



Ultrafast dynamics in Pr-doped and oxygen-deficient YBCO films by femtosecond spectroscopy

K.H. Wu*, C.W. Luo, J.Y. Juang, T.M. Uen, Y.S. Gou

Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan, Republic of China

Abstract

We have systematically measured the transient reflectivity ($\Delta R/R$) in Pr-doped YBCO ($\text{Pr}_x\text{Y}_{1-x}\text{Ba}_2\text{Cu}_3\text{O}_7$) and partially oxygen deficient YBCO ($\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$) thin films at temperatures ranging from 300 to 12 K by using the femtosecond pump–probe technique. Measurements performed at low temperatures indicate that the magnitude, polarity, and relaxation time of the transient reflectivity is strongly dependent on the photon energy and pumping intensity of the optical excitation, the ambient temperature, and the doping level of the superconducting thin films. Moreover, the temperature dependence of the Fermi-level position was also observed. © 2000 Elsevier Science B.V. All rights reserved.

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The ultrafast optical response, dealing with the role of electrons, phonons, and their interaction and the dynamics of charge carriers in either normal or superconducting state, is particularly important to study the physical properties of high- T_c superconductors (HTSCs).

However, for femtosecond pump-probe experiments at temperatures near T_c , $\Delta R/R$ may contain more complicated nonequilibrium dynamics [1,2]. The abrupt change of the density of states near the Fermi level, the photo-generation and relaxation of quasiparticles, and the opening of the superconducting gap or pseudogap [2] may all come into play simultaneously. Therefore, the relationship between the optical response and the nature of the nonequilibrium superconducting dynamics has not been clearly elucidated in this regime. It is suggestive that, further femtosecond-spectroscopy measurement using the pump and probe beams with different photon energies might be needed to reveal more information about the optical response and the band structure of HTSCs.

The $\text{Pr}_x\text{Y}_{1-x}\text{Ba}_2\text{Cu}_3\text{O}_7$ films with $x = 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.8,$ and 1.0 were prepared on (100) LaAlO_3 substrates using pulsed laser deposition. In

comparison, the oxygen stoichiometry of a sole $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film was controlled to vary δ ($\delta = 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, 0.55, < 0.6$) in a precise and reproducible fashion [3]. The pump-probe measurements used a mode-locked Ti-sapphire laser operating at 775 nm (1.55 eV) which produced a 85 MHz train of 150 fs pulses.

The temperature-dependent $\Delta R/R$ curves of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{Pr}_x\text{Y}_{1-x}\text{Ba}_2\text{Cu}_3\text{O}_7$ samples have been systematically measured. It is found that the sign of ΔR always remains positive in the whole temperature range for samples with $\delta \leq 0.2$ or $x \leq 0.1$. On the other hand, for samples with $\delta = 0.3$ (Fig. 1(a)) or $x = 0.2$ (Fig. 1(b)) and 0.3 , the sign of ΔR is positive when the temperature is larger than T_c and change gradually to negative when the temperature is lowered than $T_c - \Delta T$, where ΔT is the increase in temperature due to laser pulse. For samples with $\delta > 0.5$ or $x > 0.5$, the sign of ΔR becomes negative in the whole temperature range. However, for the films with $\delta \leq 0.2$ or $x \leq 0.1$, the present results are quite different from those reported previously [1] where higher photon energy (~ 2.0 eV) was used and larger, negative ΔR was obtained when $T < T_c$. This implies that other mechanisms may have contributed to the negative component of measured ΔR if the probe energy is near the position of Fermi level relative to the Cu upper Hubbard band UHB (E_d). Indeed, as is evident

*Corresponding author.

E-mail address: khwu@oc.nctu.edu.tw (K.H. Wu)

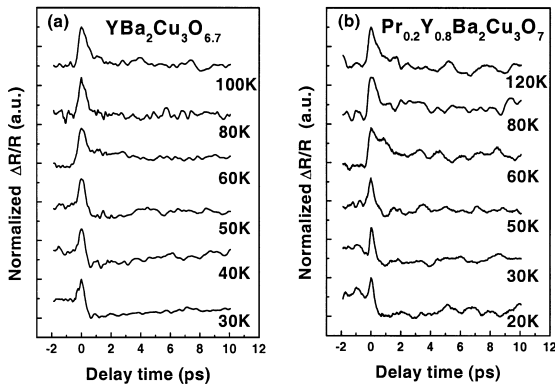


Fig. 1. The temperature dependence of the transient $\Delta R/R$ curves for (a) $\text{YBa}_2\text{Cu}_3\text{O}_{6.7}$ film ($T_c = 61$ K) and (b) $\text{Pr}_{0.2}\text{Y}_{0.8}\text{Ba}_2\text{Cu}_3\text{O}_7$ film ($T_c = 69$ K). The average pump power was 10 mW and probe beam was 2 mW.

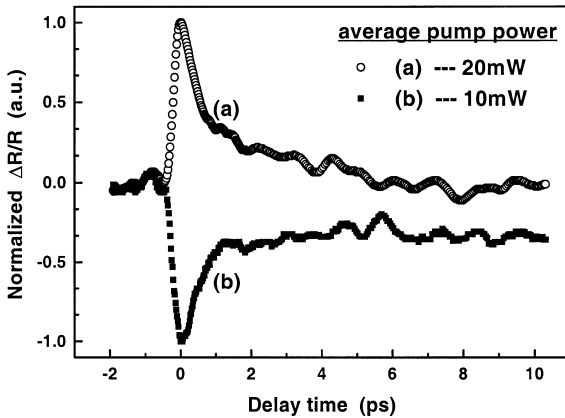


Fig. 2. The transient $\Delta R/R$ curves at room temperature for $\text{PrBa}_2\text{Cu}_3\text{O}_7$ film. The average pump power was (a) 20 mW and (b) 10 mW.

from Fig. 1, when the probing photon energy (1.55 eV in our case) was near E_d , the sign of ΔR was reversed at $T < T_c$ for samples with $\delta = 0.3$ or $x = 0.2$.

It is noted that in nonsuperconducting film, such as $\text{Pr}_x\text{Y}_{1-x}\text{Ba}_2\text{Cu}_3\text{O}_7$ film with $x > 0.6$ or $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ film with $\delta > 0.6$, the sign of ΔR were strongly related to the pumping intensity and the ambient temperature. As shown in Fig. 2, when a $\text{PrBa}_2\text{Cu}_3\text{O}_7$ film was illuminated by a pumping power of 20 mW, the sign of

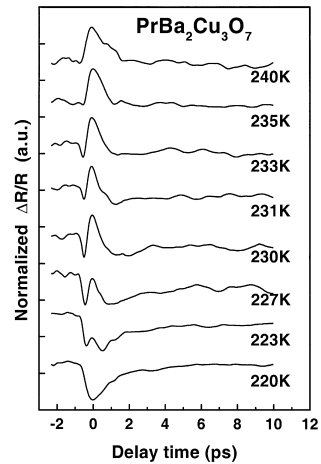


Fig. 3. The temperature dependence of the transient $\Delta R/R$ curves for a nonsuperconducting $\text{PrBa}_2\text{Cu}_3\text{O}_7$ film. The average pump power was 30 mW and probe power was 2 mW.

ΔR was positive while the pump power was reduced to 10 mW, it became negative. Moreover, Fig. 3 SHOWS THAT THE SIGN reversal of ΔR also occurred when the ambient temperature was cooled to below 233 K. The change of ΔR sign in both cases can be interpreted as the temperature dependence of the Fermi-level shift [4]. It is measurable especially when the Fermi temperature is low ($\sim 10^3$ K) for the insulating films. In principle, the temperature dependence of the Fermi-level position can be determined by the time-resolved $\Delta R/R$ measurement provided that the probe energy can be tuned over a wide range.

Acknowledgements

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