Anisotropic Ultrafast Dynamics of Quasiparticles on CuO$_2$ Planes of Y$_{0.7}$Ca$_{0.3}$Ba$_2$Cu$_3$O$_{7-\delta}$

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Abstract The orientation-resolved femtosecond spectroscopy, combined with the well-textured (110)- and (001)-Y$_{0.7}$Ca$_{0.3}$Ba$_2$Cu$_3$O$_{7-\delta}$ thin films, serves as an effective probe to quasiparticle relaxation dynamics on the $ab$ planes and along the diagonal orientations. The significant divergences in the temperature-dependent relaxation time ($\tau$) associated with the opening of superconducting gap were observed along the nodal directions and on the CuO$_2$ planes which are dominated by the $a$ axis and $b$ axis in the overdoped region. Moreover, the divergence in the temperature-dependent $\tau$ along the nodal direction disappears around optimal doped region. This implies that the superconducting gap evolves from the dominant $s$-wave symmetry in overdoped region into the dominant $d$-wave symmetry in optimal doped region.

Keywords Cuprate superconductors · Anisotropic ultrafast dynamics · Superconducting gap symmetry

1 Introduction

The symmetry of order parameters is a key issue to resolve the conundrum for high temperature superconductivity (HTSC). Since the pairing symmetry in the cuprates may vary with doping [1, 2], a detailed study of the symmetry evolution with doping could yield crucial insight into the pairing in cuprate superconductors. Recently, Ngai et al. [3] reported that the main-gap, subgap, and satellite features in the scanning tunneling spectroscopy (STS) of Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ are due to the $d_{x^2-y^2} + s$ pairing symmetry and multiple bands. Besides, many experimental methods were also applied to reveal the pairing symmetry such as the Josephson tunneling current in SIS tunneling junctions [4], the Andreev reflection between normal metal and Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ thin films [5]. However, the intrinsic pairing symmetry may be disturbed by the local probe on the inhomogeneous samples or the surface sensitivity.

The bulk-sensitive and orientation-resolved femtosecond spectroscopy could help one to solve this issue by combining the specially oriented thin films, e.g. (110) or (100). In superconductors and other strongly correlated electron systems, the opening of a gap in the density of states introduces an additional timescale for the quasiparticle (QP) dynamics. The recovering time of QP is related to the gap magnitude, meaning that interactions which perturb the gap manifest as an easily measured change in the temporal response by detecting changes in the transient reflectivity ($\Delta R/R$) of a probe beam. 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 [6, 7] and other strongly correlated electron systems [8, 9]. It provides a new avenue, i.e. the time domain, for providing information about the pairing symmetry through the QP excitations of the materials.

In this paper, we study the anisotropic QP relaxation time in (001)- and (110)-oriented Y$_{0.7}$Ca$_{0.3}$Ba$_2$Cu$_3$O$_{7-\delta}$ thin films by the orientation-resolved femtosecond spectroscopy. By varying the oxygen content in a single thin film,
the doping-dependent evolution of the superconducting gap symmetry could be clearly tracked out.

2 Experiments

In this study, (001)- and (110)-oriented Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7−δ} thin films prepared by pulsed laser deposition were used. The detailed growth conditions and structure-property characterizations of the films are similar to those reported elsewhere [10–12]. In addition, we used the encapsulated bulk annealing method [13] to manipulate the oxygen content of the Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7−δ} 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 Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7−δ} films precisely and reversibly. Furthermore, by using this method, all the measurements with various oxygen deficiencies can be performed on a single Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7−δ} 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 orientation-dependent transient reflectivity curves ($ΔR/R$) at photon energy of 1.55 eV. The change of $ΔR/R$ is assumed to originate from the subsequent relaxation dynamics of the QPs excited by the pumping laser with the same photon energy. The details of the polarized pump–probe scheme have been described previously [7]. 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 $∼4.4 \mu J/cm^2$, and the pulses were modulated at 87 KHz with an acousto-optic modulator (AOM). The weak reflected signals were detected by using a lock-in amplifier. As discussed in detail previously [7], the measurements of $ΔR/R$ along the $ab$ diagonal (oriented at 45° relative to the $a$ axis or $b$ axis on the $ab$ plane) and on the CuO$_2$ planes ($ab$ plane) could be carried out with the availability of (110)- and (001)-Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7−δ} thin films.

3 Results and Discussion

Figure 1 shows a typical $ΔR/R$ of the (001)- and (110)-Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7−δ} thin films at various oxygen deficiencies. For all of them, the significant distinction between the normal state ($T > T_c$) and superconducting state ($T < T_c$) could be easily interpreted by the following picture which is generally accepted. Namely, the pump pulse excites the electron–hole pairs that relax to the states in the vicinity of the Fermi surface ($E_F$) by scattering mechanisms (e.g., electron–electron or electron–phonon scattering). This process occurs in the normal state ($T > T_c$) within a subpicosecond timescale [14, 15]. The presence of a gap near $E_F$ leads to the carrier accumulation in the QP states above the gap at $T < T_c$. This, in turn, gives rise to a transient change in reflectivity ($ΔR/R$) which was detected by a second laser (probe) pulse as a function of delay time ($t$) between a pump pulse and a probe pulse. The amplitude and characteristic relaxation time ($τ$) of the measured $ΔR/R$ thus give important information about the number of the accumulated QPs and the magnitude of the gap.