Intermixing and Shape Transitions of PbSe Quantum Dot during Overgrowth

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The size and shape changes of self-assembled PbSe quantum dots during overgrowth by $Pb_{1-x}Eu_xTe$ barrier layers with different Eu concentrations is investigated using atomic force microscopy and in situ reflection high-energy electron diffraction. It is shown that whereas for overgrowth with pure PbTe a very strong intermixing of the PbSe dots with the cap layer material occurs, this effect is drastically suppressed with increasing Eu concentration in the $Pb_{1-x}Eu_xTe$ cap layers. As a result, the size and shape of the as-grown PbSe dot is essentially conserved during the capping process, which is important for opto-electronic device applications.

Introduction

Self-assembled semiconductor quantum dots synthesized via the Stranski-Krastanow growth mode have attracted great interest due to their excellent electronic and optical properties. For applications, quantum dots have to be embedded in a matrix material to avoid surface recombination effects. However, it is well known that *e.g.* for InAs dots embedded in GaAs there is significant intermixing between dot and matrix material [1] – [4], which leads to remarkable changes in the dot size and shape for the overgrown dots [5]. Similar effects have been also observed for SiGe dots overgrown by silicon layers, and the degree of intermixing was found to depend strongly on the growth conditions [6]. Since the optical and electronic properties depend crucially on size and shape of the embedded dots it is important to gain detailed knowledge about the changes of these parameters due to capping process.

Experimental

In the present work, we have investigated the overgrowth behaviour of self-assembled PbSe quantum dots grown by molecular beam epitaxy (MBE) on PbTe (111). The PbSe dots are formed due to strain-induced coherent islanding (–5.4 % lattice-mismatch) once the critical coverage of 1.5 PbSe monolayers (ML) is exceeded. The surface islands have a well-defined pyramidal shape with triangular base and with (100) side facets. For the overgrowth studies, a series of dot samples was prepared under identical conditions with a total PbSe thickness of 5 monolayers. Then the PbSe dots were overgrown with $Pb_{1-x}Eu_xTe$ cap material of different Eu concentration and different cap thicknesses. The evolution of the surface morphology was determined using *in situ* reflection high-energy electron diffraction (RHEED), as well as *ex situ* atomic force microscopy (AFM) under ambient conditions after rapid quenching of the partially capped samples to room temperature.

Results

In a first set of experiments, the evolution of the RHEED patterns during overgrowth of PbSe quantum dots predeposited on a PbTe buffer layer at Ts = 360 °C was investigated. Figure 1 (a) and (b) shows examples of the RHEED patterns before and after dot overgrowth. The initial average PbSe dot height of the samples before overgrowth was 105 Å as determined from reference samples. For a more detailed analysis, the integrated intensity of the 3D (224) diffraction spot was measured as a function of cap layer thickness for several different $Pb_{1-x}Eu_xTe$ cap layer compositions as shown in Fig. 1 (c). The exact position of the (224) spot is marked in the RHEED images in (a) and (b). As is evident from Fig. 1 (c), the 3D (224) diffraction spot intensity is maximal for the initial surface with 5 ML PbSe coverage. During overgrowth, the diffraction spot rapidly decreases, and the RHEED patterns transform into the usual streaked diffraction pattern corresponding to the reformation of a planar 2D surface. This indicates that a rapid replanarization takes place in all cases. However, the cap layer thickness required for complete planarization, i.e., the thickness when the 3D spot has disappeared, strongly increases as a function of the Eu content in the ternary cap layer, indicating that the planarization process is much slower for the cap layers with higher Eu content.

Similar results were obtained from the AFM images of the partially capped samples shown in Fig. 2. In this case, the dots were overgrown with different cap layer thickness of $Pb_{1-x}Eu_xTe$, x = 0.065 and pure PbTe, respectively. In Fig. 2, left-hand side, the AFM surface images of PbSe dots overgrown with $Pb_{1-x}Eu_xTe$, x = 0.065 at cap layer thicknesses of 15 Å, 35 Å, 50 Å and 85 Å are shown for (a) – (d), respectively. At a cap thickness of 50 Å, the partially capped quantum dots are still clearly visible. For a cap thickness of 85 Å, the dots are fully overgrown although the initial dot height before overgrowth was 105 Å. This indicates that a certain intermixing between dot material and matrix material takes place and reduces the remaining dot height during capping. A closer look shows that the dots have not only vanished completely, but there are small holes on the surface, which have approximately the same density as the buried dots. From STM studies it is known that these holes are due the lattice deformations, which are caused by the highly strained buried dot.

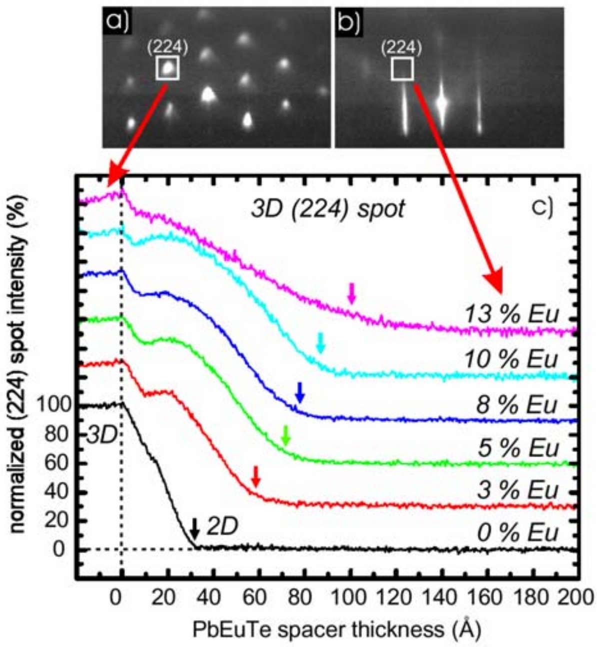

Fig. 1: Top: RHEED pattern of (a) 3D surface with PbSe quantum dots before overgrowth and (b) after complete overgrowth with $Pb_{1-x}Eu_xTe$ spacer layer. Bottom: Normalized intensity of (224) spot as a function of $Pb_{1-x}Eu_xTe$ spacer thickness for x = 0, 0.03, 0.05, 0.08, 0.1, and 0.13. Each curve has a relative offset of 20 for clarity.

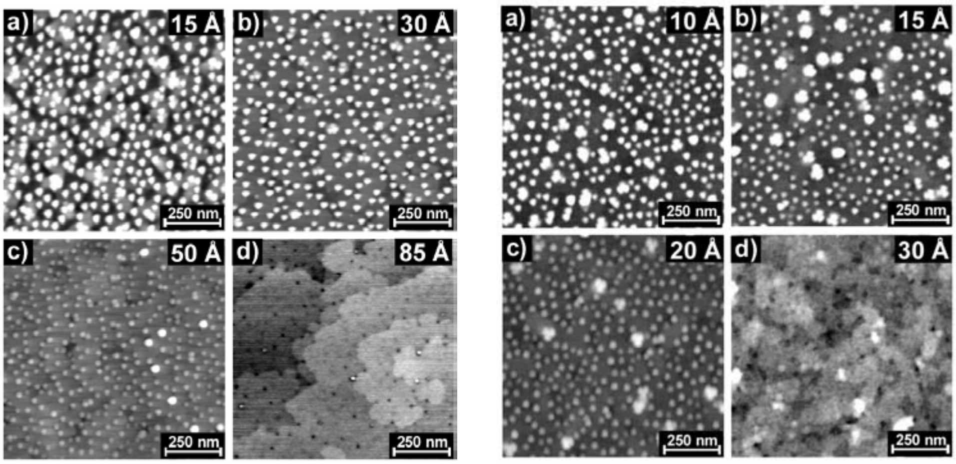

Fig. 2: Left hand side: Atomic force microscopy surface images (1 x 1 μm^2) of 5 ML PbSe quantum dot layer on PbTe buffer layer overgrown with different $Pb_{1-x}Eu_xTe$ (x = 0.065) spacer layer thickness of (a) 15 Å, (b) 35 Å, (c) 50 Å and (d) 85 Å. Right hand side: Atomic force microscopy surface images (1 x 1 μm^2) of 5 ML PbSe quantum dot layer on PbTe buffer layer overgrown with different PbTe spacer layer thicknesses of (a) 10 Å, (b) 15 Å, (c) 20 Å, and (d) 30 Å.

In Fig. 2 right-hand side (a) – (d), AFM surface images of partially overgrown dots with pure PbTe at a cap layer thickness of 10 Å, 15 Å, 20 Å, and 30 Å are shown respectively. In this case, the dots can only be detected up to a cap layer thickness of 20 Å. At 30 Å cap thickness the dots are fully overgrown, indicating that for pure PbTe the intermixing between dot material and matrix material is even more pronounced. Also for pure PbTe there are small holes detected on the surface when the dots are fully overgrown.

From the statistical evaluation of the AFM images, the remaining PbSe island height was determined as a function of the cap thickness, where the result is shown in Fig. 3. The residual dot height is plotted as a function of cap layer thickness d for x = 0, 0.03, 0.065, and 0.1. The dashed line gives the theoretical curve for the dot height, when full shape conservation is assumed and if the $Pb_{1-x}Eu_xTe$ capping material grows only around the dots and not on top. In this case the residual dot height h should be just equal to $h = h_0 - d$ with $h_0 \approx 105$ Å being the initial dot height before capping. Although, in all cases the dot height decreases essentially linearly with increasing cap thickness, the critical cap layer thickness required for planarization increases from only 30 Å for PbTe capping to about 100 Å for capping with $Pb_{1-x}Eu_xTe$ with x_{Eu} = 10%. The data obtained for x_{Eu} = 10% is remarkably close to the theoretical curve, indicating that the dot shape is almost completely conserved. For x = 0.065, there is already a clear deviation from the theoretical curve. The overgrown dots are clearly smaller than what would be expected in the case of shape conservation even for a thin cap layer thickness. It is evident that there is only little overgrowth on the top of the dots, and the intermixing at the dot base leads to further decrease in the remaining dot height. There is no evidence for discontinuities in the overgrowth and intermixing process since the measured residual dot heights follow a straight line. For x = 0.065, the dots have vanished completely after a cap layer thickness of 70 Å was deposited. For Eu contents of x = 0.03 and 0, the dots vanish already at a cap thickness of 56 and 32 Å, respectively. This indicates that while a very strong intermixing and dissolution of the initially 105 Å high dots takes place during pure PbTe overgrowth, this effect is strongly suppressed by the presence of Eu in the cap layer, *i.e.*, for sufficiently high Eu concentrations the dot shape is preserved during the overgrowth process.

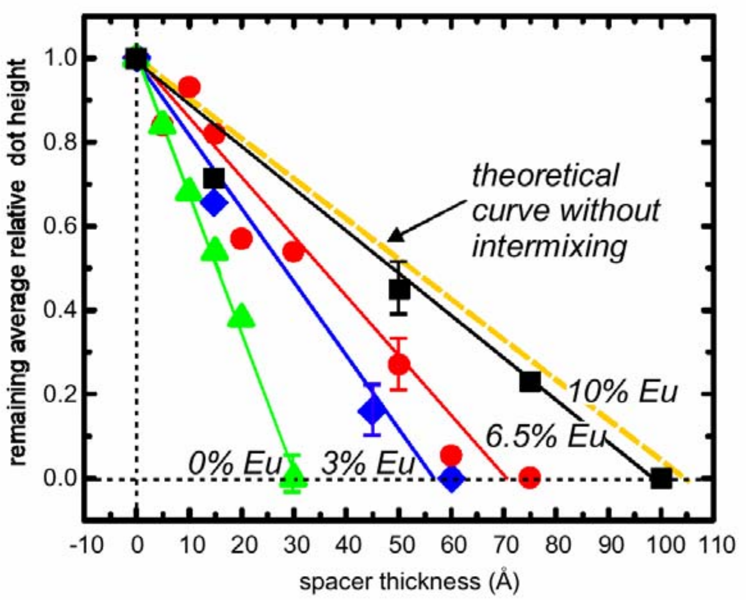

Fig. 3: Remaining average relative dot height as a function of $Pb_{1-x}Eu_xTe$ spacer thickness for different Eu contents of x=0 (▲), x=0.003 (◆), x=0.065 (●), and x=0.1 (■). The dashed curve is the theoretical curve without intermixing.

Conclusions

In conclusion, we have investigated the overgrowth behaviour of self-assembled PbSe quantum dots by $Pb_{1-x}Eu_xTe$ barrier layers with different Eu concentrations. Whereas for overgrowth with pure PbTe a very strong intermixing of the PbSe dots with the cap layer material occurs, this effect is drastically suppressed with increasing Eu concentration in the $Pb_{1-x}Eu_xTe$ cap layers. As a result, in this case the size and shape of the as grown PbSe dot is essentially conserved during the capping process. This is important for optoelectronic device applications.

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