

**IN-PLANE OPTICAL RESPONSE OF UNDERDOPED
 $\text{La}_{2-x}(\text{Ca,Sr})_x\text{CaCu}_2\text{O}_{6+?}$ SINGLE CRYSTALS: EVIDENCE
FOR INTRINSIC INHOMOGENEITY**

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In-plane optical response of underdoped $\text{La}_{2-x}(\text{Ca},\text{Sr})_x\text{CaCu}_2\text{O}_{6+\delta}$ single crystals: evidence for intrinsic inhomogeneity

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Abstract

The in-plane optical properties of two crystals of the bilayer cuprate $\text{La}_{2-x}(\text{Ca},\text{Sr})_x\text{CaCu}_2\text{O}_{6+\delta}$, one with excess Ca and $x = 0.10$ and the other with Sr and $x = 0.15$, were investigated over the frequency range of 45–25000 cm^{-1} . A metallic response both in frequency and temperature was observed for Sr=0.15 superconducting sample at low frequencies. Meanwhile, the sample also exhibits a prominent charge-transfer excitation at around 15000 cm^{-1} . This observation, together with neutron experiments performed on the same sample showing diffuse, elastic antiferromagnetic scattering, indicate that the quasi-mobile carriers coexist at low temperature with static antiferromagnetic clusters. This coexistence indicates intrinsic spatial inhomogeneity.

Key words: $\text{La}_{2-x}(\text{Ca},\text{Sr})_x\text{CaCu}_2\text{O}_{6+\delta}$; optical conductivity; inhomogeneity

High temperature superconductors are doped Mott insulators. A fundamental question is whether or not the doped holes in cuprates are homogeneously distributed. Up to now, there have been considerable experimental evidences for charge inhomogeneities in high- T_c cuprates, in particular, for samples at the low doping levels. One of such inhomogeneities is the striped phase in which the charge carriers are confined to separate linear region. Dynamical stripe fluctuations have been indicated for many high- T_c superconductors. The pinned or static stripe phases have been observed in Nd-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4+\delta}$ [1] and very underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [2]. Although it was found that the pinned charge stripes could coexist with the superconductivity, the superconducting transition temperature is usually very low.

Among all known bilayer cuprates, the La-based system $\text{La}_2\text{CaCu}_2\text{O}_{6+\delta}$ (La2126) phase could be regarded as the simplest one.[3] The structure consists of a pair

of pyramidal Cu-O layers facing one another, which are the only electronically active elements. But unlike other bilayer cuprates, whose maximum T_c of about 90 K could be easily achieved, it has proven to be difficult to raise the superconducting transition temperature of this system. The highest T_c obtained even in polycrystalline samples is only around 60 K.[3] It would be interesting to investigate the difference between La2126 and other bilayer superconducting cuprates. Here, we present the in-plane reflectivity and optical conductivity data for single crystals of $\text{La}_{1.9}\text{Ca}_{1.1}\text{Cu}_2\text{O}_{6+\delta}$ and $\text{La}_{1.85}\text{Sr}_{0.15}\text{CaCu}_2\text{O}_{6+\delta}$. We shall illustrate that there exists some sort of inhomogeneity in those samples.

Large single crystals of $\text{La}_{2-x}(\text{Ca},\text{Sr})_x\text{CaCu}_2\text{O}_{6+\delta}$ were grown by the traveling-solvent floating-zone technique. SQUID magnetization measurements indicate that the $x = 0.10$ sample is not superconducting down to the lowest measurement temperature ~ 2 K, but that the Sr-substituted sample exhibits a superconducting response with an onset temperature of 30 K. The polarized reflectance measurements from 45 to 25000 cm^{-1}

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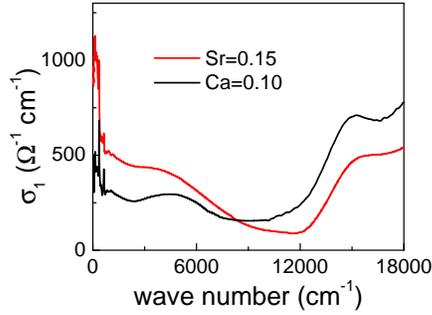


Fig. 1. The frequency dependent conductivity of $\text{La}_{2-x}(\text{Ca,Sr})_x\text{CaCu}_2\text{O}_{6+\delta}$ with $x=0.10$ (Ca-doped) and $x=0.15$ (Sr-doped) at room temperature.

for $\mathbf{E}||a$ -axis were carried out on a Bruker 66v/S spectrometer on polished surfaces of crystals. The optical conductivity spectra were derived from the Kramers-Kronig transformation.

Figure 1 shows the in-plane optical conductivity spectra of the two crystals at room temperature. In accord with the nominal hole concentrations, the Sr-doped sample has larger spectral weight at low frequency. Note that the conductivity spectra have three distinct absorption features: a Drude like response at low frequency, a mid-infrared band, and a Cu-O charge-transfer (CT) excitation near 15000 cm^{-1} . Usually, the CT excitation is prominent for parent compound, and disappears gradually with the carrier doping. The apparent CT-excitation indicates that either the sample is close to the parent compound or it contains insulating parts in the sample. However, the low- ω Drude response suggests the metallic nature of the sample.

Figure 2 shows the reflectance and conductivity spectra below 2000 cm^{-1} at different temperatures for the Sr=0.15 sample. The reflectance increases with decreasing temperature, evidencing metallic temperature dependence. In accord with this, the low- ω conductivity become significantly enhanced at low temperature. The coexistence of the CT excitation and the metallic temperature and frequency response is a significant property of the Sr-doped superconducting sample.

We note that the $\sigma_1(\omega)$ at 200 K and 300 K exhibits a peak at finite frequency, reaching 150 cm^{-1} at 300 K, in contrast to the $\omega = 0$ peak expected for standard Drude behavior. Since the low- ω peak has been observed in striped sample,[4] one might think that similar stripe phase exists in the sample. However, recent neutron scattering experiments performed on the same samples have found no evidence of any charge ordering, instead, diffuse elastic scattering from antiferromagnetic clusters has been observed.[5].

We emphasize here that the infrared data showing a prominent CT excitation at high frequencies is consistent with the neutron result. Those observations,

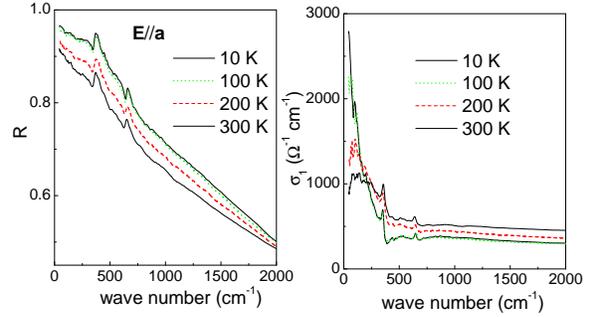


Fig. 2. The frequency dependent reflectance and conductivity of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CaCu}_2\text{O}_{6+\delta}$ at different temperatures.

together with a Drude-like response and a metallic temperature dependence at low frequencies, suggest that the mobile charge carriers coexist at low temperature with static antiferromagnetic clusters, indicating clearly the inhomogeneity of charge carrier distribution.

Additionally, we found that there is no obvious difference between the reflectance below and above superconducting transition temperature. This indicates that the condensed superconducting carrier density is very small in this sample. The small superconducting condensate is qualitatively consistent with the low T_c of the material; however, it seems to be much smaller than one would expect based on the "Uemura plot", [6] suggesting that the superconductivity is likely restricted to a small fraction of the sample volume.

To conclude, the holes in the lightly-doped La_{2126} are not homogeneously distributed. Mobile charge carriers coexist at low temperature with static antiferromagnetic clusters.

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References

- [1] J. M. Tranquada, et al., *Nature (London)* **375** (1995) 561.
- [2] H. A. Mook, et al., *Phys. Rev. Lett.* **88** (2002) 097004.
- [3] R. J. Cava, et al., *Nature (London)* **345** (1990) 602.
- [4] M. Dumm et al., *Phys. Rev. Lett.* **88** (2002) 147003.
- [5] M. Hücker, Y. J. Kim, J. M. Tranquada, G. D. Gu, and J. W. Lynn, unpublished; C. Ulrich, et al., *Phys. Rev. B* **65**, 220507(R) (2002)
- [6] Y. J. Uemura, et al., *Phys. Rev. Lett.* **62** (1989) 2317.