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Temperature dependence of lower critical fields in Y-Ba-Cu-O crystals

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The temperature dependence of the anisotropic lower critical fields H_{c1} in single-crystal Y-Ba-Cu-O is determined from the onset of flux penetration in zero-field-cooled dc magnetization versus temperature from T_c down to $0.5T_c$. An extended Bean-critical-state model is developed to define this onset consistently. The H_{c1}'s are linear in temperature near T_c as in Ginzburg-Landau theory and consistent with a clean local-limit BCS form with extrapolated $H_{c1}(T=0)=180$ Oe for $\mathbf{H}\perp\hat{c}$ and 530 Oe for $\mathbf{H}\parallel\hat{c}$.

Accurate measurement of superconducting critical fields is of vital importance to determine key length scales such as the coherence length and penetration depth. Their temperature dependence indicates the mean-field or non-mean-field nature of the transition, and also the topology of the superconducting state, that is, whether a conventional BCS-like pairing mechanism can be invoked.

Recent results have made it clearer that early reports of such critical fields in the new high-temperature superconductors have been unreliable.[1,2] What was initially thought to be the upper critical field H_{c2} now appears rather to be an "irreversibility" line related to flux depinning or, possibly, vortex-lattice melting.[1-3] The reported low-temperature lower critical fields H_{c1}, in turn, vary by at least an order of magnitude,[4-7] probably because most estimates have come from asymptotically small deviations from perfect diamagnetism in magnetization-versus-field curves, and the likelihood that these small deviations are due to sharp corners of the crystals makes such estimates highly suspect. Furthermore, information on the temperature dependence of H_{c1} has come principally from related measurements of penetration depth by muon relaxation,[8] low-field magnetization,[9] or microwave resonance[10] on ceramic or polycrystalline material where averaging of anisotropic properties and uncertainties about local demagnetizing factors make the results uncertain.

In this paper we report a novel way around these difficulties, so as to obtain what we believe are the first convincing temperature-dependent values of H_{c1} in Y-Ba-Cu-O. Our approach hinges on (1) the study of a significant number of the highest-quality crystals with measurements along both principal axes, (2) variation of the surface quality of these crystals by different annealing procedures to test and eliminate the possible role of surface effects, (3) variation of the geometry to test and eliminate the effect of demagnetization, and (4) the development of the novel technique of measurement and analysis, using temperature-dependent rather than field-dependent magnetization data to determine $H_{c1}(T)$ in the presence of bulk pinning.

The results show a conventional mean-field Ginzburg-Landau behavior of H_{c1}, surprising, perhaps, in view of some claims of large critical regions.[11] The results also show a conventional BCS temperature dependence, but of the clean-local-limit rather than of the extreme-anomalous or two-fluid type.[12] While contradicting the early muon data in this regard,[8] these results make sense because the clean local limit is appropriate when the coherence length is shorter than the electronic mean free path, and in these materials the coherence lengths are surely extremely short.[1,13] The fact that conventional BCS behavior is observed also sets a strong limit on the many exotic theories proposed for high-temperature superconductors.

dc magnetization-versus-temperature measurements have been performed on six crystals, grown by a technique described previously.[14] The sizes of the crystals varied significantly, with the thicknesses along the c axis ranging from 20 μm to 200 μm and c-axis demagnetization factors ranging from 0.5 to 0.9. Both oxygen annealed and as-grown crystals were used, the annealing causing some degradation of surface quality. Nevertheless, all crystals had T_c's above 90 K, so their bulk properties seemed excellent.

The measurements were performed in a noncommercial SQUID magnetometer[15] with the samples first cooled to low temperature (4.2 K) in the background field of 0.5 mOe ("zero field"). Zero-field-cooled (ZFC) curves, such as shown in Fig. 1, are obtained on slow warm-up after the field is turned on. Flux expulsion on field cooling through the superconducting transition is much smaller but strongly field dependent, as discussed elsewhere.[15] Lastly, after this field cooling, the field is turned off at low temperature and the resulting remanent moment is measured on slow warming through the transition. The evolution of remanent moment for the crystal of Fig. 1 and several applied fields is shown in Fig. 2.

In all the crystals of different shapes and sizes we consistently observe a very sharp (less than 0.5 K) superconducting transition in the ZFC data at temperatures around 91 K at sufficiently low fields (below 1 Oe). At higher fields, however, the data develop a characteristic shape consisting of an almost linear decrease in diamagnetic signal setting in at a relatively well-defined temperature which we call T_{c1}. At a somewhat higher temperature denoted T^*, the rate of the linear decrease is reduced and this reduced rate of decrease is maintained until diamagnetism disappears at T_c. T_{c1} and T^* both shift systematically to lower temperatures with increasing field, as shown in Fig. 1 for H applied in the a-b plane. The results

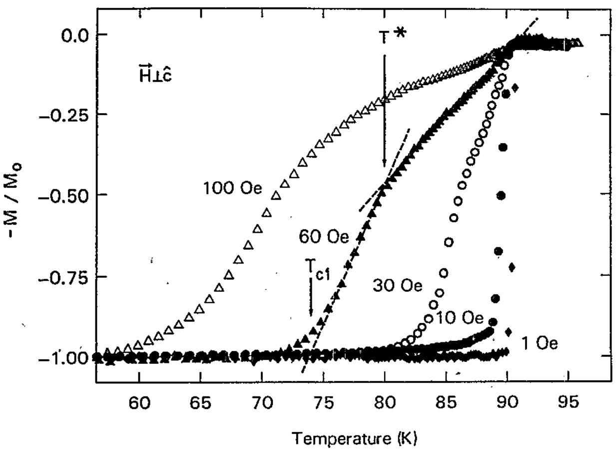

FIG. 1. Zero-field-cooled (ZFC) magnetization (normalized to its value at 4.2 K) of an Y-Ba-Cu-O crystal as a function of temperature, for several fields applied parallel to the *a-b* plane. The temperature T_{c1}, at which the applied field equals $H_{c1}/(1-N)$, is identified from the linear extrapolation of the initial flux penetration as shown by the dashed lines.

are similar for H along the c axis, but the temperature scale is compressed by about a factor of 2.

The similarity in demagnetization-corrected values over *all* our crystals, independent of surface quality, and their systematic behavior with field, suggest an obvious interpretation for T_{c1}, namely that it is the point of bulk flux penetration controlled by the bulk H_{c1}. The changing temperature reduces H_{c1} until it equals the applied field. Thus a measurement of T_{c1} versus the applied field provides a new way of determining the temperature dependence of H_{c1}. The simplest way to define T_{c1} is by extrapolating the linear slope from the principal region of flux penetration, as shown by the dashed lines in Fig. 1. This procedure seems to work fairly well down to a T_{c1} which is 40% below T_c. To go further we need a more quantitative method for extrapolating the data.

Our interpretation is reinforced by the knee at T^*, and the reduced slope above this temperature, both of which have a simple interpretation in the flux penetration picture; between T_{c1} and T^* flux fronts advance from the edge of the crystal, as the critical current J_c and the slope dB/dx decrease. At T^* the flux fronts meet at the center, and at higher temperatures, the flux distribution flattens progressively, giving a more gradual approach of the magnetization to zero.

These effects can be modeled with a conventional Bean-critical-state model,[16–19] suitably extended to include the effect of H_{c1}. Space forbids a detailed treatment here and we simply quote the results, which are derived quite straightforwardly.[20]

We derive the initial slope of flux penetration m with temperature as defined by a magnetization $4\pi M$ given by $-[H_0/(1-N)]+m\Delta t$, where H_0 is the applied field, N is the demagnetizing factor, t is the reduced temperature T/T_c, and Δt is the increase in reduced temperature above T_{c1}. Similarly, we define the slope h of H_{c1} at T_{c1} in terms of $H_{c1}(T)=H_{c1}(T_{c1})-h\Delta t$, where $H_{c1}(T_{c1})=H_0/(1-N)$. We also define J_{c1} as the value of J_c at T_{c1}. Finally, we define f_b such that f_bH_{c1} is the value of $B(x)$ below which $B(x)$ drops sharply to zero. This is a simple approximation to the "dB/dH effect," which arises physically from the exponential dropoff of intervortex forces once the vortices are separated by distances larger than the penetration depth.[17,18]

With these definitions, a slab geometry of thickness D, and field in the slab plane, the linear slope of flux penetration to lowest order in Δt is

$$m=-\frac{hcf_bH_0}{2\pi(1-N)^2DJ_{c1}+Ncf_bH_0}. \tag{1}$$

This formula suggests a simple graphical method for evaluating the accuracy of the earlier method for determining T_{c1} and for correcting it in cases where the rounding of the data becomes significant. The idea is to lean a line of slope m tangent to the experimental data and to take T_{c1} as the point where this line extrapolates to the full flux exclusion $H_0/(1-N)$.

To carry out this procedure, we of course need to determine all the parameters in Eq. (1). We obtain N from an ellipsoidal approximation to the sample shape. In the case of field parallel to the c axis, we can substitute D by r, the root mean square of the two transverse dimensions. We can also obtain f_b and J_{c1} from simple expressions derived using the same model for the ZFC and remanent magnetizations near T_c:

$$4\pi M_{\mathrm{ZFC}}=-(1-f_b)H_{c1}-(\pi DJ_{c1}/c), \tag{2a}$$

$$4\pi M_{\mathrm{rem}}=f_bH_{c1}+(\pi DJ_{c1}/c). \tag{2b}$$

These expressions explain the linear decrease of these magnetizations to zero near T_c, consistent with the expected linear dependence of H_{c1} on $1-t$ in the Ginzburg-Landau theory. The critical current terms are negligibly

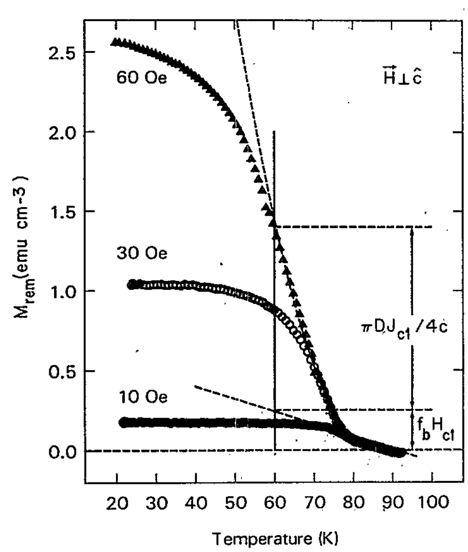

FIG. 2. Remanent magnetization of the same crystal as in Fig. 1, as a function of temperature for several fields applied parallel to the *a-b* plane. The extrapolated line separates contributions to Eq. (3b) from H_{c1} and from J_c.

small here because of their typically higher-power dependence on $1-t$ and because of the very large flux creep[21] which reduces J_c near T_c yet further.

The values for $J_c(T)$ can now be extracted from the remanent moment data of Fig. 2 by extrapolating the high-temperature linear region and using the nonlinear deviations to identify the second term in Eq. (2b). The slopes m are then constructed in an iterative manner. From the low-field data, an initial slope of $H_{c1}(T)$ is established, giving the parameter h in Eq. (1). Then the various parameters can be substituted for the next trial temperature, and the slope used to determine an improved value for T_{c1} at the given applied field.

The results of this procedure are summarized in Fig. 3, where we show temperature dependences of H_{c1} for $\mathbf{H}\perp\hat{c}$ and $\mathbf{H}\parallel\hat{c}$. Shifts due to the slope construction described above are shown by left-pointing arrows. The data for all samples superimpose, which indicates the absence of strong surface barrier effects and the proper correction for demagnetization. There is of course always the likelihood of flux penetration at sample corners, giving a small amount of additional rounding seen in the data. Here the linear dependence of the bulk flux penetration, predicted by Eq. (1), is of vital importance: It allows us to extrapolate to T_{c1} from the *bulk* flux penetration, and this gives further confidence that the results are not compromised by corner demagnetization or surface barriers. However, in higher fields the rounding becomes considerable and the tangent construction involves such a small slope for $T<T_c/2$ that we are not able to get meaningful results in this regime.

In spite of these limitations in our analysis, we can draw some interesting conclusions. First of all, the lower critical fields in Y-Ba-Cu-O are indeed linear in temperature near T_c, as expected in a Ginzburg-Landau second-order phase transition. This is not at all obvious from measurements of H_{c2} where all sorts of contradictory curvatures have been reported.[1] Furthermore, the linearity basically persists down to $0.5T_c$, which is inconsistent with the conventional two-fluid[12] form for λ if one takes $H_{c1}\propto\lambda^{-2}\propto 1-t^4$. It is, however, plausible to expect a clean-limit behavior for these materials, since the coherence length is so short.[11,13] In the weak-coupling clean local limit, with $H_{c1}\sim\lambda^{-2}$, conventional theory[12] predicts the form shown as a dashed line in Fig. 3, in reasonable agreement with the data. The low-temperature extrapolation gives $H_{c1}(T=0)=180\pm20$ Oe for $\mathbf{H}\perp\hat{c}$ and 530 ± 50 Oe for $\mathbf{H}\parallel\hat{c}$. The $H_{c1}^{\perp}(T=0)$ value is in good agreement with the value deduced recently from relaxation measurements,[7] but $H_{c1}^{\parallel}(T=0)$ is somewhat lower, which could imply a deviation from BCS behavior along this axis.

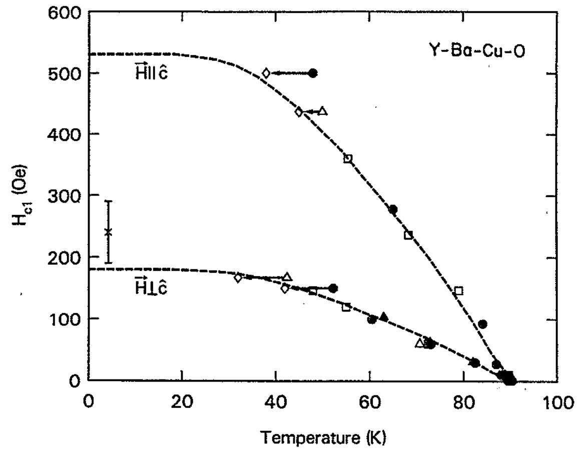

FIG. 3. H_{c1} for H applied parallel to the c axis and perpendicular to the c axis for several Y-Ba-Cu-O crystals. The T_{c1} corrections described in the text are indicated by the horizontal arrows. Low-temperature value for $\mathbf{H}_{c1}\perp\hat{c}=250\pm50$ Oe from Yeshurun *et al.* (Ref. 7) is shown as a cross (their value for $\mathbf{H}_{c1}\parallel\hat{c}$ is 900 ± 100 Oe). The dashed line represents to the clean-local-limit weak-coupling BCS prediction.

To estimate the low-temperature penetration depth, we use the approximate London formula:[22,23]

$$H_{c1}=\frac{\phi_0}{4\pi\lambda_i\lambda_j}\left(\ln\frac{\lambda}{\sqrt{\xi_i\xi_j}}+0.50\right), \tag{3}$$

where $\lambda_i=\lambda\sqrt{m_i}$, $\xi_i=\xi/\sqrt{m_i}$, the effective masses m_i obey $m_am_bm_c=1$, and ϕ_0 is the flux quantum. Using $\xi_{ab}=13$ Å and $\xi_c=2$ Å (the results are not sensitive to the precise values),[13] we find $\lambda_{ab}(T=0)=1300$ Å and $\lambda_c(T=0)=4500$ Å. Earlier estimates of λ_{ab} varied from 260 to 900 Å and of λ_c from 1200 to 8000 Å.[5,8,9] The average $T=0$ penetration depth $\lambda=(\lambda_{ab}^2\lambda_c)^{1/3}$ extrapolated from our measurements is 2000 Å, approximately the value reported for powdered samples.[8]

In summary, by using a novel procedure which minimizes errors from pinning, nonuniform degmagnetization and surface effects, we determine the temperature dependence of the anisotropic H_{c1} and penetration depth. The behavior follows conventional mean-field Ginzburg-Landau and clean-local-limit weak-coupling BCS forms.

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[1]A. P. Malozemoff, T. K. Worthington, Y. Yeshurun, F. Holtzberg, and P. Kes, Phys. Rev. B 38, 7203 (1988).
[2]M. Tinkham Phys. Rev. Lett. 61, 1658 (1988).
[3]P. L. Gammel, L. F. Schneemeyer, J. V. Waszczak, and D. J. Bishop, Phys. Rev. Lett. 61, 1666 (1988).
[4]T. R. Dinger, T. K. Worthington, W. J. Gallagher, and R. L. Sandstrom, Phys. Rev. Lett. 58, 2687 (1987); T. K. Worthington, W. J. Gallagher, and T. R. Dinger, *ibid.* 59, 1160 (1987).
[5]A. Umezawa, G. W. Crabtree, J. Z. Liu, T. J. Moran, S. K.

Malik, L. H. Nunez, W. L. Kwok, and C. H. Sowers, Phys. Rev. B **38**, 2843 (1988).

[6]I. Fruchter, C. Giovanella, G. Collin, and I. A. Campbell, Physica C **156**, 69 (1988).

[7]Y. Yeshurun, A. P. Malozemoff, F. Holtzberg, and T. R. Dinger, Phys. Rev. B **38**, 11828 (1988).

[8]D. R. Harshman, G. Aeppli, E. J. Ansaldo, B. Batlogg, J. H. Brewer, J. F. Carolan, R. J. Cava, M. Celio, A. C. Chaklader, W. N. Hardy, S. R. Kreitzman, G. M. Luke, D. R. Noakes, and M. Senba, Phys. Rev. B **36**, 2386 (1987).

[9]J. R. Cooper, C. T. Chu, L. W. Zhou, B. Dunn, and G. Grüner, Phys. Rev. B **37**, 638 (1988).

[10]J. P. Carini, A. M. Awasthi, W. Beyermann, G. Grüner, T. Hylton, K. Char, M. R. Beasley, and A. Kapitulnik, Phys. Rev. B **37**, 9726 (1988). The work on oriented films [also L. Drabeck, J. P. Carini, G. Grüner, T. Hylton, K. Char, and M. R. Beasley (unpublished)] appears to agree with our *c*-axis results in the high-T limit.

[11]A. Kapitulnik, M. R. Beasley, C. Castelani, and C. DiCastro (unpublished).

[12]M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).

[13]B. Oh, K. Char, A. D. Kent, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond, and A. Kapitulnik, Phys. Rev. B **37**, 7861 (1988).

[14]D. L. Kaiser, F. Holtzberg, M. F. Chisholm, and T. K. Worthington, J. Cryst. Growth **85**, 593 (1987).

[15]L. Krusin-Elbaum, A. P. Malozemoff, Y. Yeshurun, D. C. Cronemeyer, and F. Holtzberg, Physica C **153–155**, 1469 (1988).

[16]C. P. Bean, Phys. Rev. Lett. **8**, 250 (1962); Rev. Mod. Phys. **36**, 31 (1964).

[17]A. M. Campbell and J. E. Evetts, Adv. Phys. **21**, 199 (1972); Rev. Mod. Phys. **36**, 31 (1964).

[18]H. Ullmaier, *Irreversible Properties of Type II Superconductors* (Springer-Verlag, Berlin, 1975).

[19]J. R. Clem, J. Appl. Phys. **50**, 3518 (1979).

[20]L. Krusin-Elbaum, A. P. Malozemoff, and J. R. Clem (unpublished).

[21]Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).

[22]V. G. Kogan, Phys. Rev. B **24**, 1572 (1981).

[23]R. A. Klemm and J. R. Clem, Phys. Rev. B **21**, 1868 (1980).