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Strongly Correlated Charge Density Wave in  
 $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

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# Nature of the charge-density wave excitations in cuprates

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The discovery of charge-density wave (CDW)-related effects in the resonant inelastic x-ray scattering (RIXS) spectra of cuprates holds the tantalizing promise of clarifying the interactions that stabilize the electronic order. Here, we report a comprehensive RIXS study of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO) finding that CDW effects persist up to a remarkably high doping level of  $x = 0.21$  before disappearing at  $x = 0.25$ . The inelastic excitation spectra remain essentially unchanged with doping despite crossing a topological transition in the Fermi surface. This indicates that the spectra contain little or no direct coupling to electronic excitations near the Fermi surface, rather they are dominated by the resonant cross-section for phonons and CDW-induced phonon-softening. We interpret our results in terms of a CDW that is generated by strong correlations and a phonon response that is driven by the CDW-induced modification of the lattice.

Thirty years after the discovery of high-temperature superconductivity in the cuprates, there is still no consensus regarding the minimal set of interactions needed to describe the “normal state” from which superconductivity emerges [1]. Popular Hubbard and ‘ $t - J$ ’ theoretical models suggest that superconducting, charge-density wave (CDW), and spin-density wave (SDW) states have similar ground state energies [2–8], which is consistent with experiments that reveal widespread interplay between all three states [9–15]. This complexity motivates experimental efforts to measure collective excitations associated with CDW order that should clarify the pertinent interactions. Resonant inelastic x-ray scattering (RIXS), as illustrated in Fig. 1(a), has an enhanced sensitivity to valence charge and phonon excitations [16]. Several experiments indeed reported anomalies in cuprate RIXS spectra at the CDW wavevector ( $\mathbf{Q}_{\text{CDW}}$ ), opening new routes to understand cuprate CDWs [17–22]. Uniquely isolating and interpreting CDW-effects in RIXS is, however, complicated as CDWs inevitably modify their host crystal lattice and thus the phonons. Compounding this problem, RIXS spectra near  $\mathbf{Q}_{\text{CDW}}$  have been conceptualized in several different ways including charge excitations [20, 21, 23], momentum-dependent electron-phonon coupling (EPC) [22] and Fano-effects [19].

In this Letter, we use ultrahigh energy-resolution RIXS to examine the nature of the CDW in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO). We observe that CDW correlations and associated CDW-induced phonon-softening persist up to a

strikingly high doping level of  $x = 0.21$  before both effects disappear at  $x = 0.25$ . The large doping range  $x = 0.12 \rightarrow 0.25$  traverses a topological transition in the Fermi surface [24–26], allowing us to empirically test the relative importance of charge and lattice effects in the excitation spectra. We find that the data can be described entirely in terms of CDW-induced phonon softening and wavevector-dependent changes in the phonon displacement, without invoking any more complex electronic excitations or CDW-related modification of the EPC. Overall, our results suggest a ‘real-space’ picture in which the CDW emerges due to strong electronic correlations and modifies the underlying phonons.

LSCO single crystals with  $x = 0.12, 0.17, 0.21$ , and  $0.25$  were grown using the floating zone method and are denoted as LSCO $n$  where  $n = 12, 17, 21$ , and  $25$ , respectively [27]. Structural and electronic characterization of the samples indicates excellent quality [26, 27]. High energy-resolution RIXS measurements were performed at 2-ID at the National Synchrotron Light Source II, Brookhaven National Laboratory with a resolution of  $\Delta E = 30$  meV full-width at half-maximum (FWHM) and at I21 at Diamond Light Source featuring a resolution of  $\Delta E = 55$  meV. The RIXS process is shown in Fig. 1(a). X-rays were tuned to the Cu  $L_3$ -edge and measurements were taken with  $\sigma$  x-ray polarization perpendicular to the scattering plane (unless otherwise specified). Reciprocal lattice units (r.l.u.) are defined in terms of  $(H, K, L)$  with lattice constants  $a = b = 3.76$  Å,  $c = 13.28$  Å. Different values of  $H$  and  $K$  were accessed by rotating

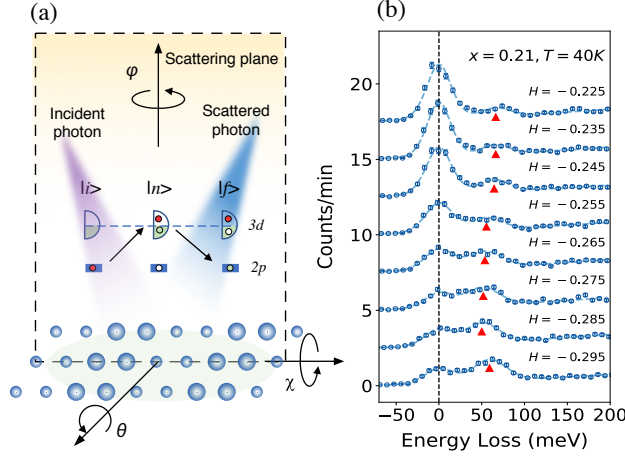


FIG. 1. RIXS process and typical low-energy RIXS spectra. (a) Schematic of the Cu  $L$ -edge RIXS process and experimental setup.  $|i\rangle$ ,  $|n\rangle$  and  $|f\rangle$  represent initial, intermediate and final states, respectively and solid (empty) circles represent occupied (unoccupied) states.  $\theta$ ,  $\chi$ , and  $\phi$  denote the sample rotations. (b) High-resolution ( $\Delta E = 30$  meV) RIXS data for LSCO21 at  $T = 40$  K (blue circles) illustrating the principle components of the spectra: Quasi-elastic scattering (marked by the vertical dotted line) and a low-energy phonon excitation (indicated by red triangles). The fit, as described in the main text, is represented by the dashed blue line.

the sample about the  $\theta$  and  $\chi$  axes without changing the scattering angle  $2\theta$ . Intensities were normalized to the intensity of the  $dd$  excitations similar to previous works [10, 18, 19, 23]. Grazing incidence geometry (defined as negative  $H$ ) was chosen to enhance the intensity of charge and lattice (phonon) excitations, and to suppress spin excitations, which are, in any case,  $> 200$  meV and outside of the energy window we focus on in this paper [28, 29].

Figure 1(b) plots RIXS data of LSCO at the Cu  $L_3$ -edge illustrating the main spectral features studied here: (i) a quasi-elastic peak and (ii) a dispersive feature around 50 – 65 meV. Feature (i) contains a component of trivial elastically scattered x-rays due to the finite disorder (defects) in the sample and surface scattering. Quasi-elastic scattering is further enhanced by static or quasi-static CDW correlations and displays a peak at  $Q_{\text{CDW}}$  whenever such correlations are present. The inelastic feature (ii) has been seen in several other cuprate RIXS experiments [19, 21–23, 30, 31] and is assigned to the in-plane Cu-O bond-stretching (BS) phonon mode in agreement with early inelastic x-ray and neutron works [32–38]. The spectra were fitted with a Pseudo-Voigt function for the elastic peak, an anti-symmetric Lorentzian function for the BS mode and a linear background. All components of the fit were convoluted with the energy resolution function.

Having assigned the basic spectral features, we use the ultra-high throughput of the I21 beamline to comprehen-

sively map out the momentum and doping dependence of LSCO  $12 \leq n \leq 25$ , (see Fig. 2). This doping range crosses over from 1/8 doping, where the CDW correlations are strongest, into the overdoped Fermi-liquid-like phase where the CDW disappears [24–26]. Importantly, this traverses a topological transition in the electronic structure where hole-pocket/arc-type states transform into an electron-like Fermi surface with a more Fermi-liquid-like scattering rate [24–26, 39]. This allows us to investigate the relationship of the RIXS spectra with the changes in electronic structure.

We first discuss the quasi-elastic CDW feature, which is summarized in Fig. 2(i) showing the integrated intensity in the  $\pm 20$  meV energy range of Fig. 2(a)-(h). The intensity enhancement of the resonant process combined with the background suppression attained by energy-resolving the scattered beam make RIXS very sensitive to even very short correlation length CDWs. A CDW around an in-plane wavevector of  $(-0.23, 0)$  is not only observed for LSCO12, where it was seen several times previously [14, 15, 40, 41], but also up to far higher dopings of LSCO21 [42]. The  $H$ -width of the quasi-elastic scattering is consistent with correlation lengths (calculated as  $2/\text{FWHM}$ ) of 25 – 45 Å with shorter values for  $x = 0.17$ . Within error, the peaks exist at the same  $H_{\text{CDW}} = -0.231 \pm 0.005$  r.l.u., consistent with the stripe phenomenology where  $H_{\text{CDW}}$  saturates for  $x > 1/8$  [43]. As expected, a substantial fraction of the CDW intensity is suppressed upon warming to 100 K, leaving only a much weaker and diffuse signal [14, 15, 26]. We further observe a non-monotonic intensity dependence as function of doping, with LSCO17 being weaker than LSCO12 and LSCO21. There are multiple potential explanations for such a behavior. Perhaps most plausible is to note that the quasi-static CDW intensity in cuprates tends to compete with superconductivity, so the reduced CDW intensity at  $x = 0.17$  may be associated with the enhanced superconducting correlations that are known to exist at this near-optimal doping level.

Next we discuss the inelastic component of the spectra in Fig. 2(a-h). In the 100-200 meV energy window, above the maximum phonon energy, flat, structure-less intensity arising from the charge continuum and the tail of the higher energy paramagnon excitations is present over all  $Q$ . This intensity shows no clear changes around  $Q_{\text{CDW}}$  and minimal changes with doping. In fact, only a slight increase in the overall intensity is found with increasing doping, which is expected as overdoped samples are more metallic. The inelastic intensity below 100 meV is dominated by the BS phonon, which shows clear energy dispersion and very strong intensity variation. Superficial inspection of the raw intensity in Fig. 2(a) may appear as if a soft mode is dispersing to zero energy at  $Q_{\text{CDW}}$ . This can be examined through a systematic study of samples with different doping levels. To separate the phonon from charge and quasielastic intensity, we fit the data using the

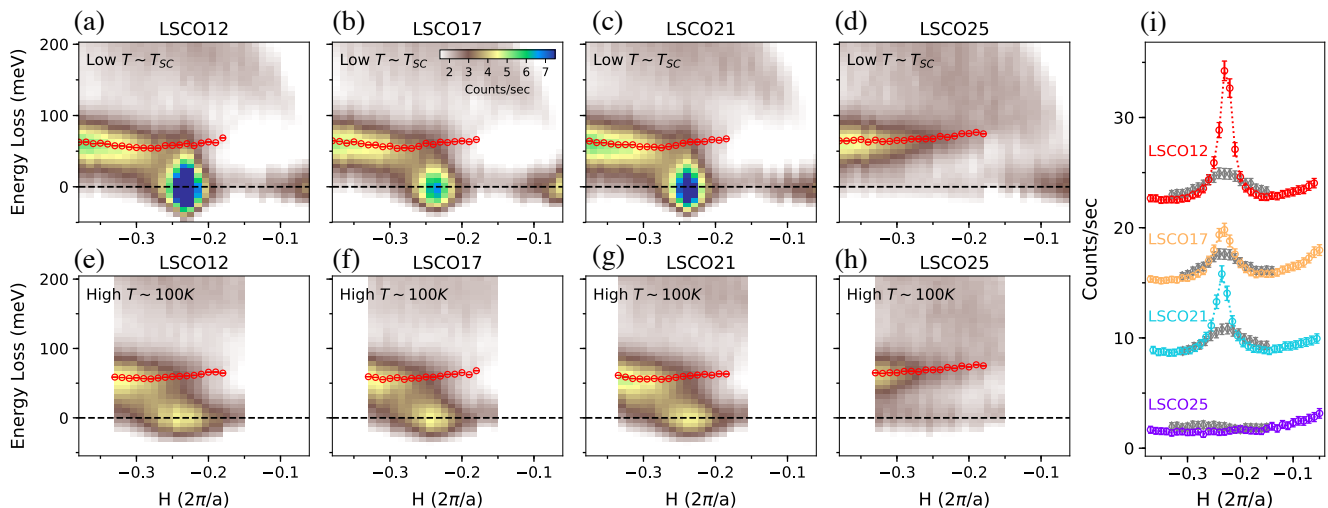


FIG. 2. Doping and temperature evolution. (a)-(d) show ( $\Delta E = 55$  meV) RIXS maps of  $\text{LSCO}_n$  ( $n = 12, 17, 21, 25$ ) at their superconducting transition temperature  $T_{\text{SC}}$ , where the CDW is strongest. All maps share the same colorscale inset in panel (b). Red circles represent the BS phonon energy. (e)-(h) The same measurement for  $T = 100$  K. (i) Comparison of integrated intensity in the  $\pm 20$  meV energy window. Data from panels (a)-(d) at  $T = T_{\text{SC}}$  are represented in color; data at  $T = 100$  K from panels (e)-(h) are plotted in gray.

previously described model and display the results as red circles in Fig. 2(a-h). Since the phonon intensity drops strongly as  $|H|$  decreases, we focus on  $|H| > 0.18$  where the BS phonon can be fitted with good precision.

Figure 3 summarizes the evolution of the BS phonon parameters. Although the phonon softening, shown in panel (a), is appreciable ( $13 \pm 4$  meV or  $19 \pm 6\%$ ), it never shows full soft-mode (i.e. zero energy) behavior. The simultaneous disappearance of both the elastic peak and the phonon softening in LSCO25 makes a strong case that the softening is intimately related to the CDW correlations. Inelastic x-ray and neutron scattering measurements of phonon softening in underdoped cuprates have also assigned the phonon softening to CDW correlations [32–38]. It is noted that the phonon energy in LSCO25 is slightly higher than other samples, which might be linked to lattice contraction associated with large Sr concentrations.

The phonon intensity dispersion is plotted in Fig. 3(c). Within error, no phonon intensity anomalies are seen around  $Q_{\text{CDW}}$ , instead the clearest feature is a strong increase with  $|H|$ . Since RIXS excites phonons via the EPC process, the measured intensity reflects this interaction strength and scales with  $g^2$  where  $g$  is the EPC [16, 30, 31, 44, 45]. As a function of  $Q$ , the “breathing-type” Cu-O bond displacement involved in the BS mode changes. Assuming a well-defined Madelung energy change associated with Cu-O bond stretching, one can predict RIXS intensity scaling  $I \propto g_{\text{br}}^2 = \sin^2(\pi H) + \sin^2(\pi K)$  [30, 31, 44, 46]. The comparison in Fig. 3(c) shows that this simple model is sufficient to describe the intensity behavior of our data, without invoking any more

complex phenomenology. It is worth adding that definitively distinguishing  $\sin^2(\pi H)$  scaling from other scaling forms is somewhat challenging. The photon energy corresponding to the Cu  $L_3$  edge intrinsically limits the highest  $|H|$  we can reach, and at low  $|H|$  leakage of specular scattering intensity overwhelms the low-energy region of the RIXS spectra. The slight flattening of the dispersion at  $x = 0.25$  might arise from some leakage of additional background at high doping levels. With these considered, the agreement with  $\sin^2(\pi H)$  scaling holds well in the reciprocal-space range measured.

*Discussion of the CDW.*— Both the quasielastic RIXS intensity and the phonon softening demonstrate the existence of CDW correlations up to a remarkably high doping level of  $x = 0.21$ , traversing the topological transition in the electronic structure [24–26], in which arc or hole-pocket like states centered around the Brillouin zone corner transform into an electron-like Fermi surface at the Brillouin zone center. This result confirms very recent non-resonant diffraction measurements and shows, due to the resonant nature of the RIXS probe, that the correlations involve the electronically active Cu states [26]. The persistence of the CDW correlations, despite very substantial Fermi surface changes, provides a vivid demonstration that the LSCO CDW cannot be described using any type of weak-coupling Fermi surface nesting picture, as previously suggested for some other cuprate materials [47]. Instead, the nearly doping-independent CDW wavevector supports mechanisms in which the periodicity of the CDW is set by the short-range electronic interactions. Here, doped holes can save super-exchange energy by clustering together and breaking fewer mag-

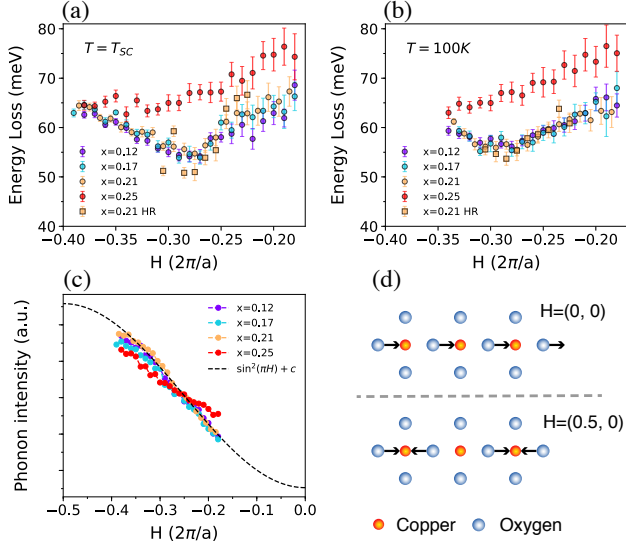


FIG. 3. Momentum dependent energy and intensity of the BS phonon mode. (a)&(b) Show the phonon energy dispersion and softening in the vicinity of  $\mathbf{Q}_{CDW}$  at (a)  $T = T_{SC}$  and (b)  $T = 100$  K. Circles and squares represent data from Fig. 2 ( $\Delta E = 55$  meV) and Fig. 1 ( $\Delta E = 30$  meV), respectively. (c) Plots the integrated intensity of the BS phonon mode in the  $[40, 120]$  meV energy window after subtracting elastic peak. The dashed line represents the  $\sin^2(\pi H) + \text{const.}$  fit to LSCO21. (d) Illustration of the oxygen atom displacements involved in the BS mode at  $H = (0, 0)$  and  $(0.5, 0)$ .

netic bonds, but by doing so, they pay a cost of increased kinetic and Coulomb energy. It has been proposed that a CDW is the optimal compromise between these two tendencies [4]. However, a fascinating question remains regarding why the CDW is so stable against increasing doping and electron itineracy. Furthermore, our results motivate a reexamination of the anomalous transport properties of the cuprates, which are often discussed in terms of strange metal physics below a critical doping level of  $x_c \approx 0.19$ , where the systems becomes increasingly more Fermi-liquid-like [1]. Since CDW correlations can exist over a more extended doping range than previously thought, it is interesting to consider their influence on transport properties [48]. These results, however, argues against a quantum critical point that is associated with a CDW transition as similar as CDW correlations exist in LSCO17 and LSCO21, either side of the putative critical doping of  $x_c \approx 0.19$ . In terms of the CDW fluctuations, we note that the size of the phonon softening is reduced when warming to  $T = 100$  K, but the magnitude of the reduction is considerably less than the reduction in the quasi-elastic CDW intensity. This suggests that only a relatively small fraction of the total CDW correlations are nucleated into the CDW-order, as otherwise the magnitude of the phonon softening would be expected to scale with the quasi-elastic intensity.

An intriguing feature of the CDW effects is that the

$\mathbf{Q}$ -vector with the largest phonon softening does not coincide with the peak in the quasi-elastic scattering at 0.235 r.l.u., but occurs at a larger wavevector of  $H = 0.275$  r.l.u. We can consider two candidate explanations for this. The first is that the interactions causing the CDW instability are not the same as the interactions that pin the CDW. Electronic interactions, for instance, may potentially stabilize a wide-range of CDW wavevectors, but lattice- or SDW-coupling might play the final role in setting the final CDW wavevector. This concept was proposed several years ago [49] and is further supported by the previously observed change in CDW wavevector with temperature [18, 23, 50]. The phonon softening does not show any obvious difference between  $T_{SC}$  and 100 K in Fig. 3(a)&(b), in contrast to the CDW peak, which also supports distinct mechanisms for CDW-formation and CDW-pinning. Another potential explanation for the shift is that the effect is due to the  $\mathbf{Q}$  dependence of the EPC. Since EPC increases with  $H$ , the softening at higher  $H$  will be enhanced, which would displace the point of maximum softening.

*Discussion of electronic CDW excitations.* — A strength of the current dataset is the opportunity to compare LSCO12 and LSCO21, which exhibit comparable levels of CDW order, despite their very different doping levels. Any electronic CDW excitations that may be present in the RIXS spectra would be expected to increase with electronic density of states and change substantially with doping. Since the overall form of the spectra is rather similar over this large doping range, our results argue against the presence of electronic CDW excitations, as suggested previously, as a ubiquitous, intrinsic feature of RIXS spectra of the cuprates [19]. It is, however, possible that the x-ray polarization is important to observing electronic CDW excitations [51].

*Electron phonon coupling.* — The concept of using RIXS to extract EPC in cuprates has generated considerable excitement recently [22, 30, 31, 44, 52]. Our results support this, in the sense that we observe intensity scaling as  $I \propto \sin^2(\pi H)$ . However, no phonon intensity anomalies related to the CDW are observed. A recent preprint reports measurements of  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_{4+\delta}$  which used an incident energy detuning method, formulated in Refs. [30, 31, 52], to suggest a very large CDW-induced modification of the EPC from  $g = 0.30 \rightarrow 0.35$  upon cooling into the CDW phase [22]. Assuming  $I \propto g^2$ , this would imply a phonon intensity change of  $(0.35/0.3)^2 \sim 1.4$ . As such, it is very difficult to justify the absence of a discernible  $\sim 40\%$  temperature-induced change in the phonon dispersion at  $\mathbf{Q}_{CDW}$  on resonance in our data or that in Ref. [22].

*RIXS as a probe of cuprate CDWs.* — Overall, our results support a real-space picture of an electronically driven CDW, without needing to invoke nesting or a van Hove singularity. RIXS measures the phonon softening occurring due to the charge modulation, but the

overall doping dependence is fully explicable without invoking more complex phason, Fano, or CDW-enhanced EPC effects that have generated considerable excitement recently [17–20, 22]. Our observation is backed, by improved energy resolution ( $\Delta E = 30$  meV) and more extensive doping dependence compared with what has been done previously [17–20, 22]. Excluding these exciting effects is at some level disappointing in terms of novel excitations, but is, however, highly important in view of the more-and-more extensive use of RIXS. Although RIXS, in this case, provides information similar to inelastic x-ray and neutron scattering, compelling applications for RIXS remain in cases where the x-ray penetration depth and resonant mode selectivity is important [45].

In conclusion, we report RIXS measurements of CDW correlations in LSCO over an extensive doping range. CDW-related quasi-elastic scattering and phonon softening is observed from  $x = 0.12$  to  $x = 0.21$ , traversing a topological transition in the Fermi-surface, before disappearing at  $x = 0.25$ . Based on these results, we conclude that the spectra have little or no direct coupling to electronic excitations. Instead the spectra are dominated by CDW-driven phonon softening and phonon intensity variations arising from changes in the phonon displacement as a function of  $Q$ . Overall, our results support a scenario in which the CDW is driven by strong correlations and clarify that the low-energy RIXS response in cuprates are driven by the CDW modifying the lattice, invoking more complex interactions.

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# Supplemental Material: Nature of the charge density wave excitations in cuprates

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This document provides additional details of the sample synthesis, sample characterization, temperature-dependent phonon intensity data, and fitting of the RIXS spectra.

## SAMPLE SYNTHESIS

$\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  samples were grown by the traveling-solvent floating-zone method. For each composition, a single feed rod with a length of 20–25 cm was used. After the growth, the first few centimeters of the crystal rod were removed and discarded, while the remainder was annealed in flowing  $\text{O}_2$  at 980°C for 1 week. Sample surfaces were prepared by cleaving the samples mechanically to expose a  $c$ -axis face. The samples were crystallographically aligned prior to the measurements RIXS using Laue diffraction. During the RIXS measurements themselves, the  $\theta$  and  $\chi$  angles were further refined by optimizing the CDW intensity as a function of both angles.

## SAMPLE CHARACTERIZATION

The superconducting transition temperatures were determined by magnetization measurements in a 1 mT applied magnetic field (after cooling in zero field) yielding the expected values of  $T_{\text{SC}} = 28, 37, 30$ , and 10 K, for LSCO12, LSCO17, LSCO21 and LSCO25, respectively. X-Ray diffraction measurements confirmed excellent sample crystallinity with crystal mosaics of the order of  $0.02^\circ$ . The doping level was characterized by angle-resolved photoemission spectroscopy (ARPES) [1]. The effective doping was determined to be consistent with the Sr concentration via a tight binding fitting of the Fermi surface. The topological change in Fermi surface topology was also observed between LSCO17 and LSCO21. Very similar results have been observed in previous ARPES studies of LSCO [2, 3].

## PHONON INTENSITY DEPENDENCE

In Fig. S1 we show that the phonon intensity dispersion is essentially identical at  $T_c$  and 100 K.

## FITTING OF THE RIXS SPECTRA

In order to illustrate the method and quality of data fitting, we present the fitting result for LSCO17 at low temperature in Fig. S2. The spectra were fitted with a Pseudo-Voigt function for the elastic peak, an anti-symmetric Lorentzian function for the bond-stretching (BS) mode and a linear background, all convoluted with resolution.

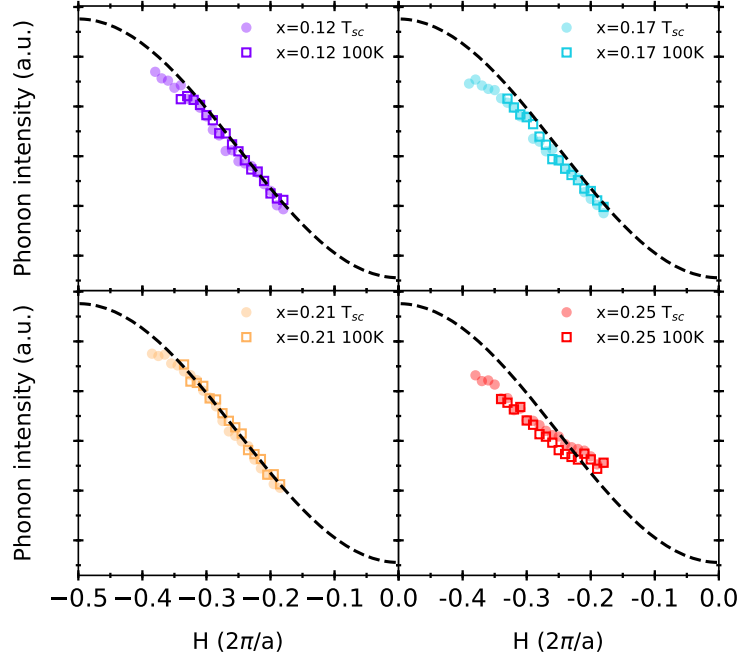


FIG. S1. Phonon intensity as a function of  $H$  similar to Fig. 3(c) of the main text, but with the 100 K data overlaid on the low-temperature data and separated onto different panels. This shows that the phonon intensity is the same at  $T_c$  and 100 K for all the different dopings.

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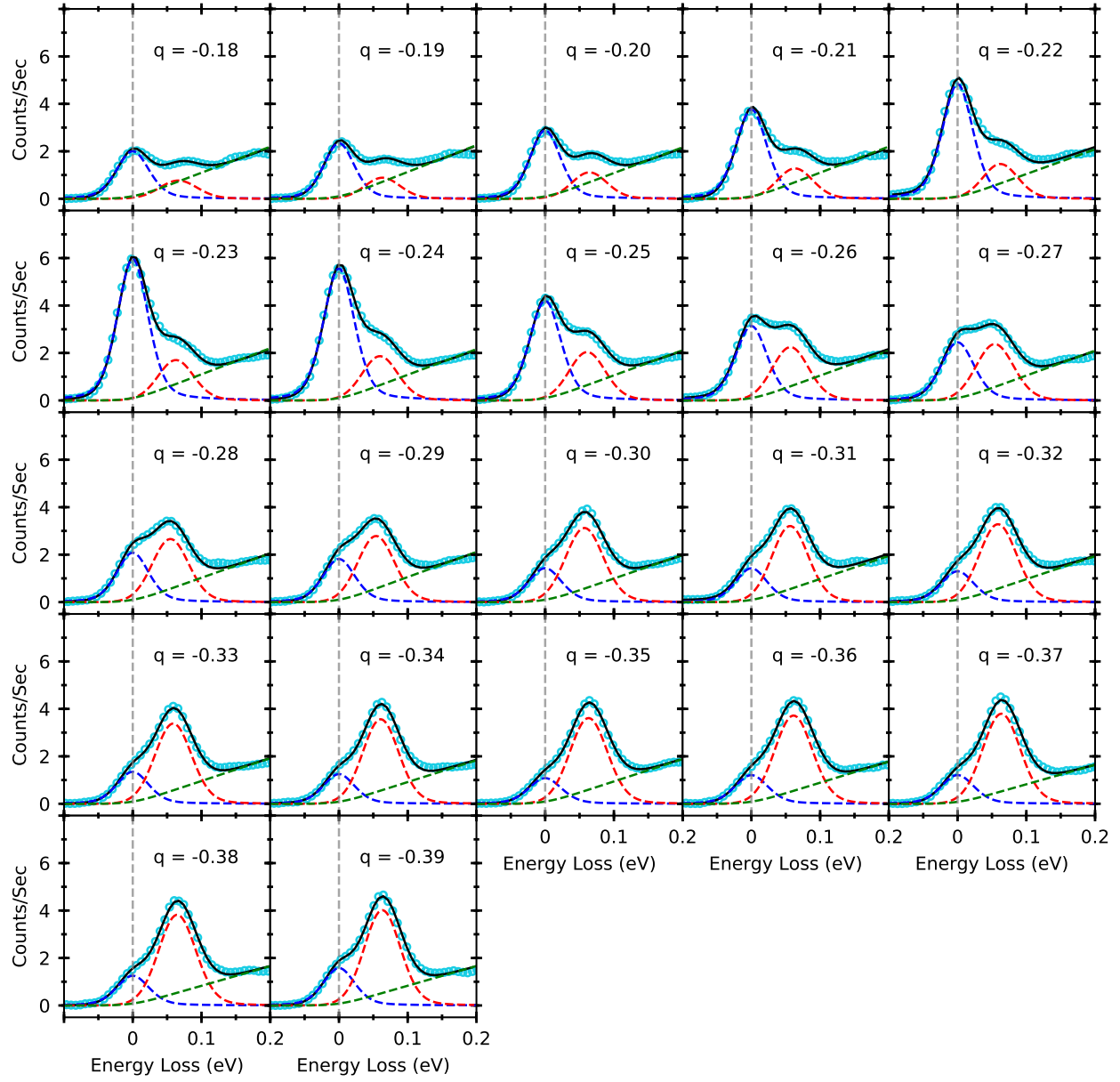


FIG. S2. Fitting results for LSCO17 at  $T = T_{SC}$ . The data is represented by cyan dots and the fit is shown as a black line. Blue, red and green dashed lines represent the elastic line, phonon mode and background components, respectively.