

## Dynamical Properties of Charged Stripes in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

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Inelastic light-scattering spectra of underdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  single crystals are presented which provide direct evidence of the formation of quasi-one-dimensional charged structures in the two-dimensional  $\text{CuO}_2$  planes. The stripes manifest themselves in a Drude-like peak at low energies and temperatures. The selection rules allow us to determine the orientation to be along the diagonals at  $x = 0.02$  and along the principal axes at  $x = 0.10$ . The electron-lattice interaction determines the correlation length which turns out to be larger in compound classes with lower superconducting transition temperatures. Temperature is the only scale of the response at different doping levels demonstrating the importance of quantum critical behavior.

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Spin-charge separation is a well-known phenomenon in one-dimensional (1D) conductors [1]. It has also been proposed to occur in the essentially two-dimensional (2D) copper-oxygen planes of high-temperature superconductors [2–4]. Under certain circumstances static charged “stripes” in an antiferromagnetically ordered matrix [5] are observed such as sketched in Fig. 1. The phenomenon can be envisaged as a periodic charge modulation best comparable to a density wave. Moreover, there are several scenarios in which charge and spin ordering play a pivotal role in explaining superconductivity [6,7]. In particular, charge-ordering fluctuations would be capable of providing an effective mechanism for the formation of Cooper pairs [6]. Therefore, apart from being an interesting phenomenon in itself, the understanding of the dynamics of stripes is an important issue in the cuprates.

In recent years a great variety of methods has been employed to study stripes [8]. In underdoped cuprates regular patterns in the electron distribution can eventually be observed in the pseudogap state [9] by scanning tunneling microscopy [10]. Infrared spectroscopy (IRS) and inelastic light scattering show the *dynamical* behavior particularly well. Strong peaks at low but finite energy develop in the conductivity  $\sigma'(\omega, T)$  and in the Raman response  $\chi''(\omega, T)$  [11–13]. Explicit calculations demonstrate the existence of an absorption at low energy due to a transverse excitation of charged stripes [14]. As in the case of IRS the Raman spectra and their temperature dependence observed in  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$  [12] are similar to the results in the ladder compound  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  [15,16]. Therefore, it is qualitatively clear which type of response one can expect in systems with a tendency to form charged stripes.

The interpretation of the Raman spectra of  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$  in terms of fluctuating stripes [12] can be tested via the selection rules. For stripe orientations along the principal axes and the diagonals of the copper-oxygen planes the response is expected in the  $B_{1g}$  ( $x^2 - y^2$ ) and in the  $B_{2g}$  ( $xy$ ) symmetry, respectively, (Fig. 1). The  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO) system itself provides the oppor-

tunity for a direct check. As found by neutron scattering, the magnetic superstructure and, hence, the orientation of the stripes, rotates from diagonal to parallel when  $x$  exceeds a critical value of  $x_s \approx 0.055$  [17].

In this Letter, we show that charged stripes can be directly observed in a Raman experiment. The high intensity of the additional response at low doping allows us to determine the temperature dependence of fluctuating stripes. The scaling behavior raises implications on quantum criticality in the cuprates.

A standard light-scattering setup equipped with a scanning spectrometer and the sample mounted on the cold finger of a He-flow cryostat was used for the experiments.

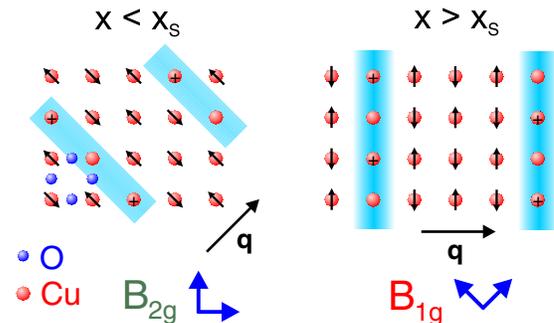


FIG. 1 (color online). Sketch of spin-charge-ordered states in the copper-oxygen plane (adopted from Ref. [5]). There are antiferromagnetic insulating areas and charged spin-free “stripes.” The modulation is characterized by the vector  $\mathbf{q}$ . If the pattern fluctuates the correlation length  $\xi_s$  is finite and can be as small as a few lattice constants. The response of 1D objects perpendicular to the modulation direction  $\mathbf{q}$  can only be observed by Raman scattering if the polarization vectors of both the incoming and the outgoing photons have a finite projection on either  $\mathbf{q}$  or the stripe direction for transverse or longitudinal excitations, respectively. This implies that stripes parallel to the principal axes can be observed only in  $B_{1g}$  and diagonal ones only in  $B_{2g}$  symmetry. In either orientation orthogonal stripes are equivalent and cannot be distinguished.

The scattering was excited by the Ar<sup>+</sup> laser line at 458 nm. The absorbed power ranged from 1 to 4 mW resulting in a local heating of 3 to 12 K as determined by comparing energy gain and loss spectra. In the figures, spot temperatures are indicated. The electronic Raman response  $\chi''_{\mu}(\omega, T) = S_{\mu}(\omega, T)/\{1 + n(\omega, T)\}$  is a two-particle correlation function related to the conductivity  $\sigma'(\omega, T)$ .  $\mu = B_{1g}, B_{2g}$ , etc.,  $S_{\mu}(\omega, T)$ , and  $\{1 + n(\omega, T)\}$  are the symmetry index, the structure factor (proportional to the measured Raman intensity), and the Bose factor, respectively. The symmetries  $\mu$  can be selected by the polarizations of the incident and the scattered photons and correspond to form factors projecting out different electron momenta. Hence, the response is a momentum-sensitive transport quantity [18,19]. In the case of free carriers, the slope of the spectra at  $\omega = 0$ ,  $\tau_0^{\mu}(T)$ , is proportional to a  $\mathbf{k}$ -resolved dc conductivity.

In Fig. 2, the  $B_{1g}$  and  $B_{2g}$  spectra of LSCO at  $x = 0.02$  and  $x = 0.10$  can be compared. At  $x = 0.10$  the  $B_{2g}$  spectra [Fig. 2(d)] and their temperature dependence are similar to those in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi-2212) and  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  (Y-123) [20]. They exhibit an increase of the slopes and hence a decrease of the dc scattering rates (“Raman resistivities”)  $\Gamma_0^{B_{2g}}(T) = \hbar/\tau_0^{B_{2g}}(T)$  upon cooling. The results are consistent with the optical and the dc resistivity to within 30% [20]. For  $B_{1g}$  symmetry [Fig. 2(c)] the spectra are relatively flat at room temperature. Upon cooling, a dramatic increase of the initial slope occurs, leading to a pileup of intensity at low energies quite similar to what is found in ladder compounds [16]. The  $B_{1g}$  results in LSCO are in clear contrast to those in Y-123 and Bi-2212 (Fig. 3) which both show an insulating type of

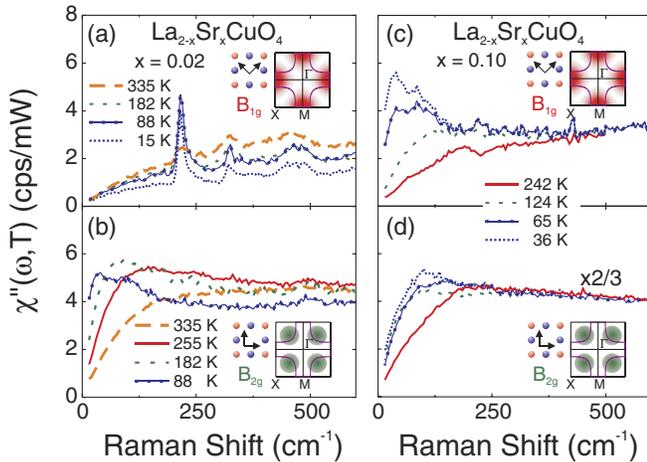


FIG. 2 (color online). Raman response  $\chi''_{\mu}(\omega, T)$  of  $\text{La}_{1.98}\text{Sr}_{0.02}\text{CuO}_4$  (a), (b) and  $\text{La}_{1.90}\text{Sr}_{0.10}\text{CuO}_4$  (c), (d). As indicated in the insets, areas around the  $M$  points and the center of the quadrant are projected out in  $B_{1g}$  and  $B_{2g}$  symmetry, respectively, on a quadratic 2D lattice. The selection rules for 1D structures are explained in Fig. 1.

behavior at  $\omega \rightarrow 0$  [20,21]. At low doping,  $x = 0.02$ , the  $B_{1g}$  behavior [Fig. 2(a)] expected from extrapolating the results of Y-123 and Bi-2212 is restored while the initial slope of the  $B_{2g}$  spectra [Fig. 2(b)] increases as strongly as that of the  $B_{1g}$  spectra at  $x = 0.10$  [Fig. 2(c)]. In either symmetry [Fig. 2(a) and 2(b)] the overall intensity decreases upon cooling, indicating a reduction of the number of free carriers as anticipated from the resistivity [22]. This is one of the reasons why there is no pileup of the  $B_{2g}$  intensity at  $x = 0.02$ . Additionally, there is inhomogeneous broadening of the peaks originating from both small variations of the Sr concentration and local distortions due to the low temperature structural transformation. At low temperature the stripes are at least partially pinned which is equivalent to static order [23].

According to the information from previous studies [12,20,21] we have to conclude that a new scattering channel opens up at low temperature in the  $B_{1g}$  and the  $B_{2g}$  symmetry at  $x = 0.10$  and  $x = 0.02$ , respectively. Since there is no sum rule as in the case of the optical conductivity, the additional spectral weight is directly superimposed on the response of the 2D  $\text{CuO}_2$  planes. At  $x = 0.02$ , the reduction of free carriers and the tendency to static order reduces the effect.

The new peaks can be separated out to a good approximation by subtracting the “background” of the 2D  $\text{CuO}_2$  planes. To this end, we use analytic approximations to the high-temperature spectra. In order to avoid any additional influence we assume that the 2D response is independent of temperature and is determined by the spectra at 335 K and 242 K for  $x = 0.02$  and  $x = 0.10$ , respectively. After subtraction, a Drude-like spectrum [24] comparable to the one in  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  [16] is obtained. The characteristic energy  $\Omega_c(x, T)$  corresponding to the peak position depends strongly on temperature [Figs. 4(a) and 4(b)]. The experimental peaks are narrower than expected for the Drude response. In spite of the different scattering symmetries at

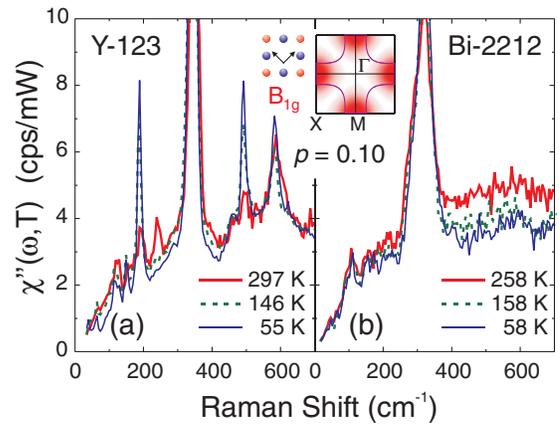


FIG. 3 (color online). Raman response  $\chi''_{B_{1g}}(\omega, T)$  of  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  (Y-123) and  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi-2212) at  $p = 0.10$ .

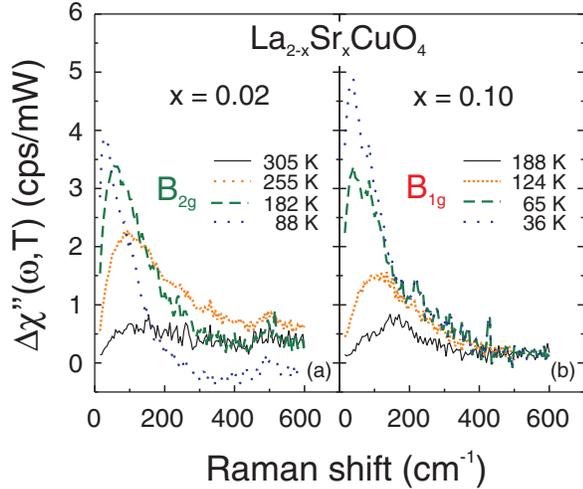


FIG. 4 (color online). Response of fluctuating charge order. A Drude-like peak [24] with a characteristic energy  $\Omega_c(x, T)$  is revealed after subtraction of the 2D response of the  $\text{CuO}_2$  planes. At  $x = 0.02$  (a) and  $0.10$  (b) the additional response is observed in  $B_{2g}$  and  $B_{1g}$  symmetry, respectively. The styles of the lines (colors) do *not* correspond to similar temperatures but rather highlight the *scaling* of the response with temperature: similar spectra are obtained if the temperatures differ by approximately a factor of 2.

$x = 0.02$  and  $0.10$  the spectral shapes are remarkably similar if a factor of approximately 2 in the temperature scales is taken into account [compare Figs. 4(a) and 4(b)]. This also indicates that the subtraction procedure has only little influence.

In Fig. 5,  $\Omega_c(x, T)$  is plotted for the two samples studied.  $\Omega_c(x, T)$  saturates below crossover temperatures  $T^*(x)$  of approximately 120 and 60 K for  $x = 0.02$  and  $0.10$ , respectively, at a doping-independent energy of 25–30  $\text{cm}^{-1}$ . The results from the two doping levels can be mapped on top of each other by scaling the temperature by a factor of approximately 2,  $\Omega_c(0.1, 2T) \approx \Omega_c(0.02, T)$  [see Fig. 5 inset (b)], implying and, hence, corroborating  $T^*(0.02) \approx 2T^*(0.10)$  as derived from the saturation of  $\Omega_c(x, T)$  (see Fig. 5). This is a strong indication that, independent of symmetry, the observed features originate from fluctuation phenomena where temperature is the only energy scale and doping is the control parameter [25–27]. For a quantum phase transition  $T^*(x)$  should approach 0 at the quantum critical point (QCP)  $x_c$ ,  $T^*(x \rightarrow x_c) = 0$ . For  $x < x_c$  the crossover temperature  $T^*(x)$  separates the fluctuation regime at  $T > T^*$  from an at least partially ordered state at  $T < T^*$ . In the case of long range static order such as in an antiferromagnetic Néel state the intensity of the new fluctuation mode is expected to vanish below  $T^*(x)$ .

In the presence of a QCP,  $\Omega_c(x, T)$  is a quantity proportional to the mass  $M(x, T)$  in the fluctuation propagator [27–29]. At sufficiently high temperatures, where thermal and quantum fluctuations dominate, the mass  $M(x, T)$  and,

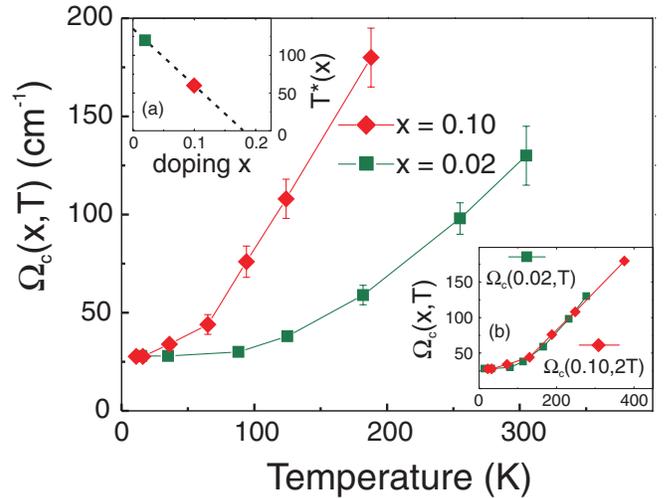


FIG. 5 (color online). Temperature dependence of the characteristic energy  $\Omega_c(x, T)$  of the stripe response (see Fig. 4) in LSCO. Inset (a) shows the determination of the quantum critical point  $x_c$  at which  $T^*(x)$  extrapolates to zero (see text). From changing the temperature scale [inset (b)] one obtains  $T^*(x = 0.02) \approx 2T^*(x = 0.10)$ . Inset (b) demonstrates that temperature is the *only* scale of  $\Omega_c(x, T)$ . For the two doping levels studied the scaling factor is approximately 2.

consequently,  $\Omega_c(x, T)$  are linear in  $T$ ,  $\Omega_c(x, T) \propto M(x, T) = \alpha(x)[T - T^*(x)]$ . Below  $T^*(x)$  in the (partially) ordered state,  $M(x, T < T^*)$  saturates [27]. Hence, the extrapolation to zero of the linear part of  $\Omega_c(x, T)$  or  $M(x, T)$  should provide estimates of  $T^*(x)$ . In fact, the data at  $x = 0.10$  have a linear part above approximately 60 K (Fig. 5). At  $x = 0.02$ , the linear part of  $T^*(x)$  is not as evident since the crossover is probably closer to the highest measuring temperature than at  $x = 0.10$ . Therefore, in order to obtain an estimate of  $x_c$  we use  $T^*(0.02) \approx 2T^*(0.10)$  (see above). With the relation  $T^*(x) \propto x$  as supported by both theory and experiment [9,27,30,31] one arrives at  $x_c = 0.18 \pm 0.02$  [see Fig. 5 inset (a)]. This analysis must be qualified by noting that the symmetry of the two “stripe” states is different, and thus the two data points in panel (a) Fig. 5 might be related in a more complicated way.

It is tempting to identify  $T^*(x)$  with the pseudogap line. Apparently, it is close to the lower boundary of values found experimentally [9,31]. If we assume that fluctuations dominate the physics, then  $T^*(x)$  is defined as the temperature at which the mass  $M$  extrapolates to zero. As a consequence,  $T^*(x)$  depends on the characteristic cutoff of the respective experimental probe and cannot be determined uniquely [27].

The results in LSCO provide direct experimental evidence of local symmetry breaking by the formation of quasi-1D structures (cf. Fig. 1) at low temperature and doping which can be interpreted in terms of a charge-ordering instability. The main support comes from the

new type of response [see Figs. 4(a) and 4(b)] superimposed on the usual spectra of the  $\text{CuO}_2$  planes and its dependence on symmetry demonstrating the reorientation of the charge modulation as a function of doping as already suggested by neutron scattering on the spin system [17]. At either doping the new response is unrelated to the dc resistivity indicating its dynamical and most probably transverse nature.

It is an important question as to why the low-energy response cannot be observed in Y-123 and Bi-2212 at  $p = 0.10$  (Fig. 3). We hypothesize that the correlation length  $\xi_s$  of the ordering phenomenon must exceed a minimal value to make the response visible. For example, in the case of phonons, translational symmetry must be established over several lattice constants to guarantee the selection rules to hold. Here, this seems to be a lower bound, and, more likely, the electronic mean free path  $\ell$  is the relevant scale. On the other hand,  $\xi_s$  corresponds to a fluctuation frequency of order  $\Omega_c \propto (\xi_s)^{-z}$  ( $z = 2$  for damped modes). If  $\xi_s$  is substantially smaller than in LSCO  $\Omega_c$  is expected to be larger. As a consequence, the intensity becomes weaker and is distributed over a bigger energy range. Then, the response of the stripes cannot be separated from the usual one of the planes.

We conclude that the charge-ordering instability may be an intrinsic feature of the copper-oxygen plane. The maximal doping level up to which the stripes can be observed directly depends on whether or not the lattice helps to stabilize the order. In LSCO  $\xi_s$  has obviously the proper magnitude to allow the observation of the stripes in an optical experiment facilitating a detailed study of the dynamical and critical behavior. If part of the La is substituted by Nd or Eu [5,32] or if Sr is replaced by Ba [23]  $\xi_s$  increases, and static order is established by a modification of the tilt of the  $\text{CuO}_6$  octahedra or, equivalently, by slightly changing the corrugation of the copper-oxygen plane. Above a critical tilt angle superconductivity is quenched [32]. Apparently, if  $\xi_s$  increases, the superconducting transition temperature  $T_c$  decreases suggesting a relation between charge ordering and superconductivity. Since the lattice determines the correlation length  $\xi_s$  it influences  $T_c$ , but, according to the results here, in a subtle and indirect way.

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