Analysis of Nanostructures by EM Techniques: Quantum Dots

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Summary: Nanostructures have gained increasing interest in materials science due to their special mechanical, electrical and optical behavior. The correlation between these extraordinary properties and the morphology and crystal structure can be analyzed by combining numerous electron-optical techniques. The present contribution demonstrates the successful application to semiconductor quantum dots. TEM techniques is focused on the determination of parameters, such as shape and size of islands, their chemical composition and the complex lattice strain fields. A successful image contrast analysis in terms of shape and strain demands the application of image simulation techniques based on the many-beam dynamical theory and on structure models refined by molecular dynamics or molecular static energy minimization.

Nanostructures are incorporated in various materials as a very interesting matter since they are characterized by properties, which strongly differ from those of corresponding bulk materials. Having a size of \( \leq 100 \) nanometers, for instance, tubes, (spherical) clusters, pyramids, and precipitates occur, showing a strong correlation between their specific properties and their morphology, size and embedding. Different electron-optical techniques have been applied successfully to analyze such structures in the nm-range: high-resolution transmission electron microscopy (HREM), conventional diffraction contrast (TEM), chemical analysis by EDX, and energy-filtered electron microscopy (EFTEM). As an example, we would like to demonstrate the possibilities of such combined investigations for the case of nanostructures in semiconductor materials, viz. the so-called quantum dots (QDs). For the InGaAs system, the scientific interest and technological aim are focused on QD arrays emitting light of about 1.3 to 1.5 \( \mu \)m, controlled by the size, shape, lattice strain, and chemical composition of the QDs, and thus strongly influenced by the thermodynamics and kinetic processes during the growth of QD samples. The possibility of arranging such semiconductor islands into complex arrays implies many opportunities for scientific investigations and technological applications (for an overview, see, e.g., [1]). The imaging of microscopic structure variations in and around QDs by HREM or TEM is well known and routinely used, but the analysis of HREM and TEM images of such nanostructures demands a structure modelling on an atomic scale valid for microscopically relevant dimensions and the application of image simulation or image matching techniques (see, e.g., [2,3] for QD image analysis) to separate the different factors controlling the physical properties.

Size and distribution: Fig. 1 shows a typical plan-view TEM image of InAs dots on a GaAs substrate. Due to the conventional diffraction contrast technique applied the dots are detectable mainly by their strain fields. From contrast analyses it can be concluded that the dots seem to have a quadratic base face and edges along <100> directions. Such images allow one to determine not only the size and dot density (here about \( 10^7 \) cm\(^{-2} \)), but also the relation between adjacent islands. A careful analysis of bright-field TEM contrasts revealed that, under varying growth conditions, the islands may occur as lens-shaped inclusions with a circular base [4] or having a more rectangular or rhombohedral base (see also Fig.2).

Chemical composition: The growth process and the post-growth annealing of the samples imply an interdiffusion of the elements between matrix and island, which influences the energy states of the excitons. The change in stoichiometry as, for instance, the In/Ga ratio is revealed as an integral measurement using PL spectroscopy. Several approaches have been made to determine the element distribution in the nm-range by image processing of HREM micrographs (see, e.g., [5-7]). The application of these techniques to the analysis of QD structures is partly restricted. Hence, first, one has to eliminate the lattice distortions, which would disturb the analyzing process. Fig.3 gives an example, where the In distribution has been analyzed in the island as well as in the InAs wetting layer.

Structural modelling: While, in principle, it is now possible to predict material properties by using quantum-theoretical ab initio calculations with a minimum of free parameters, the only method of simulating time-dependent atomic processes with macroscopic relevance is the molecular dynamics (MD) method solving Newton’s equations of motion for a molecular system and using
suitably fitted many-body empirical potentials, preferably of the Tersoff/ bond-order type [8,9]. The calculations are done using a constant volume (NVE ensemble) or a constant pressure (NpT ensemble) and time steps of the order of 0.25 fs to ensure the proper calculation of all modes.

Fig. 1 Bright-field TEM image of a single layer of InAs dots grown on a <001> GaAs substrate by the MBE growth technique.

Fig. 2 Bright-field, plan-view images of different samples. Left: capped InGaAs island, right: uncapped SiGe island.

Fig. 3 Analysis of a single layer of InAs QDs. Upper part: cross-section image. Middle part: HREM image of the wetting layer (WL) consisting of 4 monolayers of InAs. Lower part: scheme of the inhomogeneous In distribution in the island. The chemical analysis (distribution of In) is obtained from image processing (chemical sensitive {200} reflection).

Alternatively to MD for structures near the equilibrium, static energy minimization may be performed using steepest descent or conjugate gradient methods to relax the structures towards one of the nearest local energy minima. In Fig. 4, typical structure models are shown (Fig.4a: complete model with embedded pyramid, Fig.4d: examples of pyramids with facets {011}, {112}, {113}, and {136}) together with HREM - image simulations (Fig.4b,c: 400 kV parameters, see below, and thickness t= 9 nm) before and after relaxation, to demonstrate the striking influence of relaxation on the image contrast. The relaxation was calculated under periodic boundary conditions at the side faces of the GaAs supercell, with the InAs pyramids inside. Nevertheless, the difference of the energy before and after relaxation is rather small, the different wetting layers, truncations, and step structures of the facets varying due to their different inclination, yield characteristic strain fields.

**Image simulations:** The image simulations are performed via multi-slice calculations, starting with MD relaxed supercells which are sliced so that at least 4 subslices per unit cell in <100> direction are used. The imaging parameters are chosen according to two typical microscopes, viz. the JEOL 4000EX at an accelerating voltage of 400kV used for high resolution microscopy, and the Philips CM20 FEG with 200kV, equipped with a field emission gun: accelerating voltage U=400kV (200kV), spherical aberration Cₐ=1.2mm (1.0mm), defocus spread δ=8nm (5nm), and beam divergence α=0.05mrad (0.03mrad). The imaging aperture is varied between 2 nm⁻¹ and 6nm⁻¹ to have sufficient information of the QD itself and to avoid image artefacts. Fig. 5 shows the simulated zone-axis bright-field contrast of QDs with different facets (cf. Fig. 4 for the shapes) at 200kV and for two
different thicknesses of $t=7.4\text{nm}$ in (a) and $13.6\text{nm}$ (b), to demonstrate the contrast reversal with increasing thickness. Fig. 6a shows 400kV simulations of pyramidal QDs of the $\{011\}$ type in comparison with QDs shaped as spherical segments in Fig. 6b for different sample thicknesses, depths and defect sizes to characterize the contrast variations.

Fig. 4. Pyramidal-shaped InAs QDs in GaAs after applying MD-relaxations (a: complete atomic model, $\{011\}$ facets, d: different facetting wetting, and truncation, but matrix removed) and simulated [001]-HREM images of QDs before (b) and after (c) relaxation (400 kV, $C_s=1\text{ nm}$, $\alpha=1.2\text{nm}^{-1}$, $t=9\text{ nm}$, Scherzer focus $\Delta=40\text{ nm}$).

Fig. 5: Simulated 200kV bright-field diffraction contrast TEM images (with 7.4nm edges parallel to <110>) of QDs of different pyramidal shapes, truncation and relaxation using two sample thicknesses of $t = 7.4 \text{ nm}$ (a) and $13.6\text{nm}$ (b).

Fig. 6: Simulated 400kV bright-field diffraction contrast TEM images of $\{011\}$ pyramids (a) and spherical segments (b) for different thicknesses $t$, depth $t_0$ of the QD below the surface and defect sizes $d$, characterizing the base length of the pyramid or the base diameter of the segments, given as approximated parameter set by $(t/\text{nm}, t_0/\text{nm}, d/\text{nm})$. 

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**Figures:****

- **Fig. 4:** Pyramidal-shaped InAs QDs in GaAs after applying MD-relaxations (a: complete atomic model, $\{011\}$ facets, d: different facetting wetting, and truncation, but matrix removed) and simulated [001]-HREM images of QDs before (b) and after (c) relaxation (400 kV, $C_s=1\text{ nm}$, $\alpha=1.2\text{nm}^{-1}$, $t=9\text{ nm}$, Scherzer focus $\Delta=40\text{ nm}$).

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- **Fig. 6:** Simulated 400kV bright-field diffraction contrast TEM images of $\{011\}$ pyramids (a) and spherical segments (b) for different thicknesses $t$, depth $t_0$ of the QD below the surface and defect sizes $d$, characterizing the base length of the pyramid or the base diameter of the segments, given as approximated parameter set by $(t/\text{nm}, t_0/\text{nm}, d/\text{nm})$. 

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The simulations reveal that there are imaging conditions that allow a clear distinction of the different QDs by their characteristic contrast features due to their strain fields. Without strains solely a weak structure factor contrast arises. The pyramidal QDs with a steeper descent of the facets show a fourfold symmetry of their contrast features. The striations superimposed show that some of the structures are not yet completely relaxed, their contrast depends sensitively on the objective aperture.

**QD arrays:** To improve the QD structures (higher dot density, more pronounced size distribution) promising solutions have been developed, two of which will be discussed. First, the multiple stacking of QD layers has been envisaged as an attractive growth concept to provide a 3-dimensional array of islands (see, as an example: Fig.7). It has been applied successfully to several systems of semiconductors [10]. A further possibility of attaining an emission at longer wavelengths (> 1.3 µm) has been demonstrated by generating associated QDs [11]. For MBE growth at lower temperatures the QDs are arranged as complex arrays (Fig.8): laterally linked islands containing 3 to 15 dots.

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References