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Shape transformation of Ge quantum dots due to Si overgrowth

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Abstract

The optimisation of the opto-electronic properties of Ge dots embedded in Si requires the precise control of their structural properties. In the present study we investigated the initial stages of overgrowth using transmission electron microscopy (TEM), energy resolved TEM and in situ scanning tunnelling microscopy. It is found that Ge dome clusters and Ge hut clusters behave differently during the overgrowth by Si. Ge domes transform back into hut clusters after the deposition of 5 Monolayers of Si, thus they decrease in height and increase in diameter. Ge hut clusters increase in height and diameter during Si overgrowth. The changes in shape are accompanied by local changes in the strain and composition of the dot. The presented data give detailed insights into the shape and composition of Ge quantum dots before and after embedding in Si.

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Keywords: Scanning tunnelling microscopy; Transmission electron microscopy; Quantum dots

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1. Introduction

The concept of light emission from Ge quantum dots embedded in a Si matrix relies on a strong localisation of the carriers in a small quantum box with high potential barriers [1]. Ideally the Ge clusters in the Si matrix should have an aspect ratio of 1 and should not intermix with the surrounding Si, since this situation should lead to the strongest localisation. Theoretical model calculations show that the reduction in size of the clusters might lead to an increase in the matrix element and consequently to an increase of the radiative recombination processes [2]. The deposition of germanium on Si(0 0 1) surfaces leads to several types of self-assembled Si_xGe_{1-x} quantum dots [3–6]. Our experiments show, that the size and composition does not solely depend on the conditions during the growth of the Ge dots themselves. The overgrowth of Ge dots with a Si layer to embed them in a Si matrix, as it is necessary for the realisation of opto-electronic devices, can lead to a change in shape and composition of quantum dots. These changes due to the overgrowth can have a measurable effect on physical properties [7]. The present study therefore aims at a contribution to a better understanding of these shape transformations during the initial steps of Si overgrowth using high-resolution transmission electron microscopy (HRTEM), energy filtered transmission electron microscopy (EFTEM), and scanning tunnelling microscopy (STM).

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2. Experimental details

Two sets of samples were investigated. For TEM large dislocated domes formed by depositing 8.6 monolayers (ML) of Ge, capped with 0, 1, 3, and 5 ML Si were grown by molecular beam epitaxy (MBE, Balzers UMS 500). These samples were grown at 520°C using e-beam evaporation for silicon and for germanium. The growth rates amounted to 0.08 ML/s for Ge and 0.03 ML/s for Si, respectively. Growth conditions for the second set of samples were chosen such that dome and hut clusters formed on the Si(0 0 1) surface. The clusters formed after depositing 6.5 ML of Ge. The deposition temperature was adjusted to 620°C while the growth rates were the same as for the first set. Samples with bare Ge islands and after additional deposition of 5 ML Si were investigated in situ by UHV-STM and XTEM.

For XTEM the specimens were mechanically pre-thinned and subsequently milled to electron transparency by Ar ions in a precision ion polishing system from Gatan at an acceleration voltage of 4.3 kV and an edging angle of 4°. HRTEM and EFTEM were performed along the Si(1 1 0) direction in a Philips CM30ST TEM at an acceleration voltage of 300 kV (point to point resolution: 0.19 nm) and in a Philips Tecnai ST (300 kV) equipped with a Gatan imaging filter, respectively. The three windows technique was applied to the Si L-edge. For the STM investigations the samples were transferred into a home-build set-up without breaking the ultra-high vacuum [8].

3. Results and conclusions

Prior to the overgrowth the dome clusters on Si(0 0 1) surfaces have typical contact angles around 26°, as shown in the HRTEM pictures of Fig. 1. These contact angles are indicative for $\{15\,3\,23\}$ and $\{113\}$ facets, which are frequently observed for Ge domes [9,10]. After the capping with 1 ML of Si, no influence on the shape of the clusters can be seen while the capping with 3 ML of Si leads to a formation of new facets at the pedestal of the clusters. These new facets have contact angles of around 11°, which is indicative for $\{1\,1\,7\}$ facets. The rest of the clusters remain dome like. After the capping with 5 ML of Si, the clusters totally change their shape. Only facets with contact angles around of 8° can be seen, as they are typically for hut clusters. In addition a (0 0 1) top facet is detected. This indicates a shape transition from a dome cluster to a truncated hut cluster after the deposition of 5 ML of Si. The evolution process seems to be continuous.

During overgrowth the height of the domes decreases and the diameter increases. Consequently material is transported from the top of the initial dome cluster down to the pedestal of the newly formed hut cluster. Further support for this can be drawn from the elementary map of Si shown in Fig. 2. For the uncapped cluster, an almost uniform composition of the Ge cluster throughout the entire cluster is detected from the EFTEM. The relatively high amount of Si in these clusters is in good agreement with previous studies and can be explained by the high growth temperature of 520°C used in this study [5,11]. After capping the samples with Si, a strong alloying can be seen at the newly formed pedestals of the clusters (area A), indicated by the bright colour. While for the sample capped with 3 ML of Si the Ge composition throughout the former dome cluster seems to be constant (line B), the shape change initiated by the coverage with 5 ML leads to a diffusion of Si into the parts of the former cluster (line C). After the shape transformation from a dome to a hut cluster the core of the newly formed hut cluster remains Ge rich, as indicated by the dark colour. It is noticeable that no pure Si layer is detected on top of the clusters. From the TEM pictures it is concluded, that capping Ge clusters with small amounts of Si leads to a shape transition, which increases the diameter and decreases the height. In addition intermixing with Si occurs.

The STM pictures confirm the results of the TEM investigations. The sample without a cap layer is grown in a regime, where dome and hut clusters exist simultaneously, as can be seen in Fig. 3. The density of clusters is 5.3×10^{9} cm^{-2}. Especially, a low density of huts, as indicated by the ratio of huts to domes of 18:87, is established. For the capped samples in Fig. 4 no more dome clusters are visible. From the TEM pictures we know, that a shape transition from domes to huts has happened. The STM pictures proof that this occurs not only for some of these domes, but for all of them. In particular the different growth conditions for the TEM and STM samples indicate, that

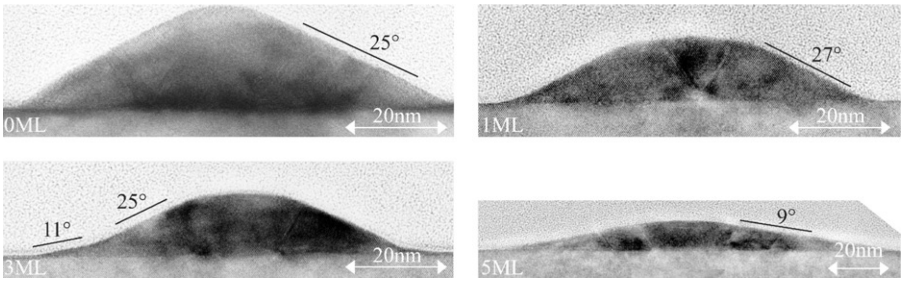

Fig. 1. HRTEM pictures of an uncapped sample and samples with a 1, 3, and 5 ML thick Si-cap. The cluster on the uncapped sample is dome-like, while the clusters on the samples with cap layers change the shape to a hut cluster in the case of the 5 ML thick cap.

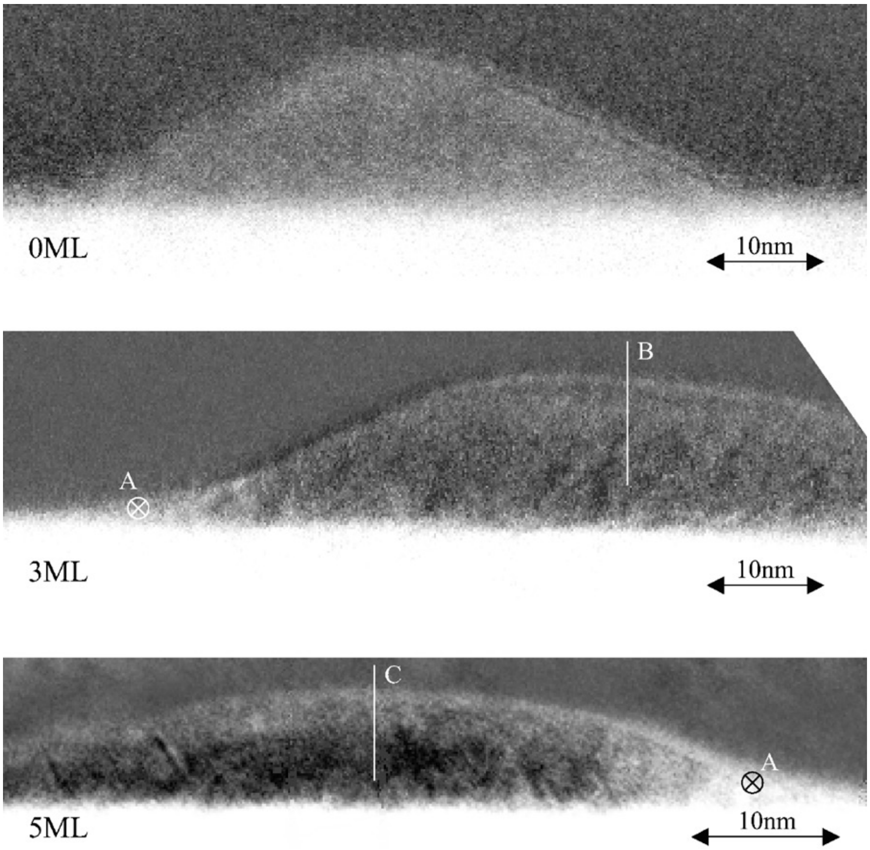

Fig. 2. Si elementary map made by EFTEM. White stands for pure Si. The colours are not calibrated. The uncapped sample has a uniform Si and therefore uniform Ge distribution. After capping with 3 ML of Si, the new-formed facets at A are highly intermixed with Si. With a cap of 5 ML, the Si penetrates into the dot itself.

this phenomenon exists in a large range of growth conditions. For the sample with a 5 ML Si cap the density of clusters has slightly increased to 7.2×10^9 cm^{-2}, compared to the uncapped sample. However, this might be within the statistical variations of the dot density on the sample.

To study the effect of overgrowth on both types of clusters, huts and domes, the height and diameter

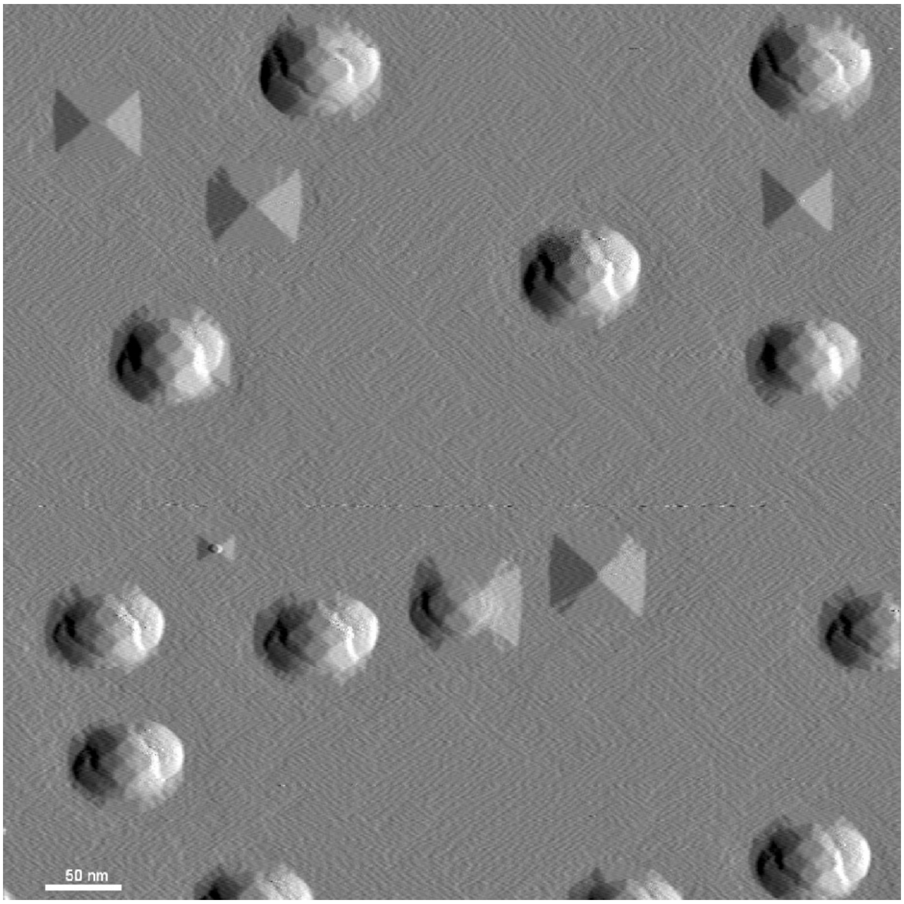

Fig. 3. STM picture of uncapped sample. Two types of clusters are visible, domes and huts. The ratio of domes to huts is 87:17.

distributions were investigated. Fig. 5 shows that on the uncapped sample the height of the huts is by a factor of two smaller than the height of the dome clusters. As the height of huts and the domes are in two different ranges, they can clearly be distinguished using statistical analysis of the STM data. The shape transition induced by capping of the dome clusters can clearly be seen in the height distribution of the sample with a cap of 5 ML of Si. Due to the capping the values of the average heights are smaller, i.e. comparing the distribution ranges, the uncapped clusters have a height of 3.8–11.5 nm and the capped of 2.9–8.9 nm. In particular the transition from dome to hut clusters can clearly be seen, the mean height of the uncapped domes (9.7 nm) is not within the distribution range of height of the capped clusters (2.9–8.9 nm). The effect of overgrowth of initial hut clusters cannot be identified, as their height range (3.8–7.6 nm) is completely within the distribution range (2.9–8.9 nm) of the capped clusters. But the EFTEM shows an intermixing of the clusters, which lets us assume, that the volume of the huts increases due to the intermixing and that therefore the height should become larger.

In Fig. 6 the diameter distribution is shown. The diameters are determined by an automated routine, which approximates the diameters of domes and also of the square shape of hut clusters by a circle touching the outermost edges of the clusters. For the hut clusters, the value of the circular diameter is divided by the square root of two, to get the side length of the square base. In the distribution it is not possible to distinguish the huts and domes by the values itself. Domes

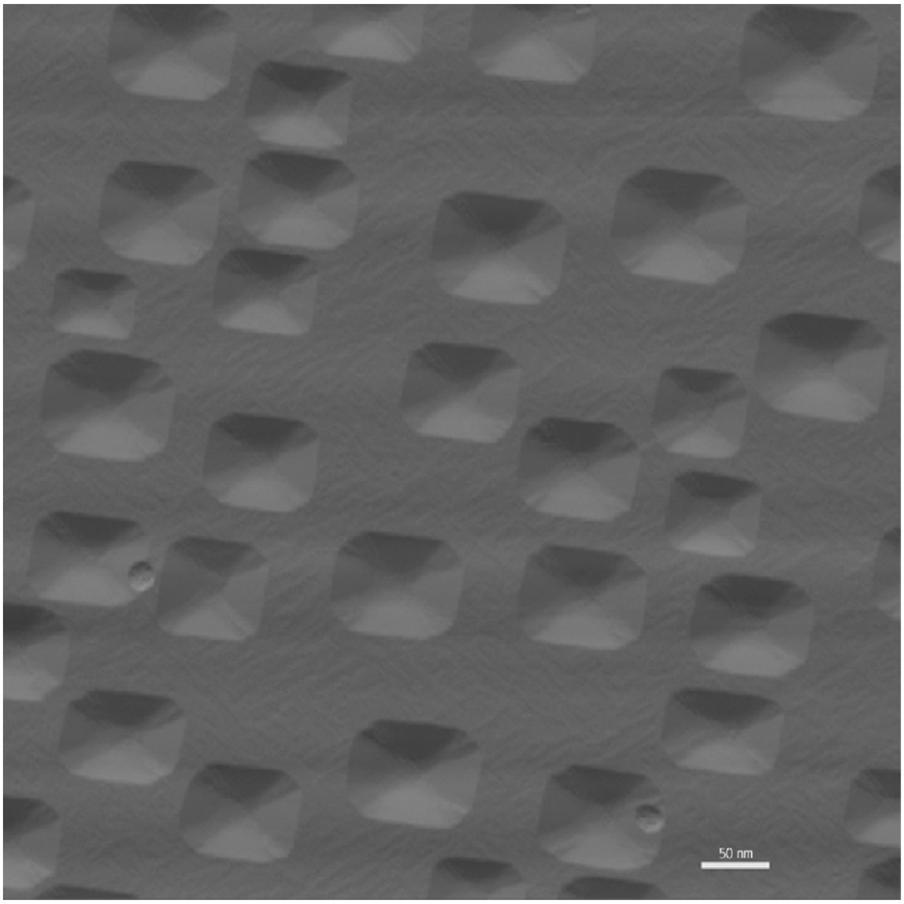

Fig. 4. STM picture of a sample with a 5 ML thick Si cap. Only hut clusters can be found. The capping with Si leads to a shape transition from dome to hut clusters.

have steeper facets and therefore the ratio of diameter to height is smaller compared to a hut. For huts, the diameter to height ratio is 10:1, thus a small change in volume is manifested in a comparatively large change in diameter. Consequently a rather broad distribution of the diameters of hut clusters is observed, which overlaps with the diameters of the dome clusters.

The average diameter of clusters after capping with a Si layer is larger as shown in Fig. 6b. For comparison the range of cluster diameters on the uncapped sample (29.3–67.5 nm) is shifted to higher values (44.7–95.1 nm) due to the capping. The mean value of 67.5 nm for capped clusters is larger than those determined on the uncapped sample for huts (45.3 nm) and domes (57.1 nm). Again, this confirms the shape transition from dome to hut clusters and the increase in size connected with this. Clearly material (Ge) from the top of the domes is transported to the pedestals and hut clusters are formed. This results in smaller heights and larger diameters. It can also be seen, that the diameter of the initial uncapped huts increases due to the capping layer. As the diameter of a hut cluster is in a certain ratio to the height (10:1), the increase in diameter leads to a larger height for the formerly uncapped hut clusters and therefore to an increase in volume, clearly indicating intermixing with Si.

The STM picture indicates, that the shape transition from dome to hut clusters occurs for all domes of the samples grown under the used conditions. The

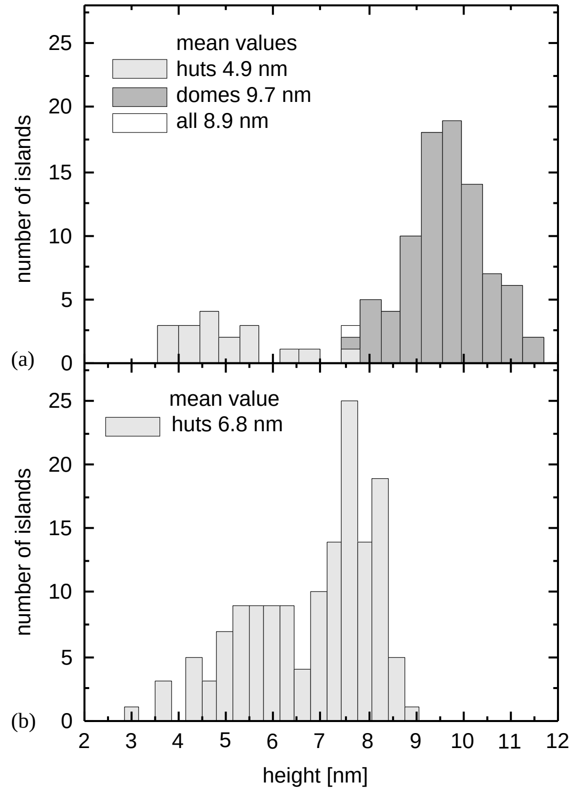

Fig. 5. Height distribution of the uncapped sample (top) and the capped sample (bottom). The shape transformation from domes to huts is accompanied by a decrease in height.

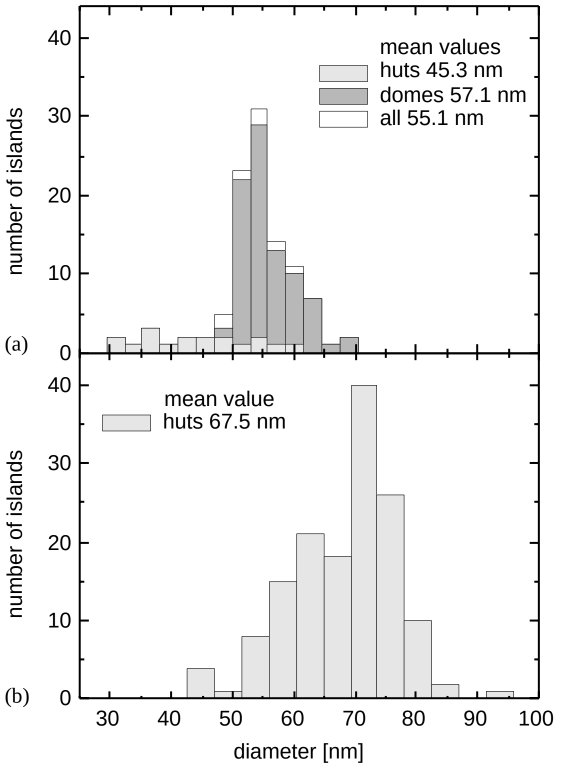

Fig. 6. Diameter distribution of the uncapped sample (top) and the capped sample (bottom). The diameter range shifts to higher value due to the capping. The shape transition of domes to huts and the intermixing of the initially uncapped hut clusters with Si can explain this.

height and diameter distributions let us conclude, that the volume of the hut clusters increases due to the capping.

In order to put these observations in perspective, one has to realise that the Ge clusters on Si surfaces are formed by the Stranski–Kastranov growth mode to reduce the strain. Thus the lattice constant in the Ge wetting layer is Si like, whereas on top of the Ge clusters it is Ge like. Si deposited on the top of the clusters will be under tensile strain. To minimise the strain, Si will intermix with the Ge, consequently, the Ge like lattice constant on top of the cluster shrinks due to the intermixing. In turn, the driving force for the formation of clusters, the relaxation of the strain stored in the Ge film, is reduced and energetically it becomes more favourable to form shallow, flat clusters, due to the reduction in surface energy. Thus even though the Si overgrowth leads to a narrower distribution in size, going from a bimodal to a mono-modal distribution, the loss in confinement due to intermixing will deteriorate the optical properties. Hence much lower deposition temperatures have to be used, to suppress intermixing and material transport from the top to the pedestals of the islands.

Acknowledgements

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