Evolution of the resistivity of single-layer Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_y$ thin films with doping and phase diagram

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Abstract

The temperature dependence of the in-plane resistivity $\rho(T)$ is measured on epitaxial $c$-axis oriented single-layer Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_y$ thin films at various oxygen concentrations. By successive annealing treatments, the oxygen content of the same film is changed from maximally overdoped to strongly underdoped non-superconducting state, passing through the optimal state with $T_{c,\text{max}} = 30$ K. The underdoped states show a downturn of the resistivity from the high $T$-linear behavior below a characteristic temperature $T^*$, signature of the pseudogap effect. $T^*$ appears near optimally doped state and increases sharply with decreasing carrier concentration. Two other characteristic temperatures are observed in $\rho(T)$ for underdoped states: the temperature $T_1$ of the inflection point in $\rho(T)$ ($T_1 \sim 0.5 T^*$) and the temperature $T_M$ corresponding to the onset of localization effects. A phase diagram $T$ versus doping is established. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The evolution with hole doping of the normal state electronic properties of cuprates is considered to be a principal line of research for understanding the underlying mechanism of high-temperature superconductivity (HTSC). Depending on temperature and doping, the electronic properties change from those of an antiferromagnetic insulator to those of a metal, what motivates detailed investigations of the rich phase diagram of cuprates.

One striking phenomena is the pseudogap which opens in the electronic excitations [1–8]. This effect, observed in underdoped HTSC, has been the center of investigations for many years without consensus about its nature and its relation with superconductivity. The pseudogap effect was first observed in CuO$_2$ bilayer YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) [9] and single-layer La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) [10] cuprates, although certain differences between these two materials [11] have lead to controversial conclusions regarding the existence of this effect in systems with one CuO$_2$ plane per cell [12]. Recently, the existence of the pseudogap was confirmed in single-layer systems by NMR measurements on HgBa$_2$CuO$_{4+\delta}$ (HgBa-CuO) [13] and by photoemission measurements...

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on Bi$_2$Sr$_{2-x}$La$_x$CuO$_y$ (BSLCO) [14]. More recently, a pseudogap effect was reported from transport measurements on single-layer Bi$_2$Sr$_{2-x}$La$_x$CuO$_y$ crystals, where the carrier concentration was changed by changing La doping $x$ [15].

On the other hand, at low doping level and low-temperature localization effects compete with the observed pseudogap effect. Onset of localization effects was investigated recently by suppressing superconductivity with 60-T pulsed magnetic fields [16,17]. It was reported that the metal–insulator (MI) crossover takes place at optimum doping in LSCO compound, i.e. at the same doping where pseudogap opens [16]. However, recent results in BSLCO indicate that the MI crossover is not universal for all cuprates [17]. The other way for inducing a MI crossover is by decreasing the carrier concentration or by increasing disorder by chemical substitution [18–20].

Here, we present results obtained by measuring the in-plane resistivity of Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_y$ (Bi(La)-2201) thin films as a function of temperature, over a wide range of carrier concentration. The oxygen content is changed on the same film by annealing treatments, which permitted to investigate almost all the phase diagram, from strongly overdoped to strongly underdoped. By reducing the carrier concentration, the superconductivity is suppressed and the MI crossover is induced.

2. Experimental details

The $c$-axis oriented epitaxial Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_y$ thin films, with thickness between 1000 and 2000 Å, are grown in situ by RF magnetron sputtering on SrTiO$_3$ substrates [21]. The films are chemically patterned and equipped with gold sputtered contact pads. The temperature dependence of the in-plane resistivity is measured at different oxygen doping levels using a standard four-probe dc method. The oxygen content of a single sample is changed by repeated annealing treatments in a controlled atmosphere going from the maximally overdoped state ($T_c(R = 0)$ = 17 K) to the strongly underdoped non-superconducting states ($T_c = 0$). The change in Hall coefficient, measured at room temperature in a magnetic field of 1 T, confirms the change in carrier concentration after each annealing procedure. The transport properties of states obtained in this way are reproducible. The partial substitution of Sr by La increases the optimum $T_{c\text{max}}$ from 18 K for pure Bi$_2$Sr$_2$CuO$_y$ (Bi-2201) to 30 K for Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_y$, which also increases the width of the underdoped region. The number of holes per Cu, $p$, is calculated from the empirical parabolic function $T_c/T_{c\text{max}} = 1 - 82.6 (p - 0.16)^2$ [22].

3. Experimental results

Fig. 1 shows three typical sets of curves representing the temperature dependence of $\rho(T)$ obtained on the same film in (a) the overdoped, (b) the underdoped and (c) the strongly underdoped non-superconducting region ($T_c = 0$). The conductivity at room temperature decreases monotonically with decreasing oxygen content in the sample and it has been used to characterize the carrier concentration. It is to be noted that while in pure Bi-2201 films the non-superconducting metallic state ($T_c = 0$) can be reached [23], the minimal critical temperature value of our Bi(La)-2201 films is $T_c(R = 0)$ = 17 K. In the overdoped region (Fig. 1(a)), $\rho(T)$ develops a positive curvature and can be described by the phenomenological law $\rho(T) = \rho_0 + zT^m$ with $m = m_{\text{max}} = 1.3$ for the maximally overdoped state. It appears that $m_{\text{max}}$ has the same characteristic value for all overdoped Bi-2201 [23] and is lower than in LSCO and Tl$_2$Ba$_2$CuO$_{6+\delta}$ (Tl-2201) compounds ($m_{\text{max}} = 1.5$ and 1.8 respectively [24,25]). This low $m_{\text{max}}$ value can be related to the low $T_{c\text{max}}$ value in Bi(La)-2201 compared to those in the other two single-layer compounds ($T_{c\text{max}} = 35$ and 85 K in LSCO and Tl-2201 respectively). The exponent $m$ decreases with decreasing doping down to 1, which corresponds to the optimally doped state (inset of the same figure). The resistivity at optimal doping shows a $T$-linear behavior above 55 K (top curve in Fig. 1(a) and bottom curve in Fig. 1(b)). In the underdoped states (Fig. 1(b)), $\rho(T)$ deviates downwards from this linear high-$T$ behavior below $T^*$, attributed to the pseudogap opening [9]. Another characteristic temperature corresponding to
a change of curvature in \( \rho(T) \), labeled \( T_1 \), is pointed in Fig. 1(b). Additional removing of oxygen out of the film leads to the loss of superconductivity and the occurrence of insulating behavior starting at the temperature \( T_M \), where \( \rho(T) \) is minimum (Fig. 1(c)). All above characteristic behaviors are also found in bilayer Bi-2212 thin films and a comparison between both phases in the pseudogap region is done in Ref. [26,28].

Fig. 2a shows the doping dependence of the Hall number \( n_H \), measured at 300 K, for the same film as in Fig. 1. The upper horizontal axis gives also the conductivity at room temperature normalized by its value at optimally doped state \( (\sigma/\sigma_{op})_{300\ K} \). The critical temperature \( T_c \) is also a parabolic function of \( (\sigma/\sigma_{op})_{300\ K} \), parameter previously used to characterize the oxygen doping in the case of Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_y\) (Bi-2212) films [27]. This quantity is used here in the same manner in order to parameterize the doping of the strongly underdoped states (\( T_c = 0 \)) where the above parabolic function cannot be applied (see further Fig. 3). The decrease of \( n_H \) with reducing carrier concentration indicates a total decrease of carriers by a factor of the order of 4, which is in good agreement with the corresponding decrease of \( p \) from 0.24 to 0.06. Moreover, it is shown that \( n_H \) varies nearly linearly with \( p \) and \( (\sigma/\sigma_{op})_{300\ K} \) which confirms that the latter can be used to characterize doping. Another quantity which allows to characterize the doping level is the \( c \)-axis lattice constant, \( c_0 \), obtained from X-ray diffraction spectra. The doping dependence of \( c_0 \) for some characteristic states for the same film
Another characteristic temperature $T_1$ is defined by the maximum value of the first derivative $d\rho/dT$ in the underdoped region (arrows pointing downward in Fig. 3(b)). This inflection point coordinate $T_1$ has the same doping dependence as $T^*$, and they are related by: $T_1 \sim T^*/2$ [28], relation earlier found in YBCO [29] and BSLCO [30] compounds. This observation has permitted to determine $T^*$ values when they exceed room temperature.

For strongly underdoped states, the temperature $T_M$ below which the insulating behavior begins is extracted from the minimum of the resistivity versus temperature (Fig. 3(c)). The insulating behavior is a manifestation of localization effects, which suppress the observed pseudogap effect. Moreover, the crossover from superconducting to insulating behavior occurs at $\rho_0^{\text{sl}} = 1.1 \pm 0.1 \text{ m}\Omega \text{cm}$. This value corresponds to a resistance per square per CuO$_2$ plane $\rho_0^{\text{sl}}/(c_0/2) \sim 9 \text{ k}\Omega$, where $c_0/2$ is the interlayer distance (see Fig. 3(c)). The normalized resistivity values in $(k_F\ell)^{-1}$ units are plotted on the right vertical axis (same figure), given by $\rho_0^{\text{sl}}/(c_0/2) = (h/e^2)/(k_F\ell)$, where $k_F$ is the Fermi wave vector and $\ell$ the mean free path between scattering events. The observed crossover value $\rho_0^{\text{sl}}/(c_0/2)$ is higher than that of conventional 2D superconductors, equal to $h/(2e)^2 = 6.5 \text{ k}\Omega$ ($k_F\ell \sim 4$) [31,32]. However, it is in very good agreement with the value estimated from $\sim h/0.69(2e)^2 = 9.4 \text{ k}\Omega$, established for 2D disordered systems with Coulomb interactions [33]. This comparison suggests that, although disorder plays an important role in these materials, one cannot neglect Coulomb interactions between carriers, which increase with the decreasing carrier concentration, shown in Fig. 2(a).

Note also in Fig. 3(c) that a log(1/T) behavior of $\rho(T)$ is only observed for the state close to the MI transition (third state from bottom curve) and for $T \lesssim 20 \text{ K}$. This insulating behavior was previously observed in both underdoped superconducting LSCO [16] and BSLCO [17] under a 60 T magnetic field. However, $\rho(T)$ for all others states [34] are in good agreement with a thermally activated conduction behavior just below $T_M$ and with 2D Mott variable-range hopping at lower temperature observed in LSCO system and associated
with phonon assisted impurity scattering [35]. The observed logarithmic behavior close to the superconducting–insulating transition and then 2D Mott variable-range hopping at low temperature are also in agreement with previous results on underdoped non-superconducting Bi-2201 single crystals [36].

4. Phase diagram

The doping dependence of all characteristic temperatures introduced above are shown in Fig. 4 for two different thin films (solid and open symbols). All three temperatures, \( T^* \), \( T_1 \) and \( T_M \), increase with decreasing carrier concentrations and their doping dependence is the same for all examined samples. Note also the parabolic behavior of \( T_c \) as a function of normalized conductivity \( (\sigma/\sigma_{op})_{300\, K} \), without suppression of the critical temperature near \( p \sim 1/8 \) (“1/8” anomaly), as observed previously in La\(_{2-x}\)Ba\(_x\)CuO\(_4\) compound [37] and recently mentioned in BSLCO single crystals [38]. The values of \( T^* \) in Bi(La)-2201 are of the same order of magnitude as in the case of BSLCO single crystals compared for the same ratio \( T_c/T_{c\, max} \) [15] and smaller than the values reported in LSCO compounds [10]. The change in slope of \( T_1(p) \) coinciding with the beginning of localization effects could induce a systematic error in \( T^*(p) \sim 2T_1(p) \) behavior in the strongly underdoped region (\( T^* \) values deduced from \( T_1 \) could be overestimated). The doping dependence of \( T^*(p) \) around optimally doped states presents a steep decrease towards the curve \( T_c(p) \). By extrapolating \( T^*(p) \) to zero, we find that \( T^* = 0 \) for \( p \sim 0.17 \pm 0.01 \) hole per Cu, which is considered as a possible quantum critical point [39,40].

The variation of the third characteristic temperature, \( T_M(p) \), provides the low-temperature boundary of the pseudogap region, as it was proposed theoretically earlier [39]. The extrapolated value at zero temperature of \( T_M(p) \) lies well inside the underdoped regime for BSLCO \( (p \sim 0.07 \pm 0.01) \) which is in good agreement with the MI boundary deduced from \( \rho(T) \) behavior of BSLCO single crystal under 60 T [17]. The only difference between the two results arises from different evaluated hole concentration values, but still it appears that the MI boundary cut \( T_c(p) \) at the same ratio \( T_c/T_{c\, max} \sim 0.4 \). The situation is quite different in LSCO compounds, where the onset of the localization behavior extends up to optimum doping [16]. So the low \( T \) beginning of the MI boundary in Bi(La)-2201 deeply in the underdoped region indicates that it depends on the cuprate family and it does not show a visible link with pseudogap effect occurring near optimum doping.

Finally, a schematic phase diagram was proposed in Ref. [41]. There the pseudogap phase is associated with 1D stripe formation, while at the MI boundary disorder disrupts the 1D stripes restoring a 2D regime for transport with
\( \rho(T) \sim \log(1/T) \) [42]. Although this insulating behavior \( \log(1/T) \) that we have seen in one state close the MI crossover might be associated with low dimensionality imposed by charge confinement into stripes as indicated in Ref. [17], it is difficult to understand why this behavior is observed in just a very narrow doping window. Moreover, until now there is no direct evidence, as in neutron scattering data on LSCO compound [43], of the existence of the stripe formation in Bi(La)-2201 which might help to clarify this issue.

5. Summary

To summarize, we have studied the evolution of the temperature dependence of the in-plane resistivity over a wide range of doping levels, from overdoped to strongly underdoped. We show that the behavior of the resistivity changes strongly with the oxygen content in the film, from metallic to semiconducting behavior. From this analysis, we have established a very detailed unique phase diagram of the single-layer \( \text{Bi}_2\text{Sr}_1\text{La}_{0.4}\text{CuO}_y \) phase. The upper boundary of the pseudogap region is given by the temperature \( T^*(p) \) and the lower boundary by the temperature \( T_M(p) \), where localization effects occur. The extrapolation to zero of \( T^*(p) \) around \( p \sim 0.17 \) may possibly indicate the existence of a quantum critical point. Moreover, our data indicate that there is no obvious relation between the onset of localization effects and the pseudogap opening occurring around optimal doping.

References