

Suppression of InAs/GaAs quantum dot decomposition by the incorporation of a GaAsSb capping layer

Citation for published version (APA):

Ulloa Herrero, J. M., Drouzas, I. W. D., Koenraad, P. M., Mowbray, D. J., Steer, M. J., Liu, H. Y., & Hopkinson, M. (2007). Suppression of InAs/GaAs quantum dot decomposition by the incorporation of a GaAsSb capping layer. *Applied Physics Letters*, 90(21), 213105-1/3. Article 213105. <https://doi.org/10.1063/1.2741608>

DOI:

[10.1063/1.2741608](https://doi.org/10.1063/1.2741608)

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:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
- The final author version and the galley proof are versions of the publication after peer review.
- The final published version features the final layout of the paper including the volume, issue and page numbers.

[Link to publication](#)

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license above, please follow below link for the End User Agreement:

www.tue.nl/taverne

Take down policy

If you believe that this document breaches copyright please contact us at:

openaccess@tue.nl

providing details and we will investigate your claim.

Suppression of InAs/GaAs quantum dot decomposition by the incorporation of a GaAsSb capping layer

J. M. Ulloa,^{a)} I. W. D. Drouzas,^{b)} and P. M. Koenraad

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

D. J. Mowbray

Department of Physics and Astronomy, University of Sheffield, S3 7RH Sheffield, United Kingdom

M. J. Steer, H. Y. Liu, and M. Hopkinson

Department of Electronic and Electrical Engineering, University of Sheffield, S1 3JD Sheffield, United Kingdom

(Received 1 March 2007; accepted 26 April 2007; published online 23 May 2007)

The influence of a GaAsSb capping layer on the structural properties of self-assembled InAs/GaAs quantum dots (QDs) is studied on the atomic scale by cross-sectional scanning tunneling microscopy. QDs capped with GaAs_{0.75}Sb_{0.25} exhibit a full pyramidal shape and a height more than twice that of the typical GaAs-capped QDs, indicating that capping with GaAsSb suppresses dot decomposition. This behavior is most likely related to the reduced lattice mismatch between the dot and the capping layer. © 2007 American Institute of Physics. [DOI: 10.1063/1.2741608]

A significant effort has been made in the last few years to extend the emission wavelength of InAs/GaAs quantum dots (QDs) to the 1.55 μm telecommunication band. Room temperature photoluminescence emission beyond 1.5 μm has been achieved by a number of different approaches.¹⁻⁸ Recently, GaAsSb capping layers have also been used to extend the emission wavelength^{9,10} and room temperature photoluminescence at 1.6 μm has recently been reported from GaAsSb-capped In(Ga)As/GaAs QDs.^{11,12} The strong redshift observed when using GaAsSb instead of GaAs capping layers has been attributed to a type-II band alignment.^{11,12} However, the structural properties of these QDs have not been studied, despite the fact that they could be significantly different from those of GaAs-capped QDs. Because of the larger lattice constant of GaAsSb compared to GaAs, the strain induced in the dot during capping should be reduced, which would further redshift the emission wavelength. In addition, the modified strain difference could induce differences in dot size, shape, and composition, since dot decomposition during capping can be influenced by the local strain. In this work, we have used cross-sectional scanning tunneling microscopy (X-STM) to analyze, at the atomic scale, how capping with GaAsSb influences the structural properties of InAs/GaAs QDs. The QD size, shape, and composition are determined and are compared to the case of GaAs capping.

The samples were grown by solid source molecular beam epitaxy on n^+ Si doped (100) GaAs substrates. In the first sample (sample A), 2.7 ML of InAs were deposited at 500 °C and 0.1 ML/s on an intrinsic GaAs buffer layer. The QDs were capped with a nominal 6 nm thick GaAs_{0.75}Sb_{0.25} layer grown at 475 °C. In the second sample (sample B), two QD layers were grown under the same conditions (2.7 ML of InAs deposited at 500 °C and 0.1 ML/s). The first layer was capped with GaAs, and the second with 6 nm GaAs_{0.75}Sb_{0.25}. The capping temperature was 500 °C in both

cases. A layer of uncapped QDs were also grown on the sample surface for atomic force microscopy measurements.

The X-STM measurements were performed at room temperature on the [1 1 0] surface plane of *in situ* cleaved samples under UHV ($p < 4 \times 10^{-11}$ Torr) conditions. Polycrystalline tungsten tips prepared by electrochemical etching were used. The images were obtained in constant current mode during which both topography and current images were recorded simultaneously. All the images shown in this letter were recorded for high voltage (~ 3 V). Under these conditions the electronic contrast is strongly suppressed and the measurements reflect mainly the topographic contrast, which is due to the outward relaxation of the cleaved surface due to the compressive strain.¹³

A number of individual QDs were analyzed in sample A in order to extract information concerning their size, shape, and composition. Figure 1 shows a high voltage filled states topography image of a QD from this sample. The measurement conditions image group V elements so that the bright spots represent Sb atoms in the As matrix. Sb segregation into the GaAs layer is clearly observed in this image. In contrast to the behavior when capping with GaAs at similar temperatures,¹⁴ the capping layer completely covers the dot.

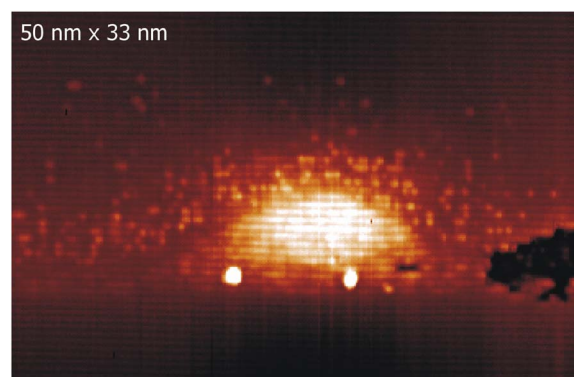


FIG. 1. (Color online) Filled state topography image of an InAs QD capped with GaAsSb ($V = -3$ V). The bright spots are Sb atoms in the As matrix. The white circles and the dark feature are cleavage induced defects.

^{a)}Electronic mail: j.m.ulloa@tue.nl

^{b)}Permanent address: Department of Physics and Astronomy, University of Sheffield, United Kingdom.

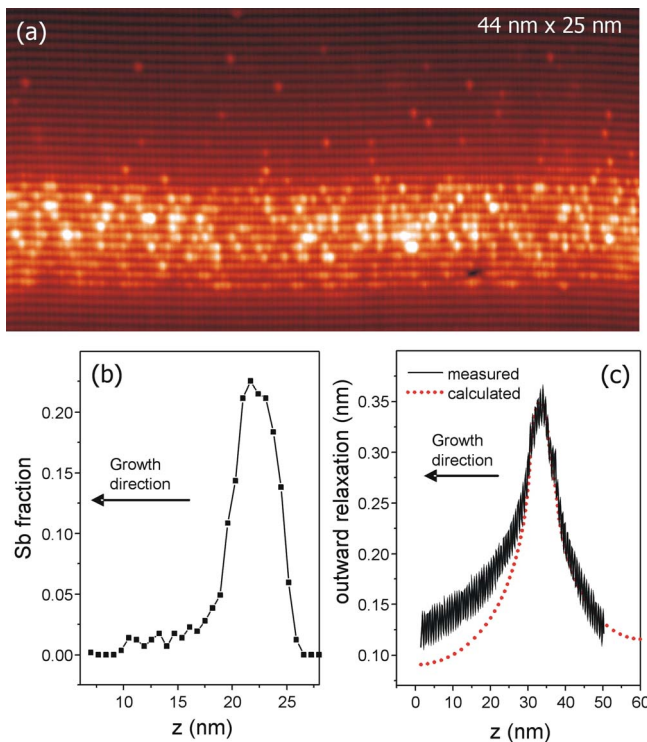


FIG. 2. (Color online) (a) Topography image of the GaAsSb capping layer ($V=-3$ V). The bright spots are Sb atoms in the As matrix. Sb segregation into the GaAs layer is clearly observed. (b) Sb concentration profile in the capping layer obtained by counting individual Sb atoms. (c) Measured (solid line) and calculated (dotted line) outward relaxation profiles of the GaAsSb capping layer. A 5 nm thick layer [as measured from (a)] with a 25% Sb composition was used in the calculation.

This behavior cannot be attributed to a larger bond strength preventing Ga migration on the growth surface, since the Ga–Sb bond is weaker than that of Ga–As (45.9 and 50.1 kcal/mol, respectively). The most likely explanation is the smaller strain that exists between the partially relaxed InAs QDs and the GaAsSb capping layer. This smaller strain can be accommodated by the system at the present growth temperature (475 °C) without inducing the migration of capping material away from the top of the dot. The dot is a full pyramid with a diagonal base length of 32 ± 2 nm. The height is 9.5 ± 0.2 nm, much larger than that of typical GaAs-capped QDs (around 4 nm).^{15–17} This result indicates that the QDs are not dissolved during capping with GaAsSb.

The Sb composition in the capping layer can be directly obtained by counting the individual Sb atoms in filled state images, for example, Fig. 2(a). The Sb concentration profile as a function of distance along the growth direction is plotted in Fig. 2(b). The average Sb content is 22%. This result can be checked by analyzing the outward relaxation of the cleaved surface. The relaxation of the capping layer was compared to calculations from continuum elasticity theory. A finite element calculation was performed to solve the three-dimensional problem, in which an isotropic material is considered. The fit shown in Fig. 2(c) was obtained for a 5 nm thick layer (as measured from the X-STM images) with a 25% Sb composition. The agreement between theory and experiment is quite good in the region of the layer itself, but deviates above the layer. This deviation is due to the effect of Sb segregation, which is not included in the model but is clearly present in the structure [see Fig. 2(a) and the asymmetry in the Sb profile obtained from atom counting]. The measured Sb composition of 22%–25% agrees well with

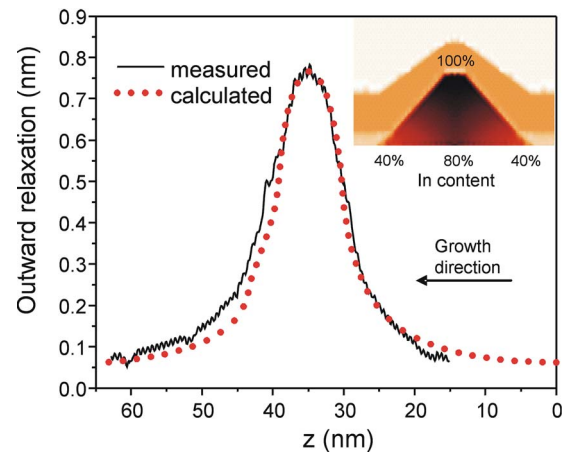


FIG. 3. (Color online) Measured (solid line) and calculated (dotted line) outward relaxation profiles through the center of a GaAsSb-capped QD. The inset shows the QD In distribution used in the calculation.

the nominal growth value. The lattice mismatch between this GaAsSb composition and relaxed InAs is 0.048, about 28% smaller than when GaAs capping is used. We propose that the reason why dot decomposition during capping is suppressed is due to the reduced strain between the dot and the capping layer, although chemical effects due to the presence of Sb could also be relevant. This interpretation is in agreement with previously reported results in which QDs capped with InGaAs instead of GaAs are shown to retain their shape during the initial stages of capping.^{18,19}

The composition of the QDs can also be estimated from an analysis of the outward relaxation.¹³ The size and shape of the dots obtained from the images are used as input parameters for the model and the composition is changed until the calculated outward relaxation profile agrees with the experimentally determined one. We assume that the dot with the largest observed base length is cleaved through its middle and that the dots have a square base with edges along the $\langle 100 \rangle$ directions. The largest measured base length was 32 nm (see Fig. 1) which corresponds to sides of length 23 nm. Figure 3 shows the best fit to the outward relaxation profile through the middle of the dot, obtained when a trumpetlike shape QD In composition is included in the model. The fit deviates in the region above the dot, probably due to the fact that Sb segregation is not included in the calculation. The In content increases from 80% at the bottom to 100% at the top of the QD and from 40% in the corners to 80% at the center of the base (see inset of Fig. 3). A similar nonuniform In composition has been previously proposed in Ref. 20 and deduced from X-STM measurements in Ref. 21 for GaAs capped InAs QDs, and is attributed to In–Ga intermixing. The presence of In-deficient corners in the present GaAsSb-capped QDs indicates that they are not necessarily created by a redistribution of material from the top of the dots to the bottom during capping, as has been recently reported.²² In the present case, the top of the dot is not dissolved during capping but the In-poor corners are still present, hence they must originate during an earlier stage of the QD formation process.

In order to confirm the observed differences between QDs capped with GaAs and GaAsSb, sample B was studied. This sample contains QDs capped with both GaAs and GaAsSb, with both capping layers grown at the same temperature as the InAs dots. From an analysis of the measured outward surface relaxation, the Sb composition of the

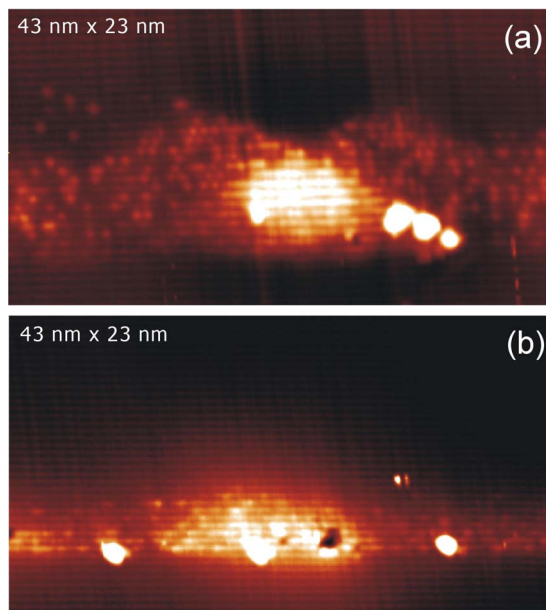


FIG. 4. (Color online) Filled state topography images of two QDs capped with (a) GaAsSb and (b) GaAs ($V = -3$ V). The white circles and the dark features are cleavage induced defects.

GaAsSb layer was found to be 24%. However, the properties are different to those of sample A because the GaAsSb capping layer does not completely cover the dots, with their apex remaining uncovered [see Fig. 4(a)]. As the only difference between samples B and A is that the capping temperature was 25 °C higher in sample B, it can be concluded that this increase in temperature is sufficient to allow the capping material to migrate from the relaxed apex of the dot, minimizing the strain.¹⁴ This observation reveals that the final dot/GaAsSb capping layer configuration is very sensitive to the temperature during the capping process. Despite changes to the capping process, significant differences in the QD structure for GaAs and GaAsSb capping remain in sample B (Fig. 4). The GaAsSb-capped QDs [Fig. 4(a)] are again full pyramids, with a height of 8.3 ± 0.2 nm, while the GaAs-capped QDs [Fig. 4(b)] are truncated pyramids, with a height of only 3.8 ± 0.2 nm. This result indicates that QDs capped with GaAs are significantly dissolved during capping, while the shape of QDs capped with GaAsSb ($\sim 25\%$ Sb) at the same temperature is preserved. This result is confirmed by atomic force microscopy measurements on uncapped surface QDs, which are found to have a height of 8 ± 1 nm, in good agreement with the height of the GaAsSb-capped QDs. The base length of the strongly dissolved GaAs-capped QDs is smaller than that of the preserved GaAsSb-capped dots (20 vs 26 nm, respectively), indicating that material redistribution during capping is not from the apex to the base of the dot,²² but occurs instead to the wetting layer (WL). This is in agreement with the observation of a significantly higher In content in the WL of GaAs-capped QDs (1.8 ML vs 0.8 ML in GaAsSb-capped QDs), obtained by counting individual In atoms in X-STM empty states images.

The present results indicate that strain may be playing an important role in inducing dot decomposition during capping, and that decreasing the dot/capping layer lattice mismatch appears to strongly reduce dot decomposition. The increased height of GaAsSb-capped QDs, as revealed by the present work, is expected to contribute to their long wavelength emission, in addition to the contribution from the for-

mation of a type-II system.¹² However, it is difficult to comment on the relative importance of these two contributions as the QD height dependence on GaAsSb composition is not known. This dependence will form the subject of future work.

In conclusion, X-STM has been used to study at the atomic scale the effect of a GaAsSb capping layer on the structural properties of self-assembled InAs QDs. QDs capped with a GaAsSb layer with $\sim 25\%$ Sb are much larger than typical GaAs-capped QDs. GaAsSb-capped QDs exhibit a full pyramidal shape of 8.3–9.5 nm height, while similar dots capped with GaAs are truncated pyramids with a height of only 3.8 nm. This finding indicates that dot decomposition during capping is suppressed by using GaAs_{0.75}Sb_{0.25}. This behavior is most likely related to the reduced dot/capping layer lattice mismatch, although chemical effects due to the presence of Sb could also be relevant.

This work has been supported by the European Union through the SANDiE Network of Excellence (Contract No. NMP4-CT-2004-500101).

- ¹M. V. Maximov, A. F. Tsatsul'nikov, B. V. Volovik, D. A. Bedarev, A. Yu. Egorov, A. E. Zhukov, A. R. Kovsh, N. A. Bert, V. M. Ustinov, P. S. Kop'ev, Zh. I. Alferov, N. N. Ledentsov, D. Bimberg, I. P. Soshnikov, and P. Werner, *Appl. Phys. Lett.* **75**, 2347 (1999).
- ²M. J. da Silva, A. A. Quivy, S. Martini, T. E. Lamas, E. C. F. da Silva, and J. R. Leite, *Appl. Phys. Lett.* **82**, 2646 (2003).
- ³Y. C. Xin, L. G. Vaughn, L. R. Dawson, A. Stintz, Y. Lin, L. F. Lester, and D. L. Huffaker, *J. Appl. Phys.* **94**, 2133 (2003).
- ⁴N. N. Ledentsov, A. R. Kovsh, A. E. Zhukov, N. A. Maleev, S. S. Mikhlin, A. P. Vasil'ev, E. S. Semenova, M. V. Maximov, Yu. M. Shernyakov, N. V. Kryzhanovskaya, V. M. Ustinov, and D. Bimberg, *Electron. Lett.* **39**, 1126 (2003).
- ⁵G. Balakrishnan, S. Huang, T. J. Rotter, A. Stintz, L. R. Dawson, K. J. Malloy, H. Xu, and D. L. Huffaker, *Appl. Phys. Lett.* **84**, 2058 (2004).
- ⁶E. C. Le Ru, P. Howe, T. S. Jones, and R. Murray, *Phys. Status Solidi C* **0**, 1221 (2003).
- ⁷T. Tatebayashi, M. Nishioka, and Y. Arakawa, *Appl. Phys. Lett.* **78**, 3469 (2001).
- ⁸W. H. Chang, H. Y. Chen, H. S. Chang, W. Y. Chen, T. M. Hsu, T. P. Hsieh, J. I. Chyl, and N. T. Yeh, *Appl. Phys. Lett.* **86**, 131917 (2005).
- ⁹K. Akahane, N. Yamamoto, and N. Ohtani, *Physica E (Amsterdam)* **21**, 295 (2004).
- ¹⁰H. Y. Liu, M. J. Steer, T. J. Badcock, D. J. Mowbray, M. S. Skolnick, P. Navaretti, K. M. Groom, M. Hopkinson, and R. A. Hogg, *Appl. Phys. Lett.* **86**, 143108 (2005).
- ¹¹J. M. Ripalda, D. Granados, Y. González, A. M. Sánchez, S. I. Molina, and J. M. García, *Appl. Phys. Lett.* **87**, 202108 (2005).
- ¹²H. Y. Liu, M. J. Steer, T. J. Badcock, D. J. Mowbray, M. S. Skolnick, F. Suarez, J. S. Ng, M. Hopkinson, and J. P. R. David, *J. Appl. Phys.* **99**, 046104 (2006).
- ¹³D. M. Bruls, J. W. A. M. Vugs, P. M. Koenraad, H. W. M. Salemink, J. H. Wolter, M. Hopkinson, M. S. Skolnick, F. Long, and S. P. A. Gill, *Appl. Phys. Lett.* **81**, 1708 (2002).
- ¹⁴Q. Xie, P. Chen, and A. Madhukar, *Appl. Phys. Lett.* **65**, 2051 (1994).
- ¹⁵G. D. Lian, J. Yuan, L. M. Brown, G. H. Kim, and D. A. Ritchie, *Appl. Phys. Lett.* **73**, 49 (1998).
- ¹⁶P. B. Joyce, T. J. Krzyzewski, P. H. Steans, G. R. Bell, J. H. Neave, and T. S. Jones, *Surf. Sci.* **492**, 345 (2001).
- ¹⁷Q. Gong, P. Offermans, R. Nötzel, P. M. Koenraad, and J. H. Wolter, *Appl. Phys. Lett.* **85**, 5697 (2004).
- ¹⁸R. Songmuang, S. Kiravittaya, and O. G. Schmidt, *J. Cryst. Growth* **249**, 416 (2003).
- ¹⁹W. M. McGee, T. J. Krzyzewski, and T. S. Jones, *J. Appl. Phys.* **99**, 043505 (2006).
- ²⁰M. A. Migliorato, A. G. Cullis, M. Fearn, and J. H. Jefferson, *Phys. Rev. B* **65**, 115316 (2002).
- ²¹P. Offermans, P. M. Koenraad, J. H. Wolter, K. Pierz, M. Roy, and P. A. Maksym, *Phys. Rev. B* **72**, 165332 (2005).
- ²²G. Costantini, A. Rastelli, C. Manzano, P. Acosta-Díaz, R. Songmuang, G. Katsaros, O. G. Schmidt, and K. Kern, *Phys. Rev. Lett.* **96**, 226106 (2006).