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Journal of Physics: Conference Series **150** (2009) 052144 doi:10.1088/1742-6596/150/5/052144

# Ultrafast dynamics in oxygen-deficient  $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$  superconductors

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Abstract. The transient reflectivity change  $(\Delta R/R)$  in oxygen-deficient Y<sub>0.7</sub>Ca<sub>0.3</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7−δ</sub> (YCaBCO) thin films have been systematically measured at various temperatures by femtosecond spectroscopy. The magnitude change of  $\Delta R/R$  from normal state to superconducting state were clearly observed in YCaBCO with various oxygen deficient, e.g. from overdoped region over optimaldoped region to underdoped region. Additionally, the carrier-phonon coupling strength was also characterized by the relaxation time of  $\Delta R/R$  at various temperatures and hole concentration.

## 1. Introduction

Owing to the anisotropic sandwiched-structure and the complex phase diagrams including insulating, antiferromagnetic, metallic, and superconducting phases for high- $T_c$  cuprates, a fundamental conundrum of the mechanism in high- $T_c$  superconductors has not been revealed yet. Therefore, figuring the phase diagram out perhaps could shed light on the high- $T_c$  mystery. Especially, a lot of essential properties found in overdoped and underdoped regions, respectively, should be illustrated by a whole picture. The most important thing is that the experiments must provide a systematic result from overdoped region to underdoped region. In this paper, we report the ultrafast responses of Y<sub>0.7</sub>Ca<sub>0.3</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7−δ</sub> (YCaBCO) which could be precisely controlled from overdoped over optimaldoped to underdoped regions.

## 2. Experiment

(001)-oriented YCaBCO thin films prepared by pulsed laser deposition were used in this study. The detailed growth conditions and structure-property characterizations of the films were reported elsewhere [1–3]. In addition, we used the encapsulated bulk annealing method [4] to manipulate the oxygen content of the YCaBCO films. Although the oxygen content of the films can only be estimated from the corresponding  $T_c$  obtained, we emphasize that this method is capable of controlling the oxygen content of the YCaBCO films precisely and reversibly. Furthermore, by using this method, all the measurements with various oxygen deficiencies can be performed on a single YCaBCO thin film. As a result, any changes in the superconducting properties should arise mainly from the effects of the oxygen content. The possible complications originated from individual film structures are minimized.



We measured the temperature-dependent transient reflectivity change  $(\Delta R/R)$  at a photon energy of 1.55 eV. The change of  $\Delta R/R$  is assumed to originate from the subsequent relaxation dynamics of the quasiparticles (QPs) excited by the pumping laser with the same photon energy. The details of the pump-probe scheme have been described previously [5]. Briefly, the optical pulses were produced by a mode-locked Ti:sapphire laser with a 75 MHz train of 20 fs pulses. The ratio between the average power of the pump and probe beams was set at 40:1. The typical energy density of the pump pulses was  $\sim$ 4.4  $\mu$ J/cm<sup>2</sup>, and the pulses were modulated at 97 KHz with an AO modulator. The small reflected signals were detected by using a lock-in amplifier.

#### 3. Results and discussion

Figure 1 shows the in-plane resistance as a function of temperature for YCaBCO films with various oxygen contents. At fully oxygenated state, the hole concentration of YCaBCO is raised by the Ca-doped and pushes it into the overdoped region in phase diagram with  $T_c = 66.3$ K. The hole concentration of YCaBCO can be easily controlled by the content of oxygen. By decreasing the content of oxygen, however, the  $T_c$  does not shrink monotonously but increases to near optimaldoped 82.1 K and then reduces to underdoped 65.5 K.



Figure 1. The resistance versus temperature curves of an (001)-oriented  $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$  thin film for various oxygen contents.

Figure 2 shows the temperature dependence of the  $\Delta R/R$  curves in a single (001)-oriented YCaBCO thin film for various oxygen contents. For the overdoped case with  $T_c = 66.3$  K as shown in Fig. 2(a), the sign of  $\Delta R/R$  change from negative to positive as decreasing temperature. The relaxation time of photon excited quasiparticles at superconducting state is around 6 ps which quite larger than the relaxation time of ∼0.6 ps at normal state. This behaviour of temperature-dependent relaxation time which is consistent with most other experimental data [6–8] is believed to be a generic manifestation of superconducting gap opening. Furthermore, the amplitude of  $\Delta R/R$  gradually raise as decreasing temperature from 33 K which is a half of  $T_c$  (= 66.3 K). It is worth noting that the growth of the negative amplitude in  $\Delta R/R$ with shorter relaxation time is assigned to the electron-electron scattering or electron-phonon scattering around  $T_c$ . Similar abnormal rise in the amplitude of  $\Delta R/R$  near  $T_c$  has been also observed in optimaldoped YCaBCO with  $T_c = 82.1$  K and underdoped YCaBCO with  $T_c$  = 65.5 K as shown in Fig.  $2(b)$  and  $2(c)$ , respectively. This abnormal characteristic appears in the vicinity of  $T_c$  may indicate that the formation of the other order parameter, such as s-wave gaps [9], competing order [10].

For partially oxygen-deficient of YCaBCO with higher  $T_c$  (= 82.1 K), no sign change was observed in temperature-dependent  $\Delta R/R$  from normal state to superconducting state. Obviously, the relaxation time of photoexcited QPs at superconducting state (e.g.  $T = 20 \text{ K}$ ) shrink to ~1.5 ps which is smaller than that in oxygen-rich case with  $T_c = 66.3$  K. This implies





Figure 2. The temperature dependence of  $\Delta R/R$  in a single (001)-oriented  $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$  thin film for various oxygen contents. (a) overdoped case with  $T_c$  $= 66.3$  K (b) optimaldoped case with  $T_c =$ 82.1 K (c) underdoped case with  $T_c = 65.5$ K.

that the strength of carrier-phonon coupling become strong at near optimally doped region [4]. For further oxygen-deficient YCaBCO with  $T_c = 65.5$  K, however, the recombination of photon excited quasiparticles takes longer time of  $\sim$ 7 ps as shown in Fig. 2(c).

#### 4. Summary

In summary, we have measured the ultrafast dynamics of  $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$  films from overdoped to underdoped region by controlling the oxygen-deficient in a single thin film. The relaxation time of photon excited quasiparticles near optimally doping is much shorter than that in underdoped and overdoped region. This may be due to the stronger carrier-phonon coupling strength at the optimally doping with the highest  $T_c$ . Moreover, the abnormal rises for the amplitude of  $\Delta R/R$  in the vicinity of  $T_c$  have been clearly observed in whole doping region. The validity of attributing the obtained experimental results to the other order parameter, however, should be judged carefully by further experiments and more developed theories.

#### Acknowledgements

This work was supported by the National Science Council of Taiwan, R.O.C. under grant: NSC95-2112-M-009-011-MY3, and by the Grant MOE ATU Program at NCTU.

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- 25th International Conference on Low Temperature Physics (LT25) IOP Publishing Journal of Physics: Conference Series **150** (2009) 052144 doi:10.1088/1742-6596/150/5/052144
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