MOVPE grown self-assembled and self-ordered InSb quantum dots in a GaSb matrix assessed by AFM, CTEM, HRTEM and PL

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Abstract

Self-assembled InSb quantum dots (QDs) were grown by metal-organic vapour phase epitaxy (MOVPE) in a GaSb matrix. Atomic force microscopy (AFM), conventional diffraction contrast transmission electron microscopy (CTEM), high resolution transmission electron microscopy (HRTEM), and photoluminescence (PL) were used for the assessment of the QDs. Reductions in the III/V ratios and growth rates resulted in a change of the morphology of the InSb islands from hillocks without facets, and a low level of order to dumbbell shaped islands with distinct facets and a higher level of order. © 2001 Published by Elsevier Science B.V.

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1. Introduction

There is currently an increasing interest in the growth and characterization of semiconductor quantum dots for opto-electronic devices. Heteroepitaxy in the Stranrski–Krstanow growth mode is one of the promising routes towards the fabrication of such QDs, because this process is self-assembling and avoids the drawbacks of the lithographic techniques. A high size uniformity of self-assembled QDs is achievable through self-ordering processes [1]. Little work has been done to extend the wavelength range at which potential QD based opto-electronic devices might work in the mid infrared region [2–5]. The aim of this paper is to expand on our earlier reports on such investigations [3–5] and present new experimental observations on InSb QDs in a GaSb matrix.

2. Experimental

All samples were grown by metal-organic vapour phase epitaxy (MOVPE) on nominal (001) oriented substrates of either GaSb or GaAs, the latter having been covered with GaSb buffer layers of typically 0.5–2.6 μm thickness prior to the growth of the InSb QDs. The susceptor temperatures of the MOVPE reactor during the growth of the InSb QDs were 490 ± 10°C. The InSb QDs formed after the deposition of an amount of InSb that was equivalent to about 4 to 8 monolayers (ML), followed by the over-growth of a GaSb capping layer of typically 0.1–0.5 μm thickness. Two different sets of growth conditions were employed, below referred to as initial and modified growth conditions. The modified growth conditions are characterised by lower III/V ratios and growth rates. More details on both sets of growth parameters are given elsewhere [4,5].

AFM was performed on uncapped samples, analysing InSb islands, i.e. predecessors of InSb QDs. CTEM and HRTEM analyses were carried out on both uncapped and capped samples, i.e. both InSb islands and InSb QDs were analysed. PL, on the other hand,
Fig. 1. High-resolution AFM image of InSb islands on GaSb, III/V ratio: 5/1, growth rate, 1–2 monolayers per second (ML s−1). The image edges are oriented parallel to (110) directions. Terraces of ML height, which are consistent with an 0.5° misorientation, and two-dimensional islands (e.g. marker D) are depicted in addition to three different types of three-dimensional islands (markers A–C). While marker A stands for a fully strained island, markers B and C stand for partially relaxed isolated and coalesced islands. Only fully strained islands are considered to be predecessors of QDs. All of the three-dimensional islands may have nucleated at the edges of ML terraces, i.e. at locations of reduced strain.

was always performed on capped samples. Atomic resolution Z-contrast images, which will be shown elsewhere, were also taken from some of the capped and uncapped samples, employing the JEOL JEM STEM/TEM 2010 of the University of Illinois at Chicago [6]. While CTEM was performed on a Philips CM 20, and a JEOL JEM 2000CX at the Department of Materials of Oxford University, a JEOL JEM 3010 at the University of Illinois at Chicago was used for the HRTEM investigations. AFM was performed using a Parks Scientific Instrument SEM-BD2 scanning probe microscope at an interdepartmental facility of Oxford University. PL and magneto-PL were performed at the Clarendon Laboratory of Oxford University, employing user built equipment and magnetic fields of up to 15 T.

3. Main results and discussion

AFM examinations on uncapped samples grown under the initial growth conditions revealed the co-existence of three different types of three-dimensional InSb islands on GaSb, Fig. 1 [3]. The strained state of the smallest islands was confirmed by CTEM [4]. PL showed QDs emission at around 1.7 μm. Only fully strained embedded InSb particles are considered to luminesce with QD characteristics. Magneto-PL experiments showed that the largest QDs were about 4 nm high and had a diameter of about 30 nm. The height of the QD-PL peak scaled with the number densities of the QDs which was up to 5 × 10^9 cm−2, as determined by AFM. The best samples of this series possessed full widths at half maximum (FWHM) of the PL peaks as low as 15 meV [3].

From CTEM we could earlier only conclude that the QDs which were grown under the initial growth conditions did not possess a uniform shape. This conclusion could be drawn from the non-uniform distribution of the direction vectors of the typically occurring ‘black–white’ contrasts with respect to the diffraction vector [4]. HRTEM in cross-section, Fig. 2a and 2b, showed that the dominant shape of the QDs in these samples is similar to ‘convex lenses with varying degrees of curvatures’, as indicated by the coarse dark fringes superimposed on the fine lattice fringes. The spacing of the coarse fringes in Fig. 2a correspond well with the

Fig. 2. HRTEM images of two InSb QDs in a GaSb matrix; III/V ratio, 5/1, growth rate, 1–2 ML s−1. The shape of these QDs is similar to that of ‘convex lenses with varying degrees of curvature’. While the QD in Fig. 2a has a lens shape, Fig. 2b shows a QD with an almost spherical shape. Both QDs are shown in cross-section geometry, and the growth direction is indicated by the arrow.
Fig. 3. Dumbbell shaped InSb islands on GaSb, III/V ratio (x), 5/5, growth rate 0.5–1 ML s⁻¹. (a) 220 bright-field diffraction contrast CTEM image. Moiré fringes that clearly reveal facets and the dumbbell shape of this island can be seen in plan-view geometry. (b) Low-resolution AFM image in plan-view geometry, 2.5 × 2.5 μm² scan area. The third dimension of this image is represented by 256 grey levels that cover the whole range from black to white within 50 nm. The AFM tip was scanned parallel to the [110] direction and the short axes of the dumbbells were roughly perpendicular to this direction.

calculated spacings for 200 and 220 moiré fringes arising from a significantly relaxed InSb QD in a GaSb matrix. These fringes show that the QD is 47 nm in width and 28 nm in height. The width corresponds well with islands of type B in the AFM image of Fig. 1, while the height is larger than expected. The convex curvature on the side of the substrate in Fig. 2, together with other evidence, suggests that the particle has locally alloyed into the substrate, and hence has a greater height when viewed in TEM cross section than as determined by AFM.

For the modified growth conditions, i.e. lower growth rates and III/V ratios, we obtained dumbbell shaped islands, Fig. 3a and 3b Fig. 4. Since Fig. 3b was taken with the AFM tip scan direction parallel to the long axis of the dumbbells and Fig. 4 with the AFM tip scan direction perpendicular to this axis, we are confident that the dumbbell shape of the islands is not an AFM tip artefact. Independent proof of this particular island shape has been obtained by CTEM, see Fig. 3a which was taken from the same uncapped sample. At the moment, HRTEM investigations are being undertaken on samples grown under the modified growth conditions in both cross-section and plan-view geometry in order to derive the shape of the QDs in these samples.

PL of the sample grown under the modified conditions showed two peaks at around 1.6 and 1.75 μm [5]. The longer wavelength peak was shown by means of magneto-PL to possess QD emission characteristics. The FWHM of the QDs PL peaks of the best samples of this series were as low as 10 meV. The size of the largest QDs in the samples grown under the modified growth conditions, as determined by magneto-PL, was comparable to the size of the largest QDs grown under the initial growth conditions.

Self-ordering of the InSb islands and QDs was observed by a combination of AFM and CTEM. We employed the phenomenological classification scheme of the hierarchy of islands and QDs self-ordering mechanisms by Bimberg et al. [1]. We are aware that this classification scheme is only qualitative and that the boundaries between the hierarchy levels are not well defined, but as far as we know, there is no better classification scheme available.

According to [1], there are four levels to the hierarchy of self-ordering mechanisms: 1st-orientation order, 2nd-shape order, 3rd-size order, and finally 4th-alignment order. Any higher level of hierarchy includes the feature(s) of the lower level(s). As mentioned
above, CTEM and HRTEM showed that there is no shape order for QDs that were grown under the initial growth conditions. There was, however, as already observed several years ago for InAs islands on GaAs [7], orientation order (1st level of the ordering hierarchy) possibly due to preferential nucleation at the edges of terraces, see Fig. 1.

For the samples grown under the modified growth conditions, we observed shape, i.e. the 2nd, and same size order, i.e. the 3rd level of the ordering hierarchy, see Figs. 3b and 4. The lowest growth rate and \( \text{III}/\text{V} \) ratio resulted in the highest level of ordering, Fig. 4. The fact that we observed the occurrence of faceted islands in conjunction with self-ordering of higher levels suggest that facet induced self-limiting growth of fully strained and partially relaxed islands [8] may be one of the self-ordering mechanisms [5]. The higher levels of self-ordering of the samples grown under the modified growth conditions correspond well with the reduced FWHM of the PL peaks, as given above. This indicates that the self-ordering is strongly dependent on the growth conditions.

4. Conclusions

Self-assembled InSb QDs were grown in a GaSb matrix and luminesced in the mid-infrared. Lower \( \text{III}/\text{V} \) ratios and growth rates led to a change in the morphology of the InSb islands from hillocks without facets and a low level of ordering to dumbbell shaped islands with distinct facets and a higher level of ordering. The full widths at half maximum of the photoluminescence peaks decreased correspondingly from 15 to 10 meV, indicating a better QDs size distribution for the samples with lower \( \text{III}/\text{V} \) ratios and growth rates. Further, optimisation of the growth conditions may lead to a better QDs size distribution and superior photoluminescence properties.

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References