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# In islands and their conversion to InAs quantum dots on GaAs (100): Structural and optical properties

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We report growth of crystalline In islands on GaAs (100) by molecular beam epitaxy at low temperatures. The islands have a pyramidlike shape with well defined facets and epitaxial relation with the substrate. They are of nanoscale dimensions with high density. Above a certain substrate temperature, associated with the melting point of In, noncrystalline round shaped islands form with larger size and lower density. Upon conversion of the In islands into InAs islands under As flux, the final shape does not depend on the original crystalline state but on the annealing temperature of the InAs islands. Clear photoluminescence is observed from InAs quantum dots after conversion of the crystalline In islands. © *2010 American Institute of Physics*. [doi:10.1063/1.3269700]

## I. INTRODUCTION

Recently, there has been a lot of interest in droplet epitaxy for the formation of III-V semiconductor quantum nanostructures. In droplet epitaxy, only the group-III element is deposited to form liquid droplets or solid islands on the substrate surface which are then recrystallized under group-V element flux.[1] This technique is very versatile and has been applied for the formation of various nanostructures such as quantum dots (QDs),[2] QD pairs,[3] or single and multiple quantum rings.[4,5] For InAs QDs grown on GaAs using droplet epitaxy, the best results in terms of QD size, shape, and optical quality have been obtained for In deposition at near-room temperature.[6] Most of the work, so far, has concentrated on the properties of the final InAs nanostructures and not much on the nature of the In islands and its possible influence on the recrystallization process.

In this work, we report the distinct crystalline structure of In islands grown on GaAs (100) at sufficiently low temperatures. Reflection high-energy electron diffraction (RHEED) and atomic force microscopy (AFM) reveal a pyramidlike shape with well defined facets and epitaxial relation with the substrate, independent on the reconstruction of the starting GaAs surface. The islands are of nanoscale dimensions with high density. Above a certain substrate temperature, associated with the melting point of In, noncrystalline round shaped islands form with larger size and lower density. After converting the In islands into InAs islands under As flux, the final shape does not depend on the original crystalline state. However, the final shape does depend on the annealing temperature of the InAs islands. InAs QDs formed by recrystallization of the small crystalline In islands reveal clear photoluminescence (PL) emission and sharp lines from individual QDs at low temperature evidencing high structural and optical quality.

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## II. EXPERIMENTAL PROCEDURE

The samples were grown by solid source molecular beam epitaxy (MBE) on undoped, singular GaAs (100) substrates. After native oxide removal under $As_4$ flux at 580 °C, a 200 nm thick GaAs buffer layer was grown. Then, the samples were cooled down to temperatures between 50 and 120 °C (thermocouple reading) and the As valve was closed around 300 °C, resulting in an As-rich $c(4\times4)$ surface reconstruction, determined by RHEED. For some samples, a Ga-rich $(4\times6)$ surface reconstruction was prepared by deposition of Ga to an equivalent of 1.75 monolayers (ML) GaAs at 400 °C after closing the As valve. In was then deposited to an equivalent of 2 and 12 ML InAs after the As background pressure was below $2\times10^{-9}$ Torr. Some samples were taken out at this stage for AFM investigations (tapping mode in air). For conversion of the In islands into InAs, the starting temperature was 80 °C for all samples and the $As_4$ beam equivalent pressure was $1\times10^{-5}$ Torr. After a few minutes, the substrate temperature was raised to 400 or 500 °C followed by 20 min annealing. Once again, some samples were cooled down and taken out at this step for AFM investigations. For PL measurements, 20 nm GaAs were grown after annealing at the annealing temperature and 80 nm at 580 °C. For PL, the samples were mounted in a low temperature cryostat and excited with the 632.8 nm line of a He–Ne laser. The PL was dispersed by a 1/4 m single monochromator and detected by a liquid nitrogen cooled InGaAs photodiode array detector.

## III. RESULTS AND DISCUSSION

Figure 1 shows the RHEED patterns recorded along the GaAs [011], $[01\bar{1}]$, and [001] directions after deposition of 2 ML In at 80 °C. Similar patterns were obtained for temperatures below 120 °C and different In amounts. The spotty RHEED pattern clearly shows the transmission diffraction pattern in agreement with the body centered tetragonal crystal structure of bulk In, indicating the formation of crystalline In islands. The characteristic transmission diffraction

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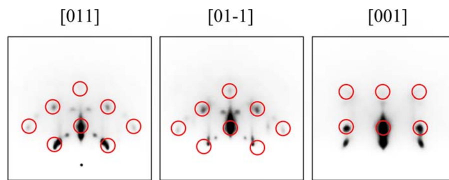


FIG. 1. (Color online) RHEED patterns recorded along GaAs [011], [01$\bar{1}$], and [001] after deposition of 2 ML In at the temperature of 80 °C.

spots are marked by circles. Crystalline In islands have also been reported for growth on GaAs (110).[7,8] Careful analysis of the RHEED patterns reveals the epitaxial relation with the substrate to be $[001]_{In} \| [011]_{GaAs}$, where $[100]_{In} \| [100]_{GaAs}$ is the high symmetry direction of In. The additional diffraction features might be attributed to twin planes in the islands,[9] surface scattering,[10] finite size effects,[11] or even electron refraction.[12] The deduced epitaxial relation is confirmed by the AFM image presented in Fig. 2(a). Most of the islands obtained after deposition of 12 ML In, to better resolve the shape, have an average side length of 100 nm, height of 35 nm, and density of 12 $\mu m^{-2}$. They exhibit a pyramidlike shape with square base oriented along [011] and distinct side facets, most likely {110} facets. Some islands adopt more complicated shapes, which most probably originate from the formation of defects, especially twin planes.

All the results discussed above are obtained for In islands grown on As-rich GaAs surfaces with $c(4\times4)$ surface reconstruction. Very similar results are obtained on Ga-rich GaAs surfaces with $(4\times6)$ surface reconstruction apart from slight variations in island density, which might well originate from slight variations in the substrate temperature. Therefore, we conclude that the reconstruction of the starting GaAs surface has no influence on the formation of the crystalline In islands.

For In islands grown at 120 °C, no streaky to spotty RHEED pattern transition is observed, but only a slight increase of diffuse background scattering, indicating that crystalline islands do not form. This is confirmed by the AFM image shown in Fig. 2(b). The islands obtained after deposition of 12 ML In have a shape of a truncated sphere with average diameter of 300 nm, height of 150 nm, and density of 0.5 $\mu m^{-2}$, and no visible facets. Upon cooling the sample in the MBE chamber, there is no change of the RHEED pattern. However, in scanning electron microscopy (not presented here), a small fraction of around 5% of the islands exhibits the faceted pyramidlike shape indicating crystallization of some islands during cooling. The larger size and smaller density of the In islands grown at 120 °C, compared to those grown at 80 °C, are consistent with the larger In adatom surface migration length at higher temperature. The different shape is explained by the fact that the islands grown at 120 °C are liquid. The melting temperature of bulk In is 156 °C. In an experiment to melt the crystalline In islands in the MBE chamber by increasing the substrate temperature from 80 °C, a melting temperature between 170–180 °C is deduced from the disappearance of the spotty RHEED pattern. This discrepancy is explained by heating of the GaAs surface by the open In effusion cell during In island growth and a slight offset of 10–20 °C between the thermocouple reading and actual GaAs surface temperature for the closed In effusion cell during melting. Then, during In island growth, the thermocouple reading of 80 °C is still below the bulk In melting temperature and the reading of 120 °C is above. It is interesting to note that the In islands observed after melting the crystalline In islands have size, shape, and density very similar to the In islands grown at 120 °C. Hence, during melting, there is no memory of the size, shape, and density of the crystalline islands and an equilibrium state according to the higher temperature is reached.

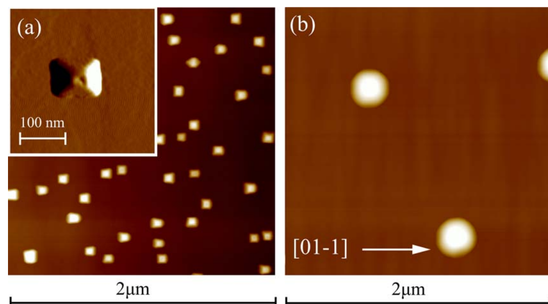


FIG. 2. (Color online) AFM image of the In islands obtained after deposition of 12 ML In at (a) 80 °C and (b) 120 °C. The scan fields are $2\times2$ $\mu m^2$. Inset in (a) shows a magnified amplitude image of a single In island formed at 80 °C.

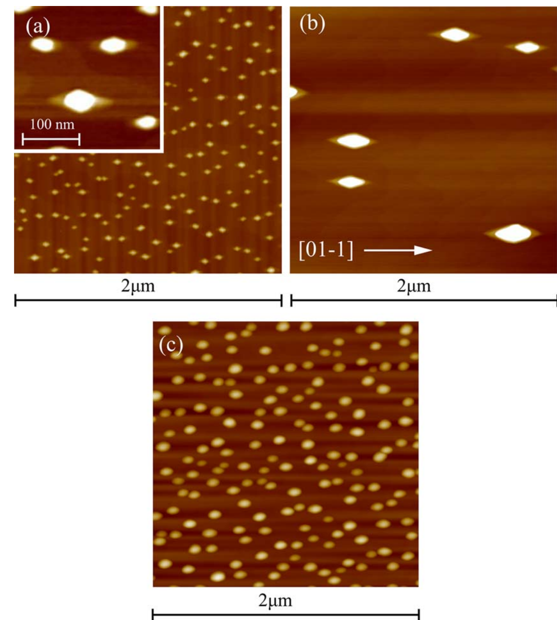


FIG. 3. (Color online) AFM images of the InAs islands formed from (a) crystalline and (b) noncrystalline In islands after exposure to $As_4$ flux and annealing at 500 °C. (c) InAs islands formed from crystalline In islands and annealed at 400 °C. The amount of deposited In is 2 ML. The scan fields are $2\times2$ $\mu m^2$. Inset in (a) shows a magnified image of InAs islands formed from crystalline In islands annealed at 500 °C.

Figures 3(a) and 3(b) show the AFM images of the InAs islands obtained after deposition of 2 ML In at 80 and 120 °C, exposure to As flux, heating up, and annealing at 500 °C, revealing islands elongated along the [01$\bar{1}$] direction. The annealing step is inevitable for obtaining InAs QDs of high structural and optical quality.[13] The In islands grown at 120 °C are first cooled down to 80 °C to avoid collapse

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of the solidified islands during the initial exposure to As which often leads to the formation of rings-like structures for liquid islands exposed to $As_4$. This can also be prevented by applying intense As fluxes at higher temperatures. However, to guarantee identical recrystallization conditions for the crystalline and noncrystalline In islands, both solid, for InAs island formation, we start the process at the same temperature for comparison. For both types of In islands, instantly after applying the As flux, the RHEED pattern changes into one typical for InAs islands. After reaching 400 °C and annealing, the intensity of the diffraction spots increases, indicating complete conversion of In into InAs. Characteristic chevrons appear in the $[01\bar{1}]$ azimuth whose intensity increases for further heating to 500 °C. This is typical for the formation of InAs islands which become more and more elongated in the $[01\bar{1}]$ direction at higher temperature. This is confirmed by the AFM image shown in Fig. 3(c) of InAs islands formed from 2 ML In deposited at 80 °C and annealed at 400 °C. The islands are less elongated and, consequently, also higher compared to those annealed at 500 °C. The RHEED patterns remain unchanged during annealing, even at 500 °C, demonstrating the formation of stable InAs islands which do not need to be overgrown at low temperatures, as has been reported in Ref. 6. This results in InAs QDs with high optical quality, as discussed below.

The AFM images and RHEED studies reveal that the shape of the InAs islands is independent on the crystalline state of the In islands. Only their size differs according to that of the In islands. This indicates that the final shape is entirely determined during the high temperature annealing step.

Figure 4(a) shows the low-temperature PL spectra of the InAs QDs formed from the nanoscale, high-density crystalline 2 ML In islands and annealed at 400 and 500 °C. InAs islands formed from the noncrystalline In islands are not discussed here as they are too large to be considered as quantum nanostructures. As expected, the PL efficiency is larger for the InAs QDs annealed at higher temperature leading to better optical quality. Moreover, the annealing temperature of the InAs QDs has a pronounced influence on the PL spectra. At higher annealing temperature, the spectrum is blue shifted and exhibits a double peak structure. The blue shift is in agreement with the larger elongation and flattening of the InAs islands annealed at higher temperature. The double peak structure is not dependent on the excitation power and the high energy peak vanishes upon increasing the measurement temperature to 100 K. Therefore, the high-energy emission does not arise from excited state transitions nor from a bimodal size distribution. It is attributed to emission from the quasitwo-dimensional, local wetting layer at the apex of the QDs formed due to the outdiffusion of InAs during annealing at 500 °C, seen in the AFM images in Fig. 3(a), which is not observed for the InAs QDs annealed at 400 °C, shown in Fig. 3(c). With increase of the measurement temperature, carriers initially localized in this wetting layer are transferred into the QDs leading to the observed rapid emission intensity quenching of the high energy line. Moreover, the local wetting layer might increase the carrier capture into the QDs, contributing to the higher PL efficiency. For the InAs QDs annealed at 400 °C, sharp peaks with resolution limited linewidth of 0.2 nm from individual QDs can easily be resolved in microPL, employing a microscope objective (~2 $\mu$m spatial resolution) for excitation and detection, shown in Fig. 4(b). This is not possible for the InAs QDs annealed at 500 °C, though the AFM measurements demonstrate a comparable QD density. Hence, the annealing at higher temperature increases the number of defect-free InAs QDs such that emission from individual QDs cannot be resolved. On the other hand, sharp emission from individual QDs for the lower annealing temperature demonstrates that droplet epitaxy is a way to obtain InAs QDs with high optical quality.

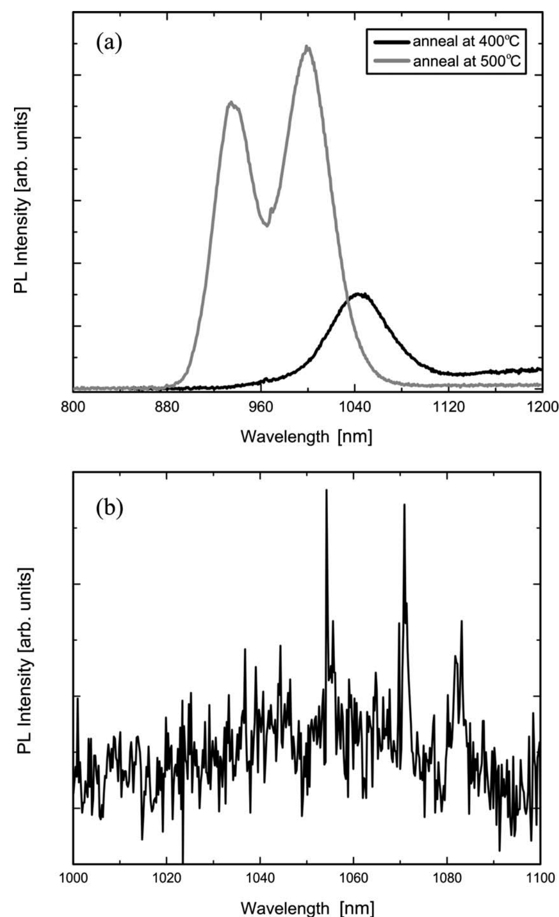


FIG. 4. (a) PL spectra taken at 10 K of the capped InAs QDs formed from the crystalline 2 ML In islands annealed at 400 °C (black line) and 500 °C (gray line). (b) MicroPL spectrum of the capped InAs QDs annealed at 400 °C.

## IV. CONCLUSIONS

In conclusion, growth of crystalline In islands on GaAs (100) by MBE at low temperatures has been reported. The islands have a pyramidlike shape with well defined facets and epitaxial relation with the substrate, determined by RHEED and AFM. The islands are of nanoscale dimensions with high density. Above a certain substrate temperature, associated with the melting point of In, noncrystalline round-shaped islands form with larger size and lower density. The formation of the In islands was found to be independent on the reconstruction of the starting GaAs surface. Upon con-

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version of the In islands into InAs islands under As flux, the final shape does not depend on the original crystalline state of the In islands but on the annealing temperature of the InAs islands. The InAs islands elongate and outdiffuse with increasing annealing temperature forming a quasitwo-dimensional wetting layer at the apex. This is reflected in the PL spectra revealing the formation of InAs QDs of high optical quality.

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