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**Citation:** Perkins, B. G., H. Y. Hwang, N. K. Grady, L. Yan, D. Trugman, Q. Jia, H. T. Chen, A. J. Taylor, and K. A. Nelson. "Nonlinear ultrafast dynamics of high temperature  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  superconductors probed with THz pump / THz probe spectroscopy." Edited by M. Chergui, A. Taylor, S. Cundiff, R. de Vivie-Riedle, and K. Yamagouchi. EPJ Web of Conferences 41 (March 13, 2013): 03010.

**As Published:** <http://dx.doi.org/10.1051/epjconf/20134103010>

**Publisher:** EDP Sciences

**Persistent URL:** <http://hdl.handle.net/1721.1/82548>

**Version:** Final published version: final published article, as it appeared in a journal, conference proceedings, or other formally published context

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## **Nonlinear ultrafast dynamics of high temperature YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> superconductors probed with THz pump / THz probe spectroscopy**

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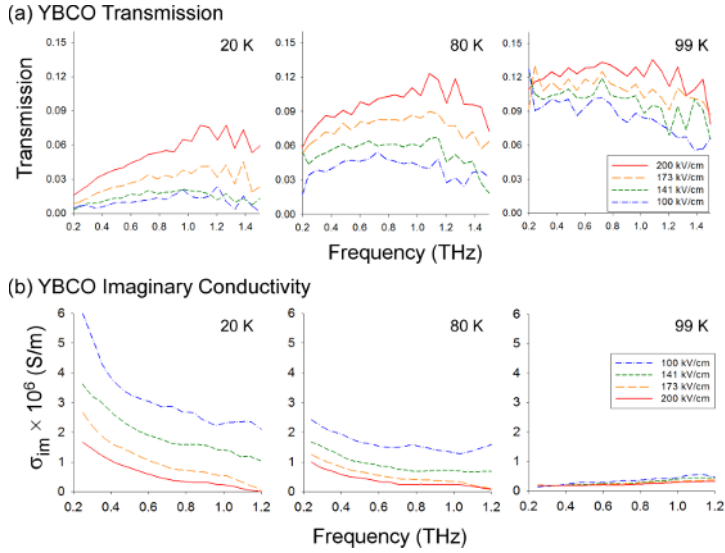
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**Abstract.** High power THz pulses induce near transparency in superconductive YBCO thin films below the critical temperature. THz pump/THz probe measurements reveal a decay of the induced transparency on the time scale of a few picoseconds.

High- $T_c$  superconductors have been studied with ultrafast pump-probe experiments to understand the mechanism through which superconductivity is destroyed by light and then recovered by recombination of electrons to form Cooper pairs [1-3]. Previous experiments have used visible or IR ultrafast pump pulses with either visible, IR, or THz probe pulses [1-5] to determine various contributions to the overall recombination mechanism in thin films of doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> (YBCO). In these studies, the pump light breaks superconductivity by exciting electrons with hundreds to thousands of meV of energy, which then relax by transferring energy into phonon modes in the crystal. To complicate matters, the excited phonons break other Cooper pairs as the system returns to thermal equilibrium [3]. The goal of our experiment is to explore recombination dynamics as strong THz pulses excite electrons in YBCO above the superconductive gap to break superconductivity in ways that limit the excess energy below that needed to populate optic phonons.

The thin films were studied with high-power time-domain THz spectroscopy in an apparatus that was configured first for nonlinear transmission measurements and second for THz pump-THz probe scans [6]. High-energy THz pulses were produced by nonlinear rectification of amplified femtosecond pulses ( $\lambda = 800$  nm,  $\tau = 100$  fs, energy = 6 mJ/pulse) in lithium niobate in a tilted pulse front configuration, yielding field strengths up to 200 kV/cm when focused at the sample. The pump and probe THz pulses were generated co-linearly through optical rectification of two variably delayed 800-nm pulses. The 50-nm YBCO thin film on LaAlO<sub>3</sub> and its matching 500  $\mu$ m reference substrate were mounted in a free-flowing liquid helium cryostat for the measurements. Transmitted time-dependent electric fields were recorded for the thin film sample and the reference for a range of temperatures and THz light intensities. Transmission spectra of a YBCO thin film at 20 K, 80K, and 99K are plotted in Figure 1a for THz field strengths that range from 100 to 200 kV/cm. The transmission spectra are nearly flat across the experimental bandwidth, and the average transmission increases with both field strength and temperature. The most dramatic field-dependent changes occur at lowest temperatures, yet nonlinear effects are also observed above  $T_c$ .

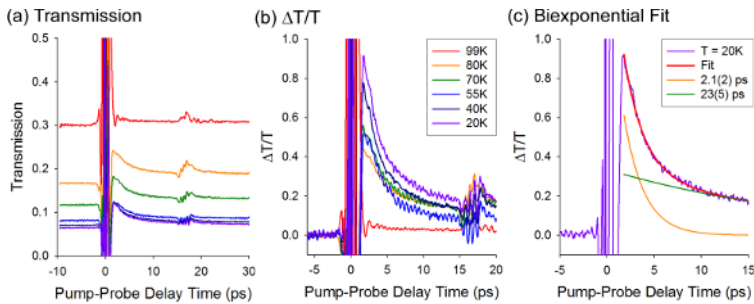
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**Fig. 1.** (a) Power-dependent transmission of 50nm YBCO thin films on LaAlO<sub>3</sub> taken at 20K, 80K, and 99K. (b) Imaginary conductivity extracted from time-dependent transmission traces that are fit with the two-fluid model presented in the text.  $T_c \approx 90$  K for this sample.

The transmission spectra in Figure 1a reveal an induced transparency in the thin films, which suggests that the THz field is reducing the number of conductive and/or superconductive carriers. To understand this affect, we employ a two-fluid model that has been used to characterize the temperature-dependent conductivity in previous YBCO thin film studies [7, 8]. In this model, the real and complex conductivity are related to the normal carrier (quasiparticle) fraction [ $f_n(T)$ ] and the superfluid fraction [ $f_s(T)$ ] by equations  $\sigma_{re} = (n \cdot e^2 / m^*) \cdot f_n(T) \cdot \tau / (1 + \omega^2 \cdot \tau^2)$  and  $\sigma_{im} = (n \cdot e^2 / m^*) \cdot [f_n(T) \cdot \tau / (1 + \omega^2 \cdot \tau^2) + f_s(T) / \omega]$ . In these equations,  $f_n(T) + f_s(T) = 1$ ,  $n$  is the carrier density,  $m^*$  is the carrier effective mass, and  $\tau$  is the quasiparticle relaxation time. The thin film has been analyzed previously in linear THz spectroscopy studies, which characterized the complex conductivity as a function of temperature [7]. For example,  $\sigma_{re}$  at 0.6 THz is nearly constant, in the  $1.5\text{--}1.0 \times 10^6$  (S/m) range over  $T = 40\text{--}100$  K. In contrast, the  $\sigma_{im}$  is approximately zero above  $T_c$ , and then increases to  $\sim 5.6 \times 10^6$  (S/m) as the temperature drops to 50 K. In our current study, we extract the complex conductivity of the sample, for which we show  $\sigma_{im}$  in Figure 1b for temperatures 20 K, 80 K, and 99 K. The analysis reveals that  $\sigma_{im}$  is highest at the lowest temperatures and lowest THz field strengths. As the THz field strength increases at a given temperature, we see  $\sigma_{im}$  monotonically decrease indicating the THz pulse reduces the superfluid fraction. These results are consistent with a physical picture in which the THz light breaks Cooper pairs to reduce superconductivity in YBCO.

The flat and featureless transmission spectra in Figure 1a indicate that dispersion in the 50 nm YBCO across the measured bandwidth is negligible. For this reason, we measured only the time-dependent change of the peak probe electric field as a function of pump-probe delay time at the maximum pump field strength (approximately 200 kV/cm). Time traces of the sample and reference substrate were recorded for temperatures ranging from 20 to 99K. From these data, we calculated the time-dependent transmission, which is plotted in Figure 2a for each temperature. The plot shows that the baseline transmission before pump-probe temporal overlap ( $t = 0$  ps) increases with temperature as in Figure 1a. The signal between times -1 and 1 ps shows evidence of the temporal overlap between the pump and probe beams in the lithium niobate. After the pump and probe are safely separated in time, we observed an induced transmission that decays on the time scale of several picoseconds. Evidence of a reflection in the sample is apparent at 16-17 ps. In addition, the signal plateaus above the baseline for the 20–100 ps time scale of our scans. The traces in Figure 2a



**Fig. 2.** (a) THz-pump / THz probe transmission measurements show induced transparency that decays on the several picosecond time scale. (b) Relative change in transmission is the largest at the lowest temperatures. Above  $T_c = 90$  K, transmission changes only slightly due to heating effects. (c) A biexponential fit of the decay reveals 2.1(2) ps and  $\approx 20$  ps components.

also show strong temperature dependence in the relative magnitude of the induced transparency. The relative transmission is plotted in Figure 2b for all temperatures. The relative change is the largest at lowest temperatures, for which  $\Delta T/T$  peaks at  $\sim 90\%$ . The signals have been fit as in Figure 2c with a biexponential function that characterizes the decay with two time constants. Extracted values for the fast decay component are nearly constant with temperature from 20 to 80 K and are consistent with 1.5–3.5 ps values from visible pump–THz probe measurements [3]. This decay represents the Cooper pair recombination time that restores superconductivity to the thin film sample. The slower component of the fit reflects a time scale that is greater than the time trace of data used in the fit. Therefore, the extracted values of 20–30 ps represent a lower limit for a second decay process that involves more complicated breaking and recombination dynamics of the superconducting electrons in the sample. Importantly, these dynamics appear to be caused by mechanisms other than thermal heating since the estimated temperature rise for this highly reflective thin film is less than 0.2 K.

In summary, power-dependent transmission measurements of YBCO thin films have shown that a strong THz electric field induces partial transparency, which is consistent with a transient reduction of superconducting electrons. Dynamics of this phenomenon have been studied with THz pump / THz probe experiments that reveal a decay of the induced transmission on the time scale of a few picoseconds due to Cooper pair regeneration. After the initial decay, transmission remains above baseline values, which may indicate the relaxation process involves additional nonthermal dynamics that reestablish superconductivity in the thin film.

Funding for this work is provided by ONR grant N00014-09-1-1103 and NSF American Competitiveness in Chemistry Fellowship 1041979. We acknowledge partial support from LANL/LDRD program. Additionally, this work was performed, in part, at the Center for Integrated Nanotechnologies, a U.S. Department of Energy, Office of Basic Energy Sciences user facility. Los Alamos National Laboratory, an affirmative action equal opportunity employer, is operated by Los Alamos National Security, LLC, for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396.

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