Unusual dynamic charge-density-wave correlations in HgBa$_2$CuO$_{4+\delta}$

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The charge-density-wave (CDW) instability in the underdoped, pseudogap part of the cuprate phase diagram has been a major recent research focus, yet measurements of dynamic, energy-resolved CDW correlations are still in their infancy. We report a high-resolution resonant inelastic X-ray scattering (RIXS) study of the underdoped cuprate superconductor HgBa$_2$CuO$_{4+\delta}$ ($T_c=70$ K). At $T=250$ K, above the CDW order temperature $T_{CDW} \approx 200$ K, we observe significant dynamic CDW correlations at about 40 meV. This energy scale is comparable to both the superconducting gap and the previously reported low-energy pseudogap. At $T=T_c$, a strong elastic CDW peak appears, but the dynamic correlations around 40 meV remain virtually unchanged. In addition, we observe a new feature: dynamic correlations at significantly higher energy, with a characteristic scale of about 160 meV. A similar scale was previously identified in other experiments as a high-energy pseudogap.

The existence of three distinct features in the charge response is highly unusual for a CDW system, and suggests that charge order in the cuprates is closely related to the pseudogap phenomenon and more complex than previously thought. We further observe the paramagnon dispersion along [1,0], across the two-dimensional CDW wavevector $q_{CDW}$, which is consistent with magnetic excitations measured by inelastic neutron scattering. Unlike for some other cuprates, our results point to the absence of a discernible coupling between CDW and magnetic excitations.

I. INTRODUCTION

The high-$T_c$ cuprates are doped charge-transfer insulators with lamellar structures that feature the quintessential CuO$_2$ plane [1]. At moderate and intermediate hole doping, these complex oxides exhibit a partial depletion of the density of states at the Fermi-level (the pseudogap, PG). The PG state is characterized by myriad ordering tendencies, including CDW order, with a modulation direction along the planar Cu-O bond and a periodicity of 3–4 lattice units [2–19]. Whereas long-range CDW order competes with superconductivity [7, 15, 16], dynamic CDW correlations have long been argued to play a key role in shaping the phase diagram (Fig. 1a) [20–22]. However, key questions remain unresolved. The mechanism of CDW formation could be related to Fermi-surface nesting (i.e., a reciprocal-space mechanism) [23], or to strong electronic correlations that lead to charge separation (i.e., a real-space mechanism) [2, 24]. There is evidence for both scenarios, but recent scattering experiments point to the former [25]. Furthermore, the relation between CDW correlations and the PG is far from understood, with suggestions that either one is the underlying phenomenon [20, 22, 26–28]. Finally, the short correlation lengths indicate that disorder plays an important role, but it is still unknown how the CDW in the cuprates becomes static on cooling and what the pinning mechanism is.

CDW correlations have been detected in numerous cuprates, primarily via resonant X-ray scattering (RXS) [6–19], including simple-tetragonal HgBa$_2$CuO$_{4+\delta}$ (Hg1201) [9, 19]. In these experiments, the incident photon energy is tuned to the Cu $L_3$ edge to enhance the scattering cross-section. The scattered photons can be directly measured in energy-integrated mode (EI-RXS), or additionally analyzed by a spectrometer in energy-resolved inelastic mode (RIXS). EI-RXS has the benefit of relatively short counting times and enabled the efficient exploration of the doping and temperature dependence of CDW correlations. In principle, the charge dynamics can be measured via RIXS, yet until recently, the best available energy resolution was well above 100 meV.

We report a RIXS study of the charge dynamics of underdoped Hg1201 ($T_c=70$ K; see Fig. 1a) across $q_{CDW}$ with nearly unprecedented energy resolution of 60 meV (FWHM). Hg1201 is a single-layer compound, with a simple tetragonal crystal structure and an optimal $T_c$ of nearly 100 K [26, 29–40]. The model nature of Hg1201 is exemplified, e.g., by the fact it is one of the few cuprates for which transport measurements reveal Shubnikov-de-Haas oscillations due to Fermi-surface reconstruction associated with the CDW order [33, 38]. However, it is important to note that the Fermi-surface reconstruction in the cuprates only appears at temperatures significantly below the CDW onset and in high magnetic fields. The zero-field CDW as seen by EI-RXS has very short in-plane correlation lengths of just a few wavelengths and there is no sharp transition temperature [9, 19]. Gaining further understanding of these emergent correlations is the primary focus of the present work. The comparatively high onset temperatures, reduced dimensionality and short length scales suggest that significant dynamic CDW correlations might be present, in analogy...
with well-known systems such as NbSe$_2$ in the presence of point disorder [41]. RIXS is a unique probe to study such effects. Indeed, at 250 K, well above the temperature ($T_{CDW}$ ≈ 200 K) below which short-range CDW order was previously observed via EI-RXS [9, 19] and time-resolved optical reflectivity [42], we only discern dynamic short-range CDW correlations, along with dynamic signal that includes a feature with a remarkably high energy of ∼ 160 meV. This scale appears in other observables (the “high-energy pseudogap”) and has been linked to the strong electronic correlations that cause the charge-transfer gap of the undoped parent insulators. We argue that the overall subtle CDW order is an emergent phenomenon that arises once the pseudogap formation is complete. Finally, we find that the magnetic excitations are nearly unaffected by the unusual CDW correlations and that the paramagnon dispersion is highly consistent with magnetic neutron scattering results.

**II. EXPERIMENTAL METHODS**

The measurements were performed with the ERIXS spectrometer at beam line ID32 of the European Synchrotron Radiation Facility (ESRF), Grenoble, France. The incident X-ray energy was tuned to the maximum of the Cu $L_\alpha$ absorption peak around 932 eV, and the X-ray polarization was set either parallel ($\sigma$) or perpendicular ($\pi$) to the scattering plane [44]. The scattered photons were analyzed without considering the final-state polarization. The energy resolution was approximately 60 meV, as determined from the full width at half maximum (FWHM) of the non-resonant spectrum of a standard polycrystalline silver sample. In order to prepare a clean and high quality surface, the Hg1201 single crystal was cleaved $ex situ$ to reveal a face parallel to the CuO$_2$ planes. Momentum scans were performed by rotating the sample about the axis perpendicular to the scattering plane, and the detector angle was set to $2\theta = 150^\circ$. The scattering wave vector is $Q = H a^* + K b^* + L c^* = (H, K, L)$ in reciprocal lattice units, where $a^* = b^* = 1.62$ Å$^{-1}$ and $c^* = 0.66$ Å$^{-1}$. $K$ was chosen to be zero, and the scans were taken along [H 0 L], with $L$ coupled to $H$. Due to the short-range nature of the observed two-dimensional correlations, the $L$ dependence of the cross section is expected to be negligible, and we quote the two-dimensional reduced wave vector $q = (H, K)$. The RIXS spectra were normalized to the integrated intensity of localized $dd$ excitations, following prior work [45].

**III. RESULTS**

Figure 1(b,c) show RIXS intensity contour plots at $T = 70$ K and 250 K, respectively, as a function of $H$ and energy transfer $\omega$ (negative $\omega$ corresponds to energy loss), obtained with $\sigma$-polarized incident X-rays directly from individual energy scans such as those shown in Fig. 1(d) [44]. The maps clearly display CDW signal around $q_{CDW} \approx (0.28, 0)$, consistent with prior EI-RXS work [9, 19]. Whereas the dominant signal at 70 K is elastic, at 250 K ($> T_{CDW}$) the response is dynamic and centered in the optic phonon range. Due to the larger phonon contribution, the 250 K spectra exhibit higher intensity away from $q_{CDW}$ than the 70 K data. CDW correlations are
(a-d) σ-polarized RIXS intensity spectra at 70 K and 250 K, integrated over the FWHM instrument resolution, centered at zero, 60, 120 and 180 meV energy loss. (e) σ and (f) π-polarized RIXS spectra at 70 K and 250 K, integrated over larger energy window, as indicated. In all cases, the 70 K curves are vertically shifted for clarity. The black lines in (a-e) are polynomial momentum dependences, and the same in each case at low and high temperature. Blue and red lines in (a-e): fits to Gaussian peaks on top of the background at 70 K and 250 K, respectively. Blue and red lines in (f): guides to the eye. Vertical dashed grey lines indicate \( q_{\text{CDW}} \).

Also observed in \( \pi \) polarization [44].

A more detailed data analysis reveals additional information. Fig. 2(a-d) shows the momentum dependence of RIXS signal (obtained from energy scans such as those in Fig. 1(d), with \( \sigma \) polarization) integrated over the FWHM energy resolution (60 meV) with different energy ranges: quasi-elastic \((-30, +30)\) meV; inelastic \((-90, -30)\) meV, \((-150, -90)\) meV and \((-210, -150)\) meV. In order to arrive at a systematic estimate of the signal strength, we fit the data to a Gaussian peak with fixed center \( q_{\text{CDW}} \) and width \( \Delta \). For \( |\omega| < 90 \text{ meV} \), this ‘background’ invariably includes phonon scattering; at both 70 K and 250 K it is indistinguishable for all energy-integration ranges.

Fig. 3(a,b) shows the energy dependence of the Gaussian amplitude obtained in this manner for \( \sigma^- \) and \( \pi^- \) polarization, respectively. This amplitude is a measure of the \( q \)-integrated CDW signal. The 250 K results in Figs. 3(a,b) show broad peaks centered at \( \sim 40 \) meV and no evidence for elastic scattering, as the energy dependence of the amplitude is fully captured by the sum of Stokes and anti-Stokes scattering. The response is broader than the energy resolution and corresponds to an intrinsic width of \( \sim 60 \) meV or a large distribution of charge modes [44]. This analysis removes featureless phonon contributions: whereas the peak at \( \sim 30 \) meV in Fig.1(c) contains phonon contributions, those at \( \sim 40 \) meV in Figs. 3(a,b) are mostly CDW-dominated.

At 70 K, the dominant CDW response is quasi-elastic. The width of this peak is larger than the energy resolution, consistent with an additional dynamic contribution at \( \sim 40 \) meV, as observed at 250 K. This distinct possibility is highlighted for both polarizations in the (Bose-factor-corrected) intensity difference plots in Fig. 3(c,d), which reveal resolution-limited elastic peaks centered at \( \omega = 0 \).

Interestingly, at 70 K we also observe dynamic CDW signal above 80 meV, the highest phonon energy of Hg1201. This is directly seen from Fig. 2, especially Fig. 2(e), where the large binning range \((-280,-120)\) meV was chosen in order to optimize signal-to-background. These data, obtained with \( \sigma \)-polarization, are contrasted in Fig. 2(f) with the equivalent result with \( \pi \)-polarization, which is more sensitive to magnetic scattering [44]. The convex momentum dependence can be attributed to paramagnons, which become prominent above \( \sim 200 \) meV (see below). This is seen from the comparison in Fig. 2(f) with the result obtained with narrower \((-200,-120)\) meV integration, which yields an approximately linear background consistent with the \( \sigma \)-polarization result in Fig. 2(e). From Fig. 3(a), the high-energy charge signal is seen to be peaked at about 100 meV; it is not discerned in \( \pi \)-polarization (Fig. 3(b,d)) due to the higher background level (proximity to paramagnon excitations; Fig. 2(f)) and lower expected charge scattering cross section (by a factor of two [44]).

Figures 1(d) and 4(a) reveal an additional broad, dispersive peak in the 0.15 to 0.4 eV range. This feature, which has been observed in a number of cuprates, signifies paramagnon scattering that evolves from well-defined antiferromagnetic excitations in the undoped parent compounds [46, 47]. These excitations are more prominent in \( \pi^- \) than in \( \sigma^- \) polarization in the present scattering geometry, as expected for magnetic scattering [44, 48, 49]. We extract the paramagnon energy by fitting the raw RIXS spectra to a resolution-limited elastic peak, a phonon peak, and a damped paramagnon excitation [44]. Figure 4(b) summarizes our result for the paramagnon dispersion along \([1,0]\) at 70 K. We compare the RIXS data with magnetic neutron scattering data near the antiferromagnetic wave vector for two Hg1201 samples (one with essentially the same doping level and \( T_c = 71 \) K [37], and the other with \( p \approx 0.064 \) and \( T_c = 55 \) K [50]) and find that these data are highly consistent and complementary. From a heuristic fit of the combined neutron and X-ray data above \( H = 0.1 \) rlu to simple linear spin-wave theory [51–53], we obtain an effective nearest-neighbor exchange of 123(3) meV. Overall, the RIXS data for the paramagnon dispersion in Hg1201 are consistent with prior measurements for other hole-doped cuprates [44, 48, 54].
FIG. 3. Energy dependence for (a) $\sigma$- and (b) $\pi$-polarization of fitted Gaussian amplitude from energy-integrated data such as those shown in Fig. 2(a–d). Insets: zoom of the range $(-250, -100)$ meV. Blue and red shaded areas indicate the instrument resolution 60 meV (FWHM). Gaussian fits to Stokes and anti-Stokes scattering at 250 K (dashed red lines; sum: solid red lines) yield 41(2) meV and 61(6) meV (FWHM) for the peak position and intrinsic (de-convoluted) peak width. Gaussian fits to three peaks at 70 K (blue lines) capture (1) quasi-elastic, (2) low-energy ($\sim 40$ meV), and (3) high-energy (peak at 155(6) meV, intrinsic width of 85(15) meV (FWHM)) contributions to the CDW response; the latter is not discerned in $\pi$-polarization, for which the overall charge response is expected and seen to be weaker [44]. (c) and (d): difference in amplitude between 70 K and 250 K from (a) and (b), respectively, after correcting the 250 K data for the Bose factor. Green and purple shaded areas indicate the instrument resolution; vertical dashed lines indicate peak centers obtained from fits to Gaussian profiles, which are consistent with zero energy transfer. The orange shaded area in (c) indicates the net signal centered at 160(6) meV.

IV. DISCUSSION

We observe CDW signal in three different energy ranges: (1) quasi-elastic order at 70 K; (2) low-energy ($\sim 40$ meV) fluctuations at 250 K, and likely also at 70 K; (3) high-energy fluctuations at 70 K with a characteristic energy of about 160 meV. Prior work indicates that the quasi-elastic CDW component develops below the characteristic temperature $T_{CDW}$ [9, 19, 42] (Fig. 1). The short correlation length of $\xi/a \sim 4-5$ of both the dynamic and quasi-elastic CDW correlations deduced from the peak widths in Fig. 2 is consistent with prior EI-RXS work [9, 19]; in the case of Hg1201, the spatial extent is potentially set by point disorder due to interstitial oxygen atoms [19].

The 40 meV scale lies in the optic phonon range, is consistent with superconducting gap and pseudogap scales for cuprates with a comparable optimal $T_c$ [55, 56], and with the lower bound of $\sim 20$ meV for the gap between the reconstructed pockets due to biaxial CDW order deduced from quantum oscillation experiments for Hg1201 [38]. Similar to observations (1) and (2), a recent RIXS study of hole-doped NdBa$_2$Cu$_3$O$_{6+\delta}$ (NBCO) [27] found quasi-elastic CDW correlations only at low temperature, and dynamic CDW correlations to persist above $T^*$ with comparable relative intensity, but significantly smaller energy [44]. In hole-doped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212) [57], dynamic CDW correlations were identified in the 40-60 meV range and associated with an optic phonon anomaly at low temperature, and with a wavevector shift near $T^*$ that is not observed in the present work. Importantly, it has been shown that even in well-known CDW systems such as NbSe$_2$, phase fluctuations of the CDW order can be relevant, especially in the presence of point disorder [41], leading to an extended precursor regime at high temperatures. It is therefore not surprising that a dynamic signature of the CDW is also present in the cuprates above the nominal ordering temperature, which in this case most likely represents a pinning temperature and not a true phase transition.

We note, however, that the difference plots in Fig. 3 suggest another possibility. Taken at face value, the differences indicate that the weight of the $\sim 40$ meV inelastic contribution is temperature-independent, since it cancels out within error for the two temperatures. Equiv-
alently, there seems to be little spectral weight transfer from the $\sim 40$ meV inelastic to the elastic contribution in cooling, which is rather unexpected in a scenario where the CDW simply becomes pinned at low temperatures. It is possible that the elastic and the inelastic contributions have a somewhat different physical origin; in particular, the elastic contribution might be due to the interference of Friedel oscillations around impurities, as was suggested theoretically [58], and tested in a quasi-one-dimensional CDW system [59]. The ‘true’ underlying CDW signal would then be inelastic even at 70 K, intriguingly suggesting that the CDW remains dynamic in Hg1201 at this temperature, consistent with the fact that transport signatures of Fermi-surface reconstruction are only seen at relatively low temperature (in the presence of a large $c$-axis magnetic field) [60]. Also, the lack of temperature dependence would imply a dynamic CDW onset temperature significantly above 250 K. Alternatively, the inelastic feature might be only indirectly related to the CDW and a signature of some other charge mode that is insensitive to temperature in the given range.

B. High-energy response, comparison with other observables

High-energy charge correlations above the optic phonon range so far have been reported only for electron-doped Nd$_{2-x}$Ce$_x$CuO$_4$+$\delta$, where they persist to $\sim 0.4$ eV [61]. The large $\sim 160$ meV energy scale identified here for hole-doped Hg1201 is consistent with the high-energy pseudogap (“hump”) scale seen in other observables [26, 55]. A recent phenomenological model, rooted in the dual empirical observations of a universal transport scattering rate [36, 62] and universal inherent inhomogeneity [39, 63, 64], associates this scale with the charge-transfer gap at zero doping and with the (de)localization of one hole per planar CuO$_2$ unit above $T^*$ (Fig. 1) [26, 40]. For Hg1201, at $p \approx 0.09$, the mean (de)localization gap is $\sim 180$ meV, and the gap distribution that best captures the transport data is 70 meV (FWHM) [26], consistent with the characteristic CDW scale and width (Fig. 3a,c)).

However, the delocalization of one hole per CuO$_2$ unit is a large effect that clearly manifests itself in, e.g., the evolution of the Fermi-surface with doping and the strong temperature and doping dependence of the Hall number [26, 40]. In contrast, the CDW order involves a tiny fraction of one hole per unit cell. While X-ray scattering experiments cannot provide the absolute CDW amplitude, a value of about 0.03 hole per CuO$_2$ unit was estimated for La$_2-x$Ba$_x$CuO$_4$ [24]. A separate, consistent estimate can be obtained from NMR: From the universal relation between oxygen hole content and NQR frequency [65] and the $^{17}$O NQR line broadening in the CDW phase [12], we estimate 0.028 hole in the Cu 3$d$ orbital for Hg1201 [66]. It therefore seems likely that the CDW is a secondary, emergent phenomenon related to the strong correlations that underlie the hole localization. This is supported by STM evidence for a qualitative change in the CDW form factor of Bi2212 at a characteristic scale comparable to the pseudogap scale [28]. Furthermore, it is known that cuprates are intrinsically inhomogeneous, with (inhomogeneous) local gaps that persist well above the pseudogap temperature [67]. Puddles of localized charge that sustain dynamic CDW correlations thus may already exist outside of the nominal PG region (above $T^*$($p$)). This can account for the present observation for Hg1201 at 250 K and for the similar result for YBCO [27].

The high-energy signal we observe has an even smaller spectral weight, i.e., charge magnitude, than the static CDW response. It therefore might not be a direct signature of the fluctuations of one hole per CuO$_2$ unit. Yet we note that the local real-space character of the hole localization should affect much of the Brillouin zone. In our experiment, the associated energy scale is only observed at the CDW wavevector, and only at relatively low temperature. It appears to be related to features seen in Raman spectra with $B_{1g}$ [68] and $B_{2g}$ symmetry [69]. In $B_{1g}$ symmetry, a broad feature with characteristic energy of $\sim 200$ meV ($\sim 1700$ cm$^{-1}$) was identified for a Hg1201 sample with similar doping level ($T_c = 77$ K) and associated with the two-magnon excitations of the undoped antiferromagnetic parent compounds. While a clear temperature dependence was observed below $\sim T^*$, this feature persists at higher temperatures [68]. In $B_{2g}$ symmetry, a feature of width $\sim 60$ meV centered at $\sim 150$ meV ($\sim 1250$ cm$^{-1}$) was observed for Hg1201 with $T_c = 72$ K and interpreted as a CDW energy scale [69]. These values are remarkably close to the peak at 155(6) meV and intrinsic width of 85(15) meV (FWHM) we extract (Fig. 3). Since Raman scattering and RIXS probe different regions of the Brillouin zone, it seems plausible that the same excitations exist in a wide range of reciprocal space. Similarly, there may exist charge fluctuations throughout the entire Brillouin zone at the lower characteristic energy scale ($\sim 40$ meV) identified in the present work. We note that optical spectroscopy efforts to extract the bosonic pairing glue revealed a robust, compound and doping independent peak around 50-60 meV, and a second feature in the 100-300 meV range [70, 71]. Furthermore, evidence for a charge collective mode has been deduced from anomalies in the optic phonon range [72]. Clearly, further RIXS studies of Hg1201 across the phase diagram are highly desirable, and should clarify these important issues.

C. Real-space vs. k-space mechanism, charge vs. magnetic correlations

The present data do not allow us to unambiguously discern if the CDW forms predominantly due to Fermi surface nesting or a real-space mechanism. We can, however, make some inferences at this point. In previous RXS work, it was argued that the doping dependence of
the CDW wave vector is consistent with a nesting scenario, and transport measurements certainly show that the Fermi surface is reconstructed at low temperatures and high magnetic fields. However, the static zero-field CDW correlations observed here clearly do not induce a reconstruction (at least above $T_c$), since transport properties are virtually insensitive to the CDW formation at temperatures above $T_c$. Furthermore, both the presence of a dynamic component with high onset temperature and the emergent high-energy scale suggest a strong-coupling scenario, making an underlying real-space mechanism more likely. This is also to be expected if the CDW is indeed an emergent phenomenon, since the correlations that cause the pseudogap are localized in real space. It seems likely that, effectively, both $k$- and $r$-space effects contribute, which could be related to the deeper question of the existence of a well-defined reciprocal space in a material that is inherently inhomogeneous at the nanoscale [64, 73].

A recent RIXS study of electron-doped Nd$_{2-x}$Ce$_x$CuO$_{4+\delta}$ suggests a coupling between dynamic magnetic and charge-order correlations [61]. In hole-doped cuprates, on the other hand, there is no clear evidence for such a coupling, except for the special case of the “214” family of materials that exhibit charge-spin stripe order [25]. This is supported by the insensitivity of the paramagnon energy and width around $q_{CDW}$ observed here for underdoped Hg1201 (Fig. 4).

V. CONCLUSION

In conclusion, we have measured the charge dynamics of underdoped Hg1201 using RIXS at the Cu $L_3$ edge with nearly unprecedented energy resolution, which has allowed us to discern dynamic from static CDW correlations. Above $T_{CDW}$, the temperature previously identified with the onset of CDW correlations, the response is purely dynamic, with an energy scale comparable to both the superconducting gap and the low-energy pseudogap. As expected, quasi-static CDW correlations are observed at low temperature. However, there also exists an additional dynamic signature with a remarkably high energy scale that appears to be an imprint of the high-energy pseudogap and associated with the strong electronic correlations that cause the charge-transfer gap of the underdoped parent insulators. The present work sets the foundation for future RIXS measurements of the detailed temperature and doping dependence of the dynamic CDW correlations in the model cuprate Hg1201, and for understanding the CDW phenomenon of the cuprates in the context of large local charge fluctuations.

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1. **Scattering Geometry**

Resonant Inelastic X-ray Scattering (RIXS) measurements were performed at the ID32 beamline of the European Synchrotron Radiation Facility (ESRF) with energy resolution of 60 meV (FWHM). The Hg1201 single crystal was cleaved ex situ prior to the RIXS measurements. Figure S1(a) shows a schematic of the scattering geometry. Momentum scans were performed by rotating the sample about the $b$ axis with the detector angle fixed at $2\theta = 150^\circ$. We obtained energy-momentum intensity maps along $[H,0]$ for the $H$ values indicated in Fig. S1(b). At each momentum transfer, we measured the excitation spectrum for approximately 45 minutes. Additionally, X-ray absorption spectra were periodically taken to ensure that the incident X-ray energy remained at the maximum of the Cu $L_3$ resonance edge. Raw data are shown in Fig. S2.

![Scattering geometry with two different incident photon polarizations: parallel ($\pi$) and perpendicular ($\sigma$) to the $(H\ 0\ L)$ scattering plane. (b) Reciprocal-space schematic with first nuclear and antiferromagnetic first Brillouin zones shown as solid and dashed lines, respectively. The $H$ values accessed in the experiment are indicated by circles.](image1)

**Fig. S1.** (a) Scattering geometry with two different incident photon polarizations: parallel ($\pi$) and perpendicular ($\sigma$) to the $(H\ 0\ L)$ scattering plane. (b) Reciprocal-space schematic with first nuclear and antiferromagnetic first Brillouin zones shown as solid and dashed lines, respectively. The $H$ values accessed in the experiment are indicated by circles.

![RIXS spectra measured in (a) $\sigma$ and (b) $\pi$ polarization.](image2)

**Fig. S2.** RIXS spectra measured in (a) $\sigma$ and (b) $\pi$ polarization.
2. RIXS spectra and paramagnon excitation

In order to extract the paramagnon response, we decompose the mid-infrared (MIR) region of the RIXS spectra into three components (Fig. S3): a resolution-limited elastic peak, a resolution-limited phonon excitation and a damped paramagnon excitation. We use the following heuristic formula to fit the “elastic line”,

\[ \frac{1}{I_{dd}}(\omega) = G(\omega) + n_B L(\omega) + n_B \chi''(\omega), \]

where \( n_B = [1 - \exp(-\hbar \omega/k_B T)]^{-1} \) is the Bose factor, \( G(\omega) \) is the resolution-limited Gaussian function, and \( L(\omega) \) the resolution-limited Lorentzian profile used to capture the phonon contribution. In addition, a linear background is assumed. The paramagnon has an asymmetric shape, and was fitted to a damped harmonic oscillator form,

\[ \chi''(\omega) = \chi_0'' \frac{\gamma \omega}{[\omega^2 - \omega_0^2 + \omega_1^2 + \gamma^2]} = \frac{\chi_0''}{2\omega_1} \left[ \frac{\gamma/2}{(\omega - \omega_1)^2 + (\gamma/2)^2} - \frac{\gamma/2}{(\omega + \omega_1)^2 + (\gamma/2)^2} \right] \]

with damping coefficient \( \frac{\gamma}{2} = \sqrt{\omega_0^2 - \omega_1^2} \). In order to model the RIXS spectra, \( G(\omega), L(\omega) \) and \( \chi''(\omega) \) are further convoluted with the instrument resolution function. The damping coefficients for Hg1201 are shown in inset of Fig. S4.

Fig. S4 also compares the (para)magnon excitations in doped and undoped La\(_{2-x}\)Sr\(_x\)CuO\(_4\) system. The paramagnon excitations in LSCO \( p = 0.11 \) [43] and Hg1201 \( p = 0.08 \) lines in the similar energy up to \( H = 0.35 \) r.l.u.

![Fig. S3. Fits to RIXS data taken at 70 K in \( \pi \) geometry, with the in-plane momentum transfer \( H \) indicated. The spectra are decomposed into three components plus a linear background (blue): a resolution-limited elastic peak (yellow), a resolution-limited effective phonon peak (purple), and a damped paramagnon excitation (green).](image-url)
Fig. S4. Dispersion of magnetic excitations in Hg1201 (p = 0.083) at T = 70 K, compared with the undoped antiferromagnetic La$_2$CuO$_4$ (LCO) [40,41,42] and 11% Sr-doped La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) [43]. Blue and purple dashed lines are fits to nearest-neighbor spin wave theory for Hg1201 and LCO, respectively. Inset: Damping coefficient for Hg1201 at 70 K and 250 K. Error bars are set by the energy resolution (30 meV - HWHM).

3. Polarization dependence

The Cu L$_3$-edge RIXS cross section can be calculated using the approximated isolated Cu$^{2+}$ ion under the tetragonal crystal field with $D_{4h}$ symmetry. Following ref. [38], we calculate the scattering amplitude for the present experimental geometry. For spin-conserving scattering,

$$F_{\sigma\sigma'} \propto \frac{1}{3}$$

$$F_{\pi\pi'} \propto -\frac{1}{3} \sin\left(\frac{tt\theta - \delta}{2}\right) \sin\left(\frac{tt\theta + \delta}{2}\right)$$

and for spin-flip scattering,

$$F_{\sigma\pi'} \propto -\frac{i}{6} \sin\left(\frac{tt\theta - \delta}{2}\right)$$

$$F_{\pi\sigma'} \propto -\frac{i}{6} \sin\left(\frac{tt\theta + \delta}{2}\right)$$

where $\sigma/\pi$ and $\sigma'/\pi'$ denote the incident and scattered photon polarization. The angles $tt\theta$ and $\delta$ are the scattering angle and the rotation angle of the sample from the specular condition, respectively. By comparing the intensity ratio of two different incident photon polarizations, $I_\sigma/I_\pi$, we can determine the charge or magnetic nature of the excitation. Fig. S5 shows the theoretical and experimental polarization dependence in the quasi-elastic and paramagnon range. The calculation confirms the dominant charge and magnetic origin for quasi-elastic and paramagnon range, respectively.
Fig. S5. (a) RIXS intensity calculation with isolated Cu\(^{2+}\) model in different scattering geometries. (b) Intensity ratio \(I_\sigma/I_\pi\) in the quasi-elastic range. (c) Intensity ratio \(I_\sigma/I_\pi\) in the paramagnon range. Yellow and purple circles are the data and the solid lines are the corresponding calculations.

4. Comparison with Nd\(_2\)Ba\(_3\)CuO\(_{7-\delta}\)

Arpaia et al. reported similar low energy CDW excitations in NdBa\(_2\)CuO\(_3\) (NBCO) [27]. Figure S6 shows a comparison of CDW excitations in Hg1201 UD70 and NBCD OP90 [27]. Although the NBCO UD60 sample studied in ref. [27] has a doping level that is closer to our Hg1201 sample, we consider the phonon-subtracted CDW data here, which are only available for NBCO OP90. The data in Fig. S6 are scaled to match the low-temperature amplitudes. Both compounds show quasi-elastic CDW correlations near \(T_c\) and purely dynamic correlations at high temperature. Although the characteristic energy of dynamic CDW is estimated to be slightly smaller in NBCO, the dynamic correlations in Hg1201 and NBCO have similar relative low-temperature amplitudes. Moreover, in addition to the analysis in ref. [27], where a standard dynamic Ginzburg-Laudau approach is used, we show that the NBCO 250 K data can also modeled as single-mode Stokes and anti-Stokes excitations. The resulting excitation energy of about 15 meV, consistent with the analysis shown in ref. [27].

Fig. S6. Comparison of Hg1201 UD70 (this work) with optimally-dope NBCO OP90 [27] after subtraction of phonon contributions. Blue and red squares: fitted Gaussian amplitude for Hg1201, as shown in Fig. 3(a). Yellow and green circles: RIXS spectra for NBCO. The solid green line is a fit to NBCO OP90 250 K data assuming single-mode Stokes and anti-Stokes contributions (dashed lines). The peak lies at 15 meV, consistent with the analysis in ref. [27].
5. Charge susceptibility

For the analysis in Fig. 3, we assumed a single mode at 250 K around 40 meV. The sum of Stokes and anti-Stokes contribution fits well to the data. We note that this low energy CDW excitation can also be modeled as a distribution of modes of different energies, known as charge susceptibility. We show such analysis in Fig. S7, where a Log-normal distribution is assumed at zero temperature. The high temperature (250 K) can be modeled using Bose-Einstein statistics and detailed balance. The line shape is further convoluted with the Gaussian resolution function.

Fig. S7. Charge susceptibility modeled by a log-normal distribution in (a) σ- and (b) π-polarization. Filled square and diamond symbols are the resulting fit amplitudes from Fig. 2 and are the same as those in Fig. 3(a, b). The solid red line is the fits to a resolution-convoluted Log-normal distribution. The dashed red dashed line indicates the deconvoluted charge susceptibility at 250 K. The blue dashes line is the Bose-corrected charge susceptibility at zero temperature.