Enhancement of superconducting coherence in $YBa_2Cu_3O_x$ by resonant lattice excitation

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Abstract: By using femtosecond pulses in the mid-infrared, we resonantly excite an infrared-active phonon mode in the high-temperature superconductor $YBa_2Cu_3O_x$. The electronic properties of the driven state, probed with ultra-broadband time-domain terahertz spectroscopy, are highly unconventional.
OCIS codes: (320.7130) Ultrafast processes in condensed matter; (300.6495) Spectroscopy, terahertz.

1. Introduction and Methods

Non-linear resonant excitation of infrared-active vibrational modes of the crystal lattice has been proven to melt magnetic or orbital orders and to induce insulator-to-metal transitions in complex solids [1-3]. In high-T_c superconducting cuprates, this technique was used to remove the competing charge- and spin-order in the so-called *stripe* phase in $La_{1.675}Eu_{0.2}Sr_{0.125}CuO_4$, thus transiently inducing superconductivity (at temperatures as high as 20 K) in a material which is not superconducting at equilibrium [4]. In the present experiment, we show that a large-amplitude modulation of the apical oxygen positions (see Fig. 1a) in $YBa_2Cu_3O_x$ can promote highly unconventional electrodynamics, which can be captured with ultra-broadband time-domain terahertz spectroscopy.

Superconductors at equilibrium display two characteristic physical properties: zero DC resistance and the expulsion of static magnetic fields. The first of these properties manifests itself as a positive imaginary part of the optical conductivity $\sigma_2(\omega)$ that diverges at low frequency as $1/\omega$. In high-T_c cuprates, the layered structure gives rise to additional *c*-axis excitations of the superfluid, with the appearance of one or more longitudinal Josephson plasma modes due to tunneling of Cooper pairs between capacitively coupled superconducting planes. In $YBa_2Cu_3O_x$, a bi-layer cuprate, two longitudinal plasma modes are found [5], reflected by two peaks in the energy loss function $-\text{Im}(1/\tilde{\varepsilon})$ at ω_1~1 THz and ω_2~14 THz (Fig. 1b and 1c, negative time delays). Within each family of cuprates, the mode frequency quantifies the strength of the Josephson coupling between pairs of CuO_2 layers.

Here, we measure the transient *c*-axis optical properties of $YBa_2Cu_3O_x$ (x = 6.45, 6.5, 6.6) after photo-excitation, both below and above the superconducting transition temperature T_c [6,7]. Mid-infrared pump pulses of ~300 fs duration, polarized along the *c* direction and tuned to ~20 THz frequency, were made resonant with the infrared-active distortion shown in Fig. 1a. Such pump pulses were generated by difference-frequency mixing in an optical parametric amplifier and focused onto the samples with a maximum fluence of 4 mJ/cm^2, corresponding to peak electric fields up to ~3 MV/cm. At these strong fields, the apical oxygen positions are driven in an oscillatory way by several percent of the equilibrium unit cell [6]. The solid was then investigated with broadband THz probe pulses generated both by optical rectification in ZnTe (0.5 – 2.5 THz) and by gas ionization (1 – 14 THz).

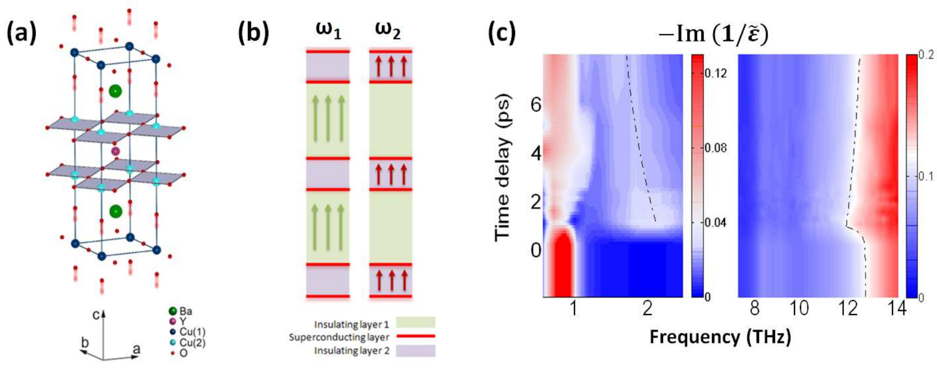

Fig. 1. (a) Crystal structure of $YBa_2Cu_3O_x$ and sketch of the optically-driven distortion of the apical oxygen. (b) Josephson plasma modes of the bi-layer structure. (c) Transient energy loss function of $YBa_2Cu_3O_{6.5}$ after mid-infrared excitation at 10 K. The dynamics of the Josephson plasma modes is displayed by exponential fits as a function of pump-probe time delay.

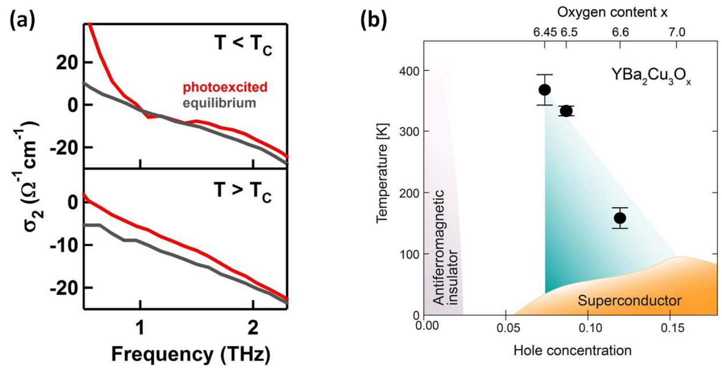

Fig. 2. (a) Transient increase in the imaginary part of the optical conductivity at low frequency, measured 0.8 ps after photo-excitation in $YBa_2Cu_3O_{6.5}$, both below and above $T_c = 50$ K (at 10 K and 100 K, respectively). (b) Temperature-doping phase diagram of $YBa_2Cu_3O_x$. The region affected by optically-enhanced coherence (shaded) has been determined by estimating the maximum temperature at which a divergence in $\sigma_2(\omega)|_{\omega\to 0}$ could be detected for each doping level (black circles).

2. Results and Discussion

The transient optical properties of $YBa_2Cu_3O_x$ were extracted from measurements of the amplitude and phase of the reflected electric field after photo-excitation, taking into account the pump-probe penetration depth mismatch [6]. In Fig. 1c, we report the changes in the inter- and intra-bilayer Josephson coupling strength by plotting the time- and frequency-dependent energy loss function of $YBa_2Cu_3O_{6.5}$ in the superconducting state [7]. The peak at ω_1~1 THz, which reflects the inter-bilayer plasma mode at equilibrium, reduces in amplitude after photo-excitation, as a second higher-frequency peak appears at ~2 THz. Simultaneously, the intra-bilayer mode, which at equilibrium is observed at ω_2~14 THz, shifts to the red. All spectral changes relax back to equilibrium within ~7 ps.

A qualitatively similar response to that described above is found in the same material above T_c (not shown). After excitation, a peak in the loss function appears at ~2 THz, while a red-shift of spectral weight is detected around ~14 THz. These observations are consistent with an optically-driven transfer of coupling strength from the bi-layers towards the inter-bilayer region, occurring both below and above T_c.

In Fig. 2a, we report the imaginary part of the optical conductivity of $YBa_2Cu_3O_{6.5}$ after excitation. Since the superfluid density at equilibrium is quantified by $\omega\sigma_2(\omega)|_{\omega\to 0}$, the increase in the slope of $\sigma_2(\omega)$ at low frequency indicates a photo-induced enhancement of superfluid density in the superconducting state (upper panel). Above T_c (lower panel) $\sigma_2(\omega)|_{\omega\to 0}$ also exhibits an increase, turning positive and diverging down to the lowest measured frequency. This behavior, combined with the other optical constants, is consistent with the response of a conductor with anomalously high mobility, which would be highly unusual for incoherent transport in oxides. An interpretation based on a transient superconducting state above T_c is in our view more plausible [7].

The phase diagram of $YBa_2Cu_3O_x$ is displayed in Fig. 2b. The shaded region indicates the non-equilibrium high-mobility phase induced by resonant lattice excitation. Remarkably, this extends even above room temperature at the lowest doping levels and tracks surprisingly well the onset temperature of the so-called *pseudogap* state in the equilibrium phase diagram of high-T_c cuprates [8].

All the above observations are compatible with previous reports of an inhomogeneous state above T_c, which would retain important properties of a superconductor [9]. In such scenario, mid-infrared light might melt a competing order, or dynamically synchronize the inter-layer phase [4,10]. The transient redistribution of coherence demonstrated here could lead to new strategies to enhance superconductivity in the steady state.

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