Molecular Beam Epitaxy Growth of 3D Quantum Dot Crystals

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The self-organization of pyramidal PbSe islands that spontaneously form during strained-layer epitaxial growth of PbSe/PbEuTe superlattices results in the formation of three-dimensional quantum-dot crystals. In these crystals, the dots are arranged in a trigonal lattice with a face-centered cubic (fcc)-like ABCABC... stacking sequence. As shown by theoretical calculations, the elastic anisotropy in these artificial dot crystals acts in a manner similar to that of the directed chemical bonds of crystalline solids.

1. Introduction

The spontaneous formation of nano-scale three dimensional (3D) islands during strained-layer heteroepitaxy has recently emerged as novel technique for fabrication of self-assembled quantum dots. It is based on the fundamental instability of highly strained surfaces that leads to nucleation of coherent islands on the surface after completion of the wetting layer. In multilayer structures, the buried dots tend to influence the dot nucleation in the subsequent layers due to long range elastic interactions. This usually results in a vertical alignment of the dots, but may also lead to an ordering in the lateral direction. This could lead to improved quantum dot size homogenities, which is of crucial importance for device applications.

2. Experimental Results

In the present work, we have investigated the evolution of vertical and lateral correlations in self-assembled PbSe/PbEuTe quantum dot superlattices grown by molecular beam epitaxy on PbTe (111) surfaces. In these superlattices, the composition of the ternary layer was adjusted to achieve a complete strain symmetrization of the superlattice stack, which allows the growth of an unlimited number of superlattice periods without misfit dislocation formation. Atomic force microscopy (AFM) investigations (Fig. 1) indicate a nearly perfect hexagonal ordering of the PbSe quantum dots within the layers as the number of superlattice periods increases. From high resolution x-ray diffraction and transmission electron microscopy (TEM) studies we find that the vertical correlation of the dots is not parallel but inclined with respect to the growth direction. As a consequence, an ABCABC... vertical dot stacking sequence is formed, similar to the stacking of the closely packed (111) planes in fcc-lattices.

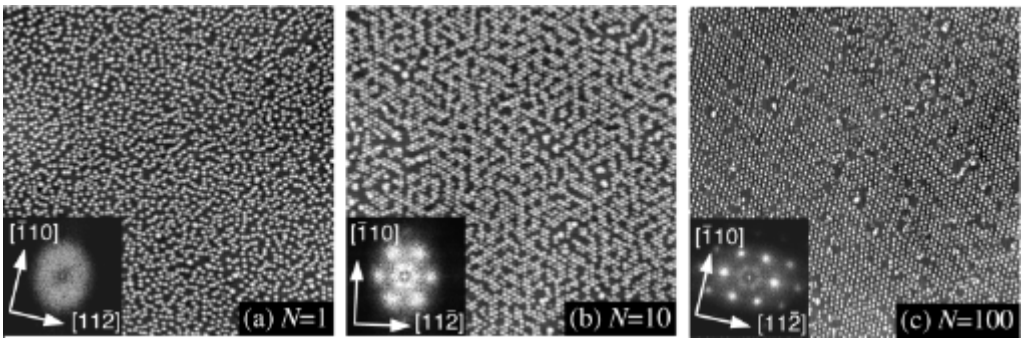

Fig. 1: AFM images of (a) a single PbSe quantum dot layer and of the top PbSe layer of strain symmetrized PbSe/PbEuTe dot superlattices after (b) 10 and (c) 100 periods. This indicates a rapid evolution of hexagonal ordering within the dot layers, which is evident also from the 2D power spectra of the AFM images shown in the inserts.

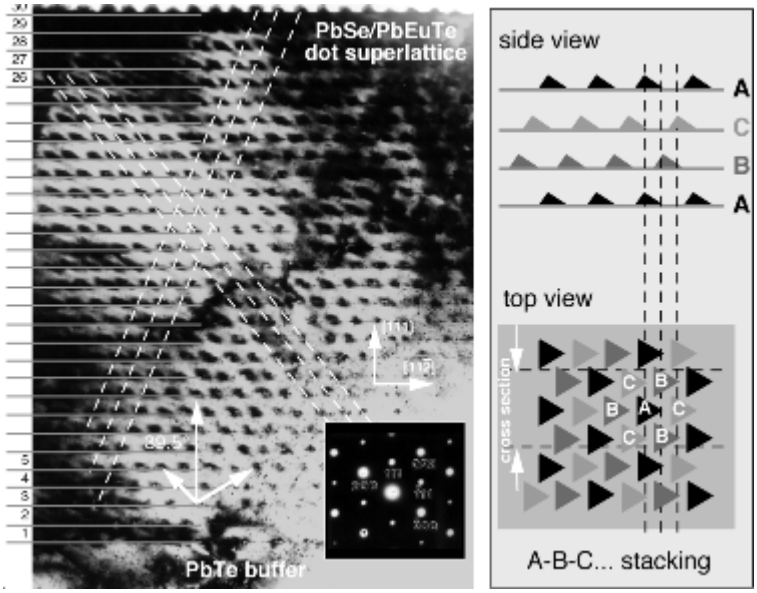

Fig. 2: Cross-sectional TEM image of a 30 period PbSe/PbEuTe quantum dot superlattice showing the high degree of lateral and vertical order in the structure that starts already within the first few superlattice periods. The PbSe thickness was 5 monolayers and the PbEuTe spacer thickness 480Å in this sample. The right hand panel shows the fcc-like ABCABC vertical dot stacking sequence deduced by TEM as well as by high resolution x-ray diffraction.

As shown in the cross-sectional TEM image in Fig. 2, the ordering in the dot superlattices is so efficient that nearly perfect trigonal quantum dot crystals are formed. Even more, for different superlattices we find that the trigonal angle of the dot lattice remains essentially constant, which means that the in-plane dot-dot separation can be tuned continuously just by changing the spacer thickness.

3. Theoretical Calculations

In order to explain the 3D dot arrangement in the superlattices we have developed a model for calculation of the elastic energy distribution on the surface above buried Stranski-Krastanow islands taking into account the elastic anisotropy as well as surface relaxation. For (111) growth of the highly anisotropic IV-VI materials we find that the energy minimum is not directly above a buried dot but instead three side minima occur. As shown in Fig. 3a, these minima are laterally displaced along certain crystallographical directions and serve as preferential nucleation sites for the dots in the subsequent layer. This leads to the ABCABC... vertical stacking as well as the in-plane hexagonal ordering that is observed in our experiments.

Similar calculations were performed for other semiconductors and other growth directions. We find a systematic dependence of the strain distributions on the elastic anisotropy ratio $A = 2c_{44}/(c_{11}-c_{12})$ that reflects the ratio between the Youngs moduli along the <111> and <100> directions. For rock salt type materials with $A < 1$, an ordering similar to the PbSe/PbEuTe system is predicted, with a linear increase of the trigonal correlation angle when the elastic anisotropy increases (Fig. 3c). For the diamond and zinc-blende type semiconductors with $A > 1$, side minima appear only for the (001) surface orientation, which could result in a centered tetragonal dot arrangement (ABAB... stacking) if the elastic anisotropy is sufficiently large (Fig. 3b).

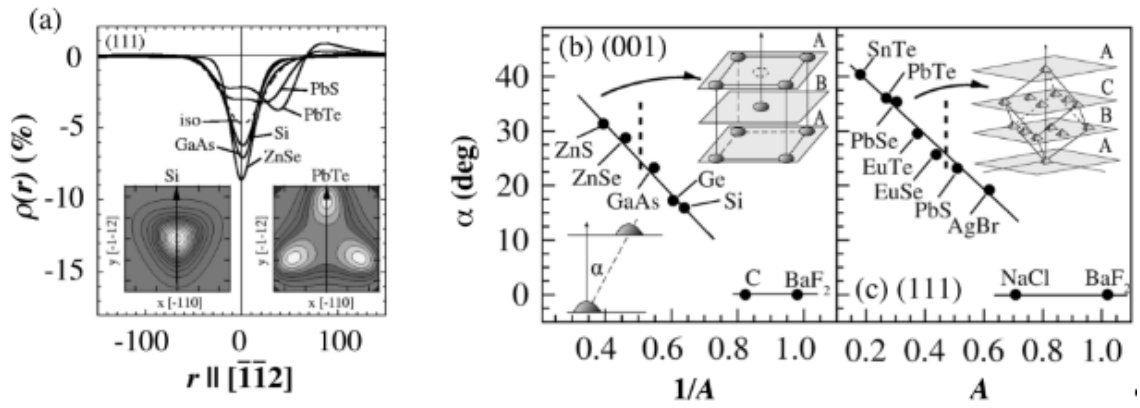

Fig. 3: (a) Normalized elastic energy density $\rho(x,y)$ above a strained quantum dot at 50 nm below the surface for various matrix materials and the (111) surface orientation. The curves represent cross-sections of ρ in the directions r indicated by arrows in the 2D contour plots. (b) Angle α relative to the surface normal under which the minima of $\rho(x,y)$ appear on the (001) and (111) surfaces vs. elastic anisotropy ratio $A = 2\,c_{44}/(c_{11}-c_{12})$. The inserts show the expected dot superlattice stacking.

References

[1] G. Springholz, V. Holy, M. Pinczolits, and G. Bauer, Science 282, 734 (1998).