



Ultrafast quasiparticle dynamics in heavy Ca-doped YBCO thin films

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ABSTRACT

The dynamics of photoinduced quasiparticle in the $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films is revealed by using the femtosecond reflection spectroscopy. For the case of $x = 0.3$, two distinct components have been clearly observed in the transient reflectivity change ($\Delta R/R$). The positive component of $\Delta R/R$ appears well below T_c . 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 $YBa_2Cu_3O_{7-\delta}$.

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1. Introduction

In recent years, although enormous progress in clarifying the mechanism of the high- T_c 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 T_c and the superconducting gap below T_c [7–11]. However, from the evolution of hole-concentration in the phase diagram of high- T_c 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 $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ (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) $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ ($x = 0.1$ 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 (T_c) are 76.6 K and 66.0 K for $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{6.92}$ and $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{6.90}$ 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 pre-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 ($\lambda/2$) plate, and the polarizer, the pump beam was focused on the surface of samples with 125 μ m in diameter by the 200 mm lens. On the other hand, the probe beam only went through the $\lambda/2$ plate and polarizer after the AOM and was focused on the surface of samples with 84 μ m in diameter by the 150 mm lens. The fluence of pump beam was kept at 4.35 μ J/cm² while the probe fluence was kept at 0.24 μ J/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

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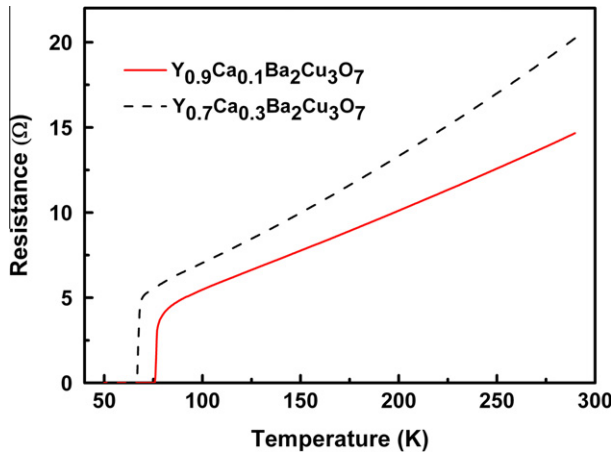


Fig. 1. Typical resistivity versus temperature curves of (001) $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films.

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 the sample chamber, and the helium gas after absorbing the heat of samples was pumped out of the isolation tube by a pump. The cooling power could be controlled by the needle valve between the transfer tube and Dewar flask. Additionally, a 25 Ω 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 (001) $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{6.92}$ film with $T_c = 77.6$ K and a (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{6.90}$ film with $T_c = 66.0$ K, respectively. The amplitude of $\Delta R/R$ in Fig. 2a increases gradually with decreasing temperature. Quan-

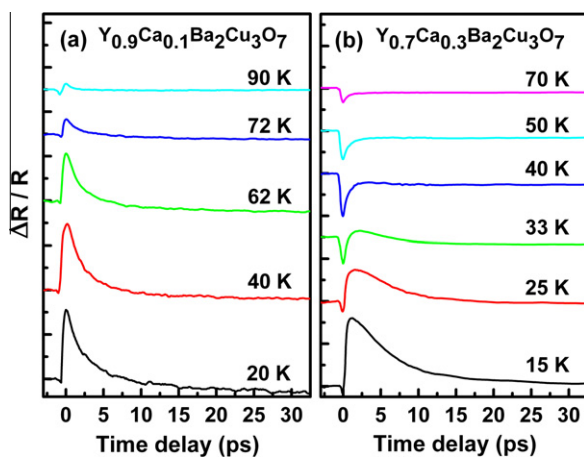


Fig. 2. The temperature dependence of $\Delta R/R$: (a) measured in a (001) $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{6.92}$ film with $T_c = 77.6$ K; (b) measured in a (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{6.90}$ film with $T_c = 66.0$ K.

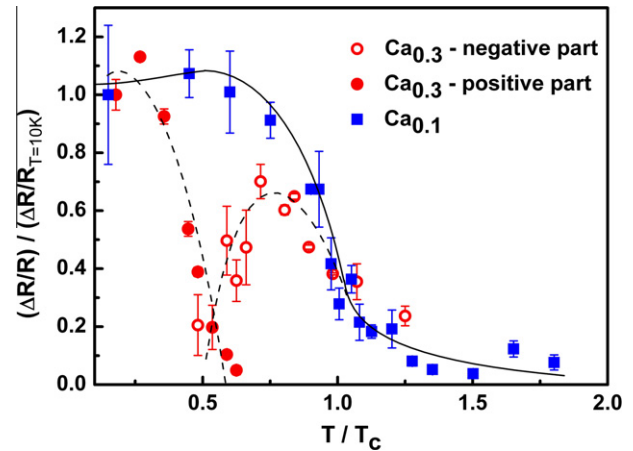


Fig. 3. The normalized $\Delta R/R$ as a function of the reduced temperature of a (001) $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{6.92}$ film with $T_c = 77.6$ K and a (001) $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{6.90}$ film with $T_c = 66.0$ K. The solid and dashed lines are guides to the eye emphasizing temperature dependences.

titatively, it is clearly presented in Fig. 3 with the anomaly increase near T_c (solid squares). This temperature dependence in (001) $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{6.92}$ is very similar with the results of (001) $YBa_2Cu_3O_{6.9}$ in Ref. [11], i.e. A- and B-type temperature dependence are involved below and above T_c , respectively. Actually, this phenomenon has been observed by Demsar et al. [8] in slightly overdoped samples of $Y_{1-x}Ca_xBa_2Cu_3O_7$. They claimed that the A-type temperature dependence is associated with the opening of the superconducting gap and the B-type temperature dependence is governed by the pseudogap.

However, the relaxation behavior of photoinduced quasiparticle (QP) in further overdoped sample, i.e. $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{6.90}$ films, is dramatically different from that in slightly overdoped samples of $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{6.92}$. An additional component with negative sign appears at temperatures just below T_c and quickly overwhelmed by the growth of the positive component shown in Fig. 2b. The anomaly change in the positive component of $\Delta R/R$ arises around $0.6T_c$ which is apparent lower than its intrinsic T_c . Fig. 3 shows that the empty region between $0.6T_c$ 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 $0.6T_c$. Moreover, this suggestion could be further elaborated in Fig. 4 by the temperature-dependent relaxation time of $\Delta R/R$, extracted by fitting a single exponential function as shown in the inset of Fig. 4. In general, the relaxation time of $\Delta R/R$ as a function of temperature will diverge near T_c such as that shown for the $Y_{0.9}Ca_{0.1}Ba_2Cu_3O_{6.92}$ with solid squares in Fig. 4, which is a signified characteristic related to the opening of superconducting gap. For the case of $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{6.90}$, it is suggesting that the superconducting gap *does not* open until around $0.6T_c$ according to the divergent peak of the temperature-dependent relaxation time of $\Delta R/R$. Although, it is not clear at present whether this behavior is unique to the heavy overdoped YBCO system due to the complications arising from significant Ca-doping or other effects, several possible scenarios are further discussed here to interpret these experimental results.

(1) Recently, Chia et al. observed a competing order that appears to depress the superconducting gap in $Tl_2Ba_2Ca_2Cu_3O_y$ (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 (ΔT_c

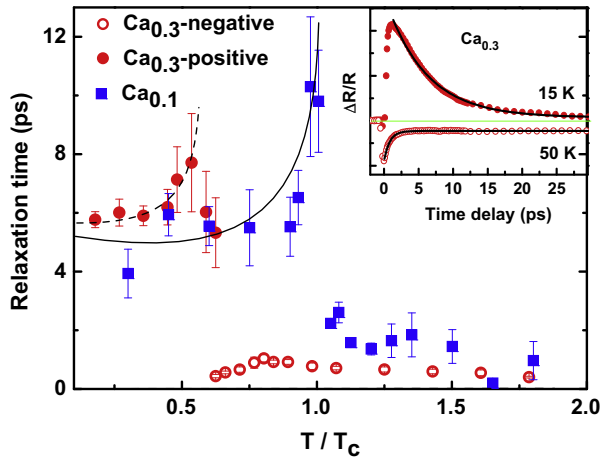


Fig. 4. The reduced temperature-dependent relaxation time of $\Delta R/R$ at various Ca contents obtained by fitting with a single exponential function as shown in the inset. The solid and dashed lines are guides to the eye emphasizing temperature dependences.

~ 1.4 K for $\text{Ca}_{0.1}$, $\Delta T_c \sim 1.2$ K for $\text{Ca}_{0.3}$), 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 $\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{6.90}$ 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 $0.6T_c$ for $\text{Y}_{0.7}\text{Ca}_{0.3}\text{Ba}_2\text{Cu}_3\text{O}_{6.90}$ 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 $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$. 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.

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