

# Random pinning elucidates the nature of melting transition in two-dimensional core-softened potential system

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(Dated: January 16, 2017)

Despite about forty years of investigations, the nature of the melting transition in two dimensions is not completely clear. In the framework of the most popular Berezinskii-Kosterlitz-Thouless-Halperin-Nelson-Young (BKTHNY) theory, 2D systems melt through two continuous Berezinskii-Kosterlitz-Thouless (BKT) transitions with intermediate hexatic phase. The conventional first-order transition is also possible. On the other hand, recently on the basis of computer simulations the new melting scenario was proposed with continuous BKT type solid-hexatic transition and first order hexatic-liquid transition. However, in the simulations the hexatic phase is extremely narrow that makes its study difficult. In the present paper, we propose to apply the random pinning to investigate the hexatic phase in more detail. The results of molecular dynamics simulations of two dimensional system having core-softened potentials with narrow repulsive step which is similar to the soft disk system are outlined. The system has a small fraction of pinned particles giving quenched disorder. Random pinning widens the hexatic phase without changing the melting scenario and gives the possibility to study the behavior of the diffusivity and order parameters in the vicinity of the melting transition and inside the hexatic phase.

PACS numbers: 61.20.Gy, 61.20.Ne, 64.60.Kw

## INTRODUCTION

The phenomenon of two-dimensional melting is one of the long-standing issues addressed by condensed matter physics. Despite numerous papers published over the last forty years, the mechanisms underlying the microscopic nature of melting in two dimensions (2D) remain unclear. In contrast to the three dimensional case, where melting develops through conventional first order transition, several microscopic scenarios have been proposed to explain microscopic 2D melting. The difference between these cases deals with dramatic increase of fluctuations in 2D in comparison with the 3D case. Landau, Peierls and Mermin [1, 2] showed that in two dimension situation the long-ranged positional order can not exist because of the thermal fluctuations, therefore the positional order transforms into the quasi-long-ranged order. On the other hand, the real long-ranged orientational order (the order in directions of the bonds binding a particle with its neighbors) does exist in this case.

Currently, the Berezinskii-Kosterlitz-Thouless-Halperin-Nelson-Young (BKTHNY) theory of 2D melting [3–9] is considered the best to describe the phenomenon. According to this theory 2D solids melt through two continuous transitions, which are activated by topological defects. For example, a disclination is an elementary topological defect in triangular lattice. In a triangular crystal lattice disclination is defined as an isolated defect having five or seven neighbours. A dislocation can be considered as a bound pair of 5- and 7-fold disclinations. In the framework of BKTHNY scenario, 2D solids melt through dissociation of bound dislocation pairs. In this case the long-ranged orienta-

tional order transforms into quasi-long-ranged order, and the quasi-long-ranged positional order becomes short-ranged. The new intermediate phase is called a hexatic phase. In turn, the hexatic phase transforms into an isotropic liquid phase having short-ranged orientational and positional orders through unbinding dislocations (5 and 7-fold bound pairs) into free disclinations. It should be noted, that the BKTHNY theory provides only limits of stability of the solid and hexatic phases and does not rule out that the first order liquid-solid transition can be preceded by other melting mechanisms.

The BKTHNY theory seems universal and it can be applied to describe all systems. It does not contain the interparticle potential in the explicit form, but, it contains two phenomenological parameters - the core energy of dislocation  $E_c$  and Frank module of hexatic phase  $K_A$  - that cannot be expressed explicitly in terms of the interparticle potential. Later it was shown that decreasing  $E_c$  can result in melting occurring through a single first-order transition resulting, for example, from forming grain boundaries [10] or "exploding" 5-7-5-7 quartets (bound dislocation pairs) into free disclinations [11].

Recently the BKTHNY scenario was confirmed in experiments with superparamagnetic colloidal particles interacting through long-range dipolar potential [12–16]. In these experiments the particles were absorbed at a liquid-air interface which restricted the out-of-plane motion. On the other hand, melting transition consistent with the BKTHNY scenario was found [18–20] in popular experimental systems of colloidal particles confined between two glass plates [17]. At the same time, the existence of the first order liquid-solid transitions [21] and even the first-order liquid-hexatic and the first-order

hexatic-solid phase transitions [22] can be expected. In 2D superconducting vortex lattices, macroscopic measurements provide evidence for melting close to the transition to the normal state. In Refs. [23, 24] the authors using the scanning tunnelling spectroscopy showed directly that the transition into an isotropic vortex liquid below the superconducting critical temperature does exist. Before that, they found a hexatic phase, characterized by the appearance of free dislocations, and a smectic-like phase, possibly formed through partial disclination unbinding. These results confirm that the melting mechanism is not universal and it depends on interparticle interactions. At the same time the ambiguity remains even in describing the same systems such as, for example, the systems of hard disks [25–34].

Recently, the other melting scenario was proposed [35–40]. In contrast to the BKTHNY theory, it was maintained that the hexatic phase does exist in the basic hard disk model, and the system melts through a continuous solid-hexatic transition but through a first-order hexatic-liquid transition [35–37]. In [39] it was shown that a first-order transition occurs between the stable hexatic phase and isotropic liquid in 2D Yukawa system. In paper [38] Kapfer and Krauth explored behavior of a soft disk system with potential  $U(r) = (\sigma/r)^n$ . The system was shown to melt in accordance with the BKTHNY theory for  $n \leq 6$ , while for  $n > 6$  the two-stage melting transition took place with continuous solid-hexatic and the first-order hexatic-liquid transition. This scenario was confirmed experimentally in a system of colloidal particles on water-decane interface [41]. Recently a paper was published [42] where at low densities the Herzian disks model was shown to demonstrate the reentering melting transition featuring the maximum on the melting line. The discontinuous liquid-hexatic transition occurs at lower than the maximum densities, while at higher densities systems melt undergoing a continuous BKT transition.

It should be noted that the simulations in Refs. [35–40] demonstrate the very narrow hexatic phase. However, the presence of disorder can widen the hexatic phase and help study its properties. Experiments usually involve two-dimensional confinement provided by slit pores having different origin or by adsorption on solid substrates. Both cases can result in the frozen-in (quenched) disorder generated by a certain roughness. Quenched disorder can change the melting scenario in 2D. A disordered substrate can have a similar destructive effect on the crystalline order as temperature and can bring about melting even at zero temperature [43–46]. It was shown in Refs. [43, 44] that the BKTHNY melting scenario persists in presence of weak disorder. It is intuitively understood that the temperature of the hexatic-isotropic liquid transition  $T_i$  is almost unaffected by disorder, whereas the melting temperature  $T_m$  decreases drastically with increasing disorder [43–46]. As a result, the stability range

of the hexatic phase gets wider. These predictions were confirmed in experiment and through simulating superparamagnetic colloidal particle systems [15, 16]. In these experiments under gravity particles formed a monolayer at the bottom of a cylindrical glass cell having 5 – mm diameter. Quenched disorder developed due to pinning a small amount of particles to the glass substrate due to van der Waals interactions and chemical reactions. In our simulations we tried to find a simulation method similar to above experimental setup.

In this paper we present results of detailed computer simulation studies of 2D phase diagram previously suggested core softened potential system [47–52] in presence of quenched disorder for small value of the width of repulsive shoulder. The different forms of core-softened potentials are widely used for qualitative description of the anomalous water-like behavior, including density, structural and diffusion anomalies, liquid-liquid phase transitions, glass transitions, and melting maxima [47–65]. In our previous publications the preliminary results on plotting phase diagrams for the system were reported [66–70].

Here we approached the case of the system having small repulsive shoulder. It was shown that in this case the behavior of the system potential was similar to that of a soft disks system [67]. The random pinning results in drastic increase of the width of hexatic phase without changing the melting scenario. It gives the possibility to study some properties of the hexatic phase including calculation of the diffusion coefficient and the behavior of the orientational and translational order parameters.

## SYSTEMS AND METHODS

In the current simulations we studied a system described by potential [47–52, 66–68, 70]:

$$U(r) = \varepsilon \left( \frac{\sigma}{r} \right)^n + \frac{1}{2} \varepsilon (1 - \tanh(k_1 \{r - \sigma_1\})). \quad (1)$$

where  $n = 14$  and  $k_1 \sigma = 10.0$ .  $\sigma$  are the hard-core diameters. We simulate systems having small soft-core diameter:  $\sigma_1/\sigma = 1.15$  (see Fig. 1).

Through the paper we used dimensionless quantities, which in 2D had the form:  $\tilde{\mathbf{r}} \equiv \mathbf{r}/\sigma$ ,  $\tilde{P} \equiv P\sigma^2/\varepsilon$ ,  $\tilde{V} \equiv V/N\sigma^2 \equiv 1/\tilde{\rho}$ ,  $\tilde{T} \equiv k_B T/\varepsilon$ ,  $\tilde{\sigma} = \sigma_1/\sigma$ . Hereinafter we omitted the tildes.

The study dealt with molecular dynamics simulations in  $NVT$  and  $NVE$  ensembles in the framework of the LAMMPS package [72] for the number of particles ranging from 20000 to 100000. We randomly choose a subset of particles at the random positions and kept them immobile for a complete simulation run [70] in order to model a quenched disorder. The simulations of 10 independent replicas of the system with different distributions of random pinned patterns were made. The thermodynamic functions were calculated by averaging over replicas. We

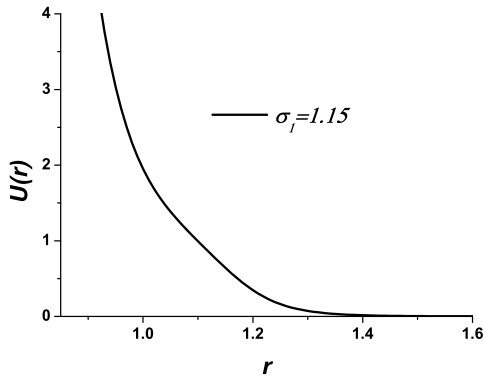


FIG. 1: The potential (1) with soft-core diameter  $\sigma_1/\sigma = 1.15$ .

obtained curves for pressure  $P$  plotted as functions of density  $\rho$  along isotherms; we plotted correlation functions  $G_6(r)$  and  $G_T(r)$  of the bond orientational  $\psi_6$  and translational  $\psi_T$  order parameters (OPs), which characterized the overall orientational and translational order [70].

The translational  $\psi_T$  (TOP) and orientational order parameters  $\psi_6$  (OOP) and the bond-orientational  $G_6(r)$  (OCF) and translational  $G_T(r)$  (TCF) correlation functions were determined in the ordinary manner [7, 8, 31, 33, 34, 42, 64, 65] with subsequent averaging over the quenched disorder [70].

In accordance with the standard definitions [7, 8, 42], TOP has the form:

$$\psi_T = \frac{1}{N} \left\langle \left\langle \left| \sum_i e^{i\mathbf{G}\mathbf{r}_i} \right| \right\rangle \right\rangle_{rp}, \quad (2)$$

where  $\mathbf{r}_i$  is the position vector of particle  $i$  and  $\mathbf{G}$  is the reciprocal-lattice vector of the first shell of the crystal lattice. The translational correlation function can be obtained from the equation:

$$G_T(r) = \left\langle \frac{\langle \exp(i\mathbf{G}(\mathbf{r}_i - \mathbf{r}_j)) \rangle}{g(r)} \right\rangle_{rp}, \quad (3)$$

where  $r = |\mathbf{r}_i - \mathbf{r}_j|$  and  $g(r) = \langle \delta(\mathbf{r}_i) \delta(\mathbf{r}_j) \rangle$  is the pair distribution function. The second angular brackets  $\langle \dots \rangle_{rp}$  correspond to averaging over the random pinning. In the solid phase without random pinning the long range behavior of  $G_T(r)$  has the form  $G_T(r) \propto r^{-\eta_T}$  with  $\eta_T \leq \frac{1}{3}$  [7, 8].

To measure the orientational order and the hexatic phase, the local order parameter determining the 6-fold orientational ordering can be defined as follows:

$$\Psi_6(\mathbf{r}_i) = \frac{1}{n(i)} \sum_{j=1}^{n(i)} e^{in\theta_{ij}}, \quad (4)$$

where  $\theta_{ij}$  is the angle of the vector between particles  $i$  and  $j$  with respect to the reference axis and the sum over  $j$  is counting  $n(i)$  neighbors of  $i$ , obtained from the Voronoi construction. The global OOP can be calculated as an average over all particles and random pinning:

$$\psi_6 = \frac{1}{N} \left\langle \left\langle \left| \sum_i \Psi_6(\mathbf{r}_i) \right| \right\rangle \right\rangle_{rp}. \quad (5)$$

The orientational correlation function  $G_6(r)$  (OCF) is provided in a similar manner in Eq. (3):

$$G_6(r) = \left\langle \frac{\langle \Psi_6(\mathbf{r}) \Psi_6^*(\mathbf{0}) \rangle}{g(r)} \right\rangle_{rp}, \quad (6)$$

where  $\Psi_6(\mathbf{r})$  is the local bond-orientational order parameter (4). In hexatic phase there is a quasi-long range order with the algebraic decay of the orientational correlation function  $G_6(r) \propto r^{-\eta_6}$  with  $0 \leq \eta_6 \leq \frac{1}{4}$  [7–9]. The stability criterion of the hexatic phase has the form  $\eta_6(T_i) = \frac{1}{4}$ .

The influence of disorder on the orientational and translational correlation functions was explored in our previous publication [70] (see Fig. 1 in [70]) for  $\sigma = 1.35$ . We expected that in presence of pinning there would be no qualitative change observed in the behavior of  $G_6(r)$ . This was in accordance with the results by Nelson and coworkers [43, 44]. On the other hand, the translational correlation function  $G_T(r)$  behaves differently in presence of random disorder. Without pinning TCF features a conventional power decay. In case of pinning the envelope gets steeper with crossover value equal to  $r_0$ . The region  $r < r_0$  corresponds to the local order unaffected by quenched disorder, whereas asymptotic TCF behavior when  $r > r_0$  is controlled by random pinning [70]. It was also shown that according to intuitive physics expectations  $r_0$  decreases with increase in pinning centers concentration accompanied by increasing slope of envelope. The equality  $\eta_T(T_m) = 1/3$  holding for the long range asymptote of TCF (for  $r > r_0$ ) can be considered as the solid-hexatic stability criterion. The hexatic-liquid stability point corresponds to  $\eta_6(T_i) = 1/4$  [43, 44].

## RESULTS AND DISCUSSION

Let us consider the behavior of the system having small repulsive shoulder  $\sigma = 1.15$ . In this case, as one can see in Fig. 1, the form of the potential is similar to that of a soft disks system with  $n = 14$ . The preliminary discussion of the phase diagram of this system can be found in Ref. [67], where isotherms with the van der Waals loops were developed and the phase diagram was calculated using double-tangent construction to the Helmholtz free energy curves [71].

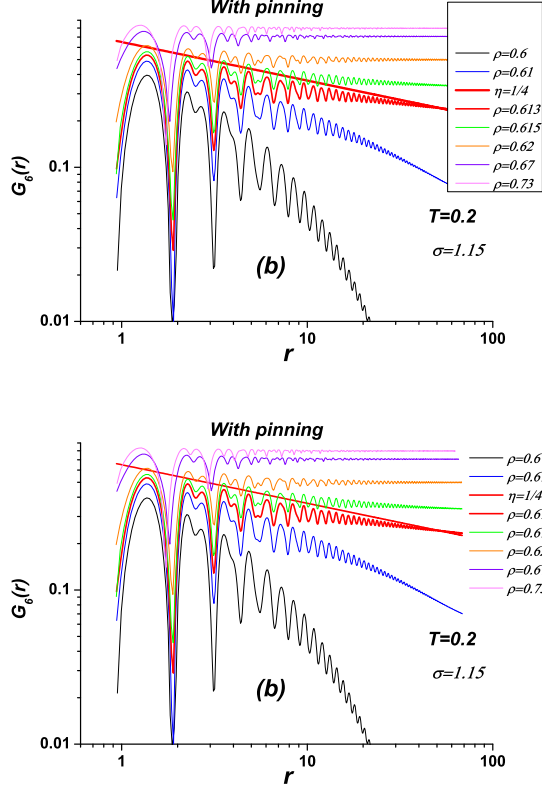


FIG. 2: OCF plotted for different densities at  $T = 0.2$  without random pinning (a) and concentration of the pinning centers 0.1% (b).

In Fig. 2 we show the orientational correlation functions (OCF) obtained for a system without quenched disorder (a) and having random pinning (concentration of pinning centers is 0.1%) (b) at temperature  $T = 0.2$ . Behavior of OCFs is identical for both cases, in line with discussions in Introduction section; the limits of hexatic phase stability determined from condition  $\eta_6 = 1/4$  also coincide. As Fig. 2 suggests the density at which hexatic phase becomes unstable is  $\rho \approx 0.613$ .

On the other hand, the translational correlation functions obtained for systems with and without random pinning are qualitatively different (see Fig. 3). In this case the densities corresponding to the loss of solid phase stability are considerably different. It can be deduced from Fig. 3 (a) that without pinning the stability limit corresponds to  $\rho \approx 0.63$  while in systems having the quenched disorder the solid-hexatic stability limit is  $\rho \approx 0.66$  (see Fig. 3 (b)).

Fig. 4 depicts the equation of state at  $T = 0.2$ . It is seen that the liquid-hexatic stability limit density is located inside the Van der Waals loop for both systems the one having the random pinning and without it. At the same time, in pinning-free systems the hexatic-solid stability limit is located outside the Van

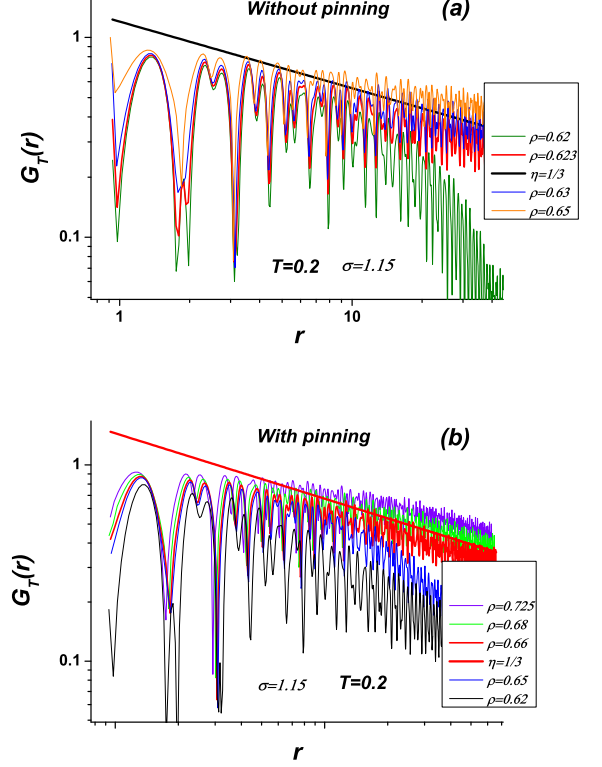


FIG. 3: TCF plotted for different densities at  $T = 0.2$  without random pinning (a) and for concentration of the pinning centers equal to 0.1% (b).

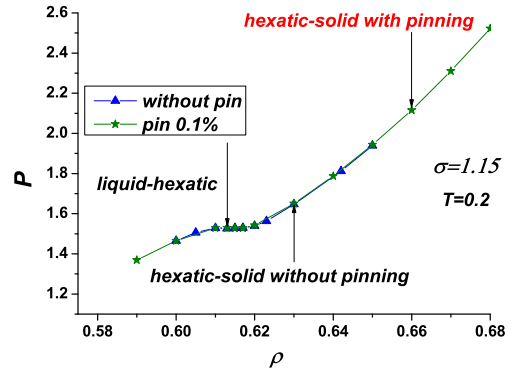


FIG. 4: Isotherms developed for a system having  $\sigma = 1.15$  without pinning (triangles) and for system having concentration of the pinning centers equal to 0.1% (stars) when  $T = 0.2$ .

der Waals loop. One can conclude that in this case the system melts according to the melting scenario proposed in Refs. [35, 36, 39].

In presence of random pinning the hexatic-solid instability density shifts to the higher densities, yet, the melting scenario remains the same. In our case we observe

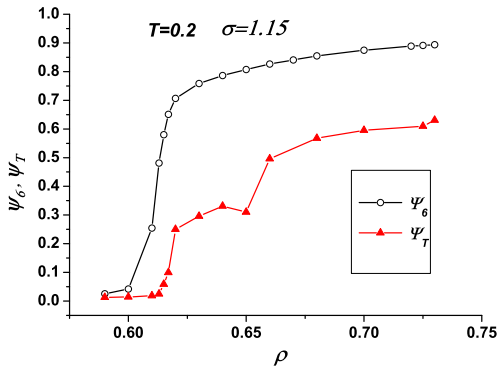


FIG. 5: The orientational  $\Psi_6$  and translational  $\Psi_T$  order parameters for a system having random pinning and  $\sigma = 1.15$ .

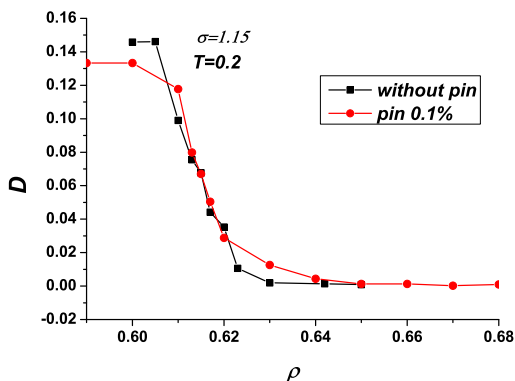


FIG. 6: The diffusion coefficient of the system with  $\sigma = 1.15$  with (red circles) and without (black squares) random pinning.

the first-order liquid-hexatic transition and continuous hexatic-solid transition. Fig. 5 provides for corresponding behavior in orientational  $\psi_6$  and translational  $\psi_T$  order parameters. The orientational order parameter  $\psi_6$  is typical for conventional behavior, for example, for systems without random pinning [67]. However, the translational order parameter  $\psi_T$  has a qualitatively different form. It can be seen in Fig. 5 that the curve  $\psi_T$  has a step-like behavior. When density decreases,  $\psi_T$  moves down at  $\rho \approx 0.66$  corresponding to the solid phase stability limit in presence of random pinning. On the other hand,  $\psi_T$  is not equal to zero at  $\rho \approx 0.66$  because the local translational order exists in this state (see Fig. 3 and discussion given in previous Section). The translational order parameter disappears only in the two-phase region.

Since the translational order parameter  $\psi_T$  is not equal to zero in hexatic phase, it is interesting to calculate the diffusion coefficient in order to be sure that the system is

in the liquid state (having quasi-long range orientational order). It should be noted that in cases when the hexatic phase was reported (see, for example, [35, 36, 39, 65]) the density range is extremely narrow, and it is very difficult to calculate the dynamic properties of the system. In presence of quenched disorder the hexatic phase gets considerably wider, and the calculation of diffusion coefficient gives more accurate results. In Fig. 6 we plot the diffusion coefficient for  $\sigma = 1.15$  having random pinning and without it. One can see that in case without pinning an increase in diffusion coefficient begins at  $\rho \approx 0.63$ , while in a system having random pinning the diffusion coefficient has non-zero values at  $\rho \approx 0.66$  in accordance with results shown in Figs. 4 and 5.

## CONCLUSIONS

In this paper we present results of computer simulations of melting transitions in 2D core softened systems having small length of the repulsive shoulder (Eq. (1) and Fig. 1) for cases when the random pinning (quenched disorder) is present and when there is no random pinning. It is shown that without quenched disorder the system having small repulsive shoulder  $\sigma = 1.15$  which is close in shape to soft disks  $1/r^n$  with  $n = 14$  melts according to the melting scenario proposed in Refs. [35, 36, 38] (first-order liquid-hexatic and continuous hexatic-solid transitions). Random pinning widens the hexatic phase without altering the melting scenario. It is shown that as one can expect the random pinning almost does not change the behavior of the orientational order parameter, orientational correlation function and equation of state of the system. This means that the first order transition between the hexatic phase and isotropic liquid is almost unchanged. On the other hand, random pinning drastically changes the behavior of the translational correlation function (Fig. 3) and translational order parameter (see Fig. 5). As it was discussed above, this change is related to the different behavior of the local and overall translational order in the system. The solid-hexatic transition shifts to the range of the higher densities. We also calculate the diffusion coefficient in the hexatic phase (Fig. 6). One can see that at the point of the BKT type solid-hexatic transition diffusion coefficient becomes nonzero and slowly grows while the diffusion coefficient of the solid phase (without pinning) at the same densities is zero. The rapid increase of the diffusion coefficient begins at the two phase region. As one can expect, the diffusion coefficient of the system with pinning is slightly lower than the one in the pinning-free isotropic liquid. It should be noted that the signs of the solid-hexatic BKT-type transition cannot be found on the equation of state plot (Fig. 4). As it was mentioned above, the melting criterion based on the behavior of the translational correlation function gives only the stability limit

of the solid phase. In this case it is important to have additional proofs of the melting transition. The behavior of the translational order parameter and diffusion coefficient shown in Figs. 5 and 6 confirms the system melting scenario.

It is interesting to note that as it was discussed in our previous publication [70], melting in system having larger repulsive step  $\sigma = 1.35$  is more complex. At high densities the conventional first-order transition takes place without random pinning. It seems that the repulsive shoulder added to the soft disk potential  $1/r^{14}$  makes the hexatic phase which does exist in soft disks with  $n > 6$  [38] unstable. The disorder, however, drastically changes this melting scenario. A single first-order transition is split into two transitions, one of them (solid-hexatic) is the continuous BKT-like transition, and the hexatic to isotropic liquid transition occurs as the first order transition in accordance with [35, 36, 38]. A possible mechanism for this transition can be associated with spontaneous proliferation of grain boundaries [10, 37, 39]. It should be noted, that melting scenario with single first-order transition corresponds to systems kept at sufficiently low temperatures. At high temperatures the repulsive shoulder of the potential becomes ineffective, and the properties of the potential will be similar to that of soft disks  $1/r^n$  with  $n = 14$ . In this case we can expect finding a critical temperature (similar to tricritical point) above which melting should occur through two transitions in accordance with scenario proposed in Ref. [38].

It should be also noted, that the nature of the first-order liquid-hexatic transition is not completely understood as conventional theories like the BKTHNY are not capable of describing the first-order liquid-hexatic transition.

The authors are grateful to S.M. Stishov and V.V. Brazhkin for valuable discussions. We are thankful to the Russian Science Center at the Kurchatov Institute and the Joint Supercomputing Center of the Russian Academy of Science for providing computational facilities. The work was supported by the Russian Science Foundation (Grant No 14-12-00820).

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