Light-enhanced Charge Density Wave Coherence in a High-Temperature Superconductor

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Quantum materials are often characterized by intertwined and competitive orders. The competition between superconductivity and charge density waves (CDW) in high-$T_C$ cuprates is a paradigmatic example. External tuning knobs, such as high-magnetic-fields and uniaxial strain, have been successfully employed to investigate their interaction. However, time-averaged steady-state experiments cannot capture the dynamical evolution of these quantum orders on the natural timescale of their interaction. Here we use ultrafast resonant soft X-ray scattering to track the transient evolution of CDW correlations in YBa$_2$Cu$_3$O$_{6+x}$ following the quench of superconductivity by an infrared laser pulse. The CDW order reacts...
to the quench of superconductivity on the picosecond timescale with a large enhancement of spatial coherence, nearly doubling the correlation length. We capture a CDW state which is profoundly non-thermal and distinct from the high-magnetic-field state observed in steady-state conditions. From this ultrafast snapshot we infer the existence of a superconductivity-induced phase modulation of CDW, yielding novel insight into the interaction between intertwined orders.

Unconventional superconductivity often emerges in the proximity of other low-energy broken-symmetry states, including antiferromagnetism, spin order, and charge density waves (CDW). These orders exist on similar energy scales and often become “intertwined” (1), showing a complex interplay arising from strong correlations. The presence of incommensurate CDW order is ubiquitous in cuprates (2, 3), and has been detected by resonant X-ray scattering (4, 5) and hard X-ray diffraction (6). This CDW order coexists with superconductivity below the critical temperature \( T_C \), and its temperature dependence suggests a tendency to compete with superconductivity (4). In YBa\(_2\)Cu\(_3\)O\(_{6+x}\) (YBCO), strong magnetic fields suppress superconductivity and enhance the underlying CDW order (6, 7). More recently, CDW enhancement has been achieved by means of applying uniaxial pressure (8), yet dynamical information about the interaction between order parameters is accessible from neither high-magnetic-field nor uniaxial pressure experiments. Therefore, the dynamical response of the CDW state following a quench of superconductivity is still unknown.

An emerging approach to explore intertwined phases is to measure their dynamical response to ultrafast photo-excitation in the time domain. Ultrafast laser pulses may enable selective quenching of phases, thereby inducing an enhancement of their competing counterparts (9-11) and launching a Higgs mode dynamic response (12). The use of free electron lasers enables such studies on CDW systems. However, ultrafast X-ray scattering studies on charge-ordered systems have focused so far mostly on photo-induced melting dynamics (13-16). In this work we employ ultrafast photo-excitation to quench the superconducting state of a YBCO single crystal, while the CDW state is not directly perturbed by the light pulse. We use ultrafast X-ray scattering to probe the reaction of CDW to the quench of superconductivity, thus exploring their dynamical interaction. We demonstrate that the laser-driven quench of the superconducting order transiently enhances the spatial coherence of CDW modulations in YBCO in a non-thermal fashion. By studying this
snapshot of the CDW spatial correlations we infer the existence of a superconductivity-induced phase modulation of CDW.

We track the CDW dynamical response to a laser-driven ultrafast quench of superconductivity by performing resonant soft X-ray scattering measurements at the LCLS free electron laser facility. To probe the electronic modulation connected to the CDW phase we used an incident photon energy of 931.5 eV, resonant with the Cu L₂-edge, using a π-scattering geometry and scanned the momentum transfer around wavevector $\mathbf{Q} = (0.31, 0, 1.4)$ (see Fig. 1A). The sample was photo-excited by 100-fs infrared laser pulses, variably delayed in time with respect to the 100-fs X-ray pulses. The doping of the sample ($x = 0.67$) was selected to be at the maximum point of the CDW dome (Fig. 1B), where the strongest competition with superconductivity occurs. The experiment was performed in the superconducting phase ($< 20$ K), and for comparison at $T_C = 65$ K, where the CDW scattering peak intensity is maximal (Fig. 1A and (17)). 800-nm laser pulses are known to strongly perturb superconductivity in cuprates, producing a non-thermal photo-induced phase transition within a few hundreds of femtoseconds (18-21). The fluence of the pump pulses was kept close to the minimum needed to quench the superconducting phase (19) ($\approx 50$ µJ/cm²), and such that parasitic local heating effects could be avoided.

The photo-induced reaction of the CDW state is readily observable in the time-domain response, by probing the peak intensity integrated around $\mathbf{Q}$ at varying time delays with respect to the pump pulse. The pump induces a prompt resolution-limited decrease of the CDW peak intensity at 65 K (Fig. 1C), followed by a slower $\approx 3$ ps recovery of the CDW state. This is similar in terms of dynamics to the well-known photo-induced melting in other charge ordered systems (13, 22-24). At 65 K, a relatively high fluence pump ($164 \pm 25$ µJ/cm²) was used to obtain good signal-to-noise for the melting process. On the contrary, at 20 K the pump yields the opposite effect, inducing an enhancement of the CDW peak signal with noticeably dissimilar dynamics (Fig. 1D). Here the low pump fluence ($45 \pm 20$ µJ/cm²) produces a large and positive CDW modulation, with much slower rise time ($\approx 1.5$ ps), and a considerably longer decay time, $\tau \approx 7$ ps. We stress that the pump fluence used here does not induce significant thermal effects, with a maximum temperature rise of $\approx 20$ K.

The dotted line in Fig.1D indicates the estimate of maximum heat-induced changes obtained by interpolating the equilibrium data. When raising the fluence up to 164 µJ/cm² the 20 K response presents a combination of both a fast quench and a slow delayed enhancement (Fig. S3).
enhancement signal is already saturated at this fluence (Fig. 1E), whereas the CDW melting signal is still growing with increasing fluence. Notably this enables full optical control over the fate of the CDW, switching between its enhancement and suppression by a mere tuning of laser power.

To better understand these results and draw a direct comparison with the dynamics of the superconducting order, we performed complementary transient optical reflectivity measurements at 800nm. At this wavelength we expect to observe the well-known spectral weight transfer from the low-energy superconducting gap to interband transitions (25, 26). The transient optical reflectivity signal is dominated by the superconducting response, as confirmed by its temperature and fluence dependence (Fig. S4), and by comparing our results to transient MIR spectroscopy resonant to the superconducting gap on an analogous sample (Fig. S5) (27). The dynamics show typical features resulting from the quench of the superconducting state – the signal saturates at a critical fluence, \( \Phi_c \) (20, 28) (Fig. 1E) and a flat-top dynamics develops when pumping well above the critical fluence (29) (Fig. S4). Remarkably, the superconducting state quenches within \( \approx 300 \) fs, while the CDW order reacts only after \( \approx 1.5 \) ps, indicating a highly non-thermal balance between the two orders within the first picoseconds. Superconductivity recovers at longer delays on a similar timescale as the CDW (Fig. 1D). When we normalize the \( \Delta R/R \) signal by the saturated value, we note that at the fluence of \( \approx 50 \) \( \mu \)J/cm\(^2\) superconductivity is almost completely quenched (> 90%) in the probing volume. Our measurements capture remarkably distinct dynamics for the superconducting and CDW orders, revealing a time window of \( \approx 2 \) ps to explore unbalanced and dynamically interacting orders beyond thermal equilibrium.

To obtain a snapshot of the CDW spatial correlations following photo-excitation, we collected cuts along the \( H \)-direction in reciprocal space (Fig. 2) at the delay times where photo-induced variations are maximal (\( \approx 0.5 \) ps at \( T_c \) and \( \approx 2 \) ps below \( T_c \)). At 65 K the CDW peak signal suppression is due to a decrease of integrated intensity (\( \approx 16\% \)), as well as a decrease of correlation length (\( \approx 25\% \)). This reaction is comparable to the CDW melting observed in tri-tellurites (30), but differs from the ultrafast charge order melting in nickelates and manganites, where no change of correlation length was observed (13, 23). When the system is cooled to well below \( T_c \) (12 K, Fig. 2B), the \( H \)-scan reveals a considerable CDW peak enhancement following photo-excitation, amounting to an increase in peak strength of \( \approx 120\% \). The CDW peak increase is mostly due to the dramatic peak narrowing, indicating a \( \approx 90\% \) increase of correlation length, from \((36 \pm 3) \) nm
to (69 ± 5) nm, i.e., almost a doubling in size of coherent CDW domains. Concurrently, the integrated intensity increases, but by a smaller amount (10-20 %). We also report a photo-induced momentum shift of the peak of ≈ 0.003 r.l.u. Similar results were obtained using a different detector and at T = 20 K (Fig. S6).

It is important to remark that the CDW transient state achieved in our experiments cannot be explained by simple thermal effects. To this end, we compare the correlation length and integrated intensities obtained at LCLS with a complete temperature dependence obtained at the Canadian Light Source (CLS) synchrotron (Figs. 2C and D). The results at negative delays obtained with the free electron laser at 65 K and 12 K (blue symbols) coincide with the equilibrium characterization performed at CLS (black symbols). The pump induced values (red symbols) for the correlation length and integrated intensity clearly do not follow an effective temperature pattern. The increase in correlation length is well beyond what can be achieved thermally, whereas the integrated intensity increase remains small compared to the temperature dependent swing. These data clearly testify the non-thermal nature of the photo-induced state created by the 800-nm laser pulse.

The ability to measure both CDW and superconductivity on the same sample with similar excitation conditions provides unprecedented insights in the dynamics of intertwined orders. Time-dependent Ginzburg-Landau (TDGL) theory provides a framework to interpret the dynamical interplay between interacting orders (9, 31). The theory predicts that for homogenous and competitive orders, the CDW order parameter amplitude would increase on a timescale of several picoseconds, driven by the quench of superconductivity (see SI). This scenario is depicted in Fig. 3A together with the expected scattering profile. The fact that the CDW integrated intensity change is much smaller than the correlation length variation is in stark disagreement with the prediction of TDGL for coupled order parameters (cfr. SI and Refs. (11)). We thus conclude that a simple competition model, assuming locally coexisting orders, is incompatible with our results.

A second scenario is that of phase separated strongly competitive orders. In this case we consider well separated CDW and superconducting domains, following for example the work of Campi et al. (32), with an average spacing between neighboring CDW domains larger than the CDW periodicity (> 1 nm). In this scenario, represented in Fig. 3B, the correlation length $\xi$ of CDW domains expands because of the suppression of neighboring superconducting domains. This inflation however implies a contemporaneous increase of integrated intensity due to the expanding
CDW filling factor, scaling as $\propto \xi^2$. Such scaling is again incompatible with our data, suggesting that domain expansion does not explain the dramatic correlation length increase we observe within $\approx 2$ ps after photo-excitation. Even in the case of a unidirectional expansion of domains, where the filling factor scales linearly with $\xi$, this scenario still does not agree with the data (Fig. 3B).

To explain our data, we resort to a third scenario, illustrated in Fig. 3C, where a CDW domain loses phase coherence due to the formation of an interstitial superconducting region within the domain. The sudden photo-quench of superconductivity allows the domain to reestablish phase coherence by a slide of the CDW pattern. In this process superconductivity acts as a topological defect for the CDW pattern, which is removed by the pump pulse allowing a fully coherent CDW domain to form again. This model, while potentially representing an oversimplification of the spatial arrangement of the superconducting and CDW regions, captures the salient aspects of the data (Fig. 3C), where the correlation length nearly doubles, without an increase in amplitude and filling factor. In this scenario, we can tentatively explain the momentum shift as originating by a $\approx 1\%$ compression of the periodicity as CDW accommodate the defect within the domain.

The scenario proposed here, in which superconductivity acts as topological defect for CDW, has been discussed theoretically (33, 34). In this theoretical picture the CDW is the dominant order, while superconductivity is sub-dominant. Superconductivity may lower the energy cost for a CDW dislocation, hence increasing their density distribution. Based on our data we can infer that the average spacing of such objects is $\approx 3$–4 nm, thus limiting the CDW correlation length below $T_c$. Importantly, our experiment directly relates this effect to superconductivity. On the contrary, as reported in (35), the effect of disorder is quite different, as it lowers the CDW amplitude, without a significant effect on the correlation length. Interestingly, we note that high-magnetic-field and uniaxial pressure experiments in steady-state conditions show an increase of the integrated intensity in addition to the increase of correlation length (6-8). Whether this different behavior is related to the fact that light-excitation does not decrease the effect of disorder, or to a distinction in the timescales involved remains to be clarified.

An alternative theoretical representation of scenario 3 can be formulated considering a phase modulation of the underlying CDW order such as that $\rho(r) \propto \exp(iQ \cdot r + \phi \cos(q \cdot r + \alpha))$, where $\rho$ is the electron density, $q$ is a supermodulation of the CDW phase, $\phi$ is the strength of the modulation and $\alpha$ is a phase offset. Such phase patterning was initially proposed by McMillan (36)
for a CDW supported by an electron fluid. Such superlattice modulation was inferred experimentally in dichalcogenides (37), and may provide a spatial texture for the formation of superconductivity. This model is consistent with our observations in YBCO, with the important distinction that the energy gain from such modulation arises directly from the interaction between superconductivity and CDW, rather than from the vicinity to a commensurate order (see SI).

In each of the theories proposed for scenario 3, an ultrafast sliding of the CDW patterns is required to restore a common phase pattern across the CDW domain. Such motion of the CDW condensate thus excludes strong pinning to defects. We note that possible CDW sliding effects have been recently reported in ultrafast experiments during the photo-induced melting in LBCO (38). The other important common trait between these theories is that superconductivity forms within a CDW domain. This is quite distinctive to a phase separated state for strongly competitive orders and it is a testament to the fundamental intertwining between superconductivity and CDW.

In summary, by combining the optical quench of superconductivity and the ultrafast probe of the CDW order parameter via ultrafast X-ray scattering, we propose a fundamental intertwining between CDW and superconducting domains, where superconductivity acts as a topological defect for CDW. This approach creates new opportunities to study competing orders in addition to current steady-state methods, such as high-fields and uniaxial pressure experiments. By observing the early state following the quench of one of the order parameters, this method allows sensitive detection of spatial patterns of intertwined orders in – and out-of – their ground state.

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Fig. 1. **Experimental conditions and time-domain results.** (A) Diagram of the experiment. An infrared laser pulse perturbs the sample while an X-ray pulse resonant with the Cu $L_3$-edge (931.5 eV) probes the underlying CDW phase around the wavevector $\mathbf{Q} = (0.31, 0, 1.4)$. The momentum space is explored by rocking the sample and acquiring the scattering maps onto a detector. Both a 2D area detector and a point detector were used for this experiment. Two representative $H$-scans are shown in the inset representing the CDW peak, after subtraction of fluorescence background, at a temperature of 12 and 65 K. The x-ray absorption spectra and the full temperature dependent scattering profiles at equilibrium are shown in Fig. S1 and S2 respectively. (B) Schematic phase diagram of YBa$_2$Cu$_3$O$_{6+x}$ showing the two temperatures of interest, within ($T = 20$ K) and just above ($T = 65$ K) the superconducting dome at the doping level $x=0.67$. (C) Pump-probe measurement of the CDW signal at 65 K with a laser pumping fluence of (164 ± 25) µJ/cm$^2$. (D) Optical (blue circles) and X-ray scattering (purple circles) signals measuring the superconducting and CDW dynamics at 20 K with a laser pumping fluence of $\approx 50$ µJ/cm$^2$. The solid purple line indicates a fit of the X-ray scattering dynamics, and the dashed line is the extrapolated curve when the residual melting signal is removed. The gray dotted line represents quasi-thermal heating effects assuming full absorption of the pump energy into material without diffusion. To match the fluence value, the optical data was obtained by averaging the two closest curves from the full fluence dependence characterization (Fig. S3). (E) Fluence dependence of the transient optical signal (blue solid circles) and the maximum X-ray scattering photo-induced variation (purple open circles) at 20 K. The blue line is a fit with an exponential function representing the saturation of the superconducting signal.
Fig. 2. X-ray scattering profiles of CDW peak before and after photo-excitation. (A-B) $H$-scan plots of the elastic X-ray scattering signal, after background subtraction, measured along wavevector $\mathbf{Q} = (0.31,0,1.4)$ with photon energy 931.5 eV in equilibrium (yellow symbols in A, and gray symbols in B) and following photo-excitation (red symbols in A, purple symbols in B) at 65K (A) and 12 K (B). The pump fluence was $\approx 50 \mu J/cm^2$. Error bars are 1 SD. (C-D) Correlation length along the $a$ crystal direction (C) and integrated intensity of the CDW peak (D) as a function of temperature measured at CLS (black squares), and at LCLS at negative delays (blue triangles), and around $\approx 2$ps delay time (red triangles). For the data at 65K the 2ps values were extrapolated from the 0.5 ps assuming the decay dynamics reported in Fig. 1C. Note: CLS data were corrected to account for the angular detector resolution, while for the case of LCLS this effect was minimal.
Fig. 3. Scenarios for SC and CDW spatial arrangements. (A-B-C) Schematics of the proposed scenarios for the spatial arrangements of CDW and SC regions. The first column represents the ground state in each case, while the middle columns represent the state following the quench of the superconducting state by a laser pulse. The third column represents the expected scattering profile for each of these cases for the equilibrium (gray solid line) and photo-excited case (dashed purple line). The profiles are compared to the experimental data following photo-excitation (purple solid symbols and line). (A) represents the case of coexisting SC and CDW orders, (B) the case of mesoscale (>1 nm) phase separation between SC and CDW domains, (C) the case of SC developing as a topological defect within the CDW domain. The data are well reproduced in this case.
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