Sign Change of *c*-Axis Magnetoconductivity in $YBa_2Cu_3O_{7-\delta}$ Single Crystals

J. Axnäs, W. Holm, Yu. Eltsev, and Ö. Rapp

Department of Solid State Physics, Royal Institute of Technology, S-100 44 Stockholm, Sweden

(Received 29 February 1996)

The out-of-plane magnetoconductivity $\Delta\sigma_c$ ($B\|I\|$crystal c axis) of two $YBa_2Cu_3O_{7-\delta}$ single crystals has been measured in magnetic fields up to 12 T at temperatures ranging from $T_c + 3$ K to $T_c + 40$ K. A change of sign, from negative $\Delta\sigma_c$ near T_c to positive at higher temperatures, is observed in a metallic sample ($d\rho_c/dT > 0$). Recent fluctuation theories can describe the main features of these observations with parameters that are in good agreement with other studies. These results suggest that fluctuations in the density of states have been observed in the magnetoconductivity. [S0031-9007(96)00941-6]

PACS numbers: 73.50.Jt, 74.40.+k, 74.72.Bk

It has recently become increasingly clear that the *c*-axis properties of anisotropic high-temperature superconductors are essential for understanding the anomalous superconducting as well as normal-state properties. A problematic observation, for instance, is a positive $d\rho/dT$ for the *ab*-plane resistivity combined with a strongly negative *c*-axis $d\rho/dT$ in underdoped $YBa_2Cu_3O_{7-\delta}$ and in Bi-based materials. Attempted descriptions include various tunneling models, fluctuations, interplanar disorder, and hopping due to resonant tunneling [1–5].

Studies of superconducting fluctuations in the magnetoconductivity, $\Delta\sigma(B,T) = 1/\rho(B,T) - 1/\rho(0,T)$, represent a powerful experimental technique to address related problems. The considerable effects of the short-lived electron pairs over a wide range of temperatures above T_c, together with the possibility to use strong magnetic fields, provide for stringent comparisons with theories. In addition to reliable estimates of the coherence lengths and the phase-breaking time [6], measurements of $\Delta\sigma$ may allow for conclusions about the pairing state [7,8], or clarify the nature of the impurity state in certain doped alloy systems [9,10]. There is also a rapid development of the fluctuation theories, with continued discussions about what terms and what limits of existing theories to use, and with an increasing number of suggested additions and new theories [11–13].

In the present study we have measured the *c*-axis magnetoconductivity $\Delta\sigma_c$ ($B\|I\|c$) of two $YBa_2Cu_3O_{7-\delta}$ single crystals in the temperature range $T_c + 3$ K to $T_c + 40$ K. A change of sign was observed, from negative $\Delta\sigma_c$ near T_c to positive above $T_c + 10$ K. In the theory by Dorin and co-workers [13], such a sign change could occur as a result of fluctuations in the normal density of states. We have adapted this theory for comparisons with experiments. Excellent descriptions of the observations with reasonable parameter values were obtained in the region where $\Delta\sigma_c \leq 0$, including the sign change at approximately the correct temperature. In the region where $\Delta\sigma_c > 0$, the uncertainties are somewhat larger.

Two single crystals of $YBa_2Cu_3O_{7-\delta}$ were grown by the self-flux method [14]. Sample A was oxygenated at 460, 400, 350, and finally 300 °C, for a few days at each temperature. Sample B was oxygenated at 400 °C. Sample A was heavily twinned, whereas sample B contained only a few twins. Sample dimensions and contact configurations are shown in the insets of Fig. 1.

The resistivity was measured with a standard four-probe technique. The contact configuration of sample B gives an inhomogeneous current distribution, and a finite clement analysis was used to obtain the resistivity [15]. In Fig. 1 the *c*-axis resistivity of both samples is plotted vs temperature, and the importance of the finite element analysis is also illustrated. The remaining small peak in the resistivity of sample B is probably a result of a lower oxygen content [16]. The magnetoconductivity was measured with $B\|I\|c$ at a series of fixed temperatures,

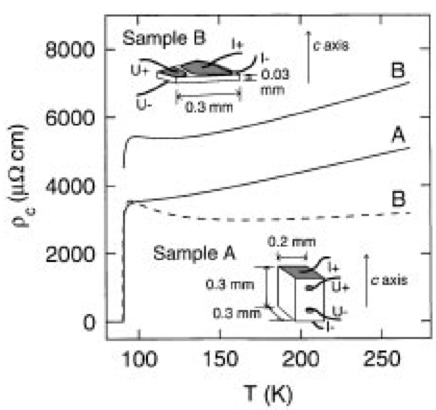

FIG. 1. Main panel: Solid curves are temperature dependences of the *c*-axis resistivities. The overall magnitude of the curve for sample B is uncertain, but the relative changes with temperature are more reliable. The dashed curve shows the resistivity deduced for sample B if inhomogeneities in the current distribution are neglected (see text). For sample A, $T_c \approx 90.9$ K and $\Delta T_c \approx 0.2$ K, and for sample B $T_c = 90.4$ K and $\Delta T_c \approx 0.15$ K. T_c was defined from the midpoint of the resistive transition and ΔT_c from 10% to 90% of the resistive drop. The measured electrical resistance at 100 K was 88 mΩ for sample A and 16 mΩ for sample B. Insets: Approximate sample dimensions and contact configurations (not to scale). Shaded area represent silver paint contacts to which gold wires are attached.

 0031-9007/96/77(11)/2280(4)$10.00

with the magnetic field swept up to 12 T. The correction to $\Delta\sigma_c$ from the finite element analysis of sample B was considerable: 150% at 95 K and 12 T, 30% at 115 K. The temperature was regulated with a platinum thermometer located in zero magnetic field 20 cm above the sample, and the temperature at the sample was measured with a diode thermometer immediately before and after each field sweep. The temperature drift during sweeps was negligible. The results are shown in Fig. 2 for $\Delta\sigma_c(B)$ and in Fig. 3 for $\Delta\sigma_c(T)$ at 12 T. A striking feature is the sign change of $\Delta\sigma_c$ at about 102 K [$\varepsilon = \ln(T/T_c) \approx 0.12$]. The curves in the figures will be described below. The estimated errors for sample B are large due to the low resistance and the unfavorable contact configuration. For sample A these problems do not occur. A change of sign of $\Delta\sigma_c$ is unambiguously verified for sample A, and the results for sample B are consistent with this finding.

There are several theoretical studies of the *c*-axis fluctuation conductivity, $\sigma_c^{\rm fl}(B,T)$, in high-temperature superconductors. Recent calculations by Dorin *et al.* [13] include four contributions to the fluctuation conductivity:

$$\sigma_c^{\rm fl} = \sigma_c^{\rm Al} + \sigma_c^{\rm DOS} + \sigma_c^{\rm MT(reg)} + \sigma_c^{\rm MT(an)}.$$

The Aslamazov-Larkin (AL) contribution represents the direct acceleration of superconducting electrons. This contribution has been derived previously under various conditions [17–20]. The DOS contribution is a result of the fluctuation of the normal density of states, and has opposite sign. This term was previously neglected, in spite of the fact that it can be comparable in magnitude to the AL contribution. In particular, it is the competition between the AL and DOS contributions that can give rise to a peak in the resistivity and a change of sign in the magnetoconductivity. The regular (reg) and anomalous (an) Maki-Thompson (MT) contributions are indirect effects on the normal-site electrons, and are usually smaller. Zeeman corrections [21,22] were neglected here.

The fluctuation magnetoconductivity, $\Delta\sigma_c^{\rm fl}(B,T)$, is given by

$$\Delta\sigma_c^{\rm fl} = \Delta\sigma_c^{\rm AL} + \Delta\sigma_c^{\rm DOS} + \Delta\sigma_c^{\rm MT(reg)} + \Delta\sigma_c^{\rm MT(an)},$$

where $\Delta\sigma_c^{\rm AL} = \sigma_c^{\rm AL}(B,T) - \sigma_c^{\rm AL}(0,T)$, etc. Since the weak-field approximation of Ref. [13] is not valid for our field strengths in $\mathrm{YBa_2Cu_3O_{7-\delta}}$, we must use the full expressions. However, the full expressions in Ref. [13] for the DOS contribution, $\sigma_c^{\rm DOS}(B,T)$ and $\sigma_c^{\rm DOS}(0,T)$, cannot be directly used for comparison with experiments: Firstly, $\lim_{B\to 0}\sigma_c^{\rm DOS}(B,T) \neq \sigma_c^{\rm DOS}(0,T)$. This inconsistency is easily removed by omitting an unnecessary simplification in the last step of the derivation of $\sigma_c^{\rm DOS}(0,T)$. Secondly, the sharp field-dependent cutoff in the sum for $\sigma_c^{\rm DOS}(B,T)$ of Ref. [13] gives a large oscillatory dependence on the magnetic field, in disagreement both with the weak-field approximation of Ref. [13] and with experiments. The precise form of this cutoff is, however, unknown and can only be approximated [23]. We used a weighted average of calculations of the sum for different sharp cutoffs, with almost all weight given to cutoffs near that of Ref. [13]. This gives a monotonic field dependence and agreement with the weak-field approximation at low fields. The resulting full expressions for the four terms in $\Delta\sigma_c^{\rm fl}$ are then

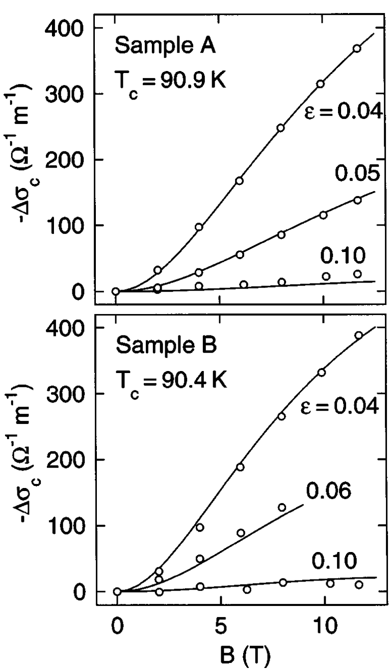

FIG. 2. The symbols are the observed magnetoconductivity $\Delta\sigma_c$ ($\approx -\Delta\rho_c/\rho_c^2$) with $B\|I\|c$ for $\varepsilon = \ln(T/T_c) = 0.04$–$0.10$ ($T = 94.2$–100.0 K). The curves are the best fits of the fluctuation theory (see text).

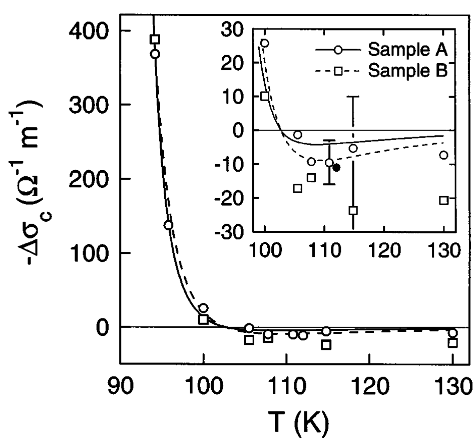

FIG. 3. Main panel: The symbols are the experimental magnetoconductitivy $\Delta\sigma_c$ for $I \parallel c \parallel B = 12$ T. The solid and dashed curves are the corresponding theoretical calculations with parameters from the best fit. Inset: Blow-up, which also gives typical experimental uncertainties in $\Delta\sigma_c$. At $T = 112$ K the measurements on sample A were made with a higher precision, and the uncertainty is within the filled circle.

$$\Delta\sigma_c^{\mathrm{AL}} = \frac{e^2 s}{128\eta\hbar}\left[r^2\beta\sum_{n=0}^{\infty}\frac{1}{[(\varepsilon_B+\beta n)(\varepsilon_B+\beta n+r)]^{3/2}} - 4\left(\frac{\varepsilon+r/2}{[\varepsilon(\varepsilon+r/2)]^{1/2}}-1\right)\right]$$

$$\Delta\sigma_c^{\mathrm{DOS}} = -\frac{e^2 sr\kappa}{16\eta\hbar}\left[\beta\frac{1}{\sum_{k=0}^{\infty}w_k}\sum_{N=0}^{\infty}\left(w_N\sum_{n=0}^{N}\frac{1}{[(\varepsilon_B+\beta n)(\varepsilon_B+\beta n+r)]^{1/2}}\right) - 2\ln\left(\frac{(1+\varepsilon)^{1/2}+(1+\varepsilon+r)^{1/2}}{\varepsilon^{1/2}+(\varepsilon+r)^{1/2}}\right)\right]$$

$$\Delta\sigma_c^{\mathrm{MT(reg)}} = -\frac{e^2 s\tilde{\kappa}}{8\eta\hbar}\left[\beta\sum_{n=0}^{\infty}\left(\frac{\varepsilon_B+\beta n+r/2}{[(\varepsilon_B+\beta n)(\varepsilon_B+\beta n+r)]^{1/2}}-1\right) - \frac{1}{2}[(\varepsilon+r)^{1/2}-\varepsilon^{1/2}]^2\right]$$

$$\Delta\sigma_c^{\mathrm{MT(an)}} = \frac{e^2 s}{16\eta\hbar}\left[\frac{\beta}{\varepsilon-\gamma}\sum_{n=0}^{\infty}\left(\frac{\gamma_B+\beta n+r/2}{[(\gamma_B+\beta n)(\gamma_B+\beta n+r)]^{1/2}} - \frac{\varepsilon_B+\beta n+r/2}{[(\varepsilon_B+\beta n)(\varepsilon_B+\beta n+r)]^{1/2}}\right) - \left(\frac{\gamma+\varepsilon+r}{[\varepsilon(\varepsilon+r)]^{1/2}+[\gamma(\gamma+r)]^{1/2}}-1\right)\right]$$

In these formulas $r = 2k_B^2 J^2\tau^2 f_0/\hbar^2$, $\beta = 4\eta eB/\hbar$, $\eta = v_F^2\tau^2 f_0/2$,

$$f_0 = -\left[\Psi\left(\frac{1}{2}+\frac{\hbar}{4\pi k_B T\tau}\right) - \Psi\left(\frac{1}{2}\right) - \frac{\hbar}{4\pi k_B T\tau}\Psi'\left(\frac{1}{2}\right)\right]$$

$$\kappa = \frac{-\Psi'(\frac{1}{2}+\hbar/4\pi k_B T\tau) + (\hbar/2\pi k_B T\tau)\Psi''(\frac{1}{2})}{\pi^2[\Psi(\frac{1}{2}+\hbar/4\pi k_B T\tau) - \Psi(\frac{1}{2}) - (\hbar/4\pi k_B T\tau)\Psi'(\frac{1}{2})]}$$

$$\tilde{\kappa} = \frac{-\Psi'(\frac{1}{2}+\hbar/4\pi k_B T\tau) + \Psi'(\frac{1}{2}) + (\hbar/4\pi k_B T\tau)\Psi''(\frac{1}{2})}{\pi^2[\Psi(\frac{1}{2}+\hbar/4\pi k_B T\tau) - \Psi(\frac{1}{2}) - (\hbar/4\pi k_B T\tau)\Psi'(\frac{1}{2})]}$$

$\gamma = 2\eta/v_F^2\tau\tau_\phi$, $\varepsilon = \ln(T/T_c)$, $\varepsilon_B = \varepsilon + \beta/2$, $\gamma_B = \gamma + \beta/2$, and w_n is a weight function, here taken to be

$$w_n = \mathrm{erf}[n - (1/\beta - 1/2) + 1] - \mathrm{erf}[n - (1/\beta - 1/2)]$$

with

$$\mathrm{erf}(x) = \frac{2}{\sqrt{\pi}}\int_0^x e^{-t^2}dt$$

T is the temperature, B the magnetic field, s the layer spacing, v_F the Fermi velocity parallel to the layers, τ the in-plane elastic scattering time, τ_ϕ the phase-breaking time, and J the hopping integral (in Kelvin). $\Psi = d[\ln\Gamma(x)]/dx$ is the digamma function. The expressions are the same for all values of τT_c, making the usual distinction between clean ($4\pi k_B\tau T_c/\hbar \gg 1$) and dirty ($4\pi k_B\tau T_c/\hbar \ll 1$) limits unnecessary. The derivation was made assuming $\beta \ll 1$ and $\varepsilon_B \ll 1$.

We compared our experimental magnetoconductivity with the above theory, including all four terms. The magnetoconductivity of the normal state was neglected, since the fluctuations are large close to T_c. In the comparison we took $s = 11.69$ Å, $v_F = 2\times10^5$ m/s [9], and T_c from the midpoints of the resistive transitions. J was assumed to be constant within our measurement range. Further it was assumed, somewhat arbitrarily, that $\tau = \tau_\phi \propto T^{-1}$. We then fitted the formulas to the experimental data by varying the two parameters J and the overall magnitude of τ_ϕ and τ [parametrized by τ(100 K)] until the lowest root mean square deviation (rms) was obtained. Since $\varepsilon \ll 1$ is assumed in the theory, only data for $\varepsilon \le 0.1$ ($T \le 100$ K) were used in the fits. The parameters giving the best fits were $J = 205$ K and $\tau(100\ \mathrm{K}) = 3.1$ fs for sample A and $J = 225$ K and $\tau(100\ \mathrm{K}) = 5.0$ fs for sample B. The uncertainties are for sample A less than 30% for J and 50% for τ if we use the criterion that the rms should be at most twice its minimum value. The agreement between experiment and theory was excellent at the lowest temperatures and still good at $\varepsilon = 0.10$ (Fig. 2). The observed change of sign in $\Delta\sigma_c$ (Fig. 3) is also obtained at approximately the correct temperature, although the calculated magnitude at higher temperatures is less than observed. This discrepancy may be due to violation of the condition $\varepsilon \ll 1$ in the theory. The choice of cutoff is, however, important for $T > 100$ K. If the weight function w_n is changed so that the cutoff is, e.g., 3 times broader, good agreement with experiments can be obtained in this region. The assumption $\tau_\phi = \tau \propto T^{-1}$ is not crucial. If, e.g., τ and τ_ϕ are assumed to be temperature independent, or a different value for τ_ϕ in the range 0.5–50 fs is used, good fits can still be obtained by changing J and τ by only 10%. This is partly explained by the fact that τ_ϕ only enters into one of the MT terms, both of which are typically several times smaller than the other terms. The AL term is approximately 4 times larger than the DOS term at $T = 94.2$ K and 2–3 times smaller than the DOS term at 112 K. If the DOS term is excluded, the theory can still describe experimental data up to 100 K, but we do not obtain the change of sign.

From the results for J and τ, one can calculate quantities that are more easily compared with other studies. Using the relation $\xi_{ab}^2(T = 0\text{ K}) = \eta(T_c)$ [13], one finds the in-plane coherence length, $\xi_{ab}(T = 0\text{ K}) = 12$ Å for sample A and 14 Å for sample B. The out-of-plane coherence length can be calculated from $r = 4k_B^2\eta J^2/\hbar^2\nu_F^2$ and $r(T_c) = 4\xi_c^2(0)/s^2$ [13], which gives $\xi_c(T = 0\text{ K}) =$ 1.8 Å for sample A and 2.4 Å for sample B. These values are in good agreement with results of several in-plane fluctuation magnetoconductivity measurements $(I\|ab)$, summarized in Ref. [6] as $\xi_{ab}(0) = 13.6 \pm 2$ Å and $\xi_c(0) = 2.0 \pm 0.5$ Å. Further, the normal-state conductivity anisotropy can be calculated as [13] $\sigma_{ab}/\sigma_c = \nu_F^2\hbar^2/k_B^2J^2s^2 \approx 40$, in rough agreement with direct measurements in, e.g., Ref. [24].

We briefly discuss alternative interpretations of the experiments. Weak localization may cause a temperature dependence of $\Delta\sigma$ as in Fig. 3. In icosahedral Al-Cu-Fe of similar resistivity, a negative $\Delta\rho(B)/\rho$ was observed in the same temperature range [25]. However, the mean free path in the c-axis direction of $YBa_2Cu_3O_{7-\delta}$ is likely smaller than the distance between planes [26], and alternatives to diffusive motion would be more appropriate, such as the hopping mechanism [13] presently considered. Another possibility is a contribution from a magnetic field depression of a pseudo gap [5]. A pseudo gap is usually expected to imply that $d\rho_c/dT < 0$, which is not observed in sample A. However, the resistivity near T_c has a slight positive deviation compared to an extrapolation of the linear temperature dependence above 200 K. This mechanism, therefore, may not be ruled out.

In summary, we have measured the c-axis magnetoconductivity of two $YBa_2Cu_3O_{7-\delta}$ single crystals above T_c. In one sample, $d\rho_c/dT > 0$ in the whole temperature interval. In both samples we observed a change of sign of $\Delta\sigma_c$ at $T_c + 10$ K. These data could be well described by a theory that considers density of states fluctuations. Approximately the correct temperature for the sign change was obtained, and the quantitative agreement below that temperature was excellent. The parameter values obtained for the two samples are within $J = 215 \pm 10$ K and $\tau = 4 \pm 1$ fs, and were found to correspond to coherence lengths that agree well with other studies.

We are grateful to S. Östlund at the Department of Solid Mechanics, Royal Institute of Technology, for access to equipment for finite element analysis. We also thank R. A. Klemm for helpful correspondence. This work has been supported by the Göran Gustafsson Foundation and the Swedish Superconductivity Consortium.

[1] N. Kumar and A. M. Jayannavar, Phys. Rev. B **45**, 5001 (1992).
[2] L. B. Ioffe, A. I. Larkin, A. A. Varlamov, and L. Yu, Phys. Rev. B **47**, 8936 (1993).
[3] A. R. Rojo and K. Levin, Phys. Rev. B **48**, 16 861 (1993).
[4] A. A. Abrikosov, Phys. Rev. B **52**, R7026 (1995).
[5] Y. F. Yan, P. Matl, J. M. Harris, and N. P. Ong, Phys. Rev. B **52**, R751 (1995).
[6] For a recent summary of several published results see, e.g., W. Holm, Ö. Rapp, C. N. L. Johnson, and U. Helmersson, Phys. Rev. B **52**, 3748 (1995).
[7] S.-K. Yip, Phys. Rev. B **41**, 2612 (1990).
[8] A. M. Gulian, Phys. Lett. A **200**, 201 (1995).
[9] K. Semba, A. Matsuda, and T. Ishii, Phys. Rev. B **49**, 10 043 (1994).
[10] J. Axnäs, W. Holm, Yu. Eltsev, and Ö. Rapp, Phys. Rev. B **53**, R3003 (1996).
[11] J. B. Bieri, K. Maki, and R. S. Thompson, Phys. Rev. B **44**, 4709 (1991).
[12] G. Balestrino, E. Milani, and A. A. Varlamov, Pis'ma Zh. Éksp. Teor. Fiz. **61**, 814 (1995) [JETP Lett. **61**, 833 (1995)].
[13] V. V. Dorin, R. A. Klemm, A. A. Varlamov, A. I. Buzdin, and D. V. Livanov, Phys. Rev. B **48**, 12 951 (1993).
[14] As in Yu. Eltsev, W. Holm, and Ö. Rapp, Phys. Rev. B **49**, 12 333 (1994), except that powders of Y_2O_3, $BaCO_3$, and CuO were used.
[15] A commercial program for finite element analysis (ANSYS) was used to solve Laplace's equation, giving ρ_c from the measured voltage between the potential contacts and previous measurements of ρ_{ab} on another crystal (SC2 of Ref. [6]). The element division of the sample was 215 hexahedra: 5 layers along the c axis, each consisting of 43 (nonequal) hexahedra.
[16] L. Forro, V. Ilakovac, J. R. Cooper, C. Ayache, and J.-Y. Henry, Phys. Rev. B **46**, 6626 (1992).
[17] K. Yamaji, Phys. Lett. **38A**, 43 (1972).
[18] R. A. Klemm, J. Low Temp. Phys. **16**, 381 (1974).
[19] K. Maki and R. S. Thompson, Phys. Rev. B **39**, 2767 (1989).
[20] C. Baraduc, V. Pagnon, A. Buzdin, J. Y. Henry, and C. Ayache, Phys. Lett. A **166**, 267 (1992).
[21] A. G. Aronov, S. Hikami, and A. I. Larkin, Phys. Rev. Lett. **62**, 965 (1989); **62**, 2336 (1989) (E).
[22] R. S. Thompson, Phys. Rev. Lett. **66**, 2280 (1991).
[23] R. A. Klemm, private communication.
[24] I. Terasaki, Y. Sato, S. Miyamoto, S. Tajima, and S. Tanaka, Phys. Rev. B **52**, 16 246 (1995).
[25] P. Lindqvist, P. Lanco, C. Berger, A. G. M. Jansen, and F. Cyrot-Lackmann, Phys. Rev. B **51**, 4796 (1995).
[26] S. Massidda, J. Yu, K. T. Park, and A. J. Freeman, Physica C **176**, 159 (1991).