Melting of Wigner-like lattice of parallel polarized dipoles

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Abstract – We show that a triangular lattice consisting of dipolar molecules pointing orthogonally to the plane undergoes a first-order defect melting transition.

About thirty years ago, Nelson and Halperin [1] extended the Kosterlitz-Thouless pair unbinding theory [2] of vortices in a thin layer of superfluid helium to the phase transitions of defects in two-dimensional crystals. They argued that melting would proceed by a sequence of two Kosterlitz-Thouless transitions, the first when dislocations of opposite Burgers vector unbind, creating a hexatic phase, and a second in which disclinations of opposite Frank vector separate. However, they never specified the physical parameter of the crystal which would decide when this melting scenario occurs, rather than a simple first-order melting transition which was previously expected on the basis of out three-dimensional experience. Such a parameter was found in ref. [3], and developed further in [4] and in the textbook [5]. It was shown that a higher-gradient elastic constant called the angular stiffness determines which scenario takes place. Only for a high angular stiffness will the two-step melting process occur. Otherwise the melting transition would be a completely normal first-order process. Computer simulations of the simplest lattice defect model on a lattice confirmed the results [6].

The theory was applied to a Lennard-Jones crystal and a Wigner crystal and it was found that in both cases the angular stiffness was too small to separate the melting transition into two successive Kosterlitz-Thouless transitions.

Here we investigate the angular stiffness for a crystal that is similar to the Wigner crystal, except that the repulsive forces are due to parallel magnetic dipoles. Thus, the potential has the behavior $1/r^3$ rather than $1/r$.

The angular stiffness parameter is defined as follows. Let $\mu$ and $\lambda$ be the usual elastic constants of a crystal, then the usual elastic energy density depends on the displacement field $u_i(x)$ via the strain tensor $u_{ij}(x) \equiv \partial_i u_j(x) + \partial_j u_i(x))/2$ as follows:

$$E = \mu u_{ij}^2 + \frac{\lambda}{2} u_{ij}^2.$$  (1)

The angular stiffness is parametrized by the second of the higher-gradient energies,

$$\Delta E = 2\mu \ell^2 (\partial \omega_j)^2 + \frac{2(\mu + \lambda)}{2} \ell^2 (\partial u_{jj})^2,$$  (2)

where $\omega_j(x) \equiv \frac{1}{2} \epsilon_{jkl} \partial_k u_l(x)$ is the local rotation field. The parameter $\ell^2$ is the length scale of the angular stiffness.

It was argued in [3] that for $\ell = 0$, dislocations are indistinguishable from neighboring pairs of disclinations of opposite Frank vector, and disclinations can be built from strings of dislocations. There the transition is of first order. For high $\ell$, on the other hand, beginning about with the lattice spacing $a_0$, the disclinations could be suppressed with the consequence that the transition based on disclination unbinding would occur later than that of dislocation unbinding. The precise location of the critical $\ell$ was found by computer simulations [6] and is plotted in fig. 1.

In order to apply this criterion to the triangular lattice formed by dipoles we must calculate the elastic constants in $E$ and $\Delta E$. This can be done for any repulsive interatomic potential $\Phi(x) = 1/|x|^p$ of power $p$, which has the value $p = 1$ for the Wigner crystal and $p = 3$ for the crystal of parallel dipoles. For the second derivatives of this potential, we calculate the Fourier transform

$$V_{ij}(k) \equiv \sum_{x \neq 0} [1 - \cos(kx)] \partial_i \partial_j \Phi(x).$$  (3)
If \( M \) is the mass of the lattice constituents, the sound waves of polarization vector \( \varepsilon_i^{(\lambda)}(k) \) have the frequencies determined by

\[
\rho \omega^{(\lambda)2} = V_{ij}(k) \varepsilon_i^{(\lambda)}(k) \varepsilon_j^{(\lambda)}(k),
\]

where \( \rho \) is the mass density of the material.

This can now be compared with the equation of motion following from the Lagrangian density associated with the Lagrangian density,

\[
\mathcal{L} = \frac{\rho}{2} \dot{u}_i^2(x, t) - \mathcal{E} - \Delta \mathcal{E},
\]

which reads

\[
\rho \omega^2 u_i^{(\lambda)}(k) - \mu k^2 (1 + \ell^2 k^2) P^{\ell^2}_i(k) u_j(k) - (2\mu + \lambda) k^2 (1 + \ell^2 k^2) P^{4}_i(k) u_j(k) = 0,
\]

where

\[
P^{\ell^2}_i(k) \equiv \left( \delta_{ij} - \frac{k_i k_j}{k^2} \right), \quad P^4_i(k) \equiv \frac{k_i k_j}{k^2}
\]

are the projections into transverse and longitudinal directions with respect to \( k \).

Thus, we merely have to calculate the transverse part of (3) and determine \( \ell^2 \) from the ratio of the \( k^2 \)-part vs. the \( k^2 \)-part. This is not straightforward. Using the tensor decomposition

\[
\partial_i \partial_j \Phi(x) = A \delta_{ij} + B x_i x_j,
\]

with

\[
A = \Phi'(r)/r = -p/r^{p+2},
\]

\[
B = \Phi''(r)/r^2 - \Phi'(r)/r^3 = p(p + 2)r^{p+4},
\]

we calculate for small \( k \)

\[
V(k) = \sum_{x \neq 0} \left[ \frac{(xk)^2}{2} - \frac{(xk)^4}{24} + \ldots \right] (A \delta_{ij} + B x_i x_j),
\]

and find

\[
V_{ij}(k) = (V_T^{(2)} k^2 + V_T^{(4)} k^4 + \ldots) (\delta_{ij} - \hat{k}_i \hat{k}_j)
\]

\[
+ (V_L^{(2)} k^2 + V_L^{(4)} k^4 + \ldots) \hat{k}_i \hat{k}_j,
\]

where

\[
V_T^{(2)} = 3A r^2/2 + 3Br^4/8 = \frac{3p(2 + 3p)}{8} r^{-p},
\]

\[
V_T^{(4)} = -3Ar^4/32 - Br^6/128 = \frac{p(10 - p)}{128} r^{-2p},
\]

\[
V_L^{(2)} = 3Ar^2/2 + 9Br^4/8 = \frac{3p(p - 2)}{8} r^{-p},
\]

\[
V_L^{(4)} = -3Ar^4/32 - 11Br^6/128 = -\frac{p(10 + 11p)}{128} r^{-2p}.
\]

If the lattice sum were carried out only over the six nearest neighbors at \( r = a_0 \), the length parameter of angular stiffness would be given by

\[
\ell^2 \equiv \frac{1}{a_0^2} \frac{V_T^{(4)}}{V_T^{(2)}} = \frac{10 - p}{p - 2} \frac{48}{45}.
\]

For dipole forces this is equal to \( \ell^2/a_0^2 = 7/48 \approx 0.145 \). Comparing this with the phase diagram of the general lattice defect model shown in fig. 1, we would conclude that the melting transition is weakly of first order.

Let us now see the effect of the full lattice sum. Inspection of (3) shows that the \( k^4 \)-part cannot be calculated directly from the sum over lattice sites since for \( p = 3 \) the extra four powers of \( x \) lead to a logarithmic divergence. To solve this problem we set \( R = |x + u| \) and use Ewald’s formula to rewrite

\[
V_p(k) = \sum_x \frac{\epsilon^{ikx}}{R^p} = \frac{1}{\Gamma(p/2)} \int_0^\infty dt t^{p/2} \sum_x e^{-tR^2 + i kx}.
\]

The sum over the lattice sites converges fast for large \( t \). For small \( t \) it is convenient to perform a duality transformation that converts lattice sums into sums over the reciprocal lattice vectors, \( c \):

\[
\sum_x e^{ikx} = \frac{(2\pi)^D}{v} \sum_c \sigma^{(D)}(k - c).
\]

Inserting this into the Fourier representation of an arbitrary function

\[
f(x) = \int \frac{d^Dk}{(2\pi)^D} \tilde{f}(k)e^{-ixk},
\]

we have
we obtain the relation
\[ \sum_x f(x) = \frac{(2\pi \nu)^D}{V} \sum_c f(c). \] (18)

The function
\[ f(x) = e^{-tR^2+i\mathbf{k} \cdot \mathbf{x}} \]
has a Fourier transform
\[ \tilde{f}(\mathbf{k}') = \frac{\pi^{D/2}}{\nu^{D/2}} e^{-(\mathbf{k}' + \mathbf{k})^2/4t} \]
so that
\[ \sum_x e^{-tR^2+i\mathbf{k} \cdot \mathbf{x}} = \frac{\pi^{D/2}}{\nu^{D/2}} \sum_c e^{-(c+k)^2/4t} \Theta \epsilon \] (20)
Inserting this into eq. (15) we see that now the small-\( t \) part of the integral converges fast. For an optimal convergence we split the integrand at some \( t \)-value \( \epsilon \) and rewrite (15) as
\[ \sum_x \frac{e^{i\mathbf{k} \cdot \mathbf{x}}}{R^p} = \frac{1}{\Gamma(p/2)} \left[ \int_0^\epsilon \frac{dt}{t} t^{p/2} \sum_x e^{-tR^2+i\mathbf{k} \cdot \mathbf{x}} \right. \\
\left. + \frac{\pi^D}{\nu^{D/2}} \int_\epsilon^\infty \frac{dt}{t} t^{p/2} \sum_c e^{-(c+k)^2/4t} \right]. \] (22)
We now introduce the Misra functions
\[ \varphi_n(z) \equiv \int_1^\infty t^n e^{-zt}. \] (23)
They are related to the incomplete Gamma functions
\[ \Gamma(n, z) \equiv \int_z^\infty t^{n-1} e^{-t}, \] (24)
by
\[ \varphi_n(z) = z^{-n-1} \Gamma(n+1, z), \] (25)
and can therefore be expanded as follows:
\[ \varphi_n(z) = z^{-n-1} \left[ \Gamma(n+1) - \sum_{k=0}^\infty (-1)^k \frac{z^{k+n+1}}{k!(k+n+1)} \right]. \] (26)
Using the relation
\[ \int_\epsilon^\infty t^n e^{-zt} = e^{z+1} \varphi_n(z), \] (27)
eq (22) can be written as
\[ V_p(\mathbf{k}) = \sum_x \frac{e^{i\mathbf{k} \cdot \mathbf{x}}}{\nu^p} = \frac{\epsilon^{p/2}}{\Gamma(p/2)} \varphi_{p/2-1}(\epsilon R^2) e^{i\mathbf{k} \cdot \mathbf{x}} \]
\[ + \frac{\pi^{D/2}}{\nu^{D/2}} \sum_c \varphi(D-p)/2-1 \left( \frac{(c+k)^2}{4\epsilon} \right) e^{i\mathbf{k} \cdot \mathbf{c}}. \] (28)
For a triangular lattice with \( D = 2 \), lattice vectors
\[ \mathbf{x} = a_0 \left( l_1 - \frac{1}{2} l_2, \frac{\sqrt{3}}{2} l_2 \right), \quad l_1, l_2 = \text{all integers}, \] (29)
cell volume \( v = \sqrt{3} a_0^2/2 \), and reciprocal lattice vectors
\[ c = \frac{2\pi}{a_0} \left( c_1, \frac{1}{\sqrt{3}} c_1 + \frac{\sqrt{2}}{3} c_2 \right), \quad c_1, c_2 = \text{all integers}, \] (30)
we choose \( \epsilon = \pi/\nu \) so that the arguments \( \epsilon x^2 \) and \( \epsilon^2/4\epsilon \) of the Misra functions run through the same values:
\[ s \equiv \epsilon x^2 = \epsilon a_0^2 \left( l_1 - \frac{1}{2} l_2, \frac{\sqrt{3}}{2} l_2 \right)^2 = \frac{2\pi}{\sqrt{3}} (l_1^2 - l_1 l_2 + l_2^2), \]
\[ s = \frac{\epsilon^2}{4\epsilon} = \frac{(2\pi)^2}{a_0^2} \frac{v}{4\pi} \left( c_1, \frac{1}{\sqrt{3}} c_1 + \frac{\sqrt{2}}{3} c_2 \right)^2 = \frac{2\pi}{\sqrt{3}} (c_1^2 + c_1 c_2 + c_2^2), \] (31)
Then eq. (28) yields the formal relation
\[ \sum_x \frac{1}{|\mathbf{x}|^p} = \frac{\epsilon^{p/2}}{\Gamma(p/2)} \times \sum_s \varphi_{p/2-1}(s) + \frac{\pi^{D/2}}{\nu^{D/2}} \varphi(D-p)/2-1 \left( \epsilon a^2 \right). \] (32)
This becomes meaningful by a subtraction of the \( x = 0 \) term, which yields, after a separate treatment of the \( s = 0 \) term on the right-hand side, if we assume \( p > D \),
\[ \sum_{x \neq 0} \frac{1}{|\mathbf{x}|^p} = \lim_{\epsilon \to 0} \left[ -\frac{1}{|\mathbf{x}|^p} + \frac{\epsilon^{p/2}}{\Gamma(p/2)} \sum_s \varphi_{p/2-1}(\epsilon x^2) \right. \\
\left. + \frac{1}{\Gamma(p/2)} \sum_{s \neq 0} \varphi_{p/2-1}(s) + \frac{\pi^{D/2}}{\nu^{D/2}} \varphi(D-p)/2-1 \left( \epsilon a^2 \right) \right]. \] (33)
We have omitted the \( s = 0 \) term of the last sum since it vanishes for \( p > D \). The limit in the brackets vanishes due to the expansion (26).
Using relation (16) we can rewrite the Fourier-transformed expression (3) as sum over reciprocal lattice vectors:
\[ V_j(k) \equiv \sum_{\epsilon \neq 0} [(c + k)_i (c + k)_j \Phi(c + k)] - c_i c_j \Phi(c). \] (34)
We are now able to calculate the effect of the full lattice sum, splitting the potential in the sum (3) as in (33) into a small- and a large-\( t \) part \( \Phi^S(r) \) and \( \Phi^L(r) \), so that
\[ V_j(k) = V_j^S(k) + V_j^L(k). \] (35)
where

\begin{align*}
V^x_{ij}(k) &= \frac{\varepsilon^{p/2}}{\Gamma(p/2)} \sum_{x \neq 0} [1 - \cos(kx)] \partial_i \partial_j \varphi_{p/2-1}(\varepsilon x^2), \quad (36) \\
V^y_{ij}(k) &= \frac{\varepsilon^{D/2} \varepsilon^{p/2}}{\Gamma(p/2) \varepsilon^D/2} \\
&\times \sum_c \left[ (c + k)_i (c + k)_j \varphi_{(D-p)/2-1} \left( \frac{(k + c)^2}{4 \varepsilon} \right) \\
&- c_i c_j \varphi_{(D-p)/2-1} \left( \frac{c^2}{4 \varepsilon} \right) \right], \quad (37)
\end{align*}

Expanding the sums up to powers \(k^4\) and using the property \(\varphi_n'(z) = -\varphi_{n+1}(z)\), these become, assuming \((D-p)/2 - 1 < 0, \)

\begin{align*}
V^x_{ij}(k) &= \frac{\varepsilon^{p/2}}{\Gamma(p/2)} \sum_{x \neq 0} \left[ \frac{1}{2} (kx)^2 - \frac{1}{24} (kx)^4 \right] \\
&\times \left[ 4\varepsilon^2 x_i x_j \varphi_{p/2+1}(\varepsilon x^2) - 2\varepsilon \partial_i \partial_j \varphi_{p/2}(\varepsilon x^2) \right], \quad (38) \\
V^y_{ij}(k) &= V^y_{ij} = 0(k) + \frac{\pi^{D/2} \varepsilon^{p/2}}{\Gamma(p/2) \varepsilon^{D/2}} \\
&\times \sum_{c \neq 0} \left\{ (c + k)_i (c + k)_j \left[ \varphi_{(D-p)/2-1} \left( \frac{c^2}{4 \varepsilon} \right) \\
&- \left( 2ck + k^2 \right) \varphi_{(D-p)/2} \left( \frac{c^2}{4 \varepsilon} \right) + \ldots \right. \right. \\
&\left. + \left( \frac{-1}{4!} \right)^4 \left( \frac{2ck + k^2}{4 \varepsilon} \right)^4 \varphi_{(D-p)/2+3} \left( \frac{c^2}{4 \varepsilon} \right) \right) \\
&- c_i c_j \varphi_{(D-p)/2-1} \left( \frac{c^2}{4 \varepsilon} \right) \right\}, \quad (39)
\end{align*}

where \(V^y_{ij} = 0(k)\) is the purely longitudinal term

\begin{align*}
V^y_{ij} = 0(k) &= \frac{\pi^{D/2} \Gamma((D-p)/2)}{\Gamma(p/2) \varepsilon^{D/2}} \frac{1}{k} k_{i} k_{j} \left( \frac{D-p}{2} \right) \\
&\times \left[ 1 - \frac{D-p}{4(D+p)} k^2 v + \ldots \right]. \quad (40)
\end{align*}

The higher Misra functions can be reduced to the lower ones by the iteration formula

\begin{align*}
\varphi_{n+1}(z) &= \frac{1}{z} \left[ (n+1) \varphi_n(z) + e^{-z} \right]. \quad (41)
\end{align*}

We now go to \(D = 2\) and \(p = 3\) and find from the nearest neighbors in (38) the sums

\begin{align*}
V^y_T^{(2)}(k) &= \sum_{s \neq 0} \frac{\pi s}{\varepsilon^{3/2}} \left[ 3s \varphi_{(2/2)}(s) - 6 \varphi_{(3/2)}(s) \right] \approx \frac{6.55}{\varepsilon^{3/2}}, \quad (42) \\
V^y_T^{(4)}(k) &= \sum_{s \neq 0} \frac{s^2}{16 \varepsilon^{1/2}} \left[ -8s \varphi_{(4/2)}(s) + 6 \varphi_{(3/2)}(s) \right] \approx \frac{0.54}{\varepsilon^{1/2}}, \\
V^y_L^{(2)}(k) &= \sum_{s \neq 0} \frac{\pi s}{\varepsilon^{3/2}} \left[ 9s \varphi_{(2/2)}(s) - 6 \varphi_{(3/2)}(s) \right] \approx \frac{28.47}{\varepsilon^{3/2}}, \\
V^y_L^{(4)}(k) &= \sum_{s \neq 0} \frac{s^2}{16 \varepsilon^{1/2}} \left[ -11s \varphi_{(4/2)}(s) + 6 \varphi_{(3/2)}(s) \right] \approx \frac{2.58}{\varepsilon^{1/2}}.
\end{align*}

If we include all neighbors, the result changes only little to

\begin{align*}
V^y_T^{(2)}(k) &\approx \frac{7.04}{\varepsilon^{3/2}}, \\
V^y_T^{(4)}(k) &\approx \frac{0.80}{\varepsilon^{1/2}}, \\
V^y_L^{(2)}(k) &\approx \frac{32.53}{\varepsilon^{3/2}}, \\
V^y_L^{(4)}(k) &\approx \frac{2.59}{\varepsilon^{1/2}}.
\end{align*}

The sum over the reciprocal lattice vectors \(c\) in (39) has a purely longitudinal contribution from \(c = 0\):

\begin{align*}
V^y_{ij}(k) &= \frac{k_i k_j}{k^2} \frac{4\pi}{\varepsilon^{1/2}} k^2 \left( 1 - \frac{k \sqrt{\varepsilon}}{2} + \frac{k^2 \varepsilon}{4 \pi} + \ldots \right). \quad (46)
\end{align*}

Of the remaining sum we include only the six smallest \(c\) vectors. We further approximate this sum isotropically by replacing it by 6 times the angular average \(\langle \ldots \rangle_\phi \equiv (2\pi)^{-1} \int_0^{2\pi} \ldots \, d\phi \ldots\), where \(c = (c \cos \phi, c \sin \phi)\). If we define the subtracted quantities

\begin{align*}
\gamma_n &\equiv 6((c + k)_i (c + k)_j (2ck + k^2)\phi - (k = 0), \quad (47)
\end{align*}

we obtain

\begin{align*}
\gamma_0 &= 6k^2, \\
\gamma_1 &= 3c^2 k^2 + (12c^2 + 6k^2)k_1^2, \\
\gamma_2 &= 3c^4 k^2 + 3c^2 k^4 + (6c^4 + 36c^2 k^2)k_1^2, \\
\gamma_3 &= 9c^4 k^4 + 54c^2 k^2 k_1^2, \\
\gamma_4 &= 6c^6 k^4 + 24c^6 k^2 k_1^2. \quad (48)
\end{align*}

Note that only \(\gamma_4\) is affected by the isotropic approximation. The others are the same as in the sum over nearest neighbors. With these \(\gamma_n\)’s we find from (39)

\begin{align*}
V^y_{ij} = 0(k) &= \frac{\pi^{D/2} \varepsilon^{p/2}}{\Gamma(p/2) \varepsilon^{D/2}} \sum_{q = 0}^4 \frac{(-1)^q \gamma_q}{q! (4\varepsilon)^p} \varphi_{(D-p)/2-1+q}(s).
\end{align*}
Summing only over the six nearest neighbors these become

\[ V^{\ell \neq 0}(k) = \frac{2\pi}{v^{3/2}} k^2 \left\{ \frac{3}{2} \left[ -s \varphi_{-1/2}(s) + s^2 \varphi_{1/2}(s) \right] \ight.
\[ + \frac{k^2 v}{\pi} \frac{1}{32} \left[ 6s \varphi_{1/2}(s) - 12s^2 \varphi_{3/2}(s) \right] \
\[ + s^3 \varphi_{3/2}(s) \right\}, \quad \text{(51)} \]

\[ V^{\ell \neq 0}(k) = \frac{2\pi}{v^{3/2}} k^2 \left\{ \frac{1}{2} \left[ 12\varphi_{-3/2}(s) - 30s \varphi_{-1/2}(s) \right] \ight.
\[ + \frac{k^2 v}{\pi} \frac{1}{32} \left[ -48 \varphi_{-1/2}(s) + 156 \varphi_{1/2}(s) \right] \
\[ - 84s^2 \varphi_{3/2}(s) + 5s^3 \varphi_{5/2}(s) \right\}. \quad \text{(52)} \]

Inserting \( s = 2\pi/\sqrt{3} \) we obtain

\[ V_T^{\ell \neq 0(2)} = 0.571 \frac{v^{3/2}}{\ell}, \quad V_T^{\ell \neq 0(4)} = 0.040 \frac{v^{3/2}}{\ell}, \quad \text{(53)} \]

\[ V_L^{\ell \neq 0(2)} = 1.019 \frac{v^{3/2}}{\ell}, \quad V_L^{\ell \neq 0(4)} = 0.250 \frac{v^{3/2}}{\ell}. \quad \text{(54)} \]

Extending the sum to the entire reciprocal lattice, these change to

\[ V_T^{\ell \neq 0(2)} = 0.537 \frac{v^{3/2}}{\ell}, \quad V_T^{\ell \neq 0(4)} = 0.044 \frac{v^{3/2}}{\ell}, \quad \text{(55)} \]

\[ V_L^{\ell \neq 0(2)} = 0.831 \frac{v^{3/2}}{\ell}, \quad V_L^{\ell \neq 0(4)} = 0.258 \frac{v^{3/2}}{\ell}. \quad \text{(56)} \]

Hence we find for \( \ell^2 \)

\[ \ell^2 = 0.0041, \quad \text{(57)} \]

which is very small, thus confirming that the melting transition will be of first order.

Recently, several different criteria for judging the type of melting transitions have been discussed in ref. [7] in connection with the possibility of studying the melting process in two-dimensional suspensions of small colloid spheres [8]. Some of them have dipole moments and show a hexatic phase. Since the interaction forces in these models are more complicated than the pure dipole forces treated here, the order of the transition does not have to follow our criterion. It will be interesting to understand the relation of the criteria in [7] with the simple stiffness criterion in [3].

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