

QED of excitons with microscopic nonlocality in arbitrary-structured dielectrics

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We have constructed complete quantum theory for an optical process of excitons with microscopic nonlocality (nonlocal susceptibility) originating from their center-of-mass motion. This theory provides a practical calculation method for arbitrary-structured nano-to-macro scale dielectrics where excitons are weakly confined. We obtain good correspondences with underlying theories, semiclassical microscopic nonlocal theory, and QED theories for dispersive and absorptive materials with local susceptibility.

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I. INTRODUCTION

In the conventional theories of optical processes in condensed matters, light has been mainly treated classically regardless of whether the matter systems are described in quantum mechanical terms (semiclassical theory) or classical ones. These theories have successfully explained a variety of optical phenomena for the classical light or the coherent states of photons. However, there is growing interest in the quantum electrodynamics (QED) of elementary excitations in condensed matters in order to discuss optical processes for nonclassical light such as entangled states, single photons, squeezed states, cavity photons, and so on. The relevant experiments have already been reported, for example, the entangled-photon generation via biexcitons (excitonic molecules),¹ triggered single photon generation from bound excitons in a semiconductor,² and the squeezing of cavity polaritons in semiconductor microcavities.³ The quantization of a radiation field has been studied for a long time not only in a vacuum⁴ but also in the medium characterized by a frequency-independent dielectric constant. On the other hand, Hopfield has systematically discussed the eigenstates of exciton-photon systems or exciton-polaritons,⁵ which have a frequency dependence in a susceptibility $\chi(\omega)$ or in a dielectric function $\epsilon(\omega) = 1 + \chi(\omega)$ as seen from their dispersion relation $\omega^2\epsilon(\omega) = c^2k^2$. Although in his treatment, $\chi(\omega)$ included only a real part, susceptibility is generally represented as a complex function satisfying the Kramers-Kronig relations. In addition, its imaginary part, causing damping effects, cannot be neglected in the discussion of resonant optical processes of elementary excitations in condensed matters.

The quantization of the electromagnetic fields in such dispersive and absorptive dielectrics has been systematically carried out for homogeneous media by Huttner and Barnett (HB),⁶ and for inhomogeneous 3D ones by Suttrop and Wubs (SW).⁷ In the former scheme, dispersive dielectrics are described using the Hopfield polariton model,⁵ i.e., polarizable harmonic oscillators in-

teracting with a radiation field, and absorption is considered using a reservoir of oscillators interacting with the polarizable ones. The electromagnetic fields are described in terms of the eigen operators derived from the diagonalization of a Hamiltonian. In the expression of those fields, there exists a complex dielectric function $\epsilon(\omega)$ represented by system parameters with satisfying the Kramers-Kronig relations. All the quantum mechanical properties of the electromagnetic fields are characterized by this dielectric function. The pioneering work of HB stimulated various theoretical studies associated with the QED of dispersive and absorptive dielectrics, for example, the spontaneous decay,^{8,9} input-output relations,^{10,11,12} and quantization in amplifying, anisotropic, magnetic, or nonlinear media.¹³ On the other hand, SW have carried out the quantization of the electromagnetic fields in arbitrary-structured 3D dielectrics by using the Laplace-transform technique,^{7,14} which is completely different from the quantization scheme of HB. Around the same time, the diagonalization of the Hamiltonian of SW has been performed by Suttrop and van Wonderen.¹⁵ In these schemes, complex dielectric function $\epsilon(\mathbf{r}, \omega)$ depends on spatial position \mathbf{r} of a medium and radiation frequency ω .

In the above QED theories and also in semiclassical ones, a dielectric function is usually treated as local form $\epsilon(\mathbf{r}, \omega)$ with respect to the spatial position. However, in general, the optical susceptibility has a nonlocal form as $\chi(\mathbf{r}, \mathbf{r}', \omega)$, which characterizes polarization $\mathbf{P}(\mathbf{r}, \omega)$ at position \mathbf{r} induced by electric field $\mathbf{E}(\mathbf{r}', \omega)$ at different position \mathbf{r}' as

$$\mathbf{P}(\mathbf{r}, \omega) = \epsilon_0 \int d\mathbf{r}' \chi(\mathbf{r}, \mathbf{r}', \omega) \mathbf{E}(\mathbf{r}', \omega). \quad (1)$$

This microscopic nonlocality originates from the spatial spreading of the wave function of elementary excitations or, particularly for excitons in semiconductors, their center-of-mass motion with a finite translational mass. Usually, such a nonlocality is not considered in the discussion of macroscopic materials; this is because the

coherence length of elementary excitations is generally much shorter than the spatial scale of materials. Therefore, only the averaged values of physical quantities over the coherence volume are reflected in observation, and the microscopic nonlocal effect is not apparent. However, in order to discuss the excitons in inhomogeneous media, we must suppose that the motion of excitons has considerably long coherence and microscopic nonlocality becomes important even for bulk materials, as explained below.

In the case of homogeneous media, nonlocal susceptibility depends only on the difference $\mathbf{r} - \mathbf{r}'$ of the two positions; then, Eq. (1) is rewritten in the reciprocal space as

$$\mathbf{P}(\mathbf{k}, \omega) = \epsilon_0 \chi(\mathbf{k}, \omega) \mathbf{E}(\mathbf{k}, \omega). \quad (2)$$

In this way, susceptibility $\chi(\mathbf{k}, \omega)$ has a wavevector dependence and the ω -dependence even for homogeneous media when it has a microscopic nonlocality. This \mathbf{k} -dependence gives more than one propagating or evanescent modes for a single frequency satisfying

$$\omega^2 \epsilon(\mathbf{k}, \omega) = c^2 |\mathbf{k}|^2. \quad (3)$$

Now, we consider a single exciton state with finite translational mass m_{ex} , transverse exciton energy E_{T} , and longitudinal one E_{L} at respective band edges. Since the transverse exciton energy is written as $E_{\text{ex}}(\mathbf{k}) = E_{\text{T}} + \hbar^2 |\mathbf{k}|^2 / 2m_{\text{ex}}$ for wavevector \mathbf{k} , we can find two propagating polariton modes for $\hbar\omega > E_{\text{L}}$, and one propagating mode at the polariton band gap $E_{\text{T}} < \hbar\omega < E_{\text{L}}$ in addition to an evanescent mode. These multiple polariton modes do not appear in the Hopfield polariton model⁵ because the excitons were assumed to have infinite translational mass. As pointed out by Pekar for the first time,¹⁶ it appears that additional boundary conditions (ABCs) should be introduced besides the Maxwell boundary conditions for the unique connection between the polariton modes inside a material and the external ones at the interface between two materials. This problem is known as the ABC problem; it arises when the translational symmetry of a system is broken due to surfaces or interfaces. Since Pekar's work, continued studies have revealed that this problem can be resolved by considering the microscopic boundary conditions of the excitonic center-of-mass motion at interfaces.^{17,18,19} Nowadays, in the semiclassical framework, a calculation method independent from the notation of ABCs is well known as an ABC-free theory²⁰ or a microscopic nonlocal theory.^{21,22} These theories systematically consider the nonlocality of susceptibility $\chi(\mathbf{r}, \mathbf{r}', \omega)$, and various linear and nonlinear phenomena in inhomogeneous materials have been discussed using them. In particular, for nano-structured materials, where the coherence of the center-of-mass motion of excitons is maintained in the entire material (weak confinement regime), anomalous size dependence of their optical processes has been elucidated. With regard to nanofilms, the nonlocal theory

has successfully explained their peculiar spectral structures originating from the polariton interference.^{19,23,24} Further, with recent development of fabrication technologies of nano samples, various peculiar effects due to long-range coherence are appearing through the interplay between the spatial structures of electromagnetic and excitonic waves, such as the resonant enhancement of a nonlinear response,²⁵ the interchange of quantized states due to giant radiative shift,²⁶ and the ultrafast radiative decay with femtosecond order.²⁷

From these theoretical and experimental results and great interest in the nonclassical states of elementary excitations^{1,2,3} as mentioned above, it is very attractive to discuss them in detail for the sake of applications to quantum information technologies. Although some studies introduced microscopic nonlocality into the QED of dispersive and absorptive dielectrics, there remains a problem of how to perform calculations in practical applications, as shown in Sec. III. The principal purpose of this paper is to construct a QED theory providing a practical calculation method for excitons weakly confined in arbitrary-structured 3D dielectrics considering radiative and nonradiative relaxations, which is necessary to discuss the effects of, for example, material interfaces, excitonic confinement in nano structures, and nonradiative relaxation processes. In this paper, we merge a microscopic nonlocal theory^{21,22} and the quantization technique of SW.⁷

We explain SW's QED theory for dielectrics with local susceptibility in Sec. II and the previously discussed QED theories with microscopic nonlocality in Sec. III. We show our Hamiltonian in Sec. IV, explain our QED theory in Sec. V, and discuss the nonradiative relaxation of excitons in Sec. VI. Last, we compare our theory with other QED ones with microscopic nonlocality in Sec. VII and summarize the discussion in Sec. VIII. We will explain only the outline of our theory in these sections and redundant calculations are shown in appendices. In App. A, the second quantization of excitonic polarization is discussed. In App. B, we derive the Hamiltonian from the microscopic point of view. In App. C, we extend the Maxwell wave equation discussed by SW so that we can consider the excitonic polarization. In App. D, we evaluate the commutation relations of ω -Fourier transformed Heisenberg operators. In App. E, we discuss the relationship between the retarded correlation functions of excitons and self-standing modes or exciton polariton modes. In App. F, we verify the equal-time commutation relations, which are expected from those of Schrödinger operators.

In this paper, we use MKS units and Coulomb gauge.

II. QED OF DIELECTRICS WITHOUT NONLOCALITY

In this section, we explain the outline of the discussion of SW,⁷ the QED theory without microscopic nonlocality.

Its Hamiltonian is shown in Eq. (B9) of the present paper. The quantization scheme is based on the motion equation of the electric field $\mathbf{E}(\mathbf{r}, t)$ whose definition in SW theory is

$$\mathbf{E}(\mathbf{r}, t) = -\frac{\partial}{\partial t}\mathbf{A}(\mathbf{r}, t) - \nabla\phi_{\text{bg}}(\mathbf{r}, t). \quad (4)$$

$\mathbf{A}(\mathbf{r}, t)$ is the vector potential and $\phi_{\text{bg}}(\mathbf{r}, t)$ is the Coulomb potential. We write the latter and the dielectric function $\epsilon_{\text{bg}}(\mathbf{r}, \omega)$ with a subscript bg for the description in the following sections. Since the Coulomb gauge is used in this scheme, the vector potential is a transverse field satisfying $\nabla \cdot \mathbf{A}(\mathbf{r}) = 0$, and the second term of Eq. (4) represents the longitudinal field. From the Laplace transform of the Heisenberg equations of the system variables, the Maxwell wave equation for the Fourier component of the electric field

$$\hat{\mathbf{E}}^{\pm}(\mathbf{r}, \omega) \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{\pm i\omega t} \mathbf{E}(\mathbf{r}, t) = \{\hat{\mathbf{E}}^{\mp}(\mathbf{r}, \omega)\}^{\dagger} \quad (5)$$

is derived as

$$\nabla \times \nabla \times \hat{\mathbf{E}}^{\pm}(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \hat{\mathbf{E}}^{\pm}(\mathbf{r}, \omega) = i\mu_0 \omega \hat{\mathbf{J}}(\mathbf{r}, \omega), \quad (6)$$

where $\hat{\mathbf{E}}^+(\mathbf{r}, \omega)$ and $\hat{\mathbf{E}}^-(\mathbf{r}, \omega)$ are called positive- and negative-frequency Fourier components of $\mathbf{E}(\mathbf{r}, t)$, respectively. We write the Fourier transformed operator with a hat ($\hat{}$) in this paper. Wave equation (6) has the same form as the one that appears in the classical electrodynamics except for operator $\hat{\mathbf{J}}(\mathbf{r}, \omega)$ on the right hand side. This operator is called the noise current density and interpreted as a source of the electromagnetic fields or the fluctuation caused by absorption. It plays an essential role in the series of QED theories for dispersive and absorptive dielectrics. The same kind of operator for homogeneous systems has been derived by HB,⁶ and the one for inhomogeneous 3D systems has been phenomenologically introduced in Ref. 28. On the other hand, from the Laplace-transformed motion equations of system variables, SW have systematically derived the representation of $\hat{\mathbf{J}}(\mathbf{r}, \omega)$, which is written in terms of the canonical variables and momenta of the system at $t = 0$ (we have performed a similar calculation in App. C). From the commutation relations between them, those for $\hat{\mathbf{J}}(\mathbf{r}, \omega)$ have been derived as

$$\begin{aligned} & [\hat{\mathbf{J}}(\mathbf{r}, \omega), \{\hat{\mathbf{J}}(\mathbf{r}', \omega')\}^{\dagger}] \\ &= \delta(\omega - \omega') \delta(\mathbf{r} - \mathbf{r}') \frac{\epsilon_0 \hbar \omega^2}{\pi} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \mathbf{1}, \end{aligned} \quad (7a)$$

$$[\hat{\mathbf{J}}(\mathbf{r}, \omega), \hat{\mathbf{J}}(\mathbf{r}', \omega')] = \mathbf{0}, \quad (7b)$$

where $[\hat{\mathbf{J}}, \hat{\mathbf{J}}^{\dagger}]$ is a 3×3 tensor and its (ξ, ξ') element implies $[\hat{J}_{\xi}, \hat{J}_{\xi'}^{\dagger}]$ for $\xi = x, y, z$ (the same kind of calculation is shown in App. D of the present paper).

Here, using Green's tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ satisfying

$$\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \mathbf{1}, \quad (8)$$

we can rewrite Maxwell wave equation (6) as

$$\hat{\mathbf{E}}^{\pm}(\mathbf{r}, \omega) = i\mu_0 \omega \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{J}}(\mathbf{r}', \omega). \quad (9)$$

From the commutation relations (7) of $\hat{\mathbf{J}}(\mathbf{r}, \omega)$, those of $\hat{\mathbf{E}}^{\pm}(\mathbf{r}, \omega)$ can be derived as

$$[\hat{\mathbf{E}}^+(\mathbf{r}, \omega), \hat{\mathbf{E}}^-(\mathbf{r}', \omega')] = \delta(\omega - \omega') \frac{\mu_0 \hbar \omega^2}{\pi} \text{Im}[\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)], \quad (10a)$$

$$[\hat{\mathbf{E}}^+(\mathbf{r}, \omega), \hat{\mathbf{E}}^+(\mathbf{r}', \omega')] = \mathbf{0}, \quad (10b)$$

where we use the equivalence shown in Eq. (1.54) of Ref. 13:

$$\begin{aligned} & \int d\mathbf{s} \frac{\omega^2}{c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{s}, \omega)] \mathbf{G}(\mathbf{r}, \mathbf{s}, \omega) \cdot \mathbf{G}^*(\mathbf{s}, \mathbf{r}', \omega) \\ &= \text{Im}[\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)] \end{aligned} \quad (11)$$

and the reciprocity relation:

$$\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \{\mathbf{G}(\mathbf{r}', \mathbf{r}, \omega)\}^{\dagger}. \quad (12)$$

Eqs. (10) can be understood by the fact that the Green's tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ for the Maxwell wave equation (6) identifies with the Fourier transform of the retarded correlation function of the electric field except for the factor $-\mu_0 \hbar \omega^2$, as discussed in Ref. 29:

$$\begin{aligned} & -\mu_0 \hbar \omega^2 \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \\ &= \int_{-\infty}^{\infty} dt e^{i\omega(t-t')} (-i)\theta(t-t') \langle [\mathbf{E}(\mathbf{r}, t), \mathbf{E}(\mathbf{r}', t')] \rangle, \end{aligned} \quad (13)$$

where $\theta(t)$ is the Heaviside step function. In this framework, all we have to do is find Green's tensor satisfying Eq. (8) in order to discuss the quantum mechanical properties of the electromagnetic fields in dielectrics. All the information of the material structure is included in dielectric function $\epsilon_{\text{bg}}(\mathbf{r}, \omega)$. The form of Green's tensor has already been known for various structures with high symmetry³⁰ and also can be numerically calculated for arbitrary 3D structures.³¹

III. PREVIOUS QED THEORIES WITH MICROSCOPIC NONLOCALITY

The above theory enables us to discuss the linear optical process in arbitrary-structured 3D dielectrics characterized by dielectric function $\epsilon_{\text{bg}}(\mathbf{r}, \omega)$. However, in order to discuss the materials with microscopic nonlocality, it is necessary to consider more general elementary excitations that cannot be described by harmonic oscillators of the Hopfield model.

As a pioneering study on a full-quantum theory with microscopic nonlocality, Jenkins and Mukamel have discussed molecular crystals in d dimensions ($d = 1, 2, 3$),³² where the relative motion of excitons is localized at a single molecule and the center-of-mass moves between molecules due to dipole-dipole interaction. While their theory concentrates on treating the resonant polarization without nonradiative relaxation, recently, microscopic nonlocality is being introduced into the field quantization in dispersive and absorptive media,^{33,34,35,36,37,38} and some studies have demonstrated the application of their theories for specific structures.^{33,38} Stefano et al. discussed excitons with the microscopic nonlocality in media with spatial translation symmetry broken along one dimension, and they practically calculated the spatial and frequency dependences of the vacuum-field fluctuation in a semiconductor quantum well structure.³³ Thereafter, they extended their theory to an arbitrary 3D structure,³⁶ and discussed the input-output relations in scattering systems.³⁷ On the other hand, Bechler performed the field quantization for the homogeneous systems with nonlocality by using the path-integral method,³⁵ and Suttrop did for the nonlocal, inhomogeneous, and anisotropic systems by using the diagonalization method.³⁴ Most recently, Raabe et al. phenomenologically discussed the nonlocal systems with both dielectric and magnetic properties.³⁸ They propose the use of the dielectric approximation with the surface impedance method for the practical application of their theory.

As seen in the above studies, it is safe to say that a consistent framework for the field quantization in dielectrics with microscopic nonlocality has already been established. Thus, the issue of current importance is to establish a general and practical calculation method applicable to arbitrary-structured 3D system, which is desired for the actual applications of the above framework though interesting applications have already been demonstrated in specific situations by Stefano et al. and Raabe et al. The essential task for this purpose is the derivation of Green's tensor for the Maxwell wave equation with nonlocal susceptibility as seen in Eq. (59) of the present paper.

In this paper, we provide a practical calculation method for Green's tensor for arbitrary structures by using the fact that the nonlocal susceptibility has a separable form with respect to two positions as seen in Eq. (54). This technique has been developed in the semiclassical microscopic nonlocal theory.^{21,22} We extend this theory to be able to consider the quantum mechanical properties of electromagnetic fields by using the Laplace transformation technique of SW.⁷ In other words, we generalize the SW theory to media with microscopic nonlocality for providing the practical calculation method. The SW theory is suitable to extend the nonlocal theory because the latter consists of two equations: the Maxwell wave equation, which is just the fundamental one in the SW theory as seen above, and the motion equation of excitonic polarization, which can easily be derived by the Laplace

transformation technique.

We provide the outline of our theory in the following sections, and the detailed explanations, including lengthy calculations, are given in Apps. A to D to keep straightforwardness of the main part. After that, we will compare our theory with some of the above-mentioned previously discussed ones.

IV. HAMILTONIAN

We describe the dielectric materials with resonant contributions from excitons with center-of-mass motion and the nonresonant ones with local dielectric function $\epsilon_{\text{bg}}(\mathbf{r}, \omega)$. This treatment is essential for including the consideration of the effects arising from the radiation mode structures modified by the practical dielectric structures (with absorption) such as a substrate, a dielectric multilayer cavity, photonic crystals, and so on, surrounding excitonic active structures. We explicitly discuss optical and nonradiative-relaxation processes of the excitons, and the nonresonant backgrounds are treated as the same procedure of SW.⁷ The total Hamiltonian discussed in the present paper is written as

$$H = H_{\text{em}} + H_{\text{int}} + H_{\text{mat}}, \quad (14)$$

where H_{em} describes the radiation field and background dielectric medium, H_{mat} represents excitons and a reservoir of oscillators, and H_{int} is the interactions between H_{em} and H_{mat} . H_{em} is just the complete Hamiltonian discussed by SW⁷, and its representation is shown in Eq. (B9) of the present paper.

As a result of the discussion in App. B, the interaction Hamiltonian is represented as

$$H_{\text{int}} = - \int d\mathbf{r} \left[\mathbf{I}_{\text{ex}}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) - \frac{1}{2} N_{\text{ex}}(\mathbf{r}) \mathbf{A}^2(\mathbf{r}) \right] + \int d\mathbf{r} \phi_{\text{bg}}(\mathbf{r}) \rho_{\text{ex}}(\mathbf{r}) + \frac{1}{2} \int d\mathbf{r} \phi_{\text{ex}}(\mathbf{r}) \rho_{\text{ex}}(\mathbf{r}). \quad (15)$$

$\mathbf{I}_{\text{ex}}(\mathbf{r})$ is the excitonic current density without radiation contribution $-N_{\text{ex}}(\mathbf{r})\mathbf{A}(\mathbf{r})$, i.e., the complete current density is written as $\mathbf{J}_{\text{ex}}(\mathbf{r}) = \mathbf{I}_{\text{ex}}(\mathbf{r}) - N_{\text{ex}}(\mathbf{r})\mathbf{A}(\mathbf{r})$ (see App. A). $\rho_{\text{ex}}(\mathbf{r})$ is the excitonic charge density and

$$\phi_{\text{ex}}(\mathbf{r}) \equiv \int d\mathbf{r}' \frac{\rho_{\text{ex}}(\mathbf{r}')}{4\pi\epsilon_0|\mathbf{r} - \mathbf{r}'|} \quad (16)$$

is the Coulomb potential. The first and second terms of Eq. (15) represent the interaction between the radiation field and excitons. The third term is the Coulomb interaction between induced charges of excitons and that of the background medium. The last term is the one between excitonic charges themselves, and is also considered as the dipole-dipole interaction between excitonic polarizations, or the exchange interaction between electrons and holes^{22,39,40} (see App. B). Although this term

usually belongs to matter Hamiltonian H_{mat} , we displace it into H_{int} because it can also be considered as the interaction between the longitudinal component of the polarization and that of the electric field. This treatment will give us the motion equation of excitons as a simple form as Eq. (28), and will take away our explicit consideration of the longitudinal-transverse (LT) splitting of the exciton eigenenergies, because the last term of Eq. (15) is the origin of the LT splitting.

With regard to excitons, generally, we should describe them starting from the basis of electrons and holes interacting with each other and themselves. However, as long as we consider a linear optical process of excitons under weak excitation, it is valid to describe electronic systems in terms of excitonic eigenstates. In addition, in order to describe a nonradiative relaxation process, we consider a reservoir of oscillators interacting with excitons. The matter Hamiltonian is written as

$$H_{\text{mat}} = \sum_{\mu} \hbar\omega_{\mu} b_{\mu}^{\dagger} b_{\mu} + \sum_{\mu} \int_0^{\infty} d\Omega \{ \hbar\Omega d_{\mu}^{\dagger}(\Omega) d_{\mu}(\Omega) + [b_{\mu} + b_{\mu}^{\dagger}] [g_{\mu}(\Omega) d_{\mu}(\Omega) + g_{\mu}^*(\Omega) d_{\mu}^{\dagger}(\Omega)] \}, \quad (17)$$

where b_{μ} is the annihilation operator of the excitons in eigenstate μ with eigenfrequency ω_{μ} , which does not include the LT splitting because we displace the exchange interaction between electrons and holes from H_{mat} to H_{int} . In this paper, we assume that the center-of-mass motion of excitons is confined in finite spaces, and index μ represents degrees of freedom of not only the relative motion but also the translational one. Instead of evaluating the commutation relations of b_{μ} from its representation (A23) with Fermi's commutation relations of electrons and holes, we consider the excitons as pure bosons satisfying

$$[b_{\mu}, b_{\mu'}^{\dagger}] = \delta_{\mu, \mu'}, \quad (18a)$$

$$[b_{\mu}, b_{\mu'}] = 0. \quad (18b)$$

This approximation is valid under weak excitation. On the other hand, in Eq. (17), $d_{\mu}(\Omega)$ is the annihilation operator of the reservoir oscillators with frequency Ω interacting with the excitons in state μ , and $g_{\mu}(\Omega)$ is its coupling parameter. The oscillators are independent of each other and satisfy the commutation relations as

$$[d_{\mu}(\Omega), d_{\mu'}^{\dagger}(\Omega')] = \delta_{\mu, \mu'} \delta(\Omega - \Omega'), \quad (19a)$$

$$[d_{\mu}(\Omega), d_{\mu'}(\Omega')] = 0. \quad (19b)$$

V. QED OF EXCITONS

In this section, from motion equations of excitons and the Maxwell wave equation for our system derived by the quantization technique of SW,⁷ we show our QED theory for excitons by using the technique of the microscopic nonlocal theory developed in the semiclassical framework.^{21,22} Since we require a lengthy calculation

based on the SW's scheme to prove the validity of the following discussion, we show only the outline of our theory and the details are discussed in Apps. C and D. We do not consider the nonradiative relaxation of excitons in this section. It will be discussed in the next section.

Since we consider excitons and also the background medium, the exciton-induced longitudinal field also contributes to the electric field as

$$\mathbf{E}(\mathbf{r}, t) = -\frac{\partial}{\partial t} \mathbf{A}(\mathbf{r}, t) - \nabla \phi_{\text{bg}}(\mathbf{r}, t) - \nabla \phi_{\text{ex}}(\mathbf{r}, t). \quad (20)$$

This definition is different from that of SW (Eq. (4) of the present paper). In addition, instead of Eq. (6), excitonic polarization density $\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega)$ appears in the Maxwell wave equation as

$$\begin{aligned} \nabla \times \nabla \times \hat{\mathbf{E}}^+(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \hat{\mathbf{E}}^+(\mathbf{r}, \omega) \\ = i\mu_0 \omega \hat{\mathbf{J}}_0(\mathbf{r}, \omega) + \mu_0 \omega^2 \hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) \end{aligned} \quad (21)$$

(see App. C), where $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ is the same kind of operator as $\hat{\mathbf{J}}(\mathbf{r}, \omega)$ in Eq. (6), and it satisfies the same commutation relations of Eq. (7):

$$\begin{aligned} [\hat{\mathbf{J}}_0(\mathbf{r}, \omega), \{\hat{\mathbf{J}}_0(\mathbf{r}', \omega')\}^{\dagger}] \\ = \delta(\omega - \omega') \delta(\mathbf{r} - \mathbf{r}') \frac{\epsilon_0 \hbar \omega^2}{\pi} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \mathbf{1} \end{aligned} \quad (22a)$$

$$[\hat{\mathbf{J}}_0(\mathbf{r}, \omega), \hat{\mathbf{J}}_0(\mathbf{r}', \omega')] = \mathbf{0}, \quad (22b)$$

(see App. D). This operator is interpreted as the source of the background electromagnetic fields and its definition is shown in Eq. (C32). Using Green's tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ satisfying Eq. (8), we can rewrite Eq. (21) as

$$\hat{\mathbf{E}}^+(\mathbf{r}, \omega) = \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) + \mu_0 \omega^2 \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}', \omega), \quad (23)$$

where

$$\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) \equiv i\mu_0 \omega \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{J}}_0(\mathbf{r}', \omega) \quad (24)$$

is the background electric field satisfying the commutation relations

$$[\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega), \hat{\mathbf{E}}_0^-(\mathbf{r}', \omega')] = \delta(\omega - \omega') \frac{\mu_0 \hbar \omega^2}{\pi} \text{Im}[\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)], \quad (25a)$$

$$[\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega), \hat{\mathbf{E}}_0^+(\mathbf{r}', \omega')] = \mathbf{0}. \quad (25b)$$

These are equivalent to relations (10) of the electric field in a local system. This equivalence indicates that the behavior of the background field in our system is exactly the same as that of the complete electric field in the local system. This natural result can be systematically derived from the representation of $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ (C32) and commutation relations of the system variables by using the Laplace

transformation technique. We have verified this equivalence for the linear optical process of excitons even with the nonradiative relaxation of excitons (see App. D).

Next, we discuss the motion of unknown variable $\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega)$ in the Maxwell wave equation (21). The second-quantized polarization density is written in terms of exciton operator set $\{b_\mu\}$ as

$$\mathbf{P}_{\text{ex}}(\mathbf{r}) = \sum_{\mu} [\mathcal{P}_{\mu}(\mathbf{r}) b_{\mu} + \mathcal{P}_{\mu}^*(\mathbf{r}) b_{\mu}^{\dagger}], \quad (26)$$

where the expansion coefficient $\mathcal{P}_{\mu}(\mathbf{r})$ is

$$\mathcal{P}_{\mu}(\mathbf{r}) = \mathcal{P}_{\mu} \mathbf{e}_{\mu} G_{\mu}(\mathbf{r}) \quad (27)$$

(see App. A). \mathcal{P}_{μ} is the transition dipole moment, \mathbf{e}_{μ} is a unit vector in the polarization direction, and $G_{\mu}(\mathbf{r})$ is the wave function of the center-of-mass motion in exciton state μ . Since we assume the weak confinement regime, \mathcal{P}_{μ} approximately depends only on the relative motion of excitons and is related with LT splitting $\Delta_{\text{LT}}^{\mu} = |\mathcal{P}_{\mu}|^2 / \epsilon_0 \epsilon_{\text{bg}}$. Neglecting the reservoir oscillators and assuming $\omega \sim \omega_{\mu}$, the Fourier-transformed Heisenberg equation of excitons is derived from Hamiltonian (15) and (17) as

$$(\hbar\omega_{\mu} - \hbar\omega - i\delta) \hat{b}_{\mu}(\omega) = \int d\mathbf{r} \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot \hat{\mathbf{E}}^+(\mathbf{r}, \omega) \quad (28)$$

(see App. D 1), where infinitesimal damping $i\delta$ is added to match the discussion with the nonradiative relaxation in the next section. Under the rotating wave approximation (RWA), the positive-frequency Fourier component of the polarization density (26) is written as

$$\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) = \sum_{\mu} \mathcal{P}_{\mu}(\mathbf{r}) \hat{b}_{\mu}(\omega). \quad (29)$$

Substituting Eq. (28) into (29), we obtain the nonlocal form of the polarization density as

$$\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) = \epsilon_0 \int d\mathbf{r}' \chi_{\text{ex}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{E}}^+(\mathbf{r}', \omega), \quad (30)$$

where the susceptibility tensor is defined as

$$\chi_{\text{ex}}(\mathbf{r}, \mathbf{r}', \omega) \equiv \frac{1}{\epsilon_0} \sum_{\mu} \frac{\mathcal{P}_{\mu}(\mathbf{r}) \mathcal{P}_{\mu}^*(\mathbf{r}')}{\hbar\omega_{\mu} - \hbar\omega - i\delta}. \quad (31)$$

The spatial spreading of the exciton state, the origin of the nonlocality, is reflected through polarization coefficient $\mathcal{P}_{\mu}(\mathbf{r})$ or center-of-mass wave function $G_{\mu}(\mathbf{r})$. On the other hand, the spatial structure of the background dielectrics is characterized by dielectric function $\epsilon_{\text{bg}}(\mathbf{r}, \omega)$ in Maxwell wave equation (21) and in commutation relations (22). In our framework, we can discuss arbitrary-structured exciton motions and background dielectrics as seen below.

In order to discuss the optical process of excitons, we must simultaneously solve Maxwell wave equation (21)

and the motion equation of polarization density (30) to determine unknown variables $\hat{\mathbf{E}}^+(\mathbf{r}, \omega)$ and $\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega)$. Substituting Eq. (30) into (21), we obtain the nonlocal wave equation as

$$\begin{aligned} \nabla \times \nabla \times \hat{\mathbf{E}}^+(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \hat{\mathbf{E}}^+(\mathbf{r}, \omega) \\ - \frac{\omega^2}{c^2} \int d\mathbf{r}' \chi_{\text{ex}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{E}}^+(\mathbf{r}', \omega) = i\mu_0 \omega \hat{\mathbf{J}}_0(\mathbf{r}, \omega). \end{aligned} \quad (32)$$

This is the same equation discussed by Savasta et al.^{36,37} and also has the same form as that of Raabe et al.³⁸ However, it appears very difficult to solve this nonlocal equation, and there remains a problem to derive Green's tensor for this equation in the practical application of their theories. This problem can be solved by using the fact that nonlocal susceptibility (31) has a separable form with respect to \mathbf{r} and \mathbf{r}' . One scheme is to directly derive Green's tensor for Eq. (32) as discussed in Ref. 41, and the other is to reduce this nonlocal problem into a simultaneous linear equation set.^{21,22} In our QED theory, we adopt the latter scheme because it provides not only Green's tensor of the former but also much interesting information on exciton-polariton systems.

Substituting the representation of the electric field (23) into the motion equation of excitons (28) by expanding $\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega)$ as Eq. (29), we obtain the linear equation set determining exciton amplitudes $\{\hat{b}_{\mu}(\omega)\}$ as

$$\begin{aligned} \sum_{\mu'} [(\hbar\omega_{\mu} - \hbar\omega - i\delta) \delta_{\mu, \mu'} + \mathcal{A}_{\mu, \mu'}(\omega)] \hat{b}_{\mu'}(\omega) \\ = (\hbar\omega_{\mu} - \hbar\omega - i\delta) \hat{b}_{\mu}^{(0)}(\omega). \end{aligned} \quad (33)$$

This has the same form of the self-consistent equation set in the semiclassical nonlocal theory.^{21,22} The operator on the right hand side

$$\hat{b}_{\mu}^{(0)}(\omega) \equiv \frac{1}{\hbar\omega_{\mu} - \hbar\omega - i\delta} \int d\mathbf{r} \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) \quad (34)$$

has the same form of Eq. (28) replacing $\hat{\mathbf{E}}^+(\mathbf{r}, \omega)$ with $\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega)$; then, it can be interpreted as the exciton amplitude directly induced by the background electric field. Here, we use the word ‘‘directly’’ to mean that $\hat{b}_{\mu}^{(0)}(\omega)$ does not include the diffusion of the exciton amplitudes via the electromagnetic fields. Such effect is reflected in the correction term of eigenenergy $\hbar\omega_{\mu}$ in Eq. (33):

$$\mathcal{A}_{\mu, \mu'}(\omega) \equiv -\mu_0 \omega^2 \int d\mathbf{r} \int d\mathbf{r}' \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathcal{P}_{\mu'}(\mathbf{r}'). \quad (35)$$

This value represents the exciton-exciton interaction via the electromagnetic fields, i.e., the polarization at \mathbf{r}' induces electric field, and later it induces another polarization at \mathbf{r} . The interaction between the transverse fields is the retarded interaction, and the one between the longitudinal fields is interpreted as the Coulomb interaction

between induced charges. The latter is just the exchange interaction between electrons and holes, which we displace from H_{mat} to H_{int} , and it gives the LT splitting of the exciton eigenenergies. The radiative relaxation of excitons and the transition between exciton states are described by the imaginary part of $\mathcal{A}_{\mu,\mu'}(\omega)$.

Writing the coefficient matrix element of Eq. (33) as

$$S_{\mu,\mu'}(\omega) \equiv (\hbar\omega_{\mu} - \hbar\omega - i\delta)\delta_{\mu,\mu'} + \mathcal{A}_{\mu,\mu'}(\omega), \quad (36)$$

the self-consistent equation set is rewritten as

$$\sum_{\mu'} S_{\mu,\mu'}(\omega) \hat{b}_{\mu'}(\omega) = \int d\mathbf{r} \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega). \quad (37)$$

By using the inverse matrix $\mathbf{W}(\omega) = [\mathbf{S}(\omega)]^{-1}$ with the basis of exciton states, we obtain the representation of exciton operators as

$$\hat{b}_{\mu}(\omega) = \sum_{\mu'} W_{\mu,\mu'}(\omega) \int d\mathbf{r} \mathcal{P}_{\mu'}^*(\mathbf{r}) \cdot \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega). \quad (38)$$

We can describe all the physical variables in terms of these operators and $\hat{\mathbf{E}}_0^{\pm}(\mathbf{r}, \omega)$. For example, the excitonic polarization is written as Eq. (29) and the electric field (23) as

$$\hat{\mathbf{E}}^+(\mathbf{r}, \omega) = \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) + \sum_{\mu} \boldsymbol{\mathcal{E}}_{\mu}(\mathbf{r}, \omega) \hat{b}_{\mu}(\omega), \quad (39)$$

where the coefficients are defined as

$$\boldsymbol{\mathcal{E}}_{\mu}(\mathbf{r}, \omega) \equiv \mu_0\omega^2 \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathcal{P}_{\mu}(\mathbf{r}'), \quad (40)$$

$$\mathcal{F}_{\mu}(\mathbf{r}, \omega) \equiv \mu_0\omega^2 \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathcal{P}_{\mu}^*(\mathbf{r}'). \quad (41)$$

The latter will appear in Eq. (44). The exciton operators are represented by the background electric field $\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega)$, whose commutation relations are described by Green's tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ as shown in Eq. (25). Since there is no problem in deriving Green's tensor as explained above, the additional works are to perform integrations (35), (40), and (41), and to derive inverse matrix $\mathbf{W}(\omega) = [\mathbf{S}(\omega)]^{-1}$. These calculations can be performed straightforwardly. Therefore, in our framework, for a given wave function of excitons' center-of-mass motion $\{G_{\mu}(\mathbf{r})\}$ and background dielectric function $\epsilon_{\text{bg}}(\mathbf{r}, \omega)$, we can discuss the QED of excitonic materials with the microscopic nonlocality.

When we do not consider the nonradiative relaxation of excitons, all the quantum mechanical properties are described by commutation relations (22) of noise current density $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$. Using relations (25) derived from Eqs. (22), the commutation relations of exciton operators (38) are evaluated as

$$\begin{aligned} & [\hat{b}_{\mu}(\omega), \{\hat{b}_{\mu'}(\omega')\}^{\dagger}] \\ &= \delta(\omega - \omega') \frac{\hbar}{i2\pi} [W_{\mu,\mu'}(\omega) - W_{\mu',\mu}^*(\omega)], \end{aligned} \quad (42a)$$

$$[\hat{b}_{\mu}(\omega), \hat{b}_{\mu'}(\omega')] = 0. \quad (42b)$$

This result indicates that the elements of inverse matrix $\mathbf{W}(\omega)$ of the self-consistent equation set identify with the Fourier transforms of the retarded correlation functions of excitons except for the factor $-\hbar$ (see App. E). In addition, we can obtain the commutation relations of electric field operators (39):

$$\begin{aligned} & [\hat{\mathbf{E}}^+(\mathbf{r}, \omega), \hat{\mathbf{E}}^-(\mathbf{r}', \omega')] = \delta(\omega - \omega') \\ & \times \frac{\mu_0\hbar\omega^2}{i2\pi} [\mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega) - \{\mathbf{G}_{\text{ren}}(\mathbf{r}', \mathbf{r}, \omega)\}^{t*}], \end{aligned} \quad (43a)$$

$$[\hat{\mathbf{E}}^+(\mathbf{r}, \omega), \hat{\mathbf{E}}^+(\mathbf{r}', \omega')] = \mathbf{0}, \quad (43b)$$

where $\mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega)$ is defined as

$$\begin{aligned} \mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega) &= \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \\ &+ \frac{1}{\mu_0\omega^2} \sum_{\mu,\mu'} \boldsymbol{\mathcal{E}}_{\mu}(\mathbf{r}, \omega) W_{\mu,\mu'}(\omega) \mathcal{F}_{\mu'}(\mathbf{r}', \omega). \end{aligned} \quad (44)$$

We can find that if this tensor satisfies the equation

$$\begin{aligned} & \nabla \times \nabla \times \mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega) \\ & - \frac{\omega^2}{c^2} \int d\mathbf{r}'' \boldsymbol{\chi}_{\text{ex}}(\mathbf{r}, \mathbf{r}'', \omega) \cdot \mathbf{G}_{\text{ren}}(\mathbf{r}'', \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \mathbf{1}, \end{aligned} \quad (45)$$

then it can be interpreted as Green's tensor for nonlocal Maxwell wave equation (32). This tensor, which renormalizes the linear optical process of excitons with the microscopic nonlocality, is also calculated directly from the above nonlocal equation.⁴¹ The breakdown of reciprocity relation (12) for $\mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega)$ arises from anisotropic susceptibility tensor (31) of the excitonic polarization. By using $\mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega)$, electric field operator (39) is also written as

$$\hat{\mathbf{E}}^+(\mathbf{r}, \omega) = i\mu_0\omega \int d\mathbf{r}' \mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{J}}_0(\mathbf{r}', \omega). \quad (46)$$

Comparing Eqs. (9), (10), (43), and (46), we can find a good correspondence between our theory and the QED theories for local systems. In addition, we also verify that commutation relations (42) and (43) satisfy the equal-time commutation relations expected in the Schrödinger representation (see App. F).

VI. WITH NONRADIATIVE RELAXATION

Next, we discuss the modulation of the above theory by considering the nonradiative relaxation of excitons. The detailed calculation is shown in App. D3, and the result is as follows. The nonradiative relaxation slightly modulates the motion equation of excitons from the original one (28):

$$\begin{aligned} & [\hbar\omega_{\mu} - \hbar\omega - i\gamma_{\mu}(\omega)/2] \hat{b}_{\mu}(\omega) \\ &= \int d\mathbf{r} \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot \hat{\mathbf{E}}^+(\mathbf{r}, \omega) + \hat{\mathcal{D}}_{\mu}(\omega). \end{aligned} \quad (47)$$

$\gamma_\mu(\omega)$ is the nonradiative relaxation width defined as Eq. (D48) in terms of exciton-reservoir interaction coefficient $g_\mu(\Omega)$. Operator $\hat{\mathcal{D}}_\mu(\omega)$ represents the fluctuation caused by the reservoir oscillators. It is defined as Eq. (D51) and satisfies the commutation relations

$$\left[\hat{\mathcal{D}}_\mu(\omega), \{\hat{\mathcal{D}}_{\mu'}(\omega')\}^\dagger\right] = \delta_{\mu,\mu'} \delta(\omega - \omega') \frac{\hbar}{2\pi} \gamma_\mu(\omega), \quad (48a)$$

$$\left[\hat{\mathcal{D}}_\mu(\omega), \hat{\mathcal{D}}_{\mu'}(\omega')\right] = 0. \quad (48b)$$

This is another source operator of our system and is independent from noise current density $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ as

$$\left[\hat{\mathcal{D}}_\mu(\omega), \hat{\mathbf{J}}_0(\mathbf{r}, \omega')\right] = \left[\hat{\mathcal{D}}_\mu(\omega), \{\hat{\mathbf{J}}_0(\mathbf{r}, \omega')\}^\dagger\right] = \mathbf{0}. \quad (49)$$

Substituting representation (23) of the electric field into Eq. (47), we obtain the self-consistent equation set as

$$\sum_{\mu'} S_{\mu,\mu'}^{\text{abs}}(\omega) \hat{b}_{\mu'}(\omega) = \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) + \hat{\mathcal{D}}_\mu(\omega), \quad (50)$$

where the coefficient matrix element is

$$S_{\mu,\mu'}^{\text{abs}}(\omega) \equiv [\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2] \delta_{\mu,\mu'} + \mathcal{A}_{\mu,\mu'}(\omega). \quad (51)$$

The above result indicates that we can easily introduce the nonradiative relaxation into the discussion of the previous section. All we have to do is introduce relaxation width $\gamma_\mu(\omega)$ into coefficient matrix (51) of the self-consistent equation set. By deriving its inverse matrix $\mathbf{W}^{\text{abs}}(\omega) = [\mathbf{S}^{\text{abs}}(\omega)]^{-1}$, we obtain the commutation relations for excitons as

$$\left[\hat{b}_\mu(\omega), \{\hat{b}_{\mu'}(\omega')\}^\dagger\right] = \delta(\omega - \omega') \frac{\hbar}{i2\pi} [W_{\mu,\mu'}^{\text{abs}}(\omega) - \{W_{\mu',\mu}^{\text{abs}}(\omega)\}^*], \quad (52a)$$

$$\left[\hat{b}_\mu(\omega), \hat{b}_{\mu'}(\omega')\right] = 0. \quad (52b)$$

These have the same forms as those of Eqs. (42).

On the other hand, substituting Eq. (47) into Eq. (29), the excitonic polarization (30) is rewritten as

$$\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) = \epsilon_0 \int d\mathbf{r}' \chi_{\text{ex}}^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{E}}^+(\mathbf{r}, \omega) + \sum_\mu \frac{\mathcal{P}_\mu(\mathbf{r}) \hat{\mathcal{D}}_\mu(\omega)}{\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2}, \quad (53)$$

where the nonlocal susceptibility is also rewritten as

$$\chi_{\text{ex}}^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega) \equiv \frac{1}{\epsilon_0} \sum_\mu \frac{\mathcal{P}_\mu(\mathbf{r}) \mathcal{P}_\mu^*(\mathbf{r}')}{\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2}. \quad (54)$$

Substituting Eq. (53) into Maxwell wave equation (21), nonlocal wave equation (32) becomes

$$\begin{aligned} \nabla \times \nabla \times \hat{\mathbf{E}}^+(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \hat{\mathbf{E}}^+(\mathbf{r}, \omega) \\ - \frac{\omega^2}{c^2} \int d\mathbf{r}' \chi_{\text{ex}}^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{E}}^+(\mathbf{r}', \omega) = i\mu_0 \omega \hat{\mathbf{J}}_0^{\text{abs}}(\mathbf{r}, \omega), \end{aligned} \quad (55)$$

where we define a new noise current density as

$$\hat{\mathbf{J}}_0^{\text{abs}}(\mathbf{r}, \omega) \equiv \hat{\mathbf{J}}_0(\mathbf{r}, \omega) - i\omega \sum_\mu \frac{\mathcal{P}_\mu(\mathbf{r}) \hat{\mathcal{D}}_\mu(\omega)}{\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2}. \quad (56)$$

From commutation relations (22), (48), and (49), we can obtain the commutation relations for $\hat{\mathbf{J}}_0^{\text{abs}}(\mathbf{r}, \omega)$ as

$$\begin{aligned} \left[\hat{\mathbf{J}}_0^{\text{abs}}(\mathbf{r}, \omega), \{\hat{\mathbf{J}}_0^{\text{abs}}(\mathbf{r}', \omega')\}^\dagger\right] \\ = \delta(\omega - \omega') \frac{\epsilon_0 \hbar \omega^2}{\pi} \text{Im}[\epsilon^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega)] \end{aligned} \quad (57a)$$

$$\left[\hat{\mathbf{J}}_0^{\text{abs}}(\mathbf{r}, \omega), \hat{\mathbf{J}}_0^{\text{abs}}(\mathbf{r}', \omega')\right] = \mathbf{0}, \quad (57b)$$

where $\epsilon^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega) \equiv \mathbf{1}\delta(\mathbf{r} - \mathbf{r}')\epsilon_{\text{bg}}(\mathbf{r}, \omega) + \chi_{\text{ex}}^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega)$ is the nonlocal dielectric tensor. This is a natural result from the fluctuation theorem as discussed in Ref. 36-38. In addition, we also obtain the commutation relations for the electric field as having the same form as that of Eq. (43) by replacing $\mathbf{G}_{\text{ren}}(\mathbf{r}, \mathbf{r}', \omega)$ with

$$\begin{aligned} \mathbf{G}_{\text{ren}}^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \\ + \frac{1}{\mu_0 \omega^2} \sum_{\mu,\mu'} \boldsymbol{\mathcal{E}}_\mu(\mathbf{r}, \omega) W_{\mu,\mu'}^{\text{abs}}(\omega) \mathcal{F}_{\mu'}(\mathbf{r}', \omega), \end{aligned} \quad (58)$$

which satisfies the nonlocal wave equation

$$\begin{aligned} \nabla \times \nabla \times \mathbf{G}_{\text{ren}}^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \mathbf{G}_{\text{ren}}^{\text{abs}}(\mathbf{r}, \mathbf{r}', \omega) \\ - \frac{\omega^2}{c^2} \int d\mathbf{r}'' \chi_{\text{ex}}^{\text{abs}}(\mathbf{r}, \mathbf{r}'', \omega) \cdot \mathbf{G}_{\text{ren}}^{\text{abs}}(\mathbf{r}'', \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \mathbf{1} \end{aligned} \quad (59)$$

and Green's tensor required for the practical application of nonlocal QED theories.^{37,38}

VII. DISCUSSION

In this paper, we have constructed a QED theory for excitons weakly confined in arbitrary-structured dielectrics with microscopic nonlocality and nonradiative relaxation of excitons. On the other hand, as mentioned in Sec. III, the QED of media with the microscopic nonlocality has already been discussed in a few studies. From the viewpoint of a practical application, we compare our theory with the studies of Stefano et al.^{33,36,37} and Raabe et al.³⁸

Stefano et al. have discussed quantum-well structures of the dispersive and absorptive dielectrics with microscopic nonlocality in Ref. 33, and their theory is generalized to enable the consideration of arbitrary structures in Ref. 36 and 37. However, there still remains a problem in deriving Green's tensors for the nonlocal wave equation as shown in Eq. (45) or (59) in the present paper. Our theory provides a solution to this problem by giving

a definite calculation method using Green's tensor satisfying wave equation (8) for local media and the fact that the nonlocal susceptibility has a separable form as shown in Eq. (31) or (54). The problem of Ref. 37 can be solved by using our theory because we derive Green's tensor (44) or (58) for arbitrary-structured excitonic polarization and background dielectrics.

On the other hand, Raabe et al. propose the use of the dielectric approximation with the surface impedance method for the practical calculation of Green's tensor for the nonlocal Maxwell wave equation. In the dielectric approximation, the characteristic length of spatial dispersion (the spatial spreading of the center-of-mass motion of excitons) is assumed to be small compared to the spatial length of materials, and the information outside a focusing region is compressed to integrations of the electromagnetic fields at the interfaces. Green's tensor can be derived using the surface impedance method for a given surface impedance or admittance, which just includes the outside information. In contrast, our theory provides Green's tensor for given $\epsilon_{bg}(\mathbf{r}, \omega)$ and center-of-mass wave functions of excitons by applying only the RWA. Since nonlocality becomes essential only under the resonance conditions, the RWA does not impose any significant restriction on our theory for the discussion of nonlocal systems. In addition, our calculation method would be generalized to the one without the RWA as performed in the semiclassical nonlocal theory.^{21,22}

As mentioned in Sec. I, there is growing interest in the QED of elementary excitations in condensed matters. For example, theoretical studies on entangled-photon generation via biexcitons have already been performed by Savasta et al.^{42,43} (though the microscopic nonlocality was not sufficiently considered in these calculations) and also by us by extending our QED theory reported in the present paper.⁴⁴ When we discuss such nonlinear processes of excitons, we must self-consistently treat their nonlinear motion equation and the Maxwell wave equation. Based on self-consistent equation set (37) or (50) as discussed in the present paper, the new task is to solve the equation set with nonlinear terms originating from nonlinear processes. On the other hand, based on Maxwell wave equation (32) or (55) with the nonlocal susceptibility as discussed in the previously discussed QED theories,^{33,34,35,36,37,38} we must solve the wave equation with both nonlinear and nonlocal susceptibility. Both approaches can be performed by applying some techniques such as successive approximation; however, they generally require much hard work. In such a case, more detailed and systematic calculation should be performed by using the Feynman diagram technique with the correlation functions of excitons derived using our QED theory. In this sense, our scheme will be a powerful tool to discuss the nonlinear quantum optics in condensed matters with the microscopic nonlocality.

VIII. SUMMARY

By using the quantization technique of Suttorp et al.,^{7,14} we have extended the microscopic nonlocal theory^{21,22} developed in the semiclassical framework to enable the consideration of quantum mechanical properties of the electromagnetic fields. This theory keeps good correspondences with both the nonlocal theory and the QED theories for dispersive and absorptive materials with local susceptibility. While microscopic nonlocality has been considered in the form of nonlocal susceptibility in some of the QED theories,^{33,34,36,37,38} we reduce the problem to a linear equation set as discussed in the semiclassical framework.^{21,22} In addition, this theory provides Green's tensor for the Maxwell wave equation with a nonlocal susceptibility, which is just required for a practical application of previous studies. By using our theory, we can discuss the QED of excitons weakly confined in nanostructures, which are known to show anomalous nonlinear optical phenomena.^{25,45,46,47,48} Although this paper is devoted to only the linear optical process of excitons, we can phenomenologically extend our theory to describe nonlinear processes, such as the entangled-photon generation via biexcitons in nanostructures.⁴⁴ In addition, our theory has a potential to systematically discuss higher order nonlinear processes of elementary excitations in condensed matters by using the Feynman diagram technique with correlation functions of excitons derived in our calculation. Based on our QED theory, we are going to discuss various optical phenomena which cannot be discussed in the semiclassical framework.

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APPENDIX A: THE SECOND QUANTIZATION OF EXCITONIC POLARIZATION

In this appendix, we provide microscopic descriptions of the current density, charge density, and polarization density of charged particles. Then, we expand them in terms of the electron or exciton operators. We write the second-quantized operators with a hat ($\hat{}$) in this appendix.

Considering the charged particles with mass m_i and charge q_i at position \mathbf{r}_i , current density $\mathbf{J}_{cp}(\mathbf{r})$ and charge

density $\rho_{\text{cp}}(\mathbf{r})$ are written as

$$\mathbf{J}_{\text{cp}}(\mathbf{r}) \equiv \sum_i \frac{q_i}{2} [\dot{\mathbf{r}}_i \delta(\mathbf{r} - \mathbf{r}_i) + \delta(\mathbf{r} - \mathbf{r}_i) \dot{\mathbf{r}}_i], \quad (\text{A1})$$

$$\rho_{\text{cp}}(\mathbf{r}) \equiv \sum_i q_i \delta(\mathbf{r} - \mathbf{r}_i). \quad (\text{A2})$$

Here, due to the interaction with the radiation field (see App. B), the momentum of the charged particles is written as

$$\mathbf{p}_i = m_i \dot{\mathbf{r}}_i + q_i \mathbf{A}(\mathbf{r}_i). \quad (\text{A3})$$

Then, current density (A1) includes a contribution from the radiation field. In order to expand it in terms of electron or exciton operators, we extract the radiation contribution from $\mathbf{J}_{\text{cp}}(\mathbf{r})$:

$$\mathbf{I}_{\text{cp}}(\mathbf{r}) \equiv \sum_i \frac{q_i}{2m_i} [\mathbf{p}_i \delta(\mathbf{r} - \mathbf{r}_i) + \delta(\mathbf{r} - \mathbf{r}_i) \mathbf{p}_i]. \quad (\text{A4})$$

By writing the coefficient of the radiation contribution as

$$N_{\text{cp}}(\mathbf{r}) \equiv \sum_i \frac{q_i^2}{m_i} \delta(\mathbf{r} - \mathbf{r}_i), \quad (\text{A5})$$

we can write complete current density (A1) as

$$\mathbf{J}_{\text{cp}}(\mathbf{r}) = \mathbf{I}_{\text{cp}}(\mathbf{r}) - N_{\text{cp}}(\mathbf{r}) \mathbf{A}(\mathbf{r}). \quad (\text{A6})$$

This subtraction of the radiation contribution is discussed in section 2.2 of Ref. 22.

Next, we expand the above variables in terms of the electron operator \hat{a}_η and its wave function $\varphi_\eta(\mathbf{r})$. The field operator is written as

$$\hat{\psi}(\mathbf{r}) = \sum_c \hat{a}_c \varphi_c(\mathbf{r}) + \sum_v \hat{a}_v \varphi_v(\mathbf{r}), \quad (\text{A7})$$

where labels c and v represent the degrees of freedom of conduction and valence electrons, respectively. Assuming optical excitation of electron-hole pairs, we obtain the second-quantized form of the above variables as

$$\hat{\mathbf{I}}_{\text{ex}}(\mathbf{r}) = \frac{(-e)}{2m} \sum_{c,v} \hat{a}_v^\dagger \hat{a}_c [\varphi_v^*(\mathbf{r}) \mathbf{p} \varphi_c(\mathbf{r}) - \varphi_c(\mathbf{r}) \mathbf{p} \varphi_v^*(\mathbf{r})] + \text{H.c.}, \quad (\text{A8})$$

$$\hat{N}_{\text{ex}}(\mathbf{r}) = \frac{(-e)^2}{m} \sum_{c,v} \hat{a}_v^\dagger \hat{a}_c \varphi_v^*(\mathbf{r}) \varphi_c(\mathbf{r}) + \text{H.c.}, \quad (\text{A9})$$

$$\hat{\rho}_{\text{ex}}(\mathbf{r}) = (-e) \sum_{c,v} \hat{a}_v^\dagger \hat{a}_c \varphi_v^*(\mathbf{r}) \varphi_c(\mathbf{r}) + \text{H.c.} \quad (\text{A10})$$

These operators are also written in terms of exciton operators $\{\hat{b}_\mu\}$ as polarization density (26):

$$\hat{\mathbf{I}}_{\text{ex}}(\mathbf{r}) = \sum_\mu \mathcal{I}_\mu(\mathbf{r}) \hat{b}_\mu + \text{H.c.}, \quad (\text{A11})$$

$$\hat{N}_{\text{ex}}(\mathbf{r}) = \sum_\mu N_\mu(\mathbf{r}) \hat{b}_\mu + \text{H.c.}, \quad (\text{A12})$$

$$\hat{\rho}_{\text{ex}}(\mathbf{r}) = \sum_\mu \rho_\mu(\mathbf{r}) \hat{b}_\mu + \text{H.c.} \quad (\text{A13})$$

Instead of evaluating the expansion coefficient of each operator, we describe them in terms of $\mathcal{P}_\mu(\mathbf{r})$, the coefficient of polarization density (26). From the relations

$$\hat{\rho}_{\text{ex}}(\mathbf{r}) = -\nabla \cdot \hat{\mathbf{P}}_{\text{ex}}(\mathbf{r}), \quad (\text{A14})$$

$$\hat{\mathbf{J}}_{\text{ex}}(\mathbf{r}) = \frac{\partial}{\partial t} \hat{\mathbf{P}}_{\text{ex}}(\mathbf{r}) = \frac{1}{i\hbar} [\hat{\mathbf{P}}_{\text{ex}}(\mathbf{r}), \hat{H}], \quad (\text{A15})$$

and considering weak exciton-photon interaction, i.e., $\hat{H} \sim \hat{H}_{\text{mat}}$ and $\hat{\mathbf{J}}_{\text{ex}}(\mathbf{r}) \sim \hat{\mathbf{I}}_{\text{ex}}(\mathbf{r})$, we can write the above coefficients as

$$\mathcal{I}_\mu(\mathbf{r}) = -i\omega_\mu \mathcal{P}_\mu(\mathbf{r}), \quad (\text{A16})$$

$$N_\mu(\mathbf{r}) = (-e/m) \rho_\mu(\mathbf{r}), \quad (\text{A17})$$

$$\rho_\mu(\mathbf{r}) = -\nabla \cdot \mathcal{P}_\mu(\mathbf{r}), \quad (\text{A18})$$

where ω_μ is the eigenfrequency of excitons. Using above operators (A11)-(A13), the excitonic current density and Coulomb potential are respectively written as

$$\hat{\mathbf{J}}_{\text{ex}}(\mathbf{r}) = \hat{\mathbf{I}}_{\text{ex}}(\mathbf{r}) - \hat{N}_{\text{ex}}(\mathbf{r}) \mathbf{A}(\mathbf{r}), \quad (\text{A19})$$

$$\hat{\phi}_{\text{ex}}(\mathbf{r}) = \int d\mathbf{r}' \frac{\hat{\rho}_{\text{ex}}(\mathbf{r}')}{4\pi\epsilon_0 |\mathbf{r} - \mathbf{r}'|}. \quad (\text{A20})$$

In order to evaluate coefficients (A16)-(A18), we derive the representation of $\mathcal{P}_\mu(\mathbf{r})$ from the microscopic description of the polarization density. Averaging the polarization at lattice point \mathbf{R}_0 over a unit cell, the microscopic description is written as

$$\mathbf{P}_{\text{cp}}(\mathbf{R}_0) \equiv \frac{1}{\Omega} \int_\Omega d\mathbf{r} \sum_i q_i \mathbf{r} \delta(\mathbf{R}_0 + \mathbf{r} - \mathbf{r}_i), \quad (\text{A21})$$

where the integration is over the unit cell and Ω is its volume. Explicitly indicating the lattice point of the electron states as (η, \mathbf{R}) and assuming their wave function as the wanner function $w_\eta(\mathbf{r} - \mathbf{R})$, we obtain the second-quantized form of the polarization density as

$$\begin{aligned} \hat{\mathbf{P}}_{\text{ex}}(\mathbf{R}_0) &= \sum_{c,v,\mathbf{R},\mathbf{R}'} \hat{a}_{v,\mathbf{R}_0+\mathbf{R}'}^\dagger \hat{a}_{c,\mathbf{R}_0+\mathbf{R}+\mathbf{R}'} \\ &\times \frac{1}{\Omega} \int_\Omega d\mathbf{r} w_v^*(\mathbf{r} - \mathbf{R}') (-e) \mathbf{r} w_c(\mathbf{r} - \mathbf{R} - \mathbf{R}') + \text{H.c.} \end{aligned} \quad (\text{A22})$$

We expand this in terms of the exciton operators

$$\hat{b}_{\mu,m} \equiv \sum_{c,v,\mathbf{R}_0,\mathbf{R}} \Phi_{\mu,c,v,\mathbf{R}}^* G_{m,\mathbf{R}_0} \hat{a}_{v,\mathbf{R}_0}^\dagger \hat{a}_{c,\mathbf{R}_0+\mathbf{R}}, \quad (\text{A23})$$

where μ and $\Phi_{\mu,c,v,\mathbf{R}}$ are, respectively, the quantum number and the wave function of the relative motion of excitons, and m and G_{m,\mathbf{R}_0} are those of the center-of-mass motion. From the completeness of the wave functions, we can rewrite the electron-hole operator set as

$$\hat{a}_{v,\mathbf{R}_0}^\dagger \hat{a}_{c,\mathbf{R}_0+\mathbf{R}} = \sum_{\mu,m} \Phi_{\mu,c,v,\mathbf{R}} G_{m,\mathbf{R}_0} \hat{b}_{\mu,m}. \quad (\text{A24})$$

Using this relation, we can expand Eq. (A22) in terms of the exciton operators as

$$\hat{\mathbf{P}}_{\text{ex}}(\mathbf{R}_0) = \sum_{\mu,m} \mathcal{P}_{\mu,m}(\mathbf{R}_0) \hat{b}_{\mu,m} + \text{H.c.}, \quad (\text{A25})$$

where the expansion coefficient is written as

$$\begin{aligned} \mathcal{P}_{\mu,m}(\mathbf{R}_0) &\equiv \sum_{c,v,\mathbf{R},\mathbf{R}'} G_{m,\mathbf{R}_0+\mathbf{R}'} \Phi_{\mu,c,v,\mathbf{R}} \\ &\times \frac{1}{\Omega} \int_{\Omega} d\mathbf{r} w_v^*(\mathbf{r}-\mathbf{R}') (-e)\mathbf{r} w_c(\mathbf{r}-\mathbf{R}-\mathbf{R}'). \end{aligned} \quad (\text{A26})$$

Supposing that the spatial variation of the center-of-mass wave function is negligible within the extent of the electron-hole relative wave function, we can consider $G_{m,\mathbf{R}_0+\mathbf{R}'} \sim G_{m,\mathbf{R}_0}$. Further, by expanding the integration range to the entire crystal region with iterating \mathbf{R}' , we obtain

$$\begin{aligned} \mathcal{P}_{\mu,m}(\mathbf{R}_0) &\sim G_{m,\mathbf{R}_0} \sum_{c,v,\mathbf{R}} \Phi_{\mu,c,v,\mathbf{R}} \\ &\times \frac{1}{\Omega} \int_{\Omega} d\mathbf{r} w_v^*(\mathbf{r}) (-e)\mathbf{r} w_c(\mathbf{r}-\mathbf{R}). \end{aligned} \quad (\text{A27})$$

Here, if we assume the wave functions to be smooth with respect to the spatial position, i.e., $G_m(\mathbf{R}_0) = G_{m,\mathbf{R}_0}/\sqrt{\Omega}$ and $\Phi_{\mu,c,v}(\mathbf{R}) = \Phi_{\mu,c,v,\mathbf{R}}/\sqrt{\Omega}$, we obtain the expansion coefficient of polarization density (A25) as

$$\mathcal{P}_{\mu,m}(\mathbf{R}_0) = \mathcal{P}_{\mu} G_m(\mathbf{R}_0), \quad (\text{A28})$$

where

$$\mathcal{P}_{\mu} \equiv \sum_{c,v,\mathbf{R}} \Phi_{\mu,c,v}(\mathbf{R}) \int_{\Omega} d\mathbf{r} w_v^*(\mathbf{r}) (-e)\mathbf{r} w_c(\mathbf{r}-\mathbf{R}) \quad (\text{A29})$$

is the transition dipole moment of exciton band μ , and its absolute value is related with the LT splitting of the exciton eigenenergy as $\Delta_{\text{LT}}^{\mu} = |\mathcal{P}_{\mu}|^2/\epsilon_0\epsilon_{\text{bg}}$.

APPENDIX B: DERIVATION OF HAMILTONIAN

As a model of the background system, i.e., radiation field and local dielectrics, we adopt the system discussed by SW.⁷ Considering the charged particles of App. A, the total Lagrangian is written as

$$L = \sum_i \left[\frac{1}{2} m_i \dot{\mathbf{r}}_i^2 - V(\mathbf{r}_i) \right] + \int d\mathbf{r} \mathcal{L}, \quad (\text{B1})$$

where $V(\mathbf{r}_i)$ is the one-body potential of the particles and \mathcal{L} is the Lagrangian density depending on the spatial

position:

$$\begin{aligned} \mathcal{L} &= \frac{1}{2} \epsilon_0 \mathbf{E}^2 - \frac{1}{2\mu_0} \mathbf{B}^2 + \frac{1}{2} \rho \dot{\mathbf{X}}^2 - \frac{1}{2} \rho \omega_0^2 \mathbf{X}^2 \\ &- (\phi_{\text{bg}} + \phi_{\text{cp}})(\rho_{\text{bg}} + \rho_{\text{cp}}) + \mathbf{A} \cdot (-\alpha \dot{\mathbf{X}} + \mathbf{J}_{\text{cp}}) \\ &+ \frac{1}{2} \rho \int_0^{\infty} d\omega \dot{\mathbf{Y}}_{\omega}^2 - \frac{1}{2} \rho \int_0^{\infty} d\omega \omega^2 \mathbf{Y}_{\omega}^2 \\ &- \int_0^{\infty} d\omega v_{\omega} \mathbf{X} \cdot \dot{\mathbf{Y}}_{\omega}. \end{aligned} \quad (\text{B2})$$

We omit the descriptions of position dependences. $\mathbf{E} = -\dot{\mathbf{A}} - \nabla\phi_{\text{bg}} - \nabla\phi_{\text{cp}}$ is the electric field, and $\mathbf{B} = \nabla \times \mathbf{A}$ is the magnetic induction. $\mathbf{X}(\mathbf{r})$ is the amplitude of polarizable harmonic oscillators with density $\rho(\mathbf{r})$ and eigenfrequency $\omega_0(\mathbf{r})$. These oscillators describe the background medium in our QED theory. The polarization density, charge density, and current density of the background are respectively written as $-\alpha\mathbf{X}$, $\rho_{\text{bg}} = \nabla \cdot (\alpha\mathbf{X})$, and $-\alpha\dot{\mathbf{X}}$ with position-dependent coefficient $\alpha(\mathbf{r})$. The background Coulomb potential is written as

$$\phi_{\text{bg}} = \int d\mathbf{r}' \frac{\rho'_{\text{bg}}}{4\pi\epsilon_0|\mathbf{r}-\mathbf{r}'|} = \int d\mathbf{r}' \frac{\nabla' \cdot (\alpha'\mathbf{X}')}{4\pi\epsilon_0|\mathbf{r}-\mathbf{r}'|}. \quad (\text{B3})$$

ϕ_{bg} is related with the longitudinal component of the polarization as

$$\nabla\phi_{\text{bg}} = -\frac{1}{\epsilon_0} [\alpha\mathbf{X}]_{\text{L}}, \quad (\text{B4})$$

and it also satisfies the Poisson equation

$$\nabla^2\phi_{\text{bg}} = -\frac{\rho_{\text{bg}}}{\epsilon_0} = -\frac{1}{\epsilon_0} \nabla \cdot (\alpha\mathbf{X}). \quad (\text{B5})$$

The damping in the background system is described by a reservoir of oscillators interacting with the polarizable ones. $\mathbf{Y}_{\omega}(\mathbf{r})$ is the amplitude of the oscillators with frequency ω , and $v_{\omega}(\mathbf{r})$ represents the coupling strength.

From Lagrangian (B1), the canonical momenta of the above variables are derived as

$$\mathbf{\Pi} \equiv \frac{\partial L}{\partial \dot{\mathbf{A}}} = \epsilon_0 \dot{\mathbf{A}}, \quad (\text{B6a})$$

$$\mathbf{P} \equiv \frac{\partial L}{\partial \dot{\mathbf{X}}} = \rho \dot{\mathbf{X}} - \alpha \mathbf{A}, \quad (\text{B6b})$$

$$\mathbf{Q}_{\omega} \equiv \frac{\partial L}{\partial \dot{\mathbf{Y}}_{\omega}} = \rho \dot{\mathbf{Y}}_{\omega} - v_{\omega} \mathbf{X}, \quad (\text{B6c})$$

$$\mathbf{p}_i \equiv \frac{\partial L}{\partial \dot{\mathbf{r}}_i} = m_i \dot{\mathbf{r}}_i + q_i \mathbf{A}(\mathbf{r}_i). \quad (\text{B6d})$$

Since \mathbf{A} and $\nabla(\phi_{\text{bg}} + \phi_{\text{cp}})$ are perpendicular to each other, from the Poisson equation (B5) and $\nabla^2\phi_{\text{cp}} = -\rho_{\text{cp}}/\epsilon_0$, the first term of Eq. (B2) is rewritten as

$$\int d\mathbf{r} \frac{\epsilon_0}{2} \mathbf{E}^2 = \int d\mathbf{r} \left[\frac{\mathbf{\Pi}^2}{2\epsilon_0} + \frac{1}{2} (\phi_{\text{bg}} + \phi_{\text{cp}})(\rho_{\text{bg}} + \rho_{\text{cp}}) \right]. \quad (\text{B7})$$

After a straightforward calculation, we obtain the Hamiltonian as

$$H = H_{\text{em}} + \sum_i \left[\frac{1}{2m_i} \{ \mathbf{p}_i - q_i \mathbf{A}(\mathbf{r}_i) \}^2 + V(\mathbf{r}_i) \right] + \int d\mathbf{r} \left[\frac{1}{2} \phi_{\text{cp}} \rho_{\text{cp}} + \phi_{\text{bg}} \rho_{\text{cp}} \right], \quad (\text{B8})$$

where H_{em} describes the complete Hamiltonian discussed by SW, representing the radiation field and background dielectrics with local susceptibility:

$$H_{\text{em}} = \int d\mathbf{r} \left[\frac{\mathbf{\Pi}^2}{2\epsilon_0} + \frac{1}{2\mu_0} (\nabla \times \mathbf{A})^2 + \frac{\mathbf{P}^2}{2\rho} + \frac{\rho \tilde{\omega}_0^2}{2} \mathbf{X}^2 + \int_0^\infty d\omega \frac{\mathbf{Q}_\omega^2}{2\rho} + \int_0^\infty d\omega \frac{\rho \omega^2}{2} \mathbf{Y}_\omega^2 + \frac{\alpha}{\rho} \mathbf{P} \cdot \mathbf{A} + \frac{\alpha^2}{2\rho} \mathbf{A}^2 + \int_0^\infty d\omega \frac{v_\omega}{\rho} \mathbf{X} \cdot \mathbf{Q}_\omega + \frac{1}{2} \phi_{\text{bg}} \rho_{\text{bg}} \right]. \quad (\text{B9})$$

The first two terms represent the radiation energy, the third term is the kinetic energy of the oscillators, and the fourth is the potential. The seventh and eighth terms represent the interaction between the oscillators and the radiation field. The eigenfrequency of the oscillators shown in the fourth term of Eq. (B9) is modified as

$$\tilde{\omega}_0^2 \equiv \omega_0^2 + \frac{1}{\rho} \int_0^\infty d\omega v_\omega^2 \quad (\text{B10})$$

by the interaction with the reservoir oscillators, which is described as the ninth term. The energy of reservoir is the fifth and sixth terms. The last term is the Coulomb interaction between the induced charges of backgrounds.

The kinetic energy of the charged particles, the second term of Eq. (B8), is expanded with the expression (B6d) of their momentum as

$$\begin{aligned} & \sum_i \frac{1}{2m_i} [\mathbf{p}_i - q_i \mathbf{A}(\mathbf{r}_i)]^2 \\ &= \sum_i \frac{1}{2m_i} \mathbf{p}_i^2 - \sum_i \frac{q_i}{2m_i} [\mathbf{p}_i \cdot \mathbf{A}(\mathbf{r}_i) + \mathbf{A}(\mathbf{r}_i) \cdot \mathbf{p}_i] \\ & \quad + \sum_i \frac{q_i^2}{2m_i} \mathbf{A}^2(\mathbf{r}_i). \end{aligned} \quad (\text{B11})$$

The first term is the kinetic energy without the radiation contribution, and the other terms represent the interaction between the charged particles and the radiation field. Here, using the variables defined in Eqs. (A4) and (A5), we can rewrite Hamiltonian (B8) as

$$H = H_{\text{em}} + \sum_i \left[\frac{1}{2m_i} \mathbf{p}_i^2 + V(\mathbf{r}_i) \right] + \frac{1}{2} \int d\mathbf{r} \phi_{\text{cp}} \rho_{\text{cp}} + \int d\mathbf{r} \phi_{\text{bg}} \rho_{\text{cp}} - \int d\mathbf{r} \left[\mathbf{I}_{\text{cp}} \cdot \mathbf{A} - \frac{1}{2} N_{\text{cp}} \mathbf{A}^2 \right] \quad (\text{B12})$$

Expanding these terms with field operator (A7), we obtain the first tree terms of interaction Hamiltonian (15)

from the exciton-associated components of the last three terms of Eq. (B12), i.e., the terms proportional to $a_v^\dagger a_c$ or $a_c^\dagger a_v$, but not to $a_c^\dagger a_{c'}$ or $a_v^\dagger a_{v'}$, which are negligible under the weak excitation regime. On the other hand, as mentioned in Sec. IV, we put the exchange interaction between electrons and holes into H_{int} . It is obtained by expanding the fourth term of (B12):

$$\begin{aligned} & \frac{1}{2} \int d\mathbf{r} \phi_{\text{cp}} \rho_{\text{cp}} \rightarrow \dots \\ & + \sum_{c,v,c',v'} a_c^\dagger a_v a_{v'}^\dagger a_{c'} \int d\mathbf{r} d\mathbf{r}' \frac{e^2 \varphi_c^*(\mathbf{r}) \varphi_v(\mathbf{r}) \varphi_{v'}^*(\mathbf{r}') \varphi_{c'}(\mathbf{r}')}{4\pi\epsilon_0 |\mathbf{r} - \mathbf{r}'|}. \end{aligned} \quad (\text{B13})$$

When we use the RWA and assume commutation relations (18) of the exciton operators, we can find that the Coulomb interaction between the excitonic charges themselves

$$\frac{1}{2} \int d\mathbf{r} d\mathbf{r}' \frac{\rho_{\text{ex}}(\mathbf{r}) \rho_{\text{ex}}(\mathbf{r}')}{4\pi\epsilon_0 |\mathbf{r} - \mathbf{r}'|} \quad (\text{B14})$$

gives exchange expression (B13) with a constant energy term by expanding $\rho_{\text{ex}}(\mathbf{r})$ using Eq. (A10). While all the other terms (\dots) in Eq. (B13) and the second and third terms of Eq. (B12) belong to H_{mat} , instead of discussing them in detail, we treat the matter Hamiltonian as Eq. (17) for a simple description of the linear optical process of excitons with nonradiative relaxation.

APPENDIX C: EXTENSION OF MAXWELL WAVE EQUATION

Here, we extend the Maxwell wave equation discussed by SW to enable the consideration of the exciton-induced polarization with nonlocal susceptibility. We derive the Heisenberg equations of the system variables in Sec. C 1, calculate their Laplace-transform in Sec. C 2, and provide the Maxwell wave equation for the electric field in Sec. C 3.

1. Heisenberg equations

We derive the Heisenberg equations of the system variables and momenta from background Hamiltonian (B9) and interaction terms (15). The commutation relations of the variables are

$$[\mathbf{A}(\mathbf{r}), \mathbf{\Pi}(\mathbf{r}')] = i\hbar \delta_{\text{T}}(\mathbf{r} - \mathbf{r}'), \quad (\text{C1})$$

$$[\mathbf{X}(\mathbf{r}), \mathbf{P}(\mathbf{r}')] = i\hbar \delta(\mathbf{r} - \mathbf{r}') \mathbf{1}, \quad (\text{C2})$$

$$[\mathbf{Y}_\omega(\mathbf{r}), \mathbf{Q}_{\omega'}(\mathbf{r}')] = i\hbar \delta(\omega - \omega') \delta(\mathbf{r} - \mathbf{r}') \mathbf{1}, \quad (\text{C3})$$

where

$$\delta_{\text{T}}(\mathbf{r} - \mathbf{r}') \equiv \mathbf{1} \delta(\mathbf{r} - \mathbf{r}') + \frac{\nabla' \nabla'}{4\pi |\mathbf{r} - \mathbf{r}'|} \quad (\text{C4})$$

is the Dirac delta function extracting the transverse component. We obtain the equations of the radiation field as

$$\dot{\mathbf{A}} = \frac{1}{\epsilon_0} \mathbf{\Pi}, \quad (\text{C5})$$

$$\ddot{\mathbf{\Pi}} = \frac{1}{\mu_0} \nabla^2 \mathbf{A} - \left[\frac{\alpha}{\rho} (\mathbf{P} + \alpha \mathbf{A}) \right]_{\text{T}} + \mathbf{J}_{\text{exT}}, \quad (\text{C6})$$

where

$$\mathbf{J}_{\text{exT}}(\mathbf{r}) \equiv \int d\mathbf{r}' \delta_{\text{T}}(\mathbf{r} - \mathbf{r}') \cdot \mathbf{J}_{\text{ex}}(\mathbf{r}') \quad (\text{C7})$$

is the transverse component of the current density (A19). The equations of the polarizable oscillators are

$$\dot{\mathbf{X}} = \frac{1}{\rho} (\mathbf{P} + \alpha \mathbf{A}), \quad (\text{C8})$$

$$\dot{\mathbf{P}} = -\rho \tilde{\omega}_0^2 \mathbf{X} - \frac{\alpha}{\epsilon_0} [\alpha \mathbf{X}]_{\text{L}} - \frac{1}{\rho} \int_0^\infty d\omega v_\omega \mathbf{Q}_\omega + \alpha \nabla \phi_{\text{ex}}, \quad (\text{C9})$$

and those of the reservoir oscillators are obtained as

$$\dot{\mathbf{Y}}_\omega = \frac{1}{\rho} (\mathbf{Q}_\omega + v_\omega \mathbf{X}), \quad (\text{C10})$$

$$\dot{\mathbf{Q}}_\omega = -\rho \omega^2 \mathbf{Y}_\omega. \quad (\text{C11})$$

From Eqs. (C5), (C6), and (C8), we obtain the Maxwell wave equation for the vector potential

$$\nabla^2 \mathbf{A} - \frac{1}{c^2} \ddot{\mathbf{A}} = \mu_0 \left[\alpha \dot{\mathbf{X}} \right]_{\text{T}} - \mu_0 \mathbf{J}_{\text{exT}}, \quad (\text{C12})$$

which has the transverse component of the excitonic current density compared to the same kind of equation in Ref. 7. Using a relation between the longitudinal components of excitonic variables

$$\mathbf{J}_{\text{exL}}(\mathbf{r}) = \dot{\mathbf{P}}_{\text{exL}}(\mathbf{r}) = \epsilon_0 \nabla \dot{\phi}_{\text{ex}}(\mathbf{r}) \quad (\text{C13})$$

and that for the polarizable oscillators (B4), we can rewrite Eq. (C12) as a wave equation for electric field (20):

$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{c^2} \ddot{\mathbf{E}} = \mu_0 \alpha \ddot{\mathbf{X}} - \mu_0 \dot{\mathbf{J}}_{\text{ex}}. \quad (\text{C14})$$

On the other hand, from Eqs. (C8) and (C9), we obtain the differential equation of the polarizable oscillators:

$$\rho \ddot{\mathbf{X}} + \rho \tilde{\omega}_0^2 \mathbf{X} = \alpha \dot{\mathbf{A}} - \frac{\alpha}{\epsilon_0} [\alpha \mathbf{X}]_{\text{L}} + \alpha \nabla \phi_{\text{ex}} - \frac{1}{\rho} \int_0^\infty d\omega v_\omega \mathbf{Q}_\omega. \quad (\text{C15})$$

2. Laplace transform

Next, we rewrite the equations of motion derived in the previous section to those for the forward Laplace transform of the variables

$$\bar{\Omega}(p) \equiv \int_0^\infty dt e^{-pt} \Omega(t), \quad (\text{C16a})$$

and for the backward Laplace transform

$$\check{\Omega}(p) \equiv \int_0^\infty dt e^{-pt} \Omega(-t). \quad (\text{C16b})$$

From these motion equations, we derive the ones for positive-frequency Fourier transform

$$\hat{\Omega}^+(\omega) = \frac{1}{2\pi} [\bar{\Omega}(-i\omega + \delta) + \check{\Omega}(i\omega + \delta)]. \quad (\text{C17})$$

From the forward Laplace transform of Eqs. (C10) and (C11) for the reservoir oscillators, we obtain

$$\begin{aligned} \bar{\mathbf{Q}}_\omega(p) &= -\frac{\omega^2}{p^2 + \omega^2} v_\omega \bar{\mathbf{X}}(p) \\ &+ \frac{1}{p^2 + \omega^2} [p \mathbf{Q}_\omega(0) - \rho \omega^2 \mathbf{Y}_\omega(0)], \end{aligned} \quad (\text{C18})$$

and, by using Eq. (C8) at $t = 0$, that for the polarizable oscillators (C15) becomes

$$\begin{aligned} &(p^2 + \tilde{\omega}_0^2) \bar{\mathbf{X}}(p) \\ &= \frac{\alpha}{\rho} \left\{ p \bar{\mathbf{A}}(p) - \frac{1}{\epsilon_0} [\alpha \bar{\mathbf{X}}(p)]_{\text{L}} + \nabla \bar{\phi}_{\text{ex}}(p) \right\} \\ &- \frac{1}{\rho^2} \int_0^\infty d\omega v_\omega \bar{\mathbf{Q}}_\omega(p) + p \mathbf{X}(0) + \frac{1}{\rho} \mathbf{P}(0). \end{aligned} \quad (\text{C19})$$

Substituting the former into the latter and using the forward Laplace-transform of the electric field

$$\bar{\mathbf{E}}(p) = -p \bar{\mathbf{A}}(p) + \frac{1}{\epsilon_0} [\alpha \bar{\mathbf{X}}(p)]_{\text{L}} - \nabla \bar{\phi}_{\text{ex}}(p) + \mathbf{A}(0), \quad (\text{C20})$$

we obtain the same equation as Eq. (20) of Ref. 7:

$$\begin{aligned} \bar{\mathbf{X}}(p) &= -\frac{\epsilon_0}{\alpha} \bar{\chi}(p) \bar{\mathbf{E}}(p) + \frac{\epsilon_0}{\alpha^2} \bar{\chi}(p) \left\{ \alpha \mathbf{A}(0) + \rho p \mathbf{X}(0) \right. \\ &\left. + \mathbf{P}(0) + \int_0^\infty d\omega \frac{v_\omega}{p^2 + \omega^2} \left[\omega^2 \mathbf{Y}_\omega(0) - \frac{p}{\rho} \mathbf{Q}_\omega(0) \right] \right\}, \end{aligned} \quad (\text{C21})$$

where

$$\bar{\chi}(p) = \frac{\alpha^2}{\epsilon_0 \rho} \left[p^2 + \tilde{\omega}_0^2 - \frac{1}{\rho^2} \int_0^\infty d\omega \frac{\omega^2 v_\omega^2}{p^2 + \omega^2} \right]^{-1} \quad (\text{C22})$$

is the background susceptibility.

Next, we derive the forward Laplace transform of Maxwell wave equation (C14) for the electric field as

$$\begin{aligned} &\nabla \times \nabla \times \bar{\mathbf{E}}(p) + \frac{p^2}{c^2} \bar{\mathbf{E}}(p) - \mu_0 \alpha p^2 \bar{\mathbf{X}}(p) \\ &= -\mu_0 p \bar{\mathbf{J}}_{\text{ex}}(p) + \mu_0 \mathbf{J}_{\text{ex}}(0) + \frac{1}{c^2} \dot{\mathbf{E}}(0) + \frac{p}{c^2} \mathbf{E}(0) \\ &- \mu_0 \alpha \dot{\mathbf{X}}(0) - \mu_0 \alpha p \mathbf{X}(0). \end{aligned} \quad (\text{C23})$$

Here, using the Eqs. (20), (B4), (C5), (C8), (C12), and (C13), we obtain the relations between the variables at $t = 0$ as

$$\dot{\mathbf{X}}(0) = \frac{\alpha}{\rho} \mathbf{A}(0) + \frac{1}{\rho} \mathbf{P}(0), \quad (\text{C24})$$

$$\dot{\mathbf{E}}(0) = c^2 \nabla \times \nabla \times \mathbf{A}(0) + \frac{\alpha^2}{\epsilon_0 \rho} \mathbf{A}(0) + \frac{\alpha}{\epsilon_0 \rho} \mathbf{P}(0) - \frac{1}{\epsilon_0} \mathbf{J}_{\text{ex}}(0), \quad (\text{C25})$$

$$\mathbf{E}(0) = -\frac{1}{\epsilon_0} \mathbf{\Pi}(0) + \frac{1}{\epsilon_0} [\alpha \mathbf{X}(0)]_{\text{L}} - \nabla \phi_{\text{ex}}(0). \quad (\text{C26})$$

Using these relations, we can rewrite the Maxwell wave equation (C23) to

$$\begin{aligned} \nabla \times \nabla \times \bar{\mathbf{E}}(p) + \frac{p^2}{c^2} \bar{\epsilon}(p) \bar{\mathbf{E}}(p) \\ = -\mu_0 p \bar{\mathbf{J}}(p) - \mu_0 p \bar{\mathbf{J}}_{\text{ex}}(p) - \frac{p}{c^2} \nabla \phi_{\text{ex}}(0), \end{aligned} \quad (\text{C27})$$

where $\bar{\epsilon}(p) = 1 + \bar{\chi}(p)$ is the background dielectric function, and the operator

$$\begin{aligned} \bar{\mathbf{J}}(p) = & -\frac{1}{\mu_0 p} \nabla \times \nabla \times \mathbf{A}(0) - \epsilon_0 p \bar{\chi}(p) \mathbf{A}(0) + \mathbf{\Pi}(0) \\ & + \alpha \left[1 - \frac{\epsilon_0 \rho}{\alpha^2} p^2 \bar{\chi}(p) \right] \mathbf{X}(0) - [\alpha \mathbf{X}(0)]_{\text{L}} - \frac{\epsilon_0}{\alpha} p \bar{\chi}(p) \mathbf{P}(0) \\ & - \frac{\epsilon_0}{\alpha} p \bar{\chi}(p) \int_0^\infty d\omega \frac{v_\omega}{p^2 + \omega^2} \left[\omega^2 \mathbf{Y}_\omega(0) - \frac{p}{\rho} \mathbf{Q}_\omega(0) \right] \end{aligned} \quad (\text{C28})$$

is the same one shown in Eq. (27) of Ref. 7. This operator depends only on the background system variables and momenta of Ref. 7.

On the other hand, the backward Laplace transform of the Maxwell wave equation is obtained as

$$\begin{aligned} \nabla \times \nabla \times \check{\mathbf{E}}(p) + \frac{p^2}{c^2} \bar{\epsilon}(p) \check{\mathbf{E}}(p) \\ = \mu_0 p \check{\mathbf{J}}(p) + \mu_0 p \check{\mathbf{J}}_{\text{ex}}(p) - \frac{p}{c^2} \nabla \phi_{\text{ex}}(0), \end{aligned} \quad (\text{C29})$$

where the operator on the RHS

$$\begin{aligned} \check{\mathbf{J}}(p) = & -\frac{1}{\mu_0 p} \nabla \times \nabla \times \mathbf{A}(0) - \epsilon_0 p \bar{\chi}(p) \mathbf{A}(0) - \mathbf{\Pi}(0) \\ & - \alpha \left[1 - \frac{\epsilon_0 \rho}{\alpha^2} p^2 \bar{\chi}(p) \right] \mathbf{X}(0) + [\alpha \mathbf{X}(0)]_{\text{L}} - \frac{\epsilon_0}{\alpha} p \bar{\chi}(p) \mathbf{P}(0) \\ & - \frac{\epsilon_0}{\alpha} p \bar{\chi}(p) \int_0^\infty d\omega \frac{v_\omega}{p^2 + \omega^2} \left[\omega^2 \mathbf{Y}_\omega(0) + \frac{p}{\rho} \mathbf{Q}_\omega(0) \right] \end{aligned} \quad (\text{C30})$$

is also independent from the variables associated with excitons.

3. Fourier transform

From the forward and backward Laplace transforms (C27) and (C29) of the Maxwell wave equation, we obtain that for the positive-frequency Fourier component of the electric field as

$$\begin{aligned} \nabla \times \nabla \times \hat{\mathbf{E}}^+(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \hat{\mathbf{E}}^+(\mathbf{r}, \omega) \\ = i\mu_0 \omega \left[\hat{\mathbf{J}}_0(\mathbf{r}, \omega) + \hat{\mathbf{J}}_{\text{ex}}^+(\mathbf{r}, \omega) \right], \end{aligned} \quad (\text{C31})$$

where $\epsilon_{\text{bg}}(\mathbf{r}, \omega) = \bar{\epsilon}(\mathbf{r}, -i\omega + \delta)$ is the background dielectric function. The noise current density operator in our system is written as

$$\begin{aligned} \hat{\mathbf{J}}_0(\mathbf{r}, \omega) = \hat{\mathbf{J}}(\mathbf{r}, \omega) - \frac{i\omega^2}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \\ \times \int d\mathbf{r}' \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \cdot \left[\check{\mathbf{J}}_{\text{ex}}(\mathbf{r}', i\omega + \delta) - \epsilon_0 \nabla \phi_{\text{ex}}(\mathbf{r}', 0) \right], \end{aligned} \quad (\text{C32})$$

where $\mathbf{G}(\mathbf{r}, \mathbf{r}, \omega)$ is Green's tensor satisfying Eq. (8), and

$$\begin{aligned} \hat{\mathbf{J}}(\mathbf{r}, \omega) = \frac{1}{2\pi} \left[\bar{\mathbf{J}}(\mathbf{r}, -i\omega + \delta) + \check{\mathbf{J}}(\mathbf{r}, i\omega + \delta) \right] \\ - \frac{i\omega^2}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \int d\mathbf{r}' \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \cdot \check{\mathbf{J}}(\mathbf{r}', i\omega + \delta) \end{aligned} \quad (\text{C33})$$

is the same operator shown in Eq. (45) of Ref. 7 satisfying the commutation relations (7). By using the relation

$$\hat{\mathbf{J}}_{\text{ex}}(\mathbf{r}, \omega) = -i\omega \hat{\mathbf{P}}_{\text{ex}}(\mathbf{r}, \omega), \quad (\text{C34})$$

we can rewrite Eq. (C31) to (21).

APPENDIX D: EVALUATION OF COMMUTATORS

Although we derive the Maxwell wave equation considering the excitons in App. C, there exists a problem in deriving the commutation relations of noise current density $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ and in describing the optical processes of excitons. These are the subjects of this appendix. In Sec. D 1, we derive the self-consistent equations for the Laplace-transformed operators, and we evaluate the commutators of the noise current density in Sec. D 2. In Sec. D 3, we consider nonradiative relaxation of excitons.

1. Laplace transform of self-consistent equations

From matter Hamiltonian (17) and interaction Hamiltonian (15), neglecting the radiation contribution of the current density $N_{\text{ex}}(\mathbf{r}) \mathbf{A}^2(\mathbf{r})/2$ under weak excitation, we obtain the Heisenberg equation of excitons as

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} b_\mu(t) \\ = \hbar \omega_\mu b_\mu(t) - \int d\mathbf{r} \left[\mathcal{I}_\mu^*(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}, t) - \rho_\mu^*(\mathbf{r}) \phi(\mathbf{r}, t) \right] \\ + \int_0^\infty d\Omega \left[g_\mu(\Omega) d_\mu(\Omega, t) + g_\mu^*(\Omega) d_\mu^\dagger(\Omega, t) \right], \end{aligned} \quad (\text{D1})$$

where $\phi(\mathbf{r}) \equiv \phi_{\text{bg}}(\mathbf{r}) + \phi_{\text{ex}}(\mathbf{r})$ is the complete Coulomb potential. In this section, we neglect nonradiative relaxation and assume $\omega \sim \omega_\mu$. Using the relations between

$\mathcal{I}_\mu(\mathbf{r})$, $\rho_\mu(\mathbf{r})$, and $\mathcal{P}_\mu(\mathbf{r})$ (Eqs. (A16) and (A18)) and the Laplace transform of the electric field

$$\bar{\mathbf{E}}(\mathbf{r}, p) = -p\bar{\mathbf{A}}(\mathbf{r}, p) - \nabla\bar{\phi}(\mathbf{r}, p) + \mathbf{A}(\mathbf{r}, 0), \quad (\text{D2a})$$

$$\check{\mathbf{E}}(\mathbf{r}, p) = p\check{\mathbf{A}}(\mathbf{r}, p) - \nabla\check{\phi}(\mathbf{r}, p) - \mathbf{A}(\mathbf{r}, 0), \quad (\text{D2b})$$

the forward and backward Laplace transforms of Eq. (D1) are respectively derived as

$$\begin{aligned} (\hbar\omega_\mu - \hbar\omega - i\delta) \bar{b}_\mu(-i\omega + \delta) &= -i\hbar b_\mu(0) \\ &+ \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot [\bar{\mathbf{E}}(\mathbf{r}, -i\omega + \delta) - \mathbf{A}(\mathbf{r}, 0)], \end{aligned} \quad (\text{D3a})$$

$$\begin{aligned} (\hbar\omega_\mu - \hbar\omega + i\delta) \check{b}_\mu(i\omega + \delta) &= i\hbar b_\mu(0) \\ &+ \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot [\check{\mathbf{E}}(\mathbf{r}, i\omega + \delta) + \mathbf{A}(\mathbf{r}, 0)]. \end{aligned} \quad (\text{D3b})$$

Adding these two equations, we obtain Eq. (28), the Fourier transform of the excitons' motion equation. On the other hand, using the relations

$$\epsilon_0 \nabla \phi_{\text{ex}}(\mathbf{r}) = \mathbf{P}_{\text{exL}}(\mathbf{r}), \quad (\text{D4})$$

and

$$\bar{\mathbf{J}}_{\text{ex}}(\mathbf{r}, p) = p\bar{\mathbf{P}}_{\text{ex}}(\mathbf{r}, p) - \mathbf{P}_{\text{ex}}(\mathbf{r}, 0), \quad (\text{D5a})$$

$$\check{\mathbf{J}}_{\text{ex}}(\mathbf{r}, p) = -p\check{\mathbf{P}}_{\text{ex}}(\mathbf{r}, p) + \mathbf{P}_{\text{ex}}(\mathbf{r}, 0), \quad (\text{D5b})$$

the Laplace transforms (C27) and (C29) of the Maxwell wave equation are rewritten as

$$\begin{aligned} \nabla \times \nabla \times \bar{\mathbf{E}}(\mathbf{r}, -i\omega + \delta) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}(\mathbf{r}, \omega) \bar{\mathbf{E}}(\mathbf{r}, -i\omega + \delta) \\ = i\mu_0\omega [\bar{\mathbf{J}}(\mathbf{r}, -i\omega + \delta) - \mathbf{P}_{\text{exT}}(\mathbf{r}, 0)] \\ + \mu_0\omega^2 \bar{\mathbf{P}}_{\text{ex}}(\mathbf{r}, -i\omega + \delta), \end{aligned} \quad (\text{D6a})$$

$$\begin{aligned} \nabla \times \nabla \times \check{\mathbf{E}}(\mathbf{r}, i\omega + \delta) - \frac{\omega^2}{c^2} \epsilon_{\text{bg}}^*(\mathbf{r}, \omega) \check{\mathbf{E}}(\mathbf{r}, i\omega + \delta) \\ = i\mu_0\omega [\check{\mathbf{J}}(\mathbf{r}, i\omega + \delta) + \mathbf{P}_{\text{exT}}(\mathbf{r}, 0)] \\ + \mu_0\omega^2 \check{\mathbf{P}}_{\text{ex}}(\mathbf{r}, i\omega + \delta). \end{aligned} \quad (\text{D6b})$$

Here, under the RWA, the Laplace transforms of the polarization density operator can be written as

$$\bar{\mathbf{P}}_{\text{ex}}(\mathbf{r}, -i\omega + \delta) \sim \sum_\mu \mathcal{P}_\mu(\mathbf{r}) \bar{b}_\mu(-i\omega + \delta), \quad (\text{D7a})$$

$$\check{\mathbf{P}}_{\text{ex}}(\mathbf{r}, i\omega + \delta) \sim \sum_\mu \mathcal{P}_\mu(\mathbf{r}) \check{b}_\mu(i\omega + \delta) \quad (\text{D7b})$$

in the same manner as its Fourier transform (29). Substituting Maxwell wave equations (D6) in motion equations (D3) of excitons, we obtain the self-consistent equation set for the Laplace transformed operators:

$$\begin{aligned} \sum_{\mu'} [(\hbar\omega_\mu - \hbar\omega)\delta_{\mu,\mu'} + \mathcal{A}_{\mu,\mu'}(\omega)] \bar{b}_{\mu'}(-i\omega + \delta) \\ = \bar{\beta}_\mu(-i\omega + \delta), \end{aligned} \quad (\text{D8a})$$

$$\begin{aligned} \sum_{\mu'} [(\hbar\omega_\mu - \hbar\omega)\delta_{\mu,\mu'} + \mathcal{A}_{\mu',\mu}^*(\omega)] \check{b}_{\mu'}(i\omega + \delta) \\ = \check{\beta}_\mu(i\omega + \delta), \end{aligned} \quad (\text{D8b})$$

where the operators on the RHS are

$$\begin{aligned} \bar{\beta}_\mu(-i\omega + \delta) \equiv i\mu_0\omega \int d\mathbf{r} \int d\mathbf{r}' \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \\ \cdot [\bar{\mathbf{J}}(\mathbf{r}', -i\omega + \delta) - \mathbf{P}_{\text{exT}}(\mathbf{r}', 0)] \\ - i\hbar b_\mu(0) - \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}, 0), \end{aligned} \quad (\text{D9a})$$

$$\begin{aligned} \check{\beta}_\mu(i\omega + \delta) \equiv i\mu_0\omega \int d\mathbf{r} \int d\mathbf{r}' \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \\ \cdot [\check{\mathbf{J}}(\mathbf{r}', i\omega + \delta) + \mathbf{P}_{\text{exT}}(\mathbf{r}', 0)] \\ + i\hbar b_\mu(0) + \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}, 0). \end{aligned} \quad (\text{D9b})$$

In order to derive (D8b), we used the representation

$$\mathcal{A}_{\mu',\mu}^*(\omega) = -\mu\omega^2 \int d\mathbf{r} \int d\mathbf{r}' \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathcal{P}_{\mu'}(\mathbf{r}'), \quad (\text{D10})$$

which comes from definition (35) and the reciprocity relation

$$G_{ij}(\mathbf{r}, \mathbf{r}', \omega) = G_{ji}(\mathbf{r}', \mathbf{r}, \omega) \quad (\text{D11})$$

shown in Eq. (1.53) of Ref. 13. Adding self-consistent equation sets (D8), we obtain the one for the Fourier transform of the exciton operators as

$$\sum_{\mu'} [(\hbar\omega_\mu - \hbar\omega)\delta_{\mu,\mu'} + \mathcal{A}_{\mu,\mu'}(\omega)] \hat{b}_{\mu'}(\omega) = \hat{\beta}_\mu(\omega), \quad (\text{D12})$$

where the operator on the RHS is

$$\begin{aligned} \hat{\beta}_\mu(\omega) \equiv \frac{1}{2\pi} \bar{\beta}_\mu(-i\omega + \delta) + \frac{1}{2\pi} \check{\beta}_\mu(i\omega + \delta) \\ + \frac{1}{2\pi} \sum_{\mu'} [\mathcal{A}_{\mu,\mu'}(\omega) - \mathcal{A}_{\mu',\mu}^*(\omega)] \check{b}_{\mu'}(i\omega + \delta). \end{aligned} \quad (\text{D13})$$

Now, we verify that this operator is equivalent to the RHS of Eq. (37). Using Eqs. (C32), (D4), (D5), and (11), the background electric field (24) is rewritten as

$$\begin{aligned} \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) = i\mu_0\omega \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{J}}(\mathbf{r}', \omega) \\ - \frac{\mu_0}{2\pi} \int d\mathbf{r}' [\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega)] \\ \cdot [\omega^2 \check{\mathbf{P}}_{\text{ex}}(\mathbf{r}', i\omega + \delta) + i\omega \mathbf{P}_{\text{exT}}(\mathbf{r}', 0)]. \end{aligned} \quad (\text{D14})$$

Since, from Eqs. (C33) and (11), we can rewrite the first term as

$$\begin{aligned} & \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{J}}(\mathbf{r}', \omega) \\ &= \frac{1}{2\pi} \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \bar{\mathbf{J}}(\mathbf{r}', -i\omega + \delta) \\ & \quad + \frac{1}{2\pi} \int d\mathbf{r}' \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \cdot \check{\mathbf{J}}(\mathbf{r}', i\omega + \delta), \end{aligned} \quad (\text{D15})$$

background field (D14) can be rewritten as

$$\begin{aligned} \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) &= \frac{i\mu_0\omega}{2\pi} \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot [\bar{\mathbf{J}}(\mathbf{r}', -i\omega + \delta) - \mathbf{P}_{\text{exT}}(\mathbf{r}', 0)] \\ & \quad + \frac{i\mu_0\omega}{2\pi} \int d\mathbf{r}' \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \cdot [\check{\mathbf{J}}(\mathbf{r}', i\omega + \delta) + \mathbf{P}_{\text{exT}}(\mathbf{r}', 0)] \\ & \quad - \frac{\mu_0\omega^2}{2\pi} \int d\mathbf{r}' [\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega)] \cdot \check{\mathbf{P}}_{\text{ex}}(\mathbf{r}', i\omega + \delta), \end{aligned} \quad (\text{D16})$$

and the RHS of Eq. (37) becomes

$$\begin{aligned} \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) &= \frac{i\mu_0\omega}{2\pi} \int d\mathbf{r} \int d\mathbf{r}' \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot [\bar{\mathbf{J}}(\mathbf{r}', -i\omega + \delta) - \mathbf{P}_{\text{exT}}(\mathbf{r}', 0)] \\ & \quad + \frac{i\mu_0\omega}{2\pi} \int d\mathbf{r} \int d\mathbf{r}' \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \cdot [\check{\mathbf{J}}(\mathbf{r}', i\omega + \delta) + \mathbf{P}_{\text{exT}}(\mathbf{r}', 0)] \\ & \quad + \frac{1}{2\pi} \sum_{\mu'} [\mathcal{A}_{\mu, \mu'}(\omega) - \mathcal{A}_{\mu', \mu}^*(\omega)] \check{b}_{\mu'}(i\omega + \delta). \end{aligned} \quad (\text{D17})$$

We can find that this is equivalent to (D13) by expanding $\bar{\beta}_\mu(-i\omega + \delta)$ and $\check{\beta}_\mu(i\omega + \delta)$ with Eqs. (D9).

2. Commutation relations

In the representation of background field (D14) or (D16), the information of excitons during $t < 0$ is reflected via operator $\check{\mathbf{P}}_{\text{ex}}(\mathbf{r}, i\omega + \delta)$. In order to evaluate the commutation relations of noise current density $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$, we first discuss the motion equations of excitons.

The exciton motion during $t < 0$ is described by Eq. (D8b) with (D9b). Using $\mathbf{W}(\omega)$, the inverse matrix of coefficient matrix $\mathbf{S}(\omega)$ defined in Eq. (36), we rewrite Eq. (D8b) to

$$\check{b}_\mu(i\omega + \delta) = \sum_{\mu'} W_{\mu', \mu}^*(\omega) \check{\beta}_{\mu'}(i\omega + \delta). \quad (\text{D18})$$

First of all, we evaluate the cummutation relations of $\check{\beta}_\mu(i\omega + \delta)$. Since, from Eqs. (C30) and (26), we obtain the relations

$$[\check{\mathbf{J}}(\mathbf{r}, i\omega + \delta), \mathbf{A}(\mathbf{r}', 0)] = i\hbar\delta_{\text{T}}(\mathbf{r} - \mathbf{r}'), \quad (\text{D19})$$

$$[\mathbf{P}_{\text{exT}}(\mathbf{r}, 0), b_\mu(0)] = -[\mathcal{P}_\mu^*(\mathbf{r})]_{\text{T}}, \quad (\text{D20})$$

the first two terms and the following terms of (D9b) are commutable as

$$\begin{aligned} & [\check{\mathbf{J}}(\mathbf{r}, i\omega + \delta) + \mathbf{P}_{\text{exT}}(\mathbf{r}, 0), \\ & \quad i\hbar b_\mu(0) + \int d\mathbf{r}' \mathcal{P}_\mu^*(\mathbf{r}') \cdot \mathbf{A}(\mathbf{r}', 0)] = \mathbf{0}. \end{aligned} \quad (\text{D21})$$

Then, since $\mathbf{P}_{\text{exT}}(\mathbf{r}, 0)$ and $\mathbf{A}(\mathbf{r}, 0)$ are commutable with themselves, we obtain the expression to be evaluated:

$$\begin{aligned} & [\check{\beta}_\mu(i\omega + \delta), \{\check{\beta}_\nu(i\omega' + \delta')\}^\dagger] = \hbar^2\delta_{\mu, \nu} \\ & \quad + \mu_0^2\omega\omega' \int d\mathbf{r} \int d\mathbf{s} \int d\mathbf{s}' \int d\mathbf{r}' \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{G}^*(\mathbf{r}, \mathbf{s}, \omega) \\ & \quad \cdot [\check{\mathbf{J}}(\mathbf{s}, i\omega + \delta), \{\check{\mathbf{J}}(\mathbf{s}', i\omega' + \delta')\}^\dagger] \cdot \mathbf{G}(\mathbf{s}', \mathbf{r}', \omega') \cdot \mathcal{P}_\nu(\mathbf{r}'). \end{aligned} \quad (\text{D22})$$

Using the relation shown in Eq. (B1) of Ref. 7

$$\begin{aligned} & [\check{\mathbf{J}}(\mathbf{s}, i\omega + \delta), \{\check{\mathbf{J}}(\mathbf{s}', i\omega' + \delta')\}^\dagger] \\ &= \frac{\hbar(\omega + \omega')}{\mu_0\omega\omega'} [\nabla\nabla - \mathbf{1}\nabla^2] \delta(\mathbf{s} - \mathbf{s}') \\ & \quad + \frac{\epsilon_0\hbar\omega\omega'}{\omega - \omega' - i\delta} [\epsilon_{\text{bg}}^*(\mathbf{s}, \omega) - \epsilon_{\text{bg}}(\mathbf{s}, \omega')] \mathbf{1}\delta(\mathbf{s} - \mathbf{s}'), \end{aligned} \quad (\text{D23})$$

we obtain

$$\begin{aligned} & \mu_0^2 \omega \omega' \int d\mathbf{s} \int d\mathbf{s}' \mathbf{G}^*(\mathbf{r}, \mathbf{s}, \omega) \\ & \cdot [\hat{\mathbf{J}}(\mathbf{s}, i\omega + \delta), \{\check{\mathbf{J}}(\mathbf{s}', i\omega' + \delta')\}^\dagger] \cdot \mathbf{G}(\mathbf{s}', \mathbf{r}', \omega') \\ & = \frac{\mu_0 \hbar}{\omega - \omega' - i\delta} [\omega^2 \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) - \omega'^2 \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega')]. \end{aligned} \quad (\text{D24})$$

From this relation, Eq. (D22) is evaluated as

$$\begin{aligned} & [\check{\beta}_\mu(i\omega + \delta), \{\check{\beta}_{\mu'}(i\omega' + \delta')\}^\dagger] \\ & = \hbar^2 \delta_{\mu, \mu'} + \frac{\hbar}{\omega - \omega' - i\delta} [\mathcal{A}_{\mu, \mu'}(\omega') - \mathcal{A}_{\mu', \mu}^*(\omega)] \end{aligned} \quad (\text{D25})$$

$$= \frac{\hbar}{\omega - \omega' - i\delta} [S_{\mu, \mu'}(\omega') - S_{\mu', \mu}^*(\omega)]. \quad (\text{D26})$$

We also obtain the relation

$$[\check{\beta}_\mu(i\omega + \delta), \check{\beta}_{\mu'}(i\omega' + \delta')] = 0, \quad (\text{D27})$$

because this commutator has a nonresonant denominator compared to (D26). Therefore, from Eqs. (D18), (D26), and (D27), we obtain the commutation relations of the backward Laplace-transformed exciton operators

$$\begin{aligned} & [\check{b}_\mu(i\omega + \delta), \{\check{b}_{\mu'}(i\omega' + \delta')\}^\dagger] \\ & = \frac{\hbar}{\omega - \omega' - i\delta} [W_{\mu', \mu}^*(\omega) - W_{\mu, \mu'}(\omega')], \end{aligned} \quad (\text{D28a})$$

$$[\check{b}_\mu(i\omega + \delta), \check{b}_{\mu'}(i\omega' + \delta')] = 0. \quad (\text{D28b})$$

On the other hand, using relations (D4) and (D5), noise current density $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ defined in Eq. (C32) is rewritten as

$$\begin{aligned} \hat{\mathbf{J}}_0(\mathbf{r}, \omega) & = \hat{\mathbf{J}}(\mathbf{r}, \omega) - \frac{\omega^3}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \int d\mathbf{r}' \mathbf{G}^*(\mathbf{r}, \mathbf{r}', \omega) \\ & \cdot \left[\check{\mathbf{P}}_{\text{ex}}(\mathbf{r}', i\omega + \delta) + \frac{i}{\omega} \mathbf{P}_{\text{exT}}(\mathbf{r}', 0) \right]. \end{aligned} \quad (\text{D29})$$

Then, the commutator with its Hermite conjugate is evaluated as

$$\begin{aligned} [\hat{\mathbf{J}}_0(\mathbf{r}, \omega), \{\hat{\mathbf{J}}_0(\mathbf{r}', \omega')\}^\dagger] & = [\hat{\mathbf{J}}(\mathbf{r}, \omega), \{\hat{\mathbf{J}}(\mathbf{r}', \omega')\}^\dagger] \\ & + \mathbf{X}_1 + \mathbf{X}_2 + \mathbf{X}_3, \end{aligned} \quad (\text{D30})$$

where

$$\begin{aligned} \mathbf{X}_1 & \equiv -\frac{\omega^3}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \\ & \times \int d\mathbf{s} \mathbf{G}^*(\mathbf{r}, \mathbf{s}, \omega) \cdot \left[\check{\mathbf{P}}_{\text{ex}}(\mathbf{s}, i\omega + \delta), \{\hat{\mathbf{J}}(\mathbf{r}', \omega')\}^\dagger \right], \end{aligned} \quad (\text{D31})$$

$$\begin{aligned} \mathbf{X}_2 & \equiv -\frac{\omega'^3}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}', \omega')] \\ & \times \int d\mathbf{s}' \left[\hat{\mathbf{J}}(\mathbf{r}, \omega), \{\check{\mathbf{P}}_{\text{ex}}(\mathbf{s}', i\omega' + \delta)\}^\dagger \right] \cdot \mathbf{G}(\mathbf{s}', \mathbf{r}', \omega'), \end{aligned} \quad (\text{D32})$$

$$\begin{aligned} \mathbf{X}_3 & \equiv \frac{\omega^3 \omega'^3}{\pi^2 c^4} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}', \omega')] \\ & \times \int d\mathbf{s} \int d\mathbf{s}' \mathbf{G}^*(\mathbf{r}, \mathbf{s}, \omega) \\ & \cdot \left[\check{\mathbf{P}}_{\text{ex}}(\mathbf{s}, i\omega + \delta) + \frac{i}{\omega} \mathbf{P}_{\text{exT}}(\mathbf{s}, 0), \right. \\ & \left. \{\check{\mathbf{P}}_{\text{ex}}(\mathbf{s}', i\omega' + \delta)\}^\dagger - \frac{i}{\omega'} \mathbf{P}_{\text{exT}}(\mathbf{s}', 0) \right] \cdot \mathbf{G}(\mathbf{s}', \mathbf{r}', \omega'). \end{aligned} \quad (\text{D33})$$

First, from Eqs. (D9) and (D18), the commutator appearing in \mathbf{X}_1 becomes

$$\begin{aligned} & \left[\check{\mathbf{P}}_{\text{ex}}(\mathbf{s}, i\omega + \delta), \{\hat{\mathbf{J}}(\mathbf{r}', \omega')\}^\dagger \right] \\ & = \sum_{\mu, \mu'} \mathcal{P}_\mu(\mathbf{s}) W_{\mu', \mu}^*(\omega) \int d\mathbf{s}' \mathcal{P}_{\mu'}^*(\mathbf{s}') \\ & \cdot \left[i\mu_0 \omega \int d\mathbf{s}'' \mathbf{G}^*(\mathbf{s}', \mathbf{s}'', \omega) \cdot \check{\mathbf{J}}(\mathbf{s}'', i\omega + \delta) + \mathbf{A}(\mathbf{s}', 0), \right. \\ & \left. \{\hat{\mathbf{J}}(\mathbf{r}', \omega')\}^\dagger \right]. \end{aligned} \quad (\text{D34})$$

Since $\hat{\mathbf{J}}(\mathbf{r}', \omega')$ is defined as Eq. (C33), what we must evaluate is

$$\begin{aligned} & \left[\check{\mathbf{J}}(\mathbf{s}'', i\omega + \delta), \{\hat{\mathbf{J}}(\mathbf{r}', \omega')\}^\dagger \right] \\ & = \frac{1}{2\pi} [\check{\mathbf{J}}(\mathbf{s}'', i\omega + \delta), \{\check{\mathbf{J}}(\mathbf{r}', -i\omega' + \delta) + \check{\mathbf{J}}(\mathbf{r}', i\omega' + \delta)\}^\dagger] \\ & + \frac{i\omega'^2}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}', \omega')] \int d\mathbf{r}'' \\ & [\check{\mathbf{J}}(\mathbf{s}'', i\omega + \delta), \{\check{\mathbf{J}}(\mathbf{r}'', i\omega' + \delta)\}^\dagger] \cdot \mathbf{G}(\mathbf{r}'', \mathbf{r}', \omega'). \end{aligned} \quad (\text{D35})$$

From Eqs. (B1) and (B3) of Ref. 7, the first term is evaluated as

$$\begin{aligned} & [\check{\mathbf{J}}(\mathbf{s}'', i\omega + \delta), \{\check{\mathbf{J}}(\mathbf{r}', -i\omega' + \delta) + \check{\mathbf{J}}(\mathbf{r}', i\omega' + \delta)\}^\dagger] \\ & = \frac{2\hbar\epsilon_0 \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}', \omega')]\omega\omega'}{i(\omega - \omega' - i\delta)} \mathbf{1}\delta(\mathbf{s}'' - \mathbf{r}'). \end{aligned} \quad (\text{D36})$$

Since the second term obeys Eq. (D24), we obtain

$$\begin{aligned} & \int ds'' \mathbf{G}^*(\mathbf{s}', \mathbf{s}'', \omega) \cdot \left[\check{\mathbf{J}}(\mathbf{s}'', i\omega + \delta), \{\hat{\mathbf{J}}(\mathbf{r}', \omega')\}^\dagger \right] \\ &= \frac{\epsilon_0 \hbar \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}', \omega')]\omega'^3}{i\pi \omega(\omega - \omega' - i\delta)} \mathbf{G}(\mathbf{s}', \mathbf{r}', \omega'). \end{aligned} \quad (\text{D37})$$

From Eqs. (52) and (53a) of Ref. 7, the last term of Eq. (D34) becomes

$$\begin{aligned} & \int ds' \mathcal{P}_{\mu'}^*(\mathbf{s}') \cdot \left[\mathbf{A}(\mathbf{s}', 0), \{\hat{\mathbf{J}}(\mathbf{r}', \omega')\}^\dagger \right] \\ &= \frac{\hbar\omega'^2}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}', \omega')] \int ds' [\mathcal{P}_{\mu'}^*(\mathbf{s}')]_{\text{T}} \cdot \mathbf{G}(\mathbf{s}', \mathbf{r}', \omega'). \end{aligned} \quad (\text{D38})$$

Therefore, \mathbf{X}_1 is evaluated as

$$\begin{aligned} \mathbf{X}_1 &= -\frac{\hbar\omega'^2\omega'^2}{\pi^2 c^4} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}', \omega')] \\ &\times \sum_{\mu, \mu'} \int ds \mathbf{G}^*(\mathbf{r}, \mathbf{s}, \omega) \cdot \mathcal{P}_{\mu}(\mathbf{s}) W_{\mu', \mu}^*(\omega) \\ &\times \int ds' \left[\frac{\omega\omega' \mathcal{P}_{\mu'}^*(\mathbf{s}')}{\omega - \omega' - i\delta} + \omega [\mathcal{P}_{\mu'}^*(\mathbf{s}')]_{\text{T}} \right] \cdot \mathbf{G}(\mathbf{s}', \mathbf{r}', \omega'). \end{aligned} \quad (\text{D39})$$

From its complex conjugate, we obtain

$$\begin{aligned} \mathbf{X}_2 &= -\frac{\hbar\omega'^2\omega'^2}{\pi^2 c^4} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega)] \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}', \omega')] \\ &\times \sum_{\mu, \mu'} \int ds \mathbf{G}^*(\mathbf{r}, \mathbf{s}, \omega) \cdot \left[\frac{\omega\omega' \mathcal{P}_{\mu}(\mathbf{s})}{\omega' - \omega + i\delta} + \omega' [\mathcal{P}_{\mu}(\mathbf{s})]_{\text{T}} \right] \\ &\times W_{\mu, \mu'}(\omega) \int ds' \mathcal{P}_{\mu'}^*(\mathbf{s}') \cdot \mathbf{G}(\mathbf{s}', \mathbf{r}', \omega'). \end{aligned} \quad (\text{D40})$$

Since, from Eqs. (D7b), (D9b), (D18), and (D28), the commutator appearing in \mathbf{X}_3 is written as

$$\begin{aligned} & \left[\check{\mathbf{P}}_{\text{ex}}(\mathbf{s}, i\omega + \delta) + \frac{i}{\omega} \mathbf{P}_{\text{exT}}(\mathbf{s}, 0), \right. \\ & \left. \{\check{\mathbf{P}}_{\text{ex}}(\mathbf{s}', i\omega' + \delta)\}^\dagger - \frac{i}{\omega'} \mathbf{P}_{\text{exT}}(\mathbf{s}', 0) \right] \\ &= \sum_{\mu, \mu'} \mathcal{P}_{\mu}(\mathbf{s}) W_{\mu', \mu}^*(\omega) \left[\frac{\hbar \mathcal{P}_{\mu'}^*(\mathbf{s}')}{\omega - \omega' - i\delta} + \frac{\hbar}{\omega'} [\mathcal{P}_{\mu'}^*(\mathbf{s}')]_{\text{T}} \right] \\ & - \sum_{\mu, \mu'} \left[\frac{\hbar \mathcal{P}_{\mu}(\mathbf{s})}{\omega - \omega' - i\delta} - \frac{\hbar}{\omega} [\mathcal{P}_{\mu}(\mathbf{s})]_{\text{T}} \right] W_{\mu, \mu'}(\omega') \mathcal{P}_{\mu'}^*(\mathbf{s}'), \end{aligned} \quad (\text{D41})$$

we can find that

$$\mathbf{X}_1 + \mathbf{X}_2 + \mathbf{X}_3 = \mathbf{0}. \quad (\text{D42})$$

Therefore, we obtain commutator (22a) of the noise current density. From the same kind of calculation and neglecting the nonresonance terms, we also obtain Eq. (22b).

3. With nonradiative relaxation

In this section, we perform the same kind of calculation as in the previous section considering a nonradiative relaxation process of excitons. Considering the reservoir oscillators, the Laplace transforms of Heisenberg equation (D1) of excitons are derived as

$$\begin{aligned} & (\hbar\omega_{\mu} - \hbar\omega - i\delta) \bar{b}_{\mu}(-i\omega + \delta) \\ &= -i\hbar b_{\mu}(0) + \int d\mathbf{r} \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot [\bar{\mathbf{E}}(\mathbf{r}, -i\omega + \delta) - \mathbf{A}(\mathbf{r}, 0)] \\ & - \int_0^{\infty} d\Omega [g_{\mu}(\Omega) \bar{d}_{\mu}(\Omega, -i\omega + \delta) + g_{\mu}^*(\Omega) \bar{d}_{\mu}^{\dagger}(\Omega, -i\omega + \delta)] \end{aligned} \quad (\text{D43a})$$

$$\begin{aligned} & (\hbar\omega_{\mu} - \hbar\omega + i\delta) \check{b}_{\mu}(i\omega + \delta) \\ &= i\hbar b_{\mu}(0) + \int d\mathbf{r} \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot [\check{\mathbf{E}}(\mathbf{r}, i\omega + \delta) + \mathbf{A}(\mathbf{r}, 0)] \\ & - \int_0^{\infty} d\Omega [g_{\mu}(\Omega) \check{d}_{\mu}(\Omega, i\omega + \delta) + g_{\mu}^*(\Omega) \check{d}_{\mu}^{\dagger}(\Omega, i\omega + \delta)]. \end{aligned} \quad (\text{D43b})$$

On the other hand, we obtain the motion equation of reservoir oscillators

$$i\hbar \frac{\partial}{\partial t} d_{\mu}(\Omega, t) = \hbar\Omega d_{\mu}(\Omega, t) + g_{\mu}^*(\Omega) [b_{\mu}(t) + b_{\mu}^{\dagger}(t)], \quad (\text{D44})$$

and its Laplace transforms

$$\begin{aligned} & (\hbar\Omega - \hbar\omega - i\delta) \bar{d}_{\mu}(\Omega, -i\omega + \delta) \\ &= -i\hbar d_{\mu}(\Omega, 0) - g_{\mu}^*(\Omega) [\bar{b}_{\mu}(-i\omega + \delta) + \bar{b}_{\mu}^{\dagger}(-i\omega + \delta)], \end{aligned} \quad (\text{D45a})$$

$$\begin{aligned} & (\hbar\Omega - \hbar\omega + i\delta) \check{d}_{\mu}(\Omega, i\omega + \delta) \\ &= i\hbar d_{\mu}(\Omega, 0) - g_{\mu}^*(\Omega) [\check{b}_{\mu}(i\omega + \delta) + \check{b}_{\mu}^{\dagger}(i\omega + \delta)]. \end{aligned} \quad (\text{D45b})$$

Substituting Eqs. (D45) into (D43) and neglecting the nonresonant terms, we obtain

$$\begin{aligned} & [\hbar\omega_{\mu} - \hbar\omega - i\gamma_{\mu}(\omega)/2] \bar{b}_{\mu}(-i\omega + \delta) \\ &= -i\hbar b_{\mu}(0) + \int d\mathbf{r} \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot [\bar{\mathbf{E}}(\mathbf{r}, -i\omega + \delta) - \mathbf{A}(\mathbf{r}, 0)] \\ & + \bar{D}_{\mu}(-i\omega + \delta), \end{aligned} \quad (\text{D46a})$$

$$\begin{aligned} & [\hbar\omega_{\mu} - \hbar\omega + i\gamma_{\mu}^*(\omega)/2] \check{b}_{\mu}(i\omega + \delta) \\ &= i\hbar b_{\mu}(0) + \int d\mathbf{r} \mathcal{P}_{\mu}^*(\mathbf{r}) \cdot [\check{\mathbf{E}}(\mathbf{r}, i\omega + \delta) + \mathbf{A}(\mathbf{r}, 0)] \\ & + \check{D}_{\mu}(i\omega + \delta), \end{aligned} \quad (\text{D46b})$$

where the operators on the RHS are defined as

$$\begin{aligned} \bar{D}_{\mu}(-i\omega + \delta) &\equiv \int_0^{\infty} d\Omega \frac{ig_{\mu}(\Omega)}{\Omega - \omega - i\delta} d_{\mu}(\Omega, 0) \\ & - \int_0^{\infty} d\Omega \frac{ig_{\mu}^*(\Omega)}{\Omega + \omega + i\delta} d_{\mu}^{\dagger}(\Omega, 0), \end{aligned} \quad (\text{D47a})$$

$$\begin{aligned} \check{D}_\mu(i\omega + \delta) &\equiv - \int_0^\infty d\Omega \frac{ig_\mu(\Omega)}{\Omega - \omega + i\delta} d_\mu(\Omega, 0) \\ &+ \int_0^\infty d\Omega \frac{ig_\mu^*(\Omega)}{\Omega + \omega - i\delta} d_\mu^\dagger(\Omega, 0), \end{aligned} \quad (\text{D47b})$$

and the relaxation width is

$$\frac{i\gamma_\mu(\omega)}{2} \equiv \int_0^\infty d\Omega \left[\frac{|g_\mu(\Omega)|^2}{\hbar\Omega - \hbar\omega - i\delta} + \frac{|g_\mu(\Omega)|^2}{\hbar\Omega + \hbar\omega + i\delta} \right]. \quad (\text{D48})$$

Substituting Laplace transformed Maxwell wave equations (D6) into Eqs. (D46), the self-consistent equation sets for the Laplace transformed exciton operators are obtained as

$$\begin{aligned} \sum_{\mu'} S_{\mu,\mu'}^{\text{abs}}(\omega) \bar{b}_{\mu'}(-i\omega + \delta) \\ = \bar{\beta}_\mu(-i\omega + \delta) + \bar{D}_\mu(-i\omega + \delta) = \bar{\beta}_\mu^{\text{abs}}(-i\omega + \delta), \end{aligned} \quad (\text{D49a})$$

$$\begin{aligned} \sum_{\mu'} \{S_{\mu',\mu}^{\text{abs}}(\omega)\}^* \check{b}_{\mu'}(i\omega + \delta) \\ = \check{\beta}_\mu(i\omega + \delta) + \check{D}_\mu(i\omega + \delta) = \check{\beta}_\mu^{\text{abs}}(i\omega + \delta), \end{aligned} \quad (\text{D49b})$$

and the one for the Fourier transform is

$$\sum_{\mu'} S_{\mu,\mu'}^{\text{abs}}(\omega) \hat{b}_{\mu'}(\omega) = \hat{\beta}_\mu(\omega) + \hat{D}_\mu(\omega) = \hat{\beta}_\mu^{\text{abs}}(\omega), \quad (\text{D50})$$

where operator $\hat{D}_\mu(\omega)$ is defined as

$$\begin{aligned} \hat{D}_\mu(\omega) &\equiv \frac{1}{2\pi} [\bar{D}_\mu(-i\omega + \delta) + \check{D}_\mu(i\omega + \delta)] \\ &- \frac{1}{2\pi} \frac{i\gamma_\mu(\omega) + i\gamma_\mu^*(\omega)}{2} \check{b}_\mu(i\omega + \delta). \end{aligned} \quad (\text{D51})$$

On the other hand, Fourier transformed motion equation (47) of the excitons is obtained by adding Eqs. (D46). Further, by substituting Eq. (23) into it, we can directly obtain self-consistent equations (D50).

From commutation relations (19) of $d_\mu(\Omega)$, the ones of the operators $\bar{D}_\mu(-i\omega + \delta)$ and $\check{D}_\mu(i\omega + \delta)$ are calculated as

$$\begin{aligned} [\bar{D}_\mu(-i\omega + \delta), \{\bar{D}_{\mu'}(-i\omega' + \delta)\}^\dagger] \\ = \frac{\hbar\delta_{\mu,\mu'}}{\omega - \omega' + i\delta} \frac{i\gamma_\mu(\omega) + i\gamma_\mu^*(\omega')}{2}, \end{aligned} \quad (\text{D52a})$$

$$\begin{aligned} [\check{D}_\mu(i\omega + \delta), \{\check{D}_{\mu'}(i\omega' + \delta)\}^\dagger] \\ = - \frac{\hbar\delta_{\mu,\mu'}}{\omega - \omega' - i\delta} \frac{i\gamma_\mu^*(\omega) + i\gamma_\mu(\omega')}{2}, \end{aligned} \quad (\text{D52b})$$

$$\begin{aligned} [\bar{D}_\mu(-i\omega + \delta), \{\check{D}_{\mu'}(i\omega' + \delta)\}^\dagger] \\ = - \frac{\hbar\delta_{\mu,\mu'}}{(\omega + i\delta) - (\omega' + i\delta)} \frac{i\gamma_\mu(\omega) - i\gamma_\mu(\omega')}{2}, \end{aligned} \quad (\text{D52c})$$

and neglecting the nonresonant terms, we obtain

$$\begin{aligned} &[\bar{D}_\mu(-i\omega + \delta), \bar{D}_{\mu'}(-i\omega' + \delta)] \\ &= [\check{D}_\mu(i\omega + \delta), \check{D}_{\mu'}(i\omega' + \delta)] \\ &= [\bar{D}_\mu(-i\omega + \delta), \check{D}_{\mu'}(i\omega' + \delta)] = 0. \end{aligned} \quad (\text{D52d})$$

From these relations, we obtain the commutation relations of $\check{\beta}_\mu^{\text{abs}}(i\omega + \delta)$:

$$\begin{aligned} &[\check{\beta}_\mu^{\text{abs}}(i\omega + \delta), \{\check{\beta}_{\mu'}^{\text{abs}}(i\omega' + \delta)\}^\dagger] \\ &= \hbar^2 \delta_{\mu,\mu'} + \frac{\hbar}{\omega - \omega' - i\delta} [\mathcal{A}_{\mu,\mu'}(\omega') - \mathcal{A}_{\mu',\mu}^*(\omega)] \\ &- \frac{\hbar\delta_{\mu,\mu'}}{\omega - \omega' - i\delta} \frac{i\gamma_\mu^*(\omega) + i\gamma_\mu(\omega')}{2} \end{aligned} \quad (\text{D53})$$

$$= \frac{\hbar}{\omega - \omega' - i\delta} [S_{\mu,\mu'}^{\text{abs}}(\omega') - \{S_{\mu',\mu}^{\text{abs}}(\omega)\}^*], \quad (\text{D54})$$

and

$$[\check{\beta}_\mu^{\text{abs}}(i\omega + \delta), \check{\beta}_{\mu'}^{\text{abs}}(i\omega' + \delta)] = 0. \quad (\text{D55})$$

They have a good correspondence with Eqs. (D26) and (D27), which are discussed without the nonradiative relaxation. Therefore, the commutation relations of $\check{b}_\mu(i\omega + \delta)$ have the same form as that of Eq. (D28) except for replacing $W_{\mu,\mu'}(\omega)$ with $W_{\mu,\mu'}^{\text{abs}}(\omega)$, and relations (22) of $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ are not changed even by considering the nonradiative relaxation.

Next, we evaluate the commutators of $\hat{D}_\mu(\omega)$ defined in Eq. (D51). Here, we apply the Markov approximation to the reservoir oscillators interacting with excitons, and assume that correction term $\gamma_\mu(\omega)$ is a real value. From Eqs. (D52) and Dirac's equivalence $(x \pm i\delta)^{-1} = P x^{-1} \mp i\pi\delta(x)$, we find

$$\begin{aligned} &[\bar{D}_\mu(-i\omega + \delta) + \check{D}_\mu(i\omega + \delta), \\ &\{\bar{D}_{\mu'}(-i\omega' + \delta) + \check{D}_{\mu'}(i\omega' + \delta)\}^\dagger] \\ &= \delta_{\mu,\mu'} \delta(\omega - \omega') 2\pi\hbar\gamma_\mu(\omega), \end{aligned} \quad (\text{D56a})$$

$$\begin{aligned} &[\bar{D}_\mu(-i\omega + \delta) + \check{D}_\mu(i\omega + \delta), \\ &\bar{D}_{\mu'}(-i\omega' + \delta) + \check{D}_{\mu'}(i\omega' + \delta)] = 0. \end{aligned} \quad (\text{D56b})$$

On the other hand, Eq. (D49b) gives the relation

$$\begin{aligned} &[\bar{D}_\mu(-i\omega + \delta) + \check{D}_\mu(i\omega + \delta), \{\check{b}_{\mu'}(i\omega' + \delta)\}^\dagger] \\ &= - \left[\frac{i\hbar}{(\omega + i\delta) - (\omega' + i\delta)} \frac{\gamma_\mu(\omega) - \gamma_\mu(\omega')}{2} \right. \\ &\left. + \frac{i\hbar}{\omega - \omega' - i\delta} \frac{\gamma_\mu^*(\omega) + \gamma_\mu(\omega')}{2} \right] W_{\mu,\mu'}^{\text{abs}}(\omega'). \end{aligned} \quad (\text{D57})$$

Then, we obtain Eq. (48a) and also Eq. (48b) by neglecting the nonresonant terms.

Finally, we verify the independence of source operators $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ and $\hat{D}_\mu(\omega)$. Since $\hat{D}_\mu(\omega)$ is defined as (D51), we

obtain the expression of the commutator

$$\begin{aligned} & \left[\hat{\mathcal{D}}_\mu(\omega), \{\hat{\mathbf{J}}_0(\mathbf{r}, \omega')\}^\dagger \right] \\ &= \frac{1}{2\pi} \left[\bar{D}_\mu(-i\omega + \delta) + \check{D}_\mu(i\omega + \delta), \{\hat{\mathbf{J}}_0(\mathbf{r}, \omega')\}^\dagger \right] \\ & \quad - \frac{i}{2\pi} \frac{\gamma_\mu(\omega) + \gamma_\mu^*(\omega)}{2} \left[\check{b}_\mu(i\omega + \delta), \{\hat{\mathbf{J}}_0(\mathbf{r}, \omega')\}^\dagger \right]. \end{aligned} \quad (\text{D58})$$

Here, from Eqs. (D29) and (D57), the first term becomes

$$\begin{aligned} & \frac{i\hbar\omega'^3}{4\pi^2 c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega')] \\ & \times \left[\frac{\gamma_\mu(\omega) - \gamma_\mu(\omega')}{(\omega + i\delta) - (\omega' + i\delta)} + \frac{\gamma_\mu^*(\omega) + \gamma_\mu(\omega')}{\omega - \omega' - i\delta} \right] \\ & \times \sum_{\mu'} W_{\mu, \mu'}^{\text{abs}}(\omega') \int d\mathbf{r}' \mathcal{P}_{\mu'}^*(\mathbf{r}') \cdot \mathbf{G}(\mathbf{r}', \mathbf{r}, \omega'). \end{aligned} \quad (\text{D59})$$

The commutator appearing in the second term of (D58) is written as

$$\begin{aligned} & \left[\check{b}_\mu(i\omega + \delta), \{\hat{\mathbf{J}}_0(\mathbf{r}, \omega')\}^\dagger \right] = \left[\check{b}_\mu(i\omega + \delta), \{\hat{\mathbf{J}}(\mathbf{r}, \omega')\}^\dagger \right] \\ & - \frac{\omega'^3}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega')] \int d\mathbf{r}' \left[\check{b}_\mu(i\omega + \delta), \right. \\ & \left. \{\check{\mathbf{P}}_{\text{ex}}(\mathbf{r}', i\omega' + \delta)\}^\dagger - \frac{i}{\omega'} \mathbf{P}_{\text{exT}}(\mathbf{r}', 0) \right] \cdot \mathbf{G}(\mathbf{r}', \mathbf{r}, \omega'). \end{aligned} \quad (\text{D60})$$

From Eq. (D39), the first term on the RHS of (D60)

$$\begin{aligned} & \frac{\hbar\omega'^2}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega')] \sum_{\mu'} \{W_{\mu', \mu}^{\text{abs}}(\omega)\}^* \\ & \times \int d\mathbf{r}' \left[\frac{\omega' \mathcal{P}_{\mu'}^*(\mathbf{r}')}{\omega - \omega' - i\delta} + [\mathcal{P}_{\mu'}^*(\mathbf{r}')]_{\text{T}} \right] \cdot \mathbf{G}(\mathbf{r}', \mathbf{r}, \omega'), \end{aligned} \quad (\text{D61})$$

and from Eqs. (D9b), (D49b), and (D28), the second term is calculated as

$$\begin{aligned} & - \frac{\hbar\omega'^2}{\pi c^2} \text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega')] \\ & \times \sum_{\mu'} \int d\mathbf{r}' \left\{ \frac{\omega' \mathcal{P}_{\mu'}^*(\mathbf{r}')}{\omega - \omega' - i\delta} [\{W_{\mu', \mu}^{\text{abs}}(\omega)\}^* - W_{\mu, \mu'}^{\text{abs}}(\omega')] \right. \\ & \left. + \{W_{\mu', \mu}^{\text{abs}}(\omega)\}^* [\mathcal{P}_{\mu'}^*(\mathbf{r}')]_{\text{T}} \right\} \cdot \mathbf{G}(\mathbf{r}', \mathbf{r}, \omega'). \end{aligned} \quad (\text{D62})$$

Therefore, commutator (D60) is evaluated as

$$\begin{aligned} & \left[\check{b}_\mu(i\omega + \delta), \{\hat{\mathbf{J}}_0(\mathbf{r}, \omega')\}^\dagger \right] = \frac{\hbar\omega'^3}{\pi c^2} \frac{\text{Im}[\epsilon_{\text{bg}}(\mathbf{r}, \omega')]}{\omega - \omega' - i\delta} \\ & \times \sum_{\mu'} W_{\mu, \mu'}^{\text{abs}}(\omega') \int d\mathbf{r}' \mathcal{P}_{\mu'}^*(\mathbf{r}') \cdot \mathbf{G}(\mathbf{r}', \mathbf{r}, \omega'). \end{aligned} \quad (\text{D63})$$

From this and Eq. (D59), we obtain Eqs. (49) by assuming $\gamma_\mu(\omega) = \gamma_\mu^*(\omega)$ and neglecting the nonresonant terms.

APPENDIX E: SELF-STANDING MODES AND RETARDED CORRELATION FUNCTION

As discussed in sections V and VI, commutation relations of exciton operators in the Fourier transformed Heisenberg representation are written as Eq. (42) in terms of inversed matrix $\mathbf{W}^{\text{abs}}(\omega)$ of coefficient $\mathbf{S}^{\text{abs}}(\omega)$ of the self-consistent equation set. As the analogue of the relation between the electric field and Green's tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ satisfying Eq. (8), we can understand that the elements of $\mathbf{W}^{\text{abs}}(\omega)$ identify with the Fourier transforms of retarded correlation functions $D_{\mu, \mu'}^{\text{Rex}}$ of exciton operators:

$$- \hbar W_{\mu, \mu'}^{\text{abs}}(\omega) = D_{\mu, \mu'}^{\text{Rex}}(\omega) = \int_{-\infty}^{\infty} dt D_{\mu, \mu'}^{\text{Rex}}(t) e^{i\omega t}, \quad (\text{E1})$$

wehre

$$D_{\mu, \mu'}^{\text{Rex}}(t - t') \equiv \begin{cases} -i \left\langle \left[b_\mu(t), b_{\mu'}^\dagger(t') \right] \right\rangle & t > t' \\ 0 & t < t' \end{cases}. \quad (\text{E2})$$

This identity indicates that $\mathbf{W}^{\text{abs}}(\omega)$ has no pole in the upper half side of the complex ω -plane. On the other hand, the poles $\{\omega_\lambda\}$ in the lower half ω -plane characterize the self-standing modes of the exciton-polaritons satisfying

$$\det[\mathbf{S}^{\text{abs}}(\omega_\lambda)] = 0, \quad (\text{E3})$$

because it makes inverse matrix $\mathbf{W}^{\text{abs}}(\omega)$ singular at $\omega = \omega_\lambda$.

APPENDIX F: EQUAL-TIME COMMUTATION RELATIONS

Commutation relations of equal-time Heisenberg operators should keep the form of those of the Shrödinger operators. This means that the relations

$$\left[b_\mu(t), b_{\mu'}^\dagger(t) \right] = \delta_{\mu, \mu'}, \quad (\text{F1})$$

$$[\mathbf{P}_{\text{ex}}(\mathbf{r}, t), \mathbf{P}_{\text{ex}}(\mathbf{r}', t)] = \mathbf{0}, \quad (\text{F2})$$

$$[\mathbf{E}(\mathbf{r}, t), \mathbf{E}(\mathbf{r}', t)] = \mathbf{0} \quad (\text{F3})$$

should be derived from the commutation relations of the Fourier transformed Heisenberg operators. Moreover, from Eqs. (20) and (C5), the electric field is represented as

$$\epsilon_0 \mathbf{E}(\mathbf{r}, t) = -\mathbf{\Pi}(\mathbf{r}, t) - \epsilon_0 \nabla \phi(\mathbf{r}, t), \quad (\text{F4})$$

and since $\mathbf{\Pi}(\mathbf{r}, t)$ and $\mathbf{A}(\mathbf{r}, t)$ satisfy commutation relation (C1), the relation

$$[\epsilon_0 \mathbf{E}(\mathbf{r}, t), \mathbf{A}(\mathbf{r}', t)] = i\hbar \delta_{\text{T}}(\mathbf{r} - \mathbf{r}') \quad (\text{F5})$$

should also be derived. For local dielectric media, the same kind of calculation has been performed by Knöll, Scheel, and Welsch (KSW).¹³

From the time-representation of exciton operator

$$b_\mu(t) = \int_{-\infty}^{\infty} d\omega \hat{b}_\mu(\omega) e^{-i\omega t} \quad (\text{F6})$$

and relations (52) for ω -representation, the equal-time commutation relation is written as

$$[b_\mu(t), b_{\mu'}^\dagger(t)] = \frac{\hbar}{i2\pi} \int_{-\infty}^{\infty} d\omega [W_{\mu,\mu'}^{\text{abs}}(\omega) - \{W_{\mu',\mu}^{\text{abs}}(\omega)\}^*]. \quad (\text{F7})$$

In the limit $|\omega| \rightarrow \infty$, as indicated in App. A.1 of KSW work,¹³ it is known that $\epsilon_{\text{bg}}(\mathbf{r}, \omega) \rightarrow 1$ and

$$\lim_{|\omega| \rightarrow \infty} \frac{\omega^2}{c^2} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = -\delta(\mathbf{r} - \mathbf{r}'). \quad (\text{F8})$$

Then, due to the orthogonality of $\mathcal{P}_\mu(\mathbf{r})$ shown in Eq. (27) and the relation with LT splitting $\Delta_{\text{LT}}^\mu = |\mathcal{P}_\mu|^2 / \epsilon_{\text{bg}}(\omega_\mu) \epsilon_0$, the limit of correction term (35) is

$$\lim_{|\omega| \rightarrow \infty} \mathcal{A}_{\mu,\mu'}(\omega) = \frac{1}{\epsilon_0} \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathcal{P}_{\mu'}(\mathbf{r}) \quad (\text{F9})$$

$$= \delta_{\mu,\mu'} \epsilon_{\text{bg}}(\omega_\mu) \Delta_{\text{LT}}^\mu. \quad (\text{F10})$$

On the other hand, the limit of nonradiative width $\gamma_\mu(\omega)$ defined in Eq. (D48) is $\gamma_\mu(\omega) \rightarrow 0$. Therefore, coefficient matrix $\mathbf{S}^{\text{abs}}(\omega)$ becomes diagonal as

$$\lim_{|\omega| \rightarrow \infty} S_{\mu,\mu'}^{\text{abs}}(\omega) = [\hbar\omega_\mu + \epsilon_{\text{bg}}(\omega_\mu) \Delta_{\text{LT}}^\mu - \hbar\omega - i\delta] \delta_{\mu,\mu'}, \quad (\text{F11})$$

and of course its inverse matrix is also diagonal:

$$\lim_{|\omega| \rightarrow \infty} W_{\mu,\mu'}^{\text{abs}}(\omega) = \frac{\delta_{\mu,\mu'}}{\hbar\omega_\mu + \epsilon_{\text{bg}}(\omega_\mu) \Delta_{\text{LT}}^\mu - \hbar\omega - i\delta}. \quad (\text{F12})$$

As mentioned in App. E, since $\mathbf{W}^{\text{abs}}(\omega)$ has no pole in the upper half ω -plane, the integration over the real axis is evaluated as

$$\int_{-\infty}^{\infty} d\omega W_{\mu,\mu'}^{\text{abs}}(\omega) = \frac{i\pi}{\hbar} \delta_{\mu,\mu'}. \quad (\text{F13})$$

We can find that this equation reproduces commutation relation (F1) from Eq. (F7).

Next, we verify commutation relation (F2) of excitonic polarization $\mathbf{P}_{\text{ex}}(\mathbf{r}, t)$. Although we approximate its positive-frequency Fourier component as Eq. (29) in Sec. V, here we describe it without the RWA as

$$\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) = \sum_{\mu} \left[\mathcal{P}_\mu(\mathbf{r}) \hat{b}_\mu(\omega) + \mathcal{P}_\mu^*(\mathbf{r}) \{\hat{b}_\mu(-\omega)\}^\dagger \right]. \quad (\text{F14})$$

This representation keeps the following relation derived from the definition of Fourier transform Eq. (5):

$$\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) = \hat{\mathbf{P}}_{\text{ex}}^-(\mathbf{r}, -\omega) = \{\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, -\omega)\}^\dagger \quad (\text{F15})$$

From this relation, the time-representation can be written as

$$\begin{aligned} \mathbf{P}_{\text{ex}}(\mathbf{r}, t) &= \int_0^{\infty} d\omega \left[\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) e^{-i\omega t} + \hat{\mathbf{P}}_{\text{ex}}^-(\mathbf{r}, \omega) e^{i\omega t} \right] \quad (\text{F16}) \\ &= \int_{-\infty}^{\infty} d\omega \hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) e^{-i\omega t} = \int_{-\infty}^{\infty} d\omega \hat{\mathbf{P}}_{\text{ex}}^-(\mathbf{r}, \omega) e^{i\omega t}. \quad (\text{F17}) \end{aligned}$$

Therefore, the equal-time commutator becomes

$$\begin{aligned} [\mathbf{P}_{\text{ex}}(\mathbf{r}, t), \mathbf{P}_{\text{ex}}(\mathbf{r}', t)] &= \int_{-\infty}^{\infty} d\omega \int_{-\infty}^{\infty} d\omega' e^{-i\omega t} \left[\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega), \hat{\mathbf{P}}_{\text{ex}}^-(\mathbf{r}', \omega') \right] e^{i\omega' t} \\ &= \sum_{\mu} \left[\mathcal{P}_\mu(\mathbf{r}) \mathcal{P}_\mu^*(\mathbf{r}') - \mathcal{P}_\mu^*(\mathbf{r}) \mathcal{P}_\mu(\mathbf{r}') \right]. \quad (\text{F18}) \end{aligned}$$

We cannot obtain the second term from ω -representation (29) with the RWA. Equation (F18) is also obtained using Eqs. (26) and (F1) directly. From Eq. (F18), we can reproduce equal-time commutation relation (F2):

$$\begin{aligned} [\mathbf{P}_{\text{ex}}(\mathbf{r}, t), \mathbf{P}_{\text{ex}}(\mathbf{r}', t)]_{\xi, \xi'} &= \sum_{\mu} \left[\mathcal{P}_\mu^\xi(\mathbf{r}) \{\mathcal{P}_\mu^{\xi'}(\mathbf{r}')\}^* - \text{c.c.} \right] \quad (\text{F19}) \end{aligned}$$

$$= \sum_{\mu} \left[\langle 0 | P_{\text{ex}}^\xi(\mathbf{r}) | \mu \rangle \langle \mu | P_{\text{ex}}^{\xi'}(\mathbf{r}') | 0 \rangle - \text{c.c.} \right] \quad (\text{F20})$$

$$= \langle 0 | \left[P_{\text{ex}}^\xi(\mathbf{r}), P_{\text{ex}}^{\xi'}(\mathbf{r}') \right] | 0 \rangle = 0, \quad (\text{F21})$$

where $\xi, \xi' = x, y, z$, $|\mu\rangle = b_\mu^\dagger |0\rangle$, and $|0\rangle$ indicates the ground state of the medium.

Next, we verify relation (F3) of the electric field operator. From the representation of the positive-frequency Fourier component of electric field (23) and that of excitonic polarization (F14), instead of Eq. (39), we can also write the electric field operator without the RWA:

$$\begin{aligned} \hat{\mathbf{E}}^+(\mathbf{r}, \omega) &= \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) \\ &+ \sum_{\mu} \left[\mathcal{E}_\mu(\mathbf{r}, \omega) \hat{b}_\mu(\omega) - \mathcal{E}_\mu^*(\mathbf{r}, -\omega) \{\hat{b}_\mu(-\omega)\}^\dagger \right], \quad (\text{F22}) \end{aligned}$$

where we use the relation derived from Eqs. (40) and (41) with $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \{\mathbf{G}(\mathbf{r}, \mathbf{r}', -\omega^*)\}^*$:

$$\mathcal{E}_\mu(\mathbf{r}, \omega) = \mathcal{F}_\mu^*(\mathbf{r}, -\omega), \quad (\text{F23})$$

$$\mathcal{F}_\mu(\mathbf{r}, \omega) = \mathcal{E}_\mu^*(\mathbf{r}, -\omega). \quad (\text{F24})$$

Since the time-representation of the electric field is also written in the same form as Eq. (F17), we can evaluate its equal-time commutator as

$$\begin{aligned}
[\mathbf{E}(\mathbf{r}, t), \mathbf{E}(\mathbf{r}', t)] &= \int_{-\infty}^{\infty} d\omega \int_{-\infty}^{\infty} d\omega' e^{-i(\omega-\omega')t} \left[\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega), \hat{\mathbf{E}}_0^-(\mathbf{r}', \omega') \right] \\
&+ \frac{\hbar}{i2\pi} \sum_{\mu, \mu'} \int_{-\infty}^{\infty} d\omega \left[\mathcal{E}_{\mu}(\mathbf{r}, \omega) W_{\mu, \mu'}^{\text{abs}}(\omega) \mathcal{F}_{\mu'}(\mathbf{r}', \omega) - \mathcal{F}_{\mu}^*(\mathbf{r}, \omega) \{W_{\mu', \mu}^{\text{abs}}(\omega)\}^* \mathcal{E}_{\mu'}^*(\mathbf{r}', \omega) \right. \\
&\left. + \mathcal{E}_{\mu}^*(\mathbf{r}, -\omega) \{W_{\mu, \mu'}^{\text{abs}}(-\omega)\}^* \mathcal{F}_{\mu'}^*(\mathbf{r}', -\omega) - \mathcal{F}_{\mu}(\mathbf{r}, -\omega) W_{\mu', \mu}^{\text{abs}}(-\omega) \mathcal{E}_{\mu'}(\mathbf{r}', -\omega) \right]. \quad (\text{F25})
\end{aligned}$$

The first term is the equal-time commutator between the electric fields in the background medium; then, it becomes zero as indicated by SW or as calculated by KSW. Based on the fact that $W_{\mu, \mu'}^{\text{abs}}(\omega)$ and $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ have no pole in the upper half ω -plane as discussed in App. E, we evaluate the second term using the residue theorem. Since $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ becomes (F8) in the limit of $|\omega| \rightarrow \infty$, from Eqs. (40) and (41), we can obtain

$$\lim_{|\omega| \rightarrow \infty} \mathcal{E}_{\mu}(\mathbf{r}, \omega) = -\mathcal{P}_{\mu}(\mathbf{r})/\epsilon_0, \quad (\text{F26})$$

$$\lim_{|\omega| \rightarrow \infty} \mathcal{F}_{\mu}(\mathbf{r}, \omega) = -\mathcal{P}_{\mu}^*(\mathbf{r})/\epsilon_0. \quad (\text{F27})$$

From these and Eq. (F12), the second term of Eq. (F25) becomes

$$\begin{aligned}
&\frac{\hbar}{i2\pi} \sum_{\mu, \mu'} \int_{-\infty}^{\infty} d\omega \mathcal{E}_{\mu}(\mathbf{r}, \omega) W_{\mu, \mu'}^{\text{abs}}(\omega) \mathcal{F}_{\mu'}(\mathbf{r}', \omega) \\
&= \frac{1}{2\epsilon_0^2} \sum_{\mu} \mathcal{P}_{\mu}(\mathbf{r}) \mathcal{P}_{\mu}^*(\mathbf{r}'). \quad (\text{F28})
\end{aligned}$$

Calculating the other terms in the same way, we obtain

$$\begin{aligned}
&[\mathbf{E}(\mathbf{r}, t), \mathbf{E}(\mathbf{r}', t)] \\
&= \frac{1}{\epsilon_0^2} \sum_{\mu} [\mathcal{P}_{\mu}(\mathbf{r}) \mathcal{P}_{\mu}^*(\mathbf{r}') - \mathcal{P}_{\mu}^*(\mathbf{r}) \mathcal{P}_{\mu}(\mathbf{r}')] = \mathbf{0}, \quad (\text{F29})
\end{aligned}$$

then, equal-time commutation relation (F3) is reproduced.

Last, we verify relation (F5). As discussed in App. A.2 of KSW work, we can write the time-representation of the vector potential as

$$\begin{aligned}
\mathbf{A}(\mathbf{r}, t) &= \lim_{\epsilon \rightarrow 0} \int_{\epsilon}^{\infty} d\omega \int d\mathbf{s} \left[\frac{e^{-i\omega t}}{i\omega} \hat{\mathbf{E}}^+(\mathbf{s}, \omega) + \text{H.c.} \right] \cdot \boldsymbol{\delta}_{\text{T}}(\mathbf{s} - \mathbf{r}), \quad (\text{F30}) \\
&= \text{P} \int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega t}}{i\omega} \int d\mathbf{s} \hat{\mathbf{E}}^+(\mathbf{s}, \omega) \cdot \boldsymbol{\delta}_{\text{T}}(\mathbf{s} - \mathbf{r}), \quad (\text{F31}) \\
&= -\text{P} \int_{-\infty}^{\infty} d\omega \frac{e^{i\omega t}}{i\omega} \int d\mathbf{s} \hat{\mathbf{E}}^-(\mathbf{s}, \omega) \cdot \boldsymbol{\delta}_{\text{T}}(\mathbf{s} - \mathbf{r}), \quad (\text{F32})
\end{aligned}$$

where P indicates the principal value integration. From this representation, the commutator between the electric field and the vector potential becomes

$$\begin{aligned}
[\epsilon_0 \mathbf{E}(\mathbf{r}, t), \mathbf{A}(\mathbf{r}', t)] &= -\epsilon_0 \int_{-\infty}^{\infty} d\omega \text{P} \int_{-\infty}^{\infty} d\omega' \frac{e^{-i(\omega-\omega')t}}{i\omega'} \int d\mathbf{s} \left[\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega), \hat{\mathbf{E}}_0^-(\mathbf{s}, \omega') \right] \cdot \boldsymbol{\delta}_{\text{T}}(\mathbf{s} - \mathbf{r}') \\
&+ \frac{\epsilon_0 \hbar}{2\pi} \sum_{\mu, \mu'} \text{P} \int_{-\infty}^{\infty} \frac{d\omega}{\omega} \int d\mathbf{s} \left[\mathcal{E}_{\mu}(\mathbf{r}, \omega) W_{\mu, \mu'}(\omega) \mathcal{F}_{\mu'}(\mathbf{s}, \omega) - \mathcal{F}_{\mu}^*(\mathbf{r}, \omega) W_{\mu', \mu}^*(\omega) \mathcal{E}_{\mu'}^*(\mathbf{s}, \omega) \right. \\
&\left. + \mathcal{E}_{\mu}^*(\mathbf{r}, -\omega) W_{\mu, \mu'}^*(-\omega) \mathcal{F}_{\mu'}^*(\mathbf{s}, -\omega) - \mathcal{F}_{\mu}(\mathbf{r}, -\omega) W_{\mu', \mu}(-\omega) \mathcal{E}_{\mu'}(\mathbf{s}, -\omega) \right] \cdot \boldsymbol{\delta}_{\text{T}}(\mathbf{s} - \mathbf{r}'). \quad (\text{F33})
\end{aligned}$$

The first term is the same kind of commutator for background medium, then it becomes $i\hbar \boldsymbol{\delta}_{\text{T}}(\mathbf{r} - \mathbf{r}')$ as verified by KSW. Therefore, all we have to do is verify that the other terms become zero. First, in the limit of $|\omega| \rightarrow \infty$, they become zero because of factor ω^{-1} comparing to

the last four terms of Eq. (F25). In the limit of $|\omega| \rightarrow 0$, due to the equation shown in KSW work,

$$\lim_{|\omega| \rightarrow 0} \frac{\omega^2}{c^2} \int d\mathbf{s} \mathbf{G}(\mathbf{r}, \mathbf{s}, \omega) \cdot \boldsymbol{\delta}_{\text{T}}(\mathbf{s} - \mathbf{r}') = \mathbf{0} \quad (\text{F34})$$

we can find that the terms also become zero:

$$\begin{aligned} & \lim_{|\omega| \rightarrow 0} \frac{\omega^2}{c^2} \int ds \delta_{\mathbf{T}}(\mathbf{s} - \mathbf{r}') \cdot \boldsymbol{\mathcal{E}}_{\mu}(\mathbf{s}, \omega) \\ &= \lim_{|\omega| \rightarrow 0} \frac{\omega^2}{c^2} \int ds \delta_{\mathbf{T}}(\mathbf{s} - \mathbf{r}') \cdot \boldsymbol{\mathcal{F}}_{\mu}(\mathbf{s}, \omega) = \mathbf{0}. \end{aligned} \quad (\text{F35})$$

Therefore, Eq. (F33) reproduces equal-time commutation relation (F5).

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