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Nonequilibrium THz conductivity of $Bi_2Sr_2CaCu_2O_{8+\delta}$

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Using high sensitivity visible-pump/THz-probe spectroscopy we investigate the dynamics of the complex optical conductivity, $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$, in optimally-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ films directly after photoexcitation. The photoinduced change in the imaginary part $\Delta\sigma_2$, indicative of a reduction in the superconducting condensate density, saturates at higher laser-fluences and shows a complete destruction of the condensate.

Keywords: THz spectroscopy; superconductivity; Bi-2212

1. Introduction

While the mechanism behind the formation of a superconducting condensate in the cuprates remains unresolved, understanding the dynamics of their low-energy excitations can reveal information about intrinsic quasiparticle interactions [1]. Ultrashort laser sources provide new tools to separate components of the response of complex many-body systems in the time domain [2,3]. For cuprate superconductors, the abundance of ultrafast laser sources in the visible has inspired several experiments in that spectral range [4,5]. The observed small optical reflectivity modulations around ~1-3 eV can be linked [5] via Kramers-Kronig relations to the mid-infrared pseudogap (~100 meV) whose dynamics was recently detected [3].

The signature of the superconducting condensate appears in the terahertz spectral range (1 THz = 4.1 meV) – orders of magnitude lower in energy than the pseudogap. The electrodynamics of superconductors, expressed using the optical conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$, is fundamentally characterized by a strong inductive response in its imaginary part, $\sigma_2(\omega) \sim (n_S/m^*)\omega^{-1}$, proportional to the superconducting condensate density n_S [6]. THz frequencies are well-suited to probe the condensate response in equilibrium [7]. Recently the technique for carrying out time-resolved experiments on superconductors in the THz range was also demonstrated, albeit over a limited range of densities [8].
Here, we present ultrafast measurements of the THz conductivity induced by near-infrared photo-excitation in optimally-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212, $T_C \approx 88$ K). Our experiments cover a large range of excitation densities. Low densities allow a study of fundamental quasiparticle interactions. At the higher excitation levels discussed below, a complete destruction of superconductivity can be observed along with a reversal of the electrodynamic response to the normal state.

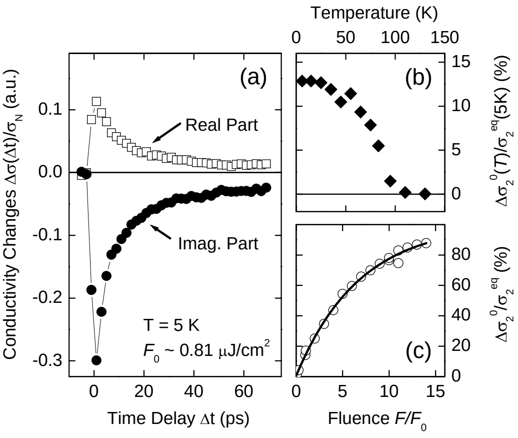

Fig. 1. Photoinduced changes in the low-energy optical conductivity at a single probe frequency $\nu = 1.3$ THz. (a) Ultrafast return to equilibrium of $\Delta\sigma_2/\sigma_N$ (solid circles) and $\Delta\sigma_1/\sigma_N$ (open squares) at $T = 5$ K following laser excitation by a 100-fs pump pulse at 1.5 eV and fluence $F_0 \sim 0.8$ μJ/cm^2. (b) Temperature-dependent signal amplitude $\Delta\sigma_2^{\,0}(T)$ of imaginary part at fixed pump fluence F_0, normalized by the low-temperature equilibrium value $\sigma_2^{eq}(5\text{ K})$. (c) Amplitude $\Delta\sigma_2^{\,0}$ as a function of fluence F/F_0, at $T = 5$ K (open symbols). Solid line: fit to $\propto (1 - e^{-F/U})$.

2. Experiment and Results

The sample is a highly crystalline, c-axis oriented thin-film grown by molecular beam-epitaxy on a $LaAlO_3$ substrate [9]. Picosecond THz probe pulses encompassing a broad frequency range (~ 0.5–3 THz) are derived from a 250-kHz Ti:sapphire regenerative amplifier alongside the near-infrared pulses used for excitation. The electric field of the THz pulses is detected coherently in the time-domain via electro-optic sampling. This scheme for detecting the electric field $E_{THz}(t)$ in the time domain yields both real and imaginary parts of the film's THz conductivity $\sigma(\omega)$ directly and on equal footing [10].

The experiment is carried out by choosing delay times Δt between the arrival of (*i*) the exciting pump pulse and (*ii*) the THz probe pulse. For each fixed Δt, the modulation of the THz electric field due to the pump is measured, which in turn provides a snapshot of the photo-induced conductivity change $\Delta\sigma(\omega,\Delta t) = \sigma(\omega,\Delta t) - \sigma_{eq}(\omega)$. Here, $\sigma_{eq}(\omega)$ is the conductivity in equilibrium (without excitation) and $\sigma(\omega,\Delta t)$ that with excitation. Scanning Δt then allows the picosecond recovery of the superconducting condensate to be followed in time.

Figure 1(a) shows the dynamical evolution of real and imaginary parts of the conductivity, as normalized to the normal state conductivity σ_N. The data plotted is given for a probe frequency of 1.3 THz and low lattice temperature $T = 5$ K. Directly after excitation, both real and imaginary parts of $\sigma(\omega)$ change rapidly to a new nonequilibrium peak value and subsequently decay back to the original conductivity on a picosecond timescale. The negative change of $\Delta\sigma_2$ signifies the depletion of the condensate density immediately after photoexcitation. This is confirmed in the strongly frequency-dependent spectral response (not shown). The corresponding positive change of $\Delta\sigma_1$ can be explained by enhanced absorption due to an increased quasiparticle fraction. The temperature dependence of the signal amplitude is shown in Fig 1(b) which plots the fraction $\Delta\sigma_2^{\,0}/\sigma_2^{eq}(T\text{=5K})$ of the initial imaginary conductivity change $\Delta\sigma_2^{\,0}(T)$ relative to its $T = 5$ K equilibrium value. It closely follows the temperature evolution of the equilibrium condensate spectral weight, and thus shows that a constant fraction of the available superconducting Cooper pairs is depleted through the near-visible excitation pulse.

At fixed temperature, the photoinduced change in the imaginary part spectral weight does not increase above the equilibrium value but rather saturates with increasing pump fluence F. Indeed, a plot of $\Delta\sigma_2^0/\sigma_2^{eq}$ at T = 5K versus pump fluence (Fig. 1c) exhibits a saturation approaching 100% of the equilibrium value. This corresponds to a complete destruction of the superconducting condensate. Interestingly, such saturation behavior can be fit with a functional form $\Delta\sigma_2^0/\sigma_2^{eq} \propto (1 - e^{-F/U})$ that yields an estimate of the saturation energy $U \approx 1$ J/cm^3. Furthermore, the conductivity spectrum $\sigma_1(\omega)$ of the maximally depleted state is characterized by a conducting response.

In summary, we discussed optical-pump THz-probe experiments that measure the ultrafast dynamics of quasiparticles and superconducting condensate in superconducting Bi-2212. A full depletion of the Cooper pair condensate is accessible which provides a new contactless measure of the condensation energy. Using this scheme, we have carried out high-sensitivity measurements of the condensate dynamics at very low densities that yield insights into two-particle interactions during the formation of the superconducting condensate, as will be reported elsewhere [11].

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References

[1] J. Orenstein and A. J. Millis, Science **288**, 468 (2000).
[2] D. S. Chemla, J. Shah, Nature **411**, 549 (2001) and references therein.
[3] R. A. Kaindl *et al.*, Science **287**, 470 (2000) and references.
[4] C. Stevens *et al.*, Phys. Rev. Lett. **78**, 2212 (1997).
[5] G. P. Segre *et al.*, Phys. Rev. Lett. **88**, 137001 (2002).
[6] see e.g. M. Tinkham, *Introduction to Superconductivity, 2nd ed.* (McGraw-Hill, New York, 1996).
[7] J. Corson *et al.*, Nature **398**, 221 (1999).
[8] R. D. Averitt *et al.*, Phys. Rev. **B63**, 140502 (2001).
[9] J. N. Eckstein and I. Bozovic, Annu. Rev. Mater. Sci. **25**, 679 (1995).
[10] M. C. Nuss and J. Orenstein, in *Millimeter and Submillimeter Wave Spectroscopy of Solids*, Ed. by G. Gruner (Springer, Berlin, 1998).
[11] M. A. Carnahan *et al.*, to be published.