

Doping evolution of the charge density wave and charge density fluctuations in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

Charles C. Tam^{1,2,*}, Mengze Zhu^{1,†,‡}, Maud C. Barthélemy¹, Lauren J. Cane¹, Oliver J. Lipscombe¹, Stefano Agrestini², Jaewon Choi², Mirian Garcia-Fernandez², Ke-Jin Zhou^{2,§} and Stephen M. Hayden^{1,||}

¹*H. H. Wills Physics Laboratory, University of Bristol, Bristol BS8 1TL, United Kingdom*

²*Diamond Light Source, Harwell Campus, Didcot OX11 0DE, United Kingdom*



(Received 12 August 2025; revised 17 February 2026; accepted 22 April 2026; published 13 May 2026)

Cuprate superconductors show various collective charge correlations that are intimately connected with their electronic properties. In particular, charge order in the form of an incommensurate charge density wave (CDW) order with an in-plane wave vector $\delta_{\text{CDW}} \approx 0.23\text{--}0.35$ reciprocal lattice units appears to be universally present. In addition to CDW, dynamic charge density fluctuations (CDFs) are also present with wave vectors comparable to δ_{CDW} . CDFs are present up to ≈ 300 K and have relatively short correlation lengths of $\xi \approx 20$ Å. Here we use Cu-L_3 and O-K resonant inelastic x-ray scattering (RIXS) to study the doping dependence of CDW and CDFs in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. We fit our data with (quasi)elastic peaks resulting from the CDW and up to four inelastic modes associated with oxygen phonons that can be strongly coupled to the CDFs. Our analysis allows us to separate the charge correlations into three components: the CDW with wave vector $\delta_{4a\text{-CDW}} \approx 0.24$ and two CDF components with $\delta_{4a\text{-CDF}} \approx 0.24$ and $\delta_{3a\text{-CDF}} \approx 0.30$. We find that for $T \approx T_c$ the CDW coexists with the CDFs for dopings near $x = p \approx 1/8$. The $4a\text{-CDW}$ disappears beyond $x = 0.16$ and the $4a\text{-CDF}$ beyond $x = 0.19$, leaving only a weak $3a\text{-CDF}$ at the highest doping studied, $x = 0.22$. Our data suggest that low-energy charge fluctuations exist up to doping $x = 0.19 = p^*$, where the pseudogap disappears; however, we find no evidence that they are associated with a quantum critical point.

DOI: [10.1103/PhysRevB.113.174506](https://doi.org/10.1103/PhysRevB.113.174506)

I. INTRODUCTION

Understanding the normal state from which superconductivity emerges is essential to constructing theories of high- T_c superconductivity in the cuprates. The normal state of the hole-doping versus temperature (p - T) phase diagram of the cuprates contains a number of intertwined phases and behaviors [1]. These include the pseudogap, strange-metal behavior, and spin and charge density wave order (SDW, CDW) and their associated fluctuations. In this paper, we focus on CDW order and charge density fluctuations (CDFs) [2]. Our aim is to determine their evolution with doping, so that they may be correlated with other physical properties.

Conventional CDWs are periodic modulations of the valence charge density, which have a corresponding atomic

displacement [3]. In conventional continuous phase transitions, there is an order parameter whose correlation length ξ diverges on cooling through the transition temperature. The charge correlations in cuprates do not follow this behavior [2]. At high temperatures, $T \approx 150\text{--}300$ K (depending on the system), short-range correlations with $\xi \approx 15$ Å and in-plane wave vectors $\mathbf{Q} = (\delta, 0)$ and $(0, \delta)$ with $\delta \approx 0.23\text{--}0.35$ reciprocal lattice units (r.l.u.) are observed [2,4–6]. We generically call these correlations “charge density fluctuations” (CDFs). They are inelastic or quasi-elastic in nature and have also been called “CDW fluctuations,” “CDW excitations,” or “charge fluctuations.” On cooling, a second component, which we call “charge density wave order” (CDW), develops below a characteristic temperature T_{CDW} . The correlation length and the intensity I_{CDW} of the CDW order grow upon cooling with ξ_{CDW} reaching approximately 70 Å in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (YBCO) [4,7], 35 Å in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) [8,9] and 160 Å in $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$ (Tl2201) [10]. In LSCO, the CDW peak is relatively narrow at low temperature $T = T_c \approx 31$ K with $\xi \approx 35$ Å. Below T_c , both ξ_{CDW} and I_{CDW} can be reduced. This behavior is interpreted as evidence of competition between CDW and superconductivity.

Resonant inelastic x-ray scattering (RIXS) can be used to investigate collective charge correlations. RIXS is more sensitive to the valence charge density than nonresonant x-ray methods. The state-of-the-art energy resolution of soft x-ray RIXS spectrometers is approaching 10 meV at the time of writing [11], meaning quasi-elastic or elastic scattering components can be resolved from inelastic scattering. Previous RIXS studies on cuprates have identified separate CDW and

*Contact author: ctam@ucsb.edu

†Contact author: zhumen@phys.ethz.ch

‡Present address: Laboratory for Solid State Physics, ETH Zürich, 8093 Zürich, Switzerland.

§Present address: National Synchrotron Radiation Laboratory and School of Nuclear Science and Technology, University of Science and Technology of China, Hefei 230026, China.

||Contact author: s.hayden@bristol.ac.uk

Published by the American Physical Society under the terms of the Creative Commons Attribution 4.0 International license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

CDF components in YBCO [12], $\text{HgBa}_2\text{CuO}_{4+\delta}$ (Hg124) [13] and LSCO [11] with the CDW contribution being sharper in reciprocal space than the CDF.

To understand the physical properties and the phase diagram of the cuprates, it is important to know how the various components of the charge correlations described above evolve with doping. LSCO is well suited for this type of study because it can be doped through the phase diagram. Thus, in this article, we investigate the nature of the CDW order and CDFs in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ as a function of doping. We measure each doping near T_c to avoid probing the competition with superconductivity [7,8]. There have been a number of studies [8,9,14–16] which focus on the evolution of the charge correlations and show that they weaken with doping. Our approach contrasts with these studies in that we collect systematic wave-vector-dependent RIXS data with high energy resolution and at both Cu- L_3 and O- K edges and fit the data to two (Cu- L_3) or four (O- K) phonon modes and a (quasi-)elastic peak. We interpret changes of the phonon intensity and dispersion anomalies as signaling the presence of CDFs. Using this method we distinguish three contributions to the charge response that change differently with doping.

The article is organized as follows: The experimental methods, including sample growth, characterization, the RIXS method, and fitting procedures, are detailed in Sec. II. The results, including RIXS data and the extracted parameters, are described in Sec. III. In Sec. IV we described how the charge order and charge density fluctuations evolve with doping. In Sec. V, we discuss how our results relate to other experiments. In Sec. VI, we discuss the implications of our observations for models of the cuprate phase diagram and the transport properties. Finally, in Sec. VII, we summarize the main conclusion of the work.

II. EXPERIMENTAL METHOD

A. Sample growth and characterization

We studied single crystals of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ with Sr doping values ranging from $x = 0.11$ to $x = 0.22$, grown by the traveling solvent floating zone method, followed by annealing in flowing oxygen at 800°C for 2 weeks. The Sr content x , which we take to be equivalent to the hole doping p , was measured by electron probe micro-analysis (EPMA). Pieces used in the current study were cut from high-quality single-crystal growths that have been used in previous inelastic neutron scattering (INS), x-ray, and transport studies [8,17,18]. T_c was determined by the midpoint of the magnetic susceptibility transition. The measured Sr doping and T_c values of the samples used in this study are listed in Table I. Samples were postcleaved in the load lock in a vacuum of 1×10^{-8} mbar or better. Samples were oriented *in situ* on the (002) reflection, combined with alignment on the CDW peak, which orders along the primary crystallographic directions or using information from *ex situ* Laue measurements.

B. Spectrometer details

RIXS measurements were taken at beamline I21, Diamond Light Source [19]. In this article, we use both Cu- L_3 and O- K edge RIXS to measure the CDW and associated charge

TABLE I. Sample T_c values measured as the midpoint of the susceptibility transition, and Sr doping concentration values, determined by EPMA.

Sr concentration, x	Midpoint, T_c (K)
0.110(2)	24.2(5)
0.125(5)	31.0(5)
0.160(2)	38.4(5)
0.19(1)	33.5(5)
0.215(5)	26.0(5)

fluctuations in LSCO. Momentum transfer is labeled in reciprocal lattice units (r.l.u.) of the tetragonal unit cell, where $\mathbf{Q} = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^*$, with $a = b = 3.86$ Å and $c = 13.1$ Å. The photon energy loss is $E = E_i - E_f = \hbar\omega$, where E_i and E_f are the initial and final photon energies. Scans were taken with grazing incidence (i.e., $\theta < \Omega$), where θ is the incidence angle and Ω is the scattering angle. Grazing incidence is denoted with negative H . We used linear vertical polarized x-rays for all measurements (σ polarization). The instrumental resolution was determined as the full-width-at-half-maximum of elastic scattering from carbon tape. For Cu- L_3 ($E_i \approx 928$ eV) this was $\Delta E_{\text{FWHM}} \approx 37$ meV and for O- K ($E_i \approx 527$ eV) this was $\Delta E_{\text{FWHM}} \approx 27$ meV. The CDW in LSCO is weakly correlated in L but is peaked at integer + 1/2 positions [8].

The RIXS H scans were taken at fixed scattering angle values of $\Omega = 117^\circ$ and $\Omega = 154^\circ$, to give $L = 1.5$ and $L = 0.7$ (r.l.u.) at the expected in-plane CDW wave vector $H = 0.23$ for the Cu- L_3 and O- K edge measurements, respectively. With these constraints on the scattering angle and the low incident energy of soft x-rays, the highest H value that can be reached in LSCO at Cu- L_3 is $|H| = 0.38$, and at O- K is $|H| = 0.3$. Measurements were normalized to the orbital excitations in range [1, 3] eV, for both O- K and Cu- L_3 data to allow for comparisons between different doping values. Measurements were performed at the superconducting transition temperature T_c of the respective samples, which are listed in Table I. The aim of the study was to see how the CDW and CDFs vary with doping without the complications entering the superconducting state [7,8].

C. RIXS measurements

Undoped cuprates are classified as charge-transfer Mott insulators [20], since the Mott gap is determined by the anion charge transfer Δ , rather than the onsite Coulomb repulsion U . Due to strong hybridization between Cu 3d and O 2p, the valence band has oxygen character and can be understood as a single band, called the Zhang-Rice singlet (ZRS) [21] [see Fig. 1(a)]. Cu- L RIXS involves excitations between the Cu 2p states and the Cu 3d upper Hubbard band (UHB). While O- K RIXS involves excitations from the O 1s state to the ZRS. The ZRS appears as a prepeak in O- K x-ray absorption spectroscopy (XAS) [see Fig. 1(b)], at lower energy than the UHB, and is where doped holes are seen to reside [22].

For the O- K RIXS measurements, we tuned the incident x-ray energy to the ZRS prepeak in O- K XAS, plotted in Fig. 1(b), at around 527.3 eV. An O- K RIXS spectrum taken at $H = -0.29$ of $x = 0.19$ LSCO is seen in Fig. 1(c). From right

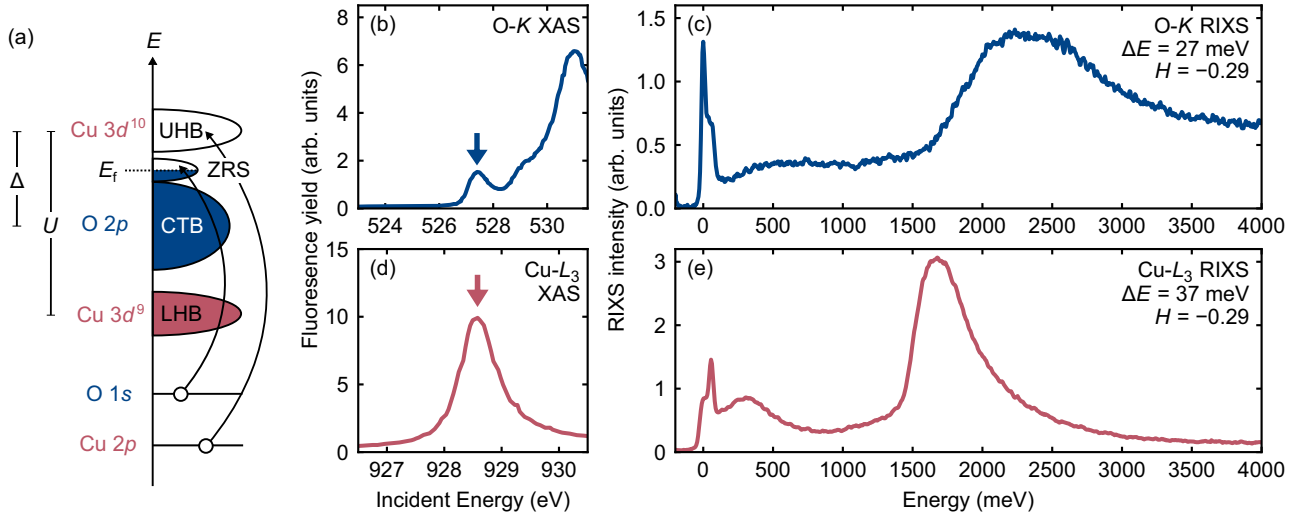


FIG. 1. RIXS at two atomic sites. (a) Schematic energy-level diagram of superconducting cuprates in the Zaanen-Allen-Swartzky scheme [20], and the excitations that Cu- L_3 and O- K RIXS creates within the scheme. UHB means upper-Hubbard band, ZRS means Zhang-Rice singlet, and LHB means lower Hubbard band. Δ is the charge-transfer energy, and U is the Coulomb energy. (b), (d) Cu- L_3 and O- K XAS spectra obtained in fluorescence yield mode. The arrows mark the incident energy for the RIXS experiments. Small discrepancies between absolute energies in the XAS spectra and published values are due to a small miscalibration in the beamline monochromator. (c), (e) RIXS spectra taken at $H = -0.29$ at Cu- L_3 and O- K edges, respectively. All measurements in this figure are taken on the same $x = 0.19$ LSCO sample.

to left, we see a broad orbital excitation centered at 2500 meV. Next, we see a very broad, nondispersing bimagnon centered at around 600 meV, and finally several low-energy features below 100 meV.

For the Cu- L_3 RIXS measurements, the incident x-ray energy was tuned to the maximum of the Cu- L_3 d^9 absorption peak, at about $E_i = 928$ eV, plotted in Fig. 1(d). A Cu- L_3 RIXS spectrum taken at $H = -0.29$ of $x = 0.19$ LSCO is seen in Fig. 1(e). From right to left, we see dd crystal field excitations centered at around 1600 meV energy loss, a dispersing paramagnon centered at around 300 meV, and again several low-energy excitations below 100 meV.

Figure 2 shows RIXS spectra collected at the O- K and Cu- L_3 edges for each of the five compositions investigated at $H = -0.19$. Figure 3 shows collections of spectra such as those in Fig. 2 plotted as H - E color intensity maps, where $E = \hbar\omega$ is the energy loss.

D. Interpretation and fitting

Measurements of charge excitations using RIXS, such as those presented here, are interpreted in terms of the dynamical structure factor $S(\mathbf{Q}, \omega)$ defined as the Fourier transform of the charge density-density correlation function $\langle n(\mathbf{r}, t)n(0, 0) \rangle$ in time and space [23]. Furthermore, $S(\mathbf{Q}, \omega)$ can be related to a generalized charge susceptibility $\chi(\mathbf{Q}, \omega)$ through the fluctuation-dissipation theorem $S(\mathbf{Q}, \omega) = 2\hbar(1 - e^{-\beta\hbar\omega})^{-1}\chi''(\mathbf{Q}, \omega)$. The actual RIXS cross section will also contain other factors and matrix elements determined by the polarization of the x-rays, orbital character, and geometry [23]. It should also be noted that the processes at the O- K and Cu- L edges differ. The indirect process at the K edge, where excitations are created through the intermediate-

state core-hole potential, means that the charge fluctuations are probed effectively [23].

A diagrammatic approach [11,24] shows that the coupling between CDFs and phonons will lead to peaks in the RIXS intensity, I_{RIXS} , where phonons cross CDFs (see schematic in Fig. 8). This picture is supported by RIXS data in the cuprates, where a number of oxygen phonons with strongly modulated intensities [11,25] are observed at the Cu- L and O- K edges. Certain phonons involving motion of the in-plane oxygen atoms are visible in RIXS because of their strong electron-phonon coupling [24]. These are the longitudinal acoustic (LA), the A_{1g} and B_{1g} c axis modes, and the bond-stretching (BS) mode. This picture is supported by very-high-resolution ($\Delta E_{\text{FWHM}} = 16$ meV) O- K RIXS measurements [11] on LSCO ($x = 0.15$), which are fitted to four inelastic components. The analysis in this paper follows this work. With the energy resolution of the data currently presented, we are able to understand the RIXS spectra in terms of these four phonons at O- K , or two phonons at Cu- L_3 .

We interpret the peaks in I_{RIXS} as peaks in $S(\mathbf{Q}, \omega)$ rather than variations of the electron-phonon coupling matrix element $g(q)$. This is justified by theoretical calculations [24] which suggest that $g(q)$ is slowly varying. For example, for the bond-stretching mode $g(q) \sim \sin(q_x a/2)$, which is peaked at $q = (1/2, 0)$ rather than $(\delta_{4a}, 0)$.

1. Five-peak model (O- K edge)

We fit the low-energy part of the O- K RIXS spectra in the range $[-100, 200]$ meV to a five-peak model. This consists of a pseudo-voigt (pV) function (a linear combination of Gaussian and Lorentzian functions, normalized such that $pV(0) = 1$) to fit the quasi-elastic (QE, $\omega = 0$) scattering, Gaussians with the experimental resolution width to fit the

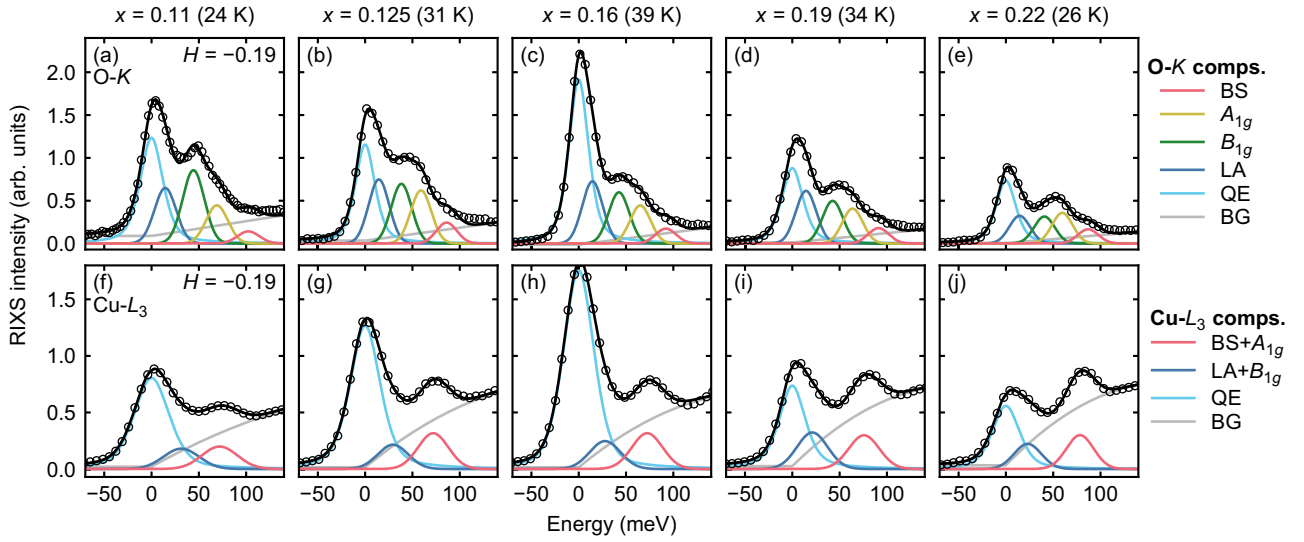


FIG. 2. RIXS fitting scheme. (a)–(e) Five-peak fitting scheme for O-K RIXS fits for the five doping values. The overall fit is plotted in black, and the quasi-elastic (QE), longitudinal acoustic (LA) mode, A_{1g} and B_{1g} bond-buckling modes, bond-stretching (BS) phonon and polynomial background (BG) components are plotted. (f)–(j) Three-peak fitting scheme for Cu- L_3 RIXS fits. The overall fit is plotted in black, and the quasi-elastic, p_1 , a combination of the longitudinal acoustic + B_{1g} mode ($LA + B_{1g}$), and p_2 , a combination of bond-stretching + A_{1g} mode ($BS + A_{1g}$), and polynomial background (BG) components are plotted. All spectra in this figure are taken at $H = -0.19$.

four phonon modes, and a polynomial background. From low to high energy [see Fig. 4(p)], the phonon modes are a ≈ 14.5 meV longitudinal acoustic (LA) mode, a ≈ 39 meV A_{1g} buckling mode, a ≈ 56 meV B_{1g} buckling mode, and a ≈ 80 meV bond stretching (BS) phonon [11]. For a cut along the trajectory $\mathbf{Q} = (H, 0)$, the fitted intensity can be written as

$$I_{\text{RIXS}}(H, \omega) = I^{\text{QE}}(H) pV(\omega) + \sum_{i=\{LA, B_{1g}, A_{1g}, BS\}} I^i(H) \times \exp \left[-\frac{(\omega - \omega_i)^2}{2\sigma_{\text{res}}^2} \right] + \text{B.G.}, \quad (1)$$

where $I_{\text{RIXS}}(H, \omega)$ is the RIXS intensity, i indexes the four phonon peaks, I^i is the intensity of the i th peak, ω_i is the peak position, B.G. is the background intensity, and the instrumental resolution $\Delta E_{\text{FWHM}} \approx 27$ meV $\approx 2.335\sigma_{\text{res}}$.

The current experimental resolution is not sufficient to allow the LA phonon frequency to vary in the fitting procedure, so it is fixed to 14.5 meV, which is the frequency measured in LSCO at $(0.25, 0, 0)$ [28] by INS and is consistent with a previous RIXS study [11]. When the frequencies were allowed to vary, the frequencies of the A_{1g} and B_{1g} buckling modes are consistent with Ref. [11]. However, when we

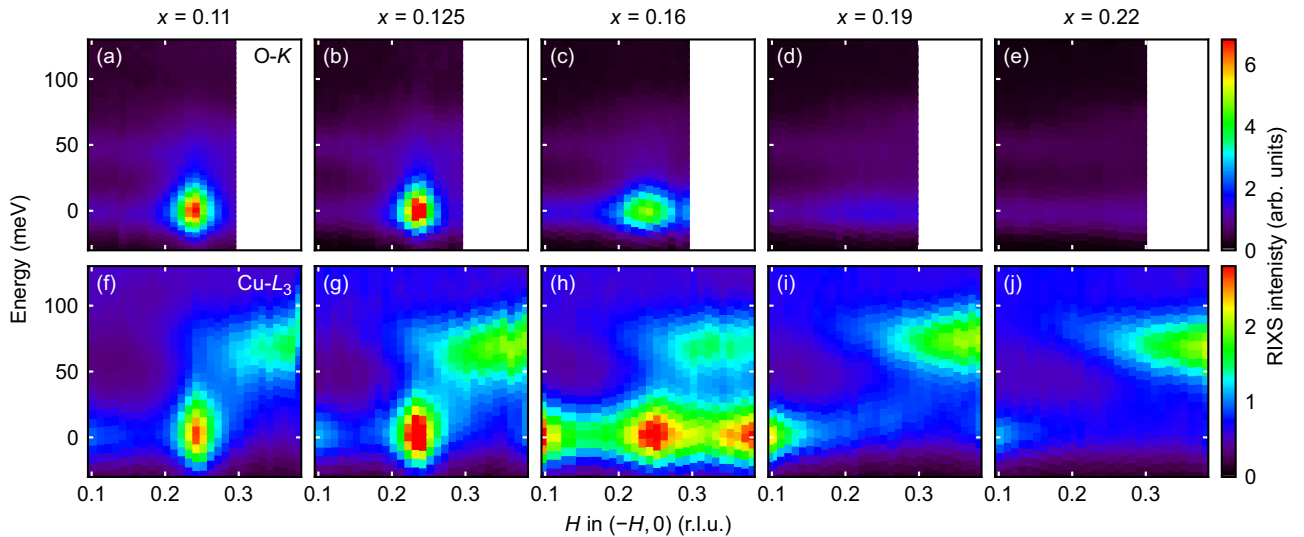


FIG. 3. H -dependent RIXS color maps. (a)–(e) Low-energy O-K RIXS color maps for samples with different Sr doping levels. (f)–(j) Low-energy Cu- L_3 RIXS color maps. The $x = 0.16$ sample used to collect data in this paper showed a larger elastic background [panels (c) and (h)] than the other samples. The background is most likely from poor surface quality resulting from a poor cleave.

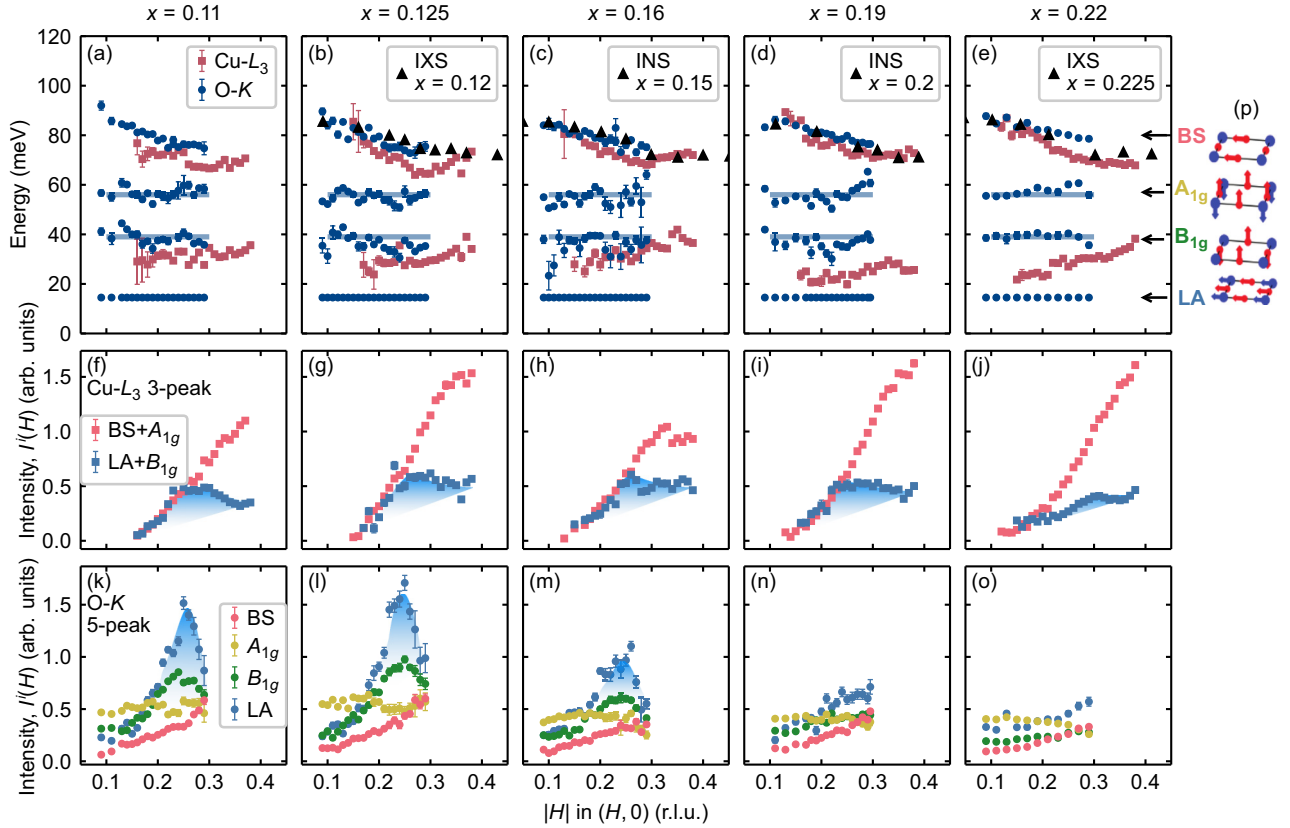


FIG. 4. Phonons measured with RIXS in LSCO. (a)–(e) Fitted phonon dispersion, measured with both O-K (blue markers) and Cu-L₃ (red markers) RIXS. The black points are the dispersion of the BS phonon from samples with similar doping levels, measured with IXS/INS, with data taken from Refs. [26,27]. The pale blue lines mark the average value of the dispersion. (f)–(j) Fitted Cu-L₃ phonon intensities (Gaussian height), using the three-peak model described in the text. (k)–(o) Fitted O-K phonon intensities, using the five-peak model described in the text. Blue shaded areas are the approximate signal due to CDFs in the LA data. (p) Illustration of the BS, A_{1g}, B_{1g} and LA phonons modes in the CuO₂ planes.

examine the H dependence of the intensity, the frequencies are fixed to their average values of 39 and 56 meV, respectively. Example fits for $H = -0.19$ are plotted in Figs. 2(a)–2(e), and the result of fitting the data displayed in Fig. 3 is shown in Fig. 4. In Figs. 2(a)–2(e), it can be seen that we cannot resolve the individual phonon peaks, such as the LA. However, we know the instrumental resolution, and in order to describe the line shape in, for example, Fig. 2(b), we require five peaks. Here, the separation of the peaks is comparable to the half-width-half-maximum resolution Δ_{HWHM} and σ_{res} . The fits can provide information about the underlying peak intensities and positions. Fits for all the O-K spectra presented in this article can be seen in Supplemental Material [29].

2. Three-peak model (Cu-L)

To fit the low-energy [−100, 200] meV Cu-L₃ spectra, a three-peak model was used. It consisted of a pseudovoigt peak to fit quasi-elastic scattering, and two resolution-limited Gaussians to fit ≈ 30 and ≈ 80 meV modes. This can be

written as

$$I_{\text{RIXS}}(H, \omega) = I^{\text{QE}}(H) \text{pV}(\omega) + \sum_{i=\{p_1, p_2\}} I^i(H) \times \exp \left[-\frac{(\omega - \omega_i)^2}{2\sigma_{\text{res}}^2} \right] + \text{B.G.}, \quad (2)$$

where i indexes the two identifiable (combined) phonon modes $p_1 = \text{LA} + B_{1g}$ and $p_2 = A_{1g} + \text{BS}$ and $\Delta_{\text{FWHM}} \approx 2.335\sigma_{\text{res}} \approx 37$ meV.

Given the phonon energies discussed in Sec. II D 1 for the O-K data, p_1 , the ≈ 30 meV mode is likely a mixture of the LA and B_{1g} buckling phonons, while the 80 meV p_2 mode is likely a mixture of the A_{1g} buckling and the bond-stretching (BS) phonon present in cuprates [25,30]. Example fits for $H = -0.29$ are plotted in Figs. 2(f)–2(j) and the H -dependent results (phonon energies and peak intensities) are displayed in Fig. 4. While a two-peak fit has been commonly used in the literature (i.e., Refs. [6,30]), adding a third significantly increases the fit quality, especially near Q_{CDW} . Three peak fits are used in more recent work, i.e., Refs. [31,32]. Additionally,

with the three-peak model, the energy of the BS phonon [Figs. 4(a)–4(e)] more closely resembles that measured with INS and inelastic x-ray scattering (IXS) (see, e.g., Refs. [26,27]), while with a two-peak fit, it is around 10 meV lower than IXS and INS [30]. Fits for all the Cu- L spectra presented in this article can be seen in Supplemental Material [29].

III. RESULTS

Color intensity $I_{\text{RIXS}}(H, \omega)$ maps of O- K and Cu- L_3 RIXS scattering are shown in Fig. 3. The most notable feature, for $x \leq 0.16$, is the strong scattering near $|H| = \delta$ and $E \approx 0$. This is due to the CDW and CDF scattering. A second strong feature in the Cu- L_3 RIXS is observed near $E \approx 70$ meV and $H \approx 0.4$ and is due to the oxygen bond-stretching phonon, while the O- K RIXS couples most strongly to a ≈ 50 meV oxygen bond-buckling mode. Inelastic features are more easily visualized in color maps with the quasielastic energy subtracted, which can be seen in Supplemental Material [29].

A. E -dependent fits of phonons

The models described in Secs. IID 1 and IID 2 were used to fit the phonon dispersions and intensities of the phonons. Example fits are shown in Fig. 2. In Figs. 4(a)–4(e) we plot the fitted phonon dispersions at both O- K and Cu- L_3 resonances for all measured samples. Here we describe the dispersion and the intensity variation of the modes phonon by phonon, starting from the BS phonon and going down in energy.

1. BS phonon

The BS mode is detected with both O- K and Cu- L_3 RIXS. Starting from the top of Figs. 4(a)–4(e), we see generally good agreement between O- K and Cu- L_3 (p_1 mode) with the ≈ 80 meV BS phonon. We have also plotted IXS and INS measurements of the BS phonon on similar sample compositions from Refs. [26,27]. We see there is good agreement for the overdoped samples. For the RIXS data, there appears to be a softening near $|H| \approx 0.3$ for $x = 0.11, 0.125$, and 0.16 [Figs. 4(a)–4(c)] suggesting the presence of charge excitations.

The fitted p_2 phonon intensity (that is, the height of the fitted resolution-limited Gaussian) of the BS + A_{1g} mode measured at Cu- L_3 are as red markers in Figs. 4(f)–4(j), while the O- K BS phonon intensities are plotted as red markers in Figs. 4(k)–4(o). The mode intensity measured at both edges shows an increasing intensity as H increases. This qualitatively fits with the $\propto \sin^2(\pi H)$ behavior of momentum dependence of the electron-phonon coupling, which the BS phonon in cuprates has been shown to obey [24].

2. A_{1g} and B_{1g}

The A_{1g} and B_{1g} buckling modes can be separated with O- K RIXS, and the energy dispersions are plotted in Figs. 4(a)–4(e). They show little dispersion with average energies of around 56 and 39 meV, respectively. The dispersion shows similar behavior between samples.

The O- K A_{1g} and B_{1g} phonon intensities are plotted as yellow and green markers, respectively, in Figs. 4(k)–4(o).

The A_{1g} mode shows a roughly constant intensity, and the B_{1g} mode (green) shows a peak near $|H| \approx 0.24$ for $x = 0.11, 0.125$, and 0.16 , indicating the presence of CDFs with $|H| \approx 0.24$ for these compositions. For the Cu- L data, the poorer resolution means that we see a combination of the LA and B_{1g} modes (denoted p_1). The (LA + B_{1g}) Cu- L data in Figs. 4(f)–4(j) shows a CDF contribution near $|H| \approx 0.24$, also seen at the O- K edge, $x = 0.11, 0.125$, and 0.16 . The data also suggest there are CDFs near $|H| \approx 0.3$ (see also Refs. [16,26,27]) for all compositions studied. These two contributions are highlighted by the shading in Figs. 4(f)–4(j).

3. LA phonon

The lowest energy mode in the five-peak model used to fit the O- K data is the LA phonon, which we have fixed to 14.5 meV. It is plotted as blue markers in Figs. 4(k)–4(o). Within the measured range of $0.1 < H < 0.3$, it has a weak dispersion that is consistent with other INS [28] and RIXS [11,28] measurements. At O- K , the LA phonon mode has a peak intensity due to CDFs at $H \approx -0.24$ for $0.11 \leq x \leq 0.19$. The peak is largest at $x = 0.125$.

In summary, our analysis demonstrates that the spectra can be largely understood in terms of coupling between CDFs and phonons [11,24]. The dynamic CDFs have two contributions: one peaked at $|H| \approx 0.24$ and a second peaked at $|H| \approx 0.3$.

B. Model for CDW and CDF components

Here, we define the nomenclature of the different types of CDW and CDFs that we use throughout the paper. As discussed in Sec. I, previous studies [4,8,12,16] have highlighted the multicomponent nature of the charge correlations in cuprates. Our O- K data with their good energy resolution contain information about the energy scales of the fluctuations associated with the individual components. Signatures of various components are contained in the H dependence of the phonon intensities $I^i(H)$, $i = \{\text{LA}, B_{1g}, A_{1g}, \text{BS}\}$ [see Eq. (1) and Figs. 4(k)–4(o)].

Our data suggest there are three components to the underlying charge correlations: (i) a quasielastic CDW contribution (4a-CDW) peaked at $H = \delta_{4a} \approx 0.24$ and $\omega = 0$; (ii) a broader inelastic CDF component (4a-CDF) peaked at $H = \delta_{4a} \approx 0.24$; and (iii) a broader inelastic CDF component (3a-CDF) peaked at $H = \delta_{3a} \approx 0.3$ (only accessible in H at the Cu- L resonance). We have denoted these 3a and 4a simply for ease of naming, because the period of the components is roughly three or four lattice units. The inelastic 3a-CDF and 4a-CDF components are not seen in $I^{A_{1g}}$ or I^{BS} , suggesting the energy scale of these components is less than 50 meV.

Due to the finite instrumental resolution, $I^{\text{QE}}(H)$ contains contributions from the CDW and CDF components. We model these as

$$I^{\text{QE}}(H) = I_{4a\text{-CDW}} \exp \left[-\frac{(H - \delta_{4a})^2}{2\sigma_{4a\text{-CDW}}^2} \right] + I_{4a\text{-CDF}}(\omega = 0) \exp \left[-\frac{(H - \delta_{4a})^2}{2\sigma_{4a\text{-CDF}}^2} \right] + \text{B.G.}, \quad (3)$$

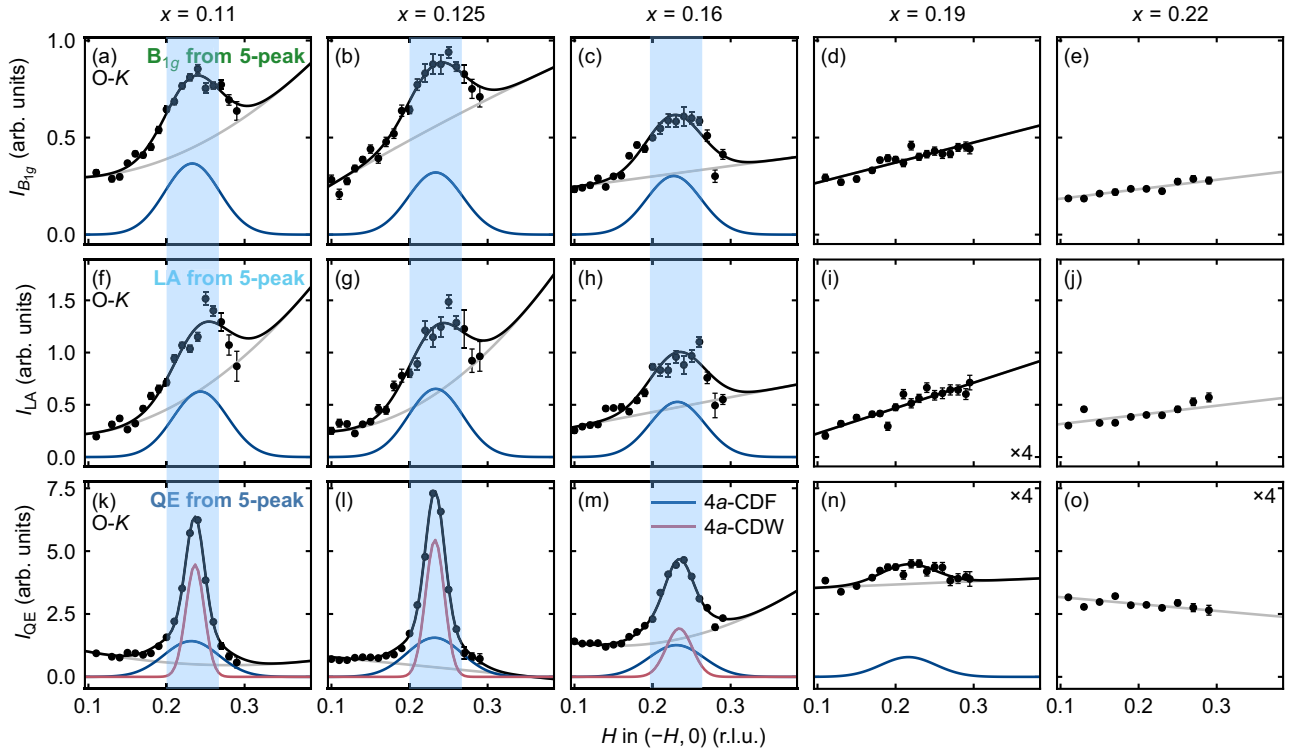


FIG. 5. Charge density fluctuations (CDFs) obtained from five-peak fit to phonons measured by O-K RIXS. H dependence of fitted phonon intensities, obtained from fitting the O-K RIXS spectra using the five-peak model described in the text. (a)–(e) B_{1g} phonons, (f)–(j) LA phonons. The H dependence of the quasi-elastic peak is plotted in panels (k)–(o). The total fit is plotted in black, with Gaussian components in blue or red, and the background is gray. Intensities in panels (i), (n), and (o) have been quadrupled for visibility. Blue shaded regions illustrate the lack of dispersion of the CDFs.

where $I_{4a\text{-CDW}}$ is the intensity of the $4a\text{-CDW}$ component and $I_{4a\text{-CDF}}(\omega = 0)$ is the intensity of the CDF component. The higher-energy data are modeled as

$$I^i(H) = I_{4a\text{-CDF}}(\omega_i) \exp\left[-\frac{(H - \delta_{4a})^2}{2\sigma_{4a\text{-CDF}}^2}\right] + I_{3a\text{-CDF}}(\omega_i) \exp\left[-\frac{(H - \delta_{3a})^2}{2\sigma_{3a\text{-CDF}}^2}\right] + \text{B.G.}, \quad (4)$$

where $I_{4a\text{-CDF}}(\omega_i)$ is measured at a phonon with frequency ω_i and the $I_{3a\text{-CDF}}(\omega_i)$ component can only be measured at the Cu- L_3 edge.

C. CDW and CDF at the O-K resonance

If we consider the $x = 0.125$ sample as an example of our data. Figure 5(l) shows a peak near $|H| \approx 0.24$ for $I^{\text{QE}}(H)$. Peaks are also seen for the LA and B_{1g} phonons in Figs. 4(l) and 5(b) and 5(g), but not obviously for the A_{1g} and BS modes. The peak width of the LA and B_{1g} phonons is larger than that of the quasi-elastic ($E = 0$) scattering. Thus, our O-K edge data can be understood using the first two terms of Eq. (3) representing a narrower QE CDW component with $\sigma_{4a\text{-CDW}} \approx 0.01$ r.l.u. ($\xi \approx 35$ Å) and a broader inelastic CDF component seen mostly in the LA and B_{1g} phonons, with $\sigma_{4a\text{-CDF}} \approx 0.04$ r.l.u. ($\xi \approx 15$ Å). Our results differ from some previous measurements at the Cu- L edge. For example, in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ [25] the data look dispersive and are

fitted as dispersive CDFs. In contrast, in Figs. 5(b), 5(g), and 5(l), the excitations do not appear to disperse strongly and the CDW and CDFs are centered on the same wave vector.

The result of fitting Eqs. (3) and (4) to the data in Figs. 4 and 5 is shown in Figs. 7(c)–7(e). The narrow $4a\text{-CDW}$ QE component is only observed from $x = 0.11$ to $x = 0.16$. The $x = 0.16$ quasi-elastic is best fit to a combination of both components, Fig. 5(m). This was done by fixing the width of the broad component at the average value of $\sigma = 0.04$ r.l.u. and allowing the narrow component to vary. This is to reduce the number of fitting parameters when the CDW contribution is weak. The $4a\text{-CDF}$ is no longer detected in the $x = 0.22$ quasi-elastic channel.

D. CDW and CDF at Cu- L_3 resonance

RIXS measurements at the Cu- L_3 resonance provide complementary information to those made at the O-K resonance: they are sensitive to phonons in a different way, and the higher energy of the Cu- L_3 resonance means that measurements can be made to larger $|H| \approx 0.4$. However, the energy resolution $\Delta E \approx 37$ meV is worse than at the O-K resonance; therefore, the phonons cannot be isolated in the same way as for the O-K data. For simplicity, we integrate our data over an energy range corresponding to the experimental resolution and fit to the resulting points. Figure 6 shows H -dependent cuts integrated over energy ranges $[-18, 18]$ meV, and $[18, 54]$ meV.

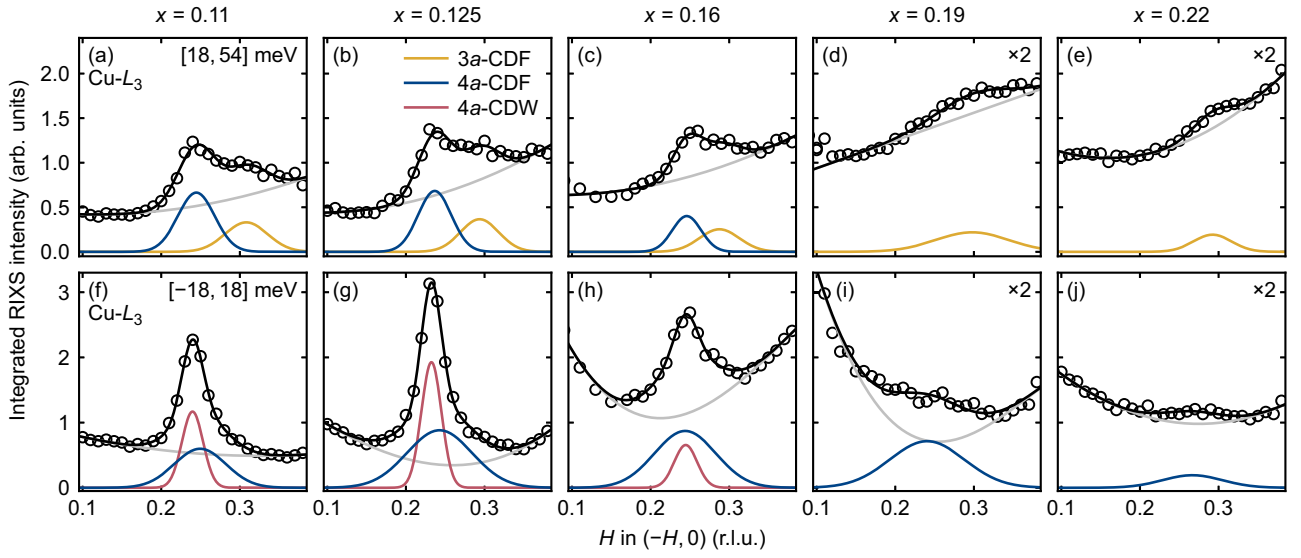


FIG. 6. Charge density fluctuations measured with Cu- L_3 RIXS. (a)–(e) [18, 54] meV Cu- L_3 RIXS cut. (f)–(j) Cu- L_3 quasi-elastic RIXS cut, obtained by integrating RIXS in $[-18, 18]$ meV. Data have been fit to either a sum of two Gaussians and a polynomial background or one Gaussian and a polynomial background. The total fit is plotted in a black line, and fit components are plotted in colored lines. Intensities in panels (d), (e), (i), and (j) have been doubled for visibility.

The cuts are consistent with those obtained by the three-peak fits shown in Figs. 4(f)–4(j).

The Cu- L_3 quasi-elastic $E \in [-18, 18]$ meV cuts for the $x = 0.11, 0.125$, and 0.16 can be fitted to the sum of Gaussian

peaks and a second-order polynomial background [Figs. 6(f) and 6(g)]. As with the two-dimensional (2D) O- K fits, one peak is broader than the other ($\sigma_{\text{CDF}} \sim 0.04$ r.l.u. compared with $\sigma_{\text{CDW}} \sim 0.01$ r.l.u.). Again, we identify the broad and

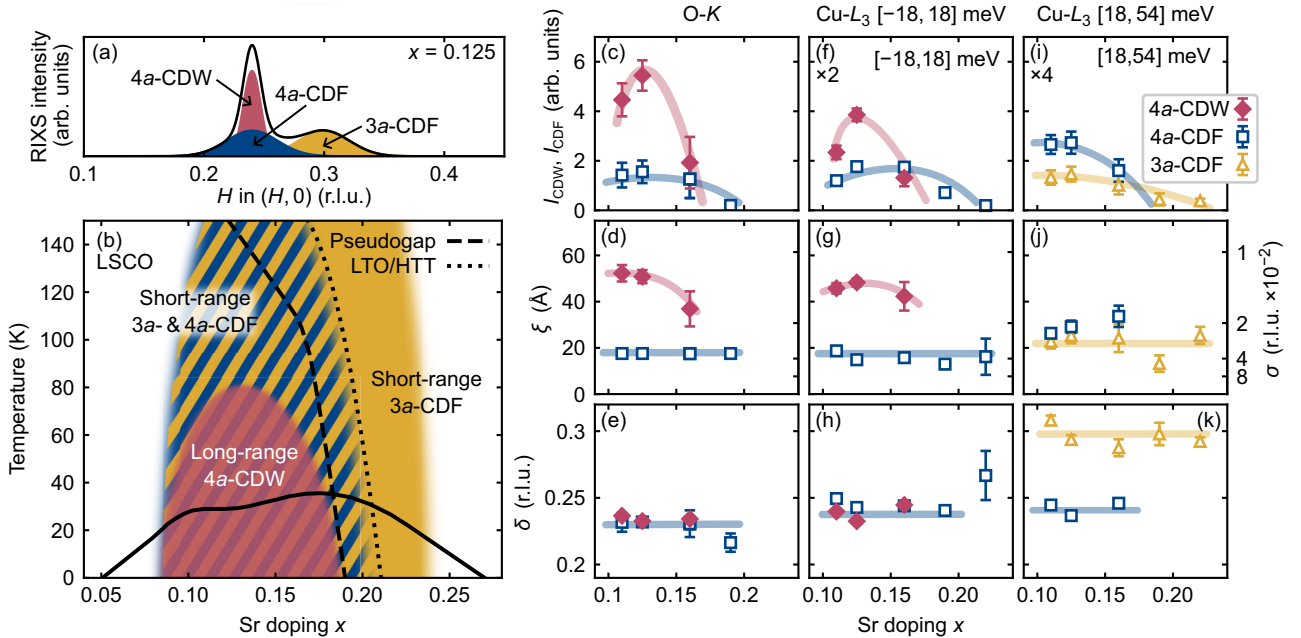


FIG. 7. CDW phase diagram in LSCO. (a) Schematic in RIXS scattering intensity and H of the three CDW components. (b) Schematic phase diagram, with regions where $4a$ -CDW order (red), $4a$ -CDF (blue) $3a$ -CDF (yellow) are present. The superconducting dome is plotted in a black solid line. The pseudogap boundary is marked by the black dashed line, which is based on Nernst effect measurements [33]. The boundary between the low-temperature orthogonal (LTO) and high-temperature tetragonal phase (HTT), is marked with a black dotted line, based on data from Ref. [34]. (c), (f), (i) Fitted peak intensities (Gaussian height); (d), (g), (j) correlation length ξ ; and (e), (h), (k) CDW incommensurability δ for the one-dimensional (1D) O- K phonon fit, the 1D $[-18, 18]$ meV Cu- L_3 cut, and the 1D [18, 54] meV Cu- L_3 cut, respectively. The secondary y axis in panels (d), (g), (j) is the fitted Gaussian width parameter σ and is related to the correlation length by $\xi = 1/\sigma$. There are two $4a$ -CDF markers at $x = 0.16$ in panels (c)–(e) because there are both a broad QE component and a broad LA fit present at this doping.

narrow components centered at $H \approx -0.24$, with the $4a$ -CDF and $4a$ -CDW contributions, respectively. For $x = 0.16$, we fix $\sigma_{\text{CDF}} = 0.04$ r.l.u., as with the O- K data. For $x = 0.19$ we fit one broad peak [Figs. 6(h) and 6(i)]. A weak peak centered at $H \approx -0.27$ is seen for $x = 0.22$ [Fig. 6(j)]. A summary of the fitted values for the quasi-elastic cut is plotted in Figs. 7(f)–7(h).

The finite energy $E \in [18, 54]$ meV cut at Cu- L_3 which probes the CDF component is plotted in Figs. 6(a)–6(e). It is possible to access a wider range in H at the Cu- L_3 resonance compared with O- K , we observe scattering centered at $H \approx -0.29$ in the $x = 0.22$ sample. For $x \leq 0.16$, there is a shoulder of scattering in addition to the $H \approx -0.24$ scattering from the $4a$ CDW or CDF. Here we choose to interpret the scattering as the sum of two peaks with at two distinct H values.

We apply a two-Gaussian fit to the $0.11 \leq x \leq 0.19$ [18, 54] meV cut. Using what we know from the O- K data, that the center of the $4a$ CDF is similar to the $4a$ -CDW, we fix one of the CDF peaks to the center of the $H \approx -0.24$ CDW measured in the quasi-elastic cut. The other peak was allowed to float. The result is a second peak centered at $H \approx -0.29$ in all the measured samples—we call this “ $3a$ -CDF” since $\lambda \approx a/0.29 \approx 3.4a$. The $3a$ CDF peak has a width $\sigma_{3a\text{-CDF}} \approx 0.04$ r.l.u., which is comparable with $\sigma_{4a\text{-CDF}}$ and approximately constant with doping x . A summary of the fitted values for the inelastic cuts is plotted in Figs. 7(i)–7(k). Note that the $3a$ -CDF can only be clearly observed at the Cu- L resonance because the large energy allows $H > 0.3$ to be easily reached.

IV. EVOLUTION OF CHARGE ORDER AND CHARGE FLUCTUATIONS WITH DOPING

A. Three components

In this work, we have identified three components contributing to the charge correlations [see schematic in Fig. 7(a) for how these appear in an H scan]. The first is longer-range $4a$ -CDW order with $\mathbf{Q} = (\delta, 0)$ and $\delta \approx 0.24$. This is elastic ($E = 0$) within the resolution of the present experiment; however, this component may fluctuate on an energy scale $\Delta E \lesssim 1$ meV, as discussed in Refs. [35,36]. Our measurements at both the O- K and Cu- L_3 resonances suggest that the $4a$ -CDW disappears at about $x = 0.16$ and that for dopings $x = 1/8$ (where it is strongest) it has a correlation length $\xi \approx 50$ Å. Our results are broadly in agreement with previous studies, which report an increase below ξ below $T_{\text{CDW}} \approx 80$ K signaling the onset of the CDW [8,9,14]. The second component ($4a$ -CDF) corresponds to shorter-range correlations that also occur with $\delta \approx 0.24$. In Sec. III C we show that the $4a$ -CDF component is dynamic and has the highest intensity for $\hbar\omega_{\text{LA}} \approx 14.5$ meV. The third component ($3a$ -CDF) is seen in the Cu- L_3 RIXS shown in Figs. 6(a)–6(e) is CDFs with $\delta \approx 0.29$, that is seen in the energy range [18, 54] meV.

B. Phase diagram

Figure 7(b) shows a sketch of the regions of x - T space where the three contributions to the charge correlations are strong. This is based on present and literature data. The $4a$ -CDW component exists in the range $0.1 < x < 0.16$,

is strongest at $x = 0.125$, and shows a strong temperature dependence. The temperature where the CDW emerges is approximately 80 K at $x = 0.125$ [8,9,14,15]. $4a$ -CDFs in LSCO exist to at least 150 K when measured with x-ray diffraction (XRD) [8], Cu- L_3 RIXS [16] and at 210 K in O- K RIXS [11]. For $0.19 < x < 0.22$, the $4a$ -CDF is no longer detected in our data, while the $3a$ -CDF, which may have a higher energy scale, is observed over the range $x \in [0.11, 0.22]$.

C. Intensities, correlation lengths, and characteristic wave vectors

Figures 7(c)–7(k) shows the peak intensity (i.e., Gaussian height), correlation length ξ and incommensurability δ describing the three components of the charge correlations described above. We see [Figs. 7(c)–7(f)] that both the $4a$ -CDW and $4a$ -CDF, represented by red diamonds and blue squares, are strong at $x = 0.125$. The $4a$ -CDW component disappears between $x = 0.16$ and 0.19 , while the $4a$ -CDF disappears between $x = 0.19$ and 0.22 . The $3a$ -CDF, represented by yellow triangles, exists over all measured doping values, and extrapolation suggests that it would disappear at $x \approx 0.25$. $x = 0.25$ is also the doping value where an anomaly in the BS phonon disappears.

The fitted correlation lengths are plotted in Figs. 7(d), 7(g), and 7(j). The correlation lengths of the $3a$ - and $4a$ -CDFs are relatively doping independent, with similar values of around $\xi \approx 20$ Å obtained for the two components. The correlation length of the $4a$ -CDW, is maximized around $x = 0.125$ at $\xi \approx 50$ Å and it decreases slightly as doping is increased toward $x = 0.16$. We find that our $\xi_{4a\text{-CDW}}$ is larger than that found [8] using hard x-rays ($\xi_{\text{x-ray}} \approx 30$ Å), this is most likely due to the multicomponent model used here which separates the CDW contribution of the CDFs.

The CDW incommensurability δ of the three components is plotted in Figs. 7(e), 7(h), and 7(k). We find the δ for the $4a$ CDF and CDW (blue and red points) are constant with doping with $\delta = 0.24$. The $3a$ CDF (yellow points) are centered consistently on $\delta = 0.29$.

D. Energy scales

A summary of the doping evolution of three components in (H , E) of our observations is shown in Fig. 8. The $4a$ -CDF component gives rise to intensity peaks in the 14.5 meV LA and the 39 meV B_{1g} phonon modes, but is not seen significantly through the 56 meV A_{1g} mode. The $4a$ -CDF gives intensity peaks in phonons of similar energies in the Cu- L data. However, the poorer resolution means that the LA and B_{1g} phonons cannot be resolved.

Figures 9(a)–9(c) plot the intensities $4a$ -CDFs seen through the phonons together with the QE scattering measured at O- K resonance. If we assume that these intensities are proportional to the dynamic structure factor of the charge fluctuations $S(\mathbf{Q}, \omega)$ then we can estimate the energy scale of the CDFs. This analysis assumes that I_{RIXS} is equally sensitive to $S(\mathbf{Q}, \omega)$ when measured at different oxygen phonon modes. Calculations in Ref. [24] suggest this is approximately true.

In Figs. 9(a)–9(c) we fit a very overdamped harmonic-oscillator response $\chi''(\mathbf{Q}, \omega) \propto \omega\Gamma/(\omega^2 + \Gamma^2)$ times a Bose

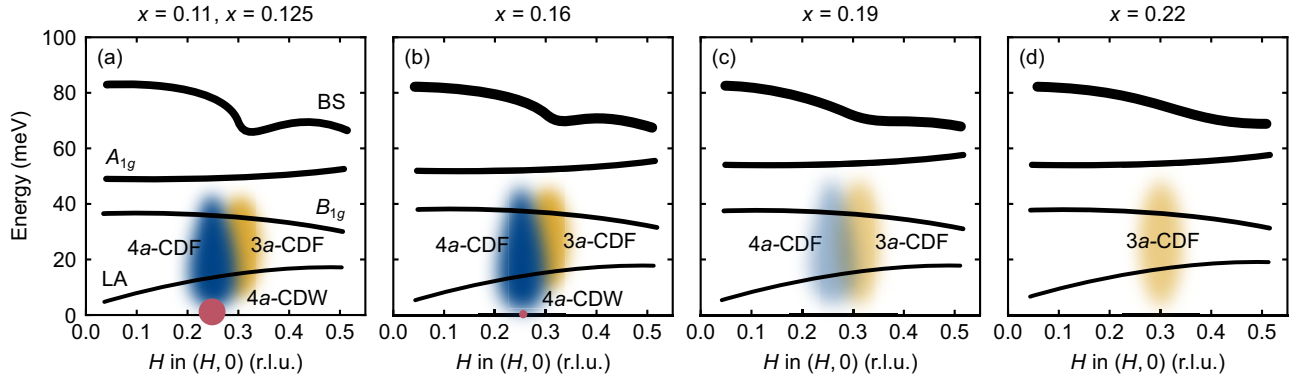


FIG. 8. Schematic doping-dependent charge fluctuations in LSCO. Doping-dependent (H, E) schematic of charge fluctuations. (a) Underdoped compositions, where $4a$ -CDW is maximized (red circle), and the $3a$ - and $4a$ -CDF (yellow, blue regions) are detected. (b) In optimally doped compositions, the $4a$ -CDW is reduced in intensity, and the $3a$ - and $4a$ -CDF are still present. (c) In overdoped compositions, $4a$ -CDW is no longer present. (d) For the most overdoped composition only a weak $3a$ -CDF remains.

factor (see Sec. II D) convolved with the instrumental resolution to the $I_{4a\text{-CDF}}(\omega_i)$ values. The resulting fit yield a doping-independent relaxation rate $\hbar\Gamma \approx 11$ meV for $x \in [0.11, 0.16]$, where there is a significant $4a$ -CDF signal.

V. RELATIONSHIP TO OTHER EXPERIMENTS

A. RIXS on cuprate superconductors

Our results agree broadly with other x-ray studies on LSCO [8,14–16,30,37], which also reveal the multicomponent nature of the charge correlations. For example, a recent RIXS study of LSCO at the Cu- L resonance [16] has characterized the disappearance of the charge correlations with doping in terms of decreasing correlation length. We conclude that the $4a$ -CDW order disappears near $x \approx 0.16$, which is in agreement with this study [16] and in disagreement with Ref. [15] which suggests that the $4a$ -CDW order persists to $x = 0.21$. Our work at the O- K resonance contrasts with the Cu- L RIXS [16,30] and diffraction [8,14,15] in that we are able to separate three components using good energy resolution, showing that the broader component is inelastic. Our work is in agreement with very high-resolution O- K RIXS experiments on LSCO ($x = 0.15$) [11] which find evidence for RIXS excitations from due

to coupling of CDW fluctuations to acoustic phonons peaked near 14 meV for $H = \delta$.

Signals of an inelastic CDFs are also seen with Cu- L_3 RIXS in Hg1210 [13] and YBCO [12,31]. In YBCO [31], the wave vectors are different; however, at low temperatures, CDFs are seen with $\hbar\Gamma = 10$ –15 meV for $p = 0.17$ –0.19, i.e., comparable to the values $\hbar\Gamma \approx 11$ meV reported here for LSCO. As mentioned earlier, our results differ from previous measurements at the Cu- L edge. In $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ [25] the data look dispersive and are fitted this dispersive CDFs. In contrast, in LSCO the $4a$ -CDFs do not appear to disperse strongly and have the same wave vector as the $4a$ -CDW.

B. Phonon anomalies in cuprates

In LSCO, there is a softening and broadening of the 80 meV BS phonon at $H \approx 0.3$ [26,27]. The phonon anomaly exists from around $0.05 \lesssim x \lesssim 0.25$. The anomaly is temperature independent up to 100 K [30]. There are a number of candidates to explain the softening, given in Ref. [27]. It is likely that the $3a$ -CDF we observe is related to the BS phonon anomaly, given both the similarity of the wave vector and its variation with doping.

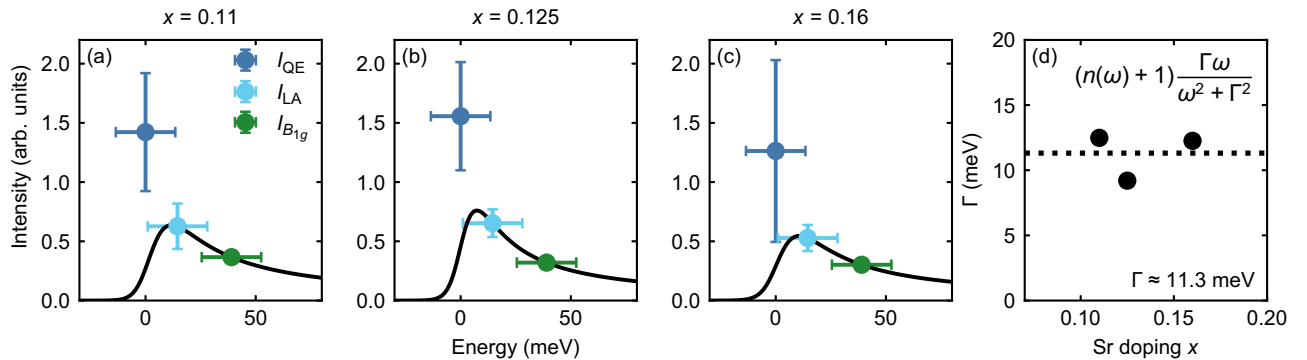


FIG. 9. Energy scales of $4a$ CDFs. (a)–(c) Fitted peak intensities from the five-peak fitting procedure for the $4a$ CDFs. The nonzero $4a$ CDF intensities, i.e., the quasielastic (QE) and LA and B_{1g} phonons, have been fitted to Lorentzian relaxation form given in Sec. IV D. Horizontal errorbars correspond to the half-width half-maximum (HWHM) energy resolution of 13.5 meV. (d) The fitted relaxation rate Γ for the three doping values, which have an average value of 11.3 meV.

C. STM on Bi2212 and the pseudogap

Spectroscopic imaging scanning tunneling microscopy (SI-STM) measures correlations in the local charge density of states (LDOS). In $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) the LDOS correlations are strongest around $p = 0.125$ and weaken as doping is increased, and are no longer detected beyond $x = 0.19$ [38]. The disappearance is accompanied by signatures of the closing of the pseudogap [38]. The correlation length of these fluctuations [2], i.e., when converted to $\xi = 1/\sigma$, is around $15 \text{ \AA}$ when measured at T_c [2,39]. When measured with RIXS, the correlation length is slightly longer at around $20 \text{ \AA}$ [2,6]. The comparison with Bi2212 suggests that the disappearance of the $4a$ -CDF in LSCO near $p = p^*$ is associated with the collapse of the pseudogap.

D. Neutron scattering and low-energy spin fluctuations

The present work suggests the presence of charge density fluctuations (CDF) with relatively low energies $\hbar\Gamma \approx 11 \text{ meV}$ across a large portion of the cuprate phase diagram. In this section we compare with the collective spin fluctuations. It is well known that the spin fluctuations in cuprates have several components that evolve with doping and have been extensively characterized. In the normal state of LSCO, for temperatures $T \approx T_c$ (i.e., comparable with the present measurements), there are strong incommensurate low-energy spin fluctuations (SF) with characteristic energies $\hbar\Gamma \approx 5\text{--}10 \text{ meV}$ according to doping [40,41]. The SF are strongly temperature dependent and have been shown to exist into the overdoped region ($x = 0.22$) where strange metal behavior occurs and are labeled as “critical spin fluctuations” [42]. The wave vectors $\mathbf{Q}_{\text{SF}}$ of the SFs are geometrically related to those of the $4a$ -CDF: $\mathbf{Q}_{\text{SF}} = (1/2 \pm \delta/2, 1/2)$ and $(1/2, 1/2 \pm \delta/2)$. It is also interesting to note that the SF and $4a$ -CDFs have similar correlation lengths with $\xi_{\text{SF}} = 20\text{--}25 \text{ \AA}$.

The fact that the SFs and $4a$ -CDFs have geometrically related wave vectors, similar correlation lengths and similar characteristic energies is consistent with a fluctuating stripe picture [43,44]. The stripe picture postulates an entity with intertwined spin and charge modulations. However, it should be noted that the incommensurate spin fluctuations persist to higher dopings $x = 0.22$, where the $4a$ -CDFs have disappeared at the sensitivity of the RIXS experiments described here.

VI. DISCUSSION

A. Multicomponent nature of the charge correlations

Calculating the charge density wave order and charge density fluctuations in cuprate superconductors is complex. The correlations emerge from the multi-orbital Cu-O electronic structure with the oxygen degrees of freedom playing a critical role [45–47]. We may also need to take into account the coupling to spin fluctuations, hot spots, phonons, material-specific physics, and other factors. To our knowledge, theory does not yet quantitatively predict the δ_{3a} and δ_{4a} CDFs and CDW observed here. However, multi-orbital calculations do predict [47] different charge susceptibilities at the Cu and O atoms and $\delta_{\text{charge}} \approx 0.3$. Experimentally, for LSCO there is evidence of charge correlations with $\delta_{4a} \approx 0.24$ from hard

x-ray diffraction and RIXS [8,9,14–16]. The $\delta_{3a} \approx 0.3$ correlations are seen in this work at the Cu- L RIXS as a (side-)peak in the intensity and a dip in the energy [Fig. 4(b)] of the “BS + A_{1g} ” phonon. The frequency of this phonon aligns [see Figs. 4(a)–4(e)] with the BS phonon suggesting that we are primarily seeing this. This is consistent with inelastic neutron scattering and (nonresonant) inelastic x-ray scattering [27] studies of the BS phonon in LSCO that show a dip in the phonon energy and a peak in the phonon width (damping) at $h \approx 0.3$. We expect the BS phonon to be particularly sensitive to oxygen charge susceptibility. This suggests the $3a$ -CDF is primarily associated with the oxygen site.

B. Critical fluctuations across the cuprate phase diagram

The possibility of a quantum critical point at the critical pseudogap doping p^* has been discussed in many papers [31,38,48]. If charge density fluctuations were closely associated with order-parameter fluctuations, a diverging correlation length and correlation time would be expected. Our observations do not support this. Instead, we find the correlation length [Figs. 7(d), 7(g), and 7(j)] and relaxation rate [Fig. 9(d)] are approximately constant with doping for $x \leq 0.19$ with $\xi_{4a\text{-CDF}} \approx 20 \text{ \AA}$ and $\hbar\Gamma_{4a\text{-CDF}} \approx 11 \text{ meV}$. In contrast, the relaxation rate associated with low-energy charge fluctuations decreases rapidly with temperature [11] for $x = 0.15$. Analogous behavior has been observed in the low-energy spin fluctuations in LSCO, where low-frequency ($5\text{--}10 \text{ meV}$) fluctuations are observed at low temperatures ($T \sim T_c$), and these fluctuations appear to be “critical” across the phase diagram.

The extended region of criticality is consistent with a scenario [42] in which the cuprate phase diagram is controlled by a quantum Griffiths phase. A Griffiths phase occurs near a continuous phase transition in systems with quenched disorder. The inclusion of disorder is logical in the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ system because of the perturbation caused by Sr doping in the plane neighboring the CuO_2 plane. The Griffiths phase corresponds to a smearing of singular behavior as a function of a control parameter. This could naturally lead to slow dynamics and low-frequency relaxation of the collective spin and charge excitations across the phase diagram in LSCO rather than a well-defined quantum critical point.

C. Linear resistivity and Planckian dissipation

The present study suggests that the $4a$ -CDW and the $4a$ -CDFs disappear (or become too weak to observe) for dopings above $x \gtrsim x^* \approx 0.19$. In contrast, the strongly T -dependent low-energy spin fluctuations with a geometrically related wave vector ($\delta_{\text{SF}} = \frac{1}{2}\delta_{4a\text{-CDF}}$) are easily observable at higher dopings, i.e., $x = 0.22$ [41,42]. It is intriguing that in the overdoped region $x \gtrsim 0.19$ where a very clear T -linear resistivity (“Planckian dissipation”) [18,49] only T -dependent low-energy spin fluctuations have been observed to date and not the corresponding charge fluctuations.

VII. SUMMARY

In this work we have measured Cu- L_3 and O- K RIXS in LSCO over $0.11 < x < 0.22$. The observed excitations are

interpreted using a model in which charge excitations are coupled to phonons. We find three components ($4a$ -CDW, $4a$ -CDF, $3a$ -CDF) in (H, E) -space (see Sec. IV A), each has a distinct doping evolution. Our data also allows us to comment on the structure of the CDF in E .

The first component, the $4a$ -CDW, has an ordering wave vector $\delta \approx 0.24$, is static within energy resolution, and has maximum intensity for $x \approx 0.125$ doping. It has a correlation length of up to $\xi \approx 50$ Å and is not detected for $x > 0.16$. The second component are the $4a$ -CDFs, which have the same wave vector as the $4a$ -CDW, but a shorter correlation length of around 20 Å. The $4a$ -CDFs couple most strongly to the LA phonon, and we estimate their energy scale is ≈ 11 meV. The $4a$ -CDF are strongest at $x \approx 0.125$, are no longer detected beyond $x = 0.19$, and therefore may be associated with the closing of the pseudogap. The energy scale and correlation length of the $4a$ -CDF are approximately constant with doping. The third component are the $3a$ -CDF, which are also dynamic and have wave vector $\delta \approx 0.29$. The $3a$ -CDF exist from $x = 0.11$ to $x = 0.22$ and show a gradual suppression of intensity with doping. This component can only be seen at the Cu- L edge and may have a higher energy scale of 30–80 meV because it is seen in a range of phonons.

Our results show that the charge correlations in LSCO have a more complex structure in H - E than previously thought, with ordering tendencies at two different Q values. Each CDW or CDF component shows a distinct evolution with doping. We find that the $4a$ -CDF component disappears at approximately the same doping as the pseudogap. However, there is no evidence of charge fluctuations associated with a quantum critical point.

ACKNOWLEDGMENTS

This work was supported by the UK EPSRC under grant EP/R011141/1. We thank the Diamond Light Source for providing beam time under proposal ID MM29150. C.C.T. acknowledges funding from Diamond Light Source and the University of Bristol under joint doctoral studentship STU0372. We thank Seamus Davis for fruitful discussions.

DATA AVAILABILITY

The data that support the findings of this article are openly available [50].

-
- [1] B. Keimer, S. A. Kivelson, M. R. Norman, S. Uchida, and J. Zaanen, From quantum matter to high-temperature superconductivity in copper oxides, *Nature (London)* **518**, 179 (2015).
 - [2] S. M. Hayden and J. M. Tranquada, Charge correlations in cuprate superconductors, *Annu. Rev. Condens. Matter Phys.* **15**, 215 (2024).
 - [3] P. Monceau, Electronic crystals: An experimental overview, *Adv. Phys.* **61**, 325 (2012).
 - [4] G. Ghiringhelli, M. Le Tacon, M. Minola, S. Blanco-Canosa, C. Mazzoli, N. B. Brookes, G. M. De Luca, A. Frano, D. G. Hawthorn, F. He, T. Loew, M. M. Sala, D. C. Peets, M. Salluzzo, E. Schierle, R. Sutarto, G. A. Sawatzky, E. Weschke, B. Keimer, and L. Braicovich, Long-range incommensurate charge fluctuations in $(Y, Nd)BaCu_3O_{6\delta}$, *Science* **337**, 821 (2012).
 - [5] W. Tabis, B. Yu, I. Bialo, M. Bluschke, T. Kolodziej, A. Kozłowski, E. Blackburn, K. Sen, E. M. Forgan, M. v. Zimmermann, Y. Tang, E. Weschke, B. Vignolle, M. Hepting, H. Gretarsson, R. Sutarto, F. He, M. Le Tacon, N. Barišić, G. Yu, *et al.*, Synchrotron x-ray scattering study of charge-density-wave order in $HgBa_2CuO_{4+\delta}$, *Phys. Rev. B* **96**, 134510 (2017).
 - [6] W. S. Lee, K.-J. Zhou, M. Hepting, J. Li, A. Nag, A. C. Walters, M. Garcia-Fernandez, H. C. Robarts, M. Hashimoto, H. Lu, B. Nosarzewski, D. Song, H. Eisaki, Z. X. Shen, B. Moritz, J. Zaanen, and T. P. Devereaux, Spectroscopic fingerprint of charge order melting driven by quantum fluctuations in a cuprate, *Nat. Phys.* **17**, 53 (2021).
 - [7] J. Chang, E. Blackburn, A. T. Holmes, N. B. Christensen, J. Larsen, J. Mesot, R. Liang, D. A. Bonn, W. N. Hardy, A. Watenphul, M. v. Zimmermann, E. M. Forgan, and S. M. Hayden, Direct observation of competition between superconductivity and charge density wave order in $YBa_2Cu_3O_{6.67}$, *Nat. Phys.* **8**, 871 (2012).
 - [8] T. P. Croft, C. Lester, M. S. Senn, A. Bombardi, and S. M. Hayden, Charge density wave fluctuations in $La_{2-x}Sr_xCuO_4$ and their competition with superconductivity, *Phys. Rev. B* **89**, 224513 (2014).
 - [9] V. Thampy, M. P. M. Dean, N. B. Christensen, L. Steinke, Z. Islam, M. Oda, M. Ido, N. Momono, S. B. Wilkins, and J. P. Hill, Rotated stripe order and its competition with superconductivity in $La_{1.88}Sr_{0.12}CuO_4$, *Phys. Rev. B* **90**, 100510(R) (2014).
 - [10] C. C. Tam, M. Zhu, J. Ayres, K. Kummer, F. Yakhov-Harris, J. R. Cooper, A. Carrington, and S. M. Hayden, Charge density waves and Fermi surface reconstruction in the clean overdoped cuprate superconductor $Tl_2Ba_2CuO_{6+\delta}$, *Nat. Commun.* **13**, 570 (2022).
 - [11] H. Y. Huang, A. Singh, C. Y. Mou, S. Johnston, A. F. Kemper, J. van den Brink, P. J. Chen, T. K. Lee, J. Okamoto, Y. Y. Chu, J. H. Li, S. Komiya, A. Komarek, A. C. Fujimori, C. T. Chen, and D. J. Huang, Quantum fluctuations of charge order induce phonon softening in a superconducting cuprate, *Phys. Rev. X* **11**, 041038 (2021).
 - [12] R. Arpaia, S. Caprara, R. Fumagalli, G. De Vecchi, Y. Y. Peng, E. Andersson, D. Betto, G. M. De Luca, N. B. Brookes, F. Lombardi, M. Salluzzo, L. Braicovich, C. Di Castro, M. Grilli, and G. Ghiringhelli, Dynamical charge density fluctuations pervading the phase diagram of a Cu-based high- T_c superconductor, *Science* **365**, 906 (2019).
 - [13] B. Yu, W. Tabis, I. Bialo, F. Yakhov, N. B. Brookes, Z. Anderson, Y. Tang, G. Yu, and M. Greven, Unusual dynamic charge correlations in simple-tetragonal $HgBa_2CuO_{4+\delta}$, *Phys. Rev. X* **10**, 021059 (2020).
 - [14] J.-J. Wen, H. Huang, S.-J. Lee, H. Jang, J. Knight, Y. S. Lee, M. Fujita, K. M. Suzuki, S. Asano, S. A. Kivelson, C.-C. Kao, and J.-S. Lee, Observation of two types of charge-density-wave

- orders in superconducting $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, *Nat. Commun.* **10**, 3269 (2019).
- [15] H. Miao, G. Fabbri, R. J. Koch, D. G. Mazzone, C. S. Nelson, R. Acevedo-Esteves, G. D. Gu, Y. Li, T. Yilmaz, K. Kaznatcheev, E. Vescovo, M. Oda, T. Kurosawa, N. Momono, T. Assefa, I. K. Robinson, E. S. Bozin, J. M. Tranquada, P. D. Johnson, and M. P. M. Dean, Charge density waves in cuprate superconductors beyond the critical doping, *npj Quantum Mater.* **6**, 31 (2021).
 - [16] K. von Arx, Q. Wang, S. Mustafi, D. G. Mazzone, M. Horio, D. J. Mukkattukavil, E. Pomjakushina, S. Pyon, T. Takayama, H. Takagi, T. Kurosawa, N. Momono, M. Oda, N. B. Brookes, D. Betto, W. Zhang, T. C. Asmara, Y. Tseng, T. Schmitt, Y. Sassa, *et al.*, Fate of charge order in overdoped La-based cuprates, *npj Quantum Mater.* **8**, 7 (2023).
 - [17] B. Vignolle, S. M. Hayden, D. F. McMorrow, H. M. Rønnow, B. Lake, C. D. Frost, and T. G. Perring, Two energy scales in the spin excitations of the high-temperature superconductor $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, *Nat. Phys.* **3**, 163 (2007).
 - [18] R. A. Cooper, Y. Wang, B. Vignolle, O. J. Lipscombe, S. M. Hayden, Y. Tanabe, T. Adachi, Y. Koike, M. Nohara, H. Takagi, C. Proust, and N. E. Hussey, Anomalous criticality in the electrical resistivity of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, *Science* **323**, 603 (2009).
 - [19] K.-J. Zhou, A. Walters, M. Garcia-Fernandez, T. Rice, M. Hand, A. Nag, J. Li, S. Agrestini, P. Garland, H. Wang, S. Alcock, I. Nistea, B. Nutter, N. Rubies, G. Knap, M. Gaughran, F. Yuan, P. Chang, J. Emmins, and G. Howell, I21: An advanced high-resolution resonant inelastic x-ray scattering beamline at diamond light source, *J. Synchrotron Radiat.* **29**, 563 (2022).
 - [20] J. Zaanen, G. A. Sawatzky, and J. W. Allen, Band gaps and electronic structure of transition-metal compounds, *Phys. Rev. Lett.* **55**, 418 (1985).
 - [21] F. C. Zhang and T. M. Rice, Effective Hamiltonian for the superconducting Cu oxides, *Phys. Rev. B* **37**, 3759 (1988).
 - [22] C. T. Chen, F. Sette, Y. Ma, M. S. Hybertsen, E. B. Stechel, W. M. C. Foulkes, M. Schuler, S.-W. Cheong, A. S. Cooper, L. W. Rupp, B. Batlogg, Y. L. Soo, Z. H. Ming, A. Krol, and Y. H. Kao, Electronic states in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4+\delta}$ probed by soft-x-ray absorption, *Phys. Rev. Lett.* **66**, 104 (1991).
 - [23] L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, and J. van den Brink, Resonant inelastic x-ray scattering studies of elementary excitations, *Rev. Mod. Phys.* **83**, 705 (2011).
 - [24] T. P. Devereaux, A. M. Shvaika, K. Wu, K. Wohlfeld, C. J. Jia, Y. Wang, B. Moritz, L. Chaix, W.-S. Lee, Z.-X. Shen, G. Ghiringhelli, and L. Braicovich, Directly characterizing the relative strength and momentum dependence of electron-phonon coupling using resonant inelastic x-ray scattering, *Phys. Rev. X* **6**, 041019 (2016).
 - [25] L. Chaix, G. Ghiringhelli, Y. Y. Peng, M. Hashimoto, B. Moritz, K. Kummer, N. B. Brookes, Y. He, S. Chen, S. Ishida, Y. Yoshida, H. Eisaki, M. Salluzzo, L. Braicovich, Z.-X. Shen, T. P. Devereaux, and W.-S. Lee, Dispersive charge density wave excitations in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$, *Nat. Phys.* **13**, 952 (2017).
 - [26] T. Fukuda, J. Mizuki, K. Ikeuchi, K. Yamada, A. Q. R. Baron, and S. Tsutsui, Doping dependence of softening in the bond-stretching phonon mode of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($0 \leq x \leq 0.29$), *Phys. Rev. B* **71**, 060501(R) (2005).
 - [27] S. R. Park, T. Fukuda, A. Hamann, D. Lamago, L. Pintschovius, M. Fujita, K. Yamada, and D. Reznik, Evidence for a charge collective mode associated with superconductivity in copper oxides from neutron and x-ray scattering measurements of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, *Phys. Rev. B* **89**, 020506(R) (2014).
 - [28] L. Pintschovius, N. Pyka, W. Reichardt, A. Rumiantsev, N. Mitrofanov, A. Ivanov, G. Collin, and P. Bourges, Lattice dynamical studies of HTSC materials, *Phys. C (Amsterdam, Neth.)* **185–189**, 156 (1991).
 - [29] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.113.174506> for fits of RIXS data to three-peak and five-peak models and $E - H$ RIXS colormaps with the quasi-elastic component subtracted.
 - [30] J. Q. Lin, H. Miao, D. G. Mazzone, G. D. Gu, A. Nag, A. C. Walters, M. García-Fernández, A. Barbour, J. Pellicciari, I. Jarrige, M. Oda, K. Kurosawa, N. Momono, K.-J. Zhou, V. Bisogni, X. Liu, and M. P. M. Dean, Strongly correlated charge density wave in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ evidenced by doping-dependent phonon anomaly, *Phys. Rev. Lett.* **124**, 207005 (2020).
 - [31] R. Arpaia, L. Martinelli, M. M. Sala, S. Caprara, A. Nag, N. B. Brookes, P. Camisa, Q. Li, Q. Gao, X. Zhou, M. Garcia-Fernandez, K.-J. Zhou, E. Schierle, T. Bauch, Y. Y. Peng, C. Di Castro, M. Grilli, F. Lombardi, L. Braicovich, and G. Ghiringhelli, Signature of quantum criticality in cuprates by charge density fluctuations, *Nat. Commun.* **14**, 7198 (2023).
 - [32] C. Zou, J. Choi, Q. Li, S. Ye, C. Yin, M. Garcia-Fernandez, S. Agrestini, Q. Qiu, X. Cai, Q. Xiao, X. Zhou, K.-J. Zhou, Y. Wang, and Y. Peng, Evolution from a charge-ordered insulator to a high-temperature superconductor in $\text{Bi}_2\text{Sr}_2(\text{Ca,Dy})\text{Cu}_2\text{O}_{8+\delta}$, *Nat. Commun.* **15**, 7739 (2024).
 - [33] O. Cyr-Choinière, R. Daou, F. Laliberté, C. Collignon, S. Badoux, D. LeBoeuf, J. Chang, B. J. Ramshaw, D. A. Bonn, W. N. Hardy, R. Liang, J.-Q. Yan, J.-G. Cheng, J.-S. Zhou, J. B. Goodenough, S. Pyon, T. Takayama, H. Takagi, N. Doiron-Leyraud, and L. Taillefer, Pseudogap temperature T^* of cuprate superconductors from the Nernst effect, *Phys. Rev. B* **97**, 064502 (2018).
 - [34] K. Yamada, C. H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R. J. Birgeneau, M. Greven, M. A. Kastner, and Y. J. Kim, Doping dependence of the spatially modulated dynamical spin correlations and the superconducting-transition temperature in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, *Phys. Rev. B* **57**, 6165 (1998).
 - [35] E. Blackburn, J. Chang, A. H. Said, B. M. Leu, R. Liang, D. A. Bonn, W. N. Hardy, E. M. Forgan, and S. M. Hayden, Inelastic x-ray study of phonon broadening and charge-density wave formation in ortho-II-ordered $\text{YBa}_2\text{Cu}_3\text{O}_{6.54}$, *Phys. Rev. B* **88**, 054506 (2013).
 - [36] M. Le Tacon, A. Bosak, S. M. Souliou, G. Dellea, T. Loew, R. Heid, K.-P. Bohnen, G. Ghiringhelli, M. Krisch, and B. Keimer, Inelastic x-ray scattering in $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ reveals giant phonon anomalies and elastic central peak due to charge-density-wave formation, *Nat. Phys.* **10**, 52 (2014).
 - [37] X. Hong, Y. Yan, L. Martinelli, I. Biało, K. von Arx, J. Choi, Y. Sassa, S. Pyon, T. Takayama, H. Takagi, Z. Li, M. Garcia-Fernandez, K.-J. Zhou, J. Chang, and Q. Wang, Observation of collective charge excitations in a cuprate superconductor, *arXiv:2503.04334*.
 - [38] K. Fujita, C. K. Kim, I. Lee, J. Lee, M. H. Hamidian, I. A. Firmo, S. Mukhopadhyay, H. Eisaki, S. Uchida, M. J. Lawler, E.-A. Kim, and J. C. Davis, Simultaneous transitions in cuprate momentum-space topology and electronic symmetry breaking, *Science* **344**, 612 (2014).

- [39] M. H. Hamidian, S. D. Edkins, C. K. Kim, J. C. Davis, A. P. Mackenzie, H. Eisaki, S. Uchida, M. J. Lawler, E.-A. Kim, S. Sachdev, and K. Fujita, Atomic-scale electronic structure of the cuprate d -symmetry form factor density wave state, *Nat. Phys.* **12**, 150 (2016).
- [40] G. Aeppli, T. E. Mason, S. M. Hayden, H. A. Mook, and J. Kulda, Nearly singular magnetic fluctuations in the normal state of a high- T_c cuprate superconductor, *Science* **278**, 1432 (1997).
- [41] M. Zhu, D. J. Voneshen, S. Raymond, O. J. Lipscombe, C. C. Tam, and S. M. Hayden, Spin fluctuations associated with the collapse of the pseudogap in a cuprate superconductor, *Nat. Phys.* **19**, 99 (2023).
- [42] J. Radaelli, O. J. Lipscombe, M. Zhu, J. R. Stewart, A. A. Patel, S. Sachdev, and S. M. Hayden, Critical spin fluctuations across the superconducting dome in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, *Nat. Commun.* (2026).
- [43] J. Zaanen and O. Gunnarsson, Charged magnetic domain lines and the magnetism of high- T_c oxides, *Phys. Rev. B* **40**, 7391 (1989).
- [44] J. M. Tranquada, Cuprate superconductors as viewed through a striped lens, *Adv. Phys.* **69**, 437 (2020).
- [45] W. A. Atkinson, A. P. Kampf, and S. Bulut, Charge order in the pseudogap phase of cuprate superconductors, *New J. Phys.* **17**, 013025 (2015).
- [46] A. Thomson and S. Sachdev, Charge ordering in three-band models of the cuprates, *Phys. Rev. B* **91**, 115142 (2015).
- [47] P. Mai, B. Cohen-Stead, T. A. Maier, and S. Johnston, Fluctuating charge-density-wave correlations in the three-band Hubbard model, *Proc. Natl. Acad. Sci. USA* **121**, e2408717121 (2024).
- [48] B. Michon, C. Girod, S. Badoux, J. Kačmarčík, Q. Ma, M. Dragomir, H. A. Dabkowska, B. D. Gaulin, J.-S. Zhou, S. Pyon, T. Takayama, H. Takagi, S. Verret, N. Doiron-Leyraud, C. Marcenat, L. Taillefer, and T. Klein, Thermodynamic signatures of quantum criticality in cuprate superconductors, *Nature (London)* **567**, 218 (2019).
- [49] S. A. Hartnoll and A. P. Mackenzie, *Colloquium: Planckian dissipation in metals*, *Rev. Mod. Phys.* **94**, 041002 (2022).
- [50] C. Tam and S. Hayden, Data for Doping evolution of the charge density wave and charge density fluctuations in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, Zenodo (2026), doi:[10.5281/zenodo.19711629](https://doi.org/10.5281/zenodo.19711629).