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## Longitudinal and transverse magnetoresistances of  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>$  in very high field up to 430 kG

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Measurements of the resistivity, together with the longitudinal and transverse magnetoresistances of  $YBa_2Cu_3O_7$  in extremely high fields up to 430 kG at different temperatures (4.2-300 K) are reported. It is shown that the application of a field as high as 430 kG does not destroy superconductivity at 50 K. Moreover, the resistance at 77 K and 430 kG does not approach the saturation limit and is only  $\sim$  50% of that of the normal phase. A large negative and anisotropic magnetoresistance  $\Delta \rho$  is observed at high temperature (T > 100 K). It is suggested that this behavior could be either of magnetic origin or due to some influence of the electromagnetic field on the short-range order of oxygen ions.

Since the recent discovery of a new class of high- $T_c$  superconductors, <sup>1-3</sup> tremendous efforts have been devoted to these materials. In this paper, we want to focus our attention on some transport properties of both the superconducting and the high-temperature phases and we report measurements of the resistivity  $[\rho(T,0)]$ , the longitudinal  $[\rho_{\parallel}(T,H)]$ , and the transverse  $[\rho_{\perp}(T,H)]$  magnetoresistances of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> as a function of field  $(0 \le H \le 430)$ kG) and temperature,  $(4.2 \le T \le 300 \text{ K})$ . As usual, by longitudinal and transverse magnetoresistances we mean that the field H is applied parallel or perpendicular to the direction of the current flow in the sample.

For simplicity, these magnetoresistances will be defined by  $\Delta \rho_{\parallel} = \rho_{\parallel} (T, H) - \rho (T, 0)$  and  $\Delta \rho_{\perp} = \rho_{\perp} (T, H) - \rho (T, 0)$ . It will be helpful for the analysis of the present results to recall that it was shown recently by Oussena, Senoussi, and Collin<sup>4</sup> and by Senoussi, Oussena, Ribault, and Collin<sup>5</sup> that the magnetic and transport properties of the new superconducting materials, including the sample investigated here, are (in the present state of the methods of preparation) those of a type-II granular superconductor in which strongly superconducting grains are coupled via weak superconducting links. In addition, there is evidence that the critical current density and the lowest critical field  $H_{c1}$  of these materials were governed by the weak links and do not represent the intrinsic parameters of the bulk state. Here too we shall see that the destruction of superconductivity by the high external field is, to a larger extent, related to the imperfection of the material (weak links). As a matter of fact, it seems that a large fraction of the grains are still superconducting in 400 kG and at liquid-nitrogen temperature.

Our experiments were performed in the "Service National des Champs Intenses de Toulouse" at different temperatures in the slowly decreasing part of a pulsed field cycle (field rise time: 200 ms, field decay time: 800 ms). The measurement is a classical four-probe ac method using a current of <sup>1</sup> mA at a frequency of 100 kHz. The sample was a small parallelepiped (length 10 mm, width  $\sim$  3 mm, thickness  $\sim$  1.5 mm).

The material was prepared starting from  $Y_2O_3$ , BaCO<sub>3</sub>, and CuO oxides reacted at 900-1000'C for 3 h. After grinding, the powder was reannealed in an oxygen flow at  $300^\circ$ ,  $500^\circ$ , and  $700^\circ$ C for 12 h at each temperature. The powder was pressed into pellets and then sintered in oxygen. Our samples were systematically characterized by x-ray diffraction with a Guinier-Lainé Caméra (Cu  $Ka$ ) and are single phase.

Figure <sup>1</sup> shows the temperature dependence of the resistance of  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>$  at 0, 200, and 400 kG. We draw attention, however, to the fact that, contrary to  $\rho(T, 0)$ ,  $\rho(T, 200 \text{ kG})$  and  $\rho(T, 400 \text{ kG})$  have not been traced point by point on raising the temperature, but have been deduced indirectly from the isothermal magnetoresistance curves displayed in Fig. 2. In agreement with other results, <sup>6,7</sup> we find that  $\rho(T, 0)$  exhibits a clear metallic conducting behavior with a linear temperature dependence over the temperature range  $(110 \le T \le 300 \text{ K})$ .<sup>8</sup> As shown in the inset in Fig. 1, negative deviation from the linear laws sets in below about 110 K. From Fig. 1, the transition temperature (at  $H = 0$ ) is estimated to be about 93 K (defined as the point where  $\rho$  is reduced to approximately half its high-temperature value). The width of the transition (defined as the temperature interval over which the resistance raises from  $\sim$  10% to 90% of its final value) is  $\sim$  2 K. It is worth noting too that the transition temperature and the associated width at 400 kG (defined as above) are about 78 and 22 K, respectively.



FIG. 1. Temperature dependence of the electrical resistances in  $H=0$ , and in transverse fields of 200 and 400 kG. It is to be stressed that  $H = 200$ - and 400-kG curves have not been measured at constant field, but have instead been deduced indirectly from the transverse magnetoresistance isotherms of Fig. 2. The dashed lines are linear extrapolations of the high-temperature resistances to low temperatures and have been traced for the purpose of comparison with the true resistances (full lines). The inset is an enlargement of the transition region showing that the resistance starts to drop toward zero value at  $\approx$  110 K.

As in Ref. 6, it is of interest to ascribe the departure from linearity [curve  $\rho(T,0)$ ] to the onset of superconducting effects near this temperature  $(110 K)$  and then check if this assumption is consistent with our magnetoresistance data around 110 K.

The transverse magnetoresistance  $\Delta \rho_{\perp}(T,H)$  at dif-



FIG. 2. Transverse magnetoresistance  $(\Delta R \text{ vs } H)$  at the indicated temperatures. Note that  $\Delta R$  becomes negative at high temperatures in the nonsuperconducting phase.

ferent constant temperatures is presented in Fig. 2. First of all, we observe that superconductivity at 50 K is not destroyed (within the present experimental precision) by the highest field applied here. This is *direct evidence that* the upper critical field at 50 K is greater than  $430$  kG  $[H_{c2}(50 K) > 430kG].$ 

Also displayed in Fig. 2 is the transverse magnetoresistance at 77 K which is now different from zero. However, the most interesting feature of the 77 isotherm is that it does not saturate as a function of  $H$  even at 430 kG. This is perhaps best illustrated in Fig. 1 where the comparison of the curves  $\rho_{\perp}(T,200 \text{ kG})$  and  $\rho_{\perp}(T,400 \text{ kG})$ , with the dashed lines representing the extrapolation of the normal-state resistivity to low temperatures, shows that  $\rho_{\perp}$ (77 K, 400 kG) is only about half of the saturation limit which would be reached if the whole sample were in the normal state. In other words, this suggests strongly that a large fraction of the sample is still superconducting at 77 K and at 430 kG. Now, assuming that the upper critical field is given approximately<sup>9</sup> by the parabolic equation

$$
H_{c2}(T) = H_{c2}(0)[1 - (T/T_c)^2]
$$
 (1)

with

$$
T_c \approx 93 \text{ K},\tag{2}
$$

we deduce that for these regions of the sample which stay superconducting at 77 K, 430 kG, we have  $H_{c2}(T) \ge 430$ kG and  $H_{c2}(0) = H_{c2}(77)/[1 - (T/T_c)^2] \ge 1200$  kG.

We now consider the 90-K magnetoresistance (Fig. 2) which shows clear tendency toward saturation with increasing field. However, here too the comparison of the curves  $\rho_{\perp}(T,200 \text{ kG})$  and  $\rho_{\perp}(T,400 \text{ kG})$  of Fig. 1 with the dashed lines of the same figure is interesting and suggests that superconducting effects (though very weak) still persist at high field. Let us assume that this is connected with some small regions of the sample and apply the same reasoning as for the 77-K isotherm. Then, from Eq. (1), we obtain the upper critical field which would be associated with these regions:  $H_{c2}(0) \ge 430 \text{ kG/[1-(90/93)^2]}$  $\approx$  7 MG. Such a value is probably too high to have a plausible meaning. It might signify that the high field data near 90 K cannot be described by the same transition temperature (93 K) as at low  $H$ .

Instead of taking  $T_c = 93$  K for the whole sample (as we have done before) and searching for  $H_{c2}$ , we can tentatively assume that the superconducting effects remaining (90 K, 400 kG) have the same origin as the departure from linearity of  $\rho(T,0)$  for  $T \le 110$  K. Then, putting  $T_c'$  = 110 K (instead of 93 K) in Eq. (1) leads to  $H'_{c2}(0) \ge 1250$  kG. This result must be regarded with caution; nevertheless it is not inconsistent with the suggestion that superconductivity effects begin at temperatures as high as 110 K. More generally, this might either be due to chemical inhomogeneities, especially variations in oxygen composition, or to more intrinsic properties such as time-dependent critical phenomena.

Returning to Fig. 2, we see that the transverse magnetoresistance of the high-temperature phase is negative and roughly quadratic in  $H$ . Moreover, its amplitude first increases with T [since  $|\Delta \rho(110 \text{ K}, H)| < |\Delta \rho(135 \text{ K}, H)|$  $H$ ) 1, and then evolves much more slowly with increasing



FIG. 3. Field dependence of the reduced transverse magnetoresistance  $\Delta \rho / \rho(T, 0)$  at different temperatures.

T, as we see no detectable difference between  $\Delta \rho_{\perp}$  (135 K, H) and  $\Delta \rho_{\perp}$ (200 K,H). This is probably fortuitous and would mean that  $\Delta \rho_{\perp}$  passes through an extremum somewhere between 135 and 200 K. This is perhaps best illustrated in Fig. 3 showing  $\Delta \rho_{\perp}(T, H)/\rho(T, 0)$  at various T. We tentatively propose that the above behavior is due to the contribution of at least three competing terms which we write as follows:

$$
\Delta \rho = \Delta \rho^s + \Delta \rho^m + \Delta \rho^n \tag{3}
$$

Here  $\Delta \rho^s$  is a positive term arising from the suppression of superconducting fluctuations by the applied field.  $\Delta \rho^m$ is a negative contribution whose origin is not clear to us: It could be due to some unusual mechanisms, typical of the new superconductors, such as localization effects or, more probably, to some coupling between the electromagnetic field and oxygen, leading to a rearrangement of the short-range order of oxygen ions. A more conventional possibility is that it is a magnetic effect. In this case,  $\Delta \rho^m$ would represent a spin-disorder resistivity of the sort obpossibility is that it is a magnetic effect. In this case,  $\Delta \rho$  "<br>would represent a spin-disorder resistivity of the sort ob-<br>served either in usual ferromagnetic metals  $^{10,11}$  above the Curie temperature or in magnetic alloys such as spin glasses.<sup>12</sup> Interestingly enough, if the magnetoresistance is of magnetic origin, then the fact that it is strongly anisotropic with  $\Delta \rho_{\perp} \ll \Delta \rho_{\parallel}$  (Fig. 4) would imply a large orbital contribution comparable to the isotropic spin contribution. It is to be emphasized that the presence of orbital effects would have large influences on the behavior of



 $\begin{bmatrix} \mathbf{5} & 135K \end{bmatrix}$  FIG. 4. Longitudinal (**H**||i) and transverse magnetoresistances  $(H \perp i)$  at 200 K. Note the strong and positive anisotropy  $(\rho_{\parallel} - \rho_{\perp}) > 0$ .

many other physical quantities such as Hall resistivity, NMR, and EPR, etc.

Finally, the last term,  $\Delta \rho^n$  of the right-hand side of Eq. (3) is the Lorentz or (normal) magnetoresistance, the most characteristic feature of which is that it is always positive with  $\Delta \rho_{\perp}^n > \Delta \rho_{\parallel}^n$ . From Figs. 2 and 4, it is clear that as long as the transverse geometry is concerned, the Lorentz magnetoresistance is completely masked by the negative contribution, whereas it is predominant in the longitudinal configuration (Fig. 4). It is worth noting that very peculiar transport data showing magnetothermal irreversibilities, time-dependent effects, and other unusual phenomena at high temperature between 80 and 300 K have been reported recently.<sup>13</sup> No effects of that sort have been detected by us. The reason might be that transport properties are very sensitive to all kinds of inhomogeneities on a microscopic scale and could therefore differ markedly from sample to sample depending on the method of preparation employed.

Our high field data confirm that  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>$  behaves like an inhomogeneous material, the superconducting properties of which are at the present time primarily limited by the badly superconducting parts (weak links) of the sample. It is likely that with the inevitable improvement of the methods of preparation of these materials the critical field  $H_{c2}(77 \text{ K})$  will attain several hundreds of kG.

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 ${}^{1}$ K. H. Bednorz and K. A. Müller, Z. Phys. B 64, 189 (1986).

<sup>3</sup>C. W. Wu, J. R. Ashburn, C. J. Torng, P. H. Hor, R. L. Meng, L. Gao, Z. J. Huang, Y. Q. Wang, and C. W. Chu, Phys. Rev. Lett. 58, 908 (1987).

- 4M. Oussena, S. Senoussi, and G. Collin, Europhys. Lett. (to be published).
- S. Senoussi, M. Oussena, M. Ribault, and G. Collin, this issue, Phys. Rev. B 36, 4003 (1987).

<sup>&</sup>lt;sup>2</sup>C. W. Chu, P. H. Hor, R. L. Meng, L. Gao, Z. J. Huang, and Y. Q. Wang, Phys. Rev. Lett. 58, 405 (1987).

- <sup>6</sup>K. Kadowski, Y. K. Huang, M. Van Sprang, and A. A. Menovsky (unpublished).
- 7R. J. Cava, B. Batlogg, R. B. van Dover, D. W. Murphy, S. Sunshine, T. Siegrist, J. P. Remeika, E. A. Rietman, S. Zahurak, and G. P. Espinosa, Phys. Rev. Lett. 58, 1676 (1987).
- <sup>8</sup>It is to be stressed that the measured  $\rho(T, 0)$  curve exhibits a very small anomaly (not detectable on Fig. 1) around  $T \approx 220$  K. However, we have not been able to decide whether it is real or due to the experimental uncertainties on the temperature.
- <sup>9</sup>We are aware that the temperature dependence of  $H_{c2}$  is very complicated and depends on many characteristics of the ma-

terials, such as the mechanism responsible for superconductivity, anisotropy of the Fermi surface, electric mean free path, spin-orbit coupling, etc. Nevertheless, the quadratic approximation of formula (1) is probably reasonable for the qualitative discussion of our experimental results.

- <sup>10</sup>P. G. de Gennes and J. Friedel, J. Phys. Chem. Solids 4, 71 (19S8).
- <sup>11</sup>M. E. Fisher and J. S. Langer, Phys. Rev. Lett. 20, 665 (1968).
- $12$ S. Senoussi, J. Phys. F 10, 249 (1980); Solid State Commun. 4\$, 407 (1983);Phys. Rev. Lett. 56, 2314 (1986).
- <sup>13</sup>S. Komiyama, S. Hikami, and T. Takamasu, Jpn. J. Appl. Phys. Lett. (to be published).