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Determination of the structural properties of multiple quantum dot ensembles based on a rigorous X-ray specular and diffuse scattering analysis and comparison with measurements

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Abstract. Multiple and multi-wave diffraction, absorption, and resonances influence significantly X-ray scattering from multiple quantum dots (QDs), and these effects are taken into account by known theories only approximately. The present report dwells on application of a rigorous theoretical analyses of X-ray reflection from In(Ga)As/GaAs QD ensembles to the investigation of their structural properties. The angle dependences of diffuse scattering exhibit very strong peaks near the angles corresponding to specular reflection from the QD facets (the so-called blaze condition for gratings). These peaks exist at nearly the same diffraction angles for QDs with the equal slope angle; their intensities, however, may differ by a few times for vertically correlated and non-correlated QDs.

Introduction

Further progress in investigation of self-assembled quantum dots (QDs) and of other low-dimension structures with typical sizes of the order of 10-nm and having unique optoelectronic properties requires development of methods that would provide more accurate characterization of structural details. Nondestructive X-ray diffraction generally has the potential to measure the chemical composition and strain field distributions. In many cases, however, the complete intensity distribution has to be calculated using a structural model with a large number of free fitting parameters [1], and it falls short from predicting absolute values exactly. For example, the widely used Takagi–Taupin formalism [2], which was developed for the analysis of strained crystals, is not capable of predicting directly the influence of the height h and width L of QDs, as well as their vertical correlation rate. Multiple and multi-wave diffraction, absorption, and resonances influence significantly X-ray scattering from multiple QDs, and these effects are taken into account by known theories only approximately. The present report dwells on application of comprehensive theoretical and experimental analysis of X-ray scattering from QD ensembles to the investigation of growth processes of QDs in In(Ga)As/GaAs systems and of their structural properties.

1. Theory

Rigorous computation of the field scattered by X-rays from rough surfaces is a problem of daunting complexity in the area of electromagnetism and optics even for modern computers because of the small wavelength-to-period λ/d and wavelength-to-height λ/h ratios on the one hand [3] and large number of accountable asperities, especially at grazing incidence, on the other [4]. The rigorous integral equation method employed in the analysis of diffraction grating efficiency [5] is extended here to the case of quasi-periodical and non-periodical structures of any kind [6]. The software developed allows one to operate with exact models (viz based on Maxwell's equations, exact boundary conditions, and radiation conditions) to study X-ray scattering from multilayer structures in real space [7], in particular ensembles of QDs having different shapes, lateral and vertical correlation lengths, surface densities, and fractal dimensions, with the use of a low- or mid-end workstation in a reasonable computation time.

It was shown that in perfect crystalline structures with vertically correlated QD ensembles there is an additional long-range QD ordering in lateral planes which gives rise to corrugation of crystalline planes and results in a quasi-periodical distribution of the elastic deformation and QDs [8]. To rigorously include the quasi-periodicity of QD ensembles, we used the model in which the rough surface is represented by a grating, the large period of which contains a few or a large number of irregularities [4]. Thus, the program deals with a structure which, while being a grating from a mathematical point of view, can model a rough surface provided d becomes large compared with the lateral correlation length (width) L of the asperities. Moreover, if L is about or greater than λ, the number of diffraction orders is large, and the continuous speckle of the rough surface is simulated by the discrete speckle of the grating. Border profiles with QDs have both a periodic and a random component, and random samples have to be averaged to obtain the exact scattered field. The model assumed permits also to use corrugated interfaces having QDs as irregularities.

The time required to calculate one reflectance for all scattering angles for a 20-border QD structure (with the energy balance error of $\sim 1.e-5$) was about 3.5 hours when using a workstation with two Intel(R) Pentium(R) 2 GHz processors, 2 Mbyte cache, 400 MHz bus clock, 2 Gbyte RAM and controlled by OS Windows(R) XP Pro.

2. Experimental

Two samples were grown by molecular beam epitaxy. Both of them consisted of 10 InAs QD sheets (two mono layers of InAs were deposited for each QD layer) separated with 10 nm (sample F667) and 40 nm (sample F668) of GaAs allowing

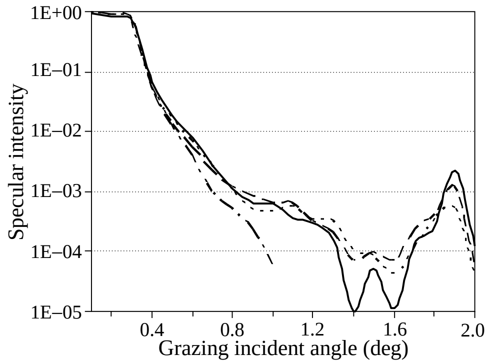

Fig. 1. Absolute specular reflectivity for sample F668 with vertically non-correlated GDs at a 0.154-nm wavelength vs grazing angle of incidence: perfect plane structure model (short-dashed line); Névot–Croce approximation with rms roughness of 1.45 nm for triangles (long-dashed line); rigorous calculation with QD height $h = 5$ nm and width $L = 16.7$ nm (full line); measurement (long-short-dashed line).

to get correlated and non-correlated QD arrays. In the first case (10 nm), the dots are perfectly aligned vertically (which is confirmed by transmission electron microscopy), in the second case they are arranged more or less randomly.

Measurements of both specular and diffuse X-ray intensities in different geometries and spaces were performed on chosen samples using the grazing incidence X-ray diffraction techniques at a wavelength of Cu $K\alpha_1$ ($\lambda = 0.1541$ nm) by the Hotbird diffractometer and position sensitive detector [9]. The beam's divergence is 0.01° and the angular resolution of 0.005° or 0.001° has been chosen for different experiments.

3. Results and discussions

By comparing the theoretical with experimental X-ray reflectances (Fig. 1), the average values of $h = 5.0$ nm and $L = 16.7$ nm determined in the Stranski–Krastanow growth model [10], as well as the superlattice parameters were borne out and refined by the fitting procedure for two different samples. Calculations have revealed a weaker dependence of specular scattered intensities on L and a stronger one on h, a feature that is especially noticeable at high grazing angles. Also, noticeable differences appearing at these angles between both approximate (Névot–Croce model) and rigorous calculation methods, on the one hand, and the theory and experiment, on the other, find adequate explanation.

The angle dependences of diffuse scattering exhibit very strong peaks near the angles θ_{diff} corresponding to specular reflection from the QD facets (Fig. 2), the so-called blaze condition for gratings:

$$2\alpha = \theta_{\text{inc}} - \theta_{\text{diff}} ,$$

where α is the facet angle of QDs ($\tan\alpha = 2h/L$), θ_{inc} is the incident angle and θ_{diff} is the diffraction (scattering) angle corresponding to a strong intensity peak. The good coincidence for the results obtained from the blaze equation with those obtained numerically indicates the validity of computations and possibility to extract QD slope angles from measurements of X-ray diffuse scattering.

These peaks exist at nearly the same diffraction angles for QDs with the equal slope angle; their intensities, however, may differ by a few times for correlated and non-correlated QD structures. This determination of the QD slope angle and vertical correlation rate, as well as of other structural parameters, can be performed with a high accuracy, provided the data sample to be averaged is statistically large enough. So the proposed approach appears promising.

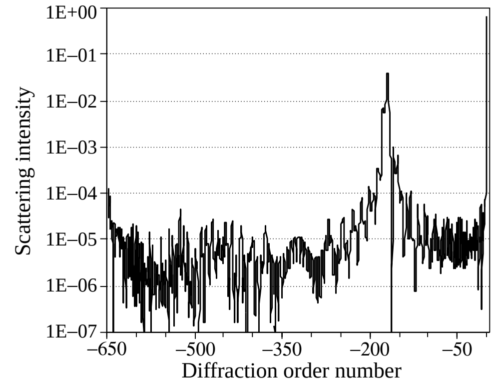

Fig. 2. Absolute specular and non-specular reflectivity calculated for sample F667 with vertically correlated GDs at a 0.154-nm wavelength and a 0.5° grazing incidence vs diffraction angle in the range from −85.7° to +89.7°. The strong peak exists near a scattering angle of 28.5° that corresponds to specular reflection from QD facets.

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