Study of the conductivity of $YBa_2Cu_3O_{7-x}$ single crystals in a wide frequency range

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The resistivity of $YBa_2Cu_3O_{7-x}$ crystals was measured by the four-contact method using direct current and by the contactless method in the megahertz and gigahertz ranges. The results of the contactless measurements suggest that the crystals have a macroscopic inhomogeneity along the direction of the c axis. The $\rho_\parallel(T)$ plot was found to be independent of the frequency.

Several experimental studies, in which the temperature dependence of the resistivity of $YBa_2Cu_3O_{7-x}$ single crystals was measured, have recently been published (see, e.g., Refs. 1–4). The results of these measurements, which were carried out with samples grown in different laboratories, differ in absolute value and in the temperature dependence.

According to the information found in the literature, the largest single crystals of $YBa_2Cu_3O_{7-x}$ grown to date are thin wafers with characteristic dimensions $1\times1\times0.05$ mm, whose plane is perpendicular to the major axis of the crystal c. The small size of the samples and the appreciable detectable anisotropy of the conductivity along the direction of the wafer plane and perpendicular to it, which increases with decreasing temperature [$\rho_\perp/\rho_\parallel \sim 10^2 - 10^3$ at $T \gtrsim T_c$ (Refs. 1 and 4)], lead us to believe that the technique used to deposit the contacts has a considerable effect on the measurement results. Since the surface of single crystals is usually covered with a poorly conducting layer, the measuring contacts are either sputtered on or brazed with a conducting paste. In each case the electrode metal diffuses into the crystals. The other known methods of depositing contacts do not allow us to obtain a contact resistance low enough to carry out measurements with confidence.

We thought that a study of the temperature dependence of $\rho_{\parallel}$ using the contact and contactless methods and a comparison of the results obtained by these methods would be of interest. The single crystals which we have studied were grown at the Institute of Crystallography, Academy of Sciences of the USSR, by a method described elsewhere.[4] The measurements were carried out using single crystals from two different batches. The structural and resistive properties of the single crystals from these two batches, which were measured by the contact method, were described in Ref. 4. The top-grade single crystals (lot A) were found to have a lower anisotropy at $T \sim T_c (\rho_\perp/\rho_\parallel \sim 100)$ and $\rho_\parallel$ (300 K)/$\rho_\parallel$ (100 K) ≈ 4. The low-grade single crystals (lot B) were found to have $\rho_\perp/\rho_\parallel \sim 700$ at $T \sim T_c$ and $\rho_\parallel$ (300 K)/$\rho_\parallel$ (100 K) ≈ 4.

1. Figure 1 shows the $\rho_\parallel (T)$ curves for two single crystals from lot A, which were annealed in oxygen under the same conditions but with contacts fabricated by different methods. The dc measurements were carried out using the four-contact method. The principal difference between these curves is the strong nonlinearity at $T \leqslant 150$–180 K of the curve corresponding to the brazed silver-paste contact. As a result, an extrapolation of $\rho_\parallel (T)$ to $T \to 0$ shows that the sputtered gold contacts have a residual resistance and in the case of brazed contacts the extrapolated $\rho_\parallel (T)$ curve crosses the abscissa. Similar results were obtained using several crystals. In all of the cases $\rho_\parallel$ (300 K) obtained with the brazed contacts was smaller by several factors than that obtained with the sputtered contacts and had a value of $\sim 200 \mu\Omega \cdot$cm.

In the contactless measurements we determined the Q of the LC circuit in the megahertz range. The natural frequency of this circuit was ~ 10 MHz. During the measurement the test sample was inserted on a moveable quartz holder into an induction coil of the circuit. This procedure allowed us to measure the Q of the circuit without a load Q_0 and with a load Q_l at each temperature. Knowing these values, we could determine the losses associated with the current in the crystal. The depth of the skin layer at the frequency ~ 10 MHz is much greater than the thickness of the crystal, so that the losses correspond to the resistance averaged over its thickness.

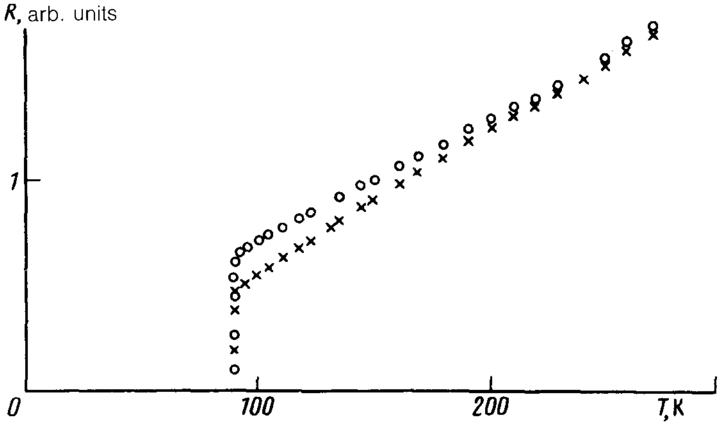

FIG. 1. Typical $\rho_\parallel (T)$ plots, of the "best" low-resistance samples, measured by the four-contact method. O—Gold-deposited contacts; ×—brazed silver paste contacts.

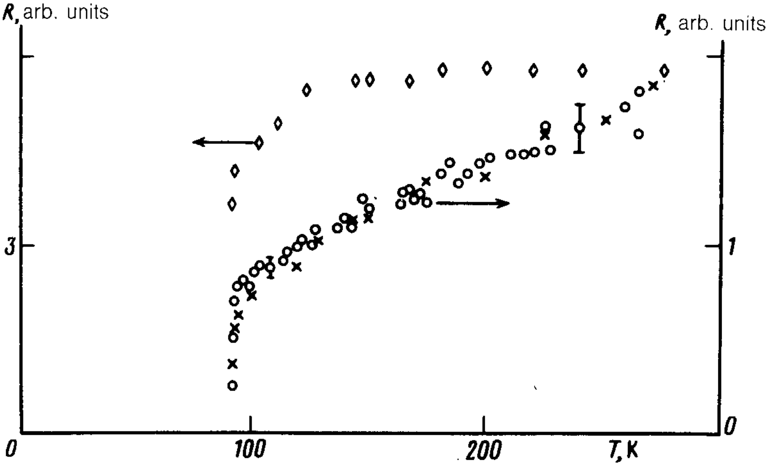

FIG. 2. Typical $\rho_\omega(T)$ plots of the "best" low-resistance samples (○, ×) and of the "worst" higher-resistance samples (◇). ○—$\omega/2\pi = 18$ Mz, ×—$\omega/2\pi = 4$ MHz; ◇—$\omega/2\pi = 18$ MHz.

Figure 2 shows typical $\rho(T)$ curves which were measured at two frequencies $\omega/2\pi = 4$ and 18 MHz. We see that ρ_ω increases linearly with the temperature. The use of this method yielded ~100% absolute measurement accuracy and ~10% relative measurement accuracy for $\rho_\parallel$. A frequency dependence of ρ was not observed within the experimental error, the absolute values of $\rho_\parallel(300)$ measured by the contactless method were 100–200 $\mu\Omega\cdot$cm. A divergence from a linear dependence was much smaller than in the case of the brazed contacts and it manifested itself at lower temperatures ($\leqslant 110$ K).

The largest discrepancy in the values of $\rho_\parallel(T)$ measured by different methods was obtained for low-grade single crystals (lot *B*): the value of $\rho_\parallel$ which was measured by the contactless method, in contrast with that reported in Ref. 4, is constant within 20% over the temperature range 110–300 K. The reason for this discrepancy will likely be cleared up when larger crystals are grown for correct measurements by the contact method and also for measurements by the contactless method over a wider frequency range.

2. Let us consider the anisotropy of the resistivity $\rho_\perp/\rho_\parallel$, which increases markedly as T_c is approached.[4] In addition, two superconducting transition temperatures seem to have been observed by Makarenko and Nikiforov[4]: one temperature, the higher of the two, was observed when $\rho_\parallel$ was measured and the other, lower by 1.5 K, was observed when $\rho_\perp$ was measured. Such a behavior of $\rho_\perp/\rho_\parallel$ may stem from the inhomogeneity of the crystal along the direction of the *c* axis, which seems perfectly natural bearing in mind the layered structure of $YBa_2Cu_3O_{7-x}$ and the appreciable difference in the growth rates in the direction of the *c* axis and at right angles to it. To resolve this problem, we built an *LC* oscillator which we used to measure the single crystal by inserting it into the induction coil and then removing it from the coil. At $T \leqslant T_c$ the oscillator frequency increased as a result of insertion of the crystal, making it possible

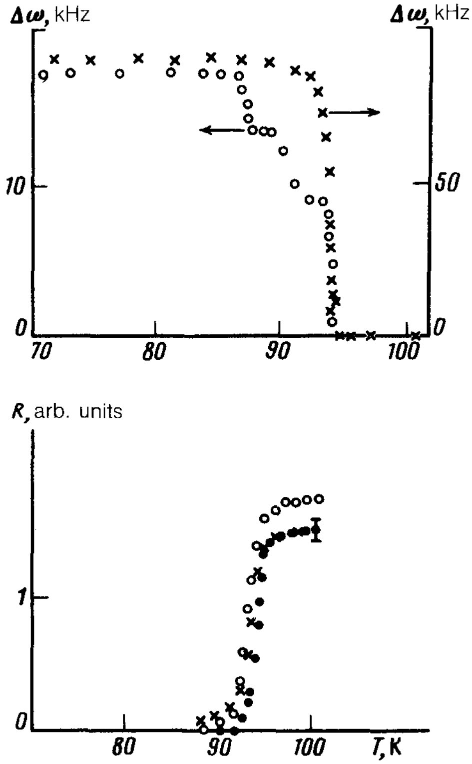

FIG. 3. (a) Resonance frequency shift of the circuit with the sample $\Delta\omega(T)$, $\omega_0/2\pi = 10$ MHz. O—h_{rf} in the plane perpendicular to the c axis; ×—h_{rf} in the plane parallel to the c axis. (b) The $R(T)$ dependence; ●—$\omega/2\pi = 26.3$ GHz, $H = 0$; ×—$\omega/2\pi = 26.3$ GHz, $H = 13$ kOe; O—$\omega/2\pi = 36$ GHz, $H = 0$.

to detect the superconducting transition temperature from the frequency shift. The oscillator was stable to within $\sim 10^4$, allowing us to measure the transition temperature in the plane of the crystal and across the layers. To asure that the sample filled the induction coil adequately, the coil was wound on two thin mica wafers between which the crystal could slide. Figure 3 is a plot of the temperature dependence of the oscillator's frequency shift for two orientations of the crystal relative to the coils. Curve *a* corresponds to the case in which the current flows in the plane of the crystal and curve *b* corresponds to the current flow across the crystal. Curve *b* shows a steplike frequency shift, which seems to indicate the presence of macroscopic layers with various T_c. All test samples were found to have a diffuse transition whose width changed from one sample to another. The measurements were conducted in the geometry corresponding to curve *b*. The transition, measured in the geometry corresponding to curve *a*, was not nearly as broad in all crystals (~ 1 K).

3. The impedance of a $YBa_2Cu_3O_{7-x}$ ceramic measured previously at microwave frequencies revealed the presence of an absorption tail which extended far into the low-temperature region.[5] We have measured the temperature dependence of the resistive component of the impedance of $YBa_2Cu_3O_{7-x}$ single crystals at frequencies of 25 and 36 GHz (Fig. 3b). The screening current flowed in a plane perpendicular to the c axis. We see that the resistance of a single crystal, in contrast with a ceramic, decreases near T_c over a fairly narrow temperature interval.

The shape of the $R(T)$ curve measured upon application of a static field H in a plane perpendicular to the c axis is the same as the shape of the curve obtained from direct-current measurements.[3] The result which we have obtained apparently indicates that the diffuseness of the superconducting transition, determined from the measurement of the microwave impedance of the ceramic, is attributable to the inhomogeneity of the crystals that make up the ceramic.

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