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Anisotropic photoexcited carrier dynamics in (100)-, (001)-, and (110)-oriented YBCO films by polarized ultrafast optical spectroscopy

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We report the time-resolved ultrafast optical spectroscopy results obtained from a series of well characterized (001)-, (100)- and (110)-oriented $YBa_2Cu_3O_{7-\delta}$ (YBCO) superconducting films prepared by pulsed laser deposition. The well-controlled film orientation has allowed us to probe the relaxation dynamics along various crystal orientations using respectively polarized pump and probe laser beams. The significant anisotropies in both the magnitude of the characteristic relaxation time and the temperature dependence of the photo-induced transient reflectance change indicate the nature of relaxation channel might be intrinsically different along different orientations. The implications of the results on the fundamental characteristics concerning superconducting gaps are discussed.

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1. INTRODUCTION

The ability to generate and time resolve nonequilibrium photo-excited carrier distributions using femtosecond pump-probe laser has produced important information about high-T_c superconductivities [1–7]. This technique is particularly powerful for studying the ultrafast carrier relaxation dynamics in materials with an energy gap in electronic density of states. In a superconductor, for instance, the opening of a gap, as the temperature is decreased through the superconducting transition temperature (T_c), is manifested by a rapid increase in the amplitude and relaxation time of the photo-induced

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transient reflectance occurring in the time scale ranging from few hundred femtosecond to picosecond range. Within this scenario, two distinct quasiparticle relaxation times have been ubiquitously identified. The longer (~2-5 ps) diverging at T_c and the shorter (~500-700 fs), which is nearly temperature independent, are generally attributed to the manifestations of superconducting gap and pseudogap, respectively [5–8]. By probing on a detwinned YBCO single crystal, Gay et al. [9] and Stevens et al. [10] had further indicated that the quasiparticle relaxation dynamics can be very anisotropic within the ab-plane, with superconducting condensates relaxation being along a-axis and the b-axis response being associated with the pseudogap. However, we note that b-axis relaxation time ~3ps reported in [9,10] is compatible to the superconducting gap related relaxation dynamics. While it is much longer than the pseudogap associated relaxation cited above [5–8] . In addition, very recently, it has been pointed out by Müller [11] that the optically probed quasiparticle dynamics, being bulk in nature, is in fact better fitted to the expected characteristics of an *isotropic gap* [12]. It appears that the rich and yet complicated physics involved in the ultrafast optically-excited quasiparticle dynamics still remains unsettled.

In this study, we present experimental results obtained from a series of well characterized (001)-, (100)- and (110)-oriented YBCO superconducting films prepared by pulsed laser deposition. The well-controlled film orientation allows us to probe the relaxation dynamics along various crystal orientation using respectively polarized pump and probe laser beams. The significant anisotropies in both the magnitude of the characteristic relaxation time and the temperature dependence of the photo-induced transient reflectance change indicate the nature of relaxation channel might be intrinsically different along different orientations. The implications of the results are discussed in terms of the d-wave gap symmetry.

2. Experimental

The samples used in this study are optimal doped superconducting YBCO films prepared by pulsed laser deposition. All films were about 300-350 nm thick. The KrF excimer laser (λ= 248 nm) was mostly operated with an estimated energy density of about 5 J/cm^2 and a repetition rate of 5 Hz. However, in order to obtain the desired epitaxial orientation, the substrate, deposition temperature (T_s), and even the oxygen partial pressure (PO_2) were varied. For (001)-oriented YBCO films $SrTiO_3$(100) substrate was used with PO_2 = 0.3 Torr and Ts = 760 ^{o}C. For (110)-oriented films $SrTiO_3$(110) substrate was used. However, in order to obtain a pure (110)-oriented film

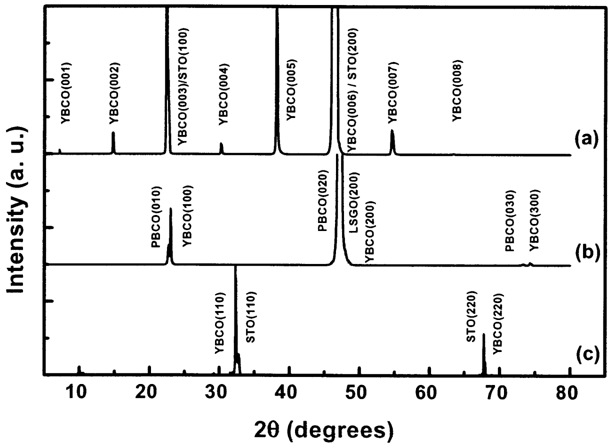

Fig. 1. The XRD results of the YBCO films investigated in this study. Traces (a), (b), and (c) are for (001)-, (100)-, and (110)-oriented films, respectively.

with acceptable superconducting properties, we first deposited a layer of (110)-$PrBa_2Cu_3O_7$ (PBCO) at 720 ^oC, then immediately raised the temperature to 790 ^oC for YBCO deposition. This process not only has successfully suppressed the growth (103)-YBCO but also improved the Tc significantly [13]. Finally, for the preparation of (100)-YBCO films, a novel process has been employed [14]. Briefly, a thin PBCO buffer layer was deposited at 660 ^oC on $LaSrGaO_4(100)$ substrate with $PO_2 = 0.1$ Torr. Without interrupting PBCO deposition, the substrate temperature was raised from 660 ^oC to 780 ^oC at a rate of 20 ^oC/min. After it reaches 780 ^oC, the PO_2 was kept at 0.28 Torr for subsequent YBCO deposition. The corresponding x-ray diffraction (XRD) results are displayed in Figure 1, where traces (a), (b), (c) represent the result of (001)-, (100)-, and (110)-oriented films, respectively.

The pump-probe measurements used a mode-locked Ti:sapphire laser which produces a 75 MHz train of 45 fs

pulses with a central wavelength of 798 nm (~1.55 eV). The details of the optics and transient reflectance (DR/R) measurements were similar to those reported previously [15,16]. We emphasize, however, in the present system, a pair of SF10 prisms were used to compensate the dispersion originated from the A-O modulator (which was used to replacing optical chopper to improve the modulation frequency), and had narrowed the pulse width to 45 fs as compared to 150 fs used before. This has, in turn, improved the time resolution of the quasiparticle relaxation significantly. The ratio between the intensity of the pump beam and the probe beam is usually set at 20:1, but is subject to change when necessary. In order to reveal the anisotropy along different crystal axes, the pump and probe beams are separately polarized. It is noted that, except for the (001)-films where a- and b-axis are indistinguishable, the amplitude of DR/R can vary by order of magnitudes when the polarized electric field of the probe beam is parallel or perpendicular to that of pump beam, suggesting that excitation and relaxation are taking place along the same channel. As a consequence, in this study we only report results obtained with Epump//Eprobe.

3. Results and discussion

Before discussing the results, it should be heuristic to note the generic processes involved in the measurements. In time-resolved pump-probe experiments, the pump pulse excites electron-hole pairs which then relax to states in the vicinity of the Fermi energy (E_F) by electron-electron and electron-phonon scattering creating a nonequilibrium quasiparticle distribution. This process occurs within a time scale of ~100 fs [4,12]. The presence of a gap near E_F requires phonons with specific energy to proceed further relaxation, making carriers accumulate in quasiparticle states above the gap. This, in turn, gives rise to a transient change in absorbance or reflectance to be detected by a second probe laser pulse. The amplitude and characteristic relaxation time of the measured DR/R thus give important information about how many quasiparticles are accumulated and how big the gap is.

Figure 2 shows the results of time-resolved DR/R measured along four different orientations at T =110K. There are several features to be noted. Firstly, the relaxation time in all cases is in the order of few tenths of ps, consistent with the absence of gap in the normal state. The relaxation of DR/R along b-axis persisting into a negative regime, nonetheless, is markedly different from that probed along other crystalline axes. Similar behavior has been observed in the single crystal experiments [9,10], except that the time constant derived from those results was ~3ps. Although the temperature independent characteristic has led the authors to identify it as manifestation of pseudogap, the magnitude was inconsistent with other experiments [5–8]. The distinct behavior, however, is indicative that we are indeed probing the b-axis.

As shown in Fig.3, when temperature was lowered to below T_c (indicated on the figure for each film), DR/R exhibits drastic changes. In all cases the amplitude of DR/R grows and the relaxation time constant appears to

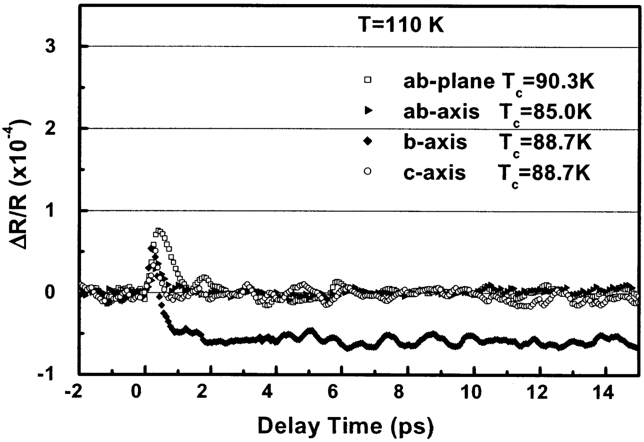

Fig. 2. DR/R measured at 110 K along various crystal orientations. The open square is probing in the ab-plane using a (001)-YBCO film; solid triangle is for probing along ab-axis diagonal using (110)-YBCO film; solid diamond and open circle are probing along the b-axis and the c-axis of the (100)-YBCO film, respectively.

prolong into ps range, indicating a gap opening near the EF has occurred [5–8]. The change in amplitude, however, is far more pronounced when probing in the ab-plane and along b-axis. This is better illustrated by the temperature dependence of DR/R amplitude shown in Fig. 4. It is interesting to note that, from the results of (100)-YBCO alone, the quasiparticle relaxation dynamics has evidently displayed significant amplitude anisotropy along b-axis and c-axis. While measurements along ab-axis diagonal and in the ab-plane show similar extent of anisotropy. Since the amplitude of DR/R has been associated with the accumulation of carriers in the quasiparticle states, which should be very much dependent on the existence and exact magnitude of a gap, the current results seem to imply certain intrinsic nature about the superconducting gap symmetry.

As noted above, the qualitative agreement observed here and in single crystal experiments [9,10] has indicated that we are indeed probing the characteristics along b-axis. However, with the improved time resolution (45 fs as compared to 100 fs used in [9,10]), we have evidently observed that both the amplitude and relaxation time of DR/R are temperature dependent. Thus, it seems somewhat equivocal to identify it as being a sole consequence of

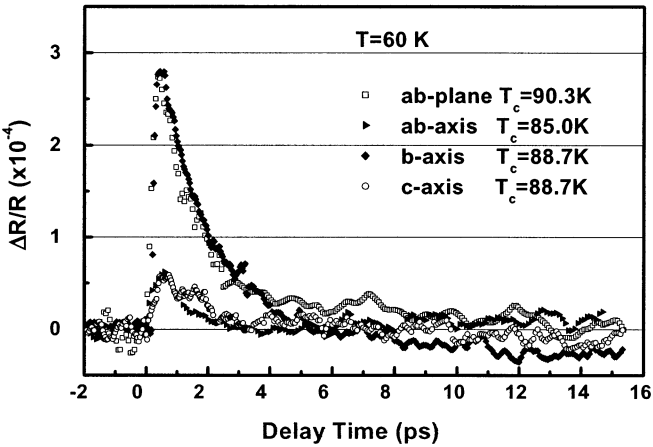

Fig. 3. DR/R measured at 60 K along various crystal orientations. The symbols are the same as those shown in Fig. 2.

pseudogap. In particular, in both cases, the samples were all near optimally doped, where pseudogap and superconducting gap are vaguely separable. On the other hand, if we interpret the results in terms of intrinsic gap anisotropy inherent in d-wave symmetry, a more consistent picture emerges. In that, the current results imply significant gap opening along a- and b-axes, resulting in drastic changes in both the amplitude and relaxation time constant in DR/R. The gradual change for ab-plane results can be regarded as an average of all in-plane orientation and perhaps some intrinsic anisotropy along a- and b-axis, as well. While for results probing along c-axis and ab-axis diagonal, it can be understood as being due to vanishing gap expected from two-dimensional d-wave superconductivity with nodal direction running along ab-diagonal.

As pointed out by Stevens et al. [10], the suggestion of attributing the pump-probe results obtained from thin films to s-wave superconductivity for YBCO proposed by Kabanov et al. [12] and later by Muller [11] is dispensable as the response obtained from (001)-oriented films might have been dominant by response along b-axis. This is also consistent with our b-axis and ab-plane results shown in Fig. 4. A more quantitative measure on the exact values of gap anisotropy and other features with detailed analyses on the temperature-dependent relaxation time with improved resolution will be given elsewhere.

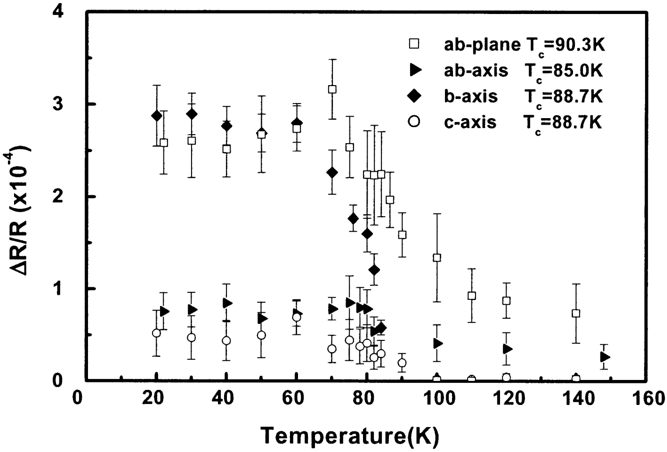

Fig. 4. Temperature dependence of DR/R measured along various crystal orientations. The symbols are the same as those shown in Fig. 2.

4. Summary

In summary, by using a series of optimally doped YBCO superconducting thin films with well-characterized orientations, the anisotropies of photo-induced quasiparticle relaxation dynamics along several major crystalline axes are unambiguously revealed by the time-resolved femtosecond pump-probe spectroscopy. With improved time resolution of the laser pulse to 45 fs, the component associated with superconducting gap opening appeared to be temperature dependent along every crystal axis. The results suggest that the d-wave nature of the superconducting gap symmetry might be responsible for the anisotropic characteristics observed.

5. ACKNOWLEDGMENTS

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REFERENCES

1. S.G. Han, Z.V. Vardeny, K.S. Wong, O.G. Symko, and G. Koren, *Phys.Rev.Lett.* **65**, 2708 (1990).
2. J.M. Chwalek, C. Uher, J.F. Whitaker, G.A. Mourou, J. Agostinelli, and M. Lelental, *Appl. Phys. Lett.* **57**, 1696 (1990).
3. G.L. Eesley, J. Heremans, M.S. Meyer, G.L. Doll, and S.H. Liou, *Phys. Rev. Lett.* **65**, 3445 (1990).
4. C.J. Stevens, D. Smith, C. Chen, J.F. Ryan, B. Podobnik, D. Mihailovic, G.A. Wagner, and J.E. Evetts, *Phys. Rev. Lett.* **78**, 2212 (1997).
5. J. Demsar, B. Podobnik, V.V. Kabanov, Th. Wolf, and D. Mihailovic, *Phys. Rev. Lett.* **82**, 4918 (1999).
6. R.A. Kaindl, M. Woerner, T. Elasaesser, D.C. Smith, J.F. Ryan, G.A. Farnan, M.P. McCurry, and D.G. Walmsley, *Science* **287**, 470 (2000).
7. D. Dvorsek, V.V. Kabanov, J. Demsar, J. Karpinski, S.M. Kazakov, and D. Mihailovic, **cond-mat**/0203614 v2, 2002.
8. R.A. Kaindl, M. Woerner, T. Elasaesser, D.C. Smith, J.F. Ryan, G.A. Farnan, M.P. McCurry, and D.G. Walmsley, *Phisica C* **341-348**, 2213 (2000).
9. P. Gay, C.J. Stevens, D.C. Smith, C. Chen, and J.F. Ryan, *Physica C* **341-348**, 2269 (2000).
10. C.J. Stevens, P. Gay, D.C. Smith, C. Chen, and J.F. Ryan, J. *Low Temp. Phys.* **117**, 1031 (1999).
11. K.A. Muller, *Phil. Mag. Lett.* **82**, 279 (2002).
12. V.V. Kabanov, J. Demsar, B. Podobnik, and D. Mihailovic, *Phys. Rev.* **B59**, 1497 (1999).
13. S.P. Chen, J.Y. Juang, K.H. Wu, T.M. Uen, and Y.S. Gou, unpublished.
14. C.W. Luo, M.H. Chen, C.C. Chiu, K.H. Wu, J.Y. Juang, T.M. Uen, Y.S. Gou, and J.Y. Lin, this conference.
15. K.H. Wu, C.W. Luo, J.Y. Juang, T.M. Uen, and Y.S. Gou, *Physica B* **284-288**, 939 (2000).
16. K.H. Wu, C.W. Luo, J.Y. Juang, T.M. Uen, and Y.S. Gou, *Chin. J. Phys.* **38**, 279 (2000).