# Femtosecond x rays link melting of charge-density wave correlations and light-enhanced coherent transport in $YBa_2Cu_3O_{6.6}$

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We use femtosecond resonant soft x-ray diffraction to measure the optically stimulated ultrafast changes of charge-density wave correlations in underdoped $YBa_2Cu_3O_{6.6}$. We find that when coherent interlayer transport is enhanced by optical excitation of the apical oxygen distortions, at least 50% of the in-plane charge-density wave order is melted. These results indicate that charge ordering and superconductivity may be competing up to the charge ordering transition temperature, with the latter becoming a hidden phase that is accessible only by nonlinear phonon excitation.

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Hole doping of cuprates removes the antiferromagnetic order of the parent compound and promotes unconventional high-temperature superconductivity. A key ingredient in determining the critical temperature $T_C$ is the competition between superconducting phase coherence and charge or spin orders. A vivid demonstration of this interplay is the frustration of interlayer coupling by charge stripes around 1/8 doping in single-layer materials such as $La_{2-x}Sr_xCuO_4$ and $La_{2-x}Ba_xCuO_4$ [1–4].

Most recently, it was shown that the "flattening" of the superconducting-to-normal-state phase boundary in $YBa_2Cu_3O_{6+x}$ near $x = 0.6$ (∼12.5% hole concentration) is coincident with the appearance of biaxial charge-density wave (CDW) order [5–9]. Similar observations have been made in other high-$T_C$ cuprates, including $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ [10] and $HgBa_2CuO_{4+\delta}$ [11]. The interplay of charge order competing with superconductivity appears then to be a general phenomenon in the physics of these systems.

Although pressure [12,13] and magnetic fields [14] have long been used to affect this interplay at low temperatures, only recently it was shown that high-frequency optical pulses achieve a qualitatively similar effect over larger temperature ranges. For example, coherent interlayer coupling was induced in the low-temperature stripe-ordered phase of $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$ [15] and $La_{2-x}Ba_xCuO_4$ [16], likely caused by the melting of the charge stripe order [17].

In $YBa_2Cu_3O_{6+x}$, optical excitation of apical oxygen distortions has been shown to cause an even more striking effect, enhancing coherent interlayer transport below $T_C$ and inducing a transient state above $T_C$ with important similarities to the equilibrium superconductor [18]. This effect was recently shown to involve redistribution of the tunneling strength from the intrabilayer to the interbilayer regions of the unit cell [19] and a rearrangement of the lattice structure that could not be achieved at equilibrium [20].

Here, femtosecond resonant soft x-ray diffraction (RSXD) is combined with time-resolved THz spectroscopy to measure the response of the in-plane charge order in $YBa_2Cu_3O_{6.6}$. We wish to establish if the enhancement of coherent interlayer coupling involves a reduction of CDW order. We show that prompt reduction of the CDW resonant soft x-ray diffraction peak occurs as the material is transformed into the coherent state, providing a key microscopic ingredient for this class of phenomena.

Detwinned samples of $YBa_2Cu_3O_{6.6}$ were synthesized by the self-flux method. The equilibrium $c$-axis optical properties at $T = 20$ K, below the superconducting transition temperature $T_C = 62$ K, are reported in Fig. 1 for frequencies between 0.5 and 2.5 THz. Quasi-single-cycle THz-frequency pulses were generated by either optical rectification or by a photoconductive antenna and measured after reflection from the sample by electro-optic sampling. The reflected field was referenced to the same measurement made above $T_C$ and to literature data. The equilibrium reflectivity displays the Josephson plasma edge [Fig. 1(b)], a signature of supercurrent oscillations between capacitively coupled $CuO_2$ bilayers. As this is a longitudinal plasma excitation and involves a zero crossing of $\mathrm{Re}[\varepsilon(\omega)]$, a peak in the loss function $-\mathrm{Im}\,1/\epsilon\,(\omega)$ is also observed, as displayed in Fig. 1(c).

Upon excitation with 300-fs-long pulses at 15-$\mu$m wavelength, made resonant with the $B_{1u}$ infrared-active lattice distortion (670 $\mathrm{cm}^{-1}$) sketched in Fig. 1(a) [21], the same optical properties observed at equilibrium below $T_C$ appeared transiently in the normal state. The lower panels in Figs. 1(b) and 1(c) report a representative example of the photoinduced

 

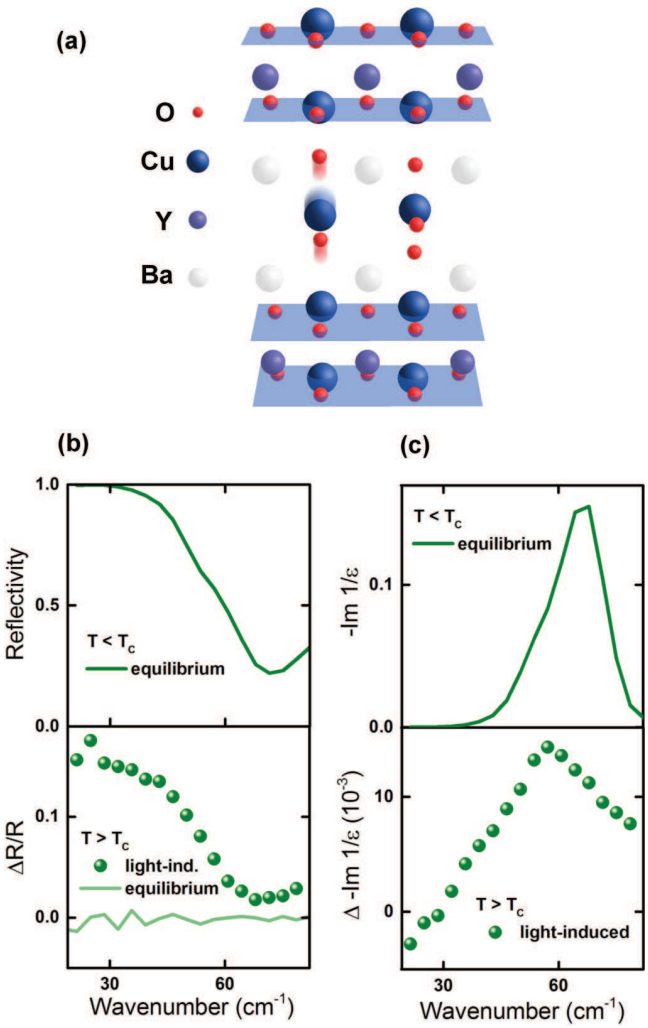


FIG. 1. (Color online) (a) Crystal structure of orthorhombic $YBa_2Cu_3O_{6.6}$ and the motion of the apical oxygen atoms (red shadows) associated with the resonantly excited $c$-axis $B_{1u}$ phonon mode. (b) Top: Below-$T_C$ (20 K) static frequency-dependent reflectivity of THz light polarized along the $c$ axis, clearly showing the Josephson plasma edge. The lower panel shows the light-induced reflectivity changes above $T_C$ (100 K, green dots). Here, the sample was excited with 300-fs pulses at 15 $\mu$m wavelength, polarized along the $c$ axis, and the data are taken at +0.8 ps time delay. At negative time delay (light green solid line), the sample does not react to the midinfrared excitation. (c) The static above-$T_C$ electron loss function is shown at the top. The lower panel depicts the light-induced change in the $T > T_C$ loss function for the same conditions as described in part (b).

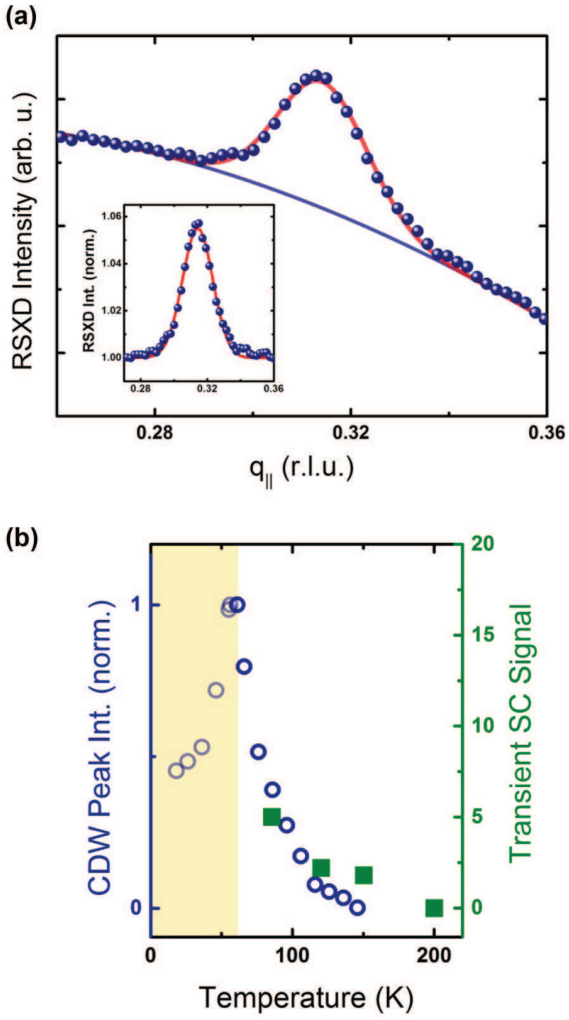


FIG. 2. (Color online) (a) RSXD scan of the $YBa_2Cu_3O_{6.6}$ CDW peak at 62 K, using $\pi$-polarized x rays at the Cu $L_3$ edge (blue data points), with $q_{||}$ the in-plane component of the diffraction wave vector along the (1 0 0) direction. The blue solid line is a polynomial fit to the fluorescence background. The inset shows the same diffraction peak normalized to this background and fitted with a Gaussian function. Data were taken using synchrotron radiation at the Diamond Light Source. (b) Temperature dependence of the integrated intensity of the CDW peak (blue circles) and of the volume fraction of the transient superconducting state.

optical properties [18], measured for $T = 100$ K and ~1 ps after excitation at a fluence of 4 mJ/cm$^2$. Details about the time-resolved THz probe experiment, including data analysis, are described in the Supplemental Material [22]. A reflectivity edge and a peak in the loss function are observed at $\omega_{JPR}$, underscoring transient interlayer (short-range) superconducting coherence. These effects can be induced only up to the temperature scale at which quasistatic charge order is observed ($T_{CO} \sim 160$ K), suggesting a link between the two phenomena.

This is clearly seen in Fig. 2(b), in which the strength of the static charge order, as revealed by $\pi$-polarized x-ray diffraction in resonance with the Cu $L_3$ edge (931.5 eV) at the in-plane wave vector $q_{||} \sim 0.31$ [see Fig. 2(a)], is plotted alongside the strength (volume fraction) of the light-induced coherent state [18].

Femtosecond resonant soft x-ray diffraction experiments were carried out at the Stanford LCLS x-ray free electron laser (FEL) under the same excitation conditions. The sample was mounted onto the same in-vacuum diffractometer used for the measurements of Fig. 2(a), cooled to immediately above the critical temperature $T_C = 62$ K and excited by the same 15-$\mu$m-wavelength pulses used for the THz probe experiments of Fig. 1. The FEL photon energy was tuned to the Cu $L_3$ edge

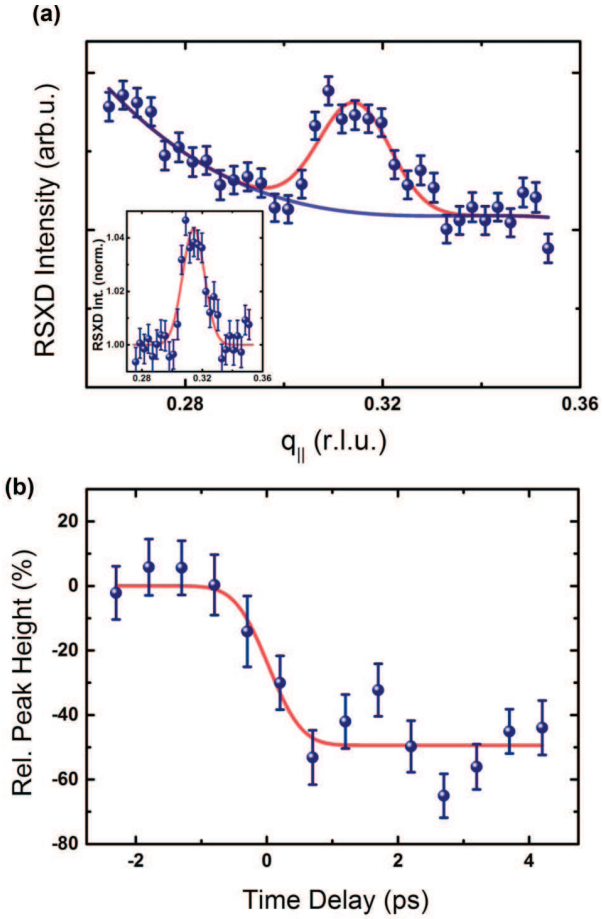


FIG. 3. (Color online) (a) The same RSXD scan as shown in Fig. 2(a), now measured at the Linac Coherent Light Source free electron laser. (b) Transient height of the CDW diffraction peak induced by direct excitation of the apical oxygen mode using 400-fs pulses at 15-$\mu$m wavelength, polarized along the $c$ axis.

and cut to 0.5-eV bandwidth by a grating monochromator. The diffracted x rays were detected as a function of pump-probe time delay using an avalanche photodiode, enabling pulse-to-pulse normalization to the incident x-ray intensity.

Figure 3(a) shows the steady-state measurement of CDW diffraction at $q_{||} \sim 0.315$ at the free electron laser. Although the noise level has clearly increased in comparison to the synchrotron radiation measurements shown in Fig. 2(a), the CDW related diffraction peak was detected at the same position in reciprocal space and with about the same amplitude and width above the fluorescence background (see inset). Figure 3(b) shows the transient change of the peak amplitude, normalized to the steady state after subtraction of the fluorescence background. Here, we assume that the fluorescence background is not altered on the ultrafast time scale. After excitation, the scattering signal reduced promptly to approximately half of its equilibrium value. Because the x-ray pulses were absorbed over a 200-nm layer and the excitation pulse was deposited over a $\sim$2-$\mu$m depth, the reduction in the scattering signal can be directly related to the melting of approximately 50% of the charge order.

The observed disappearance of charge order occurs on a time scale comparable with the appearance of the Josephson plasma edge, strongly indicating a correlation between the two phenomena. However, while the transient plasma edge survives only for 5–7 ps [18], the charge order remains melted for a significantly longer time. This is most likely due to the fact that interlayer coherence disappears immediately after the local lattice distortions are relaxed [20], but the recovery of charge order requires the buildup of correlations on longer length scales.

The reported melting of charge order is reminiscent of the physical origin of light-induced interlayer coherence in single-layer stripe-ordered cuprates at low temperatures [15,17]. In the striped compounds the effect was easily understood by considering frustrated Josephson coupling due to pair density wave order [23], yet the present result in $YBa_2Cu_3O_{6.6}$ does not lend itself to an equally simple interpretation.

The data are strongly indicative of a ground state in which charge order, or a fluctuating/intertwined state involving both charge and superconducting order [24], frustrate superconductivity at least immediately above $T_C$. The microscopic physics following the optical excitation is likely to involve anharmonic lattice motions [20,25], or alternatively a more complex stabilization effect for the light-induced coherent tunneling in the high-temperature state. Light-induced coherence appears at higher temperatures than residual phase coherence in the planes [26], indicating the presence of a hidden state invisible in equilibrium. Similarly, charge-order melting may also be invoked for the light-induced coherence obtained at lower doping values reported in Refs. [18,19], although in that case the ordering of charges appears on shorter range, and is not accessible with femtosecond x rays.

Theoretical efforts will be necessary to explain the observed experimental features in more detail. Future experimental work will focus on improvements in our ability to controlling light-induced melting of charge order, perhaps even minimizing dissipation to achieve steady-state coherence by continuous wave light excitation.

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