Anisotropy of the lower critical field, magnetic penetration depth, and equilibrium shielding current in single-crystal $YBa_2Cu_3O_7 - \delta$

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Magnetization measurements at 11 K are used to obtain anisotropic H_{c1} values for fields along the *c* direction and in the basal plane of single crystals of YBa₂Cu₃O_{7-s}. The anisotropic magnetic penetration depth and equilibrium shielding currents at H_{c1} are derived and analyzed within effective-mass theory.

The two-dimensional layered structure¹⁻³ of YBa₂- $Cu_3O_{7-\delta}$ leads to a strong anisotropy in its electronic and superconducting properties. Anisotropy in the Fermi surface, 4,5 critical current, $^{6-10}$ upper critical field, $^{11-13}$ and resistivity^{14,15} are well established. The magnitude and anisotropy of the lower critical field and the magnetic penetration depth have received much less attention. In this paper we report careful measurements of H_{c1} in single-crystal samples of $YBa_2Cu_3O_{7-\delta}$ for fields along the c direction and in the a, b plane. We analyze these data using London electrodynamics in the large- κ approximation to derive the magnitude and anisotropy of the penetration depth. We find H_{c1} to be considerably smaller and the penetration depth and κ to be considerably larger than previously thought. Finally, we derive the anisotropic shielding supercurrent density flowing in equilibrium at H_{c1} .

Single crystals of YBa₂Cu₃O_{7- δ} were prepared using an annealing method described elsewhere.¹⁶ Substantial twinning of the crystals involving the interchange of the a and b axes between twins prevented the observation of anisotropy in the a, b plane. Measurements were taken on several crystals to verify that the observed behavior was an intrinsic property independent of sample quality. Three types of crystals were selected: one as-grown, the second as-grown and annealed at 550 °C for 72 h in pure oxygen, and the third as-grown and irradiated with fast neutrons (E > 0.1 eV) to a fluence of 8.16×10^{17} n/cm². Some of the characteristics of these three crystals were reported in previous studies. 8,16,17 Magnetization measurements were taken using a SHE superconducting quantum interference device magnetometer at 11 K in fields up to 3 kG on a relatively fine grid of fields with increments of 10 G for fields along the a, b plane and 50 G for fields along the c direction.

Typical magnetization data for the field along the c direction in the as-grown crystal are shown in Fig. 1. The sharp cusp in the magnetization characteristic of H_{c1} in equilibrium is not observed due to strong pinning which effectively prevents flux from entering the sample. Such behavior is common in A15 superconductors which display strong flux pinning.¹⁸ In these cases, the entry of flux into the sample at H_{c1} can be recognized as a slight deviation from perfect diamagnetism. In order to clearly observe this deviation in our data, each point was sub-

tracted from the initial linear dependence of the magnetization on field as determined by a least-squares fit to the low-field data. The deviation plot created from the data in Fig. 1 is shown in Fig. 2. There it can be seen that the first deviation from linear behavior occurs at 425 ± 50 G, a much lower field than would be evident in Fig. 1. Indeed, at an applied field of 1500 G, the deviation from linearity is only 10 G. A comparison of the behavior for two of the crystals is shown in Fig. 3 for fields in the *a*,*b* plane. Although the flux enters the annealed crystal faster than the as-grown crystal for fields above approximately 80 G, both samples exhibit the first departure from linearity at nearly the same field.

The values of H_{c1} were obtained from the field at which the flux first enters the sample by correcting for demagnetizing effects obtained from the initial slope of the magnetization curve assuming $M/H = -1/4\pi(1-n)$. The data for the three crystals are summarized in Table I, giving weighted average values of H_{c1} of 120 ± 10 G for fields in the *a*,*b* plane and 690 ± 50 G for fields in the *c* direction.

We analyze our data using the equations $H_{c1} = (\Phi_0/4\pi\lambda^2)\ln(\lambda/\xi)$ and $H_{c2} = \Phi_0/2\pi\xi^2$. We take the latter equation to be a definition of ξ without assuming any particular temperature dependence for either H_{c2} or ξ . The first equation can be derived from the energy associated with a single vortex assuming London electrodynamics and excluding the core region $r < \xi$. This ap-



FIG. 1. Magnetization curve for the as-grown single crystal. The straight line is a least-squares fit to the first six data points.



FIG. 2. The deviation of magnetization from the initial linear behavior for the data shown in Fig. 1.

proximation is best for materials where $\kappa = \lambda/\xi \gg 1$ as is the case for these materials (see below). These two equations can be conveniently combined into a single equation for κ ,

$$\frac{H_{c1}}{H_{c2}} = \frac{\ln\kappa}{2\kappa^2},\tag{1}$$

which can be applied to uniaxial superconductors^{19,20} by allowing κ to take two values,

$$\kappa^{c} = \frac{\lambda_{a,b}^{c}}{\xi_{a,b}}, \ \kappa^{a,b} = \left(\frac{\lambda_{a,b}^{b,a}\lambda_{c}^{b,a}}{\xi_{a,b}\xi_{c}}\right)^{1/2},$$

for fields in the c direction and the a,b plane, respectively, where the subscripts denote the direction in which the field penetration or coherence is being described, and the superscripts denote field direction.

We estimate the values of H_{c2} at low temperature from Werthamer, Helfand, and Hohenberg extrapolations of measurements of the initial slope $dH_{c2}/dT|_{T_c}$ reported in the literature. For fields in the *a,b* plane we chose $dH_{c2}/dT = 3.6$ T/K, which agrees within 15% with the three measurements²¹⁻²³ available. For fields along the *c* direction we chose the values 0.55 T/K (Refs. 22 and 23) and 1.1 T/K.²¹ These values extrapolate to critical fields at T = 0 shown in Table II where Pauli limiting has not been taken into account. The values of κ and λ in Table II were derived from Eq. (1) using our measured values of H_{c1} . Because ξ enters the expression for H_{c1} only loga-



FIG. 3. The deviation of magnetization from the initial linear behavior for the as-grown (\Box) and annealed (\Box) crystals.

rithmically, the factor of 2 change in H_{c2} for $H \parallel c$, does not seriously change the final values for λ .

It is important to note the geometry describing the field penetration in uniaxial superconductors²⁴ as illustrated in Fig. 4. The penetration depth in any direction is controlled by the shielding supercurrents in a perpendicular direction. Thus if c is the hard direction for supercurrent flow, then $\lambda_{a,b}^{b} \gg \lambda_{a,b}^{c}$. Because supercurrents in the a,bplane control both the penetration along a or b for fields along c and the penetration along c for fields in the a,bplane, $\lambda_{a,b}^{c} = \lambda_{c}^{b,a}$.

The values for H_{c1} are considerably smaller and the values of λ and κ considerably larger than those estimated from single-crystal magnetization measurements reported earlier.¹³ Measurements on polycrystalline samples by ESR (Ref. 25) give values of H_{c1} of about 600 G, and by muon-spin rotation²⁶ give values of λ of 1400 Å. We find that these values are within the anisotropy range and are thus potentially consistent with our results. The condensation energy calculated from the formulas $H_c = H_{c2}/\kappa\sqrt{2}$ and $H_c = H_{c1}2\kappa/\ln\kappa$ is in the range 7-12 kG for the two directions assuming the lower value of H_{c2} in the *c* direction.

The anisotropy in H_{c1} can be treated phenomenologically with an anisotropic effective mass in either the Ginzburg-Landau (GL) equations^{19,27} or the London equations.^{24,28} In the GL approach, the mass is the only anisotropic parameter so that the anisotropy in any superconducting property can be used to infer the anisotropy in

TABLE I. Values of the applied field where the magnetization curve deviates from linearity for the three crystals. The two numbers shown for as-grown and annealed crystal refer to two perpendicular symmetry directions in the basal plane. Values of the demagnetizing factor n are taken from the initial slope of the magnetization curve.

	H∥a,b (G)	H c (G)	n	Mass (mg)	Dimensions (mm)
As-grown sample	90 ± 10 100 ± 10	425 ± 50	0.31(H a,b) 0.45(H c)	0.175	0.3×0.3×0.3
Annealed sample	80 ± 10 70 ± 10	375 ± 50	0.31(H a,b) 0.47(H c)	0.672	0.3×0.3×0.3
Irradiated sample	90 ± 10	325 ± 50	$0.27(H \ a, b)$ $0.46(H \ c)$	0.497	0.5×0.5×0.25

	H∥c		H∥a,b
H _{c1}	$690 \pm 50 \text{ G}$	······································	120 ± 10 G
H_{c2}	35 T *		230 T
	70 T [†]		
ξa,b		31 Å* 22 ņ	
ξc		4.8 Å*	
κ	29 * 44 [†]	0.7 A	230
λ <i>b,a</i>	900 Å*		8400 Å* 7800 ņ
λς	930 A		900 Å
$J_{c1,b,a} \\ J_{c1,c}$	6×10^7 A/cm ²		$1 \times 10^{7} \text{ A/cm}^{2}$ $1.2 \times 10^{6} \text{ A/cm}^{2}$

the mass. The ratio $\kappa^{a,b}/\kappa^c$ in Table II implies an effective-mass ratio of 63, while the measured values of H_{c1} and the values of κ together imply a mass ratio of 78. This is to be compared with a mass ratio inferred from the values of H_{c2} or 43 assuming the lower value of H_{c2} in the *c* direction. The mass anisotropy arising from the normal-state Fermi surface can be estimated within the isotropic relaxation-time approximation by the anisotropy in the resistivity, which gives values in the range 30-90 depending on temperature.¹⁴

The equilibrium supercurrent density flowing at H_{c1} can be estimated from Maxwell's equation and the approximation $H_{c1}/\lambda \sim dH/dx = (4\pi/c)J$. The results in Table II show a large anisotropy of as much as a factor of 50, demonstrating that even perfect samples with no pinning sites would contain "hard" and "easy" axes for supercurrent flow. These intrinsic anisotropies in maximum supercurrent flow are qualitatively consistent with the effective-mass anisotropy inferred from the critical fields.

In summary, we have measured the magnitude and anisotropy in the lower critical field at low temperature for the 90-K superconductor YBa₂Cu₃O_{7- δ}. We find significantly lower values of H_{c1} and higher values of κ than previously thought, with an anisotropy roughly consistent with those of the upper critical field and the normal-state resistivity within GL theory. We find a large magnetic penetration depth of order 8000 Å for fields in the *a,b* plane, which may lead to sample size limitations of the



FIG. 4. Spacial representation of the magnetic penetration depth and the corresponding shielding currents. Unshaded areas represent the area of magnetic penetration characterized by λ . The field direction is indicated by superscripts.

shielding and Meissner effects in thin-film and polycrystalline samples. We find considerable anisotropy in the equilibrium shielding supercurrent density flowing at H_{c1} , showing directly the existence of hard and easy directions for supercurrent flow that are related to intrinsic anisotropies in the superconductivity rather than to pinning sites in the material.

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- ¹M. A. Beno, L. Soderholm, D. W. Capone II, D. G. Hinks,
- J. D. Jorgenson, J. D. Grace, I. K. Schuller, C. U. Segre, and K. Zhang, Appl. Phys. Lett. 51, 57 (1987).
- ²J. E. Greedan, A. O'Reilly, and C. V. Stager, Phys. Rev. B 35, 8770 (1987).
- ³J. J. Capponi, C. Chaillout, A. W. Hewat, P. Lejay, M. Ma-

rezio, N. Nguyen, B. Raveau, J. L. Soubeyroux, J. L. Tholence, and R. Tournier, Europhys. Lett. 3, 1301 (1987).

- ⁴J. J. Yu, S. Massida, A. J. Freeman, and D. D. Koelling, Phys. Lett. A **122**, 203 (1987).
- ⁵L. Mathiess and D. R. Hamman, Solid State Commun. **63**, 395 (1987).
- ⁶P. Chaudhari, R. H. Koch, R. B. Laibowitz, T. McGuire, and R. Gambino, Phys. Rev. Lett. **58**, 2684 (1987).
- ⁷T. R. Dinger, T. K. Worthington, W. J. Gallagher, and R. L. Sandstrom, Phys. Rev. Lett. 58, 2687 (1987).
- ⁸G. W. Crabtree, J. Z. Liu, A. Umezawa, W. K. Kwok, C. H. Sowers, S. K. Malik, B. W. Veal, D. J. Lam, M. B. Brodsky, and J. W. Downey, Phys. Rev. B 36, 4021 (1987).
- ⁹R. N. Shelton, R. W. McCallum, M. A. Damento, and K. A. Gschniedner, Jr., Int. J. Mod. Phys. (to be published).
- ¹⁰L. F. Schneemeyer, E. M. Gyorgy, and J. V. Waszczak, Phys. Rev. B 36 8804 (1987).
- ¹¹Y. Hidaka, Y. Enomoto, M. Suzuki, M. Oda, A. Katsui, and T. Murakama, Jpn. J. Appl. Phys. 26, L726 (1987).
- ¹²Y. Iye, T. Tamagai, H. Takeya, and H. Takei, Jpn. J. Appl. Phys. 26, L1057 (1987).
- ¹³T. K. Worthington, W. J. Gallagher, and T. R. Dinger, Phys. Rev. Lett. 59, 1160 (1987).
- ¹⁴S. W. Tozer, A. W. Kleinsasser, T. Penney, D. Kaiser, and F. Holtzberg, Phys. Rev. Lett. 59, 1768 (1987).
- ¹⁵K. Murata, K. Hayashi, Y. Honda, M. Tokumoto, H. Ihara, M. Hirabayashi, N. Terada, and Y. Kimura, Jpn. J. Appl. Phys. Lett. 26, L1941 (1987).

- ¹⁶J. Z. Liu, G. W. Crabtree, A. Umezawa, and Li Zongquan, Phys. Lett. **121**, 305 (1987).
- ¹⁷A. Umezawa, G. W. Crabtree, J. Z. Liu, H. W. Weber, W. K. Kwok, L. H. Nunez, T. J. Moran, C. H. Sowers, and H. Claus, Phys. Rev. B 36, 7151 (1987).
- ¹⁸C. P. Bean, Phys. Rev. Lett. 8, 250 (1962).
- ¹⁹D. R. Tilley, Proc. Phys. Soc. (London) 85, 1177 (1965).
- ²⁰P. de Trey, S. Gygax, and J. P. Jan, J. Low Temp. Phys. 11, 421 (1973).
- ²¹Y. Iye, T. Tamagai, H. Takeya, and H. Takei, in Proceedings of the Eighteenth Yamada Conference on Superconductivity in Highly Correlated Fermion Systems, Sendai, Japan, 1987, edited by M. Tachiki, Y. Muto, and S. Maekawa (North-Holland, Amsterdam, 1987).
- ²²W. J. Gallagher, T. K. Worthington, T. R. Dinger, and R. L. Sandstrom, in Ref. 21.
- ²³Y. Hidaka, M. Oda, M. Suzuki, A. Katsui, T. Murakami, N. Kobayashi, and Y. Muto, in Ref. 21.
- ²⁴A. V. Balatskii, L. I. Burlachkov, and L. P. Gor'kov, Zh. Eksp. Teor. Fiz. **90**, 1478 (1986) [Sov. Phys. JETP **63**, 866 (1986)].
- ²⁵C. Rettori, D. Davidov, I. Belaish, and I. Felner, Phys. Rev. B 36, 4028 (1987).
- ²⁶D. R. Harshman, G. Aeppli, E. J. Ansaldo, B. Batlogg, J. H. Brewer, J. F. Carolan, R. J. Cava, M. Celio, A. C. D. Chaklader, W. N. Hardy, S. R. Kreitzman, G. M. Luke, D. R. Noakes, and M. Senba, Phys. Rev. B 36, 2386 (1987).
- ²⁷R. A. Klemm and J. R. Clem, Phys. Rev. B 21, 1868 (1980).
- ²⁸V. G. Kogan, Phys. Rev. B 24, 1572 (1981).



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