Summary of thesis: Nonlocal optical response of confined excitons in Cu_2O thin films beyond the long-wavelength approximation

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When atoms are in a coherently excited state, they cooperatively emit photons very rapidly. This phenomenon is referred to as superradiance, which was predicted by Dicke in 1954. The radiative rate of superradiance is proportional to the number, N, of atoms and the radiative intensity changes proportionally to N^2. Excitons (Coulomb-bound states of electrons and holes) confined in high-quality microcrystals or thin films can be regarded as a coherent excited state of atoms, which their center-of-mass motion wavefunction coherently spreading throughout the crystal. In this case, the sample size d corresponds to the number of atoms, and the radiative rate varies proportionally to d, according to the nonlocal response to the light. This phenomenon called "excitonic superradiance" was demonstrated by the group of Tokizaki *et al* by using CuCl microcrystals in 1989. However, the experiment was performed only with the sample sizes for which long-wavelength approximation (LWA) is satisfied. Despite interesting predictions, such as extremely fast photoresponse, selective excitation of forbidden transitions, and a crossover to the polariton (a mixed state of light and exciton), experiments on the size dependence of the nonlocal optical response have not been realized.

First, I performed the demonstrative experiment of excitonic superradiance beyond the LWA. The problem is how the radiative rate of confined excitons changes with the sample size not satisfying the LWA. I focused on the 2p state of the yellow excitons in Cu_2O, in which the radiative rate is smaller than the nonradiative rate by five orders of magnitude. The radiative rate increased in proportion to the film thickness with oscillation as shown by the squares in Fig. 1, until it saturated at the thickness of 640 nm. The behavior was found to be reproduced by calculations using a nonlocal theory, when the finite coherence length is incorporated.

Second, I examined the problem whether the radiative rate increases infinitely by the excitonic superradiance, if there is no limit on the coherence length. Several theories for this problem claim the existence of crossover to exciton-polaritons at a certain sample size. However, this phenomenon has not been observed experimentally for more than 20 years because a required large coherence length of several to tens of μm was not achieved. I focused on the blue exciton in Cu_2O having a very strong radiative interaction, to reduce the crossover thickness to a measurable size. Figure 2 shows qualitatively different thickness dependences below/above the expected crossover thickness of 177 nm (the vertical dotted line). Considering the effect of mixing with higher-order energy levels to the crossover theory, I successfully reproduced the experimental results. This is the first observation of crossover from excitonic superradiance to exciton-polaritons.

To summarize, the thickness dependence of the transmission, reflection and photoluminescence spectra was investigated for the Cu_2O thin film, in which LWA does not hold. We found the increase in the radiative rate due to excitonic superradiance and the limit of excitonic superradiance due to crossover to polariton. The manipulation of the light-matter interaction using the center-of-mass motion wavefunction of the confined excitons may give an impact to the deep understanding and vast application of the superradiant states.

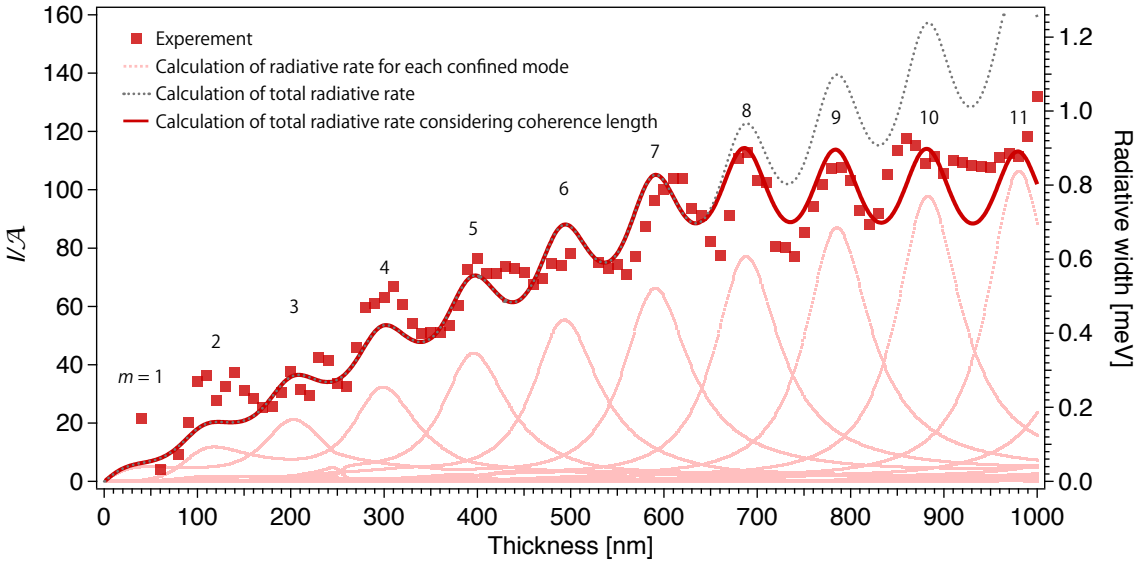

Figure 1: Thickness dependence of the radiative rate of the 2p exciton of yellow series in Cu_2O thin film. Squares are experimental results obtained from the analysis of PL spectra. The light red lines are calculation results for each confinement mode, m, and the dotted line is the sum. The red solid line represents the calculation result considering the finite coherence length.

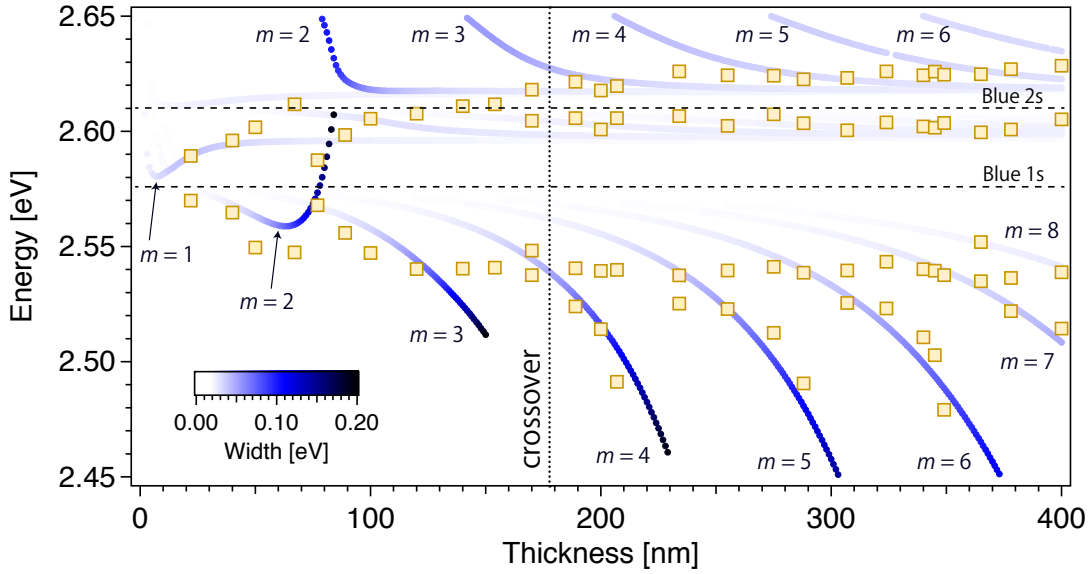

Figure 2: Thickness dependence of the resonance energy near the uncoupled blue exciton energy in Cu_2O thin film. Squares are experimental results obtained from the analysis of absorption spectra. Blue points are the calculation results of the resonance energy for each confinement mode, m.