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# $c$-axis response of single- and double-layered cuprates

D. N. Basov
*Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S 4M1*

H. A. Mook
*Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831*

B. Dabrowski
*Department of Physics, University of Northern Illinois, DeKalb, Illinois 60115*

T. Timusk
*Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S 4M1*

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We report on an infrared study of the $c$-axis response of the single-layered superconductor $La_{2-x}Sr_xCuO_4$ ($T_c$=34 K) and analyze the results in comparison with our earlier data for the double-layered $YBa_2Cu_4O_8$ [Basov *et al.*, Phys. Rev. B **50**, 3511 (1994)]. We find that in both materials nonmetallic temperature dependence of the dc resistivity along the $c$ axis results from the transfer of the spectral weight from the far-infrared region to much higher energies. The energy scale associated with this transfer in the normal state is about 200 $cm^{-1}$ in the double-layered system and exceeds 5000 $cm^{-1}$ in the single-layered material. No well-defined pseudogap is found in the latter case. The superconducting state in both systems is highly unusual since the spectral weight of the superfluid $\delta$ function peak is accumulated from the frequencies as high as 20 $kT_c$.

Although the doping and temperature dependences of the transport and magnetic properties of high-$T_c$ superconductors show a number of features common to cuprates with both one and two $CuO_2$ planes per unit cell,[2,3] a more detailed examination revealed certain differences between the two types of materials.[4] For example, a spin gap[5,6] or a pseudogap[7,1] is observed in various experiments on the $YBa_2Cu_2O_8$ (YBCO) materials in the underdoped regime. For the single-layered materials, the experimental situation is not clear. The $c$-axis resistivity of the underdoped $La_{2-x}Sr_xCuO_4$ or $Nd_{2-x}Ce_xCuO_4$ increases at low temperatures[8,9] similar to that of the underdoped YBCO compounds and is not in disagreement with the pseudogap scenario. However the nuclear magnetic resonance (NMR) and neutron-scattering data do not give conclusive support to the pseudogap idea.[10,4]

In this work, we study the far-infrared (FIR) response of single-layered $La_{2-x}Sr_xCuO_4$ crystals with the polarization along the $c$ axis and compare the results with our earlier data for the double-layered $YBa_2Cu_4O_8$.[1] In the latter system, as well as in the underdoped $YBa_2Cu_3O_{7-x}$, (Ref. 7) the $c$-axis conductivity spectra $\sigma(\omega)$ provide compelling evidence for the existence of a pseudogap—a region in the frequency dependence of the conductivity where the magnitude of $\sigma(\omega)$ is strongly suppressed but still remains at a finite value. The principal similarity in the $c$-axis response between the two types of cuprates is that the increase in the dc resistivity at low temperatures results from the transfer of spectral weight from the FIR region to higher energies. The difference between the two systems is that in $La_{2-x}Sr_xCuO_4$ crystals the region of depressed $\sigma(\omega)$ extends to much higher energies than in YBCO materials and no clear low-frequency pseudogap is observed. The superconducting state in both $YBa_2Cu_4O_8$ and $La_{2-x}Sr_xCuO_4$ is unusual since the $c$-axis polarized spectral weight of the superfluid $\delta$ function peak is accumulated from the frequencies which are as high as $20kT_c$. The similarity in the superconducting state properties in both types of materials possibly is due to a common origin of the $c$-axis superconducting response which could be a result of a Josephson coupling between $CuO_2$ layers.

Large single crystals of $La_{2-x}Sr_xCuO_4$ with dimensions $5\times5\times7$ $mm^3$ were grown by the travelling-solvent floating technique.[11] The nominal composition of Sr was $x$=0.15. The critical temperature, determined by magnetization measurements, was 34 K which is lower than the maximum $T_c$ reported for the $La_{2-x}Sr_xCuO_4$ system. The reflectance spectra $R(\omega)$ at different temperatures between 8 and 300 K were measured with the polarization along the $c$ axis in the frequency range 12–7800 $cm^{-1}$. At frequencies up to 18 000 $cm^{-1}$ only room temperature data were obtained. The reflectance is dominated by a series of infrared active phonon modes (Fig. 1, top panel). At $T$=300 K we observe a small increase of $R(\omega)$ in the FIR indicating that the sample is marginally metallic rather than insulating. With decreasing temperature the FIR reflectance declines, which is in agreement with the increase of the dc resistivity observed in different underdoped cuprates. This suggests that the actual content of Sr in our crystal is lower than the optimal doping. In the superconducting state $R(\omega)$ shows a characteristic plasma edge with a plateau below 40 $cm^{-1}$ and a sharp minimum around 50 $cm^{-1}$. The general features of the reflectance spectra are in an agreement with other data.[12,13]

The complex conductivity $\sigma(\omega)+i\sigma_2(\omega)$ was obtained from $R(\omega)$ through Kramers-Kronig analysis. We used data

 

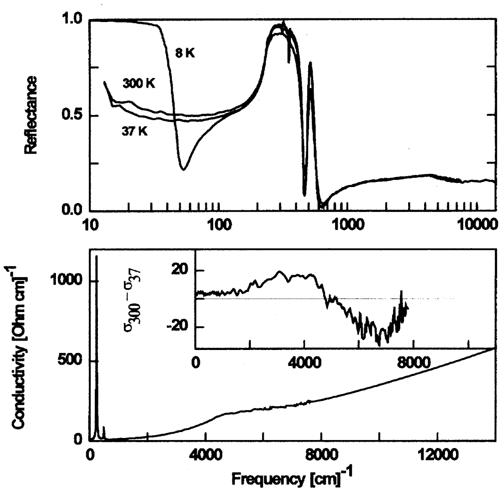


FIG. 1. Top panel: reflectance spectra for polarization along the $c$ axis of a single crystal of $La_{2-x}Sr_xCuO_4$ at different temperatures. Bottom panel: the frequency dependence of the $c$-axis conductivity at 300 K. The inset shows the difference between the conductivity spectra at room temperature and at 37 K with phonon peaks being subtracted as described in the text.

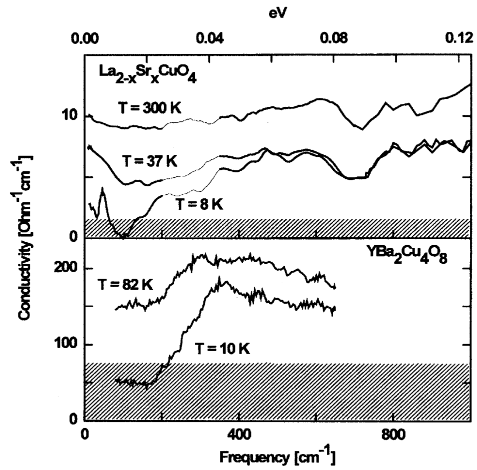


FIG. 2. The $c$-axis conductivity of the single-layered $La_{2-x}Sr_xCuO_4$ (top panel) and of the double-layered $YBa_2Cu_4O_8$ (bottom panel) at different temperatures. Phonon peaks have been subtracted for clarity. The dashed fragments of the spectra in the top panel show the frequency range where the subtraction of phonons has considerable uncertainty. Dashed areas in both panels correspond to $\omega_{ps}^2$—the squared value of the plasma frequency of the superconducting condensate determined from the imaginary part of the conductivity.

by Uchida[14] to extrapolate the experimental reflectance to high frequencies. The particular law of low-frequency extrapolation used is not important in the frequency region where actual data exist. The overall behavior of the real part of the complex conductivity $\sigma(\omega)$ is shown in the bottom panel of Fig. 1. The conductivity rises from very low values in the FIR [about 10 $(\Omega\ \mathrm{cm})^{-1}$] in a nearly linear fashion.

Both the absolute value of $\sigma(\omega\rightarrow 0)$, which is smaller than Mott's minimum metallic value $\sigma_M$ by at least an order of magnitude, and the frequency dependence of $\sigma(\omega)$ suggest that the Drude model is not applicable to the $c$-axis response. It appears that the $c$-axis transport in the normal state is completely incoherent in the single-layered $La_{2-x}Sr_xCuO_4$. The response of the double-layered $YBa_2Cu_4O_8$ is characterized by a higher degree of coherence so that the $\sigma(\omega)$ is nearly frequency independent from 200 $cm^{-1}$ up to 1 eV where the magnitude of the conductivity is comparable to the value of $\sigma_M$. It is likely that the higher conductivity along the $c$ axis in the $YBa_2Cu_4O_8$ crystal is connected with the presence of CuO chains.

In Fig. 2 we plot the conductivity spectra in the infrared region with the phonon peaks subtracted for clarity. In general, the procedures of phonon subtraction by fitting them to Lorentzian oscillators gives unambiguous results for the electronic background. However in the case of $La_{2-x}Sr_xCuO_4$, the mode at 236 $cm^{-1}$ gives rise to an absolute value of $\sigma(\omega)$ which exceeds the electronic contribution by two orders of magnitude. The error of the fit, which is better than 2%, still results in a considerable uncertainty in the vicinity of the central frequency of the mode. This part of the spectrum is shown with a dashed line.

In the normal state, the $c$-axis conductivity of both single- and double-layered cuprates exhibits quite an unusual temperature dependence. The spectral weight in the infrared part of $\sigma(\omega)$ is strongly suppressed as temperature is lowered from 300 K down to $T_c$ and $\sigma(\omega\rightarrow 0)$ is *decreasing*. Therefore, the origin of nonmetallic conductivity along the $c$ axis appears to be similar in both single- and double-layered cuprates. Besides this similarity there are important differences in the frequency dependence of $\sigma(\omega)$ between the two types of materials. In $YBa_2Cu_4O_8$ crystals, as well as in $YBa_2Cu_3O_{7-x}$ samples, the energy scale associated with the increase of the dc resistivity is well defined and is about 200 $cm^{-1}$. Indeed, the conductivity spectra of the double-layered cuprates shows a sharp threshold at about 200 $cm^{-1}$ which develops at temperatures well above the $T_c$ and above this frequency the conductivity is nearly temperature independent.[7,1]

In the single-layered $La_{2-x}Sr_xCuO_4$ the energy scale which is involved in the temperature dependance of the dc resistivity is significantly broader than in $YBa_2Cu_4O_8$ and extends at least up to 0.5 eV. In addition, no sharp onset in the frequency dependence of $\sigma(\omega)$ is found in $La_{2-x}Sr_xCuO_4$ crystals and therefore the pseudogap is not well defined in the single-layered compound.

We emphasize that the similarity in the transport properties between $La_{2-x}Sr_xCuO_4$ and the double-layered systems is not restricted just to the $c$-axis response but is also observed in the in-plane behavior. For instance, the $a$-axis resistivity in the underdoped $YBa_2Cu_3O_{7-x}$ or $YBa_2Cu_4O_8$ shows a kneelike structure at the same temperature (about 200 K) where NMR sees the formation of a spin gap.[15,16] A similar temperature dependence of the resistivity, but with a "knee" at about 500 K, has been observed in the underdoped $La_{2-x}Sr_xCuO_4$ films[17] or crystals.[9] In the optimally doped regime the $T$ dependence of the in-plane resistivity of both single- and double-layer systems follows a linear law.

The oscillator strength sum rule for the conductivity $\int_0^\infty \sigma(\omega)d\omega=\mathrm{const}$ implies that the spectral weight removed from the FIR region must be recovered at higher energies. However an exact destination of the FIR spectral weight remains unclear in the case of the double-layered cuprates.[7,1] That is because it was not possible to measure the reflectance in mid and near infrared with high enough absolute accuracy for the very small samples of YBCO. We have succeeded in performing the necessary measurements for the larger $La_{2-x}Sr_xCuO_4$ sample. These results show that the area missing at low temperatures in the FIR under the conductivity spectra is recovered at frequencies above 5000 $cm^{-1}$. This is illustrated by the inset of the bottom panel of Fig. 1 where we plot the difference between the conductivity at 300 K and 37 K. We show the differential conductivity from the spectra with phonons subtracted as described above to avoid additional complexity in the FIR.

We now turn to the analysis of the changes of $\sigma(\omega)$ that occur with the onset of superconductivity. In both materials the FIR conductivity at $T<T_c$ is further reduced. However in contrast with the normal state, where the spectral weight was transferred to higher frequencies, at $T<T_c$ the missing area is recovered under superconducting delta function at the origin. This conclusion is inferred from the characteristic frequency dependence of the imaginary part of the conductivity which follows $1/\omega$ dependence. The shadowed area in Fig. 2 corresponds to the squared value of the plasma frequency of the condensate $\omega_{ps}^2=\omega 4\pi\sigma_2(\omega)$. The area of the shadowed rectangles is equal within an experimental uncertainty to the integral of $\int(\sigma_{T=T_c}-\sigma_{T=10\,K})d\omega$ in both materials. This is consistent with the notion that the temperature dependence of the conductivity below $T_c$ in both materials results solely from the formation of condensate. Thus it appears that the process responsible for the transfer of the spectral weight to higher energies at $T>T_c$ is no longer active below $T_c$ both in the double-layered system and in the single-layered material. Further study is needed to see if this process is arrested at some temperature above $T_c$ or exactly at $T_c$. In agreement with the latter hypothesis, the NMR Knight shift shows an inflection point at $T_c$.

It is clear from Fig. 2 that the conductivity of $YBa_2Cu_4O_8$ is affected by the superconducting transition over the frequency region extending beyond 1000 $cm^{-1}$ (0.12 eV) and in $La_{2-x}Sr_xCuO_4$—up to 650 $cm^{-1}$ (0.08 eV). Surprisingly spectral weight of the superfluid is accumulated in both single- and double-layered superconductors from frequencies which are abnormally high compared to values of $kT_c$. In the weak-coupling limit most of the weight of the superconducting condensate is collected from the frequencies below $3.5kT_c$. In $La_{2-x}Sr_xCuO_4$ or $YBa_2Cu_4O_8$ less that 25% of the superfluid density comes from $\omega<3.5kT_c$ whereas the remaining 75% of the total density are gained over an energy scale as high as $20kT_c$. We emphasize that the *ab*-plane conductivity of the cuprates is in the clean limit and thus no reliable estimate of the energy scale associated with the superconducting condensation could be obtained from studies of the *ab*-plane response.

The absolute value of the *c*-axis penetration depth, obtained from the imaginary part of the conductivity $\lambda_c^2=c^2/[\omega 4\pi\sigma_2(\omega)]$, is equal to 6.2 $\mu$m for the $La_{2-x}Sr_xCuO_4$ crystal which is considerably larger than the in-plane penetration depth $\lambda_{ab}=2750$ Å, also determined with infrared spectroscopy.[18] In the double-layered $YBa_2Cu_4O_8$ $\lambda_c=9000$ Å, whereas the average in-plane value determined with infrared for a sample from the same batch is about 1400 Å.[19] Therefore, the anisotropy of $\lambda$ appears to be much smaller in this material than in $La_{2-x}Sr_xCuO_4$. However, the detailed analysis of the *c*-axis penetration data carried out in Ref. 1 suggests that the single-layered systems are not necessarily more anisotropic than their double-layered counterparts. In particular, the double-layered $Bi_2Sr_2CaCu_2O_8$ crystals show the highest anisotropy of the *c*-axis superfluid response. It is also shown in Ref. 1 that there is a systematic correlation between the *c*-axis penetration depth and the *c*-axis dc conductivity that is fulfilled for a variety of high-$T_c$ superconductors including $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_4O_8$. This correlation can be described quantitatively in terms of Josephson coupling between $CuO_2$ layers. Thus the occurrence of common features in the superfluid response of the single- and double-layered cuprates is quite natural since in both cases this response originates from the same Josephson coupling process.

The temperature dependence of $\lambda_c$ in $La_{2-x}Sr_xCuO_4$ is also consistent with the model of Josephson coupling. Within the model of a layered superconductor with weak (Josephson) coupling between the layers, the out-of-plane penetration depth is determined by the Josephson current density $J_c$ between the planes and the distance between planes $d$ as[20]

$$\lambda_\perp^2=\hbar c^2/8\pi deJ_c. \tag{1}$$

In the framework of the BCS theory the values of $J_c$ can be related to the normal state resistance $R_n=d/\sigma_{dc}$ and the energy gap $\Delta$ as[21]

$$J_c=[\pi\Delta(T)/2eR_n]\tanh[\Delta(T)/2kT]. \tag{2}$$

From Eqs. (1) and (2) one is able to calculate the temperature dependence of the penetration depth. We have taken the value of $\sigma_{dc}$ from the infrared conductivity extrapolated to zero frequency. Thus the only adjustable parameter is the gap.

The solid line in Fig. 4 has been calculated from Eqs. (1) and (2) and it is clear from the plot that this dependence reproduces the experimental data better than than the $1-(T/T_c)^2$ law which is observed in *s*-wave BCS superconductors or a $1-(T/T_c)^4$ law which is expected within the two-fluid model. This result agrees with the data by Shibauchi *et al.* obtained with microwaves.[22] We emphasize that the model of Josephson coupling not only gives a good fit for the temperature dependence of *c*-axis penetration depth but also reproduces its *absolute value*. The best fit is obtained with $\Delta(0)=41$ $cm^{-1}$. Remarkably, the spectra of $\sigma(\omega)$ show a minimum at the frequency close to $2\Delta(0)$. However, we do not observe any frequency shift of the notch at 80 $cm^{-1}$ as temperature is increased and there is even some reminiscence of this feature in the data at $T>T_c$. Also, the above value of $2\Delta(0)$ appears to be much smaller than the actual frequency range from which the superfluid density is accumulated.

The temperature dependence of $\lambda_c$ in the double-layered material falls in between the $1-(T/T_c)^2$ and $1-(T/T_c)^4$ laws[1] similar to that of $La_{2-x}Sr_xCuO_4$. A more precise comparison with a theory is not possible since $\lambda_c$ of the $YBa_2Cu_4O_8$ has been obtained only at few temperature points.

At temperatures below $T_c$, the FIR conductivity of $La_{2-x}Sr_xCuO_4$ shows a sharp resonance (Figs. 2 and 3) in contrast with the $\sigma(\omega)$ of the $YBa_2Cu_4O_8$ material which is

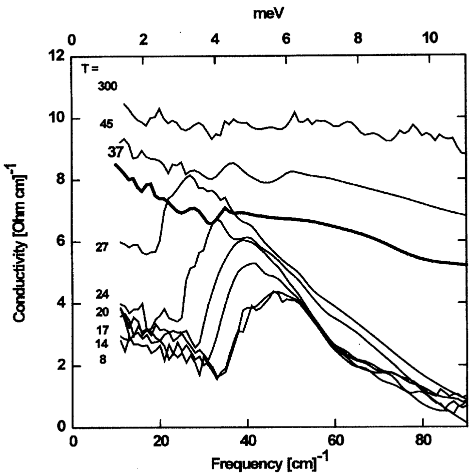


FIG. 3. Spectra of the $c$-axis conductivity of $La_{2-x}Sr_xCuO_4$ showing the temperature dependence of the resonance in the far infrared.

essentially $\omega$ independent below 200 $cm^{-1}$. The frequency of the resonance is temperature dependent and shifts down from 42 $cm^{-1}$ at $T=8$ K as temperature is increasing, as shown in Fig. 4. Neutron-scattering experiments by Mason *et al.*[10] carried out on the crystals from the same source have found a suppression of spin fluctuations below an energy of 6 meV, the energy of our peak in conductivity. The temperature dependence of this energy is also similar to what is shown in Fig 4.[10] While it would be tempting to interprete this feature as evidence for a superconducting gap, such interpretation contradicts our observations that the spectral weight of the condensate originates from the frequencies considerably higher than the position of the resonance. At present the origin of this feature in the conductivity of $La_{2-x}Sr_xCuO_4$ crystal is not well understood. Our results do not rule out the possibility that this feature is present in the normal state just above the superconducting transition, since we are not able to follow the $T$ dependence of the peak below 12 $cm^{-1}$—the position of the resonance at 32 K. Then, the peak could arise from a separate phase transition. We conclude by noting that the presence of the resonance in the FIR conductivity of $La_{2-x}Sr_xCuO_4$ is completely reproducible and is observed in the experiments by other groups.[12,13] The position and temperature dependence is found to be the same by all the investigators but the *peak value* of the conductivity varies from group to group, suggesting that perhaps impurities are involved in the activation of the mode.

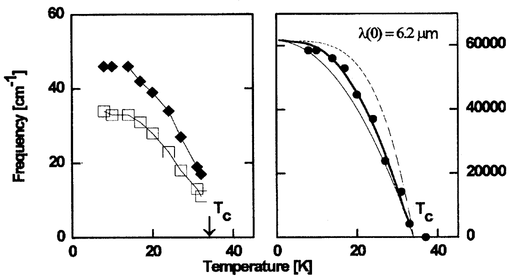


FIG. 4. Left panel: The frequency position of the far infrared resonance as a function of temperature (from Fig. 3). Filled rombs give the frequency at which $\sigma(\omega)$ shows a maximum whereas open squares correspond to the position of the onset of the peak. Right panel: the temperature dependence of squared plasma frequency $\omega_{ps}^2$ for $La_{2-x}Sr_xCuO_4$ crystal. The thick solid line is fit within the model of Josephson coupling between the $CuO_2$ layers [Eqs. (1) and (2)]. Thin solid line and the dashed line are the functional dependences $1-(T/T_c)^2$ and $1-(T/T_c)^4$, respectively.

In summary, we examined the $c$-axis response of the single-layered cuprate superconductor $La_{2-x}Sr_xCuO_4$ and compared it with our earlier data for the double-layered $YBa_2Cu_4O_8$. We found the energy scale associated with nonmetallic temperature dependence of the dc resistivity is different at least by an order of magnitude in these materials. In addition no clear low-lying pseudogap is found in the single-layered system. Despite the differences in the normal state properties, the superconducting state is quite similar in both types of materials. This includes abnormaly broad frequency region associated with the formation of superfluid and also the temperature dependence of the superfluid density. These similarities indicate on the common origin of the superconducting response along the $c$ axis in both single- and double-layered cuprates.

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