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Melting in two dimensions—the current status

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Abstract. The current status of the controversy relating to melting in two dimensions is surveyed. To begin with, a review is given of the seminal work of Kosterlitz and Thouless. This is followed by a discussion of the modifications introduced by Nelson and Halperin. The search for the continuous transitions and the intermediate hexatic phase predicted by these theories is then described, covering both the laboratory as well as simulation experiments. Alternate viewpoints to the KT theory aired recently in the literature are also briefly examined. The paper concludes with an outlook for the future.

Keywords. Two-dimensional melting; computer simulation; Kosterlitz-Thouless theory; grain-boundary mechanism.

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1. Introduction

As is well known, melting is a first-order transition. This of course refers to three-dimensional solids, but then we do live in a three-dimensional world. Be that as it may, it is interesting to speculate on the nature of melting of lower dimensional solids. The question is not purely academic since many solids actually behave as if they were of lower dimensionality.

Our attention in this paper will be directed to the melting of 2D solids, a topic which during the past decade has been the subject of active research as well as of lively controversy (as yet unsettled). After discussing why 2D melting is of interest *per se*, we will explain what the controversy is all about. We will then make a broad survey of the various attempts to shed light on the controversy, and wind up with an outlook for the future. Parenthetically we remark that although our focus is restricted to 2D melting, this topic actually forms a part of the general area of phase transitions in 2-dimensions, a subject with considerable implications far beyond condensed matter physics.

2. Some preliminaries

It is pertinent as well as useful to commence our discussion with a few general remarks on order in lower-dimensional systems. Nearly fifty years ago, Peierls (1935) and Landau (1937) demonstrated that long-range order is impossible in a 2D solid in the sense that $\langle (r-\bar{r})^2 \rangle$, the mean-square fluctuation of the distance r between two atoms separated by average distance $\bar{r}$, increases logarithmically with the size of the system, in contrast to the finite value one obtains for a three-dimensional solid. In other words, crystalline order is rendered impossible due to fluctuations. One can understand this

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intuitively by considering the number of nearest neighbours in a simple cubic, a square and a one-dimensional lattice. Whereas in the three-dimensional lattice there are 6 nearest neighbours to "hold back" every atom, there is less "check" in lower dimensional systems, facilitating the disruption of order. Slightly more formally, if $\langle u^2(R) \rangle$ denotes the mean square fluctuation of atom at R, then from a consideration of the thermal agitation caused by phonons it is found (Peierls 1935) that

$$\langle u^2 \rangle \sim \int \mathrm{d}\mathbf{q}/q^2.$$

This integral converges in three dimensions but diverges in lower dimensions, implying the absence of long-range order. In physical terms, long wavelength phonons prevent the solid from fully attaining a crystalline structure.

While the above line of reasoning is no doubt persuasive, one might legitimately wonder why it is that the famous two-dimensional Ising model does not fall into this slot but shows long-range ordering instead. Indeed, stimulated by such a line of thought, there were several studies in the sixties concerning the nature of order in various 2D systems. Mermin and Wagner (1966) for example, proved that there is no spontaneous magnetization in a 2D magnet with spins having more than one degree of freedom (-for comparison, the Ising spin has only one degree of freedom). Shortly thereafter, Hohenberg (1967) demonstrated the absence of long-range order in a 2D Bose fluid.

Even as evidence began to accumulate concerning the absence of long-range order (LRO) in several 2D systems, Stanley and Kaplan (1966) discovered by analyzing the high-temperature series expansion that the susceptibility of the 2D planar magnet (the so-called XY model) diverged at a particular temperature, suggestive of a phase transition. This was quite intriguing for, till then, one was used to a phase transition accompanied by the onset of order. Did the work of Stanley and Kaplan imply that phase transitions were possible without requiring LRO? It was at this juncture that Kosterlitz and Thouless (1973) put forth their famous idea of phase transitions accompanied by a change of "topological order" rather than the conventional LRO. Understandably, great excitement was aroused, and various 2D systems were experimentally studied to check out these novel concepts. While in 2D superfluid a Kosterlitz-Thouless (KT) transition was observed as predicted (Bishop and Reppy 1978), the results on melting have been quite controversial and it is these that we shall be dwelling upon.

3. The Kosterlitz-Thouless theory

3.1 *Aspects of the 2D XY model*

As a prelude to the KT theory, we shall briefly consider the 2D XY model in order to highlight the essential physical content of the KT mechanism. We begin by labelling systems using two integers (d, n), denoting respectively the dimensionality of the system and that of the order parameter (*i.e.* the number of independent components in it). In this scheme, the Ising model would be a (2, 1) system, the Heisenberg model investigated by Mermin and Wagner (1966) would be a (2, 3) system and so on. The models of current interest (including the XY model) belong to the (2, 2) family.

The 2D XY model refers to a system of spins constrained to rotate in a plane. The spins are often taken to be organized on a lattice. However, one eventually goes to a continuum limit, whereupon the lattice structure can be ignored. Observe that the spin vector can *continuously* rotate and has an infinity of orientations to choose from, in contrast to the Ising spin which has only two options. This leads to an important difference in the group structure of the Hamiltonian. Whereas the Ising Hamiltonian has the discrete symmetry Z_2 (group of two elements), the Hamiltonians of some of the systems we shall be discussing all have the continuous symmetry O(2). Particle physicists have strong interest in 2D O(n) models of various types (Barber 1980).

Passing reference must also be made to the Potts model (1952) which is a generalization of the Ising model wherein the order parameter can take q distinct values. For $q = 2$ (in 2D), the model reduces to the Ising model while in the limit $q \to \infty$ one recovers the XY model. The Potts model thus interpolates between the Z_2 and O(2) models.

Returning to the XY model, its Hamiltonian may be expressed as

$$H = -J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \tag{1}$$

where $J > 0$ and the sum $\langle ij \rangle$ is over-nearest neighbour pairs only (on a square lattice, say). Introducing for convenience the complex notation

$$\psi = S_x - iS_y = |\mathbf{S}| \exp(i\theta),$$

and taking $|\mathbf{S}| = 1$,

$$H = -J \sum_{\langle ij \rangle} \cos(\theta_i - \theta_j). \tag{2}$$

At low temperatures where spins on neighbouring sites are expected to be highly correlated, we may expand the cosine and retain only the quadratic term, obtaining

$$H = \frac{J}{2} \sum_{\langle ij \rangle} (\theta_i - \theta_j)^2. \tag{3}$$

The above Hamiltonian corresponds to the spin wave approximation (Wegner 1967) and is the starting point for the calculation of various physical properties. Of special interest is

$$C(r) \equiv \langle \psi(0) \psi^*(r) \rangle, \tag{4}$$

the correlation function. Analysis shows

$$C(r) = r^{-\eta(T)} \text{ for } r \to \infty, \tag{4a}$$

where

$$\eta(T) = k_B T / 2\pi J. \tag{4b}$$

This result confirms the absence of LRO, but its form is unexpected.

To appreciate this last remark, let us briefly recall the corresponding result for the Ising model (see figure 1a). In the disordered phase, correlations decay exponentially but *precisely* at the critical point, they have a power law behaviour (Stanley 1971). By contrast, the spin wave approximation to the XY model exhibits a power law behaviour at *all* temperatures (see figure 1b). KT proposed that if one goes beyond the spin wave approximation and includes appropriate topological defects, the line of critical points

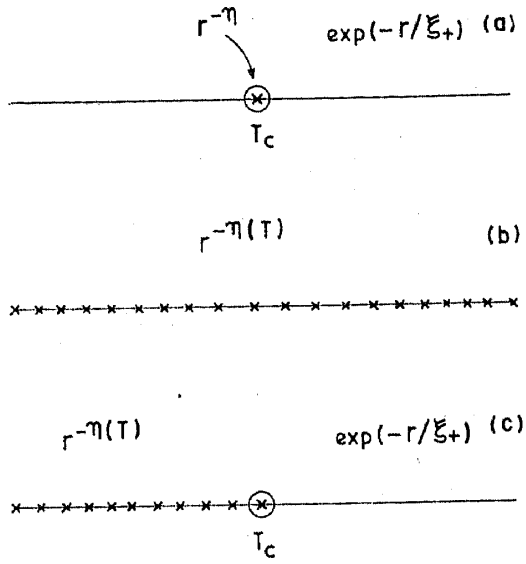

Figure 1. Schematic summary of the phase transitions of the 2D Ising (a) and XY models (b), (c). In (b) is shown the XY model in the spin wave approximation where every temperature is a critical temperature. When vortices are included, the line of critical points is terminated at T_c as shown in (c), with a transition in the correlation function (After Young 1980.)

would terminate at a finite temperature T_c (see figure 1c). Below that temperature there is power law behaviour while above there is exponential decay. The transition is thus characterized by *changes in the correlation function* rather than by the appearance or disappearance of LRO. As we shall presently see, these changes are linked to the topological order in the system.

3.2 *Role of topological defects*

How do topological defects alter the character of the system at T_c as sketched in figure 1c? To answer this question, consider the vortex in figure 2a. This and its companion in figure 2b are outside the purview of the spin wave approximation. Observe that either of these vortices is capable of strongly disturbing spins far from the region of the singularity. By contrast, in the spin wave approximation there are no such long-range perturbing agencies (see figure 2c). However, with or without vortices, $C(r)$ always decays as $r \to \infty$ *i.e.* there is no LRO.

Vortices are like charges, and can be labelled with integers N positive and negative. Those in figure 2 are the simplest, with $|N| = 1$. The energy of an isolated vortex may be calculated from (2) by using the continuum approximation, and is found to be (Kosterlitz and Thouless 1973)

$$E(\text{vortex}) = E_c + \int_{a_0}^{R} 2\pi r \mathrm{d}r \frac{J}{2} |\nabla\theta|^2$$
$$= E_c + \pi J \ln (R/a_0). \tag{5}$$

Here R is the radius of the system and a_0 is a distance of atomic dimensions characterizing the core. E_c is the core energy associated with the region $r < a_0$. Notice that the energy of an isolated vortex tends to infinity if the radius of the system goes to

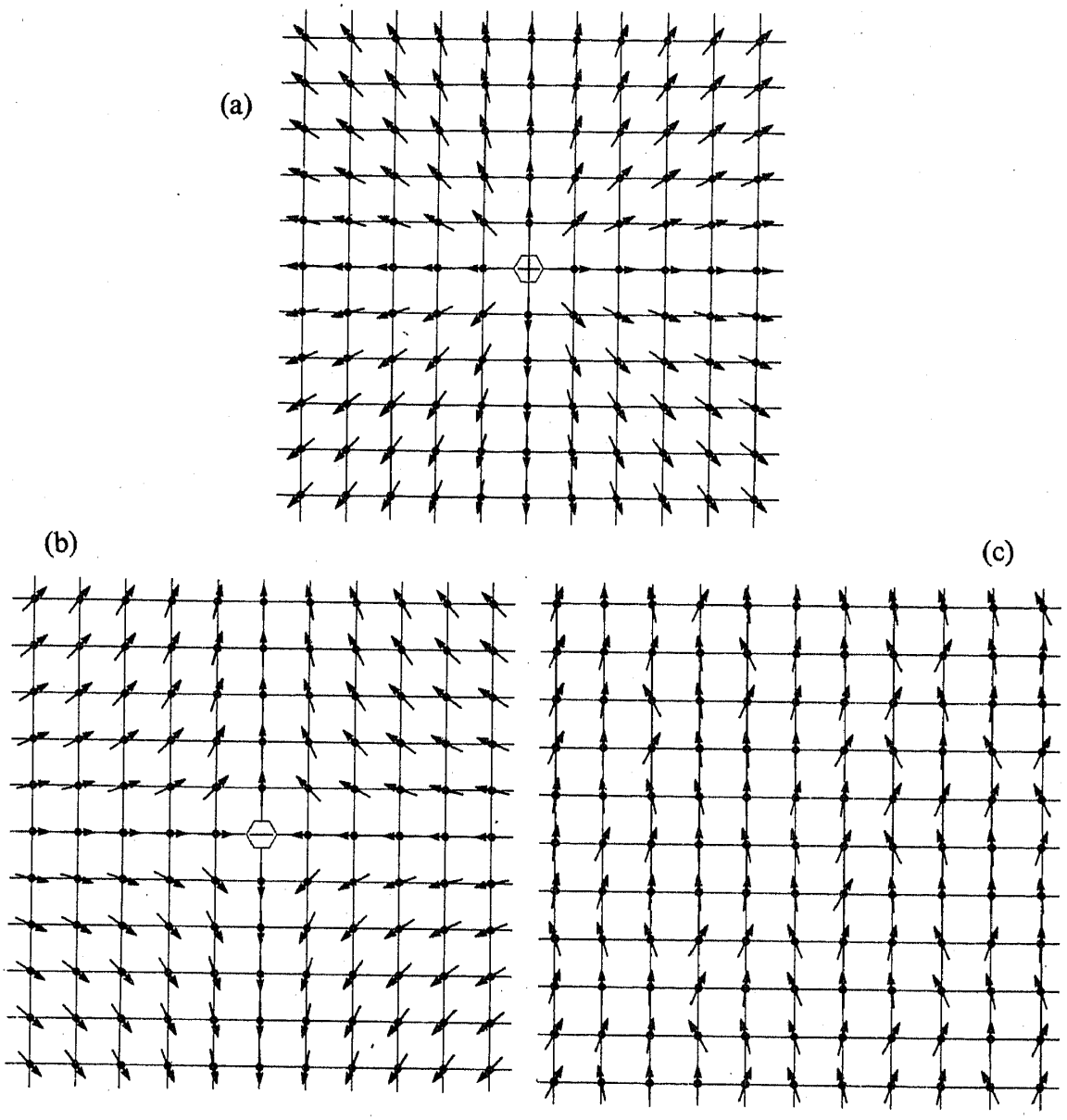

Figure 2. Some possible spin arrangements of the 2D XY system. (a) and (b) depict the situation in the presence of vortices, and clearly there is a substantial disturbance. By contrast, there is less disturbance in (c) which illustrates the spin wave approximation.

infinity, implying that it would be very difficult to create a vortex in the thermodynamic limit. As we shall presently see, this statement will require some qualification.

Next consider the vortex-antivortex pair in figure 3. On account of a mutual cancellation effect, the spins at large distances are no longer severely disturbed, the disturbance being confined to the neighbourhood of the pair. As may be expected, the energy of the pair is finite, being given by

$$E(\text{pair}) = 2E_c + 2\pi J \ln(r/a_0), \tag{6}$$

where r is the separation.

Going back to the single vortex, although its creation is energy-expensive, there is an entropy advantage associated with the fact that one can locate the vortex in various

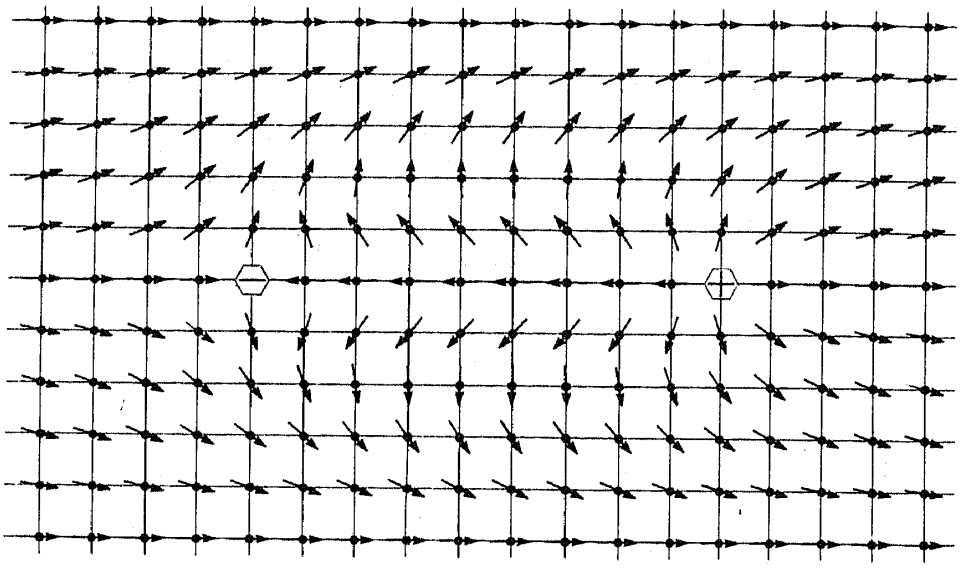

Figure 3. Vortex-antivortex pair in the 2D XY system.

ways. This entropy is $2k_B \ln (R/a_0)$ so that the free energy is

$$F = E - TS$$
$$= (\pi J - 2k_B T) \ln (R/a_0), \tag{7}$$

implying that for $T > T_c$ where

$$T_c = \pi J/2k_B, \tag{8}$$

free vortices may indeed exist even in the thermodynamic limit.

Based on the above arguments, KT proposed the following scenario: At low temperatures, only vortex pairs can exist. These are thermally generated and ride over a spin wave background. Since (bound) pairs do not cause serious perturbations at large distances (see figure 3), $C(r)$ is essentially that given by the spin wave approximation. Above T_c, the pairs dissociate since the system can now support free vortices. Technical considerations (to be amplified later) demand that the number of vortices be small. Even so, they drastically modify $C(r)$ (-see also figure 2). As already noted, with such strong fluctuations present, the spin wave approximation does not apply. Further, like their counterparts in solids namely dislocations, vortices can move freely when subjected to suitable forces. The system as a whole therefore acquires flow properties and exhibits a concomitant loss of (spin) rigidity at T_c. This then is the central feature of the transition, the unbinding of pairs at T_c and the related loss of the corresponding rigidity.

3.3 *Topological order*

Though conventional LRO is absent, KT suggest that one may differentiate between the phases on either side of T_c *via* the concept of topological order. Briefly, topological order is an assessment of whether the topological defects are bound ("ordered") or unbound ("disordered"). To make such an assessment, one makes a large closed circuit in the system and measures the phase angle all along the boundary. The total change of

the phase angle will be determined by the number and strength of the singular points enclosed by the path. In the high temperature phase (figure 4a), there will be a preponderance of isolated singularities. The number enclosed will be proportional to the area A of the contour C and the average total phase change will be proportional to the length L of C. In the low-temperature phase (figure 4b), the average total phase change will be determined by the number of pairs cut by the path, and will be proportional to $(Ld)^{1/2}$ where d is the mean separation of the pairs. Based on this criterion, one can recognize a change in the topological order at T_c. It is worth noting that a similar assessment by traversing a closed contour is also made in lattice gauge theories (Kogut 1979).

3.4 *2D melting—The KT view*

We turn now to melting in two dimensions. Analogous to the spin wave approximation, we have the harmonic (phonon) approximation. Introducing

$$\rho_{\mathbf{G}}(\mathbf{R}) \equiv \exp(i\mathbf{G}\cdot\mathbf{R}), \tag{9}$$

where $\mathbf{G}$ denotes a reciprocal lattice vector, we may as usual define the correlation function

$$C_{\mathbf{G}}(\mathbf{R}) = \langle \rho_{\mathbf{G}}(\mathbf{R})\rho_{\mathbf{G}}^*(0)\rangle, \tag{10}$$

which, for the 2D solid, varies as

$$\lim_{\mathbf{R}\to\infty} C_{\mathbf{G}}(\mathbf{R}) \sim R^{-\eta_{\mathbf{G}}(T)}. \tag{11}$$

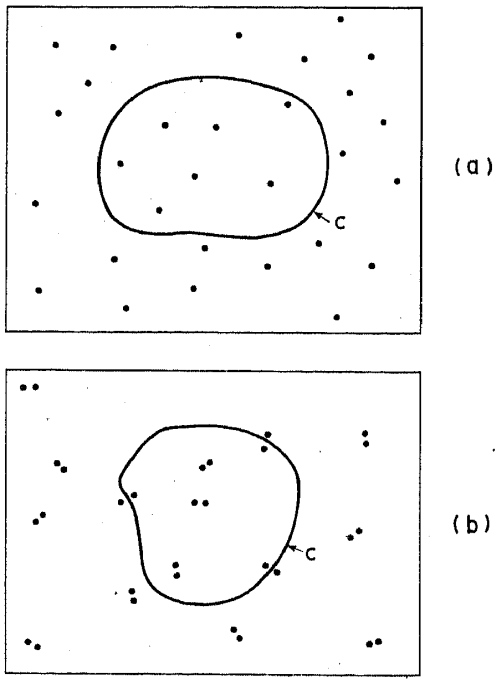

Figure 4. Assessment of topological order is made by going round a simple closed contour and measuring the overall change in phase angle. As discussed in text, the result depends on whether the topological defects are unpaired (as in (a)) or are paired (as in (b)).

One can now calculate the structure factor

$$S(\mathbf{q}) = \langle |\rho(\mathbf{q})|^2 \rangle, \tag{12}$$

whereupon one finds that the familiar δ-function Bragg singularities *i.e*

$$S(\mathbf{q}) \sim \delta(\mathbf{q} - \mathbf{G})$$

become "smeared" into power law singularities (Jancovici 1967) *i.e.*,

$$S(\mathbf{q}) \sim |\mathbf{q} - \mathbf{G}|^{-2+\eta_G(T)} \tag{13}$$

One therefore has a "quasi" crystal which, incidentally, is the reason why one may still label the diffraction peaks with the set $\{\mathbf{G}\}$. Well-defined peaks in $S(\mathbf{q})$ can be observed provided $\eta_G < 2$.

Now dislocation-mediated melting is really an old concept in three dimensions, going back to the thirties (in a sense to Mott and Gurney 1939; for a review, see Cotterill 1980). With respect to two dimensions*, KT note that at low temperatures, the system, though lacking crystallinity (see equation (13)), is a solid in that it is able to resist external shear. Besides phonons, the system also supports bound dislocation pairs which are thermally generated. The latter neither upset the power law decay of $C_G(\mathbf{r})$ (see equation (11)) nor the rigidity. Above T_m the melting temperature the pairs break, and once free dislocations appear the system loses (elastic) rigidity, signalling a transition to the fluid state. The topological order can be assessed as earlier *i.e.* by making the Burgers circuit and measuring the failure in the path closure.

Regarding the details of the transition, one clearly needs a more refined analysis than contained in equations (5)–(8). The crucial feature in such an analysis is the screening of the pair interaction (6). The underlying physics is better appreciated by considering the analogous case of a 2D gas of charged particles† with charges $\pm q$, electrically neutral on the whole, and interacting *via* the logarithmic potential

$$\begin{aligned} U(|\mathbf{r}_i - \mathbf{r}_j|) &= -2q_i q_j \ln \left| \frac{\mathbf{r}_i - \mathbf{r}_j}{a_0} \right| + 2\mu, \quad r > a_0 \\ &= 0, \qquad\qquad\qquad\qquad r < a_0. \end{aligned} \tag{14}$$

Here q_i and r_i are the charge and position of the ith particle. 2μ is the energy required to create a pair of particles of equal and opposite charge a distance a_0 apart. In 2D solids, the analogue of μ is the dislocation core energy E_c (-see also equation (5)). The quantity a_0 is an appropriate cut-off of the order of the particle diameter or, for a lattice, the lattice spacing. One expects that due to (14), opposite charges can link to form bound pairs, *i.e* dipoles. For these pairs to unbind, $U(r)$ must decrease, which can happen by screening.

Focusing attention on a particular dipole of size r, the interaction between the two members constituting the dipole will be screened by (smaller) pairs lying within the

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* For historical completeness it must be remarked that Feynman had independently proposed the idea of dislocation-assisted melting in two dimensions. Unfortunately, this work is unpublished. An outline of this theory is given by Elgin and Goodstein (1974) and by Dash (1978).

† By now the reader would undoubtedly have noticed that there are a variety of 2D systems. For elucidating various features of the KT theory, we cite different examples as convenient!

range of the field. These pairs will in turn be screened by others lying in their respective fields and so on. One thus has a scaling situation as in the case of critical fluctuations present during a conventional second-order phase transition, and not surprisingly, renormalization group techniques make their entry.

Owing to screening, (14) becomes modified to

$$U_{\text{eff}}(r) = \frac{2q^2 \ln (r/a_0)}{\varepsilon(r)}, \tag{15}$$

where $\varepsilon(r)$ is the space-dependent dielectric function, and includes the polarizability of pairs with separation less than r. Introducing $p(r)$ the probability of finding a pair with separation r, one finds that $\varepsilon(r)$ and $p(r)$ are coupled as follows (Kosterlitz 1974; Halperin 1979):

$$\begin{aligned}\varepsilon(r+\mathrm{d}r) &= \varepsilon(r) + 4\pi \times \text{pol. contributed by pairs in the range } r \text{ and } (r+\mathrm{d}r) \\ &= \varepsilon(r) + 4\pi \frac{r^2}{2T} 2\pi r\, p(r)\, \mathrm{d}r, \\ p(r+\mathrm{d}r) &= p(r) \exp\left[-\frac{2}{rT\varepsilon(r)}\right] \mathrm{d}r. \end{aligned} \tag{16}$$

Here $r^2/2T$ is the polarizability of a pair with separation r and $[2\mathrm{d}r/r\varepsilon(r)]$ is the work done in increasing the separation of the pair by the amount $\mathrm{d}r$. Using an iterative procedure, one obtains from (16) the famous Kosterlitz recursion relations (Kosterlitz 1974). Such relations can be written down for every (2, 2) system *i.e.* the XY model, 2D crystal etc.

We turn to the outcome of the recursive relations in the specific case of 2D crystal. Two crucial quantities are:

(i) $$y = \exp(-E_c/k_B T), \text{ and}$$

(ii) $$K = \frac{4\mu(\mu+\lambda)}{(2\mu+\lambda)} \frac{a^2}{k_B T}, \tag{17}$$

where μ and λ are the Lamé constants and a the lattice spacing. During the renormalization calculations, one has to rescale the dislocation core diameter from a_0 to $a_0 \exp l$ causing y and K to become l-dependent. The recursive relations are of the form (Kosterlitz 1974; Nelson 1978; Nelson and Halperin 1979; Young 1979):

$$\begin{aligned}\frac{\mathrm{d}y(l)}{\mathrm{d}l} &= f_1(y(l), K(l)) \\ \frac{\mathrm{d}K^{-1}(l)}{\mathrm{d}l} &= f_2(y(l), K(l)). \end{aligned} \tag{18}$$

The solution of these equations is too intricate to be discussed here. Of greater interest are the renormalization group flows illustrated in figure 5. The different trajectories effectively determine the system behaviour at different temperatures.

Let us follow a typical trajectory for $T < T_m$ from the starting line (see figure 5). The flow takes us to the abscissa where $y(l)$ is zero and $K(l)$ has a finite value, which implies that (i) the probability of finding a free dislocation is zero and (ii) the renormalized

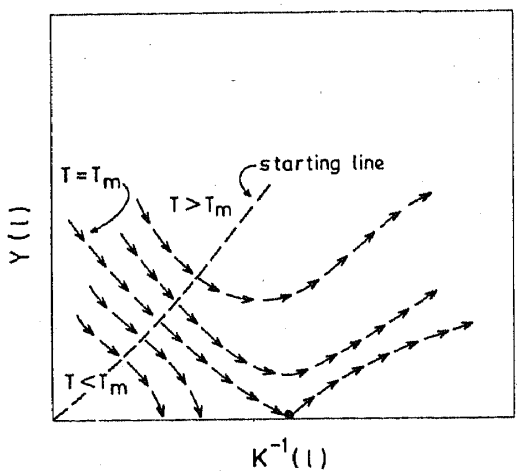

Figure 5. Renormalization group flow in the KT theory. The quantities K and y are defined in (17). The line for $T = T_m$ is the separatrix. The dislocation probability $y(l)$ tends to zero in the region to the left of the incoming separatrix which is the crystalline phase. The trajectories to the right flow away to large y-values, signalling instability and the dissociation of dislocation pairs.

stiffness is finite. The trajectory for $T = T_m$ is the separatrix, and trajectories to its right flow away, implying that for $T > T_m$ (i) free dislocations can be found and (ii) the system loses rigidity since $K(l) \to 0$. The system behaviour close to T_m may be carefully examined by zooming near the region where the separatrix curve meets the abscissa. The principal findings are:

(i) $$C_G(\mathbf{R}) \sim R^{-\eta_G(T)}, \qquad T \to T_m^-, \tag{19}$$

where $\eta_G(T)$ is related to the renormalized Lamé constants $\mu_R(T)$ and $\lambda_R(T)$ by

$$\eta_G(T) = k_B T |\mathbf{G}| \frac{(3\mu_R + \lambda_R)}{4\pi\mu_R(2\mu_R + \lambda_R)}. \tag{20}$$

At T_m, the exponent for the first Bragg point for a triangular lattice cannot exceed (1/3).

(ii) $$C_G(\mathbf{R}) \sim \exp\left[-\frac{R}{\xi_+(T)}\right], \qquad T \to T_m^+, \tag{21}$$

the correlation length ξ_+ diverging according to

$$\xi_+ \sim \exp\left[\frac{\text{const}}{(T-T_m)^{\bar{\nu}}}\right], \qquad \bar{\nu} = 0{\cdot}36963. \tag{22}$$

(iii) The renormalized stiffness constant $K_R(T)$ (see equation (14)) varies as

$$K_R^{-1}(T \to T_m^-) \sim \frac{1}{16\pi}(1 - C|t|^{\bar{\nu}}), \tag{23}$$

where C is a positive, nonuniversal constant and $t = (T - T_m)/T_m$. Above T_m the stiffness vanishes; K_R therefore abruptly jumps to zero from the value 16π at $T = T_m$.

(iv) The specific heat has only an essential singularity. Physically this is related to the fact that at T_m the density of dislocation is rather low. There is therefore very little

entropy associated with it. However above T_m there is a small bump (Berker and Nelson 1979).

Next to the order parameter, the quantities of interest in a conventional phase transition are those which exhibit singularities. In the case of the KT transition, there is no ordering in the conventional sense. The specific heat also is not very informative. The only meaningful signatures are the translational correlations $C_G(\mathbf{R})$ on either side of T_m (see (19) and (21)), and the temperature dependence of the stiffness (see (23)), all of which are conveniently studied *via* x-ray and neutron scattering.

A few other related points must also be noted at this juncture. Firstly, the calculation of the screening *via* (16) is a self-consistent one, and for this reason the KT theory is essentially a mean field type of theory. Secondly, while solving the recursive equations, the approximation is made that y is small (*i.e* defect density is small; see also remark (iv) above), implying that the core energy E_c is large (see (17)). So the picture ($T > T_m$) is that of a dilute system of dislocations floating about in an otherwise harmonic solid. Should the cores overlap, the theory collapses. In a superfluid, the vortex core size is ~ 1 Å and overlap is not a serious problem. On the other hand, the core size of a dislocation could be several lattice spacings, and overlap becomes a real possibility (Kosterlitz 1980). One must therefore be ready for surprises. We shall revert to this question later. Finally, the transition is continuous; note especially the divergence of the correlation length (equation (22)), reminiscent of a conventional second-order phase transition. (However, a significant difference is that ξ has an exponential rather than a power law behaviour, Stanley 1971.)

3.4 *Role of orientational correlations*

About five years after KT advanced their theory, Halperin and Nelson (1978; see also Nelson and Halperin 1979) drew attention to an important aspect of the problem not considered by KT*. Nelson and Halperin (NH) argued that besides translational correlations, one must also consider orientational correlations, and that when a solid melts to become an *isotropic* liquid, it sheds *both* these correlations. NH therefore introduce an orientational order parameter ψ. Most of the focus so far has been on triangular lattices which are the most close packed structures in two dimensions. For this lattice, the bond orientational parameter is given by

$$\psi(\mathbf{r}) = \exp[i6\theta(\mathbf{r})], \tag{24}$$

where $\theta(\mathbf{r})$ is the orientation relative to the bond between nearest neighbour atoms. θ is related to the displacement field $\mathbf{u}(\mathbf{r})$ by

$$\theta(\mathbf{r}) = \frac{1}{2}\left(\frac{\partial u_y(\mathbf{r})}{\partial x} - \frac{\partial u_x(\mathbf{r})}{\partial y}\right). \tag{25}$$

Analogous to (10), one now has a orientational correlation function

$$C_6(\mathbf{r}) \equiv \langle \psi(0)\psi^*(\mathbf{r}) \rangle. \tag{26}$$

Extending attention to orientational effects requires one now to consider new

* Taking into account a related paper by Young (1979), the melting theory is often referred to as KTHNY theory.

defects, namely disclinations. These are "rotational singularities", two examples of which are illustrated in figure 6. They are formed by taking a perfect triangular lattice and either removing or adding a wedge. The curvatures resulting thereof are significant (see, for example, Nelson 1983a) but we shall not consider them here. The interesting point about disclinations is that a dislocation can be viewed as resulting from the binding of a disclination pair, as illustrated in figure 6. In other words, a dislocation can be regarded as a disclination dipole.

Based on the above additional concepts, NH sketch a scenario for melting involving not only dislocations but disclinations as well. At low temperatures, the dislocations are paired, and, on account of the internal structure of the dislocation (see figure 6), the dislocation pair can be viewed as a disclination *quadrupole*. The harmonic phonons as usual disrupt positional LRO leading to a power law behaviour for the translational correlations as in (11), but, by contrast, the orientations exhibit LRO *i.e.*,

$$\lim_{r \to \infty} C_6(r) = |\langle \psi \rangle|, \tag{27}$$

a constant which is temperature-dependent.

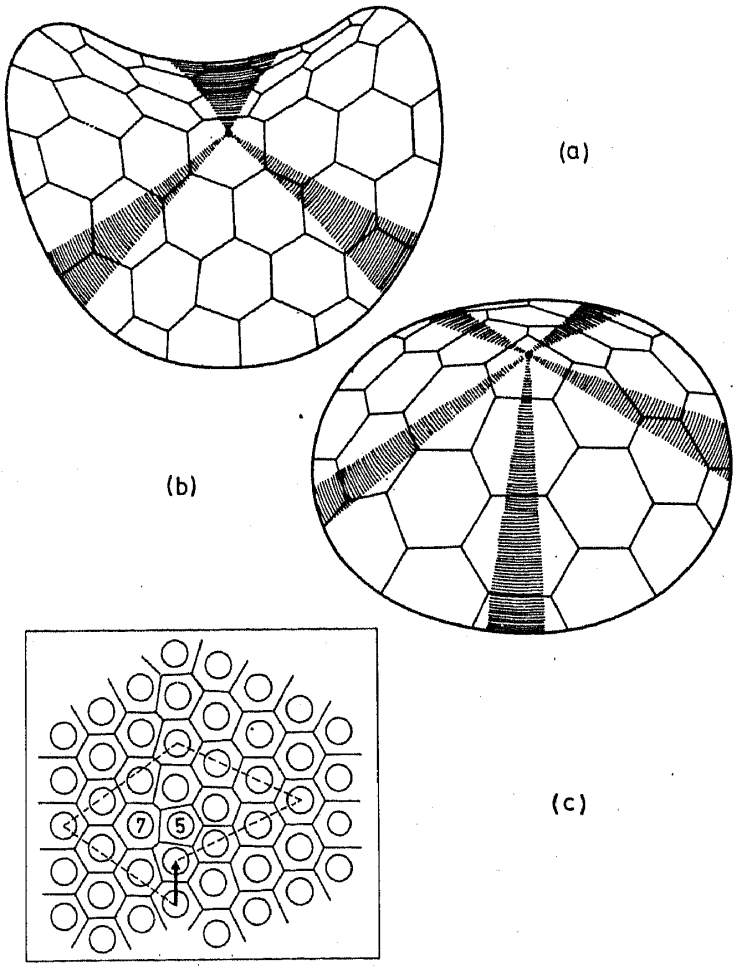

Figure 6. (a) and (b) illustrate respectively five- and seven- fold rotational singularities (*i.e.* disclinations). For convenience, the atoms are suppressed and only the surrounding Wigner-Seitz cells are shown. (c) illustrates how a pair of disclinations can combine to form a dislocation. Here the atoms are shown along with their W-S cells.

As the temperature is gradually increased, a stage is reached ($T = T_m$) where dislocations unbind. This, however, does not produce an isotropic liquid; instead the system enters an intermediate phase in which, $C_G(\mathbf{r})$ decays exponentially as in (21) but rotational correlations, though asymptotically decaying, are still strong, being given by

$$C_6(r) \sim r^{-\eta_6(T)}, \qquad T \to T_m^+, \tag{28}$$

where

$$\eta_6(T) = 18\, k_B T/\pi K_A(T). \tag{29}$$

The quantity $K_A(T)$ called the Frank constant is related to the so-called bending energy of orientational fluctuations as

$$H_A = \frac{1}{2} K_A \int \mathrm{d}^2 r (\nabla\theta)^2. \tag{30}$$

In effect, K_A is a measure of the "angular stiffness' of the system. Below T_m, $K_A = \infty$ indicative of perfect angular correlations but above T_m, there is a temperature dependence given by

$$K_A(T) \sim \xi_+^2(T), \qquad T \to T_m^+, \tag{31}$$

where $\xi_+(T)$ is the same as in (22). This divergence is of course linked to the appearance of long range *orientational* order below T_m (see (27)).

The phase with properties as in (21), (28) and (31) is like a liquid crystal in that it has greater orientational order (power law decay) than translational order (exponential decay). This phase is frequently referred to as a hexatic phase.

When heated, the hexatic phase in turn undergoes a transition at a temperature T_i, becoming an isotropic liquid. This occurs *via* the unbinding of the disclination pair, causing a loss of the orientational correlations present in the hexatic phase. Thus, in the NH picture, the free topological defects in the isotropic liquid are not the dislocations but the disclinations. As noted by NH, the transition at T_i belongs to the same universality class as the two-dimensional superfluid and the XY model, whence,

$$\eta_6 \to \tfrac{1}{4} \quad \text{as} \quad T \to T_i^-. \tag{32}$$

The renormalized Frank constant jumps discontinuously to zero at T_i from the value $72\, k_B T/\pi$ to which it decreases as $T \to T_i^-$. Above T_i,

$$C_6(r) \sim \exp(-r/\xi_6(T)) \tag{33}$$

with

$$\xi_6(T) \sim \exp\left[\frac{b}{(T-T_i)^{1/2}}\right]. \tag{34}$$

As usual, there is only an essential singularity in the specific heat. In short, at T_i there is again a crossover from a power law to exponential decay, but this time, of the orientational correlation function. Accompanying this change is a vanishing of the orientational stiffness.

Figure 7 paraphrases the features of the original KT theory and the subsequent NH modification. In passing we note that although Halperin and Nelson (1978) also predicted continuous transitions, they were cautious enough to observe that they "cannot rule out other mechanisms for melting, perhaps leading to a first-order transition".

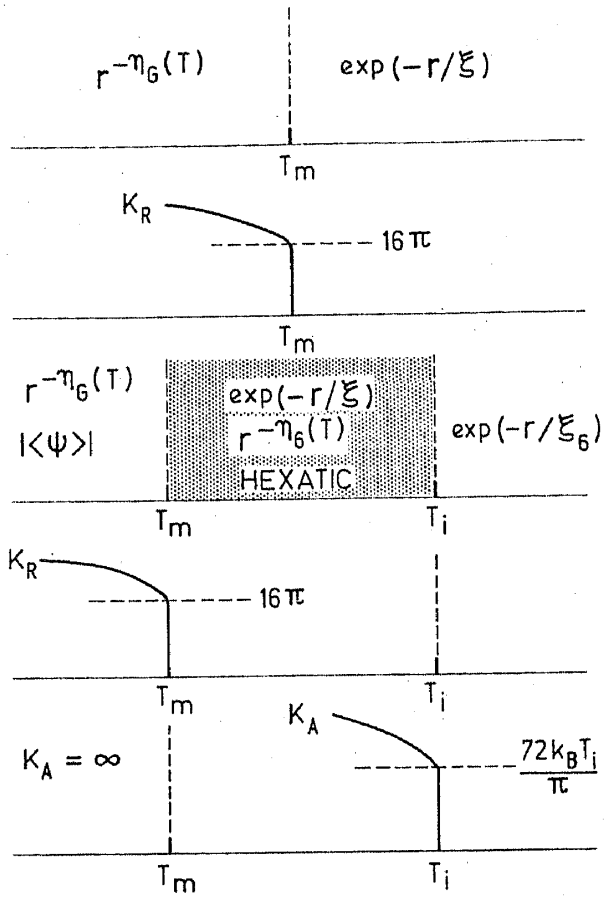

Figure 7. Comparison of the transitions predicted by the original KT theory and by the subsequent NH modification. For explanation, see text.

Laboratory experiments on 2D melting are frequently performed on layer systems supported by a substrate. NH have examined the role played by the substrate by adding an orienting field to the defect Hamiltonian, and find the following: If the substrate can be regarded as smooth, then the considerations of figure 7 continue to remain valid. On the other hand, if the periodic nature of the perturbing potential from the substrate is significant, then one must examine whether the periodicity of the substrate is commensurate or not with that of the layer above (see also figure 8). Of interest is the case where the substrate potential is weak and incommensurate. In this situation, there is only one transition, namely, that at T_m; the other one at T_i is washed out (for a triangular lattice).

A second question relates to phase diagrams. In condensed matter, we are used to diagrams as in figures 9a and 9b. One would like to know how these are modified in the light of the KTHNY theory. The outcome is sketched in figure 9c and 9d, and will become relevant later.

The prelude has been somewhat lengthy but is indicative of the fact that 2D systems are a theorist's paradise! As we have seen, topological defects in 2D are point defects. It turns out that the statistical mechanics of these is a lot simpler than that of line defects (with which one has to deal in 3D). Hence the great popularity of 2D systems with theoreticians. We turn now to the various attempts made to check the KTHNY theory.

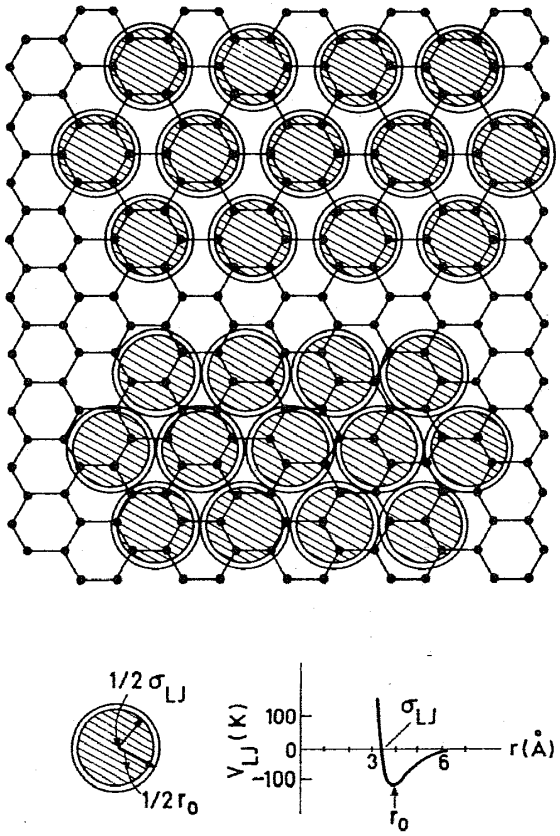

Figure 8. Schematic illustration of commensurate and incommensurate coverage of graphite by argon atoms. The Ar-Ar Lennard-Jones potential is sketched at the bottom. (After Mc Tague *et al* 1980b).

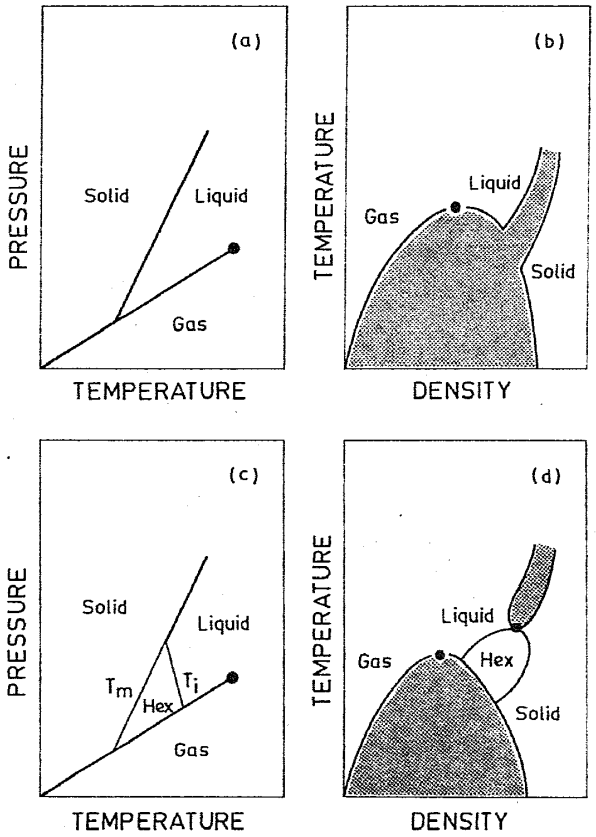

Figure 9. Sections of phase space to illustrate the modifications brought about by the KTHNY theory (see (c) and (d)). The dots indicate the critical point. The thick lines in (a) and (c) represent first-order transitions while the thin line in (c) denotes a continuous transition. The hatched regions in (b) and (d) represent regions of coexistence.

4. Experimental studies

4.1 *Laboratory experiments*

There are a number of candidate systems like liquid crystal films, electron films on liquid helium, etc which are suitable for experimentally checking out the KTHNY theory. Many of these have indeed been investigated (Sinha 1980) but the most detailed studies so far have been on rare gas films absorbed on graphite.

The basal *i.e* (0001) plane of graphite is a good substrate for forming monolayers by physisorption since it presents a relatively smooth potential to the layer absorbed. Unfortunately, large single crystals of graphite are not available but this has not deterred experimentalists. Instead they have skilfully exploited various exfoliated graphite products. Basically all of these are built up of small flakes a few μm broad and $\sim$ 100–300 Å thick. The useful size is characterized by the coherence length L (see figure 10). The *c* axes of these flakes are all partially oriented about the normal to the sheet but the azimuthal orientation about the *c* axis is random. Various types of exfoliated graphite like Grafoil, UCAR-XYZ and Papyex are now commercially available. Very popular (especially in the past), Grafoil has L $\sim$ 200 Å, $\Delta\theta \sim 25°$ (see figure 10) and a specific area for adsorption $\sim$ 30 m^2/g. The samples (for scattering experiments) typically consist of a pile of discs cut from sheets of exfoliated material. Samples with several grams of Grafoil are not uncommon. Very recently, "single crystals" of exfoliated graphite have been prepared (Clarke *et al* to be published; quoted in Rosenbaum *et al* (1983)) with L $\sim$ 400 Å and $\Delta\theta \sim 3°$.

Although notionally the graphite substrate may be deemed to exert only a weak influence, the role of the substrate cannot be entirely overlooked. As already noted in figure 8, the adsorbed layer could be commensurate* as well as incommensurate with the substrate. Our interest is in the latter situation.

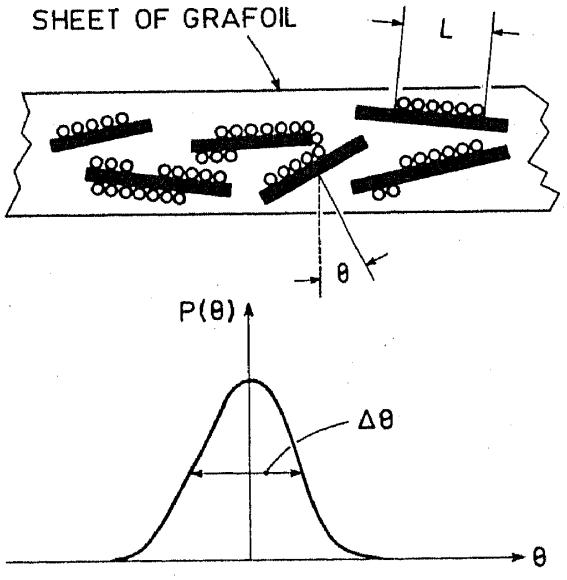

Figure 10. Schematic drawing of the Grafoil sheet. *L* indicates the typical coherence length of the adsorbed layers while $P(\theta)$ describes the distribution arising from the non-parallelism of the substrate surfaces (After Nielsen *et al* 1980).

* It is interesting that if the adsorbed atoms are in registry with the substrate, $\langle u^2 \rangle$ does not diverge (Kosterlitz 1980).

Monolayers of Ar, Kr and Xe on graphite have been studied using both neutron and x-ray scattering (Thorel *et al* 1976; Taub *et al* 1977; Horn *et al* 1978; Stephens *et al* 1979; Hammond *et al* 1980; Heiney *et al* 1982; McTague *et al* 1982; Heiney *et al* 1983) and the opinion has emerged that Xe on graphite is by far the best representative of 2D field-free system (McTague *et al* 1982). Accordingly we shall examine the recent high resolution results of Heiney *et al* (1983) obtained at the Stanford Synchrotron Radiation Laboratory (SSRL). These experiments are a sequel to an earlier series done at MIT using a rotating anode source but with somewhat lower resolution.

Heiney *et al* have criss-crossed the phase diagram along various trajectories (including constant temperature ones) in order to check out different possibilities. Figure 11 shows representative profiles observed in one of these runs. The asymmetric lineshapes are due to excess high q scattering arising from substrate azimuthal disorder. Around 152 K the lineshape changes, going over to a Lorentzian charateristic of exponential decay of the correlation function. Heiney *et al* reckon that a transition

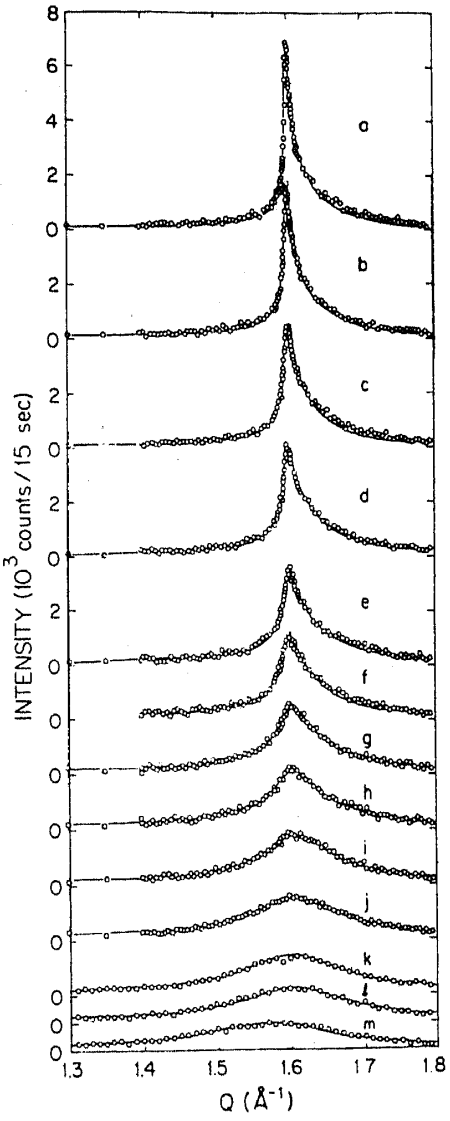

Figure 11. Diffraction profiles of the (1, 0) spot of xenon adsorbed on graphite. The scans have been made in the narrow region between 151·6 K to 160 K (After Heiney *et al* 1983).

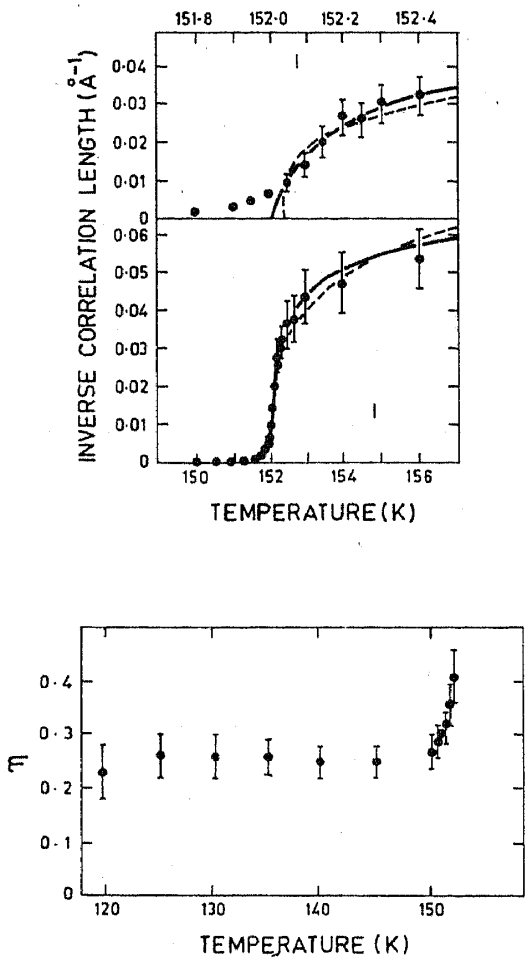

Figure 12. At the top are shown the inverse correlation lengths obtained by Lorentzian fits to the data of figure 11. The solid lines are fits to the KTHNY form while the dashed lines are power law fits. At the bottom is shown the temperature dependence of η (After Heiney *et al* 1983).

occurs at 152 K. The question whether it is of first order or continuous has been examined quite carefully. Firstly, no hysteresis is observed. Secondly, as the transition temperature is approached from below, the exponent obtained from the lineshape analysis (Dutta and Sinha 1981) has the trend shown in figure 12. At the melting temperature it is found that $0{\cdot}27 \leqslant \eta$ (melting) $\leqslant 0{\cdot}42$, reasonably consistent with KTHNY prediction $0{\cdot}25 \leqslant \eta(T_m) \leqslant 0{\cdot}33$. Above 152 K where the line shape is consistent with (22), analysis yields $\bar{\nu} = 0{\cdot}4$ (see also figure 12).

The evidence cited above no doubt strongly favours the KTHNY theory. Nevertheless Heiney *et al* have carefully examined possible consistency of their data with first-order transition. If the transition is indeed of first-order then near the transition temperature one must encounter a coexistence patch. Assuming that the solid material is converted into a liquid according to a lever law, they have analyzed their data. From the values of χ^2 obtained they conclude that the observed melting is consistent with a continuous transition although a possible two-phase coexistence in a very narrow range between 151·6 and 151·95 K cannot be ruled out.

Recently, Rosebaum *et al* (1983) studied the melting of monolayer xenon on the surface of exfoliated single crystals, using x-ray scattering. According to the NH theory,

in the solid one must get six (1, 0) Bragg spots, which, owing to the lack of LRO, will have some width both in the radial as well as angular directions with probably more in the former. On melting into the hexatic phase, the six spots still survive* with, however, enhanced width in both directions. As temperature is increased, the changes in the orientational correlations will be more rapid than the corresponding ones in the positional correlation. Consequently the angular width will increase more rapidly compared to the radial width until at T_i the spots merge to form a liquid ring. Such a behaviour (see figure 13) is in fact seen in experiments. Rosenbaum *et al* categorically rule out from the observed lineshapes, the possibility of a liquid-solid coexistence. Nor do they believe that the observed spots could be due to an isotropic liquid subjected to a substrate field. The field strength required would be three orders of magnitude too high. They are confident that their experiment offers evidence for melting of the 2D solid into an orientationally ordered liquid.

Besides adsorbed layers, melting of liquid crystal films also have been investigated to test the validity of the KTHNY theory. An important feature of these experiments is the use of freely suspended films, the preparation of which is schematically illustrated in figure 14. While a monolayer film has yet to be realized, films with as low as two layers have been successfully used.

Moncton and coworkers (Moncton and Pindak 1979, 1980; Moncton *et al* 1982) have been particularly active in the study of x-ray diffraction from such films. An early experiment (Moncton and Pindak 1980) on a material called 40·8 (butyloxybenzylidene octylaniline) seemed to suggest that the melting was continuous with η at melting in the range specified by the KTHNY theory. However, films thinner than 4 layers could not be studied owing to film rupture problems. Subsequently, they switched to another material $\overline{14}$S5 (Moncton *et al* 1982) which was amenable to the formation of films with as low as two layers. It was found that the film with two molecular layers melted by a

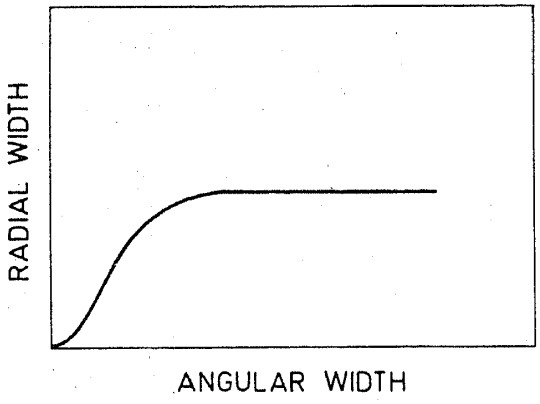

Figure 13. Schematic drawing to illustrate the outcome of the experiment of Rosenbaum *et al* (1983). Sketched here is the related variation of radial and angular widths of the (1, 0) diffraction spot as temperature is increased. As T becomes larger, the angular width increases more rapidly causing the six spots to merge into a ring.

* Technically, in the hexatic phase the mean square angular fluctuations $\langle \delta\theta^2 \rangle \to \infty$ and x-ray scattering should give a ring. However, a weak substrate field is enough to restore the six spots. On the other hand, if the orientational correlations are not strong (i.e. are liquid like as in (33)), then a very *strong* substrate field would be needed to produce the spots.

Table 1. Summary of papers dealing with computer simulation experiments.

Reference	System considered	Results	Remarks
Zollweg (1980)	1024 hard discs. Monte-Carlo.	Orientational correlations studied. Two phase coexistence suspected in transition region.	
Kalia *et al* (1981)	100 electrons. Constant density. Molecular dynamics.	First-order transition.	
Kalia and Vashista (1981)	256 colloidal particles, $(1/r^3)$ potential. Constant density. Molecular dynamics.	First order transition.	
Hansen *et al* (1979)	104, 400 electrons. Molecular dynamics. System on spherical surface.	In contrast to hard disc fluids, a diffusion coefficient does exist.	Periodic boundary conditions avoided. Might be interesting to study melting process.
Morf (1979)	780 electrons. Molecular dynamics.	Shear modulus shows a temperature dependence similar to what is indicated by KT.	
Broughton *et al* (1982)	780 particles, $(1/r^{12})$ potential. Molecular dynamics. Isothermal scan.	First-order transition. K versus P indicates a value at melting close to 16π predicted by KT.	Value of K of 16π at melting is surprising since transition is first order. A similar result has been obtained by Abraham.
Swol *et al* (1980)	3200, r^{-12} particles and 2688 LJ atoms. Isothermal scan.	First-order transition in both systems.	
McTague *et al* 1980a	256 and 2500 r^{-6} particles. Monte-Carlo. Isochore traversed.	Defect structures examined and grain boundary loops found. Unable to confirm if transition is continuous.	

Frenkel and McTague (1979)	256 LJ particles. Molecular dynamics. Isochore scan	Two continuous transitions reported, sandwiching a hexatic phase.	
Toxvaerd (1980)	Frenkel and McTague's experiment repeated	Trajectory plots show two phase coexistence.	Apparently Frenkel and McTague misinterpreted the two-phase region as the hexatic phase.
Toxvaerd (1981)	256 and 3600 LJ particles. Constant temperature scan.	First order transition seen for both systems.	Authors comment that increasing the size weakens the first-order transition.
Tobochnik and Chester (1982)	Hard disc and LJ particles. Isochore scans. Both high and low density systems studied.	Results for high density systems consistent with first-order transition. In the low density system a discontinuity in K is observed close to 16 π.	
Bakker *et al* (1984)	10864 LJ particles. Isochore scan.	First order transition reported.	Special hardware processor used for the simulation.
Abraham (1980)	256 LJ atoms. Monte Carlo. Constant pressure scan.	First order transition.	
Abraham (1981b)	256 and 529 LJ atoms. Two free surfaces present.	Solid with surface melts close to the thermodynamic melting temperature in contrast to a surfaceless solid.	Abraham feels the KT melting temperature is an upper limit for the stability of the *metastable* (i.e. superplated) 2D solid.
Koch and Abraham (1983)	576 xenon atoms on graphite. Isothermal and constant pressure scans.	First order transition.	This study was to see if substrate had any effect. Computational box as in figure 21.
Abraham (1984)	Xenon films on graphite of thickness greater than one monolayer simulated	Results compared with x-ray experiments. First-order transition observed in contrast to lab. experiments.	

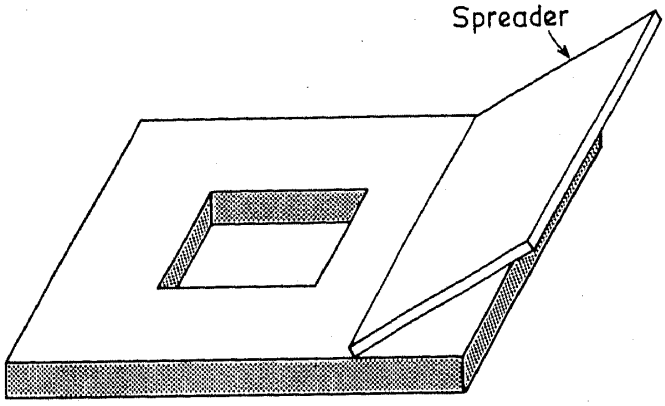

Figure 14. Ultra thin layers of self-supporting liquid crystal films are prepared by drawing the material across the open hole in the glass cover slide. The hole is about 6×6 mm^2. Films as thin as two molecular layers can be prepared (After Brinkman *et al* 1982).

single abrupt transition with hysteresis and no critical behaviour *i.e.* by a first-order transition. These results are also in accordance with the findings of Bishop *et al* (1982) in a related study in which shear mechanical measurements were made.

We have confined our survey to the most important of the several experiments done so far. While the studies on adsorbed layers seem to favour the KTHNY theory, the melting of liquid crystal films seem to point to a first order transition. Since both these classes of experiments are open to their own objections, many have sought an answer *via* computer simulation to which we now turn our attention.

4.2 *Simulation experiments*

Computer experiments on phase transitions have a long history going back to the pioneering work of Alder and Wainwright (1962) who studied hard disc systems. The experiments we review use either the Monte-Carlo approach (Metropolis *et al* 1953) or the molecular dynamics approach (Rahman 1964). A large number of papers have been published, of which table 1 offers a partial summary. Given this large volume of work, we can indicate only the highlights.

4.2*a Advantages*—An important advantage of the simulation experiment is that it enables one to follow events at the level of individual atoms. To appreciate this, we refer to figure 15. Here 15a shows an instantaneous snapshot of the system. The coordination number of each atom in this assembly can now be analyzed *via* a Dirichlet/Wigner-Seitz construction (see figure 6), and one can identify those with anomalous coordination *i.e.* those not having the normal quota of *six* neighbours. One can now suppress the "normal" atoms and project only the defect structure as done in figure 15b. Such studies have not only shed much light on how defects evolve and multiply, but more significant, have called attention to the possible role of grain boundaries in the melting process.

A second useful pointer to emerge is the superiority of the constant pressure and isothermal scans as opposed to traversal along an isochore (constant density scan). In the former, the distinction between a first order and a continuous transition is more clear-cut (as we shall presently see). In an isochore traversal on the other hand, one could strike a coexistence patch, which, besides making the transition appear

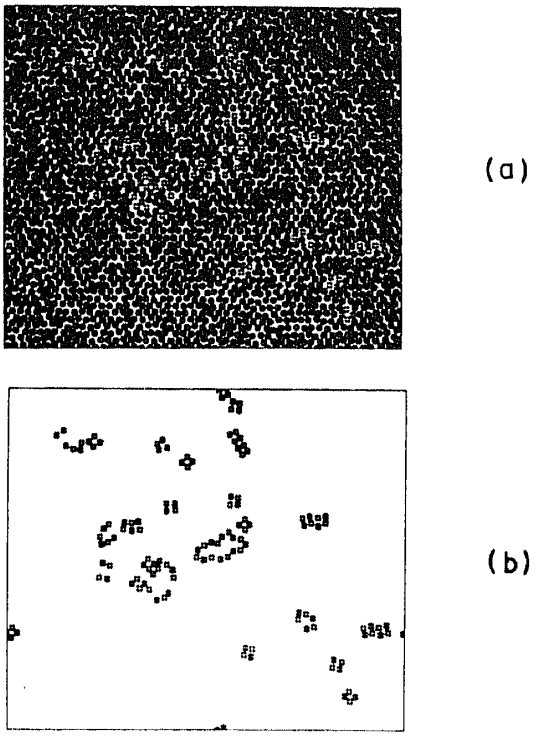

Figure 15. (a) shows a typical configuration of atoms during the simulation. + and − denote respectively seven- and five-fold coordinations. In (b) is projected the defect structure alone (After McTague *et al* 1980a).

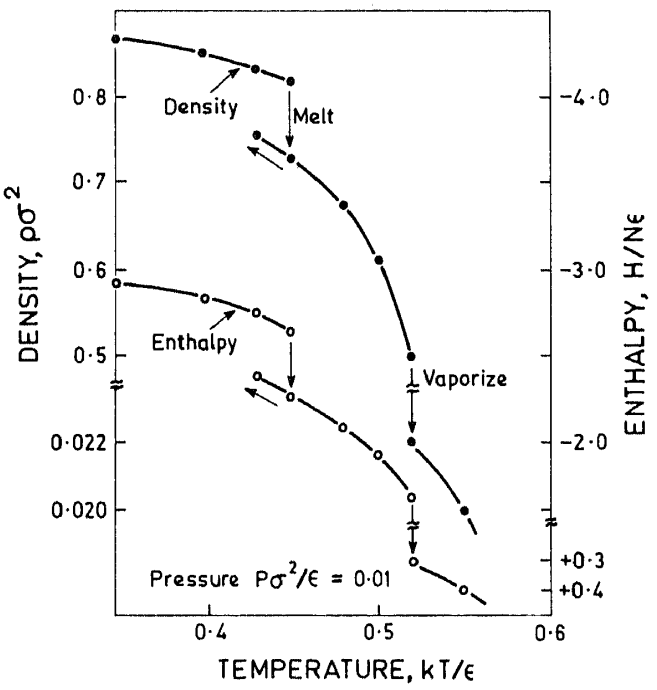

Figure 16. Equilibrium density and enthalpy per atom as a function of temperature for a 256 LJ atom system at $P^* = 0{\cdot}01$. The sharp breaks are characteristic of first-order transition (After Abraham 1980).

continuous, could also give erroneous indications about the presence of a hexatic phase.

Yet another merit of simulation is the ability to follow the trajectories of individual atoms over a fairly large number of steps. We shall shortly consider examples of such plots and their utility.

4.2*b Some results*—In a typical simulation experiment, one considers an assembly of particles interacting *via* a suitable potential. The particles are confined to a planar cell and suitable boundary conditions are imposed (usually periodic boundary conditions; exceptions however have been made: Hansen *et al* 1979; Abraham 1981b). The thermodynamic conditions like pressure, temperature etc are specified and the simulation is executed in a series of consecutive steps. Initially, a large number of steps must be gone through (with suitable "steering") so that equilibrium is attained corresponding to the desired conditions. Near a transition, special care is necessary since equilibration will require many more steps than usual. After attainment of equilibrium, the system is run through more steps, during which "measurements" are made. In a constant pressure run, for example, one would monitor the density and enthalpy. Besides evaluating thermodynamic quantities, correlation functions are also sometimes computed. In addition, defect patterns, trajectory plots etc are obtained as required. This sequence is then repeated for a new set of initial conditions and in this way a whole scan is made.

We consider now some representative results. Figure 16 shows the equilibrium density and enthalpy per atom as a function of temperature for a 256 Lennard-Jones ($V(r) = 4\varepsilon\{(\sigma/r)^{12} - (\sigma/r^6)\}$) atom system at a fixed pressure. Abraham (1980) who did this work, carried out such studies at three reduced pressures $P^* = P\sigma^2/\varepsilon = 0{\cdot}01, 0{\cdot}05$ and 1. The first two pressures were intentionally chosen low since NH had speculated that the melting could be first order at high pressures but continuous at low pressures (see also figure 9). Abraham however finds that melting is always a discontinuous process as in figure 16.

Figure 17 shows results for isothermal scan obtained by Toxvaerd (1981) for two Lennard-Jones systems with 256 and 3600 particles respectively. Both exhibit Van der Waal loops characteristic of a first order transition, the loop being smaller for the larger system. Whether the transition will continue to be first order in the thermodynamic limit is difficult to say but after investigating the size dependence from 256 to 3600 particles, Toxvaerd seems to feel that the transition will continue to remain weakly first order. Broughton *et al* (1982) investigated a system of 780 particles interacting *via* a

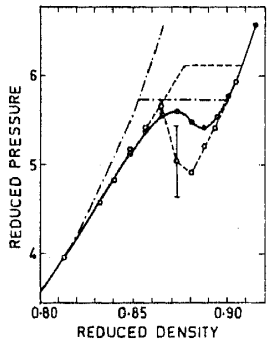

Figure 17. Pressure versus density at constant temperature for two LJ systems. The solid circles are for 3600 particles and the open circles are for 256 particles (After Toxvaerd 1981).

purely repulsive r^{-12} potential. They too find a weak, first-order transition. These authors also studied a system with a single vacancy which gave them the same loop during both forward and reverse scans, unlike the perfect system. Both Toxvaerd and Broughton *et al* have detected grain boundaries from defect structure analysis (recall figure 15). They also cautiously note that periodic boundary conditions could stabilize the perfect solid, inhibiting the formation of vacancies and interstitials. In turn this could inhibit dislocation climb and modify the character of the transition. However, taking all factors into account, the overall assessment of both authors is that the melting transition is not only weakly first order but just preempts the KT transition.

Special mention must be made of the work of Bakker *et al* (1982, 1984) who have built a dedicated hardware processor enabling them to make a quantum jump. Instead of dealing with 512 or 1024 particles as usual, they are able to study the molecular dynamics of 10864 particles. Their device had the speed of a super computer, achieving in 750 hours what would have taken 4000 hours in Amdahl 470 V/7-B. One expects this approach will set a new trend. Incidentally, Bakker *et al* (1984) have studied a Lennard-Jones system along the isochore $\rho^* = \rho\sigma^2 = 0{\cdot}88$, very close to that traversed earlier by Frenkel and McTague (1979) and Toxvaerd (1980). Whereas Frenkel and McTague interpreted their data as being indicative of two continuous transitions with an intermediate hexatic phase, Bakker *et al*, in agreement with Toxvaerd, conclude that melting is in fact first order, and that the intermediate phase is really a coexistence region.

This is a convenient juncture to introduce trajectory plots, a good example of which is given in figure 18. The plots here pertain to a 256 atom Lennard-Jones system, and were obtained during a scan along an isochore (Abraham 1981a). They show four consecutive sequences, each generated from 8×10^5 Monte-Carlo steps. In each plot one can see a crystalline as well as a liquid-like region, indicative of coexistence. Further, the melted region appears to be mobile with passage of time, suggestive of a small interfacial energy between the liquid and the solid. Such a coexistence could well mimic a hexatic phase, even if the phase were absent.

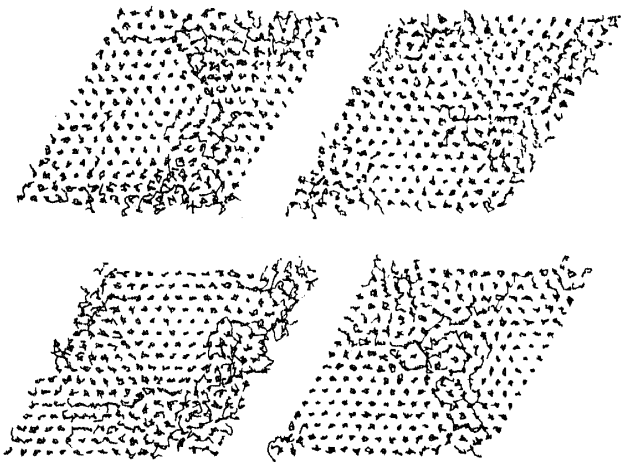

Figure 18. A consecutive sequence for four trajectory plots for a 256 LJ atom system. Each plot is generated from 8×10^5 consecutive Monte-Carlo steps, with the system held at fixed density and temperature. Observe the coexistence of solid and liquid regions (After Abraham 1981a).

The simulations discussed thus far deal with systems having no physical boundaries. Abraham (1981a, b) considered the role of surfaces since there is some evidence for surface premelting in the case of 3D crystals (Broughton and Woodcock 1978). He finds (see also figure 19) that a 2D solid with two surfaces is a stable crystal with premelted surface at reduced temperature $T^* = 0{\cdot}40$ ($T^* = k_B T/\varepsilon$) and a stable liquid at $T^* = 0{\cdot}42$. An independent free energy analysis of the 2D Lennard-Jones system yields a thermodynamic melting temperature of $T^* = 0{\cdot}415$ (Barker *et al* 1981). Abraham further finds that a solid subjected to periodic boundary conditions melts at $T^* \sim 0{\cdot}45$ with a discontinuous jump in density (*i.e. via* first order). From this Abraham concludes that a solid subjected to periodic boundary conditions superheats well beyond the thermodynamic melting temperature. As he remarks (Abraham 1981a) the melting temperature of the *surfaceless* solid "corresponds to an upper limit for the stability of the metastable 2D solid". Associated with the transition at $T^* = 0{\cdot}45$, there is the stiffness dependence shown in figure 20. At the transition temperature, the measured value of K is lower than 16π "because of defect formation in the solid constrained to remain at solid density". However, if K values obtained for $T^* < 0{\cdot}45$ are extrapolated, they yield the solid curve shown in figure 20. K thus appears to jump discontinuously to zero at the transition from the *universal* value of 16π. This of course emerges from the renormalization group analysis of KT. Why it should do so likewise in a first-order transition is not clear.

We have already alluded to some of the doubts that can be cast on the experiments on adsorbed films. One of these relates to the possible role of the second layer (which is frequently present). To make an assessment, Abraham (1983; 1984; see also Koch and Abraham 1983) simulated xenon films on graphite, the geometry of the computational box being as in figure 21. The simulations covered also the conditions of the experiments of Heiney *et al* (1982) and Rosenbaum *et al* (1983). Abraham's main conclusion is that the transition still remains first order, at variance with the findings of the x-ray experiments. Imperfections in the laboratory graphite substrate could be a possible source of the discrepancy.

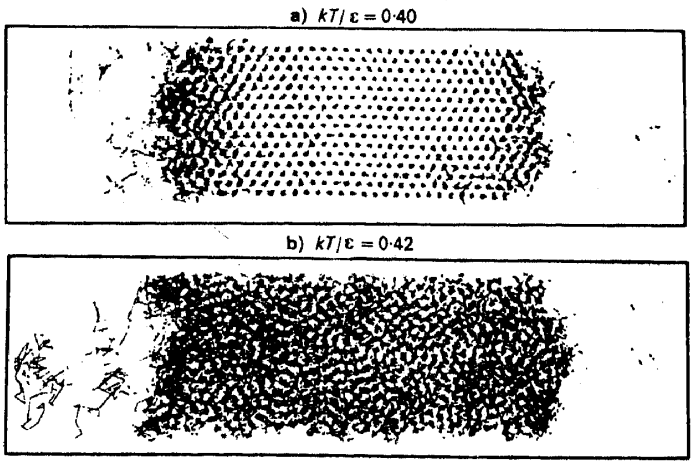

Figure 19. Trajectory plots of a 2D 512 LJ atom system with two free surfaces. The two temperatures investigated are on either side of the thermodynamic melting point of 0·415. (After Abraham 1981a).

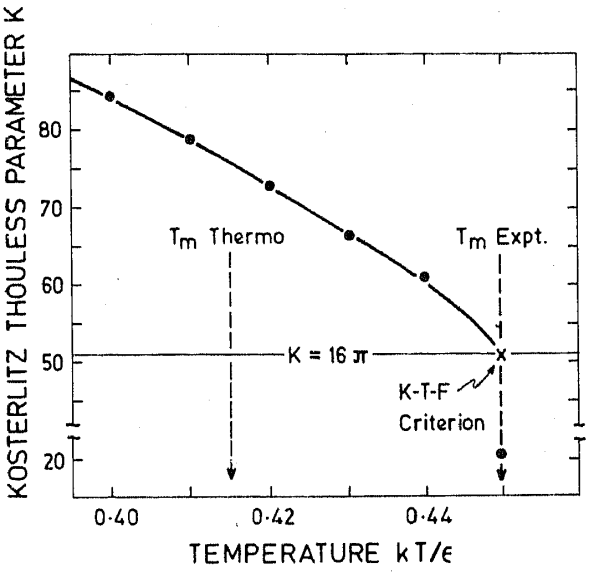

Figure 20. Parameter K as a function of temperature for a 529 LJ atom system corresponding to $P^* = 0{\cdot}05$ (After Abraham 1981a).

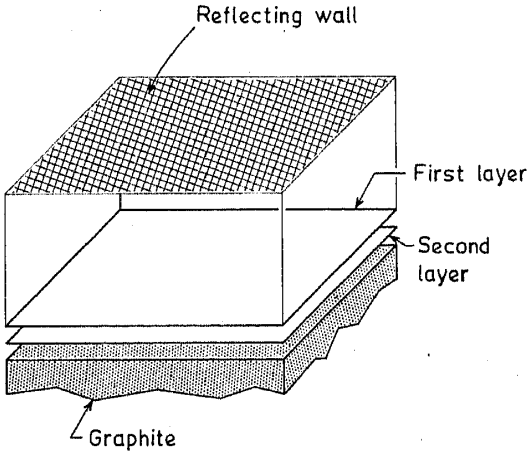

Figure 21. Computational scheme for the simulation of the freezing of xenon on graphite. The graphite surface defines the x, y plane at $z = 0$. Shown are two layers of adsorbed atoms, the four faces across which periodic boundary conditions are applied and a reflecting wall at the top (After Koch and Abraham 1983).

4.2*c* *Some doubts*—Though elegant, simulation also is not free from objections, principal among which are the following (Broughton *et al* 1982):

(i) Typical sample sizes of 500–1000 atoms are too small to be representative of thermodynamic specimens.

(ii) Even the largest computer simulation run corresponds to a very short time on a lab scale. Achievement of equilibrium near a transition is therefore a sensitive question, especially if critical slowing down is a possibility.

(iii) Periodic boundary conditions could modify the apparent nature of the transition.

(iv) Simulation without an adequate number of built-in vacancies and interstitials might not produce a sufficient amount of dislocation climb which is necessary for a proper test of the KT theory.

There are a few other supplementary queries that may be raised. It is fair to say that those engaged in simulation have given serious consideration to all these objections in carrying out their experiments and in analyzing the results. Some observers are however not still convinced.

4.2*d* *Simulation using defect Hamiltonian*—We turn now to an entirely different kind of simulation due to Saito (1982a, b) who, instead of working with atoms, used a defect Hamiltonian defined on a triangular mesh with periodic boundary conditions. In the thermodynamic limit, the defect Hamiltonian goes over to that of KT *i.e* of continuum elasticity theory (as it should). Saito uses the Monte-Carlo method. A pair of nearest neighbour sites are chosen at random from the mesh. If both sites are empty, one tries to create a pair of dislocations with opposing Burgers vectors. If both sites are occupied with dislocations of opposite Burgers vectors, an annihilation is attempted; otherwise an exchange is tried. The trial is accepted according to the usual Boltzmann weight etc.

Saito finds that if E_c the dislocation core energy is large, the melting transition is due to dislocation unbinding and is in accord with the KT theory. For a small core energy, however, the melting is of first order, being caused by the nucleation of grain boundary loops. It is natural to speculate whether a system of atoms will show similar dichotomy depending on whether the inter-atomic potential favours a large E_c or not. In collaboration with V. Sridhar and B. Chakraborty of our Centre, we are currently examining this question *via* simulation. Meanwhile, we learn that Van Himbergen (1984) has carried out a similar study for the XY model and finds both continuous as first order transition, depending on the shape of the nearest neighbour interaction. One therefore awaits Sridhar's results with interest.

5. Other mechanisms and viewpoints

In view of the doubts concerning the validity of the KT hypothesis, several alternate viewpoints have recently been proposed. Guided by the results of several simulation studies and the work of Fisher *et al* (1979) who noted that grain boundaries could appear before T_m is reached, Chui (1982, 1983) constructed a grain boundary theory of melting. Chui models the grain boundary as an array of dislocations. A pair of dislocations with Burger's vectors $\mathbf{b}$, $\mathbf{b}'$ interact with each other *via* a potential of the form

$$V = -\frac{K}{4\pi}\left(\mathbf{b}\cdot\mathbf{b}'\ln|\mathbf{r}-\mathbf{r}'| - \frac{[\mathbf{b}\cdot(\mathbf{r}-\mathbf{r}')]\,[\mathbf{b}'\cdot(\mathbf{r}-\mathbf{r}')]}{|\mathbf{r}-\mathbf{r}'|^2}\right), \tag{35}$$

where K is as in (17). The first term in (35) should be familiar to us from the corresponding term of (14). The second term of (35) represents the anisotropic part of the interaction. (The continuum Hamiltonian of NH, for example, will have a third term related to the core energy E_c. That, however, is not pertinent here.) A grain boundary configuration appropriate to a triangular lattice was next considered, and the effective potential between parallel grain boundaries at a distance Z apart was computed using

(35). In turn this formed the input for a free energy analysis of phase stability. The conclusions drawn by Chui are:

(i) grain boundaries do make an appearance before T_m,

(ii) a strongly first-order transition results if the grain boundaries are coupled to density change or bound dislocations pairs,

(iii) the transition becomes weakly first order for a core energy $< 2{\cdot}84T_m$.

In short, grain boundary melting preempts the KT transition in Chui's theory.

Ramakrishnan (1982, 1984) following his earlier work with Yussouff (1979), approaches the transition from the liquid side *i.e* as a freezing problem. According to this view, the solid emerges from the liquid primarily due to the dominance of a density wave of crystalline periodicity and symmetry. The wavevector **G** of this wave is identified with the first peak in $S(q)$ the liquid structure factor. From the quantitiative point of view, the theory considers the solid near melting as a calculable perturbation on the liquid and shows that their free-energy difference is determined by two parameters $\mu(\mathbf{G})$ and $c(\mathbf{G})$. Of these $\mu(\mathbf{G})$ is defined by

$$\rho(\mathbf{r}) = \rho_0(1 + \eta + \mu(\mathbf{G}) \exp i\mathbf{G}\cdot\Upsilon),$$

where $\rho(\mathbf{r})$ is the density. $c(\mathbf{G})$ is the Fourier transform of the fluid correlation function and is related to the structure factor as

$$S(\mathbf{G}) = (1 - c(\mathbf{G}))^{-1}.$$

A self-consistent relation for $\rho(\mathbf{r})$ is derived involving also the potential $V(r)$, from which the free energy difference is computed. The theory predicts a first-order transition at a temperature T_f.

Ramakrishnan (1982) also considers the implication of T_f being greater or less than the KT transition temperature T_m. Of course if $T_f < T_m$, then the KT mechanism has no chance. On the other hand if $T_m < T_f$, it could well happen that owing to lack of sufficient run time, computer samples continue to appear as a solid and melt only when T_f is attained (*via* first order of course). Ramakrishnan therefore advocates suitable experiments on spatial correlation to ascertain whether $T_f < T_m$ or $T_f > T_m$ in systems where T_f is clearly identified.

In a series of high-powered (!) papers, Kleinert (1983a, b and references therein) has reexamined the KT mechanism, with particular reference to the role of the core energy term in the Hamiltonian for an assembly of defects. In the KT theory, the core energy term has a form $\sim E_c \sum_i (\mathbf{b}_i^2)$ with the further stipulation $\sum_i \mathbf{b}_i = 0$. In the Coulomb model mentioned in § 3.4, the corresponding expressions would be $\sum q_i^2$ with $\sum_i q_i = 0$ (charge neutrality).

According to Kleinert, a single disclination is really a point singularity at the end of a string of dislocations, rather like the "Dirac string" of the magnetic monopole. Kleinert's view is that a core energy term of the KT type is inappropriate since it implies an infinite energy for the dislocation string, automatically ruling out dislocation pile up and a concomitant first-order transition. By following a route earlier established by Villain (1975) *vis-a-vis* the 2D XY model, Kleinert derives an expression for the partition function which, unlike that of KT, is free from this "objection". An interesting feature of Kleinert's work is the use of duality concept, already familiar in lattice gauge

theory (Kogut 1979), spin glass (Fradkin *et al* 1978) etc.*. The net outcome of Kleinert's analysis is that melting in two dimensions is a first-order process. Very recently, Janke and Kleinert (1984) have backed up this conclusion with a Monte-Carlo simulation.

6. Summary and concluding remarks

Despite the early start given by Peierls (1935) and Landau (1937) on the theoretical side and the pioneering experimental work of Langmuir (1938) on surfaces, 2D solids did not receive much attention for a long time, possibly because the departure from true crystallinity was almost notional*. However, the deep enquiry in the sixties concerning the nature of ordering in 2D systems revived interest, leading to the novel proposal by Kosterlitz and Thouless (1973) that phase transitions are possible without being accompanied by ordering in the usual sense.

Several systems (superfluid films, electrons on liquid helium etc) have been experimented upon to test the validity of the KT idea (see for example, Sinha 1980). As far as melting in 2D is concerned, the important question is whether it is prompted by long wavelength or short wavelength fluctuations. If it is the former, then the transition is not only continuous but has several unusual features. A crucial factor is the role played by topological defects, in particular the dissociation of bound pairs. On the other hand if melting occurs mainly due to a pile-up of disruption of local arrangements *i.e.* due to short wavelength fluctuations (which is facilitated by core overlap—see remarks in §3.3), then the transition is of first order as in 3D.

We have presented a sampling of the more detailed (lab) experiments on melting carried out so far. Most of these have been on adsorbed films of rare gas atoms, and the results seem to favour the KTHNY theory. The results on self-supporting films of liquid crystals seem however to favour a first-order transition.

In contrast to the lab experiments, the computer experiments do not have problems of specimen preparation and therefore have been quite popular. The evidence from such simulations appears to be in favour of a first-order transition. However, as in the case of the lab experiments, the results are not totally objection-free.† For some, no matter how detailed the simulation, these experiments come nowhere near to real life on account of the various factors discussed earlier. On the other hand, the practitioners of simulation by and large seem convinced that the transition is of first order, although some are more cautious than others in articulating their opinions. In passing, it is interesting to note that Abraham (1984) in his simulation studies is able to reinterpret the x-ray results of Heiney *et al* (1983) and of Rosenbaum *et al* (1983) in support of a first order transition, although the experimenters themselves feel their results point to a continuous transition!

At the moment there is lull of sorts, with each "camp" holding on to its own view

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* See also Jose *et al* (1977) and Nelson (1978).

* Abraham (1980), for example, estimates that for a correlation loss $\sim$ 6 Å, the 2D crystal must have a size $\sim 10^{10}$ light years! It is remarkable, however, that diffraction experiments reveal *via* line shapes even these minute differences.

† Within the simulation camp itself there has been some cross fire! See Toxvaerd (1984) and Abraham and Koch (1984).

point.* Resumption of vigorous activity is, however, very much on the cards especially on the lab experiments side, with accent on eliminating the various objections raised in the past. Besides the ever-popular diffraction studies, other supporting investigations may also be anticipated, *e.g.* on shear properties. Hopefully these will settle the issue once and for all in an unambiguous manner. Meanwhile one should not lose sight of the finding of Saito (1982a, b) that one can get either a first order or a continuous transition, depending on the value of E_c the core energy. There is also the related work on the XY model (Van Himbergen 1984) where this dichotomy has been analyzed. It could well happen that there are some 2D lattices which melt *via* a KT mechanism and others which do not!

It would indeed be a pity if in the final analysis it is established that 2D melting does not occur by a KT process. If so, it would be a narrow miss, for the transition appears to be on the edge of being a KT type, as indicated by many of the experiments done so far. In a way this underscores how sensitive the outcome of a given experiment is to "disturbances" (e.g. imperfections in the substrate).

Even if it turns out that 2D melting is first order, the KT paper would not be without significance, having exerted a strong influence on the study of phase transitions in 2D. Of special interest is the influence exerted "across the border" on particle physics. This is not altogether surprising, given the parallels between statistical mechanics and field theory (Kogut 1979), and particle theorists' interest in topological objects (Coleman 1977 and Rajaraman 1982). Thus, whatever be the final outcome as regards melting, the KTHNY and all related papers have been beneficial in their own way to the progress of physics, having made a beginning concerning the statistical mechanics of topological defects.

7. Other reviews

In view of its great popularity, there naturally have been several reviews on the KT transition from time to time. It is pertinent therefore to call attention to these, and place our own in the proper perspective.

In one of the early reviews, Kosterlitz and Thouless (1978) emphasize superfluidity, understandable in view of the forum. A subsequent survey entitled "ordering in two dimensions" by Kosterlitz (1980) is broadbased but the accent is almost entirely on theory. Halperin has reviewed the subject in his Kyoto lectures (Halperin 1979) and in the Les Housches School (Halperin 1981). The discussions are extensive and again the emphasis in on theory. Barber (1980) does not restrict himself to the KT problem but covers instead the whole gamut of O(n) models in 2D. In the process, he indicated linkages to the problem of quark confinement. Nelson (1979) has several useful and interesting comments on the XY model and its various gauge generalizations. Young

* For example, Nelson (1983b) remarks "I doubt that the simulation techniques used by Abraham *et al* could reproduce the apparently continuous melting transition of incommensurate xenon on graphite observed recently via precision x-ray diffraction by Heiney *et al.*". Abraham (1984) on the other hand holds, "From our present study the status is clear for the high-temperature xenon film on graphite . . . . We observe first-order melting of xenon . . . ".

(1980) gives a simple and crisp introduction to the central idea of the KT theory. Abraham (1981a) has a very readable article on simulation experiments but it is a paraphrase of his own extensive work. Brinkman *et al* (1982) offer a brief but popular survey of the 2D melting question. Mention should also be made of the conference volume edited by Sinha (1980) which gives a good overview of the then state of the art relating to phase transitions in 2D. Our paper restricts to the melting problem alone, but goes into some detail both on the conceptual as well as on the experimental side. The discussion of the experimental situation is reasonably uptodate. The present review should therefore be a useful complement to the various earlier surveys.

8. Dedication and acknowledgements

It is a pleasure to dedicate this article to Dr. R. Ramanna on the occasion of his sixtieth birthday for his many contributions, and especially his vigorous championing of basic research.

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Note added in proof

After this paper was sent for publication, we became aware of:

(i) A review by D R Nelson on Two Dimensional Superfluidity and Melting (in *Fundamental Problems in Statistical Mechanics* V 1980 ed E G D Cohen Amsterdam: North Holland).
(ii) a paper by A Holz 1984 (*Phys. Lett.* **105A** 472) who comments on Kleinert's work, and
(iii) a paper by S B Hurlbut and J G Dash 1984 (*Phys. Rev. Lett.* **53**, 1931) who report a first-order melting for ^{4}He monolayer on graphite.

Attention is also invited to the Conference Proceedings entitled *Melting, Localization and Chaos* (editors R Kalia and P Vashista 1982 Amsterdam: North Holland) which contains many papers relevant to the present article.