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Abnormal photoluminescence for GaAs/$Al_{0.2}Ga_{0.8}As$ quantum dot-ring hybrid nanostructure grown by droplet epitaxy

Linlin Su[a], Baolai Liang[a,b,*], Ying Wang[a], Qing Yuan[a], Qinglin Guo[a], Shufang Wang[a], Guangsheng Fu[a], Diana L. Huffaker[b], Yuriy I. Mazur[c], Morgan E. Ware[c], Yurii Maidaniuk[c], Gregory J. Salamo[c]

[a] *College of Physics Science and Technology, Hebei University, Baoding 071002, PR China*
[b] *California NanoSystem Institute, University of California, Los Angeles, CA 90095, USA*
[c] *Institute for Nanoscience and Engineering, University of Arkansas, AR 72701, USA*

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ABSTRACT

The optical properties have been investigated for the GaAs/$Al_{0.2}Ga_{0.8}As$ quantum dot-ring hybrid nanostructures grown by droplet epitaxy, in which each nanostructure consists of four quantum dots (QDs) sitting on a distinct ring of GaAs. A blueshift and narrowing of the photoluminescence (PL) spectra along with the nonlinear decay of the time-resolved PL curves of the QDs have been observed. These abnormal PL behaviors are caused by the unique state filling effect correlated with the quantum dot-ring structure feature, which is strongly affected by carrier transfer from smaller dots to larger dots via the wetting ring in the GaAs/$Al_{0.2}Ga_{0.8}As$ hybrid structure.

1. Introduction

Droplet epitaxy (DE) for semiconductor nanostructures has recently attracted much attention as the DE growth mode enables flexible control of the geometry of various nanostructures and as such control of their optical and electrical properties [1–5]. In particular, DE is suitable for fabrication of novel nanostructures from not only lattice-mismatched but also lattice-matched materials [5–10]. Up to date, versatile well-defined GaAs/AlGaAs nanostructures of "zero-strain" have been fabricated by DE growth, including quantum dots (QDs), QD-pairs, QD-clusters, quantum holes, and quantum rings (QRs) [11–19]. These nanostructures have a great deal of promising advantages for optoelectronic device applications. For example, multiple stacked zero-strained GaAs/AlGaAs QD nanostructures are obtained for infrared photodetectors and photovoltaics by vertically correlated DE growth [4,14,20–22]. Also, a lattice-matched ring-shaped GaAs/AlGaAs laser has been reported employing DE [23].

In the DE growth of lattice-matched GaAs/AlGaAs nanostructures, the transition in geometry from dot to ring can be implemented by tailoring the substrate temperature and changing the arsenic flux, which changes the balance between crystallization inside and at the edge of the droplets [3]. In our previous study of type-I to type-II band-alignment transitions for QRs grown by DE [11], we observed that each individual QR is not in a perfectly formed toroidal shape, but has fluctuations in height around the ring. This height fluctuation leads to special state filling effects for the QRs. In this paper we have carefully studied the optical performance and underlying mechanism of GaAs/$Al_{0.2}Ga_{0.8}As$ quantum dot-ring hybrid nanostructures with four identifiable QDs sitting on a wetting ring in each individual structure. Abnormal luminescent behavior from these hybrid nanostructures is observed where the photoluminescence (PL) peak blueshifts and the linewidth narrows with increasing laser excitation intensity, revealing the effect of carrier transfer from smaller dots to larger dots via the wetting ring in each hybrid structure.

2. Experiments

The samples were grown by DE on semi-insulating GaAs (100) substrates in a solid source VEECO GEN-930 molecular beam epitaxy (MBE) reactor. First, a 150 nm GaAs buffer and a 100 nm $Al_{0.2}Ga_{0.8}As$ layer were grown at 580 °C. Then, the substrate temperature was lowered from 580 °C to 400 °C while the As_2 flux was turned off at 500 °C. The remaining As_2 in the MBE chamber was thoroughly pumped out for 5 min bringing the background pressure down to ~3.5E-10 Torr. After that, Ga atoms equivalent to form 6-monolayers (ML) of GaAs were deposited to obtain Ga droplets on the $Al_{0.2}Ga_{0.8}As$ surface, followed by a growth interruption of 10-s without As_2 flux. Then, the sample surface was exposed to an As_2 molecular beam (beam equivalent pressure =

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* Corresponding author at: College of Physics Science and Technology, Hebei University, Baoding 071002, PR China.
E-mail address: liangbaolai@gmail.com (B. Liang).

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2E-6 Torr) for 5 min to turn the Ga droplets into well-defined GaAs nanostructures while the substrate temperature was increased from 400 °C to 500 °C. The GaAs nanostructure layer was initially capped by 10 nm $Al_{0.2}Ga_{0.8}As$ at 500°C and then by 90 nm $Al_{0.2}Ga_{0.8}As$ at 580°C. Finally, a 3nm GaAs capping layer was grown to protect the sample surface.

The capped sample was characterized by PL and time-resolved photoluminescence (TRPL) measurements to investigate the optical properties. For reference, one sample with uncapped GaAs nanostructures was also grown using the same conditions mentioned above for morphology study. Atomic force microscope (AFM) measurements were implemented immediately after removing the sample from the MBE growth chamber. For PL, the sample was mounted in a closed-cycle cryostat with temperature variable from 10 K to 300 K. The sample was excited by a continuous-wave 532 nm laser and detected by a liquid nitrogen cooled CCD detector array attached to an Acton SP2500 spectrometer. For TRPL, the sample was excited by a NKT super-continuum pulse laser ($\lambda = 490$ nm, pulse width ~20 ps, frequency 8 MHz). The TRPL signal was measured by a PicoHarp-300 time-correlated-single-photon-counting (TCSPC) system with an overall system resolution of ~40 ps.

3. Results and discussion

Fig. 1(a) shows a 1 μm × 1 μm AFM image of the uncapped GaAs/$Al_{0.2}Ga_{0.8}As$ nanostructures. In order to clearly see the morphologic features, Fig. 1(b) presents a three-dimensional (3D) projection and Fig. 1(c) gives the cross-sectional height profile of a representative nanostructure. It is clear that, after the growth, every Ga droplet has turned into a hybrid nanostructure with a hole in the center and a ring-like structure surrounding it. The general morphological characteristics of the hybrid nanostructures are similar to the undulated nano rings in our previous study [11]. The formation of GaAs hybrid nanostructures is mainly due to the migration of Ga atoms during the crystallization of Ga droplets under a suitable temperature and arsenic flux [24]. However, Ga-rich conditions result in the deep hole at the center of each hybrid nanostructure. This has been studied as a part of the DE growth mechanisms and has been called the nano-drill effect. [25,26] The nanoholes have an average depth of ~1.8 nm measured from the sample surface. The rings have an average top diameter of ~38 nm, an outside diameter of ~64 nm, and an areal density of ~5.2×10^9 cm^{-2}. Fig. 1(d) shows the height profile along a path around the rim of an individual ring-like structure shown by the green line in the inset. Here, it can be seen that, each ring-like structure has four QDs with the height fluctuations between 0.5 nm to 0.8 nm. Therefore, they are named quantum dot-ring hybrid nanostructures. It is expected that the height fluctuation of the QDs can lead to variations of the quantum confined energy levels and subsequently affect the optical performance of these GaAs/$Al_{0.2}Ga_{0.8}As$ hybrid quantum dot-ring nanostructures. This will be well demonstrated through PL measurements.

Fig. 2 shows two PL spectra of the hybrid structures measured at 10 K with a weak excitation intensity of 10 mW/cm^2 and a strong excitation intensity of 3000 W/cm^2, respectively. Only one peak originating from the QDs is identified at ~758 nm (1.636 eV) under the weak excitation. But three different peaks can be observed under strong excitation. The peak at 751 nm (1.651 eV) is assigned to the QDs, and the emission around 693 nm (1.789 eV) is coming from the $Al_{0.2}Ga_{0.8}As$ barrier. The energy separation between the 751 nm (1.651 eV) and 730 nm (1.698 eV) peaks is ~47 meV, which is much larger than the typical energy separation from the ground state to the excited state of GaAs/$Al_{0.2}Ga_{0.8}As$ QDs. [27] Although it is still possible for this to be from excited states of the GaAs QDs [9], due to the large energy separation we believe that the final peak at 730 nm is from the wetting ring. The pronounced PL peak of the wetting ring under strong excitation is likely due to the relatively low density of the QDs, giving a relatively stronger PL signal for the wetting ring than the QDs [25].

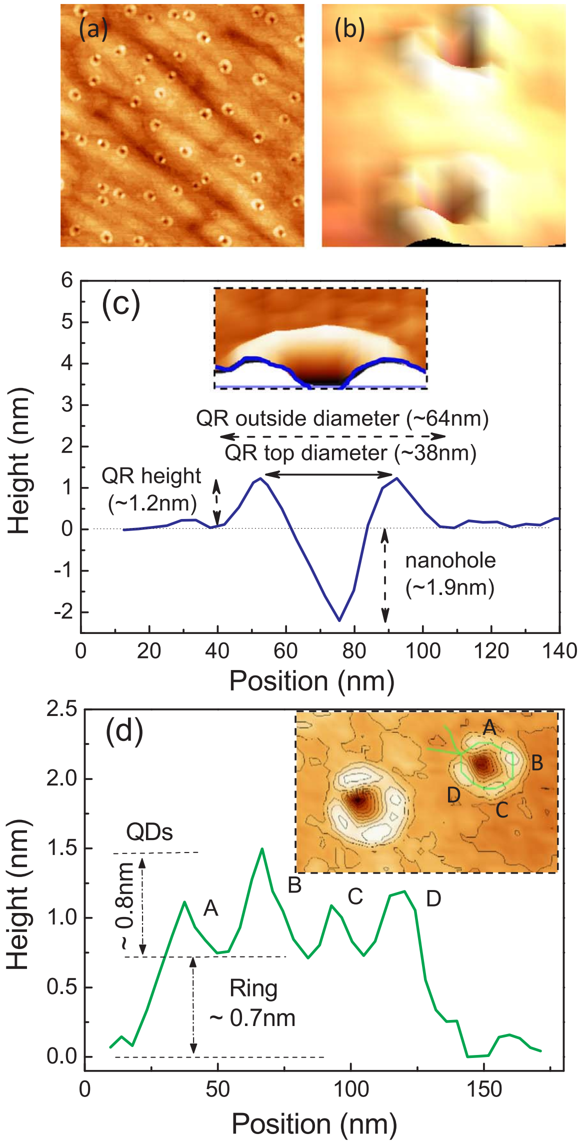

Fig. 1. AFM image and morphological property analysis of the GaAs/$Al_{0.2}Ga_{0.8}As$ hybrid nanostructures. (a) 1 μm × 1 μm AFM image; (b) 3D projection to show two individual nanostructures; (c) The cross-section profile of one nanostructure; (d) height fluctuation of an individual ring-like nanostructure along a path shown by the green line on the top of the nanostructure shown in the inset.

In order to completely understand the mechanisms underlying the PL observations, we calculate the band profile for the GaAs/$Al_{0.2}Ga_{0.8}As$ hybrid structure using an eight-band k·p model in the Nextnano software based on the detailed structural information obtained from the AFM. The material parameters for the calculations are taken from the review in Ref. [28] and evaluated at 10 K. In the calculation we neglected the strain and the small lattice-mismatch between the GaAs and the $Al_{0.2}Ga_{0.8}As$ layer. The calculated band profile of the GaAs/$Al_{0.2}Ga_{0.8}As$ hybrid structure is shown in Fig. 2(b). Fig. 2(c) presents the calculated E1-H1 transition energy as a function of the height of the hybrid structure. It can be seen that the E1-H1 emission energies match with the measured PL peak energy when we take a height of ~3 nm for the calculation. This indicates that the actual height of GaAs/$Al_{0.2}Ga_{0.8}As$ nanostructure formed during DE must be larger than the sum of the heights of the ring and QDs above the $Al_{0.2}Ga_{0.8}As$ surface. Therefore, we propose that there are GaAs structures on the sidewall of the nanohole similar to the inset of Fig. 2(c). With this model of the hybrid structure including the QDs, ring, and nanohole, the overall height of the GaAs/$Al_{0.2}Ga_{0.8}As$ nanostructure