

Selective THz excitation of collective modes in underdoped YBCO

Georgi L. Dakovski, Wei-Sheng Lee and Joshua J. Turner and Matthias C. Hoffmann

SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA

dakovski@slac.stanford.edu

Abstract: We use intense broadband THz pulses to excite underdoped YBCO exhibiting competing superconducting and charge density wave ground states. We observe pronounced coherent oscillations at 1.85 and 2.65THz, attributed to renormalized low-energy phonon modes.

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1. Introduction

Low-energy single-particle and collective excitations play a crucial role in understanding the properties of matter, especially in the formation of exotic ground states such as superconductivity (SC) and spatially-modulated e.g. charge density waves (CDW). Recent advances in the generation of strong field THz pulses [1] permit to directly and selectively drive low-energy modes in solid-state systems out of equilibrium. This is in stark contrast to relying on photoexcitation using femtosecond laser pulses with photon energies >1 eV, well above the band gap [2], obscuring the subtle effects in the low energy (meV) regime. Here we explore this selectivity by exciting underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ (YBCO) with strong single-cycle broadband THz pulses, while monitoring the response on the femtosecond time scale via the change in reflectivity in the near-infrared range. YBCO, part of the 123 family of high-temperature cuprate superconductors, has recently received a great deal of attention primarily due to the observation of a quasi-two-dimensional CDW forming at about 150K in underdoped samples, and appearing to compete with SC below T_c [3]. The microscopic mechanism of how this CDW form still remains elusive. More importantly, whether this CDW state is simply competing or intertwined with SC in a more complex way require further investigation. Revelation of its collective excitations could shed some lights on these issues.

2. Experimental

Strong single-cycle THz pulses were generated in LiNbO_3 via optical rectification using the tilted-phase front technique [Hebling2005]. The resulting THz pulses had energies up to 3 uJ with a central wavelength of about 1 THz, but carried significant spectral intensity up to ~ 3 THz. A weak near infrared probe pulse of 100 fs duration and 800 nm wavelength was propagating collinearly to the THz beam. Both pulses were incident almost normal to the surface of the sample, which was mounted inside a He cryostat. Peak THz electric fields at the sample plane were measured to be ~ 400 kV/cm using electro-optic sampling. A pair of wire grid polarizers was used before the sample to attenuate the THz beam for intensity dependent measurements. Throughout the experiment the s-polarization of the THz pulse was maintained, being perpendicular to the horizontal scattering plane. The high-quality de-twinned single crystals of YBCO were cut perpendicularly to the c-axis; experiments were performed with all four possible permutations of the pump and probe polarizations parallel and perpendicular to the a- and b crystal axes.

3. Results and discussion

The time resolved reflectivity change (Fig1a) consists of three components: a) A relative change in the reflectivity of $\sim 10^{-3}$ occurring instantaneously with the arrival of the THz pulse. Below T_c its magnitude remains relatively constant with temperature, and especially at very low temperatures it almost completely tracks the square of the THz electric field. Above T_c the initial response reverses sign depending on the polarization of the probe pulse (Fig1c). b) monotonic relaxation with a time constant of a few picoseconds, present both above and below T_c . c) pronounced coherent oscillations appear below T_c , superimposed on incoherent relaxation. With lowering of the temperature the amplitude of the oscillations grows and the incoherent relaxation almost completely vanishes. The Fourier transformation of the oscillatory signal indicates the presence of two modes centered at 1.85 and 2.65THz (Fig1b).

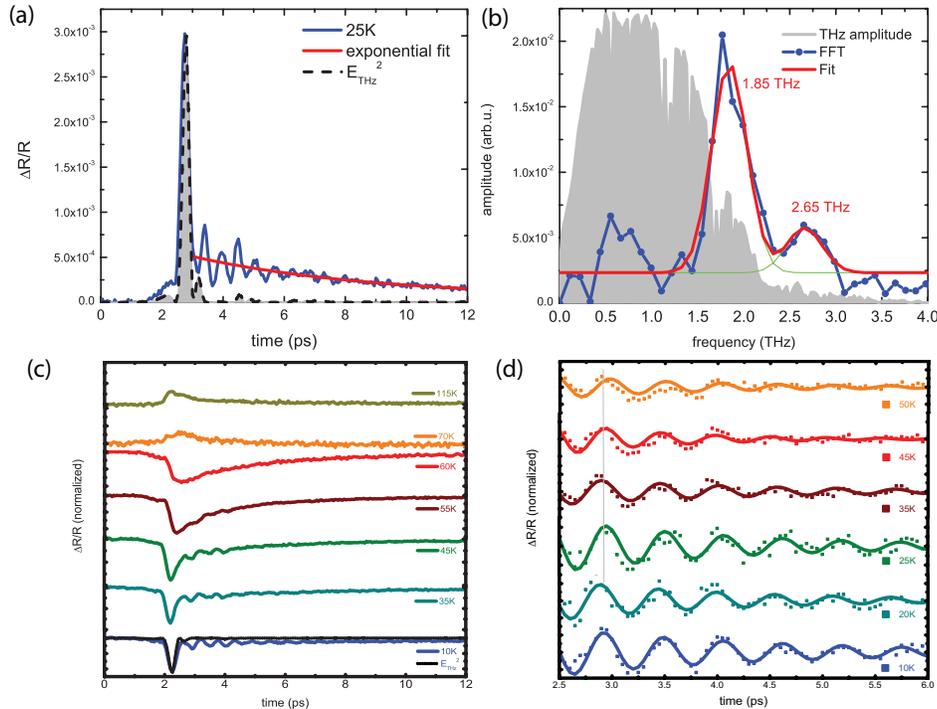


Fig. 1. (a) THz-induced reflectivity change in underdoped YBCO at 800 nm. The instantaneous response tracks the E_{THz}^2 (dashed line), and is followed by an oscillatory component on an exponentially decaying background. (b) Fourier spectrum of the oscillatory response. Two peaks at 1.85 and 2.65 THz are observed (Gaussian fit). The THz excitation pulse spectrum is shown for reference (shaded area).

The energy, bandwidth and phase of these modes remain constant with temperature within the uncertainty of our measurement (Fig1d). The pump-induced reflectivity change scaled quadratically with the THz field strength.

Recent inelastic x-ray scattering measurements [4] have shown the strong renormalization at T_c of two vibrations, a transverse acoustic and transverse optical, exactly at the wave vector q_{CDW} corresponding to the CDW modulation. The renormalized energies of these modes below T_c precisely match the frequencies extracted from the coherent oscillations we observe. Access to these vibrations is provided by the folding of the phonon Brillouin zone induced by the CDW formation mapping these modes to the zone center where they can be directly excited by our broadband THz pulse carrying sufficiently wide bandwidth.

In conclusion, we demonstrate that intense broadband THz pulses can be used to induce strong coherent oscillations at low temperatures in underdoped YBCO. These can be attributed to very strong interaction between electrons and transverse acoustical and transverse optical modes. Most importantly, the oscillations disappear near T_c , indicating that direct excitation at these THz wavelengths is a very sensitive probe to the temperature-dependent electron-phonon coupling.

4. References

- [1] M. C. Hoffmann and J.A. Fülöp, "Intense ultrashort terahertz pulses: generation and applications", *J. Phys. D*, **44** 083001 (2011)
- [2] J. P. Hinton, J. D. Koralek, Y. M. Lu, A. Vishwanath, J. Orenstein, D. A. Bonn, W. N. Hardy, and Ruixing Liang, "New collective mode in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ observed by time-domain reflectometry", *Phys. Rev. B*, **88**, 060508(R) (2013)
- [3] G. Ghiringhelli et al., "Long-Range Incommensurate Charge Fluctuations in $(\text{Y,Nd})\text{Ba}_2\text{Cu}_3\text{O}_{6+x}$ ", *Science*, **337**, 821 (2012).
- [4] M. Le Tacon, A. Bosak, S. M. Souliou, G. Dellea, T. Loew, R. Heid, K-P. Bohnen, G. Ghiringhelli, M. Krisch & B. Keimer, "Inelastic X-ray scattering in $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ reveals giant phonon anomalies and elastic central peak due to charge-density-wave formation" *Nature Physics* **10**, 52–58 (2014)