Enhancement of Nonlinear Signal of Multinode-type Excitons Using Ultrashort-pulse Excitation in CuCl Thin Films

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We have demonstrated a remarkable strong coupling between light and a multinode-type exciton in high-quality thin films with thicknesses of a few hundreds of nanometers, and succeeded in observing exceptionally high-speed superradiance much faster than the phonon-mediated dephasing process at room temperature [1,2]. However, the measured signal intensity at room temperature is three orders of magnitude smaller than that at 5 K because the intensity for the excitonic state with the ultrafast radiative decay time is restricted due to too narrow spectral width of excitation pulse (20 meV). In the present work, we investigate the effect of ultrashort-pulse excitation with large spectral width on the nonlinear optical response in the coupled system of light and multinode-type excitons.

Figure 1 shows a transient grating (TG) spectrum of a high-quality CuCl thin film with a thickness of 283 nm. The second harmonic of a mode-locked Ti:sapphire laser with a pulse duration of 60 fs and a spectral width of 44 meV was used as an excitation source. The TG spectrum exhibits a structure with several peaks peculiar to the coupled system of light and multinode-type excitons. Figure 2 shows the delay time dependence of the TG signal intensity for the excitonic state of $n = 4$ with the large radiative width of 10 meV at 283 nm. The solid line represents the exponential function convoluted by the excitation laser profile, where the radiative decay time of 32 fs calculated from the radiative width of 10 meV is substituted as the time constant. The decay profile shows ultrafast radiative decay in the order of 10 fs, which is hard to be observed under the excitation with a spectral width of 20 meV. Furthermore, the TG signal intensity at room temperature is several tens of times greater than that for 20 meV excitation [2]. These results indicate that the nonlinear optical response of the excitonic state with the large radiative width is enhanced under ultrashort-pulse excitation.

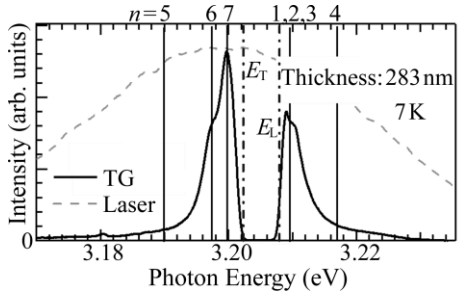

Fig. 1 TG spectrum in a CuCl thin film. E_T and E_L indicate transverse and longitudinal exciton energies of CuCl, respectively.

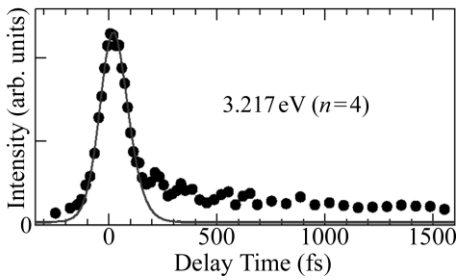

Fig. 2 Delay time dependence of TG signal intensity for $n = 4$.

[1] M. Ichimiya *et al.*, Phys. Rev. Lett. **103**, 257401 (2009).
[2] M. Ichimiya *et al.*, Phys. Status Solidi B **248**, 456 (2011).