Physica E 94 (2017) 7–14

Contents lists available at ScienceDirect

Physica E

journal homepage: www.elsevier.com/locate/physe

Influence of GaAsSb structural properties on the optical properties of InAs/GaAsSb quantum dots

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ARTICLE INFO

Keywords:
Quantum dot
Epitaxy
Heterostructure
Photoluminescence

ABSTRACT

The optical properties of InAs quantum dots with GaAsSb buffer, capping and cladding layers of different alloy compositions are studied by photoluminescence techniques. Fully strained GaAsSb layers show that the inclusion of a buffer layer gives a blue-shift to quantum dot emission, while for quantum dots capped with GaAsSb a clear red-shift is seen. Power-dependent photoluminescence suggests a transition from type-I to type-II can be achieved by GaAsSb at Sb composition between 11–13%, while the transition for the GaAsSb cladding layer occurs at around 11%. At low Sb composition, good crystal quality and energy barrier are detected by temperature-dependent photoluminescence, while high-level dislocation and defects exist under high antimony content, as evidenced by X-Ray Diffraction and Transmission Electron Microscopy.

1. Introduction

Self-assembled (SA) InAs/GaAs quantum dots (QDs) are the subject of great research interest due to the potential applications in a range of novel optoelectronic devices [1,2], such as photodetectors [3], 1.3-μm wavelength emission laser diodes [4], and photovoltaic devices [5]. Compared with conventional designs, novel devices like the Intermediate Band Solar Cell (IBSC), [6] and hot carrier solar cell [7], are predicted to provide the possibility of realizing ultra-high efficiency by offering the ability to tailor their structural and optical properties.

IBSCs allow the possible realization of solar cells with high conversion efficiencies to overcome the Shockley-Queisser (SQ) limitation [8,9]. Self-assembled quantum dots with delta-function-like density of states provide a promising way to form the intermediate band (IB) lying between the valence band (VB) and the conduction band (CB) through the ability to delocalize energy levels [10]. In particular, the InAs/GaAs quantum dots system, which is well characterized and widely grown, has attracted special interest. However, the valence band offset (VBO) existing between InAs QDs and bulk GaAs could give rise to open-circuit voltage loss [11], meaning designs that ensure a VBO of zero should be used [12].

The use of GaAsSb barriers, or cladding layers, has been suggested as a way to minimize the VBO for an IB solar cell [13], with the type I to type II heterostructure transition observed in the 12–14% Sb composition range [13–15]. Additionally, the presence of a GaAsSb buffer in the QD system leads to a higher areal density of QDs with greater size uniformity, while previous reports for samples capped by thin GaAsSb showed improvement in photoluminescence (PL) intensity with a peak red-shift indicating the capping layer provides strain relief to the QDs [16]. Long carrier lifetime and high photon absorption are preferred for the realization of IBSC, which could be achieved by zero VBO under 12–14% Sb composition. However, too high antimony content has been shown to lead to strain relaxation via extended crystalline defects like threading dislocations that can lead to the suppression of QD formation [17].

In this work, we have used photoluminescence techniques to examine samples grown with GaAsSb buffer layers, capping layers and cladding layers for various Sb compositions to observe the influence of the GaAsSb layer on the QD optical emission properties. Fully strained GaAsSb layers show that inclusion of the buffer layer gives a blue-shift to QDs emission, while for quantum dots capped with GaAsSb a clear red-shift is seen. With power-dependent PL, a heterostructure transition from type-I to type-II can be achieved by GaAsSb at Sb composition between 11–13%, while the transition for the GaAsSb cladding layer occurs at a composition lower than 11%. Temperature-dependent analysis shows a small energy barrier at the interface between InAs and GaAsSb, suggesting lower dislocations and defects level throughout the structure. Such observations are significant in terms of fabricating quantum dots solar cells which require the quantum dots to be dense and uniform but carrier lifetime to be long to avoid recombination during carrier transfer.

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http://dx.doi.org/10.1016/j.physe.2017.07.007
Received 4 June 2017; Received in revised form 17 July 2017; Accepted 17 July 2017
Available online 17 July 2017
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2. Experimental methods

All InAs/GaAs QDs structures studied were grown on GaAs (100) semi-insulating substrates in a GEN930 MBE system. The Sb_2 and As_2 fluxes were supplied by valved cracker sources and the temperature was determined by calibrating a BASF pyrometer to the deoxidation temperature of 585 °C of the GaAs substrate observed by Reflection High Energy Electron Diffraction (RHEED). After at least 200 nm of GaAs buffer layer growth, for the buffer and cladding layer samples, the growth surface underwent a 30 s soak of As and Sb [18], before approximately 17 nm of GaAsSb buffer (or capping layer) at a growth rate of 0.4 monolayers (MLs) per second was grown under various Sb_2/As_2 ratios. The substrate temperature was then ramped down to 490 °C to deposit 2 MLs of InAs to form QDs at a growth rate of ~ 0.035 ML/s by observing a consistent chevron pattern via RHEED. For the capping and cladding layer samples a further 17 nm of GaAsSb was grown immediately following QD growth. All samples were finished with a 160 nm GaAs capping layer growth that consisted of 10 nm grown at 490 °C and the remainder grown at 600 °C.

After removal from the MBE system, the thicknesses and alloy compositions were checked by ω−2θ rocking curves (RCs) using a Philips X′Pert High Resolution X-ray Diffractometer with CuKa1 radiation and a 4 bounce Ge (220) monochromators in the double crystal axis arrangement. All of the sample details are listed in Table 1.

Samples were then mounted in a closed-cycle He cryostat with a built-in temperature controller, with excitation provided by an argon ion laser (Coherent Innova 90c) at a wavelength of 488 nm with the laser light focused onto the sample using a 125 mm focal length lens. PL emission from the sample was relayed via a 4f optical telescope to a SPEX 270M scanning spectrometer fitted with a liquid nitrogen cooled InGaAs detector (EOSystems) and an SRS lock-in amplifier. The spectral resolution of the system was approximately 2 nm.

The structural characteristics of InAs/GaAs QDs with GaAsSb cladding layers under variable Sb composition were further investigated by cross-sectional high-resolution transmission electron microscopy (HRTEM) using a JEOL-JEM-3000F microscope operated at 300 kV.

3. Experimental results and discussion

3.1. GaAsSb buffer layer

The time-integrated photoluminescence measurements for InAs QDs with a GaAsSb buffer layer with variable Sb compositions were taken at 10 K, with laser excitation of 488 nm wavelength and at an incident power of 1 mW. Fig. 1(a) shows the time-integrated PL results of InAs/GaAs QDs grown on the GaAsSb (17 nm) buffer layer with various Sb compositions of 0% (S1), 1.5% (B1), 11% (B2), 13% (B3), and 22% (B4), respectively. With Sb composition increasing from 0% to 11%, it is discernible that InAs QDs experienced an enhancement of PL intensity compared with that of S1 (0%). Such result is ascribed to the GaAsSb buffer layer acting to reduce surface energy, thus generating higher areal density of the InAs QDs, as previously reported [19–21]. Furthermore, the observation of the InAs QDs ground state peak blue-shifted from approximately 1070 nm to 996 nm also indicates the reduced heights of the InAs QDs due to the surfactant function of the GaAsSb buffer layer, in keeping with previous observations [19,21]. The PL spectra for sample B2 shows a narrow InAs QDs ground state peak (FWHM 33 nm) with the largest integrated intensity counts among all the buffer layer samples, indicating a high areal QD density and uniform QD size distribution at 11% Sb. With Sb composition increasing further to 13% and 22%, a red-shift tendency for the QD ground-state peak was observed. Such red-shift behaviour can at least partly be explained by the formation of a type-II heterostructure in the interface between the InAs QDs and GaAsSb buffer at these high compositions [22], where the electrons in the QDs are likely to recombine with holes in the GaAsSb buffer layer instead of those located in the InAs.

Compared with B2 (11%), B3 (13%) had a significant peak FWHM broadening (around 100 nm), observed in the low-temperature PL result. Despite having only 2% Sb composition increment, the enlargement of the FWHM and peak red-shift suggest a likely coexistence of both type-I and type-II band structure at the interface of the InAs quantum dots and GaAsSb buffer layer, as shown in Fig. 2(d). While some electrons recombine with holes in InAs QDs (type-I), pathways for electrons in InAs QDs to recombine with holes in GaAsSb (type-II) might also occur, thus giving rise to the large FWHM value. Furthermore, the high integrated PL intensity in B3 (13%) indicates a high recombination rate for electrons in InAs QDs, giving support to the coexistence of type-I and type-II heterostructures. Such explanation corresponds with the assumption as reported [16] that a zero valence band offset between InAs QDs and GaAsSb could happen when the Sb composition reaches 12–14%.

In order to further probe the heterostructure transition, the power-dependent photoluminescence (PDPL) for all GaAsSb buffer layer samples was measured under different incident laser power settings from 1 mW to 100 mW, with an excitation wavelength of 488 nm at 10 K. The normalized spectra results for InAs QDs with GaAsSb buffer samples are plotted in Fig. 1(b) and (c), with Sb compositions of 11% (B2) and 13% (B3) respectively. As can be seen, as excitation power increases, the peak position of B2 (around 996 nm) and the line width of the ground-state emission remains unchanged, which can also be detected within the B1 sample (1.5%), indicating type-I heterostructure band alignment, as displayed in Fig. 2(c). In contrast, for sample B3 (13%) and B4 (22%), the gradually increasing laser power leads to a significant blue-shift (from 1052 nm to 1036 nm) of QD ground-state emission, as results of B3 shown in Fig. 1(c), suggesting a Type-II transition in the vicinity of InAs QDs. Fig. 1(d) displays the relationship between the peak energy of the QD ground state and the cube root of the laser power. This approach has been previously used to detect type I to type II transitions due to the exciton confinement energy increasing linearly with third root of excitation density, due to the electron field from the spatially separated electrons and holes at the interface [23–25]. As Sb composition achieves 13% (sample B3), the peak energy of the QDs shifts linearly with $P_{ex}^{1/3}$, an indicator of type-II carrier recombination [22,23,26]. These results suggest that the transition between type-I and type-II occurs at Sb compositions between 11% and 13%. Such results are consistent with previous reports where the type I to type II transition was seen around 12% of Sb composition [13,22,27].

Table 1
InAs QDs samples details with GaAsSb buffer layer, capping layer or cladding layers (both buffer and cap), under various Sb compositions of 1.5%, 11%, 13% and 22%.

Sample	GaAsSb layer	Sb composition
S1	–	–
B1	Buffer layer	1.5%
B2	Buffer layer	11%
B3	Buffer layer	13%
B4	Buffer layer	22%
C1	Capping layer	1.5%
C2	Capping layer	11%
C3	Capping layer	13%
C4	Capping layer	22%
BC1	Buffer and cap	1.5%
BC2	Buffer and cap	11%
BC3	Buffer and cap	13%
BC4	Buffer and cap	22%

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