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<b>Author(s)</b>	Watanabe, K.; Tsuchiya, S.; Toda, Y.; Kurosawa, T.; Oda, M.; Mertelj, T.; Madan, I.; Kabanov, V. V.; Mihailovic, D.
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## TIME-RESOLVED POLARIMETRY FOR PHOTOEXCITED QUASI-PARTICLE DYNAMICS IN BI2212

K. Watanabe, S. Tsuchiya, Y. Toda

*Department of Applied Physics, Hokkaido University, Kita-ku N13W8,  
Sapporo, 060-8628, Japan  
toda@eng.hokudai.ac.jp*

T. Kurosawa, M. Oda

*Department of Physics, Hokkaido University, Kita-ku N10W8,  
Sapporo, 060-0810, Japan*

T. Mertelj, I. Madan, V. V. Kabanov, D. Mihailovic

*Complex Matter Department, Jozef Stefan Institute, Jamova 39,  
Ljubljana, SI-1000, Slovenia*

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The dynamics of quasi-particle excitations with different symmetry is investigated in the superconducting (SC) and pseudogap (PG) states of the high-temperature superconductor  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+d}$  (Bi2212) using a polarization-sensitive optical pump-probe measurement. The observation of distinct anisotropies for SC excitations and for PG excitations by the probe beam polarization and absence of any dependence on the pump beam polarization evidence the existence of a spontaneous spatial symmetry breaking in the PG state.

*Keywords:* Pseudogap; Ultrafast optical spectroscopy; Cuprate superconductors; Spatial symmetry breaking.

### 1. Introduction

One of the interesting features in high- $T_c$  cuprates is a pseudogap (PG) developed in the electronic energy spectrum around  $T^*$  which is the temperature well above the superconducting (SC) transition temperature  $T_c$  in the underdoped (UD) region. The anticorrelation between  $T^*$  and  $T_c$  towards the antiferromagnetic phase is also an important issue, which can be explained in terms of competing energy gaps on the Fermi surface (i.e., FS dichotomy) based on the results of angle-resolved photoemission spectroscopy (ARPES), where the PG opens only on the antinodal parts of the FS.<sup>1,2</sup> As a result, the FS shows an arc shape in the normal state and the SC gap opens on the Fermi arc below  $T_c$ . In this case, both PG and SC gap seem to coexist in the SC state, and the

energy gap that develops on the Fermi arc below  $T_c$  will function as an effective SC gap.<sup>3-6</sup>

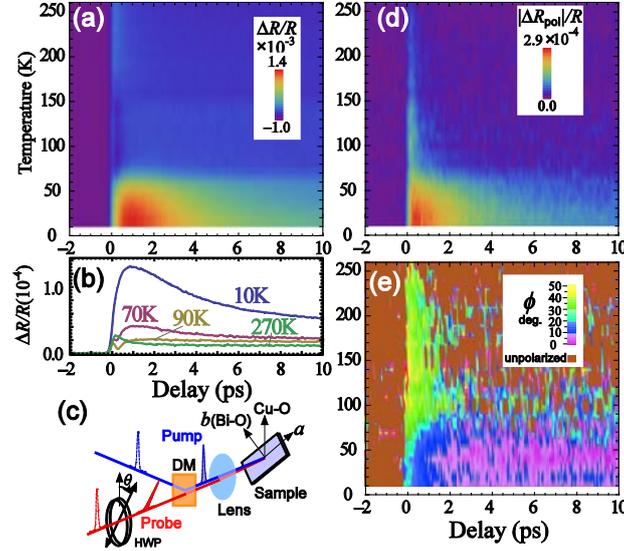


Fig. 1. (a)  $T$ -dependences of the photoinduced reflectivity ( $\Delta R/R$ ) transients at pump fluence  $F=1.6 \mu\text{J}/\text{cm}^2$ , where  $(\theta_{pu}, \theta_{pr})=(0^\circ, 90^\circ)$ . (b)  $\Delta R/R$  transients at typical temperatures. (c) A schematic illustration of the two-color pump-probe setup for time-resolved polarimetry. The probe ( $\lambda_{pr}=800 \text{ nm}$ ) was variably polarized by a half-wave plate (HWP) and was combined with the pump ( $\lambda_{pu}=400 \text{ nm}$ ) by a dichroic mirror (DM).  $T$ -dependences of (d)  $\Delta R_{pol}/R$  and (e)  $\phi$  as a function of delay time.

On the other hand, optical time-resolved pump-probe spectroscopy has revealed the dichotomous quasi-particle (QP) dynamics associated with the SC gap and PG excitations. In such time-resolved optical measurements, the dichotomous relationship can be characterized based on the nonequilibrium dynamics and on the bulk properties. Indeed, the two-component QP dynamics has been widely observed in various high- $T_c$  cuprates at low temperature below  $T_c$ . Here the two types of excitations were clearly resolved by the distinct relaxation times, temperature dependencies, and/or sign of the optical signal, depending on the material, doping level, and pump-probe conditions.<sup>6-12</sup> Furthermore, the probe polarization dependence of the two-component QP dynamics has also been reported.<sup>12-14</sup> However, the absence of a fundamental understanding of the optical processes involved in pump-probe experiments so far prevented analysis of the symmetry of excitations or detailed theoretical analysis of the excitations on a microscopic level. In this paper, we investigate a QP dynamics with different symmetry in the SC and PG of a high- $T_c$  cuprate Bi2212 using a time-resolved polarimetry (polarization-sensitive pump-probe measurement). A detailed polarization analysis of the QP transients reveals the dynamics of states associated with hidden broken symmetry and/or symmetry breaking in systems with competing orders.

## 2. Experimental

The experiments were performed on UD Bi2212 single crystal with  $T_c \approx 69$  K grown by the traveling solvent floating zone method. The time-resolved reflectivity change  $\Delta R$  was measured using a two-color pump-probe setup (pump  $E_{pu}=3.1$  eV ( $\lambda_{pu}=400$  nm) and probe  $E_{pr}=1.55$  eV ( $\lambda_{pr}=800$  nm)), where the coaxially overlapped pump and probe beams were focused onto the  $ab$ -plane of the crystal, allowing us to precisely evaluate the polarization dependence (Fig. 1(c)). The sample orientation was checked by a X-ray diffraction, in which the  $b$ -axis is determined by the direction of the multiple peaks responsible for a one-dimensional superlattice modulation. The nonequilibrium QP dynamics excited by the pump pulse can be evaluated by the reflectivity change  $\Delta R$  of the probe pulse with a variable delay time relative to the pump pulse. For the polarization measurements, the probe (and partly pump) polarization was variably rotated by an angle  $\theta$ , where  $\theta=0$  corresponds to the diagonal direction between the crystalline axes.

## 3. Results and Discussions

In Fig. 1(a) and (b), we plot the  $T$ -dependence of  $\Delta R/R$  transients at a slightly higher pump fluence of  $F=1.6$   $\mu\text{J}/\text{cm}^2$  for the probe polarization  $\theta=90$  deg. The upper panel (a) shows the density plot of the data, and the cross-sectional views at the selected temperatures are shown in (b). In the low fluence excitation condition  $F=0.2$   $\mu\text{J}/\text{cm}^2$ , the signal appears only below  $T_c$ . The signal amplitude increases with increasing the pump fluence, and then shows a saturation behavior at  $F_{th}=1.0$   $\mu\text{J}/\text{cm}^2$ . The temperature and pump fluence dependencies of the signal agree with those of the SC QPs reported previously, where the saturation behavior has been ascribed to the destruction of the superconducting condensate.<sup>8,15</sup> Above  $F_{th}$ , an additional fast relaxation component with opposite sign appears, which extends below and above  $T_c$  and disappears around  $T^*$ . This component has also been reported and assigned to the PG QPs.<sup>16</sup> When increasing the temperature above  $T^*$ , the signal with opposite sign to the PG (the same sign as SC component) becomes dominant, which has been ascribed to the electron-phonon relaxation in the metallic state.<sup>17</sup> The PG component appearing at higher  $F$  is attributed to the PG with higher saturation threshold and therefore becomes visible below  $T_c$  in the high excitation condition.<sup>16</sup>

The polarization dependences of  $\Delta R/R$  transients at various temperatures are obtained by rotating the probe polarization from  $\theta=0$  to  $360^\circ$  at each temperature. The anisotropy of  $\Delta R/R$  can be evaluated by fitting a sinusoidal function  $\Delta R(\theta) = \Delta R_{pol} \cos[2(\theta - \phi)] + \Delta R_{avg}$ . The magnitudes of the polarized component  $\Delta R_{pol}$  and their angles  $\phi$  are summarized in Fig.1 (d) and (e). The results clearly indicate the  $T$ -dependent anisotropic signal, which can be associated with the SC and PG QP components.

In order to show the details, the polarization dependences of  $\Delta R/R$  transients at selected temperatures are presented in Fig.2. The left panels show the density plots of the data together with the cross-sectional views at  $\theta_{pr}=0^\circ$ . The polar plots (right) show the probe angular dependences of the signal amplitude, where the crystalline axes are

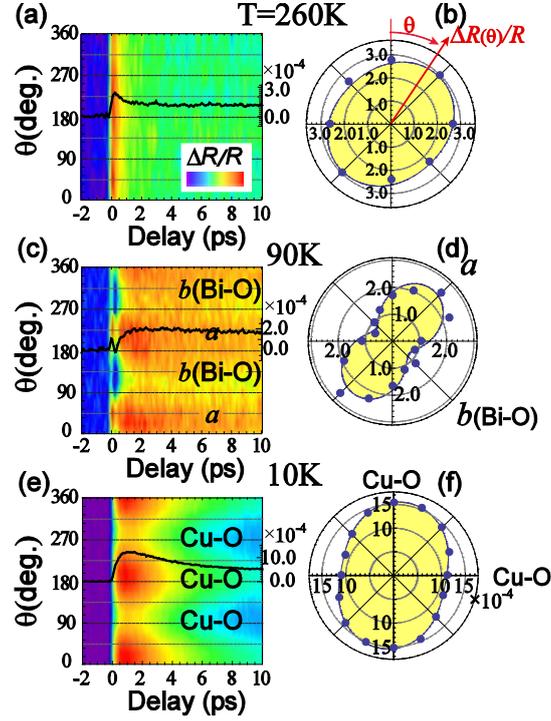


Fig. 2. (left)  $\Delta R/R(\theta)$  at typical temperatures. (right) Polar plots of the maximum values of  $\Delta R/R(\theta)$ .

oriented along the diagonal directions. Note that the  $a$  and  $b$  axes in Bi2212 are along the Bi-O bonds and rotated nearly  $45^\circ$  from the Cu-O bonds. In this case, the vertical and horizontal axes in the polar plots almost correspond to the directions along the two orthogonal Cu-O bonds. At the lowest temperature, where the SC signal is dominant, the polarization is slightly elliptic with the long axis close to the Cu-O bonds direction (Fig.2 (a) and (b)). On the other hand, the polarizations of the PG signal that is dominant above  $T_c$  is oriented along the crystalline axes (Fig.2 (c) and (d)). By taking into account the negative sign of the PG signal, the exact direction of the polarization is assigned to be  $b$ -axis (direction of the Bi-O bonds). The tilted long axis at the lowest temperature is therefore associated with the contribution of the PG signal. The  $\Delta R/R$  transients above  $T^*$  are almost independent of  $\theta$  (Fig.2 (e) and (f)) and show an isotropic electron energy relaxation. It is important to note that the probe polarization anisotropy is independent of the pump polarization, which can be briefly confirmed in Fig.3, where the polar plots of the signal amplitude at 10 K with different pump polarizations clearly show the identical polarizations. We also note that no qualitative difference was observed by rotating the pump polarization at the other temperatures.

The absence of any pump polarization anisotropy rules out stimulated Raman excitation as the excitation mechanism for any anisotropic modes. On the other hand, the

anisotropic responses observed by the probe polarization dependence suggest a contribution induced by the pump pulse. In the dissipative excitation process, the high energy photoexcited QPs induced by the pump relaxes the excess energy independently by nonelastic scattering with emission of excitations of various symmetries. Therefore the information about the incoming photon polarization is lost during this process. At the same time, the state of the system is characterized by time-dependent QP densities corresponding to different excitations, which can only couple to the totally symmetric representation of the dielectric tensor and coherently excite only the totally symmetric modes.<sup>18</sup> However, in the presence of a local, dynamic or hidden symmetry breaking, totally symmetric excitation can coherently excite the nonsymmetric modes. Therefore the anisotropic responses observed by the probe indicate that the anisotropic modes are excited coherently because the underlying tetragonal point group symmetry is spontaneously broken below  $T^*$ . In other words, we can probe the symmetry breaking by means of the totally symmetric QP excitations.

Formally, the anisotropic  $B_{2g}$  symmetry breaking of the pseudotetragonal symmetry is present at room temperature in Bi2212 due to the weak inherent orthorhombicity of the underlying crystal. However, our data shows both symmetry breakings are suppressed at room temperature and appears clearly below  $T^*$ , implying that the  $B_{2g}$  component is not simply a consequence of the underlying Bi2212 orthorhombicity. Moreover, the data show another  $B_{1g}$  symmetry breaking to occur slightly below  $T^*$  and clearly below  $T_c$ . While the BiO chain can in principle cause the  $B_{2g}$  symmetry breaking, it cannot cause the  $B_{1g}$  symmetry breaking. The appearance of the SC response in the  $B_{1g}$  symmetry can therefore be associated with a preexisting underlying order oriented along the Cu-O bonds, which is achieved by stripe ordering or similar textures.

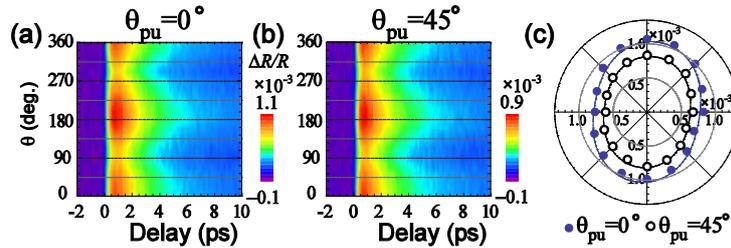


Fig. 3. (a) (b)  $\Delta R/R(\theta)$  at  $T=10$  K with different pump polarizations. (c) Polar plots of the  $\Delta R/R(\theta)$ .

#### 4. Conclusions

We investigated the polarization anisotropy of photoexcited QP dynamics in Bi2212. By rotating the probe polarization, we found that the  $\Delta R$  below  $T_c$  includes two distinct polarization components, which are associated with SC and PG QPs. The  $T$ -dependent data also show that both components are suppressed at room temperature and appears clearly below  $T^*$ , indicating that the anisotropic modes are excited coherently because the underlying tetragonal point group symmetry is spontaneously broken below  $T^*$ .

Assuming a homogeneous broken-symmetry state, the observed anisotropy would be a single stripe order. For an inhomogeneous case, the anisotropy is consistent with a picture of locally ordered commensurate charge-density-wave (CDW) patches along the Cu-O bonds, nematic order, or stripe order with different size stripes.

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