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Direct Formation of GaAs/AlGaAs Quantum Dots by Droplet Epitaxy

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1. Introduction

There are several pioneering research works on the self organizing formation of quantum dots (QDs). In the beginning of 1980s, two kinds of nucleation have been identified when InAs is grown on a GaAs substrate, depending on growth conditions[1-3]. For ultra-thin InAs films a two-dimensional (2D) growth is observed. When the layer thickness is increased, the strain in the epitaxial film induces a transition from 2D to a three-dimensional growth (3D) with island formation, which is known as Stranski-Krastanov (SK) growth mode. This has been evidenced by reflection high-energy electron diffraction (RHEED) observations[1]. Goldstein et al[4] observed photoluminescence (PL) originated from InAs islands formed in InAs/GaAs strained-layer superlattices. After these works, Petroff's group reported the direct formation of InGaAs QDs[5] based on the SK growth mode in 1993.

In 1990, we have proposed a novel self-organizing growth method, termed Droplet Epitaxy, for the direct formation of QDs[6]. The SK type growth occurs only in the strained systems. Compared with the island formation based on the SK growth mode, the Droplet Epitaxy is applicable to the formation of QDs not only in lattice-mismatched but also in lattice-matched system. The process of the Droplet Epitaxy consists of forming numerous III-column element fine droplets such as Ga or InGa with homogeneous size of around 10 nm on the substrate surface first by supplying their molecular beams, and then reacting the droplets with As molecular beam to produce GaAs or InGaAs epitaxial microcrystals.

In this paper, recent progress on the direct formation of GaAs/AlGaAs QDs system by the Droplet Epitaxy is reviewed.

2. Fabrication and Photoluminescence Properties of GaAs/AlGaAs QDs

Our original idea for fabricating compound semiconductor QDs using metal droplets of constituent component came from the observation of the phenomenon that the excess amount of III-column element, which was supplied on the substrate surface during MBE growth of III-V compound semiconductor, condensed as homogeneous size numerous fine droplets at low substrate temperature. We thought that these droplets might change to the III-V compound semiconductor epitaxial microcrystals suitable for the QDs by subsequent V-column element molecular beam supply. However, two-dimensional lateral growth of GaAs was reported[7,8] in the case of the subsequent As molecular beam supply to the Ga droplets deposited on the GaAs substrate. One of the reasons of this lateral growth is As monoatomic layer adsorption on the substrate surface during As molecular-beam supply. Then in our early efforts concentrated on searching suitable surface to prevent the monoatomic layer adsorption of V-column element[9-12], which resulted in the two-dimensional lateral growth of III-V compound semiconductor. Although we have succeeded in the fabrication of epitaxial microcrystals of InSb and GaAs by this procedure in our early experiments, strong photoluminescence from dots was not observed in these systems. This was caused by poor interface quality and/or poor crystallinity due to the low substrate temperature during crystallization of InSb or GaAs microcrystals.

Recently we modified the procedure to overcome these problems, and observed strong photoluminescence from GaAs/AlGaAs QDs system[13-17]. Another reason of the lateral growth in the case of As molecular beam supply to the Ga droplets mentioned above is Ga atom migration from Ga droplets to the As stabilized surface, which was realized by the As monoatomic layer adsorption. Figure 1 shows the RHEED pattern and surface morphology changes of the samples for the Ga droplets formation and subsequent As molecular beam supply of 4 x 10^{-7} or 4 x 10^{-5} Torr beam

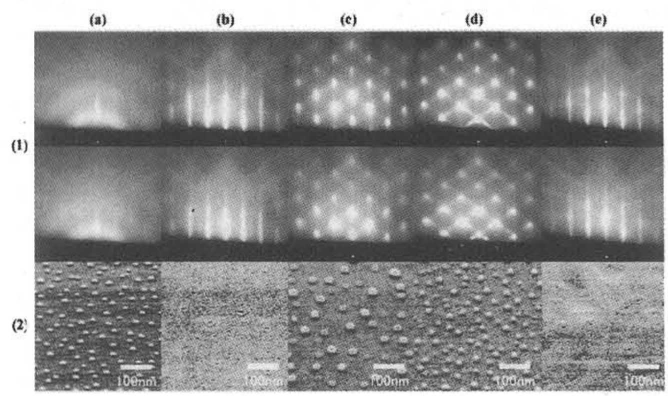

FIG. 1 (1) and (2) are the RHEED patterns and surface morphologies of the samples at each stage of the growth process, respectively. In (1), upper column: the electron beam along [110]; lower column: the electron beam along [11-0]. (a) is after the Ga deposition at 200℃. (b), (c), (d) and (e) are after subsequent As_4 molecular-beam irradiation with 4 × 10^{-7} Torr at 200℃, 4 × 10^{-5} Torr at 200℃, 4 × 10^{-5} Torr at 150℃ and 4 × 10^{-7} Torr at 150℃, respectively.

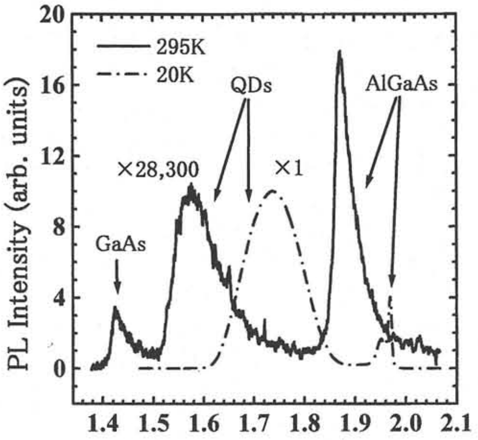

Fig. 2 PL spectra of the GaAs/AlGaAs QDs fabricated by Droplet Epitaxy after post-annealing in the As flux at 760 ℃ for 60 min.

equivalent pressure at the substrate temperature of 200 or 150℃ on the $Al_{0.30}Ga_{0.70}As$ barrier layer grown on the GaAs (001) substrates. The detailed processes of the growth were described in the Ref.14. Total amount of supplied Ga was 3.7 monolayers (ML). According to these results, it is obvious that both the supply of high As flux and the additionally low-temperature process are of significant importance for the growth of GaAs fine QDs structures. Pyramidal-shaped QDs with a typical base size of about 11 ×16 nm and a height of 6 nm are formed at even lower temperatures.

Figure 2 shows the PL spectra of the buried QDs structures by $Al_{0.30}Ga_{0.70}As$ overlayer after post-annealing in the As flux at 760℃ for 60 min. measured at 295K and 20K. The post annealing process was very effective to improve the PL properties of quantum dots. The PL intensity of quantum dots after annealing at 760℃ was enhanced by two orders of magnitude as compared to that of sample before post-annealing. Three dimensional confinement effect for excitons in this sample was confirmed by magneto-PL and micro-PL.

3. Conclusions

Recent progresses on the direct formation of GaAs/AlGaAs QDs systems by Droplet Epitaxy are reviewed. The QDs system of GaAs/AlGaAs grown by Droplet Epitaxy showed strong photoluminescence even at room temperature. Three-dimensional confinement effect for excitons in these QDs was confirmed by magneto-PL and micro-PL. This method is promising for the fabrication of compound semiconductor QDs with or without wetting layer not only in a lattice-matched system but also in a lattice-mismatched system such as InGaAs/GaAs[18-22].

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