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MELTING OF TWO-DIMENSIONAL SOLIDS

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Abstract

Experiments and computer simulation studies on two-dimensional melting are reviewed. Results are compared with predictions based on the theory of dislocation-mediated melting.

1. Introduction

Previously, it was believed that melting is quite generally a first-order phase transition. Recently, however HALPERIN and NELSON [1] developed a theory which leads to a higher-order melting transition in two-dimensional (2-d) systems. This theory is based on the idea, proposed by KOSTERLITZ and THOULESS [2], that melting is caused by an instability in the 2-d solid for the creation of free dislocations. It predicts an upper bound for the melting temperature T_m

$$k_B T_m \leq \frac{1}{4\pi} \frac{\mu(\mu+\lambda)}{2\mu+\lambda} a_0^2, \quad (1)$$

where μ and λ are the Lamé coefficients measured at T_m , and a_0 is the lattice constant. If the solid does melt via the dislocation unbinding mechanism the inequality becomes an equality and, in this case, striking predictions are made: At temperatures $T > T_m$ the system has lost its resistance to shear and translational order with a correlation length $\xi(T)$ of translational order which decreases exponentially with temperature,

$$\xi(T) \sim \exp A/(T-T_m)^\nu. \quad (2)$$

The critical exponent ν is nonuniversal and depends on the symmetry of the crystal. However orientational order is predicted to persist up to a temperature $T_i > T_m$, at which a second phase transition associated with the unbinding of another type of defect (disclinations) destroys the orientational order. The intermediate phase is characterized by an orientational order parameter

$$\psi_p(\vec{r}) = \exp ip \theta(\vec{r}), \quad (3)$$

where $\theta(\vec{r})$ is the angle relative to an arbitrary reference direction of the bond between two nearest-neighbor particles at position \vec{r} . The constant

p reflects the type of symmetry: for melting of triangular lattices $p = 6$ and for square lattices $p = 4$ corresponding to the persisting hexagonal or cubic symmetry of this phase for which the names "hexatic" and "tetratic" have been coined [1]. Defining the orientational correlation function

$$g_p(\vec{R}) = \langle \psi_p(\vec{R}) \psi_p^*(0) \rangle, \quad (4)$$

which, in the solid, for large separations \vec{R} , tends to a positive constant [3], Halperin and Nelson showed that the presence of free dislocations for temperatures $T > T_m$ leads to a power-law decay

$$g_p(\vec{R}) \sim R^{-\eta_p} \quad (5)$$

with a temperature-dependent critical exponent η_p which increases monotonically with temperature from zero at T_m to $1/4$ at T_i . This is in marked contrast to the behavior of a conventional fluid for which all correlations decay exponentially at large distances. At temperatures $T > T_i$ the presence of free disclinations leads to an exponential decay of $g_p(R)$ with a correlation length ξ_p which diverges like

$$\xi_p \sim \exp^{A/(T-T_i)^{1/2}} \quad T > T_i \quad (6)$$

as the temperature approaches the disclination-unbinding temperature T_i .

The theory developed by HALPERIN and NELSON [1] is based on continuum elasticity theory, in which long-wavelength phonons, dislocations and disclinations are taken into account. The quantities which determine the physical behavior are the Lamé-coefficients μ and λ and the core energy of dislocations E_c and disclinations E_d , which plays the role of a chemical potential for these defects. The theory is based upon an expansion in the fugacity of dislocations $\gamma = \exp - E_c/k_B T$ and similarly for disclinations. This expansion is well behaved if the core energies are large, i.e. for small defect densities. The core energies depend in a very nontrivial way on the interparticle potential [4]. If the fugacity of dislocations is not small at T_m , implying a high density of dislocations, a first-order phase transition from solid to isotropic fluid is likely to occur. Particularly in these cases, the possibility must be considered that special types of arrays of dislocations (e.g. grain boundaries) will drive the (first-order) transition [4, 5].

Other theories of 2d-melting, based on Van der Waals ideas [6] and on Landau theory [7] have been proposed. They all lead to a single first-order transition from solid to isotropic liquid.

2. Experimental Results

In order to test the striking predictions of the theory of dislocation-mediated melting - the existence of an orientationally ordered fluid and the universal relation (1), a great experimental effort has been undertaken. To summarize the present situation: In substrate-free thin film systems no evidence of a hexatic (or tetratic) phase has been obtained although in liquid crystals a 3d-stacked hexatic phase has been discovered [8]. Investigations on freely suspended liquid-crystal films by both mechanical measurements [9] and synchrotron X-ray studies [10] show first-order transitions from crystal to liquid although on 3 layer films an orientationally ordered phase has been found [9, 10]. In experiments with rare gases adsorbed on graphite both first-order and higher-order melting transitions have been seen. In these systems the situations of melting of solids which are either commensurate or incommensurate with the graphite substrate must be distinguished. While both Xenon and Argon on graphite melt from incommensurate solid to liquid [11, 12], the system of Krypton on graphite melts from commensurate solid to liquid and therefore substrate effects play an essential role [12] which we will not discuss here.

While Ar on graphite appears to have a higher-order melting transition for a wide range of coverages [12], Xe on graphite displays a first-order melting transition up to coverages close to one monolayer but clear indications of higher-order melting at a coverage of 1.1 monolayer [11]. In these systems synchrotron X-ray scattering experiments reveal correlation lengths in the fluid which become as large as the sample size (~ 100 nearest neighbor distances) as the melting point is approached and a temperature dependence consistent with (2).

Although in the incommensurate solid the graphite substrate does not directly couple to the translational order parameter it does produce orientational ordering fields [13] and it has been speculated [12] that these might be the origin of the higher order-melting transition. This point of view has been challenged very recently: An X-ray scattering experiment with Xenon on graphite at just below one-monolayer coverage [14] was interpreted as evidence for the existence of a very narrow hexatic phase ($(T_i - T_m)/T_m \approx 1\%$) which, it was argued, would be there in the absence of the weak orientational fields (without affecting the order of the transition). Clearly, to clarify this issue it would be desirable to study systems with smooth substrates and

one example that has been studied experimentally is the system of electrons trapped at the surface of liquid Helium. At the densities of the experiments, the system is in the classical regime where the thermal energy is much greater than the Fermi energy. After the first successful experimental observation of the formation of a triangular electron "solid" [15] attempts were made in order to measure the shear modulus and its behavior was found to be compatible with prediction (1) stated as an equality [16].

3. Results from Computer Simulations

A great deal of effort to clarify the nature of 2d-melting has also been devoted to computer simulations. The first simulation of 2d melting performed on the system of hard disks [17] showed evidence of a first-order melting transition very similar in appearance to melting of the 3d hard sphere system although with significantly smaller melting entropy ΔS ($\approx 0.35 k_B$ per particle). A possible connection of dislocations with 2d-melting was inferred from simulations of the Lenard-Jones system [18] at about the same time when the seminal ideas by Kosterlitz and Thouless were published [2]. The starting point of a real controversy was then the simulation of the Lenard-Jones system by Frenkel and McTague [19] who analyzed their results in the spirit of the theory of dislocation-mediated melting. They calculated the orientational correlation function $g_6(R)$ (eq. 4) the behavior of which seemed to indicate the existence of an hexatic phase. This interpretation was subsequently challenged by a number of investigators [20, 21, 22], who suggested that owing to the fact that this simulation was carried out at constant density the system was actually undergoing phase separation and the régime interpreted as the hexatic phase was actually a régime of two-phase coexistence. Another simulation of this system (constant density Monte-Carlo) by Tobochnik and Chester [23] indicated that there may be qualitatively different behavior depending on density. At relatively low densities they find a behavior of the elastic constants consistent with eq. (1) stated as an equality and a very gradual and smooth increase of the potential energy. At higher densities, however, they find a first-order transition with elastic constants at melting considerably greater than the Kosterlitz-Thouless values (1) and a steep noise rise of the energy.

Quite compelling evidence of first-order melting has also been obtained for the system with $1/r^{12}$ -interactions [24] which is consistent with

the Lenard-Jones system results at high densities [23] where the repulsive $1/r^{12}$ part dominates. The possibility has been discussed [25] that the order of the melting transition of 2d systems may depend on the "softness" of the pair-interaction potential. Indeed, in 3d-systems, it has been found that the "first-orderness" [26] (measured e.g. by the entropy of melting ΔS) is decreasing as the interaction becomes softer (e.g. for decreasing n in the pair interaction of the form $1/r^{12}$) and one might expect a similar trend for 2d systems [22, 25]. A Monte Carlo simulation of the $1/r^6$ system by McTague, Frenkel and Allen [27] indicates that this may be true. Using a microscopic definition of disclinations and dislocations they analyze the temperature dependence of defect densities and correlation functions, the behavior of which is consistent with the theory of dislocation-mediated melting. However, they were unable to determine the order of the melting transition.

Results of more recent simulations of various systems with even softer inverse-power pair interactions, e.g. the one-component plasma (log r interaction) [28, 29], the 2d-electron system ($1/r$ interaction) [30] and the system of interacting dipoles ($1/r^3$ -interaction) [31], have, in fact all been interpreted as evidence for a first-order melting transition with roughly the same melting entropy of $0.3 - 0.35 k_B$, which is consistent with the theoretical results obtained by Ramakrishnan [7]. However the interpretation of these simulation results is controversial. Both the one-component plasma and the electron system are incompressible, and as a result the density has to be continuous at melting and two-phase coexistence will not be allowed in a finite range of temperatures [25]. The observation of hysteresis and two-phase coexistence in these systems was however used in order to conclude that melting is first order. In fact this observation shows that these simulations are not really in equilibrium near the melting point, and thus conclusions about the order of the transition and the value of the melting entropy are not convincing although they may be correct.

On the other hand, both experiments [16] as well as computer simulations of the electron system [32] have found a behaviour of the shear modulus consistent with dislocation-mediated melting. Indeed, based on this theory and using the analytical values for the elastic constants λ and μ and the calculated values for the dislocation core energy [4] a prediction of the melting temperature was possible [32] which agrees with the experimental result.

At this point we should like to discuss a serious problem of all numerical simulations of 2d-melting: In simulations of thermodynamic systems one generally uses as a criterion for equilibrium the condition that for a given temperature the system evolves to the same final (and thus equilibrium) state independent of the initial configuration. This is, however, generally not the case in simulations of the crystalline state and near melting. If the system is cooled from the liquid state through the freezing transition there generally remain many defects (e.g. grain boundaries, unpaired dislocations, interstitials etc.) which cannot be annealed and which are not present in equilibrium.

This observation casts doubt on the reliability of simulations in the vicinity of the melting point, since fluctuations to the disordered (fluid) state are essentially irreversible. Also, it is important to note that, if 2d-melting is associated with dislocation unbinding, the equilibration of systems of dislocations will require extremely long time due to the fact that dislocations can move easily only in one direction, the glide direction (parallel to the Burger's vector \vec{b}), while motion in the climb direction (perpendicular to \vec{b}) requires the presence of interstitials and vacancies which should be scarce [4] in the vicinity of the melting point (particularly if a higher-order melting transition occurs).

In order to circumvent this problem of the extremely slow climb motion of dislocations in atomic systems, Saito [33] simulated a system of dislocations (with unit Burger's vectors) on a lattice. In contrast to the particle systems where dislocations are composite objects whose motion requires the correlated motions of many particles, in Saito's system the dislocations are the "particles", simulated in the grand-canonical ensemble. In this case, the diffusion of dislocations can be made essentially arbitrarily fast and independent of direction relative to the Burger's vector and thus equilibration is easier to control.

The simulation was carried out for two different values of the dislocation core energy. For the case of the larger core energy, a higher-order dislocation-unbinding transition was observed with values of the elastic constants consistent with (1) as an equality and a smooth increase of energy. For the lower value of the core energy, clear indications of a first-order transition were observed with a significantly greater

discontinuity of the elastic constants and a sharp rise of energy at T_m , as well as hysteresis. This behavior may be compared to the observations on the Lennard-Jones system [23] in which the order of the transition may depend on density. It would thus be of interest to know the dislocation core energies for different densities of the Lennard-Jones system.

So far, the core energy has only been calculated for the 2d electron solid (at $T=0$) [4]. Its value is about 50 percent larger than the larger of the core energies in Saito's simulations and thus, based on these considerations, the 2d electron solid may melt via a higher-order dislocation-unbinding transition.

Further evidence that this may be the case has been obtained from simulations by the author [34]. Two independent sets of molecular-dynamics simulations were carried out on systems with 780 and 1560 electrons subject to periodic boundary conditions and in a uniform positive background: One starting from the very-low-temperature solid ($T \approx \frac{1}{3}T_m$), the other starting from the high-temperature fluid ($T \approx 2T_m$). As the solid was heated up, the shear modulus $\mu(T)$ was determined from the frequency of transverse phonons [32]. As discussed above, agreement was obtained with the theory of dislocation-mediated melting as well as with analytical calculations of $\mu(T=0)$ and $d\mu/dT(T=0)$ [35,36,37]. At T_m , a state characterized by exponential decay of translational correlations, but by algebraically decaying orientational correlations was observed, consistent with (5). However, while this hexatic-like state was stable during the time of the simulation at this temperature (a time corresponding to 400 zone-boundary phonon periods), it is not clear that it represents a thermal-equilibrium state since I was unable to obtain such a state by cooling the liquid.

In fact, the simulations starting from the high-temperature liquid state exhibited serious equilibration problems already upon cooling to about $1.3T_m$. At temperatures between $1.3T_m$ and $1.2T_m$ a sharp increase of the orientational correlation length $\int_6(T)$ was observed [34], however with very large fluctuations between values of 2-3 a (where a is the position of the first maximum of the radial distribution function $g(r)$) and values of the order of half the system size ($>10a$ for the smaller system and $>15a$ for the larger system). These fluctuations occur in configurations separated by of the order of 100 zone-boundary phonon periods.

Since no reliable equilibrium could be obtained at temperatures as much as 20 percent above the melting point, conclusions about the order of the melting transition and about the existence of an hexatic phase cannot be convincing.

4. Simulations of the 2d - electron system in a symmetry-breaking field.

Assuming that the equilibration problems occurring around $1.2T_m$ might be caused by a large susceptibility of the system to orientational ordering fields, it may be interesting to study their effect on the system. Recently, I have carried out simulations of the 2d electron system subject to a field H_6 which couples to the orientational order parameter Ψ_6 (eq.3) [38]. This is done by adding an interaction term

$$V_6 = -\frac{1}{6} H_6 \sum_{i,j} f(|\vec{r}_{ij}|) \cos 6\theta_{ij} \quad (7)$$

where \vec{r}_{ij} is the separation vector between particles i and j and θ_{ij} is its direction. The factor $f(r)$

$$f(r) = \begin{cases} \left(\frac{c^2 - r^2}{c^2 - a_0^2} \right)^2 & r < c \\ 0 & r > c \end{cases} \quad (8)$$

cuts the summation off smoothly at a radius $c=1.37a_0$ which is approximately midway between nearest neighbor distance a_0 and second-nearest neighbor distance, and at which the radial distribution function has a minimum. Using an alternative definition of the orientational order parameter

$$M_6 = \frac{1}{6N} \sum_{i,j} f(|\vec{r}_{ij}|) \cos 6\theta_{ij} \quad (9)$$

we may write in analogy to magnetic systems

$$V_6 = -N H_6 M_6, \quad (10)$$

where N is the number of particles.

The presence of the interaction term V_6 (eq.7) leads to long-range orientational order at all temperatures and would thus eliminate the hexatic to isotropic-fluid transition at T_i (provided that the hexatic phase exists). Also, the discontinuity of $\langle M_6 \rangle$ at T_m , which is predicted even for the higher-order transition described by dislocation-mediated melting theory [1], would be replaced by a smooth variation of $\langle M_6 \rangle$ for any finite H_6 . This is in contrast to the situation one would expect if melting is a first-order transition: In this case, one would expect that the discontinuity of $\langle M_6 \rangle$ would exist even at finite values of H_6 , up to some critical value H_6^* beyond which the transition would be continuous.

One can now make predictions for the melting curve $T_m(H_6)$, assuming either that the transition is continuous at $H_6=0$ or that it is first order even for finite H_6 . In the latter case, the melting curve will be described by the Clausius-Clapeyron equation which relates the slope of the melting curve dT_m/dH_6 to the ratio $\Delta M_6/\Delta S$ of the discontinuity ΔM_6 and the melting entropy ΔS . In this case, it is also natural to expect that $\langle M_6 \rangle(T)$ has no dramatic behavior in the solid close below T_m , and the discontinuity ΔM_6 should then be obtained reliably by extrapolating its low-temperature behavior to the melting point T_m . From my simulations the discontinuity ΔM_6 at $H_6=0$ is found to be [38]

$$\Delta M_6 = 0.65 \quad . \quad (11)$$

In addition, using the value $\Delta S=0.3k_B$ [30], one predicts a slope

$$\frac{dT_m}{dH_6} = 2.2 \text{ } k_B^{-1} \quad (12)$$

On the other hand, the theory of dislocation-mediated melting predicts a melting curve $T_m(H_6)$ solely using quantities defined in the solid (λ , μ , E_c and $\langle M_6 \rangle(T)$). One so obtains [38] a slope dT_m/dH_6 which depends only weakly on H_6 and for $H_6=0$ it yields

$$\frac{dT_m}{dH_6} = 0.9 \text{ } k_B^{-1} \quad , \quad (13)$$

more than a factor of two smaller than the Clausius-Clapeyron result (12). Since the value used for ΔS is probably an upper bound for the true entropy of melting and since ΔM_6 is known quite accurately, eq.(12) is likely to be a lower bound for the slope of the melting curve at $H_6=0$, if the transition is first order.

I have carried out molecular-dynamics simulations [38] on systems with 780 and 1560 electrons subject to various different values of the symmetry-breaking field (in the range $\frac{1}{24}k_B T_m \leq H_6 \leq \frac{1}{3}k_B T_m$). Two different sets of simulations were carried out, one starting from the low-temperature solid, the other starting from the high-temperature liquid. The melting curve $T_m(H_6)$ was determined from the temperature dependence of both shear modulus (cf. [32]) and translational correlation length.

The observed melting curve $T_m(H_6)$ is in agreement with (13). Moreover, this result appears to be very reliable because for $H_6 \gtrsim 0.05k_B T_m$ the simulation of the melting transition is reversible without hysteresis in energy, correlation functions and orientational order $\langle M_6 \rangle$, and no difference between the two sets of simulations. Also, no discontinuities in energy and $\langle M_6 \rangle$ are observed and the translational correlation length ξ is found to increase dramatically in the fluid as $T_m(H_6)$ is approached from above, in a manner consistent with (2).

These results imply that at least for finite H_6 , beyond some so far unknown critical value H_6^* , melting appears to be described by the Kosterlitz-Thouless-Halperin-Nelson-Young theory. For small values of H_6 ($H_6 \lesssim 0.05k_B T_m$), the same equilibration problems as for $H_6=0$ are encountered, and thus we cannot draw definite conclusions on the asymptotic slope of the melting curve at $H_6=0$ and on the value of H_6^* , which may turn out to be zero in agreement with the theory of dislocation-mediated melting, and in this case the hexatic phase would exist.

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