Single-Dot Spectroscopy of Low Density GaAs Quantum Dots Grown by Modified Droplet Epitaxy

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ABSTRACT

Low density GaAs/AlGaAs quantum dots (QDs) have been fabricated using Modified Droplet Epitaxy (MDE) [1] for the spectroscopic study of single QDs using microphotoluminescence (μPL). In μPL measurements, the excitation/observation area is focused by a $50\times$ objective to a spot size of approximately $0.6\,\mu$m. Thus, a low density sample was necessary to limit the number of QDs in such a spot size to no more than one. This work made possible the first broad-spectrum single QD spectroscopy of GaAs/AlGaAs QDs fabricated by MDE. The excitation intensity dependence of the μPL spectrum of this sample shows multiple spectral lines which appear at higher excitation intensities. These lines appear from the recombination of electrons and holes from higher energy levels.

Keywords: modified droplet epitaxy, quantum dot, micro-photoluminescence, GaAs

1 INTRODUCTION

With the advent of high-quality crystal growth technology, the controlled fabrication of various semiconductor heterostructures has become possible. Stranski-Krastanov (SK) epitaxy made possible the appearance of self-aggregated quantum dots grown on a two dimensional wetting layer. However, self-assembling of QDs with the SK method has its drawback in the necessary presence of strain for triggering the island formation in the epitaxial growth. Recently, self-assembling of strain- and defect-free GaAs/AlGaAs QDs with no wetting layer has been achieved by MDE. This material is a promising candidate for investigating the interactions among confined carriers without any interactions with carriers confined in a wetting layer, as well as the undesirable effects of strain. In this work, efforts have been made to fabricate QDs with a density two to three orders of magnitude smaller than those used in a previous work [2] to make single-dot spectroscopy possible.

2 SAMPLE GROWTH

2.1 Growth Procedure

The sample is grown by MDE using a Riber-32P molecular beam epitaxy (MBE) system with elemental sources and an EPI (Veeco)-valved As cracking source, which enables the rapid irradiation of a high As_4 flux. After native oxide desorption, a 300 nm-thick GaAs buffer layer and a 500 nm-thick AlGaAs barrier are grown on a GaAs (001) wafer at 580°C. The substrate temperature is then reduced to 330°C. The As supply is stopped, and the minimum Ga supply (one monolayer) necessary for droplet formation on a c(4×4) surface is irradiated with a Ga flux equivalent to a GaAs growth rate of 0.05 monolayers/s. The substrate temperature is lowered to 150°C, and these droplets are crystallized by the irradiation of a high As_4 flux. The substrate temperature is raised to 200°C, and a 10 nm-thick AlGaAs barrier layer is grown over the QDs using migration enhanced epitaxy (MEE) [3] to avoid the two-dimensional regrowth of the QDs. The temperature is raised to 580°C, and a 90 nm-thick AlGaAs barrier and a 10 nm-thick GaAs capping layer are grown by standard MBE. The sample is consecutively annealed at 680°C for one hour.

2.2 Sample Surface Analysis

The reflection high-energy electron diffraction (RHEED) patterns, which give important information concerning the surface reconstruction, are observed during the growth process. A change in the RHEED pattern from that of an As-rich surface (c(4×4)) to that of a mostly Ga-terminated surface ((4×1)) with very weak 4× lines is seen during Ga droplet formation. The supplied Ga corresponds to the As coverage of the As-stabilized GaAs (001) c(4×4) surface [4]. These facts suggest that most of the Ga adatoms contribute to the formation of a Ga-terminated surface. The relatively low number of remaining Ga adatoms which coalesce into droplets, combined with the low supersaturation pressure (i.e. low nucleation rate) due to the relatively low Ga flux, contributes to the lowering of the QD density. The RHEED patterns after crystallization show weak {111} facet patterns, suggesting the formation of pyramidal QDs.

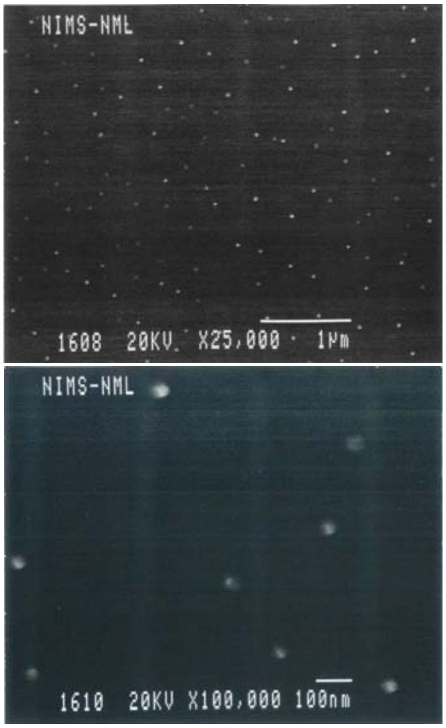

Figure 1: HRSEM image of the GaAs QD formation surface

High-resolution scanning electron microscope (HRSEM) measurements of samples fabricated under the same conditions but without a barrier and capping layer over the QDs show that the sample has an average QD base size of 40 nm and a density of $7{\times}10^8\,\mathrm{cm}^{-2}$, as shown in Figure 1. Atomic force microscopy (AFM) images of a QD sample without a barrier and capping layer show that the QDs are of a pyramidal shape. In previous works, it has been observed from cross-sectional HRSEM images of GaAs QDs made by MDE that the wetting layer possibly washes out due to group III interdiffusion during the final annealing process, and that the density of the QDs is retained after annealing. A small size reduction is also seen [5].

3 EXPERIMENTAL SETUP

In the PL measurements, the sample is cooled to 4 K by a He gas dilution refrigerator, and an Ar^+ laser (514 nm) is used as an excitation light source. The PL spectra are observed using a spectrometer and a GaAs photodetector.

In the single QD μPL measurements, the sample is placed on a continuous flow Janis ST-500 Microscopy Cryostat and cooled down to 10 K. A frequency-doubled YAG cw laser (532 nm, 20 mW) is used as an excitation light source and the light is coupled into a single mode fiber. The light from the fiber is collimated by a fiber coupler and focused onto the sample by a 50× objective lens to a spot size of 0.6 μm. The position of the excitation beam spot on the sample is controlled by adjusting the position of a concave lens placed between the fiber coupler and the objective lens. The μPL is picked up by the same objective and coupled into a different single mode fiber. The μPL spectra from the fiber are observed using a 50 cm spectrometer with a 1800g/mm grating and a liquid nitrogen-cooled CCD detector. The excitation power is changed by a neutral-density filter.

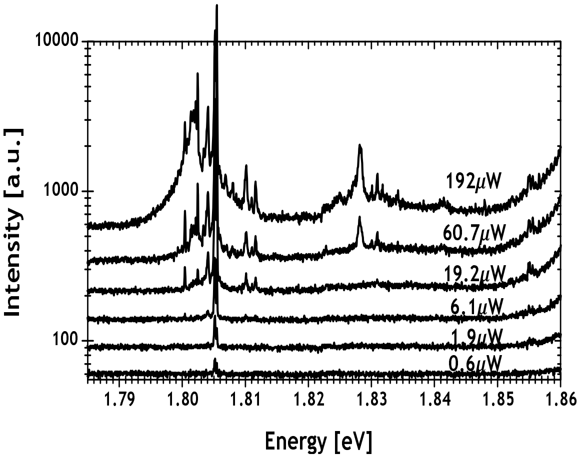

Figure 2: Excitation intensity dependence of the μPL spectrum

4 RESULTS AND DISCUSSION

The PL of this sample at 4 K shows a QD PL peak centered at 1.766 eV with a full-width half-maximum of 109 meV. The intensity of the PL peak of the QDs is low compared to that of the AlGaAs PL peak, and this reflects its low density.

The excitation intensity dependence of the μPL spectrum of this sample shows multiple spectral lines which appear at higher excitation intensities, as shown in Figure 2. The main QD peak, centered at 1.805 eV, first appears at the lowest excitation power. Various peaks surrounding the main peak begin to appear with increased excitation power. By observing the PL spectrum at high excitation power while changing the position of the excitation/observation spot in various directions on the sample with the concave lens, it is possible to differentiate the spectra of the QD in question from the spectra of separate QDs (if any exist) from the intensity change of the peaks. All the peaks' intensity in Figure 2 change in unison, thus it can be said that the observed spectral lines are all from the same QD. At 1.828 eV and its vicinity, separate peaks appear. The energy separation from the main peak is approximately 20 meV. From high pressure PL measurements of higher-density GaAs/AlGaAs QDs made by MDE, it has been determined that the heavy hole ground state and heavy hole first excited state have an energy difference of 24 meV. Thus, the peaks at 1.828 eV are possibly from the recombination of the QD conduction band ground state electron with the valence band first excited state hole. Such a recombination is possible by the coupling between optically allowed and forbidden transitions induced by Coulomb interaction among the carriers [6]. The multiple peaks which are observed several meV lower-energy side of the main peak are possibly caused by such an interaction (i.e. the electron-electron and hole-hole exchange interaction).

5 CONCLUSION

A GaAs/AlGaAs QD sample with an average QD base size of 40 nm and a density of $7\times10^{8}\,\mathrm{cm}^{-2}$ has been fabricated by MDE. From the RHEED patterns during growth, it can be seen that most of the Ga supply contributes to the formation of the Ga-terminated surface, leaving a low number of Ga adatoms to coalesce into droplets. This is the main factor in the reduction of the QD density. From μPL measurements of this sample, single QD spectroscopy has been accomplished. Most of the QD bandgap were bulk-like due to their large sizes, and their μPL spectra were indistinguishable from that of bulk GaAs. The smaller QDs with strong confinement, however, had μPL peaks at a much higher energy than that of the bulk, and the small number of small QDs in the ensemble contributed greatly to the reduction of the effective "confining" QD density, making single QD μPL observation possible. From the excitation intensity dependence of the single QD-μPL spectra, multiple spectral lines can be seen. These lines appear from the recombination of the QD conduction band ground state electron with the valence band first excited state hole.

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