c-axis magnetoconductivity of anisotropic superconducting single crystals: The density-of-states fluctuation scenario

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The *c*-axis magnetoconductivity (B||c||I) of $(Tl, Hg)_2Ba_2Ca_2Cu_3O_{10+\delta}$ (Tl 2223) and $(Hg, Cu)Ba_2CuO_{4+\delta}$ (Hg 1201) single crystals has been measured. For Tl 2223, the observed change of sign in magnetoconductivity is explained in terms of density-of-states (DOS) fluctuations and competition of this effect with the positive Aslamasov-Larkin contribution. The mercury-based compound (Hg 1201) does not show such an anomaly. Indeed, in accordance with the theory, owing to its low anisotropy, this material shows a vanishing DOS contribution preventing any change of sign in the experimental magnetoconductivity. Besides, the nature of the impurity state and the pair-breaking regime are discussed. [S0163-1829(99)03010-6]

The *c*-axis transport properties are powerful fields of investigation because they provide insight into the nature of the normal state and the origins of the anomalous superconducting properties of highly anisotropic, layered superconductors. The occurrence of a maximum in the temperature dependence of the *c*-axis resistivity near the edge of the superconducting transition is a common feature of the cuprates or organic superconductors.^{1,2} This peak was observed to have a strong field dependence with its position shifting to lower temperatures when the field increased.^{3–5} Although this has unambiguously been seen in bismuth^{6–8} and in thallium⁹-based compounds, the situation is less clear in Y-Ba-Cu-O (Refs. 10,11) and mercury¹²-based compounds.

The *c*-axis resistivity peak seems to be closely related to the anisotropy and to the doping level of the samples.^{13,14} Indeed, this peak is very pronounced in highly anisotropic and/or underdoped samples and is almost absent in overdoped samples having low anisotropy. Various theories have been proposed regarding this topic, either considering only the normal-state properties and describing the peak by means of nonmetallic normal-state conduction mechanisms^{17,15-17} or treating the normal and superconducting states together. $^{3,18-20}$ In the latter, Briceno et *al.*³ and Gray and Kim²⁰ account for the maximum of the *c*-axis resistivity assuming a competition between the decrease of quasiparticles tunneling with decreasing temperature (suppressed by the superconducting gap below T_c) and the appearence of interlayer currents due to Josephson-like shorts in magnetic fields. Besides, Ioffe et al.¹⁸ and Dorin et al.¹⁹ have evoked the role of superconducting fluctuations. This fluctuation scenario indeed gives satisfactory results and matches the experimental behavior in many papers.^{5,8,21} The authors of the theory have shown that the main effect of these fluctuations is to create a virtual gap in the electronic spectrum which reduces the quasiparticle density of states (DOS), and thus, decreases the one-electron conductivity at the edge of the superconducting transition. The *c*-axis resistivity peak is then induced by the negative contribution to conductivity arising from this fluctuation reduction of DOS.

Measurements of the magnetoconductivity, $\Delta \sigma$ $=\sigma(B,T)-\sigma(0,T)=1/\rho(B,T)-1/\rho(0,T)$ is a great experimental technique to investigate the superconducting fluctuations and related problems such as the singular negative magnetoresitivity.^{10,11,22} Comparison with theory¹⁹ does not involve assumptions concerning either a clean or a dirty state, or a strong or a weak pair-breaking regime of the material. Thus, a quantitative analysis can give reliable estimates of parameters such as the phase pair-breaking lifetime (τ_{Φ}) , the scattering lifetime (τ) , and the Fermi velocity (v_F) . This may allow us to discuss the nature of the impurity state, the impurity assisted character of the *c*-axis conductivity, and the pairing state in high-temperature superconductors HTSC's.

In the present study, we have measured *c*-axis resistivity under various magnetic fields for two compounds: $(Tl, Hg)_2Ba_2Ca_2Cu_3O_{10+\delta}$ (Tl 2223) and $(Hg, Cu)Ba_2CuO_{4+\delta}$ (Hg 1201). For Tl 2223, the magnetoconductivity shows a change of sign from positive near T_c to negative above T_c , whereas such an anomaly is not observed for Hg 1201. We account for this difference by considering the fluctuation-induced decrease in the normal density of states, as evoked by Dorin et *al.*¹⁹ and which is known to be strongly dependent on the interlayer coupling.

Crystal growth of $(Tl, Hg)_2Ba_2Ca_2Cu_3O_{10+\delta}$ and $(Hg, Cu)Ba_2CuO_{4+\delta}$ is reported elsewhere.^{12,23} These crystals have a doping level close to the optimum with respect to their critical temperatures. For each phase, two high-quality crystals were extracted from the batches. They are denoted as HgA and HgB for Hg 1201 and TlA and TlB for Tl 2223. This will allow us to check the correctness of our analysis for each phase by showing that our results are not sample dependent.

The crystals, which had typical dimensions $1 \times 1 \times 0.1 \text{ mm}^3$, were contacted in the direct cross configuration. The measured R_c was transformed to ρ_c using the crystal dimensions; this has been shown to yield reliable values of ρ_c in comparison to more sophisticated multiterminal

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FIG. 1. Normalized *c*-axis resistivity vs temperature at different magnetic fields for Tl 2223 (Sample A and Sample B).

configurations.^{24,25} Indeed, owing to the large anisotropy, the planes perpendicular to the current flow can be regarded as equipotential surfaces making a decomposition analysis unnecessary. Gold wires were attached to the "evaporated-silver stripes" with silver paint. The samples were then annealed in air at 400°C for 10 min.

The *c*-axis resistivity was measured as a function of temperature at a series of fixed fields (B||c||I). Distinct behaviors are shown for Tl 2223 and Hg 1201 (Figs. 1 and 2, respectively). For the Tl 2223, a pronounced maximum of resistivity at zero field and a strong shift of the zero resistance temperature in magnetic fields ($\Delta T_c \approx 70$ K for B = 7 T) are observed. In addition, a large field-induced broadening of the zero-field resistivity peak is seen. A smaller maximum is shown for Hg 1201 and the broadening is not as marked as in the above case. As developed below, such a difference and its consequences in terms of magneto-conductivity may be understood in the framework of the DOS fluctuation contribution to conductivity.

Hereafter, this study is focused on magnetoconductivity data. Experimentally, a striking but well-known behavior is observed for Tl 2223: a change of sign occurs for both samples TlA and TlB. (see for instance Fig. 3 for sample TlA measured at various fields). This anomalous magnetoconductivity is a common feature of very anisotropic materials such as Bi 2212.^{11,22} It has to be pointed out that although Y-Ba-



FIG. 2. Normalized *c*-axis resistivity vs temperature at different magnetic fields for Hg 1201 (Sample A and Sample B).



FIG. 3. Magnetoconductivity vs temperature at different magnetic fields (7, 5, 3, and 1 T) for Tl 2223 (Sample A).

Cu-O usually shows a weaker maximum in the *c*-axis resistivity, a change of sign in the measured magnetoconductivity may also be observed.¹⁰ Hg 1201, like Y-Ba-Cu-O, exhibits a rather small peak. However, for both studied crystals (HgA and HgB), no change of sign is evidenced.

As it was shown in the framework of a Gaussian model developed by Ioffe et al.,¹⁸ and Dorin et al.,¹⁹ c-axis conductivity fluctuations in high-temperature superconductivity is comprised of four terms. The direct contribution, initially proposed by Aslamasov and Larkin (AL),²⁶ is due to the acceleration in an electric field of short-lived Cooper pairs in thermal nonequilibrum. On the basis of various conditions, alternative expressions of this contribution have been derived.^{27,28} The DOS contribution arises from corrections to the normal quasiparticle density of states owing to fluctuations of normal quasiparticles into the superconducting state. This contribution is expected to be negative in sign since the DOS contribution in a magnetic field causes a decrease of resistivity. It is possible to observe a negative-fluctuationinduced *c*-axis magnetoresistance in the temperature region where the DOS contribution exceeds the positive AL one. The regular and anomalous Maki-Thompson (MT) contributions, respectively, result from the scattering of the normalstate particles and the superconducting pairs.^{19,28} Those contributions are usually small and in the following, the main issue is to know the relevancy of this contribution to describe the experimental results since they are not considered in many papers.^{29,30} In this paper, the interactions of the magnetic field with electron spins leading to a Zeeman effect are neglected. Finally, the fluctuation magnetoconductivity is given by

$$\Delta \sigma_c = \Delta \sigma_c^{\text{AL}} + \Delta \sigma_c^{\text{DOS}} + \Delta \sigma_c^{\text{MT(reg)}} + \Delta \sigma_c^{\text{MT(an)}}, \quad (1)$$

where $\Delta \sigma_c^{AL} = \sigma_c^{AL}(B,T) - \sigma_c^{AL}(0,T)$, etc...

The weak-field expression of Ref. 18 is valid for our field strength in Tl 2223 and Hg 1201. The resulting expressions for the four terms are then

$$\Delta \sigma_c^{\rm AL}(B,T) = -\frac{e^2 s}{32 \,\eta \hbar} \left[\frac{\beta^2 r^2(\epsilon + r/2)}{32 [\epsilon(\epsilon + r)]^{5/2}} \right],\tag{2}$$

$$\Delta \sigma_c^{\text{DOS}}(B,T) = \frac{e^2 s r \kappa}{16 \eta \hbar} \left[\frac{\beta^2 (\epsilon + r/2)}{24 [\epsilon (\epsilon + r)]^{3/2}} \right], \tag{3}$$

$$\Delta \sigma_c^{\mathrm{MT(reg)}}(B,T) = \frac{e^2 s r \tilde{\kappa}}{16 \eta \hbar} \left[\frac{\beta^2 r}{48 [\epsilon(\epsilon+r)]^{3/2}} \right],\tag{4}$$

$$\Delta\sigma_{c}^{\mathrm{MT(an)}}(B,T) = -\frac{e^{2}s}{16\eta\hbar} \left[\frac{\beta^{2}r^{2}(\epsilon+\gamma+r)\{\epsilon(\epsilon+r)+\gamma(\gamma+r)+[\epsilon(\epsilon+r)\gamma(\gamma+r)]^{1/2}\}}{96[\epsilon(\epsilon+r)\gamma(\gamma+r)]^{3/2}\{[\epsilon(\epsilon+r)]^{1/2}+[\gamma(\gamma+r)]^{1/2}\}} \right].$$
(5)

In these formulas, e is the electron charge, s is the lattice period along the c axis, and

$$\eta = -\frac{v_F^2 \tau^2}{2} \bigg[\Psi \bigg(\frac{1}{2} + \frac{\hbar}{4 \pi k_B T \tau} \bigg) - \Psi \bigg(\frac{1}{2} \bigg) - \frac{\hbar}{4 \pi k_B T \tau} \Psi' \bigg(\frac{1}{2} \bigg) \bigg],$$
(6)

where v_F is the Fermi velocity parallel to the layers, τ is the quasiparticle scattering time, $\Psi(x)$ and $\Psi'(x)$ are the digamma function and its derivative. $r=4 \eta J^2 k_B^2 / v_F^2 \hbar^2$ is the usual anisotropy parameter characterizing the dimensionality of the fluctuations and J is an effective interlayer energy in Kelvin. $\beta = 4 \eta e B/\hbar$ and $\epsilon = \ln(T/T_c)$. The constant κ and $\tilde{\kappa}$ are ruled by the impurity concentration and are function of τT :

$$\kappa = \frac{-\Psi'(\frac{1}{2} + \hbar/4\pi k_B T \tau) + (\hbar/2\pi k_B T \tau)\Psi''(\frac{1}{2})}{\pi^2 [\Psi(\frac{1}{2} + \hbar/4\pi k_B T \tau) - \Psi(\frac{1}{2}) - (\hbar/4\pi k_B T \tau)\Psi'(\frac{1}{2})]},$$
(7)
$$\tilde{\kappa} = \frac{-\Psi'(\frac{1}{2} + \hbar/4\pi k_B T \tau) + \Psi'(\frac{1}{2}) + (\hbar/4\pi k_B T \tau)\Psi''(\frac{1}{2})}{\pi^2 [\Psi(\frac{1}{2} + \hbar/4\pi k_B T \tau) - \Psi(\frac{1}{2}) - (\hbar/4\pi k_B T \tau)\Psi'(\frac{1}{2})]},$$
(8)

and

$$\gamma = \frac{2\eta}{v_F^2 \tau \tau_{\star}},\tag{9}$$

where τ_{Φ} is the pair-breaking lifetime. Since $\epsilon \ll 1$ is assumed in the theory, the normal state magnetoconductivity can be neglected since the fluctuations are large close to T_c . It has to be pointed out that the theory only accounts for a Gaussian conductivity fluctuation arising above the mean-field superconducting transition temperature. The critical fluctuations and the effects of the vortex structure on the broadening of the transition are not considered in the above description.

The first step of our analysis is to calculate numerically the prediction of the theory in order to compare with experimental data. In the comparison we took s = c/2 = 1.5 nm for Tl 2223 and s = c = 0.95 nm for Hg 1201. In the following, the result of the analysis is shown for 3 T. T_c was obtained from the midpoint of the resistive transitions and we assumed τ and $\tau_{\phi} \propto 1/T$. Figures 4(a) and Fig. 4(b) (for samples TlA and TlB, respectively) show that the weak-field theoretical predictions¹⁹ for magnetoconductivity agree with experimental data when including the fours contributions: AL, DOS, and both MT terms. In particular, the change of sign is obtained at the right temperature and the magnitude of the minimum is perfectly predicted. [See enlargements in the insets of Fig. 4a and Fig. 4(b)]. This agreement is obtained for the two samples (TIA and TIB) using the following parameters: J=4 K, $\tau(100 \text{ K}) = \tau_{\Phi}(100 \text{ K}) \approx 5.1 \times 10^{-14}$ s for both TIA and TIB and $v_F=4.4\times 10^6$ cm/s or the latter and 4.2×10^6 cm/s for the former. Such values can be trusted since there exists a very weak discrepancy from sample to sample. The same kind of analysis has been performed on the positive magnetoconductivity of Hg 1201. In this case, no change of sign is observed suggesting a vanishing (DOS+MT regular) contribution compared to the positive (AL+MT anomalous) one. This is indeed checked by numerical calculations where the (DOS+MT regular) contribution is shown to be negligibly small. On Fig. 5, the prediction of the AL+MT anomalous contributions are com-



FIG. 4. (a) Magnetoconductivity vs temperature at 3 T for Tl 2223 (Sample A). The solid line represents the theoretical calculation with parameters given in the text. The symbols are the experimental magnetoconductivity $\Delta \sigma_c$ ($B \| c \| I$). (b) Magnetoconductivity vs temperature at 3 T for Tl 2223 (Sample B). The solid line represents the theoretical calculation with parameters given in the text. The symbols are the experimental magnetoconductivity $\Delta \sigma_c$ ($B \| c \| I$).



FIG. 5. Magnetoconductivity vs temperature at 3T for Hg 1201 (Samples A and B). The solid line represents the theoretical calculation with parameters given in the text. The symbols are the experimental magnetoconductivity $\Delta \sigma_c$ ($B \| c \| I$).

pared with experiments. As expected, the use of this sole contribution gives a satisfactory result using the parameters : J=40 K, $\tau(100$ K)= 2×10^{-14} s, $\tau_{\Phi}(100$ K)= 2×10^{-13} s, and $v_F=2 \times 10^6$ cm/s for both HgA and HgB samples. Once again, no discrepancy is observed in the values of the parameters between samples.

The values of the obtained parameters cannot be directly compared with those obtained in other experiments on Tl 2223 and Hg 1201 single crystals. However, c-axis transport measurements on Bi-Sr-Ca-Cu-O 2212 gave τ to be in the range of $2-3 \times 10^{-14}$ s for single crystals and thin films; τ_{\pm} values varied from 2×10^{-13} s to 5×10^{-13} s. ^{6,8,13,21} Inplane measurements of Bi-Sr-Ca-Cu-O 2212 single-crystal whiskers³¹ estimated $\tau_{\Phi} \approx 7.5 \times 10^{-14}$ s, i.e., an order of magnitude lower than the above results. Very few results are reported for Y-Ba-Cu-O; nevertheless, τ is within the range $3-5 \times 10^{-15}$ s for single crystals.¹⁰ An important discrepancy is found in the literature for J values. J = 40 K and J = 15-20 K were reported for Bi-Sr-Ca-Cu-O thin films.^{13,6,8} A value of 4 K was given for an optimized single crystal.²¹ Such scattering reflects the differences in the doping level of the various samples. Finally, our results for Tl 2223 and Hg 1201 show an overall agreement with values obtained from the literature.

Let us now focus on the pair-breaking life time τ_{Φ} . τ_{Φ} only enters in the very small MT anomalous contribution; thus the uncertanity on this parameter is expected to be large. Figure 6 shows the sensitivity of the overall result upon the variation of τ_{ϕ} for Tl 2223 and Hg 1201, respectively. As noted above, the best descriptions are obtained for $\tau_{\Phi}(100 \text{ K}) \approx 5 \times 10^{-13} \text{ s} \approx \tau(100 \text{ K})$ for Tl 2223 and $\tau_{\phi}^{*}(100 \text{ K}) = 2 \times 10^{-13} \text{ s for Hg 1201. However, if we fur-}$ ther decrease the values of $\tau_{_{\Phi}}$ by an order of magnitude, the calculation is not appreciably modified. This is partly explained by the fact that $\tau_{_{\rm db}}$ directly controls the magnitude of the anomalous MT contribution which is expected to be several times smaller than the other contributions, such as AL or DOS. Hence, the determined values have to be considered as upper limits for this parameter. This result suggests that the best agreement with experiments, for Tl 2223 and Hg 1201 materials, is attained with moderate pair breaking $(4\pi k_B \tau_{\Phi} T_c/\hbar \approx 10 \text{ and } 4\pi k_B \tau_{\Phi} T_c/\hbar \approx 30, \text{ respectively}),$



FIG. 6. Variation of the 3 T theoretical calculation of magnetoconductivity (AL+MT anomalous +DOS+MT regular) for different values of the phase pair-breaking time. The other parameters are given in the text. Main panel: Tl 2223 (Sample B). Inset: Hg 1201 (Sample A).

close to the weak limit, rather than strong pair breaking. As a matter of fact, the regular and anomalous MT contributions are relatively small. This is evidenced in Fig. 7 where the different contributions are separated for Tl 2223. The overall magnitude of the anomalous MT contribution is small because of the weak pair-breaking mechanism. Obviously, this is also true for Hg 1201. Such a result is consistent with other experiments that showed the AL term alone adequately describes the fluctuation contribution to the in-plane magnetoconductivity in Bi-Sr-Ca-Cu-O.²⁹

Another important parameter that can be extracted from this analysis is the in-plane scattering time τ . As emphasized above, the theory is valid for an arbitrary impurity concentration making the determination of τ extremely reliable. For samples TIA and TIB, as well as for the HgA and HgB, an intermediate case is achieved $(4 \pi k_B \tau T_c / \hbar \approx 10$ for Tl 2223 and $4 \pi k_B \tau T_c / \hbar \approx 3$ for Hg 1201). From results for J, τ , and v_F , and using the relation $\xi_{ab}^2(T=0 \text{ K}) = \eta(T_c)$, one calculates values of ξ_{ab} and l, where ξ_{ab} and l are the BCS co-



FIG. 7. Decomposition of the calculation of theoretical magnetoconductivity for Tl 2223 (Sample A) at 3 T with parameters are given in the text. The different contributions are shown. Long dashed line: AL contribution; short dashed line: MT anomalous contribution; dotted line: DOS + MT regular contributions; solid line: total magnetoconductivity.

herence length and the intralayer mean free path, respectively. One obtains $\xi_{ab}(0) \approx 2.1$ nm for Tl 2223 and $\xi_{ab}(0) \approx 0.6$ nm for Hg 1201. This gives for both samples $l/\xi_{ab} \approx 1$. These results agree with other experiments showing that HTSC single crystals are not extremely clean when l/ξ_{ab} is of the order unity. So, the use of the clean-limit expression $(l \ge \xi_{ab})$ is not justified for these materials.

As discussed above, our single crystals (TI 2223 and Hg 1201) show moderate pair breaking with an intermediate impurity concentration (far from the usually assumed cleanlimit case). In such a case, according to Dorin *et al.*,¹⁹ the effective interlayer tunneling rate is of the order $k_B^2 J^2 \tau/\hbar^2$. For the compounds TI 2223 and Hg 1201, we have $k_B^2 J^2 \tau/\hbar^2 \ll \tau_{\Phi}^{-1} \approx \tau^{-1}$ and $k_B^2 J^2 \tau/\hbar^2 \ll \tau_{\Phi}^{-1} \ll \tau^{-1}$, respectively. This suggests that the phase coherence of paired quasiparticles is destroyed before tunneling to the neighboring layers. Moreover, in accordance with the Matthiesen rule, Dorin et *al.*¹⁹ assumed in their theory $\tau_{\Phi} \ge \tau$. This assumption is fullfilled for Hg 1201, whereas TI 2223 shows $\tau_{\Phi} = \tau$ involving scattering processes which break the time-reversal symmetry, as for example, localized magnetic moments.

As seen previously, our materials (TI 2223 and Hg 1201) are not vastly different in terms of impurity concentrations and pair-breaking mechanisms. Nevertheless, they exhibit basic differences regarding the *c*-axis resistivity peak and magnetoconductivity. The important point is that positive AL and MT anomalous contributions are smaller in materials with higher anisotropy ratio $\Gamma = (m_c/m_{ab})^{1/2}$; they are both proportional to $1/\Gamma^2$, while the negative DOS and MT regular are $1/\Gamma$. The out-of-plane coherence length can be calculated from $r(T_c) \approx 4 \xi_c^2(0)/s^2$. One obtains $\xi_c(0) \approx 0.038$ nm for TI 2223 and $\xi_c(0) \approx 0.09$ nm for Hg 1201. The resulting anisotropy ratio are then $\Gamma \approx 55$ for TI 2223

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and $\Gamma \approx 6$ for Hg 1201. Those values are slightly lower than results derived from other experiments ($\Gamma \approx 17$ for Hg 1201 through angular-resolved magnetoconductivity 32 and $\Gamma\!\approx\!70$ for Tl 2223 from magnetization measurements³³). Nevertheless, this unambiguously demonstrates the more anisotropic nature of Tl 2223 as compared to Hg 1201. Hence, we argue that the negative magnetoresistivity, on the one hand, and the great magnitude of the resistivity peak in Tl 2223 compared to Hg 1201, on the other hand, may be ascribed to the stronger anisotropy of Tl 2223. According to the theory, the weak anisotropy of Hg 1201 should involve vanishing DOS and MT regular contributions to magnetoconductivity (both compounds are at their optimum doping level). This is indeed verified by the experimental data where no change of sign in the magnetoconductivity is observed. Consequently, this could imply that the fluctuating scenario is not a universal explanation of the sharp increase of the *c*-axis resistivity above T_c and that the nonmetallic normal-state conduction mechanisms, such as activated behavior of the normal-state resistivity, might be relevant in this case.^{14,15,34} This will be dealt with in a forthcoming paper.

In summary, we have analyzed the magnetoconductivity of Tl 2223 and Hg 1201 single crystals in terms of fluctuation conductivity. The observed change of sign for the anisotropic Tl 2223 is explained by considering the density-ofstates fluctuations. Hg 1201 does not show any anomaly in its magnetoconductivity and only the AL contribution is necessary to describe experimental data. This is in agreement with the low anisotropy of this compound. Furthermore, the parameters deduced from this analysis (τ is the quasiparticle scattering time and τ_{ϕ} is the pair-breaking lifetime) suggest that our samples lie in the intermediate region between clean and dirty limits. Moreover, the τ_{ϕ} results show that the phase pair-breaking process has a moderate strength.

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