

Radiative decay theory: What suppresses exciton superradiance?

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Received XXXX, revised XXXX, accepted XXXX

Published online XXXX

PACS 76.20.+q, 78.67.-n, 71.35.-y, 71.36.+c

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We have theoretically studied the exciton superradiance which occurs under a phase-match between the exciton center-of-mass motion and the radiation field in a film with thickness over a light wavelength. With continuously increasing the film thickness, the radiative decay rates of excitons are calculated from extreme values of exciton correlation functions renormalizing the exciton-photon interaction in the film with finite thickness. Due to a consideration of intra- and inter-state retarded interactions, in the simplest system, we have derived the correct suppression thickness of the exciton superradiance. The correctness is verified from the general suppression condition derived from the polariton dispersion relation consisting of complex frequency and wavenumber for finite thickness.

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1 Introduction In order to obtain strong and coherent responses from nonlinear optical processes in a condensed matter, the radiative decay of elementary excitations should generally be rapid as compared to dephasing processes at their resonance conditions. In particular, the exciton superradiance, a size enhancement of the radiative decay rate of excitons, has been studied theoretically and experimentally [1–9]. In a crystal where the exciton center-of-mass motion is confined, their radiative decay rate gets larger with increasing the crystal size because of the expansion of interaction area between excitons and the radiation field [1–6]. Fig. 1 shows a thickness dependence of the radiative decay rate of excitons whose center-of-mass motion is confined in a CuCl film. The decay rate of the lowest exciton state increases with increasing thickness until about 80 nm, a half light wavelength in CuCl, but after that it reversely decreases. The origin of this superradiance suppression is the phase mismatch between the exciton center-of-mass motion and the radiation field [5, 6, 8]. In this manner, the decay rate of the higher center-of-mass motion state are maximized at their phase-matching thicknesses, and the maximum value increases with increasing thickness [7, 9, 10] as seen in Fig. 1. This is also called exciton superradiance, and should realize a rapid radiative decay faster than various dephasing processes even at the resonance conditions. In particular, a rapid decay rate

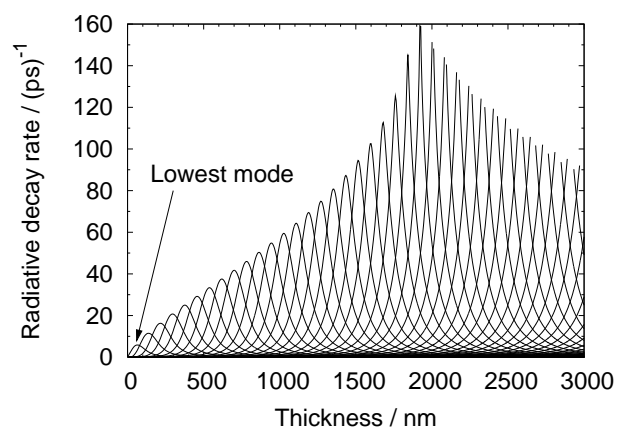


Figure 1 Thickness dependence of radiative amplitude-decay rates of excitons weakly confined in a CuCl film.

about 100 fs has recently been observed in the degenerate four-wave mixing in a CuCl film with thickness of a few hundred nanometers [11], and it is considered as a radiative decay enhanced by the exciton superradiance. Although it is usually considered that the size enhancement of the radiative decay rate is suppressed due to the dephasing processes of excitons, there is a significant question:

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What suppresses the exciton superradiance if the radiative decay rate is enhanced beyond any dephasing rates of excitons?

As an answer for this question, Knoester predicted [5] and Björk et al. theoretically demonstrated [7] that the superradiance is suppressed over a particular thickness (about $2 \mu\text{m}$ in Fig. 1) without considering any dephasing processes, and after that the decay rate of the phase-matching excitons inversely proportional to the thickness as the same manner of the polariton picture of radiative decay, where the decay time is proportional to the time-of-flight (thickness divided by group velocity). Further, Björk et al. also showed that the superradiance suppression can be interpreted as a crossover of the exciton-photon coupled modes from exciton-like (superradiant) and photon-like modes to the upper and lower branch polaritons. However, their calculation considered the retarded interaction between the same exciton states but not between the different states. Therefore, the crossover thickness may be different from the correct one, because the inter-state retarded interaction must be considered under the strong exciton-photon interaction at the superradiance. On the other hand, the inter-state retarded interaction has been considered in Refs. [8–10]. In particular, Ajiki discussed the crossover and showed that the radiative decay rate decreases with increasing the crystal size if the crystal becomes larger than a particular size [9]. However, since his calculation was performed under a pole approximation, the physical picture of the crossover was not clear. The purpose of the present paper is to grasp a correct physical picture of the superradiance suppression by means of a rigorous calculation method.

2 Calculation method We consider a material where the translational symmetry is broken in the z direction, and discuss the radiative decay of s-polarized excitons whose center-of-mass is confined in a finite region. We suppose a background system characterized by dielectric function $\varepsilon_{\text{bg}}(z, \omega)$ and a resonant contribution from excitons inducing a current density $J(z, \omega)$. The Maxwell wave equation for the vector potential $A(z, \omega)$ is represented as

$$[(\partial^2/\partial z^2) + q^2(z, \omega)] A(z, \omega) = -\mu_0 J(z, \omega), \quad (1)$$

where $q^2(z, \omega) = \varepsilon_{\text{bg}}(z, \omega)\omega^2/c^2 - k_{\parallel}^2$ and k_{\parallel} is the in-plane wavenumber. We can rewrite this wave equation as

$$A(z, \omega) = A_0(z, \omega) - \mu_0 \int dz G(z, z', \omega) J(z', \omega), \quad (2)$$

where $A_0(z, \omega)$ is the homogeneous solution of Eq. (1), and $G(z, z', \omega)$ is the Green function satisfying

$$[(\partial^2/\partial z^2) + q^2(z, \omega)] G(z, z', \omega) = \delta(z - z'). \quad (3)$$

On the other hand, supposing the exciton-photon interaction as

$$\hat{H}_{\text{int}} = - \int dz \hat{A}(z) J(z), \quad (4)$$

the expectation value $A = \langle \hat{A} \rangle$ of the vector potential is given in the same form as Eq. (2) based on the linear response theory. Therefore, the retarded correlation function of \hat{A} in the background system identifies with the Green function satisfying Eq. (3) as discussed in Ref. [12]:

$$G(z, z', \omega) = \frac{-i}{\mu_0 \hbar} \int_0^{\infty} dt e^{i\omega t} \langle [\hat{A}(z, t), \hat{A}(z', 0)] \rangle. \quad (5)$$

Furthermore, it is also identical to the time-ordered correlation function of \hat{A} under the rotating wave approximation (RWA). Therefore, under the RWA, the correlation function of \hat{A} in the background system can be obtained by finding the Green function satisfying Eq. (3), which has already been known for general multilayer systems [13].

Next, we discuss the time-ordered correlation functions of excitons. For simplicity, we consider only one relative exciton motion with eigenfrequency ω_T , and denote the center-of-mass motion by index m . The excitonic current density is quantized as $\hat{J}(z) = \mathcal{J} \sum_m g_m(z) \hat{b}_m + \text{H.c.}$, where \hat{b}_m is the annihilation operator of an exciton in state m , and $g_m(z)$ is its center-of-mass wavefunction. The absolute value of the coefficient \mathcal{J} can be estimated by the longitudinal-transverse (LT) splitting energy as $\hbar\omega_{\text{LT}} = |\mathcal{J}|^2/\varepsilon_{\text{bg}}\varepsilon_0\omega_T^2$. From the interaction Hamiltonian (4), we obtain intra- and inter-state self-energies of excitons as

$$\begin{aligned} \Sigma_{m,m'}(\omega) &= \varepsilon_{\text{bg}}\omega_{\text{LT}}(\omega_T/c)^2 \\ &\times \int_{-\infty}^{\infty} dz dz' g_m^*(z) G(z, z', \omega) g_{m'}(z'), \quad (6) \end{aligned}$$

and the time-ordered correlation function tensor of excitons is derived as the inverse of the matrix whose elements are given as $(\omega - \Omega_m)\delta_{m,m'} - \Sigma_{m,m'}(\omega)$, where Ω_m is the bare exciton frequency including the center-of-mass kinetic energy of state m . The resonance frequency ω_{res} and the radiative amplitude-decay rate γ of exciton-photon coupled modes are respectively obtained from the real and imaginary parts of extreme values $\{\omega_{\text{res}} - i\gamma\}$ of the exciton correlation function tensor. The calculation of these extreme values is just identical to that of the self-sustaining modes discussed under the microscopic nonlocal theory [9–11, 14].

Based on the above calculation method, we can consider the intra- and inter-state retarded interactions through the self-energies (6). Further, in the present paper, we numerically calculate the extreme values without any pole approximations in contrast to Ref. [9]. With regard to the RWA, it can be considered as a good approximation, because neither the frequency shift nor the radiative decay rate reaches only a few percents of the bare exciton frequency ω_T even at the maximum in the calculation of the present paper.

In Fig. 1, we consider a CuCl film with thickness d , and suppose the background dielectric constant $\varepsilon_{\text{bg}} = n_{\text{bg}}^2 = 5.59$ both inside and outside of the film. The wavefunctions

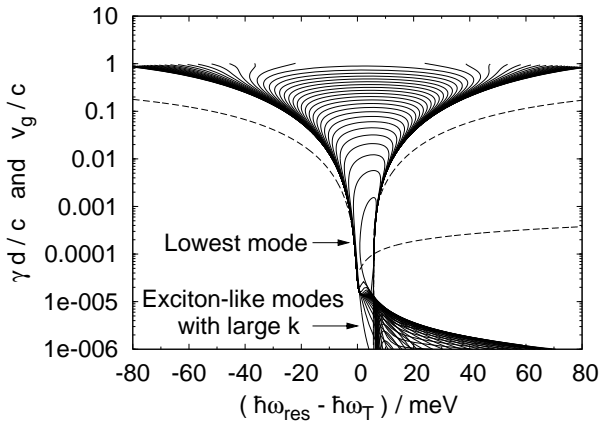


Figure 2 Frequency dependence of γd (thickness divided by radiative amplitude-decay time). The dashed lines are the group velocities of upper and lower branch polaritons in bulk CuCl.

of the exciton center-of-mass motion are sinusoidal functions whose amplitudes are zero at the surfaces: $g_m(z) = \sqrt{2/d} \sin(k_m z)$, $k_m = m\pi/d$, $m = 1, 2, \dots$ and $k_{\parallel} = 0$. The exciton translational mass is $m_{ex} = 2.3m_0$, where m_0 is the free electron mass. The bare exciton frequencies are given as $\hbar\omega_T = 3.2022$ eV and $\Omega_m = \omega_T + \hbar k_m^2 / 2m_{ex}$. The LT splitting energy is $\hbar\omega_{LT} = 5.7$ meV.

3 Calculation results In Fig. 1, we plot the radiative amplitude-decay rates γ with continuously changing the thickness d . Although any dephasing processes are not considered in the calculation, we can find that the exciton superradiance is suppressed at thickness over $2 \mu\text{m}$. Further, the behavior becomes discontinuous at the peak, and the peak value cannot be found in our numerical calculation. For the analysis of this suppression, we introduce a new value γd (thickness divided by radiative-amplitude decay time), and we find that γd reaches the vacuum light speed c at the crossover thickness, and after that the superradiance is suppressed with maintaining $\gamma d < c$. Although this behavior is just the same discussed in Ref. [7], due to the correct consideration of the inter-state retarded interaction in the present paper, the crossover condition is obtained as $\gamma d = c$, i.e., an apparent propagation speed of the polariton cannot exceed the vacuum light speed.

Fig. 2 shows the frequency dependence of γd by solid lines with continuously changing the thickness, and the dashed lines represent the group velocities v_g of the upper and lower branch polaritons in bulk CuCl. When we focus on a particular mode, ω_{res} shifts to the lower side and γ gets larger with increasing d until its phase-matching thickness, and around its thickness, ω_{res} suddenly flips to the higher side with maximizing γ . After that, ω_{res} gradually decreases to the band edge of the upper branch $\omega_T + \omega_{LT}$, and γ monotonally decreases. Although the maximum value of γ gradually increases with increasing d due to the

exciton superradiance, it is suppressed when γd reaches the vacuum light speed c , and after that γ of the phase-matching modes decreases with maintaining $\gamma d < c$. On the other hand, we can find that each γd has the same value at the same frequency before and after the superradiance. This indicates that in this phase-mismatching situation, γ obeys the polariton picture of the radiative decay [8]:

$$\gamma = (v_g/2d) \ln(1/R), \quad (7)$$

where R is the reflectance from inside to outside of the film. It is worth to note that, compared to Ref. [8], we must consider the Pekar's additional boundary condition (ABC) [15] to reproduce our calculation results from Eq. (7), because we consider the exciton center-of-mass wavefunctions whose amplitudes are zero at the surfaces. In the calculation of R , we use two complex wavenumbers \tilde{k}_i determined by the dispersion relation

$$\frac{c^2 \tilde{k}_i^2}{(\omega_{res} - i\gamma)^2} = \varepsilon_{bg} + \frac{\varepsilon_{bg} \omega_{LT}}{\Omega(\tilde{k}_i) - (\omega_{res} - i\gamma)}, \quad (8)$$

where $\Omega(\tilde{k}) = \omega_T + \hbar \tilde{k}^2 / 2m_{ex}$. Although γd depends almost only on the frequency ω_{res} just before and after the phase-matching thickness, γd of exciton-like modes with large wavenumber depends also on d because of the thickness dependence of R due to the exciton center-of-mass confinement.

4 Discussion We analyze the superradiance suppression based on the dispersion relation (8). For a given \tilde{k} , we can obtain three $\omega_{res} - i\gamma$ satisfying Eq. (8). One solution is unphysical with a negative frequency, and the other two correspond to the coupled modes in the exciton-photon system, or the upper and lower branch polaritons for bulk system. However, in contrast to the bulk calculation, there is a task to determine the imaginary part of \tilde{k} , the uncertainty of the wavenumber in a film with finite thickness, as discussed in Ref. [8]. From the exact calculation based on the correlation function method, we verify that the complex wavenumber is written as $\tilde{k} = k - i\alpha/d$, and α obeys

$$\alpha = (1/2) \ln(1/R). \quad (9)$$

This can be derived from a condition for a self-sustaining mode in the film with considering the ABC. Further, when the polariton picture is valid, we can derive a relation $\gamma = \alpha v_g / d$ from Eq. (8). Then, Eq. (9) is also obtained by a comparison with Eq. (7), and d/α can be considered as an effective thickness for the polariton. As a result, by using Eq. (9), we can obtain two physical solutions of Eq. (8) for a given real wavenumber k by a successive calculation.

As discussed in Ref. [7], we analyze the crossover of the exciton-photon coupled modes. Fig. 3 shows the dispersion relation and frequency dependence of γd for thicknesses 1.6, 2, and 3 μm . The extreme values of the exciton correlation functions are plotted with symbols and the two

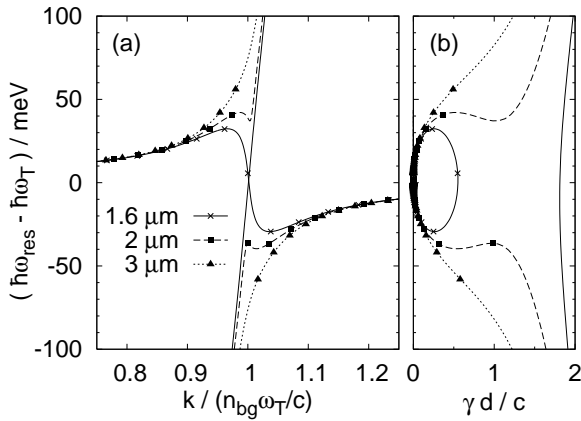


Figure 3 (a) Dispersion relation and (b) frequency dependence of γd . The lines are solutions of Eq. (8) without the ABC for a given real wavenumber k . The symbols are calculated as the extreme values of the exciton correlation functions.

solutions of Eq. (8) for given k are plotted with lines. However, in the latter calculation, we do not consider neither the k -selection rule derived from the self-sustaining condition nor the ABC in order to show the continuous behavior with respect to k . For thickness of $1.6 \mu\text{m}$, where the exciton superradiance is maintained, the two solutions of Eq. (8) respectively correspond to an exciton-like mode with $\gamma d < c$ and a photon-like one with $\gamma d > c$, and only the former is found by the correlation function method. This is because $\omega_{\text{res}} - i\gamma$ of the photon-like modes are divergent solutions for finding the extreme values of exciton correlation functions. On the other hand, increasing the thickness, γd of the two modes close to each other reaching to the vacuum light speed c . After that, as seen at $3 \mu\text{m}$, the two solutions split to the upper and lower branches, and the exciton superradiance is suppressed as discussed in Ref. [7]. Even at this bulk-like thickness, only the exciton-like modes are found by the correlation function method, because the photon-like solutions are divergent. The reason why we cannot find the peak values in Fig. 1 is the same.

The crossover between the exciton/photon-like modes and the polariton ones can be understood from Eq. (8). At the superradiance situation, i.e., $k = n_{\text{bg}}\omega_T/c$ and $\omega_{\text{res}} \simeq \omega_T$, the real part of Eq. (8) can be approximated as

$$\gamma^2 - (c\alpha/n_{\text{bg}}d)\gamma + \omega_T\omega_{\text{LT}}/2 = 0. \quad (10)$$

When the film is thin enough compared to the crossover thickness, we obtain two solutions of this equation: $\gamma = (n_{\text{bg}}\omega_T d/2c\alpha)\omega_{\text{LT}}$ for the exciton-like (superradiant) mode, and $\gamma = c\alpha/n_{\text{bg}}d$ for the photon-like mode. Since the former is proportional to d and the latter is to d^{-1} , these values gradually close to each other with increasing the thickness and reach to $\gamma = c\alpha/2n_{\text{bg}}d$, which is the degenerate solution of Eq. (10). This crossover condition can be rewritten as $(d/\alpha)/\gamma^{-1} = c/2n_{\text{bg}}$, and means that we obtain the crossover when the effective thickness d/α divided

by the radiative amplitude-decay time γ^{-1} reaches $c/2n_{\text{bg}}$, the polariton group velocity at phase-matching wavenumber $k = n_{\text{bg}}\omega_T/c$. In the simplest case $\varepsilon_{\text{bg}}(z) = \varepsilon_{\text{bg}}$ considered in the present paper, we obtain $\alpha \simeq 2n_{\text{bg}}$ for the phase-matching modes at thickness over $2 \mu\text{m}$. Therefore, we obtain the crossover condition as $\gamma d = c$ in the present paper. In more general cases, the crossover speed becomes smaller than c because of the increment of the effective thickness d/α due to the enhancement of the multiple reflection inside of the film.

5 Summary We calculate the resonance frequency and radiative decay rate of exciton-photon coupled modes in a film with finite thickness. The calculation is based on the exciton correlation function tensor renormalizing the exciton-photon interaction. Even if we do not consider any dephasing processes, the exciton superradiance is naturally suppressed, and we find the suppression condition as $\gamma d = c$, which is correctly obtained under the consideration of the inter-state retarded interaction. Further, we reproduce the resonance frequency and radiative decay rate from simple equations (8) and (9), and discuss the crossover of the exciton-photon coupled modes from these equations.

Acknowledgements The authors are grateful to Dr. H. Ajiki for helpful discussions. This work was partially supported by the Japan Society for the Promotion of Science (JSPS); a Grant-in-Aid for Creative Science Research, 17GS1204, 2005; and JSPS Research Fellowships for Young Scientists.

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