Pulsed terahertz-beam spectroscopy as a probe of the thermal and quantum response of $YBa_2Cu_3O_{7-\delta}$ superfluid

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Pulsed terahertz spectroscopy is used to determine the superfluid response of a $YBa_2Cu_3O_{7-\delta}$ film under both thermal and optical stimulation. The coherent, time-domain technique is used in a novel configuration to directly measure the complex conductivity of the film versus temperature and continuous-wave laser illumination. At 0.6 T_c, the superfluid shows an identical response regardless of whether the stimulus is thermal or optical. This contrasts with the behavior of the superfluid at 0.26 T_c, where dramatic differences are observed depending on whether the sample is heated or subjected to optical illumination. It is suggested that these differences arise from an enhanced contribution due to quantum effects, and thus also from a strong temperature dependence of the quasiparticle recombination time at low temperatures. © *1995 American Institute of Physics.*

The emergence of high critical temperature superconductors has resulted in a renewed interest in the study of the mechanisms governing the photoresponse of thin superconducting films. Although the investigation of photoresponse is motivated primarily by potential applications of superconducting films, such as in broadband optical detectors and optically controllable filters, there is also a strong desire to understand the nature of nonequilibrium superconductivity.[1–7] The photoresponse (i.e., the reaction to external optical stimulation) arises as a consequence of perturbations in the superconducting condensate of paired electrons. There are essentially two kinds of responses, and they differ by the way in which the absorbed photon energy gets dissipated within the film. In the first and more prevalent case, the quasiparticle density is altered as a consequence of heating the film—the so-called thermal, or bolometric response. The second kind of response is essentially quantum in nature and involves pair breaking and the subsequent recombination of pairs. Although the latter process is in principle faster than the former one, it is a challenging problem to devise experimental conditions which clearly distinguish between the two responses.

The generation and measurement of transient voltage pulses in a superconducting bridge illuminated by a pulsed laser is a common way to investigate the optical response of such materials. However, the interpretation of such results is not straightforward, and much controversy exists regarding the mechanisms responsible for the observed behavior. Thus, some reports attribute fast signals to a rapid bolometric response or a kinetic inductance response,[4,7] while most other researchers associate a short-duration response with nonbolometric behavior.[2,3,5] The optical response of superconductors can also be studied with sub-100-fs temporal resolution in transient reflectivity or absorption experiments,[8] although interpreting the results of such pump-probe experiments can also be difficult.[9]

In this letter, we describe a novel experimental technique in which ultrashort pulses of radiation with terahertz bandwidth are used to directly probe the response to optical radiation of a $YBa_2Cu_3O_{7-\delta}$ (YBCO) superfluid. With this unique ability to determine whether the changes in the quasiparticle density are induced by either bolometric or quantum effects, we find that the photoresponse of YBCO has a thermal character at a high reduced temperature, while at a low reduced temperature it has essentially a quantum origin.

The experiment relied on the generation, transmission, and detection of free-space, terahertz-bandwidth, electromagnetic transients,[10–12] which were both produced and measured through optoelectronic conversion of ultrashort optical pulses.

Two beams split from the output of a mode-locked Ti-sapphire laser (wtih 810-nm center wavelength and 500-mW average output power provided 75-fs pulses at a 76-MHz repetition rate) were used to drive the optoelectronic gates. Hertzian-dipolelike antennas fabricated on semiconductors having an ultrashort carrier lifetime and separated by ~4 cm were used as the transmitter and receiver. The field amplitude and phase information of the subpicosecond, free-space radiating wave form from the transmitter were obtained directly from the temporal profile of the received signal, which was measured via photoconductive sampling at the detector.[10]

A third optical beam illuminated the superconductor with an excitation beam of high photon energy ($h\nu \sim 2.3$ eV) from a cw argon laser. This beam was used to induce changes in the quasiparticle density either through direct pair breaking, thermal excitation, or both. The response of the film as probed by the terahertz beam transmission with this cw optical illumination present [as in Fig. 1] was compared to the film response when the ambient temperature of the sample was changed in the absence of the cw laser illumination. A careful inspection of the terahertz transmission at high temperatures (0.6 T_c) reveals no distinction between

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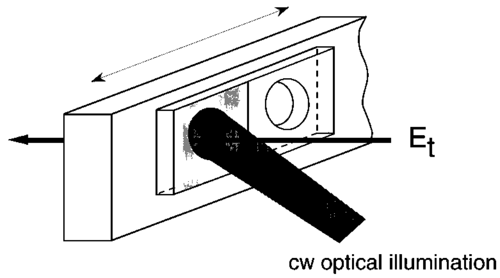

FIG. 1. Experimental configuration for terahertz-beam probing of a YBCO film under optical illumination. The film is mounted on one hole of the cold finger while the second hole is covered by a blank substrate. The cold finger may be translated to probe either the substrate or the superconductor. E_t is the electric field of the terahertz beam.

this optical-stimulation experiment and the experiment where only the substrate temperature was changed. In contrast, the same evaluation of the transmitted terahertz signal at low temperature ($0.26\ T_c$) produces a dramatic difference between the purely thermal and the optical response. The samples used in our measurements were high quality superconducting thin films of YBCO (~900 Å thick, $T_c = 90$ K) on $LaAlO_3$.[13] The film area illuminated by the argon-laser light and probed by the terahertz beam was approximately 5 mm in diameter.

The terahertz waveform transmitted through the YBCO film at three different temperatures and in the absence of cw laser illumination is shown in Fig. 2. Two features can be clearly seen: with increasing temperature, the transmitted electric field both increases in amplitude and experiences a phase shift associated with the peak position of the waveform. These are the results of a dramatic change in the quasiparticle and superfluid densities. The amplitude change at one of the frequencies in the broadband spectrum—400 GHz—is also shown as a function of the ambient temperature in the inset of Fig. 2. Progressive condensation of quasiparticles and formation of Cooper pairs below T_c dramatically decrease the transmitted field amplitude due to screening by the superfluid. This amplitude change together with the phase shift absolutely determines the change in superfluid density as a function of temperature. Figure 3 shows the terahertz waveform transmitted through the YBCO film illuminated with various cw optical powers, while the cryostat maintains the ambient substrate temperature at 24 K. With the change of optical power, both the amplitude and phase of the terahertz pulse are changing due to the optically induced changes of the superfluid. The inset shows the amplitude change at 400 GHz as a function of the optical power. Clearly, at a critical power of approximately 110 mW, the film undergoes a transition to the normal state.

In order to assess changes in superfluid density with respect to changes in either the sample temperature or the power of cw optical radiation, we make use of the transmitted terahertz pulses to extract the complex conductivity of the YBCO film.[11,12] The imaginary conductivity of the superfluid can be expressed in terms of its relative phase and amplitude compared to that of the film in the normal state (where the imaginary conductivity is close to zero) by

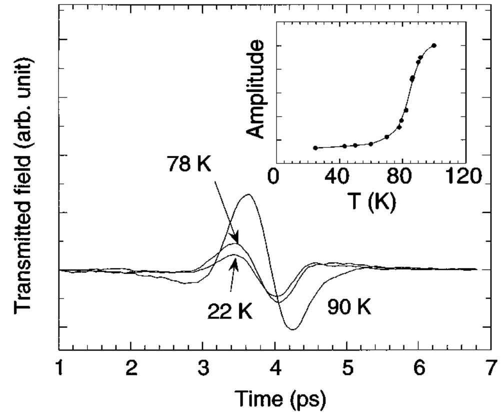

FIG. 2. Time-domain waveform for terahertz-bandwidth pulses transmitted through YBCO thin film in the configuration of Fig. 1(b) at three different temperatures. Inset: The transmitted field amplitude of the 400 GHz component of the pulse spectrum as a function of temperature.

$$\sigma_2(\omega) = C\frac{A_0(\omega)}{A(\omega)}\ \sin[\Phi(\omega)], \tag{1}$$

where C is a constant, $A_0(\omega)$ is the normal state amplitude, $A(\omega)$ is the amplitude at a certain temperature or cw power, and $\Phi(\omega)$ is the phase relative to that of the normal state. The superfluid density N_s is directly related to the imaginary conductivity by $\sigma_2 = N_s e^2/m\omega$, where ω is the angular frequency of the terahertz radiation, e is the charge of the carriers, and m is the effective mass. Using this relation, we uniquely determine the temperature and power dependence of N_s through the experimentally extracted conductivity.

To compare the dependence on temperature and power, a quantity for reduced temperature is introduced:

$$t = \frac{T}{T_c} = \frac{T_0}{T_c} + \left(1 - \frac{T_0}{T_c}\right)\frac{P}{P_c}, \tag{2}$$

where T_0 is the ambient temperature at which the power-dependence measurement is made, P_c is defined as the critical power when N_s is zero, P is the cw laser power, and T_c is

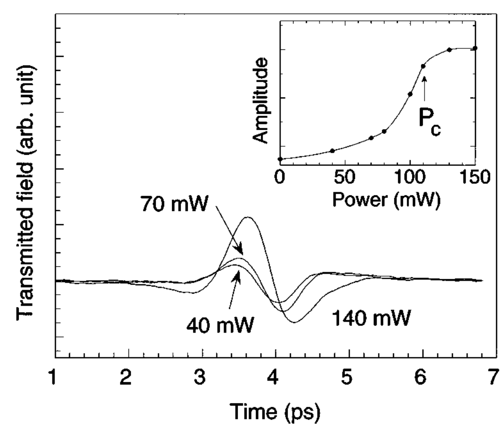

FIG. 3. Time-domain waveform for terahertz-bandwidth pulses transmitted through YBCO thin film in the configuration of Fig. 1(a) at three different power levels. Inset: The transmitted field amplitude of the 400 GHz component of the pulse spectrum as a function of power.

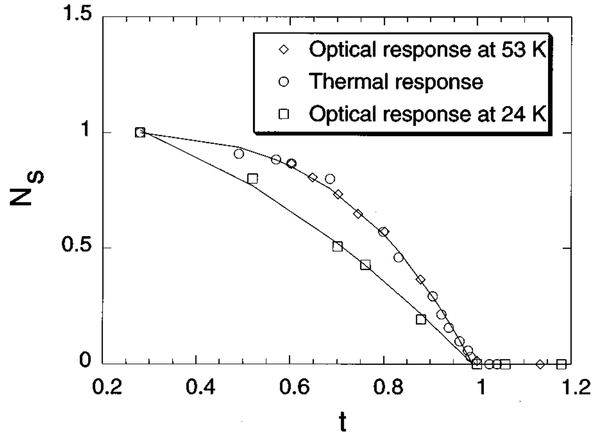

FIG. 4. The response of the YBCO superfluid density to reduce temperature (see text). Open circle: thermal response, $N_s(T)$; open square: optical response, $N_s(P)$, with $T_0/T_c=0.26$; and open triangle: optical response with $T_0/T_c=0.6$. The solid lines are curve fits as described in the text.

the transition temperature. Film heating due to optical absorption is proportional to the power of the cw laser illumination, an assumption supported by temperature measurements using a thermal diode positioned adjacent to the laser spot on the YBCO film.

Figure 4 shows how the superfluid density responds to changes in the reduced temperature in both the purely thermal and cw-illumination cases. In the absence of laser illumination, the temperature dependence of the superfluid density, $N_s(T)$, can be approximated below T_c by a power law $[1-(T/T_c)^n]$ with $n\sim4$, as shown by the fitted line. Upon closer inspection, this curve could be broken up into several regimes, where just below the critical temperature its slope is approximately 3.8, and where at low temperatures its behavior may be essentially linear. These properties differ from the isotropic, BCS *s*-wave characteristics of conventional superconductors, and they resemble closely results of Hardy *et al.*[14] on YBCO single crystals. The power dependence of the superfluid density, $N_s(P)$, is shown at two ambient temperatures: 24 K ($\sim$0.26 T_c) and 53 K ($\sim$0.6 T_c). At 0.26 T_c, $N_s(P)$ is a near-linear function below the critical power, while at 0.6 T_c, $N_s(P)$ is essentially identical to the functional dependence of the thermal response. A contrasting response of the superfluid density to the thermal and optical stimulations at low temperatures suggests that quantum effects rather than heating are the important ingredients in the photoresponse at the lower temperature of 0.26 T_c. A nearly linear functional dependence of $N_s(P)$ further substantiates this point, since the density of nonthermally generated quasiparticles should be proportional to the number of absorbed photons. The data at 0.6 T_c indicate that photon absorption in the film at high temperature is mainly dominated by thermal effects.

We believe that the key to understanding the different photoresponse in YBCO films at different temperatures lies in the nature of the nonequilibrium superconducting process. The absorption of optical photons which have energy well above the superconducting band gap results in a nonequilibrium distribution of electron energies. The high-energy electrons (quasiparticles) have two channels to dissipate their excess energy. The first (or quantum mechanical one) is through cascade pair breaking and recombination of pairs to form a condensate which will eventually reach an equilibrium state. Since the optical photons act as the source of pair breaking here, the number of pairs broken is proportional to the number of absorbed photons, and thus also to the incident power. This is very different from other nonequilibrium phenomena, such as the enhancement of superfluid density upon microwave irradiation[15] and quasiparticle tunnel injection.[16] In these cases, the quasiparticles at low-energy states are redistributed upon disturbance.

The second channel of energy dissipation (the bolometric, or thermal one) is through the coupling of electrons with phonons, which ultimately leads to heating of the sample. What determines the overall response is the time scale of the two processes. A complete understanding of the whole process would require a real understanding of the nonequilibrium recombination process and its time constant at different temperatures, neither of which are known in high T_c superconductors. The response observed in this experiment indicates a dramatic change in the time scale when going from just below T_c to well below T_c, with the recombination process becoming dominant as temperature drops.

In conclusion, we have directly observed, through a novel implementation of a terahertz-beam spectroscopy system, the different response of YBCO superconducting thin films to optical stimulation at different temperatures. This was accomplished by looking at the overall behavior of the superfluid density as extracted from the directly measured complex conductivity.

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