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ORIGINAL PAPER

Doping-Dependent Phase Diagram of Ca-Doped YBCO Observed by Femtosecond Spectroscopy

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Abstract The quasiparticle relaxation dynamics in a single (001) Ca-doped YBCO ($Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$) superconducting thin film was probed by using the femtosecond time-resolved spectroscopy. The (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ thin film was prepared on a (100)STO substrate by pulsed laser deposition. Through controlling the oxygen pressure and the annealing temperature within quartz tube, the doping level ($0.08 < p < 0.22$) in the phase diagram could be widely tuned from the overdoped region to the underdoped region with varying the hole concentration (p) in a single sample. In the overdoped region, two different components (positive/negative) were obviously identified in the transient reflectivity curves ($\Delta R/R$). The negative component in $\Delta R/R$ emerges at $T < T_c$ and quickly defeated by the development of the positive component. The dramatic change in the positive component of $\Delta R/R$ arises well below T_c. However, this anomaly change does not appear in the underdoped region. Only the positive $\Delta R/R$ was clearly observed at whole temperatures. These results indicate that the scenario of high-T_c superconductivity in the overdoped region would be different from the underdoped region.

Keywords Cuprate superconductors · Ultrafast spectroscopy · Nernst effect

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1 Introduction

In order to investigate the interesting phenomena of high-T_c superconductor cuprates, there are many themes in this domain, such as the evolution of superconducting gap (SC), pseudogap (PG), and so on [1, 2]. In recent years, femtosecond time-resolved spectroscopy has been recognized as a powerful bulk technique to study temperature-dependent changes of the low-lying electronic structure of superconductors [3, 4] and other strongly correlated electron systems [5, 6]. In superconductors and other strongly correlated electron systems, the opening of a gap in the density of states introduces an additional time scale for the quasiparticle dynamics. The significant divergences in the temperature-dependent relaxation time associated with the opening of superconducting gap were observed on the CuO_2 planes. Recently, some research groups drew the colorful phase diagram of cuprate superconductors [7, 8] by utilizing some mathematical methods to help them evidently demonstrate and deduce their results what they saw. Therefore, up to now, YBCO group is still one of the choices to debate the hot issues in condensed matter.

2 Experiments

In this paper, we have prepared a single high-quality (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ ($YCa_{0.3}BCO$) thin film grown on (100) $SrTiO_3$ substrate by pulsed laser deposition (PLD). The detail growth conditions and structure-property characterizations of the films are similar with that reported elsewhere [9–11]. The doping level of $YCa_{0.3}BCO$ can be extend to more overdoped (OD) range, which is defined that the transition temperature T_c decreases with increasing the hole concentration (p) when $p > 0.16$, by substituting Ca^{2+}

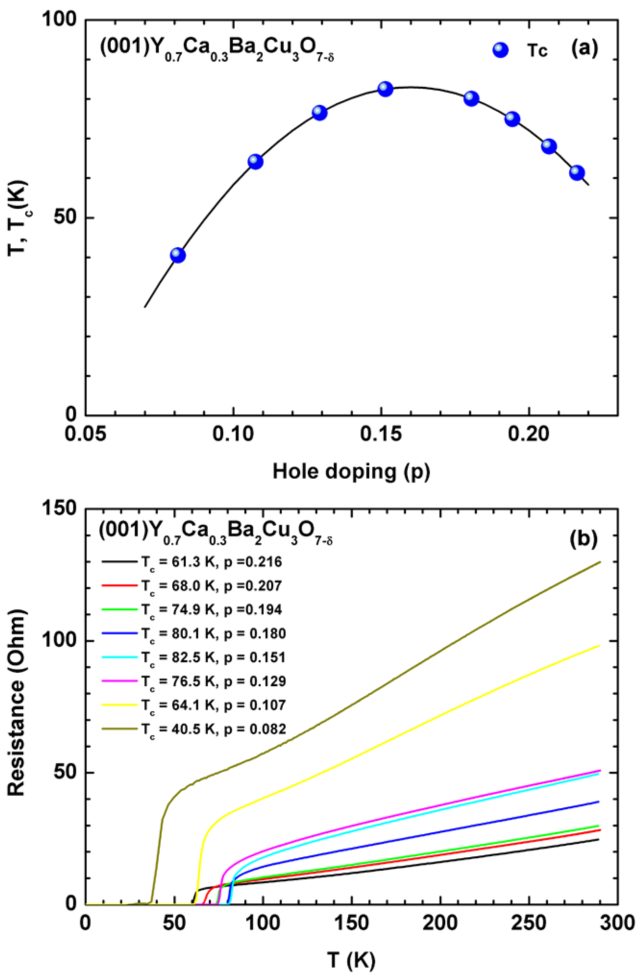

Fig. 1 (**a**) The schematic of phase diagram for $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$. (**b**) Electrical resistivity as a function of temperature for the $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ films with $p = 0.08 \sim 0.21$

ion for Y^{3+} ion site. Besides, various hole concentration of a single thin film was controlled by temperature and oxygen pressure via the oxygen-content control system and each electrical resistance was determined by four-point method. Furthermore, by using this method, all the measurements with various oxygen deficiencies can be performed on a single $YCa_{0.3}BCO$ thin film.

The schematic of phase diagram and electrical resistances as a function of temperature for a single $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ film with $p = 0.08 \sim 0.21$ were shown in Figs. 1(a) and (b). All of the value of doping level were defined from generalized $T_c(p)$ relation formula: $T_c/T_{c,\max} = 1 - 82.6(p - 0.16)^2$, where $T_{c,\max} = 83$ K.

Recently, the femtosecond time-resolved spectroscopy has been recognized as a powerful technique to study the behavior of the quasiparticle (QP) in condensed matter. From analyzing the amplitude and relaxation time of the measured transient $\Delta R/R$ curves in each sample, the important information about the number of the accumulated quasiparticles and the amplitude of the gap will be refined. We measured the orientation-dependent transient reflectivity curves ($\Delta R/R$) 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 QPs excited by the pumping laser

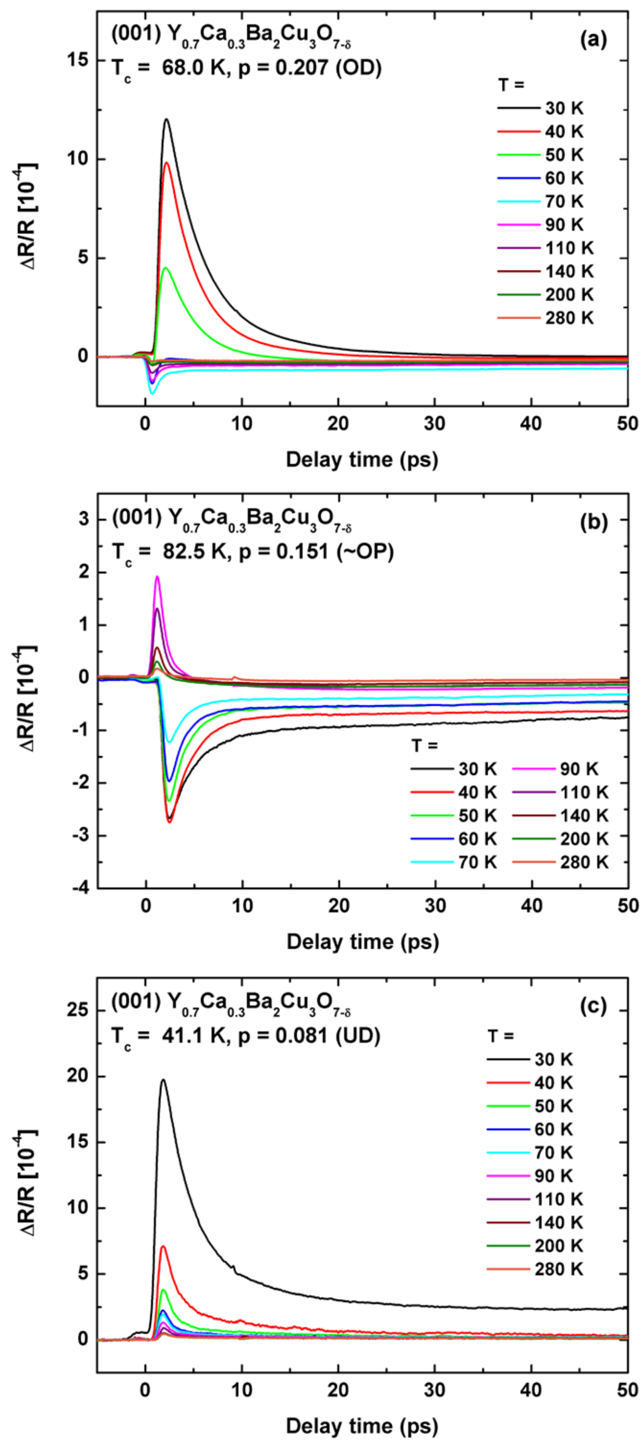

Fig. 2 The temperature dependence of the transient $\Delta R/R$ for: (**a**) measured in a (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ film with $T_c = 68.0$ K in OD region, (**b**) measured in a (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ film with $T_c = 82.5$ K in near OP region, (**c**) measured in a (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ film with $T_c = 41.1$ K in UD region

with the same photon energy. The details of the polarized pump-probe scheme have been described previously [12]. Briefly, the optical pulses were produced by a mode-locked Ti:sapphire laser with a 75 MHz train of 35 fs pulses. The ratio between the average power of the pump and probe beams was set at 20:1. The typical energy density of the pump pulses was ~5.6 $\mu J/cm^2$, and the pulses were modulated at 97 KHz with an acousto-optic modulator (AOM). The weak reflected signals were detected by using a lock-in amplifier.

3 Results and Discussion

The typical temperature dependence of $\Delta R/R$ curves on the ab-plane of a (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ film were shown in Fig. 2. From overdoped to underdoped region, the behavior of $\Delta R/R$ with doping level present very different. Each curve was separated into two kinds of amplitude (positive and negative) part. In overdoped region, the positive part reduces immediately down to zero before the occurrence of transition temperature T_c while the negative one grows up. It implies that Upper Hubbard Band (UHB) [13] does not exist in the OD region. The suggestion is consistent with the result of the previous literature [14]. However, this competing scenario does not happen in the underdoped (UD) region. During the doping level $p \sim 0.15$, the maximum positive $\Delta R/R$ above T_c were clearly observed. Comparing with the results of others literatures [15, 16], we reasonable doubt that anomalously phenomenon is relevant to vortex-Nernst effect.

4 Summary

In summary, the transient reflectivity $\Delta R/R$ a single (001) Ca-doped YBCO ($Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$) superconducting thin film from OD to UD region has been shown by the time-resolved femtosecond spectroscopy. In the overdoped region, the two (positive and negative components) kinds of the transient $\Delta R/R$ were clearly observed in the (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ thin film by the ultrafast spectroscopy. These two components exist certain relation, such as the absence of the UHB or other effect. The doping-dependent positive $\Delta R/R$ above T_c relative to Nernst effect is also clearly observed in ultrafast responses.

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