

Photoluminescence observation of electron-hole droplets in a GaAs/AlAs type-II superlattice

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Abstract. We have investigated the photoluminescence (PL) properties of a (GaAs)₁₂/(AlAs)₁₂ type-II superlattice (SL) at 10 K from the viewpoint of the formation of electron-hole droplets (EHDs). In the type-II SL, the lowest-energy exciton consists of an X-electron and a Γ -heavy-hole confined in the AlAs and GaAs layers, respectively; namely, the optical transition is indirect in momentum and real space. The excitation-power density was widely changed from ~ 1 mW/cm² to ~ 1 kW/cm² for precisely observing changes of PL spectra. It was found that a broad PL band appears with a threshold-like nature in the energy region lower than the biexciton. This suggests the occurrence of the Mott transition. The prominent feature of the broad PL band is the fact that the spectral profile hardly depends on the excitation-power density, which is one of typical properties of EHDs. From the line-shape analysis of the PL spectra, we estimated the stability energy of the EHD to be ~ 2.9 meV relative to the biexciton energy. The above results consistently demonstrate the formation of the EHD.

1. Introduction

The stability of electron-hole liquid (EHL), which is a liquid phase of electron-hole plasma (EHP) caused by a Mott transition, confined in quantum-well (QW) systems including superlattices (SLs) has attracted much attention as a prominent problem of high-density-excitation phenomena. Note that the EHL is usually observed in experiments as an electron-hole droplet (EHD) because the EHL with a finite size is surrounded by excitons and biexcitons in a coexistence phase [1]. There exist two types of potential structures from an aspect of the spatial configuration: “type I” and “type II”. In a type-I potential structure, electrons and holes are confined in the same QW layer. A type-II potential structure corresponds to a staggered lineup of conduction-band (CB) and valence-band (VB) potentials: The QW and barrier layers in the CB turn to the barrier and QW layers in the VB, respectively. Thus, electrons and holes are spatially separated in the type-II potential structure. Kleinman theoretically suggested that the EHL is unstable relative to biexcitons in GaAs type-I QWs with a layer thickness less than 22 nm [2]. In fact, there has been no convincing report on the observation of the EHD in GaAs type-I QWs. In contrast, Hawrylak [3] and Ando *et al.* [4] theoretically suggested the possibility of the EHD formation in type-II QW systems. In GaAs/AlAs QWs and SLs, the GaAs layer is the QW in the CB and VB at the Γ valley, while the AlAs layer is the QW in the CB at the X valley. The lowest-energy exciton state changes from the type-I exciton to the

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type-II one with an decrease in GaAs-layer thickness because the energy of the X-electron subband becomes lower than that of the Γ -electron subband, the so-called Γ -X subband crossover at a GaAs thickness of ~ 13 monolayers [5], where monolayer corresponds to a half of the lattice constant: 0.283 nm in GaAs. As a result, the type-II exciton consists of the X-electron in the AlAs layer and the Γ -heavy-hole in the GaAs layer. The optical transition is indirect in real and momentum space, which results in a long exciton lifetime of the order of μs [5]. This long lifetime is advantageous for the formation of EHDs from an aspect of cooling efficiency of excitons, biexcitons, and carriers. In a GaAs/AlAs type-II SL grown on a (311) GaAs substrate, the structure of which is like a quantum wire due to interface corrugation, the stability of EHDs was confirmed [6, 7]. However, there has been no clear report on the observation of EHDs in usual type-II SLs with flat interfaces.

In the present work, we have investigated the photoluminescence (PL) spectra of a GaAs/AlAs type-II SL, which was grown on a conventional (001) GaAs substrate, at 10 K in a wide excitation-power-density range from $\sim 1 \text{ mW/cm}^2$ to $\sim 1 \text{ kW/cm}^2$ for systematically investigating changes of spectral profiles. The exciton- and biexciton-PL bands were clearly observed. We found that a broad PL band appears with a threshold-like nature in the energy region lower than the biexciton, which suggests the occurrence of the Mott transition for the formation of EHP. The spectral profile of the broad PL band hardly depends on the excitation-power density. This fact is one of typical PL properties of EHDs. From the line-shape analysis of the PL spectra, we revealed the stability energy of the EHD relative to the biexciton energy.

2. Experimental details

The sample used was a $(\text{GaAs})_{12}/(\text{AlAs})_{12}$ type-II SL with 50 periods grown on a (001) GaAs substrate at 660°C by molecular beam epitaxy (MBE). The subscript of the SL structure indicates a number of monolayer in the constituent layers. In the MBE process, individual layer thicknesses were precisely controlled by observing oscillation patterns of reflection-high-energy-electron diffraction [8]. Note that the oscillation period corresponds to a growth time of one monolayer.

In the PL measurements, the excitation light source was a cw-mode Ti:sapphire laser. The excitation energy was fixed to 1.797 eV. We used a beam shaper to homogenize the spatial distribution of the excitation-laser power: The spatial power fluctuation was within $\pm 5\%$. The PL spectra were detected with a cooled charge couple device attached to a 32-cm single monochromator with a resolution of 0.15 nm. The excitation-power density was widely changed from $\sim 1 \text{ mW/cm}^2$ to $\sim 1 \text{ kW/cm}^2$. All the optical measurements were performed at 10 K.

3. Results and discussion

We first overview the PL spectra as a function of excitation-power density. Figure 1 shows the PL spectra at various excitation-power densities, where the maximum power density is $P_0 \sim 1 \text{ kW/cm}^2$, and the intensity of each PL spectrum is normalized by the maximum one. At the lowest power density of $1 \times 10^{-6} P_0$, a PL band labeled X is attributed to the type-II exciton, the decay time of which was evaluated to be $\sim 2 \mu\text{s}$ in our previous work [9]. With an increase in excitation-power density, a PL band labeled M continuously grows in the low-energy side of the X-PL band. The M band exhibits a low energy tail, which is a typical feature of the biexciton PL, the so-called inverse Boltzmann line shape. Thus, the M band is assigned to the biexciton PL, which was also confirmed in our previous work [9]. At an excitation-power density of $0.01 P_0$, a PL band labeled EHD appears with a threshold-like nature in the energy region lower than the M-PL band. This drastic change of the PL spectra suggests occurrence of the Mott transition. The coexistence of the X-, M-, and EHD-PL bands is a key result in the present work because the coexistence is one of typical characteristics of the EHD [1].

Figure 2 shows the PL spectra at excitation-power densities of $0.5 P_0$, $0.7 P_0$, and P_0 . The intensity of each PL spectrum is scaled by the maximum intensity of the EHD-PL band. Note that the EHD-PL band appears as a shoulder of the M-PL band at excitation-power densities less than $0.5 P_0$, so those could not be used for the comparison in Fig. 2. It is evident that the line shape of the EHD-PL band hardly depends on the excitation-power density. This behavior reflects that the carrier density and

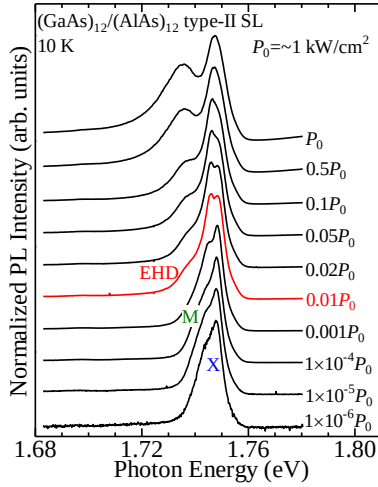


Figure 1. PL spectra at various excitation-power densities at 10 K in the (GaAs)₁₂/(AlAs)₁₂ type-II SL, where the maximum power density is $P_0 \approx 1 \text{ kW/cm}^2$, and the intensity of each spectrum is normalized by maximum one.

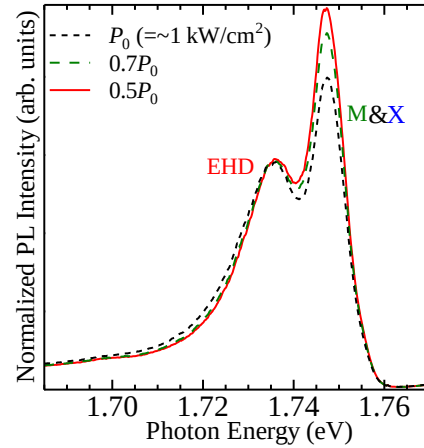


Figure 2. PL spectra at the excitation-power densities of $0.5P_0$, $0.7P_0$, and P_0 . Each spectrum is scaled by the maximum intensity of the EHD-PL band.

effective temperature contributing to the EHD formation are independent of the excitation-power density. In other words, the excitons, biexcitons, and EHDs are in quasi-thermal equilibrium.

Hereafter, we perform the line-shape analysis of the PL band to evaluate the parameters of the EHD formation. In the line-shape analysis of the EHD-PL band, we used the following two-dimensional density of states:

$$D_i(E_i) = [m_i^* / (\pi \hbar^2)] [1 + \exp(-E_i / \Gamma_i)]^{-1}, \quad (1)$$

where $i=e$ or h , m_e^* and m_h^* are the in-plane effective masses of the X-electron in the AlAs layer and the Γ -heavy-hole in the GaAs layer, respectively. The value of m_h^* was calculated using $m_h^* = m_0 / (\gamma_1 + \gamma_2)$ [10], where γ_1 and γ_2 are the Luttinger parameters: $m_h^* = 0.11m_0^*$ for $\gamma_1 = 6.8$ and $\gamma_2 = 1.9$ [11]. The value of m_e^* is taken from Ref. 12: $m_e^* = 0.19m_0^*$. Γ_e and Γ_h are the broadening factors which are fitting parameters. Assuming that the momentum conservation is relaxed, the PL line shape of EHP, which is the EHD in this case, is given by [1]

$$I_{\text{EHD}}(\hbar\omega) \propto \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} D_e(E_e) D_h(E_h) f_e(E_e, E_{F,e}, T_{\text{eff}}) f_h(E_h, E_{F,h}, T_{\text{eff}}) dE_e dE_h, \quad (2)$$

where E_e (E_h) is an electron (hole) energy relative to the conduction-band minimum (valence-band maximum), f_e , (f_h) is the Fermi distribution function of the electron (hole), $E_{F,e}$ ($E_{F,h}$) is a quasi-Fermi energy of the electron (hole), T_{eff} is an effective temperature, and E_g^* is a renormalized band-gap energy due to many body effects. The energy conservation of $\hbar\omega = E_g^* + E_e + E_h$ was taken into account.

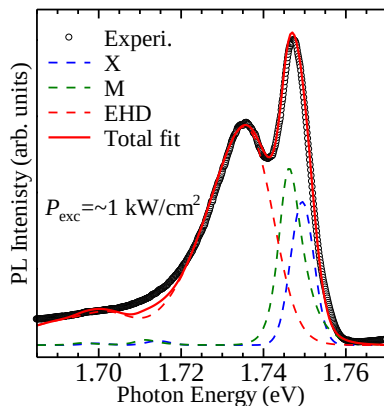


Figure 3. Results of the line-shape analysis of the PL spectrum at an excitation-power density of $P_0 \approx 1 \text{ kW/cm}^2$, where the open circles indicate the experimental spectrum, and the solid curve indicates the totally fitted result. The dashed curves depict individually fitted results of the X-, M-, and EHD-PL bands including LO-phonon sidebands of GaAs and AlAs.

Note that Eq. (2) is usually applied to the PL line-shape analysis of EHD and EHP in a gas phase [1] because the EHD and EHP(gas) consist of fermions; therefore, the Fermi distribution functions of electrons and holes dominate the PL line shapes as described by Eq. (2). For the line shape of the M-PL band, we used an inverse-Boltzmann distribution function taking account of the two-dimensional density of states [9]:

$$I_M(\hbar\omega) \propto \frac{\exp[-(E_X - E_{b,M} - \hbar\omega)/(k_B T_{\text{eff}})]}{1 + \exp[-(E_X - E_{b,M} - \hbar\omega)/\Gamma_M]}, \quad (3)$$

where E_X is the exciton energy and $E_{b,M}$ is the biexciton binding energy. We assumed that the effective temperature of the biexciton is equal to that of the EHD because of the quasi-thermal equilibrium of the EHD and excitonic system. The X-PL band was fitted by the Gaussian function.

Figure 3 shows the results of the line-shape analysis of the PL spectrum at an excitation-power density of P_0 . The LO-phonon sidebands of GaAs and AlAs were also taken into account for the line-shape analysis. For the PL bands of the exciton and biexciton, the ratios of the PL intensities of the zero-phonon band to the LO-phonon sidebands were referred to those in the PL spectrum at an excitation-power density below the threshold for the EHD. For the EHD-PL band, however, the PL intensities of the LO-phonon sidebands were treated as fitting parameters because it is expected that the interactions between the LO phonons and EHD are modified by many body effects. The estimated values of the major fitting parameters are as follows: $E_g' = 1.7280$ eV, $E_{F,e} = 6.0$ meV, $E_{F,h} = 10.6$ meV, $\Gamma_e = 2.3$ meV, $\Gamma_h = 6.3$ meV, $T_{\text{eff}} = 21$ K, $E_{b,M} = 3.8$ meV, $\Gamma_M = 1.2$ meV, and $E_X = 1.7494$ eV. These values hardly depend on the excitation-power density. The carrier density was calculated to be $4.9 \times 10^{11} \text{ cm}^{-2}$. The chemical potential of the EHD, which is given by $\mu = E_g' + E_{F,e} + E_{F,h}$, was evaluated to be 1.7446 eV. Thus, the stability energy ϕ relative to the biexciton energy, $\phi = E_M/2 - \mu = E_X - E_{b,M}/2 - \mu$, was estimated to be 2.9 meV. The stability energy is sufficiently higher than the thermal energy of $k_B T_{\text{eff}} = 1.8$ meV. The above results and analysis consistently demonstrate that the type-II EHD is stable.

4. Conclusions

We have investigated the formation of the quantum-confined EHD at 10 K in the $(\text{GaAs})_{12}/(\text{AlAs})_{12}$ type-II superlattice from an aspect of the systematic excitation-power dependence of the PL spectra. It was found that the EHD-PL band appears with a threshold-like nature in the energy region lower than the biexciton-PL band and that the line shape of the EHD-PL band hardly depends on the excitation power. In addition, the EHD-PL band coexists with the exciton- and biexciton-PL bands. The above results phenomenologically indicate the formation of the type-II EHD. From the line-shape analysis of the PL spectrum, we quantitatively demonstrated that the type-II EHD is stable: The stability energy of the EHD relative to the biexciton energy was estimated to be 2.9 meV, which is sufficiently higher than the thermal energy of $k_B T_{\text{eff}} = 1.8$ meV.

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