Optical and structural properties of InGaSb/GaAs quantum dots grown by molecular beam epitaxy

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Abstract

We present the results of an investigation into the growth of InGaSb/GaAs quantum dots (QDs) by molecular beam epitaxy using migration-enhanced epitaxy. Surface atomic force microscopy and cross-sectional transmission electron microscopy show that the QDs undergo a significant change in morphology upon capping with GaAs. A GaAs ‘cold capping’ technique was partly successful in preserving QD morphology during this process, but strong group-V intermixing was still observed. Energy-dispersive x-ray spectroscopy reveals that the resulting nanostructures are small ‘core’ QDs surrounded by a highly intermixed disc. Temperature varying photoluminescence (PL) measurements indicate strong light emission from the QDs, with an emission wavelength of 1230 nm at room temperature. Nextnano 8 × 8 k.p calculations show good agreement with the PL results and indicate a low level of group-V intermixing in the core QD.

Keywords: III–V semiconductors, quantum dots, InSb, InGaSb, infrared photonics, molecular beam epitaxy

(Some figures may appear in colour only in the online journal)

1. Introduction

The optoelectronic properties of narrow bandgap III–V semiconductors make them ideal for use in devices operating in the near infrared wavelength range. Such devices have applications including 1.3/1.55 µm telecommunications [1], photovoltaics [2] and optical gas sensing [3]. A major limiting factor in their widespread use is the availability of a suitable substrate material. In this respect, GaAs is an appealing choice due to its availability in large, high purity, low cost wafers that allow the lattice matched growth of AlGaAs, which can be used to form distributed Bragg reflectors and charge confining barriers. However, narrow bandgap III–V semiconductors typically have a large lattice mismatch with GaAs, leading to low critical thicknesses and the possibility of misfit dislocations [4], which radically degrade device performance. The growth and optical properties of arsenic [5] and phosphorous-based [6] near infrared nanostructures on numerous substrate materials have been investigated intensively, but relatively few studies have been carried out on their antimonide-based counterparts.

Previous investigations have indicated that self-assembled InSb quantum dots (QDs) can be formed on GaAs substrates by molecular beam epitaxy (MBE) [1, 7, 8]. The type-II band alignment of InSb/GaAs QDs provides strong hole confinement, preventing thermalisation of carriers at elevated temperatures and
opening the possibility of their use in charge based memory devices [9]. The delta-like zero-dimensional density of states distribution in QDs also has advantages for photonic devices by reducing spectral line width and increasing radiative recombination rates.

Here we present the results of an investigation into the growth of InGaSb/GaAs quantum dots by MBE Differences in the morphology of the QDs before and after capping are discussed, along with the effects of atomic intermixing. Photoluminescence (PL) measurements and nextnano simulations are used to evaluate the optical properties of the QDs and determine their elemental composition.

2. Experimental details

The sample was grown by the migration-enhanced epitaxy (MEE) method on a Veeco GENxplor MBE system. Following the thermal desorption of the native oxide layer, a 250 nm GaAs buffer layer was deposited on the GaAs substrate at a temperature of 560 °C. A 7 min growth interruption under As flux was used to create a group-V terminated surface. The substrate was then cooled to 400 °C and held at that temperature for 5 min without group-V flux to prepare for QD growth. Growth of the QDs then proceeded by MEE using a repeated shutter sequence, as shown in table 1. The total material deposited in the QD layer was 2.9 Ml s with a nominal composition of In0.80Ga0.20Sb. Following a 140 s growth interrupt under Sb flux, a 5 nm GaAs ‘cold cap’ layer was deposited at 0.3 Ml s−1, with a substrate temperature of 350 °C. We have previously implemented a similar cold capping method in the GaSb/GaAs system to control the evolution of nanostructure morphology during GaAs overgrowth [10, 11]. The overgrowth conditions are expected to be particularly important in the In(Ga)Sb/GaAs QD system due to the large lattice mismatch (14.6%) and low bond energy of InSb, which encourages strong intermixing during capping. A further 100 nm GaAs spacer layer was grown at a substrate temperature of 560 °C and a surface QD layer was grown to allow microscopy of uncapped nanostructures. During deposition of the 100 nm GaAs spacer layer, the in situ reflection high-energy electron diffraction (RHEED) pattern changed from a spotty pattern to a clear 2 × 4 pattern. These RHEED observations indicate that the growth surface had fully recovered prior to deposition of the surface QD layer and, therefore, the buried QD layer did not affect the formation of the surface nanostructures.

3. Results and discussion

Microscopy measurements were carried out on uncapped and capped QDs using atomic force microscopy (AFM) and transmission electron microscopy (TEM) respectively. AFM measurements were carried out using a Digital Instruments multimode scanning probe microscope with a Nanoscope IIIa controller operating in tapping mode. Silicon tipped probes with resonant frequencies of roughly 300 kHz and force constants of approximately 40 Nm−1 were used. Images were analysed using NanoScope Analysis software. TEM images were obtained with a JEOL 2100 LaB6 microscope operating at 200 kV.

Images of the uncapped QDs on the sample’s surface (figure 1) revealed a double-lobed ‘coffee bean’ shape. A similar morphology has previously been observed in InSb/InAs QDs [12]. Whilst we are unable to explain conclusively the origin of this shape, we posit that it may result from coalescence of neighbouring QDs during nanostructure formation or seeding of QDs at monatomic terraces on the GaAs growth surface. The low QD density of ∼109 cm−2 is
characteristic of In(Ga)Sb nanostructures and is caused by the high surface mobility of In atoms, which tend to agglomerate into large nanostructures [7].

Cross-sectional TEM images (figure 2) reveal a significant change in nanostructure morphology after capping. The QD volume is greatly reduced and QDs appear to be disc shaped. This transformation is believed to be due to intermixing of atoms during deposition of the GaAs capping layer. Such effects have previously been reported in InAs/GaAs QDs [13] and GaSb/GaAs quantum rings [10]. As detailed earlier, the initial 5 nm GaAs cold cap was deposited on the QD layer at a low temperature, in an attempt to minimise group-V intermixing. Surface segregation of Sb and its incorporation in layers grown immediately after a high Sb-content layer is well documented [14]. Since QDs are clearly visible in the capped layer, this cold capping with GaAs appears to have been partially successful. However, since the upper surfaces of the dots appear to coincide with that of the GaAs cold cap layer, some of the material appears to have been lost from the top of the QDs. This is perhaps unsurprising since the 5 nm GaAs cold cap is insufficiently thick to fully cover the QDs, which are 11 nm tall before capping.

Closer examination of the buried nanostructures were carried out using atomic resolution TEM and energy-dispersive x-ray spectroscopy (EDX). These images, which are shown in figure 3, revealed that the wide nanostructures shown in figure 2 are actually large discs with smaller, 5 nm diameter QDs of a different alloy at their respective cores. For clarity, a schematic representation of the nanostructure is shown in cross-section in figure 4. The atomic resolution TEM image of figure 3(a) shows that the core QD is coherent and therefore is not excessively strained. The EDX measurements shown in figure 3 confirm the presence of a core QD with high In and Sb concentration, buried within a larger outer disc. The outer disc was found to have a composition of In$_{0.08}$Ga$_{0.92}$As$_{0.95}$Sb$_{0.05}$. This illustrates the strong intermixing that occurs during capping, depleting the majority of the nanostructure of In and Sb atoms, leaving a small core QD. This could also explain why the double-lobed shape of the InGaSb sample seen in figure 1 is not observed in the capped QD layers. The small diameter of the core QDs compared to the thickness of the TEM specimen prevents accurate determination of their exact composition, but they clearly have much higher In and Sb compositions than the surrounding material and have a graded composition at their outer edges. The composition of the core QD is discussed in more detail with reference to PL and nextnano simulations in the following sections.

4.2 K PL measurements were performed using a 532 nm laser and a Peltier-cooled InGaAs array detector. Laser light was delivered using a 200 μm core optical fibre and PL emission was collected by a 550 μm core optical fibre for
measurement by a spectrometer and the InGaAs array detector. The sample was immersed in liquid helium to cool it to 4.2 K. Temperature varying measurements were carried out using a closed-cycle helium cryostat. In the temperature varying measurements, a 514 nm argon laser provided laser excitation with a power density of $0.8 \text{ W cm}^{-2}$ and PL emission was collected by a monochromator with a germanium photodetector.

At 4.2 K three main emission peaks were observed, as shown in figure 5(a), which are attributed to recombination in the InGaSb QDs, InGaSb WL and the GaAs matrix respectively. The QD peak exhibited a $\sim 4$ meV blueshift with increasing laser excitation power from $1 \times 10^{-3}$ to $10 \text{ W cm}^{-2}$. With increasing temperature the GaAs and WL peaks quenched rapidly and were not observed above 120 K and 150 K respectively (figure 5(b)). However the PL emission from the QDs was still clearly visible at 300 K. These observations demonstrate the ability of InGaSb QDs to confine charge and provide efficient radiative recombination at elevated temperatures.

A nextnano++ model was created to explore these results further. The shape and dimensions (table 2) of the modelled In$_{0.80}$Ga$_{0.20}$Sb QD was informed by the TEM images and is shown in figure 6(b). The outer, intermixed disc has a composition of In$_{0.08}$Ga$_{0.92}$As$_{0.95}$Sb$_{0.05}$ and the core QD is In$_{0.80}$Ga$_{0.20}$Sb. The wetting layer was not included in the model as the TEM images showed that it is incorporated into the outer disc that surrounds the core QD. Electron and hole energy levels within the nanostructures were calculated using the $8 \times 8\ k.p$ method. Strain was calculated based on the minimisation of elastic energy within the system and included piezoelectric effects. The simulation was run assuming a sample temperature of 4.2 K and the nextnano material database file, which contains the material parameters used in the simulation, can be found online [15].

The model predicts a type-II band alignment in the QDs, as shown in figure 6(a). The predicted recombination energy in the QDs is 1.10 eV, which is close to the experimentally observed value of 1.155 eV. The small discrepancy between these two values likely results from a low level of intermixing in the core.

![Figure 5](image1.png)

**Figure 5.** (a) Power varying and (b) temperature varying emission spectra of the QD sample.

![Figure 6](image2.png)

**Figure 6.** (a) Calculated bandgap diagram and ground-state wavefunctions in the InGaSb QD sample. (b)–(d) Are two-dimensional cross sections through the modelled nanostructure; (b) shows the nanostructure only, (c) includes an overlay of the electron ground-state wavefunction and (d) includes an overlay of the hole ground-state wavefunction.
To investigate this further the nextnano simulations were repeated with varying compositions for the core QD. These simulations showed that if group-III intermixing has occurred, i.e. an increase of the Ga composition, the recombination energy would decrease, moving it further away from the experimentally observed value. However, if intermixing was primarily of the group-V elements, i.e. an increase in the As composition, this would increase the recombination energy, as shown in figure 7. Therefore, these simulations suggest that the core QD composition is In$_{0.80}$Ga$_{0.20}$As$_{0.19}$Sb$_{0.81}$.

**4. Conclusion**

In summary we have shown that the morphology of In$_{0.8}$Ga$_{0.2}$Sb QDs grown by MEE alters significantly during capping with GaAs. During overgrowth with GaAs the double lobed QD transforms into a smaller, 5 nm diameter core QD surrounded by a highly intermixed disc with a diameter of 45 nm. PL measurements showed strong emission from the nanostructures, with the QD peak still clearly visible at 300 K. Simulations using nextnano software were in good agreement with the PL spectra and suggested a low level of group-V intermixing in the core QD. These results, along with TEM measurements, show that the resulting nanostructure have an In$_{0.80}$Ga$_{0.20}$As$_{0.19}$Sb$_{0.81}$ core surrounded by a In$_{0.08}$Ga$_{0.92}$As$_{0.95}$Sb$_{0.05}$ disc. These findings indicate that the growth conditions during capping of the QDs with GaAs strongly determines their resulting morphology and optical properties. It may be possible to extend the InGaSb QD emission to longer wavelengths by slightly modifying the MBE growth conditions immediately following nanostructure formation, e.g. by increasing the GaAs ‘cold cap’ thickness.

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