Temperature dependence of the first critical field of $YBa_2Cu_3O_x$ single crystals with various oxygen contents

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The temperature dependence of the first critical field, $H_{c1}(T)$, has been measured in $YBa_2Cu_3O_x$ single crystals by an $M_t^{1/2} \to 0$ extrapolation method (M_t is the trapped magnetization). These measurements were carried out in the orientations $H \perp C$ and $H \| c$. The anisotropy of H_{c1} increases sharply, from $H_{c1}^{\|c}(0)/H_{c1}^{\perp c}(0) = 4.1$ to $(H_{c1}^{\|c}(0) = 270\ \text{Oe})/[H_{c1}^{\perp c}(0) = 27\ \text{Oe}] = 10$, as the oxygen content decreases from $x = 6.9$ to $x = 6.64$.

The standard methods for determining the first critical field H_{c1} in type-II superconductors generally reduce to finding that value $H = H_{\text{dev}}$ of the magnetic field at which the H dependence of the magnetization M begins to deviate from a linear dependence with a slope $dM/dH = -1/(4\pi)$. If the methods used to measure $M(H)$ are overly sensitive, H_{dev} will correspond to the beginning of the penetration of vortices into those regions of the test sample in which the demagnetizing field is at a maximum, i.e., at the corners of the sample, along its edges, at clusters of normal defects, etc. In this case, H_{dev} will obviously have nothing in common with the first critical field of the bulk test sample. On the other hand, if one sets some fixed finite value $\Delta M = \Delta M_c$ of the deviation at the outset, one may "miss" the true value of H_{c1} because of an overly crude approximation of ΔM_c. All methods for determining H_{c1} from a "threshold" deviation from linearity are thus unavoidably based on a rather subjective choice of some deviation ΔM_c which is presumed to give the "true" value $H_{\text{dev}} = H_{c1}$. We believe that this arbitrary note should be avoided by using a method of extrapolating the deviation from linearity, $\Delta M^{1/2}$ (Refs. 1 and 2), or the trapped magnetization $M_t^{1/2}$ (Ref. 3) to zero. We showed in Ref. 3 that the following relations hold for a plate of thickness d of a type-II Bean superconductor with strong pinning:

$$\Delta M = \frac{(H - H_{c1})^2}{2(H^* - H_{c1})} + \frac{(1 - m_{eq})H_{c1}(H - H_{c1})}{H^* - H_{c1}}, \quad (1)$$

$$M_t = \frac{(H - H_{c1})^2}{4(H^* - H_{c1})} + \frac{(1 - m_{eq})H_{c1}(H - H_{c1})}{H^* - H_{c1}}. \quad (2)$$

Here $H^* = \pi j_c d/5$, j_c is the critical current, and the parameter m_{eq} characterizes the anomaly in $M(H)$ at $H = H_{c1}$: $m_{eq} = 1$ if there is no $M(H = H_{c1})$ discontinuity. A modified Bean model with $m_{eq} = 1$ was analyzed in Refs. 4 and 5, while arbitrary values of m_{eq} were studied in Ref. 6. A comparison with experimental data on $M(H)$ yields $m_{eq} = 0.86$–0.92 for $YBa_2Cu_3O_x$ (Ref. 6).

An important point is that ΔM and M_t have a quadratic dependence on $(H - H_{c1})$, which is valid for both the plate and a cylinder if $m_{eq} \approx 1$ (Ref. 3). In this case we can determine H_{c1} through an extrapolation to zero: $M_t^{1/2} \to 0$ (Ref. 3) or $\Delta M^{1/2} \to 0$ (Refs. 1 and 2). There is no arbitrariness in the choice of the critical deviation ΔM_c in this method, so the extrapolation $M_t^{1/2} \to 0$ or $\Delta M^{1/2} \to 0$, based on functional relations (1) and (2), which are valid for the entire bulk sample, is vastly more reliable than methods based on a subjective choice of ΔM_c. In the present study, we have used the extrapolation $M_t^{1/2} \to 0$ to find the temperature dependence of the first critical field, $H_{c1}(T)$, for the configurations $H \| C$ and $H \perp C$ in $YBa_2Cu_3O_x$ single crystals differing in oxygen content x. The extrapolation method is based on an analysis of data on the trapped magnetization in the coordinates $M_t^{1/2} = f(H)$; this method has been used in previous studies[1,2] for data on the deviation from linearity ΔM.

The single crystals serving as our test samples had dimensions of $2 \times 2 \times 1$ mm. The oxygen content was found by a gravimetric method and also by analyzing a neutron diffraction pattern by the Rietveld method, which makes it possible to determine the occupation numbers of all the crystallographic oxygen sites, O1–O4, and thus

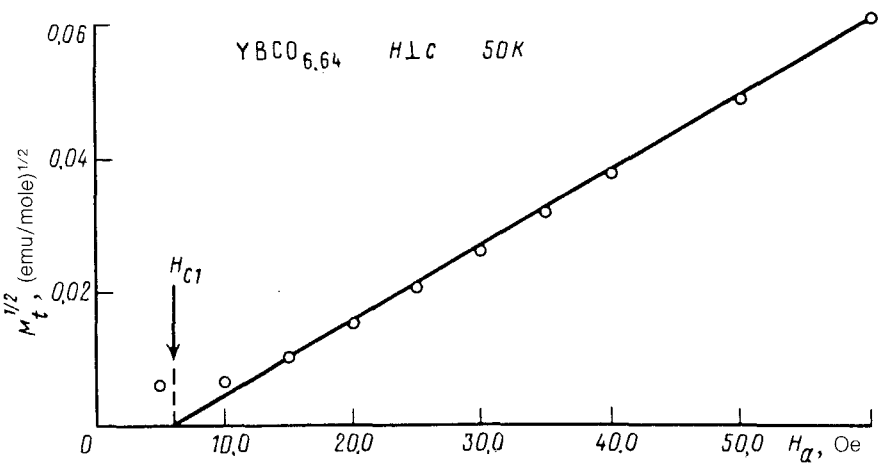

FIG. 1. Field dependence of the trapped magnetization M_t for a $YBa_2Cu_3O_{6.64}$ single crystal in the coordinates $M_t^{1/2} = f(H)$.

to calculate x in $YBa_2Cu_3O_x$. The trapped magnetization M_t was measured with the help of a Quantum Design SQUID magnetometer, in the following way: For each point $M_t(H_0, T_0)$, the sample was heated to a temperature above the transition temperature, $T > T_c$, and then cooled in a zero field to $T = T_0 < T_c$. Here, the first measurement, $M^{(1)}(0, T_0)$, was taken. At a fixed temperature $T = T_0$, a field $H = H_0$ was then applied, and $M^{(2)}(H_0 T_0)$ was measured. The field was then turned off, and $M^{(3)}(0, T_0)$ was measured. The trapped moment M_t was found from the difference $M_t(H_0, T_0) = M^{(3)}(0, T_0) - M^{(1)}(0, T_0)$. In this manner, curves of $M_t(H, T)$ and $M(H, T) = M(H_0, T_0)$ were plotted. This method requires extremely time-consuming measurements, since the sample is heated above T_c and then cooled in a zero field for each point $M_t(H_0, T_0)$.

These experiments verified that relation (2) can indeed be used to describe the functional dependence $M_t(H)$ (Fig. 1), since $M_t^{1/2}$ varies linearly with the field. It thus becomes possible to work from the abscissa intercept of $M_t^{1/2}$ to find the value of H_{c1}, after the corresponding demagnetizing fields are taken into account. For samples with dimensions of $2 \times 2 \times 1$ mm, the demagnetizing factors N found in the approxima-

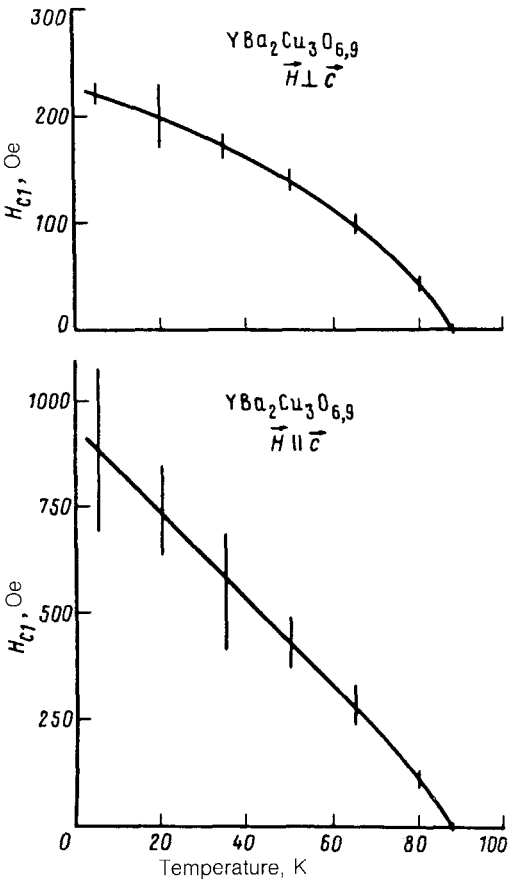

FIG. 2. Temperature dependence of the first critical field $H_{c1}(T)$ of a $YBa_2Cu_3O_{6.9}$ single crystal.

tion of an inscribed ellipsoid are[7] $N^{\|c} = 0.53$ and $N^{\perp c} = 0.234$. All the data on H_{c1} given below have been corrected for the demagnetizing factor. Strictly speaking, this procedure (and, incidentally, all other procedures) are capable of measuring only the field H_p, at which the flux begins to penetrate into the sample. Nevertheless, we assume everywhere below that $H_p = H_{c1}$.

The temperature dependence $H_{c1}(T)$ depends strongly on the orientation of the field: In the orientation $H \perp C$, the $H_{c1}(T)$ curves reach saturation as the temperature is lowered, while the $H_{c1}^{\|c}(T)$ curves remain linear (Figs. 2 and 3). An anomalous temperature dependence $H_{c1}(T)$ in a high-T_c superconductor has been seen previously,[8-11] but in many of these cases it might have been caused by a sharp increase in M accompanying an increase in the critical current j_c at low temperatures. In our own case, such an error could not have occurred, but still we find the anomalous $H_{c1}^{\|c}(T)$ dependence. Under the assumption $H_{c1} \sim \Phi_0/\lambda^2$, it is difficult to reconcile the strong low-temperature dependence $H_{c1}(T)$ with the independence of the London penetra-

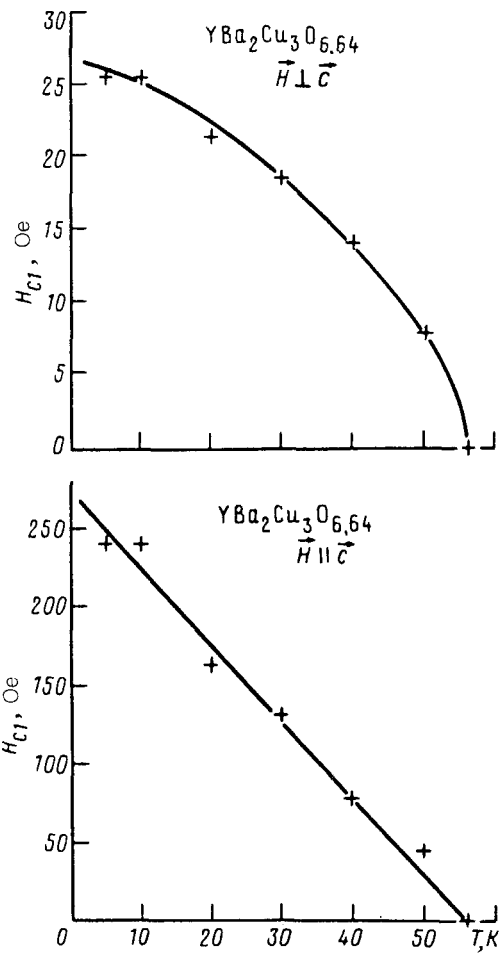

FIG. 3. Temperature dependence of the first critical field $H_{c1}(T)$ of a $YBa_2Cu_3O_{6.64}$ single crystal.

tion depth $\lambda(T)$ from the temperature in the 1-2-3 high-T_c superconductors at $T \leqslant 50$ K (Refs. 12–14). Koyama *et al.*[15] have suggested one way to resolve the contradiction in the interpretation of the results on $\lambda(T)$ and $H_{c1}^{\parallel c}(T)$. Considering layered systems in which a superconductivity is induced between layers by a proximity effect, they derived in place of the relation $H_{c1} \sim \Phi_0/\lambda^2$ the relation $H_{c1} \sim \Phi_0/\lambda\xi_P$, where the parameter ξ_P, which characterizes this induced interlayer superconductivity, has a strong temperature dependence at low temperatures. In this case the condition $\lambda(T) = \text{const}$ does not contradict the behavior $H_{c1} \sim \Phi_0/\lambda\xi_P$.

A second interesting aspect of the data on $H_{c1}(T)$ (Figs. 2 and 3) is the pronounced increase in the anisotropy $\eta = H_{c1}^{\parallel c}(0)/H_{c1}^{\perp c}(0)$ with decreasing oxygen content x in $YBa_2Cu_3O_x$: $\eta(x = 6.9) = 4.1$ and $\eta(x = 6.64) = 10.0$. The increase in η is achieved primarily as the result of a sharp decrease in the field $H_{c1}^{\perp c}(0)$, which in $YBa_2Cu_3O_{6.64}$ is 27 ± 2 Oe (Fig. 3). The increase in the anisotropy η suggests a weakening of the binding between conducting Cu–O layers as oxygen leaves the Cu–O chains. As a result, the 1-2-3 compound with $x = 6.64$ is more anisotropic than the composition with $x = 6.9$ in terms of both the lower critical field (Figs. 2 and 3) and the upper one.[16] One might expect that an even greater increase in the critical-field anisotropy would be found as we moved closer to the point of the metal-insulator transition ($x = 6.4$) in the 1-2-3 system.

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