

# Appendix A

## Multipolar Hamiltonian

There are two ways to describe the interaction between an electromagnetic field and a charged particle. One is to use the minimal coupling Hamiltonian, and the other is to employ the multipolar Hamiltonian. These two Hamiltonians are related to each other by a unitary transformation, and there are, in principle, no problems regardless of which is adopted [1–3]. The multipolar Hamiltonian has a simple form without the static Coulomb interaction and can exactly describe the retardation effects by exchanging transverse photons, which are photons possessing only a polarization component perpendicular to the wave-vector  $\mathbf{k}$ . Since these features are advantageous for the discussion in Chap. 2, this appendix reviews the multipolar Hamiltonian.

Let us consider a charged particle system confined in a nanomaterial. In the following, we choose two nanomaterials as an example and look for an appropriate Hamiltonian. When the wavelength of the electromagnetic wave is much longer than the size of the nanomaterial, the vector potential  $\mathbf{A}(\mathbf{R})$  at the center position  $\mathbf{R}$  of the nanomaterial is the same as  $\mathbf{A}(\mathbf{q})$ , independent of the position  $\mathbf{q}$  of an electric charge in the nanomaterial:

$$\mathbf{A}(\mathbf{q}) = \mathbf{A}(\mathbf{R}). \quad (\text{A.1})$$

From Eq. (A.1), it follows that the magnetic flux density is zero ( $\mathbf{B} = \nabla \times \mathbf{A} = \mathbf{0}$ ), and thus, the interaction between the charged particle and magnetic field can be neglected. Moreover, one can take only the electric dipole interaction into account because the magnetic dipole and higher multipoles can be neglected. In addition, by assuming that the electron exchange interaction is also negligible, the Lagrangian,  $L$ , for the system can be written as

$$L = L_{mol} + L_{rad} + L_{int}, \quad (\text{A.2a})$$

where

$$L_{mol} = \sum_{\varsigma} \left\{ \sum_{\alpha} \frac{m_{\alpha} \dot{\mathbf{q}}_{\alpha}^2(\varsigma)}{2} - V(\varsigma) \right\}, \quad (\text{A.2b})$$

$$L_{rad} = \frac{\epsilon_0}{2} \int \left\{ \dot{\mathbf{A}}^2 - c^2 (\nabla \times \mathbf{A})^2 \right\} d^3r, \quad (\text{A.2c})$$

and

$$L_{int} = \sum_{\varsigma} \sum_{\alpha} e \dot{\mathbf{q}}_{\alpha}(\varsigma) \cdot \mathbf{A}(\mathbf{R}_{\varsigma}) - V_{int}. \quad (\text{A.2d})$$

Here,  $\dot{\cdot}$  represents the time derivative. The index  $\varsigma$  is used for distinguishing the nanomaterials 1 and 2, and  $\alpha$  is used to specify a charged particle in a nanomaterial.  $\epsilon_0$ ,  $c$ , and  $e$  represent the dielectric constant in vacuum, the speed of light in vacuum, and the electric charge, respectively.  $L_{mol}$  denotes the energy of charged particles with mass  $m_{\alpha}$  and velocity  $\dot{\mathbf{q}}_{\alpha}$  in the Coulomb potential  $V(\varsigma)$ , while  $L_{rad}$  denotes the energy of the electromagnetic field in free space. The third term  $L_{int}$  denotes the interaction between the charged particle and the electromagnetic field. The Coulomb interaction  $V_{int}$  between the nanomaterials 1 and 2 is given by

$$V_{int} = \frac{1}{4\pi\epsilon_0 R^3} \{ \mathbf{p}(1) \cdot \mathbf{p}(2) - 3(\mathbf{p}(1) \cdot \mathbf{e}_R)(\mathbf{p}(2) \cdot \mathbf{e}_R) \}. \quad (\text{A.3})$$

Here  $R = |\mathbf{R}| = |\mathbf{R}_1 - \mathbf{R}_2|$  denotes the center-to-center separation between the nanomaterials 1 and 2, and  $\mathbf{e}_R$  is  $\mathbf{R}/R$ , the unit vector along the direction of  $\mathbf{R}$ . The electric dipole moments of the nanomaterials 1 and 2 are designated by  $\mathbf{p}(1)$  and  $\mathbf{p}(2)$ , respectively.

In order to simplify the interaction Hamiltonian, the Power–Zienau–Woolley transformation [1, 3]<sup>1</sup> is performed on the original Lagrangian  $L$ . The result is expressed as

$$L_{mult} = L - \frac{d}{dt} \int \mathbf{P}^{\perp}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d^3r, \quad (\text{A.4})$$

where  $\mathbf{P}^{\perp}(\mathbf{r})$  is the transverse component of the polarization density  $\mathbf{P}(\mathbf{r})$ .

The polarization density  $\mathbf{P}(\mathbf{r})$  is

$$\begin{aligned} \mathbf{P}(\mathbf{r}) = \sum_{\varsigma, \alpha} e (\mathbf{q}_{\alpha} - \mathbf{R}_{\varsigma}) \left[ 1 - \frac{1}{2!} \{ (\mathbf{q}_{\alpha} - \mathbf{R}_{\varsigma}) \cdot \nabla \} \right. \\ \left. + \frac{1}{3!} \{ (\mathbf{q}_{\alpha} - \mathbf{R}_{\varsigma}) \cdot \nabla \}^2 - \dots \right] \delta(\mathbf{r} - \mathbf{R}_{\varsigma}) \end{aligned} \quad (\text{A.5})$$

and only the electric dipole term is retained:

$$\mathbf{P}(\mathbf{r}) = \sum_{\varsigma, \alpha} e (\mathbf{q}_{\alpha} - \mathbf{R}_{\varsigma}) \delta(\mathbf{r} - \mathbf{R}_{\varsigma}) = \mathbf{p}(1) \delta(\mathbf{r} - \mathbf{R}_1) + \mathbf{p}(2) \delta(\mathbf{r} - \mathbf{R}_2). \quad (\text{A.6})$$

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<sup>1</sup> The Power–Zienau–Woolley transformation is a method for deriving the interaction Hamiltonian by using the electric displacement vector and magnetic flux density of the electromagnetic field instead of using the vector potential. Furthermore, measurable macroscopic quantities, such as the electric polarization and magnetization, are used.

Note that the current density is

$$\mathbf{j}(\mathbf{r}) = \sum_{\varsigma, \alpha} e \dot{\mathbf{q}}_{\alpha} \delta(\mathbf{r} - \mathbf{R}_{\varsigma}), \quad (\text{A.7})$$

and the transverse component of the current density is related to the transverse component of the polarization density as follows:

$$\mathbf{j}^{\perp}(\mathbf{r}) = \frac{d\mathbf{P}^{\perp}(\mathbf{r})}{dt}. \quad (\text{A.8})$$

Using Eqs. (A.7) and (A.8), the interaction Lagrangian  $L_{\text{int}}$  of Eq. (A.2d) can be rewritten as

$$L_{\text{int}} = \int \mathbf{j}^{\perp}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) d^3r - V_{\text{int}} = \int \frac{d\mathbf{P}^{\perp}(\mathbf{r})}{dt} \cdot \mathbf{A}(\mathbf{r}) d^3r - V_{\text{int}}, \quad (\text{A.9})$$

and thus  $L_{\text{mult}}$  given by Eq. (A.4) becomes

$$\begin{aligned} L_{\text{mult}} &= L - \int \frac{d\mathbf{P}^{\perp}(\mathbf{r})}{dt} \cdot \mathbf{A}(\mathbf{r}) d^3r - \int \mathbf{P}^{\perp}(\mathbf{r}) \cdot \dot{\mathbf{A}}(\mathbf{r}) d^3r \\ &= L_{\text{mol}} + L_{\text{rad}} - \int \mathbf{P}^{\perp}(\mathbf{r}) \cdot \dot{\mathbf{A}}(\mathbf{r}) d^3r - V_{\text{int}}. \end{aligned} \quad (\text{A.10})$$

Here, the momentum  $\mathbf{p}_{\alpha}$  conjugate to the coordinate  $\mathbf{q}_{\alpha}$  and the vector potential  $\mathbf{A}(\mathbf{r})$  conjugate to the momentum  $\mathbf{\Pi}(\mathbf{r})$  are defined by

$$\mathbf{p}_{\alpha} = \frac{\partial L_{\text{mult}}}{\partial \dot{\mathbf{q}}_{\alpha}} = \frac{\partial L_{\text{mol}}}{\partial \dot{\mathbf{q}}_{\alpha}} = m_{\alpha} \dot{\mathbf{q}}_{\alpha}, \quad (\text{A.11a})$$

$$\begin{aligned} \mathbf{\Pi}(\mathbf{r}) &= \frac{\partial L_{\text{mult}}}{\partial \dot{\mathbf{A}}(\mathbf{r})} = \frac{\partial L_{\text{rad}}}{\partial \dot{\mathbf{A}}(\mathbf{r})} - \frac{\partial}{\partial \dot{\mathbf{A}}(\mathbf{r})} \int \mathbf{P}^{\perp}(\mathbf{r}) \cdot \dot{\mathbf{A}}(\mathbf{r}) d^3r \\ &= \varepsilon_0 \dot{\mathbf{A}}(\mathbf{r}) - \mathbf{P}^{\perp}(\mathbf{r}) = -\varepsilon_0 \mathbf{E}^{\perp}(\mathbf{r}) - \mathbf{P}^{\perp}(\mathbf{r}). \end{aligned} \quad (\text{A.11b})$$

From the relation between the electric field  $\mathbf{E}(\mathbf{r})$  and electric displacement vector  $\mathbf{D}(\mathbf{r})$ , their transverse components also satisfy

$$\mathbf{D}^{\perp}(\mathbf{r}) = \varepsilon_0 \mathbf{E}^{\perp}(\mathbf{r}) + \mathbf{P}^{\perp}(\mathbf{r}) \quad (\text{A.12})$$

and thus the momentum  $\mathbf{\Pi}(\mathbf{r})$  can be rewritten as

$$\mathbf{\Pi}(\mathbf{r}) = -\mathbf{D}^{\perp}(\mathbf{r}). \quad (\text{A.13})$$

By putting them together, canonical transformation of the Lagrangian  $L_{mult}$  gives a new Hamiltonian  $H_{mult}$ :

$$\begin{aligned}
 H_{mult} &= \sum_{\zeta, \alpha} \mathbf{p}_\alpha(\zeta) \cdot \dot{\mathbf{q}}_\alpha(\zeta) + \int \boldsymbol{\Pi}(\mathbf{r}) \cdot \dot{\mathbf{A}}(\mathbf{r}) d^3r - L_{mult} \\
 &= \sum_{\zeta} \left\{ \sum_{\alpha} \frac{\mathbf{p}_\alpha^2(\zeta)}{2m_\alpha} + V(\zeta) \right\} + \left\{ \frac{1}{2} \int \left[ \frac{\boldsymbol{\Pi}^2(\mathbf{r})}{\varepsilon_0} + \varepsilon_0 c^2 (\nabla \times \mathbf{A}(\mathbf{r}))^2 \right] d^3r \right\} \\
 &\quad + \frac{1}{\varepsilon_0} \int \mathbf{P}^\perp(\mathbf{r}) \cdot \boldsymbol{\Pi}(\mathbf{r}) d^3r + \frac{1}{2\varepsilon_0} \int |\mathbf{P}^\perp(\mathbf{r})|^2 d^3r + V_{int}.
 \end{aligned} \tag{A.14}$$

It is possible to simplify Eq. (A.14) by separating  $(1/2\varepsilon_0) \int |\mathbf{P}^\perp(\mathbf{r})|^2 d^3r$  into two parts: an inter-nanomaterial part and an intra-nanomaterial part. In order to consider the inter-nanomaterial part

$$\frac{1}{2\varepsilon_0} \int \mathbf{P}_1^\perp(\mathbf{r}) \cdot \mathbf{P}_2^\perp(\mathbf{r}) d^3r, \tag{A.15}$$

by noting

$$\mathbf{P}_2(\mathbf{r}) = \mathbf{P}_2^\parallel(\mathbf{r}) + \mathbf{P}_2^\perp(\mathbf{r}), \mathbf{P}_1^\perp(\mathbf{r}) \cdot \mathbf{P}_2^\parallel(\mathbf{r}) = 0 \tag{A.16}$$

(the symbol  $\parallel$  represents the longitudinal component of the polarization, i.e., the component parallel to the wave-vector  $\mathbf{k}$ ) and

$$\mathbf{P}_1^\perp(\mathbf{r}) \cdot \mathbf{P}_2^\perp(\mathbf{r}) = \mathbf{P}_1^\perp(\mathbf{r}) \cdot \left\{ \mathbf{P}_2^\parallel(\mathbf{r}) + \mathbf{P}_2^\perp(\mathbf{r}) \right\} = \mathbf{P}_1^\perp(\mathbf{r}) \cdot \mathbf{P}_2(\mathbf{r}), \tag{A.17}$$

Equation (A.15) is rewritten as follows:

$$\begin{aligned}
 \frac{1}{\varepsilon_0} \int \mathbf{P}_1^\perp(\mathbf{r}) \cdot \mathbf{P}_2^\perp(\mathbf{r}) d^3r &= \frac{1}{\varepsilon_0} \int \mathbf{P}_1^\perp(\mathbf{r}) \cdot \mathbf{P}_2(\mathbf{r}) d^3r \\
 &= \frac{1}{\varepsilon_0} p_i(1) p_j(2) \int \delta_{ij}^\perp(\mathbf{r} - \mathbf{R}_1) \delta(\mathbf{r} - \mathbf{R}_2) d^3r \\
 &= \frac{1}{\varepsilon_0} p_i(1) p_j(2) \delta_{ij}^\perp(\mathbf{r} - \mathbf{R}_1 - \mathbf{R}_2) \\
 &= -\frac{p_i(1) p_j(2)}{4\pi\varepsilon_0 R^3} (\delta_{ij} - 3\hat{e}_{Ri}\hat{e}_{Rj}) \\
 &= -\frac{1}{4\pi\varepsilon_0 R^3} \{ \mathbf{p}(1) \cdot \mathbf{p}(2) - 3(\mathbf{p}(1) \cdot \mathbf{e}_R)(\mathbf{p}(2) \cdot \mathbf{e}_R) \},
 \end{aligned} \tag{A.18}$$

where  $\hat{e}_{Ri}$  and  $\hat{e}_{Rj}$  are the  $i$ -th and  $j$ -th Cartesian components of the unit vector  $\mathbf{e}_R$  ( $\equiv \mathbf{R}/R$ ). Equation (A.16) was used in the first row. Also, the following identities for the Dirac  $\delta$  function and the  $\delta$ -dyadics were used in the third line:

$$\begin{aligned}
\delta_{ij}\delta(\mathbf{r}) &= \delta_{ij}^{\parallel}(\mathbf{r}) + \delta_{ij}^{\perp}(\mathbf{r}), \\
\delta_{ij}^{\perp}(\mathbf{r}) &= -\delta_{ij}^{\parallel}(\mathbf{r}) = -\frac{1}{(2\pi)^3} \int \hat{e}_{ki}\hat{e}_{kj} \exp(\mathbf{i}\mathbf{k} \cdot \mathbf{r}) d^3r \\
&= \nabla_i \nabla_j \left( \frac{1}{4\pi r} \right) = -\frac{1}{4\pi r^3} (\delta_{ij} - 3\hat{e}_{ri}\hat{e}_{rj}).
\end{aligned} \tag{A.19}$$

Here,  $\hat{e}_{ki}$  and  $\hat{e}_{kj}$  are the  $i$ -th and  $j$ -th Cartesian components of the unit vector  $\mathbf{e}_k (\equiv \mathbf{k}/k)$ . Similarly,  $\hat{e}_{ri}$  and  $\hat{e}_{rj}$  are the  $i$ -th and  $j$ -th Cartesian components of the unit vector  $\mathbf{e}_r (\equiv \mathbf{r}/r)$ . Because exchanging the subscripts 1 and 2 gives the same result as Eq. (A.18), one can derive

$$\frac{1}{2\varepsilon_0} \int \mathbf{P}_1^{\perp}(\mathbf{r}) \cdot \mathbf{P}_2^{\perp} d^3r + V_{\text{int}} = 0 \tag{A.20}$$

by noting Eq. (A.3). This equation suggests that the inter-nanomaterial part given by Eq. (A.15) and  $V_{\text{int}}$  cancel each other out. Therefore, it is sufficient to consider the intra-nanomaterial part  $(1/2\varepsilon_0) \int |\mathbf{P}_{\zeta}^{\perp}(\mathbf{r})|^2 d^3r$  ( $\zeta = 1, 2$ ) only, and Eq. (A.14) can be simplified to

$$\begin{aligned}
H_{\text{mult}} &= \sum_{\zeta} \left\{ \sum_{\alpha} \frac{\mathbf{p}_{\alpha}^2(\zeta)}{2m_{\alpha}} + V(\zeta) + \frac{1}{2\varepsilon_0} \int |\mathbf{P}_{\zeta}^{\perp}(\mathbf{r})|^2 d^3r \right\} \\
&+ \left\{ \frac{1}{2} \int \left[ \frac{\mathbf{\Pi}^2(\mathbf{r})}{\varepsilon_0} + \varepsilon_0 c^2 (\nabla \times \mathbf{A}(\mathbf{r}))^2 \right] d^3r \right\} \\
&+ \frac{1}{\varepsilon_0} \int \mathbf{P}^{\perp}(\mathbf{r}) \cdot \mathbf{\Pi}(\mathbf{r}) d^3r,
\end{aligned} \tag{A.21}$$

where each row represents the charged particle motion in each nanomaterial, the free electromagnetic field, and the interaction, respectively. Because the polarization density  $\mathbf{P}^{\perp}(\mathbf{r})$  can be expanded in terms of  $2^l$  multipoles ( $l = 1, 2, 3, \dots$ ), as shown in Eq. (A.5),  $H_{\text{mult}}$  of Eq. (A.21) is called the multipolar Hamiltonian.

With the help of Eqs. (A.6) and (A.13), the interaction part in the third row of Eq. (A.21) can be more explicitly written as

$$\begin{aligned}
\frac{1}{\varepsilon_0} \int \mathbf{P}^{\perp}(\mathbf{r}) \cdot \mathbf{\Pi}(\mathbf{r}) d^3r &= -\frac{1}{\varepsilon_0} \int \mathbf{P}^{\perp}(\mathbf{r}) \cdot \mathbf{D}^{\perp}(\mathbf{r}) d^3r \\
&= -\frac{1}{\varepsilon_0} \int \mathbf{P}(\mathbf{r}) \cdot \mathbf{D}^{\perp}(\mathbf{r}) d^3r \\
&= -\frac{1}{\varepsilon_0} \left\{ \mathbf{p}(1) \cdot \mathbf{D}^{\perp}(\mathbf{R}_1) + \mathbf{p}(2) \cdot \mathbf{D}^{\perp}(\mathbf{R}_2) \right\}
\end{aligned} \tag{A.22}$$

by using the electric dipole moment  $\mathbf{p}$  and the electric displacement vector  $\mathbf{D}^{\perp}$ . The advantage of this expression is that electromagnetic phenomena occurring inside

and outside the nanomaterial can be treated equivalently, i.e., the anti-electric field inside the nanomaterial and Coulomb interaction between electrons do not have to be additionally considered, and thus, the static Coulomb interactions between the nanomaterials can be excluded. Further advantages are that the delay effect can be expressed because the interaction is described by the exchange of the transverse photons, and the origin of the electric-dipole–forbidden transition can be clearly interpreted.

When the system under study is quantized, quantities such as  $\mathbf{p}$  and  $\mathbf{D}^\perp$  should be replaced by the corresponding operators,

$$-\frac{1}{\varepsilon_0} \left\{ \hat{\mathbf{p}}(1) \cdot \hat{\mathbf{D}}^\perp(\mathbf{R}_1) + \hat{\mathbf{p}}(2) \cdot \hat{\mathbf{D}}^\perp(\mathbf{R}_2) \right\}, \quad (\text{A.23})$$

yielding the quantized multipolar Hamiltonian. The third term of Eq. (2.1), and therefore Eq. (2.25), is derived by replacing  $\hat{\mathbf{D}}^\perp(\mathbf{R}_1)$  and  $\hat{\mathbf{D}}^\perp(\mathbf{R}_2)$  by the electric displacement operators  $\hat{\mathbf{D}}^\perp(\mathbf{r}_s)$  and  $\hat{\mathbf{D}}^\perp(\mathbf{r}_p)$  at the positions of the electric charges  $\mathbf{r}_s$  and  $\mathbf{r}_p$ , respectively, and furthermore, by replacing  $\hat{\mathbf{p}}(1)$  and  $\hat{\mathbf{p}}(2)$  by  $\hat{\mathbf{p}}_s$  and  $\hat{\mathbf{p}}_p$ , respectively.

## References

1. C. Cohen-Tannoudji, J. Dupont-Roc, G. Grynberg, *Photons and Atoms* (Wiley, New York, 1989)
2. C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions* (John Wiley & Sons, New York, 1992)
3. D.P. Craig, T. Thirunamachandran, *Molecular Quantum Electrodynamics* (Dover, New York, 1998)

## Appendix B

# Elementary Excitation and Exciton-polariton

The concept of elementary excitations or quasi-particles has been discussed for a long time [1]. Excited states of a many-body system are considered as a collection of certain fundamental excited states that has been called an elementary excitation. The relation between the momentum  $\mathbf{p}$  and energy  $E$  of the elementary excitation, i.e.,  $E = E(\mathbf{p})$ , is referred to as the dispersion relation.

A well-known example of an elementary excitation in a solid is a phonon, which is a quantum of normal modes of crystal vibration. Its motion is collective, which means that the total number of phonons is independent of the number of crystal lattices. The momentum of the phonon is  $\mathbf{p} = \hbar\mathbf{k}$  in terms of the wave-vector  $\mathbf{k}$  of normal vibration, not the mechanical momentum of an individual crystal lattice. The energy is also given in terms of the angular frequency  $\omega$  of the normal vibration by  $E = \hbar\omega$ . Other examples of elementary excitations are plasmons, which correspond to the collective motion of electron density in interacting electron gas; polarons, which originate from the coupling between conduction electrons and optical phonons; and magnons, which correspond to collective modes of spin density waves.

Excitons, which describe the elementary excitation related to an electron-hole pair in a solid, are also well-known. As an extreme case, when the distance between the electron and hole in an exciton (Bohr radius of the exciton) is smaller than the interatomic distance in the crystal, it is called a Frenkel exciton; Wannier Excitons correspond to the opposite extreme case, in which the Bohr radius of the exciton is larger than the interatomic distance in the crystal.

In the following, the light-matter interaction in a macroscopic material is discussed on the basis of the exciton concept. The photon, incident on the macroscopic material is absorbed, creating the exciton. Afterward, this exciton is annihilated, creating a photon. This means that the successive creation and annihilation of the photon and exciton propagate through the macroscopic material. In the other words, the photon and exciton repeat their creation and annihilation in an out-of-phase manner, temporally and spatially. This process indicates that a new steady state with a new dispersion relation and energy is formed in the whole macroscopic material due to the photon-exciton interaction. Normal modes, or elementary excitation modes, representing this coupled oscillation are called polaritons. In particular, they are called

exciton-polaritons because they originate from the interaction between photons and excitons. The exciton-polariton is a coupled wave generated by the electromagnetic field and the polarization field of the exciton. The situation is analogous to the case in which two coupled oscillations with angular frequencies  $\omega_o$  and  $\omega_e$  produce new normal oscillations with angular frequencies  $\Omega_1$  and  $\Omega_2$ .

The following Hamiltonian is used to describe exciton-polaritons:

$$\hat{H} = \hbar\omega_o\hat{a}^\dagger\hat{a} + \hbar\omega_e\hat{b}^\dagger\hat{b} + \hbar D (\hat{a} + \hat{a}^\dagger) (\hat{b} + \hat{b}^\dagger). \quad (\text{B.1})$$

Since a macroscopic material is dealt with here, a virtual cavity can be defined for quantization. Therefore, the first term, i.e, the non-perturbed Hamiltonian for photons, is equivalent to the incident photon energy  $\hbar\omega_o$ , which is resonant with the virtual cavity. The second term corresponds to the non-perturbed Hamiltonian for excitons, having the eigenenergy  $\hbar\omega_e$ . The third term describes the photon–exciton interaction, whose interaction energy is  $\hbar D$ . The explicit expression for  $\hbar D$  is given by Eq. (2.27).  $\hat{a}$  and  $\hat{a}^\dagger$  are annihilation and creation operators for photons, respectively.  $\hat{b}$  and  $\hat{b}^\dagger$  are annihilation and creation operators for excitons, which are given by

$$\begin{cases} \hat{b} = \frac{1}{\sqrt{N}} \sum_l e^{-ik \cdot l} \hat{b}_l \\ \hat{b}^\dagger = \frac{1}{\sqrt{N}} \sum_l e^{ik \cdot l} \hat{b}_l^\dagger \end{cases} \quad (\text{B.2})$$

Here,  $N$  and  $\mathbf{k}$  are the total number of lattice sites and the wave-vector, respectively. Furthermore, we have

$$\hat{b}_l = \hat{e}_{l,c} \hat{h}_{l,v} \quad \text{and} \quad \hat{b}_l^\dagger = \hat{e}_{l,c}^\dagger \hat{h}_{l,v}^\dagger, \quad (\text{B.3})$$

where  $\hat{e}_{l,c}$  and  $\hat{e}_{l,c}^\dagger$  are the annihilation and creation operators, respectively, for the electron in the conduction band at the lattice site  $l$ , whereas  $\hat{h}_{l,v}$  and  $\hat{h}_{l,v}^\dagger$  are those for the hole in the valence band at the lattice site  $l$ .

From the Hamiltonian for the exciton-polariton given by Eq. (B.1), one can obtain the eigenstates and eigenenergies of the exciton-polaritons, or the dispersion relation. For simplicity, the rotating wave approximation is adopted to neglect terms  $\hat{a}^\dagger\hat{b}^\dagger$  and  $\hat{a}\hat{b}$ , which represent simultaneous creation or annihilation of a photon and an exciton, resulting in the following Hamiltonian:

$$\hat{H} = \hbar (\omega_o\hat{a}^\dagger\hat{a} + \omega_e\hat{b}^\dagger\hat{b}) + \hbar D (\hat{b}^\dagger\hat{a} + \hat{a}^\dagger\hat{b}). \quad (\text{B.4})$$

Next, exciton-polariton creation operators  $\hat{\xi}_1^\dagger$  and  $\hat{\xi}_2^\dagger$  and annihilation operators  $\hat{\xi}_1$  and  $\hat{\xi}_2$ , corresponding to new eigenfrequencies  $\Omega_1$  and  $\Omega_2$ , respectively, are derived. For this derivation, the Hamiltonian  $\hat{H}$  is assumed to be diagonalized in terms of these operators, and Eq. (B.4) is rewritten as



$$\begin{aligned}\hat{H} &= \hbar \left( \Omega_1 \hat{\xi}_1^\dagger \hat{\xi}_1 + \Omega_2 \hat{\xi}_2^\dagger \hat{\xi}_2 \right) = \hbar \left( \hat{b}^\dagger, \hat{a}^\dagger \right) A \begin{pmatrix} \hat{b} \\ \hat{a} \end{pmatrix} \\ &= \hbar \left( a_{11} \hat{b}^\dagger \hat{b} + a_{12} \hat{b}^\dagger \hat{a} + a_{21} \hat{a}^\dagger \hat{b} + a_{22} \hat{a}^\dagger \hat{a} \right),\end{aligned}\quad (\text{B.5})$$

where  $A$  is a  $2 \times 2$  matrix whose elements are given by

$$A = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} = \begin{pmatrix} \omega_e & D \\ D & \omega_o \end{pmatrix}. \quad (\text{B.6})$$

Applying unitary transformation  $U$

$$\begin{pmatrix} \hat{b} \\ \hat{a} \end{pmatrix} = U \begin{pmatrix} \hat{\xi}_1 \\ \hat{\xi}_2 \end{pmatrix} \text{ with } U = \begin{pmatrix} u_{11} & u_{12} \\ u_{21} & u_{22} \end{pmatrix} \quad (\text{B.7})$$

to Eq. (B.5) yields

$$\hbar \left( \hat{b}^\dagger, \hat{a}^\dagger \right) A \begin{pmatrix} \hat{b} \\ \hat{a} \end{pmatrix} = \hbar \left( \hat{\xi}_1^\dagger, \hat{\xi}_2^\dagger \right) U^\dagger A U \begin{pmatrix} \hat{\xi}_1 \\ \hat{\xi}_2 \end{pmatrix}. \quad (\text{B.8})$$

Since  $U^\dagger A U = U^{-1} A U$  is diagonalized, one can write

$$U^{-1} A U = \begin{pmatrix} \Omega_1 & 0 \\ 0 & \Omega_2 \end{pmatrix} \equiv \Lambda, \quad (\text{B.9})$$

and obtain  $AU = U\Lambda$ , which reduces to

$$\begin{pmatrix} \omega_e - \Omega_j & D \\ D & \omega_o - \Omega_j \end{pmatrix} \begin{pmatrix} u_{1j} \\ u_{2j} \end{pmatrix} = 0. \quad (\text{B.10})$$

This immediately gives the eigenvalue equation

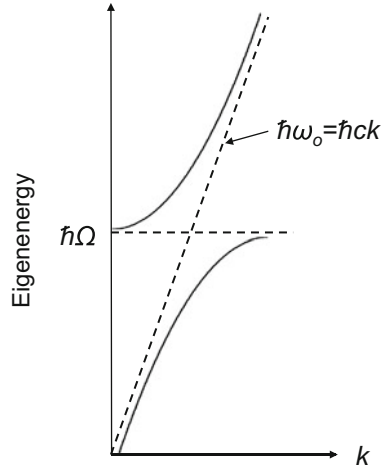
$$(\Omega_j - \omega_e)(\Omega_j - \omega_o) - D^2 = 0, \quad (\text{B.11})$$

and the eigenenergies of the exciton-polariton are

$$\hbar\Omega_j = \hbar \left[ \frac{\omega_e + \omega_o}{2} \pm \frac{\sqrt{(\omega_e - \omega_o)^2 + 4D^2}}{2} \right]. \quad (\text{B.12})$$

Equation (B.12) provides a new dispersion relation. Using the dispersion relation of photons  $\omega_o = ck$  with  $k = |\mathbf{k}|$ , the eigenenergies of the exciton-polariton can be plotted as a function of  $k$ , as shown in Fig. B.1. Here, for simplicity, the exciton dispersion is assumed to be  $\hbar\omega_e = \hbar\Omega$ , which is independent of  $k$ . From Eq. (B.10) and the unitarity of  $U$ , the components of the eigenvectors are given by

**Fig. B.1** Relation between the wavenumber  $k$  and the eigenenergy



$$\begin{cases} u_{2j} = -\frac{\omega_e - \Omega_j}{D} u_{1j} & (j = 1, 2), \\ u_{1j}^2 + u_{2j}^2 = 1 \end{cases} \quad (\text{B.13})$$

which thus reads

$$\left\{ 1 + \left( \frac{\omega_e - \Omega_j}{D} \right)^2 \right\} u_{1j}^2 = 1. \quad (\text{B.14})$$

Finally, the eigenvectors of the exciton-polariton are given by

$$\begin{cases} u_{1j} = \left\{ 1 + \left( \frac{\omega_e - \Omega_j}{D} \right)^2 \right\}^{-1/2}, \\ u_{2j} = -\left( \frac{\omega_e - \Omega_j}{D} \right) \left\{ 1 + \left( \frac{\omega_e - \Omega_j}{D} \right)^2 \right\}^{-1/2}. \end{cases} \quad (\text{B.15})$$

New steady states for the exciton-polariton can be described by Eqs. (B.12) and (B.15).

Equation (B.12) means that the sum of  $\hbar\Omega_1$  and  $\hbar\Omega_2$  is equal to the sum of the exciton and photon energies,  $\hbar(\omega_e + \omega_o)$ , because annihilation and creation of the photon occur in an out-of-phase manner to those of the exciton, as was pointed out at the beginning of this appendix. Furthermore, since the classical model of the exciton-polariton is a coupled wave of the electromagnetic wave of the light and the polarization wave of the exciton, the dependence of its amplitude on time  $t$  and position  $\mathbf{x}$  is simply expressed by a complex sinusoidal function  $\exp[i(\Omega_j t - \mathbf{k} \cdot \mathbf{x})]$ , where  $\Omega_j$  is the angular frequency in Eq. (B.12). Therefore, if the numbers  $n$  of photons and excitons are greater than one, the angular frequency of the coupled wave remains  $\hbar\Omega_j$ , even though the square of the amplitude of the coupled wave becomes  $n$  times greater than that at  $n = 1$ .

## Reference

1. D. Pines, *Elementary Excitation in Solids* (Perseus Books, Reading, Massachusetts, 1999)

# Appendix C

## Projection Operator and Effective Interaction Operator

### C.1 Projection Operator

The total Hamiltonian  $\hat{H}$  for the light–matter interaction system is given by

$$\hat{H} = \hat{H}_0 + \hat{V}, \tag{C.1}$$

where  $\hat{H}_0$  and  $\hat{V}$  describe the unperturbed and interaction Hamiltonians, respectively. The state  $|\phi_{pj}\rangle$  in Eq. (2.32) is the eigenstate of  $\hat{H}_0$ . If the eigenstate and eigenenergy of  $\hat{H}$  are written as  $|\psi_j\rangle$  and  $E_j$ , respectively, the following Schrödinger equation holds:

$$\hat{H} |\psi_j\rangle = E_j |\psi_j\rangle, \tag{C.2}$$

where the subscript  $j$  is used as a quantum number to distinguish each eigenstate. In a similar way, by denoting the eigenstate of  $\hat{H}_0$  by  $|\phi_j\rangle$ , a projection operator  $\hat{P}$  is defined as

$$\hat{P} = \sum_j |\phi_j\rangle\langle\phi_j|. \tag{C.3}$$

Applying the projection operator to an arbitrary state  $|\psi\rangle$  yields

$$\hat{P} |\psi\rangle = \sum_j |\phi_j\rangle\langle\phi_j | \psi\rangle. \tag{C.4}$$

The right-hand side of this equation is represented by the linear superposition of the eigenstates  $|\phi_j\rangle$  because the inner product  $\langle\phi_j | \psi\rangle$  is a constant. This suggests that the projection operator transforms the arbitrary state  $|\psi\rangle$  into the P space spanned by the eigenstate  $|\phi_j\rangle$ . In Eq. (C.3), the projection operator was defined based on steady states of the Schrödinger equation. The time-dependent approach of the projection operator method is reviewed in Ref. [1].

Because the eigenstate  $|\phi_j\rangle$  is orthonormalized, the projection operator  $\hat{P}$  satisfies the following relation:

$$\hat{P} = \hat{P}^\dagger, \quad (\text{C.5a})$$

$$\hat{P}^2 = \hat{P}, \quad (\text{C.5b})$$

where  $\hat{P}^\dagger$  is the Hermitian conjugate operator of  $\hat{P}$ . Equation (C.5a) means that  $\hat{P}$  is a Hermitian operator.

The complimentary operator  $\hat{Q}$  given by

$$\hat{Q} = 1 - \hat{P} \quad (\text{C.6a})$$

reads

$$\hat{Q} = \hat{Q}^\dagger, \quad (\text{C.6b})$$

$$\hat{Q}^2 = \hat{Q}. \quad (\text{C.6c})$$

Any state in the P space is orthogonal to any state in the Q space, and thus one has

$$\hat{P}\hat{Q} = \hat{Q}\hat{P} = 0. \quad (\text{C.7})$$

Noting that  $|\phi_j\rangle$  is an eigenstate of  $\hat{H}_0$ , the commutations between the projection operators and  $\hat{H}_0$  are

$$[\hat{P}, \hat{H}_0] = \hat{P}\hat{H}_0 - \hat{H}_0\hat{P} = 0, \quad (\text{C.8a})$$

$$[\hat{Q}, \hat{H}_0] = \hat{Q}\hat{H}_0 - \hat{H}_0\hat{Q} = 0. \quad (\text{C.8b})$$

## C.2 Effective Interaction Operator

The expectation value of an arbitrary physical quantity is expressed as  $\langle\psi|\hat{O}|\psi\rangle$ , where  $\hat{O}$  is the corresponding operator, and  $|\psi\rangle$  is the state of the system under discussion. In order to derive this value using only the states in the P space, i.e., to express the expectation value as  $\langle\phi_i|\hat{O}_{eff}|\phi_j\rangle$ , a new operator  $\hat{O}_{eff}$ , called an effective operator, should be derived.

The following discussion considers the eigenstates  $|\psi_j\rangle$  of  $\hat{H}$  instead of the arbitrary state  $|\psi\rangle$  because  $|\psi\rangle$  is given by the linear superposition of  $|\psi_j\rangle$  ( $j = 1, 2, 3, \dots$ ). The eigenstates  $|\psi_j\rangle$  are divided into two groups, the state  $|\psi_j^{(P)}\rangle$  in the P space and the state  $|\psi_j^{(Q)}\rangle$  in the Q space, which are defined as follows:

$$|\psi_j^{(P)}\rangle = \hat{P}|\psi_j\rangle, \quad |\psi_j^{(Q)}\rangle = \hat{Q}|\psi_j\rangle. \quad (\text{C.9})$$

By noting that  $\hat{P} + \hat{Q} = 1$ , we have

$$|\psi_j\rangle = (\hat{P} + \hat{Q}) |\psi_j\rangle = \hat{P} |\psi_j\rangle + \hat{Q} |\psi_j\rangle = |\psi_j^{(P)}\rangle + |\psi_j^{(Q)}\rangle, \quad (\text{C.10})$$

and from Eqs. (C.5b) and (C.6c), we have

$$\hat{P} |\psi_j^{(P)}\rangle = \hat{P} \hat{P} |\psi_j\rangle = \hat{P} |\psi_j\rangle = |\psi_j^{(P)}\rangle, \quad (\text{C.11a})$$

$$\hat{Q} |\psi_j^{(Q)}\rangle = \hat{Q} \hat{Q} |\psi_j\rangle = \hat{Q} |\psi_j\rangle = |\psi_j^{(Q)}\rangle. \quad (\text{C.11b})$$

Inserting Eqs. (C.11a) and (C.11b) into Eq. (C.10) yields

$$|\psi_j\rangle = \hat{P} |\psi_j^{(P)}\rangle + \hat{Q} |\psi_j^{(Q)}\rangle. \quad (\text{C.12})$$

Since Eqs. (C.1) and (C.2) give

$$(E_j - \hat{H}_0) |\psi_j\rangle = \hat{V} |\psi_j\rangle, \quad (\text{C.13a})$$

inserting Eq. (C.12) into Eq. (C.13a) yields

$$(E_j - \hat{H}_0) \hat{P} |\psi_j^{(P)}\rangle + (E_j - \hat{H}_0) \hat{Q} |\psi_j^{(Q)}\rangle = \hat{V} \hat{P} |\psi_j^{(P)}\rangle + \hat{V} \hat{Q} |\psi_j^{(Q)}\rangle. \quad (\text{C.13b})$$

Applying  $\hat{P}$  to Eq. (C.13b) from the left and using Eqs. (C.5b) and (C.7) gives

$$(E_j - \hat{H}_0) \hat{P} |\psi_j^{(P)}\rangle = \hat{P} \hat{V} \hat{P} |\psi_j^{(P)}\rangle + \hat{P} \hat{V} \hat{Q} |\psi_j^{(Q)}\rangle. \quad (\text{C.14})$$

Similarly applying  $\hat{Q}$  to Eq. (C.13b) from the left and using Eqs. (C.6b) and (C.7), Eq. (C.13b) can be rewritten as

$$(E_j - \hat{H}_0) \hat{Q} |\psi_j^{(Q)}\rangle = \hat{Q} \hat{V} \hat{P} |\psi_j^{(P)}\rangle + \hat{Q} \hat{V} \hat{Q} |\psi_j^{(Q)}\rangle. \quad (\text{C.15})$$

By moving the second term on the right-hand side of Eq. (C.15) to the left-hand side, it is possible to formally express  $\hat{Q} |\psi_j^{(Q)}\rangle$  as

$$\begin{aligned} \hat{Q} |\psi_j^{(Q)}\rangle &= (E_j - \hat{H}_0 - \hat{Q} \hat{V})^{-1} \hat{Q} \hat{V} \hat{P} |\psi_j^{(P)}\rangle \\ &= \left\{ (E_j - \hat{H}_0) \left[ 1 - (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \right] \right\}^{-1} \hat{Q} \hat{V} \hat{P} |\psi_j^{(P)}\rangle \\ &= \hat{J} (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \hat{P} |\psi_j^{(P)}\rangle, \end{aligned} \quad (\text{C.16})$$

where the operator  $\hat{J}$  is defined by

$$\hat{J} = \left[ 1 - (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \right]^{-1}. \quad (\text{C.17})$$

By inserting Eq. (C.16) into Eq. (C.14), one obtains the following equation for  $\hat{P} |\psi_j^{(P)}\rangle$ :

$$\begin{aligned} (E_j - \hat{H}_0) \hat{P} |\psi_j^{(P)}\rangle &= \hat{P} \hat{V} \hat{P} |\psi_j^{(P)}\rangle + \hat{P} \hat{V} \hat{J} (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \hat{P} |\psi_j^{(P)}\rangle \\ &= \hat{P} \hat{V} \hat{J} \left\{ \hat{J}^{-1} + (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \right\} \hat{P} |\psi_j^{(P)}\rangle. \end{aligned} \quad (\text{C.18})$$

Since

$$\hat{J}^{-1} = 1 - (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \quad (\text{C.19})$$

is derived from Eq. (C.17), inserting this equation into {} on the second row of Eq. (C.18) yields

$$(E_j - \hat{H}_0) \hat{P} |\psi_j^{(P)}\rangle = \hat{P} \hat{V} \hat{J} \hat{P} |\psi_j^{(P)}\rangle, \quad (\text{C.20})$$

which is the equation that  $|\psi_j^{(P)}\rangle$  must satisfy. On the other hand, inserting Eq. (C.16) into the second term on the right-hand side of Eq. (C.12) yields the following equation for  $|\psi_j\rangle$ :

$$\begin{aligned} |\psi_j\rangle &= \hat{P} |\psi_j^{(P)}\rangle + \hat{J} (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \hat{P} |\psi_j^{(P)}\rangle \\ &= \hat{J} \left\{ \hat{J}^{-1} + (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \right\} \hat{P} |\psi_j^{(P)}\rangle \\ &= \hat{J} \hat{P} |\psi_j^{(P)}\rangle, \end{aligned} \quad (\text{C.21})$$

where Eq. (C.19) was used to derive the third row.

Noting the normalization condition for  $|\psi_j\rangle$ , inserting Eq. (C.21) into  $\langle \psi_j | \psi_j \rangle = 1$  gives

$$\langle \psi_j^{(P)} | \hat{P} \hat{J}^\dagger \hat{J} \hat{P} | \psi_j^{(P)} \rangle = 1. \quad (\text{C.22a})$$

This can be rewritten as

$$\langle \psi_j^{(P)} | \left( \hat{P} \hat{J}^\dagger \hat{J} \hat{P} \right)^{1/2} \left( \hat{P} \hat{J}^\dagger \hat{J} \hat{P} \right)^{1/2} | \psi_j^{(P)} \rangle = 1, \quad (\text{C.22b})$$

which suggests that  $\left(\hat{P}\hat{J}^\dagger\hat{J}\hat{P}\right)^{-1/2}|\psi_j^{(P)}\rangle$  should be considered as  $|\psi_j^{(P)}\rangle$  in order to normalize  $|\psi_j^{(P)}\rangle$ . Following this suggestion, Eq. (C.21) can be rewritten as

$$|\psi_j\rangle = \hat{J}\hat{P}\left(\hat{P}\hat{J}^\dagger\hat{J}\hat{P}\right)^{-1/2}|\psi_j^{(P)}\rangle, \quad (\text{C.22c})$$

where  $|\psi_j^{(P)}\rangle$  has already been normalized, as was described above.

Using Eq. (C.22c), one can derive the effective operator  $\hat{O}_{eff}$  for an arbitrary operator  $\hat{O}$  by the following relation [2-4]:

$$\langle\psi_i|\hat{O}|\psi_j\rangle = \langle\psi_i^{(P)}|\hat{O}_{eff}|\psi_j^{(P)}\rangle. \quad (\text{C.23})$$

Inserting Eq. (C.22c) into the left-hand side of Eq.(C.23) and comparing it with the right-hand side leads to

$$\hat{O}_{eff} = \left(\hat{P}\hat{J}^\dagger\hat{J}\hat{P}\right)^{-1/2} \left(\hat{P}\hat{J}^\dagger\hat{O}\hat{J}\hat{P}\right) \left(\hat{P}\hat{J}^\dagger\hat{J}\hat{P}\right)^{-1/2}. \quad (\text{C.24})$$

By replacing  $\hat{O}$  with the bare interaction operator  $\hat{V}$  in Eq. (C.1), the effective interaction operator  $\hat{V}_{eff}$  is written as

$$\hat{V}_{eff} = \left(\hat{P}\hat{J}^\dagger\hat{J}\hat{P}\right)^{-1/2} \left(\hat{P}\hat{J}^\dagger\hat{V}\hat{J}\hat{P}\right) \left(\hat{P}\hat{J}^\dagger\hat{J}\hat{P}\right)^{-1/2}. \quad (\text{C.25})$$

This is what we are searching for. Once the bare interaction operator  $\hat{V}$  is given, it only remains to obtain the unknown operator  $\hat{J}$  for deriving  $\hat{V}_{eff}$ .

In order to obtain an explicit form of the operator  $\hat{J}$ , the states  $|\phi_{pj}\rangle$  in the P space and their eigenvalues have to be used. For this purpose, the operator  $[\hat{J}, \hat{H}_0]\hat{P}$  is considered and is applied to  $|\psi_j\rangle$ . This yields

$$\begin{aligned} [\hat{J}, \hat{H}_0]\hat{P}|\psi_j\rangle &= (\hat{J}\hat{H}_0 - \hat{H}_0\hat{J})\hat{P}|\psi_j\rangle \\ &= \left\{ (E_j - \hat{H}_0)\hat{J} - \hat{J}(E_j - \hat{H}_0) \right\} \hat{P}|\psi_j\rangle. \end{aligned} \quad (\text{C.26a})$$

Using Eqs. (C.1), (C.2), (C.11a), and (C.21), the first term  $(E_j - \hat{H}_0)$  on the second row is replaced by  $\hat{V}$ , which gives

$$[\hat{J}, \hat{H}_0]\hat{P}|\psi_j\rangle = \hat{V}\hat{J}\hat{P}|\psi_j\rangle - \hat{J}(E_j - \hat{H}_0)\hat{P}|\psi_j\rangle. \quad (\text{C.26b})$$



Also, by using Eqs. (C.1), (C.2), (C.11a), and (C.12), the second term of Eq. (C.26b), except the operator  $\hat{J}$ , can be rewritten as

$$\begin{aligned} (E_j - \hat{H}_0) \hat{P} |\psi_j\rangle &= (E_j - \hat{H}_0) \hat{P} |\psi_j^{(P)}\rangle \\ &= \hat{P} \hat{V} \hat{P} |\psi_j^{(P)}\rangle + \hat{P} \hat{V} \hat{Q} |\psi_j^{(Q)}\rangle. \end{aligned} \quad (\text{C.27a})$$

Inserting Eq. (C.16) into  $\hat{Q} |\psi_j^{(Q)}\rangle$  in the second term on the second row gives

$$\begin{aligned} (E_j - \hat{H}_0) \hat{P} |\psi_j\rangle &= \hat{P} \hat{V} \hat{P} |\psi_j^{(P)}\rangle + \hat{P} \hat{V} \hat{J} (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \hat{P} |\psi_j^{(P)}\rangle \\ &= \hat{P} \hat{V} \hat{J} \left\{ \hat{J}^{-1} + (E_j - \hat{H}_0)^{-1} \hat{Q} \hat{V} \right\} \hat{P} |\psi_j^{(P)}\rangle. \end{aligned} \quad (\text{C.27b})$$

Furthermore, inserting Eq. (C.19) into the second row, one can rewrite it as

$$(E_j - \hat{H}_0) \hat{P} |\psi_j\rangle = \hat{P} \hat{V} \hat{J} \hat{P} |\psi_j^{(P)}\rangle. \quad (\text{C.27c})$$

Since

$$(E_j - \hat{H}_0) \hat{P} |\psi_j\rangle = \hat{P} \hat{V} \hat{J} \hat{P} |\psi_j\rangle \quad (\text{C.27d})$$

is obtained by inserting Eq. (C.11a) into the right-hand side, inserting Eq. (C.27d) into the second term on the right-hand side of Eq. (C.26b) gives

$$[\hat{J}, \hat{H}_0] \hat{P} |\psi_j\rangle = \hat{V} \hat{J} \hat{P} |\psi_j\rangle - \hat{J} \hat{P} \hat{V} \hat{J} \hat{P} |\psi_j\rangle. \quad (\text{C.28a})$$

Therefore, for the operator  $\hat{J}$ , we have

$$[\hat{J}, \hat{H}_0] \hat{P} = \hat{V} \hat{J} \hat{P} - \hat{J} \hat{P} \hat{V} \hat{J} \hat{P}, \quad (\text{C.28b})$$

where all operators involved are known, except  $\hat{J}$ .

### C.3 Approximate Expression

In order to solve Eq. (C.28b) perturbatively, a polynomial form

$$\hat{J} = \sum_{n=0}^{\infty} g^n \hat{J}^{(n)} \quad (\text{C.29})$$

is assumed, where  $\hat{J}^{(n)}$  contains  $n$  operators  $\hat{V}$ . First, by noting that the first term of Eq. (C.17) is unity, and that the relation  $1 = \hat{P} + \hat{Q}$  holds due to Eq. (C.6a), one finds that

$$\hat{J}^{(0)} = \hat{P}. \quad (\text{C.30})$$

Next, by inserting Eqs. (C.29) and (C.30) into Eq. (C.28b) and equating terms of order  $g^n$  on both sides, one successively obtains  $\hat{J}^{(1)}, \hat{J}^{(2)}, \dots, \hat{J}^{(n)}$ . For example, by multiplying both sides of Eq. (C.28b) by  $\hat{Q}$  from their left, the relation

$$\hat{Q} \left[ \hat{J}^{(1)}, \hat{H}_0 \right] \hat{P} = \hat{Q} \hat{V} \hat{J}^{(0)} \hat{P} - \hat{Q} \hat{J}^{(0)} \hat{P} \hat{V} \hat{J}^{(0)} \hat{P} \quad (\text{C.31a})$$

is obtained. By inserting Eq. (C.30) into this equation, it is written as

$$\hat{Q} \left[ \hat{J}^{(1)}, \hat{H}_0 \right] \hat{P} = \hat{Q} \hat{V} \hat{P}^2 - \hat{Q} \hat{P}^2 \hat{V} \hat{P}^2 = \hat{Q} \hat{V} \hat{P}, \quad (\text{C.31b})$$

where Eqs. (C.5b) and (C.7) were used for deriving the right-hand side of this equation. The matrix element of Eq. (C.31b) with  $\langle \psi_i |$  and  $|\psi_j\rangle$  can be written as

$$\langle \psi_i | \hat{Q} \left[ \hat{J}^{(1)}, \hat{H}_0 \right] \hat{P} | \psi_j \rangle = \langle \psi_i | \hat{Q} \hat{V} \hat{P} | \psi_j \rangle. \quad (\text{C.32})$$

By noting

$$\hat{H}_0 \hat{P} | \psi_j \rangle = \hat{H}_0 \hat{P} | \psi_j^{(P)} \rangle = \hat{P} \hat{H}_0 | \psi_j^{(P)} \rangle = \hat{P} E_P^0 | \psi_j^{(P)} \rangle = E_P^0 \hat{P} | \psi_j \rangle, \quad (\text{C.33a})$$

$$\hat{H}_0 \hat{Q} | \psi_j \rangle = \hat{H}_0 \hat{Q} | \psi_j^{(Q)} \rangle = \hat{Q} \hat{H}_0 | \psi_j^{(Q)} \rangle = \hat{Q} E_Q^0 | \psi_j^{(Q)} \rangle = E_Q^0 \hat{Q} | \psi_j \rangle, \quad (\text{C.33b})$$

the left-hand side of Eq. (C.32) is rewritten as

$$\begin{aligned} \langle \psi_i | \hat{Q} \left( \hat{J}^{(1)} \hat{H}_0 - \hat{H}_0 \hat{J}^{(1)} \right) \hat{P} | \psi_j \rangle &= \langle \psi_i | \left( \hat{Q} \hat{J}^{(1)} E_P^0 \hat{P} - \hat{Q} E_Q^0 \hat{J}^{(1)} \hat{P} \right) | \psi_j \rangle \\ &= \langle \psi_i | \left\{ \hat{Q} \hat{J}^{(1)} \left( E_P^0 - E_Q^0 \right) \hat{P} \right\} | \psi_j \rangle. \end{aligned} \quad (\text{C.34})$$

On the other hand, the right-hand side of Eq. (C.32) is rewritten as

$$\langle \psi_i | \hat{Q} \hat{V} \hat{P} | \psi_j \rangle = \langle \psi_i | \hat{Q}^2 \hat{V} \hat{P}^2 | \psi_j \rangle \quad (\text{C.35})$$

by using Eqs. (C.5b) and (C.6c). Inserting them into Eq. (C.32) and comparing both sides yields

$$\hat{Q} \hat{J}^{(1)} \left( E_P^0 - E_Q^0 \right) \hat{P} = \hat{Q}^2 \hat{V} \hat{P}^2 \quad (\text{C.36})$$

Thus,  $\hat{J}^{(1)}$  is

$$\hat{J}^{(1)} = \hat{Q}\hat{V}(E_P^0 - E_Q^0)^{-1}\hat{P}, \quad (\text{C.37})$$

which contains one  $\hat{V}$ . Higher orders of  $\hat{J}^{(n)}$  are successively given in a similar way.

#### C.4 Derivation of Eq. (2.30)

Under the first-order approximation  $\hat{J} \simeq \hat{J}^{(0)} + \hat{J}^{(1)}$ , Eq. (C.25) is expressed as

$$\hat{V}_{eff} \simeq \hat{P}\hat{J}^\dagger\hat{V}\hat{J}\hat{P} \simeq \hat{P}\hat{J}^{(0)\dagger}\hat{V}\hat{J}^{(1)}\hat{P} + \hat{P}\hat{J}^{(1)\dagger}\hat{V}\hat{J}^{(0)}\hat{P}. \quad (\text{C.38})$$

Inserting Eqs. (C.30), (C.37), and  $\hat{J}^{(1)\dagger} = \hat{P}(E_P^0 - E_Q^0)^{-1}\hat{V}\hat{Q}$  into this equation gives

$$\begin{aligned} \hat{V}_{eff} &\simeq \hat{P}\hat{V}\hat{Q}\hat{V}\left(\frac{1}{E_P^0 - E_Q^0}\right)\hat{P}\hat{P} + \hat{P}\hat{P}\left(\frac{1}{E_P^0 - E_Q^0}\right)\hat{V}\hat{Q}\hat{V}\hat{P} \\ &= \hat{P}\hat{V}\hat{Q}\hat{V}\left(\frac{1}{E_P^0 - E_Q^0}\right)\hat{P} + \hat{P}\left(\frac{1}{E_P^0 - E_Q^0}\right)\hat{V}\hat{Q}\hat{V}\hat{P}, \end{aligned} \quad (\text{C.39})$$

where Eq. (C.5b) was used to derive the second row. Equation (C.39) is rewritten by using Eq. (C.6c) as

$$\hat{V}_{eff} = \hat{P}\hat{V}\hat{Q} \cdot \hat{Q}\hat{V}\left(\frac{1}{E_P^0 - E_Q^0}\right)\hat{P} + \hat{P}\left(\frac{1}{E_P^0 - E_Q^0}\right)\hat{V}\hat{Q} \cdot \hat{Q}\hat{V}\hat{P}, \quad (\text{C.40})$$

where  $\hat{Q}\hat{V}\hat{P}$  and  $\hat{P}\hat{V}\hat{Q}$  mean that  $\hat{V}$  is screened in the P and Q spaces. Using this equation, the effective interaction energy of Eq. (2.29) is given by

$$\begin{aligned} V_{eff} = \langle \phi_{Pf} | &\left\{ \hat{P}\hat{V}\hat{Q} \cdot \hat{Q}\hat{V}\left(\frac{1}{E_P^0 - E_Q^0}\right)\hat{P} \right. \\ &\left. + \hat{P}\left(\frac{1}{E_P^0 - E_Q^0}\right)\hat{V}\hat{Q} \cdot \hat{Q}\hat{V}\hat{P} \right\} | \phi_{Pi} \rangle. \end{aligned} \quad (\text{C.41})$$

The operator  $\hat{P}\hat{V}\hat{Q}$  to the left of the symbol  $\cdot$  in the first term is considered to apply to  $\langle \phi_{Pf} |$ . On the other hand, the operator  $\hat{Q}\hat{V}(E_P^0 - E_Q^0)^{-1}\hat{P}$  to the right applies to  $|\phi_{Pi}\rangle$ . Under these considerations, the eigenenergies  $E_P^0$  and  $E_Q^0$  in this term are rewritten as  $E_{P_i}^0$  and  $E_{Q_j}^0$ , respectively. Finally, the first term is transformed to

$$\begin{aligned}
& \langle \phi_{Pf} | \hat{P} \hat{V} \hat{Q} \cdot \hat{Q} \hat{V} \left( \frac{1}{E_P^0 - E_Q^0} \right) \hat{P} | \phi_{Pi} \rangle \\
& = \langle \phi_{Pf} | \hat{P} \hat{V} \hat{Q} \cdot \hat{Q} \hat{V} \hat{P} | \phi_{Pi} \rangle \left( \frac{1}{E_{Pi}^0 - E_{Qj}^0} \right).
\end{aligned} \tag{C.42a}$$

Similarly for the second term, the eigenenergies  $E_P^0$  and  $E_Q^0$  are rewritten as  $E_{Pf}^0$  and  $E_{Qj}^0$ , respectively, which transforms the second term to

$$\begin{aligned}
& \langle \phi_{Pf} | \hat{P} \left( \frac{1}{E_{Pf}^0 - E_{Qj}^0} \right) \hat{V} \hat{Q} \cdot \hat{Q} \hat{V} \hat{P} | \phi_{Pi} \rangle \\
& = \left( \frac{1}{E_{Pf}^0 - E_{Qj}^0} \right) \langle \phi_{Pf} | \hat{P} \hat{V} \hat{Q} \cdot \hat{Q} \hat{V} \hat{P} | \phi_{Pi} \rangle.
\end{aligned} \tag{C.42b}$$

This equation means that Eq. (C.41) was successfully expressed by the screened operators  $\hat{Q} \hat{V} \hat{P}$  and  $\hat{P} \hat{V} \hat{Q}$ .

By noting that the unit operator  $\hat{1}$  is expressed by using the basis  $\{|\phi_{Qj}\rangle\}$  of the Q space as  $\hat{1} = \sum_j |\phi_{Qj}\rangle \langle \phi_{Qj}|$ ,<sup>2</sup> one can derive

$$V_{eff} = \langle \phi_{Pf} | \sum_j \hat{P} \hat{V} \hat{Q} | \phi_{Qj} \rangle \langle \phi_{Qj} | \hat{Q} \hat{V} \hat{P} | \phi_{Pi} \rangle \left( \frac{1}{E_{Pi}^0 - E_{Qj}^0} + \frac{1}{E_{Pf}^0 - E_{Qj}^0} \right), \tag{C.43}$$

and thus

$$\hat{V}_{eff} = \sum_j \hat{P} \hat{V} \hat{Q} | \phi_{Qj} \rangle \langle \phi_{Qj} | \hat{Q} \hat{V} \hat{P} \left( \frac{1}{E_{Pi}^0 - E_{Qj}^0} + \frac{1}{E_{Pf}^0 - E_{Qj}^0} \right). \tag{C.44}$$

These two equations are nothing but Eqs. (2.31) and (2.30).

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<sup>2</sup> The subscript  $j = 1$  represents the states of Eqs. (2.37a) and (2.37b), containing one exciton-polariton state  $|1_{(M)}\rangle$ . Other values of  $j$  represent the states of Eq. (2.36a) and (2.36b), which contain  $|n_{(M)}\rangle$ .

# Appendix D

## Transformation from Photon Base to Polariton Base

In this appendix, Eq. (2.28) is derived by diagonalizing the photon–exciton interaction Hamiltonian. Here, the Hamiltonian of the system is expressed as

$$\hat{H} \equiv \sum_k \hat{H}_k, \hat{H}_k = \hbar\omega_k \hat{a}_k^\dagger \hat{a}_k + \hbar\Omega \hat{b}_k^\dagger \hat{b}_k - i\hbar C \left( \hat{b}_{-k} + \hat{b}_k^\dagger \right) \left( \hat{a}_k - \hat{a}_{-k}^\dagger \right), \quad (\text{D.1})$$

where  $\hat{a}_k$  and  $\hat{a}_k^\dagger$  are annihilation and creation operators of a phonon with energy  $\hbar\omega_k$ , and  $\hat{b}_k$  and  $\hat{b}_k^\dagger$  are annihilation and creation operators of an exciton with energy  $\hbar\Omega$ . The photon–exciton interaction energy is denoted by  $\hbar C$ . An exciton–polariton operator  $\hat{\xi}_k$  is defined as

$$\hat{\xi}_k = W_k \hat{a}_k + Y_k \hat{a}_{-k}^\dagger + X_k \hat{b}_k + Z_k \hat{b}_{-k}^\dagger, \quad (\text{D.2})$$

where  $W_k$ ,  $Y_k$ ,  $X_k$ , and  $Z_k$  are the expansion coefficients. It is assumed that  $\hat{\xi}_k$  and its Hermitian conjugate  $\hat{\xi}_k^\dagger$  obey the boson commutation relation. The Hamiltonian should be diagonalized in the form of  $\hat{\xi}_k^\dagger \hat{\xi}_k$ , and it follows that

$$\hat{H}_k = \hbar\Omega (k) \hat{\xi}_k^\dagger \hat{\xi}_k, \quad (\text{D.3})$$

and Heisenberg’s equation of motion

$$-i \frac{d\hat{\xi}_k}{dt} = \frac{1}{\hbar} \left[ \hat{H}, \hat{\xi}_k \right] = -\Omega (k) \hat{\xi}_k. \quad (\text{D.4})$$

Substituting Eq. (D.2) into Eq. (D.4), the left-hand side is written as

$$-iW_k \frac{d\hat{a}_k}{dt} - iY_k \frac{d\hat{a}_{-k}^\dagger}{dt} - iX_k \frac{d\hat{b}_k}{dt} - iZ_k \frac{d\hat{b}_{-k}^\dagger}{dt} \quad (\text{D.5})$$

and the right-hand side as

$$-\Omega(k) \left( W_k \hat{a}_k + Y_k \hat{a}_{-k}^\dagger + X_k \hat{b}_k + Z_k \hat{b}_{-k}^\dagger \right). \quad (\text{D.6})$$

Using the following Heisenberg's equations of motion

$$-i \frac{d\hat{a}_k}{dt} = \frac{1}{\hbar} [\hat{H}, \hat{a}_k] = -\omega_k \hat{a}_k - iC \left( \hat{b}_k + \hat{b}_{-k}^\dagger \right), \quad (\text{D.7a})$$

$$-i \frac{d\hat{a}_{-k}^\dagger}{dt} = \frac{1}{\hbar} [\hat{H}, \hat{a}_{-k}^\dagger] = \omega_k \hat{a}_{-k}^\dagger - iC \left( \hat{b}_k + \hat{b}_{-k}^\dagger \right), \quad (\text{D.7b})$$

$$-i \frac{d\hat{b}_k}{dt} = \frac{1}{\hbar} [\hat{H}, \hat{b}_k] = -\Omega \hat{b}_k + iC \left( \hat{a}_k - \hat{a}_{-k}^\dagger \right), \quad (\text{D.7c})$$

$$-i \frac{d\hat{b}_{-k}^\dagger}{dt} = \frac{1}{\hbar} [\hat{H}, \hat{b}_{-k}^\dagger] = \Omega \hat{b}_{-k}^\dagger - iC \left( \hat{a}_k - \hat{a}_{-k}^\dagger \right), \quad (\text{D.7d})$$

Equation (D.5) is rewritten as

$$\begin{aligned} & \left[ -\omega_k \hat{a}_k - iC \left( \hat{b}_k + \hat{b}_{-k}^\dagger \right) \right] W_k + \left[ \omega_k \hat{a}_{-k}^\dagger - iC \left( \hat{b}_k + \hat{b}_{-k}^\dagger \right) \right] Y_k \\ & + \left[ -\Omega \hat{b}_k + iC \left( \hat{a}_k - \hat{a}_{-k}^\dagger \right) \right] X_k + \left[ \Omega \hat{b}_{-k}^\dagger - iC \left( \hat{a}_k - \hat{a}_{-k}^\dagger \right) \right] Z_k. \end{aligned} \quad (\text{D.8})$$

Because the operators are linearly independent, from Eqs. (D.6) and (D.8) one has

$$M \begin{pmatrix} W_k \\ X_k \\ Y_k \\ Z_k \end{pmatrix} \equiv \begin{pmatrix} \Omega(k) - \omega_k & iC & 0 & -iC \\ -iC & \Omega(k) - \Omega & -iC & 0 \\ 0 & -iC & \Omega(k) + \omega_k & iC \\ -iC & 0 & -iC & \Omega(k) + \Omega \end{pmatrix} \begin{pmatrix} W_k \\ X_k \\ Y_k \\ Z_k \end{pmatrix} = 0. \quad (\text{D.9})$$

The conditions that the coefficients  $W_k$ ,  $Y_k$ ,  $X_k$ , and  $Z_k$  are not zero leads to

$$\left\{ \Omega^2(k) - \omega_k^2 \right\} \left\{ \Omega^2(k) - \Omega^2 \right\} = 4C^2 \Omega \omega_k \quad (\text{D.10})$$

as an eigenvalue equation. By setting  $\hbar\Omega(k) = E(k)$  and  $\hbar\Omega = E_m$ , Eq. (D.10) can be rewritten as

$$\left\{ E^2(k) - (\hbar\omega_k)^2 \right\} \left\{ E^2(k) - E_m^2 \right\} = 4\hbar\omega_k C^2 E_m. \quad (\text{D.11})$$

In order to determine the four coefficients,  $X_k$ ,  $Y_k$ , and  $Z_k$  are expressed in terms of  $W_k$  from Eq. (D.9):

$$Y_k = -\frac{E(k) - \hbar\omega_k}{E(k) + \hbar\omega_k} W_k, \quad (\text{D.12a})$$

$$X_k = -\frac{\{E(k) + E_m\} \{E(k) - \hbar\omega_k\}}{2i\hbar C E_m} W_k, \quad (\text{D.12b})$$

$$Z_k = -\frac{\{E(k) - E_m\} \{E(k) - \hbar\omega_k\}}{2i\hbar C E_m} W_k. \quad (\text{D.12c})$$

The boson commutation relation  $[\hat{\xi}_k, \hat{\xi}_k^\dagger] = 1$  gives the following constraint:

$$|W_k|^2 + |X_k|^2 - |Y_k|^2 - |Z_k|^2 = 1. \quad (\text{D.13})$$

Using Eqs. (D.11) – (D.13), the relation

$$\begin{aligned} W_k &= \frac{E(k) + \hbar\omega_k}{2\sqrt{E(k)\hbar\omega_k}} \sqrt{\frac{E^2(k) - E_m^2}{2E^2(k) - E_m^2 - (\hbar\omega_k)^2}} \\ &= \frac{\Omega(k) + \omega_k}{2\sqrt{\Omega(k)\omega_k}} \sqrt{\frac{\Omega^2(k) - \Omega^2}{2\Omega(k)^2 - \Omega^2 - \omega_k^2}} \end{aligned} \quad (\text{D.14})$$

is obtained. Corresponding to two eigenvalues  $E^{(\pm)}(k)$  of Eq. (D.11), the exciton-polariton operator and expansion coefficients are classified by a superscript ( $\pm$ ), such as  $\hat{\xi}_k^{(\pm)}$ ,  $\hat{\xi}_k^{(\pm)\dagger}$ , and  $W_k^{(\pm)}$ . Then, Eq. (D.2) is rewritten as follows:

$$\begin{pmatrix} \hat{\xi}_k^{(+)} \\ \hat{\xi}_k^{(-)} \\ \hat{\xi}_k^{(+)\dagger} \\ \hat{\xi}_{-k}^{(-)\dagger} \end{pmatrix} \equiv \begin{pmatrix} W_k^{(+)} & X_k^{(+)} & Y_k^{(+)} & Z_k^{(+)} \\ W_k^{(-)} & X_k^{(-)} & Y_k^{(-)} & Z_k^{(-)} \\ Y_k^{(+)*} & Z_k^{(+)*} & W_k^{(+)*} & X_k^{(+)*} \\ Y_k^{(-)*} & Z_k^{(-)*} & W_k^{(-)*} & X_k^{(-)*} \end{pmatrix} \begin{pmatrix} \hat{a}_k \\ \hat{b}_k \\ \hat{a}_{-k}^\dagger \\ \hat{b}_{-k}^\dagger \end{pmatrix}, \quad (\text{D.15})$$

which can be inversely transformed to

$$\begin{pmatrix} \hat{a}_k \\ \hat{b}_k \\ \hat{a}_{-k}^\dagger \\ \hat{b}_{-k}^\dagger \end{pmatrix} \equiv \begin{pmatrix} W_k^{(+)*} & W_k^{(-)*} & -Y_k^{(+)} & -Y_k^{(-)} \\ X_k^{(+)*} & X_k^{(-)*} & -Z_k^{(+)} & -Z_k^{(-)} \\ -Y_k^{(+)*} & -Y_k^{(-)*} & W_k^{(+)} & W_k^{(-)} \\ -Z_k^{(+)*} & Z_k^{(-)*} & X_k^{(+)} & X_k^{(-)} \end{pmatrix} \begin{pmatrix} \hat{\xi}_k^{(+)} \\ \hat{\xi}_k^{(-)} \\ \hat{\xi}_{-k}^{(+)\dagger} \\ \hat{\xi}_{-k}^{(-)\dagger} \end{pmatrix}. \quad (\text{D.16})$$

Since the coefficients with the superscripts (+) and (–) appear equivalently in the matrix on the right-hand side of Eq. (D.16), the superscripts ( $\pm$ ) can be omitted, giving

$$\begin{cases} \hat{a}_k = W_k^* \hat{\xi}_k - Y_k \hat{\xi}_{-k}^\dagger \\ \hat{a}_{-k}^\dagger = -Y_k^* \hat{\xi}_k + W_k \hat{\xi}_{-k}^\dagger, \end{cases} \quad (\text{D.17})$$

which are substituted for the photon operators in  $\hat{\mathbf{D}}^\perp(\mathbf{r})$  in Eq. (2.6), and Eq.(2.6) is then inserted into Eq. (2.25). Using Eqs. (D.12a), (D.12b), (D.12c) and (D.14), one finally obtains

$$K_\alpha(\mathbf{k}) = \sum_{\lambda=1}^2 (\mathbf{p}_\alpha \cdot \mathbf{e}_{k\lambda}(\mathbf{k})) f(k) e^{i\mathbf{k} \cdot \mathbf{r}_\alpha} \quad (\text{D.18})$$

with

$$f(k) = \frac{ck}{\sqrt{\Omega(k)}} \sqrt{\frac{\Omega^2(k) - \Omega^2}{2\Omega^2(k) - \Omega^2 - (ck)^2}}, \quad (\text{D.19})$$

which is Eq. (2.28) to be derived.



# Appendix E

## Derivation of the Equations for Size-Dependent Resonance

The right-hand side of Eq. (2.78a) is written as

$$V_{eff}(r) = -\frac{P_s P_p}{3(2\pi)\epsilon_0} W_+ \sum_{\alpha=s}^p \frac{\exp(-r/a'_\alpha)}{a_\alpha^2 r}, \tag{E.1}$$

and the first row of Eq. (2.79) as

$$I(R_{sp}) = \left| \int \nabla_{r_p} P(r_p) d^3 r_p \right|^2, \tag{E.2a}$$

$$P(r_p) = \int V_{eff}(|r_p - r_s|) d^3 r_s, \tag{E.2b}$$

where  $r_{sp}$  on the left-hand side of the first row of Eq. (2.79) is denoted by  $R_{sp}$  on the left-hand side of Eq. (E.2a) to avoid confusion in the notations in the following description.

First, in order to perform the integration of Eq. (E.2b), the  $z_s$ -Cartesian axis is fixed along the line connecting an arbitrary position on the sphere p and the center of the sphere s. Then, Eqs. (E.1) and (E.2b) give

$$\begin{aligned} P(r_p) &= \int \sum_{\alpha=s}^p \frac{\exp(-|r_p - r_s|/a'_\alpha)}{a_\alpha^2 |r_p - r_s|} d^3 r_s \tag{E.3} \\ &= \sum_{\alpha=s}^p \int_0^{a_s} dr_s \int_0^\pi d\theta_s \int_0^{2\pi} d\phi_s \\ &\quad \times \frac{\exp\left(-\sqrt{|\mathbf{R}_s - \mathbf{r}_p|^2 + r_s^2 - 2|\mathbf{R}_s - \mathbf{r}_p|r_s \cos \theta_s}/a'_\alpha}\right)}{a_\alpha^2 \sqrt{|\mathbf{R}_s - \mathbf{r}_p|^2 + r_s^2 - 2|\mathbf{R}_s - \mathbf{r}_p|r_s \cos \theta_s}} r_s^2 \sin \theta_s \end{aligned}$$

$$\begin{aligned}
&= 2\pi \sum_{\alpha=s}^p \int_0^{a_s} dr_s \int_0^\pi d\theta_s \\
&\quad \times \frac{\exp\left(-\sqrt{|\mathbf{R}_s - \mathbf{r}_p|^2 + r_s^2 - 2|\mathbf{R}_s - \mathbf{r}_p| r_s \cos \theta_s / a'_\alpha}\right)}{a'^2_\alpha \sqrt{|\mathbf{R}_s - \mathbf{r}_p|^2 + r_s^2 - 2|\mathbf{R}_s - \mathbf{r}_p| r_s \cos \theta_s}} r_s^2 \sin \theta_s.
\end{aligned}$$

By defining

$$s_s \equiv \sqrt{|\mathbf{R}_s - \mathbf{r}_p|^2 + r_s^2 - 2|\mathbf{R}_s - \mathbf{r}_p| r_s \cos \theta_s}, \quad (\text{E.4})$$

and using

$$\frac{ds_s}{d\theta_s} = |\mathbf{R}_s - \mathbf{r}_p| \frac{r_s}{s_s} \sin \theta_s, \quad (\text{E.5})$$

Equation (E.3) is rewritten as

$$\begin{aligned}
P(r_p) &= \frac{2\pi}{a'^2_\alpha |\mathbf{R}_s - \mathbf{r}_p|} \sum_{\alpha=s}^p \int_0^{a_s} dr_s r_s \int_{|\mathbf{R}_s - \mathbf{r}_p| - r_s}^{|\mathbf{R}_s - \mathbf{r}_p| + r_s} ds_s e^{-s_s / a'_\alpha} \\
&= \frac{2\pi}{a'^2_\alpha |\mathbf{R}_s - \mathbf{r}_p|} \sum_{\alpha=s}^p \int_0^{a_s} dr_s r_s a'_\alpha \\
&\quad \times \left\{ -\exp\left(-\frac{|\mathbf{R}_s - \mathbf{r}_p| + r_s}{a'_\alpha}\right) + \exp\left(-\frac{|\mathbf{R}_s - \mathbf{r}_p| - r_s}{a'_\alpha}\right) \right\} \\
&= \frac{2\pi}{a'^2_\alpha |\mathbf{R}_s - \mathbf{r}_p|} \sum_{\alpha=s}^p a'_\alpha \exp\left(-\frac{|\mathbf{R}_s - \mathbf{r}_p|}{a'_\alpha}\right) \\
&\quad \times \int_0^{a_s} dr_s r_s \left\{ \exp\left(\frac{r_s}{a'_\alpha}\right) - \exp\left(-\frac{r_s}{a'_\alpha}\right) \right\} \\
&= \frac{4\pi}{a'^2_\alpha |\mathbf{R}_s - \mathbf{r}_p|} \sum_{\alpha=s}^p a_i^3 \exp\left(-\frac{|\mathbf{R}_s - \mathbf{r}_p|}{a'_\alpha}\right) \\
&\quad \times \left\{ \frac{a_s}{a'_\alpha} \cosh\left(\frac{a_s}{a'_\alpha}\right) - \sinh\left(\frac{a_s}{a'_\alpha}\right) \right\}.
\end{aligned} \quad (\text{E.6})$$

Next, in order to perform the integration of Eq. (E.2a) with respect to  $\mathbf{r}_p$ , the volume integral is converted to a surface integral, which is expressed as

$$\begin{aligned}
\int \nabla_{r_p} P(r_p) d^3 r_p &= \int_{S_p} P(r_p) \mathbf{n}_p dS_p \\
&= 4\pi \sum_{\alpha=s}^p a_i^3 \left\{ \frac{a_s}{a'_\alpha} \cosh\left(\frac{a_s}{a'_\alpha}\right) - \sinh\left(\frac{a_s}{a'_\alpha}\right) \right\} \int_{S_p} \frac{\exp\left(-\frac{|\mathbf{R}_s - \mathbf{r}_p|}{a'_\alpha}\right)}{a'^2_\alpha |\mathbf{R}_s - \mathbf{r}_p|} \mathbf{n}_p dS_p.
\end{aligned} \quad (\text{E.7})$$

Here,  $\mathbf{n}_p$  is the unit vector normal to the surface  $S_p$ . By fixing the  $z_p$ -Cartesian axis along the line connecting the center  $\mathbf{R}_s$  of the sphere  $s$  and the center  $\mathbf{R}_p$  of the sphere  $p$ , the surface integral in Eq. (E.7) is taken, and the result is given by

$$\begin{aligned}
 & \int_{S_p} \frac{\exp\left(-\frac{|\mathbf{R}_s - \mathbf{r}_p|}{a'_\alpha}\right)}{|\mathbf{R}_s - \mathbf{r}_p|} \mathbf{n}_p dS_p \quad (\text{E.8}) \\
 &= \int_0^\pi d\theta_p \int_0^{2\pi} d\phi_p \frac{\exp\left(-\frac{\sqrt{R_{sp}^2 + a_p^2 - 2a_p R_{sp} \cos \theta_p}}{a'_\alpha}\right)}{\sqrt{R_{sp}^2 + a_p^2 - 2a_p R_{sp} \cos \theta_p}} a_p^2 \sin \theta_p \mathbf{n}_p \\
 &= \frac{a_p}{R_{sp}} \int_{R_{sp}-a_p}^{R_{sp}+a_p} ds_p e^{-\frac{s_p}{a'_\alpha}} \int_0^{2\pi} d\phi_p (\sin \theta_p \cos \phi_p, \sin \theta_p \sin \phi_p, \cos \theta_p) \\
 &= \frac{a_p}{R_{sp}} \int_{R_{sp}-a_p}^{R_{sp}+a_p} ds_p e^{-\frac{s_p}{a'_\alpha}} \frac{R_{sp}^2 + a_p^2 - s_p^2}{2a_p R_{sp}} \hat{\mathbf{R}}_{sp} \\
 &= \frac{1}{2R_{sp}^2} \int_{R_{sp}-a_p}^{R_{sp}+a_p} ds_p e^{-\frac{s_p}{a'_\alpha}} [R_{sp}^2 + a_p^2 - s_p^2] \hat{\mathbf{R}}_{sp}.
 \end{aligned}$$

where  $\hat{\mathbf{R}}_{sp}$  is a unit vector along  $\mathbf{R}_s - \mathbf{R}_p$ , and the following transformation was used:

$$s_p = \sqrt{R_{sp}^2 + a_p^2 - 2a_p R_{sp} \cos \theta_p} \quad (\text{E.9a})$$

with

$$\frac{ds_p}{d\theta_p} = \frac{a_p R_{sp}}{s_p} \sin \theta_p, \quad (\text{E.9b})$$

$$\cos \theta_p = \frac{R_{sp}^2 + a_p^2 - s_p^2}{2a_p R_{sp}}, \quad (\text{E.9c})$$

Inserting

$$\begin{aligned}
 \int_{R_{sp}-a_p}^{R_{sp}+a_p} ds_p e^{-\frac{s_p}{a'_\alpha}} &= -a'_\alpha \left\{ \exp\left(-\frac{R_{sp}+a_p}{a'_\alpha}\right) - \exp\left(-\frac{R_{sp}-a_p}{a'_\alpha}\right) \right\} \\
 &= 2a'_\alpha e^{-\frac{R_{sp}}{a'_\alpha}} \sinh\left(\frac{a_p}{a'_\alpha}\right)
 \end{aligned} \quad (\text{E.10a})$$

and

$$\begin{aligned}
 & \int_{R_{sp}-a_p}^{R_{sp}+a_p} ds_p e^{-\frac{s_p}{a'_\alpha}} s_p^2 \quad (\text{E.10b}) \\
 &= \left[ -a'_\alpha s_p^2 \exp(-s_p/a'_\alpha) \right]_{R_{sp}-a_p}^{R_{sp}+a_p} + 2a'_\alpha \int_{R_{sp}-a_p}^{R_{sp}+a_p} ds_p e^{-\frac{s_p}{a'_\alpha}} s_p
 \end{aligned}$$

$$\begin{aligned}
&= \left[ -a'_\alpha s_p^2 \exp(-s_p/a'_\alpha) - 2a'_\alpha{}^2 s_p \exp(-s_p/a'_\alpha) \right]_{R_{sp}-a_p}^{R_{sp}+a_p} \\
&\quad + 2a_i'^2 \int_{R_{sp}-a_p}^{R_{sp}+a_p} ds_p e^{-\frac{sp}{a'_\alpha}} \\
&= \left[ -a'_\alpha s_p^2 \exp(-s_p/a'_\alpha) - 2a'_\alpha{}^2 s_p \exp(-s_p/a'_\alpha) - 2a'_\alpha{}^3 \exp(-s_p/a'_\alpha) \right]_{R_{sp}-a_p}^{R_{sp}+a_p} \\
&= 2a'_\alpha e^{-\frac{R_{sp}}{a'_\alpha}} \left\{ \left( R_{sp}^2 + 2R_{sp}a'_\alpha + a_p^2 + 2a'_\alpha{}^2 \right) \sinh\left(\frac{a_p}{a'_\alpha}\right) \right. \\
&\quad \left. - 2a_p (R_{sp} + a'_\alpha) \cosh\left(\frac{a_p}{a'_\alpha}\right) \right\}
\end{aligned}$$

into Eq. (E.8) yields

$$\begin{aligned}
\int_{S_p} \frac{\exp\left(-\frac{|\mathbf{R}_s - \mathbf{r}_p|}{a_i}\right)}{|\mathbf{R}_s - \mathbf{r}_p|} \mathbf{n}_p dS_p &= 2a_i'^2 \left\{ \frac{a_p}{a_i'} \cosh\left(\frac{a_p}{a_i'}\right) - \sinh\left(\frac{a_p}{a_i'}\right) \right\} \\
&\times \left( \frac{1}{R_{sp}} + \frac{a_i'}{R_{sp}^2} \right) e^{-\frac{R_{sp}}{a_i'}} \hat{\mathbf{R}}_{sp}.
\end{aligned} \tag{E.11}$$

Furthermore, inserting this equation into Eq. (E.7) leads to

$$\begin{aligned}
\int \nabla_{r_p} P(r_p) d^3 r_p &= 8\pi \sum_{\alpha=s}^p \frac{1}{a_\alpha'^2} a_\alpha'^6 \left\{ \frac{a_s}{a'_\alpha} \cosh\left(\frac{a_s}{a'_\alpha}\right) - \sinh\left(\frac{a_s}{a'_\alpha}\right) \right\} \\
&\times \left\{ \frac{a_p}{a'_\alpha} \cosh\left(\frac{a_p}{a'_\alpha}\right) - \sinh\left(\frac{a_p}{a'_\alpha}\right) \right\} \left( \frac{1}{a'_\alpha R_{sp}} + \frac{1}{R_{sp}^2} \right) e^{-\frac{R_{sp}}{a'_\alpha}} \hat{\mathbf{R}}_{sp}.
\end{aligned} \tag{E.12}$$

Then, Eq. (E.2a) can be rewritten as

$$\begin{aligned}
I(R_{sp}) &= \left( \frac{P_s P_p}{3(2\pi)\varepsilon_0} W_+ \right)^2 \\
&\times \left[ 8\pi \sum_{\alpha=s}^p a_\alpha'^4 \left\{ \frac{a_s}{a'_\alpha} \cosh\left(\frac{a_s}{a'_\alpha}\right) - \sinh\left(\frac{a_s}{a'_\alpha}\right) \right\} \right] \\
&\times \left\{ \frac{a_p}{a'_\alpha} \cosh\left(\frac{a_p}{a'_\alpha}\right) - \sinh\left(\frac{a_p}{a'_\alpha}\right) \right\} \left( \frac{1}{a'_\alpha R_{sp}} + \frac{1}{R_{sp}^2} \right) e^{-\frac{R_{sp}}{a'_\alpha}} \right]^2.
\end{aligned} \tag{E.13}$$

Finally, by replacing  $R_{sp}$  with  $r_{sp}$ , Eq. (E.13) is reduced to Eq. (2.79).

## Appendix F

# Energy States of a Semiconductor Quantum Dot

Because electrons, holes, and electron–hole pairs in a QD are confined in a three-dimensional nanometric space, they have unique properties that cannot be achieved in bulk semiconductor materials. For example, they have discrete energy eigenvalues originating from the fact that their wave-functions are confined in the material. This is called the quantum confinement effect. The following sections review fundamental aspects of this effect for spherical or cubic QDs [1, 2].

### F.1 One-Particle States

Because a QD contains many electrons, holes, and, electron–hole pairs even though its dimensions are of nanometer order, a many-particle problem has to be solved in order to study the quantum confinement effect. For this purpose, it is useful to employ the envelope function and effective mass approximation on the assumption that the energy eigenvalues of electrons in the periodic lattice, that is, the energy bands, are not appreciably modified through the quantum confinement. This approximation allows us to determine a ground state and excited states of a one-particle (electron or hole) system, and also a ground state of a many-particle problem by successively putting particles into the lowest unoccupied energy levels.

The one-particle wave-function in a QD is given by the product of the one-particle wave-function in a bulk material and the envelope function that satisfies the boundary conditions of the QD. Thus, the eigenstate  $|\psi_e\rangle$  of an electron in the QD is expressed as

$$|\psi_e\rangle = \int d^3r \xi_e(\mathbf{r}) \hat{\psi}_e^\dagger(\mathbf{r}) |\Phi_g\rangle \quad (\text{F.1})$$

where  $\xi_e(\mathbf{r})$  is the envelope function of the single electron state,  $\hat{\psi}_e^\dagger(\mathbf{r})$  is the field operator for electron creation, and  $|\Phi_g\rangle$  is the crystal ground state (also called the electronic vacuum state  $|0\rangle$ ). Since no electrons in the conduction band exist in the

crystal ground state, applying the field operator  $\hat{\psi}_e(\mathbf{r})$  for electron annihilation to  $|\Phi_g\rangle$  gives the following relation:

$$\hat{\psi}_e(\mathbf{r})|\Phi_g\rangle = 0. \quad (\text{F.2})$$

The field operators for electron creation and annihilation satisfy the anti-commutation relation for a fermion, namely

$$\{\hat{\psi}_e(\mathbf{r}'), \hat{\psi}_e^\dagger(\mathbf{r})\} \equiv \hat{\psi}_e(\mathbf{r}')\hat{\psi}_e^\dagger(\mathbf{r}) + \hat{\psi}_e^\dagger(\mathbf{r})\hat{\psi}_e(\mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}'), \quad (\text{F.3})$$

where  $\delta(\mathbf{r} - \mathbf{r}')$  is the Dirac delta function. The equation for the envelope function  $\xi_e(\mathbf{r})$  can be obtained from the Schrödinger equation

$$\hat{H}_e|\psi_e\rangle = E_e|\psi_e\rangle \quad (\text{F.4})$$

with the Hamiltonian  $\hat{H}_e$  for a single electron in the QD

$$\hat{H}_e = \int d^3r \hat{\psi}_e^\dagger(\mathbf{r}) \left[ -\frac{\hbar^2}{2m_e} \nabla^2 \right] \hat{\psi}_e(\mathbf{r}) + E_g \int d^3r \hat{\psi}_e^\dagger(\mathbf{r}) \hat{\psi}_e(\mathbf{r}), \quad (\text{F.5})$$

and the energy eigenvalue  $E_g$ . Here,  $m_e$  and  $E_g$  denote the effective mass of an electron and the bandgap energy of the bulk semiconductor, respectively. Substituting this equation into Eq. (F.4), the left-hand side is given by

$$\begin{aligned} \hat{H}_e|\psi_e\rangle &= \int d^3r' \hat{\psi}_e^\dagger(\mathbf{r}') \left[ -\frac{\hbar^2}{2m_e} \nabla^2 \right] \hat{\psi}_e(\mathbf{r}') \int d^3r \xi_e(\mathbf{r}) \hat{\psi}_e^\dagger(\mathbf{r}) |\Phi_g\rangle \\ &\quad + E_g \int d^3r' \hat{\psi}_e^\dagger(\mathbf{r}') \hat{\psi}_e(\mathbf{r}') \int d^3r \xi_e(\mathbf{r}) \hat{\psi}_e^\dagger(\mathbf{r}) |\Phi_g\rangle \\ &= -\frac{\hbar^2}{2m_e} \int d^3r' \int d^3r \delta(\mathbf{r} - \mathbf{r}') \nabla^2 \xi_e(\mathbf{r}) \hat{\psi}_e^\dagger(\mathbf{r}') |\Phi_g\rangle \\ &\quad + E_g \int d^3r' \int d^3r \delta(\mathbf{r} - \mathbf{r}') \xi_e(\mathbf{r}) \hat{\psi}_e^\dagger(\mathbf{r}') |\Phi_g\rangle \\ &= \int d^3r \left[ -\frac{\hbar^2}{2m_e} \nabla^2 \xi_e(\mathbf{r}) \right] \hat{\psi}_e^\dagger(\mathbf{r}) |\Phi_g\rangle + E_g \int d^3r \xi_e(\mathbf{r}) \hat{\psi}_e^\dagger(\mathbf{r}) |\Phi_g\rangle \end{aligned} \quad (\text{F.6})$$

while the right-hand side is given by

$$E_e|\psi_e\rangle = E_e \int d^3r \xi_e(\mathbf{r}) \hat{\psi}_e^\dagger(\mathbf{r}) |\Phi_g\rangle, \quad (\text{F.7})$$

where Eqs. (F.2) and (F.3) were used. By equating Eqs. (F.5) and (F.6), the eigenvalue equation for the envelope function of the one-electron system is derived:

$$-\frac{\hbar^2}{2m_e} \nabla^2 \xi_e(\mathbf{r}) = (E_e - E_g) \xi_e(\mathbf{r}). \quad (\text{F.8})$$

By replacing the subscript of the envelope function  $e$  with  $h$ , the eigenvalue equation for the one-hole system is derived:

$$-\frac{\hbar^2}{2m_h} \nabla^2 \xi_h(\mathbf{r}) = E_h \xi_h(\mathbf{r}), \quad (\text{F.9})$$

where  $E_g = 0$  was used.

In order to solve these eigenvalue equations, spherical and cubic QDs are considered in the following discussions.

### (1) Spherical quantum dot

Assuming that an electron or a hole is confined in a spherical QD with radius  $R$ , its boundary condition is expressed as  $\xi_e(\mathbf{r}) = \xi_h(\mathbf{r}) = 0$  for  $|\mathbf{r}| > R$ . Noting that the Laplace operator is written in spherical coordinates as

$$\nabla^2 = \frac{1}{r} \frac{\partial^2}{\partial r^2} r - \frac{\mathbf{L}^2}{r^2}, \quad (\text{F.10a})$$

$$\mathbf{L}^2 = - \left( \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \sin \theta \frac{\partial}{\partial \theta} + \frac{1}{\sin^2 \theta} \frac{\partial^2}{\partial \phi^2} \right), \quad (\text{F.10b})$$

the envelope function  $\xi(\mathbf{r})$  can be divided into radial and angular parts as  $\xi(\mathbf{r}) = f_l(r) Y_{lm}(\theta, \phi)$ . The subscripts  $e$  and  $h$  have been removed because the envelope function  $\xi(\mathbf{r})$  depends only on  $R$  and not on specific parameters for the electron and hole. Here,  $\mathbf{L}$  denotes the operator of the orbital angular momentum and obeys the following eigenvalue equation

$$\mathbf{L}^2 Y_{lm}(\theta, \phi) = l(l+1) Y_{lm}(\theta, \phi) \quad (\text{F.11})$$

with  $|m| \leq l$ , where the functions  $Y_{lm}(\theta, \phi)$  are the spherical harmonics with  $l = 0, 1, 2, \dots$  and  $m = 0, \pm 1, \pm 2, \dots$ . The radial part  $f_l(r)$  should obey

$$\frac{d^2 f_l}{dr^2} + \frac{2}{r} \frac{df_l}{dr} + [\alpha^2 - l(l+1)] f_l = 0 \quad (\text{F.12})$$

with

$$\alpha^2 \equiv \frac{2m_e}{\hbar^2} (E_e - E_g) \text{ or } \frac{2m_e}{\hbar^2} E_h, \quad (\text{F.13})$$

and the solution has the form

$$f_{nl}(r) = \sqrt{\frac{2}{R^3}} \frac{j_l\left(\frac{\alpha_{nl} r}{R}\right)}{j_{l+1}(\alpha_{nl})}, \quad (\text{F.14a})$$

where  $j_l$  is a spherical Bessel function of  $l$ -th order, and  $\alpha_{nl}$  is determined from the boundary conditions as

$$j_l(\alpha_{nl}) = 0 \quad (n = 1, 2, 3, \dots), \text{ and } \alpha_{n0} = n\pi, \alpha_{11} = 4.4934, \dots \quad (\text{F.14b})$$

The energy eigenvalues of the electron are discrete and given by

$$E_{e,nlm} = E_g + \frac{\hbar^2}{2m_e} \left( \frac{\alpha_{nl}}{R} \right)^2. \quad (\text{F.15})$$

Those of the hole are

$$E_{h,nlm} = \frac{\hbar^2}{2m_h} \left( \frac{\alpha_{nl}}{R} \right)^2. \quad (\text{F.16})$$

## (2) Cubic quantum dot

In the case where an electron or a hole is confined in a cubic QD with side-length  $L$ , as the first step, a one-dimensional case is analyzed, where the following one-dimensional well potential

$$V(x) = 0 \quad \text{for } |x| \leq \frac{L}{2} \quad (\text{F.17a})$$

$$V(x) = \infty \quad \text{for } |x| > \frac{L}{2} \quad (\text{F.17b})$$

is assumed. The envelope function  $\xi(x)$  obeys the Schrödinger equation

$$\left[ -\frac{\hbar^2}{2m} \frac{d^2}{dx^2} + V(x) \right] \xi(x) = E_x \xi(x), \quad (\text{F.18})$$

and the boundary conditions

$$\xi\left(\frac{L}{2}\right) = \xi\left(-\frac{L}{2}\right) = 0 \quad (\text{F.19})$$

are satisfied. The solutions of the equation are given by

$$\begin{cases} \xi_{\text{even}}(x) = \sqrt{\frac{2}{L}} \cos(k_x x), \\ \xi_{\text{odd}}(x) = \sqrt{\frac{2}{L}} \sin(k_x x), \end{cases} \quad (\text{F.20})$$

and it follows from the boundary conditions that  $k_x$  has the following discrete values:

$$\begin{cases} k_x^{\text{even}} = \frac{\pi}{L} (2n - 1), \\ k_x^{\text{odd}} = \frac{\pi}{L} (2n). \end{cases} \quad (n = 1, 2, 3, \dots) \quad (\text{F.21})$$



Therefore, the energy eigenvalues are also discrete:

$$E_x = \frac{\hbar^2 k_x^2}{2m} = \frac{\hbar^2}{2m} \left( \frac{\pi}{L} n_x \right)^2, \quad (\text{F.22})$$

with  $n_x = 1, 2, 3, \dots$ , where one sets  $n_x = 2n - 1$  for  $k_x^{\text{even}}$ , and  $n_x = 2n$  for  $k_x^{\text{odd}}$ . As the second step, the envelope functions  $\xi(y)$  and  $\xi(z)$  can be similarly obtained by replacing  $x$  in Eq. (F.20) with its corresponding  $y$  and  $z$ , respectively. Finally, the envelope functions for an electron and a hole confined in a three-dimensional potential well have the form  $\xi(x)\xi(y)\xi(z)$ . The energy eigenvalues specified by a set of quantum numbers  $(n_x, n_y, n_z)$  are given by

$$E_{n_x, n_y, n_z} = \frac{\hbar^2}{2m} \left( \frac{\pi}{L} \right)^2 \left( n_x^2 + n_y^2 + n_z^2 \right) \text{ for } n_x, n_y, n_z = 1, 2, 3, \dots \quad (\text{F.23})$$

By assuming that the energy eigenvalues of an electron in a periodic potential are not modified drastically, the energies of an electron in the conduction band or valence band are given by

$$E_c = E_g + \frac{\hbar^2 k^2}{2m_c} = E_g + \frac{\hbar^2}{2m_c} \left\{ \left( \frac{\pi}{L_x} n_x \right)^2 + \left( \frac{\pi}{L_y} n_y \right)^2 + \left( \frac{\pi}{L_z} n_z \right)^2 \right\} \quad (\text{F.24a})$$

and

$$E_v = \frac{\hbar^2 k^2}{2m_v} = \frac{\hbar^2}{2m_v} \left\{ \left( \frac{\pi}{L_x} n_x \right)^2 + \left( \frac{\pi}{L_y} n_y \right)^2 + \left( \frac{\pi}{L_z} n_z \right)^2 \right\}, \quad (\text{F.24b})$$

respectively.

## F.2 Electron–Hole Pair States in a Quantum Dot

In order to analyze the electron–hole pair states in a QD, the eigenstate for an electron–hole pair is expressed as

$$|\psi_{eh}\rangle = \int \int d^3 r_e d^3 r_h \zeta_{eh}(\mathbf{r}_e, \mathbf{r}_h) \hat{\psi}_e^\dagger(\mathbf{r}_e) \hat{\psi}_h^\dagger(\mathbf{r}_h) |\Phi_g\rangle, \quad (\text{F.25})$$

where  $\zeta_{eh}(\mathbf{r}_e, \mathbf{r}_h)$  is the envelope function of the electron–hole pair,  $\hat{\psi}_e^\dagger(\mathbf{r}_e)$  and  $\hat{\psi}_h^\dagger(\mathbf{r}_h)$  are the field operators of electron creation in the conduction band and hole creation in the valence band, respectively, and  $|\Phi_g\rangle$  is the above-mentioned crystal ground state. The envelope function  $\zeta_{eh}(\mathbf{r}_e, \mathbf{r}_h)$  obeys the following equation:

$$\left[ -\frac{\hbar^2}{2m_e} \nabla_e^2 - \frac{\hbar^2}{2m_h} \nabla_h^2 + V_c + V_{conf} \right] \zeta_{eh}(\mathbf{r}_e, \mathbf{r}_h) = (E - E_g) \zeta_{eh}(\mathbf{r}_e, \mathbf{r}_h) \quad (\text{F.26})$$

with the Coulomb interaction potential  $V_c$  and the confinement potential  $V_{conf}$ . When the confinement region is a sphere with radius  $R$ ,  $V_{conf}(\mathbf{r}) = 0$  for  $|\mathbf{r}| = r \leq R$ , whereas when it is a cube with side length  $L$ ,  $V_{conf}(x, y, z) = 0$  for  $-L/2 \leq x, y, z \leq L/2$ . Here, it might be useful to examine the electron–hole pair states by comparing the confinement size ( $R$  or  $L$ ) with the Bohr radius  $a_0$ , which represents the average distance between the electron and hole in the pair. Noting that the confinement potential is proportional to  $1/R^2$  (or  $1/L^2$ ) whereas the Coulomb interaction potential is proportional to  $1/R$  (or  $1/L$ ), the following three cases are considered.

(1)  $R \ll a_0$

In this case, the Coulomb interaction between an electron and a hole is weak, and each electron (hole) in a pair independently moves in the corresponding electron (hole) confinement potential. In particular, when both the Coulomb and confinement potentials are zero in a perfectly confined area, the lowest energy of an electron–hole pair is given, in terms of energy eigenvalues in the one-particle system already described, by

$$E = E_g + \frac{\pi^2 \hbar^2}{2m_e R^2} + \frac{\pi^2 \hbar^2}{2m_h R^2} = E_g + \frac{\pi^2 \hbar^2}{2m_r R^2}, \quad (\text{F.27})$$

where  $m_r$  is the reduced mass for an electron–hole pair defined by

$$\frac{1}{m_r} = \frac{1}{m_e} + \frac{1}{m_h}. \quad (\text{F.28})$$

(2)  $R \gg a_0$

Because the Coulomb interaction between an electron and a hole becomes strong in this case, an electron–hole pair can be approximated as a single particle, which is called an exciton. Then, the motion of the center of mass of the exciton is confined within the area  $R$  (or  $L$ ). Defining the mass of the exciton, the center of mass coordinates, and the relative coordinates between the electron and hole as

$$M = m_e + m_h \quad (\text{F.29a})$$

$$\mathbf{r}_{CM} = (m_e \mathbf{r}_e + m_h \mathbf{r}_h) / M \quad (\text{F.29b})$$

$$\boldsymbol{\beta} = \mathbf{r}_e - \mathbf{r}_h \quad (\text{F.29c})$$

respectively, the envelope function of the exciton is written as

$$\psi(\mathbf{r}_e, \mathbf{r}_h) = \phi_\mu(\boldsymbol{\beta}) F_\nu(\mathbf{r}_{CM}), \quad (\text{F.30})$$

where, in particular,

$$F_V(\mathbf{r}_{CM}) = \sqrt{\frac{2}{R^3}} \frac{j_l\left(\frac{\alpha_{nl} r_{CM}}{R}\right)}{j_{l+1}(\alpha_{nl})} Y_{lm}(\Omega_{CM}) \quad (\text{F.31})$$

and

$$\phi_{\mu=1s}(\beta) = \frac{1}{\sqrt{\pi a_0^3}} \exp\left(-\frac{\beta}{a_0}\right) \quad (\text{F.32})$$

for the spherical boundary conditions, similar to the one-particle system. Here, the solid angle  $\Omega_{CM}$  for  $\mathbf{r}_{CM}$  was used, and  $\phi_{\mu}(\beta)$  is assumed to be the function for the lowest energy state (the  $1s$  state in the case of an electron in a hydrogen atom). The energy eigenvalues of the states specified by the quantum numbers  $(n, l)$  are

$$E_{nl} = E_g + E_{ex} + \frac{\hbar^2 \alpha_{nl}^2}{2MR^2} \quad (n = 1, 2, 3, \dots), \quad (\text{F.33})$$

where  $E_{ex}$  is the exciton binding energy in the bulk system.

Similarly, for the cubic boundary conditions, the envelope function for the center of mass is expressed as

$$F_V(\mathbf{r}_{CM}) = \sqrt{\frac{8}{L^3}} \begin{cases} \cos\left(\frac{\pi}{L}(2n_x - 1)x_{CM}\right) \cos\left(\frac{\pi}{L}(2n_y - 1)y_{CM}\right) \\ \quad \times \cos\left(\frac{\pi}{L}(2n_z - 1)z_{CM}\right), \\ \sin\left(\frac{2\pi}{L}n_x x_{CM}\right) \sin\left(\frac{2\pi}{L}n_y y_{CM}\right) \sin\left(\frac{2\pi}{L}n_z z_{CM}\right). \end{cases} \quad (\text{F.34})$$

The function for the relative motion is the same as Eq. (F.32). The energy eigenvalues are expressed in a similar way as

$$E_{n_x, n_y, n_z} = E_g + E_{ex} + \frac{\pi^2 \hbar^2}{2ML^2} \left(n_x^2 + n_y^2 + n_z^2\right) \quad (\text{F.35}) \\ (n_x, n_y, n_z = 1, 2, 3, \dots).$$

Note that the motion of the center of mass is confined to a sphere of radius  $R - \eta a_0$  or a cube of side-length  $L - \eta a_0$ , where the factor  $\eta$  is of the order of unity and depends on the electron-hole mass ratio [3]. This is called dead-layer correction.

(3)  $R \simeq a_0$

The situation in this case is more complicated than those of (F.1) and (F.2). Let the Bohr radii of an electron and a hole be  $a_e$  and  $a_h$ , respectively, and suppose that the confinement size is larger than  $a_h$  and smaller than  $a_e$ . Then one may assume that a hole can move in an average potential created by a free electron confined within a QD, and approximate the envelope function of the exciton by the product of those of the electron  $\xi_{nlm}(\mathbf{r}_e)$  and hole  $\psi_h(\mathbf{r}_h)$  as

$$\psi(\mathbf{r}_e, \mathbf{r}_h) = \xi_{nlm}(\mathbf{r}_e) \psi_h(\mathbf{r}_h). \quad (\text{F.36})$$

Using the orthonormalization of  $\xi_{nlm}(\mathbf{r}_e)$ , the equation for the envelope function of the hole is written as

$$\begin{aligned} & \left[ -\frac{\hbar^2}{2m_h} \nabla_h^2 - \int d\mathbf{r}_e |\xi_{nlm}(\mathbf{r}_e)|^2 V_c \right] \psi_h(\mathbf{r}_h) \\ & = \left( E - E_g - \frac{\hbar^2}{2m_e} \frac{\alpha_{nl}^2}{R^2} \right) \psi_h(\mathbf{r}_h), \end{aligned} \quad (\text{F.37})$$

where the spherical confinement is assumed to be  $V_{conf} = 0$  within the confinement region. When the cubic boundary conditions are used, the envelope function  $\xi_{nlm}(\mathbf{r}_e)$  of an electron and the discrete energy  $(\hbar^2/2m_e) \alpha_{nl}^2/R^2$  should be replaced with  $\xi_{n_x n_y n_z}(\mathbf{r}_e)$  and  $(\hbar^2/2m_e) (\pi/L)^2 (n_x^2 + n_y^2 + n_z^2)$ , respectively. In both cases, the second term on the left-hand side of Eq. (F.37) shows the Coulomb potential for the hole averaged by the electron. Numerical calculations are required to solve these equations.

### F.3 Electric-Dipole-Forbidden Transition

On the basis of the above discussion, the behavior of an electron-hole pair when it is excited by a DP or by propagating light is now examined. In order to make the difference clear, case (2) in Sect. F.2 is examined. It is then convenient to use a Wannier function basis, which is a complete set of orthogonal functions representing electrons localized at an atomic site  $\mathbf{R}$ . The Wannier function  $w_{b\mathbf{R}}(\mathbf{r})$  is defined by

$$w_{b\mathbf{R}}(\mathbf{r}) \equiv \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} \exp(-i\mathbf{k} \cdot \mathbf{R}) \psi_{b\mathbf{k}}(\mathbf{r}), \quad (\text{F.38})$$

with the Bloch function  $\psi_{b\mathbf{k}}(\mathbf{r})$ , which is a plane wave modulated by the periodicity of the lattice and is obtained from a linear combination of electron wave-functions in an isolated atom at an arbitrary site. Here,  $N$  is the number of constituent atoms. The Wannier functions for different bands (identified by the subscript  $b$ ) and different sites  $\mathbf{R}$  are orthogonal, which follows from

$$\begin{aligned} & \int w_{b\mathbf{R}}^*(\mathbf{r}) w_{b'\mathbf{R}'}(\mathbf{r}) d^3r \\ & = \frac{1}{N} \sum_{\mathbf{k}, \mathbf{k}'} \exp[i(\mathbf{k} \cdot \mathbf{R} - \mathbf{k}' \cdot \mathbf{R}')] \int \psi_{b\mathbf{k}}^*(\mathbf{r}) \psi_{b'\mathbf{k}'}(\mathbf{r}) d^3r \\ & = \frac{1}{N} \sum_{\mathbf{k}, \mathbf{k}'} \exp[i(\mathbf{k} \cdot \mathbf{R} - \mathbf{k}' \cdot \mathbf{R}')] \delta_{bb'} \delta_{\mathbf{k}\mathbf{k}'} \\ & = \frac{1}{N} \sum_{\mathbf{k}, \mathbf{k}'} \exp[i\mathbf{k} \cdot (\mathbf{R} - \mathbf{R}')] \delta_{bb'} = \delta_{bb'} \delta_{\mathbf{R}\mathbf{R}'}, \end{aligned} \quad (\text{F.39})$$

The field operator of electron creation in the conduction band and that of hole creation in the valence band can be expressed in terms of the Wannier bases. In case (2), where an electron–hole pair (an exciton) is confined in a QD, the exciton state  $|\Phi_\nu\rangle$  specified by the quantum numbers  $\nu = (m, \mu)$  is represented as a superposition of electron states at  $\mathbf{R}$  and hole states at  $\mathbf{R}'$ :

$$|\Phi_\nu\rangle = \sum_{\mathbf{R}, \mathbf{R}'} F_m(\mathbf{R}_{CM}) \varphi_\mu(\beta) \hat{e}_{c\mathbf{R}}^\dagger \hat{e}_{v\mathbf{R}'} |\Phi_g\rangle. \quad (\text{F.40})$$

Here,  $F_m(\mathbf{R}_{CM})$  is the motion of the center of mass of the exciton specified by a set of quantum numbers  $\mathbf{m} = (m_x, m_y, m_z)$ , whereas  $\varphi_\mu(\beta)$  is the relative motion specified by the quantum number  $\mu$ , and the product of them represents the envelope function of the exciton. The creation operator of an electron at  $\mathbf{R}$  in the conduction band is denoted by  $\hat{e}_{c\mathbf{R}}^\dagger$ , and the annihilation operator of an electron at  $\mathbf{R}'$  in the valence band is represented by  $\hat{e}_{v\mathbf{R}'}$ .  $|\Phi_g\rangle$  is the crystal ground operator.

### F.3.1 Excitation by Dressed Photons

In order to derive the effective interaction energy between two QDs based on Eq. (2.31), the matrix element representing the transition from the exciton state  $|\Phi_\nu\rangle$  to the crystal ground state  $|\Phi_g\rangle$  is calculated as

$$\begin{aligned} \langle \Phi_g | \hat{V} | \Phi_\nu \rangle &= \sum_{k, \lambda} \sum_{\mathbf{R}, \mathbf{R}'} F_m(\mathbf{R}_{CM}) \varphi_\mu(\beta) \\ &\times \left( \hat{\xi}(\mathbf{k}) g_{v\mathbf{R}'c\mathbf{R}, k\lambda} - \hat{\xi}^\dagger(\mathbf{k}) g_{v\mathbf{R}'c\mathbf{R}, -k\lambda} \right) \end{aligned} \quad (\text{F.41})$$

with

$$g_{v\mathbf{R}'c\mathbf{R}, k\lambda} = -i \sqrt{\frac{\hbar}{2\varepsilon_0 V}} f(k) \int w_{v\mathbf{R}'}^*(\mathbf{r}) \mathbf{p}(\mathbf{r}) w_{c\mathbf{R}}(\mathbf{r}) \cdot \mathbf{e}_\lambda(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} d^3r, \quad (\text{F.42})$$

which was derived using the fact that the expectation values of  $\langle \Phi_g | \hat{e}_{v\mathbf{R}_1}^\dagger \hat{e}_{c\mathbf{R}_2} \hat{e}_{c\mathbf{R}}^\dagger \hat{e}_{v\mathbf{R}'} |\Phi_g\rangle$  are not zero only if  $\mathbf{R}' = \mathbf{R}_1$  and  $\mathbf{R} = \mathbf{R}_2$  hold. By transforming the spatial integral to the sum of the unit cells and by noting the spatial locality of the Wannier functions, one finds that Eq. (F.42) is proportional to  $\delta_{\mathbf{R}\mathbf{R}'}$ . Defining the electric dipole moment for each unit cell as

$$\mathbf{p}_{cv} = \int_{UC} w_{v\mathbf{R}'}^*(\mathbf{r}) \mathbf{p}(\mathbf{r}) w_{c\mathbf{R}}(\mathbf{r}) d^3r \quad (\text{F.43})$$

and noting that it is the same as that of the bulk material, independent of the site  $\mathbf{R}$ , the final form of Eq. (F.41) is

$$\begin{aligned} \langle \Phi_g | \hat{V} | \Phi_v \rangle &= -i \sqrt{\frac{\hbar}{2\varepsilon_0 V}} \sum_{\mathbf{k}} \sum_{\lambda=1}^2 \sum_{\mathbf{R}} f(\mathbf{k}) [\mathbf{p}_{cv} \cdot \mathbf{e}_\lambda(\mathbf{k})] F_m(\mathbf{R}) \varphi_\mu(0) \\ &\times \left\{ \hat{\xi}(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{R}} - \hat{\xi}^\dagger(\mathbf{k}) e^{-i\mathbf{k} \cdot \mathbf{R}} \right\}. \end{aligned} \quad (\text{F.44})$$

Here it should be noted that the long-wavelength approximation  $e^{\pm i\mathbf{k} \cdot \mathbf{R}} \simeq 1$  is not applied because the energy of the DP is localized in a nanometric space.

According to the effective interaction energy between the two QDs given by Eq. (2.31), the initial and final states in the P space are set to  $|\phi_{Pi}\rangle = |\Phi_{m\mu}^A\rangle |\Phi_g^B\rangle |0\rangle$  and  $|\phi_{Pf}\rangle = |\Phi_g^A\rangle |\Phi_{m'\mu'}^B\rangle |0\rangle$ , respectively. As the intermediate states  $|\phi_{Qj}\rangle$  in the Q space,  $|\Phi_g^A\rangle |\Phi_g^B\rangle |\mathbf{k}\rangle$  and  $|\Phi_{m\mu}^A\rangle |\Phi_{m'\mu'}^B\rangle |\mathbf{k}\rangle$  are employed, where  $\mathbf{k}$  is the wave-vector of the exciton-polariton. The superscripts A and B are used to label the two QDs. Using Eq. (F.44), one can rewrite Eq. (2.31) as

$$\begin{aligned} V_{\text{eff}} &= \varphi_\mu^A(0) \varphi_{\mu'}^{B*}(0) \\ &\times \int \int F_m^A(\mathbf{R}_A) F_{m'}^{B*}(\mathbf{R}_B) [Y_A(\mathbf{R}_A - \mathbf{R}_B) + Y_B(\mathbf{R}_A - \mathbf{R}_B)] d^3 R_A d^3 R_B, \end{aligned} \quad (\text{F.45})$$

where  $\sum_{\mathbf{R}}$  in Eq. (F.44) was transformed to the integral form. The integral kernels  $Y_\alpha(\mathbf{R}_{AB})$  with  $\mathbf{R}_{AB} = \mathbf{R}_A - \mathbf{R}_B$ , which connect the two spatially isolated envelope functions  $F_m^A(\mathbf{R}_A)$  and  $F_{m'}^B(\mathbf{R}_B)$ , are defined by

$$\begin{aligned} Y_\alpha(\mathbf{R}_{AB}) &= -\frac{\hbar^2}{(2\pi)^3 \varepsilon_0} \sum_{\lambda=1}^2 \int \left[ \mathbf{p}_{cv}^A \cdot \mathbf{e}_\lambda(\mathbf{k}) \right] \left[ \mathbf{p}_{cv}^B \cdot \mathbf{e}_\lambda(\mathbf{k}) \right] \hbar f^2(k) \\ &\times \left\{ \frac{1}{E(k) + E_\alpha} + \frac{1}{E(k) - E_\alpha} \right\} e^{i\mathbf{k} \cdot \mathbf{R}_{AB}} d\mathbf{k}. \end{aligned} \quad (\text{F.46})$$

Here, the electric dipole moment  $\mathbf{p}_{cv}^\alpha$  for QD $_\alpha$  ( $\alpha = A, B$ ) is defined by Eq. (F.43), and  $E_\alpha$  denotes the exciton energy in QD $_\alpha$ . Then Eq. (F.46) can be rewritten in the same way as described in Chap. 2, for example, corresponding to Eq. (2.75):

$$Y_\alpha(\mathbf{R}_{AB}) = -\frac{p_{cv}^A p_{cv}^B}{3(2\pi) \varepsilon_0} \left( W_\alpha + \Delta_\alpha^2 + \frac{e^{-\Delta_\alpha + R_{AB}}}{R_{AB}} - W_{\alpha-} \Delta_{\alpha-}^2 - \frac{e^{-\Delta_\alpha - R_{AB}}}{R_{AB}} \right), \quad (\text{F.47})$$

where  $R_{AB} = |\mathbf{R}_{AB}|$  was used.

### F.3.2 Excitation by Propagating Light

Since the electric displacement vector of the propagating light is spatially homogeneous in the QDs, the long-wavelength approximation  $e^{\pm ik \cdot \mathbf{R}} \simeq 1$  is applied. Then, the transition matrix elements can be written in separated form in terms of  $\mathbf{R}$  and  $(\mathbf{k}, \lambda)$  as

$$\begin{aligned} & \langle \Phi_g | \hat{V} | \Phi_\nu^\alpha \rangle \\ &= -i\sqrt{\frac{\hbar}{2\varepsilon_0 V}} \sum_{\mathbf{R}} F_m^\alpha(\mathbf{R}) \varphi_\mu^\alpha(0) \sum_{\mathbf{k}} \sum_{\lambda=1}^2 f(k) [\mathbf{p}_{c\nu} \cdot \mathbf{e}_\lambda(\mathbf{k})] \left\{ \hat{\xi}(\mathbf{k}) - \hat{\xi}^\dagger(\mathbf{k}) \right\} \\ &= -i\sqrt{\frac{\hbar}{2\varepsilon_0 V}} \left[ \int F_m^\alpha(\mathbf{R}) d\mathbf{R} \right] \varphi_\mu^\alpha(0) \sum_{\mathbf{k}} \sum_{\lambda=1}^2 f(k) [\mathbf{p}_{c\nu} \cdot \mathbf{e}_\lambda(\mathbf{k})] \left\{ \hat{\xi}(\mathbf{k}) - \hat{\xi}^\dagger(\mathbf{k}) \right\}. \end{aligned} \quad (\text{F.48})$$

The integral  $\int F_m^\alpha(\mathbf{R}) d\mathbf{R}$ , as well as  $\mathbf{p}_{c\nu}$ , provide a criterion for whether the electric dipole transition between the crystal ground state  $|\Phi_g\rangle$  and the exciton state  $|\Phi_\nu^\alpha\rangle$  in the QD $_\alpha$  specified by the quantum number  $\nu = (\mathbf{m}, \mu)$  is allowed or forbidden. Thus, it follows that the electric dipole transition is forbidden if the spatial integral  $\int F_m^\alpha(\mathbf{R}) d\mathbf{R}$  is zero, whereas it is allowed if the integral is not zero. Since the spatial integral for a spherical QD, as an example, is given by

$$\begin{aligned} \int d^3 r F_m(\mathbf{r}) &= \sqrt{\frac{2}{R^3}} \int_0^R r^2 dr \frac{j_l\left(\frac{\alpha_n l r}{R}\right)}{j_{l+1}(\alpha_n l)} \int \int \sin \theta d\theta d\phi Y_{lm}(\theta, \phi) \quad (\text{F.49}) \\ &= \frac{1}{n} \sqrt{\frac{2R^3}{\pi^2}} \delta_{l0} \delta_{m0}, \end{aligned}$$

the transition only to the state specified by  $l = m = 0$  is allowed. Similarly when all of the integrands are even functions, the nonzero result

$$\begin{aligned} & \int d^3 r F_m(\mathbf{r}) \quad (\text{F.50}) \\ &= \sqrt{\frac{8}{L^3}} \int_{-\frac{L}{2}}^{\frac{L}{2}} dx \cos\left(\frac{(2n_x - 1)\pi x}{L}\right) \\ &\quad \times \int_{-\frac{L}{2}}^{\frac{L}{2}} dy \cos\left(\frac{(2n_y - 1)\pi y}{L}\right) \int_{-\frac{L}{2}}^{\frac{L}{2}} dz \cos\left(\frac{(2n_z - 1)\pi z}{L}\right) \\ &= \sqrt{\frac{512L^3}{\pi^6}} \frac{1}{(2n_x - 1)} \frac{1}{(2n_y - 1)} \frac{1}{(2n_z - 1)} \\ &\quad \times \sin\left(\frac{(2n_x - 1)\pi}{2}\right) \sin\left(\frac{(2n_y - 1)\pi}{2}\right) \sin\left(\frac{(2n_z - 1)\pi}{2}\right) \\ &= \sqrt{\frac{512L^3}{\pi^6}} \frac{1}{(2n_x - 1)} \frac{1}{(2n_y - 1)} \frac{1}{(2n_z - 1)} \end{aligned}$$

is obtained for a cubic QD, and thus the transitions are allowed if all of  $(n_x, n_y, n_z)$  are odd, whereas they are forbidden if any one of  $(n_x, n_y, n_z)$  is even.

## References

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# Appendix G

## Solutions of the Quantum Master Equations for the Density Matrix Operators

### G.1 The Case of the Two Quantum Dots

In order to derive Eqs. (3.18a)–(3.18d) in Chap. 3 for the case of  $n = 0$ ,  $\rho_{12}(t) - \rho_{21}(t)$  is denoted by  $\Delta\rho_{12}(t)$ . Then, Eq. (3.18b) can be rewritten as

$$\frac{d\Delta\rho_{12}(t)}{dt} = 2iU(r) [\rho_{11}(t) - \rho_{22}(t)] - \gamma\Delta\rho_{12}(t). \tag{G.1}$$

Equations (3.18a) and (3.18c) are also rewritten as

$$\frac{d\rho_{11}(t)}{dt} = iU(r) \Delta\rho_{12}(t), \tag{G.2}$$

$$\frac{d\rho_{22}(t)}{dt} = -iU(r) \Delta\rho_{12}(t) - 2\gamma\rho_{22}(t) + 2\gamma\rho_{33}(t). \tag{G.3}$$

Laplace transformations of these equations give

$$s\rho_{11}(s) - \rho_{11}(0) = iU(r) \Delta\rho_{12}(s), \tag{G.4}$$

$$\Delta\rho_{12}(s) - \Delta\rho_{12}(0) = 2iU(r) [\rho_{11}(s) - \rho_{22}(s)] - \gamma\Delta\rho_{12}(s) \tag{G.5}$$

$$s\rho_{22}(s) - \rho_{22}(0) = -iU(r) \Delta\rho_{12}(s) - 2\gamma\rho_{22}(s). \tag{G.6}$$

Under the initial conditions of  $\rho_{11}(0) = 1$ ,  $\Delta\rho_{12}(0) = 0$ , and  $\rho_{22}(0) = 0$ , these equations can be solved simultaneously, and the solutions are

$$\rho_{11}(s) = \frac{s^2 + 3\gamma s + 2(U^2 + \gamma^2)}{(s + \gamma)(s^2 + 2\gamma s + 4U^2)}, \tag{G.7}$$

$$\rho_{22}(s) = \frac{2U^2}{(s + \gamma)(s^2 + 2\gamma s + 4U^2)}, \quad (\text{G.8})$$

$$\Delta\rho_{12}(s) = \frac{2iU(s + 2\gamma)}{(s + \gamma)(s^2 + 2\gamma s + 4U^2)}. \quad (\text{G.9})$$

By defining a parameter

$$Z \equiv \sqrt{(\gamma/2)^2 - U^2}, \quad (\text{G.10})$$

the term  $s^2 + 2\gamma s + 4U^2$  in the denominators on the right-hand sides of Eqs. (G.7)–(G.9) is transformed to  $(s + \gamma + 2Z)(s + \gamma - 2Z)$ . Then, the inverse-Laplace transformations of Eqs. (G.7)–(G.9) give

$$\rho_{11}(t) = \frac{1}{8Z^2} e^{-(2Z+\gamma)t} \left[ -2U^2(1 + e^{2Zt}) + \gamma \left\{ 2Z(-1 + e^{4Zt}) + \gamma(1 + e^{4Zt}) \right\} \right], \quad (\text{G.11})$$

$$\rho_{22}(t) = -\frac{U^2}{4Z^2} e^{-(2Z+\gamma)t} (-1 + e^{2Zt})^2, \quad (\text{G.12})$$

$$\Delta\rho_{12}(t) = \frac{iU}{4Z^2} e^{-(2Z+\gamma)t} (-1 + e^{2Zt}) \left\{ 2Z(1 + e^{2Zt}) + \gamma(-1 + e^{2Zt}) \right\}. \quad (\text{G.13})$$

By rewriting the exponential functions in these equations as hyperbolic sinusoidal and cosinusoidal functions, Eqs. (3.19a)–(3.19c) in Chap. 3 are derived.

## G.2 XOR Logic Gate Composed of Three Quantum Dots

Laplace transformations of Eqs. (3.23a)–(3.23e) in Chap. 3 give

$$s\rho_{S_1, S_1}(s) - \rho_{S_1, S_1}(0) = i\sqrt{2}U' \left\{ \rho_{S_1, P'_1}(s) - \rho_{P'_1, S_1}(s) \right\}, \quad (\text{G.14a})$$

$$s\rho_{S_1, P'_1}(s) - \rho_{S_1, P'_1}(0) = \left\{ i(\Delta\Omega - U) - \frac{\gamma}{2} \right\} \rho_{S_1, P'_1}(s) + i\sqrt{2}U' \left\{ \rho_{S_1, S_1}(s) - \rho_{P'_1, P'_1}(s) \right\}, \quad (\text{G.14b})$$

$$s\rho_{P'_1, S_1}(s) - \rho_{P'_1, S_1}(0) = -\left\{ i(\Delta\Omega - U) + \frac{\gamma}{2} \right\} \rho_{P'_1, S_1}(s) - i\sqrt{2}U' \left\{ \rho_{S_1, S_1}(s) - \rho_{P'_1, P'_1}(s) \right\}, \quad (\text{G.14c})$$

$$s\rho_{P'_1, P'_1}(s) - \rho_{P'_1, P'_1}(0) = -\gamma\rho_{P'_1, P'_1}(s) - i\sqrt{2}U' \left\{ \rho_{S_1, P'_1}(s) - \rho_{P'_1, S_1}(s) \right\}, \quad (\text{G.14d})$$

$$s\rho_{P_1, P_1}(s) = \gamma\rho_{P_1, P_1}(s). \quad (\text{G.14e})$$

These are solved simultaneously under the initial conditions

$$\rho_{S_1, S_1}(0) = 1/2, \quad \rho_{S_1, P'_1}(0) = \rho_{P'_1, S_1}(0) = \rho_{P'_1, P'_1}(0) = \rho_{P_1, P_1}(0) = 0. \quad (\text{G.15})$$

By defining

$$\omega_{\pm} \equiv -\frac{1}{\sqrt{2}} \sqrt{(\Delta\Omega - U)^2 + W_+ W_- \pm \sqrt{\{(\Delta\Omega - U)^2 + W_-^2\} \{(\Delta\Omega - U)^2 + W_+^2\}}}, \quad (\text{G.16})$$

$$W_{\pm} \equiv 2\sqrt{2}U' \pm \frac{\gamma}{2} \quad (\text{G.17})$$

the solution of  $\rho_{P_1, P_1}(t)$  on the left-hand side of Eq. (G.14e) is expressed as

$$\begin{aligned} \rho_{P_1, P_1}(s) = & -\frac{4iU'^2\gamma}{(2s + \gamma + 2i\omega_-)(-i\gamma + 2\omega_-)(\omega_-^2 - \omega_+^2)} \quad (\text{G.18}) \\ & + \frac{4iU'^2\gamma}{(2s + \gamma - 2i\omega_-)(i\gamma + 2\omega_-)(\omega_-^2 - \omega_+^2)} \\ & + \frac{4iU'^2\gamma}{(2s + \gamma + 2i\omega_+)(-i\gamma + 2\omega_+)(\omega_-^2 - \omega_+^2)} \\ & - \frac{4iU'^2\gamma}{(2s + \gamma - 2i\omega_+)(i\gamma + 2\omega_+)(\omega_-^2 - \omega_+^2)} + \frac{16U'^2\gamma^2}{s(\gamma^2 + 4\omega_-^2)(\gamma^2 + 4\omega_+^2)}. \end{aligned}$$

Since the inverse-Laplace transformations of the first and second terms in Eq. (G.18) are

$$\frac{2e^{-(\gamma+2i\omega_-)\frac{t}{2}} U'^2\gamma \{(1 + e^{2i\omega_-t})\gamma + 2i(-1 + e^{2i\omega_-t})\omega_-\}}{(\gamma^2 + 4\omega_-^2)(\omega_-^2 - \omega_+^2)}, \quad (\text{G.19})$$

replacement of the exponential functions in this equation with sinusoidal and cosinusoidal functions gives

$$\begin{aligned} & -\frac{4U'^2}{\omega_+^2 - \omega_-^2} e^{-\frac{\gamma t}{2}} \frac{\gamma}{\sqrt{\gamma^2 + 4\omega_-^2}} \left( \frac{\gamma}{\sqrt{\gamma^2 + 4\omega_-^2}} \cos(\omega_-t) - \frac{2\omega_-}{\sqrt{\gamma^2 + 4\omega_-^2}} \sin(\omega_-t) \right) \\ & = -\frac{4U'^2}{\omega_+^2 - \omega_-^2} e^{-\frac{\gamma t}{2}} \cos(\phi_-) \cos(\omega_-t + \phi_-), \quad (\text{G.20}) \end{aligned}$$

where

$$\phi_- = \tan^{-1} \left( \frac{2\omega_-}{\gamma} \right) \quad (\text{G.21})$$

was used. Similarly, since the inverse-Laplace transformations of the third and fourth terms are

$$\frac{2e^{-\frac{i}{2}(\gamma+2i\omega_+)t}U'^2\gamma\{(1+e^{2i\omega_+t})\gamma+2i(-1+e^{2i\omega_+t})\omega_+\}}{(\gamma^2+4\omega_+^2)(-\omega_-^2+\omega_+^2)}, \quad (\text{G.22})$$

replacement of the exponential functions in this equation with sinusoidal and cosinusoidal functions gives

$$\begin{aligned} & \frac{4U'^2}{\omega_+^2-\omega_-^2}e^{-\frac{\gamma t}{2}}\frac{\gamma}{\sqrt{\gamma^2+4\omega_+^2}} \\ & \times \left( \frac{\gamma}{\sqrt{\gamma^2+4\omega_+^2}}\cos(\omega_+t) - \frac{2\omega_+}{\sqrt{\gamma^2+4\omega_+^2}}\sin(\omega_+t) \right) \\ & = \frac{4U'^2}{\omega_+^2-\omega_-^2}e^{-\frac{\gamma t}{2}}\cos(\phi_+)\cos(\omega_+t+\phi_+), \end{aligned} \quad (\text{G.23})$$

where

$$\phi_+ = \tan^{-1}\left(\frac{2\omega_+}{\gamma}\right) \quad (\text{G.24})$$

was used. The inverse-Laplace transformation of the fifth term is

$$\frac{16U'^2\gamma^2}{(\gamma^2+4\omega_-^2)(\gamma^2+4\omega_+^2)}, \quad (\text{G.25})$$

which is found to be equal to 1/2 by noting the definition of given by Eq. (G.16). Finally, summation of the five terms given above derives Eq. (3.25) in Chap. 3.

### G.3 AND Logic Gate Composed of Three Quantum Dots

Laplace transformations of Eqs. (3.33a)–(3.33d) in Chap. 3 give

$$s\rho_{S'_2,S'_2}(s) - \rho_{S'_2,S'_2}(0) = i\sqrt{2}U' \left\{ \rho_{S'_2,P'_2}(s) - \rho_{P'_2,S'_2}(s) \right\} - \gamma\rho_{S'_2,S'_2}(s), \quad (\text{G.26a})$$

$$\begin{aligned} s\rho_{S'_2,P'_2}(s) - \rho_{S'_2,P'_2}(0) &= \left\{ i(\Delta\Omega + U) + \frac{\gamma}{2} \right\} \rho_{S'_2,P'_2}(s) \\ &+ i\sqrt{2}U' \left\{ \rho_{S'_2,S'_2}(s) - \rho_{P'_2,P'_2}(s) \right\}, \end{aligned} \quad (\text{G.26b})$$

$$\begin{aligned} s\rho_{P'_2,S'_2}(s) - \rho_{P'_2,S'_2}(0) &= \left\{ i(\Delta\Omega + U) - \frac{\gamma}{2} \right\} \rho_{P'_2,S'_2}(s) \\ &- i\sqrt{2}U' \left\{ \rho_{S'_2,S'_2}(s) - \rho_{P'_2,P'_2}(s) \right\}, \end{aligned} \quad (\text{G.26c})$$

$$s\rho_{P'_2, P'_2}(s) - \rho_{P'_2, P'_2}(0) = -i\sqrt{2}U' \left\{ \rho_{S'_2, P'_2}(s) - \rho_{P'_2, S'_2}(s) \right\}. \quad (\text{G.26d})$$

The Laplace transformation of the first row of Eq. (3.34) is expressed as

$$\rho_{S_2, S_2}(s) + \rho_{P_2, P_2}(s) = \frac{\gamma}{s} \rho_{S'_2, S'_2}(s). \quad (\text{G.26e})$$

Equation (G.26e) is derived by solving the simultaneous equations of Eqs. (G.26a)–(G.26d) under the initial conditions

$$\rho_{S'_2, S'_2}(0) = 0, \quad \rho_{S'_2, P'_2}(0) = \rho_{P'_2, S'_2}(0) = 0, \quad \rho_{P'_2, P'_2} = 1. \quad (\text{G.27})$$

Inserting the solution of into Eq. (G.26e) yields

$$\begin{aligned} \rho_{S_2, S_2}(s) + \rho_{P_2, P_2}(s) &= 8U'^2\gamma(2s + \gamma) & (\text{G.28}) \\ &/s \left\{ 4s^4 + 8s^3\gamma + 8U'^2\gamma^2 + 4(\Delta\Omega)^2s(s + \gamma) + 8(\Delta\Omega)sU(s + \gamma) \right. \\ &\quad \left. + s\gamma(4U^2 + 32U^2 + \gamma^2) + s^2(4U^2 + 32U^2 + 5\gamma^2) \right\}. \end{aligned}$$

By defining

$$\omega'_\pm \equiv \frac{1}{\sqrt{2}} \sqrt{(\Delta\Omega + U)^2 + W_+ W_- \pm \sqrt{\{(\Delta\Omega + U)^2 + W_-^2\} \{(\Delta\Omega + U)^2 + W_+^2\}}}, \quad (\text{G.29})$$

$$W_\pm = 2\sqrt{2}U' \pm \frac{\gamma}{2}, \quad (\text{G.30})$$

Equation (G.28) is rewritten as

$$\begin{aligned} \rho_{S_2, S_2}(s) + \rho_{P_2, P_2}(s) &= -\frac{8iU'^2\gamma}{(2s + \gamma + 2i\omega'_-)(-i\gamma + 2\omega'_-)(\omega'^2_- - \omega'^2_+)} & (\text{G.31}) \\ &+ \frac{8iU'^2\gamma}{(2s + \gamma - 2i\omega'_-)(i\gamma + 2\omega'_-)(\omega'^2_- - \omega'^2_+)} \\ &+ \frac{8iU'^2\gamma}{(2s + \gamma + 2i\omega'_+)(-i\gamma + 2\omega'_+)(\omega'^2_- - \omega'^2_+)} \\ &- \frac{8iU'^2\gamma}{(2s + \gamma - 2i\omega'_+)(i\gamma + 2\omega'_+)(\omega'^2_- - \omega'^2_+)} \\ &+ \frac{32U'^2\gamma^2}{s(\gamma^2 + 4\omega'^2_-)(\gamma^2 + 4\omega'^2_+)}. \end{aligned}$$

Since the inverse-Laplace transformations of the first and second terms on the right-hand side are

$$\frac{4e^{-(\gamma+2i\omega'_-)\frac{t}{2}}U'^2\gamma\left\{\left(1+e^{2i\omega'_-t}\right)\gamma+2i\left(-1+e^{2i\omega'_-t}\right)\omega'_-\right\}}{\left(\gamma^2+4\omega'^2_-\right)\left(\omega'^2_--\omega'^2_+\right)}, \quad (\text{G.32})$$

replacement of the exponential functions in this equations with sinusoidal and cosinusoidal functions gives

$$-\frac{8U'^2}{\omega'^2_+-\omega'^2_-}e^{-\left(\frac{\gamma}{2}\right)t}\cos\left(\phi'_-\right)\cos\left(\omega'_-t+\phi'_-\right), \quad (\text{G.33})$$

where

$$\phi'_-=\tan^{-1}\left(\frac{2\omega'_-}{\gamma}\right) \quad (\text{G.34})$$

was used. Similarly, since the inverse-Laplace transformations of the third and fourth terms are

$$\frac{4e^{-\frac{t}{2}(\gamma+2i\omega'_+)}U'^2\gamma\left\{\left(1+e^{2i\omega'_+t}\right)\gamma+2i\left(-1+e^{2i\omega'_+t}\right)\omega'_+\right\}}{\left(\gamma^2+4\omega'^2_+\right)\left(-\omega'^2_--\omega'^2_+\right)}, \quad (\text{G.35})$$

replacement of the exponential functions in this equation with sinusoidal and cosinusoidal functions give

$$\frac{8U'^2}{\omega'^2_+-\omega'^2_-}e^{-\frac{\gamma t}{2}}\cos\left(\phi'_+\right)\cos\left(\omega'_+t+\phi'_+\right), \quad (\text{G.36})$$

where

$$\phi'_+=\tan^{-1}\left(\frac{2\omega'_+}{\gamma}\right) \quad (\text{G.37})$$

was used. The inverse-Laplace transformation of the fifth term is

$$\frac{32U'^2\gamma^2}{\left(\gamma^2+4\omega'^2_-\right)\left(\gamma^2+4\omega'^2_+\right)}, \quad (\text{G.38})$$

which is found to be equal to 1 by noting the definition of  $\omega'_\pm$  given by Eq. (G.29). Finally, summation of the five terms given above derives Eq. (3.34) in Chap. 3.

# Appendix H

## Derivation of Equations in Chap. 4

### H.1 Unitary Transformation

This section reviews how to diagonalize the Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{V} \tag{H.1}$$

where  $\hat{H}_0$  and  $\hat{V}$  are unperturbed and interaction Hamiltonians, respectively [1]. In order to transform  $\hat{H}$  to the diagonalized Hamiltonian

$$\tilde{H} = \hat{U}\hat{H}\hat{U}^\dagger \tag{H.2}$$

by a unitary transformation, the unitary operators  $\hat{U}$  and  $\hat{U}^\dagger$  are defined by

$$\hat{U} \equiv e^{\hat{S}}, \tag{H.3a}$$

$$\hat{U}^\dagger = \hat{U}^{-1}, \tag{H.3b}$$

where  $\hat{S}$  is an anti-Hermitian operator satisfying

$$\hat{S}^\dagger = -\hat{S}. \tag{H.4}$$

Inserting Eqs. (H.3a) and (H.3b) into Eq. (H.2) and performing polynomial expansion gives

$$\begin{aligned} \tilde{H} &= \hat{U}\hat{H}\hat{U}^\dagger = e^{\hat{S}}\hat{H}e^{-\hat{S}} = \left(1 + \hat{S} + \frac{1}{2!}\hat{S}^2 + \dots\right)\hat{H}\left(1 - \hat{S} + \frac{1}{2!}\hat{S}^2 + \dots\right) \\ &= \hat{H} + \hat{S}\hat{H} - \hat{H}\hat{S} + \frac{1}{2!}\left(\hat{S}^2\hat{H} - 2\hat{S}\hat{H}\hat{S} + \hat{H}\hat{S}^2\right) + \dots \end{aligned} \tag{H.5}$$

$$\begin{aligned}
&= \hat{H} + [\hat{S}, \hat{H}] + \frac{1}{2!} [\hat{S}, [\hat{S}, \hat{H}]] + \dots \\
&= \hat{H}_0 + \hat{V} + [\hat{S}, \hat{H}_0] + [\hat{S}, \hat{V}] + \frac{1}{2!} [\hat{S}, [\hat{S}, \hat{H}_0]] + \dots
\end{aligned}$$

If the anti-Hermitian operator  $\hat{S}$  satisfies the relation

$$\hat{V} = -[\hat{S}, \hat{H}_0], \quad (\text{H.6})$$

Equation (H.5) is reduced to

$$\tilde{H} = \hat{H}_0 - \frac{1}{2} [\hat{S}, [\hat{S}, \hat{H}_0]] + \dots \quad (\text{H.7})$$

In the case where the value of the interaction Hamiltonian  $\hat{V}$  is sufficiently small, Eq. (H.7) can be treated perturbatively. That is, by neglecting the terms higher than the second order of  $\hat{S}$  in this equation, the Hamiltonian  $\hat{H}$  can be diagonalized as

$$\tilde{H} = \hat{U} \hat{H} \hat{U}^\dagger = e^{\hat{S}} \hat{H} e^{-\hat{S}} \simeq \hat{H}_0. \quad (\text{H.8})$$

Based on the formulation given above, the Hamiltonian for the DP–phonon interaction

$$\hat{H}' = \sum_{i=1}^N \hbar \omega \tilde{a}_i^\dagger \tilde{a}_i + \sum_{p=1}^N \hbar \Omega_p \hat{c}_p^\dagger \hat{c}_p + \sum_{i=1}^N \sum_{p=1}^N \hbar \chi_{ip} \tilde{a}_i^\dagger \tilde{a}_i (\hat{c}_p^\dagger + \hat{c}_p) \quad (\text{H.9})$$

is diagonalized. Here, the fourth term of Eq. (4.44) in Chap. 4, representing DP-hopping, has been excluded. Operators in this equation satisfy the boson commutation relation as follows:

$$[\tilde{a}_i, \tilde{a}_j^\dagger] = \delta_{ij}, \quad (\text{H.10})$$

$$[\hat{c}_p, \hat{c}_q^\dagger] = \delta_{pq}, \quad (\text{H.11})$$

$$[\tilde{a}_i, \hat{c}_p] = [\tilde{a}_i, \hat{c}_p^\dagger] = [\tilde{a}_i^\dagger, \hat{c}_p] = [\tilde{a}_i^\dagger, \hat{c}_p^\dagger] = 0, \quad (\text{H.12})$$

$$[\tilde{a}_i, \tilde{a}_j] = [\tilde{a}_i^\dagger, \tilde{a}_j^\dagger] = [\hat{c}_p, \hat{c}_q] = [\hat{c}_p^\dagger, \hat{c}_q^\dagger] = 0.$$

The anti-Hermitian operator  $\hat{S}$  to be used for diagonalization is given by

$$\hat{S} = \sum_{i=1}^N \sum_{p=1}^N f_{ip} \tilde{a}_i^\dagger \tilde{a}_i (\hat{c}_p^\dagger - \hat{c}_p). \quad (\text{H.13})$$



Denoting the first and second terms of Eq. (H.9) by  $\hat{H}_0$ , and the third term by  $\hat{V}$ , it is found from Eq. (H.6) that the coefficient  $f_{ip}$  in Eq. (H.13) is equal to  $\chi_{ip}/\Omega_p$ . As a result, one can derive

$$\hat{S} = \sum_{i=1}^N \sum_{p=1}^N \frac{\chi_{ip}}{\Omega_p} \tilde{a}_i^\dagger \tilde{a}_i (\hat{c}_p^\dagger - \hat{c}_p), \quad (\text{H.14})$$

which corresponds to Eq. (4.26) in Chap. 4.

Since the commutation relation between  $\hat{S}$  and the Hamiltonian of Eq. (H.9) can be calculated analytically, the Hamiltonian for the DP–phonon interaction can be diagonalized without using any perturbative methods. In order to diagonalize it, the operator function

$$\hat{F}_i(t) = e^{t\hat{S}} \tilde{a}_i^\dagger e^{-t\hat{S}} \quad (\text{H.15})$$

is defined. First, differentiating it with respect to  $t$  yields

$$\begin{aligned} \frac{d}{dt} \hat{F}_i(t) &= e^{t\hat{S}} \left( \hat{S} \tilde{a}_i^\dagger - \tilde{a}_i^\dagger \hat{S} \right) e^{-t\hat{S}} \\ &= e^{t\hat{S}} \sum_{p=1}^N \frac{\chi_{ip}}{\Omega_p} \tilde{a}_i^\dagger (\hat{c}_p^\dagger - \hat{c}_p) e^{-t\hat{S}} = \hat{F}_i(t) \sum_{p=1}^N \frac{\chi_{ip}}{\Omega_p} (\hat{c}_p^\dagger - \hat{c}_p). \end{aligned} \quad (\text{H.16})$$

Second, solving this differential equation by using the initial condition  $\hat{F}_i(0) = \tilde{a}_i^\dagger$  yields

$$\hat{F}_i(t) = \tilde{a}_i^\dagger \exp \left\{ t \sum_{p=1}^N \frac{\chi_{ip}}{\Omega_p} (\hat{c}_p^\dagger - \hat{c}_p) \right\} \quad (\text{H.17})$$

and therefore

$$\hat{\alpha}_i^\dagger = \hat{U}^\dagger \tilde{a}_i^\dagger \hat{U} = \hat{F}_i(-1) = \tilde{a}_i^\dagger \exp \left\{ - \sum_{p=1}^N \frac{\chi_{ip}}{\Omega_p} (\hat{c}_p^\dagger - \hat{c}_p) \right\}. \quad (\text{H.18})$$

Similarly, for the operator function

$$\hat{G}_p(t) = e^{t\hat{S}} \hat{c}_p^\dagger e^{-t\hat{S}}, \quad (\text{H.19a})$$

one derives

$$\frac{d}{dt} \hat{G}_p(t) = - \sum_{i=1}^N \frac{\chi_{ip}}{\Omega_p} \tilde{a}_i^\dagger \tilde{a}_i, \quad (\text{H.19b})$$

and therefore

$$\hat{G}_p(t) = \hat{c}_p^\dagger - t \sum_{i=1}^N \frac{\chi_{ip}}{\Omega_p} \tilde{a}_i^\dagger \tilde{a}_i, \quad (\text{H.20})$$

$$\hat{\beta}_p^\dagger = \hat{G}_p(-1) = \hat{c}_p^\dagger + \sum_{i=1}^N \frac{\chi_{ip}}{\Omega_p} \tilde{a}_i^\dagger \tilde{a}_i. \quad (\text{H.21})$$

These derivations correspond to the proof for Eqs. (4.30) and (4.31) in Chap. 4.

In order to derive the diagonalized Hamiltonian  $\hat{H}'$  by using the equations derived above, the operators  $\tilde{a}_i^\dagger$  and  $\hat{c}_p^\dagger$  are replaced with  $\hat{F}_i(1)$  and  $\hat{G}_p(1)$ , respectively. Using them, one derives

$$\begin{aligned} \hat{H}' &= \hat{U} \hat{H}' \hat{U}^\dagger \\ &= \sum_{i=1}^N \hbar \omega \tilde{a}_i^\dagger \tilde{a}_i + \sum_{p=1}^N \hbar \Omega_p \left( \hat{c}_p^\dagger - \sum_{i=1}^N \frac{\chi_{ip}}{\Omega_p} \tilde{a}_i^\dagger \tilde{a}_i \right) \left( \hat{c}_p - \sum_{j=1}^N \frac{\chi_{jp}}{\Omega_p} \tilde{a}_j^\dagger \tilde{a}_j \right) \\ &\quad + \sum_{i=1}^N \sum_{p=1}^N \hbar \chi_{ip} \tilde{a}_i^\dagger \tilde{a}_i \left( \hat{c}_p + \hat{c}_p^\dagger - 2 \sum_{j=1}^N \frac{\chi_{jp}}{\Omega_p} \tilde{a}_j^\dagger \tilde{a}_j \right) \\ &= \sum_{i=1}^N \hbar \omega \tilde{a}_i^\dagger \tilde{a}_i + \sum_{p=1}^N \hbar \Omega_p \hat{c}_p^\dagger \hat{c}_p - \sum_{i=1}^N \sum_{j=1}^N \sum_{p=1}^N \hbar \frac{\chi_{ip} \chi_{jp}}{\Omega_p} \tilde{a}_i^\dagger \tilde{a}_i \tilde{a}_j^\dagger \tilde{a}_j. \end{aligned} \quad (\text{H.22})$$

The original Hamiltonian  $\hat{H}'$  can be derived by the inverse transformation, which is expressed by using the transformed operators  $\hat{\alpha}_i^\dagger$ ,  $\hat{\alpha}_i$ ,  $\hat{\beta}_p^\dagger$ , and  $\hat{\beta}_p$  as

$$\begin{aligned} \hat{H}' &= \hat{U}^\dagger \hat{H}' \hat{U} \\ &= \sum_{i=1}^N \hbar \omega \hat{\alpha}_i^\dagger \hat{\alpha}_i + \sum_{p=1}^N \hbar \Omega_p \hat{\beta}_p^\dagger \hat{\beta}_p - \sum_{i=1}^N \sum_{j=1}^N \sum_{p=1}^N \hbar \frac{\chi_{ip} \chi_{jp}}{\Omega_p} \hat{\alpha}_i^\dagger \hat{\alpha}_i \hat{\alpha}_j^\dagger \hat{\alpha}_j. \end{aligned} \quad (\text{H.23})$$

Since the term for DP-hopping can also be transformed by expressing  $\tilde{a}_i^\dagger$  in Eq. (H.18) by  $\hat{\alpha}_i^\dagger$  and by replacing  $\hat{c}_p^\dagger - \hat{c}_p$  with  $\hat{\beta}_p^\dagger - \hat{\beta}_p$ , the Hamiltonian and the hopping operator can be derived, as shown by Eqs. (4.35) and (4.36) in Chap. 4, respectively.

## H.2 Coherent State

In the coherent state  $|\gamma\rangle$ , an infinite number of quasi-particles cohere with each other, which can be expressed by using annihilation ( $\hat{c}$ ) and creation ( $\hat{c}^\dagger$ ) operators as

$$|\gamma\rangle = e^{\gamma(\hat{c}^\dagger - \hat{c})} |0\rangle. \quad (\text{H.24})$$

Here, for simplicity, quasi-particles of the single mode are considered and the coefficient  $\gamma$  is assumed to take real numbers. Differentiating the operator function

$$f(\gamma) = e^{\gamma(\hat{c}^\dagger - \hat{c})} \quad (\text{H.25})$$

with respect to  $\gamma$  yields<sup>3</sup>

$$\frac{df}{d\gamma} = \hat{c}^\dagger f - f \hat{c} = (\hat{c}^\dagger - \hat{c} + \gamma)f. \quad (\text{H.26})$$

Equating the solution of this differential equation,

$$f = e^{\gamma(\hat{c}^\dagger - \hat{c})} e^{\frac{1}{2}\gamma^2}, \quad (\text{H.27})$$

with Eq. (H.25), gives the relation

$$e^{\gamma(\hat{c}^\dagger - \hat{c})} = e^{-\frac{1}{2}\gamma^2} e^{\gamma\hat{c}^\dagger} e^{-\gamma\hat{c}}. \quad (\text{H.28})$$

Therefore, the coherent state can be expressed also as

$$|\gamma\rangle = e^{-\frac{1}{2}\gamma} e^{\gamma\hat{c}^\dagger} |0\rangle. \quad (\text{H.29})$$

Polynomial expansion of the exponential function gives

$$|\gamma\rangle = e^{-\frac{1}{2}\gamma^2} e^{\gamma\hat{c}^\dagger} |0\rangle = e^{-\frac{1}{2}\gamma^2} \sum_{n=0}^{\infty} \frac{(\gamma\hat{c}^\dagger)^n}{n!} |0\rangle = e^{-\frac{1}{2}\gamma^2} \sum_{n=0}^{\infty} \frac{\gamma^n}{\sqrt{n!}} |n\rangle. \quad (\text{H.30})$$

---

<sup>3</sup> If the operators  $\hat{A}$  and  $\hat{B}$  satisfy the commutation relation

$$[\hat{A}, \hat{B}] = 1, \quad (\text{a})$$

the commutation relation between  $\hat{A}$  and  $\hat{B}^n$  is derived by mathematical induction as

$$[\hat{A}, \hat{B}^n] = n\hat{B}^{n-1} = \frac{d}{d\hat{B}} \hat{B}^n. \quad (\text{b})$$

Therefore, after expanding an operator function  $f(\hat{B})$  into a power series of  $\hat{B}$ , use of Eq. (b) gives

$$[\hat{A}, f(\hat{B})] = \frac{d}{d\hat{B}} f(\hat{B}). \quad (\text{c})$$

In the case of  $\hat{A} = \hat{c}$  and  $\hat{B} = \hat{c}^\dagger - \hat{c}$ , Eq. (c) can be employed because Eq. (a) holds, and one obtains

$$\hat{c}f - f\hat{c} = \gamma f. \quad (\text{d})$$

By inserting  $f\hat{c} = (-\hat{c} + \gamma)f$ , derived from this equation, into the second term of the middle of Eq. (H.26), the right-hand side is derived.

From the right-hand side of this equation, it is confirmed that an infinite number of quasi-particles cohere in the coherent state.

The coherent state has several features, which are expressed as

$$\hat{c}|\gamma\rangle = \gamma|\gamma\rangle, \quad (\text{H.31})$$

$$\langle\gamma|\gamma\rangle = 1, \quad (\text{H.32})$$

$$\langle N\rangle = \langle\gamma|\hat{c}^\dagger\hat{c}|\gamma\rangle = \gamma^2, \quad (\text{H.33})$$

$$\Delta N = \sqrt{\langle N^2\rangle - \langle N\rangle^2} = \sqrt{\langle N\rangle} = |\gamma|. \quad (\text{H.34})$$

Equation (H.31) means that the coherent state is an eigenstate of the annihilation operator, which can be confirmed by applying the operators on the both sides of Eq. (d) in the footnote <sup>(1)</sup> to the vacuum state  $|0\rangle$ . Furthermore, since the coherent state is not the eigenstate of the number operator  $\hat{N}$  ( $= \hat{c}^\dagger\hat{c}$ ), the standard deviation of the quasi-particle number is nonzero, as is given by Eq. (H.34); i.e., the number of the quasi-particles fluctuates.

## H.3 Temporal Evolution of the Coherent State

### H.3.1 Probability of Exciting the Phonon Field

In order to analyze the temporal evolution of the coherent state, the initial condition is assumed to be  $|\psi\rangle = \tilde{a}_i^\dagger|0\rangle$ , which means that the DP is generated at site  $i$  in the probe apex by injecting the propagating light at time  $t = 0$ . Since this initial state is not the eigenstate of the Hamiltonian  $\hat{H}'$  of Eq. (H.9), phonons are excited by the DP–phonon interaction. For deriving the probability of this excitation, the creation operator  $\tilde{a}_i^\dagger$  for the DP is expressed by the operator  $\hat{\alpha}_i^\dagger$  for the DPP as

$$\tilde{a}_i^\dagger = \hat{\alpha}_i^\dagger \exp \left\{ \sum_{p=1}^N \gamma_{ip} \left( \hat{\beta}_p^\dagger - \hat{\beta}_p \right) \right\} \quad (\text{H.35a})$$

and therefore

$$|\psi\rangle = \tilde{a}_i^\dagger|0\rangle \equiv \hat{\alpha}_i^\dagger|\gamma\rangle, \quad (\text{H.35b})$$

where Eq. (H.18) and the relation  $\hat{c}_p - \hat{c}_p^\dagger = \hat{\beta}_p - \hat{\beta}_p^\dagger$  were used. In Eq. (H.35a),  $\gamma_{ip}$  is equal to  $\chi_{ip}/\Omega_p$ . The probability,  $P'$ , that the phonons are still in the vacuum state at time  $t$  is given by

$$P' = \left| \langle \psi | e^{-i\frac{\hat{H}'}{\hbar}t} | \psi \rangle \right|^2. \quad (\text{H.36})$$

It should be noted that the Hamiltonian  $\hat{H}'$  on the right-hand side is given by Eq. (H.9), which is the one excluding the fourth term (hopping term) from the Hamiltonian of Eq. (4.44) in Chap. 4. Therefore, by applying the mean field approximation of Eq.(4.45) to the third term of Eq. (4.44), one derives

$$\hat{H}' = \sum_{i=1}^N \hbar\omega\hat{\alpha}_i^\dagger\hat{\alpha}_i + \sum_{p=1}^N \hbar\Omega_p\hat{\beta}_p^\dagger\hat{\beta}_p + \sum_{i=1}^N \sum_{j=1}^N \frac{\hbar\chi}{2}\langle x_j \rangle_i \frac{1}{N}\hat{\alpha}_i^\dagger\hat{\alpha}_i. \quad (\text{H.37})$$

Inserting this into Eq. (H.36) gives the relation

$$\begin{aligned} & \langle \psi | e^{-i\frac{\hat{H}'}{\hbar}t} | \psi \rangle \\ &= \langle \gamma | \hat{\alpha}_i \exp \left( -i \sum_{i=1}^N \omega\hat{\alpha}_i^\dagger\hat{\alpha}_i t - i \sum_{i=1}^N \sum_{j=1}^N \frac{\chi}{2}\langle x_j \rangle_i \frac{1}{N}\hat{\alpha}_i^\dagger\hat{\alpha}_i t \right) \\ & \quad \times \exp \left( -i \sum_{p=1}^N \Omega_p\hat{\beta}_p^\dagger\hat{\beta}_p t \right) \hat{\alpha}_i^\dagger | \gamma \rangle \\ &= \langle \gamma | \hat{\alpha}_i \exp \left\{ -i \sum_{i=1}^N \left( \omega\hat{\alpha}_i^\dagger\hat{\alpha}_i t + \sum_{j=1}^N \frac{\chi}{2}\langle x_j \rangle_i \frac{1}{N}\hat{\alpha}_i^\dagger\hat{\alpha}_i t \right) \right\} \\ & \quad \times \hat{\alpha}_i^\dagger \exp \left( -i \sum_{p=1}^N \Omega_p\hat{\beta}_p^\dagger\hat{\beta}_p t \right) | \gamma \rangle. \end{aligned} \quad (\text{H.38a})$$

In order to transform

$$\exp \left\{ -i \sum_{i=1}^N \left( \omega\hat{\alpha}_i^\dagger\hat{\alpha}_i t + \sum_{j=1}^N \frac{\chi}{2}\langle x_j \rangle_i \frac{1}{N}\hat{\alpha}_i^\dagger\hat{\alpha}_i t \right) \right\} \hat{\alpha}_i^\dagger | \gamma \rangle \quad (\text{H.38b})$$

in the fourth row of Eq.(H.38a), a parameter  $\kappa$  is defined by

$$\kappa \equiv -i \left( \omega + \sum_{j=1}^N \frac{\chi}{2}\langle x_j \rangle_i \frac{1}{N} \right) t \equiv -i (\omega + \chi\langle x_i \rangle_i) t. \quad (\text{H.39})$$

Using this parameter, Eq. (H.38b) is transformed to

$$\begin{aligned} & \exp \left\{ -i \sum_{i=1}^N \left( \omega\hat{\alpha}_i^\dagger\hat{\alpha}_i t + \sum_{j=1}^N \frac{\chi}{2}\langle x_j \rangle_i \frac{1}{N}\hat{\alpha}_i^\dagger\hat{\alpha}_i t \right) \right\} \hat{\alpha}_i^\dagger | \gamma \rangle \\ &= \exp \left( \sum_{i=1}^N \kappa\hat{\alpha}_i^\dagger\hat{\alpha}_i t \right) \hat{\alpha}_i^\dagger | \gamma \rangle \end{aligned} \quad (\text{H.40})$$

$$\begin{aligned}
&= \left\{ 1 + \sum_{i=1}^N \kappa \hat{\alpha}_i^\dagger \hat{\alpha}_i t + \frac{1}{2!} \sum_{i=1}^N \left( \kappa \hat{\alpha}_i^\dagger \hat{\alpha}_i t \right)^2 + \dots \right\} \hat{\alpha}_i^\dagger |\gamma\rangle \\
&= \hat{\alpha}_i^\dagger \left\{ 1 + \kappa t + \frac{(\kappa t)^2}{2!} + \dots \right\} |\gamma\rangle = \hat{\alpha}_i^\dagger e^{\kappa t} |\gamma\rangle,
\end{aligned}$$

where the relations

$$\hat{\alpha}_i |\gamma\rangle = 0, \quad \langle \gamma | \hat{\alpha}_i^\dagger = 0, \quad \hat{\alpha}_i \hat{\alpha}_j^\dagger = \delta_{ij} + \hat{\alpha}_i^\dagger \hat{\alpha}_i \quad (\text{H.41})$$

were used for deriving the third row from the second row. By using Eq. (H.40), Eq. (H.38a) is transformed to

$$\begin{aligned}
&\langle \psi | e^{-i\frac{\hat{H}'}{\hbar}t} | \psi \rangle \quad (\text{H.42}) \\
&= \langle \gamma | \hat{\alpha}_i \hat{\alpha}_i^\dagger \exp \{ -i(\omega + \chi \langle x_i \rangle_i) t \} \exp \left( -i \sum_{p=1}^N \Omega_p \hat{\beta}_p^\dagger \hat{\beta}_p t \right) | \gamma \rangle \\
&= \langle \gamma | \left( 1 - \hat{\alpha}_i^\dagger \hat{\alpha}_i \right) \exp \left( -i \sum_{p=1}^N \Omega_p \hat{\beta}_p^\dagger \hat{\beta}_p t \right) | \gamma \rangle \\
&= \exp \{ -i(\omega + \chi \langle x_i \rangle_i) t \} \langle \gamma | \exp \left( -i \sum_{p=1}^N \Omega_p \hat{\beta}_p^\dagger \hat{\beta}_p t \right) | \gamma \rangle.
\end{aligned}$$

When inserting this equation into Eq. (H.36), the exponential function at the head of the fourth row is neglected because its absolute value is unity. By rewriting other time-evolving terms as

$$f = \langle \gamma | \exp \left( -i \sum_{p=1}^N \Omega_p \hat{\beta}_p^\dagger \hat{\beta}_p t \right) | \gamma \rangle, \quad (\text{H.43})$$

it represents the effect of phonon excitation triggered by the fluctuation of the number of quasi-particles. By differentiating it with respect to  $\gamma_{ip}$  ( $= \chi_{ip}/\Omega_p$ ), one obtains<sup>4</sup>

$$\frac{\partial f}{\partial \gamma_{ip}} = 2 \left( e^{-i\Omega_p t} - 1 \right) \gamma_{ip} f. \quad (\text{H.44})$$

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<sup>4</sup> Since

$$|\gamma\rangle = \exp \sum_p^N \gamma_{ip} \left( \hat{\beta}_p^\dagger - \hat{\beta}_p \right) |0\rangle \quad (\text{a})$$

holds by inserting  $\hat{c}_p^\dagger - \hat{c}_p = \hat{\beta}_p^\dagger - \hat{\beta}_p$  into Eq. (H.24), inserting it into Eq. (H.43) derives

By integrating it with respect to  $\gamma_{ip}$  and summing up over  $p = 1 - N$ , the expression

(Footnote 4 continued)

$$f = \langle 0 | \exp \left\{ - \sum_{q=1}^N \gamma_{iq} \left( \hat{\beta}_q^\dagger - \hat{\beta}_q \right) \right\} \times \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) \exp \left\{ \sum_{q=1}^N \gamma_{iq} \left( \hat{\beta}_q^\dagger - \hat{\beta}_q \right) \right\} | 0 \rangle. \quad (\text{b})$$

Here, the subscript  $p$  is replaced with  $q$  in order to avoid confusion in the following discussions. Differentiating Eq. (b) with respect to the coefficient  $\gamma_{ip}$  of the  $p$ -th term yields

$$\begin{aligned} & \frac{\partial f}{\partial \gamma_{ip}} \\ &= - \langle 0 | \exp \left\{ - \sum_{q=1}^N \gamma_{iq} \left( \hat{\beta}_q^\dagger - \hat{\beta}_q \right) \right\} \left( \hat{\beta}_p^\dagger - \hat{\beta}_p \right) \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) \\ & \times \exp \left\{ \sum_{q=1}^N \gamma_{iq} \left( \hat{\beta}_q^\dagger - \hat{\beta}_q \right) \right\} | 0 \rangle \\ &+ \langle 0 | \exp \left\{ - \sum_{q=1}^N \gamma_{iq} \left( \hat{\beta}_q^\dagger - \hat{\beta}_q \right) \right\} \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) \left( \hat{\beta}_p^\dagger - \hat{\beta}_p \right) \\ & \times \exp \left\{ \sum_{q=1}^N \gamma_{iq} \left( \hat{\beta}_q^\dagger - \hat{\beta}_q \right) \right\} | 0 \rangle \\ &= - \langle \gamma | \left( \hat{\beta}_p^\dagger - \hat{\beta}_p \right) \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) | \gamma \rangle \\ &+ \langle \gamma | \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) \left( \hat{\beta}_p^\dagger - \hat{\beta}_p \right) | \gamma \rangle. \end{aligned} \quad (\text{c})$$

Here, by noting Eq. (4.38), it is found that Eq. (H.31) is effective even when  $\hat{c}$  is replaced with  $\hat{\beta}_p$ , and resultantly, the relations

$$\hat{\beta}_p | \gamma \rangle = \gamma_{ip} | \gamma \rangle, \quad (\text{d})$$

$$\langle \gamma | \hat{\beta}_p^\dagger = \langle \gamma | \gamma_{ip} \quad (\text{e})$$

hold. By using them, in the first term in the fifth row of Eq. (c), the component having  $\hat{\beta}_p^\dagger$  on the left of the exponential function is transformed to

$$\langle \gamma | \hat{\beta}_p^\dagger \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) | \gamma \rangle = \langle \gamma | \gamma_{ip} \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) | \gamma \rangle = \gamma_{ip} f. \quad (\text{f})$$

In the second term in the seventh row, the component having  $\hat{\beta}_p$  on the right of the exponential function is transformed to

$$\langle \gamma | \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) \hat{\beta}_p | \gamma \rangle = \langle \gamma | \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) \gamma_{ip} | \gamma \rangle = \gamma_{ip} f. \quad (\text{g})$$

Next, in order to transform  $\langle \gamma | \hat{\beta}_p \exp \left( -i \sum_{q=1}^N \Omega_{qt} \hat{\beta}_q^\dagger \hat{\beta}_q \right) | \gamma \rangle$  in the second term of the fifth row, it is noted that the following relation holds:

$$f = \exp \left\{ \sum_{p=1}^N \gamma_{ip}^2 \left( e^{-i\Omega_p t} - 1 \right) \right\} \quad (\text{H.45})$$

is derived. By inserting this into Eq. (H.36), the probability of phonon excitation is expressed as

$$P = 1 - P' = 1 - |f|^2 = 1 - \exp \left\{ \sum_{p=1}^N 2\gamma_{ip}^2 (\cos \Omega_p t - 1) \right\}. \quad (\text{H.46})$$

Furthermore, the probability of exciting the specific mode  $p_0$  of the phonon while other modes are in the vacuum state is given by

$$P_{p_0} = \left[ 1 - \exp \left\{ 2\gamma_{ip_0}^2 (\cos \Omega_{p_0} t - 1) \right\} \right] \exp \left\{ \sum_{p \neq p_0}^N 2\gamma_{ip}^2 (\cos \Omega_p t - 1) \right\}. \quad (\text{H.47})$$

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(Footnote 4 continued)

$$\hat{\beta}_p \exp \left( -i\Omega_p t \hat{\beta}_p^\dagger \hat{\beta}_p \right) = \exp \left( -i\Omega_p t \hat{\beta}_p^\dagger \hat{\beta}_p \right) \hat{\beta}_p e^{-i\Omega_p t} \quad (\text{h})$$

because  $[\hat{\beta}_p, \hat{\beta}_p^\dagger] = 1$ . Using Eq. (h), one derives

$$\begin{aligned} \hat{\beta}_p \exp \left( -i \sum_{q=1}^N \Omega_q t \hat{\beta}_q^\dagger \hat{\beta}_q \right) &= \exp \left( -i\Omega_p t \hat{\beta}_p^\dagger \hat{\beta}_p \right) \hat{\beta}_p e^{-i\Omega_p t} \exp \left( -i \sum_{q \neq p}^N \Omega_q t \hat{\beta}_q^\dagger \hat{\beta}_q \right) \\ &= \exp \left( -i \sum_{q=1}^N \Omega_q t \hat{\beta}_q^\dagger \hat{\beta}_q \right) \hat{\beta}_p \gamma_{ip} e^{-i\Omega_p t}, \end{aligned} \quad (\text{i})$$

and thus, the relation

$$\begin{aligned} \langle \gamma | \hat{\beta}_p \exp \left( -i \sum_{q=1}^N \Omega_q t \hat{\beta}_q^\dagger \hat{\beta}_q \right) | \gamma \rangle &= \langle \gamma | \exp \left( -i \sum_{q=1}^N \Omega_q t \hat{\beta}_q^\dagger \hat{\beta}_q \right) \hat{\beta}_p e^{-i\Omega_p t} | \gamma \rangle \\ &= \langle \gamma | \exp \left( -i \sum_{q=1}^N \Omega_q t \hat{\beta}_q^\dagger \hat{\beta}_q \right) \gamma_{ip} e^{-i\Omega_p t} | \gamma \rangle = \gamma_{ip} e^{-i\Omega_p t} f \end{aligned} \quad (\text{j})$$

is obtained, where Eq. (d) was used to replace the first row with the second row. The component  $\langle \gamma | \exp \left( -i \sum_{q=1}^N \Omega_q t \hat{\beta}_q^\dagger \hat{\beta}_q \right) \hat{\beta}_p^\dagger | \gamma \rangle$  in the first term in the seventh row of Eq. (c) is also similarly transformed to

$$\langle \gamma | \exp \left( -i \sum_{q=1}^N \Omega_q t \hat{\beta}_q^\dagger \hat{\beta}_q \right) \hat{\beta}_p^\dagger | \gamma \rangle = \gamma_{ip} e^{-i\Omega_p t} f. \quad (\text{k})$$

Finally, Eq. (H.44) is derived by inserting Eqs. (f)–(k) into the fifth and seventh rows of Eq. (c).



### H.3.2 Fluctuations in the Number of Phonons

This section derives the magnitude of the fluctuations in the number of phonons when the phonons are in the coherent state. Since the value of the fluctuations is equal to the square root of the expectation value, as was given by Eq. (H.34), the expectation value of the number of phonons is derived in the following.

By using Eqs. (H.18) and (H.21), the phonon number operator  $\hat{N}_p$  of mode  $p$  can be expressed in terms of the unitary-transformed operators as

$$\begin{aligned}\hat{N}_p &= \hat{c}_p^\dagger \hat{c}_p \\ &= \hat{\beta}_p^\dagger \hat{\beta}_p + \sum_{i=1}^N \sum_{j=1}^N \gamma_{ip} \gamma_{jp} \hat{\alpha}_i^\dagger \hat{\alpha}_i \hat{\alpha}_j^\dagger \hat{\alpha}_j - \sum_{i=1}^N \gamma_{jp} \hat{\alpha}_i^\dagger \hat{\alpha}_i \left( \hat{\beta}_p + \hat{\beta}_p^\dagger \right).\end{aligned}\quad (\text{H.48})$$

Since time evolution of the number operator is expressed in Heisenberg representation as

$$\begin{aligned}\hat{N}_p(t) &= e^{i\frac{\hat{H}'}{\hbar}t} \hat{N}_p e^{-i\frac{\hat{H}'}{\hbar}t} \\ &= \hat{\beta}_p^\dagger \hat{\beta}_p + \sum_{i=1}^N \sum_{j=1}^N \gamma_{ip} \gamma_{jp} \hat{\alpha}_i^\dagger \hat{\alpha}_i \hat{\alpha}_j^\dagger \hat{\alpha}_j - \sum_{i=1}^N \gamma_{jp} \hat{\alpha}_i^\dagger \hat{\alpha}_i \left( e^{-i\Omega_p t} \hat{\beta}_p + e^{i\Omega_p t} \hat{\beta}_p^\dagger \right),\end{aligned}\quad (\text{H.49})$$

the expectation value of the phonon number is derived as

$$\langle N_p(t) \rangle = \langle \psi | \hat{N}_p(t) | \gamma \rangle = 2\gamma_{ip} (1 - \cos \Omega_p t). \quad (\text{H.50})$$

From Eq. (H.34), it is concluded that the value of the phonon number fluctuations is given by the square root of Eq. (H.50).

### H.3.3 Eigenvalues for the One-Dimensional Lattice Without any Impurities

Since the equation of motion for the one-dimensional lattice without any impurities has been given by Eq. (4.3) of Chap. 4, this section derives its eigenvalue. For this derivation, the triple diagonal matrix is diagonalized, and its eigenvalues and eigenvectors are derived. For these purposes, the  $N$ -dimensional triple diagonal matrix

$$C = \begin{pmatrix} A & B & & \\ B & A & \cdot & \\ & \cdot & \cdot & B \\ & & & B & A \end{pmatrix} \quad (\text{H.51})$$

is considered, where  $A$  and  $B$  are constants. In order to calculate its determinant, the determinant of the  $n$ -dimensional triple diagonal matrix is written by  $f_n$  ( $1 \leq n \leq N$ ), and its recursion relations are derived by cofactor expansion:

$$f_n - Af_{n-1} + B^2f_{n-2} = 0, \quad (\text{H.52})$$

$$f_1 = A = \alpha + \beta, f_2 = A^2 - B^2 = \alpha^2 + \alpha\beta + \beta^2, \quad (\text{H.53})$$

$$\alpha + \beta = A, \alpha + \beta = A. \quad (\text{H.54})$$

From these relations, one obtains

$$f_n = \frac{1}{\beta - \alpha} (\beta^{n+1} - \alpha^{n+1}). \quad (\text{H.55})$$

In order to derive the characteristic equation for the matrix  $C$  of Eq. (H.51),  $A$  is replaced with  $A - x$  and we set  $f_N = 0$ , where  $x$  is the eigenvalue of the matrix  $C$ . That is, by solving

$$f_N = 0, \quad (\text{H.56})$$

$$\alpha^{N+1} = \beta^{N+1}, \alpha = \beta \exp\left(2\pi i \frac{n}{N+1}\right) (1 \leq n \leq N), \quad (\text{H.57})$$

and Eq. (H.52) simultaneously with respect to  $A - x$ , the eigenvalue  $x$  is obtained:

$$x_n = A + 2B \cos\left(\frac{n}{N+1}\pi\right). \quad (\text{H.58})$$

Since the eigenvector  $\mathbf{p}_n$  satisfies

$$\begin{pmatrix} A - x_n & B & & \\ B & A - x_n & B & \\ & & \cdot & \cdot \\ & & & B & A - x_n \end{pmatrix} \begin{pmatrix} p_{1n} \\ p_{2n} \\ \cdot \\ p_{Nn} \end{pmatrix} = 0, \quad (\text{H.59})$$

the recursion relations for the elements of the eigenvector are

$$p_{k,n} - 2 \cos\left(\frac{n}{N+1}\pi\right) p_{k-1,n} + p_{k-2,n} = 0, \quad (\text{H.60})$$

$$p_{2,n} = 2 \cos\left(\frac{n}{N+1}\pi\right) p_{1,n}. \quad (\text{H.61})$$

Solving Eqs. (H.60) and (H.61) simultaneously and imposing the normalization condition

$$\sum_{k=1}^N p_{k,n}^2 = 1, \quad (\text{H.62})$$

gives

$$p_{k,n} = \sqrt{\frac{2}{N+1}} \sin\left(\frac{kn}{N+1}\pi\right) \quad (1 \leq k \leq N). \quad (\text{H.63})$$

From these results, it is found that the eigenvectors are independent of the constants  $A$  and  $B$  in the matrix  $C$ . Furthermore, the matrix  $P$ , composed by arranging the eigenvectors in line, is confirmed to be an orthonormal matrix ( $P^T = P^{-1}$ ).

By inserting  $A = 2$  and  $B = -1$  into Eq. (H.51), the eigenvalue of Eq. (4.3) in Chap. 4, i.e., the square of the eigen angular frequency, is given by

$$\Omega_p^2 = \frac{k}{m} \left\{ 2 - 2 \cos\left(\frac{p}{N+1}\pi\right) \right\} = 4 \frac{k}{m} \sin^2 \left[ \frac{p}{2(N+1)}\pi \right], \quad (\text{H.64})$$

whose square root corresponds to Eq. (4.21) in Chap. 4.

## H.4 Diagonalization of the Hamiltonian Without DP–Phonon Coupling

In the case where the DP–phonon coupling constant  $\chi$  is zero, the Hamiltonian can be written as

$$\hat{H} = \hbar \tilde{\mathbf{a}}^\dagger \begin{pmatrix} \omega & J & & \\ J & \omega & \cdot & \\ & \cdot & \cdot & J \\ & & J & \omega \end{pmatrix} \tilde{\mathbf{a}}, \quad (\text{H.65})$$

$$\tilde{\mathbf{a}} = \begin{pmatrix} \tilde{a}_1 \\ \cdot \\ \cdot \\ \tilde{a}_N \end{pmatrix}, \quad (\text{H.66})$$

and its eigenenergy is

$$\hbar\Omega_r = \hbar\omega + 2\hbar J \cos\left(\frac{r}{N+1}\pi\right). \quad (\text{H.67})$$

On the other hand, the expectation value of the number of DPs, given by Eq. (4.56) in Chap. 4, is rewritten as

$$\begin{aligned}
\langle N_i(t) \rangle_j &= \langle \psi_j | \hat{N}_i(t) | \psi_j \rangle = \sum_{r=1}^N \sum_{s=1}^N Q_{ir} Q_{jr} Q_{is} Q_{js} \cos \{ (\Omega_r - \Omega_s) t \} \\
&\times \left( \frac{2}{N+1} \right)^2 \sum_{r=1}^N \sum_{s=1}^N \sin \left( \frac{jr}{N+1} \pi \right) \sin \left( \frac{ir}{N+1} \pi \right) \sin \left( \frac{js}{N+1} \pi \right) \sin \left( \frac{is}{N+1} \pi \right) \\
&\times \cos \left\{ 2Jt \cos \left( \frac{r}{N+1} \pi \right) - 2Jt \cos \left( \frac{s}{N+1} \pi \right) \right\} \\
&= \frac{1}{(N+1)^2} \left[ \sum_{r=1}^N \left\{ \cos \left( \frac{nr}{N+1} \pi \right) - \cos \left( \frac{mr}{N+1} \pi \right) \right\} \cos \left\{ 2Jt \cos \left( \frac{r}{N+1} \pi \right) \right\} \right]^2 \\
&+ \frac{1}{(N+1)^2} \left[ \sum_{r=1}^N \left\{ \cos \left( \frac{nr}{N+1} \pi \right) - \cos \left( \frac{mr}{N+1} \pi \right) \right\} \sin \left\{ 2Jt \cos \left( \frac{r}{N+1} \pi \right) \right\} \right]^2,
\end{aligned} \tag{H.68}$$

where the notations  $m = j + i$  and  $n = j - i$  were used.

Next, in order to grasp the behavior in the limit  $N \rightarrow \infty$ , by using  $\theta_r = r\pi/(N+1)$  and replacing the summation in Eq. (H.68) with an integral, one obtains

$$\begin{aligned}
\langle N_i(t) \rangle_j &= \left\{ \frac{1}{\pi} \int_0^\pi (\cos n\theta - \cos m\theta) \cos (2Jt \cos \theta) d\theta \right\}^2 \\
&+ \left\{ \frac{1}{\pi} \int_0^\pi (\cos n\theta - \cos m\theta) \sin (2Jt \cos \theta) d\theta \right\}^2.
\end{aligned} \tag{H.69}$$

By using the integral representation of the Bessel function  $J_n(z)$  of the first kind

$$\begin{aligned}
J_n(z) &= \frac{1}{\pi i^n} \int_0^\pi e^{iz \cos \theta} \cos n\theta d\theta \\
&= \frac{1}{\pi i^n} \int_0^\pi \cos n\theta \{ \cos (z \cos \theta) + i \sin (z \cos \theta) \} d\theta,
\end{aligned} \tag{H.70}$$

the expectation value in the limit  $N \rightarrow \infty$  is obtained from Eq. (H.69) and is expressed as

$$\langle N_i(t) \rangle_j = \left\{ J_{j-i}(2Jt) - (-1)^i J_{j+i}(2Jt) \right\}^2, \tag{H.71}$$

which corresponds to Eq. (4.57) in Chap. 4.

## H.5 Expectation Value of the Displacement of Atoms

The expectation value of the displacement of atoms, as a result of DP-phonon coupling, has been presented by Eq. (4.43) in Chap. 4, i.e.,

$$\langle \hat{x}_j \rangle_i = - \sum_{p=1}^N \frac{\hbar \chi P_{ip} P_{jp}}{\sqrt{m_i m_j} \Omega_p^2}. \quad (\text{H.72})$$

By using

$$\Lambda = P^{-1} A P = P^{-1} \sqrt{M}^{-1} \Gamma \sqrt{M}^{-1} P, \quad (\text{H.73a})$$

$$(\Lambda)_{pq} = \delta_{pq} \frac{\Omega_p^2}{k}, \quad (\text{H.73b})$$

of Eqs. (4.7) and (4.8), the inverse matrix of

$$\Gamma = \sqrt{M} P \Lambda P^{-1} \sqrt{M} \quad (\text{H.74a})$$

is derived as

$$\Gamma^{-1} = \sqrt{M}^{-1} P \Lambda^{-1} P^{-1} \sqrt{M}^{-1}, \quad (\text{H.74b})$$

$$\left( \Gamma^{-1} \right)_{ij} = \frac{k}{\sqrt{m_i m_j}} \sum_{p=1}^N \frac{P_{ip} P_{jp}}{\Omega_p^2}. \quad (\text{H.74c})$$

Then, the expectation value of the displacement can be rewritten by using these equations as

$$\langle \hat{x}_j \rangle_i = - \frac{\hbar \chi}{k} (\Gamma^{-1})_{ij} \quad (\text{H.75})$$

Since the values of the diagonal elements of the matrix  $\Gamma$  are 2 and those of the adjacent off-diagonal elements are  $-1$ , as given by Eq. (4.4) in Chap. 4, it is confirmed that Eq. (H.75) is independent of the effect of impurity atoms.

By denoting the matrix used for diagonalizing  $\Gamma$  by  $R$  and the diagonalized matrix by  $W$ , the inverse matrix  $\Gamma^{-1}$  can be readily expressed as

$$R^{-1} \Gamma R = W \quad (\text{H.76a})$$

$$\left( \Gamma^{-1} \right)_{ij} = \sum_{n=1}^N R_{in} W_n^{-1} R_{nj}^{-1} = \frac{1}{N+1} \sum_{n=1}^N \frac{\sin\left(\frac{in}{N+1}\pi\right) \sin\left(\frac{jn}{N+1}\pi\right)}{1 - \cos\left(\frac{n}{N+1}\pi\right)} \quad (\text{H.76b})$$

which corresponds to Eq. (4.58) in Chap. 4.

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1. Y. Tanaka, *Theoretical Models of Optical Near Fields Interacting with Localized Phonons* (Master's Thesis, Tokyo Institute Technology, 2007)

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