Temperature dependence of the magnetic anisotropy of metallic Y-Ba-Cu-O single crystals in the normal phase

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The magnetic anisotropy measurements of metallic Y-Ba-Cu-O compounds in the normal phase reveal a temperature-dependent diamagnetic component of the susceptibility that increases with decreasing temperature. The temperature variation of the susceptibility anisotropy and its total change do not seem to be much affected by the presence of the superconductivity at some lower temperature and could not be accounted for by superconducting fluctuations. Rather, the data remind one of the behavior of some quasi-two-dimensional metals with anisotropic Fermi surfaces, reflecting the properties of the low-energy excitations in the normal phase. The anisotropy measurements above the bulk superconducting transition temperature T_c reveal the nonlinear effects, which are due to the onset of superconductivity in disconnected grains. The existence of a two-step transition, typical for granular superconductors, should be taken into consideration if the normal-phase susceptibility data are compared with the theoretical predictions in the vicinity of T_c.

The magnetic properties of high-temperature superconductors are very anisotropic[1-4] both in the normal and in the superconducting phase. The origin of the anisotropy and the possible relation between the magnetic properties and the superconductivity are not well understood. Recently, we observed[5] a large anisotropy in the magnetic susceptibility of various superconducting Y-Ba-Cu-O single crystals that exhibit a superconducting transition for temperatures between 12 and 88 K. The measurements of the anisotropy, which are defined as $\Delta\chi(T)=\chi_c-\chi_a$ with χ_c and χ_a being the principal values of the susceptibility tensor for **c** and **a** directions, are performed on the sensitive torque magnetometer.[6] The torque $\Gamma(T)$ is determined by the applied field and the magnetization of the sample and the susceptibility anisotropy is obtained from the ratio $\Gamma(T)/H^2$. It turns out that the temperature of the bulk superconducting transition T_c separates two regions with markedly different anisotropy properties: $\chi_c-\chi_a$ is positive for $T>T_c$ and negative for $T<T_c$, with the absolute values of χ_c and χ_a in the two regions differing by several orders of magnitude. The data indicate that the c-axis response is the most paramagnetic one in the normal phase while it is most diamagnetic in the superconducting phase. The temperature T_c can be accurately defined from the temperature variation of a number of properties either in the normal or in the superconducting state. Since $\Delta\chi(T)$ vanishes at T_c and varies rapidly around T_c, the question arises if the behavior of the anisotropy follows from the proximity and the properties of the superconducting state or if it reflects the properties of the low-energy excitations of the normal phase.

Typical results for $\Delta\chi(T)$ are shown in Fig. 1 for the sample having a mass of 6.82 mg and $T_c=87.6$ K. Similar results were also observed on a number of Y-Ba-Cu-O single crystals[5] and the anisotropy curves of similar shape have been reported for oriented powders of La, Y, and Bi superconductors.[3] However, most published data on $\Delta\chi(T)$ are obtained by measuring χ_c and χ_{ab} separately, which does not allow an accurate determination of the anisotropy in the temperature range where $\Delta\chi(T)\rightarrow 0$. The torque method, on the other hand, is the most accurate just in that region. The details regarding the method we use and the preparation and the properties of the samples studied in this work are given in Ref. 2.

The anisotropy exhibits four different types of behavior in various temperature regions. (i) At high temperatures the anisotropy assumes a constant value: at 300 K we have $\Delta\chi(T)\approx\Delta\chi(\infty)$, where $\Delta\chi(\infty)=1.4\times10^{-4}$ emu/mol is the anisotropic van Vleck susceptibility which appears because different transitions between the crystal-field split states of Cu ions in the Cu-O planes are induced for different orientations of the magnetic field. Note that $\Delta\chi(\infty)$ is of the order of the intrinsic susceptibility itself. (ii) Below 250 K the reduction of temperature results in the decrease of $\Delta\chi(T)$, with the slope of $\Delta\chi(T)$ increasing very fast as T_c is approached (see inset in Fig. 1). For all $T>T_c$ we have $\Delta\chi(T)<\Delta\chi(\infty)$. (iii) In the vicinity of T_c there exists a well-defined temperature interval in which $\Delta\chi(T)$, defined as $\Gamma(T,H)/H^2$, is field dependent for small values of the applied field. In the high-field limit, $\Delta\chi(T)$ becomes field independent

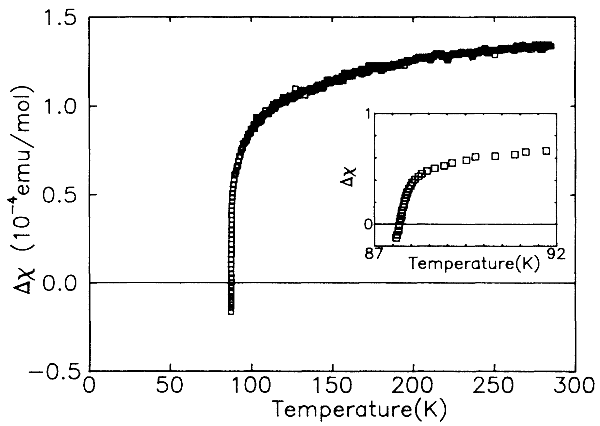

FIG. 1. Susceptibility anisotropy, measured in $H=208$ G, is plotted as a function of temperature. The inset shows the behavior in the vicinity of T_c.

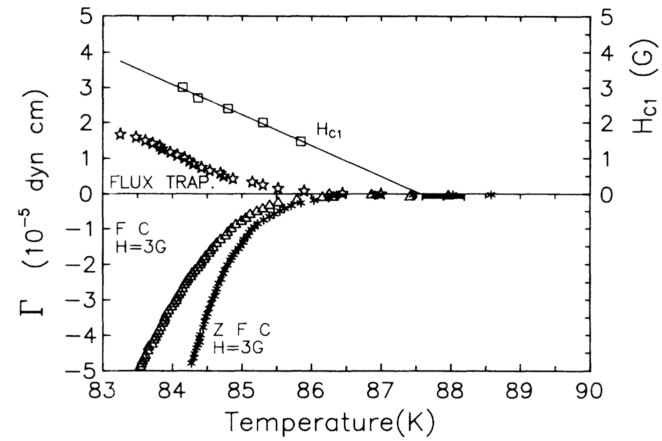

FIG. 2. The torque measurements used for the determination of T_c are shown. Stars: zero-field-cooled data. Triangles: field-cooled data. Open stars: trapped flux data. Squares: $H_{c1}(T)$ data. In the shaded temperature interval above T_c the nonlinear effects are obtained.

again and assumes the values typical of the normal phase. (iv) For $T < T_c$, a typical superconducting response is observed: the response is diamagnetic and the torque values are a few orders of magnitude larger than in the normal phase. We remark that the slope of $\Gamma(T)$ changes substantially across T_c.

The change of slope of $\Gamma(T)$ and the nonlinearities which can be detected between T_c and some higher temperature T_g can be taken as the evidence for a two-step transition into the superconducting state, typical for granular superconductors.[7] Upon cooling, at T_g, the superconductivity is induced first in isolated grains of the sample. At lower temperatures, the correlation between the phases of the superconducting wave functions of different grains increases and, in the second step, different phases lock together giving rise at T_c to the bulk superconductivity. The presence of the nonlinearities in Γ below T_g makes it difficult to define the bulk T_c very sharply by approaching the superconducting transition from above. However, detailed studies of the anisotropy in the bulk superconducting phase allow us to define T_c to a precision which greatly exceeds the temperature interval $\approx(T_c - T_g)$, in which the nonlinear effects are observed.

The procedure and the data used to estimate T_c are shown in Fig. 2. The solid stars correspond to the experiment in which the sample is cooled in zero field ($H < 0.5$ G) well below T_c, where the field of 3 G is switched on and $\Gamma(T)$ measured. The data indicate a large c-axis diamagnetism and the initial susceptibility which is a substantial fraction of the ideal Meissner value, $1/4\pi$. Recording $\Gamma(T)$, in a constant field, as a function of temperature and estimating the temperature for which $\Gamma(T)=0$, we obtain $T_c=87.6$ K. In the second experiment (triangles in Fig. 2), the sample is cooled in a constant field ($H=3$ G) and $\Gamma(T)$ is measured. The temperature dependence of $\Gamma(T)$ allows us to obtain T_c from the extrapolation $\Gamma(T_c)=0$. Note, that the line of reversibility[8] for the field-cooled and zero-field-cooled torque measurements in 3 G is very short so that the same value of T_c is obtained in both experiments. The open stars in Fig. 2 describe the data obtained in cooling the sample in 5 G to temperatures well below T_c. Then, the field is switched off so that some of the flux remains trapped in the specimen. The torque measurements in the small fields ($H < 0.5$ G) reveal the response typical of the trapped flux magnetization. The trapped flux vanishes at $T_c \pm 0.1$ K. Finally, the field dependence of Γ below T_c reveals the temperature dependence of $H_{c1}(T)$ in the bulk phase. The data (squares in Fig. 2) are obtained by estimating the field value at which the torque, at a given temperature, ceases to be a linear function of the applied field. The extrapolation of the data indicates again that $H_{c1}(T)$ vanishes at T_c.

Since the uncertainty in the T_c determination ($\Delta T_c < |0.1|$ K) is much smaller than the extension of the region where nonlinear effects are observed ($\Delta T < |T_c - T_g| < 1$ K for 90-K superconductors), the field dependence of the torque above T_c can be studied in detail. The field dependence, measured for temperatures between $T_c=87.6$ K and $T_g=88.15$ K, is shown in Fig. 3

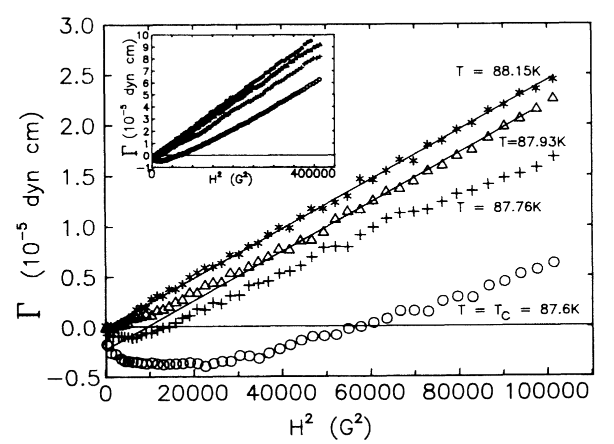

FIG. 3. Torque is plotted as a function of H^2 for various temperatures above T_c. The high-field slope (see inset) is used to estimate the anisotropy due to the normal-state electrons.

as a function of H^2. Above T_g, the torque is a linear function of H^2 for any value of the applied field and its slope gives the normal-phase anisotropy. Below T_g the high-field data can still be approximated by a straight line which, however, has the finite intercept at $H^2=0$ (see inset in Fig. 3). The slope of the high-field data decreases continuously with decreasing temperature and we take the value of that slope as the measure of the part of the anisotropy which is due to the normal-phase electrons. Thus, we obtain positive-normal-phase anisotropy even for $T<T_c$. The nonlinear part of Γ, obtained by subtracting from the data the high-field linear component, is due to the onset of superconductivity in disconnected grains.[7] The magnitude of this additional diamagnetic contribution, at a given T, decreases very slowly (probably logarithmically) with the increase of the field and for sufficiently high fields it can be suppressed completely.

The additional diamagnetic component of $\Delta\chi(T)$, which develops at low T, can be analyzed in more detail. In Fig. 4 this diamagnetic contribution, normalized by the high-temperature Van Vleck part so that it varies between 1 and 0, is plotted as a function of reduced temperature, $t=(T-T_c)/T_c$. For temperatures between $t=2.5$ and 0.2, i.e., for T between 300 and 105 K, the experimental data could be fitted by the logarithmic function $f(t)=a\ln(t)+b$ (solid line in Fig. 4), with the coefficients $a=-0.12$ and $b=0.13$. However, in the next decade, from $t=0.1$ to 0.01, the logarithmic fit (see inset in Fig. 4) requires a and b which are 1 order of magnitude smaller. Finally, for $t\to 0$, the normal-phase torque, obtained by correcting the data for the nonlinear contribution as discussed above and shown as open triangles in the inset of Fig. 4, assumes a finite value. Our data show that the total anisotropy change in the normal phase does not exceed the magnitude of the Van Vleck anisotropy. Preliminary measurements with single crystals of La, Bi, and Tl cupric superconductors have shown similar features as well.

Since the Van Vleck anisotropy depends only weakly on the value of T_c, (i.e., on the oxygen content), even when T_c is changed by 1 order of magnitude, and since the temperature interval in which the growth of the additional diamagnetic susceptibility is observed does not scale with T_c, the temperature variation of $\Delta\chi(T)$ in the normal phase is difficult to associate with the superconducting fluctuations.

At present, we are unable to explain the observed temperature variation of the normal-phase susceptibility. We would like to point out, however, that the changes remind one of the anisotropy in typical quasi-two-dimensional metals like hexamethylenetetraselenofulvalene tetracyanoquinodimethane (HMTSF-TCNQ) (Ref. 9) or Zr_3Te_5.[10] In Fig. 5 we again plotted

$$-[\Delta\chi(T)\text{-}\Delta\chi(\infty)]/\Delta\chi(\infty)\ ,$$

but now as a function of absolute temperature. For temperatures above 120 K the data are described rather accurately by a power law AT^a, where $\alpha=-2.35$. The HMTSF-TCNQ data, plotted in the same way, would be given by a similar curve with $\alpha=-1.41$; here, the increase of the diamagnetic anisotropy is mainly due to the increase of the Landau-Peierls diamagnetism[11] upon cooling. We cannot claim that the temperature variation of the anisotropy in high-T_c compounds can be explained by simple band-structure effects, but the similarity between the high-T_c data and the data corresponding to typical quasi-two-dimensional metals is striking. We think it would be of interest to measure the anisotropy of high-T_c compounds in very high fields to see if the normal-phase anisotropy at low temperatures shows the same tendency towards saturation as observed in other two-dimensional metals.

To conclude, the magnetic measurements indicate that the normal-phase susceptibility of metallic Y-Ba-Cu-O compounds has a temperature-dependent diamagnetic component which increases with decreasing temperature on a scale set by the room-temperature susceptibility. The temperature variation of the susceptibility anisotropy and its total change in the normal phase do not seem to be affected much by the presence of the superconduc-

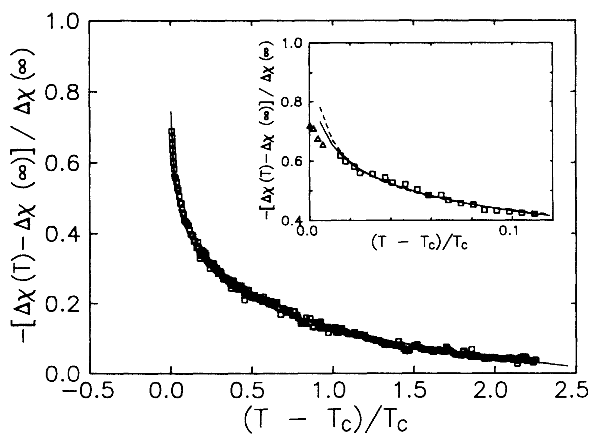

FIG. 4. Relative susceptibility anisotropy, $-[\Delta\chi(T)-\Delta\chi(\infty)]/\Delta\chi(\infty)$, is plotted as a function of reduced temperature $t=(T-T_c)/T_c$. The solid line shows the logarithmic fit (see text). The inset shows the data in the lower t decade. The open triangles are the data corrected for the nonlinear contribution.

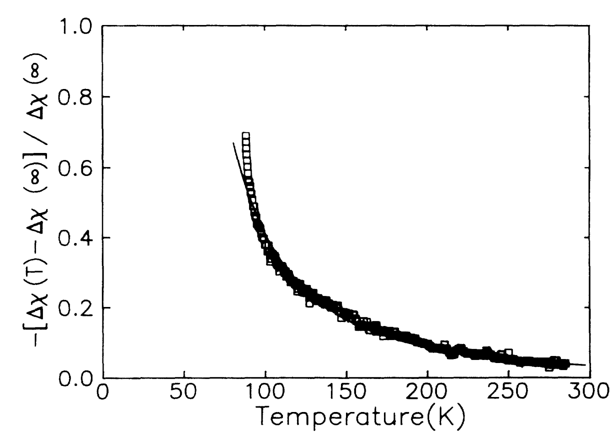

FIG. 5. Relative susceptibility anisotropy, $-[\Delta\chi(T)-\Delta\chi(\infty)]/\Delta\chi(\infty)$, is plotted as a function of absolute temperature T. The solid line shows the power-law fit (see text).

tivity at some lower temperature and could not be accounted for by superconducting fluctuations. Rather, the data remind one of the behavior of some quasi-two-dimensional metals with an anisotropic Fermi surface and we believe that they reflect the properties of the low-energy excitations in the normal phase. Our measurements of $\Delta\chi(T)$ in Y-Ba-Cu-O compounds indicate a two-step transition into the superconducting state, typical for granular superconductors, and this has to be taken into consideration if the normal-phase susceptibility data are compared with the theoretical predictions in the vicinity of T_c.

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