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Out-of-plane temperature-dependent resistivity studies on Tl-based superconductors

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Abstract
Temperature-dependent resistivities in the *ab*-plane and *c*-axis of Tl-based cuprates have been measured. Unlike the *ab*-plane properties, which are metallic, *c*-axis transport is semiconductor-like in the normal state for $Tl_2Ba_2Ca_2Cu_3O_x$ (Tl-2223) and $Tl_2Ba_2CaCu_2O_x$ (Tl-2212). In contrast, for $Tl_2Ba_2CuO_x$ (Tl-2201), transport is metal-like in both the in-plane and the *c*-axis. For multi-layered cuprates, transport properties along the *c*-axis could be described by a tunnelling model, whereas for single-layered compound Tl-2201 it would be easier for the out-of-plane transport behaviour to be coherent since the there are no insulating Ca layers in its structure. Moreover, combining the studies on Bi-2201, which has an insulating behaviour for the out-of-plane resistivity, we suggest that the Tl–O layers in Tl-based superconductors could be conducting, unlike the weakly correlated Bi–O layers in Bi-based cuprates.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

A high-temperature superconductor (HTSC) is a layered system that is highly anisotropic in-plane (*ab*-plane) and out-of-plane (*c*-axis). At optimal doping, the in-plane temperature-dependent resistivity $\rho_{ab}(T)$ is metal-like in the normal state, while the *c*-axis resistivity, $\rho_c(T)$, displays a non-metallic or metallic behaviour, and is about two orders of magnitude or more larger than $\rho_{ab}(T)$ at room temperature [1, 2]. Naturally, one can suggest that the Cu–O plane—the common structural characteristic for the HTSC cuperates—is conducting and corresponds to superconducting condensation. However, superconductivity in the *c*-axis does occur below the critical temperature T_c. Furthermore, different members of the HTSC family have different maximum T_cs ($T_c$max), even if they are all at optimal doping levels. For

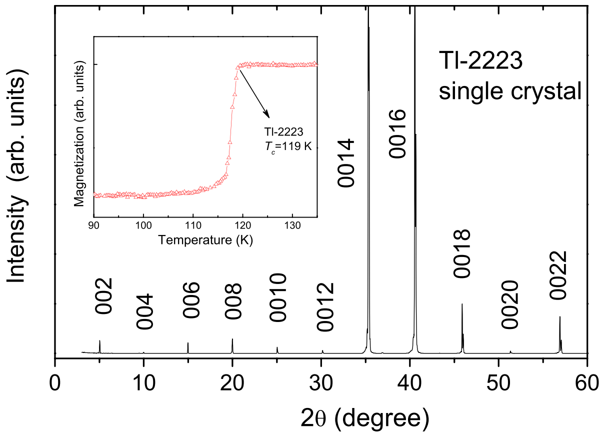

Figure 1. The x-ray diffraction pattern up to 60° for Tl-2223 crystal. Inset: the temperature-dependent ac susceptibility for the same crystal.

example, although they have nearly the same single Cu–O sheets separated by either Tl–O or Bi–O double layers, $T_c$max for Bi-2201 is no more than 40 K [3], while for Tl-2201 it can be higher than 90 K [4–7]. The reason is not clear now; although some have suggested that the carriers in the Cu–O plane couple a collective mode (the higher T_c, the higher corresponding mode energy), the contribution from the c-axis cannot be ruled out unambiguously. Moreover, a complete microscopic mechanism for HTSCs should give a defined explanation of all the phenomena, both in-plane and in the c-axis, both in normal states and in superconducting states. So, investigating the transport properties in the c-axis would be very helpful in understanding the origin of HTSCs.

Tl-based cuprates play an important role in investigating the properties in both the ab-plane and the c-axis, for there are more members with different Cu–O layers. In the TlBaCaCuO system, Tl-2201, Tl-2212 and Tl-2223 have been of especially great interest for nearly 20 years since they were synthesized in 1988, for they have exhibited much higher T_cs than most other HTSC members [8–11]. Furthermore, it would be very helpful if the studies could be combined with those of Bi-based superconductors, since they have nearly identical structures. However, up to now, there have been relatively few reports on Tl-2212 and Tl-2223 crystals. The reason may be that synthesizing the crystals for physical studies needs to resolve at least two problems: thallium volatility and the formation of inter-growth defects [5, 11].

Optimally doped Tl-2201, Tl-2212 and Tl-2223 and overdoped Tl-2201 crystals have been grown successfully by using the flux method [5, 6]. In this study, we systematically investigate the characteristics of the out-of-plane temperature-dependent resistivities of Tl-based crystals in normal states.

2. Experimental details

Our Tl-based crystals were grown by using the flux method; details have been described elsewhere [5, 6]. Each selected sample for our experiments was highly textured or had a good c-axial orientation which is perpendicular to the sample's natural growth face, since all the (0, 0, l) peaks could be identified clearly; see the x-ray diffraction patterns in figure 1. Accordingly, these samples are appropriate for our investigations. The temperature-dependent

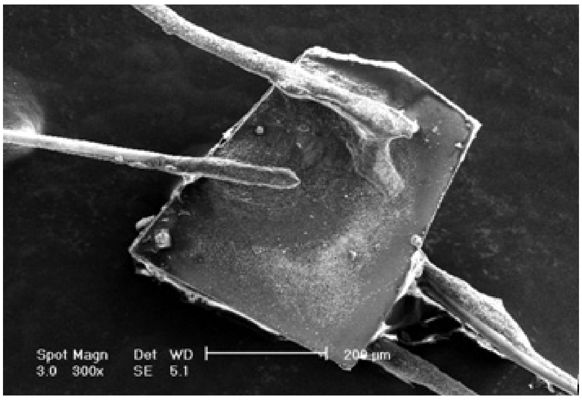

Figure 2. The distribution or the structure of the two electrodes on one side for testing the temperature-dependent resistivity parallel to the *c*-axis of Tl-2223, similar to the case on the other side.

resistivities were measured by using the standard ac four-point method with a lock-in amplifier; the frequency of the test current was 30 Hz. The typical size of the samples used for the experiment was $0.5 \times 0.4 \times 0.06\ mm^3$. For good Ohmic contact, the samples were annealed at 350 °C for about 15 min in air after the silver contact pads had been painted. The samples were mounted on sapphire using glue, and were cooled at a cooling rate of about $3\ K\ min^{-1}$. Figure 2 shows the distribution of the two electrodes on one side in testing the resistivity along the *c*-axis of Tl-2223, similar to the case on the other side. Note that, although the measurements were not carried out by using the Montgomery method, we do believe that the results should represent the primary characteristic along the out-of-plane direction for these HTSC samples, since the current should flow mostly in the *c*-axis.

3. Results and discussion

From the inset of figure 1, we can definitely determine the critical temperature of our Tl-2223 crystal sample as $T_c = 119$ K. Superconducting condensation occurs not only in the *ab*-plane but also in the *c*-axis; see figure 3. Similarly to other HTSCs, the in-plane resistivity, $\rho_{ab}(T)$, is metal-like in the normal state. Moreover, the temperature-dependent resistivity, $\rho_{ab}(T)$, could be fitted with a single line above $T_c = 119$ K, indicating the optimal doping level of the sample [2, 12], while the *c*-axis resistivity, $\rho_c(T)$, displays semiconductor-like behaviour in the normal state. As the sample was cooled down below $T_c = 119$ K, we could clearly see a sharp transition in the *c*-axis.

In a layered system of Tl-2223, the transport properties along the *c*-axis can be modelled with a series of '... CuO–insulator–CuO–insulator–CuO–insulator ...' structures. So, the carriers contributing to transport in the normal states should tunnel through the barrier formed by the insulator layers, although the Cu–O layers should be conducting, as discussed in the introduction. Since the resistivity in the normal state increased as temperature decreased, we suggest that the phonon-assisted quantum tunnelling or inter-layer hopping is rather plausible [13], for the number of phonons of a certain mode decrease when being cooled. As the temperature goes below $T_c = 119$ K, some of the carriers in the Cu–O plane condense to superfluid Cooper pairs, thus Josephson coupling occurs, namely the zero-resistivity state develops along the *c*-axis.

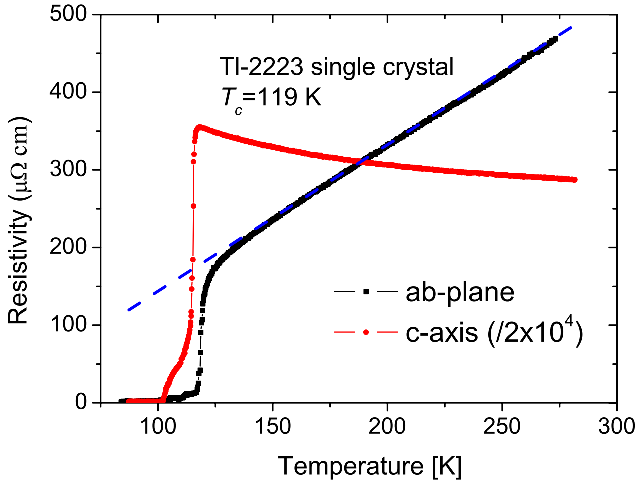

Figure 3. Temperature dependence of resistivities in both the *ab*-plane and the *c*-axis for Tl-2223.

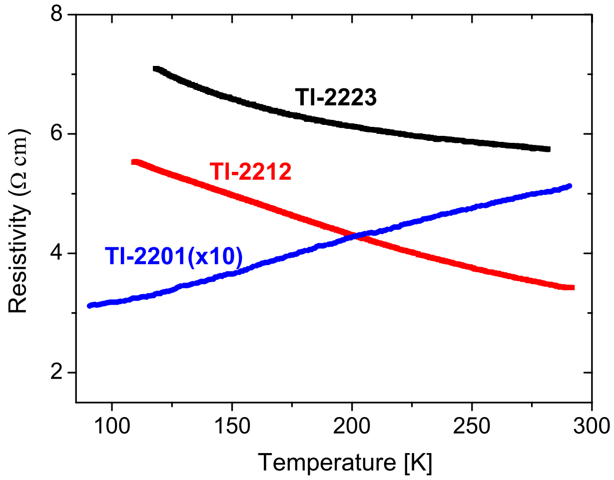

Figure 4. Normal-state temperature-dependent resistivities in the *c*-axis of Tl-based samples, Tl-2201, Tl-2212 and Tl-2223.

In our experiments, similar results have been observed for Tl-2212 samples with T_c = 109 K; see figure 4. In contrast to the properties of the two multi-layer Tl-based cuprates, $\rho_c(T)$ for Tl-2201 is metallic in the normal state, which is consistent with the earlier reports [14]. This difference will be discussed in the following context.

To our knowledge, up to now there has been no consensus about the *c*-axis transport in HTSCs [13, 15–18]. Based on electron structure calculations and symmetry analysis, it has been shown that the *c*-axis hopping integral in high-T_c cuprates with tetragonal symmetry is given by $t_c \propto (\cos k_x - \cos k_y)^2$ [19]. Clearly, t_c vanishes along the diagonals of the Brillouin zone, and exhibits its maximum at the antinodal points. In [16], ρ_c is given approximately by a universal formula $\rho_c \propto (T/\Delta)\exp(\Delta/T)$ in the limit $T_c < T < \Delta$ for all multi-layer cuprates and some single-layer cuprates, where Δ is the maximal value of the pseudogap. According to this relation, ρ_c should increase on cooling below a certain

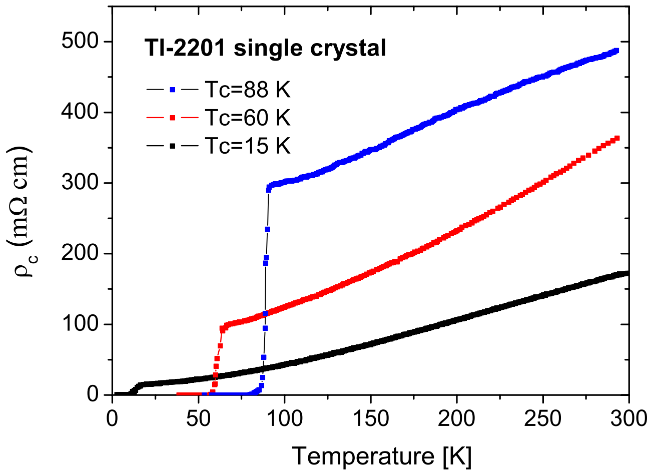

Figure 5. Temperature-dependent resistivities in the c-axis of Tl-2201 for three overdoped samples. Note that all these ρ–T relations are metal-like.

temperature, so our results are consistent qualitatively with this formula for Tl-2212 and Tl-2223; see figure 4. However, for Tl-2201 or Bi-2201, Cu atoms of two adjacent Cu–O planes do not lie collinearly along the c-axis, the hopping integral in this direction has the form $t_c \propto \cos(k_x/2)\cos(k_y/2)(\cos k_x - \cos k_y)^2$. It vanishes along both the nodal and antinodal directions. Therefore the c-axis hopping is dominated by the quasi-particles in regions between the nodal and antinodal points. In fact, the life time or the scattering rate of the quasi-particles, which can be reflected from the energy distribution curves of angle-resolved photoemission spectroscopy (ARPES) [20, 21], is also crucial for the transport behaviour. However, the above reasons are not essential, for Tl-2201 has a metallic resistivity–temperature relation in the c-axis; see figure 5.

If we consider the evolution of the crystal structures of the unit cells for Tl-2201, Tl-2212, and Tl-2223, we can find a remarkable difference, that is, there are no Ca layers in Tl-2201, whereas for Tl-2212 and Tl-2223 all the carriers contributing to the transport properties should inevitably tunnel through the insulating Ca layers. It is the Ca layers that are responsible for the semiconducting resistivity behaviour for the multi-layered high-T_c cuprates, meanwhile the Tl–O layers should not be insulating, or else Tl-2201 would have semiconductor-like resistivity along the c-axis. This viewpoint can also be supported from analysis of the infrared conductivities. It has been observed that the optical conductivities of the Bi-2212 double plane and the Tl-2201 single plane are very similar (see [22]), while for the carrier density per plane it should be 50% smaller in Bi-2212 than in Tl-2201; however, they do have nearly the same $T_c$max. One may argue that $T_c - 90$ K Tl-2201 is a strongly overdoped material with a hypothetical $T_c\text{max} \simeq 190$ K [23][3], as a peak in T_c-doping has not been observed for this material. On the other hand, if Tl-2201 has a $T_c$max of 90 K, the whole idea of T_c being determined by the normal-state free carrier density per planar Cu is put in question, unless one assumes that Tl–O planes may also be conducting. Therefore the Tl–O layers should contribute to the low-frequency ab-plane conductivity spectral weight (SW); in fact, some theoretical estimates support this possibility, although the Tl–O planes contribute only a small fraction of the total SW [22, 24]. In addition, it is clearly seen from figure 5 that the resistivity will decrease

[3] According to the parabolic relation of the T_c-doping in [23], if the systems have the same $T_c$max, then they should have nearly the same doping levels or mobile carrier densities at optimal doping levels.

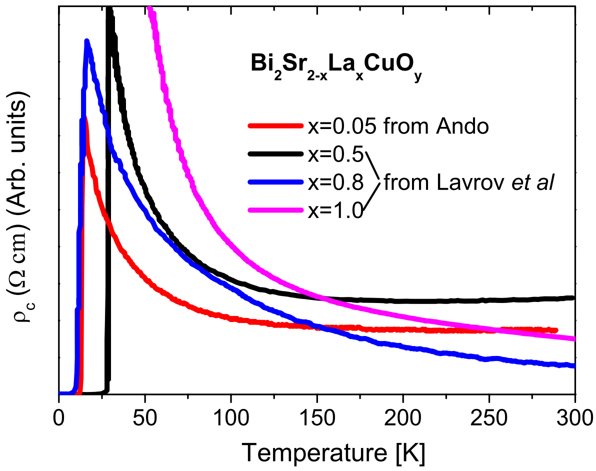

Figure 6. Temperature-dependent resistivities in the c-axis of Bi-2201 at four various doping levels. Note that all these ρ–T relations are semiconductor-like.

with overdoping [6, 25][4]; this is consistent with earlier reports [14, 26], but at the same time it also reflects that the Fermi surface of high-T_c materials is more three-dimensional [5, 27].

The above discussions should also be consistent with investigations for the out-of-plane resistivities in the Bi-based systems, Bi-2201, Bi-2212 and Bi-2223. The multi-layered cuprates Bi-2212 and Bi-2223, which have Ca layers in the unit cells, should naturally have semiconducting behaviour in the c-axis, however Bi-2201 has no Ca layers in its structure, so how can it have an insulating out-of-plane resistivity? In figure 6, the four Bi-2201 samples at various doping levels all show semiconducting resistivity behaviours [28, 29]. We suggest that the reason may come from the Bi–O layers, for we know that the two Bi–O layers have much weaker correlations (Bi-based cuprates have the biggest interplanar separation in the cuprates, so these layers are typically of van der Waals interactions) [20]; in fact, the distance between adjacent bismuth sheets is 3.25 Å, whereas for Tl-2201 it is only 2.20 Å between neighbouring Tl–O layers. This gives Bi-based cuprates the best possible cleavage in all the high-T_c families; naturally, most ARPES investigations have been performed on this system. In contrast, for Tl-based systems there is very limited ARPES data due to its poor cleavage properties; only two overdoped samples have been studied by Damascelli *et al* to this day [30]—neither optimally doped Tl-2201 nor any other Tl-based cuprates have been reported yet.

4. Summary

In conclusion, we have measured temperature-dependent resistivities by using the standard ac four-point method in the c-axis of Tl-based samples. Unlike the in-plane properties, c-axis transport is semiconductor-like in the normal state for Tl-2223 and Tl-2212. In contrast, for Tl-2201 the transport behaviour is metal-like in the c-axis direction. For multi-layered high-T_c superconductors, the transport properties along the c-axis can be described by a tunnelling

[4] Strictly speaking, the Tl-2201 samples cannot be identified as overdoped or underdoped only from their T_cs, for the T_c-doping relation has an arch feature. To the best of our knowledge, no underdoped Tl-2201 has been reported so far. Furthermore, our earlier experiments on the infrared optical properties showed that they were very similar to the results of the overdoped LSCO samples [6, 25]. In fact, the infrared probe was very powerful for investigating the properties of the carriers. Combining the above results, our Tl-2201 with T_cs no more than 90 K are overdoped samples.

model; however, for Tl-2201 it is easier for the out-of-plane transport behaviour to be coherent. We have discussed the reason for why Tl-2201 has metallic resistivity behaviour in the *c*-axis, whereas Bi-2201 and the multi-layered superconductors can be out-of-plane insulators.

Acknowledgments

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