Anisotropic Electromagnetic Response of Lightly Doped $La_{2-x}Sr_xCuO_4$ within the CuO_2 Planes

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Using infrared spectroscopy, we show that spin self-organization in untwinned $La_{2-x}Sr_xCuO_4$ (LSCO) crystals has profound consequences for the dynamical conductivity $\sigma(\omega)$. The electronic response of CuO_2 planes acquires significant anisotropy in the spin ordered state with enhancement of the conductivity along the direction of the diagonal spin stripes by up to a factor of 2. An examination of the anisotropic response indicates that the diagonal spin texture in weakly doped LSCO is also accompanied by the modulation of charge density. The electronic response of the charge stripes is found to be gapless consistent with the hypothesis of the metallic ground state. Our experiments directly show that the striped ordered systems reveal new degrees of freedom not present in ordinary one-dimensional conductors.

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Physics of doped Mott-Hubbard insulators is extraordinary rich and challenges conventional theories of metals. At least in part, the complexity of these systems is related to their intrinsic tendency towards the formation of electronic and/or magnetic inhomogeneities. Particularly interesting examples of inhomogeneities occur in layered transition-metal oxides where spins and/or charges segregate in one-dimensional (1D) self-organized regions referred to as stripes [1–6]. The relevance of these objects to electronic properties and high-temperature superconductivity have been the subject of intensive discussion. This motivates a detailed examination of the formation of the spin- and/or charge-ordered textures in prototypal doped Mott-Hubbard insulators such as $La_{2-x}Sr_xCuO_4$ (LSCO) and related compounds. Although neutron and x-ray scattering techniques have revealed static spin and charge stripes in $La_{2-x-y}Nd_ySr_xCuO_4$ (LNSCO) [2,3,6] the experimental situation in LSCO remains unresolved [7–12]. Despite the fact that static charge stripes have not been detected thus far in LSCO [13], there are clear parallels in the doping dependence of the magnetic structure seen in LSCO and LNSCO [13,14].

Apart from the above controversy, the role that spin and/or charge modulations play in the electronic transport of cuprates is yet to be systematically explored. One natural consequence of the stripes picture is that the electronic properties of CuO_2 planes are expected to acquire strong anisotropy in the stripe-ordered state. Weakly doped phases of LSCO with $x < 0.05$ which can be prepared in single-domain form indeed show anisotropic DC resistivity ρ_{DC} [9]. However, it is difficult to reconcile the relatively small (50%) anisotropy of ρ_{DC} with a hypothetical picture viewing stripes in cuprates as *rivers of charge* rigidly embedded into an antiferromagnetic (AF) insulator. Here we report on the studies of the dynamical conductivity at infrared (IR) frequencies that have allowed us to gain insights into the connections between anisotropic response and unidirectional spin structure in weakly doped LSCO.

An important virtue of IR spectroscopy pertinent to the issues of self-organized charge inhomogeneities is that the analysis of charge dynamics enables an experimental survey of the electronic anisotropy within the CuO_2 plane provided data is accumulated with polarized light. Dissipation of energy associated with both free and bound charges produces additive contributions to the spectra of the real part of the complex conductivity $\sigma_1(\omega)$ which can be readily obtained from reflectivity $R(\omega)$ measurements using Kramers-Kronig (KK) analysis. $R(\omega)$ data for untwinned LSCO crystals with $x = 0.03$ and 0.04 were collected at frequencies from 20 to 48 000 cm^{-1} (2.5 meV–6 eV) and temperatures 13 K $\leq T \leq$ 293 K. For the purpose of the KK analysis we used ρ_{DC} for the low-ω extrapolation and previously published results [15] for the high-ω extension of our data. Single crystals of LSCO were grown by the traveling-solvent floating-zone technique and detwinned under uniaxial pressure [9]. In LSCO crystals with $x < 0.06$ the spin modulation is along the b_{ortho} axis of the orthorhombic phase indicating that the spin stripes are running along the a_{ortho} axis [13]. We will employ the notation $\sigma_a(\omega)$ and $\sigma_b(\omega)$ for the real part of the conductivity determined along and perpendicular to the direction of spin stripes, respectively.

In Fig. 1 we display the optical conductivity for both studied LSCO samples in polarizations $E \parallel a$ and $E \parallel b$. At room temperature, $\sigma_a(\omega)$ and $\sigma_b(\omega)$ are nearly featureless in all panels apart from the frequency regions punctuated by sharp phonon peaks. The phonon spectrum is strongly anisotropic: we find seven modes in the $\sigma_b(\omega)$ and four modes in the $\sigma_a(\omega)$ data in accord with the

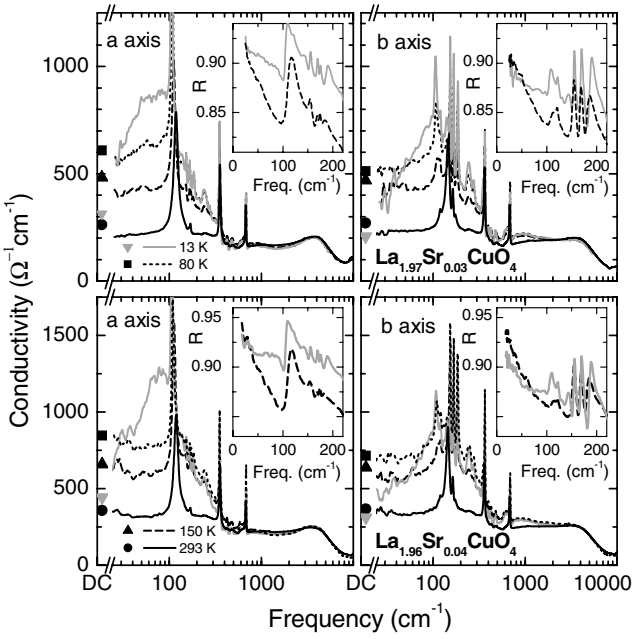

FIG. 1. Frequency dependence of the real part of the in-plane conductivity $\sigma_1(\omega)$ of $La_{1.97}Sr_{0.03}CuO_4$ (upper panels) and $La_{1.96}Sr_{0.04}CuO_4$ (lower panels) along the orthorhombic a and b axis. The symbols on the left ordinates represent the in-plane DC conductivity σ_{DC}. The insets show the spectra of the reflectivity R at 13 and 150 K for $\omega < 220\ \mathrm{cm}^{-1}$.

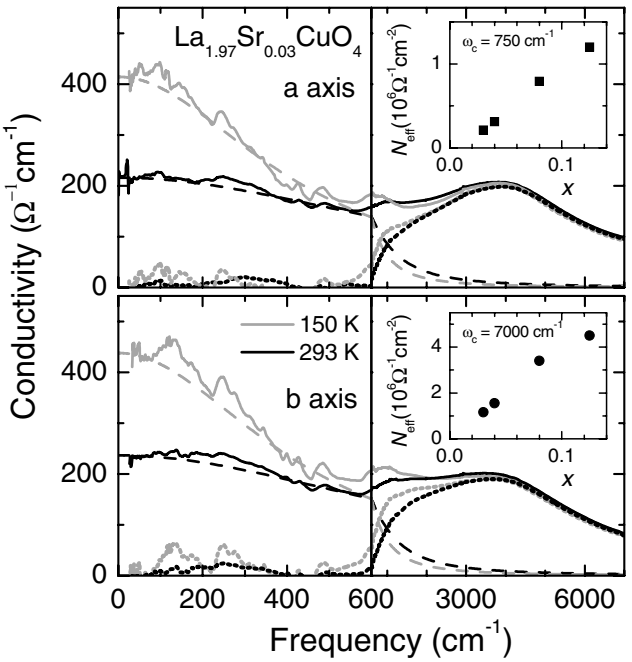

FIG. 2. Conductivity of $La_{1.97}Sr_{0.03}CuO_4$ along a and b axes at 150 and 293 K with phonon modes subtracted (solid lines). The dashed lines represent fits of the FIR conductivity with the standard Drude model. The dotted lines were calculated by subtracting the fits from the measured data. The Drude-fit parameters are $\tau^{-1} = 430\ \mathrm{cm}^{-1}$ (150 K) and $815\ \mathrm{cm}^{-1}$ (293 K) and $\omega_p = 3250\ \mathrm{cm}^{-1}$ for $\sigma_a(\omega)$, and $\tau^{-1} = 430\ \mathrm{cm}^{-1}$ (150 K) and $800\ \mathrm{cm}^{-1}$ (293 K) and $\omega_p = 3400\ \mathrm{cm}^{-1}$ for $\sigma_b(\omega)$. The insets show N_{eff} obtained from our $\sigma_1(\omega)$ data on various LSCO crystals as a function of doping ($x = 0.03, 0.04, 0.08, 0.125$) at $T = 150$ K with the integration limits $\omega_c = 750$ and $7000\ \mathrm{cm}^{-1}$. N_{eff} of the untwinned samples ($x = 0.03, 0.04$) represents the average of the a and b axes.

group theory for the orthorhombic space group D_{2h}^{18} [16]. The anisotropy gradually develops with decreasing temperatures [17]. To the best of our knowledge, untwinned single crystals of LSCO have not been previously investigated with IR methods, and data in Fig. 1 present the first observation of the anisotropic lattice response. The electronic background reveals a significant temperature dependence over a broad energy range. As T is lowered down to 80 K we observe an enhancement of $\sigma_1(\omega)$ in the far-infrared (FIR) frequency region. Changes of the conductivity observed at $T > 80$ K are consistent with narrowing of the Drude mode. This is detailed in Fig. 2 where we show data for $x = 0.03$ sample along with the fits of the electronic background using simple Drude formula. We also show the differential spectra $\sigma^{\mathrm{diff}} = \sigma_1^{\mathrm{exp}} - \sigma_1^{\mathrm{Drude}}$ for both polarizations. At $\omega < 600\ \mathrm{cm}^{-1}$, $\sigma^{\mathrm{diff}} = 0$ within the uncertainty of the conductivity data confirming that the Drude description is adequate for the FIR region. A Drude-like response in $La_{1.97}Sr_{0.03}CuO_4$ suggests that the electronic conductivity even in lightly doped La-based materials is bandlike. This conclusion is in accord with the continuous linear progression of the electronic spectral weight $N_{\mathrm{eff}} = \int_0^{\omega_c} \sigma_1(\omega) d\omega$ (ω_c is a cutoff frequency) in LSCO with doping x from lightly doped phases well into the metallic regime (insets of Fig. 2, [15]). Other confirmations of bandlike behavior come from the analysis of carrier mobility [8] as well as from observations of well-defined quasiparticles peak and the "Fermi arc" in photoemission measurements [11].

Below 80 K, the FIR conductivity undergoes qualitative changes (Fig. 1): the Drude-like behavior seen at higher T evolves into a peak centered at finite ω. Such low frequency modes are commonly found in the $\sigma_1(\omega)$ data for low-dimensional disordered conductors [19] and can be attributed to disorder-induced weak localization. Both the frequency position of the peak in the optical spectrum and the temperature position of the maximum in DC conductivity allow one to quantify the energy scales of localization in the studied materials [19]. The absence of an optical excitation gap in $\sigma(\omega)$ and the linear increase of N_{eff} with doping unravel the nature of the so-called "insulating" state of lightly doped LSCO: the crossover from metallic to "semiconducting" DC transport at $T \leq 80$ K [Figs. 3(a) and 3(b)] occurs due to modifications of the dynamics of the metallic carriers and not due to the opening of a charge gap. Combined together, transport and spectroscopic data suggest that the nature of the insulating state realized in lightly doped LSCO should be understood in terms of

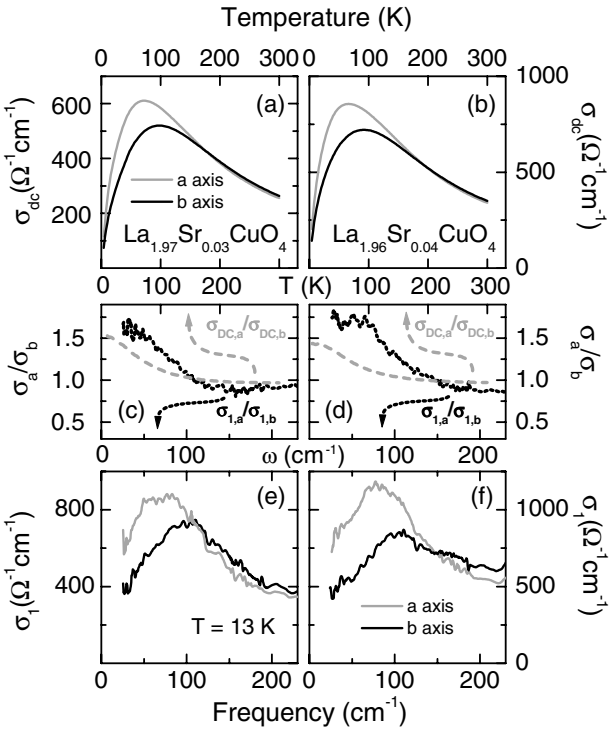

FIG. 3. Anisotropic conductivity of $La_{1.97}Sr_{0.03}CuO_4$ (left-hand panels) and $La_{1.96}Sr_{0.04}CuO_4$ (right-hand panels). Panels (a) and (b) show $\sigma_{DC}(T)$; panels (e) and (f) show $\sigma_1(\omega)$ along and across the direction of spin stripes. The lattice contribution has been subtracted from $\sigma_1(\omega)$ in (e) and (f) by fitting the phonon peaks to Lorentzian oscillators. Panels (c) and (d) display anisotropy of both DC and AC conductivities. Dashed gray line: $\sigma_{DC,a}/\sigma_{DC,b}(T)$ (upper abscissa); dotted black line: $\sigma_{1,a}/\sigma_{1,b}(\omega)$ (lower abscissa) at 13 K.

an Anderson insulator as opposed to an insulator with an energy gap.

A remarkable feature of weakly doped LSCO crystals is the anisotropy of both σ_{DC} and σ_1 at low T with unmistakable parallels between the two data sets (Fig. 3). Anisotropy starts developing in the Drude regime around 80 K (see lower panels of Fig. 1) and increases towards lower temperatures and frequencies approaching $\sigma_a/\sigma_b \leq 2$ as $(T, \omega) \rightarrow 0$. Interestingly, the data directly shows that at low T and ω the conductivity is *enhanced for charges propagating along the spin stripe direction.* Nevertheless, before this effect is assigned to direct involvement of diagonal stripes in charge transport it is imperative to explore possible roles of orthorhombicity in the observed behavior. Both $x = 0.03$ and $x = 0.04$ crystals are mildly orthorhombic at all $T < 400$ K [17,20]. However, the electronic anisotropy develops only at $T \leq 80$ K and is maximized in the limit of low T and low ω where conductivity reveals localization behavior. Anisotropy of the electronic response in orthorhombic systems is usually associated with the enhancement of the electronic spectral weight for data collected with the polarizations along the shorter crystal axis. This latter effect is obvious within a tight-binding picture directly relating the hopping integral t to the magnitude of N_{eff} defined above. Contrary to tight-binding expectations we find that N_{eff} in both LSCO systems is slightly smaller (by 5%) for the direction of higher σ_{DC} [21]. We therefore conclude that the observed anisotropy cannot be easily accounted for by orthorhombicity calling for alternative explanations.

Considerable electronic anisotropy observed within the nearly square CuO_2 planes is difficult to reconcile with a homogeneous charge distribution but is in accord with the picture of metallic charge stripes. Below, we will explore strengths and pitfalls of the stripes scenario in the context of data displayed in Figs. 1 and 2 . An important result pertinent to the stripe interpretation of the anisotropy was obtained from the period of the spin modulation detected in lightly doped LSCO [13]. Matsuda *et al.* found that diagonal stripes in this doping range are neither half-filled like parallel stripes in $La_{2-x-y}Nd_ySr_xCuO_4$ (0.5 hole/Cu) nor completely filled like diagonal stripes in insulating $La_{2-x}Sr_xNiO_4$ (1 hole/Ni) [2,23]. They suggested that the hole concentration on the stripes progressively evolves from 0.5 hole/Cu at $x = 0.08$ to 1 hole/Cu at $x = 0.02$ in $La_{2-x}Sr_xCuO_4$. A filling factor of around 0.8 hole/Cu for $x = 0.03$ and 0.04 doping permits at least in principle metallic character of stripes conductivity. As pointed out above, substantial electronic spectral weight present down to the lowest frequencies supports the notion of metal-like transport even in the case of the $x = 0.03$ system.

Radical departures of the moderately anisotropic response seen in LSCO from that of quasi-1D organic conductors [22,24] appear to be in conflict with the idea of statically ordered charge stripes rigidly embedded in and AF insulator host. This points to the relevance of the notion of the electronic liquid crystal [25] to the description of the transport/IR data where additional electronic degrees of freedom result in part from stripes meandering and transverse mobility. It is worth noting that the occurrence of charge stripes in 2D doped antiferromagnets has been devised as a purely electronic effect. On the contrary, the origin of the 1D conduction in organic materials is directly linked to the underlying crystal structure and directionality of the chemical bonds which are responsible for drastic difference between charge transport parallel and perpendicular to the molecular stacks. In cuprates each single stripe as well as the large fragments of the stripes texture can become mobile in the transverse direction [5]. Then the resulting (weak) anisotropy is a competition between the transverse degrees of freedom (not present in other 1D systems) with unidirectional conduction along the metallic stripe. Several other factors may lead to a suppression of the electronic anisotropy. Most notably, disorder of stripes [26] as well as additional transverse mobility of charges due to the finite width of stripes [7,27] will reduce the contrast between ''parallel''

and "perpendicular" response. Stripes fluctuations are commonly invoked to explain mild observable displays of such a drastic effect as formation of stripes order. However, solely fluctuating stripes cannot fully account for the data in Figs. 1 and 2 since electronic anisotropy increases at low ω and persists even for $\omega \to 0$.

A picture that emerges from the discussion above is that the formation of an ordered spin state in LSCO may be accompanied by a modulation of charge density. Specifically, the localization peak in $\sigma_1(\omega)$ provides strong support for the stripelike charge/spin self-organization scenario in LSCO. Naturally, in the regime of quasi-1D segregation charge carriers are especially susceptible to localization by disorder [28]. It is therefore not surprising that high-T transport in lightly doped LSCO is isotropic and metallic at the same time, whereas anisotropy developing at lower T promotes localization trends. Interestingly, the diagonal charge modulation in LSCO appears at higher temperatures than spin ordering [29]. This is consistent with general patterns of stripes formation in oxides [2,30].

Finally, we comment on the absorption structure in the mid-IR frequency region (right-hand panels of Fig. 2) common to many other transition-metal oxides [31,32]. Several groups discussed possible links between spin/charge stripe formation in nickelates or cuprates and the mid-IR band around 0.5 eV [23,33,34]. The feature is present both for polarization parallel and perpendicular to the spin stripes in LSCO. Differential spectra in Fig. 2 uncover some temperature dependence for $\omega < 3000\ \mathrm{cm}^{-1}$ and $T > 100$ K. This part of the absorption band has been attributed to small polarons [35]. The conductivity is slightly higher perpendicular to the stripes in this region. Remarkably, the mid-IR absorption remains unchanged for both dopings when anisotropy in FIR conductivity develops at low T.

In conclusion, our IR studies of untwinned single crystals suggest a bandlike conductivity even in lightly doped LSCO. The crossover to insulating behavior observed in these transition-metal oxides below $T \approx 80$ K is due to modification of the dynamics of conducting carriers and not due to opening of an electronic gap. In the limit of low temperatures and frequencies, we observe a substantial anisotropy of the IR conductivity which is at odds with the picture of homogeneous charge distribution. Our results point to the fact that the diagonal spin stripes in the doped Mott-Hubbard insulator LSCO are also associated with charge modulation. In contrast with the response of conventional 1D conductors we find similar magnitude of hopping integrals along and across the stripe direction. Distinctions in the phonon structure probed along and across the stripes directions can be attributed to the symmetry of the crystal unraveling only weak (or nonexistent) coupling of the charge-ordered texture to the lattice.

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