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Charge Excitations in the Stripe-Ordered $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ and $\text{La}_{2-x}(\text{Ba}, \text{Sr})_x\text{CuO}_4$ Superconducting Compounds

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Charge excitations in stripe-ordered 214 compounds $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ and 1/8-doped $\text{La}_{2-x}(\text{Ba or Sr})_x\text{CuO}_4$ are studied using resonant inelastic x-ray scattering in the hard x-ray regime. We observe ≈ 1 eV excitation with a momentum transfer corresponding to the charge stripe spatial period both for the diagonal (nickelate) and parallel (cuprates) stripes. They are interpreted as collective stripe excitations or anomalous softening of the charge excitonic modes of the in-gap states.

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There is accumulating theoretical and experimental evidence that the spatially alternating charge state, such as the charge stripe state, occurring as a result of the carrier doping into the strongly correlated electron system is closely related to the high temperature superconductivity in cuprates [1,2]. Understanding the dynamics of the charge stripe state is of fundamental importance. Whereas the spin dynamics has been studied extensively by neutron scattering [3–6], the charge dynamics is yet to be understood.

The 214 type nickelates and cuprates, in the family of the originally discovered high temperature superconductor $\text{La}_{2-x}(\text{Ba}, \text{Sr})_x\text{CuO}_4$, exhibit charge stripe order when holes are doped. The stripe order has been studied in detail by diffraction techniques [7–10] and interpreted as anti-phase antiferromagnetic domains separated by one dimensional charge stripes with localized doped holes. Upon carrier doping, $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ (LSNO) exhibits an ordered state with “diagonal” stripes propagating along the diagonal of the NiO_2 square lattice [7] [Fig. 1(a)]. Hole-doped 214 cuprates $\text{La}_{2-x}(\text{Ba or Sr})_x\text{CuO}_4$ (LBCO or LSCO) show diagonal stripe order in the low doping insulating region [11]. In the superconducting dome the systems, such as LBCO, show static order of “parallel” stripes in the vicinity of $x = 0.125$ [12,13] where the superconductivity is suppressed [14]. This is often referred to as the 1/8 anomaly. In this case the charge stripes are parallel to the Cu-O-Cu bond [Fig. 1(b)]. Understanding dynamical fluctuation of parallel stripes and their relationship to the superconductivity is of fundamental importance, and it is the main topic of the studies presented in this Letter.

Here, we report the first observation of charge excitations arising from the charge stripe-ordered state in 214 type nickelate, $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$, and 214 type supercon-

ducting cuprates $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ and $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$ using the resonant inelastic x-ray scattering (RIXS) in the hard x-ray regime. Our measurements reveal the charge excitations with the momentum transfer corresponding to the charge stripe spatial period. We observe this nontrivial feature both for the diagonal (nickelate) and parallel (cuprates) stripes.

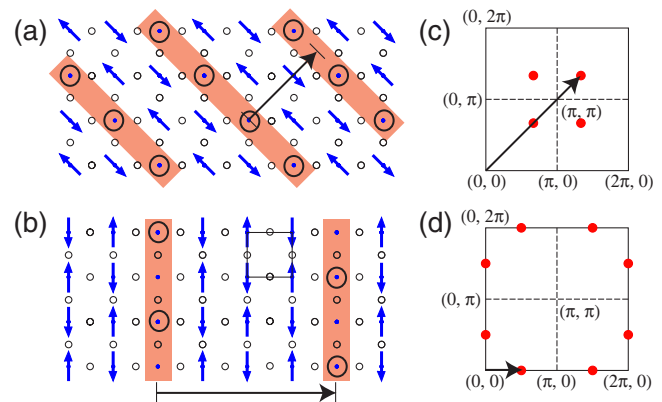


FIG. 1 (color online). Diagonal charge stripes in (a) LSNO $x = 1/3$ and parallel stripes in (b) LBCO $x = 0.125$ and LSCO $x = 0.12$ in the basal (Ni or Cu) O_2 plane. Stripes are marked by shaded belts. (c) and (d) depict momentum space for the (Ni or Cu) O_2 square lattice in unit of $1/a$ (a is the lattice constant of the square lattice). Arrows connecting the stripes in (a) and (b) indicate spatial periods of stripes. Arrows in (c) and (d) indicate characteristic momentum transfer \mathbf{q}_s . The period of $(3a/2, 3a/2)$ in (a) gives characteristic momentum transfer $\mathbf{q}_s = (2\pi/3, 2\pi/3)$, and the period of $(4a, 0)$ in (b) gives $\mathbf{q}_s = (\pi/2, 0)$. Circles in (c) and (d) indicate equivalent \mathbf{q}_s points due to the stripe domains whose stripes run perpendicular with each other.

The RIXS technique is one of the most powerful probes to measure charge-charge correlation function multiplied by a resonant enhancement factor [15,16]. RIXS in the hard and soft x-ray regime recently made remarkable progress [17] and resulted in observation of various types of charge excitations in the strongly correlated electron systems, such as the charge excitation across the charge transfer (CT) gap [18–21], intraband excitations [21], molecular orbital excitations [22], $d-d$ excitations [20,23], Zhang-Rice excitations [24], and even magnons [25,26]. The major advantage of RIXS is the possibility of studying charge excitations as a function of the momentum transfer \mathbf{q} . The stripe spacing in the real space corresponds to a characteristic momentum transfer \mathbf{q}_s as shown in Figs. 1(c) and 1(d). RIXS is an ideal probe for detecting charge excitations associated with the stripe having a specific momentum transfer \mathbf{q}_s .

RIXS in the hard x-ray regime uses incident photons with the energy at the absorption K edge of the transition metal element. Incident photons excite $1s$ core electrons into either $4p_\pi$ or $4p_\sigma$ orbitals depending on the sample geometry, and this intermediate state triggers various charge excitations. In the present studies, the incident photon energy was tuned to the energy of the $1s \rightarrow 4p_\pi$ transition, which is 8347 eV for nickelate and 8993 eV for cuprates. These energies are a few electron volts lower than the energy of the $1s \rightarrow 4p_\sigma$ transition.

The RIXS measurements of nickelate were performed at 10 K using the MERIX spectrometer installed at the Advanced Photon Source, beam line XOR-IXS 30-ID. RIXS measurements of cuprates were done at 8 K using the inelastic x-ray spectrometer at BL11XU at SPring-8. Horizontal scattering geometry was utilized for all measurements, with the scattering plane parallel to the (a, c) crystal plane.

The instrumental energy resolution of the MERIX spectrometer at Ni K edge is 150 meV. This is achieved by using a Ge(642) spherical diced analyzer, and a position sensitive microstrip detector placed on the Rowland circle with a 1 m radius. The silicon microstrip detector with 125 μm pitch is applied with the purpose of reducing the geometrical broadening of the spectral resolution function [27]. The energy resolution of the Cu K edge RIXS spectrometer at BL11XU is 400 meV. The Bent Ge(337) analyzer with 2 m curvature radius and a point silicon detector is used. The q resolution of MERIX in the present configuration is 0.26 \AA^{-1} along the $[\pi, 0]$ and 0.38 \AA^{-1} along the $[0, \pi]$ directions, respectively. The q resolution for BL11XU is 0.10 \AA^{-1} and 0.15 \AA^{-1} , respectively.

Single crystals of LSNO $x = 1/3$, LBCO $x = 0.125$, LBCO $x = 0.08$, and LSCO $x = 0.12$ were prepared by the traveling solvent floating zone method. The samples of LSNO $x = 1/3$ and LBCO $x = 0.125$ show robust charge stripe order below 180 K and 55 K, respectively. LBCO $x = 0.08$ shows no clear stripe order according to the neutron diffraction studies. LSCO $x = 0.12$ is expected

to have a stripe order below 30 K from neutron diffraction, but the charge order has not been confirmed directly. Lattice constants for LSNO and L (B or S) CO at low temperatures give the reciprocal lattice unit of 0.166 \AA^{-1} for the basal (Ni or Cu) O_2 square lattices.

Since the 214 compounds are two dimensional, we assign the $(0, 0, L)$ position as the Γ point $(0, 0)$ of the basal plane and $(1, 0, L)$ as the next Γ point $(2\pi, 0)$. Here L is chosen so that the scattering angle 2θ is $\sim 90^\circ$. In this configuration the scattered photon propagates parallel to the polarization of the initial photon, and thus, minimizes the elastic intensity. This is crucial for observing the low energy excitations below 1.5 eV reported in this Letter.

Figure 2 shows the representative RIXS spectra of LSNO $x = 1/3$ taken at the \mathbf{q} positions indicated in the right top panel. Every spectrum except the one measured at $\mathbf{q}_s = (4\pi/3, 2\pi/3)$ (labeled as F), contains three peaks: an elastic peak at zero energy transfer, and peaks at ~ 1.5 eV and ~ 4.5 eV. The 4.5 eV feature is known to be the charge excitation across the CT gap (called a CT peak), which is also observed in the nondoped La_2NiO_4 [20]. The 1.5 eV feature is attributed to the charge excitation from the valence band to the in-gap band which is known to appear when holes are doped into nickelates, based on optical measurements [28–30]. There is a dip between the elastic peak and the in-gap peak implying a gap in charge excitation, consistent with the fact that the nickelate is an insulator. However, to our surprise, the dip appears to be filled

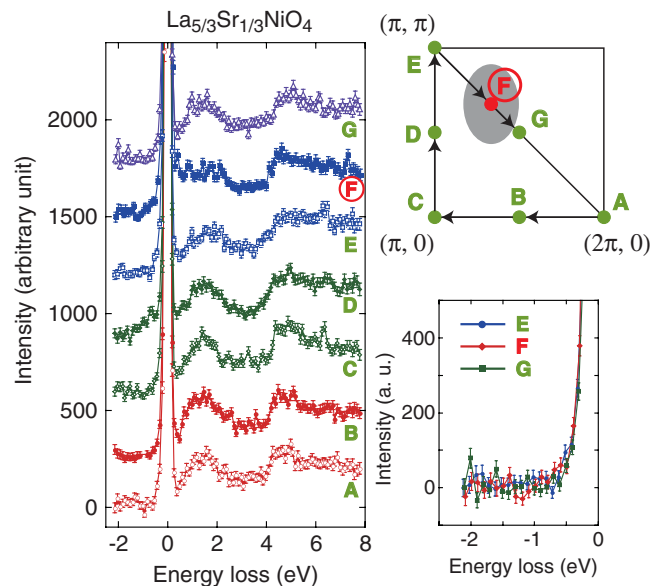


FIG. 2 (color online). RIXS spectra of LSNO $x = 1/3$ measured at different momentum transfer \mathbf{q} . The spectra are shown shifted by 300 each in intensity, for clarity. Measured \mathbf{q} positions are indicated in the right top inset by circles labeled A through G and connected by arrows, showing the order of spectra from bottom to top. Point F corresponds to \mathbf{q}_s . The shaded ellipsoid around point F shows \mathbf{q} resolution. The right bottom inset shows elastic line tails on the energy gain side of the RIXS spectra for positions E , F , and G .

at $\mathbf{q} = \mathbf{q}_s$. We note that the elastic peak at $\mathbf{q} = \mathbf{q}_s$ is twice as strong as those at the other \mathbf{q} positions due to the charge stripe order [31]. One might suspect that the additional intensity comes from the tail of the stronger elastic peak. However, as clearly seen in the right bottom panel of Fig. 2, the intensity of the elastic peak tail on the energy gain side at $\mathbf{q} = \mathbf{q}_s$ is the same as for other \mathbf{q} positions. This rules out such an explanation.

We have measured RIXS spectra at several \mathbf{q} positions between $(2\pi, 0)$ and $(0, 2\pi)$. The RIXS intensities are mapped in Fig. 3 as a contour plot. It clearly demonstrates that the dip between the elastic and the in-gap peaks is indeed filled at both \mathbf{q}_s positions: $(4\pi/3, 2\pi/3)$ and $(2\pi/3, 4\pi/3)$. Although the limited beam time did not allow us to check other \mathbf{q}_s positions, it is very likely that the filled gap is a general feature for all \mathbf{q}_s .

As a next step we will compare the LSNO results with those of cuprates. Figure 4(a) shows an intensity map of RIXS spectra of LBCO $x = 0.125$. A cross section at $(0, 0)$, along the line labeled as “A”, is shown in Fig. 4(b) as a representative RIXS spectrum. All spectra contain two peaks: the elastic peak and the CT peak at ~ 4 eV. In contrast with nickelates, cuprates show no clear in-gap peaks below the CT gap energy. This is consistent with the optical measurements [32]. The parallel stripe order with a $4a$ spacing, which is realized in LBCO $x = 0.125$ and LSCO $x = 0.12$, corresponds to momentum transfer $\mathbf{q}_s = (\pi/2, 0)$ [Fig. 1(c)]. Figure 4(c) shows the comparison of spectra measured at momentum transfer $(0, 0)$, cross section A, and at $(\pi/2, 0)$, cross section B, for LBCO with two different dopings $x = 0.125$ and 0.08 . Remarkably, we find an anomalous enhancement of the RIXS intensity near 1 eV at $\mathbf{q}_s = (\pi/2, 0)$ in the stripe-ordered sample LBCO

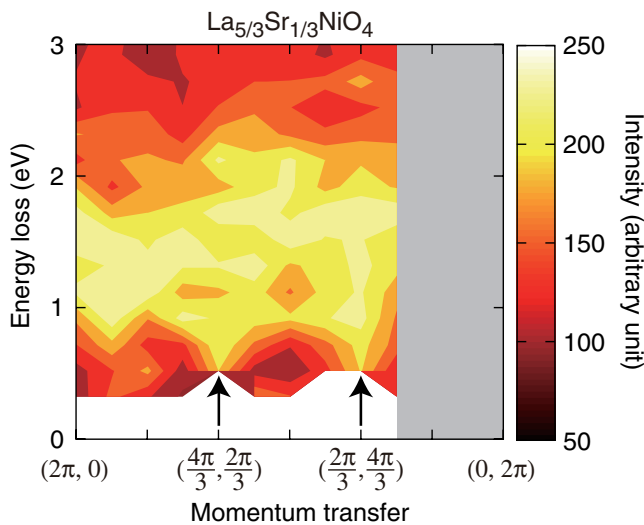


FIG. 3 (color online). A contour plot of RIXS intensity in the momentum transfer range between $(2\pi, 0)$ and $(0, 2\pi)$. The RIXS intensity is normalized to the in-gap peak intensity at ~ 1.5 eV. The white area has intensity of more than 250 due to the elastic peak or filled dip at \mathbf{q}_s positions. The shaded rectangular area has not been measured.

$x = 0.125$. This is more clearly demonstrated in Fig. 4(d), where the RIXS intensity averaged in the energy range between 1 and 1.25 eV peaks at \mathbf{q}_s in LBCO $x = 0.125$ and LSCO $x = 0.12$. By contrast, it is flat in the non-stripe-ordered LBCO $x = 0.08$. Also a common feature is observed in all measured spectra in cuprates: the CT peak energy is smallest and the elastic peak intensity is largest [31] at the $(0, 0)$ position. These facts evidence that the enhancement of the RIXS intensity at energy loss ~ 1 eV and momentum transfer \mathbf{q}_s is due to an intrinsic property of the system. It cannot be explained by the overlap of the tails of the elastic and CT peaks.

The data on nickelate and cuprates presented above demonstrate that the charge stripe state results in additional RIXS signals at energy transfer 1 eV and momentum transfer \mathbf{q}_s regardless of the stripe geometry. The charge stripes may fluctuate, i.e., experience collective charge stripe mo-

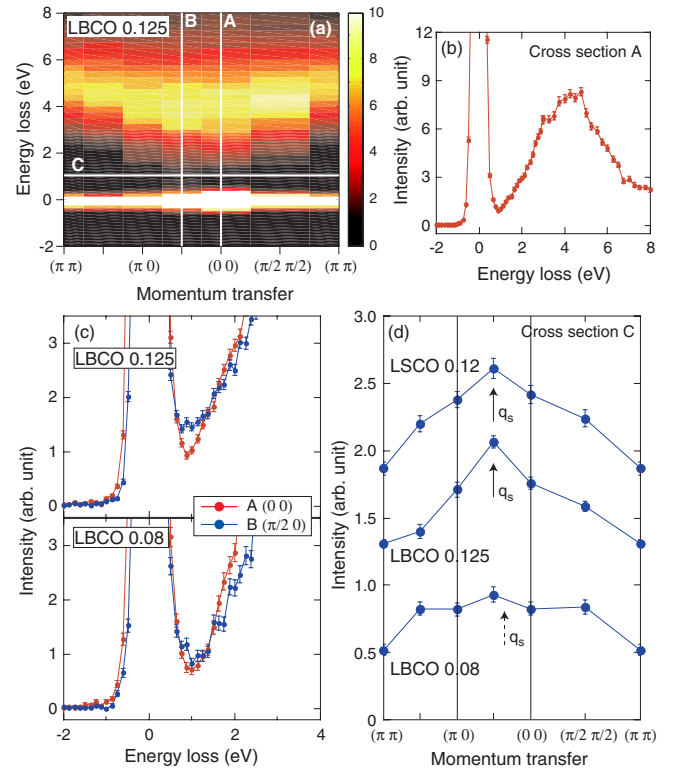


FIG. 4 (color online). (a) RIXS intensity map of LBCO $x = 0.125$ along $(\pi, \pi) \rightarrow (\pi, 0) \rightarrow (0, 0) \rightarrow (\pi, \pi)$. (b) RIXS spectrum—the cross section A in (a). (c) Close-ups of RIXS spectra around 1 eV of LBCO $x = 0.125$ (top) and LBCO $x = 0.08$ (bottom)—the cross sections A and B in (a). The cross section B corresponds to $\mathbf{q}_s = (\pi/2, 0)$ of LBCO $x = 0.125$. The RIXS intensities in (c) are normalized to the maximum of the CT peak at ~ 4 eV. (d) Momentum transfer dependence of the RIXS signal averaged in the energy region between 1 and 1.25 eV. These profiles correspond to the cross section C in (a). Each spectrum is shifted for clarity. The arrows indicate \mathbf{q}_s positions. Although LBCO $x = 0.08$ does not have a robust stripe order, we show $\mathbf{q}_s = 2\pi(2x, 0) = (0.32\pi, 0)$ by a dashed arrow estimated from a very small neutron diffraction signal reported in Ref. [35].

tion in the form of meandering and compression modes. Such collective stripe excitations have been numerically studied in Ref. [33]. Dynamical charge susceptibility with the momentum transfer \mathbf{q}_s has been predicted for parallel stripes. Qualitatively similar excitations are expected for the diagonal stripes. It is therefore natural to ascribe the additional signal observed at $\mathbf{q} = \mathbf{q}_s$ in the RIXS spectra of Figs. 2 and 4 to the collective stripe excitations. An immediate question arises of whether the observed excitation has something to do with the mechanism of the high- T_c superconductivity. The present studies show that the charge stripe excitations appear even in insulating LSNO. Therefore, the charge stripe excitations, we are observing, may not be directly related to the superconductivity. However, they might be not be sufficient but necessary for the superconductivity. Detailed study with better energy and momentum resolution is necessary to understand the role of collective motion of stripes in the superconductivity.

Another plausible explanation is that the observed anomaly at \mathbf{q}_s is due to the in-gap state of the striped system, in particular, because the excitonic mode of the in-gap state may have anomalous softening at \mathbf{q}_s . Detailed theoretical studies of the in-gap band are desirable to examine this scenario. The origin of the in-gap state has been discussed earlier by many authors [29,30,34]. It is important, however, to verify whether the theory can predict softening of the excitonic mode in the presence of charge stripe structures.

It would not be an easy task to distinguish experimentally between the above two possibilities. First, improving the energy and momentum resolution of the RIXS spectrometers would be necessary to study the structure and dispersion of the charge stripe excitations in detail and thus to better understand its nature. Second, one could apply nonresonant IXS for direct observation of the collective stripe excitations. However, the intensity of the IXS signal related to collective charge excitation might be too weak. Third, RIXS in the soft x-ray regime at the Cu L edge or O K edge could be another possibility to study the detailed structure of the excitations and understand its origin.

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