# *c*-axis resistance peak above the critical temperature in layered superconductors

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The possibility of fully describing the behavior of transverse resistivity  $\rho_c$  in cuprate superconductors exclusively in terms of some recently proposed normal-state models is discussed. It is shown that these models do not give a completely satisfactory description of the apparent singularity in  $\rho_c$  at the critical temperature in Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+x</sub>, and that they fail to describe data concerning a lower- $T_c$  cuprate superconductor as Bi<sub>2</sub>Sr<sub>2</sub>CuO<sub>6+x</sub> ( $T_c \cong 15$  K). It is argued that these difficulties cannot be overcome considering the normal-state contribution to  $\rho_c$  only. The role of the density of states fluctuations in the behavior of  $\rho_c$  just above the transition temperature is recalled. It is shown that, taking into account this contribution, it is possible to dramatically simplify the description of the normal-state transverse resistivity. This can give important hints for the understanding of the transverse electrical transport properties of these materials. [S0163-1829(96)02529-5]

# INTRODUCTION

As soon as the availability of bulk single crystals and monocrystalline films of cuprate superconductors made it possible to perform resistivity measurements both in the *ab* plane and along the *c*-axis direction, it became clear that the layered nature of these compounds leads to qualitative, and not only quantitative, differences between the temperature dependencies of in-plane and transverse resistivities. In particular, a peak in the transverse resistivity vs temperature  $\rho_c(T)$  curves is apparent just above the critical temperature  $T_{c0}$ , reminiscent of a "semiconducting" behavior of  $\rho_c$  in contrast to the well-known "metallic" temperature dependence of the in-plane resistivity  $\rho_{ab}(T)$ .<sup>1-3</sup> This peak is more pronounced for the more anisotropic samples  $[Bi_2Sr_2CaCu_2O_{8+x} (2212 BSCCO) \text{ compared to } YBa_2Cu_3O_7]$ (YBCO), reduced samples compared to oxygenated ones].<sup>4–6</sup>

Early attempts to fit the  $\rho_c(T)$  curves by means of phenomenological or physical theories always had to face the problem of an underestimation of the true resistivity close to  $T_{c0}$ , while in the high-temperature region the fit was reasonably good.<sup>7–11</sup>

These difficulties were overcome by the proposal of the fluctuation origin of the peak in  $\rho_c(T)$  by Ioffe *et al.*<sup>12</sup> According to this theory there is a competition among several fluctuation contributions to the *c*-axis conductivity. The positive Aslamazov-Larkin (AL) contribution has a temperature dependence more singular in  $T_c$  but is strongly depressed by its proportionality to the square of transparency above the Lawrence-Doniach crossover point. On the other hand, there is a fluctuation decrease of the one-electron conductivity which originates from the suppression of the one-electron density of states (DOS) at the Fermi level close to  $T_c$  (the opening of the fluctuation pseudogap). The negative DOS contribution is less singular in temperature but proportional

to the first order of transparency only. The competition between these two contributions of different sign determines the shape of the resistivity peak close to  $T_c$ . The AL contribution being more heavily dependent on the interlayer coupling than the DOS contribution, a more pronounced peak is expected for materials with higher anisotropy. The carrier density (and therefore the oxygen content in the sample) also affects the magnitude of the peak, a higher carrier concentration leading to a lower fluctuation contribution. Clearly, this description can be applied only to samples having an oxygen content not so low as to drive them close to the metal-insulator transition.

These predictions of the theory were shortly after experimentally confirmed by measurements on BSCCO and YBCO samples.<sup>6,13,14</sup> The values of some physical parameters involved in the theory were extracted from the fits and found in agreement with literature data. No phenomenological constant was necessary to fit the data.

Recently, the fluctuation contribution to transverse resistivity in layered superconductors was theoretically calculated also in the presence of an external magnetic field parallel to the *c* axis.<sup>15</sup> In this case, a well-known anomalous magnetoresistance peak is experimentally observed below  $T_{c0}$ .<sup>3</sup> Again, the experimental data were found<sup>16</sup> to be in good agreement with the theory, at least for moderate magnetic fields (fluxon dynamics and other effects were not included in the theory). A similar approach, including the effects of a magnetic field on the supercurrent in a stack of Josephson junctions, gave good results also in higher fields.<sup>17</sup>

Using the above theory it was also possible to calculate the magnetoresistance above  $T_{c0}$ , which turns out to be negative for highly anisotropic samples, in contrast to the positive one observed in the *ab* plane. A good fit of literature<sup>18</sup> data of transverse magnetoresistance in BSCCO single crystals was found<sup>19</sup> in this way. Also, the appearance of a maximum

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in the R(H) curves measured at a fixed temperature below  $T_{c0}$  was explained taking into account the DOS contribution to conductivity.<sup>20</sup>

All these experiments proved that the theory of the fluctuation origin of the transverse resistivity peak above  $T_{c0}$  in highly anisotropic cuprate superconductors is in agreement with available experimental resistivity and magnetoresistivity data on samples far from the metal-insulator transition.

Very recently, however, different phenomenological and physical models of normal-state *c*-axis conductivity have been proposed to account for the zero-field  $\rho_c(T)$  behavior in the whole temperature range above  $T_{c0}$ . The phenomenological formula proposed in Ref. 18 was found to be very accurate for both YBCO and BSCCO, while the theory proposed in Ref. 21 was checked against measurements made on YBCO. Since these two models seem to describe the data very well, their introduction could rule out a relevant role of fluctuations in determining the  $\rho_c(T)$  peak, in spite of the above-mentioned success of the fluctuation approach.

In this paper we will show that these models cannot fully describe available data on the transverse resistivity peak, and that some additional contribution which appears close to  $T_c$  (and is not, therefore, a normal-state one) is needed; fluctuations seem to be a major candidate for this contribution.

### **EXPERIMENT**

The 2212 BSCCO films used in this paper were grown by liquid-phase epitaxy (LPE) on NdGaO<sub>3</sub> substrates. Details of the growth procedure can be found elsewhere.<sup>22</sup> With this technique high-quality epitaxial BSCCO films having a mosaic spread of about 0.1° can be grown. The room-temperature resistivity measured along the *ab* plane is about 250  $\mu\Omega$  cm.

In this work an accurate measure of the ratio  $\rho_c/\rho_{ab}$  between *c*-axis and in-plane resistivities was needed (see the Discussion). Since both  $\rho_c$  and  $\rho_{ab}$  had obviously to be measured on the same sample we developed a technique to measure both  $\rho_c$  and  $\rho_{ab}$  on the same film. In previous papers<sup>6,14,16</sup> the possibility of using misori-

ented substrates (i.e., substrates cut in such a way that their physical surface makes an angle  $\theta \cong 2.5^{\circ}$  with the *ab* plane, small enough that film quality is not affected by substrate misorientation) to measure  $\rho_c$  was demonstrated. By x-ray diffractometry the direction of the projection of the c axis on the film surface can be found and a stripe of length l can be patterned along this direction. The stripe then has an effective length  $l_c = l \sin \theta$  along the *c*-axis direction. Although for  $\theta \cong 2.5^{\circ}$ ,  $l_c/l \cong 1/20$ , the high anisotropy  $\rho_c/\rho_{ab} \approx 10^4$  of BSCCO guarantees that the overall resistivity measured on this stripe is dominated by  $\rho_c$ . With this technique it is possible to measure the transverse resistance on a much longer effective path  $(l_c \approx 100 \ \mu m)$  than that available in single crystals ( $\approx 1 \ \mu m$ ), with a substantial increase in the quality of the data. The angle  $\theta$  is high enough that the mosaic spread of the film can be neglected.

To measure  $\rho_c$  and  $\rho_{ab}$  on the same film, two stripes (length l=2.5 mm, width  $w=200 \ \mu$ m, and thickness  $d\approx0.5 \ \mu$ m) were mechanically patterned on the film, one parallel and the other perpendicular to the direction of the projection of the *c* axis on the film. Measurements performed on the

oxidation states: (a) as-grown, (b) reduced, and (c) oxidized. The fitting parameters are A=8.42,  $T_0=1976$  K,  $T_1=1824$  K for the reduced sample, A=5.31,  $T_0=1972$  K,  $T_1=1829$  K for the asgrown sample, and A=3.87,  $T_0=1949$  K,  $T_1=1854$  K for the oxidized sample.

with Eq. (1) (see text) for a 2212 BSCCO film in three different

FIG. 1. Fit of the experimental  $\rho_c/\rho_{ab}$  vs temperature curves

latter one (perpendicular to the *c* axis) directly give  $\rho_{ab}$ , while on the other one it is possible to measure  $\rho_c$ . The  $\rho_c(T)$  data were corrected for the small  $\rho_{ab}$  contribution.

In this geometry the  $\rho_c$  value must be calculated considering an effective path along the *c* axis  $l_c = l \sin \theta \cong 110 \ \mu\text{m}$  and an effective cross section  $S_c = wd/\sin \theta \cong 2300 \ \mu\text{m}^2$ . Therefore the measured resistance of about 700  $\Omega$  for the as-grown sample corresponds to  $\rho_c \cong 2\Omega$  cm. To test the normal-state transverse resistivity models for different values of the anisotropy of the sample, we annealed it in both oxygen (decrease of the resistivity peak) and argon (increase of the resistivity peak) atmospheres.

## **RESULTS AND DISCUSSION**

In Fig. 1 are shown the results of the fit of the ratio between transverse and in-plane resistivities according to the formula proposed by Abrikosov in Ref. 21,

$$\frac{\rho_c}{\rho_{ab}} = A \frac{\cosh(T_0/T) + \cosh(T_1/T)}{\sinh(T_1/T)}$$
(1)

 $(k_B T_0 \text{ and } k_B T_1 \text{ are respectively the mean energy and the half-width of the energy spread of the resonant defects referred to the Fermi level, <math>k_B$  being the Boltzmann constant) for a 2212 BSCCO film in three different oxygenation states. The fit appears to be very good overall for all oxygen concentrations in the whole temperature range between the temperature  $T_m$  at which the ratio of the resistivities shows a maximum before the transition, and room temperature.

However, a closer look at the data shows that the fit tends to overestimate the experimental data at both high and low temperatures, while underestimating them at intermediate temperatures (except for the oxygen-annealed sample, showing a weak peak). This suggests that it tries to increase the curvature of the theoretical function as much as possible to match the steep increase in  $\rho_c$  just above  $T_c$ , and that therefore a good description of the region just above  $T_c$  (where fluctuation theory predicts a singular behavior) cannot be given by this model. To prove this point we plot in Fig. 2 the





FIG. 2. Curvatures of the experimental  $\rho_c/\rho_{ab}$  vs temperature curves reported in Fig. 1 (open squares, reduced sample; closed circles, as-grown sample; open diamonds, oxidized sample) compared with the curvatures of the theoretical fitting functions (from top to bottom full lines, reduced, as-grown, and oxidized sample).

curvature of the experimental data and of the fitting function above  $T_m$ . The high quality of our data permits us indeed to calculate numerically the curvature of the experimental data at each point without a scatter of the obtained values so high as to hide their behavior. It is clear from Fig. 2 that while the data are only slightly overestimated in this temperature region, the curvature is strongly underestimated for the asgrown and argon-annealed samples, in spite of the fact that the experimental curvature is partly depressed by the AL fluctuation contribution. Moreover, this underestimation disappears (within experimental resolution) as the oxygen content increases. We believe that this is a clear indication that the experimental data show a divergent trend at  $T_c$  (which cannot be accounted for by any normal-state theory) as predicted by the DOS contribution to fluctuations, and that this divergence is counterbalanced by the AL fluctuation contribution very near (a few K) to  $T_c$ . In this scenario the gradual reduction of the peak (i.e., reduced curvature) as the oxygen content is increased is consistent with the higher AL fluctuation contribution in less anisotropic samples.

We notice that when  $T_0, T_1 \ge T$ , as is the case for both our fits and those performed in Ref. 21 on YBCO, Eq. (1) reduces to

$$\rho_c / \rho_{ab} = A [1 + \exp(\Delta/T)], \qquad (2)$$

i.e., and "activated" behavior. Here  $k_B \Delta = k_B (T_0 - T_1)$  is the energy of the lowest-level resonant impurities referred to the Fermi level. It can be seen that only the difference between  $T_0$  and  $T_1$  is important and not their individual values, to which the fit is indeed insensitive as long as  $T_0, T_1 \ge T$ (the fitting parameters values are therefore only indicative). If the previous analysis is correct, i.e., the strong divergence of the exponential at T=0 K "simulates" the weaker divergence of the experimental data at  $T_c$  (except when T is very close to  $T_c$ ), it is to be expected that this model will give a curvature which is too high when applied to the lowest- $T_c$ compound of the BSCCO family (2201 BSCCO) for which  $T_c$  is only about 15 K. Although Eq. (1) was originally developed for a high- $T_c$  compound such as YBCO, it should also describe the R(T) behavior of the low- $T_c$  2201 BSCCO phase, since 2201 and 2212 BSCCO phases only differ in the



FIG. 3. Fit of the experimental  $\rho_c/\rho_{ab}$  vs temperature curves for a 2201 BSCCO crystal (data taken from Ref. 23) with Eqs. (1) (Ref. 21, full line) and (3) (Ref. 18, dotted line).

number of adjacent Cu-O<sub>2</sub> planes. The fit of literature data<sup>23</sup> relative to 2201 BSCCO crystals with Eq. (1) gives poor results, as shown in Fig. 3. Moreover, while for the 2212 phase Eq. (1) gives a lower curvature at  $T_c$  than the experimental one, the opposite is true for the 2201 phase.

The "activated" behavior of  $\rho_c$  predicted by Eq. (1) seems therefore to fail to give a detailed description of the transverse resistivity of layered superconductors for the following reasons.

(1) When  $T_c \approx 100$  K, the curvature of the exponential, which diverges at T=0, is not high enough to account for the apparent divergence of the experimental  $\rho_c$  just above  $T_c$ . This divergence is probably caused by some contribution dependent on  $T-T_c$  rather than T, i.e., some superconducting property.

(2) On the other hand, when  $T_c$  is closer to 0 K, the exponential divergence becomes much higher than the experimental one, which has been found in agreement<sup>6,14</sup> with the logarithmic behavior predicted by fluctuation theory,<sup>12</sup> and the fit becomes much worse.

This suggests that it will be very difficult for any normalstate theory, having a divergence of a given kind at T=0 K, to reconcile both situations. The same does not hold if the peak in  $\rho_c$  has a superconducting origin like the DOS fluctuation contribution, whose divergence shifts with  $T_c$ , while remaining logarithmic<sup>12</sup> in shape in all cases.

We point out that Eq. (2) is very similar to the semiphenomenological formula for  $\rho_c$  proposed in Ref. 18, namely,

$$\rho_c = A + BT + (C/T) \exp(\Delta/T) \tag{3}$$

(here  $\Delta$  is a pseudogap), which therefore faces the same difficulties as Eq. (1) in describing the curvature of the transverse resistance peak just above  $T_c$  in 2212 BSCCO, even if the additional 1/T dependence and the use of four phenomenological parameters slightly reduce the discrepancy with experimental data. On the other hand, this formula correctly describes the  $\rho_c(T)$  curves of the low  $T_c$  2201 BSCCO phase, but the best fit is for  $\Delta=0$  K, when the exponential divergence is canceled and substituted by the 1/T one. In this case, however, Eq. (3) loses its significance.

If the above analysis is correct, i.e., the DOS fluctuation correction to conductivity is responsible for the  $\rho_c(T)$  peak just above  $T_c$ , it is clear that the goal of a normal-state theory of electrical conductivity along the *c* axis in layered superconductors is *not* to give a detailed description of this peak: the normal-state resistivity  $\rho_{Nc}(T)$  curve must rather lie somewhere below the measured  $\rho_c(T)$  one.

In order to give a clue to reasonable behavior of  $\rho_{Nc}(T)$  compatible with the fluctuation origin of the transverse  $\rho_c(T)$  peak, we subtracted from our experimental  $\rho_c(T)$  the calculated contribution due to fluctuations, using reasonable values for the parameters. From formula (30) of Ref. 15 and the expression for  $\rho_{Nc}(T)$  assumed in Ref. 15 it can be shown that considering the DOS, AL, anomalous MT, and regular MT fluctuation contributions to conductivity, we have [for  $\epsilon = \ln(T/T_c) \leq 0.2$ , which is roughly the validity limit of fluctuation theory]

$$\rho_{Nc}(T) = \rho_c(T)f(T), \qquad (4)$$

where the temperature-dependent reduction factor f(T) is

$$f(T) = 1 - \frac{\pi\hbar}{2E_F\tau} \left\{ k \ln \left[ \frac{2}{\epsilon^{1/2} + (\epsilon + r)^{1/2}} \right]^2 + \tilde{k} \left[ \frac{(\epsilon + r)^{1/2} - \epsilon^{1/2}}{r^{1/2}} \right]^2 - \frac{\hbar^2 \alpha}{2\tau^2 J^2} \left[ \left( \frac{\gamma + r + \epsilon}{\epsilon^{1/2} (\epsilon + r)^{1/2} + \gamma^{1/2} (\gamma + r)^{1/2}} - 1 \right) + \frac{1}{2} \left( \frac{\epsilon + r/2}{\epsilon^{1/2} (\epsilon + r)^{1/2}} - 1 \right) \right] \right\}.$$
(5)

In this equation  $\tau$  is the scattering time,  $\tau_{\phi}$  is the phase pairbreaking lifetime, *J* is the interlayer hopping energy, and  $E_F$ is the Fermi energy, while  $\alpha = v_F^2 \tau^2/2 \eta$  ( $v_F$  is the Fermi velocity). The expressions for  $\eta$ ,  $\gamma$ , k,  $\tilde{k}$ , and r can be found in Ref. 15 (where natural units  $c = \hbar = k_B = 1$  were used, c being the speed of light and  $\hbar$  the Planck's constant).

Using Eqs. (4) and (5) we calculated three simulated  $\rho_{Nc}(T)$  curves starting from the measured  $\rho_c(T)$  of the argon-annealed sample, for three different values of  $E_F$  (namely, 0.8, 1.0, and 1.25 eV). These curves are plotted in Fig. 4 together with the experimental  $\rho_c(T)$  for T < 150 K (for  $T > 1.25T_c$  the theory is, however, no longer accurate). In this simulation the critical temperature  $T_{c0}$  is taken to be  $T_{c0}=81$  K from the  $\rho_c(T)$  data, while the values of the parameters  $\tau$ ,  $\tau_{\phi}$ , and J have been taken from literature<sup>6,16,19,24,25</sup> ( $\tau=2\times10^{-14}$  s,  $\tau_{\phi}=2\times10^{-13}$  s, and  $J/k_B=40$  K). The weak temperature dependencies of  $\tau$  and  $\tau_{\phi}$  in the narrow temperature range considered have been neglected. The Fermi energy  $E_F$  is just a scale factor for the global fluctuation contribution  $\sigma_{\rm fl}$ , and in order to keep the



FIG. 4. Sketch of possible behavior of the normal-state transverse resistance of 2212 BSCCO just above the resistivity peak after subtraction from experimental data (open circles) of the fluctuation contribution for three values of the Fermi energy  $E_F$  (from lower to higher full lines,  $E_F$ =0.8 eV,  $E_F$ =1.0 eV, and  $E_F$ =1.25 eV, respectively).

latter within the limits of validity of the theory  $(\sigma_{\rm fl} \ll \sigma_N)$  must be taken of the order of 1 eV, somewhat higher than expected in these materials.

Finally, we stress that because of the less divergent behavior of the simulated  $\rho_{Nc}(T)$  curve as compared to  $\rho_c(T)$ , simpler functional dependencies for  $\rho_{Nc}(T)$  could be compatible with this behavior. This could possibly make the search for a normal-state transverse resistivity theory in layered superconductors simpler.

### CONCLUSIONS

In summary, we presented a detailed analysis of the transverse resistivity  $\rho_c(T)$  curves in both 2212 and 2201 BSCCO samples, and found that recently proposed models for the normal-state  $\rho_{Nc}(T)$  implying an "activated" behavior cannot fully account for the observed behavior, and that some additional contribution other than the normal-state one, giving a divergence at  $T_c$  instead of T=0 K, must also be considered. Previous results concerning both transverse resistance and magnetoresistance indicate that very likely candidates for the origin of this additional contribution are thermodynamic fluctuations of the single-electron density of states. On this basis the expected shape of the normal-state transverse resistivity curve has been derived.

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