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Japanese Journal of Applied Physics **49** (2010) 060210 **RAPID COMMUNICATION**

Achievement of InSb Quantum Dots on InP(100) Substrates

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The formation of InSb quantum dots within a $GaAs_{0.51}Sb_{0.49}$ matrix lattice matched to InP is investigated. We show that the deposit of InSb on $GaAs_{0.51}Sb_{0.49}$ alloy surface, allows the achievement of a high density of InSb islands without dislocations. A strong dissolution of InSb quantum dots occurs during the capping with a GaAsSb layer. Reflection high energy electron diffraction analysis shows that InSb island dissolution occurs during the growth interruption under As and Sb. We propose a procedure based on the deposit of a thin GaSb capping layer on top of InSb islands to prevent the As/Sb exchange. Optical properties are investigated using photoluminescence. Electronic properties are discussed within an improved tight-binding model. © 2010 The Japan Society of Applied Physics

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During last decade, a great interest has been devoted to self assembled quantum dots (QDs) elaborated from the Stransky–Krastanow growth mode.[1)] Most of researches have focused on InAs QDs formed on GaAs or InP substrates. The main motivation was the achievement of high performance QD lasers working at 1.3 and 1.55 μm telecom wavelengths.[2,3)] However new applications of QDs are today considered such as in high performance mid-infrared QD lasers[4,5)] or in ultrafast universal memories.[6,7)] For these applications, low gap material and large valence band offset are required. InSb has a small band gap of 0.17 eV at room temperature and presents a valence band offset as large as 1.16 eV with AlAs. Due to these unique properties a non volatile memory storage time of 10 years has been predicted for InSb QD memory.[7)]

Few attempts of InSb QD elaboration on GaAs,[8)] InP,[9,10)] GaSb,[11)] and InAs[12)] substrates have already been reported. However due to the large lattice mismatches of InSb with the conventional substrates (14% with GaAs, 10.4% with InP, and 6.3% with GaSb) and to the low dislocation generation energy in InSb, the SK islands are often plastically relaxed and thus can not be incorporated into devices. QDs without dislocations have been only achieved on GaSb or on InAs substrates by using special growth procedures based on As/Sb exchange[12)] or on the recrystallization of an amorphous InSb layer.[11)]

In this work we studied the formation and the capping of InSb quantum dots formed on $GaAs_{0.51}Sb_{0.49}$ alloy lattice matched on InP(100). QDs in high density ($10^{10}/cm^2$) without dislocation are achieved despite the large mismatch existing between InP and InSb (i.e., 10.4%). A large dissolution of InSb QDs has been observed during capping. This has been related to As/Sb exchange at the InSb QD surface. A modified capping layer procedure in order to achieve capped InSb QDs within GaAsSb lattice is proposed and tested.

Strain induced InSb QDs were grown by solid source molecular beam epitaxy on InP(100) substrates. The arsenic and antimony flux were produced by valved cracker cells. The growth temperature was set at 450 °C during the whole growth runs. Before the InSb deposition, a 50 nm thick $GaAs_{0.51}Sb_{0.49}$ layer was grown. In order to achieve high composition control during the growth of the GaAsSb alloy, the V/III ratio was maintained close to unity. X-ray diffraction measurements were performed on test samples to control the GaAsSb layer lattice matching condition. After turning off gallium and arsenic fluxes, by closing cell shutters and arsenic valve, a 60 s growth interruption under antimony flux was performed to reduce the residual arsenic pressure in the growth chamber. Then InSb with different nominal thicknesses from 1 monolayer to 2 ML was deposited at 0.4 ML/s. The indium deposition rate has been previously calibrated from X-ray diffraction on GaInP/InP superlattices. The samples for atomic force microscopy (AFM) characterization were immediately cooled down to room temperature under Sb overpressure. For capped samples, a 5 s growth interruption under As and Sb flux was performed in order to stabilize group V pressure before the growth of GaAsSb layers. Plan view and cross sectional transmission electron microscopy (TEM) observations were performed at 200 kV.

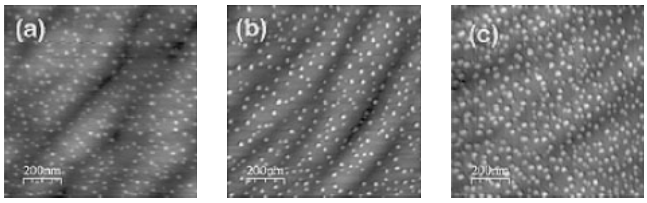

Fig. 1. AFM images recorded after the deposition of 1 ML (a), 1.5 ML (b), and 2 ML (c) of InSb.

The $1 \times 1\,\mu m^2$ AFM images recorded after the deposition of 1, 1.5, and 2 ML of InSb are shown in Fig. 1. All images reveal the formation of InSb islands. For deposits lower than 1 ML (not shown) islands are not observed. This indicates that the critical thickness of InSb island formation on GaAsSb/InP(100) is close to 1 ML. The island height increases from 2.5 to 5 nm for 1 ML to 2 ML samples. The densities for 1, 1.5, 2 ML are estimated at 4.8×10^{10}, 5.5×10^{10} and $6.1 \times 10^{10}\,cm^{-2}$, respectively. A slight increase of the average diameter is also observed as a function of the amount of InSb deposited. The average diameters are estimated at 13 and at 17 nm for 1 and 2 ML samples respectively. Note that arsenic residual pressure during InSb deposition is a key parameter during InSb QD growth. When InSb deposits are performed at residual arsenic pressure of 5×10^{-8} torr or higher, InSb

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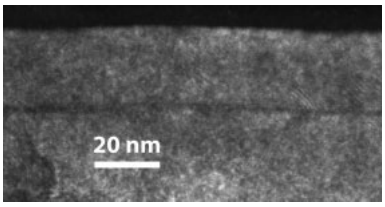

Fig. 2. Cross-sectional TEM observation [(200) dark field] taken from 1.5 ML InSb QDs after capping with GaAsSb.

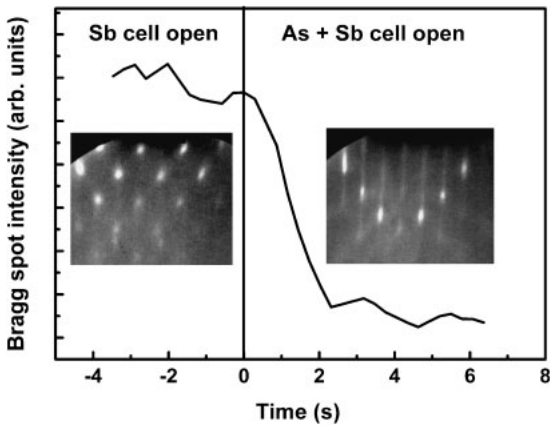

Fig. 3. RHEED patterns taken along the [1$\bar{1}$0] azimuth (a) prior the As+Sb growth interruption and (b) after a 15 s growth interrupt under As+Sb pressure. RHEED intensity measured as a function of time at the Bragg positions.

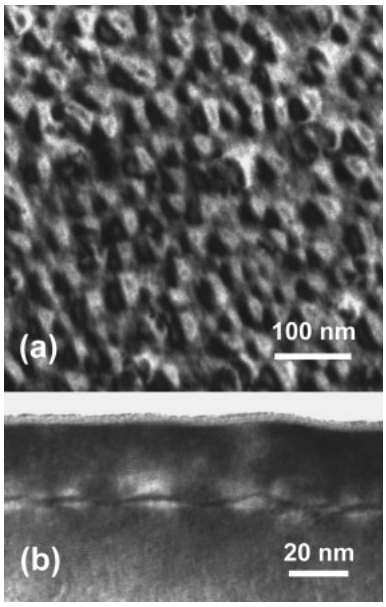

Fig. 4. TEM images taken from 1.5 ML InSb QDs capped with GaSb and GaAsSb layers with the modified capping layer procedure. (a) Plan-view observation [(022) dark field] (b) Cross-sectional observation of strained islands [(200) bright field image].

islands are not observed any longer. We assume that this is related to the high arsenic incorporation into InAsSb.[13] Thus a fine control of the arsenic residual pressure is required to achieve InSb islands on GaAsSb surfaces.

Next, we turn to the capping of InSb islands by lattice matched GaAsSb layers. Figure 2 presents a cross sectional TEM observation recorded on a sample for which 1.5 ML of InSb has been first deposited and then covered by a GaAsSb alloy layer. A dark line is observed corresponding to the InSb deposit. Despite the achievement of InSb islands, as demonstrated *ex situ* by AFM on similar samples and *in situ* by reflection high energy electron diffraction (RHEED), QDs are not observed on samples on which GaAsSb capping layers have been deposited. This result can be interpreted by a strong dissolution of InSb islands during the capping layer growth.

In order to get insights on the InSb islands capping process by GaAsSb layers, complementary RHEED experiments were performed. After the deposition of 1.5 ML of InSb, spotty patterns are observed (Fig. 3). They evidence the formation of InSb islands immediately after InSb deposition. A large change of RHEED patterns occurs when the fluxes are switched from Sb to Sb+As before the growth of GaAsSb capping layer. The patterns turn from spotty into streaky, indicating the disappearance of InSb islands. The monitoring of Bragg spots shows that this phenomenon is very fast. A complete disappearance of Bragg spots associated to InSb islands is observed less than 3 s after the arsenic cell opening. Therefore, the modification of InSb islands takes place during the group V flux switch and not during the GaAsSb capping layer growth. Such evolution has been already observed during capping of InSb QDs formed on InAs substrates[14] and has been related to Sb/As exchange occurring at the InSb QDs and wetting layer. When the arsenic flux is switched on, As/Sb exchanges lead to a change of the island composition from InSb to In (As,Sb) and thus a decrease of the lattice mismatch between islands and the substrate. Because critical thickness of InAs QDs formation on InP(100) is around 2 ML,[15] the InAsSb islands become unstable and the surface growth front is smoothed.

In order to protect InSb QDs, a modified capping procedure is used for new samples. A 3 nm thick GaSb layer is deposited after the 1.5 InSb ML deposit and before establishing the arsenic flux and to growth the GaAsSb capping layer. The onset of plastic relaxation of InAs, presenting similar lattice mismatch with InP than GaSb occurs between 4 and 9 nm.[16] Dislocation generation during GaSb deposition is thus not expected. A relatively thick layer has been chosen to facilitate the observation of GaSb capping layer by TEM (Fig. 4). The plan-view observation shows a large density of islands ($5.4 \times 10^{10}\,\mathrm{cm}^{-2}$). This density measured in capped sample is comparable to the density measured by AFM in uncapped islands ($5.5 \times 10^{10}\,\mathrm{cm}^{-2}$). Thus the thin GaSb layer grown on top of InSb islands protects them against dissolution during As+Sb growth interruption. Moreover, most of the QD are laterally strained to the matrix without dislocation within. We estimated the percentage of plastically relaxed InSb QD lower than 3%. We assume that the achievement on InSb QDs on InP substrates despite the large lattice mismatch should be related to the reduced diffusion length of indium on GaAsSb surfaces[17] which favours the formation of small islands. Similar arguments have been proposed to explain the higher densities achieved on GaSb substrate when a thin InAs layer is deposited before the InSb deposition.[18] Cross sectional observation reported in Fig. 4(b) shows that strained islands present a height between 3.5 nm and 5.3 nm and a basis width

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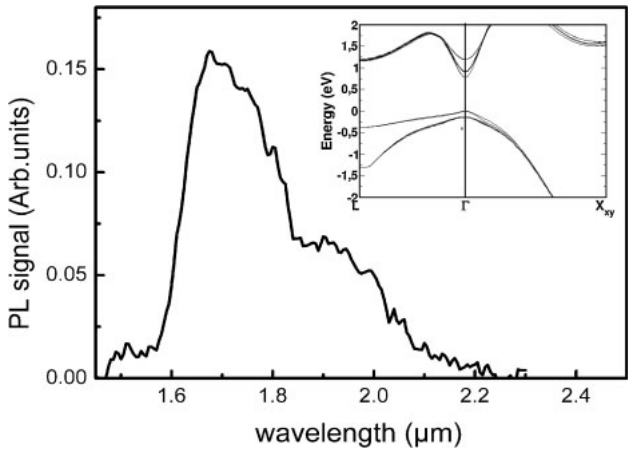

Fig. 5. PL spectrum recorded at 10 K from 1.5 ML InSb QDs capped with GaSb and GaAsSb layers and in inset the full band structure calculated for 1 ML InSb QW in GaAsSb matrix.

between 25 and 30 nm [Fig. 4(b)]. In comparison with the values deduced from AFM images, a slight increase of the height and a large increase of the diameter from 13–17 nm for uncapped islands to 25–30 nm are observed. Therefore, a large increase of the QD volume is induced by the GaSb layer. It is related to intermixing between the strained GaSb layer and InSb islands. Indeed, despite thick GaSb layer, it cannot be clearly distinguished on the TEM image. Actually, after the GaSb layer growth, the QD composition is expected to be (Ga,In)Sb rather than pure InSb, as observed in the (Ga,In)As on GaAs material system.

The photoluminescence (PL) spectrum was measured up to 4.6 μm at 10 K using liquid nitrogen cooled InSb detector. A peak around 1.7 μm was detected (Fig. 5). Recently PL emission at 1.72 μm has been observed from InSb/GaAsSb QDs and has been attributed to radiative recombination within QDs.[19] In order to assign PL peak, electronic band structure calculations were performed within the tight-binding (TB) approximation using an sp3 s*d5 nearest neighbor model that includes the spin–orbit coupling.[20] This 40-band TB model adequately reproduces measured effective masses, interband transition energies, and deformation potentials of III–V semiconductors. The usefulness of the present approach was demonstrated in the calculation of the optical properties of several III–V semiconductor strained quantum wells (QWs) and superlattices (SLs).[21] Our calculations have been performed for 1 and 13 ML thick QWs, which correspond to the wetting layer thickness and the InSb QD height.

Figure 5 inset shows the calculated band structure for the (InSb) 1 ML/($GaAs_{0.5}Sb_{0.5}$) QW. We find the conduction band minimum is characterized by a Γ derived state with a wavefunction localized on the ($GaAs_{0.5}Sb_{0.5}$) layer. Since the valence band maximum occurs at Γ, localized in the InSb monolayer, the interband transition is direct in momentum space and indirect in real space (type II) with energy of 0.77 eV, corresponding to low oscillator strength of 3. In comparison, the oscillator strength of GaAs bulk is about 10. Type II is further evidenced in the (InSb) 13 ML/($GaAs_{0.5}Sb_{0.5}$) QW where our calculations give an optical transition energy of 0.34 eV associated with a near zero (0.1) oscillator strength.

Thus we assume that PL peak at 1.7 μm (0.73 eV) is related of transitions within the WL, in accordance with the calculations. The long carrier lifetimes due to the type II band line up and the wide spectral spreading of the optical transitions induced by the size distribution lead to a very low PL signal from InSb QDs which should hardly be detectable with our experimental set up.

In summary, the formation of InSb QDs on GaAsSb alloy lattice matched to InP is investigated. A high density ($5 \times 10^{10}\,cm^{-2}$) of dislocation free islands is achieved. However a complete dissolution of the InSb islands occurs under As+Sb flux before GaAsSb capping layer growth. We relate this phenomenon to the destabilization of islands, due to the efficient As/Sb exchange taking place at the InSb QD surface. The growth of a thin layer of GaSb on the top of InSb islands prevents the island dissolution. Therefore, we show that despite large lattice mismatch and As/Sb exchanges on InSb surface, strained (Ga,In)Sb QDs can be achieved on InP(100) substrates.

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