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Max-Planck-Institut für Mikrostrukturphysik, Halle/Saale[1])

TEM/HREM Visualization of nm-Scale Coherent InAs Islands (Quantum Dots) in a GaAs Matrix

By

S. RUVIMOV[2]) and K. SCHEERSCHMIDT

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Dedicated to Professor Dr. JOHANNES HEYDENREICH on the occasion of his 65th birthday

The electron microscope determination (high resolution HREM and conventional diffraction contrast TEM) of the structure (geometry, size, shape, etc.) of nm-scale objects is of great interest for the creation of novel semiconducting materials of reduced dimensions as, e.g., quantum dots (QDs) and quantum wires. HREM contrast simulations based on molecular dynamics structure modelling are applied to check the visualization of coherently strained nm-scale InAs islands embedded in a GaAs matrix. Being of pyramidal shape, InAs islands always seem to be truncated owing to lower In content on top of the pyramid and to the high level of strains around the island. Optimum imaging conditions are analysed to reveal shape and size of such objects.

Die Untersuchung der Struktur (Geometrie, Größe, Form usw.) von Objekten im Nanometerbereich mittels Elektronenmikroskopie (Hochauflösungsabbildung HREM und konventioneller Beugungskontrast TEM) hat eine besondere Bedeutung für die Konstruktion neuer Halbleitermaterialien mit reduzierten Dimensionen, wie z. B. Quantenpunkte (QDs) und Quantendrähte. HREM Kontrastsimulationen auf der Grundlage molekulardynamisch relaxierter Strukturmodelle werden diskutiert, um die Abbildungen kohärent gedehnter InAs Inseln von Nanometergröße, die in einer GaAs Matrix eingeschlossen sind, zu analysieren. Die InAs Inseln zeigen stets den Kontrast von Pyramiden, deren Spitze gekappt scheint, was eindeutig auf den geringeren zu durchstrahlenden In-Anteil in der Pyramidenspitze und das starke Verzerrungsfeld um die QDs herum zurückzuführen ist. Es werden optimale Abbildungsbedingungen vorgeschlagen, um dennoch Größe und Form der QDs analysieren zu können.

1. Introduction

Semiconductors having structures of reduced dimensions as, e.g., quantum dots (QDs) and quantum wires are expected to exhibit special optical and electronic properties as, e.g., a δ-function-like distribution of states leading to novel electronic and photonic devices (e.g., lasers [1, 2]). One of the possible ways to form such structures without any patterning process is a self-organized growth of ordered arrays of nm-scale coherent islands on semiconducting surfaces called quantum dots (see, e.g., Ge on Si, InGaAs or InAs on GaAs etc. [3 to 12]). The islands described in the literature [6 to 12] differ in size and shape owing to differences in the growth and measuring techniques applied. The knowledge of reliable data, however, is important in connection with the influence of QDs on the strain distribution and, hence, on the electronic properties of the heterostructure [13].

[1]) Weinberg 2, D-06120 Halle/Saale, Federal Republic of Germany.

[2]) On leave from Ioffe Physico-Technical Institute, 194021 St.-Petersburg, Russia.

However, a powerful method of structure analysis, transmission electron microscopy (TEM), is restricted as to the size and shape determination of small objects, if the size is smaller than the extinction distance ξ_g. In general, three main techniques have been developed to investigate small objects showing diffraction contrast in conventional TEM with improved spatial resolution: weak-beam (WB) dark-field (DF) imaging [14, 15], high-order bright-field (BF) imaging [16, 17], and symmetrical zone-axis BF imaging [18]. The diffraction contrast is always created by both structure factors and strain fields, with the images not only depending on the shape and elastic strength of a defect, but also on elastic properties of the matrix. This implies characteristic features as, e.g., asymmetries in "black–white" or "black lobe" contrasts even for spherical precipitates in an elastically anisotropic matrix [18]. Therefore, a careful contrast interpretation based on image simulations is necessary.

High resolution electron microscopy (HREM) with its possibility of local structure imaging at an atomic level may be considered a critical method to prove the results of conventional TEM techniques. The present paper discusses the application of HREM imaging to pyramidal-shaped InAs islands coherently embedded in a GaAs matrix. Although there is a high misfit of some percent, the islands will be coherent because of their small extension, but creating a high level of strain. Molecular static calculations are used to study the relaxation of atomic models, and HREM images are simulated by means of multislice algorithms to discuss the resulting contrast. Optimum imaging conditions are analysed to reveal the shape and size of such objects which always seem to be truncated pyramids owing to the low In content on top of the pyramid and to the high level of strains around the island.

2. Experiment and Computation Technique

400 kV HREM (JEOL JEM4000 EX microscope) has been applied to investigate quantum dots of 3 ML-thick InAs layers grown by molecular beam epitaxy (MBE) on (001)-oriented GaAs substrates. The growth conditions are described elsewhere (see, e.g. [2, 12]). Both plan view and cross section specimens have been prepared for the structural characterization of the nm-scale InAs islands coherently embedded in a GaAs matrix.

The relaxed atomic structures of the InAs islands are modelled by molecular static energy minimization with the atomic interaction potentials being varied and by using mainly the CERIUS programme package (Molecular Dynamics Simulations Inc., Cambridge; for details and other programmes applied as well as the way to perform image simulations see, e.g., comparable studies of semiconductor multilayer structures in [19]). Owing to the restriction on the total number of atoms, an InAs pyramid of 6 nm base length and 3 nm height in a GaAs crystal of $10 \times 10 \times 10$ nm^3 volume was constructed. In Fig. 1 the structure model is shown (Fig. 1a) together with image simulations (Fig. 1b, accelerating voltage $U = 400$ kV, spherical aberration $C_s = 1$ mm, objective aperture $\alpha_0 = 1.2$ nm^{-1}, thickness $t = 9$ nm, defocus $\Delta = -40$ nm) before and after relaxation, to demonstrate the enormous influence of relaxations to the image contrast. The model relaxation was calculated under periodic boundary conditions at the side faces of the GaAs supercell with the InAs pyramid inside. The force-field including two-, three-, and four-body interactions, viz. distance, angle, and torsion, was parametrized so that the equilibrium crystal structures for pure GaAs and InAs were provided. Molecular statics was applied assuming that the energy difference of

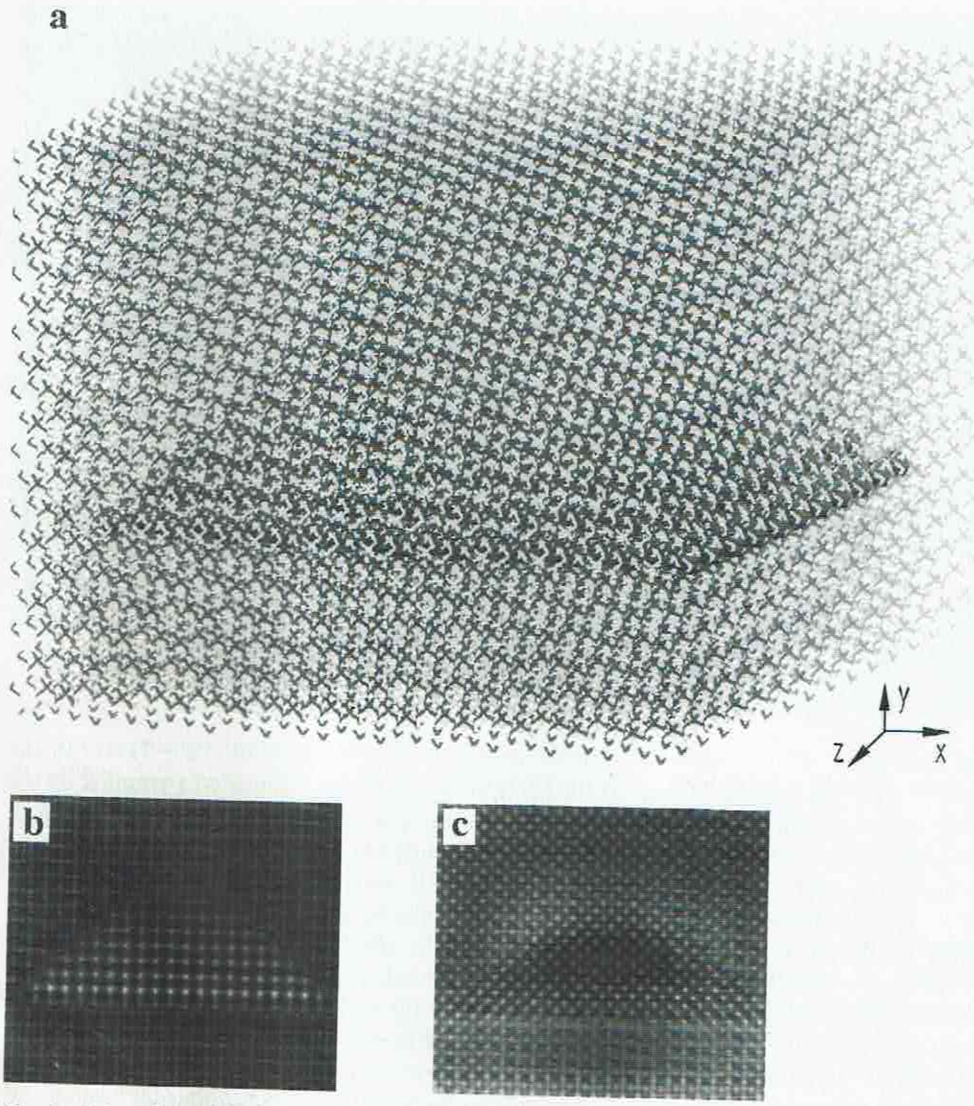

Fig. 1. a) Atomic model of pyramidal-shaped InAs QD in GaAs after applying molecular static energy minimization and simulated [001] HREM images of QDs b) before and c) after relaxation taken at Scherzer focus (400 kV, $C_s = 1$ mm, $\alpha_0 = 1.2\ \text{nm}^{-1}$, $t = 9$ nm, $\Delta = -40$ nm)

the whole structure before and after relaxation is rather small, because the geometrical model (see Fig. 1a) of the coherent embedding of the pyramidal-shaped Ga/In exchange region results in a structure not far from equilibrium.

3. Results and Discussion

Fig. 2a and b show symmetrical [001] zone-axis BF images of InAs QDs in plan view and cross section projection, respectively. The analysis of the micrographs indicates that the dots are of pyramidal shape with the square base of their principal axes being close to the

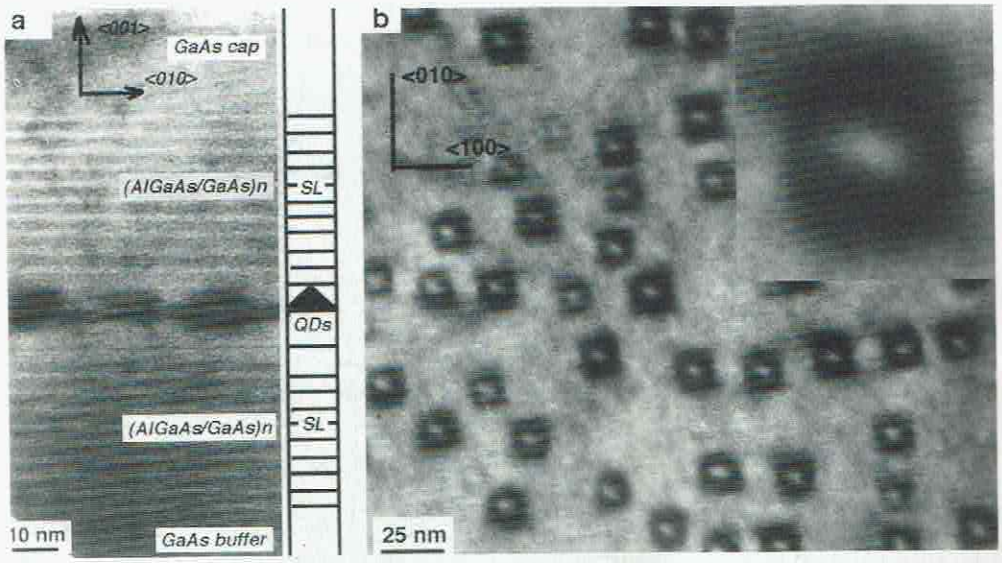

Fig. 2. Symmetrical [001] zone-axis bright-field images of a) cross section and b) plan view projection of the heterostructure with QDs of 3 ML-thick InAs layer grown by MBE on (001)-oriented GaAs substrate. Inserts: a) scheme of the heterostructure and b) a magnified image of a QD

two orthogonal ⟨100⟩ directions and the average length being about (12 ± 1) nm. In the cross section micrograph of Fig. 2b the QDs always look like truncated pyramids owing to the specific strain-contrast discussed below. In the cross section HREM image of Fig. 3, however, the pure pyramidal shape of the QDs seems to be fully proved. The height of the pyramid is about 6 nm, the sidewalls are close to {110}. Nevertheless, the top of the pyramid is weakly pronounced because of the strong strain-induced contrast around the QD, which makes it difficult to analyse the shape and size of the dot even if HREM imaging is used.

Similar contrast features are shown in the simulated HREM micrographs of Fig. 4 calculated by multi-slice image simulations based on the molecular-static relaxed pyramidal structure as shown in Fig. 1. The pyramids mostly appear to be truncated. Striations of the HREM contrast of all atomic columns around the pyramid occur, with a strong background contrast owing to the high level of strain (misfit $f = 7.5\%$ for lattice parameters of InAs and GaAs) on all the different defoci and foil thicknesses of the simulated micrographs of Fig. 4. For thicknesses of the foil comparable with the base length of the QD pyramids (left and central columns of the images), the different contrasts of the InAs and GaAs columns are visible at certain defoci and solely for the thicker parts of the pyramids, but in accordance with their focus–thickness patterns modified by stress (see, e.g., [19]). Thus, the full pyramid in such an image always looks truncated. The real shape of the pyramidal QD is best pronounced at thickness $t = 22.6$ nm and Scherzer focus ($\Delta = -40$ nm) owing to the difference in the structural factors. Some other defocus/thickness conditions, however, show pyramidal-shaped contrast, too, e.g., $\Delta = -60$ nm, $t = 9$ nm, where InAs is pronounced. The relatively small differences in the structure data of In and Ga, however, cause very weak effects even in the micrographs taken at Scherzer focus which almost vanishes

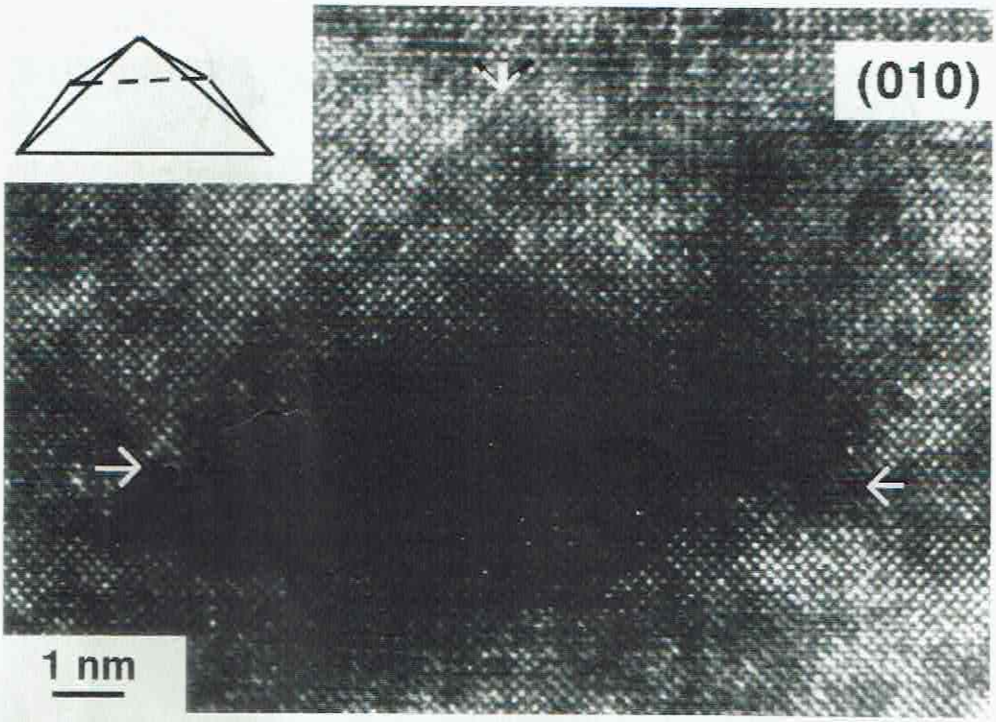

Fig. 3. Experimental 400 kV HREM image of a 3 ML InAs QD (facets of the pyramids marked by arrows, dark strain-induced contrast marking the interface at dot base). Insert: Scheme of the dot geometry (transmitted beam parallel to the side-walls of the QD which are close to {110} planes)

at the other defoci. In thicker regions of the foil (right column in Fig. 4) the image of the QD is mainly influenced by the strain-induced contrast. The strain-induced contrast is obviously independent of the defoci while it is sensitive to the foil thickness (t/ξ_g) measured in units of the extinction distance ξ_g. It has the typical shape of two branches, one above and the other below the pyramid base. The contrast is dark or bright depending on the foil thickness. Using a small aperture in order to select solely the transmitted beam, in Fig. 5 we calculated the symmetrical [001] zone-axis BF images for a set of cross section HREM images according to Fig. 4. Here, the strain-induced contrast is mainly presented. Indeed, the contrast is rather independent of defoci, showing rounded dot images in all micrographs simulated well agreeing with experimental images as, e.g., Fig. 2b.

Randomly substituting Ga for In at a composition ratio of 1 : 1 within the pyramid to create InGaAs QDs causes a reduction of the strain level, however, it does not improve the visualization of the shape of the pyramids because the difference in structure factors and absorption also decreases owing to the relatively low amount of In atoms. Calculations (not shown here) of a focus/thickness series of HREM images of the InGaAs QDs in GaAs were done under conditions similar to those for Fig. 4. In general, they demonstrate that conclusions similar to those above can be drawn.

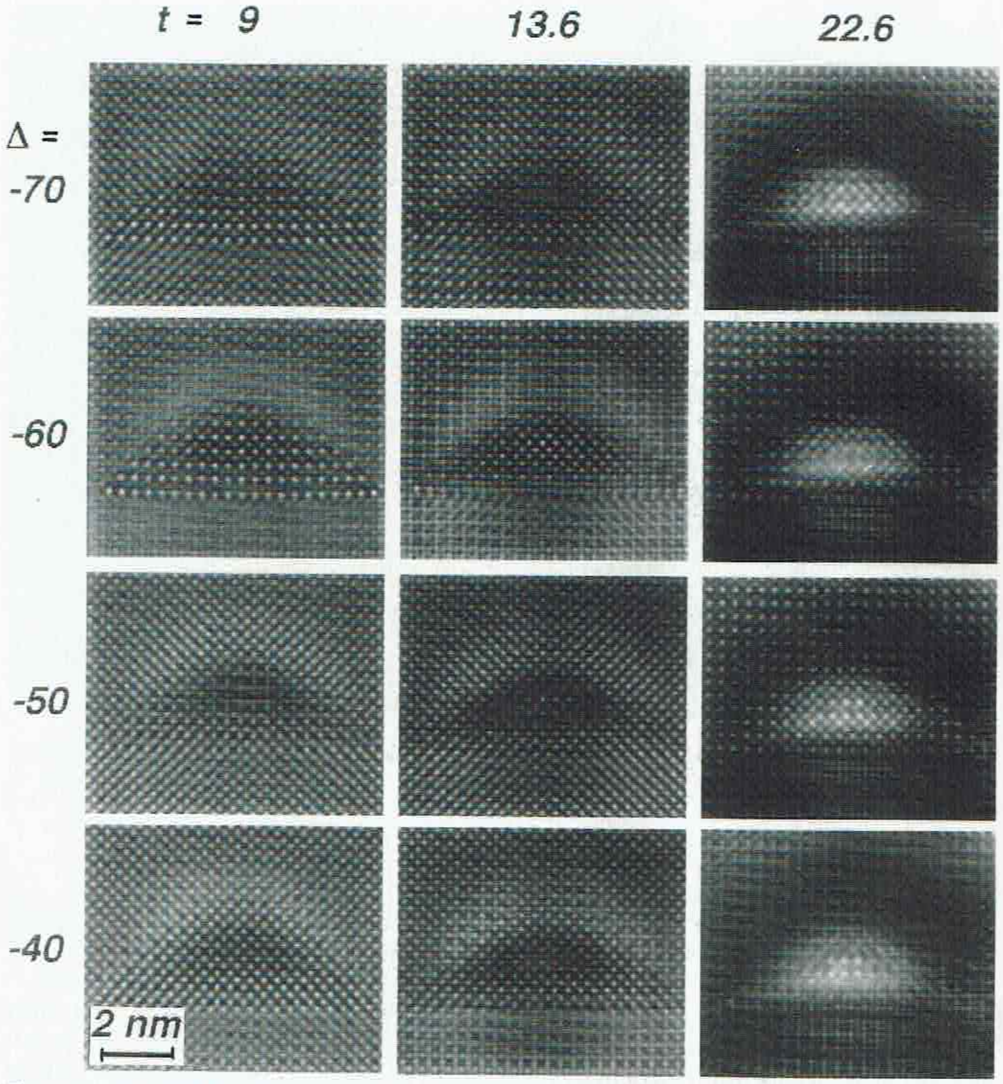

Fig. 4. Series of HREM images of QD in [001] simulated for different defoci (Δ in nm) and foil thickness (t in nm), 400 kV, C_s = 1 mm, α_0 = 1.2 nm^{-1}

4. Conclusions

The visualization of coherently strained nm-scale InAs islands embedded in a GaAs matrix by using conventional TEM is rather complicated owing to the high strain level around the QDs. Being of pyramidal shape, InAs islands always look truncated even in HREM micrographs owing to the low In content on top of the pyramid and the strong strain-induced contrast. The HREM contrast of the dots is shown to be rather sensitive to both foil thickness and defoci, whereas the symmetrical zone-axis BF contrast with lower resolution is approximately independent of usual defocus variations and thus of the strain influence,

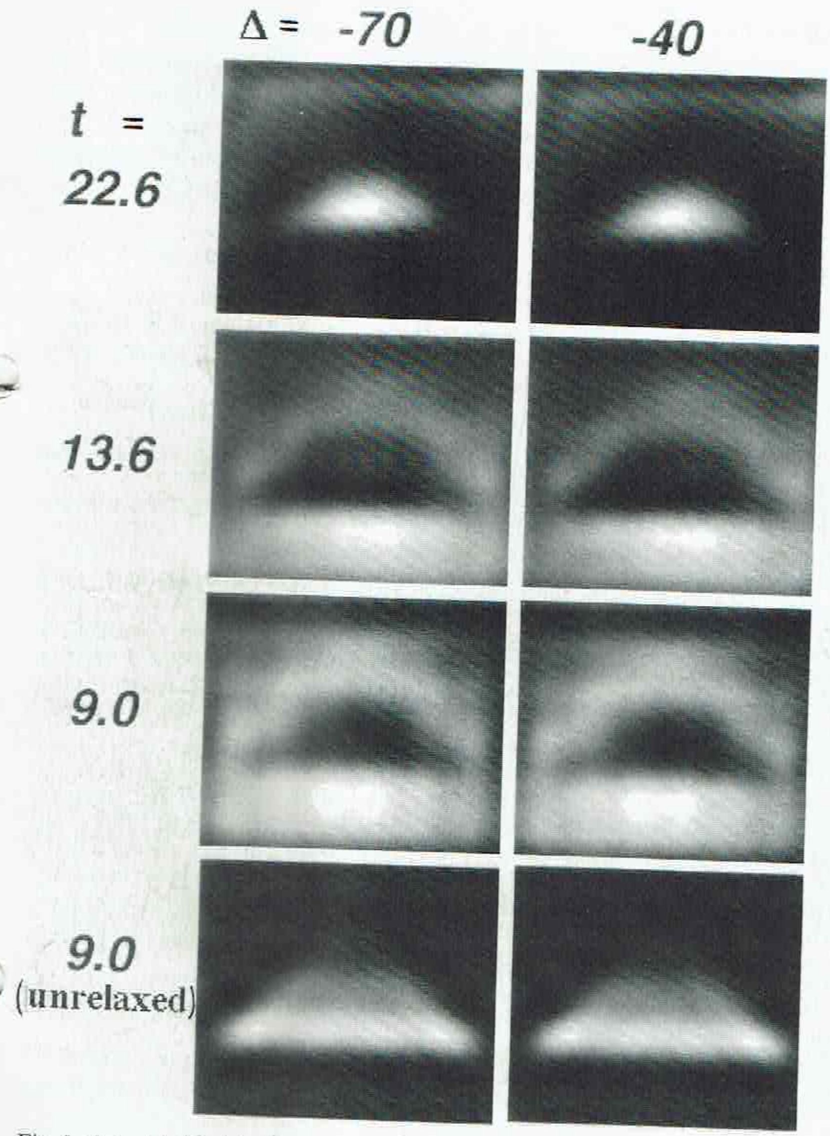

Fig. 5. Calculated bright-field [001] zone axis images of QD for different foil thickness (t in nm) and defoci (Δ in nm) showing almost no influence of focus (cf. −70 nm left column, −40 nm right column) but strong relaxation dependence (cf. calculation with unrelaxed model in lower row)

too. Optimum HREM imaging conditions (particular focus and foil thickness which is about or less than twice the base length of the dot) are found to be most favourable to reveal the shape of such objects owing to the difference in the structure factors between In and Ga atoms. The model has to be refined by using more realistic pair potentials and by including a greater number of atoms as well as of diffusion processes to provide a quantitative comparison with experimental images.

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