Magnetoresistance of c-axis-oriented epitaxial $YBa_2Cu_3O_{7-x}$ films above T_c

Y. Matsuda, T. Hirai, and S. Komiyama
Department of Pure and Applied Science, University of Tokyo, 381 Komaba, Meguro-ku, Tokyo 153, Japan

T. Terashima and Y. Bando
Institute for Chemical Research, Kyoto University, Gokasho, Uji, Kyoto 611, Japan

K. Iijima, K. Yamamoto, and K. Hirata
Research Institute for Production Development, Sakyo-ku, Shimogamo, Kyoto 606, Japan
(Received 16 January 1989)

Magnetoresistance in high-quality single-crystal films of $YBa_2Cu_3O_{7-x}$ studied in a temperature range $T_c \lesssim T \lesssim 210$ K is presented for magnetic field configurations both parallel and perpendicular to the conducting ab plane. The data are analyzed in terms of recent theories of superconducting fluctuations considering both orbital and Zeeman effects in Aslamazov-Larkin and Maki-Thompson terms. From the analysis, the phase-breaking time of carriers is determined to be $\tau_\phi = (1 \pm 0.1) \times 10^{-13}$ sec at 100 K with a T dependence of $1/\tau_\phi \propto T^{1-2}$. $\xi_c(0) = 1.5 \pm 0.5$ Å and $\xi_{ab}(0) = 11.5 \pm 0.5$ Å are also derived for the inter- and intralayer coherence lengths.

The influence of magnetic field H on resistivity has been intensively studied in high-T_c superconducting oxides at temperatures below T_c, where T_c is the transition temperature at $H = 0$.[1] These studies have elicited theoretical discussions about a critical fluctuation[2,3] or a giant flux creep.[4] On the other hand, the study of magnetoresistance $\Delta\rho(H) = \rho(H) - \rho(0)$ carried out at T well above T_c is free from those unsettled problems, and provides a reliable method to determine important physical quantities such as the anisotropic coherence lengths, $\xi_{ab}(0)$ and $\xi_c(0)$, and the phase-breaking time τ_ϕ of carriers. It also provides a probe to study the transport mechanism in the normal state.

In the previous study of $\Delta\rho(H)$ in a polycrystal of $YBa_2Cu_3O_{7-x}$,[5] we have derived $\xi_{ab}(0)$ and $\xi_c(0)$ but could not unambiguously determine τ_ϕ because the anisotropy in $\Delta\rho(H)$ could not be studied. Although Hikita and Suzuki studied $\Delta\rho(H)$ in a bulk single crystal of $YBa_2Cu_3O_{7-x}$,[6] τ_ϕ was not obtained since the study was limited to a narrow range of $T \sim T_c$ and also limited to the condition of H normal to the conducting ab plane of the crystal. Here, we report thorough studies of $\Delta\rho(H)$ in high-quality single-crystal films of $YBa_2Cu_3O_{7-x}$ performed in a wide T range up to 210 K for H both normal and parallel to the ab plane. The data are fully analyzed in terms of recent theories of superconducting fluctuations considering both orbital and Zeeman contributions in Aslamazov-Larkin and Maki-Thompson terms.[7,8]

Measurements are made on 1000- and 3000-Å-thick films of $YBa_2Cu_3O_{7-x}$, which will be designated below as samples 1 and 2. The films were epitaxially grown on the (100) surface of single crystals of $SrTiO_3$ with an activated reactive-evaporation method.[9] Structure analyses indicate that the films are excellent single crystals with the c axis perfectly oriented perpendicular to the substrate surface.[10] To apply the four-probe method, voltage probes are prepared by evaporation of silver on rectangular-shaped samples of a size of 1.5×8.0 mm^2. Similar to the previous work,[5] $\Delta\rho(H)$ and the Hall effects are studied with a magnetic-field-modulation technique ($f = 1$ kHz, $\Delta H = \pm 50 \sim 100$ Oe), by which field-induced changes in ρ of one part in 10^7 are detectable. The resistivity ρ of samples 1 and 2, and the polycrystalline sample used in the previous work, are shown in Fig. 1 together with the Hall number V_0/R_He in the inset, where $V_0 = 174$ Å^3 is the formula unit volume and R_H is the Hall coefficient. The characteristics of the samples, including the 10–90% width of the transition ΔT_c, the $d\rho/dT$ values in the range 150–300 K, are listed in Table I. The small ρ or $d\rho/dT$ values of samples 1 and 2 indicate the excellent characteristics of these film samples.

Direct recorder traces of the derivative of the magnetoconductance with respect to H, $-d\sigma/dH = (d\rho/dH)/\rho^2$, are shown as a function of H in Fig. 2 for sample 2. Here, ϵ denotes the reduced temperature $\epsilon = (T - T_c)/T_c$. The conductances $\sigma_\perp$ and $\sigma_\parallel$ refer, respectively, to the quantities studied under H perpendicular and parallel to the ab plane. Either of the quantities rapidly decreases with increasing ϵ, as will be elucidated in Fig. 4. The curves of

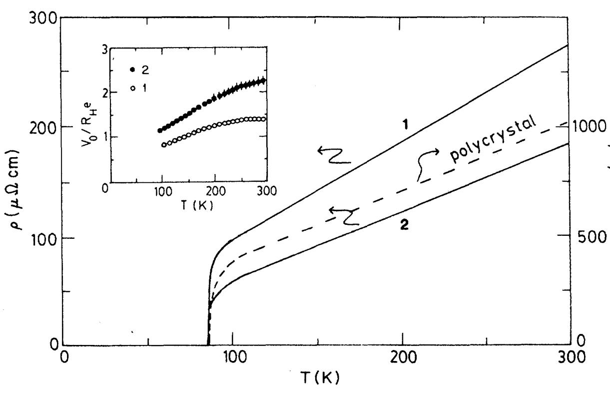

FIG. 1. Resistivity vs temperature in film samples 1 and 2 and a polycrystalline sample. The inset shows the Hall number in samples 1 and 2.

TABLE I. Characteristics of the samples. T_c and C are derived from the present analysis.

Sample	T_c (K)	ΔT_c (K)	ρ ($\mu\Omega$ cm) at 100 K	$d\rho/dT$ ($\mu\Omega$ cm K^{-1})	C	μ_H (cm^2/V sec) at 100 K
1 (1000 Å)	85.5	1.3	96	0.71	1.6 ± 0.1	14.7
2 (3000 Å)	85.5	1.3	68	0.63	1.2 ± 0.1	14.5
polycrystal	92.0	$\lesssim 0.2$	412	2.50	~ 6.8	4.4

$d\sigma_\perp/dH$ vs H deviate appreciably from straight lines at small ϵ below $\epsilon=0.06$, while they are well described by straight lines at larger ϵ implying the dependence of $\Delta\rho(H)\propto\Delta\sigma(H)\propto H^2$.

In order to compare the data of different samples, the relative magnetoresistance $\Delta\rho_\perp/\rho$ at $H=10$ kOe in the two-film samples and the earlier data $\Delta\rho/\rho$ of a polycrystalline sample are shown together as a function of ϵ in Fig. 3. Here, we deduce $\Delta\rho$ from the relation $\Delta\rho=(d\rho/dH)_{10\,\mathrm{kOe}}(10\text{ kOe}/2)$. In spite of relatively large differences in the ρ values among different samples, $\Delta\rho/\rho$ values nearly coincide and yield a single line. This supports the argument of Oh *et al.* that the sample-dependent ρ values arise from "C factors", which reflect inhomogeneous current flows in the samples.[11] Further, the sample-independent divergent behavior of $\Delta\rho/\rho$ in a small ϵ range strongly indicates that it is of an intrinsic nature of the material and arises from superconducting fluctuation. Further, a small and T-independent component remains in $\Delta\rho/\rho$ at higher ϵ in all of the samples. Figures 4(a) and 4(b) display ϵ dependences of $\Delta\sigma_\perp(H)$ and $\Delta\sigma_\parallel(H)$ at $H=10$ kOe in sample 2. Here, we have deduced $\Delta\sigma$ from the extrapolated slopes of the $d\sigma/dH$ vs H curves at the low-H limit, so that the data points represent the H^2 term in the entire ϵ range.

The superconducting fluctuations are described by the combination of Aslamazov-Larkin (AL) and Maki-Thompson (MT) terms. Orbital contributions of AL and MT terms to $\Delta\sigma_\perp(H)$ in layered compounds have been derived by Hikami and Larkin[7] by taking account of the lowest-order term of the order parameter $|\psi|^2$. Tsuneto[2] and Ikeda,[3] have considered the AL term including the higher-order term $|\psi|^4$. $\Delta\sigma_\parallel$ has not been studied in either work. Here we compare our data of $\Delta\sigma_\perp$ with the Hikami-Larkin results, the treatment of which suffices for our work in a T range well above T_c: The orbital (ALO) and the MT-orbital (MTO) terms in Ref. 7 read as

$$\Delta\sigma_{\mathrm{ALO}}=-\frac{e^2}{64\hbar d\epsilon^3}\frac{2+4\alpha+3\alpha^2}{(1+2\alpha)^{5/2}}h^2, \tag{1}$$

$$\Delta\sigma_{\mathrm{MTO}}=-\frac{e^2}{48\hbar d(1-\alpha/\delta)\epsilon^3}\times\left[\frac{\delta^2}{\alpha^2}\frac{1+\delta}{(1+2\delta)^{3/2}}-\frac{1+\alpha}{(1+2\alpha)^{3/2}}\right]h^2, \tag{2}$$

where $\alpha=2\xi_c^2(0)/(d^2\epsilon)$, $h=2e\xi_{ab}^2(0)H/\hbar$, and $\delta=16\xi_c^2(0)k_BT\tau_\phi/(\pi d^2\hbar)$ with the interlayer spacing d (11.7 Å), the coherence lengths normal to the ab plane $\xi_c(0)$ and parallel to the ab plane $\xi_{ab}(0)$, the phase breaking time τ_ϕ, the Boltzmann constant k_B, and the unit charge e. The higher-order terms in H, not included in (1) and (2), are also derived in Ref. 7. In addition to the orbital contributions above, Aronov, Hikami, and Larkin recently pointed out that the Zeeman effect adds new contributions to AL and MT terms (ALZ and the MTZ, respectively) and obtained the following terms, which are independent of the orientation of H:[8]

$$\Delta\sigma_{\mathrm{ALZ}}=-0.526\frac{e^2}{\hbar d\epsilon^2}\frac{1+\alpha}{(1+2\alpha)^{3/2}}\left[\frac{\omega_s}{4\pi k_BT_c}\right]^2, \tag{3}$$

$$\Delta\sigma_{\mathrm{MTZ}}=-\frac{e^2}{16\hbar d\epsilon}\left[\frac{1+\delta}{(1+2\delta)^{3/2}}-\frac{1+\delta+\delta/\alpha}{[(1+\delta/\alpha)(1+2\delta+\delta/\alpha)]^{3/2}}\right]\left[\frac{\omega_s\tau_\phi}{\hbar}\right]^2. \tag{4}$$

Here, $\omega_s=g\mu_BH$ is the Zeeman energy with the g factor and the Bohr magneton μ_B. Nominally, T_c and the factor C are adjustable parameters in the analysis here. However, the arbitrariness of these quantities is, in fact, small in our film samples and does not significantly affect the analysis below because of the relatively small ΔT_c and the small ρ values of the samples: T_c and C determined so as to yield the best fit in the following analysis, given in Table I, substantially agree, respectively, with the values obtained from the linear extrapolation of ρ to zero in the $\rho(T)$ curves,[12] and with $C=1.5$ (sample 1) and 1.2 (sample 2) derived from the observed $d\rho/dT$ values.

Up to the present time, Zeeman effects on superconducting fluctuations have not been observed experimentally in any material, because it is usually negligibly small in comparison to ALO and MTO terms. However, in either of the samples 1 and 2 here, the complete data of $\Delta\sigma_{\parallel,\perp}(H)$ and $\Delta\sigma_{\parallel,\perp}(\epsilon)$ as shown in Figs. 2 and 4 cannot be consistently explained by any combination of $\xi_{ab}(0)$, $\xi_c(0)$, and τ_ϕ unless the Zeeman terms given by Eqs. (3) and (4) are included in the analysis. We first compare the data of $\Delta\sigma_\parallel(\epsilon)$ with the ALZ and the MTZ terms assuming that orbital contributions to $\Delta\sigma_\parallel$ are negligibly small. Since either of the Zeeman terms is independent of $\xi_{ab}(0)$ and insensitive to $\xi_c(0)$, τ_ϕ can be thereby determined almost uniquely. The ϵ dependence of $\Delta\sigma_\parallel$ is not explained well if τ_ϕ is assumed to be T independent, as indicated by a dash-dotted line in Fig. 4(b), but is satisfactorily ex-

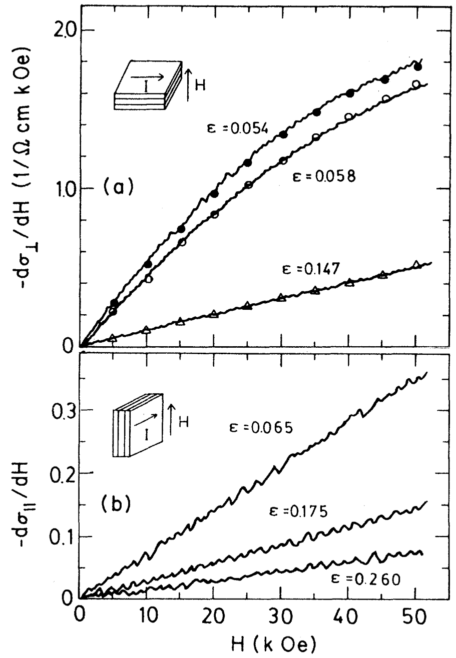

FIG. 2. Direct recorder traces of the anisotropic magnetoconductances, $d\sigma_\perp/dH$ and $d\sigma_\parallel/dH$, vs H in sample 2. Solid circles, open circles, and triangles in (a) indicate theoretical values for respective temperatures; the parameters used are $\xi_c(0)=1.7$ Å, $\xi_{ab}(0)=11.0$ Å, $\tau_\phi(100\text{ K})=1.05\times10^{-13}$ sec with $\tau_\phi\propto(1/T)^{1.0}$, $C=1.2$, and $T_c=85.5$ K.

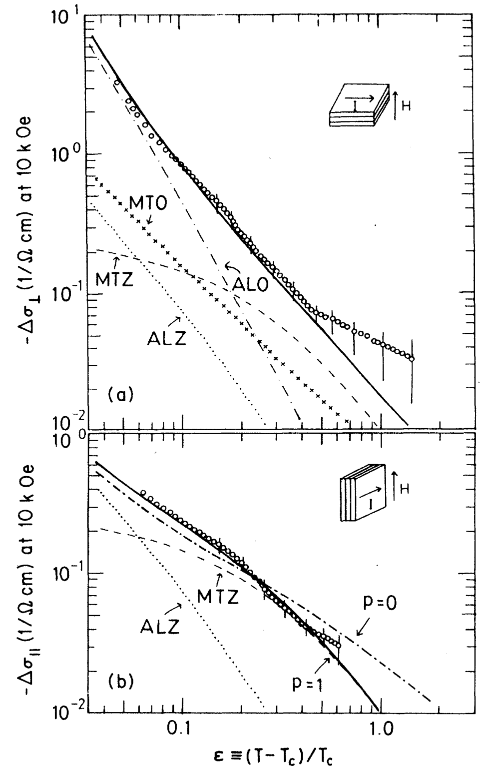

FIG. 4. Anisotropic magnetoconductances, $-\Delta\sigma=\sigma(0)-\sigma(10\text{ kOe})$, vs ϵ in sample 2. Different lines represent contributions from different origins, calculated from Eqs. (1)–(4) with the same parameter values as those used for Fig. 2(a), except for $\xi_c(0)=1.5$ Å and $\xi_{ab}(0)=11.5$ Å. The solid line indicates the sum of the different contributions. The dash-dotted line in (b) represents the sum obtained on the assumption of the T-independent τ_ϕ of $\tau_\phi=1.05\times10^{-13}$ sec.

plained if a T dependence of the form $\tau_\phi\propto(1/T)^p$ with $p=1.0$–2.0 is assumed: The solid line in Fig. 4(b), representing the sum of the Zeeman terms with $\tau_\phi(100\text{ K})=1.05\times10^{-13}$ sec and $p=1.0$, reproduces well the experimental data. [We assume $g=2.0$. A reasonable fit is obtained also with τ_ϕ (100 K) $=1\times10^{-13}$ sec and $p=2.0$.] The result is similar in sample 1. Since orbital contributions to $\Delta\sigma_\parallel$ are supposed to be smaller than those to $\Delta\sigma_\perp$ by a factor of $[\xi_{ab}(0)/\xi_c(0)]^2$, the analysis above ignoring the orbital contributions is validated by the analysis below, which derives $\xi_{ab}(0)/\xi_c(0)\sim8$.[8] We would note

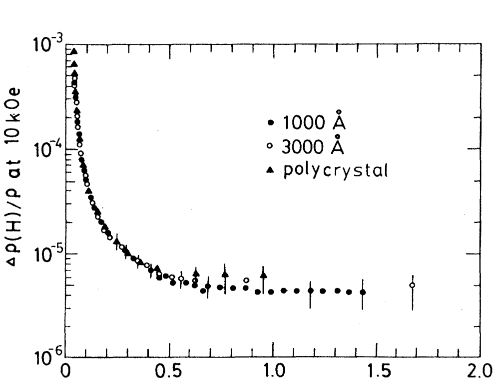

FIG. 3. Relative magnetoresistance, $\Delta\rho/\rho=[\rho(10\text{ kOe})-\rho(0)]/\rho(0)$, vs reduced temperature ϵ. $\Delta\rho=\Delta\rho_\perp$ for film samples.

that the presence of Zeeman effects in the present material indicates that the superconductivity here is "BCS-like" in the sense that the superconducting pairs are in a singlet state composed of particles *with spin.*

With τ_ϕ given as above, we analyze $d\sigma_\perp(H)/dH$ and $\Delta\sigma_\perp(\epsilon)$ in terms of the combination of ALO, MTO, ALZ, and MTZ terms. The theoretical values of $d\sigma_\perp/dH$ indicated in Fig. 2(a), which are calculated from Eqs. (1)–(4) by including the higher-order terms of H with $\xi_{ab}(0)=11.0$ Å and $\xi_c(0)=1.7$ Å, agree well with the experimental curves. The data of $\Delta\sigma_\perp(\epsilon)$ in the same sample are best fitted to the theoretical values also with similar values of $\xi_{ab}(0)$ and $\xi_c(0)$ as shown by a solid line in Fig. 4(a).[13] Similar values of τ_ϕ, $\xi_{ab}(0)$, and $\xi_c(0)$ are obtained for sample 1 as listed in Table II. Our analysis here indicates that none of the MTO, ALZ, and MTZ terms can be neglected except at very small ϵ. This fact questions existing studies of the superconducting fluctuations,[6,11,14–16] where only the ALO term was taken into account.

In our previous work on the sintered sample, where a completely random orientation of the c axis in the sample was assumed and the Zeeman terms were not included in the analysis, we obtained $\xi_{ab}(0)\sim16$ Å, $\xi_c(0)\sim2$ Å, and

TABLE II. Physical quantities derived in the present work.

	ξ_{ab} (Å)	ξ_c (Å)	τ_ϕ (10^{-13} sec) at 100 K	$1/\tau_\phi \propto T_p$
1	11.5 ± 0.5	1.5 ± 0.5	1.0 ± 0.1	$p=1-2$
2	11.2 ± 0.3	1.6 ± 0.3	1.0 ± 0.1	$p=1-2$

$\tau_\phi \lesssim 3\times10^{-14}$ sec.[5] However, we have found that an effect of a preferred orientation of the c axis is crucial in the determination of τ_ϕ. We have confirmed that the parameters determined here for film samples explain the earlier data of the sintered sample equally well, if the Zeeman terms are included and a preferred orientation is appropriately assumed. The values of $\xi_{ab}(0)$ and $\xi_c(0)$ derived here are also close to those reported by Oh *et al.*[11] and Hikita and Suzuki.[6]

We wish to consider physical implication of the short coherence length $\xi_{ab}(0)$ within a simple two-dimensional free-carrier picture. The probable hole density $n=(0.5-1)/V_0=(3-6)\times10^{21}/\text{cm}^3$ together with the corrected resistivity $\rho'=\rho/C\sim57$ Ω cm at $T=100$ K suggest $\mu=38-19$ cm^2/V sec for the mobility of holes.[17] Hence the mean free path $l=v_F\tau$ of holes at 100 K is estimated to be 110–78 Å from the relation $l=(\hbar\mu/e)\times(2\pi n_s)^{1/2}$ with $n_s=nd=(3.4-7)\times10^{14}/\text{cm}^2$. Since $l(0\text{ K})\gg l(100\text{ K})$ is expected, the inequality $l(100\text{ K})\gg\xi_{ab}(0)$ certifies that the present material is a clean-limit type-II superconductor, and strongly suggests that the measured $\xi_{ab}(0)$ is nothing but the BCS coherence length $\xi_{\text{BCS}}=(\hbar/\pi)(v_F/\Delta_0)$. From $\xi_{\text{BCS}}=11.5$ Å, together with an assumption of gap parameters of $2\Delta_0/k_BT_c=3.5-8$,[18,19] we have a Fermi velocity of $v_F=(1.6-0.7)\times10^7$ cm/s, the scattering time of $\tau=l/v_F=(0.5-1.6)\times10^{-13}$ sec, and an effective hole mass of $m^*/m_0=e\tau/\mu m_0=3.0-6.6$. The fact that $\tau\sim\tau_\phi$ strongly suggests that the scattering mechanism causing the resistivity is, by itself, of phase-breaking property. This, as well as the enhanced mass values, are consistent with a recent analysis of the optical reflection inferring that τ and m^* are strongly frequency dependent.[19,20]

$\tau_\phi(100\text{ K})=1\times10^{-13}$ sec implies $\hbar/\tau_\phi(100\text{ K})\sim6.6$ meV$=0.77k_BT$ and indicates a moderately strong pair breaking. According to a standard theory,[21] this implies a reduction in T_c by an amount of $T_c/T_{c0}=0.7$ where T_{c0} refers to the transition temperature in the absence of the pair-breaking effect. Hence, if $2\Delta_0/kT_{c0}=3.53$ is assumed, the ratio $2\Delta_0/kT_c$ would be enhanced to five, which is of comparable magnitude to those of ordinary strong-coupling BCS superconductors such as Pb and Hg.[22]

In the higher T range where $\epsilon\gtrsim0.5$, the data points of $\Delta\sigma_\perp(\epsilon)$ in Fig. 4(a) deviate from the theoretical line and give larger values. This arises from a temperature-independent component of $\Delta\rho/\rho$ seen in Fig. 3. The origin of this $\Delta\rho/\rho$ is not likely to be a classical magnetoresistance, since classical $\Delta\rho(H)$ would have a strong T dependence such that $\Delta\rho/\rho\sim(\mu_H)^2\propto T^{-2}$. It would be interesting to note that $\tau\sim\tau_\phi=1\times10^{-13}$ sec is comparable to the phonon scattering time of electrons at $T\sim100$ K in ordinary metals such as pure copper.[23] However, the presence of the T-independent component of $\Delta\rho/\rho$ may not be simply explained by the phonon scattering.

In conclusion, Zeeman effects have been observed, for the first time, in the superconducting fluctuation, evidencing that the superconducting pairs are in a singlet state composed of particles with spin. The observation enabled us to determine τ_ϕ and its probable T dependence.

---

[1] See, for example, Y. Iye, T. Tamegai, H. Takeya, and H. Takei, Jpn. J. Appl. Phys. **26**, L1057 (1987).

[2] T. Tsuneto, J. Phys. Soc. Jpn. **57**, 3499 (1988).

[3] R. Ikeda, J. Phys. Soc. Jpn. **58**, 1906 (1989).

[4] M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).

[5] Y. Matsuda, T. Hirai, and S. Komiyama, Solid State Commun. **68**, 103 (1988).

[6] M. Hikita and M. Suzuki, Phys. Rev. B **39**, 4756 (1989).

[7] S. Hikami and A. I. Larkin, Mod. Phys. Lett. B **2**, 693 (1988).

[8] A. G. Aronov, S. Hikami, and A. J. Larkin, Phys. Rev. Lett. **62**, 965 (1989).

[9] T. Terashima, K. Iijima, K. Yamamoto, Y. Bando, and H. Mazaki, Jpn. J. Appl. Phys. **27**, L91 (1988).

[10] K. Kamigaki, H. Terauchi, T. Tereshima, K. Iijima, K. Hirata, and Y. Bando, Jpn. J. Appl. Phys. **27**, L1899 (1988).

[11] B. Oh, K. Char, A. D. Kent, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond, A. Kapitulnik, and J. M. Graybeal, Phys. Rev. B **37**, 7861 (1988).

[12] Actually, T_c of the film samples in Table I are 0.2–0.3 K lower than those obtained from the ρ vs T curves, suggesting a slight inhomogeneity of T_c in the samples.

[13] Actually, the values of $\xi_{ab}(0)$ and $\xi_c(0)$ derived from the data in Fig. 2 are slightly different from those determined from the data of Fig. 4. The small discrepancies may suggest an effect of a slight T_c distribution in the samples.

[14] P. P. Freitas, C. C. Tsuei, and T. S. Plaskett, Phys. Rev. B **36**, 333 (1987).

[15] S. E. Inderhees, M. B. Salamon, N. Goldenfeld, J. P. Rice, B. G. Pazol, D. M. Ginsberg, J. Z. Liu, and G. W. Crebtree, Phys. Rev. Lett. **60**, 1178 (1988).

[16] K. Kanoda, T. Kawagoe, M. Hasumi, T. Takahashi, S. Kagoshima, and T. Mizogichi, J. Phys. Soc. Jpn. **57**, 1554 (1988).

[17] This is not inconsistent with the measured Hall mobilities $\mu_H=R_H/\rho\sim15$ cm^2/V sec, since μ_H is supposed to be lower than the true mobility because of an effect of the C factor and a presently unknown mechanism of the T-dependent R_H.

[18] Z. Schlesinger, R. T. Collins, D. L. Kaiser, and F. Holzberg, Phys. Rev. Lett. **59**, 1958 (1987).

[19] G. A. Thomas, J. Orenstein, D. H. Ropkine, M. Capizzi, A. J. Millis, R. N. Bhatt, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **61**, 1313 (1988).

[20] R. T. Collins, Z. Schlesinger, D. L. Kaiser, F. Holzberg, G. V. Chandrashkhar, and M. W. Shafer, *Fourth International Conference on Infrared Physics, Zurich, 1988* (Pergamon, Oxford, in press).

[21] K. Maki, in *Superconductivity,* edited by R. D. Parks (Marcel Dekker, New York, 1969), p. 1035.

[22] D. J. Scalapino, in *Superconductivity,* edited by R. D. Parks (Marcel Dekker, New York, 1969), p. 540.

[23] C. Kittel, *Introduction to Solid State Physics,* 6th ed. (Wiley, New York, 1986), p. 143.