RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 51, NUMBER 9 1 MARCH 1995-I

Low-temperature increase of resistive critical fields in certain superconductors: A simple fluctuation approach

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(Received 7 July 1994)

The resistive critical fields of several types of superconductor such as $Tl_2Ba_2CuO_{6+x}$, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, and $Ba_{1-x}K_xBiO_3$ show an unusual increase at low temperatures. We argue that this arises from thermodynamic fluctuations which are strongly enhanced because of the reduction in condensation energy density $U(H,T)$ by a magnetic field. This effect should be significant for any material for which $U\Omega \lesssim k_B T_c$ at low temperatures and zero field, where Ω is the coherence volume, which probably includes most cuprate superconductors.

The resistive transitions of $Tl_2Ba_2CuO_{6+x}$ (Tl 2201) (Ref. 1) and $Bi_2Sr_2CuO_y$ (Bi 2201) (Ref. 2) are reasonably sharp when T_c is low (unlike cuprates with higher T_c where there is substantial field broadening, e.g., Refs. 3 and 4). This allows a rather unambiguous estimate of the resistive critical field $H^*(T)$ at all temperatures (T). In contrast to the behavior of $H_{c2}(T)$ for conventional superconductors, which show little T dependence below $T_c/2$, $H^*(T)$ for these materials[1,2] curves upwards and continues to increase strongly below $T_c/10$. Similar behavior of $H^*(T)$ is also observed in the two-dimensional organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ (Ref. 5) and for single crystals of the three-dimensional, cubic compound $Ba_{1-x}K_xBiO_3$.[6] Here we propose an interpretation involving thermal fluctuations and make numerical estimates for Tl 2201 using high-resolution specific-heat data.[7] We briefly discuss other interpretations involving flux lattice melting[8–10] or thermally activated flux motion.

In zero applied field (H), fluctuation effects start to become important when the following condition is satisfied:[11,12]

$$|F_s - F_n|\Omega = U(T)\Omega(T) \approx \alpha k_B T. \qquad (1)$$

In this expression F_s and F_n are the free energies per unit volume of the superconducting and normal states at a temperature T, U is the condensation energy density, k_B is Boltzmann's constant, Ω is the coherence volume, and α is a model-dependent numerical factor, of order unity, to be discussed later. For isotropic compounds $\Omega = \xi^3$ where ξ is the T-dependent coherence length. For an anisotropic material[13] with layer spacing d, $\Omega = \xi_\parallel^2 d$ in the two-dimensional (2D) region $[d > \xi_\perp(T)]$ and $\Omega = \xi_\parallel^2 \xi_\perp$ in the 3D region $[d < \xi_\perp(T)]$, where $\xi_\parallel$ and $\xi_\perp$ are the in- and out-of-plane coherence lengths. Here we consider mainly the 2D limit which should be appropriate for Tl 2201 crystals which have $d = 12$ Å and effective-mass and critical-field anisotropies of 900 and 30, respectively,[3] although similar conclusions apply to the 3D case.

If Eq. (1) is satisfied then superconducting fluctuations will occur in the normal state above T_c. These can be viewed as superconducting droplets of volume Ω which spontaneously appear and disappear on time scales of $\hbar/k_B|T - T_c|$.[11] Conversely, below T_c, transient normal regions of volume Ω will exist on a similar time scale.

For $H = 0$, the width of the true critical region, in which the fluctuations are strong and universal power-law divergences are expected, is usually taken to be $T_c \pm \tau_G T_c$, where the Ginzburg parameter τ_G is defined in terms of the temperature at which the lowest-order (Gaussian) fluctuation contribution to the specific heat equals the mean-field step.[14] Using weak-coupling BCS parameters, for the 2D case this gives $\tau_G^{2D} = 0.013 k_B T_c / U(0)\Omega(0)$. At $T = T_c \pm \tau_G^{2D} T_c$, $U(T)\Omega(T) \approx 0.03 k_B T$, i.e., $\alpha \approx 0.03$. However, fluctuations will still be important at 10–$100\tau_G^{2D}$, i.e., $\alpha \approx 0.3$–3.

As described below we propose that the increase in $H^*(T)$ at low T arises from the enhancement of thermal fluctuations by a magnetic field, even though the temperatures are very low and the values of τ_G in zero field are not excessively large. The primary effect of the field is to reduce U, greatly extending the range of validity of Eq. (1) if $U(0)\Omega(0)/k_B T_c \lesssim 1$ in zero field, rather than reducing the effective dimensionality.[15] $H^*(T)$ of conventional superconductors is not unusual because they have much larger values of $U(0)\Omega(0)/k_B T_c$.

We assume that at $T = 0$, when thermal fluctuations are absent, the materials in question would display a standard magnetization-field curve described by Ginzburg-Landau (GL) mean-field theory. As discussed in many textbooks, if the applied field H is reasonably close to the upper critical field H_{c2} ($H > 0.5H_{c2}$) the magnetization M (in cgs units) is given by $(H - H_{c2})/4\pi\beta(2\kappa^2 - 1)$ where κ is the GL parameter, and $\beta = 1.16$. Integrating $M(H)$ from H_{c2} to H gives

$$F_s - F_n = -\frac{(H - H_{c2})^2}{8\pi\beta(2\kappa^2 - 1)} \approx -\frac{H_c^2}{8\pi}\left[1 - \frac{H}{H_{c2}}\right]^2 \qquad (2)$$

for materials with large values of κ, using $H_{c2} = \kappa\sqrt{2}H_c$, where H_c is the thermodynamic critical field. Equation (2) gives the correct value for $U \equiv |F_s - F_n| = H_c^2/8\pi$, at $H = 0$ (to within a factor β) and in the present context is an adequate approximation well below the mean-field $H_{c2}(T)$ line [actually, using the London formula $M \propto \ln(H/H_{c2})$ gives almost identical results]. Equation (2) can also be viewed as the product of a local free energy density

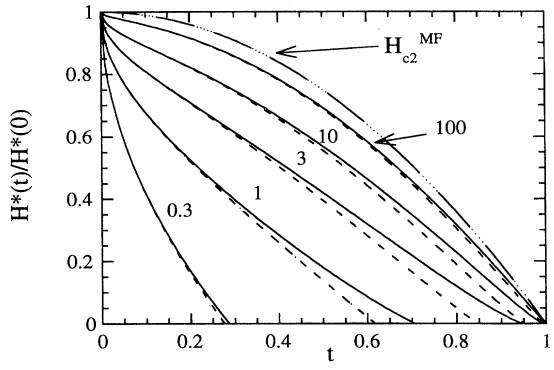

FIG. 1. Reduced critical fields versus reduced temperature $t \equiv T/T_c$ calculated from Eqs. (4) and (5) in the 2D (dashed lines) and 3D (solid lines) limits for the zero-temperature values of T_0/T_c shown.

$H_c^2(T)(1-H/H_{c2})/8\pi$ [A. M. Campbell (private communication)] and $(1-H/H_{c2})$, which represents the fraction of superconductor outside the vortex cores.

As H is increased, $U(H,T)$ falls, so that at a certain field, $H^*(T)$, Eq. (1) will be satisfied and fluctuating normal regions of volume Ω will be formed. For $H^* < H_{c2}$ the characteristic magnetic area ϕ_0/H (where ϕ_0 is the flux quantum for pairs) is always larger than $2\pi\xi_\parallel(T)^2$ and we therefore assume that Ω is the same as in zero field, i.e., $\xi_\parallel(T)^2 d$ for the 2D case and $\xi(T)^3$ for the cubic compound. (The case where the in-plane area is ϕ_0/H is discussed later.) By combining Eqs. (1) and (2), and taking $\Omega = \xi_\parallel(T)^2 d$ the field H^* at which fluctuations become important is given by

$$\left[1-\frac{H^*}{H_{c2}}\right]^2 \frac{H_c^2}{8\pi}\,\xi_\parallel(T)^2 d = \alpha k_B T \tag{3}$$

giving

$$H^*(T) = H_{c2}(T)[1-\sqrt{T/T_0}], \tag{4}$$

where

$$k_B T_0(T) = \frac{H_c(T)^2}{8\pi\alpha}\,\xi_\parallel(T)^2 d, \tag{5}$$

i.e., $k_B T_0$ is proportional to $U(T)\Omega(T)$, the condensation energy per coherence volume in zero field. This is the crucial equation; if $T_0(0) \gg T_c$ as in conventional superconductors then H^* will behave regularly, like $H_{c2}(T)$, but if $T_0 \lesssim T_c$, it will show unusual T dependence because of fluctuations. In the 2D case, with H perpendicular to the layers, $T_0(T)$ varies as $H_{c2}(T)$ and in 3D as $\sqrt{H_{c2}(T)}$. For Tl 2201, zero-field heat-capacity data[7] confirm that $H_c(T)/H_c(0)$ approximately follows the usual $(1-t^2)$ law, where $t = T/T_c$, and so neglecting the small T dependence of κ, $H^*(T)$ can be found from Eqs. (4) and (5). Calculations for the 3D and 2D cases with various values of T_0/T_c are shown in Fig. 1.

Fitting the $H^*(T)$ data for Tl 2201 (Ref. 1) to Eq. (4) [Fig. 2(a)] gives $H_{c2}(0) = 16.8$ T and $T_0(0) = 2.6$ K for a crystal with $T_c = 15.5$ K. For $T_c = 21.5$ K (Fig. 3 of Ref. 16) a similar fit (not shown) gives $H_{c2}(0) = 25$ T and $T_0(0) = 7$ K. The Bi 2201 film data[2] also fit Eq. (4) with $H_{c2}(0) = 31$ T, $T_c = 19$ K, and $T_0 = 6.2$ K (not shown). $H_c(0)$ was determined by integrating the specific-heat data[7] for Tl 2201 and $\xi_\parallel(0)^2$ is given by $\phi_0/2\pi H_{c2}(0)$. The values used were $H_c(0) = 0.059$ and 0.096 T, $\xi_\parallel(0) = 44$ and 36 Å for T_c equal to 15.5 and 21.5 K, respectively, giving $U\Omega/k_B T_c = 0.15$ and 0.19 at $T = 0$. Thus for Tl 2201, the $T_0(0)$ values determined by fitting the $H^*(T)$ data to Eq. (4) are consistent with Eq. (5) if the unknown parameter $\alpha = 0.6$–0.9.

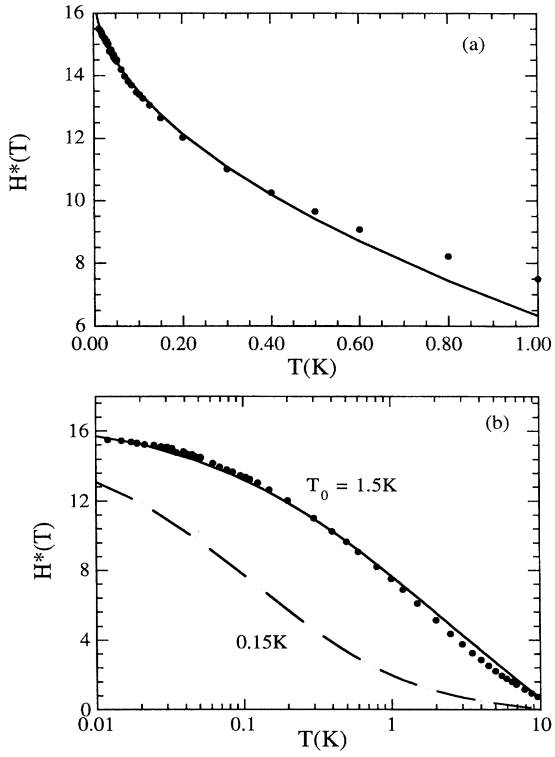

FIG. 2. (a) Fit of the experimental (Ref. 1) resistive critical-field data $H^*(T)$ for a single crystal of $Tl_2Ba_2CuO_{6+\delta}$ with $T_c = 15.5$ K and H perpendicular to the CuO_2 planes (symbols) to Eq. (4) (solid line), with $H_{c2}(0) = 16.8$ T and $T_0(0) = 2.6$ K. (b) Fit to Eq. (6) shown on a semilogarithmic scale, with $H_{c2}(0) = 17.1$ T and $T_0(0) = 1.5$ K. The long dashed line is an estimate of the melting line using the appropriate value of $T_0(0)$ (0.15 K) and the theory of Refs. 8–10.

We note that a 2D analysis for $YBa_2Cu_3O_{6.95}$, using $H_c(0) = 1.1$ T (Ref. 17) and $H_{c2}(0) = 130$ T,[18] leads to $U\Omega \approx k_B T_c$, i.e., to $T_0(0) \approx T_c$. For overdoped Tl 2201 there is a substantial linear term in the specific heat[7] below T_c, caused by an unknown "pair-breaking" effect, and this is what makes $U\Omega/T_c$ particularly small. Nevertheless the above values of $U\Omega/T_c$ give τ_G^{2D} values of 0.09 and 0.07 for this material which are comparable with estimates for some other high-T_c oxides.[19] Thus although thermal fluctuations strongly affect $H^*(T)$, in zero field the true critical region is only reached within 1–2 K of T_c. In fact the parameter $T_0(0)$ is directly related to τ_G. Using the definition of τ_G^{2D} given above[14] and with $\alpha = 1$ we have $\tau_G^{2D} = T_c/[76T_0(0)]$. From Fig. 1 there should be significant upward curvature in

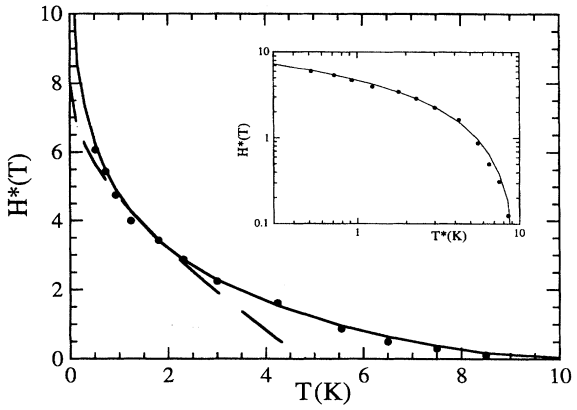

FIG. 3. Comparison of perpendicular resistive critical-field data for the 2D organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ (Ref. 5) (symbols) with Eq. (6) (solid line) for the parameter values $H_{c2}(0)=12.6$ T, $T_0(0)=1$ K and $T_c=10.7$ K. Main figure: linear scale, inset: log-log scale. The dashed line in the main figure shows a fit to Eqs. (4) and (5) with $H_{c2}(0)=8$ T, $T_0(0)=6$ K.

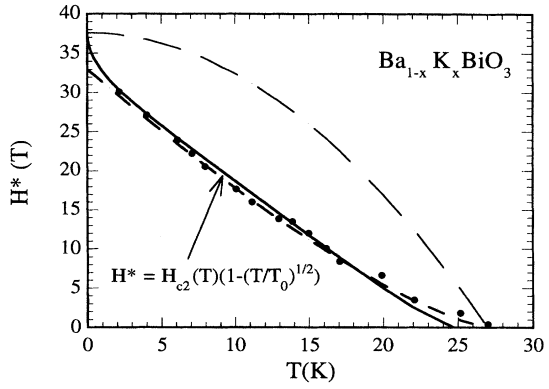

FIG. 4. Resistive critical fields of a crystal of $Ba_{1-x}K_xBiO_3$ (symbols) (Ref. 6). The solid line shows a fit to Eq. (4) with $H_{c2}(0)=37.6$ T, $T_0(0)=60$ K, and $T_0(T)/T_0(0)=\sqrt{H_{c2}(T)/H_{c2}(0)}$. The short dashed line is the $H^*=A(1-t)^{4/3}$ power law expected (Ref. 14) in the true critical region. The mean-field line used for H_{c2} is shown by the long dashed line.

$H^*(T)$ for any compound with $T_0/T_c \lesssim 1$, i.e., $\tau_G^{2D}>0.013$, and there may be an observable effect at low T even for $T_0/T_c=10$.

Tešanović and colleagues[8–10] obtain a condition for the melting of a 2D vortex lattice into a dense vortex plasma which has a similar form to Eq. (3), by finding a new solution to the GL equations in which the positions of the vortices are allowed to vary. But their condition contains the magnetic area ϕ_0/H. If we use ϕ_0/H instead of $\xi_\parallel(T)^2$ in Eq. (3) then for the 2D case,

$$H^*=H_{c2}(T)\left[1+\frac{T}{2T_0}-\sqrt{\frac{T}{T_0}\left(1+\frac{T}{4T_0}\right)}\right]. \qquad (6)$$

It is the same as Eq. (4) as $T\to 0$; Eq. (5) for T_0 still holds (with an extra factor of 2π in the numerator) and it fits the data for the Tl 2201 crystal over a wider temperature range than Eq. (4) with $T_0(0)=1.5$ K and $\alpha=6$ [Fig. 2(b)]. However, near T_c, Eq. (6) gives $H^*\propto(1-t)^n$ with $n\approx 2$, while the experimental value is certainly in the range $n=1-1.5$,[3] and appears to be close[20] to the value $n=\frac{4}{3}$ expected for critical fluctuations.[14] Therefore the deviations from Eq. (4) which are invariably observed for $H^*<H_{c2}(T)/2$ probably represent a gradual crossover to field-induced critical behavior with $n=\frac{4}{3}$, rather than clear evidence in favor of Eq. (6). The lattice melting condition[8–10] corresponds to much larger values of α (the parameter[8] $g_m^2\equiv\alpha\approx 60$) because the melting transition occurs when the free energy difference between triangular and square lattices (multiplied by $\phi_0 d/H$) is $\approx k_BT$, and this difference is $U(H,T)/50$. The expected melting line for the Tl 2201 crystal with $T_c=15.5$ K is shown by a long dashed line in Fig. 2(b).

Another possibility which has been discussed by several groups, e.g., Ref. 21, is that in the high-T_c oxides, the same condition, Eq. (6) [but with d replaced by $\xi(T)$], sets a field scale for which the vortex pinning energy is $\approx k_BT$.

In the latter two cases the resistivity just above H^* is a vortex plasma or a flux flow resistivity, respectively, while in the present picture it is the normal-state conductivity plus the field-dependent Aslamazov-Larkin fluctuation term. In 2D and at zero field this term only depends on T/T_c, the interplanar spacing d, and fundamental constants[14] and is rapidly dominated by the large normal-state conductivity of Tl 2201 crystals.[1] So the fact that the resistivity data[1,16] do seem to reach the normal-state value just above H^* is consistent with the present approach.

The bipolaron model[22] also gives an unusual increase in $H_{c2}(T)$ at low T. However, there is mounting evidence that a band picture may apply to the superconducting cuprates. We note that as shown in Fig. 3, a similar upturn in $H^*(T)$ is observed for the 2D organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ (Ref. 5) and can also be fitted to Eq. (4) or (6). For this compound there is definite experimental evidence for de Haas–Shubnikov oscillations, i.e., a Fermi surface, in the normal phase.[23]

Finally as shown in Fig. 4 the cubic compound $Ba_{1-x}K_xBiO_3$ also has an upturn in $H^*(T)$ (Ref. 6) which can be fitted to Eq. (4) and the 3D form of Eq. (5) with the parameters $H_{c2}(0)=37.6$ T, $T_c=27$ K, and $T_0(0)=60$ K. As can be seen from Fig. 4, in this case measurements to even lower temperatures are needed to distinguish between the present picture and the $\frac{4}{3}$ power law expected for true critical behavior.[14] However, the relatively large value obtained for $T_0(0)/T_c$ tends to favor our approach at low T.

In conclusion we have outlined a simple fluctuation picture which describes the increase in the resistive critical fields that is often observed in unusual superconducting materials at very low temperatures. It predicts that such behavior will be significant whenever the zero-field value of $U\Omega$ is less than k_BT_c and may be observable for values as high as $10k_BT_c$ (Fig. 1). In this case the reduction of U by a magnetic field extends the region of large fluctuations to temperatures well below T_c. In order to confirm this picture a theoretical model is needed to calculate the unknown parameter α and see whether it is indeed consistent with the value

$\alpha \approx 1$ determined experimentally. For some of the compounds mentioned, measurements of $H^*(T)$ to much lower temperatures and more specific-heat data would also be helpful.

We are grateful to A. S. Alexandrov, A. M. Campbell, A. Carrington, J. Clarke, N. E. Hussey, A. P. Mackenzie, P. Monceau, E. J. Tarte, and J. M. Wheatley for helpful discussions.

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*On leave from The Institute of Physics of the University, Zagreb, Croatia.

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