Theory of the lifetime of an exciton incoherently created below its resonance frequency by inelastic scattering

Motoaki Bamba,[1,*] Shuji Wakaiki,[2] Hideki Ichida,[2,3] Kohji Mizoguchi,[4] DaeGwi Kim,[5] Masaaki Nakayama,[5] and Yasuo Kanematsu[2,3]

[1]*Department of Physics, Graduate School of Science, Osaka University, 1-1 Machikaneyama, Toyonaka, Osaka 560-0043, Japan*

[2]*Department of Material and Life Science, Division of Advanced Science and Biotechnology, Graduate School of Engineering, Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871, Japan*

[3]*Science and Technology Entrepreneurship Laboratory, Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871, Japan*

[4]*Department of Physical Science, Graduate School of Science, Osaka Prefecture University, 1-1 Gakuen, Naka-ku, Sakai, Osaka 599-8531, Japan*

[5]*Department of Applied Physics, Graduate School of Engineering, Osaka City University, 3-3-138 Sugimoto, Sumiyoshi-ku, Osaka 558-8585, Japan*

(Received 12 November 2014; revised manuscript received 5 April 2015; published 22 June 2015)

When an exciton in semiconductor is scattered and its energy is decreased far below the resonance energy of the bare exciton state, it has been considered that an exciton-polariton is created immediately by the scattering process because there is no exciton level at that energy. However, according to the recent time-resolved measurements of P emission originating from inelastic exciton-exciton scattering, it looks rather natural to consider that the exciton-polariton is created in a finite time scale which is restricted by a coherence volume of the exciton after the scattering. In this interpretation, the exciton remains in this time scale far below its resonance energy as a transient state in a series of processes. We propose an expression of the P-emission lifetime depending on the coherence volume of the scattered excitons through the conversion process from them to the polaritons. The coherence volume of the scattered excitons appears in the calculation of the inelastic scattering process on the assumption of a finite coherence volume of the bottleneck excitons. Time-resolved optical-gain measurements could be a way for investigating the validity of our interpretation.

DOI: 10.1103/PhysRevB.91.235205 PACS number(s): 78.20.Bh, 78.47.jd, 78.45.+h, 78.55.−m

I. INTRODUCTION

We can obtain a variety of properties of condensed matters from luminescence spectra by varying sample temperature, pumping frequency, pumping intensity, etc. [1]. Time-resolved luminescence measurements give us more detailed information especially about relaxation processes of the excitations such as excitons and polaritons. However, theoretical studies of the luminescence (spontaneous emission of the excitations) is not yet well developed probably due to the complexity of the relaxation dynamics involving spatial inhomogeneities, impurities, phonon scattering, spatial diffusion, inter-excitation scattering, and so on. The relaxation, dissipation, and dephasing processes have been investigated mainly by nonlinear optical responses such as pump-probe and four-wave mixing experiments. However, even by such measurements, it is still hard to obtain the complete knowledge of the luminescence process, especially the coherence volume of the excitation, which governs the emission lifetime [2–5].

Concerning the spontaneous emission of excitations at quasiequilibrium (equilibrium only in matters excluding the radiation field), the relation between the emission lifetime and the homogeneous spectral linewidth (reflecting the coherence volume) has been investigated for quasi-two-dimensional excitons in GaAs/AlGaAs quantum wells [2]. The coherence volume also gives the limit of the so-called exciton superradiance (size enhancement of radiative decay rate or of oscillator strength) [3–6] by which the emission lifetime is shortened with an increase in interaction volume between the radiation field and the center-of-mass wave function of excitons (radius of quantum dot). There were also attempts for estimating theoretically the coherence volume of excitations such as by dephasing rate [3]. However, the understanding of the coherence volume is not yet well developed because it is usually estimated only through the emission lifetime and the luminescence is in fact influenced by many other processes and factors, such as reabsorption of photons, stimulated emission of photons, diffusion of excitation, ballistic propagation of photons, penetration depth of pumping (spatial inhomogeneity), internal reflection, etc. [1].

Although the emission frequency is almost fixed for the spontaneous emission of excitons at the quasiequilibrium (called the bottleneck region [7–9] in the picture of exciton-polaritons), we can also observe luminescence peaks at lower frequencies, which involve the emission of optical phonons, inelastic exciton-exciton scattering (P emission), exciton-carrier scattering (H emission), and excitonic molecules (M emission) [1,10]. In the P-emission process, one exciton is inelastically scattered to a higher exciton state and the other one is scattered to the photonlike polariton branch as depicted in Fig. 1. It emerges under high-power pumping exceeding a threshold, and we have also an optical gain at the P-emission frequency [11–16]. The relaxation and scattering processes toward the P emission have been investigated in time-resolved measurements performed by optical Kerr gating method [17–25] and by streak camera [26,27], and then the following facts have been revealed. (i) The onset time

*Present address: Department of Materials Engineering Science, Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan; bamba@qi.mp.es.osaka-u.ac.jp

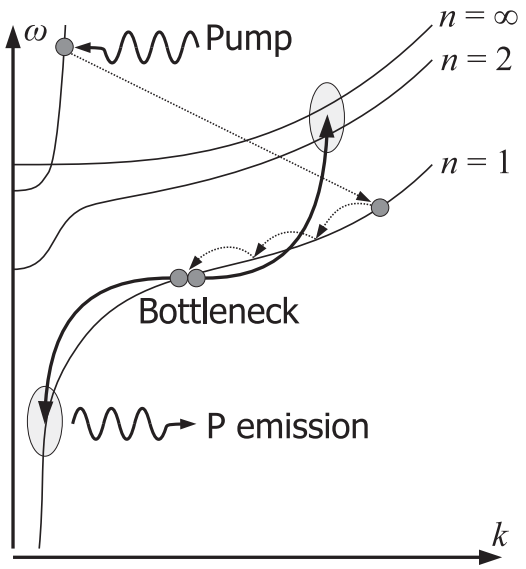

FIG. 1. Sketch of P-emission process depicted in dispersion relations. Excitons created by pumping are relaxed to the bottleneck region (dashed arrows). Then, they are scattered to higher exciton states with $n > 1$ and to photonlike polariton states with conserving the energy. The emission with the lower energy is called the P emission.

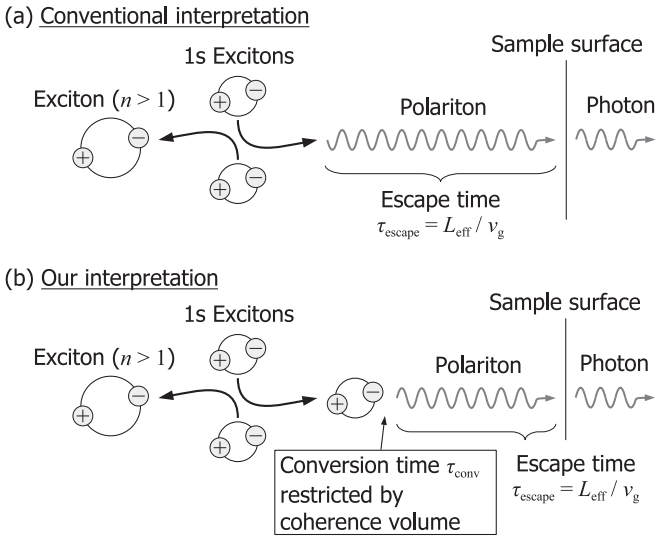

FIG. 2. Schematic diagrams of (a) conventional interpretation and (b) our interpretation of the dynamics toward the P emission. The escape time τ_{escape} of polariton is estimated to be quite short compared to the observed lifetime of the P emission. We interpret that the lifetime reflects the conversion time τ_{conv} from scattered excitons to polaritons. If the excitons after the inelastic scattering have a coherence length longer than the radiation wavelength, they can be converted quickly to polaritons as in the conventional interpretation. However, if the coherence length is quite short, it restricts the conversion time τ_{conv}, and our interpretation is rather appropriate.

reflects the time of energy relaxation of excitons toward the bottleneck region on the lower exciton-polariton branch [18,19,22]. (ii) The rise time reflects the rate of the inelastic scattering of excitons [17]. (iii) The peak frequency is changed temporally during the rise and decay periods [17,22,26]. Whereas this fact could be interpreted as the change of effective temperature (distribution) of excitons at the bottleneck, we can also interpret it as that the decay time (lifetime) of the P emission at each emission frequency strongly depends on that frequency.

The typical P-emission lifetimes are observed as a few ps [17–21,23–25] or a few tens of ps [22,26,27]. Although these lifetimes basically depend on materials of samples, they are generally much shorter than the emission lifetime τ_{emit} of bottleneck excitons at quasiequilibrium (in the order of nanoseconds). The time-resolved measurements revealed also that the P-emission lifetime is an increasing function of the emission frequency [20,21,23–25,27], and the lifetime at each emission frequency is almost independent of the pumping power [24,25]. Note that the emission-frequency dependence of the P-emission lifetime can be scaled phenomenologically by that of inverse of the group velocity of the photonlike polariton [21,23–25]. The lifetime of the spectrally integrated P-emission signal was shortened through the lowering of the peak frequency with an increase in pumping power (effective temperature) for InGaN [26]. However, it was almost unchanged for CuI [17,19] and AlGaN [20] because the peak frequencies were not strongly changed. Further, the lifetime of the spectrally integrated P-emission signal was also independent of the pumping frequency for CuI [19].

The P emission at each emission frequency shows an exponential decay in time. Its decay time is independent of the pumping power, and depends strongly on the emission frequency (inversely proportional to group velocity). Then, it is now recognized that the P-emission lifetime does not reflect the lifetime of excitons at the bottleneck, but it rather reflects the lifetime of quasiparticles (excitons or exciton-polaritons) after the inelastic scattering. In Ref. [17], the authors concluded that it reflects the lifetime of photonlike polaritons, which is considered to be shortened by the increase of photonic fraction of the polariton state. However, from the sample thickness and the group velocity of polaritons, the lifetime (escape time) of photonlike polaritons is estimated to be much shorter (tens of femtoseconds) than the P-emission lifetimes (picoseconds) observed in experiments [21,23]. On the other hand, in Ref. [20], the authors analyzed the P-emission decay as diffusive propagation of the photonlike polaritons, although the diffusion of light is usually discussed in strongly disordered media, where excitons should lose the memory of propagation direction quickly compared to the reemission time scale.

In this paper, from the viewpoint of the coherence volume, we try to propose the following interpretation of the P-emission lifetime: Just after the inelastic exciton-exciton scattering, the photonlike polariton is not immediately created, but the exciton remains with losing its energy in a time scale of picoseconds as depicted in Fig. 2. Then, the conversion time from the exciton to photonlike polariton, which is restricted by the coherence volume, is observed as the P-emission lifetime. Although the P emission has been considered as a stimulated emission of polaritons [1,11–16], we need to reconsider it as a stimulated creation (scattering) of excitons in our interpretation.

In Sec. II, we first estimate the interchange time between exciton and photon in the polariton states, the radiative recombination time of excitons, and the escape time of the polaritons. Only the radiative recombination time depends on the coherence volume. In Sec. III, we explain the detail of our interpretation of the P emission after the inelastic exciton-exciton scattering. Its justification and further discussion are performed in Sec. IV. The summary is shown in Sec. V.

II. CHARACTERISTIC TIME SCALES OF EXCITONS AND POLARITONS

We first calculate the exciton-photon interchange time in polariton states and the radiative recombination time of exciton from the Hamiltonian of light-matter coupling. We consider a homogeneous background medium with a relative dielectric constant $\varepsilon_{\rm bg}$, and the Hamiltonian of the radiation field in the background medium is written as

$$\hat{H}_{\rm rad} = \sum_{\eta=1,2}\sum_{\boldsymbol{k}} \hbar v|\boldsymbol{k}|\hat{a}^{\dagger}_{\boldsymbol{k},\eta}\hat{a}_{\boldsymbol{k},\eta}, \tag{1}$$

where $\hat{a}_{\boldsymbol{k},\eta}$ is the annihilation operator of a photon with wave vector $\boldsymbol{k}$ and polarization η, and $v = c/\sqrt{\varepsilon_{\rm bg}}$ is the speed of light in the background medium for the speed c in vacuum. The Hamiltonian of the light-matter coupling is expressed in the electric-dipole gauge as [28–30]

$$\hat{H}_{\rm LM} = -\frac{1}{\varepsilon_0\varepsilon_{\rm bg}}\int d\boldsymbol{r}\ \hat{\boldsymbol{P}}(\boldsymbol{r})\cdot\hat{\boldsymbol{D}}_{\perp}(\boldsymbol{r}). \tag{2}$$

Here, the transverse component of the electric displacement field is defined

$$\hat{\boldsymbol{D}}_{\perp}(\boldsymbol{r}) = \sum_{\eta=1,2}\sum_{\boldsymbol{k}} \boldsymbol{e}_{\boldsymbol{k},\eta} i\sqrt{\frac{\hbar\varepsilon_0\varepsilon_{\rm bg}v|\boldsymbol{k}|}{2V}}(\hat{a}_{\boldsymbol{k},\eta} - \hat{a}^{\dagger}_{-\boldsymbol{k},\eta})e^{i\boldsymbol{k}\cdot\boldsymbol{r}}, \tag{3}$$

where $\boldsymbol{e}_{\boldsymbol{k},\eta}$ is the unit vector perpendicular to $\boldsymbol{k}$, and V is the volume of the space. $\hat{\boldsymbol{P}}(\boldsymbol{r})$ represents the polarization density involving the creation and annihilation of an electron-hole pair as

$$\hat{\boldsymbol{P}}(\boldsymbol{r}) = d_{cv}\sum_{\xi}\boldsymbol{e}_{\xi}\sum_{\lambda}\delta(\boldsymbol{r}-\boldsymbol{R}_{\lambda})(\hat{\alpha}_{\xi,\boldsymbol{R}_{\lambda}}\hat{\beta}_{\xi,\boldsymbol{R}_{\lambda}} + \text{H.c.}). \tag{4}$$

Here, d_{cv} is the transition dipole moment calculated under the long-wavelength approximation, $\boldsymbol{e}_{\xi}$ is the unit vector in the direction $\xi = \{x,y,z\}$, and $\boldsymbol{R}_{\lambda}$ is the position of unit cell λ. $\hat{\alpha}_{\xi,\boldsymbol{R}_{\lambda}}$ and $\hat{\beta}_{\xi,\boldsymbol{R}_{\lambda}}$ are, respectively, annihilation operators of an electron and a hole at $\boldsymbol{R}_{\lambda}$ involving the polarization in the ξ direction. The optical interband transition is supposed to occur almost inside a unit cell. The annihilation operator of an exciton in state μ with a wave function $\psi_{\mu}(\boldsymbol{r})$ of the electron-hole relative motion and a center of mass at $\boldsymbol{R}_{\lambda}$ is written as

$$\hat{\sigma}_{\mu,\lambda} = \int d\boldsymbol{r}\ \frac{\psi_{\mu}(\boldsymbol{r})}{\sqrt{V_0}}\hat{\alpha}_{\xi_{\mu},\boldsymbol{R}_{\lambda}+(m_h/M)\boldsymbol{r}}\hat{\beta}_{\xi_{\mu},\boldsymbol{R}_{\lambda}-(m_e/M)\boldsymbol{r}}. \tag{5}$$

Here, m_e and m_h are the effective mass of the electron and hole, respectively, and $M = m_e + m_h$ is the total mass. V_0 is the volume of a unit cell. The wave function is normalized as $\int d\boldsymbol{r}\ \psi_{\mu}(\boldsymbol{r})^*\psi_{\mu'}(\boldsymbol{r}) = \delta_{\mu,\mu'}$. Due to the completeness of the exciton state $\sum_{\mu}\psi_{\mu}(\boldsymbol{r})^*\psi_{\mu}(\boldsymbol{r}') = \delta(\boldsymbol{r}-\boldsymbol{r}')$, the polarization density is rewritten as [31]

$$\hat{\boldsymbol{P}}(\boldsymbol{r}) = d_{cv}\sum_{\mu}\boldsymbol{e}_{\mu}\sqrt{V_0}\psi_{\mu}(0)\sum_{\lambda}\delta(\boldsymbol{r}-\boldsymbol{R}_{\lambda})(\hat{\sigma}_{\mu,\lambda} + \hat{\sigma}^{\dagger}_{\mu,\lambda}). \tag{6}$$

Here, we defined the wave function $\psi_{\mu}(\boldsymbol{r})$ to be real at $\boldsymbol{r} = 0$. The degree of freedom of the polarization direction ξ is included to the index μ of the exciton state. Using this expression, Eq. (2) is rewritten as

$$\hat{H}_{\rm LM} = -\sum_{\mu}\sum_{\eta=1,2}\sum_{\boldsymbol{k}}\boldsymbol{e}_{\mu}\cdot\boldsymbol{e}_{\boldsymbol{k},\eta} i\sqrt{\frac{\hbar v|\boldsymbol{k}|{d_{cv}}^2\psi_{\mu}(0)^2}{2\varepsilon_0\varepsilon_{\rm bg}N}} \times\sum_{\lambda}(\hat{\sigma}_{\mu,\lambda} + \hat{\sigma}^{\dagger}_{\mu,\lambda})(\hat{a}_{\boldsymbol{k},\eta} - \hat{a}^{\dagger}_{-\boldsymbol{k},\eta})e^{i\boldsymbol{k}\cdot\boldsymbol{R}_{\lambda}}. \tag{7}$$

Here, $N = V/V_0$ is the number of the unit cells in the whole space. For Bohr radius a_B^* of the exciton larger enough than the lattice constant (in the limit of Wannier exciton) [31], the s-orbital wave function of the electron-hole relative motion is expressed as

$$\psi_{ns}(0) = \sqrt{\frac{1}{\pi {a_B^*}^3}\frac{1}{n^3}}. \tag{8}$$

A. Exciton-photon interchange time

We define the exciton operator in the $\boldsymbol{k}$ representation as

$$\hat{\sigma}_{\mu,\boldsymbol{k}} = \frac{1}{\sqrt{N}}\sum_{\lambda}e^{-i\boldsymbol{k}\cdot\boldsymbol{R}_{\lambda}}\hat{\sigma}_{\mu,\lambda}. \tag{9}$$

The Hamiltonian of the excitons is represented as

$$\hat{H}_{\rm ex} = \sum_{\mu}\sum_{\boldsymbol{k}}\hbar\Omega_{\mu,k}\hat{\sigma}^{\dagger}_{\mu,\boldsymbol{k}}\hat{\sigma}_{\mu,\boldsymbol{k}} + \frac{1}{2\varepsilon_0\varepsilon_{\rm bg}}\int d\boldsymbol{r}\ \hat{\boldsymbol{P}}(\boldsymbol{r})\cdot\hat{\boldsymbol{P}}(\boldsymbol{r}). \tag{10}$$

Here, $\Omega_{\mu,k}$ is the eigenfrequency of exciton in state μ with wave number k. The last term is the so-called P^2 term and represents the depolarization shift [28–30]. The light-matter coupling Hamiltonian given by Eq. (7) is rewritten as

$$\hat{H}_{\rm LM} = -\sum_{\mu}\sum_{\eta=1,2}\sum_{\boldsymbol{k}}\boldsymbol{e}_{\mu}\cdot\boldsymbol{e}_{\boldsymbol{k},\eta} \times i\hbar g_{\mu,k}(\hat{\sigma}_{\mu,-\boldsymbol{k}} + \hat{\sigma}^{\dagger}_{\mu,\boldsymbol{k}})(\hat{a}_{\boldsymbol{k},\eta} - \hat{a}^{\dagger}_{-\boldsymbol{k},\eta}), \tag{11}$$

where the coupling strength is defined as

$$g_{\mu,k} = \sqrt{\frac{vk{d_{cv}}^2\psi_{\mu}(0)^2}{2\hbar\varepsilon_0\varepsilon_{\rm bg}}}. \tag{12}$$

When the fermionic nature of the exciton can be neglected in the one-body problem with respect to the exciton, the eigenstates of the electromagnetic fields in this excitonic medium are the polariton states satisfying the dispersion relation (roughly sketched in Fig. 1) as

$$\frac{c^2k^2}{\omega^2} = \varepsilon_{\rm bg} + \sum_{\mu}\frac{4\pi\beta_{\mu,k}{\Omega_{\mu,k}}^2}{{\Omega_{\mu,k}}^2 - (\omega + i0^+)^2} = \varepsilon(\omega,k), \tag{13}$$

where the nondimensional factor is defined as

$$4\pi\beta_{\mu,k} = \frac{4\varepsilon_{\rm bg}{g_{\mu,k}}^2}{\Omega_{\mu,k}vk} = \frac{2{d_{cv}}^2\psi_{\mu}(0)^2}{\varepsilon_0\hbar\Omega_{\mu,k}}. \tag{14}$$

When the polaritons exist stably in a large enough medium with negligible dissipation, the interchange rate between exciton state μ and photon one is estimated from Eq. (12) for $k = \Omega_\mu/v$ as

$$g_\mu = \sqrt{\frac{\Omega_{\mu,k} {d_{cv}}^2 {\psi_\mu(0)}^2}{2\hbar\varepsilon_0\varepsilon_{\rm bg}}} = \sqrt{\frac{\pi\beta_\mu {\Omega_\mu}^2}{\varepsilon_{\rm bg}}}. \tag{15}$$

For A exciton state with $n = 1$ $(1s)$ in ZnO [32], we have $\Omega_{{\rm A},1s} = 3.375$ eV, $\varepsilon_{\rm bg} = 4$, and $\Delta_{{\rm A},1s} = 4\pi\beta_{{\rm A},1s}\Omega_{{\rm A},1s}/\varepsilon_{\rm bg} = 5.74$ meV ($\Omega_{{\rm B},1s} = 3.381$ eV and $\Delta_{{\rm B},1s} = 6.62$ meV for B exciton). The interchange rate is then estimated as $\hbar g_{{\rm A},1s} = 70$ meV ($\hbar g_{{\rm B},1s} = 75$ meV). The interchange time $\tau_{\rm Rabi} = 2\pi/g_\mu = 0.06$ ps is one or two orders of magnitude shorter than the P-emission lifetime observed in the experiments [21,23,24].

B. Radiative recombination time of exciton

Let us next calculate the radiative recombination rate of excitons from the light-matter-coupling Hamiltonian (7). Here, we suppose an exciton in state μ as an initial state and its center of mass is localized at $\boldsymbol{R}_\lambda$. According to the Fermi's golden rule, the transition rate from the exciton state to one photon state for any $\boldsymbol{k}$ and η is obtained as

$$\begin{aligned}\gamma_\mu &= \frac{2\pi}{\hbar}\sum_{\eta,\boldsymbol{k}} |\langle 0|\hat{a}_{\boldsymbol{k},\eta}\hat{H}_{\rm LM}\hat{\sigma}^\dagger_{\mu,\lambda}|0\rangle|^2\delta(\hbar\Omega_\mu - \hbar v|\boldsymbol{k}|) \\ &= \frac{{\Omega_\mu}^3 {d_{cv}}^2 {\psi_\mu(0)}^2 V_0}{3\pi\hbar\varepsilon_0\varepsilon_{\rm bg}v^3} = \frac{2{g_\mu}^2{\Omega_\mu}^2 V_0}{3\pi v^3},\end{aligned} \tag{16}$$

where $|0\rangle$ is the vacuum state and we used the following relation for arbitrary function $F(k)$:

$$\sum_{\eta=1,2}\int d\boldsymbol{k}\ |\boldsymbol{e}_\mu\cdot\boldsymbol{e}_{\boldsymbol{k},\eta}|^2 F(|\boldsymbol{k}|) = \int_0^\infty dk\ \frac{8\pi k^2}{3}F(k). \tag{17}$$

For ZnO, the lattice constants are $a = 3.25$ Å and $c = 5.21$ Å [32], and then the volume of the unit cell is

$$V_0 = \frac{1}{2}\frac{\sqrt{3}}{2}\times(3.25\ \text{Å})^2\times 5.21\ \text{Å} = 24\ \text{Å}^3. \tag{18}$$

Therefore, the radiative recombination rate (16) is estimated for the A excitons as

$$\gamma_{{\rm A},1s} = 0.45\ (\mu\text{s})^{-1}. \tag{19}$$

This rate is quite low even compared to the spontaneous emission rate $1/\tau_{\rm emit}$ of bottleneck excitons observed in luminescence experiments (usually in the order of nanoseconds).

This is because the center of mass of exciton is in fact not localized at a unit cell, but it coherently spreads in a finite volume, which is called the coherence volume $V^\mu_{\rm coh}$. Here, we suppose that such exciton state in state μ is represented with a center-of-mass wave function $\Phi_\mu(\boldsymbol{r})$ as

$$|\text{ex}_\mu\rangle = \sum_\lambda \sqrt{V_0}\Phi_\mu(\boldsymbol{R}_\lambda)\hat{\sigma}^\dagger_{\mu,\lambda}|0\rangle, \tag{20}$$

where the wave function is normalized as $\int d\boldsymbol{r}\ |\Phi_\mu(\boldsymbol{r})|^2 = 1$. Instead of Eq. (16), the radiative recombination rate of this exciton state is written as

$$\begin{aligned}\Gamma_\mu &= \frac{2\pi}{\hbar}\sum_{\eta,\boldsymbol{k}} |\langle 0|\hat{a}_{\boldsymbol{k},\eta}\hat{H}_{\rm LM}|\text{ex}_\mu\rangle|^2\delta(\hbar\Omega_\mu - \hbar v|\boldsymbol{k}|) \\ &= \frac{2\pi}{\hbar}\sum_{\eta,\boldsymbol{k}} |\langle 0|\hat{a}_{\boldsymbol{k},\eta}\hat{H}_{\rm LM}\hat{\sigma}^\dagger_{\mu,\lambda}|0\rangle\phi_{\mu,\boldsymbol{k}}|^2\delta(\hbar\Omega_\mu - \hbar v|\boldsymbol{k}|),\end{aligned} \tag{21}$$

where the factor $\phi_{\mu,\boldsymbol{k}}$ is defined as

$$\phi_{\mu,\boldsymbol{k}} = \sum_\lambda e^{i\boldsymbol{k}\cdot\boldsymbol{R}_\lambda}\sqrt{V_0}\Phi_\mu(\boldsymbol{R}_\lambda) = \int d\boldsymbol{r}\ \frac{e^{i\boldsymbol{k}\cdot\boldsymbol{r}}}{\sqrt{V_0}}\Phi_\mu(\boldsymbol{r}). \tag{22}$$

Here, we suppose that the coherence length $(V^\mu_{\rm coh})^{1/3}$ is shorter enough than the radiation wavelength $2\pi/|\boldsymbol{k}|$. Further, the amplitude of the center-of-mass wave function $\Phi_\mu(\boldsymbol{r})$ is supposed to be almost homogeneous, i.e., $\Phi_\mu(\boldsymbol{r}) = 1/\sqrt{V^\mu_{\rm coh}}$ in the coherence volume $V^\mu_{\rm coh}$. Then, $\phi_{\boldsymbol{k}}$ does not depend on $\boldsymbol{k}$, and its absolute value is estimated as

$$|\phi_{\mu,\boldsymbol{k}}| = \int d\boldsymbol{r}\ \frac{\Phi_\mu(\boldsymbol{r})}{\sqrt{V_0}} = \sqrt{\frac{V^\mu_{\rm coh}}{V_0}}. \tag{23}$$

Substituting this into Eq. (21), the radiative recombination rate of excitons in state μ with the coherence volume $V^\mu_{\rm coh}$ is obtained as

$$\Gamma_\mu = \gamma_\mu\frac{V^\mu_{\rm coh}}{V_0}. \tag{24}$$

In this way, we get the size enhancement of the radiative recombination rate (in other words, that of oscillator strength), and it is called the exciton superradiance [3–6]. When the coherence length $(V^\mu_{\rm coh})^{1/3}$ is comparable to or larger than the wavelength of the radiation, we have to consider the crossover to the polariton picture [33].

Note that the interchange time $\tau_{\rm Rabi}$ [also the dispersion relation (13)] is obtained without the concept of the coherence volume. This means that all the atoms associate with each other coherently for the interchange, while only the atoms in the coherence volume associate for the emission from localized exciton. In other words, the interchange reflects the coherence volume of the electromagnetic fields (widely spread by the propagation), while the spontaneous emission reflects that of bare exciton. Once a photon is emitted from the bare exciton, it then gets a spatial coherence by propagating in the medium as a polariton, if dissipations and dephasing are weak enough compared to the light-matter coupling. This idea is important to understand the lifetime of the P emission in the next section.

C. Escape time of polariton

We next consider another time scale, the escape time of polaritons. We suppose a film of the excitonic medium with a thickness L, and it is thick enough compared to the radiation wavelength. When polariton states are supposed to be a good quantum state, the escape time of polariton can be estimated from its group velocity $v_{\rm g} = \partial\omega/\partial k$ [33–35], which is derived from Eq. (13). If the film surfaces directly contact to external regions, the Fresnel reflectance coefficients from inside to

outside are obtained as

$$r_j(\omega) = \frac{k(\omega) - q_j(\omega)}{k(\omega) + q_j(\omega)}. \tag{25}$$

Here, $k(\omega)$ and $q_j(\omega)$ are wave numbers perpendicular to the surfaces between the film and external regions ($j = 1,2$), respectively, and are defined as

$$k(\omega) = \sqrt{\varepsilon(\omega)\omega^2/c^2 - k_\parallel{}^2}, \tag{26}$$

$$q_j(\omega) = \sqrt{\varepsilon_j\omega^2/c^2 - k_\parallel{}^2}, \tag{27}$$

for wave number $k_\parallel$ parallel to the surfaces and relative dielectric constants ε_j of the two external regions. The escape rate $\gamma_{\rm escape}(\omega)$ of polariton at frequency ω is calculated as follows [33,35]. After a round trip in the film with a time of $2L/v_{\rm g}(\omega)$, the density of polaritons decreases by a factor of $\exp[-2\gamma_{\rm escape}(\omega)L/v_{\rm g}(\omega)]$, and it is equal to the factor $|r_1(\omega)r_2(\omega)|^2$ due to the loss at the two surfaces. Then, the escape rate of polariton in a film is obtained as

$$\gamma_{\rm escape}(\omega) = \frac{v_{\rm g}(\omega)}{2L}\ln\frac{1}{|r_1(\omega)r_2(\omega)|^2} = \frac{v_{\rm g}(\omega)}{L_{\rm eff}(\omega)}, \tag{28}$$

where

$$L_{\rm eff}(\omega) = -\frac{2L}{\ln|r_1(\omega)r_2(\omega)|^2} \tag{29}$$

is the effective length for the polariton propagation. This escape time $\tau_{\rm escape} = 1/\gamma_{\rm escape}(\omega)$ reflecting the macroscopic propagation of polaritons is another time scale in the processes of the P emission. When the effective thickness is around $L_{\rm eff} \sim 5\ \mu$m, the escape time is estimated as $\tau_{\rm escape} \sim 0.1$ ps for the P-emission frequency region in ZnO [21,23,24]. This is also quite short compared to the observed P-emission lifetime.

III. INTERPRETATION OF P-EMISSION LIFETIME

Let us consider fundamentally a series of processes after the inelastic exciton-exciton scattering at the bottleneck region until photons come out from the sample. According to the conventional interpretation of the P emission, as depicted in Fig. 2(a), one of the scattered excitons is converted to a photonlike polariton almost immediately, because there are only the photonlike polariton states (eigenstates of electromagnetic fields in medium) at the P-emission frequency. In this interpretation, when polaritons are stabilized by a large enough transition dipole, they are created in the time scale of the exciton-photon interchange time $\tau_{\rm Rabi} = 2\pi/g_\mu$ of the polariton state, and it is certainly negligible ($\tau_{\rm Rabi} \sim 0.06$ ps in ZnO) compared to the other time scales except the escape time $\tau_{\rm escape}$ of polariton (then there is a crossover around the material size comparable to the radiation wavelength [33]). Then, if the P-emission lifetimes do not originate from the lifetime of excitons at the bottleneck, obeying the conventional interpretation, we need the interpretations of the polariton diffusion [20] or of the polariton escape from a sample with an incredibly large effective thickness [21,23].

Let us examine whether this conventional interpretation is really justified or not from a fundamental viewpoint. First of all, even if the polariton states (or photons outside the sample) are the final states in the processes of the P emission, we can consider intermediate states between the inelastic scattering and the escape of polaritons from the sample. In fact, since the scattering originates from the Coulomb interaction or the fermionic nature of excitons, we originally get two excitons just after the scattering. The key problem is whether the scattered exciton is converted to the polariton in the time scale of $\tau_{\rm Rabi}$ or not.

As discussed in the previous section, the polariton picture is justified only when excitons have a long enough spatial coherence, e.g., when they are created by light irradiation or after the emission from localized excitons. In contrast, when the incoherent excitons at the bottleneck are scattered with each other, we can consider that the excitons just after the scattering have only a poor spatial coherence. The conversion from the scattered excitons to polaritons (or photons outside) is rather similar as the emission process from localized excitons, and the conversion time can be restricted by the coherence volume $V_{\rm coh}^{\mu}$ of the scattered excitons.

A. Conversion time from exciton to polariton

Obeying the above scenario, in order to estimate the conversion time from exciton to polariton, we need to extend the discussion of the radiative recombination rate of excitons with the coherence volume in Sec. II B.

Although the conversion rate from exciton to photon is calculated in Sec. II B, the created photon can be reabsorbed in the excitonic medium with a large enough size and a large enough transition dipole. This reabsorption is one of the critical problems for discussing the emission lifetime of excitons. Concerning the emission at the bottleneck region, the created photon is reabsorbed with a relatively high probability because the emission frequency is very close to the exciton resonance. Furthermore, the recreated exciton loses rapidly the memories of the phase and the propagation direction. Then, even after the creation of the photon, it gets hardly a spatial coherence, and we should consider a repetition of photon creation, reabsorption, and dephasing of exciton. This is one of the reasons why the emission lifetime of bottleneck excitons is hard to be discussed.

On the other hand, the problem can be simplified when we discuss the P emission. Since the emission frequency is far below the exciton resonance (e.g., about 0.1 eV for ZnO [21,23,24]), even if the photon is reabsorbed, the created exciton emits a photon again without losing the memories of the phase and the propagation direction, i.e., the absorption coefficient is negligible at that frequency. Then, after the conversion from a localized exciton to a photon, we can consider simply the series of absorption and creation of a photon in the excitonic medium without the dephasing or scattering process. In such a case, the propagation of the created photon can be described by that of the polariton. Then, instead of the conversion rate from an exciton to a photon calculated in Sec. II B, we here calculate the conversion rate from an exciton to a polariton because the polariton states are the eigenstates in the medium.

Under the bosonization of the exciton, the Hamiltonian $\hat{H}_{\rm pol} = \hat{H}_{\rm rad} + \hat{H}_{\rm LM} + \hat{H}_{\rm ex}$ of photons and excitons can be

diagonalized as [36,37]

$$\hat{H}_{\rm pol} = \sum_{\eta=1,2} \sum_{j} \sum_{\boldsymbol{k}} \hbar\omega_{j,k} \hat{p}^{\dagger}_{j,\boldsymbol{k},\eta} \hat{p}_{j,\boldsymbol{k},\eta}. \tag{30}$$

Here, $\hat{p}_{j,\boldsymbol{k},\eta}$ is the annihilation operator of a polariton in state j with wave vector $\boldsymbol{k}$ and polarization direction η, and it is represented approximately for $g_{\mu,k} \ll \Omega_{\mu,k}$ by the sum of the annihilation operators of photon and exciton as

$$\hat{p}_{j,\boldsymbol{k},\eta} \simeq C_{j,\boldsymbol{k}} \hat{a}_{\boldsymbol{k},\eta} + \sum_{\mu} X_{j,\boldsymbol{k},\eta,\mu} \hat{\sigma}_{\mu,\boldsymbol{k}}. \tag{31}$$

The coefficients are determined by the parameters in the Hamiltonian $\hat{H}_{\rm pol} = \hat{H}_{\rm rad} + \hat{H}_{\rm LM} + \hat{H}_{\rm ex}$ [36,37]. The eigenfrequency of the polariton state is represented as $\omega_{j,k}$, and it satisfies $c^2k^2/{\omega_{j,k}}^2 = \varepsilon(\omega_{j,k},k)$ for the dielectric function defined in Eq. (13). For frequency ω corresponding to the photonlike region of the lowest polariton branch ($j = L$), the photonic fraction of the polariton state is approximately represented by its group velocity $v_{\rm g}(\omega)$ as

$$A(\omega) = \left|C_{L,k_L(\omega)}\right|^2 \simeq \frac{v_{\rm g}(\omega)}{v}, \tag{32}$$

where $k_L(\omega)$ satisfies $c^2k_L(\omega)^2/\omega^2 = \varepsilon(\omega,k_L(\omega))$. Intuitively, if the group velocity is slowed as $v_{\rm g}(\omega) = A(\omega)v$ compared to the speed v of light in the background medium, the polariton propagates as a photon in the fraction $A(\omega)$ and as an exciton (its velocity is negligible) in the rest $1 - A(\omega)$.

After the conversion from the localized exciton to a photon, it propagates as a polariton stably in the medium in the P-emission frequency region, and the interchange rate $g_{\mu,k}$ is much higher than the conversion rate Γ_{μ}. Then, before the escape of the polariton from the sample, the final state can be supposed as the lowest polariton state $\hat{p}^{\dagger}_{L,\boldsymbol{k},\eta}|0\rangle$. The initial state is the exciton after the inelastic scattering, and here it is supposed as a mixed one concerning several exciton's relative-motion states represented in Eq. (20), i.e., the density operator of one exciton state is expressed as

$$\hat{\rho}^{\rm one}_{\rm init} = \sum_{\mu} f_{\mu} |{\rm ex}_{\mu}\rangle\langle{\rm ex}_{\mu}|, \tag{33}$$

where f_{μ} represents the probability of being in the state μ ($\sum_{\mu} f_{\mu} = 1$). Instead of Eq. (21), the conversion rate is derived as

$$\begin{aligned}\Gamma(\omega) &= \frac{2\pi}{\hbar} \sum_{\eta,\boldsymbol{k},\mu} f_{\mu} |\langle 0|\hat{p}_{L,\boldsymbol{k},\eta} \hat{H}_{\rm LM}|{\rm ex}_{\mu}\rangle|^2 \delta(\hbar\omega - \hbar\omega_{L,k}) \\ &= \frac{2\pi}{\hbar} \sum_{\eta,\boldsymbol{k},\mu} f_{\mu} |C_{L,k}|^2 |\langle 0|\hat{a}_{\boldsymbol{k},\eta} \hat{H}_{\rm LM}|{\rm ex}_{\mu}\rangle|^2 \delta(\hbar\omega - \hbar\omega_{L,k}) \\ &= A(\omega) \sum_{\mu} f_{\mu}\gamma_{\mu} \frac{V^{\mu}_{\rm coh}}{V_0} \left[\frac{vk_L(\omega)}{\Omega_{\mu,k_L(\omega)}}\right]^3. \end{aligned} \tag{34}$$

Through the factor $A(\omega)$, this conversion rate includes the contribution of reabsorption and reemission of the photon but without the dephasing or the scattering process. Note that, whereas the first line is derived from the Fermi's golden rule, the energy $\hbar\omega$ corresponds to the eigenenergy of the final state $\hat{p}^{\dagger}_{L,\boldsymbol{k}_L(\omega),\eta}|0\rangle$ but not the energy of the initial state $\hat{\rho}^{\rm one}_{\rm init}$ or $|{\rm ex}_{\mu}\rangle$, which is far above the emission energy $\hbar\omega$. This point will be discussed in Sec. IV.

The ω dependence comes from the two factors: $A(\omega) \propto v_{\rm g}(\omega)$ and $[vk_L(\omega)/\Omega_{\mu,k_L(\omega)}]^3$. Around the P-emission frequency in most of the materials, the former gives the dominant contribution than the latter, which is almost unity and gives a slight ω dependence. For example, in ZnO, we have $\hbar\Omega_{{\rm A},1s} = 3.375$ eV and $\hbar\omega \sim \hbar\Omega_{{\rm A},1s} - 0.1$ eV. Since the frequency difference 0.1 eV is in the same order as $\hbar g_{{\rm A},1s} = 70$ meV, the photonic fraction $A(\omega) \propto v_{\rm g}(\omega)$ gives the dominant contribution around this frequency region, and the conversion rate can be approximately expressed as

$$\Gamma(\omega) \simeq A(\omega) \sum_{\mu} f_{\mu}\gamma_{\mu} \frac{V^{\mu}_{\rm coh}}{V_0}. \tag{35}$$

This can be the reason why the P-emission lifetime is observed to be inversely proportional to the group velocity $v_{\rm g}(\omega)$ in the experiments for bulk materials [21,23,24]. Whereas the escape time $\tau_{\rm escape}$ of polaritons after the conversion is also inversely proportional to the group velocity $v_{\rm g}(\omega)$, it is estimated to be quite short compared to the conversion time $\tau_{\rm conv} = 1/\Gamma(\omega)$. In this way, the P-emission lifetime basically reflects the conversion time $\tau_{\rm conv}$ from the scattered excitons to the polaritons in our interpretation.

B. Estimation of coherence volume

From the experimental data for ZnO [24,25], we here estimate the coherence volume of the scattered excitons based on the expression (34) of the exciton-to-polariton conversion rate $\Gamma(\omega)$. Since the A and B excitons are the lowest two states, here we tentatively consider that these two exciton states are mostly created at the P-emission frequency region by the inelastic scattering, i.e., $f_{{\rm A},1s} + f_{{\rm B},1s} = 1$. Further, the radiative recombination rates for exciton localized in a unit cell are similar $\gamma_{{\rm A},1s} \sim \gamma_{{\rm B},1s}$ for the two exciton states because we have $g_{{\rm A},1s} \sim g_{{\rm B},1s}$ as discussed in Sec. II A. The eigenfrequencies are also similar as $\Omega_{{\rm A},1s} \sim \Omega_{{\rm B},1s}$. Then, the conversion rate is rewritten approximately as

$$\Gamma(\omega) = A(\omega) \left[\frac{vk_L(\omega)}{\Omega_{{\rm A},1s,k_L(\omega)}}\right]^3 \Gamma', \tag{36}$$

where the ω-independent decay rate is defined with an averaged coherence volume $V_{\rm coh} = f_{{\rm A},1s} V^{{\rm A},1s}_{\rm coh} + f_{{\rm B},1s} V^{{\rm B},1s}_{\rm coh}$ as

$$\Gamma' = \gamma_{{\rm A},1s} \frac{V_{\rm coh}}{V_0}. \tag{37}$$

From the experimentally obtained P-emission lifetimes, we estimated $\Gamma' = (0.8\ {\rm ps})^{-1}$. Then, from the radiative recombination rate $\gamma_{{\rm A},1s} = 0.45\ (\mu{\rm s})^{-1}$ derived for an exciton localized at a unit cell in Eq. (19), the coherence volume is estimated as

$$V_{\rm coh} = 6 \times 10^7\ ({\rm \AA})^3, \tag{38}$$

and the coherence length is $(V_{\rm coh})^{1/3} = 4 \times 10^1$ nm. Although we have currently no other way to evaluate the coherence volume (length) experimentally, this value is certainly shorter than the radiation wavelength ($\sim 2 \times 10^2$ nm for $\hbar\omega = 3.26$ eV in the background medium with $\varepsilon_{\rm bg} = 4$).

In this way, from the fundamental viewpoint, we should consider the coherence volume of the scattered excitons, and the conversion time from the exciton to the photonlike polariton can explain the observed P-emission lifetime, which is much shorter than the emission lifetime $\tau_{\rm emit}$ of the bottleneck excitons, longer than the escape time $\tau_{\rm escape}$ of polaritons, and inversely proportional to the group velocity approximately for bulk materials. Whereas the discussion in this paper does not deny the interpretation of the polariton diffusion [20], it is noted that the decrease in diffusion constant with an increase in impurity concentration reported in Ref. [20] can be explained as a decrease in coherence volume $V_{\rm coh}$ in our interpretation.

In the next section, we try to justify our interpretation against some counterintuitive points.

IV. DISCUSSION

Since the final states certainly exist as the polariton states or photon states outside the sample, the inelastic scattering to these destinations is not forbidden. However, the scattered excitons remain in the bare exciton states in the conversion time $\tau_{\rm conv} \sim 1$ ps, although the eigenfrequencies $\Omega_{\mu,k}$ of these states are far above the emission frequency ω ($\hbar\Omega_{{\rm A},1s} = 3.375$ eV and $\hbar\Omega_{{\rm A},1s} - \hbar\omega \sim 0.1$ eV for ZnO [24,25]). In the conventional interpretation, the inelastic scattering of the two excitons is resonant to both the higher exciton state with $n > 1$ and the photonlike polariton one. In contrast, in our interpretation, it is resonant only to the higher exciton state but not to the lower one (no exciton state at the P-emission frequency). However, even if one process is not resonant, it can occur in a series of processes.

Since the A and B exciton states with $n = 1$ are most resonant compared to the other exciton states ($\Omega_{{\rm A},1s}$ and $\Omega_{{\rm B},1s}$ are closest to the emission frequency ω), the scattered excitons are supposed to be mostly in the lowest two exciton states ($f_{{\rm A},1s} + f_{{\rm B},1s} \sim 1$ and $f_{{\rm A},1s} > f_{{\rm B},1s}$).[1] These facts justify the estimation of the coherence volume $V_{\rm coh}$ in Sec. III B.

In our interpretation, the scattered excitons remain in the bare exciton states not as the so-called virtual state, whose lifetime is determined by the Heisenberg uncertainty principle [38] such as $2\pi/(\Omega_{{\rm A},1s} - \omega) \sim 0.04$ ps $\ll \tau_{\rm conv}$ in our case. The P-emission process can be a good example for investigating the validity conditions of the virtual-state picture. From the experimentally observed lifetime, we conclude that the virtual-state picture is not appropriate for the P emission process. In order to investigate theoretically the validity conditions, we must consider explicitly the series of the processes including the inelastic scattering as will be discussed in Sec. IV A.

[1]In our interpretation, since the exciton is scattered not directly to the photonlike polariton state, dark exciton states can also be the transient state if the final destination exists. However, we cannot say anything about the dark excitons from the current experiments of the P emission.

Intuitively, the bottleneck excitons are scattered to unstable transient states (bare exciton states) with a lifetime of $\tau_{\rm conv}$. If the dephasing time of the higher excitons ($n > 1$) is shorter than $\tau_{\rm conv}$, the emission frequency ω is fixed during the excitons remaining in the transient states. Such transient states are surely unstable, and then $\tau_{\rm conv}$ is much shorter than the emission lifetime $\tau_{\rm emit}$ of excitons at the bottleneck region. The conversion time $\tau_{\rm conv}$ becomes shorter (less stable) with a decrease in the emission frequency (more distant from the bottleneck frequency).

The conversion rate $\Gamma(\omega)$ from exciton to polariton was calculated in Eq. (34). In this derivation, the emission energy $\hbar\omega$ corresponds to the energy of the final state (polariton) but is lower than that of the initial state $\hat{\rho}^{\rm one}_{\rm init}$, which is at least higher than the lowest exciton state ${\rm Tr}(\hat{H}_{\rm ex}\hat{\rho}^{\rm one}_{\rm init}) > \hbar\Omega_{{\rm A},1s,k=0}$. On the other hand, in the spontaneous emission from the bottleneck excitons, we can suppose $\hbar\omega \sim \hbar\Omega_\mu$, and the exciton state $|{\rm ex}_\mu\rangle$ can be supposed well as an initial state. In order to describe more rigorously the transient state in the P-emission process, we try to discuss the series of processes including the inelastic processes in the following.

A. Inelastic exciton-exciton scattering

In order to examine strictly whether the lifetime of the transient state $|{\rm ex}_\mu\rangle$ is really restricted by the coherence volume, instead of starting from the scattered excitons as in the previous section, we need to consider the series of processes of the P emission from the inelastic scattering to the creation of the polaritons. Here, we treat the exciton operator $\hat{\sigma}_{\mu,\boldsymbol{k}}$ as bosonic one and suppose the Hamiltonian of the exciton-exciton interaction as

$$\hat{H}_{\text{ex-ex}} = \sum_{\mu,\nu,\mu',\nu'}\sum_{\boldsymbol{k},\boldsymbol{k}',\boldsymbol{q}} \frac{\hbar\mathcal{V}_{\mu,\mu',\nu',\nu,\boldsymbol{q}}}{2}\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}}\hat{\sigma}^\dagger_{\mu',\boldsymbol{k}'}\hat{\sigma}_{\nu',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{\nu,\boldsymbol{k}+\boldsymbol{q}}, \tag{39}$$

where the scattering coefficient satisfies $\mathcal{V}_{\mu,\mu',\nu',\nu,\boldsymbol{q}} = \mathcal{V}_{\mu,\mu',\nu',\nu,-\boldsymbol{q}} = \mathcal{V}_{\mu',\mu,\nu',\nu,\boldsymbol{q}} = \mathcal{V}_{\nu,\nu',\mu',\mu,\boldsymbol{q}}$. From the Hamiltonian $\hat{H}_{\rm rad} + \hat{H}_{\rm LM} + \hat{H}_{\rm ex} + \hat{H}_{\text{ex-ex}}$, the Heisenberg equations of $\hat{a}_{\boldsymbol{k},\eta}$ and $\hat{\sigma}_{\mu,\boldsymbol{k}}$ are derived under the rotating-wave approximation as

$$i\frac{d}{dt}\hat{a}_{\boldsymbol{k},\eta} = v|\boldsymbol{k}|\hat{a}_{\boldsymbol{k},\eta} + \sum_\mu i g_{\mu,\boldsymbol{k},\eta}\hat{\sigma}_{\mu,\boldsymbol{k}}, \tag{40a}$$

$$i\frac{d}{dt}\hat{\sigma}_{\mu,\boldsymbol{k}} = \Omega_{\mu,k}\hat{\sigma}_{\mu,\boldsymbol{k}} - \sum_\eta i g_{\mu,\boldsymbol{k},\eta}\hat{a}_{\boldsymbol{k},\eta} + \sum_{\nu,\mu',\nu'}\sum_{\boldsymbol{k}',\boldsymbol{q}}\mathcal{V}_{\mu,\mu',\nu',\nu,\boldsymbol{q}}\hat{\sigma}^\dagger_{\mu',\boldsymbol{k}'}\hat{\sigma}_{\nu',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{\nu,\boldsymbol{k}+\boldsymbol{q}}, \tag{40b}$$

where $g_{\mu,\boldsymbol{k},\eta} = \boldsymbol{e}_\mu \cdot \boldsymbol{e}_{\boldsymbol{k},\eta} g_{\mu,k}$.

We also suppose that the excitonic system is in a quasiequilibrium at the bottleneck region, and the one-exciton density operator is represented as

$$\hat{\rho}^{\rm one}_{\rm eq} = \sum_{\mu,\lambda}\frac{P_\mu}{N}|{\rm ex}_{\mu,\lambda}\rangle\langle{\rm ex}_{\mu,\lambda}| = \sum_{\mu,\boldsymbol{k}} P_\mu|\Phi_{\mu,\boldsymbol{k}}|^2\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}}|0\rangle\langle 0|\hat{\sigma}_{\mu,\boldsymbol{k}}. \tag{41}$$

Here, P_μ is the probability of being in the μ state. The exciton exists anywhere in the whole space with an equal probability.

The center of mass of exciton spreads coherently in space with the wave function $\Phi_\mu(\boldsymbol{r})$, and the exciton state located around $\boldsymbol{R}_\lambda$ is represented as

$$|\mathrm{ex}_{\mu,\lambda}\rangle = \sum_{\lambda'} \sqrt{V_0}\Phi_\mu(\boldsymbol{R}_{\lambda'} - \boldsymbol{R}_\lambda)\hat{\sigma}^\dagger_{\mu,\lambda'}|0\rangle \tag{42a}$$

$$= \sum_{\boldsymbol{k}} e^{-i\boldsymbol{k}\cdot\boldsymbol{R}_\lambda}\Phi_{\mu,\boldsymbol{k}}\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}}|0\rangle, \tag{42b}$$

where the Fourier transform of $\Phi_\mu(\boldsymbol{r})$ is defined as

$$\Phi_{\mu,\boldsymbol{k}} = \frac{1}{\sqrt{V}}\int d\boldsymbol{r}\, e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}\Phi_\mu(\boldsymbol{r}). \tag{43}$$

Equation (41) means that the exciton spreads in the k space not as the distribution of many excitons but as the one-exciton mixed state reflecting the finiteness of the coherence volume. From this one-exciton density operator, we get the following expectation values:

$$\langle\hat{\sigma}^\dagger_{\mu,\lambda}\hat{\sigma}_{\mu',\lambda'}\rangle^{\mathrm{one}}_{\mathrm{eq}} = \delta_{\mu,\mu'}\frac{P_\mu}{N}\int d\boldsymbol{r}\,\Phi_\mu(\boldsymbol{R}_\lambda - \boldsymbol{r})^*\Phi_\mu(\boldsymbol{R}_{\lambda'} - \boldsymbol{r}), \tag{44a}$$

$$\langle\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}}\hat{\sigma}_{\mu',\boldsymbol{k}'}\rangle^{\mathrm{one}}_{\mathrm{eq}} = \delta_{\mu,\mu'}\delta_{\boldsymbol{k},\boldsymbol{k}'}P_\mu|\Phi_{\mu,\boldsymbol{k}}|^2. \tag{44b}$$

In the following, we suppose that each exciton shows this spatial distribution in the quasiequilibrium state consisting of many excitons, and the one-body correlation in the quasiequilibrium is written as

$$\langle\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}}\hat{\sigma}_{\mu',\boldsymbol{k}'}\rangle_{\mathrm{eq}} = \delta_{\mu,\mu'}\delta_{\boldsymbol{k},\boldsymbol{k}'}N_{\mu,\boldsymbol{k}}, \tag{45}$$

where $N_{\mu,\boldsymbol{k}} \propto P_\mu|\Phi_{\mu,\boldsymbol{k}}|^2$ represents the expectation number of excitons in state μ and with wave vector $\boldsymbol{k}$.

Let us discuss the inelastic exciton-exciton scattering process as a perturbation to the quasiequilibrium state. In the Heisenberg picture, the equations of deviation operators $\delta\hat{a}_{\boldsymbol{k},\eta} = \hat{a}_{\boldsymbol{k},\eta} - \hat{a}^{\mathrm{eq}}_{\boldsymbol{k},\eta}$ and $\delta\hat{\sigma}_{\mu,\boldsymbol{k}} = \hat{\sigma}_{\mu,\boldsymbol{k}} - \hat{\sigma}^{\mathrm{eq}}_{\mu,\boldsymbol{k}}$ from the quasiequilibrium are obtained from Eqs. (40) as

$$i\frac{d}{dt}\delta\hat{a}_{\boldsymbol{k},\eta} \simeq v|\boldsymbol{k}|\delta\hat{a}_{\boldsymbol{k},\eta} + \sum_\mu ig_{\mu,\boldsymbol{k},\eta}\delta\hat{\sigma}_{\mu,\boldsymbol{k}}, \tag{46a}$$

$$i\frac{d}{dt}\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}} \simeq \Omega_{1s,k}\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}} - \sum_\eta ig_{1s,\xi,\boldsymbol{k},\eta}\delta\hat{a}_{\boldsymbol{k},\eta} + \sum_{\mu\neq 1s}\sum_{\xi',\xi''}\sum_{\boldsymbol{k}',\boldsymbol{q}}[\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{q}}\delta(\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}'}\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}})$$
$$+\mathcal{V}_{\mu,\xi'',\xi',\xi,\boldsymbol{q}}\delta(\hat{\sigma}^\dagger_{1s,\xi',\boldsymbol{k}'}\hat{\sigma}_{1s,\xi'',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{\mu,\boldsymbol{k}+\boldsymbol{q}})], \tag{46b}$$

$$i\frac{d}{dt}\delta\hat{\sigma}_{\mu,\boldsymbol{k}} \simeq \Omega_{\mu,k}\delta\hat{\sigma}_{\mu,\boldsymbol{k}} - \sum_\eta ig_{\mu,\boldsymbol{k},\eta}\delta\hat{a}_{\boldsymbol{k},\eta} + \sum_{\xi,\xi',\xi''}\sum_{\boldsymbol{k}',\boldsymbol{q}}\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{q}}\delta(\hat{\sigma}^\dagger_{1s,\xi,\boldsymbol{k}'}\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}). \tag{46c}$$

Here, we supposed simply that the quasiequilibrium state consists of only the lowest excitons ($1s$) and the $1s$ exciton state has only the degrees of freedom of polarization direction $\xi = \{x,y,z\}$ and of wave vector $\boldsymbol{k}$. In the above equations, we keep only the terms involving the inelastic exciton-exciton scattering process. The third term in Eq. (46b) represents the scattering from two $1s$ excitons to $\mu \neq 1s$ and $1s$ excitons, and the last term represents the inverse process. The last term in Eq. (46c) also represents the former process. When we consider that Eq. (46b) describes the development of scattered excitons converting to the photonlike polaritons, the last two terms in it represent the creation of the scattered excitons in the $(1s,\xi,\boldsymbol{k})$ state. In this case, the most important term is the third term, and the deviation operator in it is expanded up to the lowest order as

$$\delta(\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}'}\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}) \simeq \delta\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}'}\hat{\sigma}^{\mathrm{eq}}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}^{\mathrm{eq}}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}} + \hat{\sigma}^{\mathrm{eq}\dagger}_{\mu,\boldsymbol{k}'}\delta\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}^{\mathrm{eq}}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}} + \hat{\sigma}^{\mathrm{eq}\dagger}_{\mu,\boldsymbol{k}'}\hat{\sigma}^{\mathrm{eq}}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}\delta\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}. \tag{47}$$

These $(1s,\xi',\boldsymbol{k}'-\boldsymbol{q})$ and $(1s,\xi'',\boldsymbol{k}+\boldsymbol{q})$ states correspond to the two bottleneck excitons, and $(\mu,\boldsymbol{k}')$ is the higher exciton state. For the unperturbed time development of these three states, we can neglect the coupling with photons. Then, from Eqs. (46b) and (46c), the deviation operators on the right-hand side in Eq. (47) are expressed approximately as

$$\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t) \simeq -i\int_{t_0}^t dt'\, e^{-i\Omega_{1s,k}(t-t')}\sum_{\mu\neq 1s}\sum_{\xi',\xi''}\sum_{\boldsymbol{k}',\boldsymbol{q}}[\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{q}}\delta(\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}'}\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}})(t')$$
$$+\mathcal{V}_{\mu,\xi'',\xi',\xi,\boldsymbol{q}}\delta(\hat{\sigma}^\dagger_{1s,\xi',\boldsymbol{k}'}\hat{\sigma}_{1s,\xi'',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{\mu,\boldsymbol{k}+\boldsymbol{q}})(t')], \tag{48a}$$

$$\delta\hat{\sigma}^\dagger_{\mu,\boldsymbol{k}}(t) \simeq i\int_{t_0}^t dt'\, e^{i\Omega_{\mu,k}(t-t')}\sum_{\xi,\xi',\xi''}\sum_{\boldsymbol{k}',\boldsymbol{q}}\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{q}}\delta(\hat{\sigma}^\dagger_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}\hat{\sigma}^\dagger_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}\hat{\sigma}_{1s,\xi,\boldsymbol{k}'})(t'), \tag{48b}$$

where t_0 is the starting time of the inelastic scattering process. We substitute these into the third term of Eq. (46b) through the expansion (47), and we neglect the fourth term, i.e., the inverse process. Further, we keep only the terms proportional to $\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}$, which are the dominant terms because they involve the stimulated emission of polaritons or stimulated creation of excitons (this point will be discussed in the next subsection). Finally, linearizing the equation with respect to the deviation operator, we

get

$$i\frac{d}{dt}\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t) \simeq \Omega_{1s,k}\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t) - \sum_{\eta} i g_{1s,\xi,\boldsymbol{k},\eta}\delta\hat{a}_{\boldsymbol{k},\eta}(t) + \sum_{\mu\neq 1s}\sum_{\xi',\xi''}\sum_{\boldsymbol{k}',\boldsymbol{q}} i\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{q}}\int_{t_0}^{t} dt' \sum_{\zeta,\zeta'}\sum_{\boldsymbol{k}'',\boldsymbol{q}'}$$
$$\times\Bigg[e^{i\Omega_{\mu,\boldsymbol{k}'}(t-t')}\sum_{\zeta''}\mathcal{V}_{\mu,\zeta,\zeta',\zeta'',\boldsymbol{q}'}\langle\hat{\sigma}^{\dagger}_{1s,\zeta'',\boldsymbol{k}'+\boldsymbol{q}'}(t')\hat{\sigma}^{\dagger}_{1s,\zeta',\boldsymbol{k}''-\boldsymbol{q}'}(t')\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(t)\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(t)\rangle_{\rm eq}\delta\hat{\sigma}_{1s,\zeta,\boldsymbol{k}''}(t')$$
$$- e^{-i\Omega_{1s,\boldsymbol{k}'-\boldsymbol{q}}(t-t')}\mathcal{V}_{\mu,\zeta,\zeta',\xi',\boldsymbol{q}'}\langle\hat{\sigma}^{\dagger}_{\mu,\boldsymbol{k}'}(t)\hat{\sigma}^{\dagger}_{1s,\zeta',\boldsymbol{k}''}(t')\hat{\sigma}_{\mu,\boldsymbol{k}'-\boldsymbol{q}+\boldsymbol{q}'}(t')\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(t)\rangle_{\rm eq}\delta\hat{\sigma}_{1s,\zeta,\boldsymbol{k}''-\boldsymbol{q}'}(t')$$
$$-e^{-i\Omega_{1s,\boldsymbol{k}+\boldsymbol{q}}(t-t')}\mathcal{V}_{\mu,\zeta,\zeta',\xi'',\boldsymbol{q}'}\langle\hat{\sigma}^{\dagger}_{\mu,\boldsymbol{k}'}(t)\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(t)\hat{\sigma}^{\dagger}_{1s,\zeta',\boldsymbol{k}''}(t')\hat{\sigma}_{\mu,\boldsymbol{k}+\boldsymbol{q}+\boldsymbol{q}'}(t')\rangle_{\rm eq}\delta\hat{\sigma}_{1s,\zeta,\boldsymbol{k}''-\boldsymbol{q}'}(t')\Bigg]. \tag{49}$$

In principle, the two-body correlation functions in the quasiequilibrium must be calculated by considering the elastic exciton-exciton scattering process, interaction with phonons, radiative recombination of excitons, etc. However, here we approximate them simply by products of the one-body correlations given in Eq. (45) with introducing phenomenologically dephasing rates $\gamma^{\rm deph}_{\mu,\boldsymbol{k}}$ as

$$\langle\hat{\sigma}^{\dagger}_{1s,\zeta'',\boldsymbol{k}'+\boldsymbol{q}'}(t')\hat{\sigma}^{\dagger}_{1s,\zeta',\boldsymbol{k}''-\boldsymbol{q}'}(t')\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(t)\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(t)\rangle_{\rm eq} = \delta_{\boldsymbol{k}'',\boldsymbol{k}}\delta_{\boldsymbol{q}',-\boldsymbol{q}}\delta_{\zeta',\xi''}\delta_{\zeta'',\xi'}N_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(t')N_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(t')$$
$$\times e^{[-i(\Omega_{1s,\boldsymbol{k}+\boldsymbol{q}}+\Omega_{1s,\boldsymbol{k}'-\boldsymbol{q}})-(\gamma^{\rm deph}_{1s,\boldsymbol{k}+\boldsymbol{q}}+\gamma^{\rm deph}_{1s,\boldsymbol{k}'-\boldsymbol{q}})](t-t')}, \tag{50a}$$

$$\langle\hat{\sigma}^{\dagger}_{\mu,\boldsymbol{k}'}(t)\hat{\sigma}^{\dagger}_{1s,\zeta',\boldsymbol{k}''}(t')\hat{\sigma}_{\mu,\boldsymbol{k}'-\boldsymbol{q}+\boldsymbol{q}'}(t')\hat{\sigma}_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(t)\rangle_{\rm eq} = \delta_{\boldsymbol{k}'',\boldsymbol{k}+\boldsymbol{q}}\delta_{\boldsymbol{q}',\boldsymbol{q}}\delta_{\zeta',\xi''}N_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(t')N_{\mu,\boldsymbol{k}'}(t')$$
$$\times e^{[-i(\Omega_{1s,\boldsymbol{k}+\boldsymbol{q}}-\Omega_{\mu,\boldsymbol{k}'})-(\gamma^{\rm deph}_{1s,\boldsymbol{k}+\boldsymbol{q}}+\gamma^{\rm deph}_{\mu,\boldsymbol{k}'})](t-t')}, \tag{50b}$$

$$\langle\hat{\sigma}^{\dagger}_{\mu,\boldsymbol{k}'}(t)\hat{\sigma}_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(t)\hat{\sigma}^{\dagger}_{1s,\zeta',\boldsymbol{k}''}(t')\hat{\sigma}_{\mu,\boldsymbol{k}+\boldsymbol{q}+\boldsymbol{q}'}(t')\rangle_{\rm eq} = \delta_{\boldsymbol{k}'',\boldsymbol{k}'-\boldsymbol{q}}\delta_{\boldsymbol{q}',\boldsymbol{k}'-\boldsymbol{k}-\boldsymbol{q}}\delta_{\zeta',\xi'}[1+N_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(t')]N_{\mu,\boldsymbol{k}'}(t')$$
$$\times e^{[-i(\Omega_{1s,\boldsymbol{k}'-\boldsymbol{q}}-\Omega_{\mu,\boldsymbol{k}'})-(\gamma^{\rm deph}_{1s,\boldsymbol{k}'-\boldsymbol{q}}+\gamma^{\rm deph}_{\mu,\boldsymbol{k}'})](t-t')}. \tag{50c}$$

Here, the density $N_{\mu,\boldsymbol{k}}(t)$ of excitons is supposed to depend on time. Under this approximation, keeping only the dominant terms involving the stimulated process, Eq. (49) is rewritten as

$$i\frac{d}{dt}\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t) \simeq \Omega_{1s,k}\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t) - \sum_{\eta} i g_{1s,\xi,\boldsymbol{k},\eta}\delta\hat{a}_{\boldsymbol{k},\eta}(t) + \int_{t_0}^{t} dt' \int d\omega\, i S_{\xi,\boldsymbol{k}}(\omega,t,t')e^{-i\omega(t-t')}\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t'), \tag{51}$$

where the integral kernel is expressed as

$$S_{\xi,\boldsymbol{k}}(\omega,t,t') = \sum_{\mu\neq 1s}\sum_{\xi',\xi''}\sum_{\boldsymbol{k}',\boldsymbol{q}}\delta(\omega+\Omega_{\mu,\boldsymbol{k}'}-\Omega_{1s,\boldsymbol{k}+\boldsymbol{q}}-\Omega_{1s,\boldsymbol{k}'-\boldsymbol{q}})\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{q}}{}^2$$
$$\times\left[N_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(t')N_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(t')e^{-(\gamma^{\rm deph}_{1s,\boldsymbol{k}+\boldsymbol{q}}+\gamma^{\rm deph}_{1s,\boldsymbol{k}'-\boldsymbol{q}})(t-t')} - [1+2N_{1s,\xi',\boldsymbol{k}+\boldsymbol{q}}(t')]N_{\mu,\boldsymbol{k}'}(t')e^{-(\gamma^{\rm deph}_{1s,\boldsymbol{k}+\boldsymbol{q}}+\gamma^{\rm deph}_{\mu,\boldsymbol{k}'})(t-t')}\right]. \tag{52}$$

In this way, under the above approximations, the equation is reduced to the one-body one consisting of Eqs. (46a) and (51). What we have to solve is the master equation derived from these equations as

$$\frac{d}{dt}\hat{\rho}(t) = \frac{1}{i\hbar}[\hat{H}_{\rm pol},\hat{\rho}(t)] + \mathcal{L}_{\rm diss}[\hat{\rho}] + \sum_{\xi,\boldsymbol{k}}\int d\omega\int_{t_0}^{t} dt'\, S_{\xi,\boldsymbol{k}}(\omega,t,t')\{e^{i\omega(t-t')}[\hat{\sigma}^{\dagger}_{1s,\xi,\boldsymbol{k}}\hat{\rho}(t'),\hat{\sigma}_{1s,\xi,\boldsymbol{k}}] + e^{-i\omega(t-t')}[\hat{\sigma}^{\dagger}_{1s,\xi,\boldsymbol{k}},\hat{\rho}(t')\hat{\sigma}_{1s,\xi,\boldsymbol{k}}]\}. \tag{53}$$

The second term $\mathcal{L}_{\rm diss}[\hat{\rho}]$ is introduced for the dissipation of excitons and photons. The last two terms come from the last term in Eq. (51). They originate from the inelastic exciton-exciton scattering, and give a gain for the creation of excitons or polaritons. Due to the presence of these terms, the P emission shows a threshold behavior involving the stimulated emission of polaritons or stimulated creation of excitons. The decay of the exciton density $N_{\mu,\boldsymbol{k}}(t)$ appearing in Eq. (52) should be solved together with Eq. (53). This problem will be discussed in the next subsection. Once we obtain the temporal evolution of $N_{\mu,\boldsymbol{k}}(t)$, we can calculate the correlation of polaritons $\langle\hat{p}^{\dagger}_{j,\boldsymbol{k},\eta}(t')\hat{p}_{j,\boldsymbol{k},\eta}(t)\rangle$, which gives the P-emission spectra and the exciton-to-polariton conversion time.

Since the polariton states are the eigenstates of the unperturbed Hamiltonian $\hat{H}_{\rm pol}$, the deviation operators can be approximated as $\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t) \simeq \sum_{j,\eta} X_{j,\boldsymbol{k},\eta,1s,\xi}\delta\hat{p}_{j,\boldsymbol{k},\eta}e^{-i\omega_{j,k}t}$. Substituting this into the last term in Eq. (51), we get the quasiconservation of the energy as $\omega_{j,\boldsymbol{k}}+\Omega_{\mu,\boldsymbol{k}'} \sim \Omega_{1s,\boldsymbol{k}+\boldsymbol{q}}+\Omega_{1s,\boldsymbol{k}'-\boldsymbol{q}}$, if the dephasing rate $\gamma^{\rm deph}_{\mu,k}$ is low enough compared to the oscillation frequency $\omega_{j,\boldsymbol{k}}$. Only the lowest polariton ($j=L$) can satisfy this energy conservation, and its density should be finally enhanced compared with the other polariton

states. However, it is dangerous to approximate $\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t) \simeq X_{L,\boldsymbol{k},\eta,1s,\xi}\delta\hat{p}_{L,\boldsymbol{k},\eta}e^{-i\omega_{L,k}t}$ because the exciton-to-polariton conversion can be restricted by the coherence volume of the scattered excitons as discussed in the previous section. We should solve the master equation (53) in the photon-exciton basis or in the basis consisting of all the polariton states in principle. In the conventional interpretation, the energy conservation determines the wave vector of the scattered state (destination of the inelastic scattering). On the other hand, in our interpretation, the scattered excitons spread in the k space reflecting the coherence volume, and the exciton states are forced to oscillate with the frequency ω as seen in Eqs. (51) and (53). In both interpretations, the ω dependence of $S_{\xi,\boldsymbol{k}}(\omega,t,t')$ basically gives the P-emission spectra, and it is determined by the k distribution of the bottleneck excitons $N_{1s,\xi,\boldsymbol{k}}$ and of $\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{k}}$ through $S_{\xi,\boldsymbol{k}}(\omega,t,t')$ in Eq. (52).

Although the wave vector $\boldsymbol{k}$ seems to be a good quantum number in Eqs. (46a) and (51), all the $\boldsymbol{k}$ components are in fact connected in the master equation (53). Whereas we should in principle calculate $\rho(t)$ and $N_{\mu,\boldsymbol{k}}(t)$ self-consistently, we here focus on only the exciton-to-polariton conversion process. Such a situation can be considered by assuming $N_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(t)N_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(t) = \delta(t)N_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}}(0)N_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}}(0)$ in Eq. (52) and $t_0 < 0$. For simplicity, we also suppose $N_{\mu\neq 1s,\boldsymbol{k}} = 0$, i.e., the density of the higher excitons is negligible compared to that of the bottleneck excitons. Under these approximations, the master equation (53) is simplified as

$$\frac{d}{dt}\hat{\rho}(t) = \frac{1}{i\hbar}[\hat{H}_{\rm pol},\hat{\rho}(t)] + \int d\omega \sum_{\xi,\boldsymbol{k}}\{e^{-i\omega t}S_{\xi,\boldsymbol{k}}(\omega,t,0) \times [\hat{\sigma}^{\dagger}_{1s,\xi,\boldsymbol{k}},\hat{\rho}(0)\hat{\sigma}_{1s,\xi,\boldsymbol{k}}] + \text{H.c.}\}. \quad (54)$$

The last terms create excitons during the dephasing time $1/(\gamma^{\rm deph}_{1s,\boldsymbol{k}+\boldsymbol{q}} + \gamma^{\rm deph}_{1s,\boldsymbol{k}'-\boldsymbol{q}})$, which should be long enough than the oscillation period $2\pi/\omega$. After that, the created exciton is converted to a polariton as a one-body problem in the Hamiltonian $\hat{H}_{\rm pol}$, in which the frequency mixing (nonlinear process) does not occur. Then, the problem can be reduced to the exciton-to-polariton conversion as discussed in the previous section. The initial exciton state defined in Eq. (20) or (33) is determined by the last two terms in Eq. (54), and the center-of-mass motion of excitons is distributed for emission frequency ω as

$$|\Phi_{1s,\xi,\boldsymbol{k}}|^2 \propto S_{\xi,\boldsymbol{k}}(\omega,0,0). \quad (55)$$

In this way, the coherence volume $V_{\rm coh}$ of the scattered excitons is determined mainly by the k distributions of density $N_{1s,\xi,\boldsymbol{k}}(t)$ of bottleneck excitons and of scattering coefficient $\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{k}}$ through Eq. (52). In contrast to the interpretation of the direct creation of polaritons, the delta function in Eq. (52) (energy conservation) does not determine the wave vector of the scattered exciton, and its center-of-mass wave function $|\Phi_{1s,\xi,\boldsymbol{k}}|^2$ spreads in the region of $|\boldsymbol{k}| \lesssim 1/(V_{\rm coh})^{1/3}$.

In Fig. 3(a), we calculated the k distribution of $S_{\xi,\boldsymbol{k}}(\omega,0,0)$ in the following simple model, and the estimated coherence length is plotted in Fig. 3(b). The density of the bottleneck excitons is distributed as a Gaussian function with a coherence length $\lambda^0_{\rm coh}$as

$$N_{1s,\xi,\boldsymbol{k}} \propto e^{-(k\lambda^0_{\rm coh}/2)^2}. \quad (56)$$

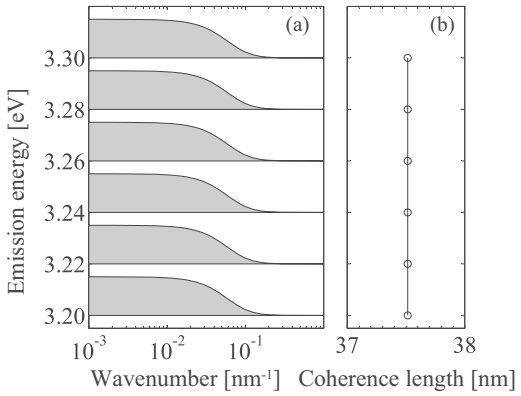

FIG. 3. (a) At each emission frequency, the k distribution of scattered exciton is plotted with gray color. It is calculated from Eq. (58) for ZnO with fitting parameters $\lambda_{\rm screen} = 14.4$ nm and $\lambda^0_{\rm coh} = 80$ nm. (b) The coherence length $\lambda_{\rm coh}$ of the scattered exciton is calculated from the width at half maximum of the distribution. At the present conditions, $\lambda_{\rm coh}$ does not strongly depend on the emission frequency.

The scattering coefficient does not depend on the exciton states and is represented as

$$\mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{q}} \propto \frac{1}{(q\lambda_{\rm screen})^2+1}. \quad (57)$$

Here, $\lambda_{\rm screen}$ is the screening length of the inelastic scattering, which is expected to be in the order of the exciton Bohr radius, which is $a^*_B = 1.8$ nm for ZnO [32]. The higher exciton states including the unbound ones are distributed continuously above the band-gap energy $E_g = \hbar\Omega_{1s,k=0} + 60$ meV [32]. The bound states below the band gap are not considered because the P emission is observed mainly for $\hbar\omega < \hbar\Omega_{1s,k=0} - 60$ meV in the experiments, i.e., the contribution of the continuous band is dominant. Then, we calculate the scattering coefficient as

$$S_{\xi,\boldsymbol{k}}(\omega,0,0) \propto \sum_{\boldsymbol{k}',\boldsymbol{q}}\int_{E_g/\hbar}^{\infty} d\omega' \frac{e^{-(|\boldsymbol{k}+\boldsymbol{q}|\lambda^0_{\rm coh}/2)^2}e^{-(|\boldsymbol{k}'-\boldsymbol{q}|\lambda^0_{\rm coh}/2)^2}}{[(q\lambda_{\rm screen})^2+1]^2} \times \delta(\omega+\omega'+\hbar|\boldsymbol{k}'|^2/2\mu - \Omega_{1s,\boldsymbol{k}+\boldsymbol{q}} - \Omega_{1s,\boldsymbol{k}'-\boldsymbol{q}}). \quad (58)$$

Here, the reduced mass $\mu = (1/m_e + 1/m_h)^{-1}$ describes the dispersion of the continuous band, while the frequency of the $1s$ exciton is $\Omega_{1s,k} = \Omega_{1s,k=0} + \hbar k^2/2M$ for the total mass $M = m_e + m_h$ ($m_e = 0.28m_0$ and $m_h = 0.59m_0$ in ZnO [32]).

The coherence length $\lambda_{\rm coh}$ of the scattered excitons plotted in Fig. 3(b) is determined from the width at half maximum of the distribution plotted in Fig. 3(a) as the Gaussian distribution in Eq. (56). The center-of-mass motion of the bottleneck exciton has a finite coherence length $\lambda^0_{\rm coh}$ and it is supposed to be longer than the screening length $\lambda_{\rm screen} \sim a^*_B$. Under this condition, the coherence length $\lambda_{\rm coh}$ of the scattered excitons is basically determined by the screening length $\lambda_{\rm screen}$, and $\lambda_{\rm coh}$ is obtained generally shorter than $\lambda^0_{\rm coh}$ of the bottleneck excitons. Due to the reabsorption problem, it is hard to estimate $\lambda^0_{\rm coh}$ from the free-exciton lifetime obtained in experiment. Here, by supposing the values of $\lambda^0_{\rm coh}$ and $\lambda_{\rm screen}$, which are listed below, we try to reproduce the coherence length of the scattered

excitons $\lambda_{\rm coh} \sim 4 \times 10^1$ nm, which was estimated in Sec. III B from the P-emission lifetime obtained experimentally. In the calculation of Fig. 3, we supposed $\lambda_{\rm screen} = 8 \times a_B^* = 14.4$ nm as a main fitting parameter for obtaining $\lambda_{\rm coh} \sim 40$ nm, and $\lambda_{\rm coh}^0 = 80$ nm is chosen simply as twice this value (e.g., we get $\lambda_{\rm coh} \sim 20$ nm for $\lambda_{\rm screen} = 4 \times a_B^*$ and $\lambda_{\rm coh}^0 = 80$ nm). Under the present model and analysis, we can only say that the coherence length of the bottleneck excitons $\lambda_{\rm coh}^0$ should be longer than that of the scattered excitons $\lambda_{\rm coh}$ estimated from the P-emission lifetime. Whereas such a long coherence length is expected for the bottleneck exciton in our calculation, its lifetime is elongated by the very small photonic fraction and also by the dephasing process of the reabsorbed photons. Note that, as seen in Fig. 3(b), the coherence length $\lambda_{\rm coh}$ of the scattered excitons does not strongly depend on the emission frequency at least under the present conditions. Then, the ω dependence of the P-emission lifetime at each emission frequency is basically determined by the photonic fraction of the polariton state.

In general, the coherence volume is determined through the last term in Eq. (51) or the last two terms in Eq. (53) with self-consistently calculating $\hat{\rho}(t)$ and $N_{\mu,\boldsymbol{k}}(t)$. Reflecting the coherence volume, the density of scattered excitons $\langle \hat{\sigma}_{1s,\xi,\boldsymbol{k}}^\dagger \hat{\sigma}_{1s,\xi,\boldsymbol{k}} \rangle$ initially spreads for $|\boldsymbol{k}| \lesssim 1/(V_{\rm coh})^{1/3}$. After a long enough time compared to the P-emission lifetime, the ω-Fourier component of $\langle \hat{p}_{j,\boldsymbol{k},\eta}^\dagger(t) \hat{p}_{j,\boldsymbol{k},\eta}(t') \rangle$ should be distributed only around $k_L(\omega)$ reflecting the large coherence volume of the propagating polaritons. Although we do not solve the master equation (53) in this paper, the exciton-to-polariton conversion time given by such a calculation should be equivalent to the one calculated in the previous section if the coherence length just after the scattering is shorter than the radiation wavelength.

At least theoretically, we can suppose any coherence volume and emission frequency in the above calculation. However, even by calculating the exciton-to-polariton conversion time around the bottleneck region, it is probably far from the spontaneous emission lifetime observed in experiments. The deviation basically originates from the two factors. (1) We must also consider the memory loss of the phase and propagation direction of the reabsorbed photons by considering the elastic exciton-exciton scattering, interaction with phonons, etc. (2) The quasiequilibrium at the bottleneck region must be discussed under considering the radiative recombination of exciton, reabsorption of the photon, and the effect (1). These problems are also remaining tasks in the future.

B. Stimulated emission of polaritons or stimulated creation of excitons

The P emission exhibits a threshold behavior with respect to the pumping power and shows also an optical gain at that frequency [11–16]. Then, the inelastic scattering has been considered as a stimulated emission [1,11–16] [or called the amplified spontaneous emission (ASE)] and lasing is also reported [12]. In contrast, instead of the stimulated emission of photons or polaritons, in this paper we interpret that the creation of excitons is stimulated by the accumulated excitons with the P-emission energy (stimulated scattering of excitons), and then those excitons are emitted in the conversion time $\tau_{\rm conv}$.

When we suppose that the polaritons are directly created by the inelastic scattering, the deviation operator in Eq. (51) is approximated as $\delta\hat{\sigma}_{1s,\xi,\boldsymbol{k}}(t') \simeq e^{i\omega_{L,k}(t-t')} \sum_\eta X_{L,\boldsymbol{k},\eta,1s,\xi}\, \delta\hat{p}_{L,\boldsymbol{k},\eta}(t)$. Then, the equation of motion of the number of polaritons $\delta N_{L,\boldsymbol{k},\eta} = \langle \delta\hat{p}_{L,\boldsymbol{k},\eta}^\dagger \delta\hat{p}_{L,\boldsymbol{k},\eta} \rangle$ is obtained as

$$\frac{d}{dt}\delta N_{L,\boldsymbol{k},\eta}(t) = -\gamma_{\rm escape}(\omega_{L,k})\delta N_{L,\boldsymbol{k},\eta}(t) + G_{\boldsymbol{k},\eta}\delta N_{L,\boldsymbol{k},\eta}(t). \tag{59}$$

Here, the loss of the polaritons with the escape rate $\gamma_{\rm escape}(\omega)$ [Eq. (28)] is introduced and the gain $G_{\boldsymbol{k},\eta}$ is represented as

$$G_{\boldsymbol{k},\eta} = \sum_{\mu\neq 1s} \sum_{\xi,\xi',\xi''} \sum_{\boldsymbol{k}',\boldsymbol{q}} \frac{4\gamma_{\rm deph} X_{L,\boldsymbol{k},\eta,1s,\xi}{}^2 \mathcal{V}_{\mu,\xi,\xi',\xi'',\boldsymbol{q}}{}^2}{(\delta\omega)^2 + (2\gamma_{\rm deph})^2} \times [N_{1s,\xi',\boldsymbol{k}'-\boldsymbol{q}} N_{1s,\xi'',\boldsymbol{k}+\boldsymbol{q}} - (1 + 2N_{1s,\xi',\boldsymbol{k}+\boldsymbol{q}}) N_{\mu,\boldsymbol{k}'}]. \tag{60}$$

Here, we simply supposed $\gamma_{\rm deph} = \gamma_{\mu,\boldsymbol{k}}^{\rm deph}$. The frequency difference $\delta\omega = \omega_{L,k} + \Omega_{\mu,\boldsymbol{k}'} - \Omega_{1s,\boldsymbol{k}+\boldsymbol{q}} - \Omega_{1s,\boldsymbol{k}'-\boldsymbol{q}}$ gives a resonance at particular $\boldsymbol{k}$ through the denominator $(\delta\omega)^2 + (2\gamma_{\rm deph})^2$. Equation (59) corresponds to the rate equation discussed in Sec. 22.1 of Ref. [1]. When the gain becomes larger than the loss, the stimulated emission of polaritons occurs, and it determines the threshold of the P emission in the conventional interpretation.

On the other hand, in our interpretation, we suppose that the excitons are created by the inelastic scattering. We define the density of scattered excitons $\delta N_{\boldsymbol{k},\xi}(\omega,t)$ converting to polaritons with a emission frequency ω as

$$\langle \delta\hat{\sigma}_{\boldsymbol{k},\xi}^\dagger(t') \delta\hat{\sigma}_{\boldsymbol{k},\xi}(t) \rangle = \int d\omega\, \delta N_{\boldsymbol{k},\xi}[\omega,(t+t')/2] e^{-i\omega(t-t')}, \tag{61}$$

$$\delta N_{\boldsymbol{k},\xi}(\omega,t) = \int d\tau \langle \delta\hat{\sigma}_{\boldsymbol{k},\xi}^\dagger(t-\tau/2) \delta\hat{\sigma}_{\boldsymbol{k},\xi}(t+\tau/2) \rangle e^{i\omega\tau}. \tag{62}$$

The rate equation is derived from Eq. (51) as

$$\frac{d}{dt}\delta N_{\xi,\boldsymbol{k}}(\omega,t) = -\Gamma(\omega)\delta N_{\xi,\boldsymbol{k}}(\omega,t) + 2\pi S_{\xi,\boldsymbol{k}}(\omega,t,t)\delta N_{\xi,\boldsymbol{k}}(\omega,t). \tag{63}$$

For deriving this equation, instead of considering explicitly the light-matter coupling, the exciton-to-polariton conversion rate $\Gamma(\omega)$ [Eq. (34)] is introduced as the loss of the scattered excitons. For deriving the second (gain) term, we supposed that the dephasing rate is low enough than the oscillation frequency as $\gamma_{\rm deph} \ll \omega$, and the density $N_{\xi,\boldsymbol{k}}(\omega,t)$ is varying slowly with respect to t compared to the dephasing time $1/\gamma_{\rm deph}$. Also in our interpretation, Eq. (63) shows a threshold behavior when the gain $2\pi S_{\xi,\boldsymbol{k}}(\omega,t,t)$ exceeds the loss $\Gamma(\omega)$, and a stimulated creation (scattering) of excitons occurs.

The stimulated emission of polaritons and the stimulated creation of excitons are different processes with different thresholds. In order to discuss theoretically which interpretation of the P emission is appropriate, we should solve the master equation (53) without the assumptions of the direct creation of polaritons or excitons by the inelastic scattering. We do not perform such a calculation in this paper. We

instead justified our interpretation from the experimental results [21,23,24] and from the discussion of the spontaneous emission from the bottleneck excitons, in which the coherence volume plays an important role. If the stimulated emission of photons or polaritons occurs and the polaritons do not propagate diffusively, we should observe the escape time τ_{escape} from the sample as the P-emission lifetime. However, the experimental data [21,23,24] show the lifetimes one or two orders of magnitude slower than τ_{escape}. Further, since the excitons at the bottleneck are incoherent (having a coherence length shorter than the radiation wavelength), the scattered excitons are also supposed to have a poor coherence length. They are the reasons why we conclude that the stimulated creation of excitons occurs, and those excitons are converted to polaritons with the rate $\Gamma(\omega)$, which is proportional to the group velocity $v_{\mathrm{g}}(\omega)$ in the experiments and also in our calculation approximately.

In order to distinguish clearly the two stimulated processes experimentally, we should perform a time-resolved measurement of the optical gain [11–16]. We obtain the stimulated emission of polaritons after the probe pulse arrives at the sample because the probe beam propagates as a polariton with a long enough spatial coherence.[2] In our interpretation, the stimulated creation of excitons occurs around the rise time of the P emission (shortened inversely proportional to the square of the pump power [17]), and the conversion from exciton to polariton occurs after that. Then, the probe beam should get the optical gain only in a time delay around the P-emission rise time plus the onset time. If the stimulated emission of photons or polariton occurs and the created polaritons escape from the sample very quickly in a time of τ_{escape}, the optical gain is obtained even in the decay period of the P emission because the P-emission lifetime corresponds to that of the bottleneck excitons. Even if the polaritons propagate diffusively and the stimulated emission of polaritons occurs only in the rise period, the decay time of the optical-gain signal should reflect the contribution of the relatively slow escape time of the diffusive polaritons. In contrast, in our interpretation, the decay time of the optical-gain signal should be shorter than the P-emission lifetime because spatial coherence is established by the probe beam and the exciton-to-polariton conversion is not restricted by the coherence volume.

[2]When the lasing occurs without the probe pulse and the radiation field gets a nonzero amplitude with temporal and spatial coherences, the inelastic scattering provides the stimulated emission of photons.

V. SUMMARY

In the conventional interpretation of the P emission, excitons at the bottleneck region are supposed to be scattered directly to photonlike polariton states. We instead propose another interpretation. The excitons are scattered to bare exciton states first, and then they are converted to polaritons in a finite conversion time, which corresponds to the P-emission lifetime observed in the recent experiments using the optical Kerr gating method [21,23,24]. We justify our interpretation by supposing that the scattered excitons should have a finite coherence volume and they are converted to polaritons as the emission process from localized exciton. Since the polariton states require a long enough spatial coherence for their establishment, they cannot be a direct destination of the inelastic scattering because of the small coherence volume of the excitons. In the calculation of the inelastic exciton-exciton scattering, the coherence volume of the scattered excitons certainly appears on the assumption that the bottleneck excitons originally have a finite coherence volume. However, more detailed experimental and theoretical investigations are required to finally conclude which interpretation is reasonable. Especially, a time-resolved optical-gain measurement would give us fruitful information for distinguishing our interpretation, the conventional one, and that of the polariton diffusion.

ACKNOWLEDGMENTS

M.B. thanks B. Deveaud for fruitful discussion. This work was supported by JSPS KAKENHI (Grants No. 24560011, No. 26287087, and No. 24-632).

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