LATTICE DEFECT MODEL WITH TWO SUCCESSIVE MELTING TRANSITIONS *

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A lattice model is presented for the rotational stiffness of molecular crystals. In contrast to models with only linear elasticity, dislocations and disclinations are independent. If l is the length scale of this stiffness, I show that for large l, melting proceeds via two Kosterlitz–Thouless transitions. For smaller l, these move closer together, possibly changing their character, until there is only one transition of first order.

1. Introduction

For two-dimensional melting of atomic point lattices the KTHNY theory [1] predicts, under certain assumption on the core energies of defects, the existence of two successive extremely smooth in phase transitions. This prediction has raised controversies, since molecular dynamics calculations have always found a single sharp transition [2]. Wherever this did not happen [3], this could be traced back to the existence of a mixed intermediate phase at constant volume [4]. The same thing was observed in lattice models made to simulate the interplay of dislocations and disclinations interacting via lowest gradient elasticity [5]. They always have a single first order transition [6] with an entropy jump per site of $\Delta s \approx 0.2$. Still, the theoretical prejudice for the two transitions was so strong that they were distinguished [7] even when there was only one transition [8]. A similar situation holds experimentally. Atomic monolayers always show a sharp peak in the specific heat. For higher coverage, the transition may become continuous, but never as smooth as required by the Kosterlitz–Thouless arguments [9]. Two clear successive transitions have been found only in layers of liquid crystalline material [11]. Since the KTHN arguments and the above lattice models do not apply to these systems, it is worthwile to find a model which is capable of describing such an experimental situation.

2. The model

We set up a lattice partition function which contains defects and their elastic interaction including the strong gradient elastic energy $\frac{1}{2}\mu l_2[\partial_i(\partial_1 u_2 - \partial_2 u_1)]^2 = 2\mu l^2(\partial_i \omega)^2$ ($u_i \equiv$ displacements). The parameter l characterizes the length scale over which the system is stiff with respect to local rotations. Any crystal with non-spherical molecules will have $l \neq 0$. Dislocations and disclinations are introduced via corresponding discrete plastic distortions and the model partition function reads

$$Z = \prod_{\boldsymbol{x}} \left(\int_{-\pi}^{\pi} \mathrm{d}\gamma_i(\boldsymbol{x}) \right) \sum_{\{n_{ij}(\boldsymbol{x}), m_i(\boldsymbol{x})\}} \exp(-\beta E), \qquad (1)$$

where $\beta \equiv a^2/2\pi^2 k_\beta T$, the vector $\boldsymbol{x}$ runs over a square lattice with spacing a, and

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$$E=\tfrac{1}{4}\sum_{x,ij}(\nabla_i\gamma_j+\nabla_j\gamma_i-4\pi n^{\mathrm{s}}_{ij})^2 + \frac{2\lambda}{\mu}\sum_x\left(\sum_i\nabla_i\gamma_i-2\pi n^{\mathrm{s}}_{ij}\right)^2 + \frac{2l^2}{a^2}\sum_x[\nabla_i\omega-\pi[2m_i+\nabla_i(n_{12}-n_{21})]\}^2 . \quad (2)$$

The symbols ∇_i are the lattice gradients ($\nabla_i f(\boldsymbol{x})\equiv f(\boldsymbol{x}+\boldsymbol{i})-f(\boldsymbol{x})$). The plastic distortions n_{ij}, m_i are integer. The superscript s denotes symmetrization. The displacement vectors $u_i(\boldsymbol{x})$ have been rescaled by a factor $2\pi/a$ (i.e. $\gamma_i\equiv 2\pi u_i/a$) so that μ, λ are the usual elastic constants.

By a duality transformation, this partition function becomes

$$Z=\prod_x\left(\int_{-\infty}^{\infty}\mathrm{d}^2\gamma\int_{-\infty}^{\infty}\mathrm{d}\omega\int_{-\infty}^{\infty}\mathrm{d}\tau_{ij}\int_{-\infty}^{\infty}\mathrm{d}\tau_i\right) \times\sum_{\{n_{ij},m_i\}}\Phi[n_{ij},m_i]\exp(-\beta E_2) , \quad (3)$$

where

$$\beta E_2=\sum_x\left\{\frac{1}{\beta}\left[\frac{1}{4}\left(\sigma^{\mathrm{s}2}_{ij}-\frac{\nu}{1+\nu}\sigma^{\mathrm{s}2}_{ii}\right)+\frac{a^2}{8l^2}\tau_i^2\right] +\mathrm{i}\sigma_{ij}(\nabla_i\gamma_j-\epsilon_{ij}\omega-2\pi n_{ij})+\tau_i(\nabla_i\omega-2\pi m_i)\right\} \quad (4)$$

and $\Phi[n_{ij},m_i]$ is a gauge fixing factor needed to remove the degeneracy with respect to the integer valued local defect gauge transformations [12]

$$n_{ij}\to n_{ij}+\nabla_i N_j-\epsilon_{ij}M , \quad m_i\to m_i+\nabla_i M ,$$
$$\gamma_j\to\gamma_j+2\pi N_j , \quad \omega\to\omega+2\pi M . \quad (5)$$

Integrating out the now independent γ_i, ω, gives the stress conservation laws,

$$\bar{\nabla}_i\sigma_{ij}=0 , \quad \bar{\nabla}_i\tau_i=\epsilon_{ij}\sigma_{ij} \quad (6)$$

(with $\bar{\nabla}_i f(\boldsymbol{x})-f(\boldsymbol{x}-\boldsymbol{i})$). They are automatically enforced by introducing the stress gauge fields A_i, h and setting

$$\sigma_{ij}=\epsilon_{ij}\bar{\nabla}_k A_k , \quad \tau_i=\epsilon_{ij}\bar{\nabla}_j h-A_i . \quad (7)$$

(Actually, A_i, h are proper gauge fields only in three dimensions, the gauge transformations being accidentally lost in two dimensions.) The energy has now the form

$$\beta E_3=\sum_x\left[\frac{1}{4\beta}\left(\frac{1}{1+\nu}(\nabla_i A_j)^2-\frac{1}{2}\frac{1-\nu}{1+\nu}(\bar{\nabla}_i A_i)^2\right) +\frac{a^2}{8\beta l^2}(\bar{\nabla}_k h-\epsilon_{kl}A_l)^2-2\pi\mathrm{i}(A_i\bar{b}_i+h\bar{\Theta})\right], \quad (8)$$

where $\bar{b}_i=\epsilon_{jk}\nabla_j n_{ki}-m_i$, $\bar{\Theta}\equiv\epsilon_{ki}\nabla_k m_i$ are the integer valued dislocation and disclination densities. They are properly defect gauge invariant under (5), as they should [13,14]. By integrating out the gauge fields one finds the pure defect representation of the partition function with an energy

$$\beta E_4=\beta\times 4\pi^2(\nu+1)\sum_x\bar{\eta}(\bar{\nabla}\cdot\nabla)^{-2}\bar{\eta} +\beta\times 4\pi^2\frac{2l^2}{a^2}\sum_x\left\{\bar{\Theta}(-\bar{\nabla}\cdot\nabla)^{-1}\bar{\Theta} +\bar{\nabla}_i\bar{b}_i\left[-\bar{\nabla}\cdot\nabla\left(1-\frac{l^2}{a^2}\bar{\nabla}\cdot\nabla\right)\right]^{-1}\bar{\nabla}_j\bar{b}_j\right\}, \quad (9)$$

where

$$\bar{\eta}\equiv\epsilon_{ij}\nabla_i\bar{b}_j+\bar{\Theta} \quad (10)$$

is the defect density. A dual representation is obtained by summing the partition function of (6) over the defects. This forces A_i and h to become integer valued $\bar{A}_i$ and $\bar{h}$, leaving a generalization of the roughening model with

$$\beta E_5=\frac{1}{4\beta}\sum_x\left[\frac{1}{1+\nu}(\nabla_i\bar{A}_j)^2-\frac{1}{2}\frac{1-\nu}{1+\nu}(\bar{\nabla}_i A_i)^2 +\frac{a^2}{2l^2}(\bar{\nabla}_k\bar{h}-\epsilon_{kl}\bar{A}_l)^2\right]. \quad (11)$$

3. Phase transition

We now observe that the present model gives naturally rise to two successive continuous phase transition of the Kosterlitz–Thouless type if the new length scale l^2 which characterizes the rotational stiffness of the system is sufficiently large. Starting point is the partition function (11) involving the integer valued stress gauge $\bar{A}_i$, $\bar{h}$.

For very small β, both $\bar{A}$ and $\bar{h}$ are squeezed to zero.

If l^2 is very large, however, i.e. if the system has a large rotational stiffness, the squeezing of the $\bar{h}$ field is relaxed and follows the effective partition function

$$Z \approx \sum_{\{\bar{h}\}} \exp\Big(- \frac{a^2}{8\beta l^2} \sum_x (\nabla_k \bar{h})^2 \Big) . \tag{12}$$

This is an ordinary roughening model. It is known to have a continuous phase transition of the Kosterlitz–Thouless type if $\beta = \beta_h$ satisfies

$$4\beta_h l^2/a^2 \approx 2/\pi . \tag{13}$$

For β of order unity, the prefactor $a^2/8\beta l^2$ is so small that the discreteness of $\bar{h}$ becomes irrelevant. It is then a good approximation to integrate over $\bar{h}$ as if it were a continuous variable. By decomposing

$$\begin{aligned} &\sum_x (\bar{\nabla}_k \bar{h} - \epsilon_{kl} \bar{A}_l)^2 \\ &= \sum_x \{ \bar{\nabla}_l [\bar{h} - (\nabla \cdot \bar{\nabla})^{-1} \epsilon_{ij} \nabla_i \bar{A}_j] \\ &- (\bar{\nabla} \cdot \nabla)^{-1} \epsilon_{lk} \nabla_k \bar{\nabla}_i \bar{A}_i \}^2 , \end{aligned} \tag{14}$$

we see that the energy separates into the squares of longitudinal and transverse parts so that, after the $\bar{h}$ integration, the partition function becomes effectively

$$\begin{aligned} Z \approx \sum_{\{\bar{A}_i\}} \exp\Big[&- \frac{1}{4\beta} \sum_x \Big(\frac{1}{1+\nu} \bar{A}_i (-\bar{\nabla} \cdot \nabla) \bar{A}_i \\ &- \frac{1}{2} \frac{1-\nu}{1+\nu} \bar{A}_i (-\nabla_i \bar{\nabla}_j) \bar{A}_j \\ &+ \frac{a^2}{2l^2} \bar{A}_i (-\bar{\nabla} \cdot \nabla)^{-1} (-\nabla_i \bar{\nabla}_j) \bar{A}_j \Big) \Big] . \end{aligned} \tag{15}$$

At infinite l, the last term can be ignored and we remain with a discrete gaussian vector field theory. For $\nu = 1$ (incompressible material) the two components decouple and there is obviously a phase transition of the Kosterlitz–Thouless type at

$$2\beta_A \approx 2/\pi . \tag{16}$$

In the defect version of the partition function we shall see the same universality class to prevail also for $\nu < 1$. The transitions at large l are displayed graphically in fig. 1.

In the opposite limit of small l, the system has only a single first order transition at

$$\beta_{h,A^T}(1+\nu) \approx 0.815 . \tag{17}$$

This is the first order melting transition discussed extensively in refs. [5,6]. Here it arises since, at small l, the third term in (11) forces the vector field $\bar{A}_k$ to be equal to $\bar{A}_k^T = -\epsilon_{kl} \bar{\nabla}_l \bar{h}$ so that the first two terms become the laplacian roughening model [13] with the associated discontinuous transition [6,8] in it, the fields $\bar{h}$ and $\bar{A}^T$ become simultaneously rough.

For β far above this transition, the effective partition function is

$$Z = \sum_{\{\bar{h}, \bar{A}_l\}} \exp\Big(- \frac{a^2}{8\beta l^2} \sum_x (\bar{\nabla}_k \bar{h} - \epsilon_{kl} \bar{A}_l)^2 \Big) . \tag{18}$$

It looks like a Villain model in which $\bar{h}$ plays the role of the phase angle and $\epsilon_{kl} \bar{A}_l$ that of the jump number n_i. There is, however, an important difference: $\bar{h}$ are integer numbers. A duality transformation shows that Z is equivalent to a sum

$$Z = \sum_{\{\bar{b}_i(x)\}} \exp\Big(-2\beta \frac{l^2}{a^2} 4\pi^2 \sum_x b_i^2(x) \Big) \tag{19}$$

Since $\bar{h}$ are integer, there is no constraint $\bar{\nabla}_i b_i(x) = 0$. For this reson there is no phase transition. If $\bar{h}$ in (18) had been a continous variable, the ensuing constraint $\bar{\nabla}_i b_i(x) = 0$ would have led to a Kosterlitz–Thouless transition at

$$\beta_A \approx 1/32\pi l^2 , \tag{20}$$

in which the $\bar{A}$ fields proliferate.

In fig. 2 we have illustrated the properties of the two phases arising for small l. The sequence of transitions can also be studied in the defect representation of the partition function involving (9). For large l, the disclinations $\bar{\Theta}$ are frozen out and only the dislocations can be excited. For $\nu = 1$, the effective action is

$$\begin{aligned} Z = \sum_{\{\bar{b}_i(x)\}} \exp\Big(&-2\beta \\ &\times 4\pi^2 \sum_x \bar{b}_i(x) (-\bar{\nabla} \cdot \nabla)^{-1} \bar{b}_i(x) \Big) . \end{aligned} \tag{21}$$

This is the partition function of two independent Coulomb gases with a Kosterlitz–Thouless transition at

$$4\beta_b \approx 2/\pi , \tag{22}$$

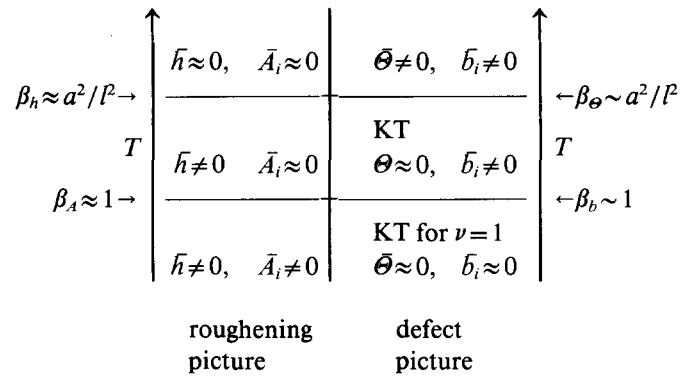

Fig. 1. Schematic characterization of the three phases for large l. The left-hand side indicates the field configurations in the roughening representations (11) (≈ 0 smooth, $\neq$ rough), the right-hand side indicates the defect excitations as deduced from the defect representation of the partition function (9).

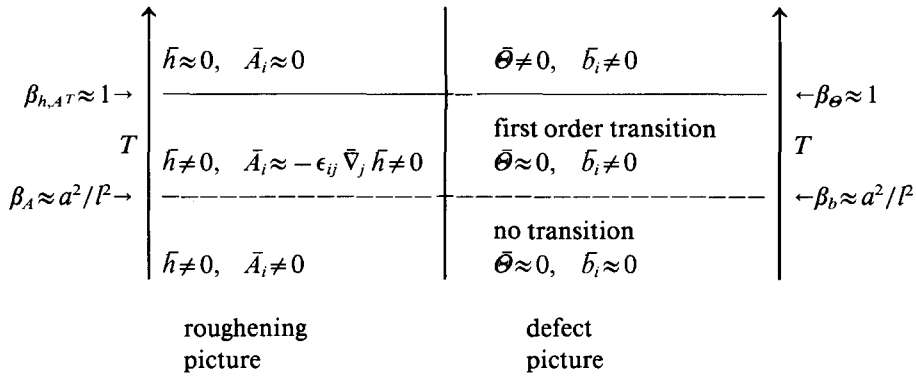

Fig. 2. At small l, there is only a single first-order melting transition at $\beta\approx 1$ where h roughens or the defects $\bar{\Theta}$ proliferate in a background of random b_i as fields.

which is to be identified with the roughening transition of the $\bar{h}$ field in (16). For $\nu\neq 1$, the long-range forces between the dislocations are modified by a factor $[2+(1+\nu)]/4$ (see eq. (9)) which enters on the left-hand side of (22). For finite l, the longitudal mode is massive and does not contribute to the critical limit, so the factor becomes $(1+\nu)/4$.

For small $\beta\approx a^2/l^2$ the effective partition function is

$$Z\approx \sum_{\{\bar{\Theta}(x)\}} \exp\Bigg(2\beta\frac{l^2}{a^2}4\pi^2 \times\sum_x \bar{\Theta}(x)(-\bar{\nabla}\cdot\bar{\nabla})^{-1}\bar{\Theta}(x)\Bigg). \tag{23}$$

It represents a Coulomb gas with a Kosterlitz–Thouless transition at

$$4\beta_\Theta l^2/a^2\approx 2/\pi\,. \tag{24}$$

This is the defect version of the roughening transitions (13). See again fig. 1 for a characterization of the phases.

Consider now the defect picture at small l. For β of order unity, the $\bar{\Theta}$ and $\bar{\nabla}_i\bar{b}_i$ terms in (9) can be dropped and the effective partition function is

$$Z\approx \sum_{\{\bar{\eta}(x)\}} \exp\Bigg(-\beta \times 4\pi^2(1+\nu)\sum_x \bar{\eta}(x)(-\bar{\nabla}\cdot\nabla)^{-2}\bar{\eta}(x)\Bigg). \tag{25}$$

This has a first-order phase transition at

$$\beta_\eta(1+\nu)\approx 0.815 \tag{26}$$

corresponding to (17) which is again the melting transition of the lowest gradient model found in ref. [6]. Another phase transition might have been expected at large $\beta\sim 1/l$, in which $\bar{\eta}$ is frozen to zero so that $\bar{\Theta}$ is equal to $-\epsilon_{kl}\nabla_k\bar{b}_l$. The second and third term in (9) can be combined and give an effective partition function

$$Z\approx \sum_{\{b_i(x)\}} \exp\left(-2\beta\frac{l^2}{a^2}4\pi^2\sum_x \bar{b}_i^2(x)\right) \tag{27}$$

just as in (19), with no phase transition, due to the absence of the constraint $\bar{\nabla}_i b_i(x)=0$ and a would-be transition at

$$\beta_b = 1/32\pi l^2 . \tag{28}$$

The characteristics of the two phases as seen from the defect point of view are shown in fig. 2.

These considerations suggest a phase diagram in the (β, l^2) plane as sketched in fig. 3. There are definitely two Kosterlitz–Thouless transitions for large l. For decreasing l, the universality classes probably remain the same unless the transition becomes of first order. It is conceivable that there is a tricritical point on the way to lower l^2 where this happens. At any rate, the two transitions move closer together. At some l, they merge into a single line which at $l\approx 0$ is certainly of first order. Probably it remains discontinuous up to the merging point.

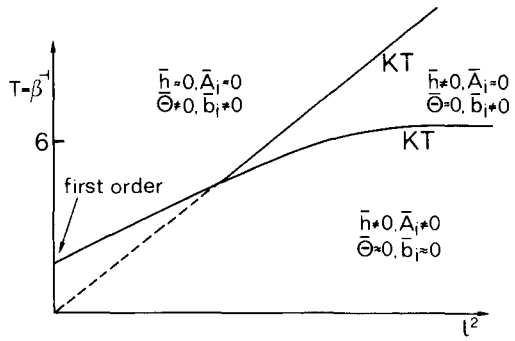

Fig. 3. Phase diagram suggested by the three known transition points found in the present discussions. It is not clear where the tricritical point lies at which the first-order transition line becomes of the Kosterlitz–Thouless (KT) type. The dashed line indicates the would-be transition (20), (26).

4. Conclusion

The model presented in this paper shows that the rotational stiffness found in two-dimensional crystals with large molecules can give rise to two successive defect generated phase transitions of the Kosterlitz–Thouless type. The model is hoped to explain the melting phenomena found in layers of liquid crystals. It should be noted, however, that such layers have a further degree of freedom, the out of plane modulations, which have been ignored in all studies, including the present one. They may well add a further transition [14,15], or modify the others.

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References

[1] J.M. Kosterlitz and D.J. Thouless, J. Phys. C 6 (1973) 1181; Prog. Low Temp. Phys. B 7 (1978) 371;
J.M. Kosterlitz, J. Phys. C 7 (1974) 1046;
D.R. Nelson, Phys. Rev. B 18 (1979) 2318;
D.R. Nelson and B.I. Halperin, Phys. Rev. B 19 (1979) 2457;
A.P. Young, Phys. Rev. B 19 (1979) 1855;
J.P. McTague, A. Als-Nielsen, J. Bohr and M. Nielsen, Phys. Rev. B 35 (1982) 7765 [mainly on Ar].

[2] J.O. Hansen and L. Verlet, Phys. Rev. 184 (1969) 15;
L. Verlet, Phys. Rev. 159 (1967) 98.

[3] D. Frenkel and J.P. McTague, Phys. Rev. Lett. 42 (1979) 1632;
J.P. McTague, D. Frenkel and M.P. Allen, in: Ordering in two dimensions, ed. S.K. Sinha (North-Holland, Amsterdam, 1980).

[4] S. Toxvaerd, J. Chem. Phys. 69 (1978) 4750;
F.F. Abraham, in: Ordering in two dimensions, ed. S.K. Sinha (North-Holland, Amsterdam, 1980).

[5] H. Kleinert, Phys. Lett. A 91 (1982) 295; 95 (1982) 38; 96 (1983) 302.

[6] W. Janke and H. Kleinert, Phys. Lett. A 105 (1984) 134; 114 (1986) 255;
S. Ami and H. Kleinert, J. Phys. (Paris) Lett. 45 (1984) 877.

[7] K.J. Strandburg, S.A. Solla and G.V. Chester, Phys. Rev. B 28 (1983) 2717;
K.J. Strandburg, Phys. Rev. B 34 (1986) 3536;
D.A. Bruce, Mater. Forum 4 (1985) 5.

[8] W. Janke and D. Toussaint, Phys. Lett. A 116 (1986) 387.

[9] M. Bretz, J.G. Dash, D.C. Hickernell, F.O. McLean and O.E. Vilches, Phys. Rev. A 8 (1973) 1589; 9 (1974) 2814 [for ^{4}He].
[10] P.A. Heiney et al., Phys. Rev. Lett. 48 (1982) 104;
S.K. Sinha, P. Vora, P. Dutta and L. Passell, J. Phys. C 15 (1982) L275.
[11] R. Pindak, D.E. Moncton, S.C. Davey and J.W. Goodby, Phys. Rev. Lett. 46 (1981) 1135;
S.B. Dierker, R. Pindak and R.B. Meyer, Phys. Rev. Lett. 56 (1986) 1819;
J.D. Brock, A. Aharony, R.J. Birgeneau, K.W. Evans-Lufferodt, J.D. Litser, P.M. Horn, G.B. Stephenson and A.R. Tajbakhsh, Phys. Rev. Lett. 57 (1986) 98;
A. Aharony, R.J. Birgeneau, J.D. Brock and J.D. Litster, Phys. Rev. Lett. 57 (1986) 1012;
D.E. Moncton, R. Pindak and S.C. Davey, Phys. Rev. Lett. 49 (1982) 1865.
[12] H. Kleinert, Phys. Lett. A 97 (1983) 511.
[13] D. Nelson, Phys. Rev. B 26 (1982) 269.