Temperature dependence of photoluminescence dynamics of exciton-exciton inelastic scattering in a GaAs/AlAs multiple-quantum-well structure

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Abstract. We have investigated the photoluminescence (PL) dynamics of exciton-exciton inelastic scattering at temperatures from 10 to 60 K. It was found that the energy dependence of the PL decay rate is scaled by that of the group velocity of the photon-like lower polariton (LP) at each temperature, taking account of the broadening factor of the polariton state. This fact demonstrates that the PL decay rate is dominated by the photon-like LP which is the final state of the exciton-exciton inelastic scattering process. The broadening factor is proportional to temperature, which indicates the influence of acoustic phonon scattering on the LP state.

1. Introduction
The photoluminescence (PL) properties of exciton-exciton inelastic scattering have attracted attention in physics and applications from the 1970s [1]. Wide-gap semiconductors such as ZnO [2-4], CdS [5], GaN [6,7], and CuI [8,9] have been samples for the investigation because of high stability of the excitonic system. In addition, the PL from exciton-exciton inelastic scattering was also observed in GaAs/AlAs multiple-quantum-well (MQW) structures because of the enhancement of the exciton binding energy by the quantum confinement effect [10,11]. In the exciton-exciton inelastic scattering process, one exciton is scattered into a higher exciton state with a hydrogenic quantum number \( n > 2 \) including the continuum state \( (n=\infty) \), while the other is scattered onto a photon-like lower polariton (LP) branch under the energy and momentum conservation. The photon-like LP is converted to a photon, the so-called P emission. Thus, the polariton nature of the P emission is one of key characteristics of the exciton-exciton inelastic scattering process. In previous works, it was reported that the energy dependence of the decay rate of the P emission is scaled by that of the LP group velocity in ZnO thin films [4] and GaAs/AlAs MQWs [11]. This suggests the polariton nature of the P emission. However, the measurement temperature was limited to a low temperature around 10 K; namely, there has been no systematic investigation of the PL dynamics of the P emission.

In this work, we have investigated the temperature dependence of the PL dynamics of the P emission in a GaAs (15 nm)/AlAs (15 nm) MQW from the viewpoint of the polariton characteristics. We discuss the energy dependence of the P-emission decay rate at temperatures from 10 to 60 K in comparison with that of the group velocity of the photon-like LP, taking account of the broadening factor of the polariton state.
2. Experimental details

The sample used was a GaAs (15 nm)/AlAs (15 nm) MQW with 20 periods grown on a (001) GaAs substrate by molecular beam epitaxy. The excitation light source was a mode-locked Ti:sapphire laser with a pulse duration of 110 fs and a repetition rate of 76 MHz. The peak energy of the excitation light was fixed at 1.698 eV. Time-resolved PL spectra were measured with a streak camera system with time and spectral resolutions of 16 ps and 0.20 nm, respectively. We also observed steady-state PL spectra with a cooled charge coupled device attached to a 32-cm single monochromator with a resolution of 0.15 nm. In the PL measurements, the sample was set into a closed-cycle helium cryostat.

3. Results and discussion

Figure 1 shows the excitation fluence dependence of the steady-state PL spectrum at 10 K in the GaAs (15 nm)/AlAs (15 nm) MQW, where the vertical dashed lines labelled $E_{H11}$ and $E_M$ indicate the first quantized heavy-hole (H11) exciton and biexciton energies, respectively, which are taken from Ref. [10]. At the lowest excitation fluence, the H11-PL band is dominant. With an increase in excitation fluence, the relative intensity of the M-PL band continuously increases. The low energy tail of the M-PL band is peculiar to the biexciton PL: the so-called inverse-Boltzmann-type line shape. At an excitation fluence of 0.15$F_0$, where $F_0$ was ~40 μJ/cm$^2$, a new PL band labelled P appears with a threshold-like nature. The energy spacing between the P-PL peak and H11-exciton energy is 7.6 meV corresponding to the energy difference between the $n=2$ (2s) and $n=1$ (1s) H11 excitons [12]. This result indicates that the P-PL band originates from the P emission due to exciton-exciton inelastic scattering because of the following reason. The P-emission energy is given by [2]

$$E_p = E_{H11, n=1} - (E_{H11, n=2} - E_{H11, n=1}) - 3\sigma k_B T_{eff},$$  \(1\)

where the subscript $n$ indicates the hydrogenic quantum number, $T_{eff}$ is an effective temperature, and $\sigma$ is a positive constant smaller than unity. At the threshold, the term of $3\sigma k_B T_{eff}$ is negligible at 10 K [11]. The vertical dashed line labelled $E_p$ indicates $E_p=E_{H11, n=1} - (E_{H11, n=2} - E_{H11, n=1})$, which agrees with the P-PL peak energy at the threshold. The P-PL peak energy shifts toward the low energy side with an increase in excitation fluence from the threshold, which reflects an increase in $T_{eff}$ in Eq. (1).
energy shift of the P-PL peak is continuous; therefore, contribution of the exciton state with \( n \geq 3 \), which will result in a discontinuous energy shift because of the binding energy difference, is not considerable.

Figure 2 shows the detection-energy dependence of the PL-decay profile (open circles) at \( \sim 40 \) \( \mu \)J/cm\(^2\) at 10 K, where the solid circles indicate the system response. It is obvious that the decay profile consists of the fast and slow components. The fast component was observed in the excitation fluence region higher than the threshold for the P emission. This fact indicates that the fast component results from the P emission. The slow component originates from the biexciton PL because the P- and M-PL bands overlap with each other as shown in Fig. 1. The PL-decay profile, \( I_{\text{PL}}(t) \), including a rise component is written as

\[
I_{\text{PL}}(t) = -I_{\text{rise}}(-t / \tau_{\text{rise}}) + I_{\text{f}}(-t / \tau_{\text{f}}) + I_{\text{M}}(-t / \tau_{\text{M}}),
\]

where the subscript P (M) indicates the P (M) PL and \( I_{\text{rise}} = I_{\text{f}} + I_{\text{M}} \). The solid curve depicts the fitted result using a convolution method for the system response and \( I_{\text{PL}}(t) \). The fitted result well explains the PL-decay profile. The estimated P-emission decay times are \( \tau = 21, 24, 26, 30, \) and 36 ps at 1.518, 1.520, 1.522, 1.524, and 1.526 eV, respectively. Note that \( \tau \) systematically increases with an increase in energy. In contrast, the M-PL decay time, \( \tau_{\text{M}} \), is independent of energy: \( 0.15 \pm 0.02 \) ns.

In Fig. 3, the black solid circles indicate the detection-energy dependence of the P-emission decay rate, \( \tau_{\text{P}}^{-1} \), at 10 K. The P emission is caused by conversion from the photon-like LP, which is the final state in the exciton-exciton inelastic scattering process, to a photon. In the framework of the polariton picture, the P-emission decay rate can be written as \( \tau_{\text{P}}^{-1}(E) = \text{Av}(E) [4,11] \), where the inverse of the scaling factor, \( 1/A \), corresponds in principle to the propagation length of the photon-like LP for the photon conversion. The LP group velocity is defined as \( v_g(E) = (1/\hbar) dE_{LP}(k_0)/dk_0 \), where \( E_{LP}(k_0) \) is the LP dispersion relation and \( k_0 \) is the in-plane wave vector along the interface in the MQW. The LP dispersion relation was calculated using the following equation [11]:

\[
\hbar c^2 k_0^2 / E^2 = \varepsilon_0 + f / \left[ E_{\text{hh1}}(k_0) - E^2 - \Gamma \right],
\]

where \( \varepsilon_0 \) is the background dielectric constant, \( c \) is the light speed in vacuum, \( f \) is the oscillator strength, \( E_{\text{hh1}}(k_0) = E_{\text{hh1}} + \hbar^2 k^2 / (2M_{\text{hh}}) \). \( M_{\text{hh}} \) is the in-plane exciton mass, and \( \Gamma \) is the broadening factor. The oscillator strength is given by \( f = 2E_{\text{T}}e_0 \Delta_{LT} [13] \), where \( E_{\text{T}} \) is the transverse exciton energy corresponding to \( E_{\text{hh1}} \) and \( \Delta_{LT} \) is the energy difference between the longitudinal and transverse excitons. The values of \( e_0 \) and \( M_{\text{hh}} \) are 10.86 [14] and 0.177\( m_0 \) [11] for GaAs, where \( m_0 \) is the free electron mass. The value of \( \Delta_{LT} \) in the MQW was estimated from the experimental GaAs-layer thickness dependence [15]: 0.31 meV. The value of \( \Gamma \) at 10 K was taken from the full width at half maximum of the H11-exciton band in the PL-excitation (PLE) spectrum [10], 2.0 meV, on the assumption that the PLE line shape of the lowest-lying H11 exciton is consistent with the absorption

**Figure 3.** Detection-energy dependence of the decay rate \( \tau_{\text{P}}^{-1} \) (solid circles) of the P emission at temperatures from 10 to 60 K in the GaAs (15 nm)/AlAs (15 nm) MQW. The solid curve indicates the scaled energy dependence of the group velocity \( v_g(E) \) of the photon-like LP at each temperature. The inset shows the temperature dependence of the broadening factor \( \Gamma \) (open circles) used as the fitting parameter. The solid line indicates the least-square fitting result.
spectrum. The black solid curve indicates the scaled energy dependence of the calculated group velocity, $v_g(E)$, at 10 K: $A = 8.4 \text{ cm}^{-1}$.

The detection-energy dependences of the P-emission decay rate (solid circles), $\tau_p^{-1}$, at temperatures of 20, 30, 40, 50, and 60 K are also depicted in Fig. 3. In the calculation of $v_g(E)$, the broadening factor was treated as a fitting parameter because we have no available data. The temperature dependence of the H11 exciton energy was calculated using the Varshni equation: $E_{\text{H11}}(T) = E_{\text{H11}}(0) - \frac{\alpha}{T + \beta}$, where $\alpha = 5.405 \times 10^{-4} \text{ eV/K}$ and $\beta = 204 \text{ K}$ for GaAs [16]. The scaling factor $A$ should be independent of temperature because $1/A$ corresponds to the propagation length of the photon-like LP as described above. Thus, the value of $A$ was fixed at 8.4 $\text{ cm}^{-1}$ obtained at 10 K. It is evident that $\tau_p^{-1}(E)$ is systematically scaled by $v_g(E)$ in the temperature range from 10 to 60 K. This fact clearly demonstrates the polariton nature of the P emission. Note that the scaling factor $A$ is a phenomenological value at present. The inset in Fig. 3 shows the temperature dependence of the broadening factor. The solid line indicates the least-square fitting result, which indicates that the broadening factor is proportional to temperature: $36 \mu \text{eV/K}$. The proportional relation indicates that acoustic phonon scattering is the dominant effect on the temperature dependence of the broadening factor of the polariton state [17]. The quantitative discussion of the proportional constant is beyond the scope of this paper.

4. Conclusions

We have revealed that the detection-energy dependence of the P-emission decay rate is systematically scaled by the energy dependence of the group velocity of the photon-like LP, which is the final state of the exciton-exciton inelastic scattering process, in the temperature range from 10 to 60 K. This fact demonstrates the polariton nature of the P emission. In addition, it was confirmed that the broadening factor is proportional to temperature, which indicates the influence of acoustic phonon scattering on the broadening factor of the polariton state.

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