Section 2
ADVANCED TECHNOLOGY DEVELOPMENTS

2.A Optical Nonlinearities in High-Temperature Superconductors

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 photon-induced 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 were found to be on a picosecond time scale,[3,5] contrary to a slow, heat-diffusion-related bolometric response observed in the films under strong laser excitation. The discovery of high-temperature superconductors (HTS) prompted a new series of transient photoexcitation measurements.[6–10] It was observed that, unlike low-T_c materials, the response of optically thick $YBa_2Cu_3O_{7-x}$ (YBCO) film was primarily bolometric with a small nonthermal (nonequilibrium) component. The relative magnitude of the nonthermal component was enhanced and could be as fast as tens of picoseconds for ultrathin films under moderate excitation.[10]

Very 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,[11–19] namely, femtosecond pump-probe measurements. In particular, femtosecond time-resolved measurements of the

differential reflectivity $\Delta R/R$ have been performed at 2 eV for various temperatures. Results interpreted using the Fermi smearing model, which was used previously to explain the dynamics of $\Delta R/R$ in metals,[20,21] indicate a different position of the Fermi level (E_F) in the oxygen-p band of oxygen-rich and oxygen-poor samples at room temperature. 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 order parameter.[18] The very recent results of Reitze *et al.*[19] imply, however, that a detailed energy band structure also has to be taken into account if any physical significance is to be assigned to these relaxation rates measured at optical frequency.

In this article we present a series of femtosecond reflectivity measurements on YBCO films. For the first time, we discuss in detail the dependence of $\Delta R/R$ on probing laser frequency, pumping laser intensity, and bias electric current. In particular, our study provides information on the nature of the transient optical response in optically thin and thick YBCO films under weak and strong laser excitations, as well as on the shift of the Fermi level between the oxygen-rich and oxygen-poor samples.

This article is organized as follows: First we provide 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 electric properties of HTS materials. We then describe the sample fabrication and experimental arrangement. Next, we discuss our experimental results and compare them with existing models, and finally, we present our conclusions and suggestions for future investigations.

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 by

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

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

Therefore, using Eqs. (1) to (3), $\Delta R/R$ can be simply expressed as

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

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

For most of the semiconducting materials, $a >> b$ and $\Delta R/R$ is thus dominated by $\Delta\varepsilon_1$. In contrast, for metals, $a << b$ and $\Delta R/R$ is essentially proportional to $\Delta\varepsilon_2$. Using unperturbed values of ε_1 and ε_2 obtained from ellipsometry measurements,[22] 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. 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 $\Delta\varepsilon_1 >> \Delta\varepsilon_2$ or $\Delta\varepsilon_2$ if $\Delta\varepsilon_1 << \Delta\varepsilon_2$.

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 than that shown in Eq. (4). 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. In particular, 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 Fermi smearing model is most commonly used in explaining the transient ΔR and ΔT signals in d-band metals and YBCO-related HTS materials, which are 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 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 cooled down below T_c, the free-carrier density of states in the superconductor is modified substantially. According to the BCS theory, in the superconducting state electrons form boson-like Cooper pairs, which occupy a single energy level, separated from E_F by the superconducting energy gap Δ, while unpaired, excited electrons (quasiparticles) occupy a continuum of states, starting Δ above E_F. The 2Δ gap is temperature dependent and, at a given temperature T below T_c but above 0 K, only a fraction $f_C(T)$ of the total number of free carriers forms Cooper pairs, while the rest $(1-f_C)$ remain as quasiparticles. Treating the superconducting condensate and normal electron gas as separate conducting "fluids,"[23] 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→p) transitions, respectively.

Under optical illumination the thermal equilibrium between the superconducting condensate and quasiparticles is disturbed (often very strongly), since photons with energies >>2Δ 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-phonon scattering, approximately 100 quasiparticles with energies widely distributed above 2Δ.[24] Thus, even a relatively weak optical excitation leads to highly nonequilibrium transient Cooper-pair/quasiparticle distributions.

As we mentioned previously, the situation is the most complicated for a relatively weak perturbation (typically 1 μJ/cm^2 to 5 μJ/cm^2 at 2 eV), since it can lead to the nonequilibrium intermediate state, resulting in the sample partitioning into separate superconducting and normal regions. Under such a condition, the superconductor is in a resistive state, which is, however, thermodynamically different from the normal (nonsuperconducting) state. According to the Elesin theory,[4] the intermediate state is stationary only for a particular excitation power that lies within the region where the energies of both superconducting and normal phases are equal. For perturbations different than critical, 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 critical and toward the normal phase in the opposite case. 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 (>>10 μJ/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 slow and can be explained using the bolometric (thermal-diffusion) model.

Experimental Procedures

A number of epitaxial YBCO films with thicknesses from 80 nm to 300 nm have been grown on $SrTiO_3$ single crystals by *in-situ* rf magnetron sputtering.[25] The films typically exhibited about 1.5-K-wide (10% to 90%) resistive superconducting transitions, with zero resistivity, T_c between 87 K (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,[26] and a 280-nm-thick sputtered film (YBCO-IV) provided by the Westinghouse Science and Technology Center.[27] The superconducting transition for the YBCO-IV sample was 0.5 K wide and its T_c was 89.5 K. For the sample YBCO-III, T_c was 86.5 K. All the tested films exhibited critical-current densities J_c greater than 10^6 A/cm^2 at liquid-nitrogen temperature. A partially oxygen-depleted YBCO sample (YBCO-V) was also fabricated. The film was deposited together with YBCO-I, but after deposition it was furnace annealed at 500°C for 20 min in pure Ar atmosphere. As a result, its T_c was lowered to 27 K, consistent with a decrease of the oxygen content of YBCO to about 6.45. For transport-current measurements, a film fabricated in the same run as the YBCO-IV sample was patterned into a 50-μm-wide and about

1-mm-long microbridge using a laser-ablation method.[28] The patterning lowered the bridge J_c to about 10^5 A/cm^2 at 77 K.

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 at 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 three orders of magnitude, as well as to use a different wavelength probe pulse selected from a white-light continuum. Time-resolved $\Delta R/R$ measurements have been 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 weak (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 in 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 K and 300 K.

Results and Discussion

1. Room-Temperature Measurements

Figure 52.12 illustrates the temporal dependence of the $\Delta R/R$ signal, measured on the YBCO-III sample at room temperature. The probe photon energy was

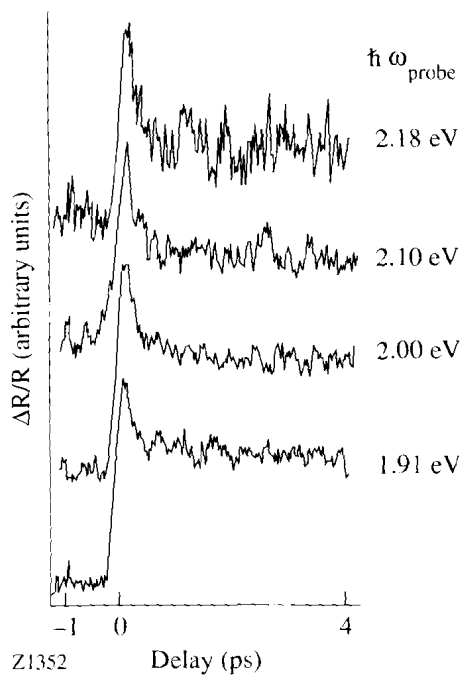

Fig. 52.12
Normalized changes in reflectivity $\Delta R/R$ (~10^{-4}) measured on the YBCO-III sample at room temperature using the probe photon energies indicated in the figure.

varied from 1.91 eV to 2.25 eV (the 2.25-eV data are not shown in Fig. 52.12), 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 "bolometric" response can be simply explained by the smearing of the electron (hole) 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 the change of $\Delta\varepsilon_2$ with probing of unoccupied hole states below E_F. A very similar behavior to that presented in Fig. 52.12 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 nm to 300 nm, implying that $\Delta\varepsilon_2 >> \Delta\varepsilon_1$. Therefore, $\Delta R/R$ is proportional to $\Delta\varepsilon_2$ and its behavior can indeed be explained by the Fermi smearing 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 0.3–100.0 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 eV to 2.25 eV, E_F must be at least 2.25 eV above the Cu-d^9/d^{10} bands. This result, which is a direct consequence of the Fermi smearing model, is inconsistent with earlier findings of Kazeroonian *et al.*,[14] who, from the dependence of $\Delta R/R$ at 2 eV on Pr-doping concentration, concluded that E_F was about 2 eV above the d-band of YBCO.

By contrast, $\Delta R/R$ measured at room temperature on the partially deoxygenated YBCO-V sample showed (see Fig. 52.13) a negative change at 2 eV. The initial decrease is again pulse-width limited, but it is then followed by a very fast (~300-fs) recovery. The sign change at 2 eV for oxygen-poor YBCO films was

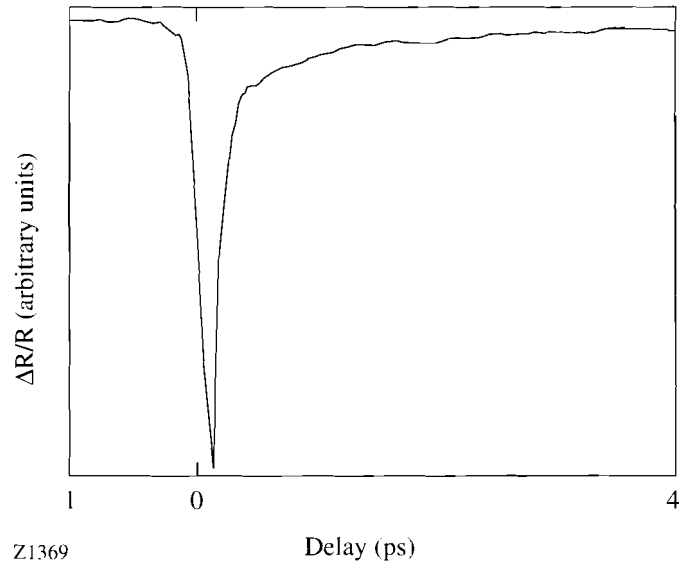

Fig. 52.13
Normalized $\Delta R/R$ measured on the YBCO-V (partially oxygen depleted) sample at room temperature using a 2-eV probe photon energy.

previously observed by Brorson *et al.*[12] and explained in the framework of the Fermi smearing model. The sign change was accounted for by a shift of the Fermi level, associated with a drastic change in the free-carrier concentration between the oxygen-rich and oxygen-poor films. Following again the Fermi smearing model, our results imply that in YBCO E_F must shift down (using the hole picture) by at least 250 meV when the sample oxygen deficiency decreases from above 6.9 to about 6.45.

2. Low-Temperature Measurements

Figure 52.14 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) is used. The negative $\Delta R/R$ followed by a moderately fast (several-picoseconds) recovery, presented in Fig. 52.14(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.[16–19] By contrast, in Fig. 52.14(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 in Ref. 19 for a 140-nm-thick YBCO sample ($T_c \approx 91$ K). We believe that the observed $\Delta R/R$ sign reversal between the optically thick and thin samples, which was not present at room temperature, is caused by a thin-film effect and suggests that $\Delta\varepsilon_1 >> \Delta\varepsilon_2$. Therefore, at $T < T_c$, 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 previously mentioned experimental conditions.

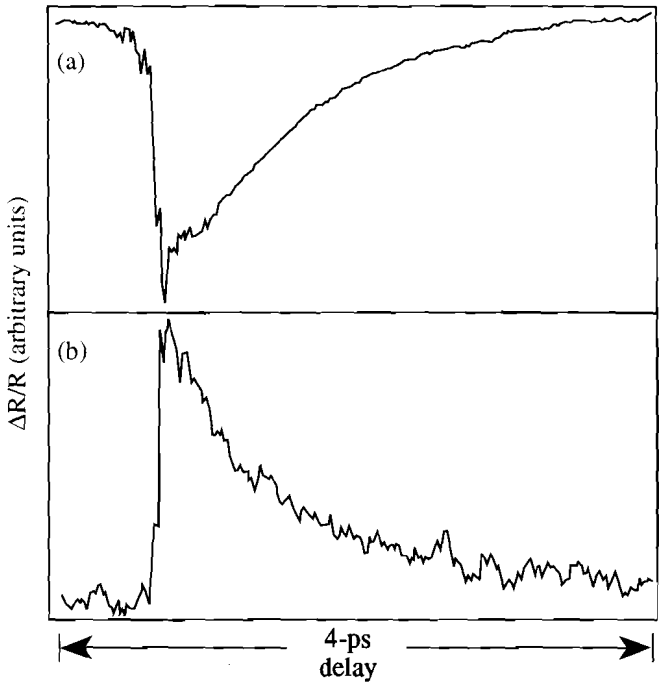

Fig. 52.14
Normalized $\Delta R/R$ ($\sim 10^{-4}$) measured on (a) YBCO-IV and (b) YBCO-II samples at $T = 25$ K using a 2-eV probe photon energy. The pump intensity was ~1 μJ/cm^2.

It was already noticed by Reitze *et al.*[19] that the amplitude of $\Delta R/R$ for YBCO films is about two orders of magnitude larger than that estimated from $\Delta\varepsilon_{1D}$ [change in the real part of the Drude component in Eq. (5)]. In fact, at I_p = 1 μJ/cm^2 the estimated density of photoexcited quasiparticles[24] δN_Q is ~4×10^{19} cm^{-3}, which is only a small fraction of the YBCO total density of free carriers. 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 the 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 $\Delta\varepsilon_1$, indicating that in YBCO the measured $\Delta R/R$ cannot be 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 completely different behavior of $\Delta R/R$ observed for weakly excited superconducting YBCO may be associated with the existence of the intermediate state, which was described previously. In this case, the optically measured signal should consist of a mixed response from both 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.

Figure 52.15 presents $\Delta R/R$ obtained at T = 12 K for the YBCO-I sample under strong 2-eV optical excitation ($I_p \approx 20$ μJ/cm^2). The response consists of a sharp, but time-resolved peak, followed by a long plateau, and is very similar to that observed at room temperature (see Fig. 52.12). We want to stress that for pump intensities ranging from ~10 μJ/cm^2 up to 100 μJ/cm^2 the behavior shown in Fig. 52.15 was characteristic of all our optically thin and thick YBCO samples in the entire temperature range from well below T_c up to room temperature.

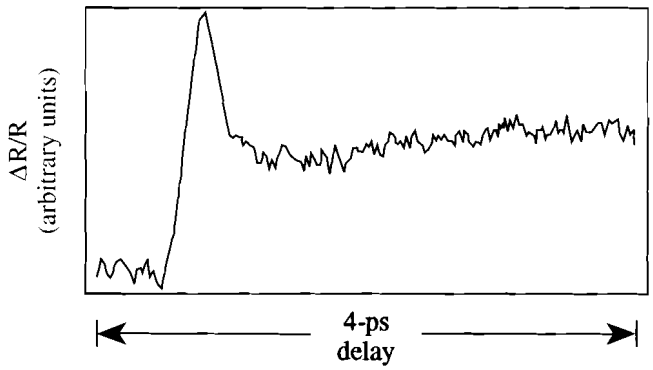

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Fig. 52.15
Normalized $\Delta R/R$ (~10^{-3}) measured on the YBCO-I sample at T = 12 K using a 2-eV probe photon energy. The pump intensity was ~20 μJ/cm^2.

We believe that under intense laser excitation ($\delta N_Q \sim 10^{21}$ cm^{-3}—of the order of the total free-carrier concentration in YBCO) YBCO undergoes a transition to the normal state.[29] Thus, all the Cooper pairs are destroyed, leaving only the $\Delta\varepsilon_D$ and $\Delta\varepsilon_H$ contributions to the measured $\Delta R/R$. The long-lasting plateau is the signature of a bolometric, heat-diffusion-determined response. This latter result is very similar to that presented previously. It is also consistent with the nanosecond photovoltage response measured for similar laser intensities.[29] On the other hand, the fact that the magnitude of the $\Delta R/R$ peak value (Fig. 52.15) is increased as compared to that measured at room temperature (Fig. 52.12) implies additional contribution from $\Delta\varepsilon_D$; in other words, from photoexcited nonequilibrium quasiparticles.

In a separate series of measurements we have investigated the impact of the electric current on the femtosecond reflectivity of YBCO. Figure 52.16 shows our preliminary data 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 52.16(a)–52.16(c) present the $\Delta R/R$ traces measured below J_c, while $\Delta R/R$ in Fig. 52.16(d) was taken above J_c. We note that the curves for $J < J_c$ are very similar to that on Fig. 52.14(a) and their

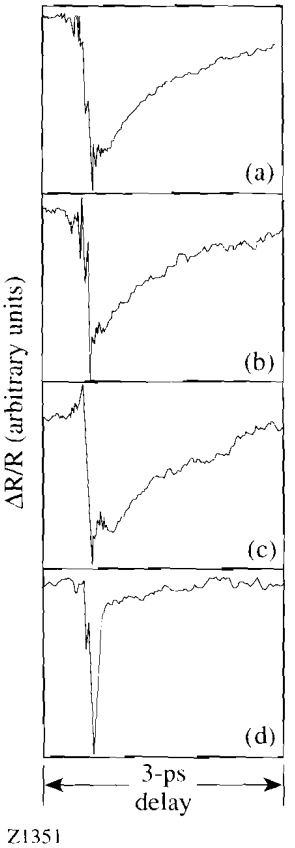

Fig. 52.16
Normalized $\Delta R/R$ measured with a 2-eV probe on the YBCO-IV microbridge biased by the following levels of electric current: (a) 0, (b) 1.5 mA, (c) 6 mA, and (d) 10 mA. The critical current of the microbridge was 8 mA. The temperature was about 72 K and the pump intensity was ~1 μJ/cm^2.

characteristic decay times change slightly with increasing the bias current. On the other hand, the decay time shows a very abrupt decrease when J exceeds J_c. This behavior appears to be correlated with the onset of nonsuperconductivity in YBCO, since earlier femtosecond time-resolved transmission measurements[18] also showed that the decay time drops very abruptly when temperature is increased to T_c. Most interestingly the very fast (negative) response of $\Delta R/R$ shown in Fig. 52.16(d) for $J > J_c$ is very similar to that presented in Fig. 52.13 for the oxygen-poor sample at room temperature, and is completely different from that in a photo-excited *nonequilibrium* normal state (Fig. 52.15). A more detailed discussion of experiments performed on the current-carrying YBCO strips will be presented in a separate publication.

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, E_F must be at least 2.25 eV above the Cu-d^9/d^{10} bands. Simultaneously, there is a large (>250-meV) energy difference between the positions of E_F in the oxygen-rich and oxygen-poor samples. These conclusions are a direct consequence of the Fermi smearing 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 dependencies of $\Delta R/R$ in optically thin and thick films are essentially the same and follow 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 pulse-width-limited rise, followed by a few-picosecond-long decay, but its sign depends on the film thickness and is negative for optically thick samples. The thickness-dependent sign change in the $\Delta R/R$ signal 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 the simple two-fluid model. Therefore, more advanced models, based on a detailed material band structure and nonequilibrium quasiparticle/Cooper-pair dynamics, should be developed.

$\Delta R/R$ measurements performed on dc-current-biased superconducting samples showed that YBCO optical responses from current-generated and photoexcited resistive states are different and cannot be easily understood using the existing models. More detailed experiments are necessary to fully characterize the optical properties of current-carrying superconducting films. In particular, it would be desirable to perform simultaneous optical and electrical transient measurements, in which the change in optical reflectivity 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-normal transition and reveal the possible role played in this process by the intermediate state.

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REFERENCES

1. See, e.g., A. Gilabert, Ann. Phys. Fr. **15**, 255 (1990).
2. G. A. Sai-Halasz *et al.*, Phys. Rev. Lett. **33**, 215 (1974).
3. R. Sobolewski, D. P. Butler, T. Y. Hsiang, C. V. Stancampiano, and G. A. Mourou, Phys. Rev. B **33**, 4604 (1986).
4. See, e.g., V. F. Elesin and Yu. V. Kopaev, Sov. Phys. Usp. **24**, 116 (1981).
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 *et al.*, Appl. Phys. Lett. **54**, 2473 (1989).
8. E. Zeldov *et al.*, Phys. Rev. B **39**, 9712 (1989).
9. A. Frenkel *et al.*, J. Appl. Phys. **67**, 3054 (1990).
10. L. Shi *et al.*, Appl. Phys. Lett. **61**, 489 (1992).
11. S. D. Brorson *et al.*, Phys. Rev. Lett. **64**, 2172 (1990).
12. S. D. Brorson *et al.*, Solid State Commun. **74**, 1305 (1990).
13. D. W. Face *et al.*, IEEE Trans. Magn. **27**, 1556 (1991).
14. A. S. Kazeroonian *et al.*, Solid State Commun. **78**, 95 (1991).
15. J. M. Chwalek *et al.*, Appl. Phys. Lett. **57**, 1696 (1990).
16. S. G. Han *et al.*, Phys. Rev. Lett. **65**, 2708 (1990); S. G. Han *et al.*, Phys. Rev. Lett. **67**, 1053 (1991).
17. S. G. Han *et al.*, IEEE Trans. Magn. **27**, 1548 (1991).
18. G. L. Eesley *et al.*, Phys. Rev. Lett. **65**, 3445 (1990); G. L. Eesley *et al.*, Phys. Rev. Lett. **67**, 1054 (1991).
19. D. H. Reitze *et al.*, to be published in Physical Review B.
20. G. L. Eesley, Phys. Rev. B **33**, 2144 (1986).
21. R. W. Schoenlein *et al.*, Phys. Rev. Lett. **58**, 1680 (1987).
22. D. E. Aspnes and M. K. Kelly, IEEE J. Quantum Electron. **25**, 2378 (1989).
23. This description 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 general enough to apply for HTS materials in equilibruim.