Magnetic properties of $YBa_2Cu_3O_x$ single crystals in the normal state

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The magnetic susceptibility χ of single-crystal $YBa_2Cu_3O_x$ samples with various oxygen contents has been measured in the normal state. At $x<7$ the magnetic susceptibility is anisotropic: $\chi_{\parallel c}/\chi_{\perp c}<1$. As $x\to 7$, the ratio $\chi_{\parallel c}/\chi_{\perp c}$ approaches unity. In a magnetic field in the orientation $H\parallel c$, an anomaly is observed near T_c which is apparently related to anisotropic effects of a fluctuation diamagnetism.

In a previous study[1] of the magnetic properties of $YBa_2Cu_3O_x$ single crystals, it was stated that their magnetic susceptibility χ in the normal state is isotropic and essentially independent of the temperature. Several subsequent studies, however, have revealed a fairly pronounced anisotropy of the normal-state magnetic properties of single crystals and textured samples of $YBa_2Cu_3O_x$ (Refs. 2 and 3) as well as other compounds in the group of high-T_c superconductors.[4] This contradiction led us to carry out a new study of the magnetic susceptibility of $YBa_2Cu_3O_x$ single crystals, including some with an oxygen content $x<7$.

As before,[1] the magnetic moment of the samples was measured with a SQUID magnetometer[5] in a magnetic field of 100 Oe. The absolute error in the determination of this moment was less than 10^{-8} cgs units. The test samples were stacks of single crystals with a total volume $\sim 0.4\ \text{mm}^3$, in identical orientations with respect to the c axis. Single crystals grown in two lots were studied. After being removed from the crucible, the crystals were annealed in flowing oxygen and then in helium. The oxygen content in the samples was estimated by x-ray measurements.[6]

The relative error in the determination of χ within each series of measurements was $\sim 3\%$. The absolute values of χ, on the other hand, could be determined only within an error $\sim 20\%$, because of the error in the determination of the volume of the samples. For convenience in comparing the experimental results (Figs. 1–4), the calculated values of χ for the different samples have been normalized to a common value at $x=7$.

The samples used in the present experiments were slightly lower in quality than those studied in Ref. 1. The width of the superconducting transition, ΔT, in the samples with the nominal composition $YBa_2Cu_3O_{7.0}$ used in the present study was $\simeq 1$ K, in comparison with the width of 0.2–0.5 K for the samples of Ref. 1, according to resistive measurements.

Figure 1 shows the experimental results. Shown here are examples of the behavior of the magnetic susceptibility of samples with the compositions $YBa_2Cu_3O_{7.0}$ and

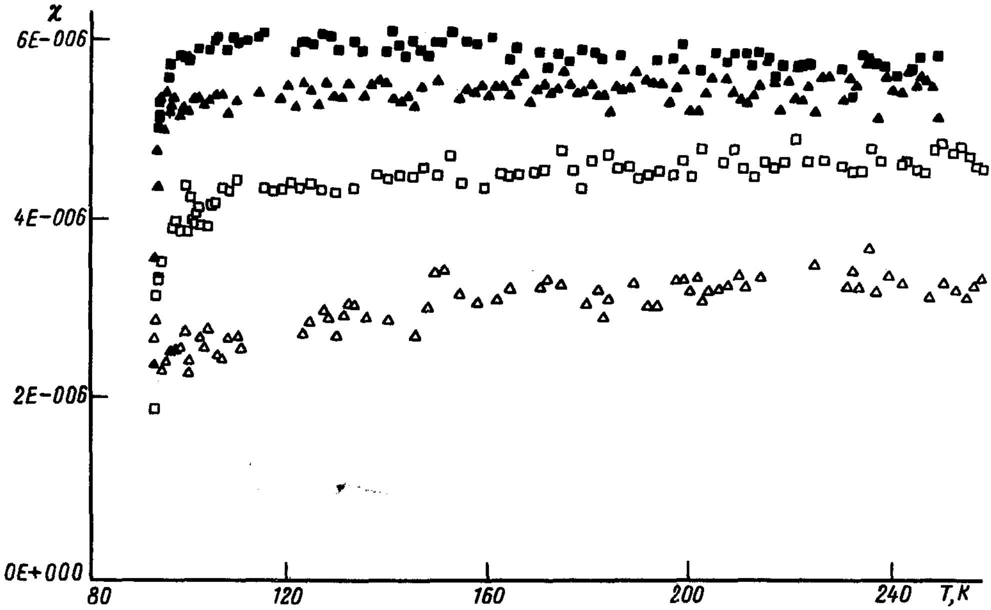

FIG. 1. Bulk magnetic susceptibility χ (cgs units) of single-crystal $YBa_2Cu_3O_{7.0}$ samples (filled points) and $YBa_2Cu_3O_{6.87}$ samples (open points) for the magnetic-field orientations $H \| c$ (squares) and $H \perp c$ (triangles).

$YBa_2Cu_3O_{6.87}$ for two orientations of the magnetic field, $H \| c$ and $H \perp c$. We see that the susceptibility of $YBa_2Cu_3O_x$ is generally anisotropic, with $\chi_{\|} > \chi_{\perp}$. However, we find $\chi_{\|}/\chi_{\perp} \to 1$ as $x \to 7$ (Figs. 2 and 3).

The weak temperature dependence observed for the susceptibility is positive

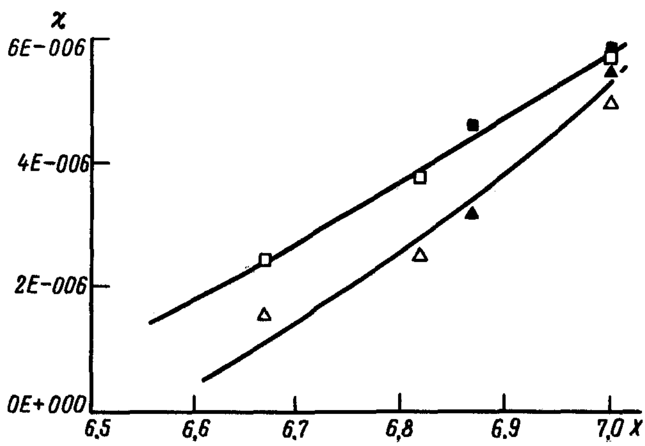

FIG. 2. Average values (over the range $T = 120$–260 K) of the bulk magnetic susceptibility of $YBa_2Cu_3O_x$ and dependence on the oxygen content x. Squares—$H \| c$; triangles—$H \perp c$. Filled points) Measurement series 1; open points) series 2.

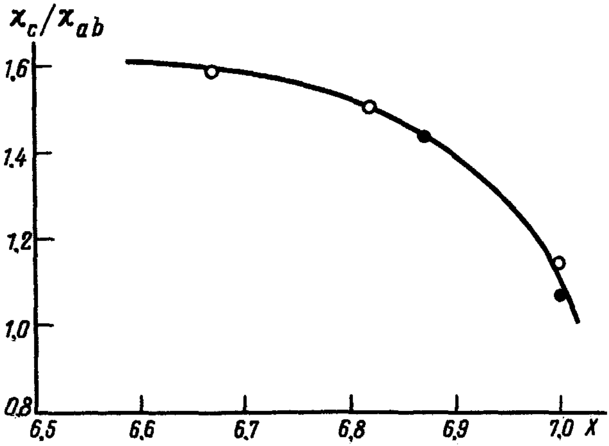

FIG. 3. Ratio of the average values (over the range $T = 120\text{–}260$ K) of the magnetic susceptibility $\chi_{\parallel}/\chi_{\perp}$ for $YBa_2Cu_3O_x$ (Fig. 2) versus the oxygen content x. Filled points—Measurement series 1; open points—series 2.

($d_\chi/dT > 0$) for both directions of the magnetic field H and for all compositions except $x = 7$ in the orientation $H \| c$, in which case we find $d_\chi/dT < 0$ (Fig. 4).

Near T_c, we see a clearly defined diamagnetic anomaly in the orientation $H \| c$ (Figs. 1 and 5). Since the derivative d_χ/dT is small, in discussing the concentration dependence of the susceptibility of $YBa_2Cu_3O_x$ we can work with values of χ averaged over the temperature range 120–260 K. It follows from Fig. 2 that the susceptibility χ decreases with decreasing oxygen content. Since the carrier density also falls

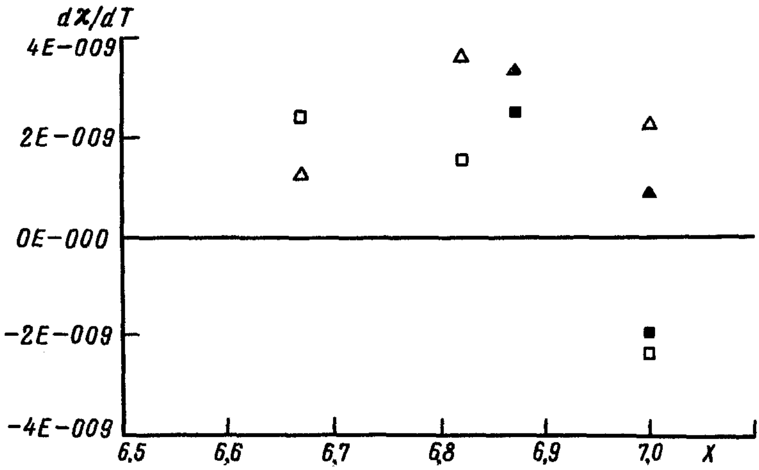

FIG. 4. Average value (over the range $T = 120\text{–}260$ K) of the slope of the $\chi(T)$ curves versus the oxygen content x. The notation is the same as in Fig. 2.

off with decreasing x, one might suggest that the Pauli component of the susceptibility of $YBa_2Cu_3O_x$ is predominant. On the other hand, the anisotropy observed in χ requires consideration of other components in the magnetic response of the system. The results of neutron studies (Ref. 7, for example) suggest that the anisotropy of χ is associated with in-plane spin correlations of an antiferromagnetic nature which vanish in the limit $x = 7$. The nature of the anisotropy in the susceptibility (i.e., $\chi_{\|} > \chi_{\perp}$) and its temperature dependence d_χ/dT agree with this picture. This conclusion does not rule out the possibility that in inhomogeneous samples one might observe effects associated with spin correlations even at a nominal oxygen concentration $x \approx 7$. The data of the present study apparently do not contradict the assertion in Ref. 1 that the susceptibility is isotropic for the composition $YBa_2Cu_3O_7$.

We turn now to the anomaly in χ near the superconducting transition temperature T_c, which is seen as a rapid decrease in $\chi(T)$ as $T \to T_c$. As we mentioned earlier, this anomaly is observed in the orientation $H \| c$. It is natural to link it with effects of a fluctuation diamagnetism, of a highly anisotropic or quasi-two-dimensional nature.

Figure 5 shows an attempt to distinguish the fluctuation component of the magnetic susceptibility from the experimental data for the sample with $x \simeq 7$ in the orientation $H \| c$. For this purpose, the set of experimental points was approximated by the expression[8]

$$\chi = \chi_{fl} + a + bT, \tag{1}$$

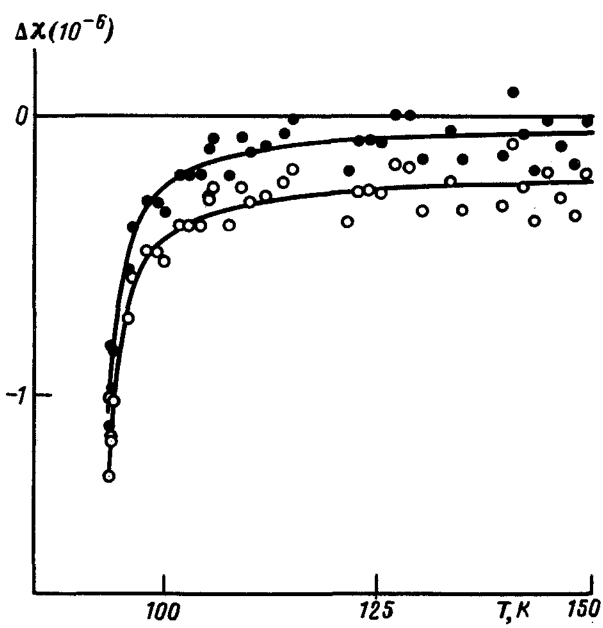

FIG. 5. Fluctuation part of the magnetic susceptibility, χ_{fl}, of a $YBa_2Cu_3O_7$ sample for $H \| c$. ○—Three-dimensional case; ●—quasi-two-dimensional case (see the text proper).

where

$$\chi_{fl} = A\left(\frac{T_c}{T - T_c}\right)^{1/2} \tag{2}$$

for the anisotropic $3D$ case or

$$\chi_{fl} = B\left(\frac{T_c}{T - T_c}\right) \tag{3}$$

for the quasi-$2D$ case. Here $A = -(1/6)\pi k_B T \Phi_0^{-2} \xi(0)(M/m)^{1/2}$, $B = -(-1/3)\pi k_B T \Phi_0^{-2} \xi^2(0)/s$, Φ_0 is the magnetic flux quantum, $\xi(0)$ is the coherence length at $T = 0$, M/m is the ratio of the masses of electron pairs for motion across and along the layers, and the numerical parameter s has the dimensionality of a length and is equal in order of magnitude to the distance between layers.

According to Fig. 5, each of these expressions can be said to agree with the experimental data, because of the large scatter in the latter. However, the assumption that the fluctuations are of a $3D$ nature leads to an extremely large fluctuation region (Fig. 5). Moreover, we should not ignore our direct observation that the anomaly is noticeable only in the orientation $H \| c$.

An estimate of the coherence length $\xi_{ab}(0)$ in the ab plane with the help of expression (3) yields a value $\simeq 7$ Å for the sample with $x = 7$ with $s = 3.8$ Å. This value looks too low in comparison with other estimates.[9] On the other hand, if we assume $(M/m)^{1/2} \approx 2$, for an anisotropic $3D$ case, we find $\xi_{ab}(0) \simeq 25$ Å. If we assume that a $2D$–$3D$ crossover occurs at small values of $\tau = \Delta T / T_c$, then these results look plausible. Note that the rapid decrease in the anomalous part of $\chi_\|$ with decreasing oxygen content x seems to imply a decrease in the effective coherence length ξ, because of the relation $(\xi_0 l)^{1/2}$, where l is the mean free path.

The anisotropic nature of the fluctuations in $YBa_2Cu_3O_x$ can explain the apparent difference between transition temperatures found in measurements of the electrical resistance of $YBa_2Cu_3O_7$ along and across the CuO_2 layers.[10]

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