Up-conversion in photoluminescence caused by radiation-mediated couplings between excitonic waves

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The light-matter coupling in nanostructures can be controlled by choosing the sample size because of the coherence of excited states over the entire volume. If the center-of-mass (c.m.) wavefunctions of excitons coherently spread across the whole crystal beyond the long wavelength approximation (LWA) regime, nonlocal correlation between the excitonic wave and the light wave clearly appears, and further the radiation-mediated couplings between different excitonic waves are enhanced. Recently, it has been revealed that the states generated by the wave-wave couplings can be monitored through the photoluminescence spectra [1]. The signals from higher energy levels of exciton-photon coupled modes including optically forbidden ones in the framework of the LWA have been observed for thin films with the thickness of a few hundreds nanometers, which is contrary to the conventional picture of photoluminescence.

In the present work, we develop a theoretical method to elucidate the above mentioned peculiar photoluminescence beyond the LWA regime by fully taking account of spatial interplay between the excitonic wave and the radiation wave. By means of this formalism, we calculate the photoluminescence spectra, and successfully reproduce the essential features of observed photoluminescence spectra beyond the LWA size regime. Furthermore, we find a new mechanism of up-converted photoluminescence based on the large radiation-mediated interaction between different excitonic waves. The study of the photoluminescence spectra for various film thickness and incident energy reveals that some portion of incident energy is up-converted according to the parity selection rule for the center-of-mass wavefunctions of excitons depending on the thickness. The underlying mechanism is based on neither multi-photon processes [2] nor on phonon-assisted processes [3]. We also discuss that the up-converted signal can be found in the results in Ref. [1].

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