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Influence of an ultrathin GaAs interlayer on the structural properties of InAs/InGaAsP/InP (001) quantum dots investigated by cross-sectional scanning tunneling microscopy

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Cross-sectional scanning tunneling microscopy is used to study at the atomic scale how the structural properties of InAs/InGaAsP/InP quantum dots (QDs) are modified when an ultrathin (0–1.5 ML) GaAs interlayer is inserted underneath the QDs. Deposition of the GaAs interlayer suppresses the influence of the As/P exchange reaction on QD formation and leads to a planarized QD growth surface. A shape transition from quantum dashes, which are strongly dissolved during capping, to well defined QDs takes place when increasing the GaAs interlayer thickness between 0 and 1.0 ML. Moreover, the GaAs interlayer allows the control of the As/P exchange reaction, reducing the QD height for increased GaAs thicknesses above 1.0 ML, and decreases the QD composition intermixing, producing almost pure InAs QDs. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2884692]

Self-assembled InAs quantum dots (QDs) grown on InP substrates are attracting a great interest due to their potential applications as the active region of optoelectronic devices operating at 1.55 μm range telecommunication wavelengths. One of the main problems in InAs/InP QD growth is the presence of the As/P exchange reaction during InAs growth on InP or InGaAsP surfaces,[1,2] which increases the QD size and shifts the wavelength far above 1.6 μm at room temperature. QD emission in the 1.55 μm range has been obtained from InAs quantum dashes[3] and quantum wires[4] grown on InP (001), as well as from InAs QDs grown on InP (311)B substrates.[5] Regarding InAs/InP (001) QDs, commonly used for production, Gong *et al.*[6] and Anantathanasarn *et al.*[7] recently demonstrated a method to reproducibly tune the emission wavelength in the 1.55 μm range of QDs grown by chemical-beam epitaxy (CBE) and metal-organic vapor-phase epitaxy (MOVPE), respectively. This is achieved by the insertion of an ultrathin (0–2 ML) GaAs interlayer between the QDs and the InGaAsP layer below.[6,7] From photoluminescence and atomic force microscopy (AFM) measurements of buried and surface QDs, it has been concluded that the GaAs interlayer suppresses the As/P exchange reaction, reducing the QD height and, consequently, blueshifting the wavelength toward 1.55 μm.

In this work, we provide a deeper insight and confirm the effect of the ultrathin GaAs interlayer on the structural properties of MOVPE grown InAs/InGaAsP/InP (001) QDs by using cross-sectional scanning tunneling microscopy (X-STM). This technique allows to study at the atomic scale how GaAs layers with different thicknesses affect the QDs.

The analyzed samples were grown by low-pressure MOVPE using trimethyl indium, trimethyl gallium, tertiarybutyl arsine (TBA), and tertiarybutyl phosphine as gas sources with hydrogen as a carrier gas. The structure consisted of four InAs (nominal thickness of 3 ML) QD layers grown on Q1.25 InGaAsP lattice matched to InP. The thicknesses of the GaAs interlayer inserted underneath the QDs in the four layers were 0, 0.5, 1.0, and 1.5 ML, respectively. The QD layers were separated by 50 nm thick Q1.25 InGaAsP layers. The QD growth was carried out at low temperature and low TBA flow rate of 510 °C and 1.5 SCCM (SCCM denotes cubic centimeter per minute at STP), respectively, optimum for device fabrication.[8]

The X-STM measurements were performed at room temperature on the (1-10) 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. All the images were obtained in constant current mode at negative voltage (filled states) so the group V elements (As and P) are directly imaged.

Figure 1(a) shows a filled states image of the four QD layers taken at −3.0 V. The QD layers without GaAs interlayer (L1) and with 0.5 (L2), 1.0 (L3), and 1.5 ML (L4) lie from right to left of the image, respectively. The inhomogeneous contrast in the InGaAsP barriers is due to short range random composition fluctuations in the alloy. At this high voltage, the electronic contrast is strongly suppressed and the measurements reflect mainly the topographic contrast, which is due to the relaxation of the cleaved surface due to the presence of strain.[9,10] Consequently, the thin GaAs interlayer appears here as a dark layer due to the tensile strain. The relaxation profile at −3.0 V (averaged in a ~50 nm wide region without QDs) across the four layers is shown in Fig. 1(b). A positive (outward) relaxation is observed in L1 (no GaAs interlayer) due to the compressive strain in the InAs layer. On the other hand, a negative (inward) relaxation is observed in the other three layers due to the presence of the GaAs interlayer. The magnitude of this inward relaxation is proportional to the amount of GaAs. Namely, the relaxation increases approximately by a factor of 2 from layer L2 (46 pm) to layer L3 (88 pm) and by a factor of 3 for layer L4 (157 pm).

Figure 2(a) shows a high voltage image of the L4 layer in a region without QDs. While the bottom interface between the GaAs layer and the InGaAsP is quite rough, the top interface between the GaAs layer and the wetting layer (WL) is

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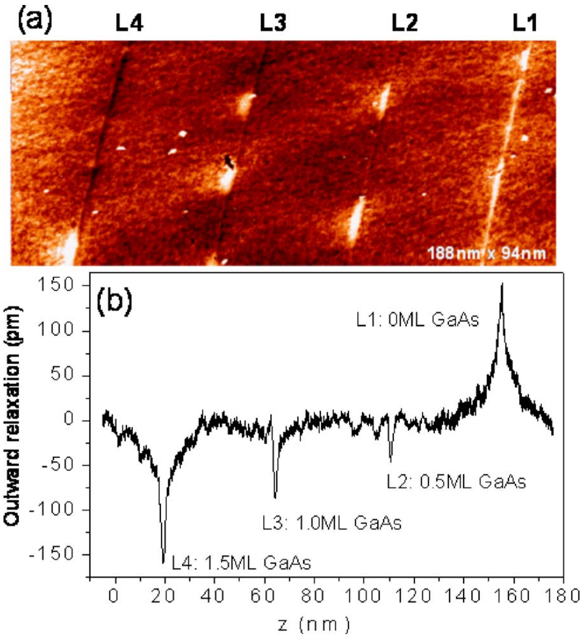

FIG. 1. (Color online) (a) High voltage ($V=-3.0$ V) filled states image showing the four QD layers. The thin GaAs interlayers appear dark due to the tensile strain. (b) Relaxation profile across four QD layers in a region without QDs. The profile is extracted from a high voltage image ($V=-3.0$ V).

very sharp [the WL only becomes visible at lower voltages when the electronic contrast due to the smaller band gap of InAs is enhanced and has a thickness of less than 2 ML, as estimated from Fig. 2(b)]. The progressive planarization of the growth surface with increasing amount of GaAs can have a strong influence on QD formation, since, for example, it has been observed that a rough InGaAsP surface leads to InAs quantum dash formation instead of QDs in CBE growth.[11] This must be taken into account in order to explain the significant differences in the resulting nanostructures between the first layer (no GaAs interlayer) and the rest of the layers (GaAs interlayer) that can already be noticed in Fig. 1(a).

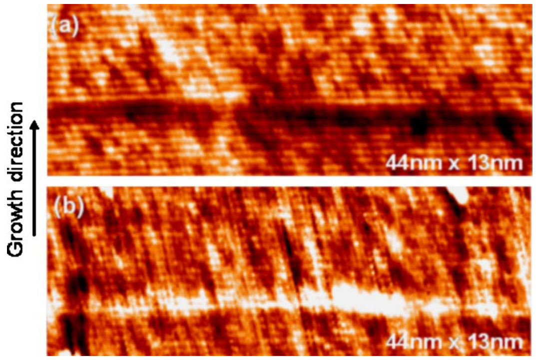

FIG. 2. (Color online) (a) High voltage ($V=-3.0$ V) filled states image of the GaAs interlayer in a region without QDs in the L4 layer (1.5 ML GaAs). (b) Low voltage ($V=-1.5$ V) filled states image of the WL in the L2 layer (0.5 ML GaAs).

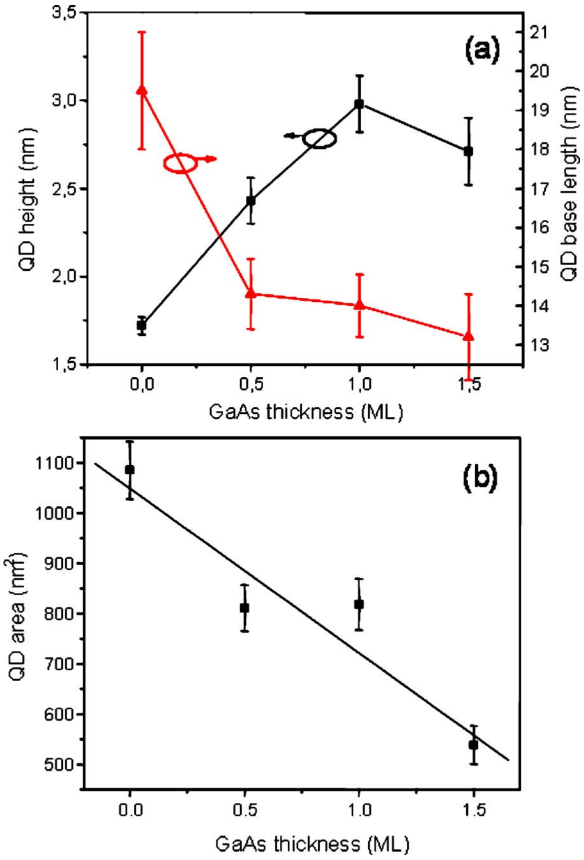

FIG. 3. (Color online) (a) Average QD height (squares) and base length (triangles) as a function of the thickness of the GaAs interlayer. (b) Total area of all the QDs found after scanning 1 μm in each layer as a function of the thickness of the GaAs interlayer. The lines are guides for the eye.

The structural parameters, including an average QD height, an average base length, and an integrated area of the QDs in the four layers are shown in Fig. 3 (~100 QDs were analyzed). Without the GaAs interlayer, the resulting nanostructures are flat and more elongated [see Figs. 3(a) and 1(a)]. Indeed, AFM measurements in similar uncapped nanostructures showed that they are quantum dashes elongated along the [1-10] direction.[7] The height of the capped quantum dashes is much smaller than that of the uncapped ones (~7 nm),[7] indicating that they strongly collapse during the capping process. This deviation was not observed in case of the QDs in other layers (the difference in height with the uncapped case is much smaller), demonstrating that the quantum dashes are more efficiently dissolved during capping. As a result of the strong collapsing, the quantum dashes are sometimes not clearly separated and the layer looks similar to a quantum well with very strong fluctuations in thickness and composition. When 0.5 ML of GaAs is inserted, the base length is strongly reduced and the height is increased, and well defined individual nanostructures are observed [see also Fig. 1(a)]. This tendency continues when the amount of GaAs is increased to 1.0 ML. In this regime (GaAs <1 ML), a morphological shape transition from quantum dashes to QDs takes place,[7] probably due to the progressive planarization of the growth surface, i.e., reduction of the anisotropic surface roughness[11] that occurs when the surface is being covered by GaAs. After the GaAs interlayer thickness exceeds 1.0 ML, a decrease in the QD height and base length is observed. This is mainly due to suppression of the As/P exchange process, which is known to create an extra amount of free In that contributes to QD formation, increasing the size of the QDs.[2,12] The effect of the reduced As/P exchange is counteracted in the submonolayer GaAs regime by the shape transition but becomes evident when the amount of GaAs is increased above 1.0 ML. The reduction of the As/P

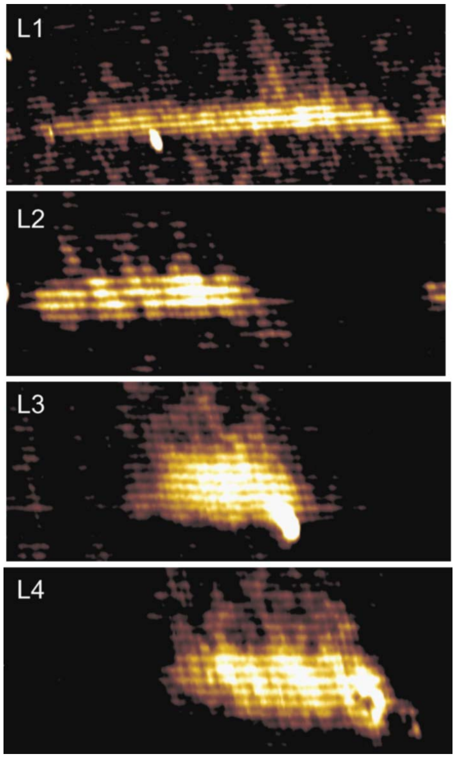

FIG. 4. (Color online) Filled state image of a representative QD in each layer. The dark spots inside the nanostructures indicate the presence of P atoms.

exchange in this regime (GaAs thickness >1.0 ML) dominates the differences in the QD structure, leading to smaller QDs with shorter emission wavelength in the 1.55 μm range.[7]

To further prove that the GaAs interlayer is progressively reducing the As/P exchange, we measured the total cross-sectional area of the QDs (without the WL) found after scanning 1 μm along the four layers, which is directly related to the total QD volume and, therefore, to the total amount of InAs. As shown in Fig. 3(b), the integrated cross-sectional area of the nanostructures decreases significantly as soon as the 0.5 ML thick GaAs layer is inserted and decreases further when the amount of GaAs is increased to 1.5 ML. This means that the total volume of InAs in the nanostructures decreases when the GaAs interlayer thickness increases. Since the same amount of InAs was deposited in every case, the difference is related to a reduction of the In available for QD formation produced by As/P exchange. This confirms that the exchange process can be controlled by the insertion of the GaAs interlayer. A contribution to the reduced amount of InAs forming the QDs coming from differences in the WL between L2, L3, and L4 cannot be completely ruled out. Nevertheless, if it exists, it is quite small because the WL looks apparently the same in L2, L3, and L4.

The composition of the nanostructures is also affected by the GaAs interlayer. Figure 4 shows atomically resolved images of a typical QD in each layer. As atoms appear bright in these images, while P atoms inside the QD appear as dark features in the As rows. The amount of dark spots is higher in the absence of a GaAs interlayer (L1), indicating a stronger intermixing, likely with P. The intermixing decreases when 0.5 ML of GaAs is inserted but is still clearly present. When the amount of GaAs is increased to 1.0 and 1.5 ML, the composition is more homogeneous and less intermixed, P is inhibited to enter the QDs, and the QDs are close to 100% InAs. Therefore, the GaAs interlayer gradually decreases QD intermixing, making it negligible for GaAs thicknesses above 1.0 ML.

In conclusion, we have used X-STM to show how an ultrathin GaAs interlayer deposited before QD formation affects the structural properties of InAs/InGaAsP/InP (100) QDs grown by MOVPE. The GaAs layer provides a planar growth surface and allows the control of the As/P exchange reaction. Increasing the GaAs thicknesses from 0 to 1.0 ML induces a shape transition from quantum dashes (which are strongly dissolved during capping) to well defined QDs, increasing the height of the nanostructures and reducing the intermixing. For GaAs thicknesses above 1.0 ML, the QDs consist of almost pure InAs with a smaller average height (and, thus, emission wavelength) due to reduced As/P exchange.

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