The influence of random pinning on the melting scenario of two-dimensional soft-disk systems

E. A. Gaiduk, Y. D. Fomin, E. N. Tsio, and V. N. Ryzhov
Institute for High Pressure Physics, Russian Academy of Sciences, 108840, Troitsk, Moscow, Russia

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We present the results of a computer simulation study of the melting scenario of two-dimensional soft-disk systems with potential $U(r) = \varepsilon(\sigma/r)^n$, $n = 12$ and $n = 1024$, both in the presence of random pinning and without it. The melting parameters have been determined from analysis of the behavior of equations of state, correlation functions of the orientational and translational order parameters, Young modulus and renormalization group equations. The transition points obtained from these criteria are in good agreement. We have shown that the systems melted in two stages - first-order hexatic phase to liquid transition and the continuous Berezinskii-Kosterlitz-Thouless type crystal-hexatic phase transition. Random pinning widened the hexatic phase, but left the melting scenario unchanged.

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I. INTRODUCTION

The problem of two-dimensional (2D) melting has been of great interest for more than 40 years already. However, even now there is no microscopic theory to satisfactorily describe the nature of two-dimensional melting, which remains among the oldest unsolved problems in condensed matter physics. In spite of a large number of publications there are still many controversies. While in three dimensional (3D) space melting is always first-order phase transition, there are several different scenarios of melting in 2D. The main reason of this difference is related to strong fluctuations in 2D compared to the 3D case. Peierls24 and Landau25 and later Mermin26 demonstrated that in 2D crystals there was no long-range translational order. The translational order of 2D crystals is in fact quasi long-range, i.e. the correlation functions decay algebraically. However, there is another type of long-range order in 2D crystals: orientational order which is the order in bonds between a particle and its nearest neighbors. At high temperatures a 2D system is in a usual state of isotropic liquid.

The Berezinskii-Kosterlitz-Thouless-Halperin-Nelson-Young (BKTHNY) theory of 2D melting is the most common. Within the framework of this theory 2D melting takes place in two stages. At the first stage a crystal transforms into a so called hexatic phase via continuous phase transition. This transformation is caused by dissociation of coupled dislocation pairs. Below we will refer to the temperature of dislocation pairs dissociation as $T_m$. This dissociation leads to a complete break of translational order: while in 2D crystals the translational order is quasi long-range, in the hexatic phase it becomes short-range. At the same time the orientational order changes from long-range in crystals to quasi long-range in the hexatic phase.

Dislocations themselves can be considered as bounded pairs of disclinations. These pairs also dissociate at higher temperature $T_i$ where the system transforms into isotropic liquid. Within the framework of the BKTHNY theory this transition is also a continuous one. The BKTHNY theory was confirmed by some experimental studies (see, for instance27–29, where systems of colloids in magnetic fields are studied).

However, 2D melting can also occur through first-order phase transition alone. In Ref.22 it was shown that at low energy of the dislocation core the dissociation of bound dislocation pairs was preempted by the proliferation of grain boundaries leading to the first-order melting transition. First-order transition takes place at dislocation core energy $E_c$ less than $2.84T_m$, where $T_m$ is obtained from equation (5) (see below). A similar mechanism leading to first-order transition was discussed in Refs.18,19. Since dislocation is a disclination dipole a bound dislocation pair can be considered as a disclination quadrupole. As shown in papers18,19 there is a critical value of disclination core energy below which the dissociation of disclination quadrupoles leads to the appearance of free disclinations and the transition of first-order to isotropic liquid. The dependence of the melting scenarios on the form of the potential was studied in Refs.20,21 within the framework of the density functional theory of crystallization. A unified approach to the description of first-order and BKT type melting of the solid phase within the framework of Landau’s theory of phase transitions was proposed in Refs.24,25.

Although the BKTHNY theory does not depend on the interaction potential, many experiments and computer simulations show that such dependence exists. It was widely believed that systems with short-range interaction potentials melted via first-order phase transition, while long-range interactions, for example, the Coulomb one, led to the BKTHNY melting scenario.

However, even for the simplest system of hard spheres the results are contradictory (see, for example26–29). This contradiction was partly resolved in the series of papers30–32, where based on the results of computer simulations it was proposed that melting took place as a two
stage process, but while transition from crystal to hexatic was a continuous one, the second transition from hexatic to isotropic liquid was a first-order transformation. This new melting scenario was experimentally confirmed in Ref.\textsuperscript{33}. In particular, Ref.\textsuperscript{34} presents a study of melting of soft disks (a system of particles interacting via inverse power potential $U \sim 1/r^n$) with a different $n$. It is shown that for small $n < 6$ the system melts in accordance with the BKTHNY theory, while for $n > 6$ the transition between the hexatic and liquid phases becomes a first-order one, i.e. melting follows the third scenario. Below we will call this scenario the BK one after Bernard and Krauth who introduced it.

The melting of systems with core-softened potentials was studied in Refs.\textsuperscript{35–40}. It was shown that the system had a complex phase diagram with different crystal structures, including a dodecagonal quasicrystal. Depending on the position on the phase diagrams, the different melting scenarios can take place, including first-order melting and the BK scenario. In Refs.\textsuperscript{41,42} computer simulations of a system of purely repulsive soft colloidal particles interacting via the Hertz potential and constrained to a two-dimensional plane were presented. This potential can be a reliable model for a qualitative description of behavior of soft macromolecules, such as globular micelles and star polymers. A large number of ordered phases was found, including the dodecagonal quasicrystal, and it was shown that depending on the position on the phase diagram, the system could melt through the first-order transition, in accordance with the BKTHNY theory as well as according to the BK scenario.

It should be noted that the region of stability of the hexatic phase is usually rather narrow. As a result this region can be almost invisible in experiments and computer simulations. In this case transition from crystal to liquid may be mistaken for a first-order one. In order to make the situation clearer it is necessary to enlarge the region of stability of the hexatic phase. One of the methods to do so is to introduce random pinning into the system, i.e. to make some particles immobile and fix them in some random positions, including interstitial lattice sites. As it was shown theoretically\textsuperscript{43–46}, the BKTHNY melting scenario persisted in this case. Random pinning almost did not affect the transition between the hexatic phase and isotropic liquid, while the solid phase could be completely destroyed by high pinning fractions. In the latter case the stability range of the hexatic phase increases due to moving the border of the solid phase to higher densities on the equation of state. This result was confirmed in experiments and simulations of 2D melting of super-paramagnetic colloidal particles with quenched disorder\textsuperscript{15,16}. In Ref.\textsuperscript{47} it was shown that the BK scenario also persisted with random pinning. An even more interesting result was obtained in Ref.\textsuperscript{48} where it was demonstrated that random pinning could transform first-order melting into the BK scenario.

It should be specially mentioned that in the article by Qi and Dijkstra\textsuperscript{49} the opposite case was considered where a random fraction of particles was pinned on the regular lattice sites of the crystal phase. In this case the pinned particles stabilized the crystal lattice and consequently decreased the range of the hexatic phase.

Below we will consider both the systems with and without random pinning. We will also call systems without pinning pure systems.

In the present work we study the melting behavior of two soft disk systems: with $n = 12$ and with $n = 1024$. In order to determine the melting scenarios more accurately we introduce random pinning into the system. It allows us to study the properties of the hexatic phase itself, for example, the diffusion coefficient of the hexatic phase. Moreover, we employ many different methods to determine phase boundaries, including equations of state, correlation functions, Young modulus and renormalization group equations. This allows us to compare different approaches in order to determine phase boundaries in 2D melting.

II. SYSTEM AND METHODS

The soft disk system studied in this paper is defined by the interaction potential:

$$U(r) = \varepsilon \left(\frac{\sigma}{r}\right)^n. \quad (1)$$

Parameters $\sigma$ and $\varepsilon$ can be used as the scales of length and energy respectively. Based on these scales one can construct the units of all other quantities of interest. For instance, the density can be written as $\rho^* = \rho \cdot \sigma^2$. Below we will use only these reduced units omitting ($^*$).

Two values of softness parameter $n$ are considered: $n = 12$ and $n = 1024$. The latter $n$ is very close to the limiting case of hard disks.

We simulate the system by means of molecular dynamics method. Mostly systems of 20000 particles are used. The system is simulated in an NVT ensemble (constant number of particles N, volume V and temperature T). Additional simulations in NVE (constant number of particles N, volume V and internal energy E) are performed to calculate the diffusion coefficient.

Firstly we perform simulations with a pure system. After that we consider systems with 0.1\% of particles pinned in random positions. In this case we consider 10 different systems with different initial positions of pinned particles. All results are averaged over these 10 replicas.

Several methods are used to find phase boundaries. First of all we analyze the equations of state. In the case of first-order phase transition it should demonstrate the Mayer-Wood loop while in the case of continuous transitions only a bend is observed. However, from the equations of state one can only find whether the system demonstrates a first-order phase transition, but it is impossible to distinguish a simple first-order transition from the BK scenario. In order to find the exact
boundaries of the crystalline phase we use the approach proposed in our recent publication. As it was demonstrated in the hexatic phase, the orientational correlation function decays algebraically at the boundary of the hexatic phase with isotropic liquid. Similarly, the translational correlation function decays algebraically at the boundary of the crystalline phase we use the approach proposed in our recent publication.

The orientational correlation function is defined as

$$\Psi_6(r_i) = \frac{1}{n(i)} \sum_{j=1}^{n(i)} e^{i\theta_{ij}},$$

(2)

where $\theta_{ij}$ is the angle between vector $r_{ij}$, connecting the $i$-th and $j$-th particles and an arbitrary axis. The sum includes all neighbors $n(i)$ of the $i$-th particle. The nearest neighbors are determined by the Voronoi construction.

One can also define the global orientational order parameter $\psi_6$, which is the average of $\Psi_6(r_i)$ over the whole system

$$\psi_6 = \frac{1}{N} \langle \langle \sum_i \Psi_6(r_i) \rangle \rangle_{rp}.$$  

(3)

The second brackets $\langle \langle \cdots \rangle \rangle_{rp}$ mean the averaging over 10 replicas with random pinning.

The translational order parameter is defined as

$$\psi_T = \frac{1}{N} \langle \langle \sum_i e^{iG \cdot r_i} \rangle \rangle_{rp},$$

(4)

where $r_i$ is the radius vector of the $i$-th particle and $G$ is a reciprocal lattice vector.

The translational correlation function is defined as

$$G_T(r) = \langle \exp(iG(r_i - r_j)) \rangle_{rp},$$

(5)

where $r = |r_i - r_j|$, and $g(r) = \langle \delta(r_i)\delta(r_j) \rangle$ is a radial distribution function. In the case of the crystalline phase without random pinning in the limit $r \rightarrow \infty$ the translational correlation function is $G_T(r) \propto r^{-\eta_4}$, where $\eta_4 \leq 0.52$.

The orientational correlation function $G_6(r)$ is defined in the same way:

$$G_6(r) = \langle \Psi_6(r) \Psi^*_6(0) \rangle_{rp},$$

(6)

where $\Psi_6(r)$ is the local orientational order parameter. The orientational correlation function decays algebraically in the hexatic phase $G_6(r) \propto r^{-\eta_6}$ where $\eta_6 \leq 0.19$.

The limit of stability of the hexatic phase with respect to isotropic liquid is determined from $\eta_6(T_c) = 1/4$.  

Figure 1. The equations of state of the system with $n = 12$ with and without random pinning at $T = 1.0$. The inset enlarges the region of the Mayer-Wood loop.

III. RESULTS AND DISCUSSION

We start discussing the equation of state of the system with $n = 12$. Fig. 1 shows the equations of state for both the pure system and the system with random pinning. At $T = 1.0$ the Mayer-Wood loop is observed at densities around unity, i.e. first-order phase transition takes place. However, it is necessary to find which phases are separated by this transition, whether it is a simple first-order phase transition or the BK scenario. It means that we need to determine which phase is located at the right branch of the Mayer-Wood loop: crystal or hexatic. In order to do this, we calculate the orientational order parameter, the translational order parameter and their correlation functions.

Fig. 2 shows the orientational order parameter and the translational order parameter both in the presence and absence of random pinning along the $T = 1.0$ isotherm. If there is no random pinning the translational order parameter becomes zero at density $\rho = 1.008$, while the orientational order parameter becomes zero at $\rho = 0.99$. It means that there is a region where the system is translationally disordered while quasi long-range orientational order is still present. This is consistent with the definition of a hexatic phase.

Let us consider the order parameters of the system with random pinning. The behavior of the orientational order parameter is the same as in the pure system. However, the translational order parameter shows qualitatively different: starting from density about 1.15 there is a jump of the translational order parameter at level about 0.45. When the system enters the two-phase re-
region the translational order parameter vanishes (Fig. 2). We believe that the step corresponds to the region of the hexatic phase. The finite value of the translational order parameter in the hexatic phase should be related to some local ordering which is not destroyed completely by random pinning.

Fig. 3 shows the correlation functions of the orientational and translational order parameters of the system without random pinning. They also confirm the presence of the hexatic phase in the system. One can see that at high densities the orientational correlation function does not decay, which corresponds to the presence of long-range orientational order in the system. At the same time the translational correlation function decays algebraically. At low densities both orientational and translational correlation functions demonstrate exponential decay, i.e. isotropic liquid is observed. The critical value of translational correlation function exponent $\eta_T = 1/3$ is reached at $\rho_{sh} = 1.014$. This density is higher than the first-order transition density obtained using the Maxwell construction for the Mayer-Wood loop. The Maxwell construction gives $\rho_l = 0.998$ for the liquid density of the liquid-hexatic transition and $\rho_{lh} = 1.006$ for the density of the hexatic phase. The limit of stability of the hexatic phase shifts to greater densities of up to $\rho_{sh} = 1.15$. One can also see that the translational correlation function of the system with random pinning demonstrates a kink. At small distances random pinning does not substantially affect the system. As a result, the limit of stability of the hexatic phase shifts to greater densities, where the crystal is stable in the pure system.

Orientional correlation function $G_6$ and translational correlation function $G_T$ of the system with random pinning are shown in Fig. 4. As it was expected random pinning does not change the location of the boundary between the hexatic phase and isotropic liquid. However, the boundary between the crystal and hexatic phases moved to higher densities of up to $\rho_{sh} = 1.15$. One can also see that the translational correlation function of the system with random pinning demonstrates a kink.
affect local ordering. At the same time random pinning does influence the asymptotic behavior of the translational correlation functions. Because of this the region after the kink should be used to determine the limit of stability of the crystal phase.

The phase boundaries obtained from $G_6$ and $G_T$ are placed on the equation of state (Fig. 5). One can see that random pinning does not influence the transition from the hexatic phase to isotropic liquid. At the same time the boundary between the crystal and hexatic phases moved to much higher densities. This observation allows us to improve our understanding of the system melting scenario. Since the Mayer-Wood loop is observed, one can be sure that a first-order phase transition takes place in the system. However, in the pure system the stability region width of the hexatic phase is very small and one can easily "miss" it. In the presence of random pinning the width of the hexatic phase region becomes substantial and one can clearly see that first-order phase transition corresponds to the transition from the liquid to hexatic phases. No peculiarities are observed at the crystal/hexatic phase boundary. This is especially obvious in the system with random pinning since here this boundary is far from first-order phase transition and does not in the least affect the equation of state behavior. Therefore, this transition is a continuous one. Our calculations confirm that the system melts via the BK scenario.

In order to be more sure of the location where the phase boundary of solid to hexatic transition is, we employ one more method based on the elastic properties of a 2D system. According to the BKTNY theory the crystal transforms into the hexatic phase at temperature $T_m$ which is determined by the BKT equation:\cite{1,7,8,10,11,12}:

$$k_B T_m = \frac{K}{16\pi}. \tag{7}$$

Here $K$ is the reduced Young modulus and $k_B$ is the Boltzmann constant.

The reduced Young modulus of a triangular crystal can be calculated as

$$K = \frac{a_0^2}{k_B T} \frac{4\mu(\mu + \lambda)}{2\mu + \lambda}, \tag{8}$$

where $\mu$ and $\lambda$ are usual Lame coefficients. $\mu$ has the meaning of a shear modulus.
In order to calculate the Young modulus we calculate shear modulus $\mu$ by the method proposed in Ref.\textsuperscript{19}. In this method the system is strained. As a result a non-diagonal component of pressure proportional to the shear modulus appears:

$$P_{xy} = \mu u_{xy} + O(u_{xy}^2),$$ \hfill (9)

where $u_{xy}$ is strain. Lame coefficient $\lambda$ can be calculated if the bulk modulus is also known:

$$B = \left(\frac{\partial P}{\partial \rho}\right)_T = \lambda + \mu.$$ \hfill (10)

Fig. 6 shows examples of $P_{xy}$ as a function of strain. One can see that at high densities far from the transition point the dependence is characterized by strong linearity. The accuracy of calculations becomes worse as transition to the hexatic phase is approached.

The resulting Young modulus is shown in Fig. 7. One can see that the Young modulus reaches magnitude $16\pi$ at $\rho = 1.014$ which is in good agreement with the results obtained from $G_T$. However, such calculations of the Young modulus can suffer from finite size effects. In order to remove these effects one needs to use renormalization group equations. These equations have the following forms:

$$\frac{dK^{-1}(l)}{dl} = \frac{3}{2} \pi y^2(l) e^{\frac{K(l)}{8\pi}} I_0 \left( \frac{K(l)}{8\pi} \right) - \frac{3}{4} \pi y^2(l) e^{\frac{K(l)}{8\pi}} I_1 \left( \frac{K(l)}{8\pi} \right)$$ \hfill (11)

and

$$\frac{dy(l)}{dl} = \left( 2 - \frac{K(l)}{8\pi} \right) y(l) + 2\pi y^2(l) e^{\frac{K(l)}{8\pi}} I_0 \left( \frac{K(l)}{8\pi} \right),$$ \hfill (12)

where $l$ is a scaling variable, $y$ is fugacity and $I_0$ and $I_1$ are Bessel functions. Fugacity can be calculated as follows

$$y = e^{-E_c/k_BT},$$ \hfill (13)

where $E_c$ is the energy of the dislocation core. It can be calculated from the probability of defect formation\textsuperscript{51}:

$$\rho_d = \frac{16\pi \sqrt{3} \pi^2}{K - 8\pi} I_0 \left( \frac{K}{8\pi} \right) e^{\frac{K}{8\pi} - \frac{2E_c}{k_BT}}.$$ \hfill (14)

Dislocation core energy is shown in Fig. 8. In Refs.\textsuperscript{17,19} it was shown that in the case of low dislocation core energies the system should melt via first-order phase transition. As it was mentioned in the Introduction, the characteristic scale of dislocation core energy was given in Ref.\textsuperscript{50}, where it was shown that transition went from strong first-order to weak first-order for dislocation core energy $E_c$ less than $2.84T_m$, where $T_m$ is taken from Eq. (7). Along isotherm $T = 1.0$ the critical core energy is $E_c = 2.84$. One can see in Fig. 8 that the core energy obtained in our work is considerably higher, therefore a continuous transition between the crystal and hexatic phases is expected.

Fig. 9 shows a solution of the renormalization group equations for the pure system with $n = 12$. The densities where $y \rightarrow 0$ as $l \rightarrow \infty$ correspond to the crystalline phase while the ones where $y \rightarrow \infty$ are in the hexatic phase.
phase. From Fig. 9 one can see that transition from crystal to hexatic takes place at \( \rho_{sh} = 1.02 \). This is somewhat higher than the results from the \( G_T \) criterion, however, the difference is within 0.5%, therefore the results can be considered as being in perfect agreement.

In order to ensure that the hexatic phase is an ordered liquid we perform calculations of the diffusion coefficient. Importantly, the hexatic phase was observed in some publications (see, for instance, \( 30,31,36,47,48 \)), but the region of stability of this phase was always vanishingly small which hindered investigation of hexatic phase properties. Because of this it appears reasonable to study the properties of the hexatic phase in a system with random pinning where it demonstrates a large enough region of stability.

Comparing the diffusion coefficients of a pure system with those of a system with random pinning one can see that in the latter case the finite value of the diffusion coefficient is preserved up to density \( \rho_{sh} = 1.15 \) (see the inset of Fig. 10 which corresponds to the boundary of crystal-hexatic transition from the \( G_T \) criterion. In isotropic liquid the diffusion coefficient is about 11% higher in the pure system compared to the system with random pinning.

The same type of calculations is performed for the pure system with \( n = 1024 \). Importantly, because of a very large \( n \) the system is very anharmonic. Therefore, sufficient statistics are required to obtain precise data. We calculated equations of state for densities from \( \rho_{min} = 0.878 \) up to \( \rho_{max} = 0.94 \) and \( T = 1.0 \). We found the Mayer-Wood loop in this region. In the region of the loop we averaged the system over 20 different replicas (10 replicas of 20000 particles, 5 replicas of 45000 particles and 5 replicas of 8000 particles). The replicas differed in the initial velocities of system particles. Only averaging over 20 replicas allowed us to remove noise from the results.

Fig. 11 shows the equation of state of the system. One can see the Mayer-Wood loop. From the Maxwell construction we obtained a region of loop existence from density \( \rho_l = 0.89 \) and \( \rho_{th} = 0.91 \). To identify phase stability regions the orientational and translational correlation functions were used (Fig. 12). One can see that the stability limit of the hexatic phase with respect to

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**Figure 8.** The dislocation core energy of the pure system with \( n = 12 \).

**Figure 9.** The solution of the renormalization group equations for the pure system with \( n = 12 \).

**Figure 10.** The diffusion coefficient in the pure system and in the system with random pinning with \( n = 12 \).
liquid is at $\rho_h = 0.894$ (Fig. 12(a)) while that of crystal with respect to the hexatic phase is $\rho_x = 0.93$ (Fig. 12(b)). These results also correspond to the BK scenario of melting. Therefore, the "hardness" of the potential does not affect the melting scenario. However, the width of the hexatic phase in the system with $n = 1024$ appears to be larger.

In conclusion, we have performed a computer simulation study of a soft-disk system with $n = 12$ and $n = 1024$. Pure systems and systems with random pinning were studied. It was found that the presence of random pinning made the stability region of the hexatic phase larger. However, random pinning did not change the melting scenario. Moreover, random pinning did not influence the orientational ordering in the system, while it strongly affected the translational order which is clearly seen from the behavior of the orientational and translational order parameters and their correlation functions. For the first time the diffusion coefficient of the hexatic phase was calculated for this system. We showed that the results for solid to hexatic transition obtained from the $G_T$ criterion, Young modulus calculation and the solution of renormalization group equations were in perfect agreement. It was shown that the diffusion coefficient became finite at the transition point of the crystal to hexatic phases. In isotropic liquid the diffusion coefficient of the system with pinning was slightly less than in the case of the pure system.

Importantly, in the vicinity of crystal to hexatic continuous transition we did not observe any peculiarities of the equation of state which was not very informative for the correct determination of the melting scenario. That is why we used other criteria such as the behavior of the orientational and translational order parameters, their correlation functions, diffusion coefficient as well as solving renormalization group equations.

Finally, it is noteworthy that the nature of first-order transition from the hexatic phase to isotropic liquid is still not understood. It should be noted that the Berezinskii-Kosterlitz-Thouless transition can be made first-order by reducing core energy $E_c$ of a corresponding topological defect (disclination) below some critical value. It should be also noted that our results confirm the melting scenario proposed in Ref. [5], and contradict the recent article. [5] This contradiction will be discussed in further publications.

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Figure 12. (a) The orientational correlation functions of the pure system with $n = 1024$. (b) The translational correlation functions of the pure system with $n = 1024$. 

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