Quantum theory for exciton polaritons in a real-space representation

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We develop a real-space method for exciton polaritons in a bounded medium with a tight-binding approximation. The creation and annihilation operators for photons are approximately constructed in a real-space representation. The quantum-mechanical derivation gives all necessary boundary conditions and complete solutions to the problem of reflection and transmission near the resonance of excitons. The present theory without the long-wavelength approximation covers the whole range of system size. In addition we calculate transient responses for short-pulse excitation. The results deeply reflect the spatial dispersion of exciton polaritons.

I. INTRODUCTION

For direct-gap semiconductors, an electronic polarization of excitons interacts strongly with electromagnetic fields at the crossing of their dispersion relations. The concept of polariton as the quantum which combines excitons and photons was first introduced by Hopfield and Thomas.[1] In the low-excitation regime the exciton polariton is regarded as a bosonic particle and accounts for many optical properties. This concept has a serious problem for responses of the bounded dispersive medium, necessity for additional boundary conditions (ABC's).[2] There have been a great number of attempts to solve the problem for many years, but most of them depend on the so-called semiclassical theory of light-matter interactions. The recent remarkable development of technologies enables us to have mutual quantum control of light and matter. Spontaneous emission in a microcavity is a good example of it.[3] Quantum electrodynamics (QED) is the best theory to understand the features of the quantized electromagnetic field. But electronic properties in condensed matters are so complicated that it is hopeless to calculate optical responses of excitons with QED. Some approximate methods of practical use are necessary. In the present paper we introduce the tight-binding photon approach. In Sec. II we develop a real-space method for photons interacting with excitons by means of the tight-binding approximation. Diagonalization of the total Hamiltonian gives upper and lower branch polariton modes. Appropriate boundary conditions (BC's) for photons and excitons are imposed to obtain optical responses of excitons in a film. In Sec. III we show numerical results of reflection and transmission for a CuCl film. This method without long-wavelength approximation can be applied for films with any thicknesses. What is more, we calculate the transient responses for short pulse excitation and show the behavior of exciton polariton propagation.

II. FORMALISM OF TIGHT-BINDING PHOTON

A. Tight-binding approximation for photons

Photons cannot be localized in real space. To be precise it is impossible to construct a position operator for photons.[4] The main reason is that the photon is a massless vector particle. Therefore QED describes photons in the $\boldsymbol{k}$-space representation. The photon field Hamiltonian is

$$H_p=\sum_{\boldsymbol{k},\lambda}\hbar\omega_{\boldsymbol{k}}(a^{\dagger}_{\boldsymbol{k}\lambda}a_{\boldsymbol{k}\lambda}+\tfrac{1}{2}), \tag{2.1}$$

where $\omega_{\boldsymbol{k}}=kc$ and $a^{\dagger}_{\boldsymbol{k}\lambda}$, $a_{\boldsymbol{k}\lambda}$ are, respectively, the photon creation and annihilation operators for the mode with wave vector $\boldsymbol{k}$ and polarization λ. For example, the energy shift of atomic levels is evaluated with QED due to emission and reabsorption of virtual photons. For that problem the contributions of both high- and low-energy frequencies must be summed up.[5]

For exciton systems the characteristic optical responses are determined by the interaction with photons near the exciton resonance. The LT splitting of CuCl Z_3 exciton is 5.7 meV and the linewidth of a 1 ps optical pulse is about 1 meV, while the resonant energy of the transverse exciton is 3.2022 eV. Thus we have only to deal with photons correctly in such a narrow energy range. Therefore, as shown in Fig. 1, we can approximately change the dispersion relation so that the group velocity agrees with that of exact photons near the exciton resonance. This new dispersion relation gives us the tight-binding Hamiltonian of photons represented in real space:

$$\begin{aligned}H_{tbp}&=\sum_k\hbar[\omega_{\rm ex}+2t_{\rm ph}\cos(k_{\rm ex}d)-2t_{\rm ph}\cos(kd)]a^{\dagger}_k a_k\\&=\sum_l\hbar[\omega_0 a^{\dagger}_l a_l-t_{\rm ph}(a^{\dagger}_{l+1}a_l+a^{\dagger}_l a_{l+1})],\end{aligned} \tag{2.2}$$

where $\hbar\omega_{\rm ex}$ is the resonant energy of the exciton, $k_{\rm ex}=\omega_{\rm ex}/c$, $\omega_0=\omega_{\rm ex}+2t_{\rm ph}\cos(k_{\rm ex}d)$, and $t_{\rm ph}$ is determined to make the dispersion relation exact near the exciton resonance,

$$t_{\rm ph}=\frac{c}{2d\,\sin k_{\rm ex}d}. \tag{2.3}$$

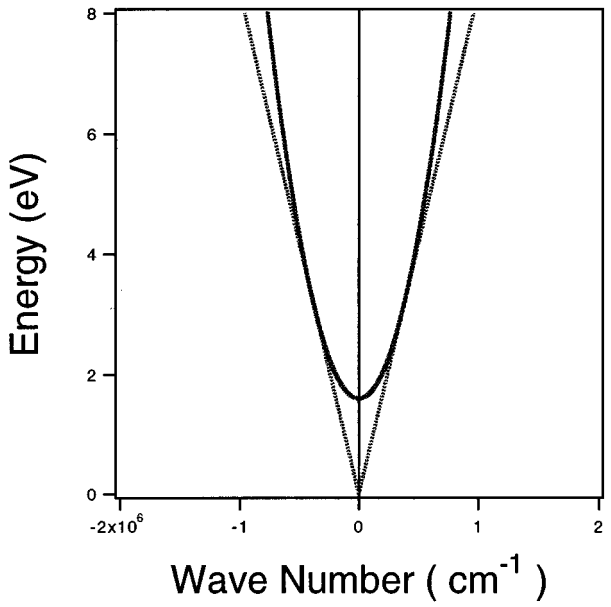

FIG. 1. The dispersion of real photons (dotted line) and tight-binding photons (solid line).

For simplicity we consider the photon modes propagating in one direction and neglect its polarization. Zero-point energy is dropped from the Hamiltonian. The operators a_l are defined as

$$a_l=\frac{1}{\sqrt{N}}\sum_k e^{ikld}a_k. \tag{2.4}$$

N is the number of modes and d is the lattice spacing which determines a cutoff frequency. This expression shows that a field mode is quantized in a one-dimensional cavity with length Nd. The $a_l^\dagger$ (a_l) is the creation (annihilation) operator of the photon localized at position ld in real space. So we call the photons created by these operators "tight-binding photons."

These operators do not work individually. However, if d is small enough that $kd\ll 1$, the field mode with wave number k becomes a superposition of localized modes approximately. In terms of localized modes electric field operators are written as

$$\begin{aligned}\boldsymbol{E}(l)&=\sum_k i\sqrt{\frac{\hbar ck}{2\epsilon_0 dN}}\,(a_k e^{ikld}-a_k^\dagger e^{-ikld})\\&\simeq i\sqrt{\frac{\hbar\omega_{\rm ex}}{2\epsilon_0 d}}\,(a_l-a_l^\dagger).\end{aligned} \tag{2.5}$$

Here we neglect the k dependence of coefficients and replace it with $k_{\rm ex}$ before Fourier transformation. As a result the local intensity of the electric field is in proportion to the number of localized photons.

These localized modes have a number of advantages. For one thing, there is only local interaction between photons and excitons in this approximation. Within the rotating wave approximation the interaction Hamiltonian is given

$$H_{\rm int}=\sum_l \hbar g(a_l^\dagger b_l+a_l b_l^\dagger), \tag{2.6}$$

where $b_l^\dagger$ (b_l) is the creation (annihilation) operator and g is a coupling constant. Although g depends on the energy as electric-field operators do, we replace it with $\omega_{\rm ex}$ as before. This local Hamiltonian reduces the eigenvalue problem of exciton-polariton to a simple equation, which is presented in the next section. Second, there is no free parameter to determine ABC's. To put it more precisely, when the system is bounded, the correct linear combination of polariton modes must be determined in the theory which treats the macroscopic polarization of excitons. BC's are locally defined, whereas the exciton polariton modes are diagonalized in the $\boldsymbol{k}$-space representation. That makes the BC's of excitons ambiguous. Thus the ABC's seems to be required in addition to Maxwell BC's. But the argument of ABC is completely avoided when the eigenvalue problems for excitons are solved with proper consideration of quantum-mechanical BC's.[6,7] The present theory is dependent on the quantum-mechanical and microscopic model in the real-space representation. We impose the BC's of photons and excitons, respectively, before diagonalization. These two are independent of each other. Accordingly, we can solve the problem without *ad hoc* conditions. Third, since the long-wavelength approximation is not assumed, this theory covers the whole range of system sizes. What is more, the tight-binding photon is not a classical wave but a quantum particle. It is easy to construct entangled states, coherent states, squeezed states, and so on. Experimental results of photon correlations are explained well by them. In the following sections we show the calculations of optical responses with the tight-binding photons.

B. Exciton-polariton system

We consider the photon propagation in a film. Light is normally incident on a film occupying the region $0<x<L$. The $1S$, Z_3 exciton of CuCl is adopted as a model for this study. The exciton is also approximated to the tight-binding boson, which has effective mass and no internal structures. The total Hamiltonian is

$$\begin{aligned}H_{\rm tot}=&\sum_{n\langle 0,n\rangle N}\hbar[\omega_0 a_n^\dagger a_n-t_{\rm ph}(a_{n+1}^\dagger a_n+a_n^\dagger a_{n+1})]\\&+\sum_{0\leqslant n\leqslant N}\hbar[\omega_0' a_n^\dagger a_n-t_{\rm ph}'(a_{n+1}^\dagger a_n+a_n^\dagger a_{n+1})]\\&+\sum_{0\leqslant n\leqslant N}\hbar[(\omega_{\rm ex}+2t_{\rm ex})b_n^\dagger b_n-t_{\rm ex}(b_{n+1}^\dagger b_n+b_n^\dagger b_{n+1})]\\&+\sum_{0\leqslant n\leqslant N}\hbar g(a_n^\dagger b_n+b_n^\dagger a_n),\end{aligned} \tag{2.7}$$

where $t_{\rm ex}=\hbar/2md^2$ and m denotes exciton mass. The first two lines are the Hamiltonian for photons inside and outside a film, respectively. The on-site energy $\hbar\omega_0'$ and transfer energy $\hbar t_{\rm ph}'$ in a film are different from those in vacuum because of the background dielectric constant ϵ_b which contains contributions from all interactions except the exciton in question. The third line represents the Hamiltonian of free

excitons and the fourth line the interaction Hamiltonian. The interactions between excitons are neglected in the low-excitation limit. The effects of scattering by phonons and defects are included as a damping constant γ of excitons. The lattice spacing d is a free parameter. The smaller it is, the better the tight-binding approximation becomes. However, exciton states with large momentum are important in exciton polariton systems. Thus d is assumed to be the lattice constant of CuCl crystal unless the system size is so small that tight-binding approximation is not valid for excitons. The parameter values used in this paper are

$$\hbar\omega_{\rm ex}=\hbar\omega_t=3.2022 \ \text{eV}, \quad \hbar\omega_l=3.2079 \ \text{eV},$$

$$\hbar m=2.3m_0, \quad \epsilon_b=5.59, \quad \hbar\gamma=0.04 \ \text{meV},$$

where $\hbar\omega_t$, $\hbar\omega_l$ are the energy of transverse and longitudinal excitons, respectively, and m_0 denotes the mass of the electron. The total Hamiltonian has a bilinear form and can be diagonalized by polariton operators as linear combination of exciton and photon operators,

$$H_{\rm tot}=\sum_k \hbar\omega_k p_k^\dagger p_k, \tag{2.8}$$

$$p_k^\dagger=\sum_n (\alpha_n a_n^\dagger+\beta_n b_n^\dagger). \tag{2.9}$$

The dispersion relation ω_k is shown in Fig. 2. Because of the tight-binding approximation the dispersion is very different from that of the real exciton polariton in the region of $\omega\ll\omega_{\rm ex}$ and $\omega\gg\omega_{\rm ex}$, but Fig. 2(b) shows that they agree very well near the exciton resonance.

So far as the modes inside a film are concerned, they are made of the upper and lower branch polaritons. On the other hand, in vacuum, eigenmodes are plane waves of photons. It seems that the presence of two propagating modes in a film with spatial dispersion requires ABC's at the interface between a film and vacuum. But this is not correct because the bounded medium breaks translational symmetry. Without an interaction between photons and excitons, two types of BC's are required to obtain eigenmodes for the system. One is the BC at the interface. The Maxwell BC's play the role for photons. As for excitons, the condition is that there is no exciton outside a film. The other is the quantum-mechanical BC for the wave function. The BC's are indispensable to quantize particles and fields. It should be noted that the surface effects on the dynamics of excitons are fully taken into account by solving the Scrödinger equation precisely and independent of the approximation for photons.

Now that the system is discrete in real space, BC's are equivalent to the boundary term of the Hamiltonian. To be more concrete, for photons, the value of the transfer energy at the interface should be changed according to the Maxwell BC's. That of excitons is zero since excitons do not exist outside a film and other BC's are needed for quantum-mechanical systems. In general, the BC's give the boundary values and the first derivative of wave functions for continuous systems. In discrete systems they determine the value of transfer and on-site energy near the boundary. ABC's are replaced with them.

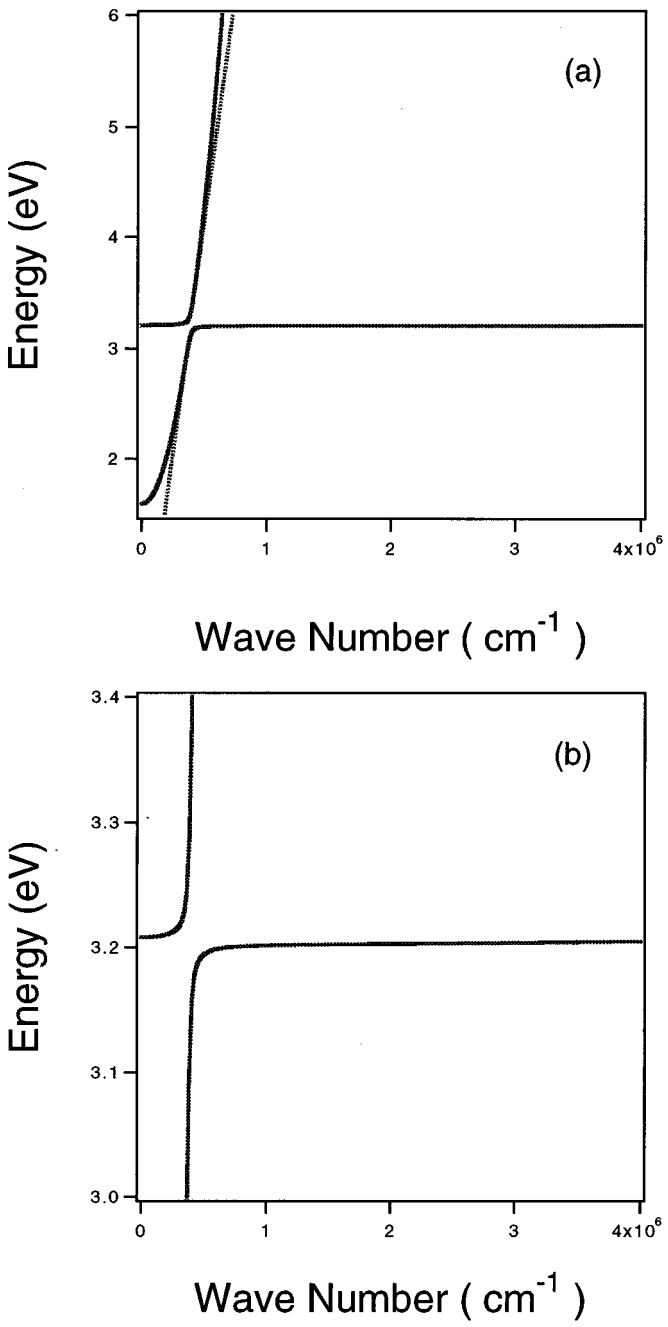

FIG. 2. The dispersion of an exciton polariton for CuCl with real photons (dotted line) and tight-binding photons (solid line).

Let us determine the coefficients α_n and β_n of the one-particle eigenstates for the Hamiltonian,

$$H_{\rm tot}|p_k\rangle=\hbar\omega|p_k\rangle, \tag{2.10}$$

$$|p_k\rangle=\sum_n (\alpha_n a_n^\dagger+\beta_n b_n^\dagger)|\text{vac}\rangle. \tag{2.11}$$

$|\text{vac}\rangle$ denotes the vacuum state of the system. From this Schrödinger equation, we get the relations between the coefficients α_n and β_n in a film:

$$(\omega-\omega_0')\alpha_n=-t_{\rm ph}'(\alpha_{n+1}+\alpha_{n-1})+g\beta_n, \tag{2.12}$$

$$(\omega-\omega_{\rm ex}+2t_{\rm ex})\beta_n=-t_{\rm ex}(\beta_{n+1}+\beta_{n-1})+g\alpha_n\,. \tag{2.13}$$

These relations are rewritten with the transfer matrix T.

$$\boldsymbol{u}_{n+1}=T\boldsymbol{u}_n \tag{2.14}$$

The vector $\boldsymbol{u}_n$ and the transfer matrix T are defined as

$$T=\begin{pmatrix} \dfrac{\omega_0-\omega}{t'_{\rm ph}} & \dfrac{g}{t'_{\rm ph}} & -1 & 0 \\ \dfrac{g}{t_{\rm ex}} & \dfrac{\omega_{\rm ex}+2t_{\rm ex}-\omega}{t_{\rm ex}} & 0 & -1 \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \end{pmatrix}, \qquad \boldsymbol{u}_n=\begin{pmatrix} \alpha_n \\ \beta_n \\ \alpha_{n-1} \\ \beta_{n-1} \end{pmatrix}. \tag{2.15}$$

As is the case with the scattering problem, the asymptotic behavior of photons outside the film is given by

$$\alpha_n=\begin{cases} e^{ikn}+\sqrt{r}e^{-ikn} & (n<0) \\ \sqrt{t}e^{ikn} & (n>N), \end{cases} \tag{2.16}$$

where $|r|$ is reflectivity and $|t|$ transmission. For excitons we impose $\beta_{-1}=\beta_{N+1}=0$ and change the values of transfer and on-site energy at the interface. After the conditions are taken into consideration, the problem is reduced to the next equations.

$$\boldsymbol{u}_{N+1}=T'T^{N-1}T''\boldsymbol{u}_0\,, \tag{2.17}$$

where T' and T'' denote the transfer matrix at the boundary:

$$T''=\begin{pmatrix} \dfrac{\omega_0-\omega}{t'_{\rm ph}} & \dfrac{g}{t'_{\rm ph}} & -\dfrac{t^b_{\rm ph}}{t'_{\rm ph}} & 0 \\ \dfrac{g}{t_{\rm ex}} & \dfrac{\omega^b_{\rm ex}+2t_{\rm ex}-\omega}{t_{\rm ex}} & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \end{pmatrix}, \qquad T'=\begin{pmatrix} \dfrac{\omega_0-\omega}{t^b_{\rm ph}} & \dfrac{g}{t^b_{\rm ph}} & -\dfrac{t'_{\rm ph}}{t^b_{\rm ph}} & 0 \\ \dfrac{g}{t_{\rm ex}} & \dfrac{\omega^b_{\rm ex}+2t_{\rm ex}-\omega}{t_{\rm ex}} & 0 & -1 \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \end{pmatrix}, \tag{2.18}$$

where constants with the superscript b denote their boundary values. Unknown parameters are r, t, β_0, and β_N. They determine the eigenmodes for photons interacting with excitons in a bounded medium.

It must be noted that the transfer matrix T has the modes of evanescent wave. As a result the largest eigenvalue makes a dominant contribution to $T^N\boldsymbol{u}_0$. Since the equations should be solved self-consistently to fix the parameters, it is difficult to get correct results by means of the numerical calculation of T^N. Therefore we get the analytical expression of r and t. The numerical results with those expressions are shown in the following section.

III. OPTICAL RESPONSES FOR CuCl FILMS

A. Reflection and transmission

We shall calculate reflection and transmission spectra in this section. First, let us consider the interference effect in a thin film. We impose the condition $\omega^b_{\rm ex}=\omega_{\rm ex}$. In the continuum limit $d\to 0$ this is equivalent to the Pekar ABC.[2] The dead layers are not taken into consideration because their effect is negligible for CuCl excitons. The validity of these conditions is discussed in many articles.[8] The BC that the on-site energy is changed at the interface is mentioned in the last part of this section.

In Fig. 3(a) the solid line shows reflection and the dotted line transmission for the 1-μm film. The interference spectra where $\omega<\omega_t$ and $\omega>\omega_l$ are due to LBP and UBP, respectively. Figure 3(b) shows the spectra for the 150-nm film. The structure of the spectrum above ω_l has an approximately doubled period in comparison with that below ω_l. That is because UBP and LBP are coupled with each other at the boundary, where they can be transformed into the other.[9] The interference in the stopband also reflects the spatial dispersion of excitons. These results are in good agreement with the experimental results by Mita and Nagasawa.[10]

Next, we consider responses for an ultrathin film. Experimental results can be explained by the frequency-dependent damping constant for excitons. To be strict, γ should be calculated from the microscopic model of scattering and include effects of system size fluctuation. For simplicity, we assume the following frequency dependence in the region of our interest:

$$\gamma=\begin{cases} 0.3 & (\omega<\omega_t) \\ 0.3+0.1(\omega-\omega_t) & (\omega\geqslant\omega_t)\,. \end{cases} \tag{3.1}$$

A unit of $\hbar\gamma$ is 1 meV. This value of the damping constant is much larger than that in the bulk crystal. For such a thin film, a center-of-mass motion of excitons is quantized due to the confinement: $E_n=\hbar\omega_t+(\hbar^2/2m_{\rm ex})(\pi n/L)^2$ with $n=1,2,3,\ldots$. According to the long-wavelength approximation (LWA), the quantized exciton wave function generates the parity selection rule. The oscillator strength for the excitons with even quantum numbers vanishes. Figure 4 shows reflection and transmission spectra for the film with the thickness of 150 Å.[11] This selection rule holds strictly although LWA is not assumed. As a film becomes thicker, the oscillator strength of those states grows up due to the breakdown of LWA. For the 150-nm film, all of the quantized states make a nearly equal contribution to the interference of the reflection spectrum in the region $\omega_t<\omega<\omega_l$. Consequently, the interval of peaks in the $\boldsymbol{k}$ space is π/L. This condition equals to that of Fabry-Perot interference $2kL=2\pi n$ with $n=1,2,\ldots$. On the other hand, the interval is $2\pi/L$ on the high-frequency side of ω_l. With the assumption that the wave number of UBP is negligible in comparison with that of LBP, the interference condition is approximately $kL=2\pi n$. Thus the contributions of excitons with even

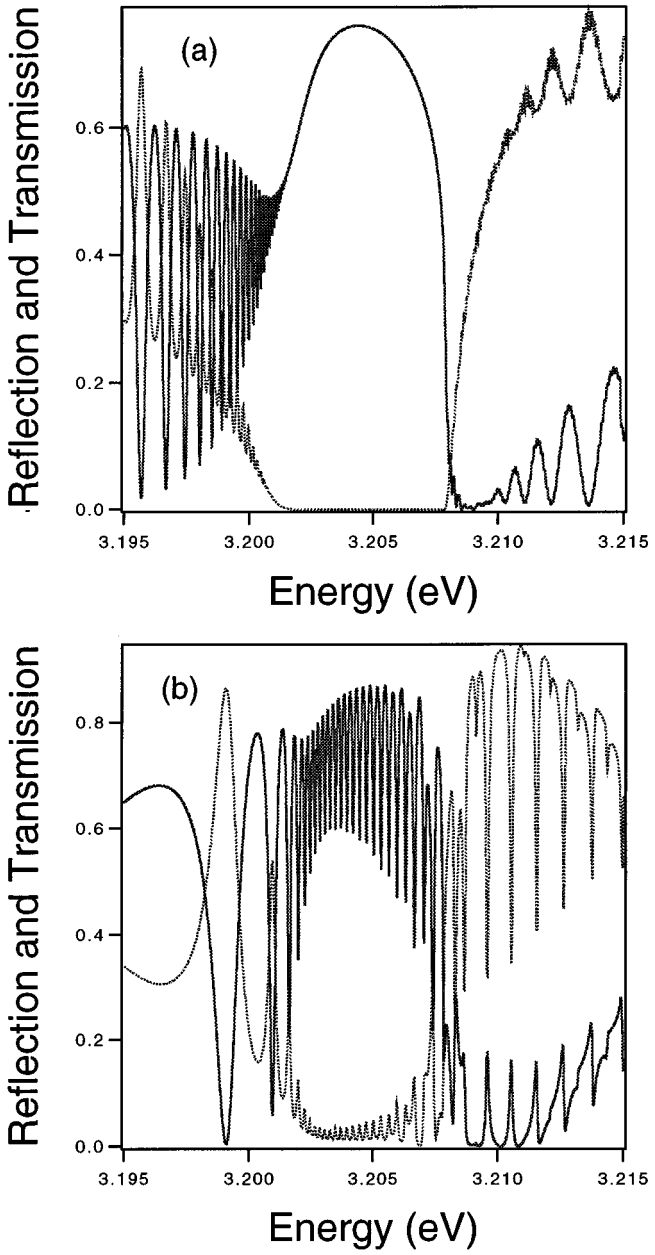

FIG. 3. Reflection (solid line) and transmission (dotted line) spectrum for a thin film for CuCl with the thickness of (a) 1 μm and (b) 150 nm.

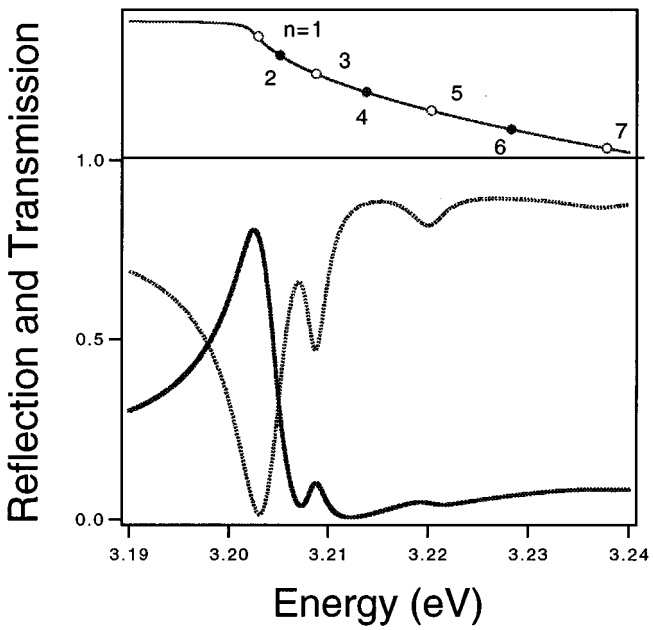

FIG. 4. Reflection (solid line) and transmission (dotted line) spectrum for a thin film for CuCl with the thickness of 150 Å. The thin line above the spectra shows the dispersion of LBP. The open and closed circles denote the energy of excitons with odd and even quantum numbers, respectively.

quantum numbers are dominant in terms of exciton confinement. As is well-known, the interference of the polariton and the exciton confinement are equivalent methods describing the optical oscillations in thin films.[12]

For a semi-infinite crystal (in the limit $N\rightarrow\infty$) we need a little modification for the method shown in the previous sections. In this case there is no reflection at the back of crystal. Thus the reflectivity are determined by the condition that there are only two modes (LBP and UBP) propagating forward (including the evanescent wave) in the crystal. To be precise the initial vector $\boldsymbol{u}_0$ is decomposed into the eigenvectors of the transfer matrix T and we set the coefficients of modes propagating backward to vanish. It is easier to solve the problem than that for a film with finite thickness because we need not calculate the Nth power of T. The reflectivity is given by

$$r=\left|\frac{1-e^{-i(k_L+k_U)d}-e^{-ikd}(2\cos k'd-e^{-ik_Ld}-e^{-ik_Ud})}{1-e^{-i(k_L+k_U)d}-e^{ikd}(2\cos k'd-e^{-ik_Ld}-e^{-ik_Ud})}\right|^2, \tag{3.2}$$

where k and k' are wave vectors of photons in a vacuum and in a film with the background dielectric constant ϵ_b. k_L and k_U are those of LBP and UBP, respectively. The result is shown as the solid line in Fig. 5. The damping constant γ is assumed to be 0 so that the effects of BC's become clear. In the continuum limit $d\rightarrow 0$ the reflectivity becomes

$$r_c=\left|\frac{1-n_{\rm eff}}{1+n_{\rm eff}}\right|^2, \quad n_{\rm eff}=\frac{\epsilon_b+n_Ln_U}{n_L+n_U}, \tag{3.3}$$

where $n_L=k_L/k$ and $n_U=k_U/k$. This is identical to the result by Pekar[2,12] and others.[9,13] The numerical results of r_c is almost equal to the solid line in Fig. 5.

Here we shall discuss the BC for excitons. A slight change of the Hamiltonian at the interface modifies the spectra. The dotted line in Fig. 5 shows the reflection spectrum where $\omega_{\rm ex}^b=\omega_{\rm ex}-t_{\rm ex}$. This is equivalent to the BC that the derivative of polarization vanishes ($\cos kx$ is used as the basis of excitons instead of $\sin kx$). In other words the reflection coefficient of excitons at the boundary is changed from -1 to 1. This figure shows that reflectivity with this BC is larger in the stopband than that with Pekar's ABC.[14] Other values of $\omega_{\rm ex}^b$ give other ABC's. Especially, the condition $\omega_{\rm ex}^b=\omega_{\rm ex}-(1-d\Gamma)t_{\rm ex}$ corresponds to

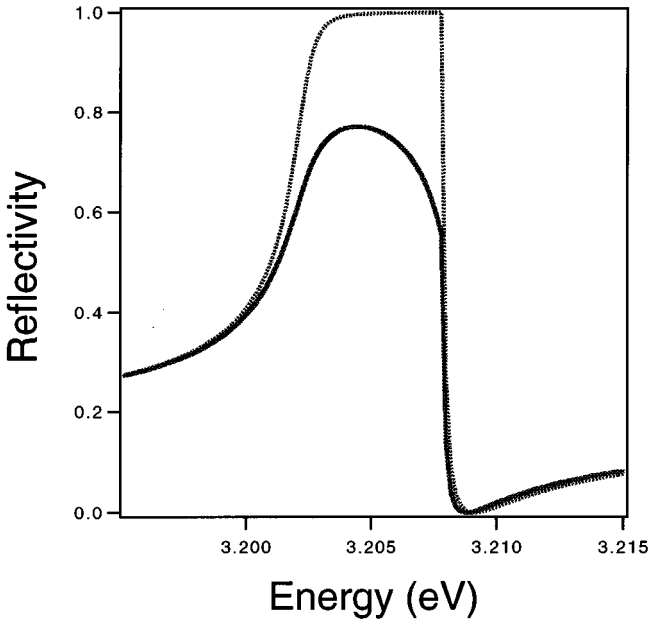

FIG. 5. The reflectivity for CuCl bulk crystal with two different BC's.

$$\Gamma P(x)+\frac{\partial P}{\partial x}(x)\bigg|_{\text{at the boundary}}=0 \tag{3.4}$$

in the continuum limit where $P(x)$ denotes the polarization of excitons. In real systems the lattice spacing d has a finite value. Consequently, as the value of ω_{ex}^{b} decreases from ω_{ex} to $\omega_{\text{ex}}-t_{\text{ex}}$, the reflectivity continuously changes between the solid line and the dotted line in Fig. 5. It is worth noting that, in this method, what we need are not *ad hoc* constraints but the conditions for excitons near the boundary, to be concrete, the values of on-site energy.

B. Transient responses for short-pulse excitation

In this section we calculate the time response of excitons in a thin film for short-pulse excitation. Because we consider only linear processes, the incident optical pulses can be described in terms of one-photon states. The initial state $|\Psi(t=0)\rangle$ is assumed to be

$$|\Psi(t=0)\rangle=\sum_{n}\exp\left[-\left(\frac{n-n_0}{\Delta}\right)^2+ik_0nd\right]|n\rangle, \tag{3.5}$$

where $|n\rangle$ is the photon state localized at the site n. This state describes the Gaussian pulse with width Δ centered at the site n_0. The purpose in this section is to calculate the time evolution of this initial state. There are several points that we should notice. First, the eigenmode with energy $\omega=kc$ is doubly degenerate because of propagating forward and backward. It is necessary to transform them to orthogonal basis. In this paper we use the symmetric and antisymmetric modes $\{|p_k\rangle_S,|p_k\rangle_A\}$ with regards to the spatial inversion, which is equal to $\{\cos kx,\sin kx\}$ in free space. Next, it is possible that there are bound states in a film other than propagating modes. The energy of bound states is lower than that of continuum (propagating) states. Thus the value of Δ should be large enough to exclude the modes in the region where the tight-binding approximation does not hold. We estimate $\Delta d/c>50$ fs from the approximated dispersion relation. After consideration of these conditions, we calculate

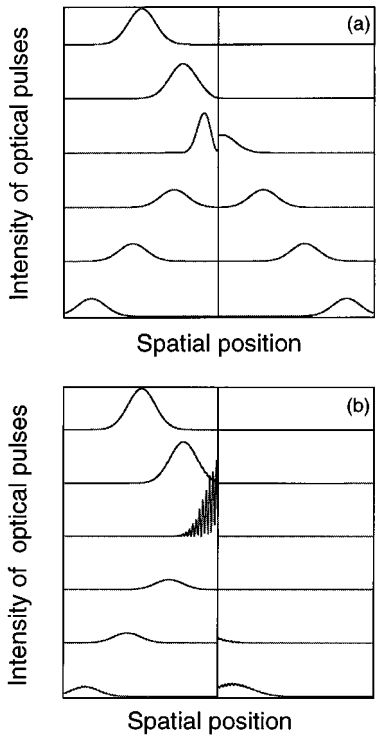

FIG. 6. The time evolutions of 1.8-ps Gaussian optical pulse with the peak at (a) 3.15 eV and (b) 3.195 eV. The time interval is 2 ps. The horizontal axis shows spatial position. The center line denotes the CuCl film with the thickness of 5.4 μm.

$$|\Psi(t)\rangle=\sum_{k}e^{i\omega_k t}(c_k^S|p_k\rangle_S+c_k^A|p_k\rangle_A) \tag{3.6}$$

and obtain time evolutions of an optical pulse in a thin film. c_k^S and c_k^A are the coefficients in an expansion of the initial state (3.5).

Figure 6(a) shows the transient response for the 1.8-ps pulse in the film with the thickness of 5.4 μm. The energy of the incident pulse is 3.15 eV far below ω_t. The time interval is 2 ps. Because the exciton polariton is almost photonlike, the optical pulse reflects and transmits as if no interaction takes place between photons and excitons.

For the incident pulse with 3.195 eV, the polariton takes on more excitonic character. As a result the delay of the transmitted pulse appears in Fig. 6(b) due to the small group velocities for the exciton polariton near ω_t.[15]

Figure 7(a) shows the propagation in a film for excitons and photons, respectively, for the incident pulse with 3.195 eV. Although the BC makes the amplitude of excitons vanish at the interface, the form of amplitude for excitons is very similar to that for photons inside a film. Namely, for the picosecond incident pulse, the pulses of excitons and photons propagate in the same way, not separately. The pulse width becomes much narrower than that of the incident optical

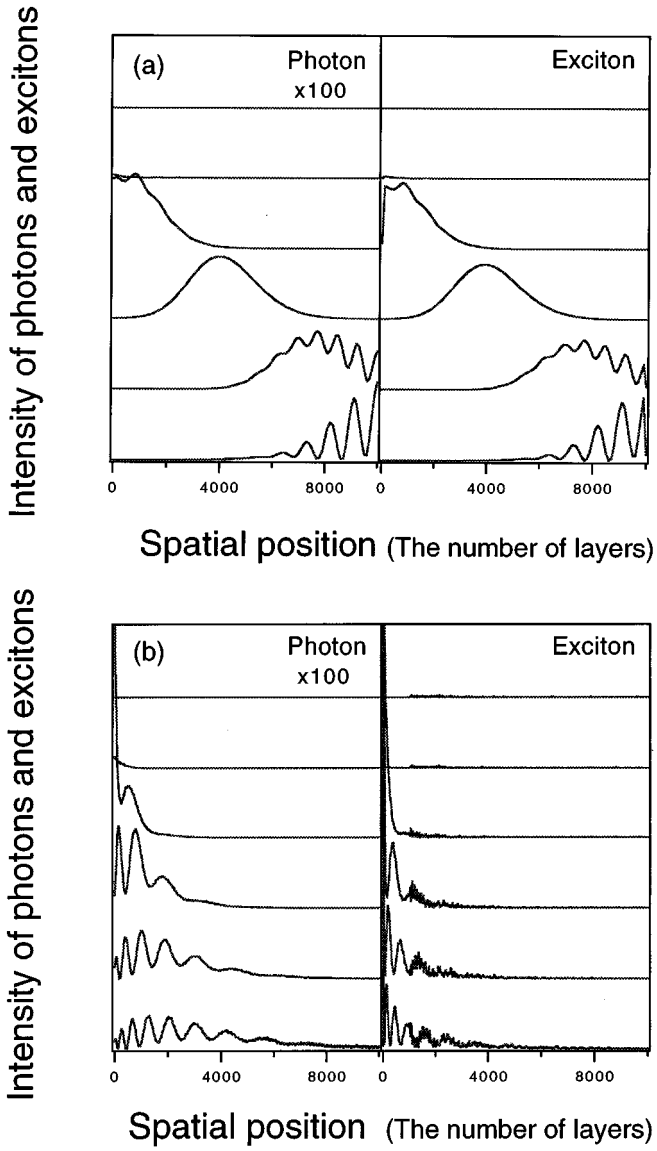

FIG. 7. The time evolutions of the exciton polariton in the film. The intensity of photons and excitons is plotted separately. The widths of incident pulses are (a) 1.8 ps and (b) 90 fs. (a) shows the time evolution in a film of Fig. 6(b). In (b) the time interval is 100 fs. The horizontal line shows the number of CuCl layers in the film.

pulse. On the other hand, this is not the case with the femtosecond pulse. Figure 7(b) shows the propagation of excitons and photons for the 90-fs incident pulse with 3.195 eV. It appears that excitons and photons propagate separately in a film. The linewidth of the 90 fs pulse is about 100 meV and covers the stopband $\omega_t<\omega<\omega_l$ completely. The pulse contains slow (excitonlike) and fast (photonlike) components in a film due to the dispersion relation. That is why the ultrashort pulse cannot keep its shape while propagating in spatially dispersive media. A number of excitons propagate very slowly and are collected near the interface. What is more, the envelope function of photons in a film is described by an Airy function in the frequency region where the group velocity of exciton polariton has a minimum.[16] In Fig. 7(b) this is reflected by the fact that the interval of intensity peaks becomes smaller as the pulse propagates.

IV. CONCLUDING REMARKS

Let us briefly discuss the quantum nature for photons in terms of tight-binding approximations. For the present we shall concentrate on the state of two photons with wave vectors $\boldsymbol{k}_A$ and $\boldsymbol{k}_B$. The two photon Fock state is $a^\dagger_{\boldsymbol{k}_A}a^\dagger_{\boldsymbol{k}_B}|\mathrm{vac}\rangle$. The real-space method enables us to construct the wave function for it,

$$\Psi(\boldsymbol{x}_1,\boldsymbol{x}_2)=\frac{1}{\sqrt{2}}\langle\mathrm{vac}|a_{\boldsymbol{x}_2}a_{\boldsymbol{x}_1}a^\dagger_{\boldsymbol{k}_A}a^\dagger_{\boldsymbol{k}_B}|\mathrm{vac}\rangle$$
$$=\frac{1}{\sqrt{2}}\,(e^{-i\boldsymbol{k}_A\cdot\boldsymbol{x}_1-i\boldsymbol{k}_B\cdot\boldsymbol{x}_2}+e^{-i\boldsymbol{k}_A\cdot\boldsymbol{x}_2-i\boldsymbol{k}_B\cdot\boldsymbol{x}_1}). \tag{4.1}$$

In terms of this wave function the normally ordered correlation function of the photon (electric field) intensity $I(\boldsymbol{x})\propto a^\dagger_{\boldsymbol{x}}a_{\boldsymbol{x}}$ is given by

$$G_2(\boldsymbol{x}_1,\boldsymbol{x}_2)=\langle:I(\boldsymbol{x}_1)I(\boldsymbol{x}_2):\rangle$$
$$\propto|\Psi(\boldsymbol{x}_1,\boldsymbol{x}_2)|^2,$$
$$=1+\cos[\Delta\boldsymbol{k}\cdot(\boldsymbol{x}_1-\boldsymbol{x}_2)], \tag{4.2}$$

where $\Delta\boldsymbol{k}=\boldsymbol{k}_A-\boldsymbol{k}_B$. This interference shows an essentially quantum effect.[17] According to the classical theory the visibility is less than or equal to 50%. Thus the tight-binding approximation does not change the quantum nature for photons at all. $\Psi(\boldsymbol{x}_1,\boldsymbol{x}_2)$ is regarded as the symmetrized wave function of two photons in real space. For the states with more than two photons the tight-binding photon approach gives the symmetrized wave functions in the same way. Therefore the quantum interference of multiphotons can be interpreted as the result of symmetrization in the real-space representation within the present theory. It is very interesting to investigate propagation of the pulse with many photons in an exciton system with nonlinear interactions.

In conclusion, we introduce a real-space and quantum-mechanical theory for photons which do not extend so much in $\boldsymbol{k}$ space. We considered the exciton polariton system in a film. The interaction Hamiltonian has the local form in terms of localized operators for photons and excitons. Reflection and transmission problem is made into a very simple equation including the quantum-mechanical BC's for excitons. The solutions near the exciton resonance are in good agreement with the experimental results in the whole range of film thickness. For transient responses the present theory demonstrates pulse propagation of exciton polaritons. The results deeply reflect the spatial dispersion. Moreover, the tight-binding approximation preserves the quantum nature for photons and explains the nonclassical effect in the interference of multiphotons. The tight-binding photon approach has many another applications: squeezed states, dressed excitons, photonic bands, and so on. In particular, it is important to clarify the quantum dynamics in optical devices. The present theory will be a useful tool for those problems.

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