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Optical properties of GaSb type-II dots by droplet epitaxy

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1. Introduction

Self-assembled quantum dots (QDs) play important roles in basic physics study as well as in device applications of zero dimensional electron systems. Although the majority of works have dealt with type-I QDs, strong interests have been attracted recently by type-II QDs in which either electrons or holes are confined in the QD, as this unique situation can lead to unique properties and functions. In case of a GaSb QD embedded in GaAs matrix, for example, holes are strongly confined in the QD by the valence band offset of about 0.81 eV, while electrons are loosely bound around the QD by the Coulomb attractive force of the holes. This results in the formation of spatially indirect excitons [1-8]. The radiative lifetime of such excitons can get far longer than that of excitons in type-I QDs [2-3], which is attractive for novel optical device applications [8].

Though GaSb QDs on GaAs have been formed mainly in Stranski–Krastanov (SK) mode [1-8], one can also grow GaSb QDs by droplet epitaxy, in which Ga droplets are first formed and then exposed to Sb molecules. This second approach has a couple of advantages, since the density and shapes of QDs can be controlled in different manners, and the thickness of a wetting layer can be tuned [9-13].

In the present work, we report on optical properties of GaSb QDs grown by droplet epitaxy. By systematic photoluminescence (PL) studies, we demonstrate that the band lineup has a type-II staggered alignment. We also reveal unique optical properties of GaSb QDs, indicative of droplet epitaxy, including the fact that PL of QDs is much stronger than that from the wetting layer (WL).

2. Droplet epitaxy of GaSb QDs and PL measurements

For our study, we formed GaSb QD samples on a (100) semi-insulating GaAs substrate by using a molecular beam epitaxy system equipped with a reflection high-energy electron diffraction (RHEED) set up. A valved cell was employed to control the flux of As_4 precisely. For Sb_4, we used a conventional Knudsen cell. The beam flux of As_4 and Sb_4 were 1.2×10^{-5} and 6.9×10^{-7} Torr, respectively. The deposition rate of Ga was determined by using the RHEED oscillations and set to about 0.7 monolayer (ML) per second. The sample surface was studied by atomic force microscope (AFM) in a non-contact mode.

After growing a 300 nm-thick GaAs buffer layer at the substrate temperature T_s = 590 ºC, we deposited 3.75 ML of Ga at T_s= 200 ºC to form Ga droplets on the c(4×4) surface of (100)GaAs. The formation of Ga droplets was confirmed by observing a halo RHEED pattern [9]. Then, the wafer was exposed to Sb_4 molecules. During this exposure process to the Sb_4 beam, the halo RHEED pattern changed to a ring-like pattern, indicating that a polycrystalline layer of Sb was formed. An AFM study of the sample at this stage would show that the surface was covered with granular poly-crystals of Sb, as will be described elsewhere. This excess granular layer can be removed by annealing the sample at T_s above 330 ºC. Indeed, we annealed the sample at T_s = 380 ºC for about a minute. During this process, the ring-like RHEED pattern changed to spotty one, indicating that GaSb QDs are formed. GaSb QDs thus formed were embedded by depositing a 10nm-thick GaAs layer at 380 ºC and a 140 nm-thick GaAs layer at 590 ºC.

Note that the formation of this polycrystalline layer of Sb is characteristic of droplet epitaxy of Sb-based materials at relatively low temperatures. In case of droplet epitaxial growth of As-based materials, such as GaAs[10,12] and InAs [11,13], QDs are formed right away, as soon as As_4 molecules are supplied to Ga droplets.

To evaluate the density and shapes of GaSb QDs by AFM, QDs were formed one more time on the sample surface by following the growth steps similar to those used for buried QDs. It was found that GaSb QDs thus formed were on average 9.2 nm in height, 74 nm in diameter and 7.8×10^{9} cm^{-2} in concentration.

PL measurements were done by mounting samples in a closed cycle helium cryostat and exciting them with a frequency doubled Nd:YAG laser light of 532nm in wavelength. PL spectra were dispersed by a grating monochromator and detected by using a liquid nitrogen cooled InGaAs photomultiplier tube, covering up to 1.6 μm.

3. Optical properties and discussion

Figure 1 (a) shows the PL spectrum of our QD sample measured at 4 K by setting the excitation power density P to be about 1.0 W/cm^2. Two PL peaks at 1.02 and 1.25eV are respectively attributed to QDs and the WL; their full widths at half-maximum (FWHM) are about 89 and 69meV. The thickness of the WL can be estimated from the PL energy and is 2~3 ML [19]. Note in Fig. 1 (a) that the integrated PL intensity I_{QD} from QDs is about 13 times stronger than that I_{WL} from the WL. This is not the case with GaSb QDs grown in the SK mode as the PL intensity of such QDs is normally comparable with or weaker than that of the WL[1-8].

When GaSb QDs are formed by droplet epitaxy, a 2~3 ML-thick WL is normally formed, which indicates the occurrence of a diffusion process such as the incorporation of Sb atoms into the substrate [14]. This diffusion was supposed to occur only on small parts of the sample surface, since the growth of the GaSb QDs was carried out at a relatively low temperature (T_s= 200~380 ºC). In addition, such

diffusion processes may cause non radiative defects or dislocations. Thus, the luminescence of the WL is weak with relatively large FWHM (69 meV).

Figure 1 (b) shows how PL spectra of GaSb QDs at 4K change when the excitation power density P is raised from 53 mW/cm^2 to 550 mW/cm^2 and finally to 5,900 mW/cm^2. Note first that both the PL intensity I_{QD} of QDs and that I_{WL} of the WL increase sub-linearly with P or tend to saturate. The ratio I_{QD}/I_{WL} is 17.5 at P = 53 mW/cm^2 but decreases to 11.5 at P = 5,900 mW/cm^2. These trends are understandable, because with the increase of P more holes will be trapped in each QD, which slows down the incoming rate of additional holes from the GaAs matrix to the QD. Note also that the PL peak from QDs shifts towards higher energies, as P increases. This blue shift is characteristic of type-II band lineup, since the trapping of more holes in a dot leads to the increase in electric fields, which results in the rise of quantum levels of field-confined electrons.

Next, the temperature dependence of PL was studied between 5K and 80K; the results are shown in Fig. 1(b) for P ~ 610 mW/cm^2 . Both the PL intensity I_{QD} of QDs and that I_{WL} of the WL decreased as T increases. Note that I_{WL} decreased more rapidly than I_{QD}; hence, the ratio I_{QD}/I_{WL} increased from 14 at 5 K to 51 at 80 K. This suggests that excitons in QDs are thermally more stable than those in the WL.

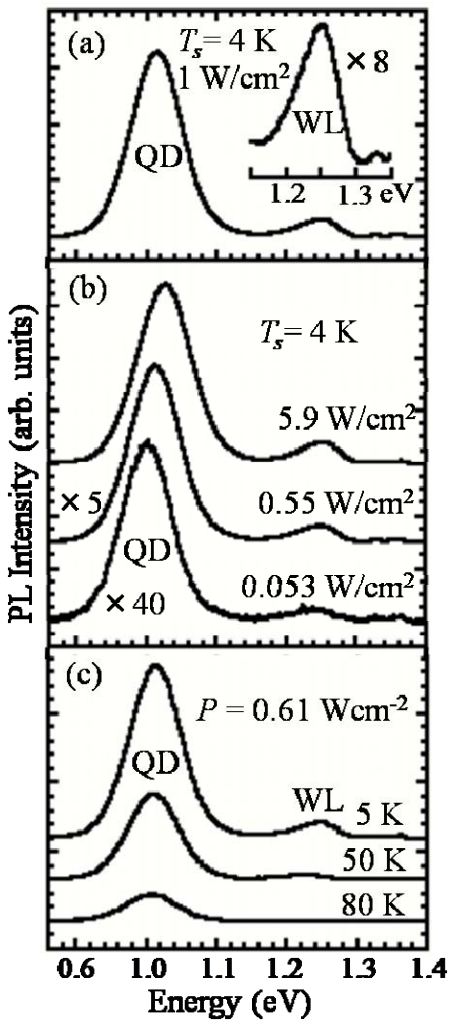

Fig. 1 PL spectrum of GaSb/GaAs QDs at T_s= 4 K, P= 1 W/cm^{-2}(a). Excitation power (b) and temperature (b) dependences of PL spectra of GaSb/GaAs QDs.

3. Summary

GaSb quantum dots embedded by GaAs with a type II band lineup were formed by a droplet epitaxy method. First, Ga droplets were covered with polycrystalline Sb grains by supplying Sb_4 beam at 200 ºC. Then the sample was annealed at 380 ºC to enhance the reaction of Ga droplets with Sb and to evaporate the excess granular layer; this two step process yielded high quality GaSb QDs. PL from QDs and the WL was found at 1.02 and 1.25 eV. The PL intensity of QDs was much stronger than that of the WL. PL peaks of the QDs and WL shifted towards higher energies as the excitation power density P increased. The thermal stability of the WL PL was far weaker than that of QDs.

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