Ultrafast quasiparticle dynamics in heavy Ca-doped YBCO thin films
C.W. Luo*
Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan

**A R T I C L E   I N F O**
Article history:
Received 7 March 2009
Received in revised form 25 July 2010
Accepted 28 July 2010
Available online 5 August 2010

Keywords:
Ultrafast dynamics
Ca-doped YBCO
Femtosecond spectroscopy
High-Tc superconductivity

**A B S T R A C T**
The dynamics of photoinduced quasiparticle in the Y1–xCaxBa2Cu3O6+x thin films is revealed by using the femtosecond reflection spectroscopy. For the case of x = 0.1, two distinct components have been clearly observed in the transient reflectivity change (ΔR/R). The positive component of ΔR/R appears well below Tc. Moreover, the divergent peak of temperature-dependent relaxation time (τ) which is believed to intimately relate to the opening of superconducting gap, shifts to lower temperature as increasing the Ca-doping in YBa2Cu3O7−δ.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction
In recent years, although enormous progress in clarifying the mechanism of the high-Tc superconductivity including the relation between superconducting gap and pseudogap, many issues remained open [1]. Since the characteristics of superconducting gap and pseudogap are very close in energy scale and the symmetry of both gaps seems to be the same, it is extremely difficult to distinguish them. Although not yet employed as extensively as the spectroscopic studies, e.g. the corner-junction SQUID experiments [2], flux quantization measurements in tricrystal rings consisted of grain-boundary Josephson junctions [3], angle-resolved photoemission spectroscopy (ARPES) [4–6], the time-domain spectroscopy can potentially delineate the electronic states leading to superconductivity and, hence, could potentially provide information on the possible connections between the pseudogap above Tc and the superconducting gap below Tc [7–11]. However, from the evolution of hole-concentration in the phase diagram of high-Tc cuprate superconductors, most studies focus on the underdoped region and relatively less in overdoped region especially in the heavy overdoped cases. In this paper, we report that the dynamics of photoinduced quasiparticle (QP) in the Y1–xCaxBa2Cu3O7−δ (YCBCO) thin films by using the femtosecond reflection spectroscopy.

2. Experimental
All of the well characterized thin films with (0 0 1) orientations used in this study were prepared by pulsed laser deposition (PLD).

A KrF excimer laser operating at a repetition rate 3–8 Hz with an energy density of 2–4 J/cm² was used. The oxygen partial pressure during deposition was maintained at 0.25 Torr. The substrate temperature was kept at 780–790°C for 300-nm-thick (0 0 1) Y1–xCaxBa2Cu3O7−δ (x = 0.1 and 0.3) films deposited (1 0 0) STO substrates. After the deposition, the films were cooled in 600 Torr of oxygen to room temperature with the heater off. The crystallographic axes of these thin films have been examined by the X-ray diffraction (XRD) patterns. Additionally, Fig. 1 shows that the zero resistance transition temperatures (Tc) are 76.6 K and 66.0 K for Y0.8Ca0.2Ba2Cu3O7.92 and Y0.7Ca0.3Ba2Cu3O7.90 films, respectively.

The time-resolved femtosecond spectroscopy was carried out by the pump–probe scheme with 20 fs pulses train at repetition rate of 75 MHz and a central wavelength of 800 nm. The femtosecond pulses from Ti:sapphire laser went through two prisms to chirp the pulses. Then, we used two lenses to reduce the diameter of the laser beam enlarged by prisms. The beamsplitter reflected 70% of light in the pump channel, whereas the remnant was transmitted and served as the probe. Both pump and probe beams went through two acousto-optic modulators (AOM), respectively. But, only one in the pump beam was driven by the RF driver and modulated the pump beam at 87 kHz. After passing the delay stage, the half-wave (λ/2) plate, and the polarizer, the pump beam was focused on the surface of samples with 125 μm in diameter by the 200 mm lens. On the other hand, the probe beam only went through the λ/2 plate and polarizer after the AOM and was focused on the surface of samples with 84 μm in diameter by the 150 mm lens. The fluence of pump beam was kept at 4.35 J/cm² while the probe fluence was kept at 0.24 μJ/cm². The spatial overlap of pump and probe beams on the samples was monitored by the CCD camera. The reflection of the probe beam was received by the photodiode and the signal was taken by the lock-in amplifier. We utilized
the computer to control the delay stage (delay time, \( t \)) and measured the data \( \Delta R/R(t) \). We carried out the femtosecond time-resolved spectroscopy in thin films at various temperatures.

A low temperature system gives the control of the sample temperature. Samples were mounted on the cold finger of a Janis cryostat connected with transfer tube. The liquid helium was transferred from the Dewar flask to the isolation tube isolated by a pump. The cooling power could be controlled by the needle valve between the transfer tube and Dewar flask. Additionally, a 25 \( \Omega \) heater was used to control the temperature, which was monitored by the thermometer embedded in the finger below samples. The variable temperature range in this system is from 10 K to 300 K. Furthermore, there are four quartz windows with \( \Phi = 50 \) mm on the vacuum jacket allowing the optical pulses into and out of the low temperature chamber.

### 3. Results and discussion

Fig. 2 shows the typical \( \Delta R/R \) curves on the ab-plane of a (0 0 1) \( Y_{0.6}Ca_{0.3}Ba_2Cu_3O_{7,92} \) film with \( T_c = 77.6 \) K and a (0 0 1) \( Y_{0.6}Ca_{0.3}Ba_2Cu_3O_{7,90} \) film with \( T_c = 66.0 \) K, respectively. The amplitude of \( \Delta R/R \) in Fig. 2a increases gradually with decreasing temperature. Quantitatively, it is clearly presented in Fig. 3 with the anomaly increase near \( T_c \) (solid squares). This temperature dependence in (0 0 1) \( Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{6.92} \) is very similar with the results of (0 0 1) \( YBa_2Cu_3O_6 \), which is apparent lower than its intrinsic \( T_c \). Fig. 3 shows that the empty region between 0.6\( T_c \) and \( T_c \) is incidentally filled by the variation of the negative component. It implies that the negative component might represent some kind of order that suppresses the superconductivity above 0.6\( T_c \). Therefore, it is possible that this competing order is intrinsic in the high-\( T_c \) superconductors and could be clearly disclosed by either the ultrafast spectroscopy or other experiments.

(1) Recently, Chia et al. observed a competing order that appears to depress the superconducting gap in \( \text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_2\text{O}_{6+y} \) (Tl-2223) [12]. Therefore, it is possible that this competing order is intrinsic in the high-\( T_c \) superconductors and could be clearly disclosed by either the ultrafast spectroscopy or other experiments.

(2) Although the superconducting transition of both samples in the temperature-dependent resistance (Fig. 1) is sharp \( (\Delta T_c \)
1.4 K for Ca0.1, $\Delta T_c \sim 1.2$ K for Ca0.3), the inhomogeneity due to Ca-doping is not completely excluded. More heavily Ca-doped clusters with lower $T_c$ may constitute the major part of Y0.7Ca0.3Ba2Cu3O6.90 thin films. Thus, the slow relaxation in $\Delta R/R$ related to the clusters with low $T_c$ dominates at low temperatures. On the other hand, these low $T_c$ clusters will show the metallic characteristics in the $\Delta R/R$ with fast relaxation at the temperatures above their low $T_c$.

3) Very recently, Peets et al. [13] observed the disappearance of the upper Hubbard band (UHB) at heavy overdoped region by the oxygen K-edge X-ray absorption spectra. This suggests a breakdown of the Zhang-Rice singlet approximation at heavy overdoped region and a clear change in the nature of the electronic structure. Therefore, the divergent peak in the temperature-dependent relaxation time around 0.6$T_c$ for Y0.25Ca0.3Ba2Cu3O6.90 thin films may be caused by a change of electronic structure in heavy overdoped region.

4. Conclusion

We have measured the ultrafast dynamics of the photoinduced QPs on the CuO$_2$ planes of Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$. The coexistence of two ordered phase were clearly observed by the ultrafast spectroscopy which can temporally resolve the dynamics of different degrees of freedom. The validity of attributing the obtained experimental results with the suppression of superconductivity to the several scenarios, however, should be judged carefully by further experiments and more developed theories.

Acknowledgements

This work was supported by the National Science Council of Taiwan, ROC under Grant: NSC98-2112-M-009-008-MY3, NSC96-2923-M-009-001-MY3, and by the Grant MOE ATU Program at NCTU.

References