Transient atomic structure of vibrationally excited YBCO with enhanced superconducting coherence above Tc

R. Mankowsky[1], A. Subedi[2], M. Först[1], S.O. Mariager[3], M. Chollet[4], H. Lemke[4], J. Robinson[4], J. Glownia[4], M. Minitti[4], A. Frano[5], M. Fechner[6], N. Spaldin[6], T. Loew[5], M. Le Tacon[5], B. Keimer[5], A. Georges[2] and A. Cavalleri[1,7]

[1] Max Planck Institute for the Structure and Dynamics of Matter, Hamburg
[2] Ecole Polytechnique, Strongly Correlated Quantum Materials Group, Paris
[3] Paul Scherrer Institut, FEMTO Group, Villigen
[4] Linac Coherent Light Source, SLAC, Stanford
[5] Max Planck Institute for Solid State Research, Stuttgart
[6] Eidgenössische Technische Hochschule Zürich, Materials Theory, Zürich
[7] Department of Physics, Oxford University, Clarendon Laboratory, Oxford
roman.mankowsky@mpsd.cfel.de

Abstract: Nonlinear lattice excitation in $YBa_2Cu_3O_{6.5}$ has been shown recently to transiently induce signatures of superconducting coherence up to room temperature. Here, we present ultrafast x-ray diffraction measurements used to determine the corresponding atomic structure.

Introduction

Strong-field THz pulses can be used to resonantly drive the lattice modes of a solid, steering the atoms into transient arrangements [1, 2, 3] to induce new phenomena. Ultrafast insulator-metal transitions [4, 5], melting of electronic and magnetic orders [6], and light-induced superconductivity [7] have been demonstrated with this technique.

In $YBa_2Cu_3O_{6.5}$, the same approach has recently been shown to transiently enhance superconducting transport below Tc=52 K and to induce signatures of superconducting coherence up to room temperature [8, 9]. Here, ultrafast X-ray diffraction is used in combination with the same mode selective vibrational excitation to determine the corresponding atomic structure. Picometer c-axis expansion of the apical oxygens and unbuckling of the Cu-O bonds are found in the CuO_2 plane at sites with oxygen vacancies in the chains, while an increase in buckling is observed at other sites. We speculate that by reproducing the key elements of this transient crystal structure in an equilibrium solid, possibly though strain engineering, one may achieve higher temperature steady-state superconductivity.

Theory

The response of a crystal to infrared radiation tuned to a specific vibrational resonance can be described by separating the crystal Hamiltonian into its linear and a nonlinear term $H = H_{lin} + H_{NL}$. The first term dominates the response at low fields, where only the displacements along the directly driven phonon coordinate Q_{IR} are considered leading to a harmonic expression of the type $H_{lin} = (\mu_{IR}/2)\omega_{IR}^2 Q_{IR}^2$.

At higher driving fields, coupling to other modes with generic coordinates Q_R becomes important. Limiting the interaction to lowest order and considering centrosymmetric crystals, the nonlinear Hamiltonian can be written as $H_{NL} = (\mu_{IR}/2)\omega_R^2 Q_R^2 - a_{21} Q_{IR}^2 Q_R$. The key aspect of this coupling is that the anharmonically coupled mode experiences a *directional* force that is proportional to Q_{IR}^2, effectively producing a displacement along Q_R that survives as long as Q_{IR} is coherent. This process is the equivalent of optical rectification in nonlinear optics.

$YBa_2Cu_3O_{6+x}$ crystallizes in a centrosymmetric orthorhombic structure with D_{2h} symmetry. Each bilayer is comprised of two conducting CuO_2 layers in the *ab* plane. Free charge carriers are provided by partially ordered Cu-O chains along the *b*-axis, with a density that scales with the oxygen stoichiometry. We excited the infrared-active 20 THz mode of B_{1u} symmetry. This mode is associated with motion of the oxygen deficient sites, and involves movement of the apical oxygen atoms perpendicular to the CuO_2 planes, i.e. along the c axis.

Following the nonlinear lattice Hamiltonian introduced above $Q_{B1u}^2 Q_R$ is non-zero only if Q_R is of A_g symmetry, as the square of the irreducible representation of B_{1u} is A_g. The specific geometric distortions and the coupling strengths to the 11 Raman modes of A_g symmetry were computed using first-principle DFT calculations based on frozen Q_{B1u} distortions. While these calculations do not give information about the dynamics, the results allow us to identify the relative coupling strength of these Raman modes to the excited B_{1u} mode.

Experiment and results

To experimentally determine the transient lattice response, we exploited time resolved diffraction using 50-fs hard X-ray pulses from the Stanford free electron laser. We measured the time resolved intensity change of four diffraction peaks that are particularly sensitive to distortions along A_g coordinates. With the relative coupling strengths from the DFT calculations, we can fit the experimental data with the B_{1u} amplitude as the only free parameter to get information on the transient structure attained by the crystal in the light-induced superconducting phase.

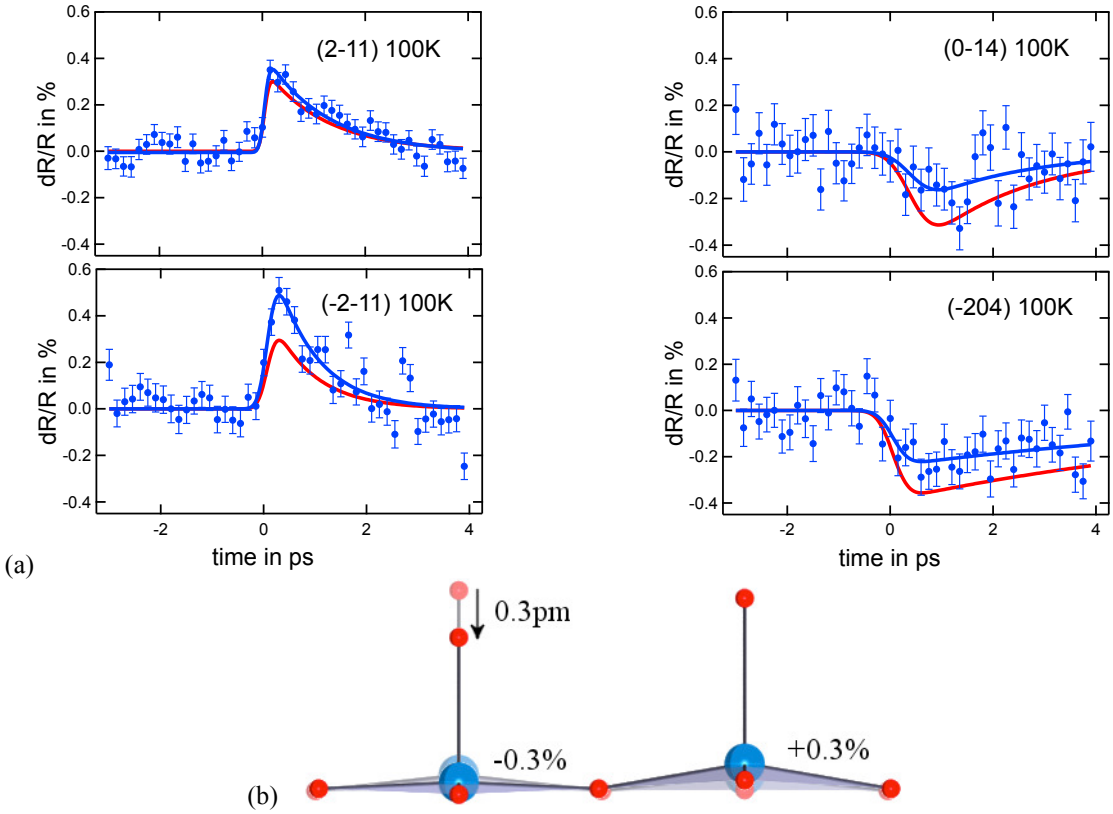

Fig 1. (a) Shown in blue are the changes in diffracted intensity of the Bragg reflections showing a shift of the atoms upon excitation of the infrared active B1u mode that relaxes at a timescale of 1ps. The red curve in the same picture is the calculated intensity change due to the atomic movements along the Ag modes as predicted by DFT calculations. (b) The most important changes in the CuO_2 plane of the crystal structure are shown. At sites with an oxygen vacancy, the apical oxygen moves by 0.3pm towards the CuO_2 plane and we observe an unbuckling of the Cu-O bond in the chain by 0.3%. At sites without an oxygen vacancy, the Cu-O buckling increases by 0.3%.

Figure 1 depicts the experimental data in blue with the fit of the lattice distortions from the DFT calculations in red. The signal shows a shift of the atoms within the unit cell that decays with the lifetime of the excited infrared active B1u mode. Comparing the dynamics with the lifetime of the induced superconducting state as measured by time-resolved optical spectroscopy, we find the same decay rates in the structural and the optical measurements. On the right hand side, the most important changes from the equilibrium structure are shown. At sites with oxygen in the Cu-O chains, we observe an expansion of the apical oxygen atoms by 0.6pm along the c-axis, while the Cu atoms in the CuO_2 plane contract by 0.2pm. This leads to an unbuckling of the Cu-O bonds by 0.3%. At other sites, we observe an expansion of the Cu atoms accompanied with an increase in the buckling by 0.3%.

References

[1] M.Först et al., "Nonlinear phononics as an ultrafast route to lattice control" Nature Physics **7** 854-856 (2011).
[2] M.Först et al., "Displacive lattice excitation through nonlinear phononics viewed by femtosecond X-ray diffraction" SSC 169, 24–27 (2013).
[3] A. Subedi et al., "Theory of nonlinear phononics for coherent light-control of solids" arXiv:1311.0544 (2013).
[4] M. Rini et al., "Control of the electronic phase of a manganite by mode-selective vibrational excitation", Nature 449, 72 (2007).
[5] A. D. Caviglia et. al., "Ultrafast Strain Engineering in Complex Oxide Heterostructures", Phys. Rev. Lett. 108, 136801 (2012).
[6] R.I. Tobey et al., Phys. Rev. Lett. 101, 197404 (2008).
[7] D. Fausti et al., "Light-Induced Superconductivity in a Stripe-Ordered Cuprate", Science 331, 189 (2011).
[8] S. Kaiser, D. Nicoletti, C. Hunt et. al, "Light-induced inhomogeneous superconductivity far above Tc in $YBa_2Cu_3O_{6+x}$", arXiv:1205.4661
[9] W. Hu et. al., arXiv:1308.3204