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LOWER CRITICAL FIELD MEASUREMENTS IN $YBa_2Cu_3O_{6+x}$ SINGLE CRYSTALS

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The temperature dependence of the lower critical field in $YBa_2Cu_3O_{6+x}$ single crystals has been determined by magnetization measurements with the applied field parallel and perpendicular to the c-axis. Results were compared with data from the literature and fitted to Ginzberg-Landau equations by assuming a linear dependence of the parameter κ on temperature. A value of 7 ± 2 kOe was estimated for the thermodynamic critical field at $T = 0$ by comparison of calculated H_{c2} values with experimental data from the literature.

INTRODUCTION

The lower critical field (H_{c1}) of the high temperature superconductor $YBa_2Cu_3O_{6+x}$ (YBCO) is an intrinsic material property which depends upon temperature and crystallographic orientation. Accurate measurement of H_{c1} is complicated by flux pinning and edge effects (as illustrated in Fig. 1) and by uncertainty in the demagnetizing factor. Consequently, early reported H_{c1} values in single crystals[1-5] were up to an order of magnitude larger than later values.[6-12]

In this study, H_{c1} measurements were made on twinned and detwinned crystals and the data were compared with results of previous investigations for orientations with the applied field parallel and perpendicular to the c-axis of the crystal. The experimental data were fitted to Ginzberg-Landau equations for the dependence of H_{c1} on temperature and upper critical field (H_{c2}) values were calculated for the two orientations of interest.

EXPERIMENTAL PROCEDURE

The YBCO crystals used in the present study (Fig. 2) were grown from Y-Ba-Cu-O melts[13] and subsequently annealed in oxygen gas at 420°C for 80 h to obtain superconducting transition temperatures $T_c > 90$ K. (Thus, the oxygen content $6+x > 6.85$[14]). Two crystals were selected for measurement. The first crystal (AN3-5) exhibited characteristic {110} twin planes and was nearly cubic with dimensions $120 \times 135 \times 120\ \mu m^3$ (c-dimension = 120 μm). Due to the cubic morphology, the demagnetizing factors were nearly identical in all three dimensions. The second crystal (AN9-5) was fully detwinned via a thermomechanical process developed in our laboratory[15] and had dimensions $a \times b \times c = 200 \times 250 \times 100\ \mu m^3$.

Magnetic measurements were made using a superconducting quantum interference device (SQUID) magnetometer, with the c-axis of the crystal aligned either perpendicular or parallel to the applied field H. The crystal was first cooled in zero field to a predetermined temperature and the magnetization was then measured as the applied field was increased to a value in excess of H_{c1}. For temperatures greater than about 60 K, sharp breaks from linearity in the M vs. H curves were observed for both crystals, making the estimation of H_{c1} relatively precise. Below 60 K, the

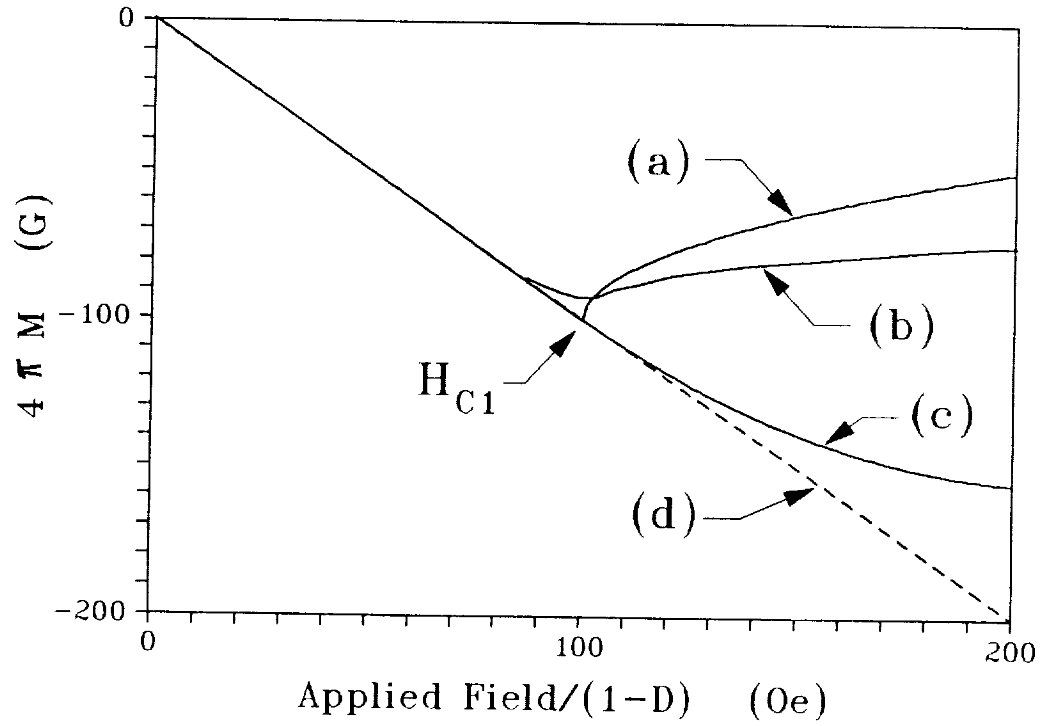

Fig. 1. Hypothetical magnetization curves after cooling in zero field for a sample: (a) at equilibrium (i.e., an ellipsoid with no flux pinning); (b) with some pinning and edge effects; (c) with strong pinning; and (d) with perfect diamagnetic character (i.e., magnetization is proportional to the applied field after correction for the demagnetizing factor, D). For the equilibrium case (a), there is a well-defined, sharp break at H_{c1}, which allows for an accurate determination of H_{c1}. Pinning and edge effects (b and c) make it difficult to estimate the true H_{c1}. In case (b), the observed onset occurs at applied fields below H_{c1} due to flux penetration at the sharp edges and corners of the crystal. In case (c), pinning causes a gradual departure from linearity, making the estimate of H_{c1} less accurate.

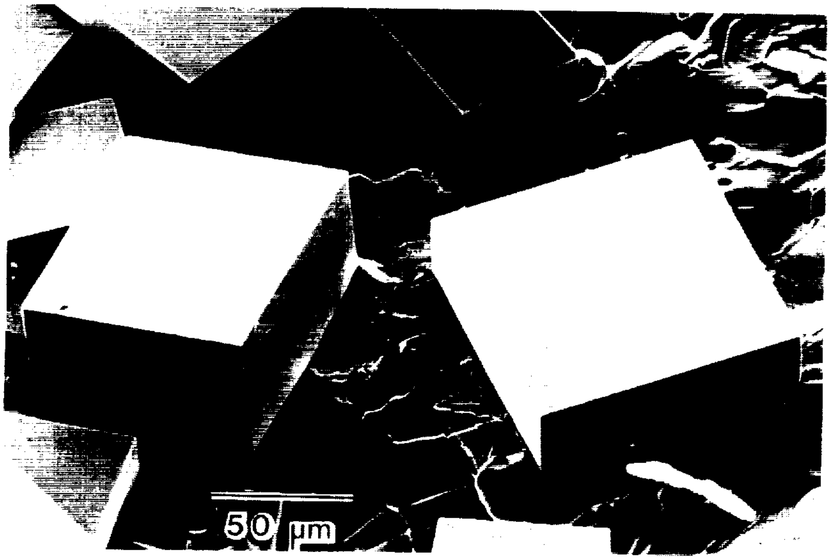

Fig. 2. Scanning electron micrograph of as-grown YBCO single crystals. The smallest dimension of a crystal generally lies along the c-axis of the unit cell.

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departure from linearity was more gradual and H_{c1} was estimated from the initial point of departure from linearity.

RESULTS AND DISCUSSION

Temperature-dependent H_{c1} data from the present study and previous investigations[4,6-8,10-12] for H ∥ c and H ⊥ c are presented in Fig. 3. The earliest reported H_{c1} values[1-3,5] were erroneously high due to difficulties in defining H_{c1} and are not included in the two plots. The curves shown in each plot were obtained by fitting all displayed data points using the Ginzberg-Landau equation[16]

$$H_{c1} = H_c(\ln \kappa + 0.08)/\sqrt{2}\,\kappa, \qquad (1)$$

where H_c is the thermodynamic critical field at temperature T as given by

$$H_c = H_{c0}\ (1 - t^2). \qquad (2)$$

Here H_{c0} is H_c at $T = 0$, t is the reduced temperature T/T_c and κ is the Ginzberg-Landau parameter (the ratio of the penetration depth λ to the coherence length ξ). Data for both H ∥ c and H ⊥ c can be well-fitted by assuming that κ varies linearly with temperature:

$$\kappa = a + bt. \qquad (3)$$

Good fits of Eq. (1) may be obtained for a wide range of H_{c0} values, leading to a wide range of values for the parameters a and b in Eq. (3). The range of permissible H_{c0} values is limited considerably by requiring that H_{c2} values calculated from a second Ginzberg-Landau equation

$$H_{c2} = \sqrt{2}\,\kappa\, H_c \qquad (4)$$

be in reasonable agreement with experimental H_{c2} data from the literature. This comparative analysis yields a value of 7 kOe for H_{c0}, $\kappa = 100 + 85t$ for H ⊥ c and $\kappa = 22 + 22t$ for H ∥ c. The calculated curves and data for H_{c2} are compared in Fig. 4. Considering the large uncertainties in the experimental H_{c2} data and the obvious differences in T_c for samples from different studies, agreement of the calculated curves with the data is a matter of judgement. The maxima seen in the calculated H_{c2} curves are unphysical, indicating that the linear form used for κ should be modified, e.g., by the addition of a quadratic term in t. However, the large uncertainty in the H_{c0} value used here (7 ± 2 kOe) does not justify such an additional term. Considering the large variations in the experimental H_{c2} data, our H_{c0} value is in reasonable agreement with the value of 10 kOe estimated by Worthington et al.[6] The resulting uncertainties in κ values calculated from our equations are also of order ±30%.

Our H_{c1} data for the detwinned crystal AN9-5 shown in Fig. 3a (H ∥ c) are in good agreement with the data of Krusin-Elbaum et al.[10] for twinned crystals. This result indicates that twin boundaries have only a small effect on H_{c1}, as noted in our earlier investigation.[17] Our H_{c1} data for the twinned crystal AN3-5 shown in Fig. 3b (H ⊥ c) are in reasonable agreement with the data from previous investigations.[4,6-8,10-12] Anisotropy in H_{c1} (H_{c1} ∥ c/H_{c1} ⊥ c) as calculated from the two curves in Fig. 3 was 3.1 ± 0.1 for 10 K < T < 80 K.

The calculated H_{c2} values for H ∥ c (Fig. 4a) are in reasonable agreement with the data of Welp et al.[18] near the superconducting transition and follow the general trend of the data of Iye et al.[19] and Worthington et al.[2]. For H⊥c (Fig. 4b) the calculated values at high temperature show good agreement with the experimental data of Gallagher et al.[3] and Welp et al.[18] Anisotropy in H_{c2} (H_{c2} ⊥ c/H_{c2} ∥ c) as calculated from the two curves in Fig. 4 was 4.3 ± 0.2 for 10 K < T < 80 K.

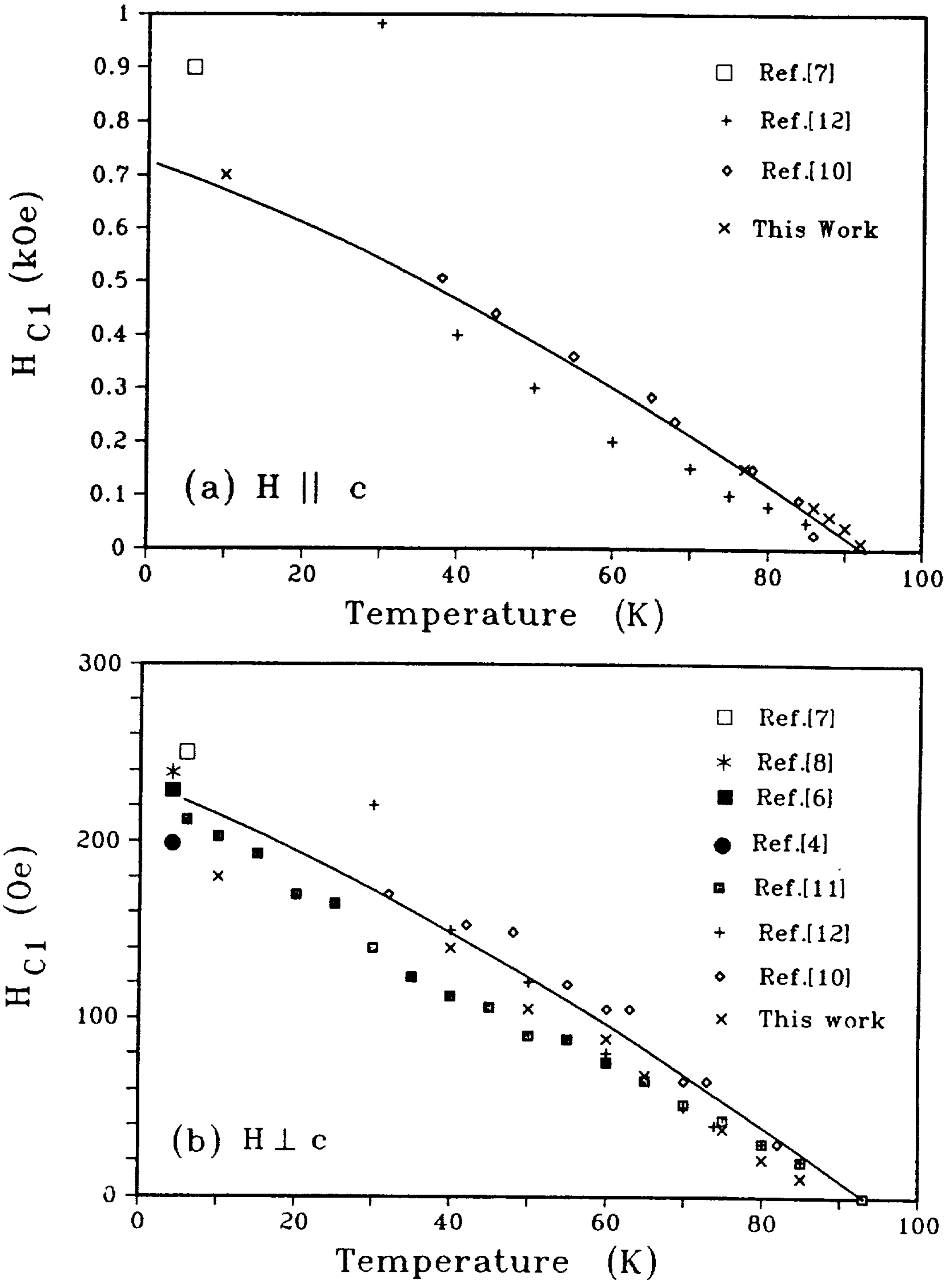

Fig. 3. Temperature-dependent H_{c1} data for (a) $H \parallel c$ (strong pinning) and (b) $H \perp c$ (weak pinning). Our data are for (a) the detwinned crystal AN9-5 and (b) the twinned crystal AN3-5. The curves were generated by fitting the data to the Ginzberg-Landau equation for H_{c1} and temperature-dependent κ equations given by (a) $\kappa = 22 + 22t$, and (b) $\kappa = 100 + 85t$ ($t = T/T_c$).

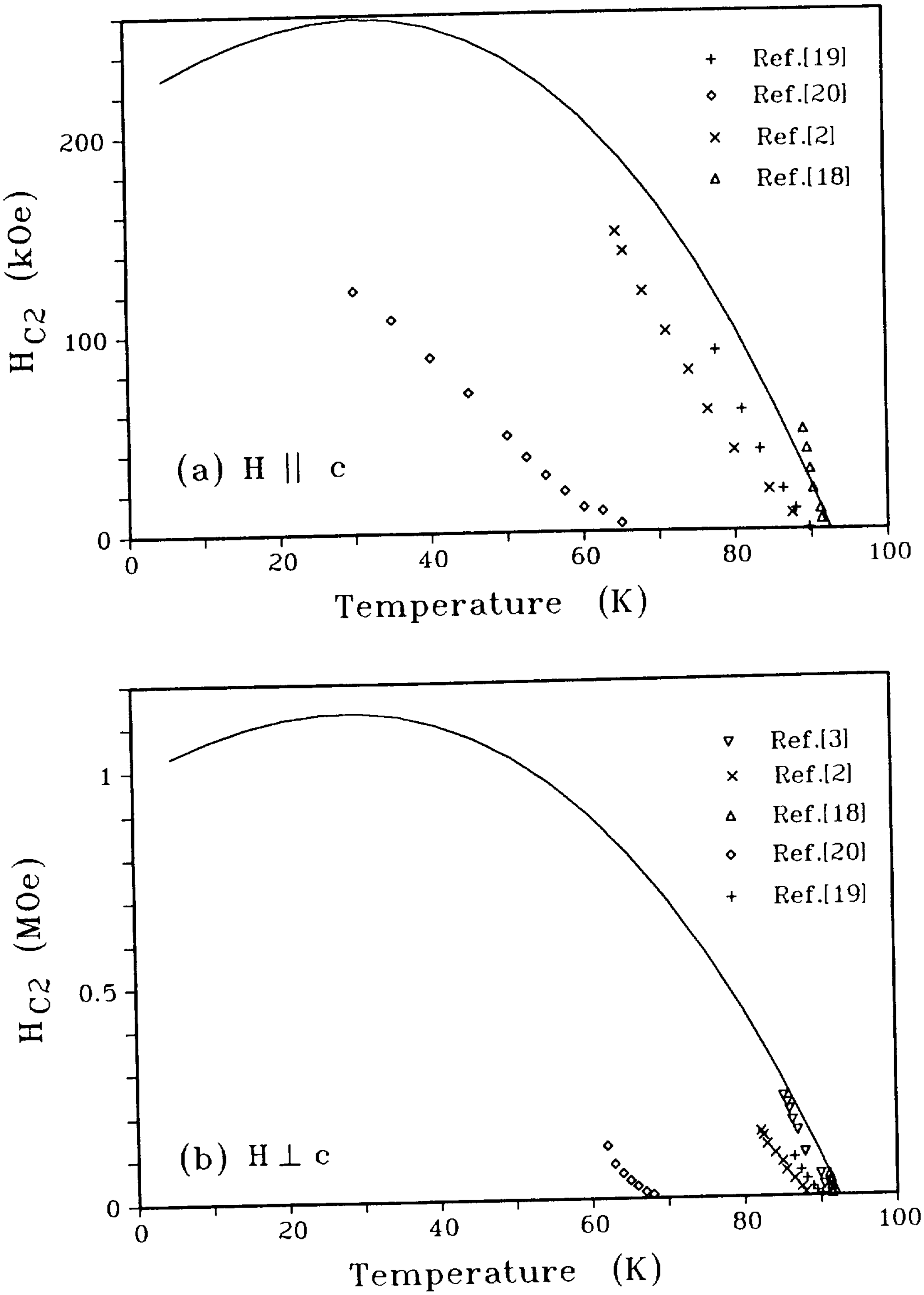

Fig. 4. Temperature-dependent H_{c2} curve (solid line) calculated from the Ginzberg-Landau equation for H_{c2} and the temperature-dependent κ equation for (a) $H \parallel c$ and (b) $H \perp c$. Experimental H_{c2} data from the literature are shown for comparison. The extrapolations to lower temperatures ($T < 60$ K) are unreliable and the maxima in the curves probably do not exist.

CONCLUSIONS

Temperature-dependent H_{c1} results were determined from magnetization measurements on detwinned and twinned single crystals of YBCO for $H \parallel c$ and $H \perp c$. The results from the present study and previous investigations for each orientation were fitted to Ginzberg-Landau equations assuming a linear temperature dependence for the parameter κ. H_{c2} values calculated from the Ginzberg-Landau equation and the temperature-dependent κ relations were in reasonable agreement with experimental H_{c2} data from the literature near the superconducting transition temperature. Values of $H_{c0} = 7 \pm 2$ kOe, $\kappa = 100 + 85t$ for $H \perp c$, and $\kappa = 22 + 22t$ for $H \parallel c$ were estimated from the analysis.

Acknowledgments

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References

1. T.R. Dinger, T.K. Worthington, W.J. Gallagher, and R.L. Sandstrom, "Direct Observation of Electronic Anisotropy in Single-Crystal $Y_1Ba_2Cu_3O_{7-x}$," *Phys. Rev. Lett.* **58**, 2687-2690 (1987).
2. T.K. Worthington, W.J. Gallagher, and T.R. Dinger, "Anisotropic Nature of High-Temperature Superconductivity in Single Crystal $YBa_2Cu_3O_{7-x}$," *Phys. Rev. Lett.* **59**, 1160-1163 (1987).
3. W.J. Gallagher, T.K. Worthington, T.R. Dinger, F. Holtzberg, D.L. Kaiser, and R.L. Sandstrom, "Anisotropy in the Magnetic Properties of Single-Crystal $Y_1Ba_2Cu_3O_{7-x}$," *Physica* **148B**, 228-232 (1987).
4. T.R. McGuire, T.R. Dinger, P.J.P. Freitas, W.J. Gallagher, T.S. Plaskett, R.L. Sandstrom, and T.M. Shaw, "Magnetic Properties of Y-Ba-Cu-O Superconductors," *Phys. Rev. B* **36**, 4032-4035 (1987).
5. R.N. Shelton, R.W. McCallum, M.A. Damento, K.A. Gschneider, Jr., H.C. Ku, H.D. Yang, J.W. Lynn, W.-H. Li, and Q. Li, "Effects of Crystal Anisotropy on Magnetization and Magnetic Order in Superconducting $RBa_2Cu_3O_{7-x}$," *Physica B* **148B**, 285-288 (1987).
6. T.K. Worthington, Y. Yeshurun, A.P. Malozemoff, R.M. Yandrofski, F. Holtzberg, and T.R. Dinger, "The Effect of Flux Pinning and Flux Creep on Magnetic Measurements of Single Crystal $Y_1Ba_2Cu_3O_{7-x}$," *J. de Phys.* **49**, C8-2093-2098 (1988).
7. Y. Yeshurun, A.P. Malozemoff, F. Holtzberg, and T.R. Dinger, "Magnetic Relaxation and the Lower Critical Fields in a Y-Ba-Cu-O Crystal," *Phys. Rev. B* **38**, 11828-11831 (1988).
8. L. Fuchter, C. Giovannella, G. Collin, and I.A. Campbell, "Lower Critical Fields and Pinning in $Y_1Ba_2Cu_3O_{7-x}$," *Physica C* **156**, 69-72 (1988).
9. A. Umezawa, G.W. Crabtree, J.Z. Liu, T.J. Moran, S.K. Malik, L.H. Nunez, W.L. Kwok, and C.H. Sowers, "Anisotropy of the Lower Critical Field, Magnetic Penetration Depth, and Equilibrium Shielding Current in Single-Crystal $YBa_2Cu_3O_{7-x}$," *Phys. Rev. B* **38**, 2843-2846 (1988).
10. L. Krusin-Elbaum, A.P. Malozemoff, Y. Yeshurun, D.C. Cronemeyer, and F. Holtzberg, "Temperature Dependence of Lower Critical Fields in Y-Ba-Cu-O Crystals," *Phys. Rev. B* **39**, 2936-2939 (1989).
11. A. Umezawa, G.W. Crabtree, K.G. Vandervoort, U. Welp, W.K. Kwok, and J.Z. Liu, "Temperature Dependence of the Lower Critical Field of Single Crystal $YBa_2Cu_3O_{7-x}$," *Physica C* **162-164**, 733-734 (1989).
12. H. Adrian, W. Assmus, A. Hohr, J. Kowalewski, H. Spille, and F. Steglich, "Anomalous $H_{c1}(T)$ Behavior of Single Crystalline $YBa_2Cu_3O_{7-x}$," *Physica C* **162-164**, 329-330 (1989).
13. D.L. Kaiser, F. Holtzberg, B.A. Scott, and T.R. McGuire, "Growth of $YBa_2Cu_3O_x$ Single Crystals," *Appl. Phys. Lett.* **51**, 1040-1042 (1987); D.L. Kaiser, F. Holtzberg, M.F.

Chisholm, and T.K. Worthington, "Growth and Microstructure of Superconducting $YBa_2Cu_3O_x$ Single Crystals," *J. Cryst. Growth* **85**, 593-598 (1987).

14. R.J. Cava, B. Batlogg, C.H. Chen, E.A. Rietman, S.M. Zahurak, and D. Werder, "Single-phase 60-K Bulk Superconductor in Annealed $Ba_2YCu_3O_{7-x}$ $(0.3 < x < 0.4)$ with Correlated Oxygen Vacancies in the Cu-O Chains," *Phys. Rev. B* **36**, 5719-5722 (1987).
15. D.L. Kaiser, F.W. Gayle, L.J. Swartzendruber, and R.S. Roth, "Thermomechanical Detwinning of Superconducting $YBa_2Cu_3O_{7-x}$ Single Crystals," *J. Mater. Res.* 4, 745-747 (1989).
16. A.A. Abrikosov, Fundamentals of the Theory of Metals, p. 325 (North Holland, Amsterdam, 1988).
17. L.J. Swartzendruber, A. Roitburd, D.L. Kaiser, F.W. Gayle, and L.H. Bennett, "Direct Evidence for an Effect of Twin Boundaries on Flux Pinning in Single-Crystal $YBa_2Cu_3O_{6+x}$," *Phys. Rev. Lett.* **64**, 483-486 (1990).
18. U. Welp, W.K. Kwok, G.W. Crabtree, K.G. Vandervoort, and J.Z. Liu, "Magnetic Measurements of the Upper Critical Field of $YBa_2Cu_3O_{7-x}$ Single Crystals," *Phys. Rev. Lett.* **62**, 1908-1911 (1989).
19. Y. Iye, T. Hiroyuki, H. Takeya, and H. Takei, "The Anisotropic Upper Critical Field of Single Crystal $YBa_2Cu_3O_x$," *Jap. J. Appl. Phys.* **26**, 1057-L1059 (1987).
20. Y. Hidaka, Y. Enomoto, M. Suzuki, M. Oda, A. Katsui, and T. Murakami, "Anisotropy of the Upper Critical Magnetic Field in Single Crystal $YBa_2Cu_3O_{7-x}$," *Jap. J. Appl. Phys.* **26**, L726-L728 (1987).