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DYNAMICAL X-RAY DIFFRACTION IN MULTILAYERED STRUCTURES

01.04.07 - solid state physics

ABSTRACT

of Ph.D. thesis on physico-mathematical sciences

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Key organization:	G.V. Kurdumov Institute of Metelophysics

The defence of the thesis will be on 15.12.2006 at 14^{15} in the V.E.Lashkarev Institute of Semiconductor Physics: Ukraine, Kiev, 03028, pr. Nauki 41.
The full text of the thesis could be found on `http://www.x-ray.net.ua`.

Kiev-2006

THE MAIN CONTENTS OF THE WORK

In the **introduction** the information on thesis theme actuality and its relation with scientific programs is described; the main objectives of the work are formulated; scientific novelty and practical value of the thesis are underlined; the information on personal contribution of the author to the work and on publications is presented.

The review of the dynamical X-ray diffraction in multilayers simulation methods is given in **chapter 1**. Algorithms like: Abele's matrixes, 1D Takagi equation [1], TMA [2], Bartels' recursion formula [3], RMA [4], multibeam TMA [5], 3D multibeam Stetsko's algorithm for uniform parallel plate [6] and Souvorov's algorithm for multilayered structures [7] are considered. The main characteristics, merits and demerits of these methods are described. All these algorithms were software-programmed and tested. One section is devoted to generally applicable methods of wave vectors direction calculation. Besides in chapter 1 short historical information on X-ray dynamical theory is given.

In the **chapter 2** the main equations of X-ray dynamical theory for plane monochromatic waves in crystalline structures are derived from Maxwell's equations without simplifications (the only two are: $\mu = 1$ and absence of charges, but it is legitimate for X-rays). For any Bloh wave the propagation equation takes the form:

$$\frac{(k_h^2 - K^2)}{K^2}\mathbf{E}_h = \frac{(\mathbf{k}_h \cdot \mathbf{E}_h)\mathbf{k}_h}{K^2} + \sum_{g \neq h}^{N} \chi_{h-g}\mathbf{E}_g \qquad (1)$$

where $\mathbf{E}$ is electric field, $\mathbf{k_h}$ and $\mathbf{K}$ are wave vectors in crystal and air ($K = 1/\lambda$), χ_{h-g} are Fourier components of the crystal polarizability and N is the quantity of reciprocal space points take part in the diffraction.

In this chapter the dispersion equation for N-beam case is introduced and general form of boundary conditions between layers and at the structure surfaces are stated:

$$\mathbf{E}_t = const \qquad \mathbf{D}_n = const \qquad \mathbf{H} = const \qquad (2)$$

Besides exact form of main equations commonly applicable approximate equations are written. Simplifying assumptions are as follows: small polarizability, the factor for Pi-polarization is $C = \cos(2\theta_0)$, vectors $\mathbf{E}$ and $\mathbf{D}$ are collinear, etc.

The **chapter 3** is devoted to dispersion equation solving in two-, three-, four- and N-beam cases. The algorithm of numerical calculation of dispersion surface by Laguerre's method for these cases is described. For two-beam case an approximate (for quadratic equation) and exact (for quartic equation) analytical solutions are given.

Dispersion equation could be obtained from the condition for the determinant of the system of N equations (1) to be equal to zero: $\det \Delta_E = 0$. For two-beam case ($N = 2$) in simplified form it could be written as:

$$(k_0^2 - (1 + \chi_0)K^2)(k_h^2 - (1 + \chi_0)K^2) = C^2K^4\chi_{barh}\chi_h \qquad (3)$$

where $\mathbf{k_h} = \mathbf{k_0} + \mathbf{h}$, and $\mathbf{h}$ is a diffraction vector.

The solution of this equation are two spheres with origins in points O and H (the beginning and the end of vector $\mathbf{h}$) and with radii $k = nK$, where n is the index of refraction (fig. 1).

The most interesting effects (dynamical) are observed near the point of cross-section of these Ewald spheres (Lorentz point). But this region in Bragg case in most publications is ploted in simplified form (fig. 2 a), but for the absorbing crystal it should look like shown on fig. 2 b).

In the table 1 different limit cases of dispersion curve for Bragg case are shown.

To solve the dispersion equation (3) some kind of variable changing must be done because it should have only one unknown. As far as $\mathbf{k_h} = \mathbf{k_0} + \mathbf{h}$ and longitudial component of wave vectors on any boundary should stay constant, it could be done. The first method is to express all wave vectors in Cartesian coordinates x and y, where x is parallel to the surface and is constant. The second method is to introduce a new variable ϵ: $\mathbf{k}_h = \mathbf{K}_0 + \mathbf{h} + K\epsilon\mathbf{n}$, where $\mathbf{n}$ is outer normal to the surface. For two-beam case both methods give the same results, but for three or more-beam cases the first method can give wrong results (fig. 3), while the second method have no such limitations (fig. 2 b). This is due to the fact that computer precision is limitd and even for three-beam case and real numbers of *long double* type problems could arise.

For bulk materials approximate methods of diffracted wave vectors finding could be applied only near exact Bragg position (about one degree) and far from grasing geometry. But for multilayers consisting of two materials with very different Bragg angle problems with approximate formula could be observed even near exact Bragg positions for these materials. For example for GaAs substrate and fully strained InAs layer the difference between Bragg angles is about 5 degrees and this situation calculated by exact and approximate formulas is shown on the fig. 4. So for accurate structure analysis precise calculation of wave vectors is needed.

In the **chapter 4** two methods of dynamical N-beam diffraction calculation are described. The first method is capable to calculate the most common case of coplanar many-beam diffraction in samples with any layer thicknesses. But it is derived from commonly used approximate propagation equation with approximate boundary conditions.

The second method is much more interesting and original. It is based on developed in [6] the best for today method of non-coplanar N-beam dynamical diffraction in parallel plate, but could be applied to multilayered structures with arbitrary layers thicknesses. It should be pointed out that in the article [7] rather similar algorithm is proposed, but described in the thesis method was developed independently and could be used for layers of any thickness, because it has no mistakes in phase factors.

The main features of proposed model are:

plane waves are considered; propagation equation and boundary conditions are solved without simplifications; suitable for any geometry (Bragg, Laue, Brag-Laue); 3D modeling in reciprocal and 1D in real space; covers the whole angular range; precise calculation of any polarization with information on diffracted and reflected beams polarization; no principal limitation on quantity of reciprocal lattice points taken part in diffraction; no limitations on layers thickness; all calculation are carried out for x,y,z projections of wave vectors, diffraction vectors and fields.

Some examples of using of the developed algorithm are shown on fig. 5 and fig. 6.

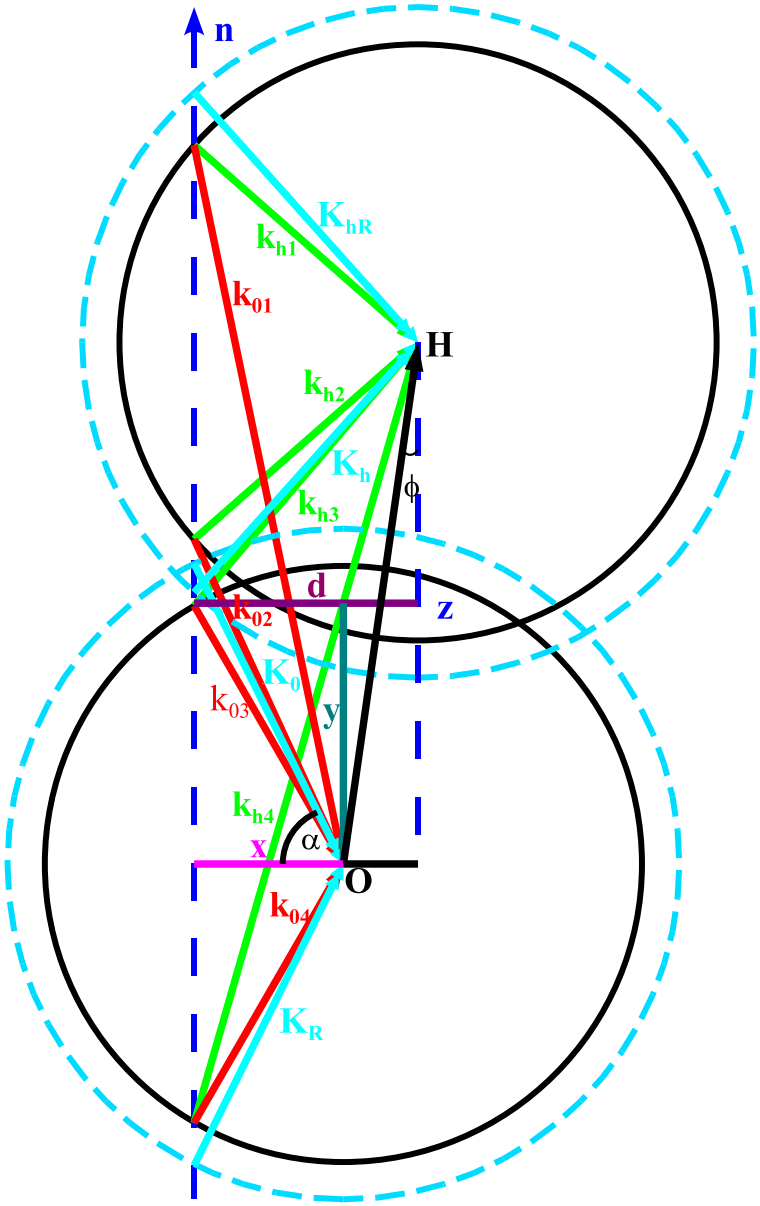


Figure 1. Dispersion curve (2 waves).

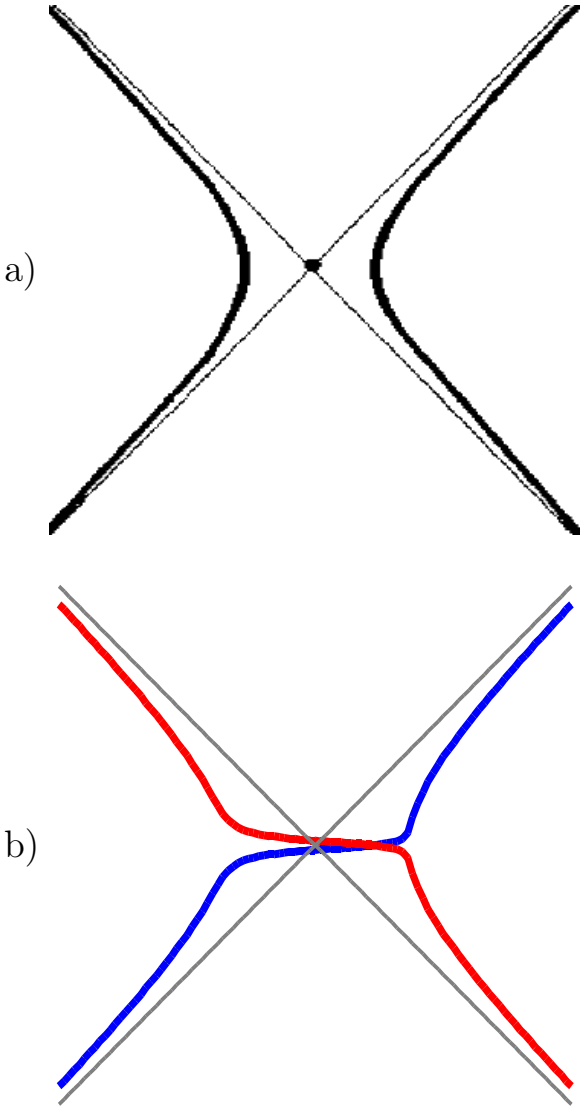


Figure 2. Dispersion curve in symmetric Bragg geometry: a) typical curve presented in most books (this figure is from [8]); b) precisely calculated.

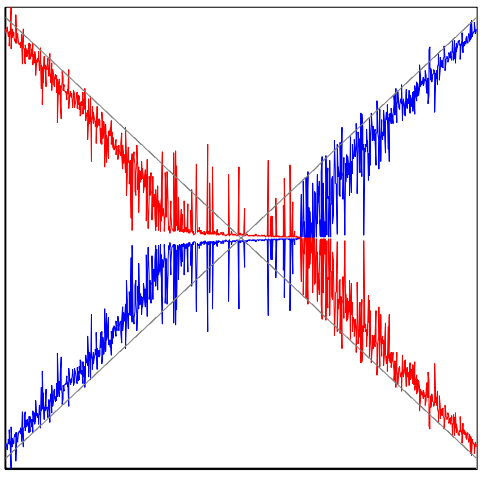


Figure 3. The solution for 4 waves in Cartesian coordinates.

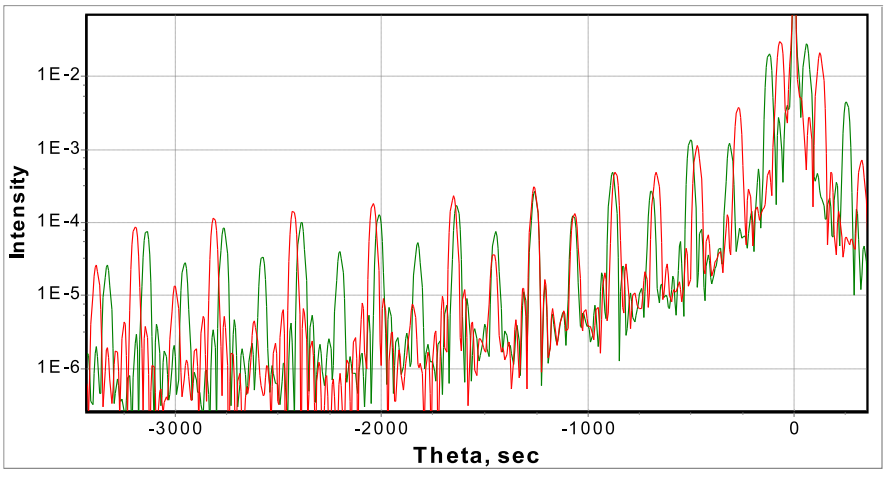
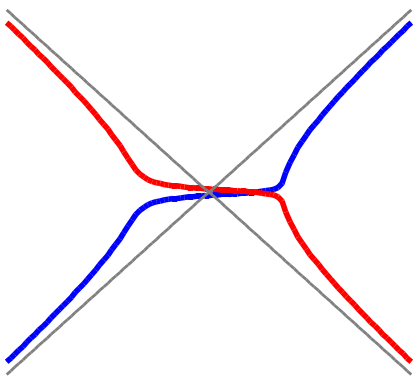
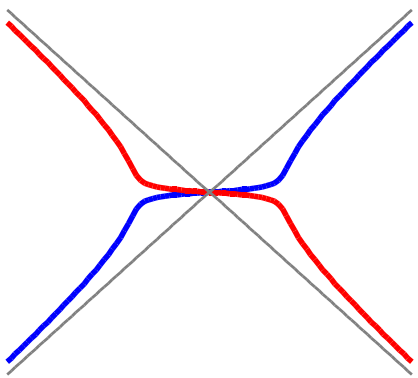
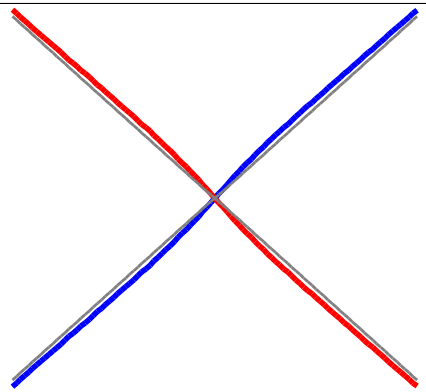
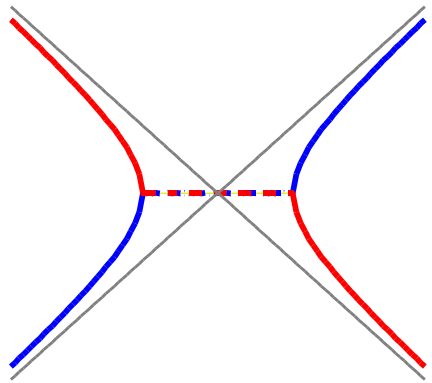
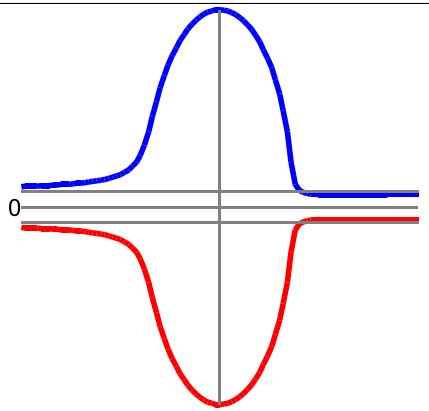
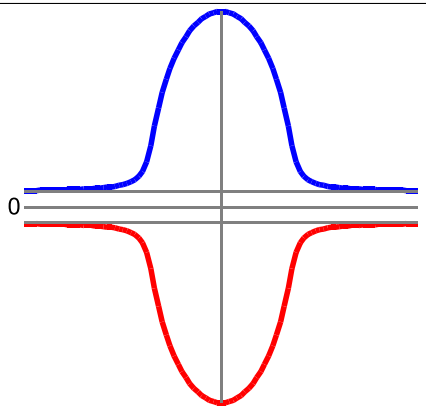
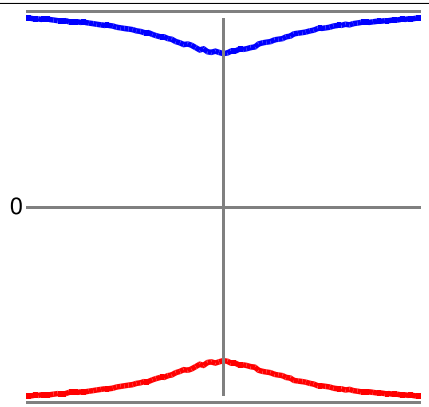
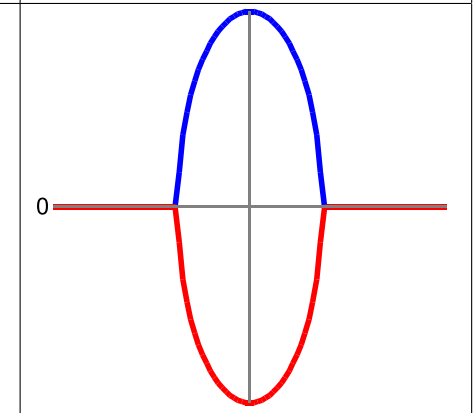
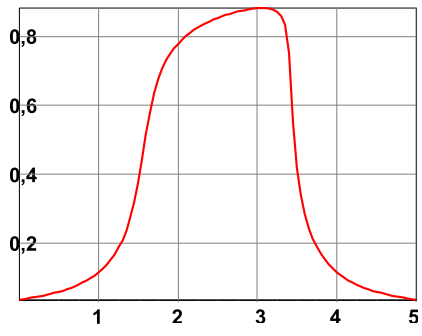
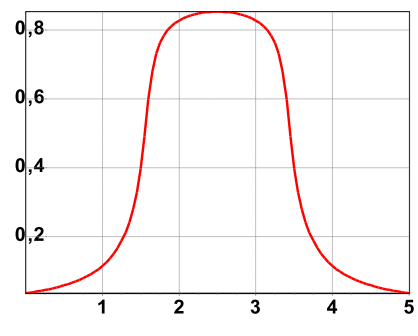
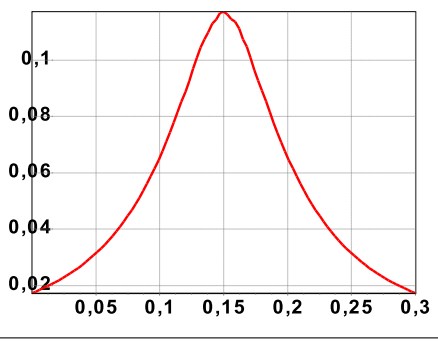
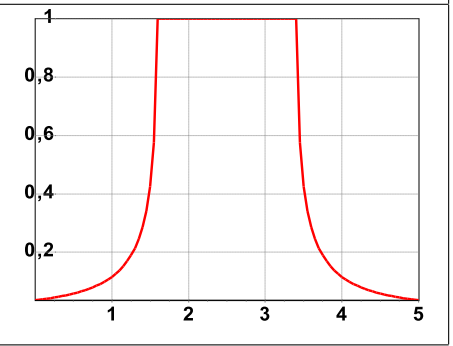


Figure 4. DC from supperlattice InAs/GaAs (5 pair of layers 50 nm each) on GaAs substrate.

Table 1. Dispersion curve in Bragg geometry.

	Normal conditions	$\Im(\chi_h) = 0$	$\Re(\chi_h) = 0 \quad (\Delta\alpha = 0{,}3$ sec)	$\Im(\chi_0) = 0, \quad \Im(\chi_h) = 0$
$\Re$				
$\Im$				
R				

Chapter 5 is devoted to applications of X-ray dynamical diffraction in multilayers simulation algorithms. Examples of using developed algorithms for simulation of specular reflection; for exact polarization calculation of incident, diffracted and reflected beams (in all equations Cartesian coordinates of all electric fields are specified); for Renninger scan simulation. In this chapter open-source program, developed as a part of the thesis, for calculation of polarizability of complex structures is described (more information and the program itself could be find on the site `http://www.x-ray.net.ua`).

One more application of developed simulation algorithms is the analysis of composition gradient on the boundary between two layers in superlattice (SL). A technique for different border zones modeling is described. Sharp, linear, quadratic and quadratic-hyperbolic composition gradient functions are considered (insertion in fig. 7). The influence of such gradients on diffraction curves is shown (fig. 7). Smooth boundaries doesn't influence on structure period determination, but it does influence on thicknesses ratio finding in analyzed $In_{0.2}Ga_{0.8}As/GaAs$ superlattices. With composition gradient introduction good agreement between experimental and simulated diffraction curves intensities could be achieved, but for unequivocal determination of sample structure different reflections should be analyzed simultaneously. For better experimental and simulated intensities coincidence different gradients on different boundaries should be considered [11]. For this reason in thesis some sort of curve autofitting was applied and described.

Next application of developed algorithm is the analysis of anisotropic deformation in multilayered structures. For such investigations diffraction curves for different azimuthal positions are measured and then by comparing experimental and simulated DC, the information on anisotropic deformations could be obtained (fig. 9). Such structure deformations are because of the presence of missorientation angle between surface and atomic layers.

One more application of developed 3D algorithm is azimutally dependent measuring. The example of such simulated and experimentally measured high-resolution reciprocal space maps is shown on fig. 10.

The method developed in the thesis gives full information on diffracted waves (amplitudes, polarizations and directions), so it is possible to use it for reciprocal space maps simulation. On the fig. 11 an example of simulated and measured maps is shown.

In the appendix some information on developed software for X-ray diffraction simulation is presented.

MAIN RESULTS AND CONCLUSIONS

1. Precise theoretical algorithm for dynamical N-beam diffraction in multilayered structures with layers of arbitrary thickness is developed. The main advantages of proposed model are: it could be used far from exact Bragg angle, i.e. for structures consisting from layers with very different composition or for very thin layers. It also could be used for any geometry and for any angles of incidence.

2. Solutions of dispersion equation are analyzed (its real and imaginary parts) and dispersion surface is built for Bragg and Laue cases for 2-beam case and for 3, 4 and N-beam cases. The fact that dispersion surface for Bragg case has principal differences

from Laue case is underlined. The difference between precisely calculated dispersion curve and printed in most publications is shown

3. A technique of layers' structure analyzing is proposed. It is based on azimuthally dependent measuring of diffraction curves. It allows to determine parameters of lattice anisotropic deformations.

4. The influence of composition gradient on layers boundaries on diffraction curve

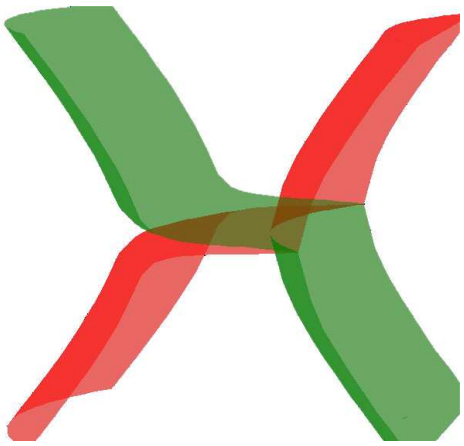


Figure 5. A part of dispersion surface for GaAs 004.

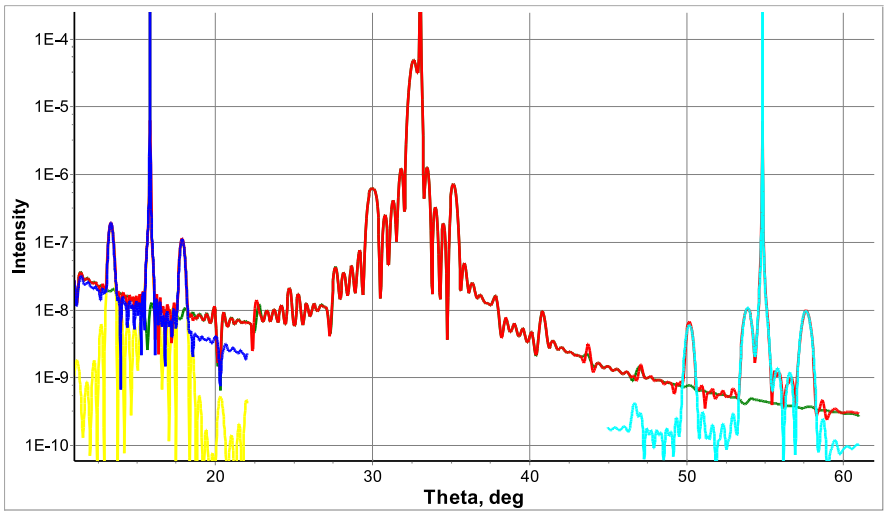


Figure 6. Wide-angle diffraction from InGaAs/GaAs SL (red), 002–blue (yellow – w/o specular reflected), 004–green, 006–cyan.

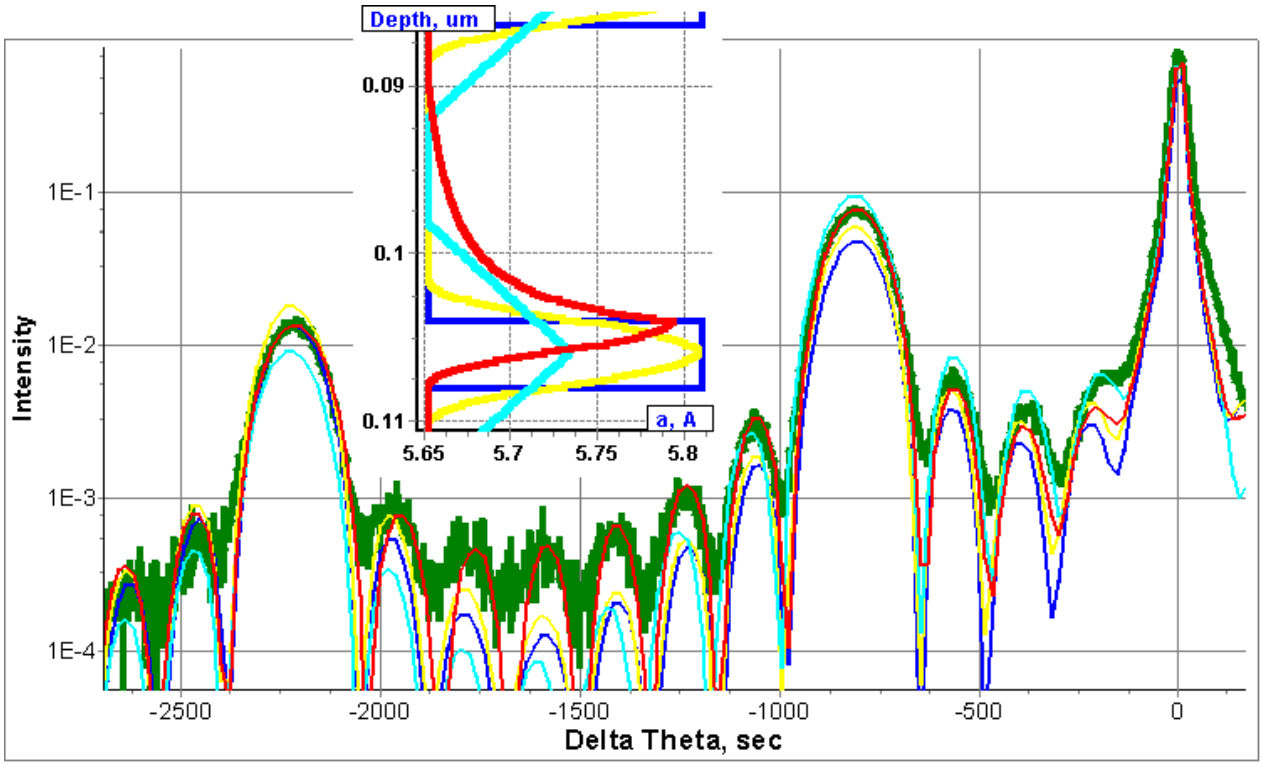


Figure 7. Experimental (green) and simulated DC for 113 reflex from 8-period superlattice $In_{0.2}Ga_{0.8}As/GaAs$ (40/113 Å) with: sharp (blue), linear (cyan), quadratic (yellow) and quadratic-hyperbolic (red) composition gradient on layers boundaries (on the insertion axis x is normal lattice parameter and axis y is depth of sublayer deposition).

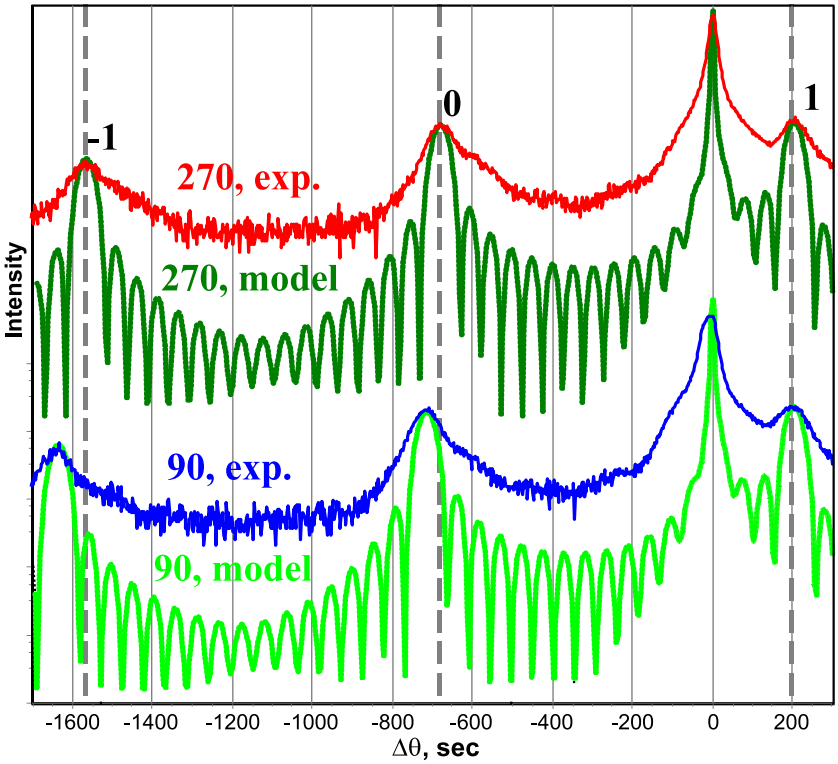


Figure 8. Experimental (red and blue) and simulated (green and light-green) DC for InGaAs/GaAs superlattice. Azimuthal angles 90 ° (2 upper curves) and 270 ° (2 lower).

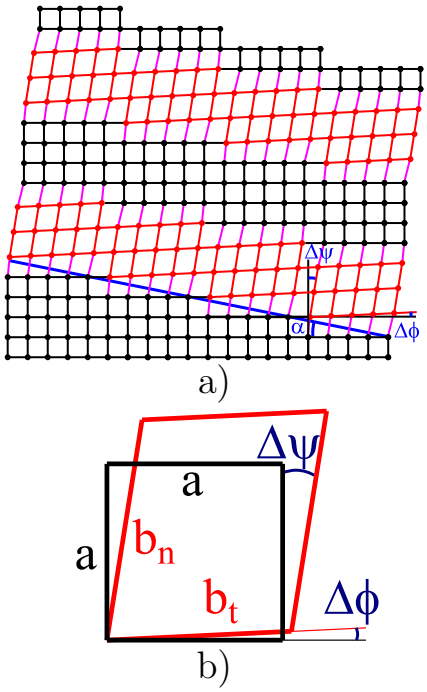


Figure 9. Deformed lattice of $In_{0.3}Ga_{0.7}As/GaAs$. Black – GaAs, red – InGaAs. a) whole structure, b) unit cell.

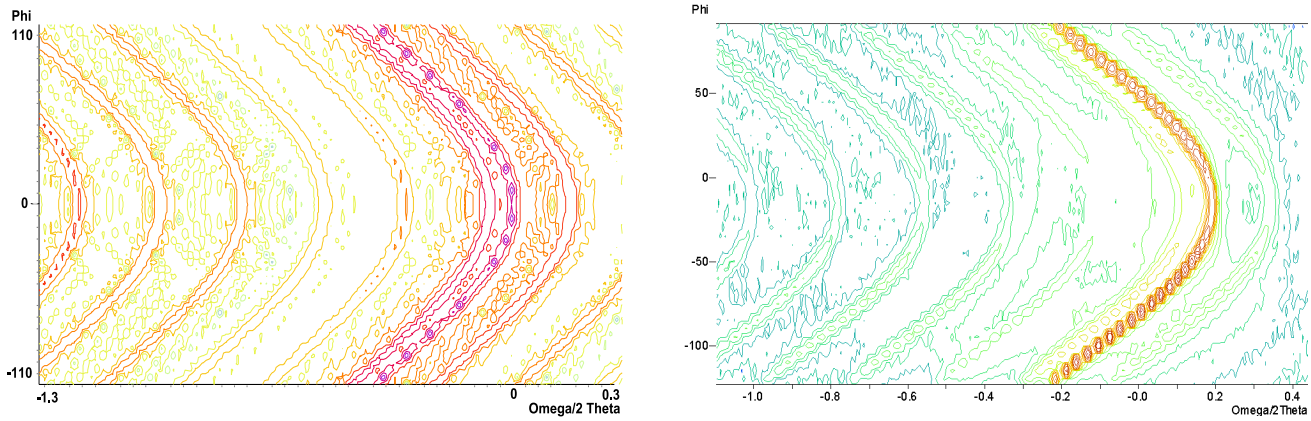


Figure 10. Azimuthal maps from 8-period superlattice $In_{0.2}Ga_{0.8}As/GaAs$ (40/113 Å) on GaAs substrate. Left – simulation, right – experiment. Phi – azimuthal angle.

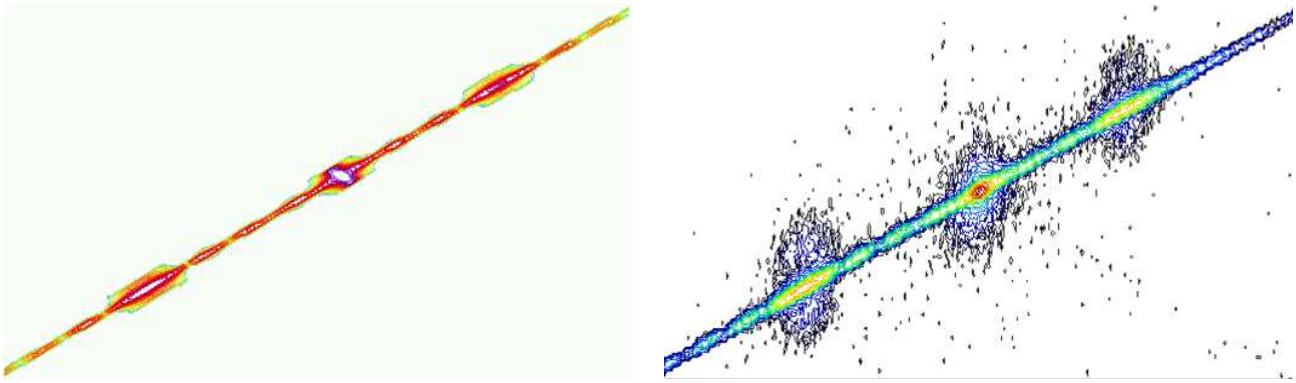


Figure 11. Reciprocal map for 113 reflex from the structure, described under fig. 10. Left – simulation, right – experiment. Axis x– $\omega/2\theta$ DC, axis y – ω.

in Bragg case is analyzed. Four types of composition gradients were analyzed but the best one was hyperbolic gradient on the InGaAs/GaAs layers boundary. The fact that layers thicknesses determination could be done incorrectly with assumption of sharp borders between layers is underlined.

5. The algorithm of correlation of experimental curves with simulated is proposed for better structure parameters (thickness, strain and Debay-Waller factor) determination.

6. Methodological basis, algorithms and software programs were designed for quantitative diagnostics of multilayered structures with the help of high-resolution X-ray diffractometry.

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[11] *Lomov A.A., Chuev M.A., etc.* //JTF letters – 2004.– Vol.30, N 10.– P.89–95.

PUBLICATIONS

On the materials of thesis 21 publication were made. Among them 9 articles and 12 proceedings of conferences. The most intresting publications are listed below.

1. Strelchuk V.V., Kladko V.P., Yefanov O.M., Kolomys O.F., Gudymenko O.Yo., Valakh M.Ya., Mazur Yu.I., Wang Z.M., Salamo J.G. Anisotropy of elastic deformations in multilayered (In, Ga)As/GaAs structures with quantum wires: X-ray diffractometry study. // Semicond.Phys.Quantum&Optoelectronics – 2005. – V.8, N1. – P.36-45.

2. Yefanov O., Kladko V., Gudymenko O., Strelchuk V., Mazur Yu., Wang Z.M., Salamo J. Fields of deformation anisotropy exploration in multilayered (In,Ga)As/GaAs structures by high-resolution X-ray scattering. // Phys. Status Sol.(a). – 2006. – V.203, Issue 1. – P.154-157.

3. Yefanov O.M., Kladko V.P. Dispersion equation solving for two-beam case. (rus) // Met.Phys. and New Techn. – 2006. – V.28, N2. – P.231-248.

4. Yefanov O.M., Kladko V.P. Simulation of X-ray diffraction from multilayered structures with different composition gradient on the layers boundaries. (rus) // Met.Phys. and New Techn. – 2006. – V.28, N5. – P.619-629.

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6. Borkovska L., Yefanov O., Gudymenko O., Johnson S., Kladko V., Korsunska N., Kryshtab T., Sadofyev Yu., Zhang Y.-H. Effect of Growth Temperature on the Luminescent and structural properties of InGaAsSbN/GaAs quantum wells for 1.3um telecom application. // Thin Solid Films. – 2006. – V.515, Issue 2. – P.786-789.

7. Yefanov O.M., Kladko V.P., Saveleva I.S. Dynamical calculation of reciprocal space maps for partially relaxed multilayered structure by noncoplanar N-beam algorithm. (rus) //Proceedings of V.Novgorod seminar. – 2006. – P.148-152.

8. Kladko V.P., Yefanov O.M., Slobodyan M.V., Machulin V.F. Lateral composition modulation and 2D-3D structure transformation investigation in multilayered periodical structures InGaAs/GaAs by high-resolution X-ray difractometry. (rus) //Proceedings of V.Novgorod seminar. – 2006. – P.167-169.

9. Yefanov O.M., Kladko V.P., Slobodyan M.V., Machulin V.F., Strelchuk V.V. A new approach for anisotropic deformation in multilayered structures analysis. (rus) //Proceedings of V.Novgorod seminar. – 2006. – p.233.

10. Kladko V.P., Yefanov O.M., Slobodyan M.V., Strelchuk V.V., Mazur Yu.I., Wang Zh.V., Salamo G.J. Structural Anisotropy and Optical Properties InGaAs/GaAs(100) Quantum Dot Chains Structures. // Proceedings of XTOP-2006. – p. 144.

11. Kladko V., Yefanov O., Slobodyan M., Machulin V., Borkovska L., Sadofyev Yu. HPXRD Investigations of Deformation Fields InGaAsSbN/GaAs Heterostructures With Quantum Well. // Proceedings of XTOP-2006. – p. 162.

12. Yefanov O.M., Kladko V.P. Simulation of Reciprocal Space Maps for Partially Relaxed Multilayered Structures by 3d Dynamical Multi-Beam Theory. // Proceedings of XTOP-2006. – p.174.

ABSTRACT

Yefanov O.M. Dynamical X-Ray Diffraction in Multilayered Structures. – Manuscript.

Dissertation for the Ph.D. degree by speciality 01.04.07 – solid state physics. V. Lashkaryov Institute of Semiconductor Physics of National Academy of Sciences of Ukraine, Kyiv, 2006.

The thesis is devoted to investigation of X-ray diffraction in multilayered structures. A new approach for calculation of N-beam dynamical diffraction in planar multilayers is developed and its applications for experimental data analysis are shown. The main features of proposed model are: plane waves are considered; propagation equation, derived from Maxwell equations, and boundary conditions are solved without simplifications; suitable for any geometry (Bragg, Laue, Bragg-Laue); 3D simulation in reciprocal and 1D in real space; covers the whole angular range, including grazing angles, backscattering and specular reflection; precise calculation of any polarization with information about diffracted and reflected beams polarization; no principal limitation on quantity of reciprocal lattice points taken part in diffraction; arbitrary layers thickness (valid for both thick and thin layers); all calculation are carried out for x,y,z projections of wave vectors, diffraction vectors and electric field. The examples of developed approach application, such as diffraction curves (DC), reflection curves,

Renninger scan and reciprocal space maps are shown.

One more method for N-beam diffraction in multilayers calculation is presented. This method is less common and is suitable only for coplanar case and only sigma polarization, but it is faster then 3D algorithm described above.

Dispersion equation for 2, 3, 4 and N-beam diffraction are numerically solved and dispersion surfaces for these cases are drawn. For two-beam case the simplified (quadratic) and exact (quartic) equations are solved analytically. Absorption is taken into account and different geometries as well. For many-beam cases (more than two) the way for avoiding numerical problems is shown. These problems arise if the wave vectors are expressed in Cartethian coordinates with the origin in (000) point. This is explained by the fact that there could be many origins of wave vectors near the same Lorentz point, so the precision of computer calculations is not enough to distinguish the difference between solutions (its order is about 10^{-5}) while the value of these roots is about 1. That's why the origin of coordinate system must be placed in Lorentz point.

The influence of composition gradient on boundaries between GaAs and InGaAs layers in 8-period superlattice is analyzed. Four type of gradient functions were considered: sharp, linear, quadratic and quadratic-hyperbolic. With the last function the best coincidence between simulated and experimental DC near substrate peak was achieved. But for good intensity correlation on the far tails of DC different parameters of composition gradient must be set for each boundary in superlattice. For this reason autofit procedure was used and this gave better results.

Anisotropic deformations investigation was done with the help of measuring azimuthaly dependent diffraction curves. It allows to explain satellite peaks period dependence on azimuthal angle and shift of the zero satellite. The first fact is well known to be explained by the structure diffraction vector misorientation to the surface normal, while the latter is more interesting and is explained by local InGaAs layers misorientation.

A new method of structure analysis via calculation of two-dimensional maps of azimuthal dependent intensity distribution is presented (one axis is usual $\omega/2\theta$ and the other is azimuthal angle). The advantages of such approach for multilayers analysis is underlined.

The program developed for diffraction calculation is described. Among with original methods mentioned above it incorporates the best algorithms for X-ray diffraction in multilayers calculation, such as Bartels recursion formula (Bartels, Acta Cr.A, 1986), Transfer Matrix Algorithm (Stepanov, Cr.Rep., 1994), Recursion Matrix Algorithm (Stepanov, Phys.Rev.B, 1998), 1D Takagi recursion equation (Fewster, 2003), N-Beam Transfer Matrix Algorithm (Stepanov, Acta Cr.A, 1994), Souvorov 3D algorithm (Souvorov, Phys.Rev.B, 2004). The part of the program is "open source" C++ library developed to calculate the polarizability of complex structures (http://x-ray.net.ua/software.php). The program is equipped with comfortable interface and it was applied for all simulations described in the thesis.

Keywords: dispersion surface, X-ray diffraction, quantum layers, multi-beam diffraction, deformation, superlattices.