Nonlinear polarization optical response to entangled state of biexciton

T Tsuji, G Oohata*, and K Mizoguchi
Department of Physical Science, Graduate School of Science, Osaka Prefecture University, 1-1 Gakuen-cho, Naka-ku, Sakai 599-8531, Japan

E-mail: oohata@p.s.osakafu-u.ac.jp

Abstract. We have investigated the nonlinear optical responses of biexcitons which show the polarization entanglement by using pump-probe spectroscopy. Measuring all polarization combinations of pump-probe signals and precise analysis using the idea of quantum tomography enable us to extract the specific property of the entanglement. The obtained results indicate the two kinds of the third-order nonlinear response, namely the photo-induced absorption and the optical Kerr effect. From the analysis with the optical Kerr effect signals, the polarization correlations of the optical responses are clearly derived.

1. Introduction
Quantum entanglement is an important physical phenomenon on the field of the quantum information science. Recently, it is reported that the quantum entanglement plays important roles also in various physical phenomena including biological systems [1,2]. Correspondingly, efficient and selective detection methods for the quantum entanglement are highly desired for the demonstration and further applications of the entanglement in the biological systems. Most of conventional detection methods for the quantum entanglement are implemented by direct measurement of the emitted entangled photons [3,4]. An example of them is measurements of photon pairs emitted in the optical process called Resonance Hyper Parametric Scattering (RHPS) in CuCl, which is the two-photon emission by the decay of the biexciton [5]. In the RHPS process, because the emitted photon-pairs are closely related to the quantum state of the biexciton, we can investigate the quantum entanglement of the biexciton by coincidence counting of the photon-pairs. However, since extremely efficient photon counting techniques are required for the direct detection of the photon-pairs, it is difficult to apply the detection methods to studies for the entanglement properties of various materials.

In this paper, we report on the observation of the entanglement of the biexciton in a CuCl single crystal. Based on the concept of the quantum tomography which is an analytical method to estimate a quantum state from the results of all projection measurements [6,7], pump-probe measurements for biexcitons were implemented in all polarization combinations. In addition, the obtained spectra by the pump-probe measurements were decomposed into the elements of optical Kerr effect (OKE) and photo-induced absorption (PIA) by using a simple theoretical model. Then we discuss the polarization correlation of the OKE and demonstrate the entanglement of the biexciton.

2. Theoretical background
The optical responses related to the biexciton have the third-order nonlinearity. Therefore, it is
predicted that the pump energy dependence of the pump-probe signals for the biexcitons correspond to the profile of the third-order nonlinear susceptibility $\chi^{(3)}$. In general, the $\chi^{(3)}$ function can be expressed as

$$\chi^{(3)} \propto \left[ \Omega_{bx} - (\omega_{pu} + \omega_{pr}) + i\Gamma \right]^{-1},$$

where $\omega_{pu}$ and $\omega_{pr}$ are the energies of the pump and probe beam respectively, and $\Omega_{bx} (=6.372$ eV in case of a CuCl) and $\Gamma$ are the resonance energy and the damping constant of the biexciton respectively [8]. The profiles of the real and imaginary parts of the $\chi^{(3)}$ are shown in figure 1(a), that means the change of the refractive index and the absorption coefficient respectively which are caused by the pump beams. These optical processes are referred to as the OKE and the PIA respectively.

Here, let us consider how these optical responses affect the polarizations. For the simplification, we assume that the polarization of the initial state is the diagonal polarization, that is, $|D\rangle \propto |H\rangle + |V\rangle$, where $|H\rangle$ and $|V\rangle$ are the state of horizontal and vertical polarizations respectively and the third-order nonlinear responses are applied only to the H polarization component. Considering the contribution of the OKE and PIA, the initial state $|D\rangle$ changes to $|D'\rangle_{\text{OKE}} \propto e^{-i\Delta\theta} |H\rangle + |V\rangle$ and $|D'\rangle_{\text{PIA}} \propto \sqrt{1-\Delta\alpha} |H\rangle + |V\rangle$, where $\Delta\theta$ is the variation of the phase difference between H and V components which originate from the variation of the refractive index and $\Delta\alpha$ is the variation of the absorption coefficient of H component. Then, the initial state $|D\rangle$ is slightly modulated to the state of right-circular polarization ($|R\rangle \propto |H\rangle + i|V\rangle$) by the OKE, and to the state $|V\rangle$ by the PIA. Similarly, we consider the change of the 6 polarizations: H, V, D, anti-diagonal A, R, and left-circular L polarizations. The polarization change caused by the OKE and PIA can be expressed as the change of a Bloch sphere for the 6 polarizations shown in Fig. 1 (b), (c) respectively. It can be seen that the OKE corresponds to the rotation of the sphere on the particular axis and the PIA does to the deformation in one direction of the sphere.

3. Experimental setup and results
In the experiment, we used a CuCl single crystal prepared by vapor deposition. The second harmonic

![Figure 2](https://example.com/figure2.png)

**Figure 2.** The schematic diagram of the experimental setup. The polarizations of the pump and probe beam are set to 6 polarizations: H, V, D, A, R, and L polarizations. The intensity differences of the transmitted probe beam with H-V, D-A, and R-L bases are measured.
wave of a mode-locked Ti:Sapphire laser was used as an excitation pulse source and its energy was controlled by 4f optical systems with diffraction gratings. The energy of a probe beam, $\omega_{pr}$ was fixed at 3.176 eV and the energy of the pump beam, $\omega_{pu}$ was changed from 3.154 to 3.217 eV. The schematic diagram of the experimental setup is shown in figure 2. Each beam was set to 6 polarizations: H, V, D, A, R, and L polarizations by using polarizers and waveplates. Then, the beams were focused on the sample at 10 K. The transmitted probe beam was split into two beams with three bases of polarizations: H-V, D-A, and R-L bases by using waveplates and a polarizing beam splitter. The intensity differences between the two beams in the respective bases were obtained by the pump-probe signals using a lock-in amplifier.

In the case that the polarization of the pump beam is H, the intensity difference observed in the respective polarization bases are plotted as a function of pump energy in figure 3. Each spectrum shows typical three structures at the resonance energy of the biexciton corresponding to the fixed energy of the probe beam: the peak (dip) profile, differential profile of Lorentz function, and nearly no signal. These optical responses correspond to the profiles of the real part and the imaginary part of the third-order nonlinear susceptibility $\chi^{(3)}$ shown in figure 1(a). Therefore, these results suggest that the OKE and the PIA occur at the resonance energy of the biexciton.

4. Discussion

We discuss each spectrum in detail using a simple theoretical model. As previously mentioned and experimentally confirmed, optical responses of the biexciton correspond to the profile of the third-order nonlinear susceptibility $\chi^{(3)}$. Therefore, we decomposed the results into the real part (differential shape) and imaginary part (peak (dip) shape) of the $\chi^{(3)}$. In figure 4, we show the pump energy dependence of the intensity differences and the fitting result in D-A basis in the case of the polarization combination of the H-pump and the R-probe beams. From the fitting with the theoretical model, the component of the real part of the $\chi^{(3)}$ is relatively large, indicating that OKE is dominant. The values of the OKE and the PIA can be estimated from the coefficients of each part of the $\chi^{(3)}$ obtained in the comparison between the experimental results and the theoretical model. In the following discussion, we focus only on the OKE signals.

As previously mentioned in section 2, the probe polarization change can be expressed as the change in the Bloch sphere for the 6 polarizations. Therefore, from the experimental results it is possible to consider the values of the OKE on each rotation axis independently. Then, the normalized relative values of the OKE on each axis are obtained from 12 experimental results for each pump polarization. The results are shown in figure 5 as a bar chart. For each polarization of the pump beam, it is found that the value on the particular rotation axis is clearly large which normalized to ± 1, whereas the values on the other rotation axes are almost zero. Therefore, these results demonstrate that the rotation axes and values of the OKE have obvious correlations with the polarization combinations of the pump and probe beam.
Finally, we discuss the rotation axes and the values of the OKE related to the entangled state of the biexciton. Considering the spin state of the biexciton, $|\psi_{bx}\rangle = \frac{1}{\sqrt{2}}(|\uparrow\rangle|\downarrow\rangle + |\downarrow\rangle|\uparrow\rangle)$, the wave function of the photons which interact with the biexcitons is expressed in terms of polarizations as

$$|\psi_{ph}\rangle = \frac{1}{\sqrt{2}}(|H\rangle|H\rangle + |V\rangle|V\rangle) = \frac{1}{\sqrt{2}}(|D\rangle|D\rangle + |A\rangle|A\rangle) = \frac{1}{\sqrt{2}}(|R\rangle|L\rangle + |L\rangle|R\rangle).$$  

This equation determines the combinations of the pump and probe polarizations that can interact with the biexciton. We can find that the relation of equation (2) is analogous to the experimental results shown in figure 5. Consequently, it is clear that the obtained polarization correlations of the optical responses express the spin state of the biexciton.

5. Conclusion
We have observed the OKE and the PIA due to the biexciton state in 72 combinations of polarization. From the obtained results, we demonstrate that the nonlinear optical response related to the quantum entanglement of the biexciton appears in the pump energy dependence of the pump-probe signals by the analysis based on the quantum tomography.

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