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Terahertz Probing of Carrier Dynamics in Hg-Based High-Temperature Superconducting Thin Films

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Abstract: We report on our investigation of time-resolved carrier dynamics in a Hg-based high-temperature superconducting film (Hg-Ba-Ca-Cu-O), using optical excitation and THz probing. The observed picosecond time-scale photoresponse suggests the material's potential applications for high-speed photodetectors.

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Among high-temperature superconductors (HTSs), the family of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ (Hg-based; HBCCO) compounds are characterized by the record-high critical temperature T_c of 134 K at ambient pressure,[1] making them extremely attractive from the applied point of view. At the same time, like the other members in the HTS family, the very basic question of the origin of the microscopic paring mechanism in the Hg-based superconductors has yet to be determined,[2] despite a very large number of experimental and theoretical studies performed so far.[3]

All-optical, femtosecond, pump-probe experiments have been employed to study carrier dynamics of HTS materials since shortly after their discovery.[4] In these experiments, electronic nonequilibrium states are generated by femtosecond pulses and the subsequent relaxation dynamics of excited carriers are coherently monitored by time-delayed, femtosecond probe pulses from the same source through the transient change of either normalized reflectivity or transmissivity.[5] The main drawback of the all-optical pump-probe technique is that the probe photon energy is typically between 1 and 2 eV and is much larger than the relevant energy scale in HTS materials, e.g., the superconductor energy gap, which is up to tens of meV. In addition, the *d*-wave of the gap, can lead to uncontrolled pair breaking by the probe beam, requiring a very weak probe power to minimize such effect. In this context, the pump-probe method based on the optical pump and THz probe (OPTP)[6] is much preferred, as the energy of the probing THz photons is small and does not break Cooper pairs. Another advantage of OPTP manifests in the simultaneous measurements of the amplitude and phase of the photoresponse (associated with optical constants) without resorting to the complex Kramer-Kronig relationship, resulting in the pure optically excited carrier dynamics without perturbation from the probe itself.

In this work, we have implemented the OPTP technique to study the carrier dynamics of a 500-nm-thick Hg-based HTS film, fabricated on a 500-μm-thick $LaAlO_3$ substrates. Our films were synthesized from 500-nm-thick Re-Ba-Ca-Cu-O precursor films, RF-magnetron sputtered on $LaAlO_3$ at room temperature from a single target. Next, the mercuration process was performed *ex-situ* in a sealed, evacuated quartz ampule, using an unreacted Hg-1223 pellet as a source of mercury. The precursor films were placed on top of the pellet and were wrapped together using a gold foil. The loaded ampoule was placed inside a furnace, and the samples were kept at 800°C for 180 min. Finally, they were cooled at a rate of 120°C/hour to the ambient temperature. X-ray diffraction analysis of our films demonstrated that they were predominantly composed of the *c*-axis-oriented Hg-1212 phase, together with some amount of the Hg-1223 phase and other intergrowths. The resistively measured superconducting transition of our films was rather broad, with the onset $T_{c,\mathrm{on}}$ at 123 K and the zero-resistance $T_{c,0}$ at 110 K.

Our OPTP experiments (see Fig. 1) were based on a 1-kHz, 800-nm-wavelength, 50-fs-duration amplified laser system with the total output of ~500 mW. The output from the laser was split to three beams: one beam was used to optically pump the sample to generate quasiparticles, the second was used to generate THz radiation via optical rectification in a 1-mm-thick ZnTe crystal, while the third beam (very weak) was used to detect the THz transmission through the HTS film via electro-optic sampling[7] based on a 2-mm-thick ZnTe crystal. To avoid serious heating problems and at the same time take the achievable signal-to-noise ratio (SNR) into consideration, we used a pump power of ≤ 5.5 mW (corresponding to fluence of ~13 μJ/cm^2), which was about four times smaller that reported by Averitt *et al.* in their OPTP studies of Y-Ba-Cu-O superconductors.[8] The HBCCO film was placed inside a compact continuous-flow, nitrogen cryostat with the temperature controlled between ~78 K and room temperature.

In terms of the two-fluid[2] interpretation for superconductivity below T_c, external optical excitation breaks Cooper pairs, leading to a decrease of the superfluid density and an increase of normal-state quasiparticle density. In other words, the optical excitation transiently increases the real part σ_1 of the complex conductivity σ and decreases the imaginary part σ_2. By probing the change of the THz transmission as a function of temporal delay with regard to the arrival of the optical excitation, transient values of $\Delta\sigma_1$ and $\Delta\sigma_2$ and, accordingly, the quasiparticle dynamics can be extracted.

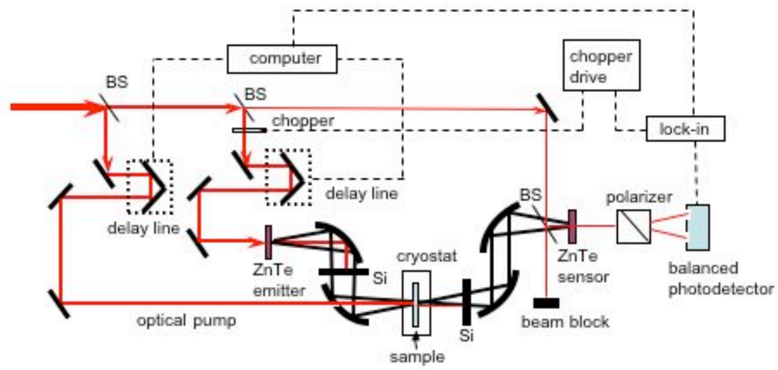

Figure 1. Schematic of the OPTP set-up.

During the course of our experiments, we fixed the optical delay line associated with the THz probe signal generation at the position where the positive, maximum peak of the THz electric-field waveform occurred. By varying a second optical delay line (associated with the optical pump), we changed the temporal distance between the optical pump and the fixed THz probe. In this manner, the transient THz transmission was recorded. This type of OPTP is also called one-dimensional (1-D) OPTP, as compared to 2-D OPTP where both delay lines are varied.[9]

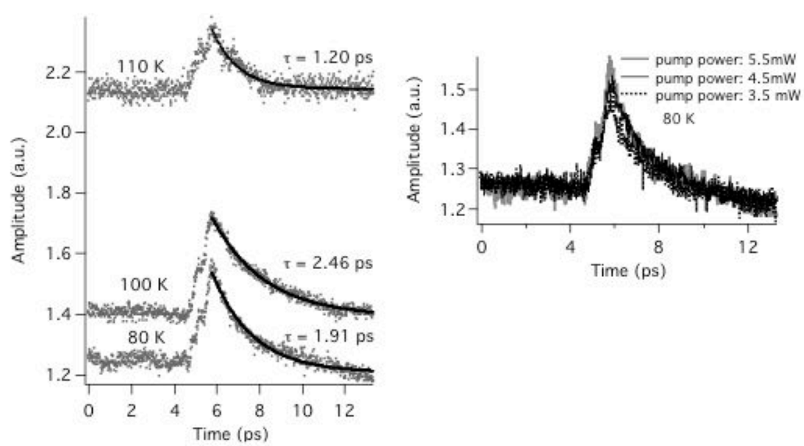

Figure 2. OPTP results for the Hg-Ba-Ca-Cu-O film recorded at various temperatures (a); and for different pump powers at 80 K (b).

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For $T > T_C$ we did not observe any signal, apparently, because of a weak overall change in the density of free carriers (normal electrons) and the resulting low signal. The OPTP measurements below T_C were performed at three fixed temperatures: 80 K, 100 K, and 110 K. Figure 2(a) shows the experimental results obtained for a constant optical pump power of ~5.5 mW. We note that without the optical pump (see the pre-signal part on each waveform), there is higher THz transmission as the temperature increases. It can be derived that the superconducting film transmission t is dependent on σ: $t \propto 1/(1 + n + Z_o \sigma d)$, where n is the refractive index of the substrate, $Z_o \approx 377\ \Omega$ is the intrinsic impedance of vacuum, and d is the film thickness. Furthermore, we note that σ_2 (associated with Cooper pairs) dominates in the complex σ when $T < T_C$. According to the two-fluid model, σ_2 drops rapidly as the temperature increases, and accordingly the magnitude of t increases rapidly. This explains our experimental results shown here. At 110 K, which is the closest value to the T_c, the change of the transmission Δt is much smaller than at either 80 K or 100 K. Tentatively, we explain this by resorting to the relationship between t and σ again. Near T_c, the density of Cooper pairs is very low [recall the free-carrier density $n_s \propto 1 - (T/T_c)^2$] and the incident optical pulse is likely to break all pairs, resulting in a transient normal state. The transition from the superconducting state to the normal state involves a small number of Cooper pairs, such that σ_2 changes only slightly and, accordingly, the measured signal is small.

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The other important information that we want to extract from the OPTP experiments is the relaxation time τ of the photoinduced quasiparticles, which can be obtained by fitting the falling edge of a signal with a single or double exponential function. We have found that the single exponential fitting worked the best [solid curves in Fig. 2(a)], and the fitted τ's were in the single picosecond time scale. It should be also stressed that no phonon bottleneck effect exhibited in the photoresponse signals, suggesting that Hg based HTS materials are very promising for ultrafast photodetectors. [10]

We also studied the pump power dependence of the photoresponse, as shown in Fig. 2(b). With the pump power increasing from ~3.5 mW to ~5.5 mW at 80 K, we saw the corresponding increase of the signal amplitude. No obvious change of carrier lifetime was observed. Thus, the signal amplitude increase could be simply explained as the higher generated density of quasiparticles resulted from the higher pump fluence. The independence of carrier lifetime vs. pump fluence also supports that the heating effect is indeed negligible in our experiments.

In conclusion, we have studied the carrier dynamics in the superconducting Hg-based thin films, by the means of time-resolved THz probe. This specific HTS material exhibits the single-picosecond photoresponse with no phonon bottleneck, suggesting they are potentially very promising for photodetector applications. A more complete investigation, involving with a wider temperature range and a higher system SNR, is currently under way in our laboratories, in order to more fully understand the physical (microscopic) mechanism of the ultrafast photoresponse in HBCCO, and HTS materials in general.

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