Pulse Excitation of Exciton-Radiation Coupled Modes with Ultrashort Radiative Lifetimes in ZnO Thin Films

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In recent years, one of unconventional coupling schemes of radiation and spatially extended excitons has been clarified for CuCl thin films in which the excitonic center-of-mass (c. m.) motions are quantized. Especially in nano-to-bulk crossover size regime, the long-wavelength approximation is violated due to submicron order of the coherence length of the c. m. wavefunctions, which actualizes wave-wave coupling of the excitons and radiation. In this situation, not only dipole-type but also multipole-type excitons receive the large radiative correction, and well phase-matched modes obtain ultrashort radiative lifetimes with increase of the film thickness [1]. These results stimulate us to further examine more potential materials such as ZnO because of its wide bandgap and high excitonic stability.

An essential difference of ZnO excitons from CuCl ones is the multi-component nature of excitons; namely A- and B-exciton branches are generated from the degenerate valence-band structure at Γ point. Taking account of these two branches, we have theoretically revealed exciton-radiation coupled mode structures of ZnO. In contrast to the conventional understanding of this material, where A- and B-excitons are independently reflected on the optical responses, two branches are exotically coupled via radiation depending on the film thickness. Due to this effect, particular modes monopolize radiative correction and obtain ultrashort radiative decay times as shown in Fig. 1. In order to maximize the functions of these modes, ultrashort pulse excitation is considered to be effective. In this report, we theoretically demonstrate nonlinear optical responses by pulse excitation of these peculiar excitonic resonances, and find that the pulse width comparable to the radiative decay times of such modes is highly effective to enhance the integrated intensity of the nonlinear signal.

[1] M. Ichimiya *et al*., Phys. Rev. Lett. **103**, 257401 (2009).

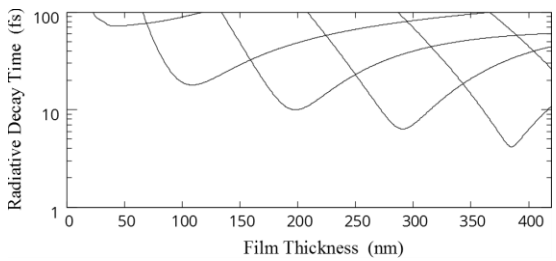

Figure 1 Film thickness dependence of radiative decay times of exciton-radiation coupled modes.