QED of excitons with nonlocal susceptibility in arbitrarily structured dielectrics

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We have constructed a complete quantum theory for an optical process of excitons with nonlocal susceptibility originating from their center-of-mass motion. This theory provides a practical calculation method for arbitrary-structured nanoscale to macroscale 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 regarding classical light or the coherent states of photons. However, there is growing interest in the quantum electrodynamics (QED) of elementary excitations in condensed matter 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 vacuum[4] but also in a medium characterized by a frequency-independent dielectric constant. On the other hand, Hopfield[5] systematically discussed the eigenstates of exciton-photon systems or exciton polaritons; those have a frequency dependence in the susceptibility $\chi(\omega)$ or dielectric function $\epsilon(\omega)=1+\chi(\omega)$, as seen in their dispersion relation $\omega^2\epsilon(\omega)=c^2k^2$. Although in his treatment $\chi(\omega)$ only included a real part, susceptibility is generally represented as a complex function satisfying the Kramers-Kronig relations. In addition, its imaginary part, which causes damping effects, cannot be neglected in the discussion of the resonant optical processes of elementary excitations in condensed matter.

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 three-dimensional (3D) ones by Suttorp and Wubs (SW).[7] In the former scheme, dispersive dielectrics are described using the classical Hopfield polariton model,[5] i.e., polarizable harmonic oscillators interacting 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 eigenoperators derived from the diagonalization of a Hamiltonian. In the expression of these fields, there exists a complex dielectric function $\epsilon(\omega)$ represented by system parameters satisfying the Kramers-Kronig relations. This function characterizes the quantum fluctuation of the electromagnetic fields in the materials. The pioneering work of HB[6] stimulated various theoretical studies associated with the QED of dispersive and absorptive dielectrics, for example, the spontaneous decay,[8,9] input-output relations,[10–12] and quantization in amplifying, anisotropic, magnetic, or nonlinear media.[13–17] On the other hand, SW[7] generalized the quantization of the electromagnetic fields in arbitrary-structured 3D dielectrics by using the Laplace transformation technique.[18] Around the same time, the diagonalization of the Hamiltonian of SW[7] has been performed by Suttorp and van Wonderen.[19] In these schemes, the complex dielectric function $\epsilon(\mathbf{r},\omega)$ depends on the spatial position $\mathbf{r}$ of a medium and the radiation frequency ω.

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

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

This 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, the nonlocality is not considered to be important for macroscopic materials; this is because the coherence length of elementary excitations is usually much shorter than the spatial scale of the light wavelength. Therefore, only the averaged values of physical quantities over the coherence volume are reflected in observation, and the nonlocal effect is not apparent. However, in high-quality media, the motion of excitons should have a considerably long coherence, and the electromagnetic fields should vary in a considerably short distance in the resonance condition. In such cases, the nonlocality becomes important even for bulk materials, as will be explained below.

In the case of homogeneous media, the 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

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

In this manner, the susceptibility $\chi(\mathbf{k},\omega)$ has a wave-vector dependence as well as ω dependence even for homogeneous media when it has the nonlocality. This $\mathbf{k}$ dependence leads to more than one propagating or evanescent modes for a single frequency satisfying

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

Now, we consider a single exciton state with a finite translational mass $m_{\rm ex}$, transverse exciton energy E_T, and longitudinal one E_L at the respective band edges. Since the transverse exciton energy is represented as $E_{\rm ex}(\mathbf{k}) = E_T + \hbar^2 |\mathbf{k}|^2 / 2m_{\rm ex}$ for wave vector $\mathbf{k}$, we can find two propagating polariton modes for $\hbar\omega > E_L$ and one propagating mode at the polariton band gap $E_T < \hbar\omega < E_L$ in addition to an evanescent mode. These multiple polariton modes do not appear in the classical Hopfield polariton model[5] because the excitons were assumed to have infinite translational mass. As first reported by Pekar,[20] it appears that additional boundary conditions (ABCs) should be introduced in addition to the Maxwell boundary conditions for the unique connection between the polariton modes within 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 study, subsequent studies have revealed that this problem can be resolved by considering the microscopically determined boundary conditions of the excitonic center-of-mass motion at interfaces.[21–23] Nowadays, in the semiclassical framework, a calculation method independent of the notation of ABCs is well known as an ABC-free theory[24] or a microscopic nonlocal theory.[25,26] These theories consider the nonlocality of the susceptibility $\chi(\mathbf{r},\mathbf{r}',\omega)$ from the microscopic point of view, in contrast to the macroscopic consideration of the nonlocality such as the phenomenological introduction of ABCs into the excitonic polarization.[20] Under these microscopic theories, various linear and nonlinear phenomena in inhomogeneous materials have been discussed. In particular, for nanostructured materials, where the coherence of the center-of-mass motion of excitons is maintained in the entire material (weak confinement regime), the anomalous size dependence of their optical processes has been elucidated.[27–31] With regard to nanofilms, the nonlocal theory has successfully explained their peculiar spectral structures originating from the polariton interference.[23,32,33] Further, with the recent development of fabrication technologies for nanosamples, 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,[30] interchange of quantized states due to giant radiative shift,[34] and ultrafast radiative decay with femtosecond order.[35]

From these theoretical and experimental results and due to the considerable interest in the nonclassical states of elementary excitations,[1–3] 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 the nonlocality into the QED of dispersive and absorptive dielectrics, there remains a problem of how to perform calculations in practical applications, as will be shown in Sec. II. The principal purpose of this study is to construct a QED theory providing a practical calculation method for the nonlocal systems from the microscopic point of view. In particular, we discuss excitons weakly confined in arbitrary-structured 3D dielectrics considering the radiative and nonradiative relaxations, which is necessary to discuss the effects of, for example, material interfaces, excitonic confinement in nanostructures, and exciton relaxation processes. For this purpose, we have developed a theory integrating a microscopic nonlocal theory[25,26] and the quantization technique of SW[7] in this study. Further, we have already applied the present theory to the practical calculation of entangled-photon generation via biexcitons in nanostructures.[36]

In Sec. II, we explain the previously discussed QED theories with the nonlocality. The Hamiltonian of the present paper is shown in Sec. III, and two fundamental equations of our theory, the Maxwell wave equation and motion equations of excitons, are, respectively, shown in Secs. IV and V. In Sec. VI, we derive a wave equation with the nonlocal susceptibility as discussed in previous studies. Our QED theory is explained in Sec. VII, and the commutation relations of excitons and the electric field are shown in Sec. VIII. We discuss the validity of the rotating wave approximation (RWA) for our theory in Sec. IX and explain the practical calculation scheme in Sec. X. Finally, we compare our QED theory with other theories with the nonlocality and summarize the discussion in Sec. XI. We only explain the outline of our theory in the above sections and present detailed calculations of the derivation in Appendixes A–G. We discuss the second quantization of excitonic polarization in Appendix A and then derive the Hamiltonian from the microscopic point of view in Appendix B. In Appendix C, we extend the Maxwell wave equation discussed by SW[7] so that we can consider the excitonic polarization and derive the motion equations of excitons in Appendix D. We evaluate commutation relations in Appendix E and prove that the derived Green's tensor satisfies the wave equation with the nonlocal susceptibility in Appendix F. In Appendix G, in order to verify the validity of our theory, we calculate the equal-time commutation relations, which are expected from those of the Schrödinger operators. In this paper, we use mks units and Coulomb gauge.

II. PREVIOUS QED THEORIES WITH NONLOCAL SUSCEPTIBILITY

The series of QED theories enables us to discuss the linear optical process in arbitrary-structured 3D dielectrics characterized by a dielectric function $\epsilon(\mathbf{r},\omega)$. However, in order to discuss the materials with the nonlocality, it is necessary to consider more general elementary excitations that cannot be described by harmonic oscillators of the classical Hopfield model.[5]

As a pioneering study on a full-quantum theory with the nonlocality, Jenkins and Mukamel[37] discussed molecular crystals in d dimensions ($d=1,2,3$), where the relative motion of excitons is localized at a single molecule and the center of mass moves between molecules due to the dipole-dipole interaction. While their theory concentrates on treating the resonant polarization without nonradiative relaxation, recently, the nonlocality has been introduced into the field quantization in dispersive and absorptive media,[16,17,38–41] and some studies have demonstrated the application of their theories for specific structures.[17,38] Di Stefano *et al.*[38] discussed excitons with the nonlocality in media with spatial translation symmetries broken along one dimension, and they practically calculated the spatial and frequency dependences of the vacuum-field fluctuation in a semiconductor quantum-well structure. Thereafter, they extended their theory to an arbitrary 3D structure[39] and discussed the input-output relations in scattering systems.[40] On the other hand, Bechler[41] performed the field quantization for homogeneous systems with the nonlocality by using the path-integral method, and Suttorp[16] performed the same for nonlocal, inhomogeneous, and anisotropic systems by using the diagonalization method. Most recently, Raabe *et al.*[17] phenomenologically discussed the nonlocal systems with both dielectric and magnetic properties. They proposed 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 can be considered that a consistent framework for the field quantization in dielectrics with nonlocal susceptibility has already been established. Thus, the issue of current importance is to establish a general and practical calculation method applicable to arbitrary-structured 3D systems; this is desired for the actual applications of the above framework, although interesting applications have already been demonstrated in specific situations by Di Stefano *et al.*[38] and Raabe *et al.*[17] The essential task for this purpose is the derivation of Green's tensor for the Maxwell wave equation with the nonlocal susceptibility, as seen in Eq. (45) of the present paper.

In this paper, we provide a practical calculation method for the Green's tensor for arbitrary structures by using the fact that the nonlocal susceptibility is represented as a summation of separable functions with respect to two positions, as seen in Eq. (43). This technique has been developed in the semiclassical microscopic nonlocal theory.[25,26] We extend this theory to consider the quantum-mechanical properties of electromagnetic fields by using the Laplace transformation technique of SW[7] and introducing commutation relations of noise operators. In other words, we generalize the SW theory[7] to media with the nonlocality with providing a practical calculation method. Our theory consists of two equations in ω representation: the Maxwell wave equation and the motion equation of excitonic polarization. Both of them are derived by the Laplace transformation technique.

In Secs. III–X, we focus on the main part of our theory, and the lengthy derivations of the equations are given in Appendixes A–D for the purpose of brevity in the main part. We will then compare our theory with some of the above-mentioned theories in Sec. XI.

III. HAMILTONIAN

We describe the dielectric materials with resonant contributions from excitons with center-of-mass motion and the nonresonant ones with the local dielectric function $\epsilon_{bg}(\mathbf{r},\omega)$. 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 the optical and nonradiative-relaxation processes of the excitons, and the nonresonant backgrounds are treated by the same procedure as that of SW.[7] In Appendix B, we derive our Hamiltonian from the Lagrangian and obtain it as

$$H = H_{em} + H_{int} + H_{mat}, \tag{4}$$

where H_{em} describes the radiation field and background dielectric medium, H_{mat} the excitons and a reservoir of oscillators, and H_{int} the interactions between H_{em} and H_{mat}. H_{em} is the complete Hamiltonian discussed by SW,[7] and its representation is shown in Eq. (B9) of the present paper.

Based on the discussion in Appendix B, the interaction Hamiltonian is represented as

$$H_{int} = -\int d\mathbf{r}\left[\mathbf{I}_{ex}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) - \frac{1}{2}N_{ex}(\mathbf{r})\mathbf{A}^2(\mathbf{r})\right] + \int d\mathbf{r}\,\phi_{bg}(\mathbf{r})\rho_{ex}(\mathbf{r}) + \frac{1}{2}\int d\mathbf{r}\,\phi_{ex}(\mathbf{r})\rho_{ex}(\mathbf{r}). \tag{5}$$

$\mathbf{A}(\mathbf{r},t)$ is the vector potential and $\phi_{bg}(\mathbf{r},t)$ is the Coulomb potential induced in the background. $\mathbf{I}_{ex}(\mathbf{r})$ is the excitonic current density without radiation contribution $-N_{ex}(\mathbf{r})\mathbf{A}(\mathbf{r})$, i.e., the complete current density is written as $\mathbf{J}_{ex}(\mathbf{r})=\mathbf{I}_{ex}(\mathbf{r}) - N_{ex}(\mathbf{r})\mathbf{A}(\mathbf{r})$ (see Appendix A). $\rho_{ex}(\mathbf{r})$ is the excitonic charge density and

$$\phi_{ex}(\mathbf{r}) \equiv \int d\mathbf{r}'\frac{\rho_{ex}(\mathbf{r}')}{4\pi\epsilon_0|\mathbf{r}-\mathbf{r}'|} \tag{6}$$

is the Coulomb potential. The first and second terms of Eq. (5) represent the interaction between the radiation field and excitons. The third term represents the Coulomb interaction between the induced charges of the excitons and that of the background medium. The last term represents the interaction between the excitonic charges themselves, and it is also considered as the dipole-dipole interaction between excitonic polarizations or the exchange interaction between electrons and holes[26,42,43] (see Appendix 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 in a simple form as seen in Eq. (33) and will eliminate the explicit consideration of the longitudinal-transverse (LT) splitting of the exciton eigenenergies because the last term of Eq. (5) is just the origin of the LT splitting.

With regard to the excitons, we generally describe them starting from the basis of electrons and holes interacting with each other and themselves, as discussed in Appendix B. However, as long as we consider the optical processes of excitons under weak excitation, it is valid to describe electronic systems in terms of excitonic eigenstates and put the nonresonant (background) contributions into $H_{\rm em}$. In addition, in order to describe the nonradiative-relaxation process, we consider a reservoir of oscillators interacting with excitons. Then, we consider the matter Hamiltonian represented as

$$H_{\rm mat}=\sum_\mu \hbar\omega_\mu b_\mu^\dagger b_\mu+\sum_\mu\int_0^\infty d\Omega\{\hbar\Omega d_\mu^\dagger(\Omega)d_\mu(\Omega)+[b_\mu+b_\mu^\dagger]\times[g_\mu(\Omega)d_\mu(\Omega)+g_\mu^*(\Omega)d_\mu^\dagger(\Omega)]\}, \tag{7}$$

where b_μ is the annihilation operator of the excitons in eigenstate μ with eigenfrequency ω_μ; this does not include the LT splitting because we displace the exchange interaction between electrons and holes from $H_{\rm mat}$ to $H_{\rm int}$. In this paper, we assume that the center-of-mass motion of excitons is confined in finite spaces, and index μ represents the 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

$$[b_\mu, b_{\mu'}^\dagger]=\delta_{\mu,\mu'}, \tag{8a}$$

$$[b_\mu, b_{\mu'}]=0. \tag{8b}$$

This approximation is valid under weak excitation. On the other hand, in Eq. (7), $d_\mu(\Omega)$ 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 following commutation relations:

$$[d_\mu(\Omega), d_{\mu'}^\dagger(\Omega')]=\delta_{\mu,\mu'}\delta(\Omega-\Omega'), \tag{9a}$$

$$[d_\mu(\Omega), d_{\mu'}(\Omega')]=0. \tag{9b}$$

IV. MAXWELL WAVE EQUATION

One of the fundamental equations of our theory is the Maxwell wave equation, which is derived in Appendix C along with the quantization scheme of SW[7] and has a noise current-density operator. In this section, we show the equation and commutation relation of the noise operator and discuss its similarities and differences as compared to the SW theory.[7]

Since we consider the matter system to be a combination of excitons and the background medium, the electric field contains Coulomb potentials induced by excitons and the background, and it is represented as

$$\mathbf{E}(\mathbf{r},t)=-\frac{\partial}{\partial t}\mathbf{A}(\mathbf{r},t)-\nabla\phi_{\rm bg}(\mathbf{r},t)-\nabla\phi_{\rm ex}(\mathbf{r},t). \tag{10}$$

Because the Coulomb gauge is used in our scheme, the vector potential is a transverse field satisfying $\nabla\cdot\mathbf{A}(\mathbf{r})=0$, and the second and third terms represent the longitudinal fields. In Appendix C, from the Laplace transform of the Heisenberg equations of the system variables, we derive the Maxwell wave equation for the Fourier component of the electric field as

$$\nabla\times\nabla\times\hat{\mathbf{E}}^+(\mathbf{r},\omega)-\frac{\omega^2}{c^2}\epsilon_{\rm bg}(\mathbf{r},\omega)\hat{\mathbf{E}}^+(\mathbf{r},\omega)=i\mu_0\omega\hat{\mathbf{J}}_0(\mathbf{r},\omega)+\mu_0\omega^2\hat{\mathbf{P}}_{\rm ex}^+(\mathbf{r},\omega), \tag{11}$$

where $\hat{\mathbf{E}}^+(\mathbf{r},\omega)$ is the positive-frequency Fourier component of $\mathbf{E}(\mathbf{r},t)$ and $\hat{\mathbf{E}}^-(\mathbf{r},\omega)$ is the negative-frequency one. They are defined as

$$\hat{\mathbf{E}}^\pm(\mathbf{r},\omega)\equiv\frac{1}{2\pi}\int_{-\infty}^{\infty}dt e^{\pm i\omega t}\mathbf{E}(\mathbf{r},t) \tag{12}$$

and satisfy

$$\hat{\mathbf{E}}^\pm(\mathbf{r},\omega)=\hat{\mathbf{E}}^\mp(\mathbf{r},-\omega)=\{\hat{\mathbf{E}}^\pm(\mathbf{r},-\omega^*)\}^\dagger. \tag{13}$$

We represent Fourier-transformed operators with a hat (ˆ) in this paper. In Eq. (11), the matter information is described by the dielectric function $\epsilon_{\rm bg}(\mathbf{r},\omega)$ of the background system and the excitonic polarization density $\hat{\mathbf{P}}_{\rm ex}^+(\mathbf{r},\omega)$, which is an additional polarization as compared to the SW theory.[7] On the other hand, as compared to the classical electrodynamics, Eq. (11) has an additional operator $\hat{\mathbf{J}}_0(\mathbf{r},\omega)$ on the right-hand side (RHS). This is called the noise current density and it is interpreted as a source of the electromagnetic fields or the fluctuation caused by absorption in the background. It plays an essential role in the series of QED theories for dispersive and absorptive dielectrics. The same type of operator for homogeneous systems has been derived by HB,[6] and the one for inhomogeneous 3D systems has been phenomenologically introduced in Ref. 44. On the other hand, from the Laplace-transformed motion equations of system variables, SW[7] systematically derived the representation of the operator, which is represented in terms of the canonical variables and momenta of the system at $t=0$. In the present paper, $\hat{\mathbf{J}}_0(\mathbf{r},\omega)$ is derived as Eq. (C26) and satisfies

$$\hat{\mathbf{J}}_0(\mathbf{r},\omega)=\{\hat{\mathbf{J}}_0(\mathbf{r},-\omega^*)\}^\dagger \tag{14}$$

as the electric field satisfies Eq. (13). Although SW[7] calculated the commutation relation of their noise operator from those of the system operators, we introduce the commutation relation of $\hat{\mathbf{J}}_0(\mathbf{r},\omega)$ as

$$[\hat{\mathbf{J}}_0(\mathbf{r},\omega),\{\hat{\mathbf{J}}_0(\mathbf{r}',\omega'^*)\}^\dagger]=[\hat{\mathbf{J}}_0(\mathbf{r},\omega),\hat{\mathbf{J}}_0(\mathbf{r}',-\omega')]=\delta(\omega-\omega')\delta(\mathbf{r}-\mathbf{r}')\times\frac{\epsilon_0\hbar\omega^2}{\pi}\mathrm{Im}[\epsilon_{\rm bg}(\mathbf{r},\omega)]\mathbf{1}, \tag{15}$$

where $[\hat{\mathbf{J}}_0,\{\hat{\mathbf{J}}_0\}^\dagger]$ is a 3×3 tensor and its (ξ,ξ') element implies $[\{\hat{J}_0\}_\xi,\{\hat{J}_0^\dagger\}_{\xi'}]$ for $\xi=x,y,z$. In the present paper, we will discuss the validity of commutation relation (15) from the validity of those for excitons and the electric field derived in Sec. VIII.

Next, we discuss the relation between the QED theories and the correlation function theory. Using Green's tensor $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ satisfying

$$\nabla\times\nabla\times\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)-\frac{\omega^2}{c^2}\epsilon_{\rm bg}(\mathbf{r},\omega)\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)=\delta(\mathbf{r}-\mathbf{r}')\mathbf{1}, \tag{16}$$

we can rewrite Eq. (11) as

$$\hat{\mathbf{E}}^{+}(\mathbf{r},\omega)=\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega)+\mu_0\omega^2\int d\mathbf{r}'\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{P}}_{\rm ex}^{+}(\mathbf{r}',\omega), \tag{17}$$

where $\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega)$ is the background field, the electric field in the $H_{\rm em}$ system, and it is defined as

$$\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega)\equiv i\mu_0\omega\int d\mathbf{r}'\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{j}}_0(\mathbf{r}',\omega). \tag{18}$$

In the classical electrodynamics, this is usually introduced as a homogeneous solution satisfying

$$\nabla\times\nabla\times\langle\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega)\rangle-\frac{\omega^2}{c^2}\epsilon_{\rm bg}(\mathbf{r},\omega)\langle\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega)\rangle=\mathbf{0} \tag{19}$$

and is considered as an incident field for excitons. On the other hand, since the background dielectric function satisfies

$$\epsilon_{\rm bg}(\mathbf{r},\omega)=\{\epsilon_{\rm bg}(\mathbf{r},-\omega^*)\}^*=\epsilon_{\rm bg}^*(\mathbf{r},-\omega), \tag{20}$$

the Green's tensor also satisfies

$$\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)=\{\mathbf{G}(\mathbf{r},\mathbf{r}',-\omega^*)\}^*=\mathbf{G}^*(\mathbf{r},\mathbf{r}',-\omega). \tag{21}$$

Then, the negative-frequency Fourier component of the electric field is represented as

$$\hat{\mathbf{E}}^{-}(\mathbf{r},\omega)=\hat{\mathbf{E}}^{+}(\mathbf{r},-\omega)=\hat{\mathbf{E}}_0^{-}(\mathbf{r},\omega)+\mu_0\omega^2\int d\mathbf{r}'\mathbf{G}^*(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{P}}_{\rm ex}^{-}(\mathbf{r}',\omega), \tag{22}$$

and the background field is given as

$$\hat{\mathbf{E}}_0^{-}(\mathbf{r},\omega)\equiv -i\mu_0\omega\int d\mathbf{r}'\mathbf{G}^*(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{j}}_0(\mathbf{r}',-\omega). \tag{23}$$

Since $\hat{\mathbf{j}}_0(\mathbf{r},\omega)$ satisfies Eq. (14), $\hat{\mathbf{E}}_0^{\pm}(\mathbf{r},\omega)$ also satisfies the following relation as $\hat{\mathbf{E}}(\mathbf{r},\omega)$ satisfies Eq. (13):

$$\hat{\mathbf{E}}_0^{\pm}(\mathbf{r},\omega)=\hat{\mathbf{E}}_0^{\mp}(\mathbf{r},-\omega)=\{\hat{\mathbf{E}}_0^{\pm}(\mathbf{r},-\omega^*)\}^{\dagger}. \tag{24}$$

Furthermore, from the commutation relation (15) of $\hat{\mathbf{j}}_0(\mathbf{r},\omega)$, that of $\hat{\mathbf{E}}_0^{\pm}(\mathbf{r},\omega)$ can be derived as

$$\begin{aligned}[\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega),\hat{\mathbf{E}}_0^{-}(\mathbf{r}',\omega')]&=[\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega),\hat{\mathbf{E}}_0^{+}(\mathbf{r}',-\omega')]\\&=\delta(\omega-\omega')\frac{\mu_0\hbar\omega^2}{i2\pi}[\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)-\mathbf{G}^*(\mathbf{r},\mathbf{r}',\omega)],\end{aligned} \tag{25}$$

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

$$\int d\mathbf{s}\frac{\omega^2}{c^2}[\epsilon_{\rm bg}(\mathbf{s},\omega)-\epsilon_{\rm bg}^*(\mathbf{s},\omega)]\mathbf{G}(\mathbf{r},\mathbf{s},\omega)\cdot\mathbf{G}^*(\mathbf{s},\mathbf{r}',\omega)=\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)-\mathbf{G}^*(\mathbf{r},\mathbf{r}',\omega), \tag{26}$$

and the reciprocity relation,

$$\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)=\{\mathbf{G}(\mathbf{r}',\mathbf{r},\omega)\}^t. \tag{27}$$

Equations (25) can be understood by the fact that in the background system, Green's tensor $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ identifies with the retarded correlation function of the electric field.[45] This means that when we define the background field in the time domain as

$$\begin{aligned}\mathbf{E}_0(\mathbf{r},t)&\equiv\int_0^\infty d\omega[e^{-i\omega t}\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega)+e^{i\omega t}\hat{\mathbf{E}}_0^{-}(\mathbf{r},\omega)]\\&=\int_{-\infty}^\infty d\omega e^{-i\omega t}\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega),\end{aligned} \tag{28}$$

the Green's tensor can be represented as

$$-\mu_0\hbar\omega^2\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)=-i\int_{t'}^\infty dte^{i\omega(t-t')}\langle[\mathbf{E}_0(\mathbf{r},t),\mathbf{E}_0(\mathbf{r}',t')]\rangle. \tag{29}$$

We can verify that this representation satisfies Eq. (25). Therefore, because of the causality seen in Eq. (29), $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ satisfies the Kramers-Kronig relation and has no pole in the upper half of the complex ω plane.

V. MOTION EQUATION OF EXCITONS

Next, we discuss the motion of the excitonic polarization $\hat{\mathbf{P}}_{\rm ex}^{+}(\mathbf{r},\omega)$ in the Maxwell wave equation [Eq. (11)]. In Appendix A, we derive the second-quantized form of it in terms of the exciton operator set $\{b_\mu\}$ as

$$\mathbf{P}_{\rm ex}(\mathbf{r})=\sum_\mu[\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})b_\mu+\boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r})b_\mu^\dagger], \tag{30}$$

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

$$\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})=\mathcal{P}_\mu\mathbf{e}_\mu G_\mu(\mathbf{r}). \tag{31}$$

$\mathcal{P}_\mu$ is the transition dipole moment; $\mathbf{e}_\mu$, a unit vector in the polarization direction; and $G_\mu(\mathbf{r})$, the wave function of the center-of-mass motion in exciton state μ. Since we assume the weak confinement regime, $\mathcal{P}_\mu$ approximately depends only on the relative motion of excitons and is related to the LT splitting as $\Delta_{\rm LT}^\mu=|\mathcal{P}_\mu|^2/\epsilon_0\epsilon_{\rm bg}$.

In Appendix D, we derive the Heisenberg equation of excitons from Hamiltonians (5) and (7) and transform it into an equation for the Fourier component of the exciton operator,

$$\hat{b}_\mu(\omega)\equiv\frac{1}{2\pi}\int_{-\infty}^\infty dte^{i\omega t}b_\mu(t). \tag{32}$$

The transformed equation is obtained as

$$[\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2]\hat{b}_\mu(\omega) + [-i\gamma_\mu(\omega)/2]\{\hat{b}_\mu(-\omega^*)\}^\dagger = \int d\mathbf{r}\boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r})\cdot\hat{\mathbf{E}}^+(\mathbf{r},\omega) + \hat{\mathcal{D}}_\mu(\omega), \quad (33)$$

where $\gamma_\mu(\omega)$ is the nonradiative-relaxation width defined as Eq. (D7) in terms of the exciton-reservoir interaction coefficient $g_\mu(\Omega)$, and it satisfies

$$\gamma_\mu(\omega) = -\{\gamma_\mu(-\omega^*)\}^* = -\gamma_\mu^*(-\omega). \quad (34)$$

In the calculation for analyzing practical materials, we usually give real values to $\{\gamma_\mu(\omega)\}$ themselves as fitting parameters, but we do not estimate them from definition (D7) for given coefficients $\{g_\mu(\Omega)\}$, which enter into our scheme only via $\{\gamma_\mu(\omega)\}$. On the other hand, operator $\hat{\mathcal{D}}_\mu(\omega)$ represents the fluctuation caused by the reservoir oscillators. Its definition is shown in Eq. (D8) and it also satisfies

$$\hat{\mathcal{D}}_\mu(\omega) = \{\hat{\mathcal{D}}_\mu(-\omega^*)\}^\dagger. \quad (35)$$

From the fluctuation dissipation theorem, we introduce their commutation relation as

$$[\hat{\mathcal{D}}_\mu(\omega),\{\hat{\mathcal{D}}_{\mu'}(\omega'^*)\}^\dagger] = [\hat{\mathcal{D}}_\mu(\omega),\hat{\mathcal{D}}_{\mu'}(-\omega')] = \delta_{\mu,\mu'}\delta(\omega-\omega')\frac{\hbar}{i2\pi}\frac{i\gamma_\mu(\omega)+i\gamma_\mu^*(\omega)}{2}. \quad (36)$$

$\hat{\mathcal{D}}_\mu(\omega)$ is another noise operator of our system and is independent of the noise current density $\hat{\mathbf{J}}_0(\mathbf{r},\omega)$, and it satisfies

$$[\hat{\mathcal{D}}_\mu(\omega),\hat{\mathbf{J}}_0(\mathbf{r},\omega')] = [\hat{\mathcal{D}}_\mu(\omega),\{\hat{\mathbf{J}}_0(\mathbf{r},\omega'^*)\}^\dagger] = \mathbf{0}. \quad (37)$$

The validities of Eqs. (36), (37), and (15) will be discussed in Sec. VIII.

Motion [Eq. (33)] of the excitons is rewritten with its Hermite conjugate as

$$\underline{\mathbf{S}}_\mu(\omega)\begin{bmatrix}\hat{b}_\mu(\omega)\\ \{\hat{b}_\mu(-\omega^*)\}^\dagger\end{bmatrix} = \int d\mathbf{r}\begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r})\\ \boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\end{bmatrix}\cdot\hat{\mathbf{E}}^+(\mathbf{r},\omega) + \begin{bmatrix}1\\1\end{bmatrix}\hat{\mathcal{D}}_\mu(\omega), \quad (38)$$

where the coefficient matrix is

$$\underline{\mathbf{S}}_\mu(\omega) = \begin{bmatrix}\hbar\omega_\mu-\hbar\omega-i\gamma_\mu(\omega)/2 & -i\gamma_\mu(\omega)/2\\ -i\gamma_\mu(\omega)/2 & \hbar\omega_\mu+\hbar\omega-i\gamma_\mu(\omega)/2\end{bmatrix}. \quad (39)$$

Here, by introducing an inverse matrix $\underline{\mathbf{W}}_\mu(\omega)\equiv\underline{\mathbf{S}}_\mu^{-1}(\omega)$, Eq. (38) becomes

$$\begin{bmatrix}\hat{b}_\mu(\omega)\\ \{\hat{b}_\mu(-\omega^*)\}^\dagger\end{bmatrix} = \int d\mathbf{r}\underline{\mathbf{W}}_\mu(\omega)\begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu^*(r)\\ \boldsymbol{\mathcal{P}}_\mu(r)\end{bmatrix}\cdot\hat{\mathbf{E}}^+(\mathbf{r},\omega) + \underline{\mathbf{W}}_\mu(\omega)\times\begin{bmatrix}1\\1\end{bmatrix}\hat{\mathcal{D}}_\mu(\omega). \quad (40)$$

Now, we substitute Eq. (40) into the ω representation of the polarization density (30),

$$\hat{\mathbf{P}}_{\mathrm{ex}}^+(\mathbf{r},\omega) = \sum_\mu[\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\hat{b}_\mu(\omega) + \boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r})\{\hat{b}_\mu(-\omega^*)\}^\dagger], \quad (41)$$

then, we obtain

$$\hat{\mathbf{P}}_{\mathrm{ex}}^+(\mathbf{r},\omega) = \epsilon_0\int d\mathbf{r}'\boldsymbol{\chi}_{\mathrm{ex}}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{E}}^+(\mathbf{r},\omega) + \sum_\mu\begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\\ \boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r})\end{bmatrix}^t\underline{\mathbf{W}}_\mu(\omega)\begin{bmatrix}1\\1\end{bmatrix}\hat{\mathcal{D}}_\mu(\omega), \quad (42)$$

where the susceptibility has a nonlocal form as

$$\boldsymbol{\chi}_{\mathrm{ex}}(\mathbf{r},\mathbf{r}',\omega) = \frac{1}{\epsilon_0}\sum_\mu\begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\\ \boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r})\end{bmatrix}^t\underline{\mathbf{W}}_\mu(\omega)\begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r}')\\ \boldsymbol{\mathcal{P}}_\mu(\mathbf{r}')\end{bmatrix}. \quad (43)$$

Since this function can be directly derived from the motion [Eq. (D1)] of excitons and that [Eq. (D3)] of reserver oscillators with satisfying the causality, $\boldsymbol{\chi}_{\mathrm{ex}}(\mathbf{r},\mathbf{r}',\omega)$ satisfies the Kramers-Kronig relations and has no pole in the upper half of the ω plane, and it also satisfies

$$\boldsymbol{\chi}_{\mathrm{ex}}(\mathbf{r},\mathbf{r}',\omega) = \{\boldsymbol{\chi}_{\mathrm{ex}}(\mathbf{r},\mathbf{r}',-\omega^*)\}^*. \quad (44)$$

However, it does not have a reciprocity relation $\boldsymbol{\chi}_{\mathrm{ex}}(\mathbf{r},\mathbf{r}',\omega)\neq\{\boldsymbol{\chi}_{\mathrm{ex}}(\mathbf{r}',\mathbf{r},\omega)\}^t$ because of the general anisotropy of excitons. The spatial spreading of the exciton state, the origin of the nonlocality, is reflected through the polarization coefficient $\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})$ or the center-of-mass wave function $G_\mu(\mathbf{r})$. On the other hand, the spatial structure of the background dielectrics is characterized by the dielectric function $\epsilon_{\mathrm{bg}}(\mathbf{r},\omega)$ in the Maxwell wave equation [Eq. (11)] and in commutation relation [Eq. (15)]. In our framework, we can discuss arbitrary-structured exciton motions and background dielectrics through these functions.

VI. MAXWELL WAVE EQUATION WITH NONLOCAL SUSCEPTIBILITY

In order to discuss the optical processes of excitons, we must simultaneously solve the Maxwell wave equation [Eq. (11)] and the motion equation [Eq. (42)] of the polarization density to describe the unknown variables $\hat{\mathbf{E}}^+(\mathbf{r},\omega)$ and $\hat{\mathbf{P}}_{\mathrm{ex}}^+(\mathbf{r},\omega)$ in terms of the noise operators $\hat{\mathbf{J}}_0(\mathbf{r},\omega)$ and $\hat{\mathcal{D}}_\mu(\omega)$. Substituting Eq. (42) into Eq. (11), we obtain a wave equation with the nonlocal susceptibility as

$$\nabla \times \nabla \times \hat{\mathbf{E}}^{+}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\epsilon_{\text{bg}}(\mathbf{r},\omega)\hat{\mathbf{E}}^{+}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\int d\mathbf{r}'\boldsymbol{\chi}_{\text{ex}}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{E}}^{+}(\mathbf{r}',\omega) = i\mu_0\omega\hat{\mathbf{J}}_0'(\mathbf{r},\omega), \tag{45}$$

where we define another noise operator comprising $\hat{\mathbf{J}}_0(\mathbf{r},\omega)$ and the second term of Eq. (42) as

$$\hat{\mathbf{J}}_0'(\mathbf{r},\omega) = \hat{\mathbf{J}}_0(\mathbf{r},\omega) - i\omega\sum_{\mu}\begin{bmatrix}\boldsymbol{\mathcal{P}}_{\mu}(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_{\mu}^{*}(\mathbf{r})\end{bmatrix}^{t}\underline{\mathbf{W}}_{\mu}(\omega)\begin{bmatrix}1\\1\end{bmatrix}\hat{\mathcal{D}}_{\mu}(\omega). \tag{46}$$

From Eqs. (14), (34), and (35), this operator also satisfies

$$\hat{\mathbf{J}}_0'(\mathbf{r},\omega) = \{\hat{\mathbf{J}}_0'(\mathbf{r},-\omega^*)\}^{\dagger}. \tag{47}$$

In Appendix E, from commutation relations (15), (36), and (37), we calculate that for $\hat{\mathbf{J}}_0'(\mathbf{r},\omega)$ as

$$[\hat{\mathbf{J}}_0'(\mathbf{r},\omega),\{\hat{\mathbf{J}}_0'(\mathbf{r}',\omega'^*)\}^{\dagger}] = \delta(\omega-\omega')\frac{\epsilon_0\hbar\omega^2}{i2\pi}[\boldsymbol{\epsilon}(\mathbf{r},\mathbf{r}',\omega) - \boldsymbol{\epsilon}^{*t}(\mathbf{r}',\mathbf{r},\omega)], \tag{48}$$

where $\boldsymbol{\epsilon}(\mathbf{r},\mathbf{r}',\omega)$ is the dielectric tensor defined as

$$\boldsymbol{\epsilon}(\mathbf{r},\mathbf{r}',\omega) = \delta(\mathbf{r}-\mathbf{r}')\epsilon_{\text{bg}}(\mathbf{r},\omega)\mathbf{1} + \boldsymbol{\chi}_{\text{ex}}(\mathbf{r},\mathbf{r}',\omega). \tag{49}$$

The wave equation [Eq. (45)] and commutation relation [Eq. (48)] are just the ones discussed by Savasta and co-workers[39,40] and they also have the same forms as those of Raabe *et al.*[17] Further, Eq. (48) can be understood as a natural result from the fluctuation theorem, as discussed in Refs. 17, 39, and 40. Along the lines of these studies, the problem reduces to finding a Green's tensor satisfying

$$\nabla \times \nabla \times \mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega) - \frac{\omega^2}{c^2}\epsilon_{\text{bg}}(\mathbf{r},\omega)\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega) - \frac{\omega^2}{c^2}\int d\mathbf{r}''\boldsymbol{\chi}_{\text{ex}}(\mathbf{r},\mathbf{r}'',\omega)\cdot\mathbf{G}_{\text{ren}}(\mathbf{r}'',\mathbf{r}',\omega) = \delta(\mathbf{r}-\mathbf{r}')\mathbf{1}. \tag{50}$$

This tensor renormalizes the linear optical process of excitons with the nonlocality and enables us to rewrite Eq. (45) as

$$\hat{\mathbf{E}}^{+}(\mathbf{r},\omega) = i\mu_0\omega\int d\mathbf{r}'\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{J}}_0'(\mathbf{r}',\omega). \tag{51}$$

However, it appears very difficult to solve this nonlocal equation in the practical application of their theories. This problem can be solved by using the fact that the nonlocal susceptibility (43) is represented as a summation of separable functions with respect to $\mathbf{r}$ and $\mathbf{r}'$. One scheme is to directly derive $\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)$, as discussed in Ref. 46, and the other is to reduce this integrodifferential equation into a simultaneous linear equation set.[25,26] In our QED theory, we adopt the latter scheme because it provides not only the Green's tensor of the former but also considerable interesting information about exciton-polariton systems.

VII. SELF-CONSISTENT EQUATION SET

Instead of solving the integrodifferential equation [Eq. (50)], we reduce the problem into a linear equation set by using the same technique as the microscopic nonlocal theory developed in the semiclassical framework.[25,26] Substituting representation (17) of the electric field into the motion equation [Eq. (33)] of excitons by expanding $\hat{\mathbf{P}}_{\text{ex}}^{+}(\mathbf{r},\omega)$ as Eq. (41), we obtain a linear equation with respect to exciton amplitudes $\hat{b}_{\mu}(\omega)$ and $\{\hat{b}_{\mu}(-\omega^*)\}^{\dagger}$ as

$$\sum_{\mu'}[S_{\mu,\mu'}(\omega)\hat{b}_{\mu'}(\omega) + T_{\mu,\mu'}(\omega)\{\hat{b}_{\mu'}(-\omega^*)\}^{\dagger}] = \int d\mathbf{r}\boldsymbol{\mathcal{P}}_{\mu}^{*}(\mathbf{r})\cdot\hat{\mathbf{E}}_0^{+}(\mathbf{r},\omega) + \hat{\mathcal{D}}_{\mu}(\omega), \tag{52}$$

where the coefficient matrix elements are defined as

$$S_{\mu,\mu'}(\omega) \equiv [\hbar\omega_{\mu} - \hbar\omega - i\gamma_{\mu}(\omega)/2]\delta_{\mu,\mu'} + \mathcal{A}_{\mu,\mu'}(\omega), \tag{53}$$

$$T_{\mu,\mu'}(\omega) \equiv [-i\gamma_{\mu}(\omega)/2]\delta_{\mu,\mu'} + \mathcal{B}_{\mu,\mu'}(\omega). \tag{54}$$

The first term on the RHS of Eq. (52) can be interpreted as an exciton amplitude directly induced by the background electric field. Here, we use the word "directly" to mean that the term does not include the diffusion of the exciton amplitudes via the electromagnetic fields. Such an effect is reflected through the correction terms appearing in Eqs. (53) and (54),

$$\mathcal{A}_{\mu,\mu'}(\omega) = -\mu_0\omega^2\int d\mathbf{r}d\mathbf{r}'\boldsymbol{\mathcal{P}}_{\mu}^{*}(\mathbf{r})\cdot\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\boldsymbol{\mathcal{P}}_{\mu'}(\mathbf{r}'), \tag{55}$$

$$\mathcal{B}_{\mu,\mu'}(\omega) = -\mu_0\omega^2\int d\mathbf{r}d\mathbf{r}'\boldsymbol{\mathcal{P}}_{\mu}^{*}(\mathbf{r})\cdot\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\boldsymbol{\mathcal{P}}_{\mu'}^{*}(\mathbf{r}'). \tag{56}$$

These can be interpreted as follows: the polarization at $\mathbf{r}'$ induces an 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 gives the LT splitting of the exciton eigenenergies.

From Eq. (52) and its Hermite conjugate, we obtain a linear equation set for $\hat{b}_{\mu}(\omega)$ and $\{\hat{b}_{\mu}(-\omega^*)\}^{\dagger}$ as

$$\sum_{\mu'}\begin{bmatrix} S_{\mu,\mu'}(\omega) & T_{\mu,\mu'}(\omega) \\ T^*_{\mu,\mu'}(-\omega) & S^*_{\mu,\mu'}(-\omega)\end{bmatrix}\begin{bmatrix}\hat{b}_{\mu'}(\omega) \\ \{\hat{b}_{\mu'}(-\omega^*)\}^\dagger\end{bmatrix} = \int d\mathbf{r}\begin{bmatrix}\boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\end{bmatrix}\cdot\hat{\mathbf{E}}^+_0(\mathbf{r},\omega) + \begin{bmatrix}1\\1\end{bmatrix}\hat{\mathcal{D}}_\mu(\omega). \quad (57)$$

This equation is called the self-consistent equation set and has the same form as that in the semiclassical theory.[26] By calculating the inverse of the coefficient matrix, the exciton operators are written as

$$\begin{bmatrix}\hat{b}_\mu(\omega) \\ \{\hat{b}_\mu(-\omega^*)\}^\dagger\end{bmatrix} = \sum_{\mu'}\begin{bmatrix} W_{\mu,\mu'}(\omega) & Z_{\mu,\mu'}(\omega) \\ Z^*_{\mu,\mu'}(-\omega) & W^*_{\mu,\mu'}(-\omega)\end{bmatrix} \times\left\{\int d\mathbf{r}\begin{bmatrix}\boldsymbol{\mathcal{P}}^*_{\mu'}(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_{\mu'}(\mathbf{r})\end{bmatrix}\cdot\hat{\mathbf{E}}^+_0(\mathbf{r},\omega) + \begin{bmatrix}1\\1\end{bmatrix}\hat{\mathcal{D}}_{\mu'}(\omega)\right\}, \quad (58)$$

where the inverse matrix has the same symmetry as the original one, and they satisfy

$$\sum_{\nu}\begin{bmatrix} S_{\mu,\nu}(\omega) & T_{\mu,\nu}(\omega) \\ T^*_{\mu,\nu}(-\omega) & S^*_{\mu,\nu}(-\omega)\end{bmatrix}\begin{bmatrix} W_{\nu,\mu'}(\omega) & Z_{\nu,\mu'}(\omega) \\ Z^*_{\nu,\mu'}(-\omega) & W^*_{\nu,\mu'}(-\omega)\end{bmatrix} = \sum_{\nu}\begin{bmatrix} W_{\mu,\nu}(\omega) & Z_{\mu,\nu}(\omega) \\ Z^*_{\mu,\nu}(-\omega) & W^*_{\mu,\nu}(-\omega)\end{bmatrix}\begin{bmatrix} S_{\nu,\mu'}(\omega) & T_{\nu,\mu'}(\omega) \\ T^*_{\nu,\mu'}(-\omega) & S^*_{\nu,\mu'}(-\omega)\end{bmatrix} = \delta_{\mu,\mu'}\mathbf{1}. \quad (59)$$

We can describe the other physical variables in terms of these exciton operators and $\hat{\mathbf{E}}^+_0(\mathbf{r},\omega)$ in our system. For example, we can represent the excitonic polarization by Eq. (41), and the electric field (17) as

$$\hat{\mathbf{E}}^+(\mathbf{r},\omega) = \hat{\mathbf{E}}^+_0(\mathbf{r},\omega) + \sum_\mu [\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)\hat{b}_\mu(\omega) - \boldsymbol{\mathcal{E}}^*_\mu(\mathbf{r},-\omega)\{\hat{b}_\mu(-\omega^*)\}^\dagger], \quad (60)$$

where the coefficients are defined as

$$\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega) \equiv \mu_0\omega^2\int d\mathbf{r}'\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\boldsymbol{\mathcal{P}}_\mu(\mathbf{r}'), \quad (61)$$

$$\boldsymbol{\mathcal{F}}_\mu(\mathbf{r},\omega) \equiv \mu_0\omega^2\int d\mathbf{r}'\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\cdot\boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r}'), \quad (62)$$

and because of Eq. (21), there exists a relation

$$\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega) = \boldsymbol{\mathcal{F}}^*_\mu(\mathbf{r},-\omega) = \{\boldsymbol{\mathcal{F}}_\mu(\mathbf{r},-\omega^*)\}^*. \quad (63)$$

VIII. COMMUTATION RELATIONS

In Sec. VII, we describe the exciton operators in terms of $\hat{\mathbf{E}}^+_0(\mathbf{r},\omega)$ and $\hat{\mathcal{D}}_\mu(\omega)$. In Appendix E, we calculate the commutation relations of excitons from those of $\hat{\mathbf{E}}^\pm_0(\mathbf{r},\omega)$ and $\hat{\mathcal{D}}_\mu(\omega)$ [Eqs. (25), (36), and (37)]. As a result, those are represented by elements of the inverse matrix (59),

$$[\hat{b}_\mu(\omega),\{\hat{b}_{\mu'}(\omega'^*)\}^\dagger] = \delta(\omega-\omega')\frac{\hbar}{i2\pi}[W_{\mu,\mu'}(\omega) - W^*_{\mu',\mu}(\omega)], \quad (64a)$$

$$[\hat{b}_\mu(\omega),\hat{b}_{\mu'}(-\omega')] = \delta(\omega-\omega')\frac{\hbar}{i2\pi}[Z_{\mu,\mu'}(\omega) - Z_{\mu',\mu}(-\omega)]. \quad (64b)$$

In addition, we also derive the commutation relation of the electric-field operators [Eq. (60)],

$$[\hat{\mathbf{E}}^+(\mathbf{r},\omega),\hat{\mathbf{E}}^-(\mathbf{r}',\omega')] = [\hat{\mathbf{E}}^+(\mathbf{r},\omega),\hat{\mathbf{E}}^+(\mathbf{r}',-\omega')] = \delta(\omega-\omega')\frac{\mu_0\hbar\omega^2}{i2\pi}\times[\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega) - \mathbf{G}^{*t}_{\text{ren}}(\mathbf{r}',\mathbf{r},\omega)], \quad (65)$$

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

$$\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega) = \mathbf{G}(\mathbf{r},\mathbf{r}',\omega) + \frac{1}{\mu_0\omega^2}\sum_{\mu,\mu'}\{\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)W_{\mu,\mu'}(\omega)\times\boldsymbol{\mathcal{F}}_{\mu'}(\mathbf{r}',\omega) + \boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)Z_{\mu,\mu'}(\omega)\boldsymbol{\mathcal{E}}_{\mu'}(\mathbf{r}',\omega) + \boldsymbol{\mathcal{E}}^*_\mu(\mathbf{r},-\omega)W^*_{\mu,\mu'}(-\omega)\boldsymbol{\mathcal{F}}^*_{\mu'}(\mathbf{r}',-\omega) + \boldsymbol{\mathcal{E}}^*_\mu(\mathbf{r},-\omega)Z^*_{\mu,\mu'}(-\omega)\boldsymbol{\mathcal{E}}^*_{\mu'}(\mathbf{r}',-\omega)\}. \quad (66)$$

We can find that this tensor satisfies

$$\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega) = \{\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',-\omega^*)\}^* = \mathbf{G}^*_{\text{ren}}(\mathbf{r},\mathbf{r}',-\omega) \quad (67)$$

but does not satisfy the reciprocity relation (27) because of the anisotropic susceptibility tensor (43) of the excitonic polarization. In Appendix F, we verify that $\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)$ satisfies the wave equation [Eq. (50)], and it can be interpreted as the Green's tensor for Maxwell wave equation [Eq. (45)] with the nonlocal susceptibility. This fact shows the validities of commutation relations (15), (36), and (37) introduced from the fluctuation dissipation theorem.

As discussed in Sec. IV, the Green's tensor $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ is the retarded correlation function for the electric field in the background system. In the same manner as this relation, commutation relation (65) indicates that the Green's tensor $\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)$ identifies with the Fourier transform of the retarded correlation function of the electric field in our entire system,

$$-\mu_0\hbar\omega^2\mathbf{G}_{\rm ren}(\mathbf{r},\mathbf{r}',\omega) = -i\int_{t'}^{\infty} dt e^{i\omega(t-t')}\langle[\mathbf{E}(\mathbf{r},t),\mathbf{E}(\mathbf{r}',t')]\rangle. \tag{68}$$

In addition, commutation relations (64a) and (64b) means that $W_{\mu,\mu'}(\omega)$ and $Z_{\mu,\mu'}(\omega)$ identify with Fourier transforms of the retarded correlation functions of exciton operators,

$$-\hbar W_{\mu,\mu'}(\omega) = -i\int_{t'}^{\infty} dt e^{i\omega(t-t')}\langle[b_\mu(t),b^\dagger_{\mu'}(t')]\rangle, \tag{69}$$

$$-\hbar Z_{\mu,\mu'}(\omega) = -i\int_{t'}^{\infty} dt e^{i\omega(t-t')}\langle[b_\mu(t),b_{\mu'}(t')]\rangle. \tag{70}$$

Therefore, $\mathbf{G}_{\rm ren}(\mathbf{r},\mathbf{r}',\omega)$, $W_{\mu,\mu'}(\omega)$, and $Z_{\mu,\mu'}(\omega)$ have no pole in the upper half of the complex ω plane, and they satisfy the Kramers-Kronig relation.

In Appendix G, for the verification of the validity of the introduced commutation relations (15), (36), and (37) for $\hat{\mathbf{J}}_0(\mathbf{r},\omega)$ and $\hat{\mathcal{D}}_\mu(\omega)$, we derive that commutation relations (64) and (65) satisfy the equal-time ones expected in the Schrödinger representation.

IX. VALIDITY OF RWA

In the semiclassical framework, the microscopic nonlocal theory has mostly been discussed under the RWA. In this section, we discuss the validity and usefulness of the RWA in our QED theory.

The RWA means that nonresonant terms proportional to $(\omega+\omega_\mu)^{-1}$ are negligible as compared to resonant terms $(\omega-\omega_\mu)^{-1}$. In discussing the resonant optical processes of elementary excitations in condensed matter and also in atoms and molecules, the RWA is usually considered to be a valid approximation. This is because the width of the energy range of interest is usually sufficiently small as compared to the eigenenergies of the elementary excitations. In particular, in the case of excitons, the width is of the order of LT splitting, center-of-mass motion energy, or radiative- and nonradiative-relaxation widths, which are usually more than 3 orders of magnitude smaller than the excitons' eigenenergies. Since the 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 the following paragraphs, we apply the RWA to the excitons' motion and derive simplified equations and commutation relations.

Under the RWA, i.e., $\omega\sim\omega_\mu$, the excitons' motion equation [Eq. (33)] can be written as

$$[\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2]\hat{b}_\mu(\omega) = \int d\mathbf{r}\boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r})\cdot\hat{\mathbf{E}}^+(\mathbf{r},\omega) + \hat{\mathcal{D}}_\mu(\omega) \tag{71}$$

because a contribution from $\{\hat{b}_\mu(-\omega^*)\}^\dagger=\hat{b}^\dagger_\mu(\omega)$ is negligible as compared to that of $\hat{b}_\mu(\omega)$. For the same reason, the excitonic polarization (41) is also rewritten as

$$\hat{\mathbf{P}}^+_{\rm ex}(\mathbf{r},\omega) = \sum_\mu \boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\hat{b}_\mu(\omega). \tag{72}$$

Substituting Eq. (71) into this equation, we obtain the excitonic polarization, instead of Eq. (42), as

$$\hat{\mathbf{P}}^+_{\rm ex}(\mathbf{r},\omega) = \epsilon_0\int d\mathbf{r}'\tilde{\boldsymbol{\chi}}_{\rm ex}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{E}}^+(\mathbf{r},\omega) + \sum_\mu \frac{\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\hat{\mathcal{D}}_\mu(\omega)}{\hbar\omega_\mu-\hbar\omega-i\gamma_\mu(\omega)/2}, \tag{73}$$

where the susceptibility tensor (43) is simplified as

$$\tilde{\boldsymbol{\chi}}_{\rm ex}(\mathbf{r},\mathbf{r}',\omega) = \frac{1}{\epsilon_0}\sum_\mu \frac{\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r}')}{\hbar\omega_\mu-\hbar\omega-i\gamma_\mu(\omega)/2}. \tag{74}$$

This function also satisfies the Kramers-Kronig relation and has no pole in the upper of the complex ω plane because it is also derived from motion equation [Eq. (D1)] of excitons and that [Eq. (D3)] of reservoir oscillators under the RWA. However, while the susceptibility (43) satisfies Eq. (44) without the RWA, we have $\tilde{\boldsymbol{\chi}}_{\rm ex}(\mathbf{r},\mathbf{r}',\omega)\neq\{\tilde{\boldsymbol{\chi}}_{\rm ex}(\mathbf{r},\mathbf{r}'-\omega^*)\}^*$ because we discuss only under $\omega\sim\omega_\mu$.

Substituting Eq. (73) into the Maxwell wave equation [Eq. (11)], we obtain another wave equation, instead of Eq. (45), as

$$\nabla\times\nabla\times\hat{\mathbf{E}}^+(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\epsilon_{\rm bg}(\mathbf{r},\omega)\hat{\mathbf{E}}^+(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\int d\mathbf{r}'\tilde{\boldsymbol{\chi}}_{\rm ex}(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{E}}^+(\mathbf{r}',\omega) = i\mu_0\omega\hat{\mathbf{J}}''_0(\mathbf{r},\omega), \tag{75}$$

where the noise current density (46) is rewritten as

$$\hat{\mathbf{J}}''_0(\mathbf{r},\omega) = \hat{\mathbf{J}}_0(\mathbf{r},\omega) - i\omega\sum_\mu \frac{\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\hat{\mathcal{D}}_\mu(\omega)}{\hbar\omega_\mu-\hbar\omega-i\gamma_\mu(\omega)/2}. \tag{76}$$

From commutation relations (15), (36), and (37), we obtain that for $\hat{\mathbf{J}}''_0(\mathbf{r},\omega)$ as

$$[\hat{\mathbf{J}}''_0(\mathbf{r},\omega),\{\hat{\mathbf{J}}''_0(\mathbf{r}',\omega'^*)\}^\dagger] = \delta(\omega-\omega')\frac{\epsilon_0\hbar\omega^2}{i2\pi}[\tilde{\boldsymbol{\epsilon}}(\mathbf{r},\mathbf{r}',\omega) - \tilde{\boldsymbol{\epsilon}}^{*t}(\mathbf{r}',\mathbf{r},\omega)], \tag{77}$$

where the nonlocal dielectric tensor is represented as

$$\tilde{\boldsymbol{\epsilon}}(\mathbf{r},\mathbf{r}',\omega) = \delta(\mathbf{r}-\mathbf{r}')\epsilon_{\rm bg}(\mathbf{r},\omega)\mathbf{1} + \tilde{\boldsymbol{\chi}}_{\rm ex}(\mathbf{r},\mathbf{r}',\omega). \tag{78}$$

Therefore, even under the RWA, the commutation relation has the same form as the original one [Eq. (48)], which is described by the nonlocal dielectric tensor (49).

On the other hand, substituting Eq. (17), the representation of the electric field, into the approximated motion equation [Eq. (71)] of excitons with expanding $\hat{\mathbf{P}}^+_{\rm ex}(\mathbf{r},\omega)$ as the approximated form (72), we obtain a linear equation set with respect to only $\{\hat{b}_\mu(\omega)\}$, instead of Eq. (52), as

$$\sum_{\mu'} S_{\mu,\mu'}(\omega)\hat{b}_{\mu'}(\omega) = \int d\mathbf{r}\boldsymbol{\mathcal{P}}^*_{\mu}(\mathbf{r})\cdot\hat{\mathbf{E}}^+_0(\mathbf{r},\omega) + \hat{\mathcal{D}}_{\mu}(\omega). \tag{79}$$

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

$$\hat{b}_{\mu}(\omega) = \sum_{\mu'} \tilde{W}_{\mu,\mu'}(\omega)\left[\int d\mathbf{r}\boldsymbol{\mathcal{P}}^*_{\mu'}(\mathbf{r})\cdot\hat{\mathbf{E}}^+_0(\mathbf{r},\omega) + \hat{\mathcal{D}}_{\mu'}(\omega)\right]. \tag{80}$$

From the commutation relations (25), (36), and (37), we obtain those of the excitons under the RWA as

$$[\hat{b}_{\mu}(\omega),\{\hat{b}_{\mu'}(\omega'^*)\}^\dagger] = \delta(\omega-\omega')\frac{\hbar}{i2\pi}[\tilde{W}_{\mu,\mu'}(\omega) - \tilde{W}^*_{\mu',\mu}(\omega)], \tag{81a}$$

$$[\hat{b}_{\mu}(\omega),\hat{b}_{\mu'}(-\omega')] = 0. \tag{81b}$$

Since $\tilde{W}_{\mu,\mu'}(-\omega)\ll\tilde{W}_{\mu,\mu'}(\omega)$, as obtained from $\omega\sim\omega'\sim\omega_\mu>0$, Eq. (81b) is zero. In addition, we have $W_{\mu,\mu'}(\omega)\gg Z_{\mu,\mu'}(\omega)$ and $W_{\mu,\mu'}(\omega)\sim\tilde{W}_{\mu,\mu'}(\omega)$ under the RWA. Therefore, the commutation relations (81a) and (81b) can be considered as approximations of Eqs. (64), which are derived without the RWA.

On the other hand, instead of Eq. (60), the electric field is written under the RWA as

$$\hat{\mathbf{E}}^+(\mathbf{r},\omega) = \hat{\mathbf{E}}^+_0(\mathbf{r},\omega) + \sum_{\mu}\boldsymbol{\mathcal{E}}_{\mu}(\mathbf{r},\omega)\hat{b}_{\mu}(\omega). \tag{82}$$

From the commutation relations (25), (81a), and (81b), we obtain those of the electric field in the same form as Eq. (65) as

$$[\hat{\mathbf{E}}^+(\mathbf{r},\omega),\hat{\mathbf{E}}^-(\mathbf{r}',\omega')] = \delta(\omega-\omega')\frac{\mu_0\hbar\omega^2}{i2\pi} \times[\tilde{\mathbf{G}}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega) - \tilde{\mathbf{G}}^{*t}_{\text{ren}}(\mathbf{r}',\mathbf{r},\omega)], \tag{83}$$

where $\tilde{\mathbf{G}}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)$ is represented, instead of Eq. (66), as

$$\tilde{\mathbf{G}}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega) = \mathbf{G}(\mathbf{r},\mathbf{r}',\omega) + \frac{1}{\mu_0\omega^2}\sum_{\mu,\mu'}\boldsymbol{\mathcal{E}}_{\mu}(\mathbf{r},\omega) \times\tilde{W}_{\mu,\mu'}(\omega)\boldsymbol{\mathcal{F}}_{\mu'}(\mathbf{r}',\omega). \tag{84}$$

Since $W_{\mu,\mu'}(\omega)\gg Z_{\mu,\mu'}(\omega)$ and $W_{\mu,\mu'}(\omega)\gg W^*_{\mu,\mu'}(-\omega)$, this tensor can be considered as an approximation of Eq. (66). Further, this also satisfies the wave equation [Eq. (50)] by replacing $\chi_{\text{ex}}(\mathbf{r},\mathbf{r}',\omega)$ with $\tilde{\chi}_{\text{ex}}(\mathbf{r},\mathbf{r}',\omega)$. However, Eq. (67) is not maintained under the RWA as $\tilde{\mathbf{G}}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)\neq\{\tilde{\mathbf{G}}_{\text{ren}}(\mathbf{r},\mathbf{r}',-\omega^*)\}^*$ because we discuss under $\omega\sim\omega_\mu$.

Since the commutation relations of excitons and the electromagnetic field maintain their forms from the general ones, $\tilde{W}_{\mu,\mu'}(\omega)$ and $\tilde{\mathbf{G}}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)$ can also be considered as retarded correlation functions under the RWA. Furthermore, since these functions are considered as approximations of the general ones, it is safe to say that the RWA is valid in our QED theory, and it is useful in the practical application from the view point of the simplicity of the self-consistent equation set and the Green's tensor.

X. CALCULATION SCHEME

In this section, we explicitly show a calculation scheme for practical applications of our theory. First, we describe practical materials in terms of our system parameters, i.e., background dielectric function $\epsilon_{\text{bg}}(\mathbf{r},\omega)$, excitons' eigenfrequencies $\{\omega_\mu\}$, transition dipole moments $\{\mathcal{P}_\mu\}$, polarization direction $\{\mathbf{e}_\mu\}$, center-of-mass wave functions $\{G_\mu(\mathbf{r})\}$, and nonradiative-relaxation widths $\{\gamma_\mu\}$. We usually determine $\{\omega_\mu\}$ and $\{G_\mu(\mathbf{r})\}$ from boundary conditions for exciton center-of-mass motion. The absolute value of $\mathcal{P}_\mu$ is determined by the LT splitting energy $\Delta^\mu_{\text{LT}}=|\mathcal{P}_\mu|^2/\epsilon_0\epsilon_{\text{bg}}$, and $\{\gamma_\mu\}$ and the phase differences of $\{\mathcal{P}_\mu\}$ between different relative-motion states are treated as fitting parameters for experimental results. However, we often consider only the lowest relative motion of excitons when the energies of higher states are far from our energy region of interest. In such a case, the phase of $\mathcal{P}_\mu$ does not appear in the calculation of observables under the RWA, and only $\{\gamma_\mu\}$ remain as fitting parameters.

Next, we derive the Green's tensor $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ that satisfies Eq. (16), which is uniquely determined by $\epsilon_{\text{bg}}(\mathbf{r},\omega)$. The form of $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ has already been known for various structures with high symmetry,[47] and it can also be numerically calculated for arbitrary 3D structures.[48] Then, we perform integrations in $\mathcal{A}_{\mu,\mu'}(\omega)$, $\mathcal{B}_{\mu,\mu'}(\omega)$, $\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)$, and $\boldsymbol{\mathcal{F}}_\mu(\mathbf{r},\omega)$ [Eqs. (55), (56), (61), and (62)] and numerically calculate the inverse of the coefficient matrix of the self-consistent equation set (57) [or Eq. (79) under the RWA]. Next, we obtain the Green's tensor $\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)$, Eq. (66) [or $\tilde{\mathbf{G}}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)$, Eq. (84) under the RWA].

The size of the coefficient matrix is $2N\times2N$ (or $N\times N$ under the RWA), where N is the number of exciton states to be considered in the calculation. The above numerical calculation has been performed for semiconductor quantum dots, films, multilayers, and so on in the semiclassical framework.[26] Furthermore, we have already applied our QED theory with the RWA for the analysis of the entangled-photon generation from a semiconductor film with a thickness of a few hundred nanometers.[36] In this numerical calculation, we considered 200 exciton states. In this manner, our QED theory is definitely feasible for practical applications.

XI. DISCUSSION

In this study, by using the quantization technique of Suttorp and Wubs[7,18] and the technique of the microscopic nonlocal theory,[25,26] we have constructed a QED theory for excitons in arbitrary-structured dielectrics with considering the nonlocal susceptibility and nonradiative relaxation of excitons. This theory keeps good correspondences with both the nonlocal theory and the QED theories for dispersive and absorptive materials. On the other hand, as mentioned in Sec. II, the QED of media with the nonlocality has already been discussed in a few studies. From the viewpoint of practical applications, we compare our theory with the studies of Di Stefano and co-workers[38–40] and Raabe *et al.*[17]

Di Stefano *et al.* discussed the quantum-well structures of dispersive and absorptive dielectrics with the nonlocality in Ref. 38, and their theory is generalized to enable the consideration of arbitrary structures in Refs. 39 and 40. However, there still remains a problem in deriving the Green's tensors for the generalized wave equation, as shown in Eq. (50) in the present paper. On the other hand, our theory provides a solution to this problem by providing a definite calculation method based on a linear equation set, which is derived from the Green's tensor $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ and the fact that the nonlocal susceptibility is represented as a summation of separable functions with respect to two positions, as seen in Eq. (43). The problem of Ref. 40 can be solved by using our theory because we derive the Green's tensor (66) for arbitrary-structured excitonic polarization and background dielectrics.

On the other hand, Raabe *et al.*[17] proposed the use of the dielectric approximation with the surface impedance method for the practical calculation of the Green's tensor for the Maxwell wave equation with the nonlocal susceptibility. In the dielectric approximation, the characteristic length of spatial dispersion (the spatial spreading of the excitons' center-of-mass motion) is assumed to be small as 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. The Green's tensor can be derived using the surface impedance method for a given surface impedance or admittance, which includes the outside information. In contrast, our theory provides the Green's tensor, without any significant approximations, for given $\epsilon_{\mathrm{bg}}(\mathbf{r},\omega)$ and microscopic information of excitons.

As mentioned in Sec. I, there is a 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.*[49,50] (although the nonlocality was not sufficiently considered in these calculations) and, by extending our QED theory reported in the present paper, we discuss it in the excitonic system weakly confined in nanostructures,[36] which are known to exhibit anomalous nonlinear optical phenomena.[27–31] In addition, Scheel and Welsch[14,15] discussed the QED of nonlinear media with absorption and dispersion (but without the nonlocality). When we discuss the nonlinear processes of excitons with the nonlocality, we must self-consistently treat their nonlinear motion equation and the Maxwell wave equation. Based on the self-consistent equation set (57) or (79), as discussed in the present paper, the new objective is to solve the equation set with nonlinear terms originating from nonlinear processes, as we have done.[36] On the other hand, based on the Maxwell wave equation [Eq. (45)] with the nonlocal susceptibility, as discussed in the previously discussed QED theories,[16,17,38–41] we must solve the wave equation with nonlinear and nonlocal susceptibility. Both approaches can be performed by applying some techniques such as successive approximation, and the expectation values of the observables are calculated based on the commutation relations (64) of excitons and those [Eq. (65)] of the electric field [Eqs. (81a), (81b), and (83) under the RWA] that are described in terms of $W_{\mu,\mu'}(\omega)$, $Z_{\mu,\mu'}(\omega)$, and $\mathbf{G}_{\mathrm{ren}}(\mathbf{r},\mathbf{r}',\omega)$ derived in the present paper. However, such calculations are usually difficult. In such a case, more detailed and systematic calculations should be performed by using the Feynman diagram technique with the correlation functions, which identifies with the retarded correlation functions under the RWA and can then be derived in our QED theory. In this sense, our scheme will be a powerful tool to discuss the nonlinear processes of elementary excitations in condensed matter with the nonlocality. Based on our QED theory, we are going to discuss various optical phenomena that 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 represent the second-quantized operators with a hat (ˆ) in this appendix.

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

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

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

Here, due to the interaction with the radiation field (see Appendix B), the momentum of the charged particles is represented as

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

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 $\mathbf{J}_{\mathrm{cp}}(\mathbf{r})$,

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

By representing the coefficient of the radiation contribution as

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

we can represent complete current density (A1) as

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

This subtraction of the radiation contribution is discussed in Sec. 2.2 of Ref. 26.

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

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

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

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

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

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

These operators are also represented in terms of exciton operators $\{\hat{b}_\mu\}$ in the same form as the polarization density (30),

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

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

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

Instead of evaluating the expansion coefficient of each operator, we describe them in terms of $\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})$, the coefficient of polarization density (30). From relations

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

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

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

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

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

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

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

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

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

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

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

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

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

We expand this in terms of the exciton operators,

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

where μ and $\Phi_{\mu,c,v,\mathbf{R}}$, respectively, denote 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 represent the electron-hole operator set as

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

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

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

where the expansion coefficient is represented as

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

Assuming that the spatial variation of the center-of-mass wave function is negligible within the extent of the electron-hole relative motion, 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 by iterating $\mathbf{R}'$, we obtain

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

Here, 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 $\Phi_{\mu,c,v}(\mathbf{R})=\Phi_{\mu,c,v,\mathbf{R}}/\sqrt{\Omega}$, and we then obtain the expansion coefficient of polarization density (A25) as

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

where

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

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

APPENDIX B: DERIVATION OF HAMILTONIAN

As a model of the background system, we adopt the system discussed by SW,[7] i.e., polarizable harmonic oscillators interacting with the radiation field and the reserver oscillators. Considering the charged particles of Appendix A, the total Lagrangian is represented as

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

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,

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

We omit the descriptions of the position dependences. $\mathbf{E}=-\dot{\mathbf{A}}-\nabla\phi_{\mathrm{bg}}-\nabla\phi_{\mathrm{cp}}$ 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, represented as $-\alpha\mathbf{X}$, $\rho_{\mathrm{bg}}=\nabla\cdot(\alpha\mathbf{X})$, and $-\alpha\dot{\mathbf{X}}$ with a position-dependent coefficient $\alpha(\mathbf{r})$. The background Coulomb potential is represented as

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

and is related to the longitudinal component of the polarization as

$$\nabla\phi_{\mathrm{bg}} = -\frac{1}{\epsilon_0}[\alpha\mathbf{X}]_L. \quad (B4)$$

Further, it satisfies the Poisson equation,

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

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

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

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

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

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

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

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

Then, after a straightforward calculation, we obtain the Hamiltonian as

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

where H_{em} describes the complete Hamiltonian discussed by SW,[7] representing the radiation field and background dielectrics with local susceptibility,

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

The first two terms represent the radiation energy, the third term the kinetic energy of the oscillators, and the fourth 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

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

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

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

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

The first term represents 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 three terms of interaction Hamiltonian (5) 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. III, we put the exchange interaction between electrons and holes into H_{int}. It is obtained by expanding the fourth term of Eq. (B12),

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

When we assume commutation relations (8a) and (8b) 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 $(\cdots)$ in Eq. (B13) and the second and third terms of Eq. (B12) should belong to H_{mat}, instead of discussing them in detail, we treat the matter Hamiltonian as Eq. (7) for a simple description of the linear optical process of excitons with nonradiative relaxation and put the contribution of charged particles except the focusing excitonic modes into H_{em} as a part of nonresonant backgrounds.

APPENDIX C: EXTENSION OF MAXWELL WAVE EQUATION

In this appendix, we extend the Maxwell wave equation discussed by SW (Ref. 7) to enable the consideration of the exciton-induced polarization with nonlocal susceptibility. We derive the Heisenberg equations of the system variables and momenta from background Hamiltonian (B9) and interaction term (5). The commutation relations of the variables are

$$[\mathbf{A}(\mathbf{r}),\mathbf{\Pi}(\mathbf{r}')] = i\hbar\boldsymbol{\delta}_T(\mathbf{r}-\mathbf{r}'), \quad \text{(C1)}$$

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

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

where $\boldsymbol{\delta}_T(\mathbf{r}-\mathbf{r}')$ is the Dirac delta function extracting the transverse component,

$$\boldsymbol{\delta}_T(\mathbf{r}-\mathbf{r}') \equiv \mathbf{1}\delta(\mathbf{r}-\mathbf{r}') + \frac{\nabla'\nabla'}{4\pi|\mathbf{r}-\mathbf{r}'|}. \quad \text{(C4)}$$

We obtain the motion equations of the radiation field as

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

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

where

$$\mathbf{J}_{\text{ex}\,T}(\mathbf{r}) \equiv \int d\mathbf{r}\,\boldsymbol{\delta}_T(\mathbf{r}-\mathbf{r}')\cdot\mathbf{J}_{\text{ex}}(\mathbf{r}') \quad \text{(C7)}$$

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

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

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

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,

$$\nabla^2 \mathbf{A} - \frac{1}{c^2}\ddot{\mathbf{A}} = \mu_0[\alpha\dot{\mathbf{X}}]_T - \mu_0 \mathbf{J}_{\mathrm{ex}\,T}, \tag{C12}$$

which has a transverse component of the excitonic current density as compared to the same type of equation in the study by SW.[7] Using the relation between the longitudinal components of excitonic variables

$$\mathbf{J}_{\mathrm{ex}\,L}(\mathbf{r}) = \dot{\mathbf{P}}_{\mathrm{ex}\,L}(\mathbf{r}) = \epsilon_0 \nabla \dot{\phi}_{\mathrm{ex}}(\mathbf{r}) \tag{C13}$$

and that for the polarizable oscillators (B4), we can rewrite Eq. (C12) as a wave equation for electric field [Eq. (10)],

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

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

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

Next, we rewrite the above motion equations in time domain into those for the forward Laplace transform of the variables,

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

and for the backward Laplace transform,

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

From their motion equations, we derive those for the positive-frequency Fourier transform,

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

This Laplace transformation technique is essential to derive the representations of noise operators $\hat{\mathbf{J}}_0(\mathbf{r},\omega)$ and $\hat{\mathcal{D}}_\mu(\omega)$ and is useful for verifying their complex-frequency relations (14) and (35) and to derive commutation relations (15), (36), and (37) as SW,[7] although we introduce the latter from the fluctuation dissipation theorem.

The forward Laplace transform of the electric field [Eq. (10)] is represented as

$$\bar{\mathbf{E}}(p) = -p\bar{\mathbf{A}}(p) + \frac{1}{\epsilon_0}[\alpha\bar{\mathbf{X}}(p)]_L - \nabla\bar{\phi}_{\mathrm{ex}}(p) + \mathbf{A}(0). \tag{C18}$$

From Eqs. (C14) and (C15), we obtain a wave equation for the transform as

$$\nabla\times\nabla\times\bar{\mathbf{E}}(p) + \frac{p^2}{c^2}\bar{\epsilon}(p)\bar{\mathbf{E}}(p) = -\mu_0 p\bar{\mathbf{J}}(p) - \mu_0 p\bar{\mathbf{J}}_{\mathrm{ex}}(p) - \frac{p}{c^2}\nabla\phi_{\mathrm{ex}}(0), \tag{C19}$$

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

$$\bar{\chi}(p) = \frac{\alpha^2}{\epsilon_0\rho}\left[p^2 + \tilde{\omega}_0^2 - \frac{1}{\rho^2}\int_0^\infty d\omega\frac{\omega^2 v_\omega^2}{p^2+\omega^2}\right]^{-1}. \tag{C20}$$

The operator on the RHS of Eq. (C19) is

$$\begin{aligned}\bar{\mathbf{J}}(p) = &-\frac{1}{\mu_0 p}\nabla\times\nabla\times\mathbf{A}(0) - \epsilon_0 p\bar{\chi}(p)\mathbf{A}(0) + \mathbf{\Pi}(0)\\ &+ \alpha\left[1 - \frac{\epsilon_0\rho}{\alpha^2}p^2\bar{\chi}(p)\right]\mathbf{X}(0) - [\alpha\mathbf{X}(0)]_L - \frac{\epsilon_0}{\alpha}p\bar{\chi}(p)\mathbf{P}(0)\\ &- \frac{\epsilon_0}{\alpha}p\bar{\chi}(p)\int_0^\infty d\omega\frac{v_\omega}{p^2+\omega^2}\left[\omega^2\mathbf{Y}_\omega(0) - \frac{p}{\rho}\mathbf{Q}_\omega(0)\right],\end{aligned} \tag{C21}$$

which is just the one shown in Eq. (27) of the study by SW (Ref. 7) and depends only on the system variables and momenta in the background. On the other hand, the backward Laplace transform of the electric field is represented as

$$\check{\mathbf{E}}(p) = p\check{\mathbf{A}}(p) + \frac{1}{\epsilon_0}[\alpha\check{\mathbf{X}}(p)]_L - \nabla\check{\phi}_{\mathrm{ex}}(p) - \mathbf{A}(0), \tag{C22}$$

and a wave equation is obtained as

$$\nabla\times\nabla\times\check{\mathbf{E}}(p) + \frac{p^2}{c^2}\bar{\epsilon}(p)\check{\mathbf{E}}(p) = \mu_0 p\check{\mathbf{J}}(p) + \mu_0 p\check{\mathbf{J}}_{\mathrm{ex}}(p) - \frac{p}{c^2}\nabla\phi_{\mathrm{ex}}(0), \tag{C23}$$

where the operator on the RHS is also independent of the variables associated with the excitons,

$$\begin{aligned}\check{\mathbf{J}}(p) = &-\frac{1}{\mu_0 p}\nabla\times\nabla\times\mathbf{A}(0) - \epsilon_0 p\bar{\chi}(p)\mathbf{A}(0) - \mathbf{\Pi}(0)\\ &- \alpha\left[1 - \frac{\epsilon_0\rho}{\alpha^2}p^2\bar{\chi}(p)\right]\mathbf{X}(0) + [\alpha\mathbf{X}(0)]_L - \frac{\epsilon_0}{\alpha}p\bar{\chi}(p)\mathbf{P}(0)\\ &- \frac{\epsilon_0}{\alpha}p\bar{\chi}(p)\int_0^\infty d\omega\frac{v_\omega}{p^2+\omega^2}\left[\omega^2\mathbf{Y}_\omega(0) + \frac{p}{\rho}\mathbf{Q}_\omega(0)\right].\end{aligned} \tag{C24}$$

From the forward and backward Laplace transforms (C19) and (C23) of the Maxwell wave equation, we obtain that for the positive-frequency Fourier component of the electric field as

$$\nabla \times \nabla \times \hat{\mathbf{E}}^{+}(\mathbf{r},\omega) - \frac{\omega^2}{c^2}\epsilon_{\mathrm{bg}}(\mathbf{r},\omega)\hat{\mathbf{E}}^{+}(\mathbf{r},\omega) = i\mu_0\omega[\hat{\mathbf{J}}_0(\mathbf{r},\omega) + \hat{\mathbf{J}}_{\mathrm{ex}}^{+}(\mathbf{r},\omega)], \tag{C25}$$

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

$$\hat{\mathbf{J}}_0(\mathbf{r},\omega) = \hat{\mathbf{J}}(\mathbf{r},\omega) - \frac{i\omega^2}{\pi c^2}\mathrm{Im}[\epsilon_{\mathrm{bg}}(\mathbf{r},\omega)] \times \int d\mathbf{r}'\mathbf{G}^{*}(\mathbf{r},\mathbf{r}',\omega)\cdot[\check{\mathbf{J}}_{\mathrm{ex}}(\mathbf{r}',i\omega+\delta) - \epsilon_0\nabla'\phi_{\mathrm{ex}}(\mathbf{r}',0)], \tag{C26}$$

where $\mathbf{G}(\mathbf{r},\mathbf{r},\omega)$ is the Green's tensor satisfying Eq. (16), and

$$\hat{\mathbf{J}}(\mathbf{r},\omega) = \frac{1}{2\pi}[\bar{\mathbf{J}}(\mathbf{r},-i\omega+\delta) + \check{\mathbf{J}}(\mathbf{r},i\omega+\delta)] - \frac{i\omega^2}{\pi c^2}\mathrm{Im}[\epsilon_{\mathrm{bg}}(\mathbf{r},\omega)]\int d\mathbf{r}'\mathbf{G}^{*}(\mathbf{r},\mathbf{r}',\omega)\cdot\check{\mathbf{J}}(\mathbf{r}',i\omega+\delta) \tag{C27}$$

is the same operator shown in Eq. (45) of the study by SW.[7] Because of the relation

$$\hat{\mathbf{J}}_{\mathrm{ex}}(\mathbf{r},\omega) = -i\omega\hat{\mathbf{P}}_{\mathrm{ex}}(\mathbf{r},\omega), \tag{C28}$$

we can rewrite Eq. (C25) into Eq. (11).

APPENDIX D: DERIVATION OF EXCITONS' MOTION EQUATION

In Appendix C, we derive the Maxwell wave equation considering the excitons. In this appendix, we derive the motion equation of excitons and rewrite it in the ω representation.

From matter Hamiltonian (7) and interaction Hamiltonian (5), neglecting the radiation contribution of the current density $N_{\mathrm{ex}}(\mathbf{r})\mathbf{A}^2(\mathbf{r})/2$ under weak excitation, we obtain the Heisenberg equation of excitons as

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

where $\phi(\mathbf{r}) \equiv \phi_{\mathrm{bg}}(\mathbf{r}) + \phi_{\mathrm{ex}}(\mathbf{r})$ is the complete Coulomb potential. Using relations (A16) and (A18) between $\boldsymbol{\mathcal{I}}_\mu(\mathbf{r})$, $\rho_\mu(\mathbf{r})$, and $\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})$ and Laplace transforms (C18) and (C22) of the electric field, the forward and backward Laplace transforms of Eq. (D1) are, respectively, derived under $\omega \sim \omega_\mu$ as

$$(\hbar\omega_\mu - \hbar\omega - i\delta)\bar{b}_\mu(-i\omega+\delta) = -i\hbar b_\mu(0) + \int d\mathbf{r}\boldsymbol{\mathcal{P}}_\mu^{*}(\mathbf{r})\cdot[\bar{\mathbf{E}}(\mathbf{r},-i\omega+\delta) - \mathbf{A}(\mathbf{r},0)] - \int_0^\infty d\Omega[g_\mu(\Omega)\bar{d}_\mu(\Omega,-i\omega+\delta) + g_\mu^{*}(\Omega)\bar{d}_\mu^{\dagger}(\Omega,-i\omega+\delta)], \tag{D2a}$$

$$(\hbar\omega_\mu - \hbar\omega + i\delta)\check{b}_\mu(i\omega+\delta) = i\hbar b_\mu(0) + \int d\mathbf{r}\boldsymbol{\mathcal{P}}_\mu^{*}(\mathbf{r})\cdot[\check{\mathbf{E}}(\mathbf{r},i\omega+\delta) + \mathbf{A}(\mathbf{r},0)] - \int_0^\infty d\Omega[g_\mu(\Omega)\check{d}_\mu(\Omega,i\omega+\delta) + g_\mu^{*}(\Omega)\check{d}_\mu^{\dagger}(\Omega,i\omega+\delta)]. \tag{D2b}$$

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

$$i\hbar\frac{\partial}{\partial t}d_\mu(\Omega,t) = \hbar\Omega d_\mu(\Omega,t) + g_\mu^{*}(\Omega)[b_\mu(t) + b_\mu^{\dagger}(t)], \tag{D3}$$

and its Laplace transforms,

$$(\hbar\Omega - \hbar\omega - i\delta)\bar{d}_\mu(\Omega,-i\omega+\delta) = -i\hbar d_\mu(\Omega,0) - g_\mu^{*}(\Omega)[\bar{b}_\mu(-i\omega+\delta) + \bar{b}_\mu^{\dagger}(-i\omega+\delta)], \tag{D4a}$$

$$(\hbar\Omega - \hbar\omega + i\delta)\check{d}_\mu(\Omega,i\omega+\delta) = i\hbar d_\mu(\Omega,0) - g_\mu^{*}(\Omega)[\check{b}_\mu(i\omega+\delta) + \check{b}_\mu^{\dagger}(i\omega+\delta)]. \tag{D4b}$$

Substituting Eqs. (D4) into Eqs. (D2a) and (D2b), we obtain

$$[\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2]\bar{b}_\mu(-i\omega+\delta) + [-i\gamma_\mu(\omega)/2]\bar{b}_\mu^{\dagger}(-i\omega+\delta) - \bar{D}_\mu(-i\omega+\delta) = -i\hbar b_\mu(0) + \int d\mathbf{r}\boldsymbol{\mathcal{P}}_\mu^{*}(\mathbf{r})\cdot[\bar{\mathbf{E}}(\mathbf{r},-i\omega+\delta) - \mathbf{A}(\mathbf{r},0)], \tag{D5a}$$

$$[\hbar\omega_\mu - \hbar\omega + i\gamma_\mu^{*}(\omega)/2]\check{b}_\mu(i\omega+\delta) + [i\gamma_\mu^{*}(\omega)/2]\check{b}_\mu^{\dagger}(i\omega+\delta) - \check{D}_\mu(i\omega+\delta) = i\hbar b_\mu(0) + \int d\mathbf{r}\boldsymbol{\mathcal{P}}_\mu^{*}(\mathbf{r})\cdot[\check{\mathbf{E}}(\mathbf{r},i\omega+\delta) + \mathbf{A}(\mathbf{r},0)], \tag{D5b}$$

where the operators on the left-hand side (LHS) are defined as

$$\bar{D}_\mu(-i\omega+\delta) \equiv \int_0^\infty d\Omega \frac{ig_\mu(\Omega)}{\Omega-\omega-i\delta} d_\mu(\Omega,0) - \int_0^\infty d\Omega \frac{ig_\mu^*(\Omega)}{\Omega+\omega+i\delta} d_\mu^\dagger(\Omega,0), \quad \text{(D6a)}$$

$$\check{D}_\mu(i\omega+\delta) \equiv -\int_0^\infty d\Omega \frac{ig_\mu(\Omega)}{\Omega-\omega+i\delta} d_\mu(\Omega,0) + \int_0^\infty d\Omega \frac{ig_\mu^*(\Omega)}{\Omega+\omega-i\delta} d_\mu^\dagger(\Omega,0), \quad \text{(D6b)}$$

and the relaxation width is represented as

$$\frac{i\gamma_\mu(\omega)}{2} \equiv \int_0^\infty d\Omega \left[\frac{|g_\mu(\Omega)|^2}{\hbar\Omega-\hbar\omega-i\delta} + \frac{|g_\mu(\Omega)|^2}{\hbar\Omega+\hbar\omega+i\delta} \right]. \quad \text{(D7)}$$

Because of this definition, $\gamma_\mu(\omega)$ satisfies Eq. (34). By adding Eqs. (D5a) and (D5b), we obtain motion equation [Eq. (33)] of excitons and the fluctuation operator defined as

$$\hat{D}_\mu(\omega) \equiv \frac{1}{2\pi}[\bar{D}_\mu(-i\omega+\delta)+\check{D}_\mu(i\omega+\delta)] - \frac{i\,\mathrm{Re}[\gamma_\mu(\omega)]}{2\pi}[\check{b}_\mu(i\omega+\delta)+\check{b}_\mu^\dagger(i\omega+\delta)]. \quad \text{(D8)}$$

Since there exist the relations $\check{b}_\mu(i\omega+\delta)=\{\check{b}_\mu^\dagger(-i\omega^*+\delta)\}^\dagger$, $\bar{D}_\mu(-i\omega+\delta)=\{\bar{D}_\mu(i\omega^*+\delta)\}^\dagger$, $\check{D}_\mu(i\omega+\delta)=\{\check{D}_\mu(-i\omega^*+\delta)\}^\dagger$, and Eq. (34), $\hat{D}_\mu(\omega)$ satisfies Eq. (35).

APPENDIX E: EVALUATION OF COMMUTATION RELATIONS

In this appendix, we present a detailed calculation of the commutation relations of the noise current density $\hat{\mathbf{J}}_0'(\mathbf{r},\omega)$, exciton operators $\hat{b}_\mu(\omega)$, and electric field $\hat{\mathbf{E}}(\mathbf{r},\omega)$.

First, we evaluate the commutation relations of $\hat{\mathbf{J}}_0'(\mathbf{r},\omega)$ defined in Eq. (46). From fundamental commutation relations (15), (36), and (37), we obtain

$$[\hat{\mathbf{J}}_0'(\mathbf{r},\omega),\{\hat{\mathbf{J}}_0'(\mathbf{r}',\omega'^*)\}^\dagger] = \delta(\omega-\omega')\frac{\epsilon_0\hbar\omega^2}{i2\pi}[\epsilon_{\mathrm{bg}}(\mathbf{r},\omega)-\epsilon_{\mathrm{bg}}^*(\mathbf{r},\omega)]\mathbf{1} + \delta(\omega-\omega')\frac{\hbar\omega^2}{i2\pi}\sum_\mu \begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\\ \boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r})\end{bmatrix}^t \underline{\mathbf{W}}_\mu(\omega) i\,\mathrm{Re}[\gamma_\mu(\omega)] \times \begin{bmatrix}1&1\\1&1\end{bmatrix}\underline{\mathbf{W}}_\mu^{*t}(\omega)\begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r}')\\ \boldsymbol{\mathcal{P}}_\mu(\mathbf{r}')\end{bmatrix}. \quad \text{(E1)}$$

Here, a part of the last term is represented as

$$\underline{\mathbf{W}}_\mu(\omega) i\,\mathrm{Re}[\gamma_\mu(\omega)]\begin{bmatrix}1&1\\1&1\end{bmatrix}\underline{\mathbf{W}}_\mu^{*t}(\omega) = \underline{\mathbf{W}}_\mu(\omega)[\underline{\mathbf{S}}_\mu^{*t}(\omega)-\underline{\mathbf{S}}_\mu(\omega)]\underline{\mathbf{W}}_\mu^{*t}(\omega) = \underline{\mathbf{W}}_\mu(\omega)-\underline{\mathbf{W}}_\mu^{*t}(\omega), \quad \text{(E2)}$$

and we can verify that Eq. (E1) becomes Eq. (48).

Next, we verify excitons' commutation relations (64a) and (64b). Commutator (64a) is evaluated as

$$[\hat{b}_\mu(\omega),\{\hat{b}_{\mu'}(\omega'^*)\}^\dagger] = \delta(\omega-\omega')\frac{-\hbar}{i2\pi}\sum_{\nu,\nu'}\{W_{\mu,\nu}(\omega)\{\mathcal{A}_{\nu,\nu'}(\omega)-\mathcal{A}_{\nu',\nu}^*(\omega)-i\,\mathrm{Re}[\gamma_\nu(\omega)]\delta_{\nu,\nu'}\}W_{\mu',\nu'}^*(\omega) + W_{\mu,\nu}(\omega)\{\mathcal{B}_{\nu,\nu'}(\omega)-\mathcal{B}_{\nu',\nu}(-\omega)-i\,\mathrm{Re}[\gamma_\nu(\omega)]\delta_{\nu,\nu'}\}Z_{\mu',\nu'}^*(\omega)+Z_{\mu,\nu}(\omega)\{\mathcal{B}_{\nu,\nu'}^*(-\omega)-\mathcal{B}_{\nu',\nu}^*(\omega) + i\,\mathrm{Re}[\gamma_\nu(-\omega)]\delta_{\nu,\nu'}\}W_{\mu',\nu'}^*(\omega)+Z_{\mu,\nu}(\omega)\{\mathcal{A}_{\nu,\nu'}^*(-\omega)-\mathcal{A}_{\nu',\nu}(-\omega)+i\,\mathrm{Re}[\gamma_\nu(-\omega)]\delta_{\nu,\nu'}\}Z_{\mu',\nu'}^*(\omega)\}. \quad \text{(E3)}$$

We can rewrite this as

$$[\hat{b}_\mu(\omega),\{\hat{b}_{\mu'}(\omega'^*)\}^\dagger] = \delta(\omega-\omega')\frac{-\hbar}{i2\pi}\sum_{\nu,\nu'}\{[W_{\mu,\nu}(\omega)S_{\nu,\nu'}(\omega)+Z_{\mu,\nu}(\omega)T_{\nu,\nu'}^*(-\omega)]W_{\mu',\nu'}^*(\omega) - W_{\mu,\nu}(\omega)[W_{\mu',\nu'}^*(\omega)S_{\nu',\nu}^*(\omega)+Z_{\mu',\nu'}^*(\omega)T_{\nu',\nu}(-\omega)]+[W_{\mu,\nu}(\omega)T_{\nu,\nu'}(\omega)+Z_{\mu,\nu}(\omega)S_{\nu,\nu'}^*(-\omega)]Z_{\mu',\nu'}^*(\omega) - Z_{\mu,\nu}(\omega)[W_{\mu',\nu'}^*(\omega)T_{\nu',\nu}^*(\omega)+Z_{\mu',\nu'}^*(\omega)S_{\nu',\nu}(-\omega)]\}. \quad \text{(E4)}$$

Here, using relation (59) between the coefficient matrix for the self-consistent equation set and its inverse matrix, we can obtain Eq. (64a) from Eq. (E4). By the same procedure, commutator (64b) is evaluated as

$$[\hat{b}_\mu(\omega),\hat{b}_{\mu'}(-\omega')] = \delta(\omega-\omega')\frac{-\hbar}{i2\pi}\sum_{\nu,\nu'}\{[W_{\mu,\nu}(\omega)T_{\nu,\nu'}(\omega)+Z_{\mu,\nu}(\omega)S^*_{\nu,\nu'}(-\omega)]W_{\mu',\nu'}(-\omega) - W_{\mu,\nu}(\omega)[W_{\mu',\nu'}(-\omega)T_{\nu',\nu}(-\omega)+Z_{\mu',\nu'}(-\omega)S^*_{\nu',\nu}(\omega)] + [W_{\mu,\nu}(\omega)S_{\nu,\nu'}(\omega)+Z_{\mu,\nu}(\omega)T^*_{\nu,\nu'}(-\omega)]Z_{\mu',\nu'}(-\omega) - Z_{\mu,\nu}(\omega)[W_{\mu',\nu'}(-\omega)S_{\nu',\nu}(-\omega)+Z_{\mu',\nu'}(-\omega)T^*_{\nu',\nu}(\omega)]\}, \quad \text{(E5)}$$

and we obtain Eq. (64b) using Eq. (59).

Finally, we verify commutation relation (65) of the electric field. The commutator is expanded as

$$[\hat{\mathbf{E}}^+(\mathbf{r},\omega),\hat{\mathbf{E}}^-(\mathbf{r}',\omega')] = [\hat{\mathbf{E}}_0^+(\mathbf{r},\omega),\hat{\mathbf{E}}_0^-(\mathbf{r}',\omega')] + \sum_{\mu'}\{[\hat{\mathbf{E}}_0^+(\mathbf{r},\omega),\{\hat{b}_{\mu'}(\omega'^*)\}^\dagger]\boldsymbol{\mathcal{E}}^*_{\mu'}(\mathbf{r}',\omega') + [\hat{\mathbf{E}}_0^+(\mathbf{r},\omega),\hat{b}_{\mu'}(-\omega')]\boldsymbol{\mathcal{E}}_{\mu'}(\mathbf{r}',-\omega')\} + \sum_\mu\{\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)[\hat{b}_\mu(\omega),\hat{\mathbf{E}}_0^-(\mathbf{r}',\omega')] + \boldsymbol{\mathcal{E}}^*_\mu(\mathbf{r},-\omega)[\{\hat{b}_\mu(-\omega^*)\}^\dagger,\hat{\mathbf{E}}_0^-(\mathbf{r}',\omega')]\} + \sum_{\mu,\mu'}\{\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)[\hat{b}_\mu(\omega),\{\hat{b}_{\mu'}(\omega'^*)\}^\dagger] + \boldsymbol{\mathcal{E}}^*_\mu(\mathbf{r},-\omega)[\{\hat{b}_\mu(-\omega^*)\}^\dagger,\{\hat{b}_{\mu'}(\omega'^*)\}^\dagger]\}\boldsymbol{\mathcal{E}}^*_{\mu'}(\mathbf{r}',\omega') + \sum_{\mu,\mu'}\{\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)[\hat{b}_\mu(\omega),\hat{b}_{\mu'}(-\omega')] + \boldsymbol{\mathcal{E}}^*_\mu(\mathbf{r},-\omega)[\{\hat{b}_\mu(-\omega^*)\}^\dagger,\hat{b}_{\mu'}(-\omega')]\}\boldsymbol{\mathcal{E}}_{\mu'}(\mathbf{r}',-\omega'). \quad \text{(E6)}$$

Using Eqs. (64a), (64b), and (25) and the following equations, we can obtain Eq. (65). Hence,

$$[\hat{\mathbf{E}}_0^+(\mathbf{r},\omega),\{\hat{b}_{\mu'}(\omega'^*)\}^\dagger] = \delta(\omega-\omega')\frac{\hbar}{i2\pi}\sum_\mu\{[\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)-\boldsymbol{\mathcal{F}}^*_\mu(\mathbf{r},\omega)]W^*_{\mu',\mu}(\omega) + [\boldsymbol{\mathcal{F}}_\mu(\mathbf{r},\omega)-\boldsymbol{\mathcal{E}}^*_\mu(\mathbf{r},\omega)]Z^*_{\mu',\mu}(\omega)\}, \quad \text{(E7)}$$

$$[\hat{\mathbf{E}}_0^+(\mathbf{r},\omega),\hat{b}_{\mu'}(-\omega')] = \delta(\omega-\omega')\frac{\hbar}{i2\pi}\sum_\mu\{[\boldsymbol{\mathcal{E}}^*_\mu(\mathbf{r},-\omega) - \boldsymbol{\mathcal{F}}_\mu(\mathbf{r},-\omega)]W_{\mu',\mu}(-\omega) + [\boldsymbol{\mathcal{F}}^*_\mu(\mathbf{r},-\omega)-\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},-\omega)]Z_{\mu',\mu}(-\omega)\}, \quad \text{(E8)}$$

$$[\hat{b}_\mu(\omega),\hat{\mathbf{E}}_0^-(\mathbf{r}',\omega')] = \delta(\omega-\omega')\frac{\hbar}{i2\pi}\sum_{\mu'}\{W_{\mu,\mu'}(\omega)[\boldsymbol{\mathcal{F}}_{\mu'}(\mathbf{r}',\omega)-\boldsymbol{\mathcal{E}}^*_{\mu'}(\mathbf{r}',\omega)] + Z_{\mu,\mu'}(\omega)[\boldsymbol{\mathcal{E}}_{\mu'}(\mathbf{r}',\omega)-\boldsymbol{\mathcal{F}}^*_{\mu'}(\mathbf{r}',\omega)]\}, \quad \text{(E9)}$$

$$[\{\hat{b}_\mu(-\omega^*)\}^\dagger,\hat{\mathbf{E}}_0^-(\mathbf{r}',\omega')] = \delta(\omega-\omega')\frac{\hbar}{i2\pi}\sum_{\mu'}\{W^*_{\mu,\mu'}(-\omega) \times[\boldsymbol{\mathcal{F}}^*_{\mu'}(\mathbf{r}',-\omega)-\boldsymbol{\mathcal{E}}_{\mu'}(\mathbf{r}',-\omega)] + Z^*_{\mu,\mu'}(-\omega)[\boldsymbol{\mathcal{E}}^*_{\mu'}(\mathbf{r}',-\omega)-\boldsymbol{\mathcal{F}}_{\mu'}(\mathbf{r}',-\omega)]\}. \quad \text{(E10)}$$

APPENDIX F: VERIFICATION OF GREEN'S TENSOR FOR NONLOCAL SYSTEM

In this appendix, we verify that $\mathbf{G}_{\mathrm{ren}}(\mathbf{r},\mathbf{r}',\omega)$ defined in Eq. (66) satisfies wave equation [Eq. (50)]. Here, we define operator $\hat{L}$ that operates on the arbitrary vector $\boldsymbol{f}(\boldsymbol{r})$ as

$$\hat{L}\boldsymbol{f}(\mathbf{r}) \equiv \nabla\times\nabla\times\boldsymbol{f}(\mathbf{r}) - \frac{\omega^2}{c^2}\epsilon_{\mathrm{bg}}(\mathbf{r},\omega)\boldsymbol{f}(\mathbf{r}) - \frac{\omega^2}{c^2}\int d\mathbf{r}''\boldsymbol{\chi}_{\mathrm{ex}}(\mathbf{r},\mathbf{r}'',\omega)\boldsymbol{f}(\mathbf{r}''). \quad \text{(F1)}$$

First of all, applying $\hat{L}$ into $\mathbf{G}(\mathbf{r},\mathbf{r},\omega)$, we obtain

$$\hat{L}\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) = \delta(\mathbf{r}-\mathbf{r}') - \sum_\mu\begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\\ \boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r})\end{bmatrix}^t \underline{\mathbf{W}}_\mu(\omega)\begin{bmatrix}\boldsymbol{\mathcal{F}}_\mu(\mathbf{r}',\omega)\\ \boldsymbol{\mathcal{E}}_\mu(\mathbf{r}',\omega)\end{bmatrix}. \quad \text{(F2)}$$

On the other hand, $\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)$ becomes

$$\hat{L}\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega) = \mu_0\omega^2\boldsymbol{\mathcal{P}}_\mu(\mathbf{r}) + \mu_0\omega^2\sum_\nu\begin{bmatrix}\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\\ \boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r})\end{bmatrix}^t\underline{\mathbf{W}}_\mu(\omega) \times\begin{bmatrix}\mathcal{A}_{\nu,\mu}(\omega)\\ \mathcal{B}^*_{\nu,\mu}(-\omega)\end{bmatrix}. \quad \text{(F3)}$$

Here, because of definition (39) of $\underline{\mathbf{S}}_\mu(\omega)$, there exists the following relation:

$$\boldsymbol{\mathcal{P}}_\mu(\mathbf{r}) = \begin{bmatrix} \boldsymbol{\mathcal{P}}_\mu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r}) \end{bmatrix}^t \begin{bmatrix} 1 \\ 0 \end{bmatrix} = \begin{bmatrix} \boldsymbol{\mathcal{P}}_\mu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r}) \end{bmatrix}^t \underline{\mathbf{W}}_\mu(\omega) \times \begin{bmatrix} \hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2 \\ -i\gamma_\mu(\omega)/2 \end{bmatrix}. \tag{F4}$$

Therefore, Eq. (F3) is rewritten as

$$\hat{L}\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega) = \mu_0\omega^2 \sum_\nu \begin{bmatrix} \boldsymbol{\mathcal{P}}_\nu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\nu^*(\mathbf{r}) \end{bmatrix}^t \underline{\mathbf{W}}_\nu(\omega) \begin{bmatrix} S_{\nu,\mu}(\omega) \\ T_{\nu,\mu}^*(-\omega) \end{bmatrix}. \tag{F5}$$

By the same procedure, we obtain

$$\hat{L}\boldsymbol{\mathcal{E}}_\mu^*(\mathbf{r},-\omega) = \mu_0\omega^2 \sum_\nu \begin{bmatrix} \boldsymbol{\mathcal{P}}_\nu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\nu^*(\mathbf{r}) \end{bmatrix}^t \underline{\mathbf{W}}_\nu(\omega) \begin{bmatrix} T_{\nu,\mu}(\omega) \\ S_{\nu,\mu}^*(-\omega) \end{bmatrix}. \tag{F6}$$

Then, the additional terms in Eq. (66) become

$$\begin{aligned} &\hat{L}\frac{1}{\mu_0\omega^2}\sum_{\mu,\mu'} [\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)W_{\mu,\mu'}(\omega)\boldsymbol{\mathcal{F}}_{\mu'}(\mathbf{r}',\omega) + \boldsymbol{\mathcal{E}}_\mu^*(\mathbf{r},-\omega)Z_{\mu,\mu'}^*(-\omega)\boldsymbol{\mathcal{E}}_{\mu'}^*(\mathbf{r}',-\omega)] \\ &= \sum_{\mu,\mu',\nu} \begin{bmatrix} \boldsymbol{\mathcal{P}}_\nu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\nu^*(\mathbf{r}) \end{bmatrix}^t \underline{\mathbf{W}}_\nu(\omega) \begin{bmatrix} S_{\nu,\mu}(\omega) & T_{\nu,\mu}(\omega) \\ T_{\nu,\mu}^*(-\omega) & S_{\nu,\mu}^*(-\omega) \end{bmatrix} \times \begin{bmatrix} W_{\mu,\mu'}(\omega) \\ Z_{\mu,\mu'}^*(-\omega) \end{bmatrix} \boldsymbol{\mathcal{F}}_{\mu'}(\mathbf{r}',\omega) \\ &= \sum_\mu \begin{bmatrix} \boldsymbol{\mathcal{P}}_\mu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r}) \end{bmatrix}^t \underline{\mathbf{W}}_\mu(\omega) \begin{bmatrix} \boldsymbol{\mathcal{F}}_\mu(\mathbf{r}',\omega) \\ 0 \end{bmatrix}, \end{aligned} \tag{F7}$$

$$\begin{aligned} &\hat{L}\frac{1}{\mu_0\omega^2}\sum_{\mu,\mu'} [\boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega)Z_{\mu,\mu'}(\omega)\boldsymbol{\mathcal{E}}_{\mu'}(\mathbf{r}',\omega) + \boldsymbol{\mathcal{E}}_\mu^*(\mathbf{r},-\omega)W_{\mu,\mu'}^*(-\omega)\boldsymbol{\mathcal{F}}_{\mu'}^*(\mathbf{r}',-\omega)] \\ &= \sum_{\mu,\mu',\nu} \begin{bmatrix} \boldsymbol{\mathcal{P}}_\nu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\nu^*(\mathbf{r}) \end{bmatrix}^t \underline{\mathbf{W}}_\nu(\omega) \begin{bmatrix} S_{\nu,\mu}(\omega) & T_{\nu,\mu}(\omega) \\ T_{\nu,\mu}^*(-\omega) & S_{\nu,\mu}^*(-\omega) \end{bmatrix} \times \begin{bmatrix} Z_{\mu,\mu'}(\omega) \\ W_{\mu,\mu'}^*(-\omega) \end{bmatrix} \boldsymbol{\mathcal{E}}_{\mu'}(\mathbf{r}',\omega) \\ &= \sum_\mu \begin{bmatrix} \boldsymbol{\mathcal{P}}_\mu(\mathbf{r}) \\ \boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r}) \end{bmatrix}^t \underline{\mathbf{W}}_\mu(\omega) \begin{bmatrix} 0 \\ \boldsymbol{\mathcal{E}}_\mu(\mathbf{r}',\omega) \end{bmatrix}. \end{aligned} \tag{F8}$$

Adding Eqs. (F2), (F7), and (F8), we can find that $\mathbf{G}_{\rm ren}(\mathbf{r},\mathbf{r}',\omega)$ satisfies Eq. (50).

APPENDIX G: EQUAL-TIME COMMUTATION RELATIONS

In order to verify the validity of introduced commutation relations (15), (36), and (37), we calculate the equal-time commutation relations from those in the ω representation. The commutation relations of equal-time Heisenberg operators should keep the form of those as the Schrödinger operators. This means that relations

$$[b_\mu(t), b_{\mu'}^\dagger(t)] = \delta_{\mu,\mu'}, \tag{G1}$$

$$[b_\mu(t), b_{\mu'}(t)] = 0, \tag{G2}$$

$$[\mathbf{P}_{\rm ex}(\mathbf{r},t), \mathbf{P}_{\rm ex}(\mathbf{r}',t)] = \mathbf{0}, \tag{G3}$$

$$[\mathbf{E}(\mathbf{r},t), \mathbf{E}(\mathbf{r}',t)] = \mathbf{0} \tag{G4}$$

should be derived from the commutation relations of the Fourier-transformed Heisenberg operators. Moreover, from Eqs. (10) and (C5), the electric field is represented as

$$\epsilon_0\mathbf{E}(\mathbf{r},t) = -\mathbf{\Pi}(\mathbf{r},t) - \epsilon_0\nabla\phi(\mathbf{r},t), \tag{G5}$$

and since $\mathbf{\Pi}(\mathbf{r},t)$ and $\mathbf{A}(\mathbf{r},t)$ satisfy commutation relation (C1), the following relation should also be derived:

$$[\epsilon_0\mathbf{E}(\mathbf{r},t), \mathbf{A}(\mathbf{r}',t)] = i\hbar\boldsymbol{\delta}_T(\mathbf{r}-\mathbf{r}'). \tag{G6}$$

For local dielectric media, the same type of calculation has been performed by Knöll, Scheel, and Welsch (KSW).[13]

From the time representation of the exciton operator

$$b_\mu(t) = \int_{-\infty}^{\infty} d\omega \hat{b}_\mu(\omega) e^{-i\omega t} \tag{G7}$$

and commutation relations (64a) and (64b) for the ω representation, the equal-time ones are written as

$$[b_\mu(t), b_{\mu'}^\dagger(t)] = \frac{\hbar}{i2\pi}\int_{-\infty}^{\infty} d\omega [W_{\mu,\mu'}(\omega) - W_{\mu',\mu}^*(\omega)], \tag{G8}$$

$$[b_\mu(t), b_{\mu'}(t)] = \frac{\hbar}{i2\pi}\int_{-\infty}^{\infty} d\omega [Z_{\mu,\mu'}(\omega) - Z_{\mu',\mu}^*(-\omega)]. \tag{G9}$$

In the limit of $|\omega|\to\infty$, as indicated in Appendix A.1 of the study by KSW,[13] it is known that $\epsilon_{\rm bg}(\mathbf{r},\omega)\to 1$ and

$$\lim_{|\omega|\to\infty} \frac{\omega^2}{c^2}\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) = -\delta(\mathbf{r}-\mathbf{r}')\mathbf{1}. \tag{G10}$$

Then, due to the orthogonality of $\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})$, as shown in Eq. (31), and the relation with LT splitting $\Delta_{\rm LT}^\mu = |\boldsymbol{\mathcal{P}}_\mu|^2/\epsilon_{\rm bg}(\omega_\mu)\epsilon_0$, the limits of correction terms (55) and (56) are

$$\lim_{|\omega|\to\infty} \mathcal{A}_{\mu,\mu'}(\omega) = \frac{1}{\epsilon_0}\int d\mathbf{r}\boldsymbol{\mathcal{P}}_\mu^*(\mathbf{r})\cdot\boldsymbol{\mathcal{P}}_{\mu'}(\mathbf{r}), \tag{G11}$$

$$= \delta_{\mu,\mu'}\epsilon_{\rm bg}(\omega_\mu)\Delta_{\rm LT}^\mu, \tag{G12}$$

$$\lim_{|\omega|\to\infty} \mathcal{B}_{\mu,\mu'}(\omega) = \delta_{\mu,\mu'}\epsilon_{\rm bg}(\omega_\mu)\Delta_{\rm LT}^\mu e^{i2\theta}, \tag{G13}$$

where θ is the phase of $\boldsymbol{\mathcal{P}}_\mu$. On the other hand, the limit of the nonradiative width $\gamma_\mu(\omega)$ defined in Eq. (D7) is $\gamma_\mu(\omega)$

$\to +0$. Therefore, the coefficient matrix of the self-consistent equation set (57) becomes block diagonal with respect to the exciton states as

$$\lim_{|\omega|\to\infty} S_{\mu,\mu'}(\omega) = \delta_{\mu,\mu'}[\hbar\omega_\mu + \epsilon_{\rm bg}(\omega_\mu)\Delta^\mu_{\rm LT} - \hbar\omega - i\delta], \tag{G14}$$

$$\lim_{|\omega|\to\infty} T_{\mu,\mu'}(\omega) = \delta_{\mu,\mu'}\epsilon_{\rm bg}(\omega_\mu)\Delta^\mu_{\rm LT}e^{i2\theta}. \tag{G15}$$

The inverse of this matrix gives the limits of $W_{\mu,\mu'}(\omega)$ and $Z_{\mu,\mu'}(\omega)$, and it is also diagonal with respect to the exciton states. As mentioned in Sec. VIII, since they have no pole in the upper half of the complex ω plane, the integration over the real axis is evaluated as

$$\int_{-\infty}^{\infty} d\omega W_{\mu,\mu'}(\omega) = \frac{i\pi}{\hbar}\delta_{\mu,\mu'}, \tag{G16a}$$

$$\int_{-\infty}^{\infty} d\omega Z_{\mu,\mu'}(\omega) = 0. \tag{G16b}$$

We can find that these reproduce commutation relations (G1) and (G2) from Eqs.(G8) and (G9).

Next, we verify commutation relation (G3) of the excitonic polarization $\mathbf{P}_{\rm ex}(\mathbf{r},t)$. From Eq. (41), the time representation can be written as

$$\mathbf{P}_{\rm ex}(\mathbf{r},t) = \int_0^\infty d\omega[\hat{\mathbf{P}}^+_{\rm ex}(\mathbf{r},\omega)e^{-i\omega t} + \hat{\mathbf{P}}^-_{\rm ex}(\mathbf{r},\omega)e^{i\omega t}], \tag{G17}$$

$$= \int_{-\infty}^{\infty} d\omega\hat{\mathbf{P}}^+_{\rm ex}(\mathbf{r},\omega)e^{-i\omega t} = \int_{-\infty}^{\infty} d\omega\hat{\mathbf{P}}^-_{\rm ex}(\mathbf{r},\omega)e^{i\omega t}. \tag{G18}$$

Since we have already derived Eqs. (G16a) and (G16b), the equal-time commutator becomes

$$[\mathbf{P}_{\rm ex}(\mathbf{r},t),\mathbf{P}_{\rm ex}(\mathbf{r}',t)] = \int_{-\infty}^{\infty} d\omega \int_{-\infty}^{\infty} d\omega' e^{-i\omega t}[\hat{\mathbf{P}}^+_{\rm ex}(\mathbf{r},\omega),\hat{\mathbf{P}}^-_{\rm ex}(\mathbf{r}',\omega')]e^{i\omega' t} = \sum_\mu [\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r}') - \boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r})\boldsymbol{\mathcal{P}}_\mu(\mathbf{r}')]. \tag{G19}$$

This is also obtained from Eqs. (30) and (G1) directly. From Eq. (G19), we can reproduce equal-time commutation relation (G3),

$$[\mathbf{P}_{\rm ex}(\mathbf{r},t),\mathbf{P}_{\rm ex}(\mathbf{r}',t)]_{\xi,\xi'} = \sum_\mu [\mathcal{P}^\xi_\mu(\mathbf{r})\{\mathcal{P}^{\xi'}_\mu(\mathbf{r}')\}^* - \mathrm{c.c.}], \tag{G20}$$

$$= \sum_\mu [\langle 0|P^\xi_{\rm ex}(\mathbf{r})|\mu\rangle\langle\mu|P^{\xi'}_{\rm ex}(\mathbf{r}')|0\rangle - \mathrm{c.c.}], \tag{G21}$$

$$= \langle 0|[P^\xi_{\rm ex}(\mathbf{r}),P^{\xi'}_{\rm ex}(\mathbf{r}')]|0\rangle = 0, \tag{G22}$$

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

Next, we verify relation (G4) of the electric-field operator. Since the time representation of the electric field is also written in the same form as Eq. (G18), we can evaluate its equal-time commutator as

$$[\mathbf{E}(\mathbf{r},t),\mathbf{E}(\mathbf{r}',t)] = \int_{-\infty}^{\infty} d\omega \frac{\mu_0\hbar\omega^2}{i2\pi}[\mathbf{G}_{\rm ren}(\mathbf{r},\mathbf{r}',\omega) - \mathbf{G}^{*\rm t}_{\rm ren}(\mathbf{r}',\mathbf{r},\omega)]. \tag{G23}$$

Since $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ becomes Eq. (G10) in the limit of $|\omega| \to \infty$, from Eqs. (61) and (62), we can obtain

$$\lim_{|\omega|\to\infty} \boldsymbol{\mathcal{E}}_\mu(\mathbf{r},\omega) = -\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})/\epsilon_0, \tag{G24}$$

$$\lim_{|\omega|\to\infty} \boldsymbol{\mathcal{F}}_\mu(\mathbf{r},\omega) = -\boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r})/\epsilon_0. \tag{G25}$$

By the same procedure used to derive Eqs. (G16a) and (G16b), the integration of Eq. (66) becomes

$$\int_{-\infty}^{\infty} d\omega \frac{\hbar\mu_0\omega^2}{i2\pi}\mathbf{G}_{\rm ren}(\mathbf{r},\mathbf{r}',\omega) = \int_{-\infty}^{\infty} d\omega \frac{\hbar\mu_0\omega^2}{i2\pi}\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) + \frac{1}{2\epsilon_0^2}\sum_\mu [\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r}') - \boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r})\boldsymbol{\mathcal{P}}_\mu(\mathbf{r}')]. \tag{G26}$$

Therefore, Eq. (G23) is evaluated as

$$[\mathbf{E}(\mathbf{r},t),\mathbf{E}(\mathbf{r}',t)] = \int_{-\infty}^{\infty} d\omega \frac{\hbar\mu_0\omega^2}{i2\pi}[\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) - \mathbf{G}^*(\mathbf{r},\mathbf{r}',\omega)] + \frac{1}{\epsilon_0^2}\sum_\mu [\boldsymbol{\mathcal{P}}_\mu(\mathbf{r})\boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r}') - \boldsymbol{\mathcal{P}}^*_\mu(\mathbf{r})\boldsymbol{\mathcal{P}}_\mu(\mathbf{r}')]. \tag{G27}$$

The first two terms are zero as indicated by SW (Ref. 7) or as calculated by KSW,[13] and the other terms are also zero, as discussed above. Then, equal-time commutation relation (G4) is reproduced.

Last, we verify relation (G6). As discussed in Appendix A.2 of the study by KSW,[13] we can write the time representation of the vector potential as

$$\mathbf{A}(\mathbf{r},t) = \lim_{\epsilon\to 0}\int_\epsilon^\infty d\omega \int d\mathbf{s}\left[\frac{e^{-i\omega t}}{i\omega}\hat{\mathbf{E}}^+(\mathbf{s},\omega) + \mathrm{H.c.}\right]\cdot \boldsymbol{\delta}_T(\mathbf{s}-\mathbf{r}), \tag{G28}$$

$$= P\int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega t}}{i\omega}\int d\mathbf{s}\hat{\mathbf{E}}^+(\mathbf{s},\omega)\cdot\boldsymbol{\delta}_T(\mathbf{s}-\mathbf{r}), \tag{G29}$$

$$= - P\int_{-\infty}^{\infty} d\omega \frac{e^{i\omega t}}{i\omega}\int d\mathbf{s}\hat{\mathbf{E}}^{-}(\mathbf{s},\omega)\cdot\boldsymbol{\delta}_T(\mathbf{s}-\mathbf{r}), \tag{G30}$$

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

$$[\epsilon_0\mathbf{E}(\mathbf{r},t),\mathbf{A}(\mathbf{r}',t)] = P\int_{-\infty}^{\infty} d\omega \frac{\epsilon_0\mu_0\hbar\omega}{2\pi}\int d\mathbf{s}[\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{s},\omega) - \mathbf{G}^{*t}_{\text{ren}}(\mathbf{s},\mathbf{r},\omega)] \cdot \boldsymbol{\delta}_T(\mathbf{s}-\mathbf{r}'). \tag{G31}$$

First, in the limit of $|\omega|\to\infty$, $\mathbf{G}_{\text{ren}}(\omega)/\omega$ becomes zero because of factor ω^{-1} as compared to the above discussion. On the other hand, in the limit of $|\omega|\to 0$, due to the equation shown in the study by KSW,[13]

$$\lim_{|\omega|\to 0}\frac{\omega^2}{c^2}\int d\mathbf{s}\mathbf{G}(\mathbf{r},\mathbf{s},\omega)\cdot\boldsymbol{\delta}_T(\mathbf{s}-\mathbf{r}') = \mathbf{0}, \tag{G32}$$

the following limits are also zeros,

$$\lim_{|\omega|\to 0}\frac{\omega^2}{c^2}\int d\mathbf{s}\boldsymbol{\delta}_T(\mathbf{s}-\mathbf{r}')\cdot\boldsymbol{\mathcal{E}}_\mu(\mathbf{s},\omega) = \lim_{|\omega|\to 0}\frac{\omega^2}{c^2}\int d\mathbf{s}\boldsymbol{\delta}_T(\mathbf{s}-\mathbf{r}')\cdot\boldsymbol{\mathcal{F}}_\mu(\mathbf{s},\omega) = \mathbf{0}. \tag{G33}$$

From these results, we can find that the last four terms of Eq. (66), the representation of $\mathbf{G}_{\text{ren}}(\mathbf{r},\mathbf{r}',\omega)$, do not contribute to the result of commutator (G31), and it becomes

$$[\epsilon_0\mathbf{E}(\mathbf{r},t),\mathbf{A}(\mathbf{r}',t)] = P\int_{-\infty}^{\infty} d\omega \frac{\epsilon_0\mu_0\hbar\omega}{2\pi}\int d\mathbf{s}[\mathbf{G}(\mathbf{r},\mathbf{s},\omega) - \mathbf{G}^{*}(\mathbf{r},\mathbf{s},\omega)] \cdot \boldsymbol{\delta}_T(\mathbf{s}-\mathbf{r}'). \tag{G34}$$

This reproduces equal-time commutation relation (G6) as verified by KSW.[13]

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