Light-Induced Charge Order Mode in a Metastable Cuprate Ladder

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We report the observation of an emergent charge order mode in the optically excited cuprate ladder $Sr_{14}Cu_{24}O_{41}$. Near-infrared light in the ladder plane drives a symmetry-protected electronic metastable state together with a partial melting of the equilibrium charge order. Our time-resolved resonant inelastic x-ray scattering measurements at the upper Hubbard band reveal a collective excitation dispersing from the charge order wave vector up to 0.8 eV with a slope on the order of the quasiparticle velocity. These findings reveal a regime where correlated carriers acquire itinerant character at finite momentum, and charge order becomes dynamically fluctuating, offering a platform to explore light-induced pairing instabilities.

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Optically excited quantum materials display striking nonequilibrium phenomena, ranging from photoinduced magnetic [1,2] and charge-ordered phases [3] to transient topological [4–6] and superconducting states [7–9]. Yet, these driven states are often short-lived, rapidly relaxing back to equilibrium due to dissipation and decoherence. In rare cases, light can steer materials into nonequilibrium configurations that evade thermalization and acquire metastable character. Such long-lived states generally arise from cooperative structural and electronic effects, defect dynamics, or trapping by impurities [10–24]. Metastability, however, may also emerge from purely electronic dynamical bottlenecks, and realizing electronic long-lived, or "hidden," states is a key challenge in the study of dynamical phase transitions.

Strongly correlated materials provide a natural platform for realizing electronic metastability [25]. Theoretical works have proposed the creation of light-induced states protected against rapid decay by the presence of a Mott gap [26,27], by approximate conservation laws [28], or by transient trapping in the presence of competing orders [29,30]. Electronic nonthermal phases may also be engineered by transiently modifying the electronic Hamiltonian, which involves coherently renormalizing electronic interactions via optical dressing [31] (either transiently or in a steady state) and enabling persistent changes in carrier distributions. These nonequilibrium states are predicted to exhibit novel electronic phases including excitonic order [32,33] and η-pairing condensates [34–36]. However, long-lived nonequilibrium electronic states have remained experimentally elusive, hindering the exploration of novel charge correlations.

Recent experiments have observed electronic metastability in the model cuprate ladder $Sr_{14}Cu_{24}O_{41}$ [37]. Starting from a self-doped ($p = 0.06$) charge-ordered ground state [38,39], ultrafast 1.55-eV pump pulses induce a prompt transfer of holes between the chain charge reservoir and ladder subunits, which are effectively decoupled at equilibrium [see Fig. 1(a)]. The ladder hole concentration increases ($\Delta p = 0.03$), partially melting charge order and driving the system toward a nonthermal, optically gapless regime. This nonequilibrium charge redistribution arises from a coherent optical dressing of Zhang–Rice singlet states in the ladders and the activation of a symmetry-forbidden chain-to-ladder

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hopping term. When the optical field vanishes, the equilibrium symmetry is restored, suppressing relaxation and trapping holes in a nonthermal state persisting for several nanoseconds. These results prompt the question of whether trapped carriers in the electronic metastable state exhibit emergent collective dynamics or incipient pairing, as expected from the intrinsic tendency of cuprate ladders toward hole binding and superconductivity under pressure [40–44].

Here, we investigate this by probing the charge dynamics of driven $Sr_{14}Cu_{24}O_{41}$ using O K-edge time-resolved resonant inelastic x-ray scattering (trRIXS). Following optical excitation into the metastable state, we observe a novel collective mode emerging from the partially melted charge order. This excitation disperses up to 0.8 eV from the ordering wave vector with a slope on the order of the quasiparticle velocity, as expected for a charge collective mode. Based on its energy scale and dispersion, and supported by numerical simulations, we attribute this mode to charge order fluctuations in the ladders. These measurements indicate that the electronic metastable state hosts fluctuating charge order dynamics that are unique to the driven system and distinct from those of charge-ordered cuprates at equilibrium.

We conducted trXAS and trRIXS measurements at the Furka endstation of the Athos beamline at SwissFEL, Paul Scherrer Institut [52]. We cleaved high-quality single crystals of $Sr_{14}Cu_{24}O_{41}$ *in situ* along the ac plane and maintained them at 100 K throughout the experiment. We acquired trXAS spectra in fluorescence yield mode with x-rays at near-normal incidence, detecting the signal with an avalanche photodiode positioned at $2\theta = 78°$. We recorded O K-edge trRIXS spectra with an incident energy of 530 eV, keeping the scattering angle fixed at $2\theta = 136°$ while varying the incident angle θ from 70° to 130°. This geometry corresponds to momentum transfers from 0 to 0.28 r.l.u. (in units of $2\pi/c_L$, $c_L = 3.95$ Å) along the ladder legs and accesses the tail of the charge-order peak at $q_{CO} = (0, 1, 0.2)$ r.l.u. The RIXS spectrometer provided a total energy resolution of 160 meV. We used σ-polarized x-rays to enhance the intensity of charge excitations and focused the beam to 300 μm$(H) \times 10$ μm(V). We monitored shot-to-shot x-ray intensity fluctuations with a photodiode and used them to normalize the data. We excited the samples with 800 nm (1.55 eV), 100-fs pulses, polarized along the ladder rungs and focused to 500 μm to achieve a fluence of 6 mJ/cm^2. The pump penetration depth exceeds that of the soft x-rays at all measured incident angles, ensuring a homogeneously excited volume.

Resonant x-ray scattering at the O K edge ($1s \to 2p$ transition) of copper oxides is a direct probe of hole ordering and collective excitations. The x-ray absorption spectrum of $Sr_{14}Cu_{24}O_{41}$ in this energy range exhibits two prominent preedge features at 528 and 529.7 eV, corresponding to the Zhang-Rice singlet (ZRS) and upper Hubbard band (UHB),

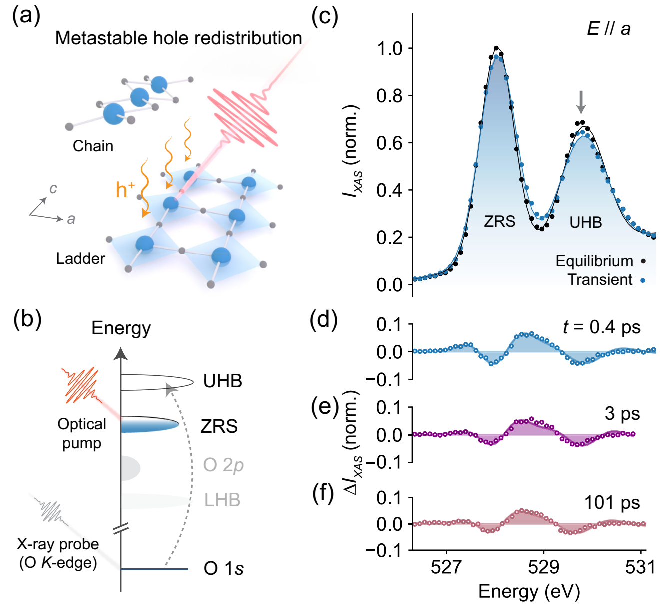

FIG. 1. (a) Crystal structure of $Sr_{14}Cu_{24}O_{41}$ featuring chain and ladder sublattices. Effectively independent at equilibrium, these units become transiently coupled by near-infrared (1.55 eV) excitation, giving rise to a metastable chain-to-ladder hole transfer. (b) Time-resolved x-ray absorption spectroscopy (trXAS) at the O K edge probes the low energy electronic structure of chains and ladders, including upper Hubbard band (UHB) and Zhang-Rice singlet (ZRS) states. (c) Equilibrium (black) and transient (blue, 6 mJ/cm^2 fluence) O K-edge trXAS at $t = 0.4$ ps. Pump and probe pulses are polarized along the ladder rungs ($E \| a$). (d)–(f) Differential trXAS intensity $[I_{XAS}(t) - I_{XAS}(t < 0)]$ at $t = 0.4$, 3, and 101 ps, respectively. The pump-induced spectral reshaping is metastable. The solid lines are fits to the data (see Supplemental Material Sec. 1 [45]).

respectively [Figs. 1(b) and 1(c)]. The relative intensities of these peaks encode the distribution of holes between chains and ladders, as established in equilibrium studies with Ca substitution [38,53]. Following optical excitation, the trXAS spectra [Fig. 1(c)] display a prompt reshaping (also see Fig. S2), which remains largely unchanged for hundreds of picoseconds [Figs. 1(d)–1(f)]. This reshaping is consistent with our previous report of metastable chain-to-ladder hole transfer [37] and with the equilibrium behavior of Ca-substituted compounds [53]. Crucially, these spectral changes reveal that correlated carriers in the UHB undergo pronounced dynamical changes in the metastable state.

We probe the collective excitations of these correlated carriers using high-resolution trRIXS [54,55] [Fig. 2(a)]. To suppress fluorescence that overlaps with the low energy RIXS signal and directly probe the dynamics of the UHB states (see RIXS energy map in Fig. S3), we tune the incident x-rays to the 529.7-eV resonance. With the scattering geometry set in the ac plane, we measure the dispersion along the ladder legs (q_{leg}) while simultaneously intercepting the tail of the charge-order peak q_{CO} [38,56],

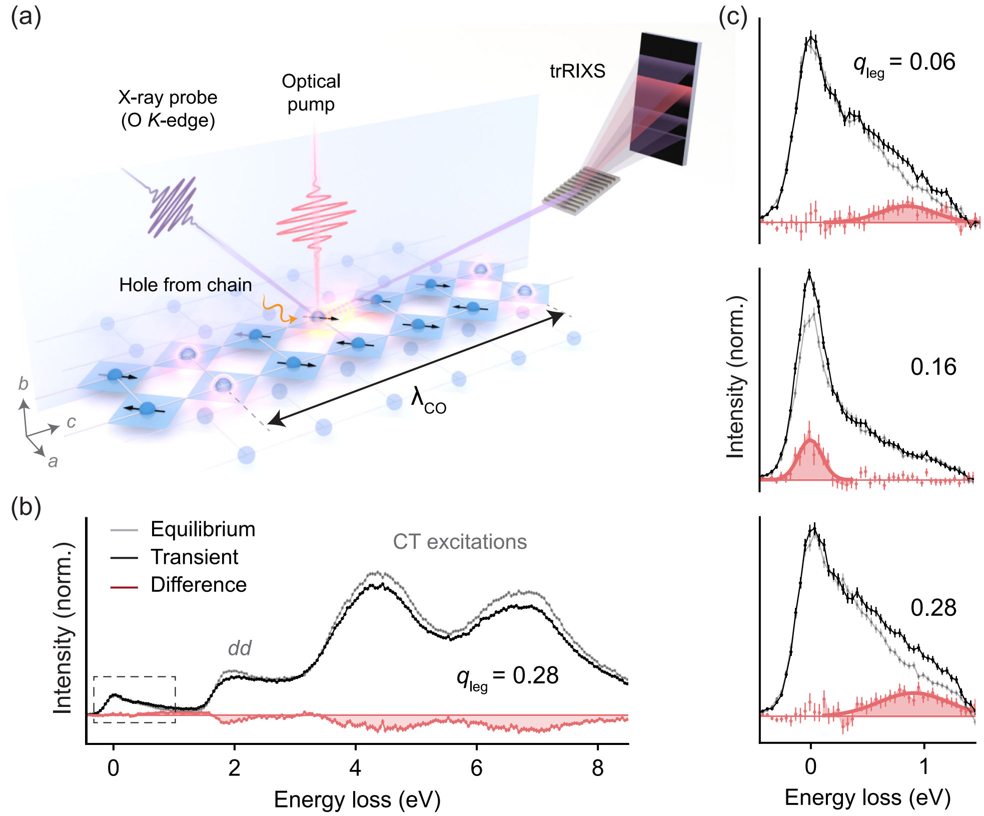

FIG. 2. (a) Sketch of the time-resolved resonant inelastic X-ray scattering (trRIXS) experiment. Following optical excitation, O K-edge x-ray pulses probe charge dynamics in the ladder via scattering into a grating spectrometer. Black arrows represent spins, pink halos represent charge-ordered holes in the ground state, and the yellow halo a hole transferred from the chain to the ladder. λ_{CO} denotes the charge-order wavelength. (b) Representative equilibrium (gray), transient (black), and difference (red) raw trRIXS spectra at resonance with the UHB peak of the XAS spectrum, at momentum transfer $q_{\mathrm{leg}} = 0.28$ reciprocal lattice units (r.l.u.), and $t = 0.4$ ps. The dd and charge transfer (CT) excitations are indicated with labels. The dashed box denotes the spectral range of interest for collective charge excitations. (c) trRIXS spectra after subtraction of the dd excitations (see Supplemental Material Sec. 2.2) at $q_{\mathrm{leg}} = 0.06$, 0.16, and 0.28 r.l.u.

which contributes to the quasielastic intensity. Although charge order is most prominent at resonance with the ZRS peak, it is still visible at the UHB resonance. We show in Fig. 2(b) representative raw trRIXS spectra at $q_{\mathrm{leg}} = 0.28$ r.l.u., where the dashed box marks the spectral region of interest in this Letter. Since the tails of the dd excitations extend into this region, we first fit and subtract the dd contributions to isolate features that emerge from the optical excitation (see Supplemental Material Sec. 2.2 and Figs. S4–S7 [45]), and use these dd-subtracted spectra as the basis for all subsequent analysis. Upon excitation into the metastable phase, the trRIXS spectra exhibit pronounced momentum-dependent reshaping at energies below 1.2 eV [Fig. 2(c)]. At $q_{\mathrm{leg}} = 0.16$ r.l.u., the quasielastic peak is enhanced while the finite-energy-loss spectrum remains unchanged. By contrast, at $q_{\mathrm{leg}} = 0.06$ and 0.28 r.l.u., the quasielastic response is largely unaltered and the inelastic spectral weight becomes strongly enhanced up to 1.2 eV. These momentum-dependent spectral changes are consistent with the emergence of a new collective excitation.

We construct energy-momentum maps of the trRIXS spectra to resolve its dispersion. We rotate the sample angle while keeping the x-ray analyzer fixed, thereby scanning the momentum transfer along the ladder direction. Figures 3(a)–3(c) display the resulting spectra as a function of q_{leg} (parallel to the ladder legs) and $q_{\perp}$ (normal to the ladder planes). The equilibrium map [Fig. 3(a)] reveals the tail of the charge order reflection, together with a broad continuum of excitations up to ~0.6 eV, attributed to the $\Delta S = 0$ two-triplon continuum [57]. The transient map in the metastable phase [Fig. 3(b)] shows an enhanced quasielastic charge order intensity. Taken together with prior trXRD measurements [37], this indicates a reduction of out-of-plane correlations (along $q_{\perp}$) that enhances the tail of the charge-order reflection, while leaving the in-plane correlation length unchanged. More strikingly, the transient RIXS signal exhibits a momentum-dependent enhancement of spectral weight up to 1.2 eV [Fig. 3(b)]. To highlight these changes, we show the difference between transient and equilibrium maps in Fig. 3(c), revealing a well-defined and strongly dispersive mode

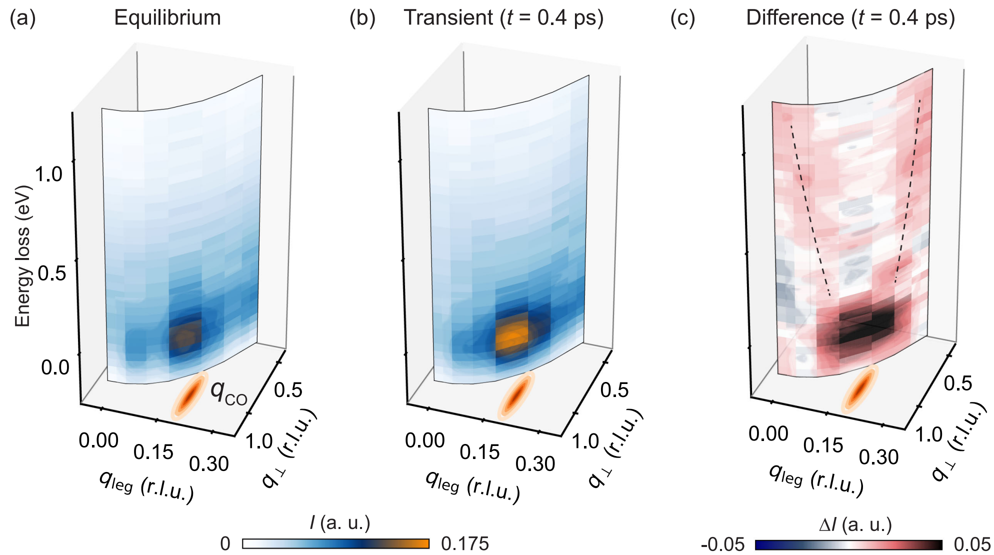

FIG. 3. (a) Equilibrium, (b) transient, and (c) difference trRIXS momentum-energy intensity maps at $t = 0.4$ ps, as a function of $q_{leg} \| c$ and $q_\perp \| b$. q_{CO} marks the charge order wave vector [38]. Dashed lines in (c) are guides to the eye.

centered at the tail of q_{CO}. The time evolution of the trRIXS spectra (Fig. 4) shows that the transient reshaping, including the high-energy enhancement, remains unchanged at all positive time delays, mirroring the trXAS signal at the UHB and indicating that the collective mode emerges in the metastable state.

We quantify the mode dispersion by fitting the momentum-dependent trRIXS spectra. Our prior Cu L-edge trRIXS experiments showed that metastable hole transfer disrupts ladder spin singlets, suppressing the $\Delta S = 1$ two-triplon continuum intensity without any measurable energy shift and indicating that the exchange coupling constants remain unchanged [37]. This observation, together with prior O K-edge RIXS measurements at equilibrium [57] and theoretical expectations for multi-triplon continua [58], constrains the O K-edge trRIXS analysis (see Supplemental Material Sec. 2.3 [45]). We constrain the equilibrium two-triplon parameters to ensure a nondispersive peak center, consistent with prior high-resolution RIXS measurements [57]. In the transient spectra, we include an additional Gaussian to capture the high-energy enhancement. To ensure the transient fits converge to physically meaningful parameters, the two-triplon frequency, linewidth, and asymmetry are constrained to vary by less than 20% relative to their equilibrium values, and the two-triplon amplitude is restricted to not exceed its

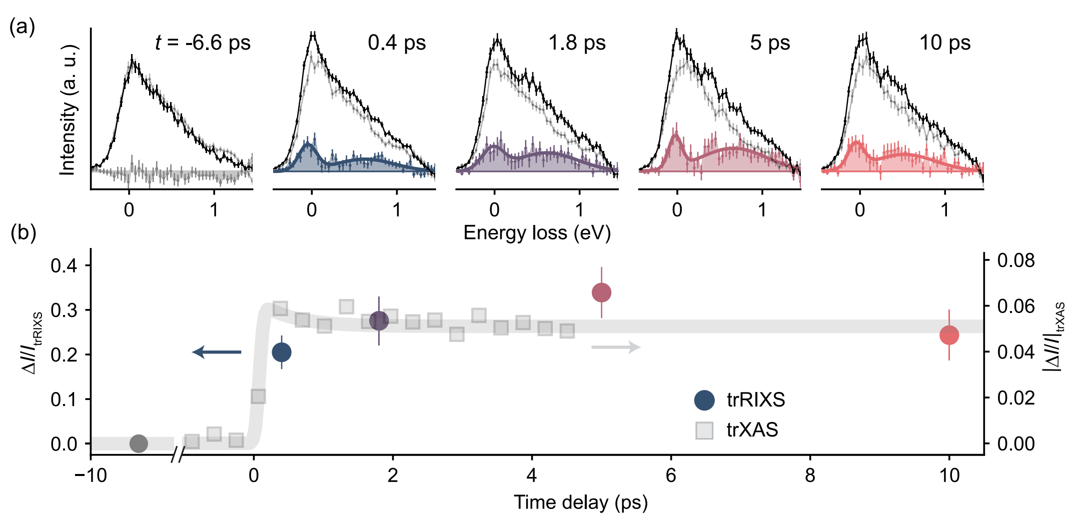

FIG. 4. (a) Equilibrium (gray), transient (black), and difference (color) trRIXS spectra at $q_{leg} = 0.25$ r.l.u. for time delays spanning $t = -6.6$ to 10 ps. (b) Differential trRIXS intensity integrated from −0.5 to 1.2 eV, plotted as a function of t. The absolute value of the differential trXAS signal at resonance with the UHB peak is reproduced from Ref. [37]. The gray line is a fit to the trXAS data.

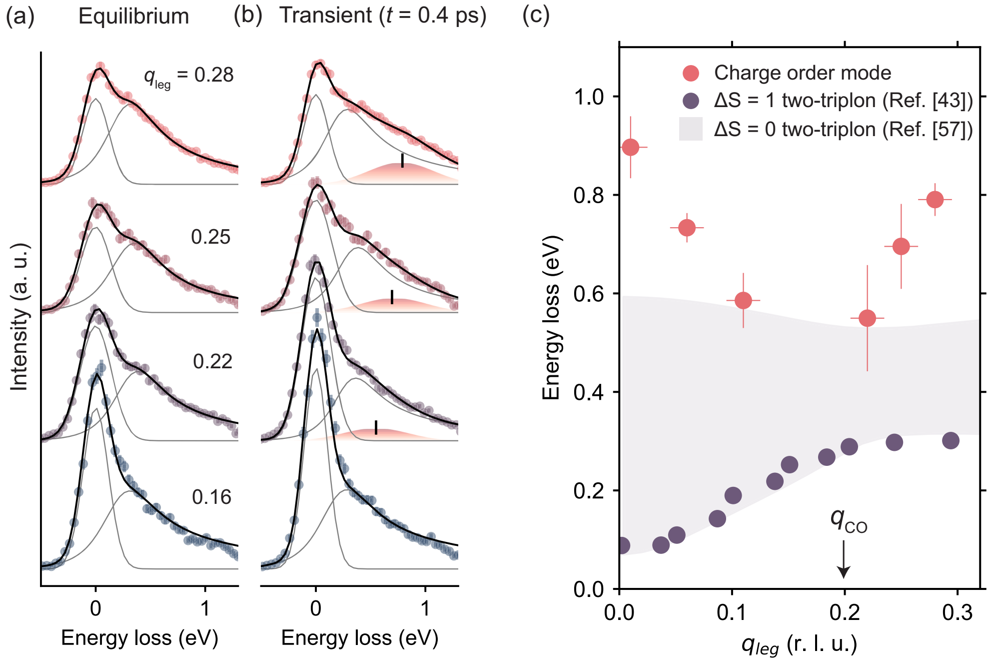

FIG. 5. (a),(b) Momentum-dependent RIXS spectra in and out of equilibrium, vertically offset for clarity. Momentum is given in r.l.u. along the ladder direction. Equilibrium spectra in (a) are fit with a Gaussian for the quasielastic peak and an asymmetric Lorentzian for the $\Delta S = 0$ two-triplon (thin black lines). Transient spectra in (b) include an additional Gaussian component (shaded red). (c) Dispersion of the emergent collective mode (red markers) extracted from fits in (b). The shaded gray area denotes the theoretical $\Delta S = 0$ two-triplon continuum from Ref. [57], while purple markers indicate the lower boundary of the $\Delta S = 1$ two-triplon continuum from Ref. [43]. Error bars represent instrumental uncertainties in momentum transfer and energy-loss reference point.

equilibrium value. We plot a subset of the fits in Fig. 5 (see Fig. S9 for the full dataset), while the resulting best-fit parameters are reported in Tables S3 and S4.

The resulting dispersion of the emergent mode is shown in Fig. 5(c). These fits reproduce the observed spectral redistribution without any systematic shift of the magnetic continuum. In the metastable state, the intensity of the $\Delta S = 0$ continuum is reduced by ~10%, consistent with the suppression of the $\Delta S = 1$ spin-excitation intensity at the Cu L edge [37,58]. In contrast, fitting the transient spectra without the additional term requires an overall blueshift of the $\Delta S = 0$ continuum to reproduce the high-energy enhancement, at odds with our prior measurements [37]. The energy scale and dispersion of the light-induced mode are also incompatible with a magnetic origin. Its propagation velocity (2.1 ± 0.3 eV Å along c) far exceeds that of $\Delta S = 1$ magnetic excitations in this ladder compound [43] [lower boundary shown in Fig. 5(c)] and those observed in other cuprates. Moreover, it approaches energy scales larger than any magnetic excitation, including the $\Delta S = 0$ two-triplon continuum that dominates the equilibrium RIXS spectrum at the O K-edge UHB [57]. Taken together, these observations identify the feature as a charge collective mode.

We now discuss the assignment of this emergent charge mode. This excitation has not been observed in previous equilibrium RIXS studies [57], raising the question of its origin. Its energy scale and propagation velocity resemble those of acoustic plasmons recently reported in electron- and hole-doped cuprates [59–62]. However, acoustic plasmons disperse symmetrically about $q = 0$, in sharp contrast to our observation of a branch centered at the charge order wave vector q_{CO}. No counterpart is detected at $q_{leg} = 0$, arguing against a zone-folding replica as the origin of this mode. Although weak coupling theories are not necessarily accurate for this material, for completeness, we performed random phase approximation (RPA) calculations (see Fig. S10), which support our conclusion that the observed mode is inconsistent with acoustic plasmons. We furthermore exclude the lower edge of the particle-hole continuum near $2k_F$ as the origin. Such excitations are indeed detectable by RIXS, as shown in recent measurements [63]. However, $2k_F = (1 - p)/2$ in one-dimensional systems [64,65], which for our experimental hole densities $p < 0.10$ corresponds to $q_{leg} > 0.45$ r.l.u., well beyond the momentum range where we observe the charge mode.

The dispersion centered on q_{CO} is consistent with a charge order collective mode. In a static charge-ordered state, the dynamical charge structure factor carries spectral weight predominantly at zero energy, whereas fluctuating order shifts spectral weight to finite energies [66,67]. Ultrafast resonant soft x-ray diffraction reveals a partial suppression of the static charge-order peak in the metastable state [37], while our current O K-edge trRIXS measurements show a concurrent enhancement around q_{CO} at finite energy loss. The absence of any measurable shift in the charge-order wave vector [37] rules out a sliding charge-ordered state [68], indicating that the melting of charge order proceeds through fluctuating, rather than translational, dynamics. The propagation velocity of our collective mode is comparable to that of dispersive charge-order excitations detected at equilibrium by RIXS at both the Cu L edge [69–73] and the O K edge [72,74]. Notably, it is also of the same order as the ladder quasiparticle dispersion (1.4 eV Å) measured by angle-resolved photoemission [75], suggesting that free and periodically modulated carriers propagate similarly within the metastable state. However, in contrast to equilibrium charge-order fluctuations, these dispersive charge excitations emerge only in the optically induced metastable state and are observed directly in the transient trRIXS spectra, rather than indirectly via Fano interference with phonons.

To understand the enhanced charge fluctuations in the metastable state, we performed density matrix renormalization group (DMRG) calculations on a single-band Hubbard ladder as a function of hole doping p (see Supplemental Material Sec. 4 [45]). We adopt model parameters from previous fits to experimental data [43], which have been shown to capture the spin dynamics of cuprate ladders at equilibrium [43,44] and in the metastable state [37]. We examine the charge properties of the ground state by calculating the real-space distribution of charge correlations. These charge correlations exhibit a power-law decay at long distances, characterized by the charge exponent K_ρ. Upon transiently doping the ladder with holes from the chains, our calculations reveal a doping-driven crossover accompanied by an order-of-magnitude enhancement of charge fluctuations. For $p < 0.08$, the ground state features rapidly decaying charge correlations, as evidenced by the large K_ρ, and indicative of suppressed charge fluctuations. As the hole density increases, these fluctuations grow markedly, as signaled by a sharp decrease of K_ρ. We identify a crossover between these regimes near $p = 0.1$ (Fig. S11). While charge order in the parent compound $Sr_{14}Cu_{24}O_{41}$ cannot be quantitatively reproduced by the single-band Hubbard model [76,77], our DMRG results nevertheless provide a theoretical basis to understand the charge mode observed in the metastable state [37]. At equilibrium, the ladder subunits in $Sr_{14}Cu_{24}O_{41}$ are self-doped with $p = 0.06$, while in the metastable state, the photoinduced hole transfer $\Delta p = 0.03$ drives the system toward a highly fluctuating regime. These fluctuations, in the presence of the partially melted charge order, manifest as the dispersive collective mode observed at q_{CO}.

Our experiments reveal an emergent charge order mode in the electronic metastable phase of $Sr_{14}Cu_{24}O_{41}$. Following the optically induced transfer of holes from the chain reservoirs to the ladders, the charge-ordered state partially melts and evolves into a fluctuating phase featuring dispersive charge excitations extending up to 0.8 eV. This collective mode appears as a distinct hallmark of the light-induced metastable state, absent from the equilibrium RIXS spectrum and unreported in prior studies [57]. Our findings have implications for the physics of metastable phases and dynamical ordering phenomena in correlated systems. The observation of a collective mode emanating from the charge-order wave vector indicates that the charge-ordered phase enters a fluctuating regime in which correlated carriers in the UHB acquire itinerant character at finite momentum. This nonequilibrium state provides a promising platform for exploring dynamical pairing instabilities in cuprates. In two-dimensional cuprates at equilibrium, charge order competes with superconductivity, and its optical suppression leads to transient superconducting-like states [7,8,78,79], consistent with theoretical expectations for nonequilibrium competing orders [29,80]. Cuprate ladders display similar competition [39,81–83] together with strong hole pairing [42–44], suggesting that their fluctuating charge-ordered state could be driven toward a metastable superconducting or η-paired condensate under suitable conditions. Our results thus motivate future optical and x-ray scattering experiments to search for signatures of such hidden condensates [84] in the spin and charge structure factors of light-driven metastable electronic states [25].

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Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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