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

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(Dated: November 14, 2014)

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 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 without violating the quantum physics.

PACS numbers: 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. 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-exciton 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.

Concerning the spontaneous emission of excitations at quasi-thermal equilibrium (thermal 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. The coherence volume also gives the limit of exciton superradiance by which the emission lifetime is shortened with an increase in interaction volume between the radiation field and center-of-mass wavefunction of excitons (radius of quantum dot). However, the coherence volume of excitations has not yet been well investigated, because it is usually estimated only through the emissionlifetime 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.

Although the emission frequency is almost fixed for the spontaneous emission of excitons at the quasi-thermal equilibrium (called the bottleneck region), 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). In the P-emission process, one exciton is inelastically scattered to a higher exciton state and the other one is scattered to the photon-like 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. The relaxation and scattering processes toward the P emission has been investigated in time-resolved measurements performed using optical Kerr gating method and by streak camera, and then the following facts have been revealed. 1) The onset time reflects the time of energy relaxation of excitons toward the bottleneck region on the lower exciton-polariton branch. 2) The rise time reflects the rate of the inelastic scattering of excitons. 3) The temporal change of the peak energy reflects the change of effective temper-
than the emission lifetime on materials of samples, they are generally much shorter than the P-emission decay times observed in experiments.\textsuperscript{12,16,18} On the other hand, in Ref. 13, the authors analyzed the P-emission decay as diffusive propagation of the photon-like 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 decay time: Just after the inelastic exciton-exciton scattering, the photon-like 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 photon-like polariton, which is restricted by the coherence volume, is observed as the P-emission decay time. Although the P-emission has been considered as a stimulated emission of polaritons\textsuperscript{16,18} we need to reconsider it as a stimulated creation (scattering) of excitons in our interpretation.

In Sec. II, we first estimate the emission lifetime of excitons, interchange time between exciton and photon in the polariton states, and escape time of the polaritons. Only the emission lifetime 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.

\section{CHARACTERISTIC TIME SCALES OF EXCITONS AND POLARITONS}

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

$$\hat{H}_{rad} = \sum_{\eta=1,2} \sum_k \hbar \nu |\mathbf{k}| \hat{a}^\dagger_{k,\eta} \hat{a}_{k,\eta},$$

where $\hat{a}_{k,\eta}$ is the annihilation operator of a photon with wavevector $\mathbf{k}$ and polarization $\eta$, and $\nu = c/\sqrt{\varepsilon_{bg}}$ 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

$$\hat{H}_{LM} = -\frac{1}{\varepsilon_0 \varepsilon_{bg}} \int \, dr \, \mathbf{P}(r) \cdot \mathbf{D}_\perp(r).$$
Here, the transverse component of the electric displacement field is defined

\[ \mathbf{D}_\perp (r) = \sum_{\eta=1,2} \sum_k \mathbf{e}_{k,\eta} i \sqrt{\frac{\hbar \varepsilon_{bg} e |k|}{2V}} (\hat{a}_{k,\eta} - \hat{a}^\dagger_{-k,\eta}) e^{i k \cdot r}, \]  

where \( \mathbf{e}_{k,\eta} \) is the unit vector perpendicular to \( k \), and \( V \) is the volume of the space. \( \mathbf{P} (r) \) represents the polarization density involving the creation and annihilation of excitons. Since optical inter-band transitions occur almost inside a unit cell, the inter-band transition dipole moment \( d_{cv} \) is modified by the wavefunction \( \psi_\mu (r = 0) \) for state \( \mu \) of the electron-hole relative motion at zero distance \( r = 0 \). Thus, the polarization density involving the light-matter coupling is expressed under the long-wavelength approximation as

\[ \mathbf{P} (r) = d_{cv} \sum_\mu \mathbf{e}_\mu \psi_\mu (0) \sum_\lambda \delta (r - \mathbf{R}_\lambda) \left( \hat{\sigma}_{\mu,\lambda} \right), \]  

where \( \hat{\sigma}_{\mu,\lambda} \) annihilates an exciton in state \( \mu \) with a center-of-mass placing at position \( \mathbf{R}_\lambda \). \( \mathbf{e}_\mu \) is the unit vector of the polarization direction of state \( \mu \). Then, Eq. (2) is rewritten as

\[ \hat{H}_{LM} = - \sum_\mu \sum_{\eta=1,2} \sum_k \mathbf{e}_\mu \cdot \mathbf{e}_{k,\eta} \sqrt{\frac{\hbar |k| d_{cv}^2 \psi_\mu (0)^2}{2 \varepsilon_{bg} V}} \]
\[ \times \sum_\lambda \left( \hat{\sigma}_{\mu,\lambda} \left( \hat{a}_{k,\eta} - \hat{a}^\dagger_{-k,\eta} \right) e^{i k \cdot R_\lambda} \right). \]  

For the nanometer-scale orbital with Bohr radius \( a_B^s \), the wavefunction of the electron-hole relative motion is expressed as

\[ \psi_{ns} (0) = \frac{V_0}{\pi a_B^s 3 n^3}, \]  

where \( V_0 \) is the volume of a unit cell.

When we define the exciton operator in the \( k \)-representation for number \( N \) of unit cells as

\[ \hat{\sigma}_{\mu,k} = \frac{1}{\sqrt{N}} \sum_\lambda e^{-i k \cdot R_\lambda} \hat{\sigma}_{\mu,\lambda}, \]  

the Hamiltonian of the excitons is represented as

\[ \hat{H}_{EX} = \sum_\mu \sum_k \hbar \Omega_{\mu,k} \hat{\sigma}_{\mu,k} \hat{\sigma}_{\mu,k}^\dagger + \frac{1}{2 \varepsilon_{bg}} \int \mathbf{d}r \mathbf{P} (r) \cdot \mathbf{P} (r). \]  

Here, \( \Omega_{\mu,k} \) is the eigenfrequency of exciton in state \( \mu \) with wavenumber \( k \). The last term is the so-called \( P^2 \) term and represents the depolarization shift.\textsuperscript{23, 24, 25} The light-matter coupling Hamiltonian given by Eq. (3) is rewritten as

\[ \hat{H}_{LM} = - \sum_\mu \sum_{\eta=1,2} \sum_k \mathbf{e}_\mu \cdot \mathbf{e}_{k,\eta} \]
\[ \times i \hbar g_{\mu,k} \left( \hat{\sigma}_{\mu,-k} + \hat{\sigma}_{\mu,k}^\dagger \right) \left( \hat{a}_{k,\eta} - \hat{a}^\dagger_{-k,\eta} \right), \]  

where the coupling strength is defined as

\[ g_{\mu,k} = \sqrt{\frac{v k d_{cv}^2 \psi_\mu (0)^2}{2 \hbar \varepsilon_{bg} V_0}}. \]  

The eigenstates of the electromagnetic fields in this excitonic medium are the polariton states satisfying the dispersion relation (roughly sketched in Fig. 11) as

\[ \frac{\Omega^2 k^2}{\omega^2} = \varepsilon_{bg} + \sum_\mu \frac{4 \pi \beta_{\mu,k} \Omega_{\mu,k}^2}{\Omega_{\mu,k} v k} = \varepsilon (\omega, k), \]  

where the non-dimensional factor is defined as

\[ 4 \pi \beta_{\mu,k} = \frac{4 \varepsilon_{bg} g_{\mu,k}^2}{\Omega_{\mu,k} v k} = \frac{2 d_{cv}^2 \psi_\mu (0)^2}{\varepsilon_0 h \Omega_{\mu,k} v k}. \]  

When polaritons stably exist in a large enough medium with negligible dissipation, the interchange rate between exciton state \( \mu \) and photon one is estimated from Eq. (13) for \( k = \Omega_{\mu} / v \) as

\[ g_\mu = \sqrt{\frac{\Omega_{\mu,k} d_{cv}^2 \psi_\mu (0)^2}{2 \hbar \varepsilon_{bg} V_0}} = \sqrt{\frac{\pi \beta_{\mu,k} \Omega_{\mu,k}^2}{\varepsilon_{bg}}}. \]  

For A excitons with \( n = 1 \) (1s) in ZnO\textsuperscript{22}, we have \( \Omega_{A,1s} = 3.375 \) eV, \( \varepsilon_{bg} = 4 \), and \( \Delta A_{1s} = 4 \pi \beta_{A,1s} \Omega_{A,1s} / \varepsilon_{bg} = 5.74 \) meV (\( \Omega_{B,1s} = 3.381 \) eV and \( \Delta B_{1s} = 6.62 \) meV for B exciton). The interchange rate is then estimated as \( h g_{A,1s} = 70 \) meV. The interchange time \( \tau_{\text{Habi}} = 2 \pi / g_\mu = 0.06 \) ps is one or two orders of magnitude shorter than the P-emission decay times observed in the experiments.

Let us next calculate the emission rate of exciton from the light-matter coupling Hamiltonian \( \hat{H}_{LM} \). Here, we suppose an exciton in state \( \mu \) as an initial state and its center-of-mass is localized at \( \mathbf{R}_\lambda \). According to the Fermi’s golden rule, the transition rate from the exciton state to one photon state with any \( k \) and \( \eta \) is obtained as

\[ \gamma_{\mu} = \frac{2 \pi}{\hbar} \sum_{\eta,k} |\langle 0 | \hat{a}^\dagger_{k,\eta} \hat{H}_{LM} \hat{a}_{k,\eta} | 0 \rangle|^2 \delta (\hbar \Omega_{\mu} - \hbar \omega) |k| \]
\[ = \frac{\Omega_{\mu}^3 d_{cv}^2 \psi_\mu (0)^2}{3 \pi \hbar \varepsilon_{bg} V_0} = \frac{2 g_{\mu}^2 \Omega_{\mu}^2 V_0}{3 \pi v^3}. \]  

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

\[ \sum_{\eta=1,2} \int |\mathbf{e}_\mu \cdot \mathbf{e}_{k,\eta}|^2 F(|k|) = \int_0^\infty dk \frac{8 \pi k^2}{3} F(k). \]  

For ZnO, the lattice constants are \( a = 3.25 \)\textsuperscript{Å} and \( c = 5.21 \)\textsuperscript{Å} and then the volume of the unit cell is

\[ V_0 = \frac{\sqrt{3}}{2} \times (3.25 \text{Å})^2 \times 5.21 \text{Å} = 24 \text{Å}^3. \]
Therefore, the emission rate \( \Gamma_\mu \) is estimated for the A excitons as
\[
\gamma_{A,1s} = 0.45(\mu s)^{-1}.
\] (17)

This rate is quite slow even compared to the spontaneous emission rate \( 1/\tau_{\text{emiss}} \) 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 \( V_{\text{coh}} \). Obeying the picture of the exciton superradiance,\textsuperscript{3,29} the emission rate of excitons in state \( \mu \) is enhanced as
\[
\Gamma_\mu = \gamma_\mu V_{\text{coh}}^\mu V_0
\] (18)

When the coherence length \((V_{\text{coh}})^{1/3}\) is comparable to or larger than the wavelength of the radiation, we have to consider the crossover to the polariton picture.\textsuperscript{29}

On the other hand, the interchange time \( \tau_{\text{coh}} \) \textsuperscript{[also the dispersion relation (11)]} 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 decay time of the P emission in the next section.

We next consider another time scale, the escape time of polaritons. We suppose a film of the excitonic medium as a polariton, if dissipations and dephasing are weak enough compared to the light-matter coupling. However, if the coherence length is quite short, it restricts the conversion time \( \tau_{\text{conv}} \), and our interpretation is rather appropriate.

FIG. 2. Schematic diagrams of (a) conventional interpretation \[(a)\] and (b) our interpretation of the dynamics toward the P emission. The escape time \( \tau_{\text{escape}} \) of polariton is estimated to be quite short compared to the observed decay time of the P emission. We interpret that the decay time reflects the conversion time \( \tau_{\text{conv}} \) 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 \( \tau_{\text{conv}} \), and our interpretation is rather appropriate.

\[ \begin{align*}
\gamma_{\text{escape}}(\omega) & = \frac{v_g(\omega)}{2L} \ln \frac{1}{|r_1(\omega) r_2(\omega)|^2} = \frac{v_g(\omega)}{L_{\text{eff}}(\omega)}, \quad (22)
\end{align*} \]
where
\[ L_{\text{eff}}(\omega) = \frac{2L}{\ln |r_1(\omega) r_2(\omega)|^2} \] (23)
is the effective length for the polariton propagation. This escape rate \( \tau_{\text{escape}} = 1/\gamma_{\text{escape}}(\omega) \) reflecting the macroscopic propagation of polaritons is another time scale in the processes of the P emission. When the effective thickness is around \( L_{\text{eff}} \sim 5 \mu m \), the escape time is estimated as \( \tau_{\text{escape}} \sim 0.1 \) ps for the P-emission frequency region in ZnO.\textsuperscript{16,18,19} This is also quite short compared to the observed P-emission decay time.
III. INTERPRETATION OF P-EMISSION DECAY

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 photon-like polariton almost immediately, because there are only the photon-like 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 $\tau_{\text{Rabi}} = 2\pi/g_{\mu}$ of the polariton state, and it is certainly negligible ($\tau_{\text{Rabi}} \sim 0.06$ ps in ZnO) compared to the other time scales except the escape time $\tau_{\text{escape}}$ of polariton (then there is a crossover around the material size comparable to the radiation wavelength). Then, if the P-emission decay times do not originate from the lifetime of excitons at the bottleneck, obeying the conventional interpretation, we need the interpretations of the polariton diffusion or of the polariton escape from a sample with an incredibly large effective thickness.

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 $\tau_{\text{Rabi}}$ 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 $V_{\text{coh}}$ of the scattered excitons.

Obeying this scenario, in order to estimate the conversion time (emission lifetime) from exciton to polariton, we need to extend the discussion of the emission rate in Sec. 11. Since the excitons collapse by feeling only the photonic component of the polariton state (destination) at frequency $\omega$, the conversion rate is modulated by the photonic fraction $A(\omega)$ of the polariton state. In other words, we need to consider the reabsorption process after the emission of photons. For photon-like polariton states in bulk material, the photonic fraction can be estimated by the group velocity $v_{g}(\omega)$ and the speed $v$ of light in the background medium as

$$A(\omega) = \frac{v_{g}(\omega)}{v},$$

and the excitonic fraction is given by $B(\omega) = 1 - A(\omega)$. The conversion rate from exciton to polariton at emission frequency $\omega$ is then estimated as

$$\Gamma(\omega) = A(\omega) \sum_{\mu} \frac{V_{\text{coh}}^{\mu}}{V_{0}} f_{\mu} \gamma_{\mu},$$

where $f_{\mu}$ is the fraction of excitons in state $\mu$ of the electron-hole relative motion after the inelastic scattering ($\sum_{\mu} f_{\mu} = 1$).

Then, in our interpretation, the P-emission decay times are governed by the conversion time $\tau_{\text{conv}} = 1/\Gamma(\omega)$ and the escape time $\tau_{\text{escape}}$ of polaritons after the conversion. Note that both of the two times are inversely proportional to the group velocity $v_{g}(\omega)$ as observed in the experiments for bulk ZnO. Since $\tau_{\text{escape}}$ is estimated to be quite short ($\sim 0.1$ ps), the P-emission decay times basically reflect the conversion time $\tau_{\text{conv}}$ from the scattered excitons to the polaritons in our interpretation.

From the experimental data for ZnO, the emission-frequency-independent decay rate is estimated as

$$\Gamma'' = \sum_{\mu} \frac{V_{\text{coh}}^{\mu}}{V_{0}} f_{\mu} \gamma_{\mu} = \frac{1}{0.8 \text{ ps}},$$

Since the A exciton is the lowest exciton state, here we tentatively consider that the A excitons are mostly created at the P-emission frequency region by the inelastic scattering, i.e., $f_{A,1s} = 1$. Then, from the emission rate derived for an exciton localized at a unit cell, the coherence volume is estimated as

$$V_{\text{coh}} = 6 \times 10^{7}(\text{Å})^{3},$$

and the coherence length is $(V_{\text{coh}})^{1/3} = 40$ nm. Although we have currently no other way to evaluate the coherence volume (length) experimentally, this value is certainly shorter than the radiation wavelength ($\sim 200$ nm for $h\omega = 3.26$ eV in the background medium with $\varepsilon_{bg} = 4$).

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 photon-like polariton can explain the observed P-emission decay time, which is much shorter than the emission lifetime $\tau_{\text{emit}}$ of the bottleneck excitons, longer than the escape time $\tau_{\text{escape}}$ of polaritons, and inversely proportional to the group velocity for bulk materials.

In the next section, we try to justify our interpretation against some counter-intuitive points.

IV. DISCUSSION

Since the final states certainly exist as the polariton states or photon states outside the sample, the in-
elastic scattering to these destinations is not forbidden. However, the scattered excitons remain in the bare exciton states in the conversion time $\tau_{\text{conv}} \sim 1$ ps, although the eigenfrequencies $\Omega_n$ of these states are far above the emission frequency $\omega$ ($h\Omega_{A,1s} = 3.375$ eV and $h\Omega_{A,1s} - \hbar\omega \sim 0.1$ eV for ZnO$^{23-29}$). Although the inelastic scattering of the two excitons is resonant to both the higher exciton state with $n > 1$ and the photon-like polariton one in the conventional interpretation, it is resonant only to the higher exciton state but not to the lower one (no exciton state at the P-emission frequency) in our scenario. However, even if one process is not resonant in a series of processes, it can occur in the quantum physics. Since the A and B exciton states with $n = 1$ are most resonant compared to the other exciton states ($\Omega_{A,1s}$ and $\Omega_{B,1s}$ are closest to $\omega$), the scattered excitons are supposed to be mostly in the lowest two exciton states ($f_{A,1s} + f_{B,1s} \sim 1$ and $f_{A,1s} > f_{B,1s}$)$^{30}$.

In our scenario, 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$^{33}$ such as $2\pi/(\Omega_n - \omega) \sim 0.04$ ps $< \tau_{\text{conv}}$ in our case. Instead, the problem should be interpreted as that of the forced oscillation in a composite system as explained below without violating the quantum physics. We interpret that the bottleneck excitons are scattered to unstable transient states (bare exciton states) with a lifetime of $\tau_{\text{conv}}$. If the dephasing time of the higher excitons ($n > 1$) is shorter than $\tau_{\text{conv}}$, the emission frequency $\omega$ is fixed during the excitons remain in the transient states. Such transient states are surely unstable, and then $\tau_{\text{conv}}$ is much shorter than the emission lifetime $\tau_{\text{emit}}$ of excitons at the bottleneck region. The conversion time $\tau_{\text{conv}}$ becomes shorter (less stable) with a decrease in the emission frequency (more distant from the bottleneck frequency). If we can generate excitons (with finite coherence volume) and tune their energy $\hbar\omega$ freely, we can observe the $\omega$-dependence of their emission lifetime. These lifetimes should correspond to the P-emission decay time observed in the experiment, and it is probably connected continuously to the spontaneous emission lifetime $\tau_{\text{emit}}$ of excitons at the bottleneck.

In contrast to the spontaneous emission of the bottleneck excitons, we cannot suppose the scattered excitons (with energy $\hbar\omega$ far below its resonance $h\Omega_\mu$) as an initial state in the emission process. We instead need to describe it as a transient state in the series of processes. For example, we can suppose an equation of motion of exciton (polarization) amplitude $\psi_k$ such as

$$\frac{d}{dt}\psi_k(t) = -i\Omega_k \psi_k(t) + g_k a_k(t) + f_k(t)e^{i\omega t}.$$  \hspace{1cm} (28)

Here, $a_k$ is the amplitude of photon, and its motion is determined by the Maxwell equations. The last term is the force with a frequency $\omega$ and a slowly-varying amplitude $f_k(t)$, which originates from the inelastic scattering. The coherence volume is described by the spatial distribution of this force $\{f_k\}$, and it restricts the conversion from the exciton state to the polariton one, while the conversion time is simply $2\pi/\gamma_k$ for a long enough coherence length compared to the radiation wavelength. Since the polariton states are the eigenstates in the system, the exciton state can be described by a superposition of upper and lower polaritons, and such a superposition can also be supposed as the transient state in the P-emission process. However, the amplitudes of excitons and photons (also of polaritons and of photons outside the sample) oscillate with a frequency $\omega$, which is determined by the energies of the bottleneck excitons and the $n > 1$ exciton under the energy conservation. Then, the problem is described as that of the forced oscillation in the exciton-photon composite system as Eq. (28). This kind of demonstration and detailed investigation are remaining tasks in the future.

Whereas the discussion in this paper does not deny the interpretation of the polariton diffusion$^{30}$ it is note that the decrease in diffusion constant with an increase in impurity concentration reported in Ref. $^{15}$ can be explained as a decrease in coherence volume $V_{\text{coh}}$ in our interpretation. A theoretical investigation of the crossover between the ballistic propagation and diffusion of polaritons is also a remaining task.

The P emission exhibits a threshold behavior with respect to the pumping power and also an optical gain at that frequency$^{6-11}$ Then, the inelastic scattering has been considered as a stimulated emission$^{6,11}$ or called the amplified spontaneous emission (ASE) and lasing is also reported$^{2}$ In contrast, instead of the stimulated emission of photons or polaritons, in this paper we interpret that the creation of excitons are stimulated by the accumulated excitons with the P-emission energy (stimulated scattering of excitons), and then those excitons are emitted in the conversion time $\tau_{\text{conv}}$. Obeying the discussion of the stimulated emission of photons in Sec. 22.1 of Ref. $^{1}$ we need to replace the density of photons by the density $N_{\text{ex}}$ of the excitons in our interpretation. The rate equation is rewritten as

$$\frac{d}{dt}N_{\text{ex}}^1(q) = -\Gamma(\omega)N_{\text{ex}}^1(q) + \sum_{i,j} \frac{2\pi}{\hbar} \delta(\Delta E)|W|^2 Q. \hspace{1cm} (29)$$

Here, $q$ is the center-of-mass wavevector, $\delta(\Delta E)$ stands for the energy conservation, $W$ is the transition matrix element involving the exciton scattering, and $Q$ is called the population factor. The stimulated part of $Q$ is expressed as

$$Q_{\text{stim}} = \{N_{\text{ex}}^1(k_1)N_{\text{ex}}^1(k_2) - N_{\text{ex}}^{n>1}(k_1 + k_2 - q)[1 + N_{\text{ex}}^1(k_1) + N_{\text{ex}}^1(k_2)]\}, \hspace{1cm} (30)$$

where $k_{1,2}$ are the wavevectors of the excitons at the bottleneck and $N_{\text{ex}}^{n>1}(k_1 + k_2 - q)$ is the density of higher excitons. We cannot find any reasons that prohibit such a replacement in the discussion of the stimulated emission of photons, and the stimulated creation of excitons can occur under the same logic in the series of processes.
toward the P emission. Once the lasing occurs and the radiation field gets a non-zero amplitude with temporal and spatial coherences, the inelastic scattering surely provides the stimulated emission of photons. In the measurement of the optical gain, since the probe beam propagates as a polariton with a long enough spatial coherence, we can also interpret the process as the stimulated emission of polaritons. In this case, the lifetime of the gain should be different from the P-emission decay time observed in the luminescence measurement discussed in this paper.

V. SUMMARY

Although excitons at the bottleneck region are supposed to be scattered directly to photon-like polariton states in the conventional interpretation of the P emission, we instead propose another scenario. 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 decay time observed in the recent experiments using the optical Kerr gating method. 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 excitons. However, more detailed experimental and theoretical investigations are required to finally conclude which interpretation is reasonable.

ACKNOWLEDGMENTS

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

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