Resonant Optical Kerr Response with Ultrashort Decay Time by Nonlocal Wave Coupling of Light and Excitons

M. Ichimiya[1], T. Umakoshi[1], H. Murata[1], T. Kinoshita[2], H. Ishihara[2], and M. Ashida[1]
[1]*Department of Materials Engineering Science, Osaka University, Osaka 560-8531, Japan*
[2]*Department of Physics and Electronics, Osaka Prefecture University, Osaka 599-8531, Japan*
ichimiya@laser.mp.es.osaka-u.ac.jp

Abstract: Resonant optical Kerr effects have been investigated in high-quality CuCl thin films. The peculiar spectral feature and ultrafast response below 200 fs due to a long-range coherent coupling between light and multinode-type excitons are observed.
OCIS codes: (320.7110) Ultrafast nonlinear optics; (310.6860) Thin films, optical properties

1. Introduction

An ultrafast optical response in semiconductor materials as well as a large nonlinearity is essential to improve the performance of optical switching devices, while it has been considered to be difficult due to the long radiative decay time of excitons. Nanostructures have attracted much attention as one of the promising candidates for realizing ultrafast optical response with large nonlinearity. In nanostructures, the optical responses are described by the long wavelength approximation (LWA), where the confined polarization wave coherently interacts with light wave, and the optical response due to the coupling with the lowest excitonic state ($n=1$) is dominantly observed. The oscillator strength increases with the system size [1–3], while the increase has been believed to be saturated in the size region larger than the excitonic coherent length or the wavelength of light. On the other hand, the size-resonantly enhanced nonlinear optical response in a system with high crystalline quality is theoretically proposed, where the self-consistent interaction between the internal field and the induced polarization causes an enhancement of the response field at a particular size and photon energy [4].

Recently, we reported experiments of degenerate four-wave mixing (DFWM) in high-quality CuCl thin films [5]. CuCl is a useful wide-gap semiconductor with huge exciton binding energy (about 200 meV) and a material that manifests remarkable nonlinear effects in the non-LWA regime. We observed a strong coupling between light and a multinode-type exciton combined with the ultrafast radiative decay of 100 fs. The calculated result of the radiative corrections predicts large radiative width, corresponding to exceptionally short response time for excitons (below 10 fs in CuCl) [6]. Such a large radiative correction is caused by the long-range coupling between light and excitons over several wavelengths and is expected to appear in other optical phenomena. The optical Kerr effect is one of nonlinear optical effects and suitable for demonstrating the ultrafast switching action. In particular, the nonlinearity of optical Kerr effect is much enhanced by using light in the exciton resonance region. In the present work, we investigate the resonant optical Kerr effect of confined excitons using CuCl thin films with high crystalline quality and with thicknesses in the non-LWA regime.

2. Experimental procedures

CuCl thin films were grown on (111)-oriented CaF_2 substrates by molecular beam epitaxy (MBE). Their crystalline qualities were significantly improved by our newly developed technique involving electron beam irradiation before the MBE growth [7]. The grown films were mounted in a helium flow cryostat and cooled to below 10 K. Resonant optical Kerr spectra were measured with the second harmonic of a mode-locked Ti:sapphire laser, whose pulse duration and repetition rate were 110 fs and 80 MHz, respectively. The photon energy of the light was set around the exciton energy in CuCl, and the spectral width was 20 meV which covered the whole exciton resonance region. The light was divided into linearly polarized pump and probe beams, which were simultaneously injected into the sample surface. The polarization of the probe beam was set to vertical, and that of the pump beam was set at 45° to that of the probe beam. The transmitted probe beam was sent through a polarizer with the horizontal polarization, and the horizontal component of the probe beam was detected as the Kerr signal by a monochromator equipped with a charge-coupled device. The spectral resolution was 0.1 meV. The film thickness and the phase decay constant of excitons (Γ) at the focused spot were derived by fitting to the reflection spectrum measured with the same geometry.

3. Results and discussion

The blue line shown in Fig. 1(a) indicates a resonant optical Kerr spectrum of a high-quality CuCl thin film with a thickness of 224 nm and Γ of 0.50 meV. E_T and E_L indicated by vertical dashed lines show the transverse and

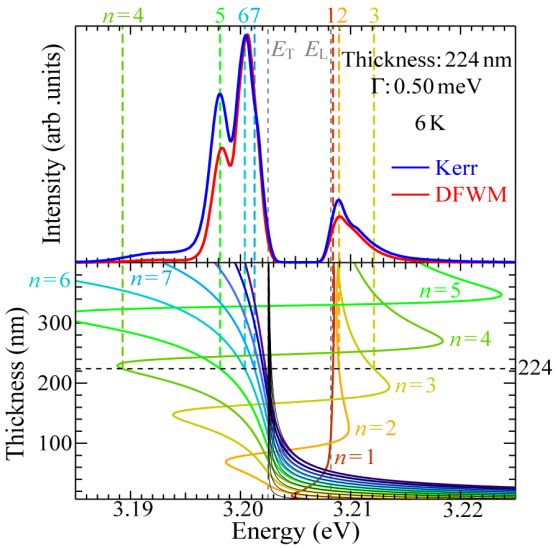

Fig. 1 (a) Observed Kerr spectrum (blue line) and DFWM spectrum (red line) in a CuCl thin film. (b) Film thickness dependences of calculated eigenenergies including the radiative shift and uncoupled excitonic modes (thin black lines) [6].

longitudinal exciton energies of CuCl bulk crystal, respectively [8]. The spectrum exhibits a peculiar structure with several peaks, which have never been observed in any Kerr spectrum. Figure 1(b) shows the film thickness dependence of the eigenenergies including the radiative shift in the coupled system of light and multinode-type excitons [6]. The energy at the intersection of the calculated curve and a horizontal dashed line corresponds to the eigenenergy at the film thickness of 224 nm. The eigenenergies precisely coincide with the energies of the peaks in the resonant optical Kerr spectrum. The DFWM spectrum measured using the same geometry as that of resonant optical Kerr effect is shown as red line in Fig. 1(a) for the comparison. The spectral feature with several peaks is in good agreement with the feature of Kerr spectrum. The photon energy at each peak corresponds to the eigenenergy including the radiative shift, and the width of each component reflects the radiative width, which is the imaginary part of the radiative corrections, due to long-range coupling between light and exciton as our previous reports [5]. A similar agreement between the Kerr spectrum and the DFWM spectrum is observed for CuCl thin films with other thicknesses. These results suggest that the shape of resonant optical Kerr spectrum closely reflects the radiative corrections similar to that of DFWM, where 100 fs-class response and signal at room temperature are observed [9]. Since the decay profile of DFWM spectrum reflects the radiative decay time calculated from the radiative widths, the Kerr response may be affected by the radiative width. Figure 2 shows the delay time dependence of the probe beam for the excitonic states of $n = 2$, 5. The green lines represent the exponential functions convoluted by the excitation laser profile, where the time constants of the calculated radiative decay time ($\tau_r = \hbar/2\gamma$) derived from the radiative width (γ) are substituted [9]. At the film thickness of 224 nm, γ of $n = 2$, 5 are 1.9 meV and 0.90 meV [6], and τ_r are 170 fs and 370fs, respectively. The measured resonant optical Kerr response reflects the calculated radiative decay profile of exciton.

In conclusion, we have successfully verified that the resonant optical Kerr spectrum shows the anomalous mode structures similar to that of DFWM in a high-quality CuCl thin film. The spectral shape reflects the radiative shift and width due to harmonized wave-wave coupling between light and multinode-type excitons. The components for the excitonic states with the large radiative width in the Kerr spectrum show ultrafast responses with the order of 100 fs, which are consistent with the calculated radiative decay times of excitons. These agreements between experimental results and the calculated mode structures suggest that the resonant optical Kerr response below 10 fs will be achieved for a CuCl thin film with high crystalline quality and appropriate film thickness.

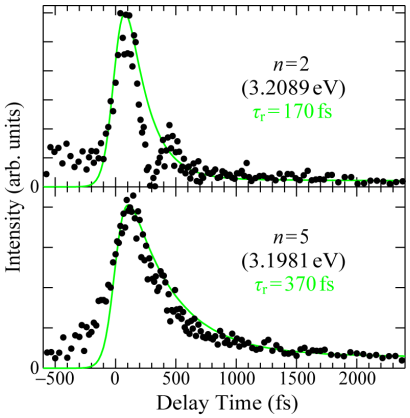

Fig. 2 Observed delay time dependence of the Kerr intensity in a CuCl thin film. The green lines represent the exponential functions convoluted by the excitation laser profile.

Acknowledgments

The present work was supported by a Grant-in-Aid for Young Scientists (B) (24740210) and a Grant-in-Aid for Scientific Research (A) (60273611) from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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