Freezing of a Stripe Liquid

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The existence of a stripe-liquid phase in a layered nickelate, $La_{1.725}Sr_{0.275}NiO_4$, is demonstrated through neutron scattering measurements. We show that incommensurate magnetic fluctuations evolve continuously through the charge-ordering temperature, although an abrupt decrease in the effective damping energy is observed on cooling through the transition. The energy and momentum dependence of the magnetic scattering are parametrized with a damped-harmonic-oscillator model describing overdamped spin waves in the antiferromagnetic domains defined instantaneously by charge stripes.

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One of the key issues in current debates over copper-oxide superconductors concerns the nature and relevance of charge stripes [1]. It has been demonstrated in one cuprate family that the holes doped into the CuO_2 planes can order in an array of periodically spaced stripes, separating antiferromagnetic domains [2]. For charge stripes to be relevant to superconductivity, they must be ubiquitous among the cuprates, and for the latter to be true, they must be able to exist in a liquid state. Indeed, theoretical models for such electronic liquid crystal phases have been proposed [3,4]. The inelastic incommensurate magnetic scattering observed in $La_{2-x}Sr_xCuO_4$ [5,6] and $YBa_2Cu_3O_{6+x}$ [7] has sometimes been interpreted as evidence for dynamic stripes; however, this interpretation has been controversial [8].

Here we present evidence for a stripe liquid phase in a related system, $La_{2-x}Sr_xNiO_4$. Ordered stripes have been observed in this system over a large range of hole concentrations [9]. Although the maximum charge and spin ordering temperatures occur for $x = 0.33$ [10], we have chosen to study crystals of $x = 0.275$, which have the advantage that the spin and charge-ordering wave vectors do not coincide. Using neutron scattering, we follow the magnetic inelastic scattering to temperatures as high as 1.7 times the charge-ordering transition, T_{co}, and demonstrate the existence of a stripe liquid. ($La_{2-x}Sr_xNiO_4$ is nonmetallic even at $T \gg T_{co}$, as indicated by a peak in its optical conductivity at ~0.5 eV [11,12], so there can be little controversy over possible alternative interpretations involving Fermi-surface nesting.) We show that the ω and $\mathbf{Q}$ dependence of the data can be effectively parametrized with a damped-harmonic-oscillator (DHO) model describing overdamped spin waves associated with the antiferromagnetic domains defined instantaneously by the charge stripes. Using a spin-wave energy dispersion with an effective gap energy, we find that the gap energy is proportional to the inverse correlation length and varies linearly with temperature through T_{co}. On the other hand, the behavior of the damping parameter changes abruptly near T_{co}.

The neutron scattering measurements were performed on the SPINS triple-axis spectrometer in the cold-neutron guide hall at the NIST Center for Neutron Research (NCNR). Initially, a single crystal of 4.4 g was used; later, a second crystal of 6.3 g was mounted alongside the first. Both crystals were grown at Kyoto University and annealed at the University of Delaware in order to achieve a stoichiometric oxygen concentration. (The first crystal was used previously for the work reported in [13], but without annealing.) The neutron spectrometer was equipped with a vertically focusing monochromator and horizontally focusing analyzer, both utilizing (002) reflections of pyrolytic graphite. Elastic scans of superlattice peaks were performed with 5-meV neutrons. Inelastic measurements were done in energy-gain mode, with incident energies of either 5 or 13.7 meV. The sample temperature was controlled with a displex refrigerator.

The stripe order in this sample is consistent with that observed in previous studies of $La_{2-x}Sr_xNiO_4$ [9]. Figure 1(a) shows the temperature dependence of the spin and charge-order superlattice peaks. The spin order approaches zero at $T_{so} \sim 120$ K, while the charge order disappears by $T_{co} \sim 190$ K.

Our focus here is on the dynamic correlations, especially those at $T > T_{co}$. Figure 2 shows examples of constant-energy-transfer scans ($\hbar\omega = -4$ meV) through the incommensurate magnetic peak positions $(1, \pm\epsilon, 0)$ at temperatures (a) $T > T_{co}$, (b) $T_{co} > T > T_{so}$, and (c) $T_{so} > T$. We clearly observe well-resolved peaks at all temperatures. Given that the ordered state at $T < T_{co}$ is already well characterized in terms of periodically spaced charge stripes separating antiferromagnetic domains,

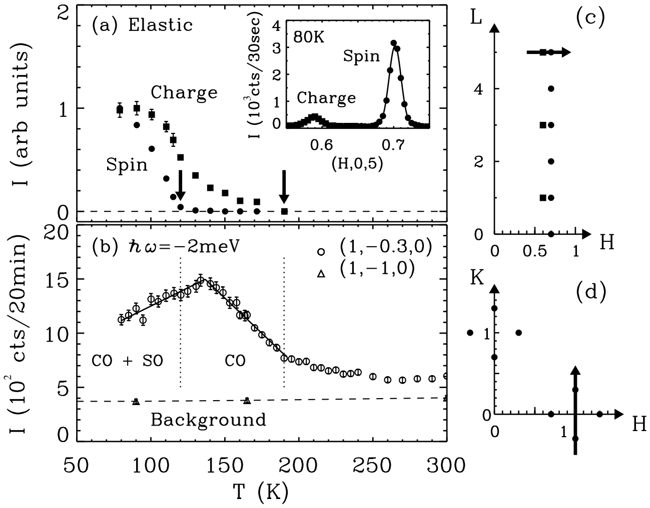

FIG. 1. (a) Temperature dependence of spin-order (SO) and charge-order (CO) superlattice intensities. Arrows indicate approximate transition temperatures. Inset: scan of intensity vs **Q** through superlattice peaks at $(2\epsilon, 0, 5)$ and $(1 - \epsilon, 0, 5)$ showing that $\epsilon \approx 0.29$ at 80 K. (b) Intensity at $(1, -\epsilon, 0)$ for an energy gain of 2 meV as a function of temperature. Points sampled at $(1, -1, 0)$ indicate the background. The lines through points are guides to the eye. (c) $(H0L)$ zone of reciprocal space, showing positions of magnetic (circles) and charge-order (squares) peaks. Arrow indicates direction of scan in inset of (a). (d) $(HK0)$ zone, showing magnetic peaks. Arrow indicates scan direction corresponding to Figs. 2(a)–2(c).

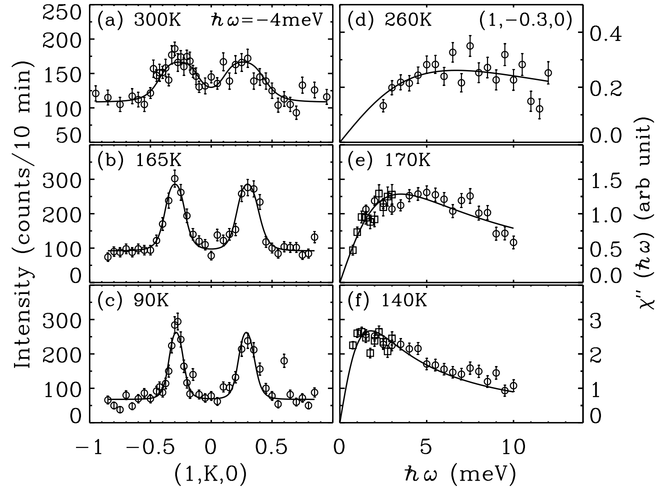

FIG. 2. Left side: Constant-energy scans for $\hbar\omega = -4$ meV for three different conditions: (a) 300 K, no static order; (b) 165 K, charge but no spin order; (c) 90 K, spin and charge order. Right side: constant-**Q** scans at $\mathbf{Q} = (1, -0.3, 0)$ for temperatures (d) 260 K, (e) 170 K, (f) 140 K. The curves through the data are fits as described in the text.

the continuous evolution of the inelastic magnetic scattering [see Fig. 1(b)] provides direct evidence for the existence of instantaneously correlated magnetic domains in the disordered state. The fact that the scattering at the commensurate antiferromagnetic wave vector $(1, 0, 0)$ is always a local minimum indicates that neighboring antiferromagnetic domains maintain their antiphase relationship, which is caused by the segregation of the doped holes to the domain walls. Thus, we feel that Fig. 2(a) is firm evidence for a stripe-liquid phase.

The variation of the Q widths of the peaks in the constant-E scans for $\hbar\omega = 0$, -4, and -8 meV is plotted in Fig. 3. For $T \lesssim T_{\rm so}$, the widths are roughly temperature independent but vary substantially with energy. The variation with energy is similar to what one might expect from spin-wave dispersion, as observed previously [14]. At higher temperatures, the widths appear to vary linearly with temperature, and the dependence on frequency is reduced.

The distribution of the magnetic-scattering strength with frequency also evolves with temperature. Figures 2(d)–2(f) show the variation of the imaginary part of the dynamic susceptibility, $\chi''(\mathbf{Q}, \omega)$, at several temperatures; here, $\mathbf{Q} = \mathbf{Q}_0$, where $\mathbf{Q}_0$ stands for an incommensurate magnetic wave vector. χ'' is related to the experimentally measured $S(\mathbf{Q}, \omega)$ via

$$S(\mathbf{Q}, \omega) = (1 - e^{-\hbar\omega/k_B T})^{-1} \chi''(\mathbf{Q}, \omega); \qquad (1)$$

the correction for the detailed-balance factor was applied after subtracting the background, measured at $\mathbf{Q} = (1, -0.9, 0)$. The position of the maximum of χ'' as a function of energy corresponds to a characteristic damping energy Γ, and one can see that Γ increases with temperature.

To describe the data more quantitatively, we have chosen to use the DHO model:

$$\chi''(\mathbf{Q}, \omega) = \sum_{\mathbf{Q}_0} \frac{2\omega\gamma\chi_0}{(\omega^2 - \omega_{\mathbf{Q}}^2)^2 + (2\omega\gamma)^2}, \qquad (2)$$

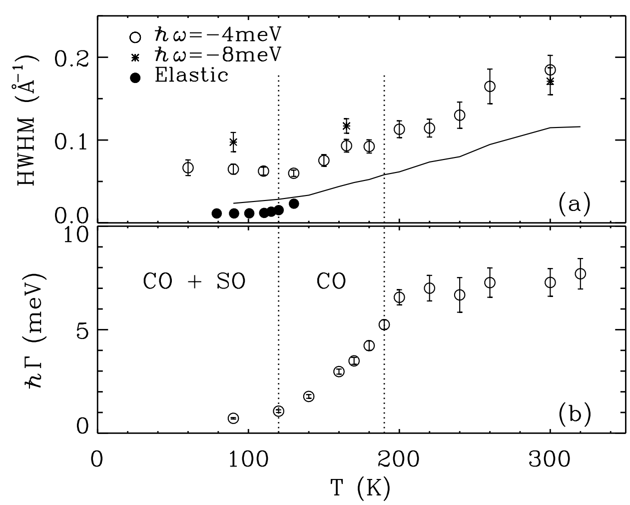

FIG. 3. (a) Half-width at half maximum (HWHM), without resolution correction, for constant-E scans through magnetic peaks: $\hbar\omega = -8$ meV (stars); -4 meV (open circles); elastic (filled circles). The solid line corresponds to fit parameter ω_0/c ($\approx\kappa$). (b) Results for effective spin-wave damping, Γ, from fits of DHO model (see text).

where the excitations are assumed to have the dispersive form

$$\omega_{\mathbf{Q}}^2 = \omega_0^2 + c^2(\mathbf{Q} - \mathbf{Q}_0)^2. \quad (3)$$

(Direct evidence for dispersive modes at high energies will be presented in a complementary study [16].) To fit the data, the model $S(\mathbf{Q}, \omega)$ was convolved with the spectrometer resolution function. The fitting parameters are ω_0, γ, and χ_0, with the effective spin-wave velocity held fixed at $\hbar c = 300$ meV Å, the approximate value determined for pure La_2NiO_4 [15]. To describe the $\mathbf{Q}$ dependence of the data, the relevant combinations are ω_0/c and γ/c; for the energy dependence, as we will discuss, it is only the ratio $\omega_0^2/2\gamma$ that matters.

The solid curves in Fig. 2 represent the fits to the data. The parameter values obtained from such fits are displayed in Fig. 4. For $T > T_{\rm so}$, ω_0 varies linearly with temperature. In contrast, γ is roughly constant up to $T \sim T_{\rm co}$ where it begins to increase rapidly. χ_0 has a fairly weak temperature dependence which is described approximately by $A/(1 + \gamma/B)$, where $B \approx 120$ meV. The temperature dependence of γ suggests that a major contribution to this damping factor comes from the fluctuations of the charge stripes [17]; it is consistent with the growth in the inverse correlation length for charge stripes observed by x-ray scattering in $La_{2-x}Sr_xNiO_4$ with $x = \frac{1}{3}$ [18]. The saturation of γ below $T_{\rm co}$ might be due in part to the quenched disorder associated with the randomly distributed Sr dopants.

Our scans versus frequency (Fig. 2) are measured at $\mathbf{Q} = \mathbf{Q}_0$, and most of the data correspond to $\omega^2 \ll \omega_0^2$, in which case Eq. (2) simplifies to

$$\chi''(\mathbf{Q}_0, \omega) \approx \frac{\chi_0}{2\gamma} \cdot \frac{\omega}{\omega^2 + \Gamma^2}, \quad (4)$$

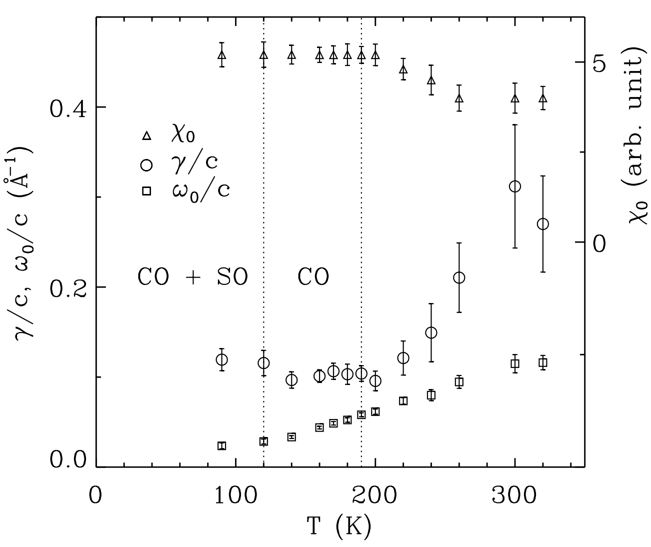

FIG. 4. Results for fitting parameters vs temperature: χ_0 (triangles, right-hand scale), γ/c (circles), and ω_0/c (squares).

where $\Gamma = \omega_0^2/2\gamma$. The values of Γ obtained from the values of the fitted parameters are plotted in Fig. 3(b). At $T > T_{\rm co}$, Γ changes slowly and has a value of roughly 7 meV. On cooling through $T_{\rm co}$, Γ abruptly starts to decrease, and becomes rather small by $T_{\rm so}$. Since ω_0 varies smoothly through this regime, the change in behavior at $\sim T_{\rm co}$ is controlled by γ. If γ is a measure of charge-stripe fluctuations, as discussed above, then it appears that the abrupt decrease in Γ below $T_{\rm co}$ may be a direct result of charge order. Low-frequency probes such as nuclear magnetic resonance [19,20] and muon-spin relaxation [21,22] should be sensitive to the variations in Γ, and it appears that such variations above $T_{\rm so}$ have been detected in studies of related nickelates [19,21,22]. The sensitivity of magnetic damping to charge order may also be relevant to the mechanism of the "wipe-out" effect observed in nuclear-quadrupole-resonance studies in cuprates [23–25].

In order to get further insight into the significance of the model parameters, we numerically integrated $S(\mathbf{Q}, \omega)$ over frequency. The result corresponds to $\mathbf{Q}$-dependent peaks with a line shape that is approximately Lorentzian. The half-width-at-half-maximum of this function should correspond to the instantaneous inverse correlation length, $\xi^{-1} \equiv \kappa$, and we find that, to within 1%, $\kappa = 0.86\omega_0/c$. This result, that the effective spin-excitation gap ω_0 is approximately equal to κc, is equivalent to the form proposed for the paramagnetic state of the undoped Heisenberg antiferromagnet [26,27].

It is of interest to consider the transport properties of $La_{2-x}Sr_xNiO_4$ in the regime $T > T_{\rm co}$ which we now associate with the stripe-liquid phase; numerous single-crystal studies of transport and optical properties have been reported previously (see, e.g., [11,12,28]). Figure 5

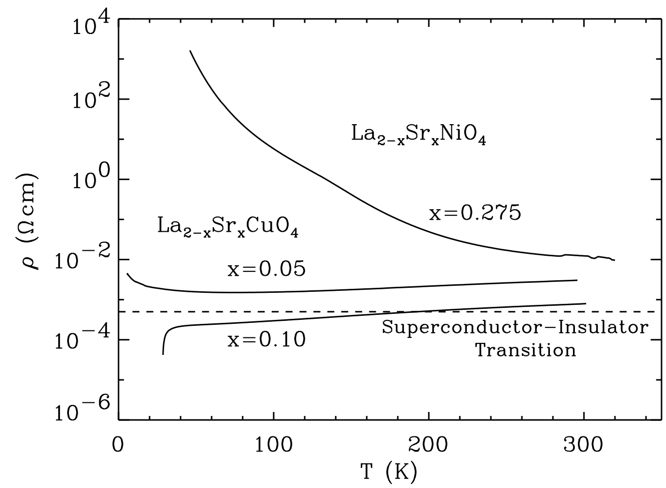

FIG. 5. Resistivity of the present sample compared with that of $La_{2-x}Sr_xCuO_4$ with $x = 0.05$ [29] and $x = 0.10$ [2]. The dashed line indicates the empirically determined critical resistivity for the superconductor-insulator transition in layered cuprates [31,32]. (If the in-plane resistivity does not drop below the critical range of $\rho \sim 0.4$–0.8 mΩ cm, then the sample will not go superconducting at any temperature.)

compares the temperature dependence of the in-plane resistivity measured on our crystal with similar measurements on two $La_{2-x}Sr_xCuO_4$ samples [2,29]. We observe that the resistivity of our nickelate crystal is only an order of magnitude greater than that of underdoped cuprates at 300 K, and we expect that it will develop a "metallic" temperature derivative at higher temperatures, as observed above 400 K in $La_{2-x}Sr_xNiO_4$ with $x = 0.33$ [11]. Furthermore, at high temperatures the resistivities of all three samples exceed a critical limit; to the extent that $d\rho/dT$ is positive, they qualify as "bad" metals [30]. We believe it is plausible that the "bad" metallic behavior of the cuprates might be associated with a stripe-liquid phase. (Resistivity in stripe-ordered cuprates can be low [2].)

The comparable magnitudes of room-temperature resistivity in the nickelates and cuprates make it plausible that a stripe-liquid phase could be relevant to transport properties in both materials. There are certainly differences in the two systems: the stripes inevitably order in the nickelates, whereas stripe order is generally avoided in the cuprates. However, these differences might be associated more with the magnitude of fluctuations rather than the nature of instantaneous correlations. In $La_{2-x}Sr_xCuO_4$ with $x = 0.14$, the measurements of Aeppli and co-workers [33] indicate that, using the present parametrization, $\hbar\Gamma \approx$ 10 meV at $T = 35$ K, with Γ increasing substantially at higher temperatures. In that case, the relevance of quantum critical phenonema has been proposed [34,35].

To conclude, we have presented experimental evidence for the existence of a liquid phase of charge stripes in a hole-doped nickelate. The magnetic correlations evolve smoothly through the freezing transition, although the damping of the fluctuations does not. The in-plane resistivity, though nonmetallic, is relatively low in the stripe-liquid phase.

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