Nonequilibrium superconductivity in $Y_{1-x}Pr_xBa_2Cu_3O_7$ thin films

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Abstract

We have measured the picosecond conductivity dynamics in $Y_{1-x}Pr_xBa_2Cu_3O_7$ thin films from 0.3-2.0 terahertz. Our experimental technique measures the complex conductivity $\sigma_{re}(\omega) + i\sigma_{im}(\omega)$ permitting the simultaneous observation of superconducting pair and quasiparticle dynamics. We emphasize aspects of the conductivity dynamics which extend our previous results [1]. In particular, the recovery of $\sigma_{re}$ is faster than $\sigma_{im}$, and the recovery of $\sigma_{im}$ decreases with increasing frequency. This suggests another carrier relaxation pathway, in addition to superconducting pair recovery, following optical excitation.
Terahertz time-domain spectroscopy (THz-TDS) is an ultrafast optical technique that has found wide application in the study of many systems having far-infrared excitations. In the context of correlated electron materials, THz-TDS has been successfully applied to study high-$T_c$ superconductors and, more recently, materials such as the ferromagnetic metal SrRuO$_3$ [2, 3]. We can expect an increase in the use of THz-TDS to study a variety of other correlated electron materials given its unique ability to directly and easily measure $\sigma_{re}(\omega) + i\sigma_{im}(\omega)$ from $\sim 50$ GHz to several THz.

Importantly, the freely propagating THz pulses generated in THz-TDS are temporally coherent with the generating optical pulses - this permits measurement of the THz conductivity with picosecond resolution following optical excitation of the sample. Several groups have been developing this technique, termed time-resolved THz spectroscopy (TRTS), to study various systems including photogenerated electrons in liquids such as hexane, or semiconductors such as GaAs [4, 5]. Our work has focused on using TRTS to study high-$T_c$ superconductors and colossal magnetoresistance manganites [1, 6, 7]. Here we present our most recent measurements on Y$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_7$ thin films.

Figure 1(a) and (b) show the conductivity at 60K and 95K respectively ($T_c = 89$K) for the near optimally doped film. The phenomenological two-fluid model fits the data quite well (shown as a dashed line in Fig. 1(a)) below $T_c$ where the imaginary conductivity is dominated by the $1/\omega$ dependence of the superfluid [1, 8]. Above $T_c$, a standard Drude model fits the data (dashed line, Fig. 1(b)). Upon optical excitation, there is a decrease in the imaginary conductivity due to superconducting pair breaking with a corresponding increase in the real component (not shown). The induced change in $\sigma_{im}(\omega)$ is shown at 60K in figure 1(c). There is a decrease that rapidly recovers on a ps timescale that is due in large part to superconducting pair reformation. Figure 1(d) shows the induced change in $\sigma_{im}(\omega)$ at 95 K (above $T_c$) which is due quasiparticle relaxation.

The dynamics can be followed by plotting the induced change in the conductivity as a function of time at a specified frequency. The induced change in the imaginary conductivity (60K) is shown in Figure 2. With increasing frequency, the lifetime decreases (see inset). In the limit of zero quasiparticle fraction, this induced change would be solely due to superconducting pair recovery. However, there are quasiparticles present (at 60K the initial quasiparticle fraction is $\sim 40\%$) so the quasiparticle fraction makes a nonnegligible contribution to $\sigma_{im}(\omega)$. At higher frequencies this fraction becomes increasingly important since...
the superconducting pair fraction response goes as $1/\omega$. This offers a potential explanation for the decrease in the lifetime of $\sigma_{im}(\omega)$ with increasing frequency: at low frequencies $\sigma_{im}(\omega)$ is dominated by the superconducting pair recovery, but at higher frequencies the relaxation is increasingly influenced by an additional relaxation pathway associated with the quasiparticles. This is further supported in that the lifetime of $\sigma(\omega)_{re}$ is quite short ($\sim 1.5$ ps independent of frequency). Speculating on the origin of this additional relaxation pathway, it could be due to the relaxation of the excited quasiparticles into the nodes of the superconducting gap along $k_x = k_y$. Since this process is faster than the superconducting pair recovery, this suggests that the excited quasiparticles relax into the nodes of the gap followed by pair recovery.

Finally, the results of the superconducting pair recovery time as a function of temperature are consistent with our previous results [1]. For optimal doping, the lifetime at 20 K is about 1.5 ps (at 1.0 THz) increasing to 3.0 ps near $T_c$. Above $T_c$, the lifetime of $\sigma_{im}(\omega)$ (which is no longer a measure of the superconducting recovery time, but rather the initial quasiparticle cooling) drops to 1.5 ps (this is also consistent with the discussion in the previous paragraph). In contrast, in the $x=0.3$ films the lifetime is $\sim 3.5$ ps independent of temperature even above $T_c$. This lifetime is the same as that measured in our YBa$_2$Cu$_3$O$_{6.5}$ films. These results suggest that for the optimally doped films, the dynamics are influenced by the closing of the superconducting gap, and that for the underdoped films, the pseudogap plays a role in determining a observed dynamics.

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Figure 1: (a)-(b) Conductivity at 60K and 95K, respectively for YBa$_2$Cu$_3$O$_7$. The imaginary conductivity is plotted with the real conductivity in the insets. The data is fit using a two-fluid model which reduces to the Drude model above $T_c$ \[1, 3\]. The thick solid line is the experimental data. The dashed lines are the overall fit to the data. In (a), the dotted line is the superconducting pair component and the thin solid line is the quasiparticle component. (c)-(d) Optically induced changes in the imaginary conductivity at 60 and 95 K at an excitation fluence of 12 $\mu J/cm^2$.

Figure 2: Normalized induced change in the imaginary conductivity (60 K) as a function of time at various frequencies. The curves are displaced vertically for clarity. The solid lines are fits to the function \(y = a \times \exp(-t/\tau) + b\). The inset shows the measured lifetime as a function of frequency.
(a) 60 K

\[ \sigma_{im} (\Omega \text{ cm})^{-1} \]

\[ \times 10^4 \]

\[ 6 \]

\[ 4 \]

\[ 2 \]

\[ 0 \]

\[ 0.4 \]

\[ 0.8 \]

\[ 1.2 \]

\[ 1.6 \]

\[ 2 \]

Frequency (THz)

\[ \tau = 175 \text{ fs} \]

(b) 95 K

\[ \sigma_{im} (\Omega \text{ cm})^{-1} \]

\[ \times 10^3 \]

\[ 6 \]

\[ 4 \]

\[ 2 \]

\[ 0 \]

\[ 0.2 \]

\[ 0.6 \]

\[ 1 \]

\[ 1.4 \]

\[ 1.8 \]

\[ 2 \]

Frequency (THz)

\[ \tau = 51 \text{ fs} \]

(c) 60 K

Unpumped

\[ 10 \text{ ps} \]

\[ 1 \text{ ps} \]

\[ 2 \text{ ps} \]

(d) 95 K

Unpumped

\[ 50 \text{ ps} \]

\[ 4 \text{ ps} \]

\[ 1 \text{ ps} \]
The graph shows the normalized change in absorption coefficient $\Delta\sigma_{im}/\sigma_{im}$ as a function of time (ps) for different frequencies: 0.5 THz, 1.0 THz, and 1.5 THz. Each frequency has a distinct curve, indicating different decay rates. The inset graph displays the lifetime $\tau_{\sigma}$ (ps) as a function of frequency (THz). As the frequency increases, the lifetime decreases, suggesting faster decay rates at higher frequencies.