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Capping process of InAs/GaAs quantum dots studied by cross-sectional scanning tunneling microscopy

Q. Gong,^{a)} P. Offermans, R. Nötzel, P. M. Koenraad, and J. H. Wolter
*eiTT/COBRA Inter-University Research Institute, Eindhoven University of Technology,
 5600 MB Eindhoven, The Netherlands*

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The capping process of self-assembled InAs quantum dots (QDs) grown on GaAs(100) substrates by molecular-beam epitaxy is studied by cross-sectional scanning tunneling microscopy. GaAs capping at 500 °C causes leveling of the QDs which is completely suppressed by decreasing the growth temperature to 300 °C. At elevated temperature the QD leveling is driven in the initial stage of the GaAs capping process while it is quenched during continued overgrowth when the QDs become buried. For common GaAs growth rates, both phenomena take place on a similar time scale. Therefore, the size and shape of buried InAs QDs are determined by a delicate interplay between driving and quenching of the QD leveling during capping which is controlled by the GaAs growth rate and growth temperature. © 2004 American Institute of Physics. [DOI: 10.1063/1.1831564]

Self-assembled quantum dots (QDs) formed in the Stranski–Krastanov growth mode attract great efforts due to their enormous potential for basic physics studies and device applications. For most cases, the QDs are embedded in a semiconductor matrix, i.e., they are capped subsequent to their formation. Both the formation and the capping process are crucial for the structural and electronic properties of the QDs. In particular the capping process may induce drastic changes in size and shape of the QDs, as has been observed in the Ge/Si(100)¹ and InAs/GaAs(100)^{2–4} material systems. Leveling of InAs/GaAs QDs after deposition of thin GaAs cap layers has been clearly revealed by top-view scanning tunneling microscopy (STM)^{2,3} and atomic force microscopy.⁴ The leveling process has been attributed to the additional strain build up between the cap layer and the partially relaxed InAs QDs^{2,4} to destabilize the QDs. These experiments, however, lack information about the shape and, in particular, the residual height of the QDs, which is the most important parameter determining the electronic properties. In this letter, we study the capping process of InAs QDs on GaAs(100) by cross-sectional STM (X-STM). Detailed and accurate results of the QD leveling are presented, which are essential for understanding the capping process and the control of the structural and electronic properties of InAs QDs.

The samples were grown by solid source molecular-beam epitaxy on Si-doped *n*-type GaAs(100) substrates. After oxide desorption at 580 °C, a 150-nm-thick GaAs buffer layer was grown. Then the substrate temperature was lowered to 500 °C for deposition of 2.1 monolayers (ML) InAs. Formation of InAs QDs was verified by the sharp transition from streaky to spotty of the reflection high-energy electron diffraction pattern. Thereafter, for samples A–D different capping procedures were applied: (A) Deposition of 10 nm GaAs at 500 °C followed by 150 nm GaAs growth at 580 °C; (B) cooling down the sample to 300 °C before capping the InAs QDs by 10 nm GaAs at the same temperature and growth of 150 nm GaAs at 580 °C; (C) capping the InAs QDs by 3 ML GaAs at 500 °C, then cooling down the sample to 300 °C for deposition of 150 nm GaAs; and (D)

capping the InAs QDs by 3 ML GaAs at 500 °C followed by a growth interruption (GI) of time *t*, deposition of 10 nm GaAs at the same temperature, and growth of 50 or 150 nm GaAs at 580 °C. In sample D five such InAs QD layers were inserted with GI times *t* of 0, 20, 40, 60, and 90 s, separated by 60 nm GaAs and 150 nm GaAs on top. An extra 30 nm GaAs spacer was grown as a marker between the QD layers with GI times *t* of 40 and 60 s. The time for cooling down samples B and C from 500 to 300 °C was 4 min. The growth rates were 0.58 and 0.06 ML/s for GaAs and InAs, respectively, and the As₄ beam equivalent pressure was 1×10^{-5} Torr. After growth, the samples were taken out and loaded into the STM ultrahigh vacuum (UHV) chamber with a base pressure of less than 2×10^{-11} Torr. The samples were cleaved in the UHV chamber and the STM measurements were carried out on the (110) cleaved facet with polycrystalline tungsten tips prepared by electrochemical etching and self-sputtering.⁵ Large areas were scanned and analyzed⁶ to select InAs QDs with the cleavage plane nearly across their centers.

Figure 1(a) shows the filled states topography X-STM image of the InAs QDs in sample A, which are capped in the

FIG. 1. (a) Filled states topography X-STM image of the InAs QDs in sample A with conventional capping by 10 nm GaAs at 500 °C and 150 nm GaAs at 580 °C. $V_{\text{sample}} = -3.0$ V. Part of the image (a) marked by four corners is treated by a local mean equalization filter and shown in (b). In (a) the arrow indicates the growth direction. The black-to-white height contrast in (a) is 0–0.5 nm.

^{a)}Electronic mail: q.gong@tue.nl

FIG. 2. (a) Filled states topography X-STM image of the InAs QD in sample B with the GaAs cap grown at 300 °C. $V_{\text{sample}} = -3.0$ V. Part of the image (a) marked by four corners is treated by a local mean equalization filter and shown in (b). In (a) the arrow indicates the growth direction. The black-to-white height contrast in (a) is 0–0.5 nm.

conventional way by 10 nm GaAs at 500 °C and 150 nm GaAs at 580 °C. Part of the image shown in Fig. 1(b) is treated with a local mean equalization filter⁷ to enhance atomic details by removing the large scale background contrast. The bright horizontal lines are the top zig-zag rows of the (110) surface, which are separated by one bilayer (BL), i.e., 2 ML. The topographical contrast in Fig. 1(a) is due to the outward relaxation of the cleaved surface of the compressively strained InAs QDs, revealing their cross-sectional shape.⁶ The bright spots in Fig. 1(b) correspond to In atoms in the top layer of the cleaved surface. The height of the InAs QDs in sample A is measured as 8 BL by counting the number of atomic rows.

It is well established that InAs QDs buried in the conventional way of sample A exhibit a reduced height compared to unburied ones due to QD leveling.^{2–4} In order to determine the QD height reduction, the shape change during overgrowth of the InAs QDs in sample B is strongly suppressed by capping them at 300 °C. The filled states topography X-STM image of an InAs QD in sample B is shown in Fig. 2(a) with the filtered image in Fig. 2(b). The InAs QD exhibits very sharp and well-defined interfaces confirming the suppressed QD leveling, atom diffusion, and segregation⁸ and, thus, the preservation of the QD shape.^{1,9} The height of the InAs QD is 12 BL, which is 4 BL larger than that of the QDs in sample A. This indicates that during conventional capping at 500 °C the QD height is reduced by about one-third of the original one.

When InAs QDs are capped only by a very thin GaAs layer, strong QD leveling or QD collapse occurs.^{2,4} Figures 3(a) and 3(b) show the filled states topography and filtered images of such InAs QDs in sample C. The QDs are capped at 500 °C by 3 ML GaAs and subsequently overgrown at 300 °C to further maintain the shape. During the thin GaAs capping and cooling down, the leveling of the InAs QDs leads to a rather homogeneous (In,Ga)As layer in-between the QDs due to In detachment from the QD tops, Ga/In intermixing, and In segregation with a thickness of about 4 BL, which is much thicker than the original InAs wetting layer. Intermixing with the GaAs substrate¹⁰ expected during the growth of InAs at 500 °C additionally contributes approximately 3 ML of GaAs to the (In,Ga)As layer, which is derived by subtracting the thicknesses of deposited InAs (2.1 ML) and GaAs (3 ML) from the total (In,Ga)As thickness of 4 BL. After leveling of the QDs, unincorporated In floating

FIG. 3. (a) Filled states topography X-STM image of the InAs QD in sample C capped by 3 ML GaAs at 500 °C, followed by 150 nm GaAs grown at 300 °C. $V_{\text{sample}} = -3.0$ V. Part of the image (a) marked by four corners is treated by a local mean equalization filter and shown in (b). In (a) the arrow indicates the growth direction. The black-to-white height contrast in (a) is 0–0.4 nm. The arrow in (b) points to the In-rich layer.

on the surface is pinned there by the low-temperature GaAs capping and forms an In-rich layer marked by the arrow in Fig. 3(b). The sharp interface between this layer and the low-temperature GaAs cap confirms that In segregation and diffusion in growth direction are strongly suppressed for GaAs overgrowth at 300 °C. Most interestingly, the InAs QDs are completely leveled to the thickness of the (In,Ga)As layer in-between them, which is much smaller than the height of the QDs observed in sample A. This suggests that the conventional, continuous GaAs capping at 500 °C in sample A not only drives QD leveling during the initial stage, like the thin GaAs capping in sample C, but also quenches the leveling process when the QDs become buried.

To assess the time scale of QD leveling which is, thus, crucial for the final size and shape of the buried QDs, varying GI times are inserted in sample D after deposition of 3 ML GaAs at 500 °C on the InAs QDs prior to GaAs overgrowth. Figure 4 shows the filled states topography image of the InAs QDs in sample D with GI times of 0, 20, 40, 60, and 90 s in subsequent layers. Clearly, the height of the InAs QDs capped without GI (first layer) is significantly larger than that of the QDs with insertion of 20 s GI. No significant further decrease in QD height is observed when the GI time

FIG. 4. Filled states topography X-STM image of the five InAs QD layers in sample D. The InAs QDs are capped by 3 ML GaAs at 500 °C, followed by a GI and a 10 nm GaAs cap grown at 500 °C plus a GaAs separation layer at 580 °C. $V_{\text{sample}} = -3.0$ V. The increasing GI times of 0, 20, 40, 60, and 90 s are noted in the image. The arrow indicates the growth direction. The black-to-white height contrast is 0–0.5 nm.

is increased to 90 s. Hence, QD leveling during thin GaAs capping and GI takes place on a time scale of less than 20 s. For the present GaAs growth rate, this is comparable to the time required for growing several nanometers of GaAs to fully bury the QDs by continuous overgrowth. This indicates that both the driving and quenching of the QD leveling in conventional capping take place on a similar time scale. The size and shape of the buried QDs are therefore determined by a delicate interplay between driving and quenching of the QD leveling, which is controlled by the GaAs growth rate and growth temperature.

A model based on the above-mentioned experimental results is proposed for the growth of InAs QDs embedded in GaAs. The growth of InAs commences in the two-dimensional (2D) layer-by-layer mode until the InAs thickness reaches the critical value of 1.7 ML for InAs QD nucleation to reduce the accumulated strain. The InAs QDs are formed by In atoms transported massively from the 2D InAs layer, leaving a thin wetting layer on the surface. The whole system is stable at the minimum of the total energy composed of the surface energy, the strain energy, and the interface energy. Subsequent capping of the InAs QDs by GaAs, on the other hand, introduces extra strain energy between the GaAs cap and the InAs QD layer, resulting in an unstable system and the consequent QD leveling process. In atoms are redistributed from the InAs QD tops to the areas in-between them during the QD leveling. They contribute to a several nanometers thick (In,Ga)As layer with an exponential In composition decay due to In segregation and Ga/In intermixing during overgrowth,¹¹ reducing the lattice mismatch and, hence, the total energy of the system. Thus, the thickness and the In composition profile of the (In,Ga)As layer in-between the InAs QDs strongly depends on the QD leveling and In segregation. It is important to note that the QD leveling is very sensitive to the substrate temperature and is strongly suppressed at low growth temperatures, where it becomes more and more difficult to thermally break the In–As bonds. In addition to inducing QD leveling, the GaAs cap buries the InAs QDs, thereby quenching the leveling process during

continued overgrowth. Therefore, the size and shape of the embedded InAs QDs are determined by a delicate interplay between driving and quenching of the QD leveling during capping, which depends strongly on the growth rate and growth temperature of the GaAs cap.

In summary, we have investigated the capping process of InAs QDs grown by molecular-beam epitaxy on GaAs(100) substrates by X-STM. In its initial stage, GaAs capping induces leveling of the QDs to drastically decrease their height. During continuous capping the QD leveling is quenched when the QDs become buried. Both phenomena—driving and quenching of the QD leveling—take place on a similar time scale and are very sensitive to the GaAs growth rate and growth temperature. This understanding opens up an efficient route for controlling the size and shape of buried QDs.

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⁷This method subtracts the mean value of the nearest neighbor pixels within the filter window: $O(x,y)=I(x,y)-(1/MN)\sum_{i=-N/2}^{N/2}\sum_{j=-M/2}^{M/2}I(x+i,y+j)$. By choosing a size of about $1\times 1\text{ nm}^2$ for the filter window, long-range fluctuations in the image are suppressed while atomic details are preserved.

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