Genetic algorithm application for solving inverse problems of X-ray diffraction on systems with quantum dots

D V Sivkov\textsuperscript{1,2}, V I Punegov\textsuperscript{1}
\textsuperscript{1}IPM of Komi SC UrB RAS, Syktyvkar, 167982, Russia
\textsuperscript{2}Immanuel Kant Baltic Federal University, Kaliningrad, 236041, Russia

Abstract. For solving inverse problem of X-ray diffraction on GaAs-AlAs superlattices with InAs quantum dots within the statistical theory of X-ray diffraction the Genetic Algorithm in the form of Differential Evolution method was used. The calculation results were compared with experimental data.

1. Introduction
Modern nanotechnologies allow to create artificial materials with new properties for different applications in physics, chemistry, biology, and medicine. Creating nanostructures using templates is recognized as a very promising way to achieve accurate positioning of nano-objects, in particular, arrays of quantum dots. Epitaxial self-organized quantum dots (QDs) are unique objects that are widely used in the electronic, optical, and structural properties of semiconductor systems [1]. High-resolution x-ray diffraction methods have been successfully used to characterize different types of epitaxial structures, including multilayer structures with QDs [2].

In addition to developing a theory describing the interaction of X-ray with matter, an effective algorithm for minimizing the residual functional is needed. The feature of this problem is in the large number of local minima in the parameter space and large number of parameters. Therefore, traditional iterative optimization methods are not well suited due to the considerable time required to find a solution. Evolutionary algorithms, including genetic ones [3], showed their high efficiency for searching solutions in the space of a large number of parameters in a wide range, particularly in problems of x-rays scattering [4].

Within the work to analyze the experimental diffraction data, both coherent and diffuse x-ray scattering components bounded together by a static Debye-Waller factor was being taking into account.

2. Experiment
The studied structures were grown on the GaAs (001) substrate and GaAs buffer layer with a thick (~2 $\mu$m) bottom Al\textsubscript{0.3}Ga\textsubscript{0.7}As barrier layer, a GaAs waveguide with the thickness of 240 nm and multilayers of InGaAs layers with InAs quantum dots 5 nm thickness separated by GaAs layers 15 nm thickness, repeated 20 times, finally capped with the same GaAs waveguide and a 450 nm thick Al\textsubscript{0.3}Ga\textsubscript{0.7}As barrier layer. The main structures were grown on a 300 nm thick GaAs buffer layer.

The x-ray measurements were performed on a high-resolution x-ray diffractometer X'Pert MRD (PANalytical) with multilayer focusing mirror, Ge (011) Bartels monochromator and a three-bounce Ge (011) analyzer crystal. Double- and triple- crystal $\omega-2\theta$ rocking curves (RC) and triple-crystal $\omega$ RCs at the maximum of the main diffraction peaks of the GaAs substrate and AlGaAs layer, and the superlattice (SL) peaks of the periodic QDs multilayer structure (“0SL”, “± 1SL ”and others) were...
measured. Reciprocal space maps (RSM) of the scattered radiation were recorded around symmetric (004) scattering spot.

3. Diffraction theory

In the X-ray scattering experiment, the total intensity obtained at the output of the detector is written as

\[ I_{\text{sim}}(q_x, q_z) = K \cdot \overline{I}_h(q_x, q_z) + I_{bg}, \]

where \( I_{bg} \) – background intensity, \( K \) – intensity scaling parameter. The expression for the scattering intensity at the input of the detector, taking into account the influence of the instrumental function of the monochromator and the analyzer, has the form [1]

\[ \overline{I}_h(q_x, q_z) = \frac{\int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} dq_x dq_z R^M(\mathbf{q}_x \cdot \mathbf{q}_z) R^A(\mathbf{q}_x \cdot \mathbf{q}_z) I_h(q_x, q_z) I_h(\mathbf{q}_x \cdot \mathbf{q}_z)}{\int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} dq_x dq_z R^M(\mathbf{q}_x \cdot \mathbf{q}_z) R^A(\mathbf{q}_x \cdot \mathbf{q}_z)}, \]

where \( R^M, R^A \) – monochromator and analyzer reflection coefficients, \( I_h \) – beam scattering intensity. For multilayer heterostructure with buried QDs the intensity can be represented in two terms, reflecting the nature of the scattering, connected through the Debye-Waller factor \( f = f_{QD}f_{s} \), where \( f_{QD} \) - a static factor, caused by the presence of QDs without accounting for the elastic deformation near them, and \( f_{s} \) is static factor caused by elastic strain in the matrix around the QDs.

\[ I_h(q_x, q_z) = (1 - f^2)I_h^c(q_x, q_z) + f^2I_h^l(q_x, q_y)F_L(q_x, q_y)F_V(q_z), \]

\( I_h^c \) – the coherent part, describing the ideal crystal scattering. It contains information on the thicknesses and periods of the layers in the superlattice structure; \( I_h^l \) – the diffuse part, containing information on the parameters of structural defects, such as quantum dots, pores, dislocations of different types (their size, shape, orientation, distribution in the sample, etc.). The diffuse scattering intensity was calculated using the ellipsoidal quantum dots model [5]. \( F_L(q_x, q_y) \) – lateral structural factor, depending on the average QDs spatial distribution in superlattice period; \( F_V(q_z) \) – vertical interference structural factor, depending on the superlattis period.

The scattering intensity calculations were performed within the dynamical theory of X-ray diffraction expanded to the special case of transversely restricted wavefronts of the incident and reflected waves taking into account the effect of the diffractometer’s instrumental function [4].

4. Residual functional

For the minimization procedure in the paper we considered the residual functional in the form

\[ \rho_{sq}(q_x^i, q_z^i) = \frac{1}{N_q} \sum_{i=1}^{N_q} \left[ \frac{I_{\text{sim}}(q_x^i, q_z^i; x) - I_{\exp}(q_x^i, q_z^i)}{I_{\text{sim}}(q_x^i, q_z^i; x)} \right]^2, \]

where \( x \) – vector of parameters in the parameter space, \( I_{\text{sim}} \) – diffracted intensity theoretical data, \( I_{\exp} \) – experimental data. For our calculations the number of angle points at which scattered radiation was collected was 50 for \( \omega \) scans and 460 for \( \omega-2\theta \) scans. Minimization of the residual function was carried out for \( \omega \) scans of all superlattice satellites. Total number of points for search was 450.

The root-mean-square deviation between theoretical and experimental data (4) is effective for analyzing highly noisy signals because it is slightly affected by the statistical noise, and represents the best compromise between speed and accuracy compared to comparison in absolute value, logarithms and the root-mean-square deviation of the logarithms comparisons [6].

5. Differential Evolution algorithm

Differential Evolution (DE) algorithm is described in detail in [7]. For the X-ray scattering intensity calculation DE-strategy in the form “rand/1/exp” was used. That means that population vector to be
mutated was chosen randomly, one difference vector was used and trial vector was generated using exponential crossover.

For our task in obtaining mutant vectors, the absolute values of the corresponding expression were taken in order to exclude the search for a solution among the parameter vectors with negative parameters. DE algorithm parameters were chosen as the best for searching their values $F = 0.1$ and $C_r = 0.4$ were chosen according to the lowest goodness of fit (residual functional) $P_{sqr}$ [3]. The target vector was chosen randomly from the population. Number of cycles was restricted by the value of 1000.

6. Results

In the work, the calculation of X-ray scattering on an 20-period GaAs(001)-AlGaAs-{InAs QDs-GaAs}SL superlattice was performed for (004) GaAs reflection $\sigma$-polarized Cu $K_{\alpha1}$ radiation. Searching parameters were: layers thicknesses $l_{\text{InGaAs}}, l_{\text{GaAs}}$ (characterize $I_{\text{K}}$); lattice mismatch $\Delta\varepsilon$, the average height and radius of the QDs $H_{\text{QD}}, R_{\text{QD}}$ (characterize $I_{\text{K}}$); the average distance between the centers of the QDs in lateral plane $d_{xy}$, variances distance in lateral plane and vertical direction $\sigma_{xy}, \sigma_z$ and the average number of QDs in stack $n$, determine vertical correlation length $l_{V} = n(l_{\text{InGaAs}} + l_{\text{GaAs}})$ – the distance between stacked QDs (characterize $F_L(q_x, q_y)$ and $F_V(q_z)$); static Debye-Waller factors of layers in periodic structure, substrate and barrier layers $f_{\text{QD}}, f_{s}, f_{\text{sub}}, f_{\text{AlGaAs}}$ (characterize total intensity).

The statistical noise was rated and calculated according to the [3]. Calculations were performed using C++. Deviation of the calculated results from the experimental data was about 5%. Structure parameters using for modeling of the scattering experiment, their initial values search ranges, and obtained solutions are shown in the table 1.

**Table 1. Structure parameters, their expected values, search ranges and obtained solutions.**

| Structure parameters | Expected values | Search ranges | Solutions |
|----------------------|----------------|---------------|-----------|
| $l_{\text{InGaAs}}$ | 5 nm           | (4-6) nm      | 5.2 nm    |
| $l_{\text{GaAs}}$   | 15 nm          | (14-16) $\mu$m | 14.8 nm   |
| $\Delta\varepsilon$ | -              | (1-3)$\times 10^3$ | $2.2\times 10^3$ |
| $H_{\text{QD}}$     | 5 nm           | (3-6) nm      | 5 nm      |
| $R_{\text{QD}}$     | -              | (5-30) nm     | 12.5 nm   |
| $d_{xy}$            | -              | (15-100) nm   | 65 nm     |
| $\sigma_{xy}$       | -              | -             | 29 nm     |
| $\sigma_z$          | -              | -             | 47 nm     |
| $n$                 | -              | 1-20          | 7 nm      |
| $f_{\text{QD}}$     | -              | 0.7-1         | 0.927     |
| $f_{s}$             | -              | 0.7-1         | 0.917     |
| $f_{\text{sub}}$    | -              | 0.7-1         | 0.96      |
| $f_{\text{AlGaAs}}$ | -              | 0.7-1         | 0.95      |

The results of solving the inverse problem of X-ray diffraction within the framework of the proposed approach are presented in figure 1.
Figure 1. $\omega$-2$\theta$ rocking curves. Experimental results (solid dots on the red line) shifted by an order in the intensity scale and residual functional minimization (solid blue line) $\omega$-2$\theta$ rocking curves near the (004) GaAs reflection for [GaAs(001)-AlGaAs-[InAs QDs-GaAs]SL]x20 structure.

7. Conclusion
Periodic structures with quantum dots were quantitatively analyzed by simultaneous simulation of the experimentally observed coherent and diffuse scattered x-ray radiation. A priori technological information about the investigated structures, the parameters of the epitaxial layers and the QDs has been used as initial approximations for numerical simulations. The developed method may be successfully used for x-ray diffraction investigation of different nano-structured objects.

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