ISSN 1063-7850, Technical Physics Letters, 2010, Vol. 36, No. 2, pp. 108–111. © Pleiades Publishing, Ltd., 2010.
Original Russian Text © L.I. Goray, N.I. Chkhalo, Yu.A. Vainer, 2010, published in Pis'ma v Zhurnal Tekhnicheskoĭ Fiziki, 2010, Vol. 36, No. 3, pp. 31–38.

Detecting Quasi-Periodic {11*n*} (*n* = 7–11) Faces in Samples with Ge/Si Quantum Dots by Grazing X-ray Reflectometry

L. I. Goray*, N. I. Chkhalo, and Yu. A. Vainer
Saint Petersburg Academic University, St. Petersburg, 194021 Russia
Institute for Analytical Instrumentation, Russian Academy of Sciences, St. Petersburg, 190103 Russia
Institute for Physics of Microstructures, Russian Academy of Sciences, Nizhni Novgorod, 603950 Russia
**e-mail: lig@pcgrate.com*

Received May 8, 2009

Abstract—High-resolution grazing X-ray reflectometry is used to obtain experimental and theoretical data on the intensity of specular and diffuse reflection from self-organized structures grown by molecular beam epitaxy with single-layer (non-overgrown) and multilayer (overgrown) Ge/Si quantum dots (QDs). Using the positions of diffuse scattering peaks in the direct space, the slopes of quasi-periodic faces have been determined to within ±0.1° by a method employed previously for the investigation of In(Ga)As/GaAs quantum dots. Finding the quasi-periodic {11*n*} (*n* = 7–11) faces (typical of the growth of ordered QDs) in the samples with disordered Ge/Si quantum dots is evidence of the generality of mechanisms of QD formation in different systems.

DOI: 10.1134/S1063785010020057

Transmission electron microscopy, atomic force microscopy (AFM), and near-field scanning optical microscopy techniques, which are widely used for the characterization of nanodimensional objects, have some disadvantages that hinder detailed investigation of the structure of systems with quantum dots (QDs). An integral nondestructive tool for the investigation of nanodimensional multilayer structures, including those with atomically-rough and/or diffusive interfaces, is offered by X-ray probing. Methods of X-ray diffractometry and reflectometry based on an analysis of the distribution of the intensity of specular and diffuse components of scattering in the direct and reciprocal space are most informative for the investigation of heterostructures with QDs.

Because it is independent of the crystalline structure of samples and the presence of internal stresses, X-ray reflectometry is among the most important methods for investigation of the morphology of boundaries of nanoobjects [1]. Recently, we successfully used [2] high-resolution grazing X-ray reflectometry (HGXR) for the analysis of X-ray scattering from samples with multilayer ensembles of QDs grown by molecular beam epitaxy (MBE) in the In(Ga)As/GaAs system. It was established that the positions of experimentally observed peaks of the intensity of diffuse scattering are determined entirely by the inclination angles α of the pyramidal faces of QDs (blaze diffraction grating condition), in agreement with the previous theoretical prediction [3]. The general approach to determining the α value, which can differ on average from the corresponding crystallographic angle, is based on the numerical solution of the inverse problem of scattering with rigorous boundary and radiation conditions. Solving this task in the range of short radiation wavelengths requires vary large computational resources even for two-dimensional (2D) models [4]. For this reason, the problem of X-ray scattering is solved using various simplifications, including the Born approximation and the distorted-wave Born approximation, which significantly facilitate the solution but not always yield correct results [5]. A comparison to the results of simulations based on the rigorous theory of scattering shows that, using the simple geometric condition $2\alpha = \theta_{inc} - \theta_{diff}$ or $2\alpha = 180° - (\theta_{inc} + \theta_{diff})$, it is possible to determine α from the position of the peak of scattered radiation intensity measured for the incidence angle θ_{inc} and diffraction angle θ_{diff}. The shape of this peak depends on a number of parameters [6]. Thus, the laborious solution of the inverse problem of scattering for determining α is reduced to calculations using an elementary formula. In addition, the known positions and amplitudes of the Bragg peaks can be used to determine the values of the roughness/interdiffusion range and the heights of QDs.

In the present study, we used an approach analogous to that outlined above and characterized for the first time the MBE-grown heterostructures with single-layer (non-overgrown) and multilayer (overgrown) Ge/Si quantum dots. The structures with QDs were grown on the vicinal Si(001) substrate surface by MBE

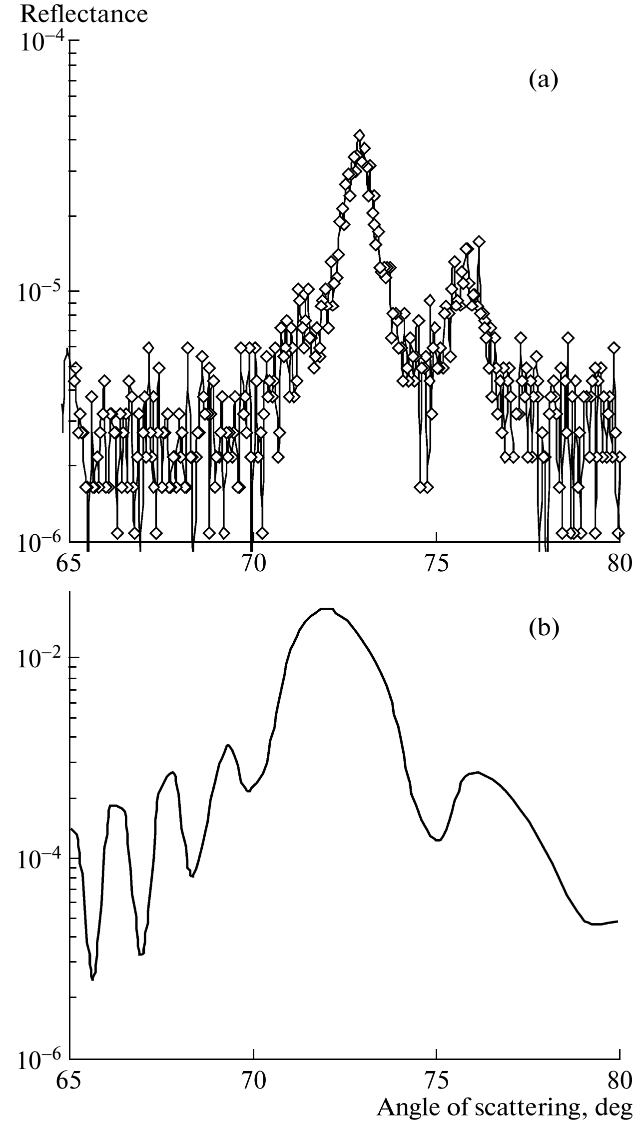

Fig. 1. Curves of diffuse X-ray scattering at a grazing incidence angle of 0.392° for sample 1: (a) measured for the sample misoriented by −5° relative to the [110] or [1−10] direction; (b) numerical calculation.

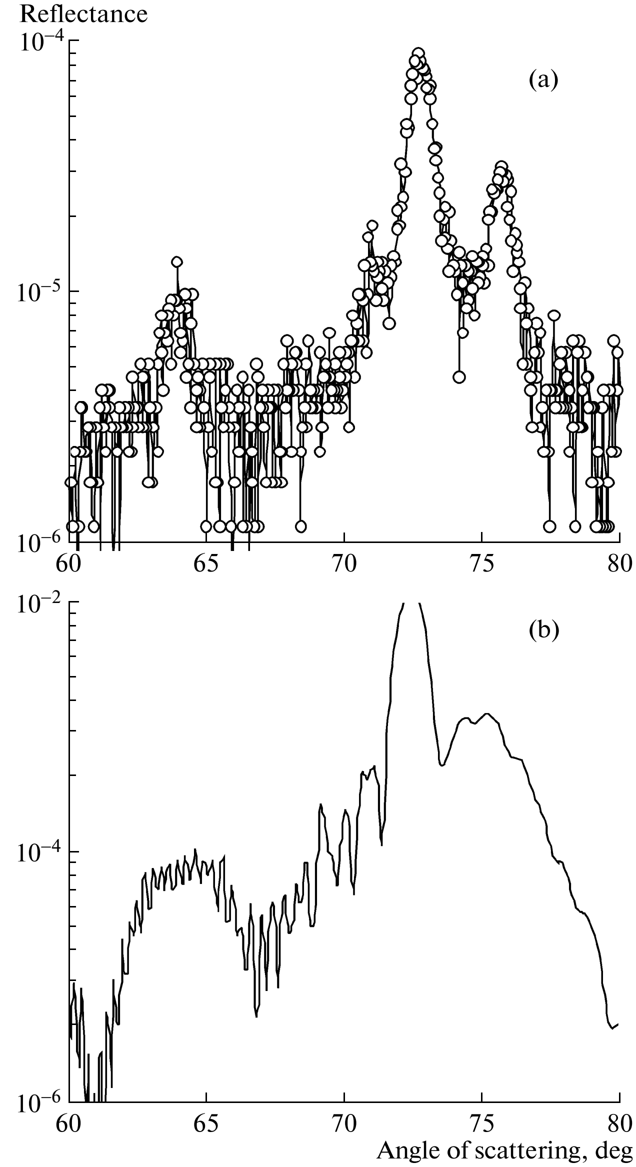

Fig. 2. Curves of diffuse X-ray scattering at a grazing incidence angle of 0.304° for sample 2: (a) measured for the sample misoriented by +5° relative to the [110] or [1−10] direction; (b) numerical calculation.

on a Balzers UMS 500P setup [7]. The single-layer samples 1 and 2 with germanium QDs without cupping layers were obtained be depositing germanium at a temperature of 700°C onto stressed SiGe layers containing 10% and 20% of germanium, respectively, which were predeposited on a 100-nm-thick buffer layer of silicon. The multilayer sample 3, which contained dome-shaped QDs and 20 layers of Ge/Si superlattices with a period of 30 nm, was grown at 650°C on a 50-nm-thick silicon sublayer and had the like cupping layer. Multilayer sample 4, which contained pyramid- and hut-shaped QDs and 20 layers of Ge/Si superlattices with a period of 11.7 nm, was grown at 550°C on a 50-nm-thick silicon sublayer and had the like cupping layer.

The surface morphology of substrates and samples 1−4 was studied by AFM. The AFM data were used to evaluate the dimensions and surface densities of self-organized nanoislands and to determine the roughness of substrates and buffer silicon layers. The AFM measurements in the tapping mode were performed ex situ in air on Solver Pro and Nanoscope III instruments.

The specular and diffuse X-ray scattering was studied by the HGXR method on a Philips Expert Pro diffractometer equipped with a four-crystal Ge monochromator. The rocking curves were recorded in the $\theta/2\theta$ scan mode, while the scattering indicatrices were measured in the θ and 2θ modes. All measurements were performed using Cu$K_{\alpha 1}$ radiation with a wavelength of $\lambda = 0.154$ nm. The detector was a gas-discharge counter possessing an ultimately low intrinsic noise (on the order of 0.1 quantum/s) with a controlled entrance slit [2]. For a detector to sample distance of 320 mm, an intense scattering signal was obtained at the crystal-monochromator slit width of 100 μm and heights within 1–5 mm. The angle scan step was chosen within 0.001°–0.005°, depending on the desired resolution at a beam angular divergence of 0.003°. The detector slit width was varied within 0.1–3 mm.

Calculations within the framework of a rigorous electromagnetic theory were performed using a modified boundary-integral-equation method (MIM) [8, 9], which proved to be rather accurate and quite rapidly converging for large ratios of the characteristic period D and the QD height h to X-ray wavelength λ ($\sim 10^3$ and $\sim 10^2$, respectively) [3, 5]. The relative error (estimated from the energy balance) amounted to $\sim 10^{-6}$ for 400–1600 collocation points at each boundary of the model structure. Data were processed by a workstation with two QuadCore Intel Xeon 5355/2.66 GHz processors, 1333 MHz front side bus, and 16 Gbyte RAM. For this system operating under 64-bit Windows Vista Ultimate with eightfold-parallel path, calculation of the curve of scattering intensity for a single statistical set of parameters required about 2 min.

The face inclination angles of islands (pits) were determined with high precision (±0.1°) from measured positions of the peaks of diffuse scattering in the direct space, mostly for the predominant quasi-periodicity directions [110] and [1–10]. Figures 1 and 2 show the typical experimental and theoretical curves of the diffuse scattering intensity, which were obtained by the HGXR method for samples 1 and 2, respectively. Both positions and shapes of the main peaks in these curves, which correspond to the reflection from {11n} (n = 7–11) faces, well coincide for the two samples. The comparison of magnitudes requires reducing the 3D problem to a 2D variant [6].

The observed reflections from {11n} (n = 7–11) faces are most likely indicative of the presence of pits with the shape of inverted pyramids. The formation of these faces is also possible in dome-shaped QDs [10, 11]. Reflections from the {119} faces (crystallographic angle, 8.9°) were reliably observed in both samples with single-layer QDs and in multilayer structures with cupping layers (containing QDs of pyramid and hut shapes or those of the dome shape). As the grazing angle was increased, the peaks shifted toward greater angles of scattering, in agreement with the theory.

The presence of pits near the base of some QDs was also confirmed by AFM data, but determining their morphology requires additional investigations. The observation of pits with quasi-periodic {11n} (n = 7–11) faces (typical of the growth of ordered QDs) in the samples with disordered Ge/Si quantum dots is evidence for the generality of mechanisms of the QD formation in different systems.

We have detected and studied a small (within a few degrees) deviation from the [110] or [1–10] direction for the mean normals to the reflecting faces. This deviation significantly modifies the scattering peak intensity. Experimental investigation of the faces of pits and QDs by the HGXR method showed that (i) the long-range order in the distribution of self-organized QDs in various structures grown on vicinal Si(001) substrates is absent in the [110] and [1–10] directions and (ii) the corresponding faces of QDs may exhibit no well-pronounced planar character [1]. Using the obtained HGXR and AFM data, we refined the MIM-based model so as to make allowance for the generation of plausible boundary profiles with random roughness.

Thus, the traditional HGXR method of determining the parameters of layers and boundary roughness has been expanded to determining the geometry of islands, pits, and QDs grown in various structures grown by epitaxial techniques. In order to obtain high-intensity diffuse X-ray reflection from faces of the Ge/Se quantum dots in the selected directions, including [100] and [010], the QDs should be grown on periodic masks. In order to eliminate the formation of lens-shaped QDs, it is necessary to deposit cupping layers at reduced temperatures.

Acknowledgments. The authors are grateful to A.V. Novikov for kindly providing the samples and fruitful discussions.

This study was supported in part by the Russian Foundation for Basic Research, project no. 06-02-17331.

REFERENCES

1. J. Stangl, V. Holy, and G. Bauer, Rev. Mod. Phys. **76**, 725 (2004).
2. L. I. Goray, N. I. Chkhalo, and G. E. Cirlin, Zh. Tekh. Fiz. **79** (4), 117 (2005) [Tech. Phys. **54**, 561 (2009)].
3. L. I. Goray, G. E. Cirlin, E. Alvers, Yu. B. Samsonenko, A. A. Tonkih, N. K. Polyakov, and V. A. Egorov, *Proceedings of the 15th Intern. Symp. "Nanostructure: Physics and Technology"* (Novosibirsk, 2007), pp. 118–119.
4. H. Gross and A. Rathsfeld, Waves Random Complex Media **18**, 129 (2008).
5. L. I. Goray, Proc. SPIE **7390**, 739 00V (2009).
6. L. I. Goray, Proc. SPIE **6617**, 661 719 (2007).

7. Z. F. Vostokov, Yu. N. Drozdov, D. N. Lobanov, A. V. Novikov, M. V. Shaleev, A. N. Yablonskii, Z. F. Krasilnik, A. N. Ankudinov, M. S. Dunaevskii, A. N. Titkov, P. Lytvyn, V. U. Yukhymchuk, and M. Ya. Valakh, in *Quantum Dots: Fundamentals, Applications, and Frontiers*, Ed. by B. A. Joyce et al. (Springer, 2005), pp. 333–351.

8. L. I. Goray, J. F. Seely, and S. Yu. Sadov, J. Appl. Phys. **100**, 094 901 (2006).

9. L. I. Goray, Nucl. Instr. Meth. Phys. Res. A **536**, 211 (2005).

10. O. P. Pchelyakov, Yu. B. Bolkhovityanov, A. V. Dvurechenskii, L. V. Sokolov, A. I. Nikiforov, A. I. Yakimov, and B. Voigtländer, Fiz. Tekh. Poluprovodn. (St. Petersburg) **34**, 1281 (2000) [Semiconductors **34**, 1229 (2000)].

11. G. Bauer and F. Schaffler, Phys. Status Solidi A **203**, 3496 (2006).

Translated by P. Pozdeev