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Atomic scale study of the impact of the strain and composition of the capping layer on the formation of InAs quantum dots*

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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, single electron transistors, etc. InAs QDs are commonly created by Stranski–Krastanov growth mode when InAs is deposited on a substrate with a bigger lattice constant, like GaAs or InP. 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. 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, 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. For InAs/InP QDs, capping materials other than InP, like InGaAsP, have also successfully been used for laser applications.

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...
that were capped by either InP, InGaAs, or InGaAsP (the last two materials being nominally lattice matched to InP). Then we studied the capping with strained layers to reveal the role of strain. For that we used InAs QDs grown on (100) GaAs capped with InGaAs and GaAsSb strain reducing layers. While strong morphological differences are found when strained layers are used, the difference between various lattice matched capping materials is more subtle, although also relevant for device applications.

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. Indeed, room temperature lasers with low threshold current density emitting at 1.5 μm were recently demonstrated. 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. 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 In0.53Ga0.47As in the second layer (followed by 20 nm of InP), and lattice matched In0.35Ga0.65As0.285P0.715 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 In0.15Ga0.85As layer grown at 500 °C, while in the second a 6 nm GaAs0.75Sb0.25 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 in situ cleaved samples under ultrahigh vacuum (p < 4 × 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.

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-
In Fig. 2, we observe a significant increase in the amount of deposited InAs by approximately twice the nominal 2.1 ML value. 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, the structure is kept under an As flux promoting the exchange of P by As and thus increasing the amount of deposited 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. 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 As concentration. The agreement is quite good in the range of validity of the counting method.

The effect of the As/P exchange reaction would be stronger 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 grown with a higher As flux 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 substrate. 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 material 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 dots in a sample similar to layer A but grown with a higher As flux ($V_{\text{sample}}=-3.0$ V, $I_{\text{tunnel}}=100$ pA) is also shown in Fig. 4.

FIG. 3. (Color online) 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.

FIG. 4. Current image of two dots and the WL in a sample similar to layer A but grown with a higher As flux ($V_{\text{sample}}=-2.0$ V, $I_{\text{tunnel}}=50$ pA). The WL goes deep into the substrate (compare with Fig. 2) and it is not aligned with the base of the dots. Growth direction: [113], lateral direction: [1–10].

FIG. 5. (Color online) 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. Growth direction: [113], lateral direction: [1–10].
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 In(As,P)/InP quantum well width 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 of InAs/InP QDs is such that the higher the growth rate, the bigger the dot height.31 Since the QDs capped with InP (higher growth rate) are smaller than the others, the difference should be due to the capping material itself.

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. 5 and 2). This is likely due to phase separation in the capping layer, which we analyze next. The inhomogeneous topographic contrast in the InGaAs layer (Fig. 5) 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 toward the regions on top of the dots to minimize the strain, creating a columnar-like In rich region above the dots. This process has been observed in columnar InGaAs QDs grown on GaAs,32 as well as in InAs/GaAs QDs, where capping with InGaAs has been shown to induce an increase of the dot size.33 In our case, the phase separation directly affects the QDs by creating a rough top interface in which the In content decreases gradually.

In the third layer, a lattice matched InGaAsP alloy was used as the capping material (Fig. 6). The average dot height was 3.4±0.2 nm, and the maximum measured base length 27±2 nm. As in the case of InGaAs, the height of the dots is higher as those capped with InP where the dot partially dissolved due to the As/P exchange. This means that, although there is phosphorous in the capping layer, there is no dot height reduction due to an As/P exchange. The shape resembles that of a truncated pyramid with a flat top interface. More remarkable is the fact that 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 μm with low threshold current density.22

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

As mention in Sec. I, previous studies have already shown a significant reduction of the InAs/GaAs dot height after capping7,8,10–12 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.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).3,11,12 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,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...
clarified by using GaAsSb as the capping material because in this case the InAs dot height cannot increase due to phase separation in the capping layer.

Figure 9 shows a filled states image of an InAs/GaAs QD capped with GaAsSb. The group V elements are imaged in this measurement so the bright spots are Sb atoms in the As matrix. Contrary to what happens when capping with GaAs, the capping layer perfectly wets the dots. Sb segregation into the GaAs layer is also observed. The dot height is now 9.5±0.2 nm, much higher than that of GaAs-capped dots, and the dot is almost a full pyramid with a diagonal base length of 32±2 nm. This clearly indicates that the dots are preserved during capping. By comparing the measured outward relaxation to the calculated one [see Fig. 10(a)], we can deduce the Sb content in the capping layer. 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 InGaAs 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.