Femtosecond optical response of Y-Ba-Cu-O thin films: The dependence on optical frequency, excitation intensity, and electric current

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We have performed a series of femtosecond reflectivity experiments on various Y-Ba-Cu-O thin films at temperatures ranging from 12 to 300 K. In particular, the dependence of the optical response on probing laser frequency, pumping laser intensity, and bias electric current has been measured. Results obtained at room temperature provide quantitative information on the position of the Fermi level in films with different oxygen content. Systematic analysis of the measurements performed in the superconducting state indicates that the optical response associated with nonequilibrium properties of Y-Ba-Cu-O depends strongly on excitation intensity, sample thickness, and bias current. The results cannot be satisfactorily interpreted as the relaxation dynamics of quasiparticles, and a simple two-fluid model is shown to fail to explain data obtained under low laser excitation. Several tentative explanations are proposed, which provide a more comprehensive understanding of the transient optical response of Y-Ba-Cu-O.

I. INTRODUCTION

Superconductor photoexcitation studies have been a subject of intense investigations for the last 20 years.[1] Early experiments were performed on metallic superconductors, using nanosecond and picosecond laser pulses, and were focused on the dynamics of the photoinduced superconducting-to-normal transition. It has been demonstrated experimentally[2,3] and explained theoretically[4] that under moderately weak optical excitation a superconducting thin film undergoes a transition into a nonequilibrium (transient) intermediate state, which is characterized by the coexistence of spatially separated superconducting and normal domains in the film. The dynamics of the intermediate state was found to be on a picosecond time scale,[3,5] in contrast to the slow, heat-diffusion related bolometric response observed in films under strong laser excitation. The discovery of high-temperature superconductors (HTS) prompted a series of transient photoexcitation measurements.[6–10] It was observed that, unlike in low-T_c materials, the response of optically thick $YBa_2Cu_3O_{7-x}$ (YBCO) films to moderate excitations was primarily bolometric with a small nonthermal component. Preliminary experimental work demonstrated that for ultrathin films the relative magnitude of the nonthermal component was enhanced and could be as fast as tens of picoseconds.[10]

Recently, a different class of laser excitation experiments has been performed to investigate transient nonequilibrium properties of both low- and high-T_c superconducting materials,[11–20] namely, femtosecond pump-probe measurements. In particular, femtosecond time-resolved measurements of the differential reflectivity, $\Delta R/R$, and differential transmissivity, $\Delta T/T$, have been performed on YBCO at various temperatures. The results obtained at room temperature were interpreted using the thermomodulation model, which was used previously to explain the dynamics of $\Delta R/R$ in metals,[21,22] and indicated different positions of the Fermi level (E_F) in the oxygen p band of oxygen-rich and oxygen-poor samples. On the other hand, the picosecond response of $\Delta R/R$, measured on optically thick YBCO films at temperatures below T_c, has been interpreted as a direct indication of the relaxation dynamics of quasiparticles[16,17] and/or the superconducting order parameter.[18]

Even though pump-probe measurements provide an excellent temporal resolution, it appears to be difficult to directly relate the purely optical (high-frequency) response to the change of electrical (low-frequency) properties of the material when its detailed band structure is not included. The very recent results of Reitze *et al.*[20] have implied that a detailed energy band structure has to be taken into account if any physical significance is to be assigned to these relaxation rates measured on YBCO at optical frequencies. Unfortunately, in HTS materials the energy band structure and its temperature dependence have not been well defined, making the transient optical signals difficult to explain.

Varying the probe wavelength can provide a more reliable way to solve the above problem than varying the material. Chekalin *et al.*[19] recently claimed to have observed the superconducting energy gap using the technique of pump and continuum probe. However, there are two problems associated with their measurement: first, the pump laser intensity was high enough to possibly cause stochiometric changes in the YBCO sample; second, there was no observation of the Fermi level at room temperature around the wavelength where they claimed to observe the opening of the energy gap at low temperatures.

In this study we present a series of femtosecond reflectivity measurements on YBCO films. We discuss in detail the dependence of $\Delta R/R$ on probe laser frequency, pump laser intensity, and bias electric current. In particular, our study provides information on the shift of the

Fermi level between the oxygen-rich and oxygen-poor samples, as well as on the nature of the transient optical response in optically thin and thick YBCO films under weak and strong laser excitations.

Our paper is organized as follows: The next section provides a brief survey of the relation between the optically measured $\Delta R/R$ and the change of the dielectric permittivity, $\Delta\varepsilon$, for optically thick and thin films, in connection with the models commonly used to explain the change of optical and electrical properties of HTS materials. Section III describes the sample fabrication and experimental arrangement. The discussion of our experimental results and a comparison with existing models are presented in Sec. IV. Finally, conclusions and suggestions for future investigations are presented in Sec. V.

II. BACKGROUND

The normal-incidence reflectivity, R, for an optically thick sample can be described by

$$R=\frac{(n-1)^2+k^2}{(n+1)^2+k^2}\ , \tag{1}$$

where n and k are the real and imaginary parts of the index of refraction. The real and imaginary parts of the dielectric permittivity (ε_1 and ε_2, respectively) are related to n and k as

$$\varepsilon_1=n^2-k^2\ , \tag{2}$$

$$\varepsilon_2=2nk\ . \tag{3}$$

Therefore, using Eqs. (1)–(3), $\Delta R/R$ can be simply expressed as[23]

$$\Delta R/R=a\Delta\varepsilon_1+b\Delta\varepsilon_2\ , \tag{4}$$

where a and b are given by the unperturbed values of ε_1 and ε_2.

For most semiconducting materials, $a\gg b$, and $\Delta R/R$ is thus dominated by $\Delta\varepsilon_1$. In contrast, for most metals, $a\ll b$, and $\Delta R/R$ is essentially proportional to $\Delta\varepsilon_2$. Using unperturbed values of ε_1 and ε_2 obtained from ellipsometry measurements,[24] we have calculated the values of a and b for $YBa_2Cu_3O_{6.85}$ at the energy of 2 eV and found them to be 0.16 and 0.25, respectively, which are similar to the values calculated in Ref. 25. Thus, for YBCO, $\Delta R/R$ could, in principle, depend strongly on both $\Delta\varepsilon_1$ and $\Delta\varepsilon_2$. However, one must remember that the magnitudes of $\Delta\varepsilon_1$ and $\Delta\varepsilon_2$ themselves can vary substantially, making $\Delta R/R$ dominated by $\Delta\varepsilon_1$ if $\Delta\varepsilon_1\gg\Delta\varepsilon_2$ or $\Delta\varepsilon_2$ if $\Delta\varepsilon_1\ll\Delta\varepsilon_2$.

The dependence of the optical reflectivity on sample properties changes when the measurement is performed on an optically thin film, fabricated on a non-index-matched substrate. In this case, multiple reflections from the sample substrate interface must be taken into account. As a result, the relation between $\Delta R/R$ and $\Delta\varepsilon$ is much more complicated. In particular, the values of a and b vary with the sample thickness. It is very important to note that when $\Delta\varepsilon_1$ dominates the $\Delta R/R$ signal, the sign of $\Delta R/R$ depends on the sample thickness, d, whereas when $\Delta\varepsilon_2$ dominates the $\Delta R/R$ signal, the sign (either positive or negative) of $\Delta R/R$ is independent of d. For example, when $\Delta R/R$ measured for samples with different d's is dominated by $\Delta\varepsilon_1$, $\Delta R/R$ changes sign for a thickness difference, Δd, given by the relation $\Delta d/\lambda=1/(4n)$. For 2-eV optical excitation energy ($\lambda=620$ nm), the calculated Δd for YBCO is ~ 75 nm, which is within the range of thickness variations of the samples used in our study.

The thermomodulation model is most commonly used in explaining the transient ΔR and ΔT signals in d-band metals and YBCO-related materials in the normal state. In this model, the pump pulse heats carriers (either electrons or holes) and modifies the electronic occupancy near the Fermi level on a time scale typically shorter than the excitation pulse width. This "smearing" decreases (increases) the occupancy of states below (above) E_F. Thus, a probe monitoring the interband transition from the filled d band to the p states below (above) E_F measures a positive (negative) $\Delta\varepsilon_2$.

When the ambient temperature is lowered below T_c, the free-carrier density of states in the superconductor is modified substantially. According to BCS theory, in the superconducting state electrons form bosonlike Cooper pairs occupying a single energy level separated from E_F by the superconducting energy gap Δ; while unpaired electrons (quasiparticles) occupy a continuum of states, starting Δ above E_F. The 2Δ gap is temperature dependent. At a given temperature ($0<T<T_c$) only a fraction $f_C(T)$ of the total number of free carriers forms Cooper pairs, while the rest $(1-f_C)$ remains as quasiparticles. Treating the superconducting condensate and normal electron gas as separate conducting "fluids",[26] one can obtain ε of the superconducting sample below T_c as

$$\varepsilon=(1-f_C)\varepsilon_D+f_C\varepsilon_C+\varepsilon_H\ , \tag{5}$$

where ε_D, ε_C, and ε_H are the contributions from the quasiparticles (based on the Drude model), the Cooper pairs, and the high-frequency interband (e.g., $d\rightarrow p$) transitions, respectively.

Under optical illumination the thermal equilibrium between the superconducting condensate and quasiparticles is disturbed (often very strongly), since photons with energies $\gg 2\Delta$ can break Cooper pairs and create quasiparticles. The absorption of a single 2-eV photon in YBCO creates through the cascading processes of carrier-carrier and carrier-photon scattering many quasiparticles with energies widely distributed above 2Δ.[27] Thus, even a relatively weak optical excitation leads to highly nonequilibrium transient Cooper-pair–quasiparticle distributions.

As we have already mentioned in Sec. I, the situation is the most complicated for a relatively weak perturbation (typically 1 to 10 μJ/cm^2 at 2 eV), since it can lead to the nonequilibrium intermediate state, resulting in an excitation-intensity-dependent sample resistivity. The intermediate state was first observed by Sai-Halasz *et al.*[2] in metallic superconducting materials, and is characterized by the coexistence of superconducting and normal regions in the excitation area. According to the Elesin theory,[4] this nonequilibrium state is stationary only for a

particular laser excitation power, called the critical power. For perturbations different than the critical one, the intermediate state is nonstationary, with the dynamics directly related to the temporal dependence of the excitation pulse.[3] The superconducting-normal region boundaries move toward the superconducting state (decreasing the volume of this phase) for a perturbation above the critical power and toward the normal phase in the opposite case. This explains the experimental observations[2,3] that the resistivity of a photoexcited superconducting film appears at a certain excitation threshold level, and then rises smoothly with excitation intensity, finally reaching the value corresponding to the normal-state resistivity. The intermediate state was found to be an intrinsic feature of optically driven nonequilibrium metallic superconductors and is expected to exist in HTS materials.

Finally, very intense ($\gg\mu J/cm^2$) optical excitation of superconductors (both metallic and HTS) always results in a thermally induced transition to the normal state. The overall response is expected to be slow and can be explained using the bolometric and heat diffusion model.

III. EXPERIMENTAL PROCEDURES

A number of epitaxial YBCO films with thicknesses from 80 to 300 nm have been grown on $SrTiO_3$ single crystals by *in situ* rf magnetron sputtering.[28] The films typically exhibited about 1.5-K-wide (10–90 %) resistive superconducting transitions, with zero resistivity, T_{c0} between 87 (YBCO-I: 200 nm thick) and 83 K (YBCO-II: 80 nm thick). In addition, we have tested a 200-nm-thick laser-ablated sample (YBCO-III) fabricated at the New York State Institute on Superconductivity,[29] and a 280-nm-thick sputtered film (YBCO-IV) provided by the Westinghouse Science and Technology Center.[30] The superconducting transition for the YBCO-IV sample was 0.5 K wide and its T_{c0} was 89.5 K. For the sample YBCO-III, T_{c0} was 86.5 K. All the above films exhibited critical current densities, J_c, greater than 10^6 A/cm^2 at liquid-nitrogen temperature. A partially oxygen-depleted sample (YBCO-V) and an oxygen-poor sample (YBCO-VI) were also fabricated. The YBCO-V film was deposited together with YBCO-I, and then was furnace annealed at 500 °C for 20 min in a pure Ar atmosphere. As a result, its T_c was lowered to 27 K, consistent with a decrease of the oxygen content to about 6.6.[31] The YBCO-VI sample was deposited together with YBCO-IV and then was annealed at 650 °C for 2 h in a pure Ar atmosphere. As a result, YBCO-VI exhibited a semiconducting behavior with thermally activated electrical transport for the entire temperature range. The estimated oxygen content for the YBCO-VI sample is less than 6.45. For transport-current-related measurements, the optically thick and thin films, fabricated in the same run as the YBCO-IV and YBCO-II samples, were patterned into 50-μm-wide and about 1-mm-long microbridges using a laser-ablation method.[32]

Femtosecond measurements were performed using a colliding pulse mode-locked (CPM) laser, which was either unamplified or amplified by a copper vapor laser. Depending on the amplification arrangement, the CPM provided a train of pulses with either a high repetition rate (~100 MHz) and low energy (~0.1 nJ/pulse), or with a moderate repetition rate (~8.5 KHz) and high energy (>1 μJ/pulse). This arrangement allowed us to change the pump laser intensity, I_p, for 2-eV photons by more than 3 orders of magnitude, as well as to use a probe pulse at different wavelengths selected from a white light continuum. Time-resolved $\Delta R/R$ measurements were performed with a temporal resolution of 100±20 fs, using a conventional pump-probe arrangement. Lock-in and differential detection techniques were used to obtain a good signal-to-noise ratio. The polarization of the probe beam was rotated by 90° relative to that of the pump beam to reduce the coherent artifact. Both beams were focused on the sample using a 5-cm lens, resulting to a focusing spot size of 40±10 μm. The measurements have been performed in a temperature-controlled, continuous-flow optical cryostat in the temperature range between 12 and 300 K.

IV. RESULTS AND DISCUSSION

A. Room-temperature measurements

Figure 1 illustrates the temporal dependence of the $\Delta R/R$ signal, measured on the YBCO-III sample at room temperature. The probe photon energy was varied form 1.91 to 2.25 eV (the 2.25-eV data are not shown in Fig. 1), while the pump photon energy was kept constant at 2 eV with I_p about 10 μJ/cm^2. We note that for all probe photon energies, $\Delta R/R$ exhibits a similar behavior—a positive pulse-width-limited rise, followed by a fast decrease, and then a long-lasting (several nanoseconds) plateau. Following Refs. 12 and 14, such a

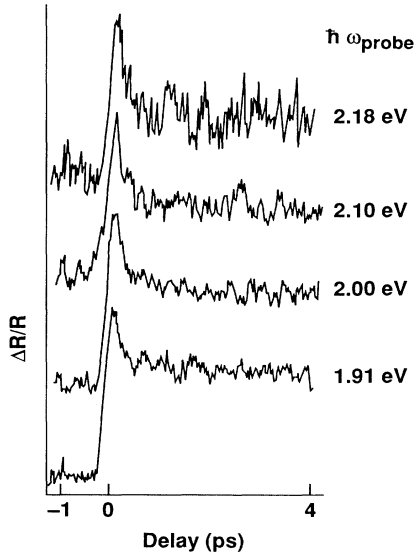

FIG. 1. Normalized changes in reflectivity $\Delta R/R$ ($\sim 10^{-4}$) measured on the YBCO-III (oxygen-rich) sample at room temperature using the probe photon energies indicated in the figure.

"bolometric" response can be simply explained by the smearing of the distribution near E_F, which, in the case of YBCO, lies in the oxygen p band. The positive $\Delta R/R$ at all wavelengths results from positive $\Delta\varepsilon_2$ with probing smeared hole states below E_F. A behavior very similar to that presented in Fig. 1 was also obtained for sputtered films (YBCO-I sample).

At 2 eV, $\Delta R/R$ was positive for all our samples with thickness from 80 to 300 nm, implying (see Sec. II) that $\Delta\varepsilon_2 \gg \Delta\varepsilon_1$. Therefore, $\Delta R/R$ is proportional to $\Delta\varepsilon_2$, and its behavior can indeed be explained by the thermomodulation model; in this sense, YBCO exhibits a behavior similar to typical d-band metals, such as Cu. We have also observed that the temporal response of $\Delta R/R$ at 2 eV has little dependence on pump pulse intensity from 0.3 to 100 μJ/cm^2. Another important observation is that since in our experiments $\Delta\varepsilon_2$ does not show a sign reversal in the energy range from 1.91 to 2.25 eV, E_F must be at least 2.25 eV above the Cu d^9/d^{10} band. This result, which is a direct consequence of the thermomodulation model, differs from earlier findings of Kazeroonian *et al.*,[14] who concluded that E_F was about 2 eV above the Cu d^9/d^{10} band.

It is worth discussing the temporal response shown in Fig. 1 in more detail. As in most d-band metals, the measured $\Delta R/R$ for YBCO can be described by

$$\Delta R/R = c\Delta T_h + d\Delta T_L \ , \tag{6}$$

where ΔT_h and ΔT_L are the change of the hole temperature and the lattice temperature, respectively, and c and d are constants. The sign of c depends on the probing frequency with respect to E_F and is positive at about 2 eV for oxygen-rich YBCO. Thus, the fast decay following the pulse-width-limited rise in Fig. 1 is attributed to phonon emission that reduces the hole temperature. The long-lasting plateau is caused by lattice heating, which decays on a slow time scale determined by the rate at which heat diffuses away from the excited region. Note that the normal meaning of the term "bolometric" applies only to $d\Delta T_L$, which is usually the dominant part of the $\Delta R/R$ signal.

Figure 2 illustrates the temporal dependence of the $\Delta R/R$ signal, measured on the YBCO-VI (oxygen-poor) sample at room temperature. The probe photon energy was varied from 1.46 to 1.91 eV, while the pump photon energy was kept constant at 2 eV with I_p about 30 μJ/cm^2. In contrast to Fig. 1, all the $\Delta R/R$ traces exhibit negative changes. This behavior results from a negative $\Delta\varepsilon_2$ when probing smeared hole states above E_F and indicates that E_F must be shifted down by more than 800 meV when the oxygen content decreases from ≥ 6.9 to < 6.45. Our result is consistent with the drastic decrease of the total free-carrier concentration in the Cu-O planes. It is also interesting to note some other differences between the traces in Figs. 2 and 1. First, the decay time resulting from the hole-phonon coupling appears to be much longer for the oxygen-poor YBCO, a result similar to the trend observed in metallic superconductors of different T_c's.[12] Second, there is a much smaller bolometric component for the oxygen-poor YBCO. Both

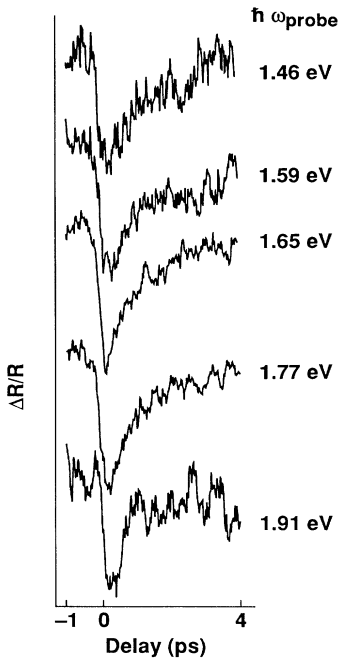

FIG. 2. Normalized changes in reflectivity $\Delta R/R$ ($\sim 10^{-3}$) measured on the YBCO-VI (oxygen-poor) sample at room temperature using the probe photon energies indicated in the figure.

results can be traced to different carrier-phonon and phonon-phonon coupling rates between the superconducting and nonsuperconducting phases, which may, in turn, affect the superconducting pairing.

Since it is difficult to accurately determine from simple electrical measurements the oxygen content in a completely deoxygenated YBCO, we have also performed measurements on the partially deoxygenated YBCO-V sample (oxygen content ~ 6.6, $T_c = 27$ K). As shown in Fig. 3, we observed a negative change at 2 eV. The initial decrease was again pulse width limited, however, in this

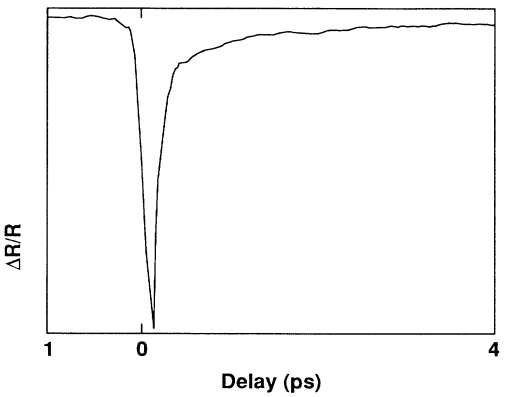

FIG. 3. Normalized $\Delta R/R$ measured on the YBCO-V (partially oxygen depleted) sample at room temperature using a 2-eV probe photon energy.

case it was followed by a very fast (~ 300 fs) recovery. This result implies that the downshift of E_F (using the hole picture) to a value less than 2 eV above the Cu d^9/d^{10} band seems not related to the material's superconductor-insulator transition, but rather follows the reduction of the oxygen content and T_c.

B. Low-temperature measurements

Figure 4 shows $\Delta R/R$ at 2 eV measured at $T=25$ K on two superconducting samples when a weak excitation ($I_p \approx 1$ μJ/cm^2) was used. The negative $\Delta R/R$, followed by a moderately fast (several picoseconds) recovery, presented in Fig. 4(a), was measured on the 280-nm-thick YBCO-IV film and is very similar to results obtained on optically thick (from 250-nm- to 500-nm-thick) samples by others.[16,18,20] By contrast, in Fig. 4(b) we observe a *positive* $\Delta R/R$ with similar temporal response, obtained on the optically thin YBCO-II sample. A positive $\Delta R/R$ was also found for a 140-nm-thick YBCO sample ($T_c \approx 91$ K).[33] Referring to Sec. II, we interpret the sign reversal between the optically thick and thin samples, which was not present at room temperature, as a thin-film effect. This suggests that $\Delta\varepsilon_1 \gg \Delta\varepsilon_2$. Therefore, for $T < T_c$, it is $\Delta\varepsilon_1$ and not $\Delta\varepsilon_2$ that dominates $\Delta R/R$ at 2 eV. The correct sign of $\Delta\varepsilon_1$ can be only obtained from an optically thick film, and is indeed negative under the above experimental conditions. This conclusion is further supported by the fact that the sign of $\Delta T/T$ remains the same (negative) with the onset of superconductivity.[15,34]

It was already noticed by Reitze *et al.*[20] that the amplitude of $\Delta R/R$ for YBCO films is more than 1 order of magnitude larger than that estimated from a simple two-fluid model including only the Drude contribution. In fact, at $I_p=1$ μJ/cm,^{2} the estimated density of photoexcited quasiparticles,[27] δN_Q, is $\sim 4\times 10^{19}$ cm^{-3}, which is only a small fraction of the YBCO total free-carrier concentration in the Cu-O plane. Thus, under such a weak excitation, the superconducting film may not be driven entirely to the normal state, and the contribution from $\Delta\varepsilon_{1C}$ cannot be neglected. Breaking only a fraction of Cooper pairs causes a reduction of the imaginary part of the conductivity (σ_2) and, correspondingly, a positive $\Delta\varepsilon_1$, since $\Delta\varepsilon_1 = -4\pi\Delta\sigma_2/\omega$. Obviously, this is not consistent with the experimentally observed negative $\Delta\varepsilon_1$. Therefore, the measured $\Delta R/R$ in YBCO cannot be fully explained by a simple two-fluid model, and a more advanced model that includes the material's band structure must be developed.

It is worth noting that this unexplained behavior of $\Delta R/R$, observed for weakly excited superconducting YBCO, may be associated with the existence of the intermediate state, which was described in Sec. II. In this case, the optically measured signal should consist of a mixed response from the superconducting state and the normal state, leading to a complicated behavior of $\Delta\varepsilon$. The existence of the transient intermediate state in optically excited YBCO needs, however, a solid experimental confirmation, before any conclusions about its influence on the optical reflectivity can be made.

Figures 5(a) and 5(b) present $\Delta R/R$ at 2 eV obtained at $T=12$ K for the YBCO-IV sample under relatively intense excitations ($I_p \approx 20$ μJ/cm^2 and 100 μJ/cm^2). In each case, the temporal response is very similar to that observed at room temperature (see Fig. 1). It is worth stressing that for pump intensities ranging from ~ 10 up to 100 μJ/cm,^{2} a similar bolometric temporal response was observed for all our optically thin and thick YBCO samples over the entire temperature range (from well below T_c up to room temperature). Interestingly, the magnitude of the signal at $T < T_c$ is significantly larger than that at room temperature.

At the intense excitation level of ~ 25 μJ/cm,^{2} the estimated δN_Q is $\sim 10^{21}$ cm^{-3} and is comparable to the total free-carrier concentration in YBCO. Thus, the sample must undergo a transition to the normal state. After the pump pulse optically induces this superconducting-to-normal transition and all the Cooper pairs are destroyed, only the $\Delta\varepsilon_D$ and $\Delta\varepsilon_H$ contributions to the measured $\Delta R/R$ remain. It is clear that the magnitude of

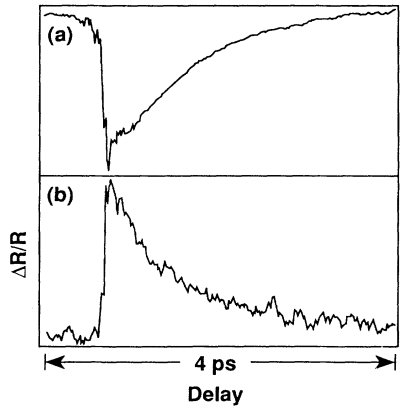

FIG. 4. Normalized $\Delta R/R$ ($\sim 10^{-4}$) measured on (a) YBCO-IV and (b) YBCO-II samples at $T=25$ K using a 2-eV probe photon energy. The pump intensity was ~ 1 μJ/cm^2.

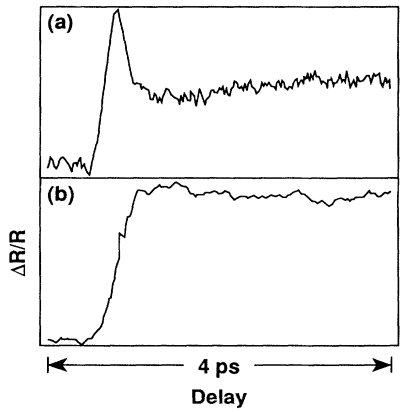

FIG. 5. Normalized $\Delta R/R$ ($\sim 10^{-3}$) measured on the YBCO-I sample at $T=12$ K using a 2-eV probe photon energy. The pump intensity was (a) ~ 20 μJ/cm^2 and (b) ~ 100 μJ/cm^2.

$\Delta\varepsilon_H$, which is directly related to the amount of smearing at 2 eV, is not expected to have an abrupt change over a relatively narrow range of change of ambient film temperature (if one assumes that the density of states at 2 eV does not change abruptly with temperature). Therefore, the abrupt increase in the magnitude of the signal observed at the onset of superconductivity implies an additional contribution from $\Delta\varepsilon_D$, or, in other words, from nonequilibrium quasiparticles generated by photoexcitation. On the other hand, the long-lasting plateau in the $\Delta R/R$ transient is a signature of a bolometric, heat-diffusion determined response. This latter result is consistent with the nanosecond response of photovoltage signals measured for similar laser intensities.[35]

In a separate series of measurements we have investigated the impact of the electric current on the femtosecond reflectivity of YBCO. Figure 6 shows results obtained under weak laser excitation ($I_p \approx 1$ μJ/cm^2) on a dc current biased 50-μm-wide superconducting microbridge patterned from the YBCO-IV sample. Figures 6(a)–6(c) present the $\Delta R/R$ traces measured below J_c, while $\Delta R/R$ in Fig. 6(d) was taken above J_c. We note that the curves for $J < J_c$ are very similar to that in Fig. 4(a) but their characteristic decay times decrease slightly with increasing the bias current. On the other hand, the decay time shows a very abrupt decrease when J exceeds J_c, as shown in Fig. 6(d). This behavior appears to be correlated with the onset of nonsuperconductivity in YBCO, a result similar to earlier femtosecond time-resolved transmission measurements[15] which showed that the decay time drops abruptly when temperature is increased to T_c. The effect observed by us is nonreversible in the sense that after the bias current is reduced, the newly measured signals do not resemble those shown in Figs. 6(a)–6(c). We have also observed that after prolonged laser excitation, J_c was reduced significantly, implying the formation of optically created "weak spot" in this stripe. It is interesting to note that the very fast (negative) response of $\Delta R/R$ shown in Fig. 6(d) for $J > J_c$ is very similar to that presented in Fig. 3 for the partially deoxygenated YBCO-V sample and is completely different from that in a photoexcited *nonequilibrium* normal state (Fig. 5). These observations suggest that this sample was a subject of the recently discovered current-induced electromigration of oxygen.[36]

To avoid the complexity involved with weak excitation in an optically thick YBCO stripe with nonuniform current flow, we also performed measurements on an optically thin stripe with an intense laser excitation. Figures 7(a) and 7(b) show $\Delta R/R$ measured at a temperature near T_c on another dc-biased 50-μm-wide superconducting microbridge patterned from the YBCO-II (thin) sample. By contrast to Fig. 6, the temporal response of the signal obtained at $J > J_c$ is essentially identical to that at $J = 0$. We have also verified that the J_c of the stripe remained the same even after prolonged intense laser excitation and many cycles of varying bias current, suggesting an absence of weak links in the patterned microbridge. Thus, the temporal response measured optically in the weak-link-free superconducting YBCO stripe under high intensity excitation is similar to that observed for an unpatterned film [Fig. 5(b)], and is not sensitive to the magnitude of the stripe's dc transport current. A

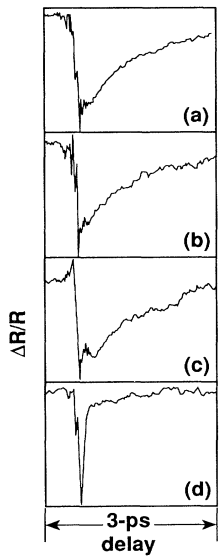

FIG. 6. Normalized $\Delta R/R$ measured with a 2-eV probe on the YBCO-IV microbridge biased by the following levels of electric current: (a) 0, (b) 1.5 mA, (c) 6 mA, and (d) 10 mA. The critical current of the microbridge was 8 mA. The temperature was about 72 K and the pump intensity was ~ 1 μJ/cm^2.

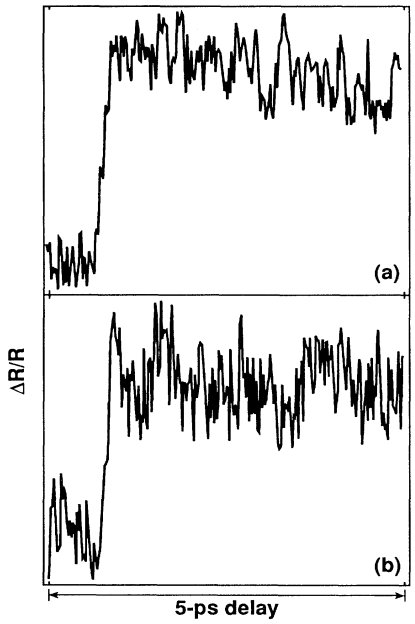

FIG. 7. Normalized $\Delta R/R$ measured with a 2-eV probe on the YBCO-II microbridge biased by the following levels of electric current: (a) 0 and (b) 15 mA. The critical current of the microbridge was 12 mA. The temperature was about 79 K and the pump intensity was ~ 100 μJ/cm^2.

more detailed discussion of experiments performed on the current-carrying YBCO stripes will be presented in a separate publication.

V. CONCLUSIONS

Our results obtained at room temperature provide quantitative information on the position of the Fermi level in YBCO with different oxygen contents. We have found that in superconducting YBCO ($T_c \approx 90$ K), E_F must be at least 2.25 eV above the Cu d^9/d^{10} band. Simultaneously, there is a large (>800 meV) energy difference between the positions of E_F in the oxygen-rich and oxygen-poor samples. Partially oxygen-depleted films with suppressed T_c (~27 K) have optical responses similar to the oxygen-poor films and their E_F is shifted down to a value less than 2 eV above the Cu d^9/d^{10} band. These conclusions are a direct consequence of the thermomodulation model, which has been generally accepted to explain experimental data for d-band metals, as well as for HTS materials at room temperature.

Measurements performed at temperatures below T_c indicate that in YBCO the optical response, associated with nonequilibrium properties of quasiparticles and Cooper pairs, is strongly dependent on the intensity of optical excitation. Under strong excitation, the temporal response of $\Delta R/R$ in optically thin and thick films is essentially the same and follows those measured at room temperature. In both cases, $\Delta R/R$ is dominated by $\Delta\varepsilon_2$ and the response is primarily bolometric. A completely different behavior is observed for weakly excited films. The $\Delta R/R$ signal exhibits a fast (a few picoseconds) recovery, but its sign depends on the film thickness and is negative for optically thick samples. The thickness-dependent sign change of $\Delta R/R$ below T_c indicates that $\Delta R/R$ is dominated by $\Delta\varepsilon_1$. However, its negative sign and relatively large magnitude (measured for the optically thick samples) cannot be explained by a simple two-fluid model. Therefore, more advanced models, based on a detailed material band structure and nonequilibrium quasiparticle Cooper-pair dynamics should be developed.

Measurements performed on dc current biased superconducting YBCO stripes showed that their $\Delta R/R$ response is sensitive to the presence of weak links in the stripe. More detailed experiments are necessary to fully characterize differences in optical properties of current-carrying epitaxial and granular YBCO stripes. In particular, it would be desirable to perform experiments, in which the change in optical reflectivity, transmissivity, and the transient voltage generated during the superconducting-resistive transition in the film are measured simultaneously. These measurements should allow us to directly observe the dynamics of the photogenerated superconducting-resistive transition, and reveal the possible role played in this process by the intermediate state.

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[1]See, e.g., A. Gilabert, Ann. Phys. (Paris) **15**, 255 (1990).

[2]G. A. Sai-Halasz, C. C. Chi, A. Denenstein, and D. N. Langenberg, Phys. Rev. Lett. **33**, 215 (1974).

[3]R. Sobolewski, D. P. Buttler, T. Y. Hsiang, C. V. Stancampiano, and G. A. Mourou, Phys. Rev. B **33**, 4604 (1986).

[4]V. F. Elesin and Yu. V. Kopaev, Usp. Fiz. Nauk **133**, 259 (1981) [Sov. Phys. Usp. **24**, 116 (1981)], and references therein.

[5]X.-H. Hu, T. Juhasz, and W. E. Bron, Appl. Phys. Lett. **59**, 3333 (1991).

[6]W. R. Donaldson, A. M. Kadin, P. H. Ballentine, and R. Sobolewski, Appl. Phys. Lett. **54**, 2470 (1989).

[7]H. S. Kwok, J. P. Zheng, Q. Y. Ying, and R. Rao, Appl. Phys. Lett. **54**, 2473 (1989).

[8]E. Zeldov, N. M. Amer, G. Koren, and A. Gupta, Phys. Rev. B **39**, 9712 (1989).

[9]A. Frenkel, M. A. Saifi, T. Venkatesan, P. England, X. D. Wu, and A. Inam, J. Appl. Phys. **67**, 3054 (1990).

[10]L. Shi, G. L. Huang, C. Lehane, J. P. Zheng, and H. S. Kwok, Appl. Phys. Lett. **61**, 489 (1992).

[11]S. D. Brorson, A. Kazeroonian, J. S. Moodera, D. W. Face, T. K. Cheng, E. P. Ippen, M. S. Dresselhaus, and G. Dresselhaus, Phys. Rev. Lett. **64**, 2172 (1990).

[12]S. D. Brorson, A. Kazeroonian, D. W. Face, F. K. Cheng, G. L. Doll, M. S. Dresselhaus, G. Dresselhaus, E. P. Ippen, T. Venkatesan, X. D. Wu, and A. Inam, Solid State Commun. **74**, 1305 (1990).

[13]D. W. Face, S. D. Brorson, A. Kazeroonian, J. S. Moodera, T. K. Cheng, G. L. Doll, M. S. Dresselhaus, G. Dresselhaus, E. P. Ippen, T. Venkatesan, X. D. Wu, and A. Inam, IEEE Trans. Magn. **MAG-27**, 1556 (1991).

[14]A. S. Kazeroonian, T. K. Cheng, S. D. Brorson, Q. Li, E. P. Ippen, X. D. Wu, T. Venkatesan, S. Etemad, M. S. Dresselhaus, and G. Dresselhaus, Solid State Commun. **78**, 95 (1991).

[15]J. M. Chwalek, C. Uher, J. F. Whitaker, G. A. Mourou, J. Agostinelli, and M. Lelental, Appl. Phys. Lett. **57**, 1696 (1990).

[16]S. G. Han, Z. V. Vardeny, K. S. Wong, O. G. Symko, and G. Koren, Phys. Rev. Lett. **65**, 2708 (1990); S. G. Han, Z. V. Vardeny, O. G. Symko, and G. Koren, *ibid.* **67**, 1053 (1991).

[17]S. G. Han, Z. V. Vardeny, O. G. Symko, and G. Koren, IEEE

Trans. Magn. **MAG-27**, 1548 (1991).
[18]G. L. Eesley, J. Heremans, M. S. Meyer, G. L. Doll, and S. H. Liou, Phys. Rev. Lett. **65**, 3445 (1990); G. L. Eesley, J. Heremans, M. S. Meyer, and G. L. Doll, *ibid.* **67**, 1054 (1991).
[19]S. V. Chekalin, V. M. Farztdinov, V. V. Golovlyov, V. S. Letokhov, Yu. E. Lozovik, Yu. A. Matveets, and A. G. Stepanov, Phys. Rev. Lett. **67**, 3860 (1991).
[20]D. H. Reitze, A. M. Weiner, A. Inam, and S. Etemad, Phys. Rev. B **46**, 14 309 (1992).
[21]G. L. Eesley, Phys. Rev. B **33**, 2144 (1986).
[22]R. W. Schoenlein, W. Z. Lin, J. G. Fujimoto, and G. L. Eesley, Phys. Rev. Lett. **58**, 1680 (1987).
[23]This relation has ignored the integrated effect over the entire penetration depth of the pump and probe beam. It is appropriate to use this relation when the penetration depth of the probe beam is close to that of the pump beam, a situation applied to our experimental conditions.
[24]D. E. Aspnes and M. K. Kelly, IEEE J. Quantum Electron. **QE-25**, 2378 (1989).
[25]M. E. Gershenzon, V. V. Golovlev, I. B. Kedich, V. S. Letokhov, Yu. E. Lozovik, Yu. A. Matveets, E. G. Sil'kis, A. G. Stepanov, V. D. Titov, M. I. Falei, V. M. Farztdinov, S. V. Chekalin, and A. P. Yartsev, Pis'ma Zh. Eksp. Teor. Fiz. **52**, 1189 (1990) [JETP Lett. **52**, 602 (1990)].
[26]The above picture may not be totally accurate for HTS materials; however, there is no doubt that in YBCO hole pairs form the superconducting condensate and the two-fluid model is generally accepted to apply for HTS materials in equilibrium.
[27]We assumed that the quasiparticle multiplication factor is 100, which is similar to that used in Ref. 16.
[28]P. H. Ballentine, A. M. Kadin, and D. S. Mallory, IEEE Trans. Magn. **MAG-27**, 997 (1991).
[29]S. Witanachchi, H. S. Kwok, X. W. Wang, and D. T. Shaw, Appl. Phys. Lett. **53**, 234 (1988).
[30]J. R. Gavaler, J. Talvacchio, T. T. Braggins, M. G. Forrester, and J. Greggi, J. Appl. Phys. **70**, 4383 (1991).
[31]E. Osquiguil, M. Maenhoudt, B. Wuyts, and Y. Bruynseraede, Appl. Phys. Lett. **60**, 1627 (1992).
[32]P. H. Ballentine, A. M. Kadin, M. A. Fisher, D. S. Mallory, and W. R. Donaldson, IEEE Trans. Magn. **MAG-25**, 950 (1989).
[33]D. H. Reitze (private communication).
[34]L. X. Zheng, T. Gong, R. Sobolewski, and P. M. Fauchet (unpublished).
[35]Photovoltage measurements [A. D. Semenov, G. N. Gol'tsman, I. G. Gogidze, A. V. Sergeev, E. M. Gershenzon, P. T. Lang, and K. F. Renk, Appl. Phys. Lett. **60**, 903 (1992)] confirm that YBCO is indeed in the normal state under an optical excitation at the level used in our measurements.
[36]B. H. Moeckly, D. K. Lathrop, and R. A. Buhrman, Phys. Rev. B **47**, 400 (1993).