Vortex-Antivortex Crystallization in Thin Superconducting and Superfluid Films

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A new scenario, based on crystallization of oppositely charged vortex-antivortex pairs into a 2D ionic crystal, is proposed for the transition in thin superconducting films with high pair density. We also discuss possible melting of this crystal and the emergence of dissipation. This model is also shown to apply to superfluid He films in cases where the energy to create a vortex is reduced.

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While the Kosterlitz-Thouless (KT) transition [1] has been accepted as pertinent for neutral superfluids such as liquid He films, its applicability for superconducting films was first suggested by Beasley, Mooij, and Orlando [2]. They noted that for a 2D superconductor the relevant penetration depth is $\Lambda=\lambda^2/d$, which can be as large as a typical sample size; hence vortices in very thin superconductors interact logarithmically for all distances (here λ is the London penetration depth and d is the thickness of the film). Thus, the necessary ingredient for the formation of the topological defects of the KT theory is satisfied. The most striking feature of the KT theory is the universal jump stating that the ratio of the superfluid density to the KT transition temperature is universal, i.e., $k_B T_{KT}=1/2(\Phi_0/4\pi)^2/\Lambda(T_{KT})$ [3]. Although the general features of the KT transition were observed in superconducting films, the universal jump was observed only in isolated cases, mostly for granular systems, while discrepancies were found whenever a homogeneous system with large penetration depth was studied [4]. To overcome this difficulty, a dielectric constant ϵ that renormalizes the charge of the vortices has been invoked. In recent experiments on $YBa_2Cu_3O_7$ [5] and MoGe [6] thin films, ϵ was found to be more than 5, a very large enhancement that in principle invalidates the applicability of the vortex-antivortex unbinding KT theory. In none of these systems has any hysteretic behavior or any other indication for a first order transition been found. Although it is acceptable that the background of vortex-antivortex pairs will polarize the medium for any specific pair, and hence an introduction of ϵ is needed, this effect should disappear when the universal ratio is considered. This is because the KT theory is a low density theory, manifested by an $\epsilon=1$ fixed point (in other words, the renormalized fugacity is zero at the transition) [7]. In fact, we claim that the conditions to observe a KT transition in superconducting films in the way suggested by Beasley, Mooij, and Orlando [2] are almost never met. Instead, in this dense state a crystallization of the vortex-antivortex fluid into an "ionic" crystal is inevitable, resulting in the true zero resistance superconducting state. The resistive state is then achieved by melting of the crystal presumably via a new universal KT transition as a result of the unbinding of dislocation pairs. We expect our analysis to equally apply to thin superfluid films.

The thermal energy needed to create a vortex is of order of the condensation energy. For a superconducting film with a coherence length ξ, this energy is of the order of $E_c=0.39(\Phi_0/4\pi)^2/\Lambda$ [8]. A first clue to the understanding of the difficulties in applying the KT theory to thin superconducting films arises if one calculates the "critical" fugacity $y_c=\exp(-E_c/k_B T_{KT})$. For a superconductor (with $\epsilon=1$), we find $y_c=\exp(-2\times 0.39)\approx 0.46$. This is to be contrasted with superfluid He, granular superconductors, or conventional XY ferromagnets [$y_c=\exp(-6.47)\approx 0.0015$ [9]]. The consequence of this difference is dramatic. It is the large fugacity which implies high defects density at the KT transition that prohibits us from using the KT theory in its straightforward form, thus requiring the introduction of a dielectric constant. ϵ can be estimated as a function of y using the renormalization group (RG) approach [10]. Denoting the polarizability and density of dipoles (i.e., vortex-antivortex pairs) by α and n, respectively, one can find in the dilute limit [11] $\epsilon=1+2\pi n\alpha$. The correction to ϵ is proportional to y^2, so that not too close to T_{KT}, $\epsilon=1$ for ferromagnet or for films of superfluid ^{4}He with an extremely good accuracy. In contrast, for superconductors, $\pi n\alpha$ is large and the dilute limit does not apply any more. Thus, Minnhagen [10] has argued that in such a situation RG analysis will give a nonuniversal jump at T_{KT} or even a first order transition. His prediction is that this should occur when y exceeds a value $y^*=0.054$. The corresponding critical value $k_B T_{KT}/(\Phi_0/4\pi)^2/\Lambda(T_{KT})\approx 0.144$ implies a dielectric constant $\epsilon\approx 1.74$, giving a correction to the dielectric constant of order unity. Thus, in such a case it is not clear that Minnhagen's equations hold at all. A more accurate estimate of the dielectric constant needs to take into account the fact that the local field is very different than the external field, i.e., a strong polarization limit. A better way to estimate ϵ is by using the Clausius-Mossotti formula [11] which for 2D gives $\epsilon=(1+\pi n\alpha)/(1-\pi n\alpha)$. A preliminary RG analysis, using the above ϵ and large y, shows a runaway from the KT fixed point which may indicate the flow towards a new fixed point that is consistent with the dense limit discussed here.

In conventional situations, the chemical potential E_c

needed to nucleate defects at $T=0$ is very large, typically of the order of the mean field order-disorder transition. Thus, below the KT transition, the thermodynamics of the defect-antidefect pairs is essentially dominated by energy considerations. By contrast, in superconducting thin films, $\Lambda(0)$ is very large and, as a result, E_c is small even at $T=0$. A consequence of this is that T_{KT} is not much lower than the BCS T_c. In that limit, many vortices will be thermally generated below T_{KT} and their thermodynamics will thus be entropy dominated. For a situation like this, we propose that vortex-antivortex dipoles will crystallize and that the KT transition will be replaced by a melting transition of this crystal at a new temperature $T^* < T_{KT}$ and most probably via the KT dislocation unbinding transition. We propose further that upon cooling the system below a yet lower temperature T_s, the crystal will sublimate into a gas of bound pairs which is identical in structure to the "ordinary" KT phase for superfluids. Figure 1 summarizes our proposal for the new phase diagram. In this figure we also include the predicted hexatic phase that separates the solid and the liquid phases [12] although, as pointed out by Fisher [3], we expect its observation to be difficult experimentally.

Kosterlitz and Thouless [1] have estimated the average size for vortex-antivortex pairs $R=\sqrt{\langle r^2\rangle^{1/2}}$ and the mean separation R_s between pairs. The fact that R grows from $r_0\sim\xi$ to the size of the system (or infinity in the thermodynamic limit) is clearly due to entropy. At a given temperature T, for R not too large, we may define the "trapping" probability as

$$\tau=\left(\frac{R}{R_s}\right)^2=\frac{\pi\exp(-0.78q^2/k_BT\tilde{\epsilon})}{q^2/k_BT\tilde{\epsilon}-2}, \quad (1)$$

where $\tilde{\epsilon}$ is an effective dielectric constant. As shown by Gilman [13], a dipole will trap another dipole if $\tau\sim\frac{1}{16}$. It was then suggested by Li [14] that a row of such dipoles will be formed below a temperature T_{row} satisfying $q^2/\tilde{\epsilon}k_BT_{row}\sim4.1$. A similar effect was suggested by Sommerfeld in the context of the Von Karman vortex row [15]. When these rows of dipoles are formed, they will interact and the resulting structure will be obtained by comparing the energies of the possible 2D solids. For $\tau\sim0.25$, the corresponding structure is that of an ionic crystal and it is obtained for $q^2/\tilde{\epsilon}k_BT_i\sim3.2$. Possible groupings of the vortex rows yielding 2D structures are shown in Fig. 2. If such a crystal is formed, it will melt via dislocation mediated transition *a la* KT. The chemical potential of a dislocation in this ionic crystal is given by $E_c=\eta\mu b^2/2\pi$, where $\eta\approx2.33$, μ is the rigidity modulus of the lattice, and b is the magnitude of the Burgers vector, $b=a\sqrt{2}$ for a square lattice and $b=a\sqrt{3}$ for triangular or hexagonal lattices [16,17]. The melting temperature is given by the relation $k_BT^*=\mu(T^*)b^2/4\pi$. Note that the fugacity for the creation of a single dislocation at this temperature is $y\sim0.0095$ and therefore the corresponding melting transition may be universal.

The scenario we outlined above depends in a crucial way on the actual potential between a vortex and its antivortex pair in the dense phase. Although the pair potential energy is always attractive [10], temperature will stabilize the crystal for $T_s<T<T^*$ and an equilibrium lattice parameter will only be found by minimizing the free energy that includes the potential energy and phonon contributions. The former is obtained by summing up the two-body terms given in Ref. [10] over the lattice. At large distances this self-energy of the structure γ can be calculated as a Madelung sum. For the row structures we consider here, this energy depends on the angle between two successive dipoles (Fig. 2) in the row. If one considers a dilute arrangement of rows, the straight rows (180°) are the most stable ones. When these rows are put together densely to form the crystal, we have to recalculate the Madelung energies. We find $\gamma_{sq}=-1.6155$ for the square lattice and $\gamma_{tr}=-1.36803$ for the frustrated triangular lattice [18,19]. The phonon part is computed in a Debye approximation, given the linear dispersion relation of lattice vibrations found below. The result of our calculation is depicted in Fig. 3 where we plot the bare free energy per vortex (in units of k_BT^*) as a function of a/ξ for various temperatures. A full derivation of

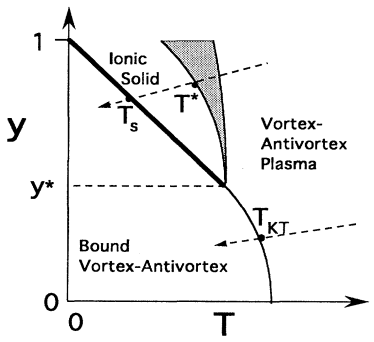

FIG. 1. The predicted phase diagram y is the bare fugacity calculated at T_{KT}. The thick solid line denotes the first order sublimation transition. We mark the two possible scenarios of transition depending on the initial fugacity. The thick dots mark the relevant transition occurring as T is lowered. The shaded area allows for a possible hexatic phase.

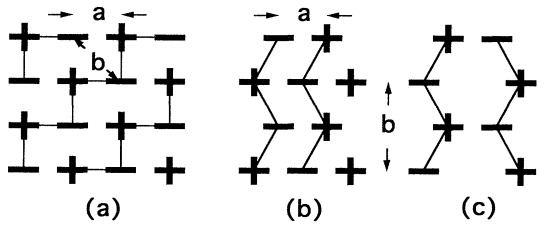

FIG. 2. The possible dipole groupings and lattices. (a) Square, (b) triangular, and (c) hexagonal lattices.

the free energy which takes the self-consistent renormalization of the phonon velocities into account will be given in a separate publication. It shows indeed that a lattice structure can exist in a range of temperatures between T_s and T^*. The above argument implies that the short range repulsion has to contribute to the lattice "spring constant" and hence to $\mu(T^*)$. If we use the attractive potential only, we can find a lower bound on T^*. This bound can be expressed in terms of the universal jump for the low-density KT theory and it gives a 4π times larger jump for the square lattice and a $4\pi/\sqrt{3}\approx 7.25$ times larger jump for the triangular lattice.

We now turn to the calculation of the vibrational spectrum of the crystal and first consider the situation where, in equilibrium, the velocity field due to all the lattice on a single vortex (of any sign) is zero. The frustrated cases will always yield some nonzero local current as will be discussed below. Following Fetter and Hohenberg [20], we denote the displacements of the lattice points as $\mathbf{u}_L^{\pm}(t)=\mathbf{s}^{\pm}\exp i(\mathbf{q}\cdot\mathbf{r}_L^{\pm}-\omega t)$ where + and − correspond to positively and negatively charged vortices, respectively. The velocity field of a vortex located at $\mathbf{r}_L$ is $\mathbf{v}(\mathbf{r}_L)=(-1)^L(\bar{K}/2\pi)(\hat{\mathbf{z}}\cdot\mathbf{x}\hat{\mathbf{r}}_L)f(\mathbf{r}_L)$; $\bar{K}=h/2m$ is the quantum diffusion constant. Note that the above equation will apply to a similar state in He films, if we write $\bar{K}=h/m_{\mathrm{He}}$. The eigenvalue problem can be solved by solving for the coefficients of $s_x^{\pm}$ and $s_y^{\pm}$ for each type of lattice. The exact determination of the modes depends on the function $f(\mathbf{r})$ which for our case contains the information about the long range attraction and the short range repulsion. In general we expect four modes corresponding to transverse and longitudinal modes, each optical or acoustic. In a continuum approximation where we neglect the repulsive interaction, the transverse and longitudinal modes, ω_T and ω_L, are degenerate [21]. However, beyond this approximation and including short range repulsion and the existence of a lattice, these modes have to split. Since the short range repulsion is obtained with a Debye approximation we can only estimate the splitting from the size of the jump at T^*. The closer T^* is to the lower bound calculated above, the less is the splitting. We may use the continuum approximation to calculate the gap between the optical and acoustic modes. Solving the equivalent equations as in Ref. [21], we find two solutions:

$$\omega_1^2=\tfrac{1}{2}n\bar{K}\{4e(e-1)+\tfrac{1}{4}[\lambda-e(2e-1)]q^2a^2\},$$
$$\omega_2^2=\tfrac{1}{8}n\bar{K}[\lambda-e]q^2a^2, \qquad (2)$$

where the first is the optical branch and the second is the acoustic branch. $\lambda=2$ for the square lattice and $\lambda=3$ for the triangular lattice and n is the density of like type vortices. In Eq. (2) $(e-1)$ is the small parameter that determines the gap [$(e-1)=0$ corresponds to the solution for only one type of vortices [20]]. By evaluating the lattice sums to first order in q^2a^2 we find $(e-1)\approx 0.04$ for the square lattice, but this parameter is zero within the accuracy of the calculation for the triangular and hexagonal lattices. The above analysis suggests that if the short range interaction will be included, distinct longitudinal and transverse modes will be present but possibly degenerate acoustic and optical modes. Such a picture is of course consistent with a Euler dynamics that governs the motion of vortices. To determine the preferred lattice structure, it is easy to see that the square lattice will not be stable against shear fluctuations since its (1,1) direction contains the same charges in each row [17]. The same applies for the triangular condenser [18] and the hexagonal lattices. However, the frustrated triangular crystal always has $(+-)$, $(++--)$ arrangements and their opposites for any lattice direction. This ensures stability against shear fluctuations and against local annihilations. For this lattice, $\tilde{\epsilon}(T_{\mathrm{Tr}}^*)=8\pi/3.2\sqrt{3}\sim 4.5$. Taking again $n\alpha\sim q^2(R/R_s)^2/2$ we find that $\tilde{\epsilon}^{-1}=0$ for $T\ll T_{\mathrm{Tr}}^*\equiv T^*$ and T_{row}. This means that $\tilde{\epsilon}$ is very large in all the temperature ranges where melting of the ionic crystal occurs. Note that for this lattice the microscopic velocity field is nonzero but pinning; boundaries and domain formation will always ensure global zero local velocity. The stability of the triangular arrangement is further ensured by the coupling between the short and long wavelength modes [19].

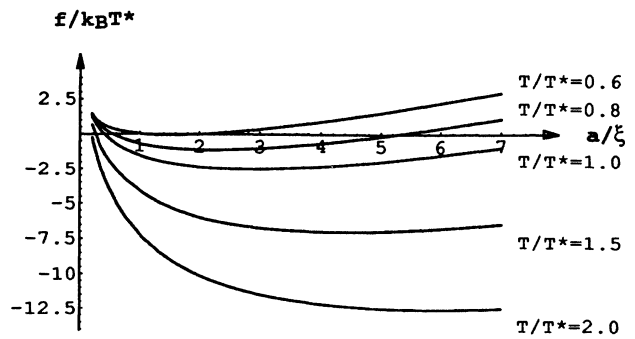

FIG. 3. Free energy of the vortex-antivortex solid (in units of k_BT^*) vs lattice parameter (in units of ξ) for temperatures T/T^* between 0.6 and 2. The lattice is unstable at low T/T^* ($a/\xi\approx 1$) and at high T/T^* (no minimum exits).

The consequences of our theory are rather dramatic. A new phase diagram is predicted on the temperature axis. Assuming a material with $y>y^*$, we find that the flow will be towards the new, "crystalline" fixed point. However, as the temperature is lowered further, a new transition will occur to the ordinary KT phase with bound vortex-antivortex pairs. The sublimation of the solid phase at this T_s temperature is expected to be a first order transition since it reflects a gas-solid phase transition when the density is varied. Recently, Lee and Teitel [19] have carried out Monte Carlo simulations of the 2D neutral Coulomb gas of integer charges on a triangular lattice as a function of temperature and chemical potential.

They indeed found a KT phase for low fugacities and an ionic crystal phase at higher fugacities. The transition between these two phases is indeed first order.

Our analysis above should also apply for the analogous situation of liquid ^{4}He films, in cases where the energy to create a vortex is very low. Such an experimental realization was recently investigated by Chen, Roesler, and Mochel [22] where they studied thin ^{4}He films on hydrogen. As in their experiment, we expect that a third sound measurement will detect two modes corresponding to the superfluid density and the elastic response of the vortex lattice consisting of an admixture of the longitudinal and transverse modes depending on the boundary conditions of the experiment. It is interesting to note that such an argument may suggest that in a dynamical measurement, the superfluid density mode and the lattice modes will appear 90° out of phase from each other as a result of the magnus force that governs the dynamics of the vortices. Indeed, the two effects are both observed in all the experiments of ^{4}He on hydrogen [23]. Moreover, a new set of results where ^{3}He is introduced to the ^{4}He film to decrease the bare chemical potential (^{3}He acts as an impurity that reduces further the already low energy to produce a vortex core and hence will increase y) shows that indeed T^* is hardly affected but T_s is strongly reduced in accord with our proposed phase diagram. A similar analysis has been recently given to this experiment by Zhang [24]. Although such experiments are carried out for He films and show results consistent with our scenario, no equivalent experiment was done to date for superconducting films.

Finally we need to discuss the influence of disorder. For the ordinary KT transition one does not expect disorder to be relevant by the Harris criterion [25] ($a_{xy} = -\infty$). This is easy to understand from energy considerations since the vortex-antivortex energy is $U_{v\bar{v}} \sim \log(R)$ whereas the pinning energy is constant for the pair. On the other hand, Larkin and Ovchinikov [26] have shown that disorder will disrupt long range positional order beyond a characteristic length $L_{\mathrm{LO}} \sim \mu^{3/2}/W$ where W is the width of the pinning force distribution. For the ionic crystal the melting temperature is less than the vortex-antivortex transition. The average intervortex distance is several times the coherence length. Thus, μ is rather large and we expect a very large positional correlation length. In fact, we may find that for small enough samples order is maintained throughout the whole system. If, however, L_{LO} is small, we may at most expect a hexatic phase that preserves the orientational order [27]. In such a case, it is not clear that the triangular arrangement will prevail. A lattice may win with a yet lower T^* transition.

In conclusion, we predict a new phase diagram for superfluids with small vortex creation energy. We find a new "ionic solid" phase that forms below the superfluid transition. This phase sublimates to a bound vortex-antivortex KT phase at lower temperatures. Our analysis shows that the most stable crystal structure for the crystal phase is the frustrated triangular lattice.

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[1] J. M. Kosterlitz and D. J. Thouless, J. Phys. C **6**, 1181 (1973).
[2] M. R. Beasley, J. E. Mooij, and T. Orlando, Phys. Rev. Lett. **42**, 1165 (1979).
[3] S. Doniach and B. Huberman, Phys. Rev. Lett. **42**, 1169 (1979); D. S. Fisher, Phys. Rev. B **22**, 1190 (1980).
[4] J. E. Mooij, in *Percolation, Localization, and Superconductivity*, edited by A. M. Goldman and S. A. Wolf (Plenum, New York, 1984), p. 325.
[5] A. T. Fiory, A. F. Hebard, P. M. Mankiewich, and R. E. Howard, Phys. Rev. Lett. **61**, 1419 (1988).
[6] A. Yazdani, M. R. Hahn, W. White, P. Lerch, M. R. Beasley, and A. Kapitulnik (to be published).
[7] See, e.g., A. P. Young, Phys. Rev. B **19**, 1855 (1979).
[8] J. Hu, Phys. Rev. B **6**, 1756 (1972).
[9] J. V. Jose, L. P. Kadanoff, S. Kirpatrick, and D. R. Nelson, Phys. Rev. B **16**, 1217 (1977).
[10] P. Minnhagen, Rev. Mod. Phys. **59**, 1001 (1987); A. Jonsson, P. Minnhagen, and M. Nylen, Phys. Rev. Lett. **70**, 1327 (1993).
[11] See, e.g., N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Rhinehart, and Winston, New York, 1976), p. 539.
[12] B. I. Halperin and D. R. Nelson, Phys. Rev. Lett. **41**, 121 (1978).
[13] J. J. Gilman, in *Dislocations in Solids*, Discussions of the Faraday Society Vol. 38 (University Press, Aberdeen, 1964), p. 123.
[14] J. C.M. Li, *Dislocations in Solids* (Ref. [12]), p. 138.
[15] A. Sommerfeld, *Mechanics of Deformable Bodies* (Academic, New York, 1950), p. 231; J. L. McCauley, Jr. and C. W. Allen, Phys. Rev. B **25**, 5680 (1982).
[16] H. B. Huntington and J. E. Dickey, Phys. Rev. **100**, 1117 (1955).
[17] J. Friedel, *Dislocations* (Pergamon, New York, 1964).
[18] Another possibility is to collect the dipoles with a 60° angle. In this less favorable case $\gamma_{\mathrm{Tr}} = -1.1533$.
[19] J. R. Lee and S. Teitel, Phys. Rev. Lett. **66**, 2100 (1991).
[20] A. L. Fetter and P. C. Hohenberg, Phys. Rev. **159**, 330 (1967).
[21] A. L. Fetter, P. C. Hohenberg, and P. Pincus, Phys. Rev. **147**, 140 (1966).
[22] M. T. Chen, J. M. Roesler, and J. M. Mochel, J. Low Temp. Phys. **89**, 125 (1992).
[23] J. M. Mochel (private communication).
[24] S. C. Zhang, following Letter, Phys. Rev. Lett. **71**, 2142 (1993).
[25] A. B. Harris, J. Phys. C **7**, 1671 (1974).
[26] A. I. Larkin and Yu. N. Ovchinikov, J. Low Temp. Phys **34**, 409 (1979).
[27] E. M. Chudnovsky, Phys. Rev. B **40**, 11 355 (1989).