Growth and Optical Properties of GaSb/GaAs type-II Quantum Dots with and without Wetting Layer

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Abstract

We investigated the effect of a wetting layer (WL) on the optical properties of GaSb/GaAs type-II Quantum Dots (QDs). By adjusting annealing conditions, we fabricated GaSb type-II QDs with and without a WL-like structure in droplet epitaxy. Photoluminescence (PL) studies showed that the PL energy shift with increasing temperature is much less for GaSb QDs with the WL than without it.

1. Introduction

Stranski–Krastanov (SK) mode of growth is one of the easiest methods to fabricate quantum dots (QDs), where 10 nm-scale QDs are automatically fabricated just by depositing source materials on a lattice-mismatched substrate. In the SK growth, a thin wetting layer (WL) is spontaneously formed in the process of the QD formation and may affect the electric and optical properties of QDs. An alternative approach is droplet epitaxy; tiny droplets of a group III element are first formed on a semiconductor substrate and then exposed to the molecular beam of a group V element to form III-V QDs. This second approach has several advantages, such as high controllability of QD density and shape, abrupt boundary between each QD and the matrix, and wider choices of constituting materials. Moreover, the wetting layer (WL) thickness can be tuned [1].

In this work, we grow GaSb/GaAs type-II QDs with and without a WL-like structure in droplet epitaxy by adjusting annealing temperatures. By photoluminescence (PL) studies, we discuss how the WL affects the optical properties of GaSb type-II QDs.

2. Experiments

For our study, GaSb QDs were formed on semi-insulating GaAs (100) substrates by using a molecular beam epitaxy. By employing an As-valved cell, we precisely controlled As₄ flux, while a conventional Knudsen cell was used for Sb₄. The beam flux of Sb₄ and As₄ were about 5×10^{-7} and 1.2×10^{-5} Torr, respectively.

First, we grew a 300-nm-thick GaAs buffer layer at 590 °C. After the As background was depleted below 1×10^{-8} Torr by closing the As valve at 330 °C, 3.75 mono-layers (MLs) of Ga atoms were deposited at 200 °C to form Ga droplets. Then, Sb₄ molecules were supplied to let Ga droplets react with Sb atoms. We then annealed the samples at $T_a = 380 \sim 420$ °C for 1 minute to enhance the reaction between Ga droplets and Sb atoms and also to let the excess Sb layer be removed by desorption. These QDs were first covered by a 10 nm-thick GaAs grown at the annealing temperature T_a . Then, a 140 nm-thick GaAs layer was grown at 590 °C.

Photoluminescence (PL) measurements were performed by

mounting the samples in a closed cycle helium cryostat. The PL was excited using a frequency doubled Nd:YAG (yttrium aluminum garnet) laser with a wave length of 532 nm, dispersed by a grating monochromator, and detected by a liquid nitrogen cooled InGaAs photomultiplier tube, covering up 1.6 μ m.

3. Results and Discussion

3.1 PL Spectra for GaSb QDs with and without WL

Figure 1 shows the PL spectra of the QD samples annealed at $T_a = 380 \sim 420$ °C. A PL peak is observed at around 1 eV in all the samples, which is attributed to GaSb QDs. When the annealing temperature is relatively high ($T_a = 400, 420$ °C), another strong PL peak is found at $1.2 \sim 1.3$ eV, which arises from a WL-like structure. Such strong WL luminescence are also observed in GaSb/GaAs type-II QDs grown by the SK mode [2-4]. From the luminescence energy, the thickness of the WL is estimated to be $2 \sim 3$ ML [5]. When the annealing temperature is relatively low ($T_a = 380, 390$ °C), only a weak luminescence is observed at around $1.2 \sim 1.3$ eV. In the case of droplet epitaxy, there is no reason to form a WL by necessity. The WL formation may result from the diffusion or the incorporation of Sb atoms into the substrate during the thermal annealing. Such processes may largely occur, only when the annealing temperature is relatively high. This enables us to form the QD samples with and without a well-defined WL-like structure by adjusting the annealing temperature T_a . We speculate that there is a certain critical temperature, above which the diffusion process is strongly enhanced, since the WL luminescence abruptly rises above $T_a = 400$ °C.



Fig. 1 PL spectra of GaSb QD samples annealed at $T_a = 380 \sim 420$ °C.

3.2 Temperature dependence

The solid squares and circles in Fig. 2 show the temperature T dependences of the QD PL peak energy $E_{S1}^{(T)}$ and $E_{S2}^{(T)}$ for the samples annealed at $T_a = 380$ °C (S1) and 400 °C (S2); S1 and S2 are the QD samples without and with the well-defined WL structure. Note that the T dependence of the QD PL energy is much weaker for S2 than S1. The GaSb/GaAs hetero-interface exhibits a type-II band lineup, where only holes are strongly confined in GaSb, while electrons are pushed away from GaSb. In S2, the WL may form a potential barrier to expel the electrons and cause continuum electron states expanding outside the GaSb QD layer, as in the case of type-II QWs [5]. In type-II QWs, the absorption coefficient behaves like $(\hbar\omega - \hbar\omega_0)^{3/2}$ in the vicinity of the effective band gap $\hbar\omega_0$ [6]. Because of the energy dependent absorption coefficient, the PL peak is expected to shift toward higher energies by $3/2 k_B T$, when the temperature T is raised [5]. We speculate that a similar effect occurs and raises the PL energy by 3/2 k_BT in S2. To eliminate this effect, we subtract $3/2 k_BT$ from the QD PL energy $E_{S2}^{(T)}$ in S2. As shown by solid triangles, the slope of $\widetilde{E}_{S2}^{(T)} = E_{S2}^{(T)} - 3/2k_BT$ is quite similar to that of $E_{S1}^{(T)}$.



Fig. 2 PL peak energy of GaSb QDs as a function of T.

3.3 PL energy shifts for Excitation Power Density and Temperature

Next, we measured the PL peak energies $E_{S1}^{(P)}$, $E_{S2}^{(P)}$ of the GaSb QDs in S1 and S2 as functions of the excitation power densities *P* ranging from about 1 to 8 W/cm² at 6.7 K. We observed the increase of $E_{S1}^{(P)}$ and $E_{S2}^{(P)}$ with increasing *P*, which is characteristic of type-II band lineup and agrees with the earlier works [7]. The blue shift of GaSb QD luminescence is explained by the Coulomb charging and the QD state filling of holes [7] or by the rise of quantum levels of field-confined electrons resulted from the hole induced electric field [4]. In both cases, the energy shift is attributed to the hole population N_h in the QDs. Since the hole population N_h is approximately proportional to the integral PL intensity I_{QD} , we examine the I_{QD} dependence of the QD PL peak energy. As shown by open squares and circles in Fig. 3, $E_{S1}^{(P)}$ and $E_{S2}^{(P)}$ shift toward higher energies with increasing I_{QD} .

Next, we investigate the I_{QD} dependence of the QD PL peak energies $E_{S1}^{(T)}$ and $E_{S2}^{(T)}$ deduced from the PL data

measured at various temperatures *T*. As shown by solid squares in Fig. 3, $E_{S1}^{(T)}$ exhibits the I_{QD} dependence similar to that for $E_{S1}^{(P)}$. This indicates that the PL peak shift with the temperature *T* is predominantly caused by the hole population N_h for the GaSb QDs (S1) with no WL (i.e. $E_{S1}^{(T)}$ in Fig. 2); as *T* increases, N_h decreases, leading to the decrease of $E_{S1}^{(T)}$.

In contrast, the I_{QD} dependence of $E_{S2}^{(T)}$ does not agree with that of $E_{S2}^{(P)}$, as shown by solid circles in Fig. 3. For the GaSb QD sample with the WL (S2), the rise of the temperature T not only decreases the hole population N_h but also shifts the PL energy by $3/2 k_B T$ as discussed in Section 3.2. To eliminate the latter effect, we subtract $3/2 k_B T$ from $E_{S2}^{(T)}$ and plot $\tilde{E}_{S2}^{(T)}$ = $E_{S2}^{(T)}$ - $3/2k_B T$ in Fig. 3. As shown by solid triangles, $\tilde{E}_{S2}^{(T)}$ agrees well with $E_{S1}^{(P)}$, supporting the interpretation in Section 3.2.



Fig. 3 PL peak energy of GaSb QDs as a function of IOD.

4. Summary

We fabricated GaSb/GaAs type-II QDs with and without a WL-like structure in droplet epitaxy by adjusting annealing conditions. Optical studies showed that the PL energy of GaSb QDs with the WL has little dependence on temperature T, while the PL energy decreases with increasing T for GaSb QDs without the WL. By comparing the dependences of the PL on temperature T and excitation power density P, we discussed the origin of the PL energy shifts with T for the GaSb/GaAs type-II QDs with and without the WL.

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