Bulk nature of the superconductivity of $YBa_2Cu_3O_x$ single crystals

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Resistive and magnetic measurements were carried out using $YBa_2Cu_3O_x$ single crystals which were grown from a melt and which undergo a superconducting transition at 80–90 K. The Meissner effect showed that these single crystals have a bulk superconductivity.

The answers to many questions concerning the nature and mechanism of superconductivity of the new high-temperature superconductors of the type Y-Ba-Cu-O or La-Ba-Cu-O cannot be found without the study of single-crystals. This study would consider, in particular, the question of whether the superconductivity of these compounds is a bulk superconductivity or whether it is traceable, as was asserted by Oda *et al.*,[1] for example, to the interfaces between the crystallites in the polycrystalline samples.

In the present letter we report the results of measurements of the Meissner effect in $YBa_2Cu_3O_x$ single crystals synthesized by us. The results of these measurements show that the single crystals have a resistive superconducting transition of width 2–5 K with T_c near 90 K. The single crystals were grown from a nonstoichiometric melt of a mixture of Y_2O_3, BaO, and CuO oxides by slow cooling in air. The crystals are square-shaped black wafers with mirror-smooth surfaces up to 2 mm wide and 30–50 μm thick.

X-ray-diffraction study of these crystals (based on rotating-crystal x-ray photographs and Weissenberg rotating-crystal photographs) showed that they are single crystals of the $YBa_2Cu_3O_x$ phase, which was detected previously in superconducting ceramic samples,[2] and that they have an orthorhombic (pseudotetragonal) lattice with the lattice constants $a \approx b = 3.85$ Å and $c = 11.68$ Å. According to the data from x-ray spectral analysis, the yttrium, barium, and copper content in the single crystals was also found to be 1:2:3. All crystals studied by x-ray diffraction were found to have twins.

The resistive properties of the single crystals were studied by a standard four-contact method with *dc* current, using platinum wires 10 μm in diameter cemented with a graphite paste. The resistance of the contacts was reduced no lower than ≈ 100 Ω apparently because of the traces of the dielectric film on the surface of the samples. At 300 K the resistivity is 400–600 $\mu\Omega\cdot$cm in any direction in the *ab* plane.

The temperature dependence of the resistivity normalized to 300 K near the superconducting transition is shown in Fig. 1. The temperatures indicating the beginning of the transition, T_b, are 83, 88, and 90 K, respectively, for samples 1, 2 and 3.

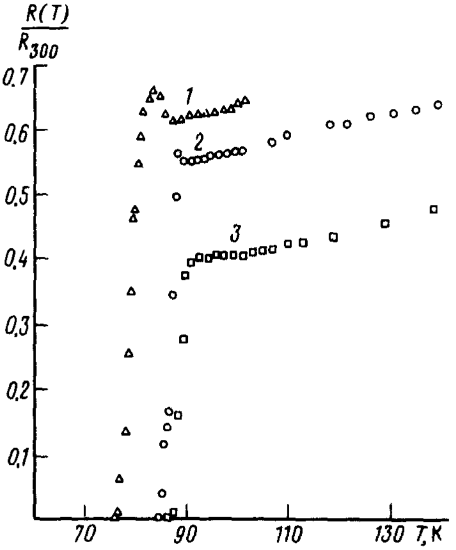

FIG. 1. Temperature dependence of the resistivity of three $YBa_2Cu_3O_x$ single crystals near the superconducting transition.

The ratio R_{300}/R_{T_b}, which characterizes the temperature decrease of the resistivity at the time the superconducting transition begins, may be used as a measure of the quality of the single crystals. In Fig. 1 we see as the ratio R_{300}/R_{T_b} is increased, the point at which the transition begins shifts toward higher temperatures, while the transition itself narrows its range. For sample 3 with $R_{300}/R_{T_b} = 2.5$ we see that T_b reaches ≈ 90 K and that the total transition width is ≈ 2 K.

We notice that with decreasing R_{300}/R_{T_b}, there is an increase in the resistivity which precedes the superconducting transition. With the degradation of the quality of the crystals, the time at which the resistivity begins to increase is shifted, as is the superconducting transition itself, toward the low temperatures. It can therefore be assumed that here we are dealing with a pretransitional phenomenon which may possibly be related to the redistribution of the current paths in the strongly anisotropic or defective crystals.

To estimate the part of the superconducting phase contained in the crystals under study, we measured the temperature dependence of the static magnetization of these crystals in the *ab* plane using a SQUID magnetometer. Figure 2a shows the results of diamagnetic screening for sample 3. In these experiments the magnetic field was applied at a temperature $T \approx 10$ K and the change in the magnetic moment of the sample was detected during its warming to $T \approx 100$ K. At $T = 7.2$ K the amplitude of the magnetic-moment variation was nearly equal to the magnitude of the magnetization of a specially prepared lead reference. Here the total change in the susceptibility as a result of diamagnetic screening is denoted by χ_d.

The results obtained from measurements of the Meissner effect in the same sample are shown in Fig. 2b. In these experiments the samples at a temperature above T_b were cooled in the presence of a magnetic field. We see that in small magnetic fields

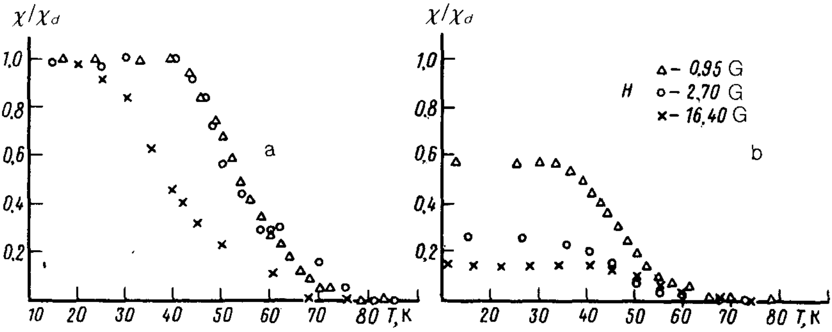

FIG. 2. Temperature dependence of the reduced magnetic susceptibility of sample 3 for (a) diamagnetic screening and (b) the Meissner effect.

the ratio of the signals for the two indicated types of experiments is greater than 50%.

In the case of screening, the diamagnetic moment is proportional to the magnetic field H to within $H \approx 40$ G. At the same time, the diamagnetic moment measured in the presence of the Meissner effect changes by only 20–25% when the applied field is increased form 1 G to 16 G. This change is equivalent to a decrease in the fraction of the forced-out magnetic flux from 55% to 14%. Such a behavior can be attributed to flux pinning on twinning boundaries and on other possible structural defects. Structural imperfections and anisotropy of the superconducting properties are also most likely responsible for the considerable broadening of the superconducting transition, which can be measured from the magnetic moment, and for its appreciable displacement in small magnetic fields.

On the other hand, the results of magnetic measurements using all the test samples show that the magnetic field can be forced out from a large part of the sample by lowering the temperature below T_c. This fact indisputably shows that the superconductivity of $YBa_2Cu_3O_x$ single crystals studied by us is a bulk superconductivity. As for the possible effect of the twinning boundaries and other defects of the crystal structure on the particular features of the superconductivity and the magnetic behavior of the single crystals studied, we note that this topic requires further study.

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[1] Y. Oda, I. Nakada, and K. Asayama, Jpn. Journ. Appl. Phys. **26**, L608 (1987).
[2] K. Semba, S. Tsurumi, M. Hikita, *et al.*, Jpn. Journ. Appl. Phys. **26**, L429 (1987).

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