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Properties of InAs quantum dots on silicon(001) and (111)

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Abstract. The self assembled formation of InAs Quantum Dots (QDs) on silicon has been studied by RHEED, AFM, Raman and TEM. On Si(001) up to 10^{11} cm^{-2} InAs QDs can either be grown in a conventional growth mode, or by utilizing a postgrowth dewetting transition at decreasing substrate temperatures. On Si(111) instead, QDs only form on an As-passivated surface. Otherwise InAs grows in large 2D clusters.

Introduction

Besides first successes with the (heterovalent) growth of germanium QDs on Si, the most popular material of semiconductor industry, silicon, has remained to be a great challenge as a material for optoelectronic devices due to its indirect bandgap [1]. Recent results on the self-organized growth of InAs QDs embedded in a silicon matrix seem to be very promising. An intense photoluminescence peak at a wavelength of $1.3 \mu\text{m}$ could be observed up to room temperature [2]. In a former study on the embedding of the QDs in a silicon matrix we report on the possibility of growing up to 10^{11} cm^{-2} InAs QDs and the formation of large Indium clusters between the InAs QDs [3]. Further, we investigated the QD size distribution, ways of improving their uniformity as well as the defect structure in the QDs, information which is essential for a potential application of this system [4]. So far, the problem arises that RHEED, Raman and TEM indicate that after the 2D/3D growth mode transition of InAs on Si, the InAs QDs are fully relaxed with the inclusion of misfit dislocations. It seems that an annealing after the growth of a Si-cap layer finally leads to the formation of strained InAs inclusions in the Si matrix [5]. Besides that, the investigation of QD growth on differently terminated silicon substrates is believed to give new insight into the physics of self-organization in the InAs/Si system. A dewetting transition of a single monolayer (ML) of InAs during the postgrowth lowering of the temperature has been observed and being described on the basis of a thermodynamic model in [4]. For this paper, we compare the properties of InAs growth on Si(001) with Si(111) where growth depends strongly on the surface termination. InAs forms large clusters when grown directly on Si(111), whereas on the arsenic terminated surface QDs show up.

1. Experimental details

Growth experiments were carried out on 2 inch Si-substrates using a Riber 2300 MBE system. (001) substrates were Hydrogen-passivated, details on their preparation and optimized growth conditions can be found in [3, 4]. (111) substrates were either also H-passivated or after an RCA-etch the protective SiO_2 layer was desorbed in a separate chamber belonging to the same UHV-MBE-cluster. Growth was monitored in-situ by RHEED, the Raman

measurements were performed without breaking the UHV via an UHV transport-box. TEM images were obtained with a Phillips CM 200 FEG at 200 kV. AFM images, taken with a Rasterscope 4000, were evaluated by SPM image magic software.

2. Results and discussion

On Si(001) the 2D–3D transition in growth mode occurs after depositing 1.7 ML of InAs at a growth temperature of 370°C. The critical thickness has been observed to depend on the growth temperature [2] which qualitatively matches with our experiments, though we always determine a lower critical thickness than reported in [2]. Figure 1(a) shows an AFM image of a sample with nominally 5 ML of InAs on Si(001) leading to approximately 10^{11} cm^{-2} QDs. Statistics on radii and heights were evaluated and show a Gaussian distribution with mean values of 14.2 nm and 10 nm, respectively. Cross sectional TEM images let us assume, however, that the real size of the nanostructures is smaller (Fig. 1(b)). Indeed, the shape of the AFM tip has not been accounted for, and thus lateral sizes of QDs are overestimated by AFM. Information on the coherence of the QDs can be achieved by an analysis of RHEED images obtained during the growth which directly shows a projection of the reciprocal space of the epitaxial surface. At the growth-mode change, we observe an abrupt 10% decrease of the lateral distance of the appearing 3D spots compared to the former 2D streak pattern of Si(001). This indicates the full relaxation of the dots. The lattice mismatch between InAs ($a = 6.058 \text{ \AA}$) and Si ($a = 5.431 \text{ \AA}$) makes up 11.5%. As a consequence TEM images (Fig. 1(b)) show the inclusion of misfit dislocations. An averaged information on the whole QD ensemble can be achieved by Raman spectroscopy. Those spectra, taken on a sample with 3 ML InAs, show optical phonon modes of the InAs dots in the range from 216 cm^{-1} to 238 cm^{-1} (TO and LO), which corresponds to relaxed bulk frequencies of InAs (218 cm^{-1} and 243 cm^{-1}). In contrast, a fully strained state would result in a shift of 30 cm^{-1} to higher wavenumbers, which therefore can be excluded in these structures.

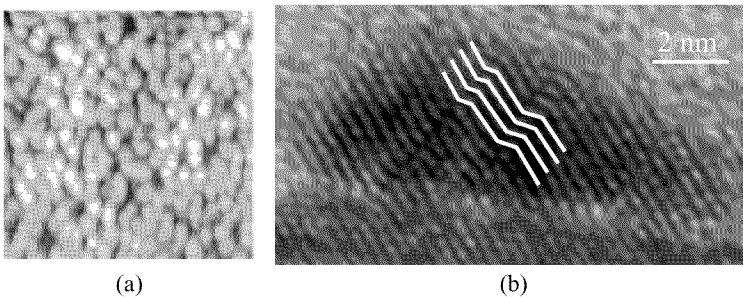


Fig. 1. (a) 500 nm^2 AFM image of 5 ML InAs/Si. (b) TEM image of the same sample.

To avoid nonradiative recombination of electron hole pairs at defects inside the QDs the structural quality of the QDs has to be improved. An interesting phenomenon in this context is the observation of a dewetting transition of a thin (below the critical thickness) InAs-film on Si(001). When reducing the substrate temperature after growth the 2D layer gets thermodynamically unstable and a spotty RHEED image indicates an abrupt transition into 3 to $8 \times 10^{10} \text{ cm}^{-2}$ islands. Though the average size of those QDs determined by AFM is smaller than for the dots grown in the conventional growth mode, TEM images again show misfit dislocations inside of the dots.

It has always been pointed out that strain is a necessary but not sufficient criterion for the

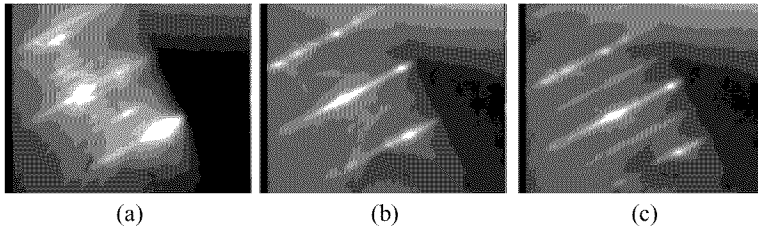


Fig. 2. RHEED images during growth of InAs on Si(111). (a) (7×7) reconstruction of Si(111), (b) 2 ML of InAs, (c) 30 ML of InAs.

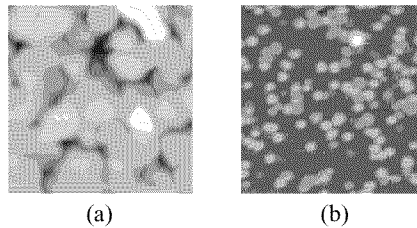


Fig. 3. 500 nm^2 AFM images of InAs on differently treated Si(111) surfaces, (a) on Si(111) InAs forms large clusters with a 2D surface even up to 30 ML, max height 13.7 nm. (b) 3 ML InAs/Si(111):As, dot size: $r = 35 \text{ nm}$, $h = 20 \text{ nm}$.

selforganized formation of QDs. For InAs on GaAs the formation of QDs is only observed for the (001) surface, whereas on GaAs (110) and (111) growth occurs in a 2D way under inclusion of many misfit dislocations [6]. We now have grown InAs on 2 differently treated Si(111) surfaces. The first procedure was that after an anneal in a separate chamber the samples were transferred via UHV into the InAs growth chamber. Figure 2(a) shows the RHEED image of the (7×7) reconstruction of the starting surface. In contrast to a deposition on Si(001) the RHEED image remains streaky and shows even a twofold reconstruction after deposition of 30 ML. In Fig. 3(a) an AFM image shows the structure of large InAs clusters separated by a network of misfit dislocations, but nevertheless consisting of a more or less “flat” surface according to RHEED.

The second procedure is to grow on an inert surface. From [7] it is known that the Si(111) surface can be terminated with Arsenic by heating the sample up to 700°C under As-flux. The uppermost Si-layer is removed and exchanged by an As-layer. The 5 bonding electrons of the As lead to 3 bonds to Si and one fully occupied electron orbital reaching perpendicularly out of the surface. The samples were prepared by an additional anneal at approximately 700°C under As-flux and then cooled to growth temperature for the InAs growth. As soon as the In was opened in addition to As the RHEED image turned spotty. AFM images (Fig. 3(b)) show nanocrystals of approximately 70 nm in diameter and 20 nm in height which formed on Si(111):As after depositing 3 ML of InAs. The morphology on this inert surface, where the InAs is rather van-der-Waals bonded than chemisorbed is very different from the growth modus on the untreated Si(111) surface. Due to the weakness of physisorption we expect the InAs to form crystals with its own lattice constant.

3. Conclusion

A broad range of parameters can be varied to finally achieve a self organized growth of InAs QDs on Si. In this study we investigated the influence of different substrate orientations

and substrate treatments. In contrast to InAs on GaAs, QDs can also be grown on arsenic passivated (111) oriented silicon. So far, the InAs nanostructures all seem to include crystal defects. Further work has to be done to optimize the growth and reduce the dimensions of the QDs.

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