appears in 2D systems, but also in 3D systems, such as in layered smectic liquid crystals [11], dense solutions of DNA [12, 13] and high temperature superconductors [14].

Although the KTHNY theory is currently preferred, a different theoretical approach, evoking grain-boundary-induced melting, was a first-order transition suggested by Chui [15]. Using a low-density approximation, Chui found that the grain boundaries might be generated before the dislocations unbind when the core energy of dislocations is sufficiently small ($E_c < 2.84 k_BT$); thus, he predicted a first-order transition. One may note that the condensation of geometrical defects is also a first-order transition [16, 17].

Several computer simulations on 2D melting have favored a first-order phase transition or, at most, a weak first-order transition. In these simulations, the hexatic phase was not observed. It was argued that this transition might depend on the specific properties of systems being studied such as their inter-particle potential. The transition seems to be first-order in hard-core systems [18], but to be second-order behavior in dipole–dipole interactions [19]. Some simulations for Lennard-Jones systems have discovered that the hexatic phase is metastable [20]. In the case of Yukawa systems, Naidoo and Schnitker found that the defect topology was very complicated, and the predictions of the KTHNY theory were violated [21].

The hexatic phase has indeed now been observed in 2D colloidal crystals [22–24]. Two-dimensional colloidal particles are a good experimental system for studying the 2D melting. The advantage of such systems is that the charged colloidal crystals in aqueous suspensions are observable under the microscope where, due to the particle size, the individual colloid motions can be directly observed. At the same time, the particles are still small enough to perform thermally driven motion and can be considered as a statistical ensemble in thermal equilibrium. In 1987, Murray et al. experimented on these colloidal suspensions of simple polystyrene spheres, and the results indicated that the two-step melting was the same as prescribed by the KTHNY theory. However, the experimental evidence for 2D colloidal suspensions remains a hotly debated subject of controversy. Tang et al. also observed the two-step melting process, but they indicated that the melting was first-order, which is consistent with the picture developed by Chui.

One commonly encounters the following essential question: does the melting of 2D screened coulomb colloidal systems (or Yukawa systems) follow the scenario of KTHNY theory? In [21], the answer is that they found an intermediate phase, but not a true hexatic phase, in their simulations. On the other hand, Murray et al. observed the hexatic phase in the screened Coulomb system. A similar result has been obtained by Tang et al., but it is worth noticing that although the hexatic phase was observed, the possibility of the coexistence of the hexatic phase with the isotropic liquid phase could not be ruled out in their experiments [25].

In this paper, we study the melting of 2D charged colloidal crystals, where we present a Brownian-dynamics simulation on a 2D Yukawa system. We focus on the existence of the stable hexatic phase and the coexistence of an isotropic–hexatic phase. Furthermore, we study the defect structure and compare it with former results from experiments and simulations. In Murray and Van Winkle’s work, the two-step KTHNY melting was found by a correlation length analysis, however, the paired dislocations in the solid phase and the free dislocations in the hexatic phase were not found. Tang et al. have also obtained a similarly contradictory result. By Voronoi construction analysis, we find that the paired dislocations exist in the solid phase and only a few unstable free dislocations are found in the hexatic phase.

The organization of this paper is as follows. In section 2, we describe the Brownian-dynamics simulation methods. The results are presented and discussed in section 3. Here, we calculate the orientational and pair correlation functions, and then examine the two-phase coexistence regions and defect topology. In the last section, we summarize our results.

2. The model

The well-known DeJaegher–Landau–Verwey–Overbeek (DLVO) theory gives us a good description for the effective pair interaction of the one-component model in the colloidal systems. The DLVO potential consists of an electrostatic repulsion and a van der Waals attraction. Normally this is managed in a Yukawa or screened Coulomb form, which only retains the electrostatic part [26]. For a dilute charged stabilized colloidal system in which many-body interactions can be ignored [27–29], a pairwise Yukawa potential is defined as

$$V(r) = U_0 \frac{\sigma}{r} \exp \left( -\frac{r - \sigma}{\lambda} \right),$$

where $U_0$ is the energy and $\sigma$ is the scale length. The screening parameter $\lambda$ describes the ‘softening’ of the particles: when $\lambda$ increases from zero to infinity, the cores of interacting particles change from very soft to extremely hard [30–32]. In our simulations, we assumed that the Yukawa potential between the particles is very soft (the screening parameter $\lambda = 8$).

Now we briefly describe the standard Brownian-dynamics simulation, which is based on a finite difference integration of the irreversible Langevin equations. The equation of motion for an individual colloid $i$ is

$$\xi \dot{r}_i (t) = F_i (t) + R_i (t),$$

where $i = 1, \ldots , N$ labels the $N$ particles, $\xi$ is the friction coefficient, and $\xi = 1$ in simulation units. $R_i (t)$ is the Langevin random force of the solvent, and $F_i (t)$ is the total inter-particle force on particle $i$. Here the hydrodynamic interaction is ignored. The finite difference integration is

$$r_i (t + \Delta t) = r_i (t) + F_i (t) \Delta t + (\Delta r)_R + O(\Delta t)^2,$$

where $(\Delta r)_R$ is a random displacement sampled from a Gaussian distribution of zero mean and variance

$$(\Delta r)^2_R = 4 D_0 \Delta t.$$

Here $D_0 = k_B T/\eta$ is the short-time diffusion coefficient, $k_B$ is the Boltzmann constant and $T$ is the temperature. The
coefficient in equation (4) is 2 in one-dimensional systems. We used reduced units such that \( U_0 = 1, \sigma = 1, \) and \( \rho = N/V = 1. \) In all simulations, we tuned the reduced temperature \( T^* = k_B T / U_0 \) and the other parameters \( \sigma, U_0, \rho, \) and \( \lambda \) were fixed. We used a periodically repeated rectangular simulation box with \( N = 2500 \) particles and started from a triple lattice. The cutoff \( r_c \) was set as 4.1. We considered only a triangular lattice since it is the most densely packed lattice in two-dimensions and is thus favored by nature. The scale of the simulation box is in the ratio \( 2: \sqrt{3} \) with the length of the \( x \)-axis of our simulation box 55.836 in order to minimize the finite-size effects. One can find more details about this simulation in [30].

To characterize the translational order, we calculate the pair correlation function. It is defined by

\[
g(r) = \rho^{-2} \left\{ \sum_{i \neq j} \delta(r_i) \delta(r_j - r) \right\},
\]

where \( \rho \) is the 2D particle density. The bond-orientational function is

\[
g_b(r) = \langle \psi^*_b(r) \psi_b(r' - r) \rangle,
\]

where \( \psi_b(r) \) is the local bond-orientational order parameter

\[
\psi_b(r_m) = \frac{1}{N_b} \sum_{i=1}^{N_b} e^{i \vec{r}_m \cdot \vec{r}_i}
\]

Here \( N_b \) denotes the number of the nearest neighbor of the \( m \)-th particle, and \( \vec{r}_m \) is the angle between the bonds of particles \( i \) and \( j \) with an arbitrary, but fixed, reference axis. According to the KTHNY theory, the bond-orientational function \( g_b(r) \) will have an algebraic decay in a hexatic phase, and an exponential decay in the liquid phase. Before the dislocation unbinding transition occurs, the hexatic phase is anisotropic and the bond-orientational correlation function is

\[
g_b(r) \propto r^{-\eta_b(T)}
\]

where \( \eta_b = \frac{18k_B T}{\pi K_A} \). \( K_A \) is called the Frank constant which describes the coupling constant related to distortions of the bond-angle field. The KTHNY theory predicts that the disclination unbinding transition is also continuous, and that it occurs when the value of the Frank constant falls below \( 72k_B T/\pi \).

3. Results and discussions

In order to study the phase behaviors and identify the existence of the hexatic phase, we computed the pair distribution functions and bond-orientational correlation functions. As there are large fluctuations near the critical point, it is difficult to obtain the phase boundaries precisely, however, what we are interested in is the process of melting transition and phase behavior in the middle of the intermediate region. In our simulations, it takes a sufficient amount of time to reach equilibrium when a two-stage continuous melting transition occurs. Normally, the simulation reaches equilibrium after about \( 50t_B \), and the simulation results are gathered within the range of \( 10t_B \) (\( t_B = \sigma^2 \xi / U_0 \)).

![Figure 1](https://example.com/figure1.png)

**Figure 1.** The pair correlation function \( g(r) \) and its FFT analysis in Yukawa systems with \( N = 2500 \) and \( \rho = 1.0 \) for different temperatures, (a) \( T^* = 0.200 \), (b) \( T^* = 0.500 \), (c) \( T^* = 0.605 \), (d) \( T^* = 0.630 \).

3.1. The translational and orientational order

Figures 1(e)–(h) show the pair correlation functions \( g(r) \) for different temperatures. In this setting (the 2D crystal), there is only an orientational symmetry; here there is no true translational order as it can never really be ‘long-range’. The systems are in the solid phase for \( T^* = 0.200 \) and 0.500 and the oscillations of the pair distribution function persist over the entire range. It was found that the translational correlation function decays algebraically. The behavior was different, however, for \( T^* = 0.605 \) and 0.630, and the oscillations died out quickly, which indicates that short-range translational order does exist in the system.

The fast Fourier transforms (FFT) of the pair distribution functions are also presented in figures 1(a)–(d). At low temperature (solid phase), the first peak that indicates the periodic structure of the system is very sharp. The second and third peaks also are obvious (see figures 1(a), (b)). As we increase the temperature, the third peak becomes unnoticeable. For \( T^* = 0.605 \), the second peak is not obvious, but the first peak was lower than it was for the systems in the solid phase. At the temperature \( T^* = 0.630 \), the system is in the liquid phase. Here, none of the peaks were obvious and the first peak was much lower than it was for the system in the solid phase (see figures 1(c), (d)). With the increasing of temperature, it show that the first peak moves considerably in figures 1(a)–(d). Due to the crystal lattice being periodic, the first peak is very sharp in the solid phase. When the system melts into liquid phase, the crystal lattice is broken. So it indicates that the quasi-long-range position order in the solid phase becomes short-range in the liquid phase.
In figure 2, we plotted the simulation results of the bond-orientational functions $g_6(r)$ for different reduced temperatures. When $g_6(r)$ does not decay, it means that the system is in the solid phase with a long-range bond-orientational order. When the reduced temperature rises to near the disclination unbinding regions, $g_6(r)$ decays algebraically with an exponent near $1/4$, which implies the existence of the hexatic phase as predicted by the KTHNY theory. As in our simulations, it is easy to see the hexatic phase where the bond-orientational functions decay algebraically with $\eta(T^*)$ near to $1/4$ (see $T^* = 0.605$ in figure 2). With further increases in temperature, the system becomes a disordered liquid and $g_6(r)$ decays exponentially. It should be noted that $g_6(r)$ decays algebraically with an exponent 0.54 at $T^* = 0.61$, which is faster than $1/4$ from the KTHNY theory.

To locate the solid-to-hexatic phase transition temperature, we use the 2D Lindemann melting criterion introduced by Bedanov and Gadiyak [33]. As the mean square displacement $\langle u_j^2 \rangle$ diverges in a 2D crystal, they suggested the Lindemann parameter

$$\gamma_m = \frac{\langle (u_j - u_{j+1})^2 \rangle}{a^2}$$

where the indices $j$ and $j + 1$ refer to neighboring particles. At the melting point, these authors found a critical value $\gamma_m^c = 0.033$. Here we measure the Lindemann parameter at different temperatures. At the melting point $T_m^*$, the Lindemann parameter grows sharply, indicating a vanishing of the positional symmetry. In our simulations, a sharp growth of $\gamma_m$ is observed at $T_m^* = 0.520$ (see figure 3).

3.2. The coexistence of hexatic and isotropic phases

Indeed, there exists an algebraic decay of the bond-orientational correlation function $g_6(r)$. However, we observed that $\eta_6$ is larger than $1/4$ (see figure 2), which is not consistent with the prediction of KTHNY theory. In light of this, it has been conjectured that a coexistence of hexatic and isotropic phases may appear; this was pointed out by von Grünberg et al [25]. The coexistence region is important evidence of a first-order transition [30], however, in order to identify this coexistence we need to work carefully.

Figure 4 plots the trajectory of the particles at $T^* = 0.609$ for a section of the simulation box during $6\tau_B$. The existence of solid-like and liquid-like patches in figure 4 has been interpreted as evidence for phase coexistence, however, a similar path also appears in the purely hexatic phase. Given this, we have to present a more reliable and measurable method to identify whether the system is the pure hexatic phase or in a two-phase coexistence.

In order to confirm the phase coexistence, we used the recipe suggested by Strandburg et al [34]. The angular susceptibility $\chi_6$ for different length scales is defined by

$$\chi_6 = \left\langle \frac{1}{N} \sum_i \frac{1}{N_i} \sum_n e^{i l \cdot \theta_{in}} \right\rangle^2$$

where the sum on $l$ is over all particles, the sum on $n$ is over the nearest neighbors, $N_l$ is the number of nearest neighbors.
of particle \( l \), and \( N \) is the number of particles in the system. In the solid phase, the angular susceptibility \( \chi_6 \) is large due to the long-range order. On the other hand, \( \chi_6 \) is small in the fluid phase. If the system exhibits a two-phase coexistence, one might expect that the distributions of \( \chi_6 \) for sufficiently small length scales could be modeled by a combination of solid and fluid distributions. However, in the case of a homogeneous hexatic phase, varying the size of the subsystems should not lead to any qualitative changes in the distribution of \( \chi_6 \) (as shown in [19] for systems with dipole–dipole interactions).

We calculate \( \chi_6 \) in our simulations for many different length scales by dividing the system of 2500 particles into subsystems containing an average of 128, 64, 16, and 4 particles. These subsystems are also periodically repeated as rectangular simulation boxes. \( \chi_6 \) is calculated every 100 passes. If the system is homogeneous, varying the size of the subsystems should not lead to any qualitative changes in the distribution of \( \chi_6 \). Contrarily, for a two-phase region, the probability distribution of \( \chi_6 \) at a sufficiently small length scale could be modeled by a curve with two peaks, reflecting a combination of two-phase distributions.

Figure 5 shows the distributions of \( \chi_6 \) for subsystems of 128, 64, 16, and 4 particles with different temperatures. In the region of low temperature or crystal phase, there is no qualitative change (see figure 5(a)). The probability distributions of \( \chi_6 \) always remain singly peaked as the size of the subsystems is varied. This indicates that the system is in the homogeneous solid phase for low temperatures. In figures 5(b) and (c), we presented the probability distribution of \( \chi_6 \) at \( T^* = 0.600 \) and \( T^* = 0.602 \) where a power law decay of \( g_6(r) \) was found. By varying the size of the subsystems, the probability distribution of \( \chi_6 \) has a single peak, which means that there is only a homogeneous phase and the two-phase coexistence can be ruled out. One can conclude that there is a stable hexatic phase at \( T^* = 0.600 \) and 0.602.

In figures 5(d) and (e), the probability distributions of \( \chi_6 \) at \( T^* = 0.605 \) and 0.609 could be modeled by a curve with two peaks for \( n = 64 \) and 128. This clearly demonstrates the
two-phase coexistence since the peaks reflect a combination of solid and fluid distributions. It should be noted that the $g_6(r)$ decays algebraically at both temperatures.

Figure 5(f) depicts the probability distribution of $\chi_6$ at $T^* = 0.630$. In the fluid phase, the distributions peak near zero. Varying the size of the subsystems, the distribution of $\chi_6$ cannot lead to any qualitative changes so we can conclude that the 2D Yukawa systems melt to a pure liquid phase.

3.3. Topological defects

In the KTHNY theory, the 2D melting is caused by the unbinding of topological defect pairs [3–5]. First, dislocation pairs unbind and then, the disclination pairs unbind. Since dislocations are associated with an additional half row of atoms, they can be quite effective at breaking up translational order. However, dislocations are less disruptive of orientational correlations, and the disclination breaks the orientational order. Thus it is important to determine whether these defects are indeed important in the melting process. Murray et al observed that islands of sixfold coordinated particles are surrounded by a network of grain boundaries of fourfold and sevenfold coordinated particles in the fluid phase [14]. In the hexatic phase, they failed to find free dislocations, but these grain boundaries did not completely disappear. The neighboring grains began to orient with respect to each other. In the solid phase, they also failed to find paired dislocations.

Normally, the defect structure can be pictured by using Voronoi polygons where we identify disclinations as particles with five or seven nearest neighbors. A Voronoi polygon is defined as the boundary of a region enclosing a particle, which is closer to every point of the region than to any others [35, 36]. A disclination is located at a particle with five or seven vertices in its Voronoi polygon. In a perfect triangular colloidal crystal, all particles are sixfold. Disclinations are described as particles having five and seven neighbors. A disclination with positive unit strength is located at a particle with five near neighbors and that with negative unit strength is located near seven neighbors. A dislocation may be viewed as a tightly bound pair of disclinations. In the solid phase, dislocations are bound very strongly to a potential that increases as the square of the separation.

We studied the topological configurations by using Voronoi cell pictures to identify the position of the defect. We did this by showing the positions of disclinations for a single configuration after a long run. For a single configuration in a solid phase at $T^* = 0.500$ (see figure 6(a)), nearly all of the dislocations occur in pairs. Figure 6(b) shows the defect structure in an intermediate region between the isotropic liquid and the solid at $T^* = 0.600$. It was found that there exist free dislocations (see arrow 1), which supports the KTHNY theory that melting is the unbinding of dislocation pairs.

In figure 6(c), the system is clearly in an isotropic liquid phase. There are a large number of defects and free disclinations, and the defect structure is very complicated. Clustering of dislocations is observed [21]: there exists a pair of fivefold coordination (5-coordination) bindings with an 8-coordination (shown in figure 6(c) by arrow 1) or a pair of 7-coordination bindings with a 4-coordination (in figure 6(c) shown by arrow 2). These topological defects are unstable, however, and vanish very quickly.

The distributions of topological defects in the two-phase coexistence region are shown in figure 7. There are a large number of free dislocations, and only a few free disclinations. Notice that the translational order is broken by the emergence of free dislocations. Since the number of disclinations is not enough to break up the bond-orientational order, the bond-orientational function still decays algebraically at the two-phase coexistence region. We observe that defects are probably a characteristic of grain boundaries, that is, the clusters consisting of dislocations and dislocations pair up, or
small dislocations form a loop, such defects were also shown in Tang's work. The appearance of grain boundaries leads to the first-order transition suggested by Chui [15].

Figure 8 plots the fractions of 6-coordinated, 5-coordinated, and 7-coordinated particles. At low temperature, the number of defects is very small. When the temperature is above 0.500, $N_6/N$ decreases rapidly. At $T^* = 0.605$, almost 20% of particles are attached to the defects, which is consist with the results in [24]. As the defect fraction rises above 30%, the system melts into a liquid phase. At the liquid phase, the number of 5-coordinated particles is much more than the number of 7-coordinated particles due to the emergence of the 8-coordinated particles and the effect of boundaries.

We have observed the mechanism of defects in the 2D Yukawa system [37]. At low temperature, the paired dislocation is formed or annihilated. The formation of a binding dislocation pair can be viewed as the simultaneous formation of two sevenfold coordinated particles and two fivefold coordinated particles from four sixfold coordinated particles. When the temperature rises to the hexatic phase, the dislocation pair dissociates. In the solid phase, the unbinding of dislocations is unstable, and these unbound dislocations will quickly bind. This is in contrast to the stable free dislocation found in the hexatic phase. Grain boundaries and unstable disclinations appears throughout the region of two-phase coexistence. Given this, one may conjecture that the isotropic liquid phase can be characterized by the existence of stable defect clustering.

4. Conclusions

In this paper, we performed Brownian-dynamics simulations to study the melting of 2D colloidal crystals with Yukawa interactions, and two-stage melting is found. The hexatic phase in melting of 2D charged colloidal crystals was indeed observed [23]. Moreover, the hexatic-liquid phase coexistence was observed as well. Such coexistence was also confirmed by Tang et al [24].

We calculated the pair correlation function and bond-orientational correlation functions. At low temperature, due to the quasi-long-range positional order in 2D systems, the oscillations of the pair correlation function persist over the entire range. On the other hand, the oscillations of the pair correlation function died quickly at higher temperature, and it was shown that the positional order becomes shot-range. By using the 2D Lindemann melting criterion, we found that the melting temperature is 0.530 ($\pm$0.01). An algebraic decay with $\eta$ near 1/4 of the bond-orientational correlation function was observed at the temperature 0.605 ($\pm$0.01). By ruling out the coexistence, we verified that this is a pure phase at temperatures between 0.530 and 0.605. As the bond-orientational correlation function decays algebraically, we concluded that the pure phase is a stable hexatic phase with the quasi-long-range bond-orientational order.

We found that the quasi-long-range bond-orientational order still exists in the coexistence region in finding the algebraic decays of the bond-orientational functions. The emergence of unstable free disclinations and grain boundaries is a characteristic representative of an isotropic liquid phase, and a large number of free dislocations is a characteristic representative of a hexatic phase. This indicates that there was indeed a coexistence of hexatic–isotropic liquid phases. In a word, the melting of 2D Yukawa systems is a two-stage melting. Firstly, the system first undergoes a transition induced by the formation of free dislocations, the system then goes through a phase coexistence, and finally moves into an isotropic fluid phase.

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