Excitation of Coherent Oscillations in Underdoped Cuprate Superconductors by Intense THz Pulses

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ABSTRACT

We use intense broadband THz pulses to excite the cuprate superconductors YBCO and NBCO in their underdoped phase, where superconducting and charge density wave ground states compete. We observe pronounced coherent oscillations attributed to renormalized low-energy phonon modes. These oscillation features are much more prominent than those observed in all-optical pump-probe measurements, suggesting a different excitation mechanism.

Keywords: Nonlinear THz, high-temperature superconductivity, charge density wave

1. INTRODUCTION

In order to gain insight into the mechanism of high-temperature superconductivity in the cuprate materials family, understanding of the spin, charge and orbital correlations in both the normal and superconducting states is necessary. Well-known examples are the antiferromagnetic ordering in the undoped, insulating parent compounds and the stripe order at 1/8 doping in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and related systems [1,2]. Renewed interest has been focused on the $R\text{Ba}_2\text{Cu}_3\text{O}_{6+x}$ family (where $R=\text{Y}$ or another rare earth element), which possess much higher critical temperature (T_c) and show perplexing features such as the “pseudogap” region of the phase diagram.

A recent development is the discovery of charge density wave (CDW) order in the $R\text{Ba}_2\text{Cu}_3\text{O}_{6+x}$ system using resonant soft x-ray scattering [3] and hard-X-ray scattering methods [4]. The charge density wave amplitude was shown to initially increase with lower temperature but to subsequently decline for temperatures below T_c , hinting at competition between the superconducting and charge density order.

Dynamics related to the charge density have been observed with ultrafast laser spectroscopy methods in the time domain [5, 6]. Specifically, Hinton et al observed 1.8 THz oscillations in a bulk sample of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ (YBCO) at $x=0.67$. Their results are based on pump-probe experiments with femtosecond laser pulses at 800 nm, corresponding to photon energy of 1.55 eV, and excitation densities of $1.5 \mu\text{J}/\text{cm}^2$. Under these conditions, the oscillatory component of the pump probe signal is superimposed on an incoherent and exponentially decaying background that is about 1 order of magnitude larger. This incoherent background is due to the fact that the excitation photon energy is above the band gap and many excitation pathways are accessible. In this work, extending our results of [7], we investigate excitation of underdoped YBCO and NBCO samples by an electromagnetic field pulse with frequency content on the energy scale of the CDW order. The photon energy of these single-cycle THz pulses is in the range of 1 to 10 meV and well below any band gap energy or known optical phonon frequency. Simultaneously, the electrical field strength and the intensity of the THz pulses is large enough to trigger a coherent response of the sample.

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2. EXPERIMENTAL

2.1 THz pump – near infrared probe apparatus

Single-cycle THz pulses with 4 μJ energy and peak fields up to 400 kV/cm were generated by optical rectification in LiNbO_3 using the tilted pulse front method [8,9]. The THz beam was focused onto the sample located inside a closed-cycle helium cryostat using off-axis parabolic mirrors. The maximum pump fluence was on the order of 0.2 mJ/cm^2 and could be attenuated continuously using a combination of two wire grid polarizers. The repetition rate of the laser system was 1 kHz. Weak probe pulses at 800 nm wavelength and 100 fs duration were reflected off of the sample and detected with a photo-diode (Fig 1). The pump pulses were modulated at 500 Hz and a lock-in amplifier was used to acquire the change in sample reflectivity. The sample reflectivity at 800 nm is a proxy for overall conductivity since the photon energy is above any free-carrier Drude resonance.

The shape THz field at the position of the sample was characterized in the time domain using a 100 μm thick $\langle 110 \rangle$ GaP crystal on a $\langle 100 \rangle$ substrate mounted in the sample plane. The peak THz field strength can be inferred directly from the polarization modulation of the transmitted probe pulses caused by the electro-optic effect.

The orientation of the sample relative to the THz polarization as well as the direction of the probe polarization with respect to the THz polarization was changed for different set of data.

Figure 1: Experimental geometry: single-cycle THz pulses with energy up to 4 μJ are incident on the sample mounted inside a cryostat. Weak 800 nm probe pulses with 100 fs duration are incident collinearly and reflected off the sample which is kept at a slight angle. The orientation of the sample and the probe polarization was varied with respect to the THz polarization.

2.2 Samples

We studied a number of different samples from the “123” family of cuprate superconductors. The first sample was a $\text{YBa}_2\text{Cu}_3\text{O}_{6.75}$ ortho-III single crystal with $x=0.75$, hole doping $p=0.13$, and T_c of 75.2 K [10,11] (UD-YBCO sample). The second sample was an optimally doped YBCO orthorhombic (twinned) thin film on LaAlO_3 substrate. The film thickness was ~ 200 nm and $T_c \sim 90$ K [3]. The last sample was underdoped $\text{Nd}_{1.2}\text{Ba}_{1.8}\text{Cu}_3\text{O}_7$ tetragonal (untwinned) thin film on SrTiO_3 . The thickness was ~ 100 nm with the critical temperature $T_c \sim 65$ K, and the charge order temperature $T_{\text{CDW}} \sim 150$ K [12].

3. RESULTS

3.1 Single crystalline UD-YBCO

Figure 2a shows the THz-induced pump-probe signal at 10K for both pump and probe fields parallel to the sample b-axis. We observe an initial peak that tracks the square of the THz field. This is followed by coherent oscillations with a period of 540 fs. The maximum reflectivity change $\Delta R/R$ is about to 10^{-3} , which is about two times higher than what was reported in Ref [6]. The oscillatory part of the signal is much larger than any incoherent background at this temperature.

Figure 2: (a) THz-induced reflectivity change in single crystal $\text{YBa}_2\text{Cu}_3\text{O}_{6.75}$ at 10K for E_{THz} and E_{probe} both parallel to the crystallographic b-axis. The sign of the signal changes above T_c . E_{THz}^2 is shown for reference. (b) FFT amplitude spectra of the 10K data for delay > 2.5 ps and after background subtraction. The dashed line is the amplitude spectrum of the pump THz pulse.

The frequency spectrum of the pump probe signal for $t > 2.5$ ps is shown in Fig 2b. A strong mode at 1.85 THz is clearly visible. The full-width half maximum is 0.5 THz corresponding to a lifetime of about 2 ps. We note that the sign of the pump-probe signal is inverted if we change the probe laser pulse polarization from the b-axis to the a-axis.

Figure 3 shows the temperature dependence of the THz induced reflectivity change. While the 1.85 THz oscillations are clearly pronounced at low temperature, they are getting damped with increasing temperature and disappear around 55 K, well below $T_c = 75$ K. Simultaneously, we observe the appearance slow relaxation dynamics in the form of an exponential decay. The time constant of this decay and the total amplitude of the signal increase dramatically when approaching the critical temperature.

We still observe a pump probe signal above T_c that persists up to approximately 200K, but it is much weaker and largely featureless. It shows no oscillatory components and can be described by an exponential decay convoluted with the THz pump shape. If the probe polarization is along the b-axis, the sign of the reflectivity signal changes when crossing T_c . The period and hence the central frequency of the observed oscillation mode does not change appreciably with temperature. Figure 3b shows damped sine fits to the oscillatory components of the pump probe signal (after background subtraction) for several sample temperature. Contrary to Ref [6] we do not observe a phase shift when approaching T_c from low temperatures before the oscillations disappear entirely.

Figure 3: (a) Normalized THz-induced reflectivity change in single-crystal UD-YBCO at various temperatures. THz pump polarized along b axis, 800 nm probe along b-axis. E_{THz}^2 is shown for reference in the 10 K data (b) Coherent oscillations in the time domain at different temperatures. Solid line is a damped sine fit after background subtraction. We do not observe a $\pi/2$ phase shift in the signal when approaching T_c .

The dependence of the dynamical reflectivity change on the THz field strength, was studied via controlled attenuation of the THz pump beam by a pair of THz linear polarizers. Figure 4 shows the scaling of the peak pump-probe signal with the THz field strength at 8K and 55K. In both cases the dependence is quadratic.

Figure 4: THz field dependence of the time resolved reflectivity signals at (a) 8K and (b) 55K for $E_{THz}||E_{probe}||b$ -axis. The THz field was attenuated with a pair of wiregrid polarizers.

When the polarization of the THz pump beam is along the sample a-axis, we observe a qualitatively similar signal but with the opposite sign of $\Delta R/R$. The Fourier transformation of the oscillatory signal reveals the presence of an additional mode centered at 2.65 THz. This higher frequency mode matches the energy shift recently found in observations of phonon anomalies coupled to the CDW mode [13].

Figure 5: (a) time domain pump probe data for THz pump and optical probe polarization parallel to the a-axis of the sample (blue curve). The oscillatory part of the signal is superimposed on an incoherent background that can be described by a single-exponential decay (red curve) (b) amplitude spectrum of the oscillatory part of the signal (blue curve) and Gaussian fits (red curve). The THz amplitude spectrum is shown for reference (black)

3.2 Optimally doped YBCO thin film

In addition to the underdoped single crystalline YBCO sample, we also carried out a series of measurements on an optimally doped YBCO thin film on LaAlO₃ substrate. The film thickness was ~ 200 nm and the superconducting transition temperature $T_c \sim 90$ K. Here, the THz-induced reflectivity change was markedly different from the underdoped case (Figure 6). The sample response follows the square of the THz pulse, but no oscillatory component is visible at any temperature or probe condition. Above T_c , the signal decreases and a slow rising component followed by long-lived offset is observed after the THz excitation.

Figure 6: (a) THz pump/800 nm probe signal from optimally doped YBCO on LAO at 8K for two different probe polarizations. The signals are compatible with the envelope of the pump pulse and show no other features. (b) signal at 8K and above T_c at 100K.

3.3 Results for UD-NBCO

In order to gain insight into the dynamics of charge density wave order in a closely related materials system, we carried out a series of measurements on underdoped Nd_{1.2}Ba_{1.8}Cu₃O₇ (UD-NBCO). The sample was a 100 nm thick untwinned thin film on a SrTiO₃ substrate with $T_c \sim 65$ K, and the charge order temperature $T_{CDW} \sim 150$ K [12]. The results are summarized in Figure 7.

At low temperature, we observe an initial peak with a rising edge shape that tracks the square of the THz field. The peak reflectivity change $\Delta R/R$ is on the order of 10^{-3} , which is similar to the values in our previous experiment on YBCO [7] and about 2x higher than reported in [6]. This is followed by coherent oscillations corresponding to frequencies at 1.5 THz and possibly 0.45 THz, superimposed on an exponential decay (Figure 7b). The main oscillation mode has a full width half maximum corresponding to a lifetime of about 2 ps.

Figure 7: (a) UD-NBCO reflectivity change for THz-pump/800 nm probe at 8K and (b) spectral components of the observed coherent oscillations after subtraction of the exponentially decaying background.

Figure 8 shows the temperature dependence of the THz-induced reflectivity change. The absolute strength of the signal decreased with increasing temperature, but a small signal persists above T_c . The oscillatory part of the signal gets rapidly damped when approaching T_c and disappears entirely around 50 K. The central frequency of the observed oscillation mode does not change appreciably with temperature. We note that the exponentially decaying part of the signal gets faster with higher temperature, contrary to the UD-YBCO measurements where we saw the slowest dynamics just below T_c . The relationship between temperature and the time constant of the incoherent decay from single-exponential fits is shown in Figure 8b.

Figure 8 Temperature dependence of the THz pump/optical probe signal in underdoped NBCO. The oscillatory feature disappears in the vicinity of T_c . (b) Decay constants from a single-exponential fit to the signal for time delays >3 ps. The solid line is a guide to the eye.

4. DISCUSSION

The coherent oscillations observed by transient reflectivity in both underdoped systems are likely related to their charge density wave ordered state. In UD-YBCO, the 1.85 THz and 2.65 THz modes match the energies of the strongly renormalized phonon modes at the wave vector corresponding to the CDW modulation, measured by inelastic x-ray scattering [13]. While these softened modes are located at a finite momentum, they are still optically accessible at the zone center due to Brillouin zone folding with the CDW breaking the translational symmetry. While CDW-related oscillations have been observed in optical pump experiments on YBCO and LSCO before [6,5], strong-field THz pulses prove to be a surprisingly effective way to trigger this effect. Since the photon energy of the THz pulses is in the range of meV, direct creation of Cooper pair breaking and quasiparticle generation is not possible. Instead, the excitation mechanism must be driven by the electric field component of the THz pulse. Given the considerable field strength, the local current may approach the de-pairing current density of 3×10^{12} A/m² in YBCO [14].

We note that the temperature dependent dynamics are found to be very different than in previous optical pump experiments. First, while the collective oscillations induced by optical excitation persist to temperatures well above T_c , and are detectable up to T_{cdw} , the THz-induced collective dynamics disappear slightly below T_c (Figure 3a). Secondly, unlike optical excitations, neither the frequency nor the phase of the oscillations shift appreciably as a function of temperature. Lastly, the temperature dependence of the incoherent dynamics is also very different. In UD-YBCO the temperature evolution of the recovery time displays an increase just below T_c , a behavior often ascribed to the presence of a “phonon bottleneck” where photoexcited quasiparticles and low-energy phonons are in a state of quasi-equilibrium [15]. On the other hand, the amplitude of incoherent background signal vanishes at low temperatures, in contrast with the temperature-independent behavior observed in all-optical experiments. This may be due to the increase in de-pairing current density with lower temperature.

In UD-NBCO the temperature dependent dynamics show astonishingly different behavior. At low temperature an oscillation mode at 1.5 THz superimposed on an exponentially decaying background is clearly visible, with a possible side band at 0.45 THz. However, the decay constant of the incoherent portion of the signal decreases with higher temperature, in sharp contrast to the observations in UD-YBCO.

5. CONCLUSION

We demonstrate that intense broadband THz pulses can be used to induce strong coherent oscillations at low temperatures in underdoped YBCO and NBCO. These can be attributed to the strong interaction between the superconducting state and the CDW mode. The excitation mechanism is likely based on the instantaneous current driven by the THz field in contrast to direct photon absorption in all optical spectroscopy methods. This unique excitation pathway will enable future investigations that may provide more information by directly monitoring the CDW dynamics with advanced probe methods like ultra-fast resonant x-ray scattering. These could reveal the interplay between density wave physics and SC, thought to be of importance in all families of cuprates, and can be used as a novel way of exploring the properties of high-temperature superconductors.

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