

Critical Fluctuations and Quenched Disordered Two-Dimensional Charge Stripes in $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$

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Using high-resolution x-ray scattering, we have demonstrated the existence of quenched disordered charge stripes in a single crystal of $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ at low temperatures. Above the second-order transition critical scattering was observed due to fluctuations into the charge stripe phase. The charge stripes are shown to be two dimensional in nature both by measurements of their correlation lengths ($\xi_a \approx 185 \text{ \AA}$, $\xi_b = 400 \text{ \AA}$, and $\xi_c \approx 25 \text{ \AA}$) and by the critical exponents of the charge strip transition. The charge stripe ordering did not develop long-range order even at low temperatures, indicating that the charge stripes are disordered and that the length scale of the disorder is quenched.

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Strontium-doped La_2NiO_4 (LSN) is isostructural with the high- T_C cuprates, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSC) and $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ (LBC). With substitution of 15% Sr, LSC shows a superconducting transition at $T_{SC} \approx 35 \text{ K}$, but LSN remains insulating at doping levels up to 70% [1]. It has been suggested that stronger electron-phonon interactions in LSN are responsible for this behavior as it was recognized that electron-phonon interactions are critical for the persistent insulating character of the Mott insulator [2]. The hole-doped La_2NiO_4 is a prototypical system for the understanding of transport phenomena observed in compounds of LSC or LBC [3].

The substitutions of La with Sr provide hole carriers, and the doped holes tend to order in a stripelike structure acting as domain walls within the NiO_2 planes. The interplay between this ordered state and the spins of Ni dominates the transport behavior at low temperatures. Transport and optical conductivity measurements have demonstrated the anomalous phenomena resulting from the formation of the charge stripes [4,5]; in particular, a pronounced change in conductivity has been observed in $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$.

In $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ at doping levels of $x \approx 1/3$ and $1/2$ commensurate charge modulations have been observed by Chen *et al.* using electron diffraction and were explained to originate from ordering of polarons [6]. Using neutron scattering the charge ordering and the spin ordering have been evidenced to be the common features in nickelates [7]. Based on these experimental findings, Zaanen and Littlewood proposed a model of polaronic instabilities, suggesting that the polarons bind to, and thereby stabilize,

the domain walls formed due to either Sr doping or oxygenation, and that these domain walls order into a striped phase [8].

Despite the demonstrations of charge ordering and spin ordering in related colossal magnetoresistance manganites by neutron scattering, x-ray scattering has the merits of superior spatial resolution and direct probing of the charge density modulation. In this Letter, we report x-ray scattering studies that show that below T_{CO} the charge stripes are two-dimensional and disordered. This disorder is quenched at low temperature such that the length scale of the disorder does not increase at temperatures substantially below T_{CO} . Above the transition temperature we have observed charge stripe critical fluctuations. Measurements show that within these regions the charge stripes are highly disordered but remain two-dimensional up to $T_{CO} + 20 \text{ K}$.

The single crystalline sample of $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ was grown at Bell Laboratories using the floating zone method [9]. The single crystalline sample was cut into small pieces, and one of them was polished to get a shiny and even surface with an area of $\sim 2 \times 1 \text{ mm}^2$ using $1 \mu\text{m}$ diamond paste. The synchrotron experiment was performed on the beam line XMaS (BM28) at the European Synchrotron Radiation Facility (ESRF) (Grenoble, France). The incident x-ray was selected to be $1.2715(1) \text{ \AA}$ using a Si (111) monochromator, and most of the high-order contamination was rejected by the use of a focusing mirror. A Ge (111) single bounce analyzer was used to reduce the background and increase the wave vector

resolution. The sample was mounted on a Displex closed-circle cryostat with the [111] direction perpendicular to the sample surface. All the measurements were performed in the (hkl) zone of reciprocal space. The use of the multicircle diffractometer allows measurement of the scattering to be performed along the principle axes H , K , and L in reciprocal space. In keeping with the same crystallographic notation used in earlier experiments with Sr-doped La_2NiO_4 [7,9–11], the crystal structure was indexed in the tetragonal phase with lattice parameters of $a = b = 5.4145 \text{ \AA}$ and $c = 12.715 \text{ \AA}$. No realignment of the crystal was undertaken during measurements because $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ does not display any structural phase transition at low temperatures. The experimental resolution function was determined to be $\xi_H^{-1} \sim 0.0015 \text{ \AA}^{-1}$, $\xi_K^{-1} \sim 0.0015 \text{ \AA}^{-1}$, and $\xi_L^{-1} \sim 0.0044 \text{ \AA}^{-1}$ as measured on the Bragg peak $(4 \ 2 \ 4)$ which is near the charge ordering peaks measured at $T = 220 \text{ K}$, and the sample mosaic width was found to be $\sim 0.05^\circ$. Measurements of the Bragg peak and charge stripe satellites were taken as a function of temperature around the charge ordering transition temperature.

Satellite reflections were observed at positions of $(h \pm 2\epsilon \ 0 \ l)$, $\epsilon \approx 1/3$, $h = \text{even}$, and $l = \text{odd}$, at low temperatures in accord with the neutron scattering studies reported in [7]. We note that our results, however, are not consistent with the $(1/3, 1/3, 1)$ wave vector reported in [9]. In Sr-doped La_2NiO_4 (LSNO), the characteristic wave vector for the charge density modulation is $q_{\text{CO}} = (2\epsilon \ 0 \ 1)$, and $q_{\text{SO}} = (1 + \epsilon \ 0 \ 0)$ for the spin density modulation. Both electronic and magnetic structural phase distortions are responsible for the unusual transport behavior observed in LSNO compounds [4]. The incommensurability ϵ has been studied by many authors. The incommensurability has been observed to increase as the concentration of Sr is increased, and is temperature dependent, for instance, $\epsilon \approx 0.275$ for $x \approx 0.225$, $\epsilon \approx 0.25$ for $x \approx 0.2$, and $\epsilon \approx 0.12$ for $x \approx 0.135$ [7,9–11]. In $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ the transition temperature for charge ordering has been reported to be $T_{\text{CO}} \sim 240 \text{ K}$, while the spin ordering $T_{\text{SO}} \sim 180 \text{ K}$ [9]. This is in agreement with our observation for the charge ordering temperature. Figure 1 shows a plot of the integrated intensity, along each of the reciprocal space directions, of the charge ordering peak $(4.66 \ 0 \ 5)$ versus temperature. Clearly, the data can be fitted to a power law $I(t) \approx [(T_{\text{CO}} - T)/T_{\text{CO}}]^{2\beta}$, with $2\beta \approx 0.23 \pm 0.02$ and $T_{\text{CO}} \approx 239.2 \pm 0.2 \text{ K}$, and show the transition to be second order in nature. Table I lists the predicted exponents for 2D and 3D models [12]. The measured exponent of 2β clearly demonstrates that below T_{CO} the charge stripes in $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ are in the 2D universality class. The transition temperature of about 240 K is in accord with the susceptibility and resistivity measurements where the susceptibility decreases and resistivity increases, suggestive of a second-order phase transition due to the formation of charge stripes [4]. While electron diffraction images of

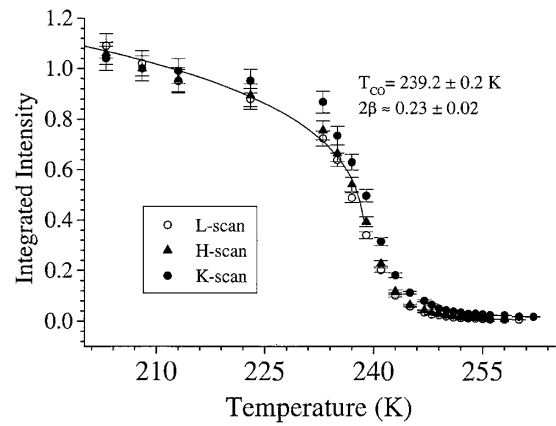


FIG. 1. Temperature dependence of the integrated intensities of the charge stripe satellite $(4.66 \ 0 \ 5)$ along all three principle directions in reciprocal space. The solid line is the best fit to a power law $I(t) \propto [(T_{\text{CO}} - T)/T_{\text{CO}}]^{2\beta}$.

charge stripes clearly show them to be arranged in two-dimensional planes, this is the first experimental evidence that the stripes behave themselves as two dimensional and that the charge stripes melt with a critical exponent expected of a 2D system.

The 2D character of charge stripes is also reflected in the anisotropic correlation lengths of the charge stripe satellites. Figure 2 displays linear scans through the charge ordering peak $(4.66 \ 0 \ 5)$ along the H ($= 2\pi/a$), K ($= 2\pi/b$), and L ($= 2\pi/c$) directions at temperatures well below and above $T_{\text{CO}} \approx 239.2 \text{ K}$, respectively. The dotted lines show the best fits convoluted with the resolution function. It is clear that the profile of the charge stripe satellite along the H and K directions are nearly identical, but it is much wider in the L direction. Even in the high temperature range where the charge stripe shows critical fluctuations, the charge stripe also displays two-dimensional behavior as shown in Figs. 2(c) and 2(d). This behavior can also be observed in the charge stripe satellite $(5.33 \ 0 \ 7)$. The correlation lengths in all three principal directions obtained by fitting are listed in Table II.

La_2NiO_4 is a 2D antiferromagnetic insulator, and transport and optical conductivity spectra have demonstrated an anisotropy ($> 10^2$) between the in-plane (a - b) and c -axis components [5], and this may limit the formation of 3D charge ordering. Scans along the L direction at low temperatures exhibit extending wings on both sides of the

TABLE I. Predicted exponents and the measured percolation exponents ν_T for 2D and 3D models [12].

	2β	ν	ν_T
2D Heisenberg	0.3	≈ 0.95	0.9
2D Ising	0.25	1	1.32
3D Ising	0.624	0.64	0.85
3D Heisenberg	0.76	0.702	0.96
3D XY	0.692	0.669	

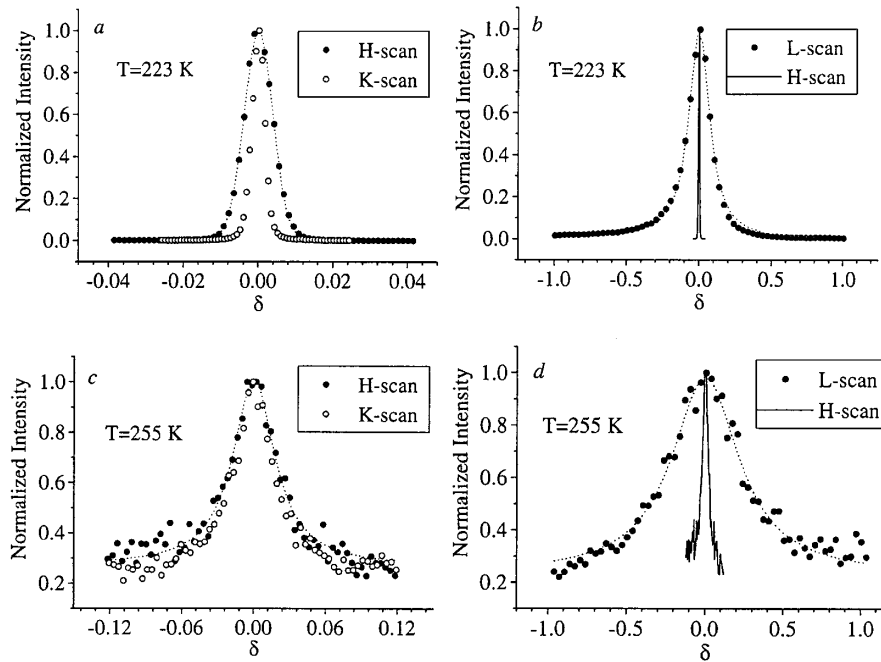


FIG. 2. Linear scans through the charge ordering peak (4.66 0 5) at $T = 223$ and 255 K in the H , K , and L directions. It is clear that the charge stripe is two-dimensional in nature even in the critical fluctuations. The dotted line in (a) is a Gaussian function convoluted with the experimental resolution function, and the dotted lines in (b), (c), and (d) are Lorentzian functions convoluted with the resolution function.

charge stripe satellite as shown in Fig. 2(b), resulting in a peak profile which was fitted well by a Lorentzian line shape convoluted with the experimental resolution function. Diffuse streaks along the L direction have been observed to be a common feature in Sr-doped La_2NiO_4 [13]. This short-range ordering along the c axis, with a length scale of approximately 2 unit cells along the c axis, may be helpful to minimize the long-range part of the Coulomb interaction [7].

In Fig. 3, we display the evolution of the inverse correlation lengths with temperature. Above the transition temperature $T_{\text{CO}} \approx 240$ K, only very weak scattering is observed at the expected positions in reciprocal space. Such scattering is caused by critical scattering due to dynamic spatial fluctuations into the charge stripe phase. We

TABLE II. Correlation lengths of charge ordering peaks (5.33 0 7) and (4.66 0 5) at temperatures below and above the transition temperature $T_{\text{CO}} \approx 240$ K.

	ξ_H (Å)	ξ_K (Å)	ξ_L (Å)	
233 K	157	331	24	(5.33 0 7)
241 K	130	208	22	(5.33 0 7)
245 K	88	116	16	(5.33 0 7)
250 K	64	75	14	(5.33 0 7)
255 K	48	47	8	(5.33 0 7)
233 K	181	383	24	(4.66 0 5)
241 K	138	230	23	(4.66 0 5)
245 K	91	121	15	(4.66 0 5)
250 K	58	73	11	(4.66 0 5)
255 K	37.5	39	8	(4.66 0 5)

believe this to be the first observation of charge stripe critical fluctuations existing above T_{CO} . The size of these clusters varies with temperature displaying a marked divergence close to T_{CO} , and a power-law-like behavior over a temperature range from T_{CO} to $T_{\text{CO}} + 20$ K. Fitting the data of Fig. 3 to a power law for the inverse correlation length, $\xi(t)^{-1} \approx [(T - T_{\text{CO}})/T_{\text{CO}}]^\nu$, allows the extraction of ν for the exponent of the correlation length of charge ordering. Such fitting gave a value of

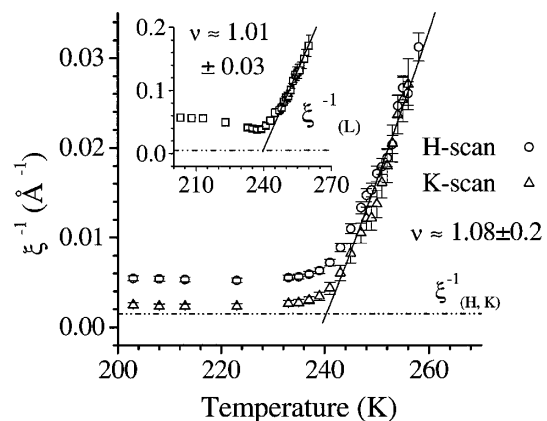


FIG. 3. The evolution of the inverse correlation lengths of the charge stripe satellite (4.66 0 5) along the H and K directions. The data can be fitted to a power law $\xi(t)^{-1} \propto [(T - T_{\text{CO}})/T_{\text{CO}}]^\nu$. The value of T_{CO} was fixed to be 239.2 K for all three principle directions. The inset shows the evolution and the best fit along the L direction drawn with the same axes as the main diagram.

$\nu = 1.08 \pm 0.2$, in excellent agreement with the predicted value of $\nu = 1$ expected for a two-dimensional system [12]. We observed the critical scattering at temperatures up to $T_{CO} + 20$ K, an abnormally high range which gives an accurate determination of the exponent ν . As compared to the experimental resolution, it is clear that the charge stripes remain only short-range ordered, indicating that the disordered charge ordering is quenched below the transition temperature T_{CO} . By studying the frustrated electronic phases in high- T_c superconductors, Emery *et al.* suggested that a local charge inhomogeneity should exist in the cluster spin glass, and that the spins in the hole deficient regions are locally Néel ordered [14]. Therefore, the spins can be considered to be the pinning centers for the localized charge density modulations below the transition temperature. A glasslike structure in $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ was proposed by Lee and Cheong in their neutron scattering study [9] but this is not supported by our study, which observed long-range correlations along the H and K directions below the charge stripe transition. They also pointed out that the charge stripes could be pinned by the defects induced by Sr dopants. Comparisons made to the changes in the correlation lengths of charge ordering below and above T_{CO} indicate an anisotropic increase between the $a \times b$ plane and the c axis, i.e., $\xi_{223}^H/\xi_{255}^H \approx 5$, $\xi_{223}^K/\xi_{255}^K \approx 10$, and $\xi_{223}^L/\xi_{255}^L \approx 3$.

In conclusion, we have shown that two-dimensional disordered charge stripes are formed below T_{CO} in a single crystal of $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$. The correlation length of these charge stripes does not increase as the temperature is lowered significantly below T_{CO} , indicating that the length scale of the disorder is quenched at low temperatures. We have obtained quantitative measurements of the correlation lengths and critical exponents that demonstrate this reduced dimensionality. In addition, we have, for the first time, observed critical fluctuations above T_{CO} and up to $T_{CO} + 20$ K. Measurements of the correlation lengths and critical exponent also demonstrate that within these fluctuations the stripes are still two dimensional. The doping with Sr provides extra holes to form the striped structure in the NiO planes. It is conceivable that the stripes are affected, or even disordered by the strain energy resulting from the mismatch of the radius of cations of La ($r_A = 1.216 \text{ \AA}$) and Sr ($r_A = 1.31 \text{ \AA}$). With doping different sizes of cations on the La site, Rodriguez-Martinez and Attfield [15] have demonstrated that large disorder effects due to size differences suppress the ferromagnetic metal–paramagnetic insulator transition temperature in $(R_{1-x}M_x)\text{MnO}_3$ perovskite manganites due to strain fields. It is likely that a similar effect is occurring in the isostructural nickelates and that such strain fields disrupt the formation of long-range charge stripe order. The strain energy distributes inhomogeneously in the La-Sr planes so that the disordered stripes are anisotropic. Below T_{CO} , such disordered charge stripes are then quenched by

the pinning potential provided by the Ni ions, the defects induced by the Sr dopants, or the combination of both. Our results are in agreement with recent theoretical predictions, which indicate a disordered pinned stripe phase [16].

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