励起子 QED 論における付加的境界条件の有用性

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Usefulness of Additional Boundary Conditions in Exciton QED Theory

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When we consider center-of-mass kinetic energy of excitons, additional boundary conditions (ABCs) are required to connect the electromagnetic fields inside and outside of excitonic medium. Although some theoretical schemes do not require ABCs, we must treat large number of exciton states in order to discuss macroscopic materials, and then much time is required for calculation. This paper provides a calculation method of exciton correlation functions in inhomogeneous exciton-photon system from the calculation result of boundary problem with ABCs, which do not require so much time.

1 Introduction

It is well known that the classical electromagnetism is completely described by the Maxwell equations together with the Lorentz force law. However, in boundary problems for connecting the electromagnetic fields between different media, we sometimes require additional boundary conditions (ABCs) [1] in addition to the Maxwell boundary conditions, which are derived from the Maxwell equations. This ABC problem emerges when we consider materials which provide multiple light modes for a given frequency. The ABC problem was first pointed out by Pekar [2] for exciton-polariton systems with exciton center-of-mass kinetic energy, and the subsequent studies have elucidated that ABCs are determined by microscopically considering boundary conditions of the exciton center-of-mass motion at interfaces [3–6]. Afterward, some calculation methods without deriving ABCs have been established, and are they called ABC-free theory [7] or microscopic nonlocal theory [8]. Based on the same idea of these theories, we have constructed a quantum electrodynamics (QED) theory for excitons in inhomogeneous systems [9].

However, our theory requires the complete set of elementary excitations in excitonic materials, and, in order to derive exciton correlation functions renormalizing the interaction with the electromagnetic fields, we must numerically calculate the inverse of a large matrix whose size is the number of all excitation states. One method to reduce the matrix size is to describe the contribution from non-resonant excitations, such as phonons and higher excitons, by the background dielectric function in the Maxwell equations as $\varepsilon = \varepsilon_{\rm bg} + \chi_{\rm ex}$. However, even by using this technique, we must consider large number of exciton center-of-mass motion states in order to discuss macroscopic materials. The purpose of the present paper is to construct the exciton correlation functions in inhomogeneous exciton-photon systems from the calculation result of boundary problem with ABCs, which do not require the numerical calculation of large matrices.

2 Matrix inversion method

We consider a material where the translational symmetry is broken in the z direction, and s-polarized excitons are weakly confined in a finite region. For simplicity, we consider only

one relative exciton state with eigenfrequency $\omega_{\rm T}$, and denote the center-of-mass motion by index m. The excitons are pure bosons, and the system is linear as

$$\hat{H}_{\rm ex} = \sum_m \hbar\Omega_m \hat{b}_m^\dagger \hat{b}_m, \tag{1}$$

where $\hat{b}_m$ is the annihilation operator of an exciton in state m, and Ω_m is the eigenfrequency including the center-of-mass kinetic energy. On the other hand, we consider the interaction between the excitons and the electromagnetic fields as

$$\hat{H}_{\rm int} = -\int {\rm d}z\ \hat{A}(z)\hat{J}(z), \tag{2}$$

where $\hat{A}(z)$ is the vector potential and $\hat{J}(z) = \sum_m \mathcal{J} g_m(z)\hat{b}_m + {\rm H.c.}$ is the excitonic current density. The coefficient $\mathcal{J}$ has the relation with the longitudinal-transverse (LT) splitting energy as $\hbar\omega_{\rm LT} = |\mathcal{J}|^2/\varepsilon_{\rm bg}\varepsilon_0{\omega_{\rm T}}^2$, and $g_m(z)$ is the exciton center-of-mass wavefunction. The whole Hamiltonian is written as $\hat{H} = \hat{H}_{\rm ex} + \hat{H}_{\rm int} + \hat{H}_{\rm em}$, and $\hat{H}_{\rm em}$ describes the electromagnetic fields and the background dielectrics as treated in our previous work [9].

The time-ordered exciton correlation function renormalizing the exciton-photon interaction is defined as

$$\mathrm{i}\mathfrak{G}_{m,m'}(\omega) \equiv \int_{-\infty}^{\infty} {\rm d}t\ {\rm e}^{\mathrm{i}\omega t} \left\langle {\rm T}\hat{b}_m(t)\hat{b}_{m'}^\dagger(0)\right\rangle, \tag{3}$$

where T is the time-ordering operator, and $\hat{b}_m(t) = {\rm e}^{\mathrm{i}\hat{H}t/\hbar}\hat{b}_m{\rm e}^{-\mathrm{i}\hat{H}t/\hbar}$ is the Heisenberg representation of the exciton operator. As a result of our QED theory [9], under the rotating wave approximation (RWA), this correlation function is calculated by matrix inversion (Dyson equation) as

$$[\mathfrak{G}^{-1}(\omega)]_{m,m'} = (\omega - \Omega_m)\delta_{m,m'} - \Sigma_{m,m'}(\omega), \tag{4}$$

where the self-energy is written as

$$\Sigma_{m,m'}(\omega) = \varepsilon_{\rm bg}\omega_{\rm LT}({\omega_{\rm T}}/{c})^2 \times \int_{-\infty}^{\infty} {\rm d}z{\rm d}z'\ g_m^*(z)G(z,z',\omega)g_{m'}(z'), \tag{5}$$

and $G(z,z',\omega)$ is the Green's function satisfying

$$\left[\frac{\partial^2}{\partial z^2} + {k_\parallel}^2 + \varepsilon_{\rm bg}(z)\frac{\omega^2}{c^2}\right] G(z,z',\omega) = \delta(z-z').$$

Here, $k_\parallel$ is the in-plane wavenumber. In deriving the self-energy (5), we use the fact that, according to Ref. 10, the Green's function corresponds to the retarded correlation function of the vector potential in $\hat{H}_{\rm em}$ system:

$$\mu_0 G(z,z',\omega) = \frac{1}{\mathrm{i}\hbar}\int_0^\infty {\rm d}t\ {\rm e}^{\mathrm{i}\omega t}\left\langle[\hat{A}_0(z,t),\hat{A}_0(z',0)]\right\rangle_{\rm em}, \tag{6}$$

where the time representation is defined as

$$\hat{A}_0(z,t) \equiv {\rm e}^{\mathrm{i}\hat{H}_{\rm em}t/\hbar}\hat{A}(z){\rm e}^{-\mathrm{i}\hat{H}_{\rm em}t/\hbar}. \tag{7}$$

However, we must treat large matrices to discuss macroscopic materials in this calculation scheme.

3 Exciton-photon coupled modes

Instead of the numerical matrix inversion in the previous section, we can also derive $\mathfrak{G}(\omega)$ by the following calculation. First, we derive the extreme value $\tilde{\omega}$ of the exciton correlation function. The real and imaginary parts of $\tilde{\omega}$ respectively represent the resonance frequency and radiative decay rate of exciton-photon coupled mode in the system. The complex frequency $\tilde{\omega}$ and its corresponding complex wavenumber $\tilde{k}$ satisfy the dispersion relation as

$$\frac{c^2({k_\parallel}^2 + \tilde{k}^2)}{\tilde{\omega}^2} = \varepsilon_{\rm bg} + \frac{\varepsilon_{\rm bg}\omega_{\rm LT}}{\Omega(\tilde{k}) - \tilde{\omega}} = \varepsilon_{\rm bg} + \chi_{\rm ex}(\tilde{k},\tilde{\omega}). \tag{8}$$

Further, they also satisfy the self-sustaining condition in the system. When we suppose a non-excitonic layer of thickness d, the self-sustaining condition is simply written as

$$r_L\ r_R\ {\rm e}^{\mathrm{i}2\tilde{k}d} = 1, \tag{9}$$

where $r_{L/R}$ is the reflection coefficient at left/right interface. This condition means that there is neither amplitude decay nor phase shift

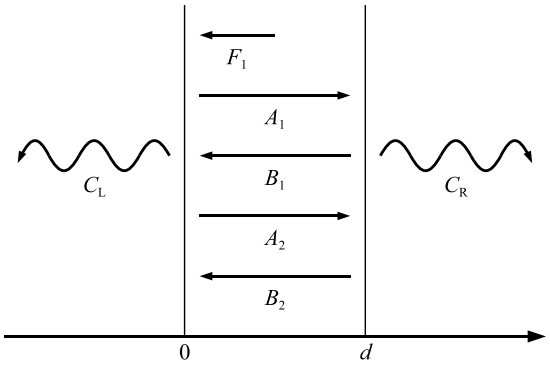

Fig. 1: System for calculating self-sustaining condition with considering ABCs.

after a round trip inside the layer. However, when we consider the exciton center-of-mass kinetic energy, there are two wavenumbers $\tilde{k}_1$ and $\tilde{k}_2$ satisfying Eq. (8) for a given $\tilde{\omega}$, and then we must consider ABCs. Therefore, we suppose the system as seen in Fig. 1 to derive the self-sustaining condition for the mode $\tilde{k}_1$. The electric field of each mode is written as

$$\begin{aligned}
E_L(z) &= C_L \mathrm{e}^{-\mathrm{i}k_L z}, \\
E_1(z) &= A_1 \mathrm{e}^{\mathrm{i}\tilde{k}_1(z-d)} + B_1 \mathrm{e}^{-\mathrm{i}\tilde{k}_1 z} \\
&\qquad (+F_1 \mathrm{e}^{-\mathrm{i}\tilde{k}_1 z} \text{ only at } z=0), \\
E_2(z) &= A_2 \mathrm{e}^{\mathrm{i}\tilde{k}_2(z-d)} + B_2 \mathrm{e}^{-\mathrm{i}\tilde{k}_2 z}, \\
E_R(z) &= C_R \mathrm{e}^{\mathrm{i}k_R z},
\end{aligned}$$

where $k_{L/R} = (\varepsilon_{L/R}\tilde{\omega}^2/c^2 - {k_\parallel}^2)^{1/2}$ is the wavenumber in left/right medium. A polariton field F_1 incidents on the left interface from the excitonic layer, and $A_{1,2}$, $B_{1,2}$, and $C_{L,R}$ are unknown. The Maxwell boundary conditions at $z=0$ and $z=d$ are obtained as

$$\begin{aligned}
C_L &= F_1 + A_1 \mathrm{e}^{-\mathrm{i}\tilde{k}_1 d} + B_1 + A_2 \mathrm{e}^{-\mathrm{i}\tilde{k}_2 d} + B_2, \\
C_R &= A_1 + B_1 \mathrm{e}^{-\mathrm{i}\tilde{k}_1 d} + A_2 + B_2 \mathrm{e}^{-\mathrm{i}\tilde{k}_2 d}, \\
k_L C_L &= k_1 F_1 - k_1 A_1 \mathrm{e}^{-\mathrm{i}\tilde{k}_1 d} \\
&\quad + k_1 B_1 - k_2 A_2 \mathrm{e}^{-\mathrm{i}\tilde{k}_2 d} + k_2 B_2, \\
k_R C_R &= k_1 A_1 - k_1 B_1 \mathrm{e}^{-\mathrm{i}\tilde{k}_1 d} \\
&\quad + k_2 A_2 - k_2 B_2 \mathrm{e}^{-\mathrm{i}\tilde{k}_2 d}.
\end{aligned}$$

Further, we require two ABCs to determine the six unknowns. In the present paper, we adopt the Pekar's ABC [2], and they are written as

$$\begin{aligned}
0 &= \chi_1 F_1 + \chi_1 A_1 \mathrm{e}^{-\mathrm{i}\tilde{k}_1 d} + \chi_1 B_1 \\
&\quad + \chi_2 A_2 \mathrm{e}^{-\mathrm{i}\tilde{k}_2 d} + \chi_2 B_2, \\
0 &= \chi_1 A_1 + \chi_1 B_1 \mathrm{e}^{-\mathrm{i}\tilde{k}_1 d} + \chi_2 A_2 + \chi_2 B_2 \mathrm{e}^{-\mathrm{i}\tilde{k}_2 d},
\end{aligned}$$

where $\chi_i = c^2({k_\parallel}^2 + \tilde{k}_i^2)/\tilde{\omega}^2 - \varepsilon_{\mathrm{bg}}$ is the excitonic susceptibility. By solving these six boundary conditions, the six unknowns are determined for given F_1. Here, in order to define the reflection coefficients r'_L and r'_R in this system, we consider that B_1/F_1 is written as

$$B_1/F_1 = r'_L r'_R \mathrm{e}^{\mathrm{i}\tilde{k}_1 d} + (r'_L r'_R \mathrm{e}^{\mathrm{i}\tilde{k}_1 d})^2 + \cdots . \quad (12)$$

Further, due to the correspondence to Eq. (9), we consider the self-sustaining condition for mode $\tilde{k}_1$ as

$$r'_L \; r'_R \; \mathrm{e}^{\mathrm{i}\tilde{k}_1 d} = \frac{B_1/F_1}{1 + B_1/F_1} = 1. \quad (13)$$

Although this condition is satisfied only in the limit of $|B_1/F_1| \to \infty$, we can renew $\tilde{k}_1$ as

$$\tilde{k}_1 := \frac{-1}{\mathrm{i}2d} \ln(r'_L \; r'_R) = \frac{-1}{\mathrm{i}2d} \ln \frac{(B_1/F_1)\mathrm{e}^{-\mathrm{i}2\tilde{k}_1 d}}{1 + (B_1/F_1)} \quad (14)$$

in the numerical successive calculation. Actually, by simultaneously solving Eqs. (8) and (14), we can reproduce $\tilde{\omega}$ obtained by the correlation function method.

4 Reconstruction of correlation function

Using the extreme value set $\{\tilde{\omega}_\lambda\}$, the exciton correlation function tensor should be written as

$$\mathfrak{G}(\omega) = \sum_\lambda \frac{\boldsymbol{v}^*(\tilde{\omega}_\lambda)\boldsymbol{v}(\tilde{\omega}_\lambda)}{\omega - \tilde{\omega}_\lambda}, \quad (15)$$

where the vectors $\{\boldsymbol{v}(\tilde{\omega}_\lambda)\}$ are orthonormal as

$$\boldsymbol{v}(\tilde{\omega}_\lambda) \cdot \boldsymbol{v}^*(\tilde{\omega}_{\lambda'}) = \delta_{\lambda,\lambda'}. \quad (16)$$

On the other hand, according to Ref. 11, it is also approximately derived from Eq. (4) as

$$\mathfrak{G}_{m,m'}(\omega) \simeq \sum_k \frac{\{g_m(k)\}^* g_{m'}(k)}{\omega - \Omega(k) - \Sigma(\omega,k)} \quad (17)$$

where $g_m(k)$ is the Fourier transform of exciton center-of-mass wavefunction:

$$g_m(k) = \frac{1}{\sqrt{L}} \int_{-\infty}^{\infty} \mathrm{d}z \ \mathrm{e}^{-\mathrm{i}kz} g_m(z), \qquad (18)$$

and the self-energy is written as

$$\Sigma(\omega, k) = \frac{\varepsilon_{\mathrm{bg}} {\omega_{\mathrm{LT}}} {\omega_{\mathrm{T}}}^2 / c^2}{\varepsilon_{\mathrm{bg}} (\omega + \mathrm{i}\delta)^2 / c^2 - {k_{\parallel}}^2 - k^2}. \qquad (19)$$

By comparing Eqs. (15) and (17) and our intuitive consideration, we find the form of $\boldsymbol{v}(\tilde{\omega}_\lambda)$ as

$$v_m(\tilde{\omega}_\lambda) = \sum_{i=1}^{2} \frac{\chi_{\lambda,i}}{N} \big[A_{\lambda,i} g_m(\tilde{k}_{\lambda,i}) + \mathrm{e}^{-\mathrm{i}\tilde{k}_{\lambda,i} d} B_{\lambda,i} g_m(-\tilde{k}_{\lambda,i}) \big], \qquad (20)$$

where $A_{i,\lambda}$ and $B_{i,\lambda}$ are the coefficients obtained in the self-sustaining condition, and N is the normalization factor. Actually, Eq. (20) satisfies the orthogonal relation (16), and provides a good approximation of the exciton correlation function at thickness smaller than about 500 nm for CuCl film.

5 Discussion

In order to obtain a good numerical precision in calculating the exciton correlation functions, in the case of CuCl film with 1 μm thickness, we must consider 500 states of exciton center-of-mass motion. The required state number is doubled, if we double the film thickness. In the matrix inversion method, the calculation time is proportional to the third power of the number of considered exciton states. In contrast, in the extreme value method, the calculation time is linearly proportional to the state number. Furthermore, we can also perform analytical calculations by using the extreme values and the form (15) of the exciton correlation function. For example, we can derive the temperature correlation function of excitons by the analytic continuation with the retarded one.

However, Eqs. (15) and (20) cannot be applied to films thicker than 1 μm. We must derive more accurate expression of the exciton correlation functions for future applications of this calculation method, as well as considering the effects of RWA. Further, we must also elucidate the correspondence with the correlation function of ideal exciton-polaritons in the limit of infinite crystal.

Although we adopt the Pekar's ABC [2] in the present paper, the other ABCs can be applied to the extreme value method. In this situation, we must microscopically derive the form of ABCs from the center-of-mass wavefunctions of excitons [3–6]. Further, in addition to the excitonic layer considered in the present paper, this method can be applied to other structures, such as multilayers, spheres, photonic crystals and so on, if we can obtain proper expressions of ABCs. In this way, the extreme value method provides the connection between the classical field expansion method with ABCs and the microscopic nonlocal theory [8] or our QED theory [9].

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