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Fluctuation conductivity in cuprate superconductors

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Abstract. We have measured the in-plane resistivity of $Bi_2Sr_2CaCu_2O_{8+\delta}$ and $Tl_2Ba_2CaCu_2O_{8+\delta}$ single crystals in the temperature range 70–300 K. The thermodynamic fluctuations in the conductivity of both the samples start around ~ 125 K. We find the Lawrence and Doniach [1] model to be inadequate to describe the fluctuation conductivity in these materials. The modification suggested by Ramallo *et al* [4] where by the conductivity is enhanced due to the presence of two superconducting layers in each unit cell is also not adequate. We suggest the fluctuation conductivity to be reduced due to the reduction in the density of states (DOS) of the quasiparticles which results due to the formation of Cooper pairs at the onset of the fluctuations. The data agrees with the theory proposed by Dorin *et al* [5] which takes into account this reduction in DOS.

Keywords. BSCCO; TBCCO; resistivity; superconductive order parameter; fluctuations.

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1. Introduction

Despite the large amount of effort put into its study, the paraconductivity ($\Delta\sigma$) in cuprates is not well-understood. In polycrystalline materials, it could be described adequately in terms of the Lawrence and Doniach (LD) theory [1]

$$\Delta\sigma^{\mathrm{LD}} = \frac{A_{\mathrm{AL}}}{\varepsilon}\left[1 + \frac{B_{\mathrm{LD}}}{\varepsilon}\right]^{-1/2} \tag{1}$$

with $A_{\mathrm{AL}} = e^2/16s\hbar$, $B_{\mathrm{LD}} = (2\xi_{\mathrm{c}}(0)/s)^2$ and $\varepsilon = 1 - T/T_{\mathrm{c}}^{\mathrm{mf}}$ where s is the interlayer separation, $\xi_{\mathrm{c}}(0)$ the zero temperature coherence length along the direction perpendicular to the layers, and $T_{\mathrm{c}}^{\mathrm{mf}}$ the mean field critical temperature. In this model, though the layers were assumed to be Josephson coupled, the interaction was treated in terms of an effective mass tensor. This approximation is valid [2] when the coherence length $\xi_{\mathrm{c}}(0)$ is long, and breakes down when it is smaller than the interlayer separation, as is the case in cuprates. Most magnetic measurements [3] yield $\xi_{\mathrm{c}}(0)$ to be ≤ 0.5 Å whereas s is more than 5 Å. Additionally, some cuprates have more than one superconducting layers in each unit cell from where the pairs tunnel to the neighboring layers. Ramallo *et al* [4] have calculated the spectrum of fluctuations of such multiply connected layers within the Ginzberg–Landau (GL) formalism using the free energy expression as generalized by Klemm. They found the original form of $\Delta\sigma^{\rm LD}$ remained unchanged but the conductivity was enhanced by a number $N(\varepsilon)$ which measured the effective number of independently fluctuating superconducting layers. The enhanced conductivity $\Delta\sigma^{\rm LD}$ is given by

$$\Delta\sigma^{\rm LD} = N(\varepsilon)\frac{A'_{\rm AL}}{\varepsilon}\left[1+\frac{B'_{\rm LD}}{\varepsilon}\right]^{-1/2}. \tag{2}$$

$N(\varepsilon)$ depends sensitively on ε and the strength γ_j with which a given layer is coupled to other layers. When one layer of the bilayers in a unit cell interacts very strongly with the other layer than with a layer in the next unit cell ($\gamma_1 \gg \gamma_2$), then the bilayers effectively act as a single layer ($N(\varepsilon) = 1$) for all $\varepsilon \le 1$. However $N(\varepsilon)$ increases when γ_1/γ_2 decreases and approaches a value of 2 for $\gamma_1 = \gamma_2$. This change in the value of $N(\varepsilon)$ is also reflected in the exponent ν which changes from 1 to 0.5 with ε. This change is spread over a wider temperature interval than it is in the case of LD theory.

Alternatively, Josephson interaction has been treated as a pair potential. It has been shown by various models which consider several conducting layers per unit cell, with either interlayer or intralayer coupling, that the interaction is equivalent to a pair potential that is periodic in k_z (the wave-vector component parallel to the c-axis) with the period equal to the c-axis repeat distance d. Using this potential, Dorin *et al* [5] have presented the most complete and most sophisticated calculations of the fluctuation conductivity in materials containing one superconducting layer in each unit cell. However, one important aspect of the fluctuation conductivity has been ignored in all the calculations so far. The density of states (DOS) of the quasiparticles decreases due to the formation of the Cooper pairs. This changes the normal state resistivity which in turn affects the fluctuation conductivity. This reduction has been incorporated by Dorin *et al* in their calculations. They have obtained four contributions to the fluctuation conductivity. With the magnetic field B applied perpendicular to the layers, these contributions are given by

$$\begin{aligned}\Delta\sigma_{ab}^{\rm LD} = \frac{e^2}{4s\hbar}\sum_{n=0}^{\infty}(n+1)\Bigg[&\frac{1}{[(\varepsilon_B+\beta n)(\varepsilon_B+\beta n+r)]^{1/2}}\\ &-\frac{2}{[(\varepsilon_B+\beta(n+\frac{1}{2})(\varepsilon_B+\beta(n+\frac{1}{2})+r)]^{1/2}}\\ &+\frac{1}{[(\varepsilon_B+\beta(n+1))(\varepsilon_B+\beta(n+1)+r)]^{1/2}}\Bigg]\end{aligned} \tag{3}$$

$$\Delta\sigma_{ab}^{\rm DOS} = -\frac{e^2\beta\kappa}{4s\hbar}\sum_{n=0}^{1/\beta}\frac{1}{[(\varepsilon_B+\beta n)(\varepsilon_B+\beta n+r)]^{1/2}} \tag{4}$$

$$\Delta\sigma_{ab}^{\rm MT(reg)} = -\frac{e^2\beta\tilde{\kappa}}{4s\hbar}\sum_{n=0}^{1/\beta}\frac{1}{[(\varepsilon_B+\beta n)(\varepsilon_B+\beta n+r)]^{1/2}} \tag{5}$$

$$\begin{aligned}\Delta\sigma_{ab}^{\rm MT(an)} = -\frac{e^2\beta}{8s\hbar(\varepsilon-\gamma)}\sum_{n=0}^{\infty}\Bigg[&\frac{1}{[(\gamma_B+\beta n)(\gamma_B+\beta n+r)]^{1/2}}\\ &-\frac{1}{[(\varepsilon_B+\beta n)(\varepsilon_B+\beta n+r)]^{1/2}}\Bigg]\end{aligned} \tag{6}$$

where the superscript MT represents the contributions from the Maki–Thompson processes in which the fluctuations get scattered by the quasiparticles. Here, $\gamma = 7\zeta(3)\hbar^2/16\pi^2T^2\tau_\phi\tau k_B^2$, the anisotropy constant $r(T_c) = 7\zeta(3)J^2/8\pi^2k_B^2T^2$, and the constants κ and $\tilde{\kappa}$ in the clean limit are defined as $9.384(\tau T)^2k_B^2/\hbar^2$ and $\pi^2/[14\zeta(3)]$ respectively, where τ is the elastic scattering time, τ_ϕ the time for which the electrons remain locked in phase after the break up of the pair and J the interlayer tunneling energy. The pair breaking effect of the magnetic field has been included in the magnetic pair breaking term γ_B while its effect of reducing the T_c is incorporated in ε_B. At low strengths, since these effects of the field are small, ε_B and γ_B have been expanded to give $\varepsilon_B = \varepsilon + \beta/2$ and $\gamma_B = \gamma + \beta/2$, with $\beta = 4\pi\xi_{ab}(0)^2B/\Phi_0$.

In low fields, the above series can be summed using Euler–MacLaurin theorem. In the absence of the field, the expression in eq. (3) is identical to that of LD conductivity (eq. (1)). However the anisotropy constant r here is a function of temperature and increases as the temperature decreases. This results in the exponent ν to be dependent on temperature.

Ramallo *et al* [4] have shown that the in-plane fluctuation conductivity of $YBa_2Cu_3O_7$ (YBCO) can be completely described in terms of eq. (2) with $N(\varepsilon) = 2$. The DOS or other indirect contributions were not required. Similarly, Pomar *et al* [6] did not require to invoke these terms in the paraconductivity of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO). On the other hand, Livanov *et al* [7] found significant amounts of $\Delta\sigma^{\mathrm{DOS}}$ to be present in their studies on thin films of BSCCO. We had earlier shown [8] that the peak in the c-axis resistivity of BSCCO resulted due to the fluctuations of the order parameter and the DOS conductivity was present in significant amount in the entire mean field region. We have now measured the in-plane resistivity of single crystals of BSCCO and $Tl_2Ba_2CaCu_2O_{8+\delta}$ (TBCCO) and find their fluctuation conductivities also contain the DOS contribution and conform better to the model proposed by Dorin *et al* [5].

2. Experimental details

Single crystals of BSCCO were grown by flux method using Li_2CO_3 as the charge. For TBCCO, the technique of reacting Tl_2O_3 with the precursor $Ba_2Ca_2Cu_3O_\delta$ had to be deployed to reduce the evaporation of Tl_2O_3 at the reaction temperature. The in-plane and out-of-plane resistivities were measured by employing the six terminal technique. Crystals were platelets with typical dimensions $1.5 \times 1 \times 0.02$ mm^3. Indium micrometer soldering was used to attach silver wires on the samples. Further details of the sample preparation and measurement techniques are discussed by Chowdhury *et al* [9].

3. Results and analysis

For BSCCO, above $\sim$130 K, the in-plane resistivity was seen to vary linearly with temperature and could be fitted to $\rho_{abn}(T) = \rho_{ab}(0) + BT$ with an RMS deviation of 0.2%. The deviations from linearity observed below this temperature have been considered to arise from the thermodynamic fluctuations in the order parameter. This $\rho_{abn}(T)$ was subtracted from the measured resistivity $\rho_{ab}(T)$ and the paraconductivity $\Delta\sigma_{ex}(T)$ was obtained as

$$\Delta\sigma_{ex}(T) = 1/\rho_{ab}(T) - 1/\rho_{abn}(T)\,. \tag{7}$$

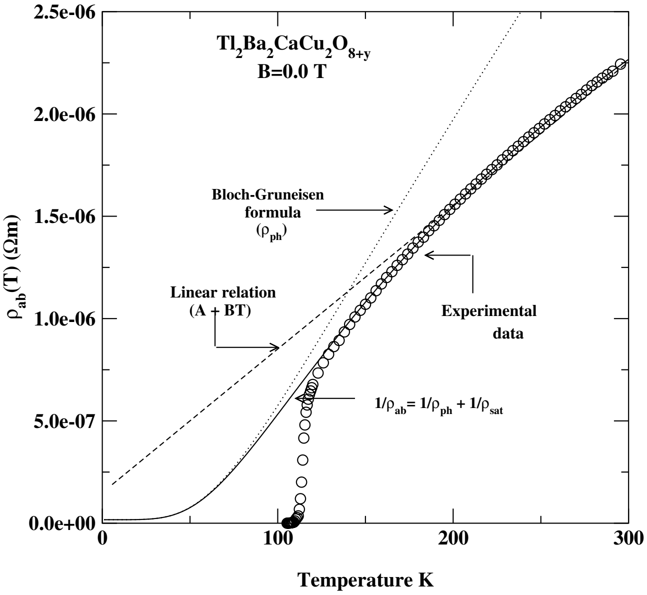

Figure 1. The temperature dependence of the in-plane resistivity, $\rho_{ab}(T)$, measured on a TBCCO single crystal. Open circles are the experimental data. The solid curve is calculated from the model discussed in the text.

In TBCCO, a similar linear behavior was not observed in $\rho_{ab}(T)$ (figure 1). Above $\sim$ 200 K, the curve bends downwards. This is not due to the (oxygen) under-doping of the samples, as a similar curvature was observed in polycrystalline samples also, where the excess oxygen δ was measured by the standard titration technique and was determind to be $\sim$0.24, which is the value observed in optimally doped samples of BSCCO. The high value of T_c obtained here also does not suggest the samples to be under-doped. This leaning of $\rho_{ab}(T)$ curve was attributed [9] to the presence of another scattering process which gives a temperature independent resistivity and operates in parallel with the conventional electron–phonon scattering process governed by the Bloch–Gruneisen relation. The paraconductivity obtained for these samples resembles the $\Delta\sigma_{ex}(T)$ obtained for BSCCO samples both in magnitude as well as in its dependence on temperature as can be seen from $\Delta\sigma_{ex}(T)$ displayed in figures 2 and 3 for BSCCO and TBCCO respectively. This suggests that the background resistivity $\rho_{abn}(T)$ has been estimated correctly for these samples.

As a first step the data was analyzed in terms of the LD theory (eq. (1)). According to this theory, since $B_{\rm LD}$ is independent of T, plots of $(\Delta\sigma_{ex})^{-1}$ vs. T and $(\Delta\sigma_{ex})^{-2}$ vs. T should show straight lines in the 2D and 3D regions with the intercept on the T-axis equal to $T_c^{\rm mf}$. For TBCCO, a straight line for the former plot was obtained with the intercept at 114.2 K. This temperature also matches with the temperature of the inflection point (T_{ci}) in the ρ vs. T curve within the experimental uncertainties. However, the straight line in the latter plot was restricted to just a few points only. This can imply the 3D region to be very narrow, but the value of $\xi_c(0)$ calculated from the cross-over temperature works out to be unsatisfactory. This value, 0.6 Å, does not agree with $\xi_c(0)$ (=1.2 Å) estimated from the magnetization measurements [3]. Similar results were obtained for BSCCO also. With the

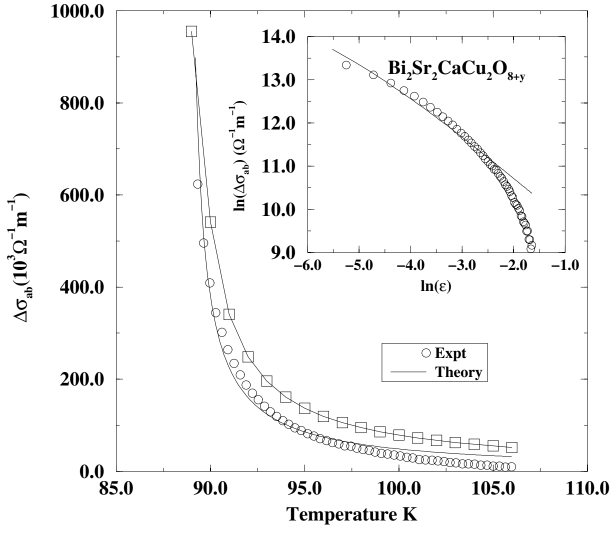

Figure 2. Experimental paraconductivity, $\Delta\sigma_{ab}$, for BSCCO plotted against temperature. The curve is the result of fitting the data to the LD theory (eq. (1)) only. Squares show the fluctuation conductivity calculated from eq. (2). In this and the other plots of $\Delta\sigma_{ab}$ which follow, only a third of the points are shown for clarity of the plots.

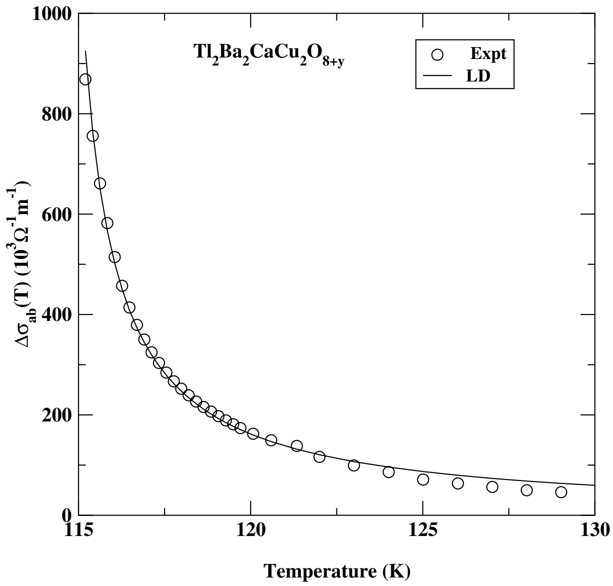

Figure 3. Experimental paraconductivity, $\Delta\sigma_{ab}$, for TBCCO plotted against temperature. The curve is the result of fitting the data to the LD contribution (eq. (2)) only.

T_c^{mf} obtained here, $\Delta\sigma_{ab}$ was plotted against ε on a log–log graph and is shown in the inset of figure 2 for BSCCO. This plot clearly shows a lack of linear behavior between the two quantities. The experimental data falls below the predicted straight line and implies B_{LD} to increase as ε decreases.

A non-linear least square fit of this data with eq. (1) was attempted, by treating T_c^{mf} and $\xi_c(0)$ as adjustable parameters. To improve the agreement, d also had to be considered as a variable parameter. As shown in figure 2 for BSCCO and figure 3 for TBCCO, the agreement between the theory and the data is satisfactory near T_c^{mf} but above $\sim$ 120 K for BSCCO and $\sim$ 130 K for TBCCO the deviations become significantly large. Though the value of T_c^{mf} obtained here agreed with the T_{ci}, $\xi_c(0)$ was found to be small ($\sim$ 0.1 Å). A value $\sim$ 18 Å was obtained for d which is roughly half the value of the c-axis cell parameter.

Identical results were obtained from eq. (2). Its predictions are also shown in figure 2. γ_1 and γ_2 were allowed to vary but a good agreement could only be obtained with $\gamma_1 \sim \gamma_2$ and two interacting layers per unit cell ($N(\varepsilon)$=2) within the temperature range $10^{-2} \le \varepsilon \le 0.1$.

Similarly, the curves calculated for $\Delta\sigma^{LD}$ from eq. (3) were found to lie consistently above the experimental data for temperatures above $\sim$ 120 K for TBCCO (as shown in figure 3) and $\sim$ 98 K for BSCCO. This suggests some other contribution(s) to be present. Accordingly, when $\Delta\sigma^{DOS}$ term was added to $\Delta\sigma^{LD}$, the agreement improved. The RMS deviation dropped from 0.24% to 0.16% (for TBCCO). $\Delta\sigma^{DOS}$ required for the best fit is shown by the dots in figure 4. It is negative as predicted by eq. (3) and is seen to depend on the temperature very weakly. This could be the reason why it was not required in other analyses. $\Delta\sigma^{DOS}$ depends primarily on the elastic scattering time τ. Its value obtained for the two systems are 25fs (TBCCO) and 19fs (BSCCO) and agrees with their values obtained from infra-red conductivities [10].

When both the MT contributions $\Delta\sigma^{MT(reg)}$ and $\Delta\sigma^{MT(an)}$ were added to $\Delta\sigma^{LD}$, the agreement further improved but the improvement was only marginal. The RMS deviation decreased to 0.11% for TBCCO and 0.13% for BSCCO. The total conductivity comprising of all the four contributions is shown in figure 4 by the solid curve. Its agreement with the data is seen to be excellent. As the regular contribution is negative and anomalous term positive, the net MT conductivity works out to be small and nearly temperature independent. Near T_c^{mf}, the MT conductivity changes more rapidly than $\Delta\sigma^{DOS}$. MT-terms added to $\Delta\sigma^{LD}$ do not yield as good results as when $\Delta\sigma^{DOS}$ is added to $\Delta\sigma^{LD}$. The parameter that controls the magnitude of $\Delta\sigma^{MT}$ is τ_ϕ. Its value obtained here is slightly greater than the corresponding τ for both the systems and implies that the quasiparticles obtained from the break up of the pair suffer at least one scattering before they loose phase coherence.

The fluctuation conductivity obtained for BSCCO in the present work agrees in magnitude with that obtained by Pomar *et al* [6] in a similar study. However, they have been able to explain their data in terms of the LD model (eq. (2)) with two interacting layers in a unit cell ($N(\varepsilon) = 2$) and $\xi_c(0) \le 0.5$ Å with an RMS deviation of 3% for both the samples of BSCCO that they studied. Despite this large error, they have not considered the presence of any other contribution to conductivity. Below $\varepsilon \le 0.03$, their data shows appreciable deviations from the fitted curve. This gap can clearly be filled with $\Delta\sigma^{DOS}$.

$\Delta\sigma_{ab}$ obtained in a field of 1 tesla is shown in figure 5. This conductivity is suppressed by three orders of magnitude (approximately) compared to its zero-field value. It was observed that the gap between the data and the predictions of the LD theory (eq. (2)) has increased. Consequently, the proportion of the DOS conductivity will increase. All the four

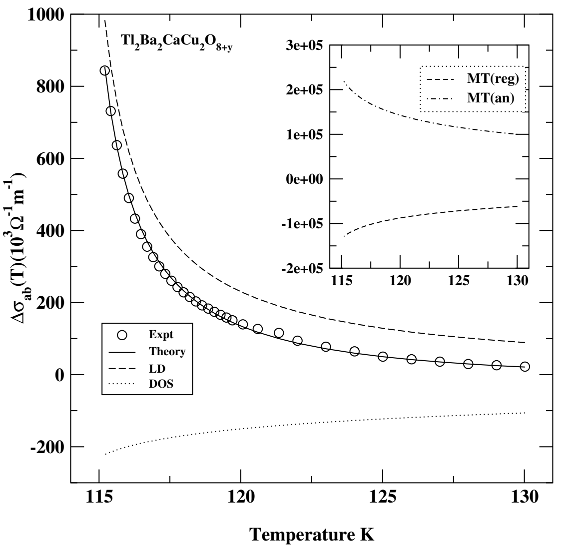

Figure 4. Experimental in-plane excess conductivity, $\Delta\sigma_{ab}(T)$, for TBCCO is plotted against temperature together with the fitted curve (solid line) calculated using LD and DOS and the two MT contributions. The inset shows the MT conductivities varying as a function of temperature.

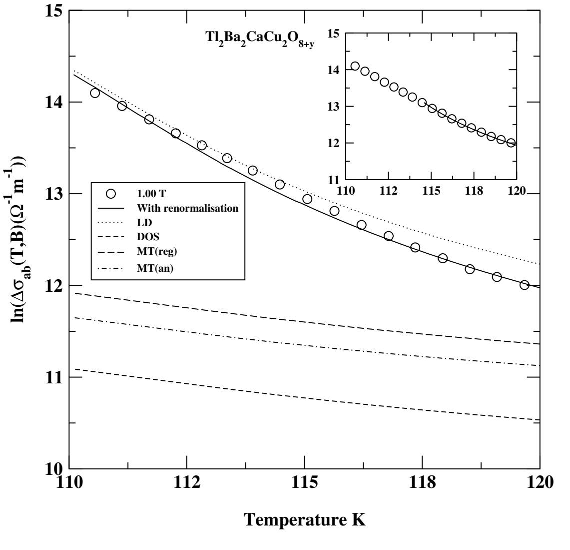

Figure 5. Experimental in-plane excess conductivity data measured in a field of 1 tesla. The calculated curves include the LD, DOS, MT(reg) and MT(an) terms. Inset shows the conductivity calculated without taking into account the renormalization effects.

contributions given by the model of Dorin *et al* are shown here. They have been calculated using the values of the parameters r, $\xi_c(0)$, T_c^{mf} and s obtained from the zero-field data.

Pomer *et al* [6] have observed a large discrepancy of their data measured at 1 tesla from the prediction of eq. (1). The discrepancy exceeds two orders of magnitude for ε below ~ 0.11. They have attributed it to the presence of an inhomogeneous distribution of O_2 within the sample which gives rise to inhomogeneities in the critical temperatures. Their data requires a Gaussian distribution in T_c^{mf} with a standard deviation of 0.6 K and 1.1 K (for the two samples) to match the prediction of the theory. Though the same samples have been used for measurements with and without field, such a distribution of T_c^{mf}'s was not required in the zero-field data.

The values of the parameters obtained here appear to support our analysis. From a similar analysis of the out-of plane fluctuation conductivity [9], we obtain $v_f \sim 4 \cdot 10^5$ cm/s. The anisotropy parameter $r(T_c)$ gives the interlayer tunneling constant J as $13.5k_B$. The in-plane carrier density n calculated from $n = \sqrt{3\pi}[\hbar/(v_f e^2 \tau \rho_{ab})]^{3/2}$ works out to be $4.3 \cdot 10^{20}$ cm^{-3} and agrees with the results obtained from the Hall data on optimally doped BSCCO. These values of v_f and J give the resistivity anisotropy ratio $\rho_c/\rho_{ab} = (v_f \hbar / Js)^2$ to be 5400 at 300 K for BSCCO which agrees with its value 5500 measured at room temperature.

4. Conclusion

We see from the analysis presented here that if we allow deviations of the calculated values from the experimental data to be as large as seen in figure 2, the LD theory alone can account for the fluctuation conductivity. In that case, contributions from indirect processes, particularly $\Delta\sigma^{\rm DOS}$, are not required. Though the absolute value of $\rho_{ab}(T)$ has large uncertainties in its magnitude due to the large errors involved in measuring the dimensions of the samples, its temperature dependence can be determined very accurately. Therefore, only those models whose predictions fall within the errors of measurements of the data should be accepted. From this point of view, we find reduction in DOS of the quasiparticles is contributing sufficiently to the fluctuation conductivity of BSCCO as well as TBCCO.

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