Anisotropy of resistivity in Tl-based single crystals: Direct evidence for the influence of the blocking-layer thickness and correlation with superconducting properties

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The in- and out-of-plane resistivities ρ_{ab} and ρ_c have been measured on single crystals of Tl₂Ba₂CaCu₂O₈ $(Tl-2212)$ and $Tl_{2/3}Bi_{1/3}Sr_2CaCu_2O_7$ [(Tl, Bi)-1212]. The ρ_c values were found to be much lower in the Tl monolayer $[$ (Tl, Bi)-1212] than in the Tl bilayer (Tl-2212). These measurements allowed us to test quantitatively the predictions of the model proposed by Kim *et al.* [Physica C 177, 431 (1991)] about the influence of the thickness of the blocking layers on ρ_c , as well as the effect on the location of the irreversibility line. $[S0163 - 1829(97)00526 - 2]$

High- T_c superconductors have large electronic anisotropies which result from their highly layered structures. These features are particularly pronounced in the case of the Bi-, Tl- and Hg-based compounds whose structures consist of intergrowths along *c* between oxygen-deficient perovskite layers containing the $CuO₂$ planes and rocksalt-type layers containing the Bi, Tl, or Hg cations. These latter layers are known to be intrinsically less favorable than the blocks of $CuO₂$ multilayers for charge-carrier transport and they strongly affect the out-of-plane conductivity yielding a noticeable electrical transport anisotropy. Closely related to this anisotropy of resistivity, the anisotropy parameter γ , defined by the square root of the ratio of the effective carrier masses in the superconducting state, is involved in a lot of properties related to vortex dynamics. An important issue is to get more quantitative information about this electronic anisotropy and to specify the influence of the structural and chemical features.

Among the relevant parameters, one of the most important is the thickness d_i of the rocksalt-type layers which separate the blocks containing the $CuO₂$ planes. A correlation between the value of d_i and some physical properties such as the irreversibility line was already clearly established.^{1–5} Nevertheless, there is no direct measurement demonstrating the influence of d_i on the value of the resistivity anisotropy. The family of the Tl-based compounds is well suited to study the role of d_i since one can compare Tl monolayers with Tl bilayers (containing one or two TlO layers within the rocksalt-type blocks, respectively). In the present paper, we report in- and out-of-plane resistivity measurements on single crystals of one mixed (Tl,Bi) monolayer $\left[\text{TI}_{2/3}\text{Bi}_{1/3}\text{Sr}_{2}\text{CaCu}_{2}\text{O}_{7}\right]$ denoted (Tl, Bi)-1212] and one Tl bilayer $(Tl_2Ba_2CaCu_2O_8$ denoted Tl-2212).

The experimental conditions for the crystal growth of Tl-2212 and $(Th, Bi)-1212$ have been reported elsewhere.^{6,7} The values of d_i considered as the interplanar $(Cu-Cu)$ distance through the rocksalt-type layer are 8.79 and 11.55 Å for (Th) , Bi)-1212 and Tl-2212, respectively.^{7,8} Note that one TlO or (Tl, Bi) O layer corresponds to two rocksalt type layers, while two TlO layers correspond to three rocksalt type layers. It must be pointed out that the smaller d_i value in (Th, Bi) -1212 compared to Tl-2212 is not only due to the presence of two rocksalt type layers instead of three, but also to the substitution of Ba by Sr since the ionic radius of Sr^{2+} (1.31 Å) is lower than that of Ba²⁺ (1.47 Å).

Because of the large anisotropy itself, the determination of ρ_{ab} and ρ_c is not straightforward. Multiterminal techniques as in the Montgomery⁹ and Busch¹⁰ methods imply the measurement of a ''bottom'' voltage, i.e., the voltage drop between two points of one face while the current is injected through two points of the other face. It turns out that such a voltage can hardly be measured in our Tl-based crystals which are at the same time rather anisotropic and rather thick ($\sim 100 \ \mu \text{m}$). Instead, we have used two configurations of contacts depicted in Fig. 1 which allow a more direct determination of each resistivity. For the (C) configuration, one can directly derive a good estimate of ρ_c from such a ''cross'' measurement providing that the sample is small enough to ensure that the current and voltage contacts are close together and close to the edges of the crystal. Several comparative studies with more sophisticated methods have shown that the direct combination of R_{cross} with the dimensions of the crystal yields reliable values of ρ_c .^{10–12} Gold wires were attached to conductive pads made by ultrasonic diffusion of pure indium, leading to very robust contacts with a resistance of the order of 10 Ω . The resistive measurements were carried out with conventional dc technique. Errors in determining the absolute resistivities from uncertainties in the dimensions are estimated to be of the order of 30%. To illustrate the obtained transition curves, normalized $\rho_c(T)$ curves of several samples for Tl-2212 and (Tl, Bi)-1212 are shown in Fig. 2. Among the Tl-2212 crystals, the T_c (midpoint) varies between 108 and 112 K, while it varies between 86 and 90 K for $(Th, Bi)-1212$. In all cases the transition widths are around 5 K. The observed T_c 's lie near the highest values which were reported for both compounds, $6,7$ indicating that these samples are close to the optimized doping state.

By contrast to the out-of-plane resistivity, the $\rho_{ab}(T)$ curves show a metalliclike behavior in all cases for both compounds. The *T* dependence of ρ_c in Tl-2212 indicates a metalliclike or a semiconductinglike behavior depending on the sample. Such a variation from sample to sample about the *T* dependence of ρ_c was reported many times for different compounds such as Bi-2201,¹² Bi-2212,¹³ or Hg-1223.¹⁴ The great sensitivity of $\rho_c(T)$ to the doping state^{15,16} can be

FIG. 1. Schematic representations of the two contact configurations *AB* and *C* used to measure ρ_{ab} and ρ_c , respectively.

involved in this scattering but more extrinsic reasons such as stacking defects cannot be ruled out. In contrast, one observes that the shapes of the $\rho_c(T)$ curves in (Tl, Bi)-1212 are all similar : ρ_c decreases with decreasing temperature down to a characteristic temperature where it presents a shallow minimum and then increases slightly until T_c where it drops abruptly. Such a mixed behavior with a semiconductorlike *T* dependence at low *T* and a metalliclike dependence at high *T* has been previously observed in several compounds: Bi-2212,^{17,18} Bi-2201,^{12,19,20} as well as underdoped $YBa_2Cu_3O_{7-\delta}$ and $(La, Sr)_2CuO_4$ (see references in Ref. 21). This peculiar T dependence is related by some authors to a competition between different dissipative processes,^{19,21} while other explanations involving misalignments of the $CuO₂$ planes have also been proposed.¹²

The main goal of the present study is to compare the absolute values of ρ_{ab} and ρ_c in a Tl monolayer [(Tl, Bi)-1212] and a Tl bilayer (Tl-2212). For each resistivity component and for each compound, five crystals at least have been studied. In each case, the experimental interval and the mean value at room temperature are reported in Table I. The ρ_{ab} (290 K) of Tl-2212 and (Tl, Bi)-1212 are close to each other and lie in the same range of values found in all high- T_c superconductors. The situation is more contrasted

FIG. 2. Normalized $\rho_c(T)/ \rho_c(290 \text{ K})$ plots for several single crystals of Tl-2212 (a) and (Th, Bi) -1212 (b).

TABLE I. Averaged values of ρ_{ab} , ρ_c and $\gamma_N = \sqrt{\rho_c / \rho_{ab}}$ in both compounds (are reported in brackets the lowest and highest values found from the measurement of five samples at least in each case). The repeat distance of the structure (d_c) and the thickness of the blocking layers d_i are also given for each compound.

	TI-2212	$(Tl, Bi) - 1212$
	ρ_{ab} (290 K) 220 $\mu\Omega$ cm (170–290)	270 $\mu\Omega$ cm (150–600)
ρ_c (290 K)		2.4 Ω cm (1.8–3.1) 0.077 Ω cm (0.062–0.080)
γ_N (290 K)	~ 104	\sim 17
$\gamma_N(T_c)$	~ 130	\sim 31
d_i (Å)	11.55	8.79
$d_c(\AA)$	14.72	12.07

about ρ_c (290 K) since the value in Tl-2212 is found to be much larger than in $(Th, Bi)-1212$. This is an experimental determination of ρ_c in a Tl-monolayer compound. There was one previous study of ρ_c in Tl-2212 which reported a value close to 0.1 Ω cm.²² The present study yields higher values which are very similar to the results obtained in Bi-2212 single crystals near the optimized doping state.¹⁵⁻¹⁸ The only available data in a compound with a double rocksalt-type layer like in (Tl, Bi)-1212 deals with Hg-1223 (HgBa₂Ca₂Cu₃O₈).¹⁴ The ρ_c values for this Hg -based compound are slightly higher than in (Tl, Bi) -1212 since they range between 0.3 and 0.7 Ω cm.¹⁴ It can be noted that ρ_c in (Tl, Bi)-1212 is very close to the value in optimized $La_{1.85}Sr_{0.15}CuO₄$ but still larger than in optimized $YBa_2Cu_3O_{7-\delta}$ (see Fig. 3).

Kim *et al.* have proposed a model in which the location of the irreversibility line (IL) for $H\|c$ is intimately related to ρ_c .¹ The temperature dependence of ρ_c is neglected in this model. According to the weak *T* dependence displayed in Fig. 2, one can reasonably consider ρ_c (290 K) as a relevant characteristic value for the ρ_c parameter used in the model. This model involves first a Josephson decoupling of the vortices followed by a melting or a depinning process within the resulting two-dimensional vortex lattice. When comparing different compounds, one can consider as a first approximation that the value of the Josephson decoupling field is the main parameter, yielding the relation

$$
H_{\rm irr} \propto \frac{1}{\rho_c \times d_c},\tag{1}
$$

FIG. 3. Comparison of the ρ_c values at room temperature in Tl-2212 and (Tl, Bi)-1212 with other superconducting cuprates: $YBa_2Cu_3O_7$ (YBCO) (Refs. 19, 29); La $_{1.85}Sr_{0.15}CuO_4$ (La-Sr-Cu-O) (Refs. 19, 30); $Bi₂Sr₂CaCu₂O₈$ (Bi-2212) (Refs. 15–19).

FIG. 4. Irreversibility lines (H $\vert c$) of Tl-2212 and (Tl, Bi)-1212 in a scale of reduced temperature. The arrow corresponds to a shift by a factor of 40.

where d_c is the repeat distance of the structure along c . Within this model, Kim *et al.* have successfully fitted the IL's of different materials including Tl-based compounds and they derived values for various parameters such as ρ_c .¹ Nevertheless there was no direct test of Eq. (1) by using independent measurements of ρ_c .

The IL's of Tl-2212 and (Th, Bi) -1212 single crystals have been determined by magnetic measurements in the orientation H||c. The irreversibility fields $H_{irr}(T)$ have been derived from the closing points of isothermal hysteresis loops, corresponding to a J_c criterion around 50 A/cm². Figure 4 shows these IL's as a function of the reduced temperature $t=T/T_c$. One can note that the IL of (Tl, Bi)-1212 is clearly above that of Tl-2212. In the temperature range where they can be quantitatively compared, these IL's are shifted by a factor around 40. On the other hand, the comparison of ρ_c and d_c in both compounds (see Table I) leads to: ρ_c [Tl- 2212 \sim 32 ρ_c [(Tl, Bi)-1212] and d_c [Tl-2212] \sim 1.22 d_c $[$ (Tl, Bi)-1212]. By combining these two relations it is found that the quantity ($\rho_c \cdot d_c$) is about 40 times larger in Tl-2212 than that in (Tl, Bi) -1212. Accordingly, it can be stated that the basic relation of the model of Kim $et al.¹$ is remarkably well verified at least for the two investigated compounds. Moreover, the data are consistent with the predicted relationship between ρ_c and d_i : $\rho_c \propto \exp(d_i/d_0)$, where d_0 is related to the tunneling barrier height.¹ Indeed, the quantitative comparison between Tl-2212 and (Th, Bi) -1212 yields a plausible value of $d_0 \sim 0.8$ Å.¹

A parameter more often used than ρ_c to characterize the interlayer coupling is the anisotropy of resistivity: γ_N = $\sqrt{\rho_c / \rho_{ab}}$. The values for γ_N at 300 K and just above T_c are given in Table I for Tl-2212 and (Tl, Bi)-1212. One observes that γ_N (Tl-2212) is much larger than γ_N [(Tl, Bi)-1212]. Actually, this difference of γ_N reflects essentially the difference of ρ_c since the ρ_{ab} values are rather close in both compounds. Note that the metallicity of ρ_{ab} makes the γ_N values to increase as the temperature decreases. Kishio *et al.*²³ have reported a "universal" relation between H_{irr} at high temperatures and γ_N (evaluated at T_c): for Y-Ba-Cu-O, La-Sr-Cu-O, and Bi-2212 with different carrier doping states in each case, it was found that H_{irr} varies continuously with γ_N^2 . Moreover the curves $H_{irr}(\gamma_N^2)$ correlate rather well in the three systems and exhibit roughly a variation law: *H*irr $\propto \gamma_N^{-2}$. Actually, this phenomenological scaling law is very close to Eq. (1) since the main parameter in each case is ρ_c .

Although the anisotropy of resistivity $\gamma_N = \sqrt{\rho_c / \rho_{ab}}$ is strongly influenced by the interlayer coupling, it cannot be identified to the electronic anisotropy γ which is related to the ratio of the effective masses of the charge carriers in the superconducting state. Even in the framework of an oversimple band theory for the transport within the *ab* planes as well as along the *c* axis, it must be underlined that the diffusion characteristic times yields an additional source of anisotropy for the resistivity. The experimental determination of γ in rather anisotropic superconductors such as the Tlbased compounds is a very difficult problem and one can note that there is considerable scatter in the literature. For instance the estimates of γ in Tl-2212 range from values²⁴ lower than 5 up to values²⁵ higher than 300. By relating the ''fishtail effect'' observed in hysteresis loops²⁶ to phase transitions in the vortex state, we have derived estimates of γ in different Tl-based compounds. We have obtained a range 26–37 for γ [(Tl, Bi)-1212] and 77–96 for γ (Tl-2212). It can be noticed that these estimates of γ are not so different from the values of γ_N . Such a rather good experimental agreement between γ and γ_N was reported many times in different compounds.^{27,28} A closer inspection of the fundamental relationship between these two anisotropies would deserve to be addressed in the case of layered superconductors.

In summary, in- and out-of-plane resistivity measurements have been performed on Tl-2212 and (Tl, Bi)-1212 single crystals. Mean values of ρ_{ab} (290 K) equal to 220 and 270 Ω cm have been found in Tl-2212 and (Tl, Bi)-1212, respectively. These values are very close to what is generally found in all high- T_c superconductors. As for ρ_c (290 K), a much larger value was found for Tl-2212 compared to (Tl, Bi)-1212, since ρ_c (Tl-2212) \sim 2.4 Ω cm and ρ_c [(Tl, Bi)-1212] $\sim 0.077 \Omega$ cm. This difference in ρ_c can be quantitatively related to the observed shift of the irreversibility line between these two compounds, as expected in the frame of the model of Kim et al.

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