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Citation for published version (APA):

DOI:
10.1063/1.2722738

Document status and date:
Published: 01/01/2007

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
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Download date: 03. Apr. 2019
Atomic scale study of the impact of the strain and composition of the capping layer on the formation of InAs quantum dots*

J. M. Ulloa,1 C. Çelebi, and P. M. Koenraad
COBRA Inter-University Research Institute, Eindhoven University of Technology, P.O. Box 513, NL-5600MB Eindhoven, The Netherlands
A. Simon, E. Gapihan, A. Letoublon, and N. Bertru
Laboratoire d'études des Nanostructures Semiconductrices (LENS), INSA-Rennes, F-35043 Rennes Cedex, France
I. Drouzas and D. J. Mowbray
Department of Physics and Astronomy, University of Sheffield, Sheffield S3 7RH, United Kingdom
M. J. Steer and M. Hopkinson
Department of Electronic and Electrical Engineering, University of Sheffield, Sheffield S1 3JD, United Kingdom

(Received 14 September 2006; accepted 27 December 2006; published online 27 April 2007)

The impact of the capping material on the structural properties of self-assembled InAs quantum dots (QDs) was studied at the atomic scale by cross-sectional scanning tunneling microscopy. Capping with lattice matched layers and with strained layers was analyzed. When the different capping materials are lattice matched to the substrate, the differences in the QD properties can be dominated by chemical effects: InAs/InP QDs capped with InP have a 2 ML smaller height than those capped with InGaAs or InGaAsP due to As/P exchange induced decomposition. The height of the dots is found to be much more strongly affected when strained capping layers are used. InAs/GaAs, QDs capped with InGaAs are considerably taller than typical GaAs-capped dots. When GaAsSb is used as the capping layer, the dots are almost full pyramids with a height of 9.5 nm, indicating that dot decomposition is almost completely suppressed. This indicates that the dot/capping layer strain plays a major role in inducing dot decomposition during capping. © 2007 American Institute of Physics. [DOI: 10.1063/1.2722738]

I. INTRODUCTION

Self-assembled InAs quantum dots (QDs) have attracted much attention in the last years due to promising applications such as QD lasers,1,2 single electron transistors,3 etc. InAs QDs are commonly created by Stranski–Krstanov growth mode when InAs is deposited on a substrate with a bigger lattice constant, like GaAs or InP.4 Once created, the QDs are subsequently capped, a step which is required for any device application. Although a lot of effort has been dedicated to understand the QD growth mechanism, there are relatively few studies focused on the effect of the capping process.5–13 Some of these studies have already shown significant differences in size, shape, and composition between uncapped and capped QDs. For example, an important collapse of the QD height has been reported for InAs/GaAs QDs capped with GaAs,7,8,10–12 revealing the big influence of the capping process on the structural properties of the QDs. The existing structural studies of buried InAs/GaAs QDs have been mainly devoted to GaAs-capped QDs. Nevertheless, different materials such as InGaAs and GaAsSb are nowadays used to cap InAs/GaAs QDs in an effort to extend its emission wavelength to the technologically interesting 1.3–1.55 μm region.14–19 For InAs/InP QDs, capping materials other than InP, like InGaAsP, have also successfully been used for laser applications.20–22

The use of different capping materials strongly affects the emission wavelength and therefore should strongly affect the QD electronic and/or structural properties, such as size, shape, composition, strain, band-offsets, etc. Indeed, critical issues occurring during capping like dot decomposition, intermixing, segregation, As/P exchange, and phase separation in the capping layer depend on the capping material. To understand the impact of the capping material on the structural properties of the QDs is consequently of crucial importance. Cross-sectional scanning tunneling microscopy (XSTM) is a suitable technique for this purpose, because it allows to assess the structure of the capped dots at the atomic scale.

In this work, we have used XSTM to analyze at the atomic scale how capping with different materials influence the structural properties of InAs QDs in GaAs and InP. The role of the different effects occurring during capping (intermixing, segregation, As/P exchange, compositional modulation in the capping layer, etc.) is determined. First we study the capping with lattice matched layers (with respect to the substrate), where the chemical effects could dominate the process. For that we study InAs dots grown on (311)B InP

*This paper is based on a talk presented by the authors at the 28th International Conference on the Physics of Semiconductors, which was held 24–28 July 2006, in Vienna, Austria. Contributed papers for that conference may be found in "Physics of Semiconductors: 28th International Conference on the Physics of Semiconductors," AIP Conference Proceedings No. 893 (AIP, Melville, NY, 2007); see http://proceedings.aip.org/proceedings/confproceed/893.jsp.

1Electronic mail: j.mulloa@tue.nl
II. EXPERIMENTAL DETAILS

The samples used in the study of the lattice matched capping (Sec. III A) were grown by gas source molecular beam epitaxy (MBE) on a Si doped (311)B oriented InP substrate. This substrate orientation is very attractive for laser applications because, in comparison with InAs dot formation on conventional (100) InP substrates, a higher density of dots having a smaller size dispersion has been achieved on (311)B InP substrates.\(^{20,21}\) Indeed, room temperature lasers with low threshold current density emitting at 1.5 \(\mu\)m were recently demonstrated.\(^{22}\) In the analyzed sample the growth temperature was set to 480 °C. The QDs were formed by depositing 2.1 (100) equivalent monolayers (ML) of InAs at 0.33 ML/s on InP buffer layers. A low As flux was supplied to the surface during the InAs deposition to enhance the formation of small QDs.\(^{21}\) 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 \(\text{In}_{0.85}\text{Ga}_{0.15}\text{As}\) capped with \(\text{In}_{0.13}\text{As}_{0.87}\text{P}_{0.15}\) in the third one. The growth rates were 1.4, 0.6, and 1.1 ML/s, respectively.

The samples used in the study of the capping by strained layers (Sec. III B) were grown by solid source MBE on a \(n^+\) Si doped (100) GaAs substrate. In the two analyzed samples, 2.7 ML of InAs were deposited at 500 °C and 0.1 ML/s on an intrinsic GaAs buffer layer. In the first one, the QD layer was capped with a 6 nm thick \(\text{In}_{0.15}\text{Ga}_{0.85}\text{As}\) layer grown at 500 °C, while in the second a 6 nm \(\text{GaAs}_{0.75}\text{Sb}_{0.25}\) layer grown at 475 °C was used. The growth rate was 0.5 ML/s in both cases.

The XSTM measurements were carried out at room temperature on the [110] or [1–10] surface plane of \textit{in situ} cleaved samples under ultrahigh vacuum (\(p < 4 \times 10^{-11}\) Torr) conditions. Polycrystalline tungsten tips prepared by electrochemical etching were used. The images were obtained in constant current mode during which both the topography and current images were recorded simultaneously. All the images shown in this article were taken at high voltage (\(-3\) V). 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.\(^{23}\)

III. RESULTS

A. Capping with lattice matched layers (InAs/InP QDs)

A large-scale filled states XSTM image of the studied structure (described in Sec. II) is shown in Fig. 1, in which the three QD and capping layers can be observed. The QD layers capped with InP, InGaAs, and InGaAsP are labeled A, B, and C, respectively.

A number of individual QDs were analyzed within each layer in order to extract information relative to its composition, size, and shape. Figure 2 shows the high voltage filled states image of a single dot in layer A. 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.

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 difference in the outward relaxation of the dots and the wetting layer (WL), as indicated by the bright-

![Image](https://example.com/image.png)

**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). Growth direction: [113], lateral direction: [1–10].

![Image](https://example.com/image.png)

**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. Growth direction: [113], lateral direction: [1–10].
posited InAs. Such an exchange has also been reported to
exchange of P by As and thus increasing the amount of de-
InP interface during the dot formation process. During the
of this extra InAs is the As/P exchange reaction at the InAs/
almost twice the nominal value of deposited InAs. The origin
3. We obtained a total amount of InAs of 4 ML. The latter is
this fitting and the corresponding As profile are shown in Fig.
phosphorus to arsenic flux or by As carryover.26 This can be seen
which is likely created during the switching between phos-
model of Muraki
profile due to segregation, we used the phenomenological
of an InAs WL in GaAs, in which there is an asymmetric In
ness 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 is
much higher than that observed in InAs/GaAs WLs.24 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 pres-
ence 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. 25 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,
which is likely created during the switching between phosphorus to arsenic flux or by As carryover.26 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.27 to model the As profile. The total
amount of deposited As is determined by fitting the calculated
relaxation profile to the measured one.24 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 de-
positioned InAs. Such an 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 an As flux without the additional In
deposition.28,29 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 effect of the As/P exchange reaction would be stron-
ger if the growth interrupt time after dot formation would be
longer, or if the As flux would be higher. A sample with a
similar QD layer to the previously analyzed one (layer A) but
in which the As flux was increased was also studied. Figure
4 shows a filled states image of a dot in this sample. The
effect of the As/P exchange is stronger now, as evidenced by
the fact that the InAs WL is going deeper into the InP sub-
strate. The dots are protecting the InP from the As flux and
the result is that their base is not aligned with the base of the
WL. This is a good example of how a chemical effect like
As/P exchange can modify the grown structure.

The small height of the dots capped with InP suggests an
As/P exchange during the capping process. This effect
should be eliminated by using InGaAs as the capping mate-
rial and therefore lattice matched In_{0.53}Ga_{0.47}As was used in
the second QD layer. Two dots in this layer are shown in Fig.
5. The height and base length distribution of a number of
single dots was again investigated, giving an average height of 3.5 ± 0.2 nm and a maximum length of 29 ± 2 nm. 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. This value is in a good agreement with the 2 ML reduction of the InGaAsP capping layer reported in Ref. 30. The observed difference cannot be due to the different growth rates. It has been shown that the effect of the growth rate on the height reduction due to an As/P exchange in the capping layer is now 0.56, about 17% smaller than in the InAs/GaAs case in which GaAs capping is used. This small difference has been successfully used as capping layer in InAs/GaAs (311)B QD lasers emitting near 1.55 μm with low threshold current density.\(^\text{32}\)

B. Capping with strained layers (InAs/GaAs QDs)

As mention in Sec. I, previous studies have already shown a significant reduction of the InGaAs dot height after capping\(^\text{7,8,10-12}\) with GaAs, the most commonly used capping material. The amount of strain between the partially relaxed dot and the capping layer could play an important role in this process. It has already been reported that QDs capped with InGaAs instead of GaAs tend to retain their shape during initial stages of capping.\(^\text{34,35}\) Nevertheless, those studies were performed using plan-view STM and atomic force microscopy, so the final buried dots could not be observed. To study the role of strain, we used XSTM to analyze samples in which the dots were capped with InGaAs and GaAsSb strain reducing layers (described in Sec. II).

Figure 7 shows a high voltage filled states image of an InAs/GaAs QD capped with InGaAs. The dot is 7.0 ± 0.5 nm high, considerably higher than the typical GaAs-capped QDs (around 4 nm high).\(^\text{8,11,12}\) The In composition in the capping layer was deduced by the analysis of the outward relaxation. The measured outward relaxation was compared to the one calculated using continuum elasticity theory. A finite element calculation is performed to solve the three-dimensional problem, in which an isotropic material is considered. The fit shown in Fig. 8 was found when a 8.5 nm thick layer (as measured from the XSTM images) with 17% In content is considered in the calculation. The In content is close to the nominal value (15%). The lattice mismatch between the top of the dot (considering that it is completely relaxed) and the capping layer is now 0.056, about 17% smaller than in the case in which GaAs capping is used. This small difference seems to be enough to reduce considerably the dot decomposition during capping and the result is that the dot height is increased by almost a factor of 2. It could be argued that the increase in dot height is due to strain induced compositional modulation in the capping layer,\(^\text{33}\) as we mentioned in Sec. II. Although part of the increase in dot height could be due to that effect, we think that its contribution should be very small, since no traces of compositional modulation are observed in the capping layer. The role of the strain can be
The fit shown in Fig. 10(a) is obtained for a 25% Sb content. The fit is quite good in the region of the layer itself, but deviates above the layer. This is likely due to the effect of the Sb segregation, which is not included in the model but is clearly present in the structure, as can be seen in Fig. 10(b). The lattice mismatch between the top of the dot (considering that it is completely relaxed) and the capping layer is now 0.048, smaller than for the GaAs-capped QDs analyzed before. That is probably the reason why the GaAsSb-capped QDs are bigger. This seems to indicate that strain plays a major role in inducing dot decomposition during capping and that decreasing the dot/capping layer lattice mismatch can strongly reduce dot decomposition. The fact that InAs/GaAs QDs capped with InGaAs, and especially with GaAsSb, are bigger than those capped with GaAs is a main factor to explain the observed increase of the emission wavelength.

IV. CONCLUSIONS

XSTM has been used to analyze at the atomic scale the effect of the capping material on the structural properties of self-assembled InAs QDs. Capping with lattice matched layers was studied by analyzing InAs/InP (311)B QDs capped with InP, InGaAs, and InGaAsP. 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. 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. The impact of the capping material was found to be much stronger when it implies a change in strain. Capping with strained layers was studied using InAs/GaAs QDs capped with InGaAs and GaAsSb. In both cases the QDs are bigger than the typical GaAs-capped QDs. In the case of GaAsSb-capped QDs, in which the lattice mismatch between the dot and the capping layer is the smallest, dot decomposition is...
almost completely suppressed. This indicates that the dot/capping layer strain plays a major role in inducing dot decomposition during capping.