Magnetic properties of $YBa_2Cu_3O_x$ single crystals in weak magnetic fields

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The magnetic moment of $YBa_2Cu_3O_x$ single crystals has been measured at temperatures higher than the superconducting transition temperature T_c. Above $T_c \approx 93$ K the magnetic susceptibility of single crystals was found to have a Pauli nature, i.e., it is temperature independent and isotropic. The numerical values of the susceptibility lie within the limits $(5–9) \times 10^{-5}$ structural change units.

In this letter we are reporting the results of an experimental study of the magnetic properties of $YBa_2Cu_3O_x$ single crystals. Attention is focused principally on the temperature region above the superconducting transition temperature T_c, since similar studies previously were conducted only with polycrystalline samples (see, e.g., Refs. 1–4).

The magnetic moment was measured by means of a SQUID magnetometer[5] in 100-Oe magnetic fields. The absolute error in measuring the magnetic moment was ~10%. The noise level was within 2×10^{-11} A·m². Since the individual crystals were too small for reliable measurements at $T > T_c$, we used stacks of single crystals with a total mass of 1.5–2 mg, oriented in the same direction.

We studied crystals from two lots taken from the same batch. The first lot of samples included crystals annealed in flowing oxygen ($T_c \approx 92–93$ K, $\rho_{\perp c} \approx 250$ $\mu\Omega \cdot$ cm at 300 K). The crystals from the second lot were not treated thermally or chemically after being grown ($T_c = 50–60$ K, $\rho_{\perp c} \approx$ 10 m$\Omega \cdot$ cm at 300 K). The samples from lot I were orthorhombic twinned crystals with the unit cell parameters $a = 3.823$, $b = 3.879$, and $c = 11.7–11.735$ Å. Lott II contained tetragonal crystals with $a = 3.857$ Å and $c = 11.828$ Å.

Because of the nature of these experiments, we have first studied the magnetization curves of the test crystals at temperatures below T_c. In these studies we used one single crystal from lot I (Fig. 1). As can be seen from a comparison of the corresponding magnetization curves measured in two regimes (cooling in a 10-Oe field and cooling in a virtually zero field), the Meissner effect in the sample under study is appreciable, reaching ~30% with $\mathbf{H} \perp \mathbf{c}$ and ~50% with $\mathbf{H} \| \mathbf{c}$[2]) in a 10-Oe field. This result is in qualitative agreement with the data of Refs. 2 and 7, although the specific values characterizing the fraction of the magnetic flux that is forced out, which we obtained experimentally, are much higher than those obtained in Refs. 2 and 7. This result apparently suggests that our samples are of a higher quality.

Figure 2 is a plot of the magnetic susceptibility $\varkappa_V$, calculated from measurements of the magnetic moment M, for three samples of lot I, which were annealed indepen-

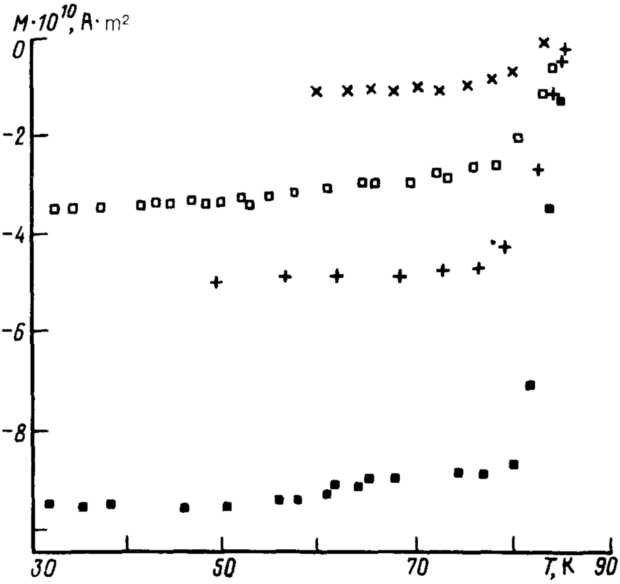

FIG. 1. Magnetization of $YBa_2Cu_3O_x$ single crystals versus the temperature for the orientation of the magnetizing field $H = 10$ Oe parallel (■, +) to the crystallographic c axis of the samples and perpendicular (□, ×) to it. ■,□—After the crystals are cooled in a field $H' < 0.1\ H$ ("shielding"); +, ×—the same situation with $H' = H = 10$ Oe (the Meissner effect). The samples are ~100×100×20 μm in size.

dently. The measurements of M were carried out in a field of 100 ± 1 Oe, with the samples oriented in two directions: **H**⊥**c** and **H**∥**c**. We see that the $\varkappa_V(T)$ curve is of a Pauli nature: there is no anisotropy or temperature dependence of $\varkappa_V$ within the error of the experiment. The numerical values of the magnetic susceptibility $(9 \pm 1) \times 10^{-5}$, $(5 \pm 1) \times 10^{-5}$, and $(7 \pm 2) \times 10^{-5}$ (expressed in structural change units) (these val-

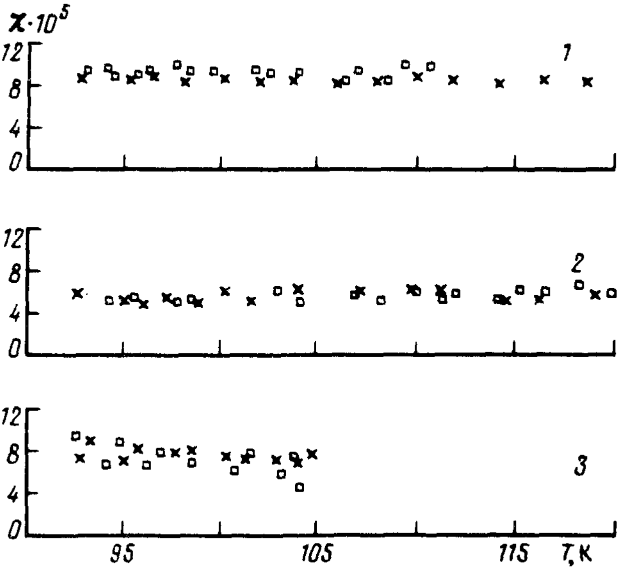

FIG. 2. Bulk magnetic susceptibility of three different sets of annealed single crystals of mass 1.5 mg (1), 2.1 mg (2), and 1.4 mg (3) in a 100-Oe magnetizing field with the field orientation **H**∥**c** (■) and **H**⊥**c**(×).

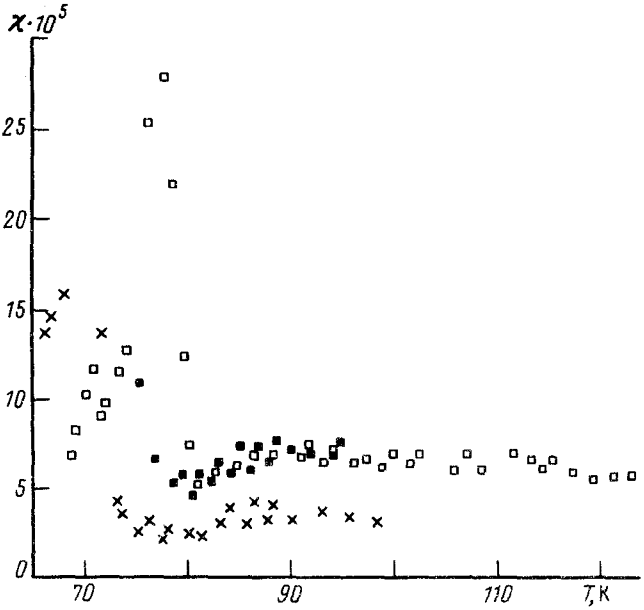

FIG. 3. Bulk magnetic susceptibility of two sets of unannealed single crystals of mass 3.15 mg (□, ×) and 1.6 mg (■). ×—H‖c; □,■—H⊥c.

ues conform to the labels on the samples; see Fig. 2) differ slightly from sample to sample. This difference is difficult to explain solely in terms of the experimental error. The maximum error obtained in calculating $\varkappa$, which stems from the error in determining the volume of the sample, amounts to 15–20%.

We should point out that the absence of a temperature dependence of the susceptibility is also characteristic of polycrystalline single-phase samples.[2,3] The numerical values of the susceptibility obtained in Refs. 2 and 3 are in general agreement with our results, although they are lower, on the average: $(3.4–7)\times 10^{-5}$.

Figure 3 is a plot of the magnetic susceptibility as a function of temperature for samples that were not annealed (lot II). We see that the temperature range of the $\varkappa_V(T)$ dependence in this case is clearly divided into two regions. At temperatures above 75–80 K, $\varkappa_V$ does not depend on the temperature, as in the case of the annealed samples, but it does exhibit a pronounced anisotropy. At lower temperatures we see a clear but slightly irregular increase in the susceptibility to the superconducting transition temperature. Such a behavior of the magnetic susceptibility of $YBa_2Cu_3O_x$ crystals which are not annealed and which are therefore defective can apparently be described in terms of partially localized spins and in terms of the trapping of carriers at defects.

Turning again to the magnetic susceptibility of the samples of lot I with $T_c \approx 93$ K, we emphasize that the appreciable differences in $\varkappa$ for the three test samples (see Fig. 2) apparently are not a consequence of the different densities of the electronic states, since the values of T_c of these samples are virtually the same. A very simple explanation of this result could be predicated on the random paramagnetic impurities. The most reliable value for single crystals of the given type should then be the smallest value of those that were obtained ($\sim 5\times 10^{-5}$, expressed in structural change units, or $\sim 3.9\times 10^{-6}$, in cgs units).

The electron state density $N(\epsilon_f) = 19\ \mathrm{eV}^{-1}$ per unit cell, calculated using the latter value with allowance for the diamagnetic contribution of ions, even in this case is too large in comparison with the "band"-calculation data.[8]

This circumstance can apparently be used as evidence in support of the considerable "Stoner" intensification of the paramagnetic susceptibility of $YBa_2Cu_3O_x$ (Ref. 3), although it should be kept in mind that the results of theoretical calculations of the state density have a large variance (see Ref. 8).

If, on the other hand, the superconducting transition temperature still turns out to be insensitive to the state density $N(\epsilon_f)$, then a special group of excitations, possibly of the Bose type, which do not contribute to the paramagnetic susceptibility, would have to be introduced in order to explain the superconducting state.

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2) We should point out an error in Ref. 6. With **H**||**c**, the Meissner effect was in fact measured in a 100-Oe field.

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[1] A. Junod, A. Bezinge, T. Graf *et al.*, Europhys. Lett. **4**, 247 (1987).
[2] T. R. McGuire, T. R. Dinger, P. J. P. Freitas *et al.*, Phys. Rev. **B36**, 4032 (1987).
[3] S. W. Cheong, S. E. Brown, Z. Fish *et al.*, Phys. Rev. **B36**, 3913 (1987).
[4] R. J. Cava, B. Batlogg, C. H. Chen *et al.*, Nature **329**, 423 (1987).
[5] L. Z. Avdeev, O. V. Snigirev, and V. V. Khanin, IEEE Trans. Magn. **21**, 914 (1985).
[6] L. Z. Avdeev, A. B. Bykov, L. N. Dem'yanets *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **46**, 196 (1987) [JETP Lett. **46**, 249 (1987)].
[7] T. R. Dinger, T. K. Worthington, W. J. Gallagher, and R. L. Sandstrom, Phys. Rev. Lett. **58**, 2687 (1987).
[8] F. Herman, R. V. Kasowski, and W. Y. Hsu, Phys. Rev. **B36**, 6904 (1987).
[9] S. Massidda, Yu. J. Freeman, and D. D. Koelling, Phys. Lett. **A122**, 198 (1987).

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