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Quantum description of exciton-light coupled states for a thin film

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The superposition coefficients of the coupled states of a photon and an exciton confined in a thin film are provided using Fano's method [U. Fano, Phys. Rev. **124**, 1866 (1961)], in which the exciton interacts with continuum photon states. The coupled states are classified into leaky and surface modes, the latter of which are investigated with a focus on the dispersion relation, the exciton component, and their film-thickness dependence. The obtained coupled states enable us to study exciton-mediated optical properties that cannot be described by a semiclassical scheme, such as photoluminescence and the radiative decay of a biexciton. As applications of the coupled states, we calculate the population decay dynamics and photoluminescence spectra of an exciton in a thin film.

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I. INTRODUCTION

Various types of exciton-light coupled states have been predicted and observed since the seminal work by Hopfield on the bulk exciton polariton [1]. For a quantum well (QW) or a semiconductor thin film embedded in a microcavity, the exciton-light coupled state is called a cavity polariton [2–5]. The dispersions of the bulk and cavity polaritons consist of upper and lower branches showing anticrossing behavior, and the level splitting of the cavity polariton is called a vacuum Rabi splitting. Recently, the vacuum Rabi splitting has been observed for a single quantum dot (QD) in a microcavity [6–8], and the exciton-light coupled states are called dressed states.

These exciton-light coupled states are intermediate states in the radiative decay processes of a biexciton, and polarization entangled photon pairs are generated through these processes [9–12]. The entangled photons are the key components of quantum computation [13], quantum teleportation [14], and quantum key distribution [15]. The cavity polaritons and dressed states provide the characteristic features of entangled photons and their generation conditions [16–23], which have been studied using the quantum description of the coupled states. The quantum description enables us to calculate the other optical properties of excited states in matter, such as photoluminescence and the radiative lifetime of a biexciton. In a semiclassical scheme, the induced exciton polarization causing light emission is calculated as the expectation value with respect to the ground state. Consequently, the photoemission processes starting from the exciton cannot be described in the semiclassical scheme, and a full-quantum treatment is indispensable.

Among the exciton–photon coupled states, an exciton polariton in a bulk semiconductor consists of an exciton and a photon with the same wave vector, and a cavity polariton or dressed state in a microcavity consists of an exciton and a single-mode cavity photon. For these coupled states, the energies and superposition coefficients of the exciton and photon can be calculated by diagonalizing a small-size matrix. For a semiconductor film, however, confined excitons interact with a photon having any wave-vector component q_z in the thickness direction (z direction). Namely, the exciton-light coupled state consists of discrete exciton states and continuous photon modes with respect to q_z. So far, such an exciton-light coupled state has not been obtained in a semiconductor film.

In this paper, the exciton and photon components of the exciton-light coupled states in the film structure are derived. The coupled states are designated by a wave vector $\boldsymbol{q}_\parallel$ parallel to the film surface because of the translational symmetry in this direction (x and y directions). Each coupled state with $\boldsymbol{q}_\parallel$ is classified into leaky and surface modes, the latter of which is also called a surface exciton polariton. Both leaky and surface modes contain continuum photon states and discrete exciton states, where only the lowest exciton state is considered in this study. The theory for the eigenstates of the interacting discrete and continuum states has been developed by Fano [24] to analyze the asymmetric peak of a discrete autoionization level in continuous spectra, which is observed in forward inelastic electron scattering by He. The exciton-light coupled states are obtained using this theory.

This paper is organized as follows. After introducing the Hamiltonian of the exciton-light coupled states in Sec. II A, the coupled states of the leaky and surface modes are described in Secs. II B and II C, respectively, and the thickness dependence of the energy dispersion and the exciton component of the surface modes are numerically investigated in the latter section. In Sec. III, a quantum description of the leaky modes is applied to the calculations of the population decay dynamics and photoluminescence spectra of an exciton in a semiconductor film. A summary and conclusion are given in Sec. IV.

II. EXCITON-LIGHT COUPLED STATES

A. Hamiltonian

We assume that the background dielectric constant outside the film is the same as that inside, and is denoted by ϵ_b. In addition, we consider the exciton-light coupled states with the s polarization, which allows us to neglect the polarization degree of freedom for light and exciton in the following calculations. In this situation, the electric field of light is quantized as

$$\hat{E}(\boldsymbol{r}) = \sum_{\boldsymbol{q}_\parallel} \int_{-\infty}^{\infty} dq_z i \sqrt{\frac{\hbar\omega_{\boldsymbol{q}}}{S\epsilon_b}} e^{-i\boldsymbol{q}\cdot\boldsymbol{r}} (a_{-\boldsymbol{q}} - a_{\boldsymbol{q}}^\dagger), \quad (1)$$

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where S is the surface area of the film, $\hbar\omega_{\boldsymbol{q}}$ is the one-photon energy with wave vector $\boldsymbol{q}$:

$$\hbar\omega_{\boldsymbol{q}} = \frac{\hbar c}{\sqrt{\epsilon_b}}\sqrt{q_\parallel^2 + q_z^2}, \tag{2}$$

and $a_{\boldsymbol{q}}$ is the annihilation operator of the photon. The continuous wave vectors $\boldsymbol{q}_\parallel$ in the x and y directions are discretized by enclosing the photons in a square with S and by imposing periodic boundary conditions, while wave vectors q_z in the z direction remain continuous. The commutation relations are given by

$$[a_{\boldsymbol{q}}, a_{\boldsymbol{q}'}^\dagger] = \delta_{\boldsymbol{q}_\parallel,\boldsymbol{q}'_\parallel}\delta(q_z - q'_z), \tag{3}$$

$$[a_{\boldsymbol{q}}, a_{\boldsymbol{q}'}] = [a_{\boldsymbol{q}}^\dagger, a_{\boldsymbol{q}'}^\dagger] = 0. \tag{4}$$

Here, we consider an inorganic semiconductor thin film whose electronic states near the surface are not appreciably modified. In addition, the film thickness is larger than the Bohr radius of an exciton in bulk (weak confinement regime). In this situation, the exciton wave function is approximately expressed as $\Psi(\boldsymbol{\rho},\boldsymbol{r}) = \phi(\boldsymbol{\rho})G(\boldsymbol{r})$, where $\phi(\boldsymbol{\rho})$ and $G(\boldsymbol{r})$ are the wave functions of relative and center-of-mass (CM) motions, respectively. The transition polarization $\boldsymbol{d}(\boldsymbol{r})$ of an exciton is calculated as [25]

$$\boldsymbol{d}(\boldsymbol{r}) = \boldsymbol{\mu}_{cv}\phi(0)G(\boldsymbol{r}), \tag{5}$$

with

$$\boldsymbol{\mu}_{cv} = \int d\boldsymbol{r}\, q\varphi_c^*(\boldsymbol{r})\varphi_v(\boldsymbol{r})\boldsymbol{r}, \tag{6}$$

where $\varphi_c(\boldsymbol{r})$ and $\varphi_v(\boldsymbol{r})$ are the Wannier functions of conduction and valence bands, respectively. We define the intensity of the transition polarization as $\mu = |\boldsymbol{\mu}_{cv}\phi(0)|$. In the weak confinement regime, μ in the film is approximately the same as that in bulk, and is related to the longitudinal and transverse splitting energy $\Delta_{\rm LT}$ in bulk. The relationship is obtained as follows: in bulk with volume V, the wave function of the CM motion is given by $G(\boldsymbol{r}) = \exp(i\boldsymbol{k}\cdot\boldsymbol{r})/\sqrt{V}$, and the transition polarization is written as $\boldsymbol{d}(\boldsymbol{r}) = \boldsymbol{\mu}\exp(i\boldsymbol{k}\cdot\boldsymbol{r})/\sqrt{V}$. There are longitudinal ($\boldsymbol{\mu} \parallel \boldsymbol{k}$) and transverse ($\boldsymbol{\mu} \perp \boldsymbol{k}$) excitons, and the energy of the longitudinal exciton is higher than that of the transverse one by $\Delta_{\rm LT}$. This energy difference comes from the electron-hole (e-h) exchange interaction, which has the same form as the Coulomb interaction between polarization charge $-\nabla\cdot\boldsymbol{d}(\boldsymbol{r})$ [26]. The e-h exchange energy of the transverse exciton is zero, while the exchange energy of the longitudinal exciton is $4\pi\mu^2/\epsilon_b$. Therefore we have the relationship $\mu^2 = \epsilon_b\Delta_{\rm LT}/4\pi$ [25,27].

The wave function of the CM motion $G(\boldsymbol{r})$ is obtained by imposing boundary conditions at the surfaces. The film thickness d is sufficiently less than the light wavelength of the exciton resonance; thus we can safely restrict ourselves to the lowest confinement of the CM motion, and $G(\boldsymbol{r})$ is given by

$$G(\boldsymbol{r}) = \sqrt{\frac{2}{Sd}}e^{i\boldsymbol{k}_\parallel\cdot\boldsymbol{r}_\parallel}\sin\left(\frac{\pi}{d}z\right), \tag{7}$$

where $\boldsymbol{k}_\parallel$ and $\boldsymbol{r}_\parallel$ are the wave vector and position parallel to the surface, respectively. Neglecting the polarization degree of freedom and using Eq. (5), the transition polarization operator $\hat{d}$ is written as

$$\hat{d}(\boldsymbol{r}) = \mu\sqrt{\frac{2}{Sd}}\sum_{\boldsymbol{k}_\parallel} e^{i\boldsymbol{k}_\parallel\cdot\boldsymbol{r}_\parallel}\sin\left(\frac{\pi}{d}z\right)(b_{\boldsymbol{k}_\parallel} + b_{-\boldsymbol{k}_\parallel}^\dagger), \tag{8}$$

where $b_{\boldsymbol{k}_\parallel}$ is the annihilation operator of the exciton.

The interaction between the exciton and photon is given by

$$\begin{aligned} H_{\rm int} &= \int d\boldsymbol{r}\hat{d}(\boldsymbol{r})\hat{E}(\boldsymbol{r}) \\ &= \sum_{\boldsymbol{q}_\parallel}\int_{-\infty}^{\infty} dq_z g_{\boldsymbol{q}}[b_{\boldsymbol{q}_\parallel}(a_{\boldsymbol{q}}^\dagger - a_{-\boldsymbol{q}}) + b_{-\boldsymbol{q}_\parallel}^\dagger(a_{\boldsymbol{q}}^\dagger - a_{-\boldsymbol{q}})], \end{aligned} \tag{9}$$

where

$$g_{\boldsymbol{q}} = -i\sqrt{\frac{\Delta_{\rm LT}\hbar\omega_{\boldsymbol{q}}}{2\pi d}}\int_0^d dz e^{-iq_z z}\sin\left(\frac{\pi}{d}z\right). \tag{10}$$

Because the spatial variation in the electric field is taken into account in $g_{\boldsymbol{q}}$, the following calculations are beyond the long-wavelength approximation (LWA), which is valid for $d \ll \lambda$, where λ is the wavelength of light. The total Hamiltonian is given by

$$H = \sum_{\boldsymbol{q}_\parallel}\int_{-\infty}^{\infty} dq_z\hbar\omega_{\boldsymbol{q}}a_{\boldsymbol{q}}^\dagger a_{\boldsymbol{q}} + \sum_{\boldsymbol{k}_\parallel} E_{\boldsymbol{k}_\parallel}^{\rm ex} b_{\boldsymbol{k}_\parallel}^\dagger b_{\boldsymbol{k}_\parallel} + H_{\rm int}, \tag{11}$$

where $E_{\boldsymbol{k}_\parallel}^{\rm ex}$ is the energy of the lowest exciton with $\boldsymbol{k}_\parallel$ and is given by

$$E_{\boldsymbol{k}_\parallel}^{\rm ex} = E_{\rm ex} + \frac{\hbar^2}{2M_{\rm ex}}\left[k_\parallel^2 + \left(\frac{\pi}{d}\right)^2\right], \tag{12}$$

where $E_{\rm ex}$ and $M_{\rm ex}$ are the band-edge energy and the translational mass of an exciton, respectively

The eigenstates of the Hamiltonian H (exciton-light coupled states) are designated by $\boldsymbol{q}_\parallel$, while q_z is not a good quantum number because of the lack of the translational symmetry in the z direction. For a fixed $\boldsymbol{q}_\parallel$, the photon energy continuously distributes greater than or equal to $(\hbar c/\sqrt{\epsilon_b})q_\parallel$ as found from Eq. (2). The exciton-light coupled states consisting of such photons have continuous eigenenergy $\mathcal{E} \geqslant (\hbar c/\sqrt{\epsilon_b})q_\parallel$, and the coupled states expand outside the semiconductor film. We call these coupled states leaky modes. By introducing a wave vector $q_{\mathcal{E}}$ defined as

$$q_{\mathcal{E}} = \frac{\sqrt{\epsilon_b}}{\hbar c}\mathcal{E}, \tag{13}$$

the leaky-mode condition is expressed as $q_{\mathcal{E}} \geqslant q_\parallel$. For $q_{\mathcal{E}} < q_\parallel$, on the other hand, q_z satisfying $\mathcal{E} = \hbar\omega_{\boldsymbol{q}}$ becomes imaginary; thus, the coupled states exponentially decay outside the film and are called surface modes.

B. Leaky modes

Because the leaky modes have continuous energy for a fixed $\boldsymbol{q}_\parallel$ as mentioned before, their states are uniquely designated by $\boldsymbol{q}_\parallel$ and $\mathcal{E}$. To obtain the eigenstates of the leaky modes with the use of Fano's prescription [24], we introduce the annihilation operator $A_{\boldsymbol{q}_\parallel}(\mathcal{E})$ of the leaky modes. The leaky modes are represented as superposition of photons with various q_z and the exciton; thus, $A_{q_\parallel}(\mathcal{E})$ is expressed as

$$A_{q_\parallel}(\mathcal{E}) = \int_{-\infty}^{\infty} dq_z[\alpha_q(\mathcal{E})a_q + \alpha'_q(\mathcal{E})a^\dagger_{-q}] + \beta_{q_\parallel}(\mathcal{E})b_{q_\parallel}, \quad (14)$$

where $\alpha_q(\mathcal{E})$, $\alpha'_q(\mathcal{E})$, and $\beta_{q_\parallel}(\mathcal{E})$ are superposition coefficients. In the expression (14) the rotating-wave approximation (RWA) is applied, and the counter-rotating term $\beta'_{q_\parallel}(\mathcal{E})b^\dagger_{-q_\parallel}$ is neglected. The second term including $\alpha'_q(\mathcal{E})$, which corresponds to the counter-rotating term of a photon, is usually neglected in the RWA. However, we also take into account the second term, because the integration in $F_{q_\parallel}(\mathcal{E})$ [see Eq. (25)] can be analytically performed owing to the counter-rotating term. Because both a_q and $a^\dagger_{-q}$ are included in $A_{q_\parallel}$, the Maxwell's equations are rigorously treated in the following calculations.

Since the leaky modes are eigenstates of the Hamiltonian in Eq. (11), the operator $A_{q_\parallel}(\mathcal{E})$ satisfies the eigenoperator equation [28]:

$$i\hbar\frac{dA_{q_\parallel}(\mathcal{E})}{dt} = [A_{q_\parallel}(\mathcal{E}),H] = \mathcal{E}A_{q_\parallel}(\mathcal{E}). \quad (15)$$

It is easily found from this equation that the time evolution of $A_{q_\parallel}(\mathcal{E})$ is described by only a dynamic phase $\exp[-i(\mathcal{E}/\hbar)t]$, which indicates $A_{q_\parallel}(\mathcal{E})$ to be the eigenoperator. We impose the standard commutation relations:

$$[A_{q_\parallel}(\mathcal{E}),A^\dagger_{q'_\parallel}(\mathcal{E}')] = \delta_{q_\parallel,q'_\parallel}\delta(\mathcal{E}-\mathcal{E}'), \quad (16)$$

which represent the orthonormality conditions for the leaky modes. The superposition coefficients $\alpha_q(\mathcal{E})$, $\alpha'_q(\mathcal{E})$, and $\beta_{q_\parallel}(\mathcal{E})$ can be determined from Eqs. (15) and (16).

By substituting Eq. (14) into Eq. (15) and taking the commutator with a_q, $a^\dagger_{-q}$, and $b_{q_\parallel}$, we obtain the following equations:

$$\hbar\omega_q\alpha_q(\mathcal{E}) + g^*_q\beta_{q_\parallel}(\mathcal{E}) = \mathcal{E}\alpha_q(\mathcal{E}), \quad (17)$$

$$-\hbar\omega_q\alpha'_q(\mathcal{E}) - g^*_q\beta_{q_\parallel}(\mathcal{E}) = \mathcal{E}\alpha'_q(\mathcal{E}), \quad (18)$$

$$E^{\rm ex}_{q_\parallel}\beta_{q_\parallel}(\mathcal{E}) + \int_{-\infty}^{\infty} dq_z g_q[\alpha_q(\mathcal{E}) + \alpha'_q(\mathcal{E})] = \mathcal{E}\beta_{q_\parallel}(\mathcal{E}), \quad (19)$$

where we use $g_{-q} = -g^*_q$ and $\hbar\omega_{-q} = \hbar\omega_q$.

From Eq. (17), $\alpha_q(\mathcal{E})$ is obtained as

$$\alpha_q(\mathcal{E}) = \left[\frac{1}{\mathcal{E}-\hbar\omega_q} + z_{q_\parallel}(\mathcal{E})\delta(\mathcal{E}-\hbar\omega_q)\right]g^*_q\beta_{q_\parallel}(\mathcal{E}), \quad (20)$$

where the function $z_{q_\parallel}(\mathcal{E})$ is determined later. Using the property of the delta function, $\alpha_q(\mathcal{E})$ is rewritten as

$$\alpha_q(\mathcal{E}) = \frac{g^*_q\beta_{q_\parallel}(\mathcal{E})}{\mathcal{E}-\hbar\omega_q} + p_\mathcal{E} z_{q_\parallel}(\mathcal{E})\sum_{s=\pm} g_q\beta_{q_\parallel}(\mathcal{E})\delta(q_z - sq_{\mathcal{E}z}), \quad (21)$$

where $q_{\mathcal{E}z} = \sqrt{q^2_\mathcal{E} - q^2_\parallel}$ and

$$p_\mathcal{E} = \frac{\sqrt{\epsilon_b}}{\hbar c}\frac{q_\mathcal{E}}{q_{\mathcal{E}z}}. \quad (22)$$

From Eq. (18), we have

$$\alpha'_q(\mathcal{E}) = -\frac{g^*_q\beta_{q_\parallel}(\mathcal{E})}{\mathcal{E}+\hbar\omega_q}. \quad (23)$$

By substituting Eqs. (20) and (23) into Eq. (19), we obtain

$$\left[E^{\rm ex}_{q_\parallel} + F_{q_\parallel}(\mathcal{E}) + \frac{1}{\pi}\Gamma_{q_\parallel}(\mathcal{E})z_{q_\parallel}(\mathcal{E})\right]\beta_{q_\parallel}(\mathcal{E}) = \mathcal{E}\beta_{q_\parallel}(\mathcal{E}), \quad (24)$$

where

$$F_{q_\parallel}(\mathcal{E}) = P\int_{-\infty}^{\infty} dq_z\frac{2\hbar\omega_q}{\mathcal{E}^2-(\hbar\omega_q)^2}|g_q|^2, \quad (25)$$

which is analytically expressed in Appendix A, and

$$\Gamma_{q_\parallel}(\mathcal{E}) = 2\pi p_\mathcal{E}|g_{q_\mathcal{E}}|^2 = \Delta_{\rm LT}\frac{(2\pi)^2}{d^3}\frac{q^2_\mathcal{E}}{q_{\mathcal{E}z}}\frac{\cos^2(q_{\mathcal{E}z}d/2)}{[q^2_{\mathcal{E}z}-(\pi/d)^2]^2}, \quad (26)$$

where $|g_{q_\mathcal{E}}|^2 \equiv |g_{(q_\parallel,sq_{\mathcal{E}z})}|^2$ is independent of s. It is noted that $\Gamma_{q_\parallel}(\mathcal{E})$ corresponds to the radiative width of the exciton with $q_\parallel$. In fact, Eq. (26) can be derived by evaluating the radiative lifetime of the exciton. We denote the state consisting of a photon with q and the material ground state by $|g;q\rangle$ and denote the exciton with $q_\parallel$ by $|X_{q_\parallel}\rangle$. From Fermi's golden rule, the radiative lifetime τ of the exciton is calculated as

$$\frac{1}{\tau} = \frac{2\pi}{\hbar}\int dq_z|\langle g;q|H_{\rm int}|X_{q_\parallel}\rangle|^2\delta(\mathcal{E}-\hbar\omega_q) = \frac{2\pi}{\hbar}\int dq_z|g_q|^2\delta(\mathcal{E}-\hbar\omega_q) = 2\frac{2\pi}{\hbar}p_\mathcal{E}|g_{q_\mathcal{E}}|^2. \quad (27)$$

From the relationship between τ and the radiative width Γ, i.e., $\tau = \hbar/(2\Gamma)$, Eq. (26) is derived. The function $z_{q_\parallel}(\mathcal{E})$ is obtained from Eq. (24) as

$$z_{q_\parallel}(\mathcal{E}) = \frac{\pi}{\Gamma_{q_\parallel}(\mathcal{E})}[\mathcal{E} - E^{\rm ex}_{q_\parallel}(\mathcal{E}) - F_{q_\parallel}(\mathcal{E})]. \quad (28)$$

Let us determine $\beta_{q_\parallel}(\mathcal{E})$. By substituting Eq. (14) into Eq. (16), we obtain

$$\int_{-\infty}^{\infty} dq_z[\alpha_q(\mathcal{E})\alpha^*_q(\mathcal{E}') - \alpha'_q(\mathcal{E})\alpha'^*_q(\mathcal{E}')] + \beta_{q_\parallel}(\mathcal{E})\beta^*_{q_\parallel}(\mathcal{E}') = \delta(\mathcal{E}-\mathcal{E}'). \quad (29)$$

From Eqs. (20) and (23), the sum of the first and second terms of Eq. (29) is calculated as

$$\int_{-\infty}^{\infty} dq_z[\alpha_q(\mathcal{E})\alpha^*_q(\mathcal{E}') - \alpha'_q(\mathcal{E})\alpha'^*_q(\mathcal{E}')] = \frac{1}{\pi}\Gamma_{q_\parallel}(\mathcal{E})|\beta_{q_\parallel}(\mathcal{E})|^2[\pi^2 + z^2_{q_\parallel}(\mathcal{E})]\delta(\mathcal{E}-\mathcal{E}') + \frac{\beta_{q_\parallel}(\mathcal{E})\beta^*_{q_\parallel}(\mathcal{E}')}{\mathcal{E}-\mathcal{E}'}\left\{E^{\rm ex}_{q_\parallel} + F_{q_\parallel}(\mathcal{E}') + \frac{1}{\pi}\Gamma_{q_\parallel}(\mathcal{E}')z_{q_\parallel}(\mathcal{E}') - \left[E^{\rm ex}_{q_\parallel} + F_{q_\parallel}(\mathcal{E}) + \frac{1}{\pi}\Gamma_{q_\parallel}(\mathcal{E})z_{q_\parallel}(\mathcal{E})\right]\right\}, \quad (30)$$

where the relationships

$$\frac{1}{(\mathcal{E}-\hbar\omega_{\boldsymbol{q}})(\mathcal{E}'-\hbar\omega_{\boldsymbol{q}})} = \pi^2\delta(\mathcal{E}-\mathcal{E}')\delta(\mathcal{E}-\hbar\omega_{\boldsymbol{q}}) + \frac{1}{\mathcal{E}-\mathcal{E}'}\left(\frac{1}{\mathcal{E}'-\hbar\omega_{\boldsymbol{q}}}-\frac{1}{\mathcal{E}-\hbar\omega_{\boldsymbol{q}}}\right), \quad (31)$$

and $\hbar\omega_{(\boldsymbol{q}_\parallel,sq_{\mathcal{E}z})}=\mathcal{E}$ are used. By substituting Eq. (24) into Eq. (30), we obtain

$$\int_{-\infty}^{\infty} dq_z[\alpha_{\boldsymbol{q}}(\mathcal{E})\alpha^*_{\boldsymbol{q}}(\mathcal{E}')-\alpha'_{\boldsymbol{q}}(\mathcal{E})\alpha'^*_{\boldsymbol{q}}(\mathcal{E}')] = \frac{1}{\pi}\Gamma_{\boldsymbol{q}_\parallel}(\mathcal{E})|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2\left[\pi^2+z^2_{\boldsymbol{q}_\parallel}(\mathcal{E})\right]\delta(\mathcal{E}-\mathcal{E}') - \beta_{\boldsymbol{q}_\parallel}(\mathcal{E})\beta^*_{\boldsymbol{q}_\parallel}(\mathcal{E}'). \quad (32)$$

Therefore Eq. (29) is calculated as

$$\frac{1}{\pi}\Gamma_{\boldsymbol{q}_\parallel}(\mathcal{E})|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2\left[\pi^2+z^2_{\boldsymbol{q}_\parallel}(\mathcal{E})\right]=1. \quad (33)$$

From Eq. (28), $|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2$ is expressed by a normalized Lorentz function:

$$|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2=\frac{1}{\pi}\frac{\Gamma_{\boldsymbol{q}_\parallel}(\mathcal{E})}{\left[\mathcal{E}-E^{\rm ex}_{\boldsymbol{q}_\parallel}-F_{\boldsymbol{q}_\parallel}(\mathcal{E})\right]^2+\Gamma^2_{\boldsymbol{q}_\parallel}(\mathcal{E})}. \quad (34)$$

The expression for $\alpha_{\boldsymbol{q}}(\mathcal{E})$ [Eq. (20)] is obtained using Eqs. (28) and (34), and that for $\alpha'_{\boldsymbol{q}}(\mathcal{E})$ [Eq. (23)] is obtained using Eq. (34). In this way, the operator $A_{\boldsymbol{q}_\parallel}(\mathcal{E})$ [Eq. (14)] for the exciton-light coupled state is determined.

C. Surface modes (surface exciton polariton)

Here, we consider the surface modes appearing for $q_{\mathcal{E}} < q_\parallel$. The surface modes are classified into X, Y, and Z modes in accordance with the light polarization [29,30]. The s-polarized surface mode in this study corresponds to the Y mode. Although the dispersion relation of the surface exciton polariton has been derived [29–32], the superposition coefficients of the exciton and photon have not been obtained. In this section, we calculate the superposition coefficients in addition to the dispersion relation of the surface exciton polariton.

In contrast to the leaky modes, the surface modes are designated by only $\boldsymbol{q}_\parallel$ because the eigenenergy is uniquely determined for each $\boldsymbol{q}_\parallel$. Thus the eigenenergy and annihilation operator for the surface mode are represented by $\mathcal{E}_{\boldsymbol{q}_\parallel}$ and $A_{\boldsymbol{q}_\parallel}$, respectively. The eigenoperator equation for $A_{\boldsymbol{q}_\parallel}$ is given by

$$[A_{\boldsymbol{q}_\parallel},H]=\mathcal{E}A_{\boldsymbol{q}_\parallel}. \quad (35)$$

Similar to the case of the leaky modes, we impose the commutation relations

$$[A_{\boldsymbol{q}_\parallel},A^\dagger_{\boldsymbol{q}'_\parallel}]=\delta_{\boldsymbol{q}_\parallel,\boldsymbol{q}'_\parallel}. \quad (36)$$

The operator $A_{\boldsymbol{q}_\parallel}$ is expressed as

$$A_{\boldsymbol{q}_\parallel}=\int_{-\infty}^{\infty}dq_z\alpha_{\boldsymbol{q}}a_{\boldsymbol{q}}+\int_{-\infty}^{\infty}dq_z\alpha'_{\boldsymbol{q}}a^\dagger_{-\boldsymbol{q}}+\beta_{\boldsymbol{q}_\parallel}b_{\boldsymbol{q}_\parallel}, \quad (37)$$

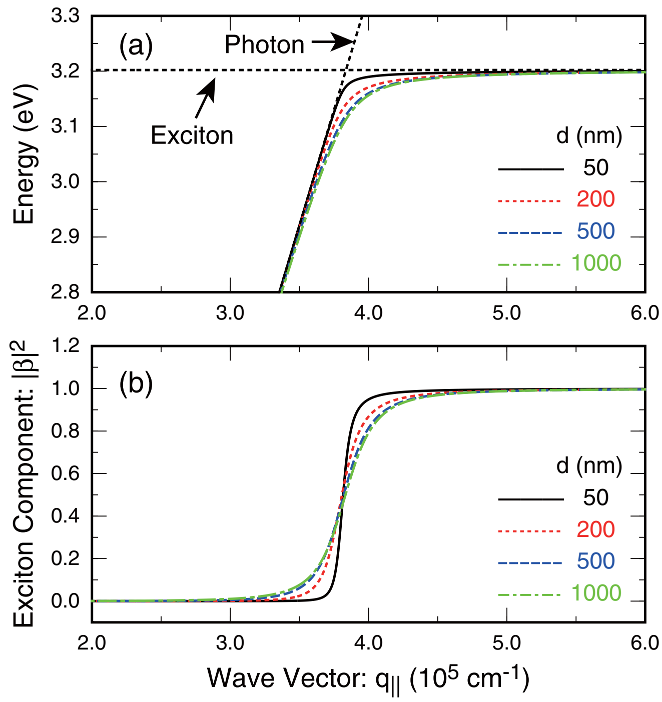

FIG. 1. (Color online) (a) The dispersions and (b) $|\beta_{\boldsymbol{q}_\parallel}|^2$ of the surface modes of a CuCl film with $d = 50$, 200, 500, and 1000 nm.

where $\alpha_{\boldsymbol{q}}$, $\alpha'_{\boldsymbol{q}}$, and $\beta_{\boldsymbol{q}_\parallel}$ are superposition coefficients of the surface modes. These coefficients can be determined from the following equations:

$$\hbar\omega_{\boldsymbol{q}}\alpha_{\boldsymbol{q}}+g^*_{\boldsymbol{q}}\beta_{\boldsymbol{q}_\parallel}=\mathcal{E}_{\boldsymbol{q}_\parallel}\alpha_{\boldsymbol{q}}, \quad (38)$$

$$-\hbar\omega_{\boldsymbol{q}}\alpha'_{\boldsymbol{q}}-g^*_{\boldsymbol{q}}\beta_{\boldsymbol{q}_\parallel}=\mathcal{E}_{\boldsymbol{q}_\parallel}\alpha'_{\boldsymbol{q}}, \quad (39)$$

$$E^{\rm ex}_{\boldsymbol{q}_\parallel}\beta_{\boldsymbol{q}_\parallel}+\int_{-\infty}^{\infty}dq_z g_{\boldsymbol{q}}(\alpha_{\boldsymbol{q}}+\alpha'_{\boldsymbol{q}})=\mathcal{E}_{\boldsymbol{q}_\parallel}\beta_{\boldsymbol{q}_\parallel}. \quad (40)$$

From Eq. (38), $\alpha_{\boldsymbol{q}}$ is obtained as

$$\alpha_{\boldsymbol{q}}=\frac{g^*_{\boldsymbol{q}}\beta_{\boldsymbol{q}_\parallel}}{\mathcal{E}_{\boldsymbol{q}_\parallel}-\hbar\omega_{\boldsymbol{q}}}. \quad (41)$$

In contrast to Eq. (20), the term including the delta function does not appear because there is no $\boldsymbol{q}_\parallel$ satisfying $\mathcal{E}_{\boldsymbol{q}_\parallel}=\hbar\omega_{\boldsymbol{q}}$ for $q_{\mathcal{E}} < q_\parallel$. From Eq. (39), we have

$$\alpha'_{\boldsymbol{q}}=-\frac{g^*_{\boldsymbol{q}}\beta_{\boldsymbol{q}_\parallel}}{\mathcal{E}_{\boldsymbol{q}_\parallel}+\hbar\omega_{\boldsymbol{q}}}. \quad (42)$$

By substituting Eqs. (41) and (42) into Eq. (40), we obtain

$$E^{\rm ex}_{\boldsymbol{q}_\parallel}+F(\mathcal{E}_{\boldsymbol{q}_\parallel})=\mathcal{E}_{\boldsymbol{q}_\parallel}, \quad (43)$$

where

$$F(\mathcal{E}_{\boldsymbol{q}_\parallel})=\int_{-\infty}^{\infty}dq_z\frac{2\hbar\omega_{\boldsymbol{q}}}{\mathcal{E}^2_{\boldsymbol{q}_\parallel}-(\hbar\omega_{\boldsymbol{q}})^2}|g_{\boldsymbol{q}}|^2. \quad (44)$$

An analytical expression for $F(\mathcal{E}_{\boldsymbol{q}_\parallel})$ is presented in Appendix B.

The dispersion relation of the surface mode is calculated by solving the nonlinear equation in Eq. (43). Figure 1(a) shows the dispersions of the surface modes of CuCl films with various thicknesses. We choose $E_{\mathrm{ex}} = 3.202$ eV, $M_{\mathrm{ex}} = 2.3m_e$, where m_e is the electron mass, $\Delta_{\mathrm{LT}} = 5.7$ meV, and $\epsilon_b = 5.59$ as the parameters of CuCl. With an increase in the film thickness, the dispersion largely deviates from photon and exciton dispersions in the vicinity of their crossing region because of the increase in the exciton-light interaction, and the dispersion approaches that of the surface polariton of a semi-infinite crystal, as expected.

The superposition coefficients $\alpha_{\boldsymbol{q}}$, $\alpha'_{\boldsymbol{q}}$, and $\beta_{\boldsymbol{q}_\parallel}$ are evaluated by using Eq. (36) in a similar manner for the leaky-mode case. The resulting coefficients are given by

$$\alpha_{\boldsymbol{q}} = \frac{g^*_{\boldsymbol{q}}}{\mathcal{E}_{\boldsymbol{q}_\parallel} - \hbar\omega_{\boldsymbol{q}}} \frac{1}{\sqrt{C_{\boldsymbol{q}_\parallel} + 1}}, \tag{45}$$

$$\alpha'_{\boldsymbol{q}} = -\frac{g^*_{\boldsymbol{q}}}{\mathcal{E}_{\boldsymbol{q}_\parallel} + \hbar\omega_{\boldsymbol{q}}} \frac{1}{\sqrt{C_{\boldsymbol{q}_\parallel} + 1}}, \tag{46}$$

$$\beta_{\boldsymbol{q}_\parallel} = \frac{1}{\sqrt{C_{\boldsymbol{q}_\parallel} + 1}}, \tag{47}$$

where

$$C_{\boldsymbol{q}_\parallel} = 4\int_{-\infty}^{\infty} dq_z \frac{\mathcal{E}_{\boldsymbol{q}_\parallel}\hbar\omega_{\boldsymbol{q}}}{\left[\mathcal{E}^2_{\boldsymbol{q}_\parallel} - (\hbar\omega_{\boldsymbol{q}})^2\right]^2} |g_{\boldsymbol{q}}|^2. \tag{48}$$

An analytical expression for $C_{\boldsymbol{q}_\parallel}$ is presented in Appendix C.

Figure 1(b) shows the calculated exciton component $|\beta_{\boldsymbol{q}_\parallel}|^2$ of the surface polariton for the CuCl films with various thicknesses. Surface polaritons have both exciton and photon components in the vicinity of the crossing region of the photon and exciton dispersions, whereas they are almost photonic ($|\beta_{\boldsymbol{q}_\parallel}|^2 \sim 0$) or almost excitonic ($|\beta_{\boldsymbol{q}_\parallel}|^2 \sim 1$), apart from the crossing region. The region where the surface polariton has both exciton and photon components is extended with an increase in the film thickness because of the increase in the exciton-light interaction.

The dispersions of the surface modes have been calculated without the RWA in the LWA [29,30]. In the present calculation, the RWA is partially applied, i.e., the counter-rotating terms of a photon in Eqs. (14) and (37) are taken into account. This means that Maxwell's equations are rigorously treated. Figure 2 shows the dispersion relations calculated including and excluding the counter-rotating term, and we find that these dispersions show good agreement with each other. Nevertheless, the counter-rotating term is taken into account in this study because $F_{\boldsymbol{q}_\parallel}(\mathcal{E})$, $F(\mathcal{E}_{\boldsymbol{q}_\parallel})$, and $C_{\boldsymbol{q}_\parallel}$ can be analytically calculated. It is also noted that the present method is applicable beyond the LWA in contrast to the conventional method in Refs. [29,30]. The dispersions of the surface modes have been calculated at the same level approximation, in which the susceptibility in the RWA and the full Maxwell's equations are used [31,32].

The exciton component of the surface mode has been calculated in a bipolariton picture [33], which qualitatively shows the same behavior as Fig. 1(b). In contrast to Ref. [33], we can also calculate the photon components of the surface modes as Eqs. (45) and (46) in the present method.

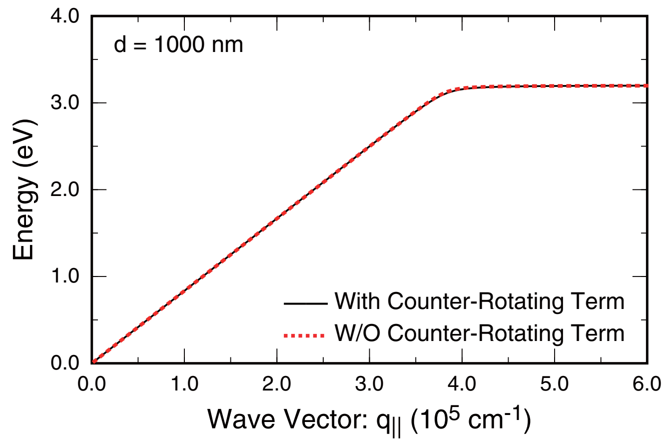

FIG. 2. (Color online) The dispersions of the surface modes of a CuCl film with $d = 1000$ nm calculated including (black solid line) and excluding (red dotted line) the counter-rotating term of a photon.

III. APPLICATIONS OF EXCITON-LIGHT COUPLED STATES

In a light scattering process by an exciton, the incident light induces exciton polarization, and the induced polarization emits light. Such a light scattering process can be calculated semiclassically. However, the photon emission process starting from an exciton as the initial state cannot be described in the semiclassical scheme, and a full-quantum description is necessary. In this section, the population-decay dynamics of an exciton and its photoluminescence are obtained using the derived exciton-light coupled states.

A. Population decay dynamics of an exciton

Let us consider the population decay dynamics of the exciton $|X_{\boldsymbol{q}_\parallel}\rangle$ with the use of the exciton-light coupled state $|\mathcal{E}\boldsymbol{q}_\parallel\rangle$ for the leaky mode. From the completeness relation of $|\mathcal{E}\boldsymbol{q}_\parallel\rangle$ with respect to $\mathcal{E}$ and the conservation law of $\boldsymbol{q}_\parallel$, $|X_{\boldsymbol{q}_\parallel}\rangle$ is expanded as

$$|X_{\boldsymbol{q}_\parallel}\rangle = \int d\mathcal{E}\, |\mathcal{E}\boldsymbol{q}_\parallel\rangle \langle\mathcal{E}\boldsymbol{q}_\parallel|X_{\boldsymbol{q}_\parallel}\rangle = \int d\mathcal{E}\beta^*_{\boldsymbol{q}_\parallel}(\mathcal{E})\, |\mathcal{E}\boldsymbol{q}_\parallel\rangle\,, \tag{49}$$

and its time development is given by

$$|X_{\boldsymbol{q}_\parallel}(t)\rangle = \int d\mathcal{E} e^{-i(\mathcal{E}/\hbar)t}\beta^*_{\boldsymbol{q}_\parallel}(\mathcal{E})\, |\mathcal{E}\boldsymbol{q}_\parallel\rangle\,. \tag{50}$$

The time dependence of the exciton population $P_{X_{\boldsymbol{q}_\parallel}}(t)$ is calculated as

$$\begin{aligned} P_{X_{\boldsymbol{q}_\parallel}}(t) &= |\langle X_{\boldsymbol{q}_\parallel}|X_{\boldsymbol{q}_\parallel}(t)\rangle|^2 \\ &= \left|\int d\mathcal{E}|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2 e^{-i(\mathcal{E}/\hbar)t}\right|^2. \end{aligned} \tag{51}$$

Here, we approximate as $F_{\boldsymbol{q}_\parallel}(\mathcal{E}) \approx F_{\boldsymbol{q}_\parallel}(E^{\mathrm{ex}}_{\boldsymbol{q}_\parallel}) \equiv F_{\boldsymbol{q}_\parallel}$ and $\Gamma_{\boldsymbol{q}_\parallel}(\mathcal{E}) \approx \Gamma_{\boldsymbol{q}_\parallel}(E^{\mathrm{ex}}_{\boldsymbol{q}_\parallel}) \equiv \Gamma_{\boldsymbol{q}_\parallel}$, and the poles of $|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2$ are approximately obtained as $\mathcal{E} = E^{\mathrm{ex}}_{\boldsymbol{q}_\parallel} + F_{\boldsymbol{q}_\parallel} \pm i\Gamma_{\boldsymbol{q}_\parallel}$ [see Eq. (34)]. By integrating with respect to $\mathcal{E}$ using a contour integral in the complex plane shown in Fig. 3(a), we obtain

$$P_{X_{\boldsymbol{q}_\parallel}}(t) = \exp[-2(\Gamma_{\boldsymbol{q}_\parallel}/\hbar)t], \tag{52}$$

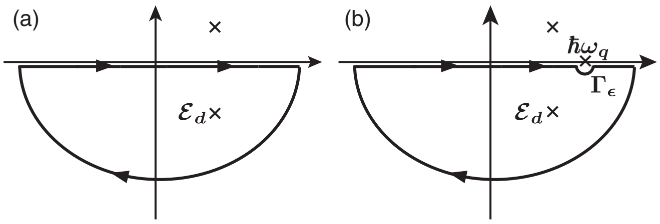

FIG. 3. The contours in the complex plane for evaluating the integrals (a) in Eq. (51) and (b) in the first term of Eq. (54), where $\mathcal{E}_d = E^{\rm ex}_{\boldsymbol{q}_\parallel} + F_{\boldsymbol{q}_\parallel} - i\Gamma_{\boldsymbol{q}_\parallel}$.

and thus, the exciton exponentially decays with the radiative decay rate $\Gamma_{\boldsymbol{q}_\parallel}(E^{\rm ex}_{\boldsymbol{q}_\parallel})$.

B. Photoluminescence of an exciton

A photon $|\boldsymbol{q}\rangle$ emitted from an exciton $|X_{\boldsymbol{q}_\parallel}\rangle$ has the same $\boldsymbol{q}_\parallel$ but has various values of q_z, and the probability of photon emission in the range of dq_z around q_z is given by $|\langle \boldsymbol{q}|X_{\boldsymbol{q}_\parallel}(t)\rangle|^2 dq_z$. This probability of photon emission is converted to that in the range of $d(\hbar\omega_{\boldsymbol{q}})$ around $\hbar\omega_{\boldsymbol{q}}$ as

$$P(\omega_{\boldsymbol{q}},t)d(\hbar\omega_{\boldsymbol{q}}) = 2\frac{\sqrt{\epsilon_b}}{\hbar c}\frac{q}{q_z}|\langle \boldsymbol{q}|X_{\boldsymbol{q}_\parallel}(t)\rangle|^2 d(\hbar\omega_{\boldsymbol{q}}). \quad (53)$$

The factor 2 comes from the fact that q_z and $-q_z$ provide the same value of $\hbar\omega_{\boldsymbol{q}}$. The photon probability density $P(\omega_{\boldsymbol{q}},t)$ in the $t \to \infty$ limit corresponds to the photoluminescence spectrum $P(\omega_{\boldsymbol{q}})$.

By inserting the completeness relation for $|\mathcal{E}\boldsymbol{q}_\parallel\rangle$ to $\langle \boldsymbol{q}|X_{\boldsymbol{q}_\parallel}(t)\rangle$, we have

$$\begin{aligned} P(\omega_{\boldsymbol{q}}) &= \lim_{t\to\infty} 2\frac{\sqrt{\epsilon_b}}{\hbar c}\frac{q}{q_z}\left|\int d\mathcal{E}\alpha_{\boldsymbol{q}}(\mathcal{E})\beta^*_{\boldsymbol{q}_\parallel}(\mathcal{E})e^{-i(\mathcal{E}/\hbar)t}\right|^2 \\ &= \lim_{t\to\infty} 2\frac{\sqrt{\epsilon_b}}{\hbar c}\frac{q}{q_z}\left|P\int d\mathcal{E}\frac{|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2}{\mathcal{E}-\hbar\omega_{\boldsymbol{q}}}g^*_{\boldsymbol{q}}e^{-i(\mathcal{E}/\hbar)t} \right. \\ &\quad \left. + z_{\boldsymbol{q}_\parallel}(\hbar\omega_{\boldsymbol{q}})|\beta_{\boldsymbol{q}_\parallel}(\hbar\omega_{\boldsymbol{q}})|^2 g^*_{\boldsymbol{q}}e^{-i\omega_{\boldsymbol{q}}t}\right|^2. \end{aligned} \quad (54)$$

The principal value of the integral of the first term can be evaluated by integrating along the contour shown in Fig. 3(b), in which the first-order singularity $\hbar\omega_{\boldsymbol{q}}$ is avoided by going around it along the infinitesimally small semicircle Γ_ϵ. Inside the contour, there is a singular point of $\mathcal{E}_d$ originating from $|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2$. The residue at $\mathcal{E}_d$ contains the damping factor of $\approx\exp[-2\Gamma_{\boldsymbol{q}_\parallel}(\mathcal{E}^{\rm ex}_{\boldsymbol{q}_\parallel})t/\hbar]$, as shown in the calculation of the population decay dynamics, and the contribution becomes zero in the $t \to \infty$ limit. By evaluating the contour integral along Γ_ϵ, the principal value in the $t \to \infty$ limit is obtained as

$$P\int_{-\infty}^{\infty} d\mathcal{E}\frac{|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2}{\mathcal{E}-\hbar\omega_{\boldsymbol{q}}}g^*_{\boldsymbol{q}}e^{-i(\mathcal{E}/\hbar)t} = i\pi|\beta_{\boldsymbol{q}_\parallel}(\hbar\omega_{\boldsymbol{q}})|^2 g^*_{\boldsymbol{q}}e^{-i\omega_{\boldsymbol{q}}t}. \quad (55)$$

As a result, we obtain

$$P(\omega_{\boldsymbol{q}}) = 2\frac{\sqrt{\epsilon_b}}{\hbar c}\frac{q}{q_z}\left[\pi^2 + z^2_{\boldsymbol{q}_\parallel}(\hbar\omega_{\boldsymbol{q}})\right]|\beta_{\boldsymbol{q}_\parallel}(\hbar\omega_{\boldsymbol{q}})|^4|g_{\boldsymbol{q}}|^2. \quad (56)$$

From $\Gamma(\hbar\omega_{\boldsymbol{q}}) = 2\pi(\sqrt{\epsilon_b}/\hbar c)(q/q_z)|g_{\boldsymbol{q}}|^2$ and Eq. (28), the photoluminescence is obtained as

$$P(\omega_{\boldsymbol{q}}) = |\beta_{\boldsymbol{q}_\parallel}(\hbar\omega_{\boldsymbol{q}})|^2, \quad (57)$$

which has Lorentzian form, as shown in Eq. (34). The peak energy of the luminescence spectrum is determined as a solution $\mathcal{E}_L$ of $\mathcal{E} = E^{\rm ex}_{\boldsymbol{q}_\parallel} - F_{\boldsymbol{q}_\parallel}(\mathcal{E})$ and is shifted from $E^{\rm ex}_{\boldsymbol{q}_\parallel}$. This peak shift $F_{\boldsymbol{q}_\parallel}(\mathcal{E}_L)$ is called a radiative shift. The half width at the half maximum (HWHM) $\Gamma_{\boldsymbol{q}_\parallel}(\mathcal{E}_L)$ of the spectrum comes from the radiative width. Because $E^{\rm ex}_{\boldsymbol{q}_\parallel} \gg F_{\boldsymbol{q}_\parallel}(\mathcal{E}_L)$, the radiative width is approximated as $\Gamma_{\boldsymbol{q}_\parallel}(E^{\rm ex}_{\boldsymbol{q}_\parallel})$, which agrees with the radiative decay rate of the exciton.

Although the photoluminescence spectra cannot be obtained with a semiclassical treatment, the elastic light-scattering spectra can be calculated semiclassically. Among the semiclassical methods, microscopic nonlocal theory makes it possible to include the effects of the radiative shift and width beyond the LWA [34,35]. In Appendix D, the reflection spectrum of the semiconductor thin film is calculated using microscopic nonlocal theory, and the radiative shift and width are shown to be the same as those of the photoluminescence spectrum.

IV. SUMMARY AND CONCLUSION

The superposition coefficients of an exciton and a photon of exciton-light coupled states have been obtained for a semiconductor film. Using the coupled states, we can theoretically investigate exciton-mediated optical properties that cannot be described by a semiclassical scheme. The coupled states are classified into leaky modes for $q_\mathcal{E} \geqslant q_\parallel$ and surface modes or surface exciton polaritons for $q_\mathcal{E} < q_\parallel$. The leaky modes are uniquely designated by $\mathcal{E}$ and $q_\parallel$. The exciton component $|\beta_{\boldsymbol{q}_\parallel}(\mathcal{E})|^2$ of the leaky modes is given by a Lorentz function, which has a peak at an energy slightly shifted from $E^{\rm ex}_{\boldsymbol{q}_\parallel}$ and has an HWHM corresponding to the radiative width of the exciton. As applications of the leaky modes, the population decay dynamics and photoluminescence spectra of an exciton are calculated.

The surface modes are only designated by $q_\parallel$, and their dispersion relations and superposition coefficient of exciton have been studied as a function of the film thickness. The superposition coefficient of photon has also been obtained for the first time. The radiative decay process of a biexciton toward the surface mode can be calculated using the present results. This problem is important to study the generation efficiency of entangled photons from the film, because the entangled photons cannot be generated outside the film in the radiative decay processes via surface modes. The decay rate is calculated using the Fermi's golden rule. In the calculation, both coefficients of the exciton and photon are necessary to evaluate the transition matrix elements between the biexciton and surface modes, and the dispersion relation of the surface mode is necessary to find $\boldsymbol{q}_\parallel$ satisfying the energy conservation. The radiative decay rate of the biexciton via surface modes will be presented near future.

We only consider the lowest exciton as a material excitation to obtain the exciton-light coupled states. Although the coupled states are calculated beyond the LWA, higher-level exciton states become significant with an increase in the film thickness [36,37]. Therefore the present study is valid for a thin film. An extension of the present study will include higher-level exciton states as future work.

ACKNOWLEDGMENTS

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APPENDIX A: ANALYTICAL EXPRESSION FOR $F_{q_\parallel}(\mathcal{E})$

The function $F_{q_\parallel}(\mathcal{E})$ related to the radiative shift in the leaky mode, which is defined in Eq. (25), is rewritten as

$$F_{q_\parallel}(\mathcal{E}) = \frac{\Delta_{\rm LT}}{\pi d} P \int_{-\infty}^{\infty} dq_z \frac{q_\parallel^2 + q_z^2}{q_{\mathcal{E}z}^2 - q_z^2} \left| \int_0^d dz e^{-iq_z z} \sin\left(\frac{\pi}{d} z\right) \right|^2 = \frac{\Delta_{\rm LT}}{\pi d} \int_0^d dz \int_0^d dz' \sin\left(\frac{\pi}{d} z\right) \sin\left(\frac{\pi}{d} z'\right) I_a(z,z'), \quad \text{(A1)}$$

where

$$I_a(z,z') = P \int_{-\infty}^{\infty} dq_z \frac{q_\parallel^2 + q_z^2}{q_{\mathcal{E}z}^2 - q_z^2} e^{iq_z(z-z')}. \quad \text{(A2)}$$

To evaluate $I_a(z,z')$ for $z > z'$, we integrate along the contour in the complex plane, which is shown in Fig. 4(a). There is no singular point inside the contour, and the integral along $\Gamma_{\rm AB}$ is zero. Using the residue theorem, $I_a(z,z')$ is given by

$$I_a(z,z') = -\int_{\Gamma_{\epsilon-}+\Gamma_{\epsilon+}} \frac{q_\parallel^2 + q_z^2}{q_{\mathcal{E}z}^2 - q_z^2} e^{iq_z(z-z')} = \pi \frac{q_\mathcal{E}^2}{q_{\mathcal{E}z}} \sin[q_{\mathcal{E}z}(z-z')] \quad (\text{for } z > z'). \quad \text{(A3)}$$

For $z < z'$, we integrate along the similar contour shown in Fig. 4(a) in the lower half plane, and we have

$$I_a(z,z') = -\pi \frac{q_\mathcal{E}^2}{q_{\mathcal{E}z}} \sin[q_{\mathcal{E}z}(z-z')] \quad (\text{for } z < z'). \quad \text{(A4)}$$

Using these results, we obtain

$$F_{q_\parallel}(\mathcal{E}) = \Delta_{\rm LT} \frac{2\pi^2}{d^3} \frac{q_\mathcal{E}^2}{q_{\mathcal{E}z}} \frac{\sin(q_{\mathcal{E}z} d)}{\left[q_{\mathcal{E}z}^2 - (\pi/d)^2\right]^2} + \Delta_{\rm LT} \frac{q_\mathcal{E}^2}{q_{\mathcal{E}z}^2 - (\pi/d)^2}. \quad \text{(A5)}$$

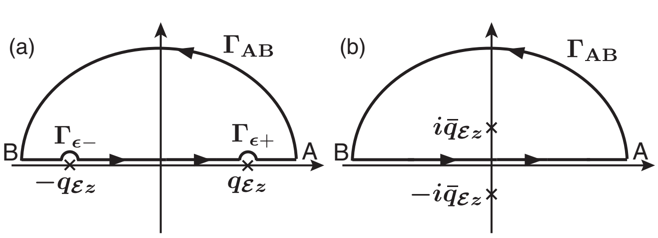

FIG. 4. The contours for evaluating the integrals (a) $I_a(z,z')$ for $z > z'$ and (b) $I_b(z,z')$ for $z > z'$.

APPENDIX B: ANALYTICAL EXPRESSION FOR $F(\mathcal{E}_{q_\parallel})$

The function $F(\mathcal{E}_{q_\parallel})$ related to the radiative shift in the surface mode, which is defined in Eq. (44), is rewritten as

$$F(\mathcal{E}_{q_\parallel}) = -\frac{\Delta_{\rm LT}}{\pi d} \int_{-\infty}^{\infty} dq_z \frac{q_\parallel^2 + q_z^2}{\bar{q}_{\mathcal{E}z}^2 + q_z^2} \times \left| \int_0^d dz e^{-iq_z z} \sin\left(\frac{\pi}{d} z\right) \right|^2 = -\frac{\Delta_{\rm LT}}{\pi d} \int_0^d dz \int_0^d dz' \sin\left(\frac{\pi}{d} z\right) \times \sin\left(\frac{\pi}{d} z'\right) I_b(z,z'), \quad \text{(B1)}$$

where $\bar{q}_{\mathcal{E}z} = \sqrt{q_\parallel^2 - q_\mathcal{E}}$ and

$$I_b(z,z') = \int_{-\infty}^{\infty} dq_z \frac{q_\parallel^2 + q_z^2}{\bar{q}_{\mathcal{E}z}^2 + q_z^2} e^{iq_z(z-z')}. \quad \text{(B2)}$$

To evaluate $I_b(z,z')$ for $z > z'$, we integrate along the contour shown in Fig. 4(b). There is a singular point $q_z = i\bar{q}_{\mathcal{E}z}$ inside the contour, and the integral along $\Gamma_{\rm AB}$ is zero. Using the residue theorem, we obtain

$$I_b(z,z') = \pi \frac{q_\mathcal{E}^2}{\bar{q}_{\mathcal{E}z}} e^{-\bar{q}_{\mathcal{E}z}(z-z')} \quad (\text{for } z > z'). \quad \text{(B3)}$$

For $z < z'$, we integrate along the similar contour shown in Fig. 4(b) in the lower half plane, and we have

$$I_b(z,z') = \pi \frac{q_\mathcal{E}^2}{\bar{q}_{\mathcal{E}z}} e^{\bar{q}_{\mathcal{E}z}(z-z')} \quad (\text{for } z < z'). \quad \text{(B4)}$$

Using these results, we obtain

$$F(\mathcal{E}_{q_\parallel}) = -\Delta_{\rm LT} \frac{2\pi^2}{d^3} \frac{q_\mathcal{E}^2}{\bar{q}_{\mathcal{E}z}} \frac{1 + e^{-\bar{q}_{\mathcal{E}z} d}}{\left[(\pi/d)^2 + \bar{q}_{\mathcal{E}z}^2\right]^2} - \Delta_{\rm LT} \frac{q_\mathcal{E}^2}{(\pi/d)^2 + \bar{q}_{\mathcal{E}z}^2}. \quad \text{(B5)}$$

APPENDIX C: ANALYTICAL EXPRESSION FOR $C_{q_\parallel}$

The superposition coefficients $\alpha_{\boldsymbol{q}}, \alpha'_{\boldsymbol{q}}$, and $\beta_{q_\parallel}$ of the surface mode are calculated from $C_{q_\parallel}$, which is defined in Eq. (48) and calculated as

$$C_{q_\parallel} = \frac{2}{\pi d} \frac{\epsilon_b}{(\hbar c)^2} \Delta_{\rm LT} \mathcal{E}_{q_\parallel} \int_{-\infty}^{\infty} dq_z \frac{q_\parallel^2 + q_z^2}{\left(\bar{q}_{\mathcal{E}z}^2 + q_z^2\right)^2} |I_c(q_z)|^2, \quad \text{(C1)}$$

where

$$I_c(q_z) = \int_0^d dz e^{-iq_z z} \sin\left(\frac{\pi}{d} z\right) = -\frac{\pi}{d} \frac{e^{-iq_z d} + 1}{q_z^2 - (\pi/d)^2}. \quad \text{(C2)}$$

Thus, $C_{q_\parallel}$ is rewritten as

$$C_{q_\parallel} = \frac{2\pi}{d^3} \frac{\epsilon_b}{(\hbar c)^2} \Delta_{\rm LT} \mathcal{E}_{q_\parallel} (J_+ + J_-), \quad \text{(C3)}$$

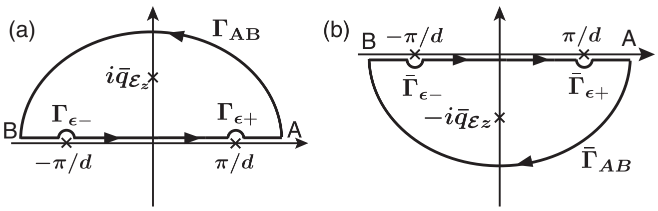

FIG. 5. The contours for evaluating the integrals (a) J_+ and (b) J_-.

where

$$J_\pm = \int_{-\infty}^{\infty} dq_z \frac{q_\parallel^2 + q_z^2}{\left(\bar{q}_{\mathcal{E}z}^2 + q_z^2\right)^2} \frac{1 + e^{\pm i q_z d}}{\left[q_z^2 - (\pi/d)^2\right]^2} \equiv \int_{-\infty}^{\infty} dq_z f_\pm(q_z). \tag{C4}$$

To evaluate the integrals J_+ and J_-, we integrate along the contours shown in Figs. 5(a) and 5(b), respectively, where two first-order singular points $\pm\pi/d$ are avoided by going around them along infinitesimally small semicircles $\Gamma_{\epsilon\pm}$. First, we calculate J_+. There is a second-order singularity at $q_z = i\bar{q}_{\mathcal{E}z}$ inside the contour in Fig. 5(a), and the integral along $\Gamma_{\rm AB}$ is zero. Therefore J_+ is given by

$$J_+ = \oint dq_z f_+(q_z) - \int_{\Gamma_{\epsilon-}+\Gamma_{\epsilon+}} dq_z f_+(q_z). \tag{C5}$$

The first integration is calculated as

$$\oint dq_z f_+(q_z) = \frac{\pi}{2} \frac{q_\mathcal{E}^2 + 2\bar{q}_{\mathcal{E}z}^2}{\bar{q}_{\mathcal{E}z}^3} \frac{1 + e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} + \frac{\pi}{2} \frac{q_\mathcal{E}^2 d}{\bar{q}_{\mathcal{E}z}^2} \frac{e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} + 2\pi \frac{q_\mathcal{E}^2}{\bar{q}_{\mathcal{E}z}} \frac{1 + e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^3}, \tag{C6}$$

and the second and third integrations are calculated as

$$\int_{\Gamma_1} dq_z f_+(q_z) = \int_{\Gamma_2} dq_z f_+(q_z) = -\frac{d^3}{4\pi} \frac{q_\parallel^2 + (\pi/d)^2}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2}. \tag{C7}$$

Using these results, we obtain

$$J_+ = \frac{d^3}{2\pi} \frac{q_\parallel^2 + (\pi/d)^2}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} + \frac{\pi}{2} \frac{q_\mathcal{E}^2 + 2\bar{q}_{\mathcal{E}z}^2}{\bar{q}_{\mathcal{E}z}^3} \frac{1 + e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} + \frac{\pi}{2} \frac{q_\mathcal{E}^2 d}{\bar{q}_{\mathcal{E}z}^2} \frac{e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} + 2\pi \frac{q_\mathcal{E}^2}{\bar{q}_{\mathcal{E}z}} \frac{1 + e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^3}. \tag{C8}$$

In a similar way, we have

$$J_- = \frac{d^3}{2\pi} \frac{q_\parallel^2 + (\pi/d)^2}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} + \frac{\pi}{2} \frac{q_\mathcal{E}^2 + 2\bar{q}_{\mathcal{E}z}^2}{\bar{q}_{\mathcal{E}z}^3} \frac{1 + e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} + \frac{\pi}{2} \frac{q_\mathcal{E}^2 d}{\bar{q}_{\mathcal{E}z}^2} \frac{e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} + 2\pi \frac{q_\mathcal{E}^2}{\bar{q}_{\mathcal{E}z}} \frac{1 + e^{-\bar{q}_{\mathcal{E}z} d}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^3}. \tag{C9}$$

Thus we obtain $C_{q_\parallel}$ as

$$C_{q_\parallel} = \frac{2\pi}{d^3} \frac{\epsilon_b}{(\hbar c)^2} \frac{\Delta_{\rm LT}\mathcal{E}_{q_\parallel}}{\left[\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2\right]^2} \left\{ \frac{d^3}{\pi}\left[q_\parallel^2 + (\pi/d)^2\right] + \pi \frac{1 + e^{-\bar{q}_{\mathcal{E}z} d}}{\bar{q}_{\mathcal{E}z}} \left[2 + \frac{q_\mathcal{E}^2}{\bar{q}_{\mathcal{E}z}^2} + \frac{4 q_\mathcal{E}^2}{\bar{q}_{\mathcal{E}z}^2 + (\pi/d)^2}\right] + \pi \frac{q_\mathcal{E}^2 d}{\bar{q}_{\mathcal{E}z}^2} e^{-\bar{q}_{\mathcal{E}z} d} \right\}. \tag{C10}$$

APPENDIX D: REFLECTION SPECTRUM CALCULATED USING A SEMICLASSICAL TREATMENT

In this Appendix, the reflection spectrum for s-polarized light is calculated using microscopic nonlocal theory [34,35]. The nonlocal theory is a semiclassical method, and linear and nonlinear optical responses can be calculated beyond the LWA. In the nonlocal theory, the scattered light and exciton polarization are obtained in a self-consistent manner; thus, the effects of the radiative shift and width are included in the resulting spectra. The purpose of this Appendix is to analytically compare the radiative shift and width of the reflection spectrum and those of the photoluminescence spectrum.

We consider a uniform background dielectric with ϵ_b. We set the electric field of the s-polarized incident light to be $\boldsymbol{E}_{q\parallel}^0 = (0, E_{q\parallel}^0, 0)$, and the electric field and induced exciton polarization can be treated as scalar functions. Because of the translational symmetry in the direction parallel to the surface, the induced exciton polarization $d_{q_\parallel}(\boldsymbol{r},\omega)$ has the same $\boldsymbol{q}_\parallel$ as that of the incident light. Within the linear response regime, the induced polarization is expressed as

$$d_{q_\parallel}(\boldsymbol{r},\omega) = \int d\boldsymbol{r}' \chi(\boldsymbol{r},\boldsymbol{r}';\omega) E_{q_\parallel}(\boldsymbol{r}'), \tag{D1}$$

where $\chi(\boldsymbol{r},\boldsymbol{r}';\omega)$ is the susceptibility in the RWA:

$$\chi(\boldsymbol{r},\boldsymbol{r}';\omega) = \frac{\langle g|\,\hat{d}(\boldsymbol{r})\,|X_{q_\parallel}\rangle\,\langle X_{q_\parallel}|\,\hat{d}(\boldsymbol{r}')\,|g\rangle}{E_{q_\parallel}^{\rm ex} - \hbar\omega - i\gamma}, \tag{D2}$$

$|g\rangle$ is the material ground state, and γ is the phenomenological dissipation constant excluding the radiative decay.

From Maxwell's equations, the electromagnetic wave equation is obtained as

$$\nabla \times \nabla \times \boldsymbol{E}_{q_\parallel}(\boldsymbol{r}) - q_b^2 \boldsymbol{E}_{q_\parallel}(\boldsymbol{r}) = 4\pi q^2 \boldsymbol{d}_{q_\parallel}(\boldsymbol{r}), \tag{D3}$$

where $q_b = \sqrt{\epsilon_b} q$ and $q = \omega/c$. Using $\nabla \cdot \boldsymbol{E}_{q_\parallel} = \partial E_{q_\parallel}/\partial y = 0$, this wave equation is rewritten as

$$\left(\frac{\partial^2}{\partial z^2} + \frac{\partial^2}{\partial x^2} + q_b^2\right) E_{q_\parallel}(\boldsymbol{r}) = -4\pi q^2 d_{q_\parallel}(\boldsymbol{r}). \tag{D4}$$

A formal solution of this equation is expressed as

$$E_{q_\parallel}(\boldsymbol{r}) = E_{q_\parallel}^0(\boldsymbol{r}) + \int d\boldsymbol{r}' G(\boldsymbol{r},\boldsymbol{r}') d_{q_\parallel}(\boldsymbol{r}'), \tag{D5}$$

where $G(\boldsymbol{r},\boldsymbol{r}')$ is the Green's function satisfying the following equation:

$$\left(\frac{\partial^2}{\partial z^2} + \frac{\partial^2}{\partial x^2} + q_b^2\right) G(\boldsymbol{r},\boldsymbol{r}') = -4\pi q^2 \delta(\boldsymbol{r} - \boldsymbol{r}'). \tag{D6}$$

By substituting Eq. (D1) into Eq. (D5), we have

$$E_{\boldsymbol{q}_\parallel}(\boldsymbol{r}) = E^0_{\boldsymbol{q}_\parallel}(\boldsymbol{r}) + \int d\boldsymbol{r}' \int d\boldsymbol{r}'' G(\boldsymbol{r},\boldsymbol{r}')\chi(\boldsymbol{r}',\boldsymbol{r}'')E_{\boldsymbol{q}_\parallel}(\boldsymbol{r}''). \tag{D7}$$

By multiplying $\langle X_{\boldsymbol{q}_\parallel}|\hat{d}(\boldsymbol{r})|g\rangle$ from the left-hand side of Eq. (D7) and integrating both sides with respect to $\boldsymbol{r}$, the following linear algebraic equation is obtained:

$$(E^{\rm ex}_{\boldsymbol{q}_\parallel} - \hbar\omega - i\gamma)F_{\boldsymbol{q}_\parallel} + A(\omega)F_{\boldsymbol{q}_\parallel} = F^0_{\boldsymbol{q}_\parallel}, \tag{D8}$$

where

$$F_{\boldsymbol{q}_\parallel} = \frac{1}{E^{\rm ex}_{\boldsymbol{q}_\parallel} - \hbar\omega - i\gamma}\int d\boldsymbol{r}\,\langle X_{\boldsymbol{q}_\parallel}|\hat{d}(\boldsymbol{r})|g\rangle\, E_{\boldsymbol{q}_\parallel}(\boldsymbol{r}), \tag{D9}$$

$$F^0_{\boldsymbol{q}_\parallel} = \int d\boldsymbol{r}\,\langle X_{\boldsymbol{q}_\parallel}|\hat{d}(\boldsymbol{r})|g\rangle\, E^0_{\boldsymbol{q}_\parallel}(\boldsymbol{r}), \tag{D10}$$

$$A(\omega) = -\int d\boldsymbol{r}\int d\boldsymbol{r}'\,\langle X_{\boldsymbol{q}_\parallel}|\hat{d}(\boldsymbol{r})|g\rangle \times G(\boldsymbol{r},\boldsymbol{r}')\,\langle g|\hat{d}(\boldsymbol{r}')|X_{\boldsymbol{q}_\parallel}\rangle, \tag{D11}$$

where Eq. (D2) is used. The solution of Eq. (D8) is obtained as

$$F_{\boldsymbol{q}_\parallel} = \frac{F^0_{\boldsymbol{q}_\parallel}}{E^{\rm ex}_{\boldsymbol{q}_\parallel} - \hbar\omega + A(\omega) - i\gamma}. \tag{D12}$$

The light reflected from the semiconductor film is obtained from Eq. (D7) with $E^0_{\boldsymbol{q}_\parallel}(\boldsymbol{r}) = 0$:

$$E_{\boldsymbol{q}_\parallel}(\boldsymbol{r}) = \int d\boldsymbol{r}' G(\boldsymbol{r},\boldsymbol{r}')\frac{\langle g|\hat{d}(\boldsymbol{r}')|X_{\boldsymbol{q}_\parallel}\rangle}{E^{\rm ex}_{\boldsymbol{q}_\parallel} - \hbar\omega + A(\omega) - i\gamma}F^0_{\boldsymbol{q}_\parallel}, \tag{D13}$$

where Eqs. (D2) and (D12) are used.

Here, we calculate the Green's function by solving Eq. (D6). The Fourier expansions of $G(\boldsymbol{r},\boldsymbol{r}')$ and $\delta(\boldsymbol{r}-\boldsymbol{r}')$ are written as

$$G(\boldsymbol{r},\boldsymbol{r}') = \sum_{\boldsymbol{q}_\parallel}\int dq_z G(\boldsymbol{q})e^{i\boldsymbol{q}_\parallel\cdot(\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel)}e^{iq_z(z-z')}, \tag{D14}$$

$$\delta(\boldsymbol{r}-\boldsymbol{r}') = \frac{1}{S}\sum_{\boldsymbol{q}_\parallel}e^{i\boldsymbol{q}_\parallel\cdot(\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel)}\frac{1}{2\pi}\int dq_z e^{iq_z(z-z')}. \tag{D15}$$

By substituting these Fourier expansions into Eq. (D6), we obtain the Fourier component of the Green's function as

$$G(\boldsymbol{q}) = \frac{2q^2}{S}\frac{1}{q_z^2 - q_{bz}^2}, \tag{D16}$$

where we choose the x axis in the direction of $\boldsymbol{q}_\parallel$ and define $q_{bz} = \sqrt{q_b^2 - q_\parallel^2}$. By substituting it into Eq. (D14) and integrating with respect to z with the use of the residue theorem, the Green's function is obtained as

$$G(\boldsymbol{r},\boldsymbol{r}') = \frac{2\pi i}{S}\frac{q^2}{q_{bz}}\sum_{\boldsymbol{q}_\parallel}e^{i\boldsymbol{q}_\parallel\cdot(\boldsymbol{r}_\parallel - \boldsymbol{r}'_\parallel)}e^{iq_{bz}|z-z'|}. \tag{D17}$$

From Eq. (8), the matrix element of the induced exciton polarization is given by

$$\langle X_{\boldsymbol{q}_\parallel}|\hat{d}(\boldsymbol{r})|g\rangle = \mu\sqrt{\frac{2}{Sd}}e^{-i\boldsymbol{q}_\parallel\cdot\boldsymbol{r}_\parallel}\sin\left(\frac{\pi}{d}z\right), \tag{D18}$$

and $F^0_{\boldsymbol{q}_\parallel}$ defined in Eq. (D10) is calculated as

$$F^0_{\boldsymbol{q}_\parallel} = -\mu\sqrt{\frac{2S}{d}}\frac{\pi}{d}\frac{e^{iq_{bz}d}+1}{q_{bz}^2 - (\pi/d)^2}, \tag{D19}$$

for an incident plane wave of $E^0_{\boldsymbol{q}_\parallel}(\boldsymbol{r}) = e^{i\boldsymbol{q}_\parallel\cdot\boldsymbol{r}_\parallel}e^{iq_{bz}z}$. When the incident field is applied from the left-hand side of the semiconductor film, the position detecting the reflected field z is smaller than the position z' in the film region. For $z < z'$, we perform the integration of $\boldsymbol{r}'$ in Eq. (D13) as follows:

$$\int d\boldsymbol{r}' G(\boldsymbol{r},\boldsymbol{r}')\,\langle g|\hat{d}(\boldsymbol{r}')|X_{\boldsymbol{q}_\parallel}\rangle = -2\pi i\mu\sqrt{\frac{2}{Sd}}\frac{\pi}{d}\frac{q^2}{q_{bz}}\frac{e^{iq_{bz}d}+1}{q_{bz}^2-(\pi/d)^2}e^{i\boldsymbol{q}_\parallel\cdot\boldsymbol{r}_\parallel}e^{-iq_{bz}z}. \tag{D20}$$

Using Eqs. (D19) and (D20) and $\mu^2 = \epsilon_b\Delta_{\rm LT}/4\pi$, we obtain the reflection spectrum

$$|E_{\boldsymbol{q}_\parallel}(\boldsymbol{r})|^2 = \frac{\left\{\Delta_{\rm LT}\frac{(2\pi)^2}{d^3}\frac{q_b^2}{q_{bz}}\frac{\cos^2(q_{bz}d/2)}{[q_{bz}^2-(\pi/d)^2]^2}\right\}^2}{\left[\hbar\omega - E^{\rm ex}_{\boldsymbol{q}_\parallel} - A^{\rm Re}(\omega)\right]^2 + [\bar{A}^{\rm Im}(\omega)+\gamma]^2}, \tag{D21}$$

where $A^{\rm Re}(\omega) = {\rm Re}[A(\omega)]$, and $\bar{A}^{\rm Im}(\omega) = -{\rm Im}[A(\omega)]$. The function of $A(\omega)$ defined in Eq. (D11) represents the self-interaction of the induced exciton polarization via the electromagnetic field, i.e., the self-energy of an exciton, and is calculated as

$$A^{\rm Re}(\omega) = \Delta_{\rm LT}\frac{2\pi^2}{d^3}\frac{q_b^2}{q_{bz}}\frac{\sin(q_{bz}d)}{\left[q_{bz}^2-(\pi/d)^2\right]^2} + \Delta_{\rm LT}\frac{q_b^2}{q_{bz}^2-(\pi/d)^2}, \tag{D22}$$

and

$$\bar{A}^{\rm Im}(\omega) = \Delta_{\rm LT}\frac{(2\pi)^2}{d^3}\frac{q_b^2}{q_{bz}}\frac{\cos^2(q_{bz}d/2)}{\left[q_{bz}^2-(\pi/d)^2\right]^2}. \tag{D23}$$

Thus the reflection spectrum is expressed by

$$|E_{\boldsymbol{q}_\parallel}(\omega)|^2 = \frac{[\bar{A}^{\rm Im}(\omega)]^2}{\left[\hbar\omega - E^{\rm ex}_{\boldsymbol{q}_\parallel} - A^{\rm Re}(\omega)\right]^2 + [\bar{A}^{\rm Im}(\omega)+\gamma]^2}, \tag{D24}$$

where $E_{\boldsymbol{q}_\parallel}(\boldsymbol{r})$ is renamed $E_{\boldsymbol{q}_\parallel}(\omega)$. The peak energy of the reflection spectrum is determined as the solution of $\mathcal{E}$ for $\mathcal{E} = E^{\rm ex}_{\boldsymbol{q}_\parallel} + A^{\rm Re}(\mathcal{E})$. The peak value of the reflection becomes 1 for $\gamma = 0$, and the HWHM of the spectrum is $\bar{A}^{\rm Im}(\mathcal{E})$. The value of $A^{\rm Re}(\mathcal{E})$ indicates the peak-energy shift in the reflection spectrum. When we set $q_b = q_{\mathcal{E}}$, it leads to $q_{bz} = q_{\mathcal{E}z}$, and consequently, $A^{\rm Re}(\omega)$ and $A^{\rm Im}(\omega)$ agree with $F_{\boldsymbol{q}_\parallel}(\mathcal{E})$ given by Eq. (A5) and $\Gamma_{\boldsymbol{q}_\parallel}(\mathcal{E})$ given by Eq. (26), respectively. In other words, the analytical expressions of the peak energy and spectral width of the photoluminescence have the same form as those of the reflection spectrum. It is noted that the reflection spectrum does not have the Lorentzian form and has a different shape from that of the photoluminescence spectrum.

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