Fluctuations of Vortices in Layered High-T_c Superconductors

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(Received 3 March 1992)

The temperature T_s of spontaneous creation of vortex lines in Josephson coupled layered superconductors is obtained by taking into account the entropy contribution to the free energy due to thermal distortions. For Bi- and Tl-based high-T_c superconductors and superlattices, the superconducting critical temperature T_s lies noticeably below the mean-field transition temperature T_{c0} ($T_{c0}-T_s \approx 4$ K or more). The contribution of thermal distortions to the free energy causes significant changes in the temperature and field dependences of magnetization below T_s seen in Bi compounds.

PACS numbers: 74.60.Ge

The importance of topological excitations in the thermodynamics of two-dimensional (2D) magnets and superconductors was first discussed by Berezinskii and Kosterlitz and Thouless [1]; for a review see [2]. Above the Kosterlitz-Thouless (KT) transition temperature T_{KT}, which lies below the mean-field transition temperature T_{c0}, thermally induced free vortices are present in 2D superconductors giving rise to dissipation and persistent-current decay. The spontaneous creation of free vortices above T_{KT} occurs due to the entropy contribution to the free energy $F=\varepsilon-T\sigma$, where $\varepsilon=(\phi_0^2 d/16\pi^2\lambda^2)\ln(R/\xi)$ and $\sigma=\ln(R^2/\xi^2)$ are the vortex energy and its entropy (ϕ_0 is the flux quantum, λ is the London penetration depth, ξ is the coherence length, πR^2 and d are the system area and thickness). Above T_{KT}, which is given implicitly by

$$T=\frac{\phi_0^2 d}{32\pi^2\lambda^2(T)}, \tag{1}$$

the free energy is negative, and spontaneous creation of vortices is favorable.

Generalizations of the KT approach to layered (but still three-dimensional) high-T_c superconductors [3–5] were stimulated by experiments that show the KT scaling behavior of resistivity; Bi-2:2:1:2 and superlattices YBaCuO/PrBaCuO are perhaps the best example (see review [6] and references therein). The idea behind the generalizations was brought about by the observation that in the absence of interlayer superconducting coupling, 2D vortices of different layers interact only via the magnetic field, which provides the same logarithmic dependence of the vortex energy ε on the sample size as in 2D superconductors [5, 7]. This made feasible the KT transition [3–5], i.e., spontaneous creation of 2D vortices and antivortices within the same layer above the temperature obtained by solving Eq. (1) with d replaced by the interlayer distance s. However, even in the case of weak Josephson coupling [8], ε increases linearly with R [9], while the entropy σ of a 2D vortex still changes only logarithmically. In these circumstances, thermally induced spontaneous creation of free 2D vortices (in the low concentration limit) is virtually impossible.

In the following, taking into account the entropy contribution to the free energy due to thermal distortions of vortices, we show that thermally induced spontaneous creation of vortex lines should occur in Josephson coupled layered materials above some temperature $T_s < T_{c0}$. We estimate the difference $T_{c0}-T_s$ to be as high as 5–10 K for Bi- and Tl-based high-T_c compounds and superlattices. We also show that in highly anisotropic superconductors, the entropy contribution to the free energy cannot be neglected in a broad temperature domain below T_s. This causes the field and temperature dependence of the magnetization M to deviate from the standard London behavior, as observed in Bi-2:2:1:2 [10, 11] and Bi-2:2:2:3 [12].

Let us consider a lattice of vortices in a magnetic field $H \ll H_{c2}(T)$ applied perpendicular to the layers; $H_{c2}(T)$ is the corresponding upper critical field. We introduce the induction B and denote the distortions of 2D vortices from their equilibrium positions by $\mathbf{u}(n,\mathbf{r}_\nu)$, where $\mathbf{r}_\nu$ is the 2D coordinate of the vortex pancake ν in the layer n, $\mathbf{u}=(u_x,u_y)$, $\mathbf{r}=(x,y)$. This description implies that vortex lines within the Lawrence-Doniach (LD) approach are, in fact, correlated stacks of 2D vortices rather than *continuous* lines. For the undistorted lattice, the stacks are situated along straight lines. The very concept of correlated stacks of pancakes assumes that distortions within a line do not exceed the interline distance:

$$\langle[\mathbf{u}(n,\mathbf{r}_\nu)-\mathbf{u}(n+1,\mathbf{r}_\nu)]^2\rangle \le \phi_0/B, \tag{2}$$

where $\langle\cdots\rangle$ stands for thermodynamic averaging. Also, we assume that the Josephson coupling between pancakes in different layers is strong in comparison with their electromagnetic interaction: $\lambda_J \ll \lambda_{ab}$. Here $\lambda_J=\gamma s$ is the Josephson length, $\gamma=\lambda_c/\lambda_{ab}$ is the anisotropy ratio, λ_{ab} and λ_c are the penetration depths. [Under this condition, vortex *lines* (rather than weakly correlated 2D pancakes) are spontaneously created above the critical temperature T_s.]

In the following we use the harmonic approximation in vortex distortions to obtain the free energy of the mixed state in the LD framework. The approximation is valid provided $\langle[\mathbf{u}(n,\mathbf{r}_\nu)-\mathbf{u}(n+1,\mathbf{r}_\nu)]^2\rangle \ll \lambda_J^2$; we check *a posteriori* that this condition is fulfilled at all relevant temperatures and fields. The free energy density functional in terms of B and $\mathbf{u}(n,\mathbf{r}_\nu)$ is

$$\mathcal{F} = F_0(B) + \mathcal{F}_{\rm el}\{B,\mathbf{u}(n,\mathbf{r}_\nu)\}\,, \tag{3}$$

where $F_0(B)$ is the free energy density of the undistorted lattice:

$$F_0(B) = \frac{B\phi_0}{16\pi^2\lambda_{ab}^2}(\ln\kappa + 0.5)\,, \quad B\to 0\,, \tag{4}$$

$$F_0(B) = \frac{B\phi_0}{32\pi^2\lambda_{ab}^2}\ln\frac{\eta\phi_0}{2\pi\xi_{ab}^2 B}\,, \quad \frac{\phi_0^2}{4\pi\lambda_{ab}^2} \ll B \ll H_{c2}\,. \tag{5}$$

Here η is a parameter of the order unity, $\kappa = \lambda_{ab}/\xi_{ab}$, and the term $B^2/8\pi$ (irrelevant here) is omitted in Eq. (5). The term $\mathcal{F}_{\rm el}$ represents the elastic energy density:

$$\mathcal{F}_{\rm el} = \frac{1}{2s}\int\frac{d\mathbf{k}}{(2\pi)^2}\int_0^{2\pi}\frac{dq}{2\pi}\sum_{i,j}(c_L k^2 P_{L,ij} + c_{66}k^2 P_{T,ij} + \delta_{ij}c_{44}Q^2)u_i(q,\mathbf{k})u_j^*(q,\mathbf{k})\,. \tag{6}$$

Here $\mathbf{k}=(k_x,k_y)$, $i,j = x,y$, c_{66}, c_L, and c_{44} are the shear, bulk, and tilt moduli, and $P_{L,ij} = k_ik_j/k^2$ and $P_{T,ij} = \delta_{ij} - P_{L,ij}$ are longitudinal and transverse projection operators [13]. Further, $Q^2 = 2(1-\cos q)/s^2$ and $\mathbf{u}(q,\mathbf{k})$ is the Fourier component of distortions; the integration over $\mathbf{k}$ is performed in the region $k^2 < k_0^2 = 4\pi B/\phi_0$. The elastic moduli of a Josephson coupled system ($\xi_c \ll s$) with $\xi_{ab} \ll \lambda_J \ll \lambda_{ab}$ (moderate anisotropy) have been evaluated in Ref. [14]:

$$c_{66} = \frac{\phi_0 B}{(8\pi\lambda_{ab})^2}\,, \quad c_{44} = \frac{B\phi_0}{2(4\pi\lambda_c)^2}\ln\frac{\xi_{ab}^{-2}}{k_0^2+\lambda_J^{-2}}\,, \tag{7}$$

$$c_{11} = c_L + c_{66} = \frac{B^2\gamma^2}{4\pi(1+\lambda_c^2k^2+\lambda_{ab}^2Q^2)}.$$

The main contribution of thermal distortions to the free energy comes from *large* $Q\sim 1/s$ and $k\sim k_0$.

We now evaluate the statistical sum

$$\ln Z = \sum_{q,\mathbf{k}}\ln\int\frac{d\mathbf{u}(q,\mathbf{k})k_0^2}{4\pi^2\alpha s\xi_{ab}^2}\exp\left(-\frac{\mathcal{F}}{T}\right)\,. \tag{8}$$

The elementary phase area for the variable $\mathbf{u}(n,\mathbf{r}_\nu)$ is $\alpha\pi\xi_{ab}^2$, the area of the normal core (α is a parameter of the order unity). In other words, similar to the Kosterlitz-Thouless theory (see [2]), we consider the pancakes as classical particles of the area $\alpha\pi\xi_{ab}^2$; the number of different configurations of a pancake within the layer is $\int du_x du_y/\alpha\pi\xi_{ab}^2$. Performing the integration over $\mathbf{u}(q,\mathbf{k})$ and summation over $(q,\mathbf{k})$, we obtain the free energy density $F(B,T) = F_0(B,T) + F_{\rm th}(B,T)$:

$$F_{\rm th} = -\frac{TB}{\phi_0 s}\ln\frac{32\pi^2T\kappa^2\lambda_J^2}{\alpha\phi_0^2 s\ln(\lambda_J/\xi_{ab})}\,, \quad B \ll B_{\rm cr}\,, \tag{9}$$

$$F_{\rm th} = -\frac{TB}{\phi_0 s}\ln\frac{16\pi\sqrt{e}T\kappa^2}{\alpha\phi_0 sB}\,, \quad B \gg B_{\rm cr}\,, \tag{10}$$

with the crossover field

$$B_{\rm cr} = \frac{\phi_0}{\pi\lambda_J^2}\ln\frac{\lambda_J/\xi_{ab}}{4\sqrt{\ln(\lambda_J/\xi_{ab})}}\,. \tag{11}$$

The limit $B\to 0$ yields the free energy of a single vortex. The line energy of a single vortex $\epsilon(T)$ renormalized by thermal fluctuations has been discussed in Refs. [4, 15]; here we obtain

$$\epsilon(T) = \frac{\phi_0^2}{16\pi^2\lambda_{ab}^2(T)}(\ln\kappa+0.5) - \frac{T}{s}\ln\frac{32\pi T\kappa^2\lambda_J^2}{\phi_0^2 s\alpha\ln(\lambda_J/\xi_{ab})}. \tag{12}$$

Above a temperature T_s defined by $\epsilon(T_s) = 0$, the free energy of a single vortex is negative. In other words, *spontaneous creation of vortex lines* becomes possible at $T \geq T_s$. Thus we have for T_s

$$f(T_s) = \frac{1}{2(\ln\kappa+0.5)}\ln\frac{(\lambda_J/\xi_{ab})^2}{f(T_s)\alpha\pi\ln(\lambda_J/\xi_{ab})}\,, \tag{13}$$

$$f(T) = \phi_0^2 s/32\pi^2 T\lambda_{ab}^2(T). \tag{14}$$

For $\lambda_J \ll \lambda_{ab}(0)$ the right-hand side of (13) is smaller than unity, and therefore T_s lies above T^*, given by $f(T^*) = 1$. Assuming $T_{c0} - T_s \ll T_{c0}$ and taking $\lambda_{ab}(T) = 0.7\lambda_{ab}(0)/\sqrt{t}$, $t = 1 - T/T_{c0}$, we obtain

$$t_s = \frac{t^*}{2(\ln\kappa+0.5)}\ln\frac{t^*(\lambda_J/\xi_{ab})^2}{\alpha\pi\ln(\lambda_J/\xi_{ab})}\,, \tag{15}$$

$$t^* = 16\pi^2T_{c0}\lambda_{ab}^2(0)/\phi_0^2 s. \tag{16}$$

In the absence of applied field, in the temperature interval $T_s < T < T_{c0}$, the superconducting order parameter is nonzero, and the quasiparticle density of states has a gap or a pseudogap which can be probed by NMR Knight shift, tunneling, or optical measurements. The presence of thermally induced vortex and antivortex lines in this temperature interval (*in zero applied field*) results in fluctuations of the magnetic field in the sample. The amplitude of these fluctuations is of the order $\phi_0 t_s/4\pi\lambda_{ab}^2(0)$ (≈ 1 Oe). They can be, in principle, detected using muon spin rotation, neutron scattering, or the Hall probe technique. The thermally induced vortex and antivortex lines give rise to dissipative properties at temperatures above T_s similar to the case of a 2D superconductor above $T_{\rm KT}$; the resistivity and lower critical field $H_{c1}(T)$ became zero at T_s rather than at T_{c0}. One can speculate that above a certain temperature, the lines will dissociate into free 2D vortices causing the KT scaling behavior of resistivity.

Below T_s, in the presence of an applied field we can calculate the magnetization renormalized by thermal fluctuations: $M = -\partial(F_0 + F_{\rm th})/\partial B$. The results for M so obtained are certainly valid below the melting temperature T_m of the vortex lattice. We argue that in a dense vortex liquid, $B \gg \phi_0/\lambda_{ab}^2$, they are valid above T_m as well. Actually, F_0 depends weakly on the lattice structure due to the long-range interaction of vortices in a dense vortex phase; only the factor η under the logarithm of Eq.(5) differs slightly for different lattices. The contribution of thermal distortions to the free energy is determined mainly by the short wave distortions which depend on the short-range order in the system of vortex lines (it is the long-range order that changes at the melting transition, while the short-range order does not). The absence of any noticeable anomaly in the experimental T dependence of M [10–12] confirms this picture. Thus the discussion of the magnetization which follows is irrespective of whether the lattice is melted or not.

Let us consider the magnetization changes due to the contribution of thermal distortions to the free energy. It is seen from Eqs. (9) and (10) that $M_{\rm th} = -\partial F_{\rm th}/\partial B$ is linear in $\ln B$ for $B \gg B_{\rm cr}$, whereas it is B independent for $B \ll B_{\rm cr}$. Hence, the slope of M vs $\ln B$ changes in the vicinity of $B_{\rm cr}$:

$$\left(\frac{\partial M}{\partial \ln B}\right)_{B\ll B_{\rm cr}} - \left(\frac{\partial M}{\partial \ln B}\right)_{B\gg B_{\rm cr}} = \frac{T}{\phi_0 s}. \qquad (17)$$

Therefore, it is possible to determine s from the field dependence of magnetization. Moreover, the value of the crossover field $B_{\rm cr}$ provides information on the Josephson length λ_J and, hence, on the anisotropy parameter γ. For Bi-2:2:1:2 with anisotropy parameter $\gamma \approx 55$ [16] the value $B_{\rm cr}$ should be about 0.1 T.

For fields $B_{\rm cr} \ll B \ll H_{c2}(T)$, we have

$$-M = \frac{T}{\phi_0 s}\left[f(T)\ln\frac{\eta\phi_0}{2\pi e\xi_{ab}^2 B} - \ln\frac{16\pi T\kappa^2}{\alpha\phi_0 sB\sqrt{e}}\right], \qquad (18)$$

and

$$\frac{\partial M}{\partial \ln B} = \frac{T}{\phi_0 s}[f(T)-1] \approx \left(1-\frac{T}{T^*}\right)\frac{\phi_0}{16\pi^2\lambda_{ab}^2(0)}. \qquad (19)$$

It is worth noting that M is *field independent* at $T = T^*$:

$$-M(T^*) = \frac{T^*}{\phi_0 s}\ln\frac{\eta\alpha}{\sqrt{e}}. \qquad (20)$$

Thus all $M(T)$ curves for different $B \gg B_{\rm cr}$ cross at $T = T^*$, the feature clearly seen in the data on Bi-2:2:1:2 [10–12]. Theoretical curves $M(T)$ for different fields along with the data [10] are shown in Fig. 1. Above T^* the magnetization value, $-M$, for fields $B \gg B_{\rm cr}$ *increases* logarithmically with field, while below T^* it behaves "normally," i.e., *decreases* logarithmically with

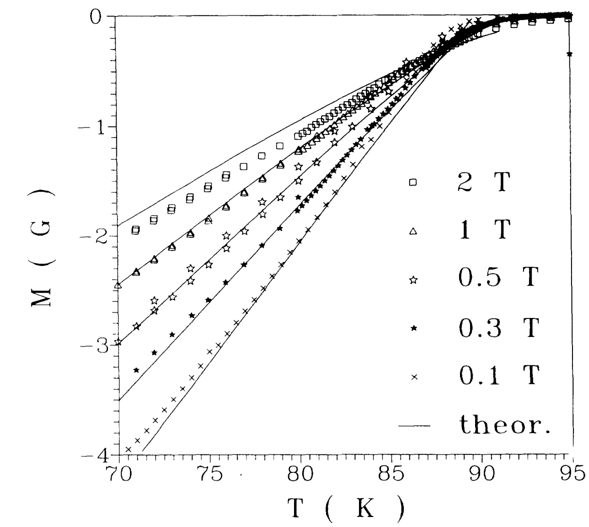

FIG. 1. The calculated magnetization $M(T)$ for different fields and the experimental data [10] for Bi-2:2:1:2.

B. This peculiar behavior is clearly seen in the data [10–12]. The experimental data for the T dependence of $\partial M/\partial \ln B$ in fields $B_{\rm cr} \ll B \ll H_{c2}(T)$ [10] are in agreement with Eq. (19) which makes it possible to determine the system parameters. For the sample of Bi-2:2:1:2 [10] with T^*=88.3 K we obtain $\lambda_{ab}(0) = 1500$ Å, $\xi_{ab}(0)/\sqrt{\eta} = 17$ Å, and T_{c0}=95 K. For another sample of the same material [11] with T^*=87.2 K the parameters are $\lambda_{ab}(0) = 1700$ Å, $\xi_{ab}(0)/\sqrt{\eta} = 15$ Å, and T_{c0}=96 K. Evaluating these parameters we set $\ln \eta\alpha/\sqrt{e} = 1$ [17] and $s = 15$ Å. The parameters obtained were used to calculate theoretical curves in Fig. 1. For Bi-2:2:1:2 we estimate $T_s \approx 92$ K.

The results obtained are valid provided the Josephson model for the interlayer coupling can be applied. In particular, this implies that both T_s and T^* should be situated in the temperature interval where $\xi_c(T) \ll s$. Using Eq. (16) we see that this is fulfilled for sufficiently strong anisotropies: $\gamma^2 \gg \gamma_0^2 = \phi_0^2/8\pi^2\kappa^2 sT_{c0}$. For high-$T_c$ materials we estimate $\gamma_0^2 \approx 100$. Hence, our theory applies to Bi and Tl compounds and superlattices; for $YBa_2Cu_3O_7$ the above condition is not satisfied [18]. We note also that due to the condition (2) we cannot apply our model well above T^*. This condition is fulfilled if $B \ll B_{\rm cr}$; when $B \gg B_{\rm cr}$, we have in addition a temperature restriction $\pi f(T) \geq \ln(\lambda_J^2 B/\phi_0)$. If this condition is violated one may expect the vortex lines to dissociate into an ensemble of 2D pancakes in which vortex lines can no longer be defined.

Thus, in layered superconductors with moderate anisotropy $\xi_{ab} \ll \lambda_J \ll \lambda_{ab}$ the spontaneous creation of vortex and antivortex lines should occur above T_s, which differs considerably from T_{c0}. If $H = 0$ and $T_s < T < T_{c0}$, a liquid phase of vortex and antivortex lines should exist similar to what occurs above the KT transition in 2D superconductors. The substantial difference between this phase and the KT "plasma" of 2D vortices is that, in the Josephson coupled layered materials, the thermally activated topological excitations are *vortex lines*. The field and temperature dependences of magnetization below T_s are strongly affected by the contribution of thermal distortions to the free energy. The characteristic point here is T^* where all $M(T)$ curves for different fixed fields $B \geq B_{\rm cr}$ intersect.

In conclusion we note that spontaneous creation of vortex lines above T_s is not an exclusive property of Josephson coupled layered superconductors. It should also occur in anisotropic 3D materials (e.g., in Y-1:2:3 and Y-1:2:4), but there T_s is closer to T_{c0}, the situation analogous to the $T_{\rm KT}$ for thick superconducting films [18].

The authors acknowledge stimulating discussions with J. Clem. We thank D. Finnemore, M. Maley, and K. Kadowaki for providing the data for our analysis. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-Eng-82. The work was supported by the Director for Energy Research, Office of Basic Energy Sciences, and in part through the Midwest Superconductivity Consortium, Grant No. DE-FG02-90ER45427. One of us (M.L.) acknowledges support of the NSF Grant No. JF 898 under U.S.–Yugoslavia Cooperative research program and the ORNL Pilot Center for Superconductivity.

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[17] For high fields $H \gtrsim H_{c2}(T)/3$ the magnetization $M^*(T^*)$ obtained within a scaling theory [Z. Tešanović, L. Xing, L. Bulaevskii, Q. Li, and M. Suenaga (to be published)] is given by (20) without the factor $\ln(\eta\alpha/\sqrt{e})$. According to experimental data in both Bi-2:2:1:2 and Bi-2:2:2:3, the crossing point (T^*, M^*) is approximately the same for low $[H \ll H_{c2}(T)]$ and high fields. This is possible only if $\ln(\eta\alpha/\sqrt{e}) \approx 1$. The value $4\pi M^* = -0.37$ G for Bi-2:2:1:2 [11] corresponds to $s = 16.5$ Å which is quite close to half of the cell size c ($c \approx 30$ Å). This suggests that in Bi-2:2:1:2, two CuO_2 layers separated by Ca are strongly coupled and can be considered as a single superconducting layer; these combined layers form the Josephson coupled system. In the unit cell there are two such combined layers; they are equivalent as far as the electron properties are concerned. Thus, $s = c/2$. To determine $\lambda_{ab}(0)$ we have used the relation $M^* = T^*/\phi_0 s$ and s=15 Å.

[18] For superconductors described by the Ginzburg-Landau functional with anisotropic masses, all expressions obtained above are retained, but s should be replaced by $C\xi_c(T)$, with a numerical factor C of the order unity, and the factor $\ln(\lambda_J/\xi_{ab})$ should be omitted. We estimate T_s to be about 1 K below T_{c0} for Y-1:2:3 with $\gamma \approx 8$.