Electrical, magnetic, and structural properties of $YBa_2Cu_3O_x$ single crystals

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The electrical, magnetic, and structural properties of $YBa_2Cu_3O_x$ crystals, which had been previously grown, are studied. These crystals exhibit a clearly defined anisotropy of the Meissner effect and of other magnetic properties. The values of the critical current j_c in the direction parallel to the *ab* plane are calculated.

Single crystals of the high-temperature superconductor $YBa_2Cu_3O_x$ are grown from a solution in a melt at $T \approx 900$–$1000\,^\circ C$. The crystals have a clear lamellar habit and their average size is $1 \times 1 \times 0.030$ mm. X-ray spectral microanalysis and x-ray structural and electron-microscope studies showed that these crystals do in fact have the necessary chemical composition and the crystal structure of an oxygen-deficient perovskite with a unit cell which is tripled along one of the axes and which corresponds to the ordered position of large Y and Ba cations. The electron micrographs of the atomic resolution clearly show double layers of heavy atoms and single layers of light atoms, in accordance with the sequence . . . YBaBaY . . . .

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The single crystals extracted from the melt have a tetragonal symmetry, with the unit-cell parameters $a = 3.863(1)$ and $c = 11.837(2)$ Å, and show no evidence of superconductivity down to liquid-helium temperatures (which was ascertained with the use of contactless rf measurements). Electron-microscope studies show that the structure of tetragonal crystals is highly disordered and defective and that the sequence of the Y and Ba layers is disrupted at approximately 70-Å intervals. The stacking faults, i.e., the sequence errors of the Y and Ba layers, disappear almost entirely after annealing the crystals in oxygen. The crystals acquire an orthorhombic structure with the following unit cell parameters: $a = 3.826(2)$, $b = 3.890(2)$, and $c = 11.705(3)$ Å.

The characteristic feature of the annealed crystals is the existence of a twinning domain structure which is easily seen in the *ab* plane (001) in the electron-microscope studies. The width of the individual domain is approximately equal to 1000 Å.

The splitting of the reflections which has been detected in the electron-diffraction and x-ray structural analyses is also evidence for the existence of an ordered twinning structure with the twinning occurring in the (110) plane. Different splitting multiplicities in the electron diffraction and x-ray structural experiments allow us to assume, however, that the annealed orthorhombic crystals have two identical domain systems which are rotated 90° with respect to each other (a detailed discussion of this subject may be found in the literature; see also Ref. 1).

Orthorhombic crystals annealed in oxygen undergo a sharp ($\Delta T < 0.5$ K) transition to the superconducting state at a temperature of ~ 93 K (see Fig. 1). It should be noted that under an electron microscope the crystals annealed in air are virtually

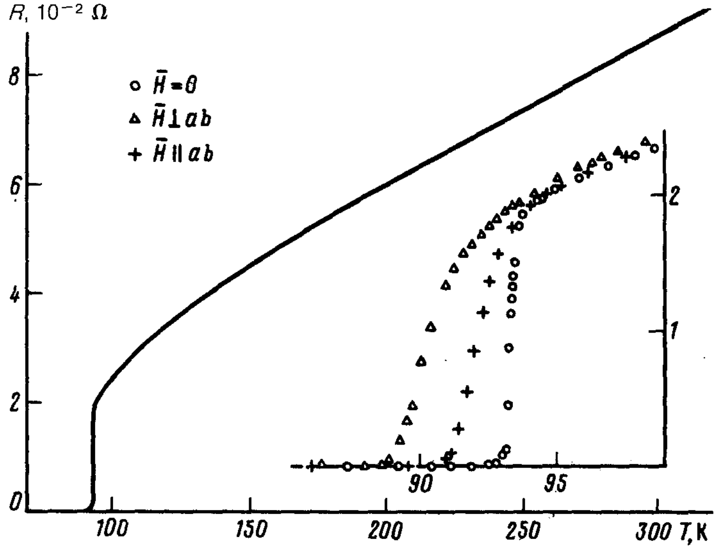

FIG. 1. Temperature dependence of the electrical resistance of a $YBa_2Cu_3O_x$ single crystal in the *ab* plane in a magnetic field $H = 0$; $H \perp ab = 1.1$ T and $H \parallel ab = 1.1$ T.

indistinguishable from the "oxygen" crystals, but they undergo a transition to the superconducting state at considerably lower temperatures ($T_c \approx 50$ K).

Figure 1 shows the results of measurements of the electrical resistance of a single-crystalline $YBa_2Cu_3O_x$ sample in the direction parallel to the *ab* plane, in the absence of a magnetic field and in a field $H = 1.1$ T with $\mathbf{H} \| ab$ and $\mathbf{H} \perp ab$. The measurements were carried out with a four-probe method in a direct current. We used a sample measuring $0.4 \times 0.2 \times 0.03$ mm, which corresponded to a high-quality part of a larger single crystal. Note that the $R(T)$ curve is highly nonlinear at temperatures between T_c and ~200 K. The value $R \approx 2 \times 10^{-2}$ Ω directly above the transition to the superconducting state corresponds to the resistivity $\rho \cong 50$–100 $\mu\Omega \cdot$cm. Although a jump in $R(T)$ near the transition is rather sharp, a small "tail," which extends several tenths of a degree, is seen in the region of the low-temperature transition. Such a behavior of the $R(T)$ curve seems to suggest that low-dimensionality fluctuational phenomena occur over a large temperature interval (paraconductivity). The 1.1-T magnetic field has virtually no effect on T_b (the temperature at which the transition begins), but it shifts far enough the temperature T_c at which the resistivity vanishes completely. In this respect, the action of the magnetic field is strongly anisotropic: with $\mathbf{H} \| ab$, we have $\partial T_c / \partial H \approx 1.9$ K/T and with $\mathbf{H} \perp ab$, we have $\partial T_c / \partial H \approx 3.6$ K/T. It is clear that definite conclusions about the derivatives $\partial H_{c_2} / \partial T$ can hardly be drawn in this case. The factors that give rise to the "diffuseness" of the transition in a magnetic field apparently should first be clarified.

A general-purpose apparatus with a SQUID was used to study the magnetic

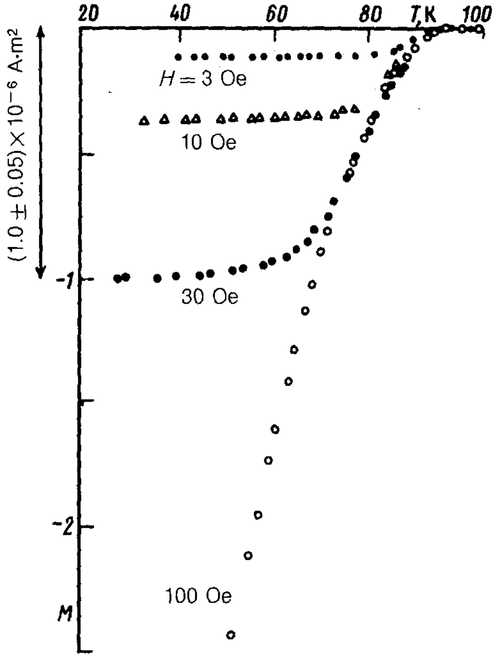

FIG. 2. Temperature dependence of the magnetization M of a $YBa_2Cu_3O_x$ single crystal for various values of the magnetic field H perpendicular to the *ab* plane. The shape of the crystal is that of an almost perfect square, 1 ± 0.1 mm on a side and 0.045 mm average thickness, in the *ab* plane.

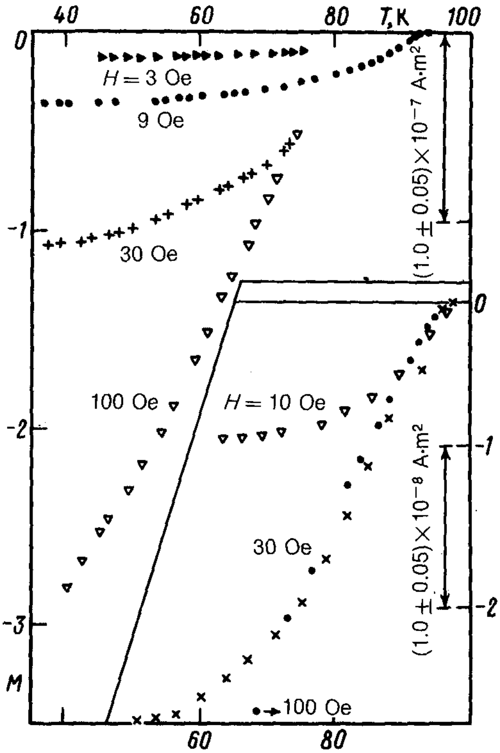

FIG. 3. The same as in Fig. 2, but for a field H parallel to the ab plane. (Inset—the same plot for a crystal with a shape resembling a half-disk of average thickness 0.050 mm and 0.45 mm in diameter.)

properties of crystals.[2] Figures 2 and 3 show typical families of the temperature dependences of the magnetic moment M of a single crystal for two orientations of a fixed external magnetic field H. To obtain each dependence, the sample was first cooled from $T \sim 100$ K to $T \sim 10$ K in a low residual field $H_0 \cong 0.1$ H and after the application of the field H it was slowly warmed, with the detection of the moment.

The low-temperature magnetization plateau was found, within absolute determination of M (about $\pm 5\%$), to correspond to the total field screening in the bulk of the sample. An increase of the temperature in the field $\mathbf{H} \perp ab$ (Fig. 2) causes all the curves to approach the universal curve $M_c(T)$, which is nearly independent of H. Such a behavior is consistent with the model which describes strong pinning of the Abrikosov vortices at the unsaturated centers, where the density modulus of the critical current j_c in the critical state does not depend on the local magnetic induction and hence on the coordinates (see, e.g., Ref. 3). Under this assumption the curve in Fig. 2 gives the directly reduced dependence $j_c(T)$. For a square sample with dimensions $L \times L \times d$ a straightforward calculation gives the equation $M_c(T) = (L^3 d/24) j_c(T)$. For our sample [$L = (1.0 \pm 0.1)$ mm, $d_{\mathrm{av}} = 45\ \mu\mathrm{m}$] this equation gives for $T = 50$ K the value $j_c = (1.0 \pm 0.3) \times 10^5$ A/cm^2 and for $T = 77$ K it gives the value $j_c = (5 \pm 2) \times 10^4$ A/cm^2 (for $j \| ab$, $\mathbf{H} \perp ab$, $3\ \mathrm{Oe} \leqslant H \lesssim 100\ \mathrm{Oe}$). The first of these values is approximately equal to the value measured for the domain-free single crystals at the same temperature.[4]

With fields $\mathbf{H}\perp ab$ (Fig. 3), the curves $M(T)$ corresponding to various H cross each other, indicating that j_c decreases markedly with increasing H, typical of a weaker pinning.[3] Although this behavior makes it impossible to accurately calculate the critical current density from our data, at $T=50$ K it is estimated to be less than 0.3×10^5 A/cm^2 ($\mathbf{j}\|ab$, $\mathbf{H}\|ab$, $\mathbf{j}\perp\mathbf{H}$), which is in fact much lower than the value found for $H\perp ab$.

The existence of pinning anisotropy and its nature can also be confirmed by measuring the $M(T)$ curves in a constant field while lowering the temperature, beginning with $T>T_c$: when $\mathbf{H}\|ab$, the Meissner effect amounts to approximately 25% of the total effect and when $\mathbf{H}\perp ab$, the Meissner effect amounts to $\sim3\%$ (both figures are for $T\ll T_c$ and $H\approx10$ Oe). According to the data of Ref. 4, the domain-free single crystals have an inverse Meissner-effect-induced anisotropy (this is, in our view, inconsistent with the anisotropy of j_c). In our crystals with a domain structure, the strong pinning anisotropy apparently is attributable to the fact that the domain walls serve as effective pinning centers for vortices parallel to the c axis.

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