Multiorbital charge-density wave excitations and concomitant phonon anomalies in Bi$_2$Sr$_2$LaCuO$_{6+\delta}$

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Charge-density waves (CDWs) are ubiquitous in underdoped cuprate superconductors. As a modulation of the valence electron density, CDWs in hole-doped cuprates possess both Cu-3d and O-2p orbital character owing to the strong hybridization of these orbitals near the Fermi level. Here, we investigate underdoped Bi$_2$Sr$_2$La$_x$CuO$_{6+\delta}$ using resonant inelastic X-ray scattering (RIXS) and find that a short-range CDW exists at both Cu and O sublattices in the copper-oxide (CuO$_2$) planes with a comparable periodicity and correlation length. Furthermore, we uncover bond-stretching and bond-buckling phonon anomalies concomitant to the CDWs. Comparing to slightly overdoped Bi$_2$Sr$_1$La$_{0.6}$CuO$_{6+\delta}$ where neither CDWs nor phonon anomalies appear, we highlight that a sharp intensity anomaly is induced in the proximity of the CDW wavevector ($Q_{\text{CDW}}$) for the bond-buckling phonon, in concert with the diffused intensity enhancement of the bond-stretching phonon at wavevectors much greater than $Q_{\text{CDW}}$. Our results provide a comprehensive picture of the quasistatic CDWs, their dispersive excitations, and associated electron-phonon anomalies, which are key for understanding the competing electronic instabilities in cuprates.

**Charge-density waves | resonant inelastic X-ray scattering | phonon anomaly | high-temperature superconducting cuprates**

Cuprate superconductivity is achieved through doping into the stoichiometric parent compound, which is a charge-transfer Mott insulator with strong electronic correlations. Here, the injected holes can segregate into periodically spaced domain walls, rapidly suppressing static antiferromagnetic (AFM) order. A classic example is the ordered stripe phase in La$_{1.875}$Ba$_{0.125}$CuO$_4$ (LBCO), where both spin and charge form a quasistatic order near a doping level of ~1/8 (1). Recently, quasi-two-dimensional (2D) short-range charge-density waves (CDWs) have been discovered in most hole-doped cuprates (2–12). Although hole carriers are primarily doped into the O-2p orbitals in the CuO$_2$ planes, resonant elastic X-ray scattering (REXS) experiments have detected the quasistatic CDW order within both the Cu and O sublattices in La$_{1.875}$Ba$_{0.125}$CuO$_4$ (LBCO) owing to the strong hybridization between the Cu-3d and O-2p orbitals (2, 13, 14). The CDW in LBCO is, however, relatively long-range, leaving an open question on whether generic short-range CDWs (such as the Bi-based compounds) also project onto multiple orbitals. Subsequently to the discovery of the stripe phase in LNSCO, inelastic neutron scattering (INS) identified a pronounced softening in the Cu-O bond-stretching phonon branch close to the CDW wavevector in La-based cuprates and YBa$_2$Cu$_3$O$_{6.6}$ (YBCO) (15). In Bi$_2$Sr$_2$LaCuO$_{6+\delta}$ nonresonant inelastic X-ray scattering (IXS) studies also revealed softening in the bond-stretching phonon (16). Most recently, a similar electron-phonon anomaly was uncovered by resonant inelastic X-ray scattering (RIXS) in underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{6+\delta}$ (Bi2212), where a short-range CDW occurs (17). These studies suggest an intimate link between electron-phonon coupling (EPC) and CDW correlations. Interestingly, a dynamical dispersive CDW excitation is inferred in Bi2212 as an anomalously enhanced phonon intensity upon the interference between phonons and underlying charge excitations (17). It is unclear, however, whether such dispersive CDW excitations are ubiquitous to other cuprate families.

**Results and Discussion**

We initially used RIXS to study an underdoped superconducting single-layer Bi$_2$Sr$_1.4$La$_{0.6}$CuO$_{6+\delta}$ (Bi2201) (superconducting temperature $T_c$ = 23 K, UD23) (Materials and Methods and SI). The nature of quasistatic CDWs and their dynamical excitations is crucial for understanding their origin—similar to the study of AFM in cuprates. Although the quasistatic properties of CDWs have been investigated extensively, their dynamics (i.e., the collective CDW excitations) are largely unexplored due to the limited availability of suitable experimental tools. RIXS is one of the few techniques that directly probe quasistatic CDWs, their excitations, and electron-phonon coupling, which are important elements for identifying the CDW mechanism.

**Significance**

Charge-density waves (CDWs) are a ubiquitous form of electron density modulation in cuprate superconductors. Unveiling the nature of quasistatic CDWs and their dynamical excitations is crucial for understanding their origin—similar to the study of antiferromagnetism in cuprates. However, dynamical CDW excitations remain largely unexplored due to the limited availability of suitable experimental probes. Here, using resonant inelastic X-ray scattering, we observe dynamical CDW excitations in Bi$_2$Sr$_2$LaCuO$_{6+\delta}$ (Bi2201) superconductors through its interference with the lattice. The distinct anomalies of the bond-buckling and the bond-stretching phonons allow us to draw a clear picture of funnel-shaped dynamical CDW excitations in Bi2201. Our results of the interplay between CDWs and the phonon anomalies shed light on the nature of CDWs in cuprates.

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Appendix, Fig. S1). Fig. 1A and B show the Cu $L_3$ and O K X-ray absorption spectra (XAS) of UD23 with the incoming linear polarization (σ) parallel to the CuO$_2$ planes (σ polarized light is used throughout unless otherwise stated) (see SI Appendix, Fig. S2 for the experimental geometry). The Cu $L_3$ and O K XAS represents a projection of the unoccupied states of planar Cu-3d and O-2p orbitals, respectively. The momentum dependence of the Cu $L_3$ RIXS measurements was acquired at the Cu $L_3$ resonance ($E \sim 931.6$ eV) and over a broad range of in-plane momentum transfers $q_{//}$ (Fig. 1C) presents the low-energy excitations ($<100$ meV) in the Cu $L_3$ RIXS spectra as a function of $q_{//} = (H, 0)$. A quasielastic scattering peak is clearly visible at $q_{//} \sim 0.25$ reciprocal lattice units (r.l.u.). Such a peak was also seen in RIXS spectra under different configurations (SI Appendix, Fig. S3). O K RIXS spectra were collected with incident photon energy tuned to the resonance of the mobile carrier (hole) peak ($E \sim 528.4$ eV) as a function of $q_{//} = (H, 0)$ and up to $\sim 0.3$ r.l.u. Fig. 1D shows the corresponding data, where a quasielastic scattering peak also appears at $q_{//} \sim 0.25$ r.l.u.

To clarify the origin of these peaks, we performed an energy-dependent RIXS scan at a fixed $q_{//}$ at both resonances (Cu $L_3$ and O K). The integrated intensities of the quasielastic peak are superimposed on top of the XAS data in Fig. 1A and B. The resonance enhancement of the intensities at the Cu $L_3$- and O K hole peaks unambiguously proves the existence of a CDW on both the Cu and O sublattices of the CuO$_2$ planes (2, 6, 11, 14). Notably, an energy offset exists between the maximum of the XAS spectra and the peak of the energy-dependent scattering intensities of the quasistatic CDW. A comparable offset has been seen in multiple cuprate CDW studies where the XAS connects with the imaginary part of the atomic form factor while the scattering relates to the real part of the form factor (2, 14, 18). For a more quantitative assessment, we plot the integrated intensity of the quasistatic peak in Fig. 1E. The peak position of the CDW is extracted to be $\sim 0.259 \pm 0.006$ r.l.u. and $\sim 0.25 \pm 0.003$ r.l.u. at Cu $L_3$- and O K edges, respectively. The full width at half-maximum (FWHM) of the CDW peak is $\Gamma \sim 0.078 \pm 0.006$ r.l.u. at the Cu $L_3$ and $\Gamma \sim 0.076 \pm 0.006$ r.l.u. at O K edges. The corresponding CDW correlation length is $2/\Gamma \sim 15$ Å. Note that a comparable difference of the CDW wavevector between the Cu and O sublattices was also observed in REXS results from LBCO and is due to a tiny change in the refractive index of X-rays between the O K- and the Cu $L_3$ edges (2). We confirmed these observations on a second sample of UD23 and obtained comparable results (SI Appendix, Fig. S4). The in-plane CDW wavevector and the FWHM of the CDW peak at the Cu $L_3$ are consistent with data obtained on similar Bi2201 compounds (6, 11). We conclude that the observed CDWs at the Cu and O sublattices have comparable periodicity and correlation length, reflecting the projection of the same electronic order onto multiple orbitals.

In Fig. 1C, we highlight an excitation at $\sim 60$ meV whose intensity is enhanced for $q_{//}$ between $Q_{CDW}$ and the zone boundary. At the O K (Fig. 1D) the excitation is much weaker in comparison to the CDW peak. To quantify the $q_{//}$ dependence of the inelastic component, we fitted the quasielastic peak and then subtracted it from the RIXS spectra (Materials and Methods and SI Appendix, Figs. S5 and S6). Fitting examples of the excitation spectra at the Cu $L_3$- and O K edges are shown in Fig. 2A and B, respectively. The $q_{//}$-dependent inelastic excitations are displayed in Fig. 2 C and D with selected spectra depicted in Fig. 2 E and F covering $q_{//}$ of 0.2 to 0.3 r.l.u. Most notably, the line profile of the inelastic spectra at the Cu $L_3$ edge is single-peak while a double-peak structure appears at the O K edge. To elucidate the latter, we collected higher-energy resolution RIXS spectra from the same sample and were able to clearly resolve the lower-energy peak (SI Appendix, Fig. S7). The dispersion and integrated spectral weight were extracted and summarized in Fig. 2 G-J. At the Cu $L_3$ resonance (Fig. 2G), the inelastic peak is located around 65 meV at small $q_{//}$ then softens in the $q_{//}$ range

![Fig. 1. CDW in underdoped UD23. (A and B) Solid black lines: Cu $L_2$- and O K-edges XAS of UD23 sample collected with $\pi$ incoming polarization in normal incidence geometry (see the experimental setup in SI Appendix, Fig. S2). Blue circles (squares): Energy-dependent scattering intensities of the quasistatic peaks at the Cu $L_3$-edge (O K edge) at $q_{//} = 0.26$ r.l.u. (0.25 r.l.u.). (C and D) RIXS intensity maps excited at the Cu $L_3$ resonance of 931.6 eV and the O K-edge hole-peak resonance of 528.4 eV as a function of energy and $q_{//}$ along the (H, 0) direction. (E) Integrated intensity of the quasielastic peak, at the Cu $L_3$- and O K edges, within an energy window ($\pm 30$ meV) marked by the two white dashed lines in C and D.](https://www.pnas.org/cgi/doi/10.1073/pnas.2001755117)
of 0.2 ~ 0.4 r.l.u., with a broad “dip” developing at about 50 meV near \( Q_{\text{CDW}} \). The extracted dispersion matches well with those obtained using \( \pi \)-polarized light and they are reminiscent of the phonon softening observed in Bi2212 (17) (SI Appendix, Fig. S8). Although the softening wavevector is close to \( Q_{\text{CDW}} \) in Bi2201 and Bi2212, a recentRIXS study of CDW correlations in LSCO shows that the phonon softening develops at \( Q > Q_{\text{CDW}} \), possibly implying a delicate relationship between the momentum of the phonon softening and the CDW wavevectors (19). In Fig. 2H, the integrated intensity has a nonmonotonic increase as a function of \( q_{\parallel} \) with a maximum around 0.35 r.l.u. Noticeably, both the dispersion and the intensity profiles of the bond-stretching phonon excitations at the Cu \( L_3 \) edge as a function of momentum. The phonon spectral weight is integrated within the energy window (0–100 meV) illustrated by the white dotted line in C. (I and J) The dispersion and integrated intensity of the bond-stretching phonon (the high-energy peak, filled markers) and the bond-buckling phonon (the low-energy peak, open markers) at the O \( K \) edge. The dispersion is extracted from the fitting. Phonon intensity is the area of the curve fitting each phonon mode.

Fig. 2. Phonons in underdoped UD23. (A and B) Fitting examples of the inelastic excitation at the Cu \( L_3 \) and O \( K \) edges. Fitting details are described in Materials and Methods. (C and D) Cu \( L_3 \) and O \( K \) RIXS intensity maps as shown in Fig. 1C and D with the fitted elastic peak subtracted. (E and F) Cu \( L_3 \) and O \( K \) RIXS spectra after removing the fitted elastic peaks at selected momentum transfer values ranging from \( q_{\parallel} = 0.2 \) to 0.3 r.l.u. along the (H, 0) direction. (G and H) The fitted dispersion and the integrated intensity of the bond-stretching phonon excitations at the Cu \( L_3 \) edge as a function of momentum. The phonon spectral weight is integrated within the energy window (0–100 meV) illustrated by the white dotted line in C. (I and J) The dispersion and integrated intensity of the bond-stretching phonon (the high-energy peak, filled markers) and the bond-buckling phonon (the low-energy peak, open markers) at the O \( K \) edge. The dispersion is extracted from the fitting. Phonon intensity is the area of the curve fitting each phonon mode.
diagram by investigating a slightly overdoped Bi_{2}Sr_{1.6}La_{0.4}CuO_{6+δ} superconducting compound (Tc = 30 K, OD30), anticipating that the CDW correlations and EPC may be rather different from its underdoped counterpart. In Fig. 3A and B, Cu L_3- and O K RIXS spectra of OD30 are plotted as a function of q// (H, 0). No scattering peak is observed in the quasielastic region at either edge across the whole accessible q_κ range. To expand on this observation, we studied RIXS under various configurations including along the (H, 0) direction, the (H, H) direction, and in off-resonance conditions (SI Appendix, Fig. S9). None of them reveal any CDW signatures, making this system distinct from underdoped and the extremely overdoped Bi2201, where the CDW was found (6, 25). We illustrate these remarkable results in Fig. 3C and D. In contrast to the well-defined CDW peak in the UD23 sample, the integrated intensity of the quasielastic peak in OD30 has a simple backgroundlike profile demonstrating the complete obliteration of CDW correlations.

Correspondingly, the phonon excitations manifest differently in the absence of CDW correlations. First, the bond-stretching phonon softening is largely suppressed, regardless of whether it is probed on the Cu- (Fig. 3E) or O sublattices (Fig. 3F). Second, the EPC anomaly of the bond-stretching mode, i.e., the bond-buckling phonon (black open squares) excitations at the O edge. For E, F, and I, phonon peak positions extracted from UD23 sample are displayed in blue for comparison. (G) Integrated intensity of the bond-stretching phonon at the Cu L_3 edge within the same energy window (0–100 meV) as for UD23 sample after the removal of the elastic peak. (H and J) Bond-stretching and bond-buckling phonon intensities through the integration of the area of each fitted phonon mode at O K edge. For G, H, and J, phonon intensities in UD23 are displayed in blue for comparison. Fits of the phonon intensities are superimposed on top of experimental data in G and J.

Fig. 3. CDW and phonons studies in overdoped OD30. (A and B) Cu L_3- and O K RIXS intensity map as a function of energy and q_κ along the (H, 0) direction, respectively. The map on the right-hand side of B is the residual excitations after subtraction of the fitted elastic peaks. (C and D) Integrated intensities of the quasielastic peaks at the Cu L_3- (black dots) and O K (black squares) within the energy window (±30 meV) defined by the two white dashed lines in A and B. Integrated elastic peaks intensities in UD23 from the same energy range are shown in blue for comparison. (E) Dispersion of the bond-stretching phonon excitations at the Cu L_3- in OD30 (black dots) extracted from the fitting. (F and G) Dispersion of the bond-stretching phonon (black filled squares) and bond-buckling phonon (black open squares) excitations at the O K edge. For E, F, and I, phonon peak positions extracted from UD23 sample are displayed in blue for comparison. (G) Integrated intensity of the bond-stretching phonon at the Cu L_3 edge within the same energy window (0–100 meV) as for UD23 sample after the removal of the elastic peak. (H and J) Bond-stretching and bond-buckling phonon intensities through the integration of the area of each fitted phonon mode at O K edge. For G, H, and J, phonon intensities in UD23 are displayed in blue for comparison. Fits of the phonon intensities are superimposed on top of experimental data in G and J.
Q_{CDW} = 0.259 r.l.u. at the Cu L3 edge, we extracted the velocity of the CDW excitations, \( V_{CDW-stretching} \sim 0.45 \pm 0.05 \) eV Å, close to the bond-stretching phonon (\( \sim 60 \) meV). Near the bond-buckling phonon (\( \sim 30 \) meV), we joined the intensity anomalies at \( Q_A = 0.238 \) and 0.267 r.l.u. with \( Q_{CDW} = 0.25 \) r.l.u. from the O K edge, and obtained an averaged velocity of the CDW excitations, \( V_{CDW-buckling} \sim 1.3 \pm 0.3 \) eV Å. Remarkably, \( V_{CDW-buckling} \) is about four times larger than \( V_{CDW-stretching} \) demonstrating unambiguously that the CDW excitation disperses steeply after arising from the quasistatic CDW, then gradually flattens at higher energy. Overall, the trend extracted by the bond-stretching and the bond-buckling phonon describes funnel-shaped dispersive CDW excitations as highlighted in Fig. 4. We notice that the \( V_{CDW-stretching} \) in Bi2201 is comparable to that of the underdoped Bi2212 (\( \sim 0.6 \pm 0.2 \) eV Å) at the energy \( \sim 60 \) meV (17). Interestingly, at the energy \( \sim 30 \) meV, the deduced CDW excitations velocity in Bi2201 is close to that of the electron band dispersion, \( \sim 1.7 \pm 0.2 \) eV Å, retrieved from angular-resolved photoemission data in underdoped Bi2212 (27). The similarity of the velocities may indicate comparable self-energies between the ordinary and the periodically modulated charge carriers. However, this connection should not be simply viewed as an evidence of the weak-coupling (i.e., Fermi surface nesting) picture to describe the emergence of CDWs correlations. Rather, it is crucial for theoretical models to take into account the velocity values for the description of the CDWs in cuprates.

Lin et al. recently reported little signature of dispersive CDW excitations in LSCO compounds despite the phonon-softening across a wide doping range (19). The bond-stretching phonon intensities, with and without the presence of CDW correlation, show similar momentum dependence, in contrast to our Bi2201 RIXS data (Fig. 3 G, H, and J). As the bond-buckling phonon intensity anomaly is much more confined around \( Q_{CDW} \) differentiating drastically from the bare EPC, we corroborate the highly sensitive O K RIXS in the detection of the coupling of CDW excitations with phonons and its complementary with Cu L RIXS.

The coexistence of CDW at the Cu and O sublattices in Bi2201 demonstrates that the modulated charge density carries both Cu-3d and O-2p orbital character. REXS studies on LBCO and YBCO showed that the CDW order has \( s' \)- or \( d' \)-wave symmetry which depict a bond-centered charge order naturally explaining the projection onto Cu and O sublattices (14, 28). Given the correlation length of the CDW in Bi2201 is more than an order of magnitude shorter than in LBCO (29), we suggest that the multiorbital nature is universal to the CDWs in all hole-doped cuprates. From a theoretical perspective it is unclear whether the CDWs can be captured properly using a single-band model given the significant oxygen character of the density modulation and the phonons involved. Our work, therefore, highlights the need to use multiorbital Hubbard models (specifically three-bands Hubbard models) to describe the cuprates in terms of electron dynamics and orders (13, 30–32).

Our findings on the rich interplay between the multiorbital CDW and the electron-phonon anomalies in Bi2201 substantiate the existence of funnel-shaped CDW excitations dispersing in the energy-momentum space. This is a major step forward in characterizing the dispersive CDW excitations comparing to the previous study on Bi2212, which was solely based on the interference effect from a single-phonon branch (17). The dispersive CDW excitations are in line with the short-range dynamical charge-density fluctuations found in a large portion of the phase diagram in YBCO, postulating its ubiquity in cuprates (12). Further experiments aimed at uncovering dynamical CDW excitations in different cuprates may help elucidate the relevance of CDWs for the anomalous normal state and the unconventional superconducting properties. Concerning the underlying mechanism of CDWs, the most recent CDW studies on YBCO and LSCO add mounting evidence that the Fermi-surface nesting alone is unlikely to be the primary driving force (12, 19). Moreover, the fact that the CDW dynamics interfere with the phonons points toward an important role of the EPC in the formation of the CDWs (33–36). As suggested by refs. 20, 21, 23, 37, 38, the phonon intensity measured by RIXS is scaled to \( M^2 \), where \( M \) is the EPC matrix element. A simple comparison of phonon intensities between UD23 and OD30 (Fig. 3 G, H, and J) would imply that \( M \) at momenta far away from the phonon anomalies is comparable between two Bi2201 compounds. However, determining \( M \) at \( Q_{CDW} \) faces difficulties as the RIXS intensity is not a simple proportion to \( M^2 \) but rather reflects a complex interplay with CDW excitations. It is currently challenging to extract reliably the EPC at CDW wavevector and a much more sophisticated theoretical modeling is required in the future.
Conclusion
We combined high-resolution RIXS at the O K- and Cu L2 edges to study the quasistatic CDWs, the collective CDW excitations, and the electron-phonon coupling in Bi2201. The quasistatic CDWs are present at both Cu and O sublattices and carry comparable periodicity and correlation length implying their multiorbital nature to be universal in hole-doped cuprates. Both the bond-stretching and the bond-buckling phonons exhibit strong anomalies concomitant to CDWs. The confined intensity anomaly of the bond-buckling phonon, together with the diffused intensity enhancement of the bond-stretching phonon, uncovered unambiguously funnel-shaped dispersive CDW excitations. The significant interference effects suggest that CDWs are intimately connected to the electron-phonon coupling, which needs to be considered as a crucial element for the underlying mechanism leading to CDWs in cuprates.

Materials and Methods
Sample Growth and Characterization. High-quality single crystals of UD23 and OD30 Bi2Sr2−xLaxCuxOy cuprates with x = 0.6 and 0.2, respectively, were grown by the traveling-solvent floating-zone method. The as-grown samples were annealed at 650 °C in oxygen atmosphere for 2 d to improve sample homogeneity. The samples were precharacterized and aligned using Laue diffraction prior to RIXS experiments (SI Appendix, Fig. S1). Superconducting transition temperature Tc, are 23 and 30 K for UD23 and OD30 (SI Appendix, Fig. S1) which are consistent with a doping concentration of ∼0.13 and ∼0.18, respectively (39).

High Resolution RIXS Measurements on UD23 and OD30. High-resolution RIXS experiments were performed at the I21-RIXS beamline at Diamond Light Source, United Kingdom. Samples were cleaved in air prior to the transfer into the sample load-lock vacuum chamber. The experimental geometry is sketched in SI Appendix, Fig. S2. All samples were aligned with the surface normal (001) lying in the scattering plane. X-ray absorption was measured using the total electron yield method by recording the drain current from the sample using the RIXS spectrometer designed, built, and owned by Diamond Light Source. The RIXS spectrometer was positioned at a fixed scattering angle of 154° resulting in a maximal total momentum transfer value Q, of −0.92 Å−1 (0.52 Å−1) at the Cu L2 (O K) edge. The projection of the momentum transfer, qxy, in the a-b plane was obtained through varying the grazing incident angle of the sample owing to the quasi-2D CDW in Bi2201 system. We use the pseudotetragonal unit cell with a = b = 3.86 Å and c = 24.69 Å for the reciprocal space mapping. The momentum transfer Q is defined in r.l.u. as Q = Ha + Kb + Lc where a* = 2π/a, b* = 2π/b, and c* = 2π/c. All measurements were done at 20 K under a vacuum pressure of about 5 × 10−10 mbar. To confirm the observation of CDW, the RIXS measurements were repeated on a second sample for UD23 with the same experimental setup. Detailed results are summarized in SI Appendix, Fig. S4.

Data Analysis and Data Fitting. All RIXS spectra have been normalized by the counting time and corrected for self-absorption effects through the procedure described in SI Appendix. The zero-energy positions of RIXS spectra were determined by comparing to reference spectra recorded from the amorphous carbon tapes next to the sample for each qxy position. They were finely adjusted through the Gaussian fitting of each elastic peak. The intensity of CDW as a function of qxy results from the integration of spectra within an energy window of ±30 meV and was fitted with a Gaussian profile and a power-law function as a background. To quantify the phonon excitations, the fitting model for the Cu L2 spectra (from ~100 up to 800 meV) consists of a Gaussian (elastic peak) with a width constrained to the instrumental energy resolution, a Lorentzian (bond-stretching phonon), a damped harmonic oscillator model to account for the paramagnon at high energy and a linear background. For the O K RIXS data where the elastic peaks are generally much stronger, an instrumental energy resolution-limited Gaussian function is firstly used to fit the spectra ranging from ~100 to 25 meV, then two Gaussians (for the buckling and bond-stretching phonon modes) and a linear trend (for the background) are applied to fit the residual spectra from ~100 to 150 meV after removing the fitted elastic peaks. We note that the fitted phonon parameters in the O K spectra are generally consistent with a global fitting containing three Gaussian functions, except the central position of the bond-buckling mode is ~10 meV lower than the fitted central position of ~30 meV presented in the main text. Error bars of the phonon dispersions are determined by a combination of the uncertainty of determining the zero-energy position and SDs from the fit. For the integrated RIXS intensities of CDW and phonons, error bars are based on the noise level of measured spectra.

Data Availability. All relevant data have been deposited in the Zenodo repository at http://doi.org/10.5281/zenodo.3890415; all materials are available in the SI Appendix.

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