

QED theory for excitons with microscopic nonlocality

Motoaki Bamba^{1,3,*} and Hajime Ishihara^{2,3}

¹ Department of Materials Engineering Science, Osaka University, 1-3 Machikaneyama-cho, Toyonaka, Osaka 560-8531, Japan

² Department of Physics and Electronics, Osaka Prefecture University, 1-1 Gakuen-cho, Naka-ku, Sakai, Osaka 599-8531, Japan

³ CREST, Japan Science and Technology Agency, 4-1-8 Honmachi, Kawaguchi, Saitama 332-0012, Japan

Received XXXX, revised XXXX, accepted XXXX

Published online XXXX

PACS 42.50.Nn, 71.35.-y, 78.67.-n

* Corresponding author: e-mail bamba@pe.osakafu-u.ac.jp, Phone +81-72-254-9271, Fax +81-72-254-9271

We have constructed QED theory for excitons with microscopic nonlocality or nonlocal susceptibility originating from their center-of-mass motion. The nonlocality must be properly considered in excitons' weak (center-of-mass) confinement regime where the long wavelength approximation is breakdown. This theory keeps good correspondences with its underlying theories, the QED theory for dispersive and absorptive local dielectrics and

the microscopic nonlocal theory developed in the semiclassical framework. Our theory provides correlation functions of excitons, which renormalize radiative shift, transition between exciton states via electromagnetic fields, radiative and nonradiative relaxation processes. By using these functions, we can systematically study various nonlinear, dephasing, and emission processes of excitons in weak-confinement to bulk-like systems.

Copyright line will be provided by the publisher

1 Introduction In the conventional theories of optical processes in condensed matters, the light has been mainly treated classically regardless whether the matter systems are described in quantum mechanical terms (semiclassical theory) or in classical ones. These kinds of theories have successfully explained a variety of optical phenomena for the classical light or the coherent states of photons. However, there is growing interest in the quantum electrodynamics (QED) of elementary excitations in condensed matters both experimentally [1–3] and theoretically [4–8].

The quantization of the electromagnetic fields in dispersive and absorptive dielectrics has been systematically carried out for homogeneous media by Huttner and Barnett [4]. Their pioneering work stimulated various theoretical studies associated with the QED in such materials [5]. On the other hand, Suttorp and Wubs (SW) have systematically discussed the one applicable to arbitrary 3D structures [6]. In this scheme, all the matter information is characterized by a complex dielectric function $\varepsilon(\mathbf{r}, \omega)$, which depends on the spatial position \mathbf{r} of the medium as well as the radiation frequency ω . However, in general as seen in Eq. (12) of the present paper, the optical susceptibility 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 the electric

field $\mathbf{E}(\mathbf{r}', \omega)$ at the different position \mathbf{r}' . This microscopic nonlocality originates from the spatial spreading of wave functions of elementary excitations or, especially for excitons in semiconductors, their center-of-mass motion with a finite translational mass. The nonlocality must be properly considered in the excitons' weak (center-of-mass) confinement regime where the excitons' spatial spreading is comparable to the wavelength of the self-consistently determined electromagnetic fields (i.e. beyond the long wavelength approximation). It is known that anomalous nonlinear optical phenomena appear in nano-structures where excitons are weakly confined [9–13]. Savasta, Stefano, and Girlanda (SSG) have introduced the nonlocality to the QED theory for dispersive and absorptive dielectrics or excitonic systems [7]. Their theory consists of the Maxwell wave equation with the nonlocal susceptibility as seen in Eq. (14). Although they have previously discussed the same kind of systems with quantum-well structures [8], in the case of the other structures, there remains a problem to derive a Green function for the nonlocal wave equation. In the present paper, based on the microscopic nonlocal theory developed in the semiclassical framework [14] and the QED theory for dispersive and absorptive local systems by SW [6], we show a practical calculation method for the QED of exci-

Copyright line will be provided by the publisher

tons weakly confined in arbitrary-structured 3D dielectrics with considering their microscopic nonlocality. The detailed underlying calculation of our theory is given in the other paper [15].

2 Hamiltonian We describe the matter systems with resonant contributions from excitons with center-of-mass motion, and non-resonant (background) ones with the local dielectric function $\varepsilon_{\text{bg}}(\mathbf{r}, \omega)$. We explicitly discuss the optical and nonradiative relaxation processes of the former, and the latter is treated in the same procedure of SW [6]. The total Hamiltonian discussed in the present paper is written as $H = H_{\text{em}} + H_{\text{int}} + H_{\text{ex}}$. The H_{em} describes the radiation field and background dielectric medium, and is just the total Hamiltonian discussed by SW. The H_{ex} represents the excitons with nonradiative relaxation, and H_{int} is the interaction between H_{em} and H_{ex} .

With regard to the H_{ex} , we suppose excitons whose center-of-mass motion is confined in finite spaces, and consider a reservoir of oscillators interacting with them in order to describe their nonradiative relaxation process. The excitonic Hamiltonian is written as

$$H_{\text{ex}} = \sum_{\mu} \hbar\omega_{\mu} b_{\mu}^{\dagger} b_{\mu} + \sum_{\mu} \int_0^{\infty} d\Omega \{ \hbar\Omega d_{\mu}^{\dagger}(\Omega) d_{\mu}(\Omega) + [b_{\mu} + b_{\mu}^{\dagger}] [g_{\mu}(\Omega) d_{\mu}(\Omega) + g_{\mu}^*(\Omega) d_{\mu}^{\dagger}(\Omega)] \}. \quad (1)$$

Here, b_{μ} is the annihilation operator of excitons in the eigenstate μ with eigenfrequency ω_{μ} , which do not include the longitudinal-transverse (LT) splitting because the exchange interaction between electrons and holes is included in H_{int} as explained below. The index μ represents degrees of freedom of not only the excitons' relative motion but also the translational one. The exciton operator satisfies the bosonic commutation relations as $[b_{\mu}, b_{\mu'}^{\dagger}] = \delta_{\mu, \mu'}$ and $[b_{\mu}, b_{\mu'}] = 0$. On the other hand, $d_{\mu}(\Omega)$ is the annihilation operator of reservoir oscillators with frequency Ω interacting with excitons in state μ , and $g_{\mu}(\Omega)$ is the coupling coefficient. The oscillators are independent of each other, and satisfy the commutation relations as $[d_{\mu}(\Omega), d_{\mu'}^{\dagger}(\Omega')] = \delta_{\mu, \mu'} \delta(\Omega - \Omega')$ and $[d_{\mu}(\Omega), d_{\mu'}(\Omega')] = 0$.

With regard to the H_{int} , although it is primarily represented as the interaction between excitonic current density and vector potential, and the Coulomb interaction between excitons and the background medium [15], we describe H_{int} as the interaction between the electric field $\mathbf{E}(\mathbf{r})$ and the excitonic polarization $\mathbf{P}_{\text{ex}}(\mathbf{r})$ as

$$H_{\text{int}} = - \int d\mathbf{r} \mathbf{P}_{\text{ex}}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r}). \quad (2)$$

This treatment is valid for the linear optical process under the excitonic resonance. However, we must keep in mind that Eq. (2) contains the exchange interaction between electrons and holes, the origin of the LT splitting of excitons, then it is not included in the exciton eigenenergies

$\hbar\omega_{\mu}$ appeared in Eq. (1). The excitonic polarization is expanded by the exciton operators: $\mathbf{P}_{\text{ex}}(\mathbf{r}) = \sum_{\mu} \mathcal{P}_{\mu}(\mathbf{r}) b_{\mu} + \text{H.c.}$, where the expansion coefficient is written as $\mathcal{P}_{\mu}(\mathbf{r}) = \mathcal{P}_{\mu} e_{\mu} G_{\mu}(\mathbf{r})$. The \mathcal{P}_{μ} is the transition dipole moment, e_{μ} is a unit vector in the polarization direction, and $G_{\mu}(\mathbf{r})$ is the center-of-mass wave function of exciton state μ . In the weak confinement regime, the \mathcal{P}_{μ} approximately depends only on the relative motion of excitons, and is related with the LT splitting as $\Delta_{\text{LT}}^{\mu} = |\mathcal{P}_{\mu}|^2 / \varepsilon_0 \varepsilon_{\text{bg}}$.

3 Motion equations By using the same quantization technique of SW [6], we have derived motion equations of the electric field and excitons, and commutation relations of source operators. The equations are described in terms of the Fourier transform of physical variables: $\hat{\Omega}^{\pm}(\omega) \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{\pm i\omega t} \Omega(t)$, where $\hat{\Omega}^{+}(\omega)$ and $\hat{\Omega}^{-}(\omega)$ are respectively called positive- and negative-frequency Fourier component of variable $\Omega(t)$ in Heisenberg picture.

The motion equation of the electric field $\mathbf{E}(\mathbf{r})$ is the Maxwell wave equation:

$$\nabla \times \nabla \times \hat{\mathbf{E}}^{+}(\mathbf{r}, \omega) - (\omega/c)^2 \varepsilon_{\text{bg}}(\mathbf{r}, \omega) \hat{\mathbf{E}}^{+}(\mathbf{r}, \omega) = i\mu_0 \omega \hat{\mathbf{J}}_0(\mathbf{r}, \omega) + \mu_0 \omega^2 \hat{\mathbf{P}}_{\text{ex}}^{+}(\mathbf{r}, \omega), \quad (3)$$

where $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ is called the noise current density, the source of the background electromagnetic fields, and satisfies the commutation relations as

$$[\hat{\mathbf{J}}_0(\mathbf{r}, \omega), \{\hat{\mathbf{J}}_0(\mathbf{r}', \omega')\}^{\dagger}] = \delta(\omega - \omega') \delta(\mathbf{r} - \mathbf{r}') (\varepsilon_0 \hbar \omega^2 / \pi) \text{Im}[\varepsilon_{\text{bg}}(\mathbf{r}, \omega)] \mathbf{1}, \quad (4a)$$

$$[\hat{\mathbf{J}}_0(\mathbf{r}, \omega), \hat{\mathbf{J}}_0(\mathbf{r}', \omega')] = \mathbf{0}. \quad (4b)$$

Eqs. (3) and (4) have the same forms of those in SW theory [6] except the excitonic polarization $\hat{\mathbf{P}}_{\text{ex}}^{+}(\mathbf{r}, \omega)$ appearing in the RHS of Eq. (3). Here, using a Green function $\mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega)$ satisfying

$$\nabla \times \nabla \times \mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) - (\omega/c)^2 \varepsilon_{\text{bg}}(\mathbf{r}, \omega) \mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \mathbf{1}, \quad (5)$$

we can rewrite the Maxwell wave equation (3) as

$$\hat{\mathbf{E}}^{+}(\mathbf{r}, \omega) = \hat{\mathbf{E}}_0^{+}(\mathbf{r}, \omega) + \mu_0 \omega^2 \int d\mathbf{r}' \mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{P}}_{\text{ex}}^{+}(\mathbf{r}', \omega), \quad (6)$$

where $\hat{\mathbf{E}}_0^{+}(\mathbf{r}, \omega)$ is the background electric field defined as

$$\hat{\mathbf{E}}_0^{+}(\mathbf{r}, \omega) \equiv i\mu_0 \omega \int d\mathbf{r}' \mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{J}}_0(\mathbf{r}', \omega). \quad (7)$$

From the commutation relations (4) of $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$, we can derive those for $\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega)$ as

$$[\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega), \hat{\mathbf{E}}_0^-(\mathbf{r}', \omega')] = \delta(\omega - \omega') (\mu_0 \hbar \omega^2 / \pi) \text{Im}[\mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega)], \quad (8a)$$

$$[\hat{\mathbf{E}}_0^+(\mathbf{r}, \omega), \hat{\mathbf{E}}_0^+(\mathbf{r}', \omega')] = \mathbf{0}. \quad (8b)$$

These relations can be understood by the fact that the Green function $\mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega)$ for the Maxwell wave equation (3) identifies with the ω -Fourier transform of the retarded correlation function for the electric field [16] in the H_{em} system. The spatial structure of the background medium is characterized by the local dielectric function $\varepsilon_{\text{bg}}(\mathbf{r}, \omega)$ in the Maxwell wave equation (3) and in the commutation relations (4), or by the Green function $\mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega)$ satisfying Eq. (5). The form of the Green function has already been known for various structures with high symmetry [17] and also can be numerically calculated for arbitrary 3D structures [18].

Next, we discuss the motion of the unknown variable $\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega)$ in the Maxwell wave equation (3). The motion equation for the exciton operator is derived as

$$[\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2] \hat{b}_\mu(\omega) = \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot \hat{\mathbf{E}}^+(\mathbf{r}, \omega) + \hat{D}_\mu(\omega). \quad (9)$$

Here, $\gamma_\mu(\omega)$ is the nonradiative relaxation width defined in terms of the coupling coefficient $g_\mu(\Omega)$. The operator $\hat{D}_\mu(\omega)$ represents the fluctuation by the reservoir, and satisfies the commutation relations:

$$[\hat{D}_\mu(\omega), \{\hat{D}_{\mu'}(\omega')\}^\dagger] = \delta_{\mu, \mu'} \delta(\omega - \omega') \frac{\hbar}{2\pi} \gamma_\mu(\omega), \quad (10a)$$

$$[\hat{D}_\mu(\omega), \hat{D}_{\mu'}(\omega')] = 0. \quad (10b)$$

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

$$[\hat{D}_\mu(\omega), \hat{\mathbf{J}}_0(\mathbf{r}, \omega')] = [\hat{D}_\mu(\omega), \{\hat{\mathbf{J}}_0(\mathbf{r}, \omega')\}^\dagger] = \mathbf{0}. \quad (11)$$

Under the rotating wave approximation (RWA), the positive-frequency Fourier transform of the excitonic polarization can be written as $\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) = \sum_\mu \mathcal{P}_\mu(\mathbf{r}) \hat{b}_\mu(\omega)$ for $\omega > 0$. Substituting Eq. (9) into this, we obtain the nonlocal form in the electric-field contribution of the polarization:

$$\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega) = \varepsilon_0 \int d\mathbf{r}' \chi(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{E}}^+(\mathbf{r}', \omega) + \dots, \quad (12)$$

where the nonlocal susceptibility tensor is defined as

$$\chi(\mathbf{r}, \mathbf{r}', \omega) \equiv \frac{1}{\varepsilon_0} \sum_\mu \frac{\mathcal{P}_\mu(\mathbf{r}) \mathcal{P}_\mu^*(\mathbf{r}')}{\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2}. \quad (13)$$

We can treat any spatial spreading of the exciton states, the origin of the nonlocality, through the polarization coefficient $\mathcal{P}_\mu(\mathbf{r})$ or the center-of-mass wave function $G_\mu(\mathbf{r})$.

In order to discuss the optical process of excitons, we must simultaneously solve the Maxwell wave equation (3) and the motion equation of the polarization (12) to determine the unknown variables $\hat{\mathbf{E}}^+(\mathbf{r}, \omega)$ and $\hat{\mathbf{P}}_{\text{ex}}^+(\mathbf{r}, \omega)$. Substituting Eq. (12) into (3), we obtain the nonlocal wave equation as

$$\nabla \times \nabla \times \hat{\mathbf{E}}^+(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \int d\mathbf{r}' \varepsilon(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{E}}^+(\mathbf{r}', \omega) = i\mu_0 \omega \hat{\mathbf{J}}_0(\mathbf{r}, \omega) + \dots, \quad (14)$$

where $\varepsilon(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \varepsilon_{\text{bg}}(\mathbf{r}, \omega) \mathbf{1} + \chi(\mathbf{r}, \mathbf{r}', \omega)$. This has the same form of the one discussed in SSG theory [7]. However, it seems very difficult to solve this nonlocal equation, and in the discussion of SSG, there remains a problem to derive the Green function $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ satisfying

$$\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \int d\mathbf{s} \varepsilon(\mathbf{r}, \mathbf{s}, \omega) \cdot \mathbf{G}(\mathbf{s}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \mathbf{1}. \quad (15)$$

In the next section, we solve this nonlocal problem in the same procedure of the semiclassical nonlocal theory [14].

4 QED of excitons Substituting the representation of the electric field (6) into the motion equation of excitons (9) with the RWA, we obtain a linear equation set with respect to the exciton amplitudes $\{\hat{b}_\mu(\omega)\}$ as

$$\sum_{\mu'} S_{\mu, \mu'}(\omega) \hat{b}_{\mu'}(\omega) = \int d\mathbf{r} \mathcal{P}_\mu^*(\mathbf{r}) \cdot \hat{\mathbf{E}}_0^+(\mathbf{r}, \omega) + \hat{D}_\mu(\omega), \quad (16)$$

where the coefficient is defined as

$$S_{\mu, \mu'}(\omega) \equiv [\hbar\omega_\mu - \hbar\omega - i\gamma_\mu(\omega)/2] \delta_{\mu, \mu'} - \mu_0 \omega^2 \int d\mathbf{r} d\mathbf{r}' \mathcal{P}_\mu^*(\mathbf{r}) \cdot \mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathcal{P}_{\mu'}(\mathbf{r}'). \quad (17)$$

These have the same forms of the self-consistent equation set in the semiclassical microscopic nonlocal theory [14]. The first term in the RHS of Eq. (16) can be interpreted as the exciton amplitude directly induced by the background electric field. Here, we use the word ‘‘directly’’ to mean that it does not include the diffusion of exciton amplitudes via the electromagnetic fields. Such an effect is reflected in the last term of Eq. (17), the correction terms of the exciton eigenenergies $\{\omega_\mu\}$ describing the radiative shift, LT splitting, radiative relaxation, and the photon-assisted transition between the exciton states.

Inverting the coefficient matrix of Eq. (16) as $\mathbf{W}(\omega) \equiv \mathbf{S}^{-1}(\omega)$, we can represent the exciton amplitudes $\{\hat{b}_\mu(\omega)\}$ in terms of the source operators $\hat{\mathbf{J}}_0(\mathbf{r}, \omega)$ and $\hat{D}_\mu(\omega)$, then

all the physical variables can also be represented by them. On the other hand, from the commutation relations (8) of $\hat{\mathbf{E}}_0(\mathbf{r}, \omega)$ and (10) of $\hat{D}_\mu(\omega)$, those of the exciton operators can be calculated as

$$\begin{aligned} & [\hat{b}_\mu(\omega), \{\hat{b}_{\mu'}(\omega')\}^\dagger] \\ &= \delta(\omega - \omega') (\hbar/i2\pi) [W_{\mu, \mu'}(\omega) - W_{\mu', \mu}^*(\omega)], \quad (18a) \end{aligned}$$

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

This result indicates that the elements of the inverse matrix $\mathbf{W}(\omega)$ of the self-consistent equation set identify with the Fourier transforms of the excitons' retarded (and also causal) correlation functions:

$$-\hbar W_{\mu, \mu'}(\omega) = -i \int_{t'}^{\infty} dt e^{i\omega(t-t')} \langle [b_\mu(t), b_{\mu'}^\dagger(t')] \rangle. \quad (19)$$

This renormalizes the exciton-photon and exciton-reservoir interaction, i.e., the exciton-polariton effects with radiative and nonradiative relaxation. In addition, we can obtain the commutation relations of the electric field operators:

$$\begin{aligned} & [\hat{\mathbf{E}}^+(\mathbf{r}, \omega), \hat{\mathbf{E}}^-(\mathbf{r}', \omega')] = \delta(\omega - \omega') (\mu_0 \hbar \omega^2 / i2\pi) \\ & \times [\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \{\mathbf{G}(\mathbf{r}', \mathbf{r}, \omega)\}^{t*}], \quad (20a) \end{aligned}$$

$$[\hat{\mathbf{E}}^+(\mathbf{r}, \omega), \hat{\mathbf{E}}^+(\mathbf{r}', \omega')] = \mathbf{0}, \quad (20b)$$

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

$$\begin{aligned} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) &\equiv \mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) + \mu_0 \omega^2 \int ds ds' \mathbf{G}_0(\mathbf{r}, \mathbf{s}, \omega) \\ &\cdot \sum_{\mu, \mu'} \mathcal{P}_\mu(\mathbf{s}) W_{\mu, \mu'}(\omega) \mathcal{P}_{\mu'}^*(\mathbf{s}') \cdot \mathbf{G}_0(\mathbf{s}', \mathbf{r}', \omega). \quad (21) \end{aligned}$$

We can find that the function $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ satisfies the Eq. (15), therefore it can be interpreted as the Green function for the nonlocal Maxwell wave equation (14), and is just the one required in SSG theory [7]. Furthermore, Eq. (20) indicates that $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ identifies with the retarded correlation function for the electric field:

$$\begin{aligned} & -\mu_0 \hbar \omega^2 \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \\ &= -i \int_{t'}^{\infty} dt e^{i\omega(t-t')} \langle [\mathbf{E}(\mathbf{r}, t), \mathbf{E}(\mathbf{r}', t')] \rangle. \quad (22) \end{aligned}$$

Eqs. (20) and (15) have the same forms of Eqs. (8) and (5) respectively (there is a reciprocity relation $\mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) = \{\mathbf{G}_0(\mathbf{r}', \mathbf{r}, \omega)\}^t$ for isotropic media). This shows a good correspondence between our QED theory and SW's [6].

5 Summary We have constructed a QED theory for excitons with microscopic nonlocality by using the semiclassical microscopic nonlocal theory [14] and the QED theory for dispersive and absorptive local dielectrics [6]. Our

theory keeps good correspondences with them. Although only the linear process has been discussed in this paper, we can phenomenologically extend our theory to describe nonlinear processes, such as the entangled-photon generation via biexcitons in nano-structures [19]. In addition, our theory has a potential to systematically investigate higher order nonlinear processes of elementary excitations in condensed matters by using the Feynman diagram technique with correlation functions derived in the present paper. We are going to study various optical phenomena which cannot be discussed in the semiclassical framework in future works.

Acknowledgements The authors are grateful to Prof. K. Cho, Dr. H. Ajiki, and Dr. K. Koshino for helpful discussions. This work was partially supported by the Japan Society for the Promotion of Science (JSPS), a Grant-in-Aid for Creative Science Research, 17GS1204, 2005, and JSPS Research Fellowships for Young Scientists.

References

- [1] K. Edamatsu, G. Oohata, R. Shimizu, and T. Itoh, *Nature* **431**, 167 (2004).
- [2] S. Strauf, P. Michler, M. Klude, D. Hommel, G. Bacher, and A. Forchel, *Phys. Rev. Lett.* **89**, 177403 (2002).
- [3] J. P. Karr, A. Baas, R. Houdré, and E. Giacobino, *Phys. Rev. A* **69**, 031802 (2004).
- [4] B. Huttner and S. M. Barnett, *Phys. Rev. A* **46**, 4306 (1992).
- [5] L. Knöll, S. Scheel, and D. G. Welsch, *QED in dispersing and absorbing dielectric media*, in: *Coherence and Statistics of Photons and Atoms*, edited by J. Peřina, (Wiley-Interscience, 2001), chap. 1, pp. 1–64.
- [6] L. G. Suttrop and M. Wubs, *Phys. Rev. A* **70**, 013816 (2004).
- [7] S. Savasta, O. D. Stefano, and R. Girlanda, *Phys. Rev. A* **65**, 043801 (2002).
- [8] O. D. Stefano, S. Savasta, and R. Girlanda, *Phys. Rev. A* **60**, 1614 (1999).
- [9] H. Ishihara and K. Cho, *Phys. Rev. B* **53**, 15823 (1996).
- [10] K. Akiyama, N. Tomita, Y. Nomura, and T. Isu, *Appl. Phys. Lett.* **75**, 475 (1999).
- [11] H. Ishihara, T. Amakata, and K. Cho, *Phys. Rev. B* **65**, 035305 (2002).
- [12] H. Ishihara, K. Cho, K. Akiyama, N. Tomita, Y. Nomura, and T. Isu, *Phys. Rev. Lett.* **89**, 017402 (2002).
- [13] H. Ishihara, *Phys. Rev. B* **67**, 113302 (2003).
- [14] K. Cho, *Optical Response of Nanostructures: Microscopic Nonlocal Theory* (Springer-Verlag, 2003).
- [15] M. Bamba and H. Ishihara, arXiv:0708.2952v1 [physics.optics].
- [16] A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics*, (Dover, 1975), chap. 6.
- [17] W. C. Chew, *Waves and Fields in Inhomogeneous Media*, (IEEE PRESS, 1995).
- [18] O. J. F. Martin, C. Girard, and A. Dereux, *Phys. Rev. Lett.* **74**, 526 (1995).
- [19] M. Bamba and H. Ishihara, *phys. stat. sol. (c)* **3**, 3460 (2006).