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Capping of InAs quantum dots grown on (311)B InP studied by cross-sectional scanning tunneling microscopy

C. Çelebi,^{a)} J. M. Ulloa, and P. M. Koenraad

COBRA Inter-University Research Institute, Eindhoven University of Technology, P.O. Box 513, NL-5600MB Eindhoven, The Netherlands

A. Simon, A. Letoublon, and N. Bertru

Laboratoire d'Études des Nanostructures Semiconductrices (LENS), INSA-Rennes, F-35043 Rennes Cedex, France

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Cross-sectional scanning tunneling microscopy was used to study at the atomic scale the impact of the capping material on the structural properties of self-assembled InAs quantum dots (QDs) grown on a high index (311)B InP substrate. Important differences were found in the capping process when InP or lattice matched InGaAs(P) alloys are used. The QDs capped with InP have a smaller height due to As/P exchange induced decomposition. This effect is not present when InGaAs is used as the capping material. However, in this case a strong strain driven phase separation appears, creating In rich regions above the QDs and degrading the dot/capping layer interface. If the InAs dots are capped by the quaternary alloy InGaAsP the phase separation is much weaker as compared to capping with InGaAs and well defined interfaces are obtained. © 2006 American Institute of Physics. [DOI: 10.1063/1.2221884]

It has been shown that InAs quantum dots (QDs) grown on InP offer a promising opportunity to apply the functional advantages of zero-dimensional self-assembled nanostructures in optoelectronic devices such as lasers operating in the 1.3–1.5 μm spectral range.^{1,2} In comparison with InAs dot formation on conventional (100) InP substrates, a higher density of dots, having a smaller size dispersion, has been achieved by deposition of InAs layers on high index (311)B InP substrates.^{3,4} Indeed, room temperature lasers with low threshold current density emitting at 1.5 μm were recently demonstrated.⁵

Despite this progress, the effect of the capping material on the structural properties of the QDs is still under investigation. Capping is of crucial importance because processes such as dot decomposition, As/P exchange, and phase separation in the capping layer depend on the capping material and growth procedure. Therefore, in order to achieve the desired emission wavelength from InAs QDs on (311)B InP, a precise understanding and control of the capping procedure are needed. It has been shown that cross-sectional scanning tunneling microscopy (XSTM) can resolve many of these questions in detail.^{6,7} In this work, we analyze at the atomic scale InAs dots on (311)B InP that were capped with either InP, InGaAs, or InGaAsP by XSTM to unravel the effects of capping induced intermixing, segregation, exchange, dot decomposition, and phase separation processes in the capping layer.

The samples were grown by gas source molecular beam epitaxy on a Si doped (311)B oriented InP substrate. The specific details of the growth on such a high index substrate are described elsewhere.⁴ The growth temperature was set to 480 °C. The QDs were formed by depositing 2.1 (100) equivalent monolayers (MLs) of InAs at a growth rate of 0.33 ML/s on InP buffer layers. A low As flux was supplied to the surface during the InAs deposition to enhance the for-

mation of small QDs.⁴ After island formation, a 30 s growth interruption under As flux was performed before the growth of the capping layer. Three QD layers, separated by 40 nm, were grown under the same conditions but capped with different materials: 40 nm of InP in the first layer, 20 nm of lattice matched $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ in the second layer, followed by 20 nm of InP, and lattice matched $\text{In}_{0.87}\text{Ga}_{0.13}\text{As}_{0.285}\text{P}_{0.715}$ in the third one. The XSTM measurements were carried out on the $[0 - 1 1]$ surface plane of *in situ* cleaved samples under UHV ($p < 4 \times 10^{-11}$ Torr) conditions by using polycrystalline tungsten tips prepared by electrochemical etching technique. A large-scale filled state XSTM image of the structure is shown in Fig. 1, in which the three capping layers and InAs QD layers (labeled with A, B, and C) can be observed. The measured distances between dot layers of 35 and 37 nm are in good agreement with the nominal values. The images of the InAs dot layers and the capping layers were obtained in constant current mode during which both the topography and current images were recorded simultaneously.

A number of individual QDs were analyzed within each layer in order to extract information relative to its composi-

FIG. 1. (Color online) Large scale XSTM image of the structure showing the entire layer stack which comprises the InAs dot layers labeled with A, B, and C ($V_{\text{sample}} = -2.5$ V, $I_{\text{tunnel}} = 100$ pA).

^{a)}Electronic mail: c.celebi@tue.nl

FIG. 2. (Color online) (a) XSTM topography image and (b) current image of a single InAs dot and the wetting layer (layer A) capped with InP ($V_{\text{sample}} = -3.0$ V, $I_{\text{tunnel}} = 100$ pA). Both of the images show the low aspect ratio of the dot having a flat top facet. In the current image As atoms can be clearly seen within the P rows of the InP capping layer.

tion, size, and shape. Figure 2(a) shows the high voltage filled state image of a single dot in layer A. Under these conditions the electronic contrast is strongly suppressed and the measurement reflects mainly the topographic contrast, which is due to the outward relaxation of the cleaved surface due to compressive strain.⁸ Atomic details are resolved in this image, in which the group V elements, i.e., As and P, are imaged. From the homogeneity of the contrast it can be deduced that the QD composition is quite uniform and close to 100% InAs except for the lower left and right corners of the dot.

All the observed dots in this layer have a similar truncated pyramidal shape, with a flat top facet. The height and base length distribution of a number of dots showed an average height of 2.9 ± 0.2 nm and a maximum base length of 32 ± 2 nm. The base length was found to vary between 22 and 32 nm. The existence of various base lengths with an approximately constant height suggests that the dots are cleaved in the diagonal direction and not parallel to the sides of the square base.⁸ The maximum measured length of 32 nm [Fig. 2(a)] gives an actual base length of 23 nm and a very low aspect ratio of 0.09. The difference in the outward relaxation of the dots and the wetting layer (WL), as indicated by the brightness in the STM image that reflects the local height of the surface under the STM tip, is quite small [see Fig. 2(a)]. This indicates a high As concentration in the WL. Moreover, the measured magnitude of the outward relaxation of the WL [see Fig. 2(a)] is much higher than that observed in InAs/GaAs WLs.⁷ This is surprising as the compressive strain in the InAs/GaAs case is more than twice that of the InAs/InP system, and thus for equally thick wetting layers in both systems a reduction by a factor of more than 2 in the outward relaxation for the case of InAs/InP should be expected. We explain this by the presence of a very thick WL which contains much more InAs material than the nominal 2.1 ML that were deposited during the dot formation.

The outward relaxation of the WL was calculated by means of the analytical expression derived in Ref. 9 for the outward relaxation of a quantum well, which assumes that the elastic response is linear and isotropic. The WL was modeled including the effect of the asymmetric As profile,

FIG. 3. Distribution profile of As atoms from the InAs WL (layer A). Above a concentration of 25%, atom counting is not reliable and only a lower limit of the real value is obtained. The inset depicts the measured (dotted line) and the calculated (solid line) outward relaxation of the InAs wetting layer based on the As profile that is indicated by the solid line in the main figure.

which is likely created during the switching between phosphorus to arsenic flux or by As carryover.¹⁰ This can be seen in Fig. 2(b) in which the bright spots in the capping layer correspond to As atoms in the InP matrix. Just as in the case of an InAs WL in GaAs, in which there is an asymmetric In profile due to segregation, we used the phenomenological model of Muraki *et al.*¹¹ to model the As profile. The total amount of deposited As is determined by fitting the calculated relaxation profile to the measured one.⁷ The result of this fitting and the corresponding As profile are shown in Fig. 3. We obtained a total amount of InAs of 4 ML. The latter is almost twice the nominal value of deposited InAs. The origin of this extra InAs is the As/P exchange reaction at the InAs/InP interface during the dot formation process. During the growth interrupts used before and after the dot formation process, the structure is kept under an As flux promoting the exchange of P by As and thus increasing the amount of deposited InAs. Such exchange has also been reported to give rise to the formation of InAs QDs and InAs quantum wires on InP surfaces where only a growth interrupt was used under As flux without the additional In deposition.^{12,13} The large amount of InAs in the wetting layer is supported by the distribution of As in the WL obtained by directly counting the As atoms in the XSTM images. In Fig. 3 we plot the number of counted As atoms as a function of the distance in the growth direction. This method is accurate for As concentrations lower than about 25%, because above those values it becomes complicated to distinguish individual As atoms in InP. The profile based on the counting is shown in Fig. 3 together with the profile that was used to fit the outward relaxation. The agreement is quite good in the range of validity of the counting method.

The small height of the dots capped with InP suggests As/P exchange during the capping process. This effect should be eliminated by using InGaAs as the capping material. This alloy is frequently used to cap InAs QDs grown on GaAs substrate, where it acts as a strain reducing layer.¹⁴ In our case, lattice matched $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ was used in the second QD layer. The height and base length distribution of a number of single dots were again investigated, giving an average height of 3.5 ± 0.2 nm and a maximum length of 29 ± 2 nm, corresponding to a square base of 20 ± 2 nm side.

FIG. 4. (Color online) (a) XSTM topography image of two adjacent InAs dots (in layer B) capped with InGaAs ($V_{\text{sample}} = -3.0$ V, $I_{\text{tunnel}} = 100$ pA). The image reveals the presence of a strain induced lateral phase separation in the InGaAs capping layer. (b) High-pass filtered XSTM image of another set of InAs dots in the same layer showing a more rounded top facet (lens shaped) and higher aspect ratio.

These dots are in average 0.6 nm higher than those capped with InP, which corresponds to ~ 2 ML of InAs. This indicates that the height of the dots was reduced by ~ 2 ML due to dot decomposition induced by As/P exchange during InP capping. Nevertheless, the top facet of the dots capped with InGaAs is less well defined and is more curved than that of the dots capped with InP [compare Figs. 2 and 4]. This is likely due to phase separation in the capping layer, which we analyze next.

The inhomogeneous topographic contrast in the InGaAs layer [Fig. 4(a)] reveals the presence of an inhomogeneous strain distribution, which must be due to the presence of In rich (brighter) and Ga rich (darker) regions. This phase separation is a strain driven process in which the In adatoms on the growth surface migrate towards the regions on top of the dots to minimize the strain, creating a columnarlike In rich region above the dots. This process has been observed in columnar InGaAs QDs grown on GaAs,¹⁵ as well as in InAs/GaAs QDs, where capping with InGaAs has been shown to induce an increase of the dot size.¹⁶ In our case, the phase separation directly affects the QDs by creating a rough top interface in which the In content decreases gradually. InGaAs seems therefore to be less suitable capping material for InAs QDs grown on InP (311)B substrate.

In the third layer, a lattice matched InGaAsP alloy was used as the capping material (Fig. 5). The average dot height was 3.4 ± 0.2 nm, and the maximum measured base length was 27 ± 2 nm (corresponding to a 19 ± 2 nm side square base). As in the case of InGaAs, the height of the dots is higher as those capped with InP where the dot partially dis-

FIG. 5. (Color online) XSTM image of two InAs dots (in layer C) capped with InGaAsP alloy showing a flat base and top ($V_{\text{sample}} = -2.5$ V, $I_{\text{tunnel}} = 100$ pA).

solved due to the As/P exchange. The shape resembles that of a truncated pyramid, with a flat top interface. Remarkably, the phase separation in the capping layer is much weaker than in the InGaAs. We think that the Ga–P bond strength (54.9 kcal/mol) which is stronger than the In–As bond strength (48.0 kcal/mol) and Ga–As bond strength (50.1 kcal/mol) limits the phase segregation in InGaAsP as compared to InGaAs. The resulting weak phase separation does not affect the dot shape or size, as is evidenced by the well defined dot facets. Indeed, this material has been successfully used as capping layer in InAs/InP (311)B QD lasers emitting near $1.55 \mu\text{m}$ with low threshold current density.⁵

In summary, XSTM has been used to analyze at the atomic scale the effect of the capping material on the structural properties of self-assembled InAs/InP (311)B QDs. The As/P exchange on the InAs/InP interface during the growth interrupts is shown to increase the amount of InAs in the wetting layers by ~ 2 ML. The As/P exchange takes place also on the dot surface when the QDs are capped with InP, reducing the dot height by about 2 ML. This phenomenon can be avoided by using InGaAs as the capping material, but in that case a strong strain driven phase separation appears, giving rise to In rich regions above the dots and a degradation of the dot interface. If the quaternary alloy InGaAsP is used instead of InGaAs, the phase separation is much weaker and well defined interfaces are obtained.

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