INVESTIGATION OF GaAs/AlAs SHORT-PERIODIC SUPERLATTICES BY HIGH-RESOLUTION X-RAY DIFFRACTOMETRY

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Scattering of X-rays in GaAs/AlAs short-periodic superlattices (SL) is studied by high-resolution diffractometry. The diffraction parameters and characteristics of SL, i.e., the thickness of separate lattices and the level of their deformation, are determined by fitting the calculated diffraction curves to experimental ones. The semikinematic theory of X-ray scattering was used in calculations. Relatively high crystal coherence and structure perfection of the neighbor layers are established to occur despite the high level of elastic deformation in every layer.

Introduction

In recent years, a considerable attention was paid to the study of properties of quantum dimensional SL GaAs/AlAs, which is related to the perspective of their practical use, in particular in devices of optoelectronics. In this aspect, of great importance is the information on the structure of these objects concerning the planarity of the boundaries of layers the uniformity of the thickness of layers, the diffusion processes on boundaries, impurities and microdefects in periodic structures. All these structural imperfections influence the electrophysical and optical characteristics of SL to a significant extent.

The most sensitive and informative method of nondestructive study of a real structure of SL is high-resolution X-ray diffractometry [1—3]. In this approach, the diffraction of X-rays on structures, in which the lattice constant or scattering intensity is modulated along one of the coordinates, is characterized by the presence of satellites near the Bragg peak of an averaged lattice [4, 5]. The scattering intensity of X-rays which can be characterized by satellite peaks is proportional to the Fourier-components of the coordinate-dependent modulation of the system composition [6]. Therefore, the mutual diffusion of GaAs and AlAs in SL, e.g., can be studied by measuring the intensity of satellites as a function of the annealing duration at a constant temperature [7]. In addition, a change in the width of satellite peaks in the reciprocal space bears the information on perfection of films and, in particular, on coherence of layers of SL.

The goal of this work is to study the structural defects, geometric parameters, and elastic state of layers of short-periodic SL GaAs/AlAs by methods of high-resolution X-ray diffractometry. We compare the experimentally derived curves of diffraction reflection (CDR) with theoretically calculated ones (by using the so-called semikinematic approximation of the theory of scattering of X-rays [4] when the thicknesses of separate layers of SL are small as compared with the extinction length). The purpose of such a procedure was a determination of the parameters of real SL, their separate components, and the characteristics of structural incommensurability in heteroboundaries.

Theoretical Foundation of the Method

In the calculation of X-ray diffraction patterns, the first iteration of the Topin equation proposed in [8] is usually used. X-ray scattering by a uniform epitaxial layer is described with the following parameters: the thickness t, Fourier coefficients of the crystal polarizability χ, stresses ε, and the coefficient of absorption μ. The main characteristics of diffraction in an epitaxial layer with thickness t are A, which is inversely proportional to the extinction length, and the parameter Y defined by

$$A = \frac{\pi \gamma_h t}{\lambda \sqrt{|\gamma_0 \gamma_h|}}, \quad (1)$$

$$Y = -\sqrt{\frac{\gamma_0}{|\gamma_0|}} \frac{\sin(2\vartheta_\mathrm{B})}{|\chi_h|} \Delta\omega. \quad (2)$$

Here, λ, ϑ_B, γ_0 and γ_h stand for, respectively, the X-ray wavelength, the Bragg angle for a substrate, and the diffraction cosines for a primary and diffracted beams; $\Delta\omega$ is the value of deviation of the crystal from the Bragg position.

Analyzing the reflectivity of real SL for the given X-ray emission, it is necessary to consider mechanical stresses (deformations) in the epitaxial film, because they can be directly determined if the shape of CDR is known. We denote the characteristics of the film and substrate by indices f and s, respectively. The difference of interplanar distances depends on a specific deformation and an orientation of atomic planes $\Delta d = d_f - d_s$. In elasticity theory, the above-mentioned stresses (deformations) normal, $\varepsilon_\perp$, or parallel, $\varepsilon_\parallel$, to the crystal surface are defined by a relative change of interatomic distances in the film and substrate:

$$\Delta d/d_s = \varepsilon_\perp \cos^3\Psi + \varepsilon_\parallel \sin^2\Psi, \quad (3)$$

where Ψ is the angle between the planes and the crystal surface. The differential angle $\Delta\omega$ equals

$$\Delta\omega = \vartheta - \vartheta_B + (\varepsilon_\perp \cos^2\Psi + \varepsilon_\parallel \sin^2\Psi)\times$$

$$\times \mathrm{tg}\vartheta_B \pm (\varepsilon_\perp + \varepsilon_\parallel)\sin\Psi\cos\Psi, \quad (4)$$

where ϑ is the incident angle for X-rays.

An arbitrary distribution of the stresses ε and the structural factor F of the epitaxial film over depth can be represented by a discrete structure composed from N sublayers. The normalized diffraction amplitude of this structure reads

$$E_N = i\sqrt{\frac{\gamma_0}{|\gamma_h|}}\sum_j^N a_j \exp[-i(A_jY_j + \Phi_j)]\frac{\sin(A_jY_j)}{Y_j}, \quad (5)$$

where $a_j = \exp\left[-\mu\left(\frac{\gamma_0+|\gamma_h|}{2|\gamma_0\gamma_h|}\right)\sum_{i=j+1}^N t_i\right]$ is the absorption factor, $a_N = 1$, and $\Phi_j = 2\sum_{i=1}^{j-1} A_iY_i$ is the phase factor such that $\Phi_1 = 0$.

Here, SL represents an arbitrary epitaxial layered structure. In the simplest case, its period is formed by two layers a and b. Each of them is characterized by its thickness, stresses, structural factor, and the parameters A and Y. The diffraction amplitude for an M-periodic two-layer SL is [5]

$$E_M = i\sqrt{\frac{\gamma_0}{|\gamma_h|}}\exp(-i\beta)F_S\frac{\sin[M(A_aY_a + A_bY_b)]}{\sin(A_aY_a + A_bY_b)}, \quad (6)$$

where $\beta = (M-1)(A_aY_a + A_bY_b) + A_aY_a$, and $F_S = \frac{\sin(A_aY_a)}{Y_a} + \exp[-i(A_aY_a + A_bY_b)]\frac{\sin(A_bY_b)}{Y_b}$ can be considered as a structural factor for one period of SL. The sine in the numerator of (6) becomes zero with period $\Delta\vartheta_M$ which is determined from the condition $M(A_aY_a + A_bY_b) = \pi$. This factor describes the so-called rapid pendular oscillations of the intensity on CDR. In view of (1) and (2), we get

$$\Delta\vartheta_M = \frac{\lambda|\gamma_h|}{M(t_a + t_b)\sin(2\vartheta_B)}, \quad (7)$$

which allows us to determine the full thickness of the superlattice $T = M(t_a + t_b)$.

The sine in the denominator of (6) corresponds to other (slower) oscillations of the scattering amplitudes in registering a CDR. The zero positions of oscillations are determined from the relation

$$A_aY_a + A_bY_b = n\pi, \quad (8)$$

and are denoted as $n = 0, \pm1, \pm2....$ Having known the angle distance between these peaks $\Delta\vartheta_p$, we may determine the period of SL as

$$p = t_a + t_b = \frac{\lambda|\gamma_h|}{\Delta\vartheta_p \sin(2\vartheta_B)}. \quad (9)$$

The superlattice-induced peak of a zero-order satellite is located from the peak from the substrate at the angle distance defined by $\Delta\vartheta_0$,

$$-\Delta\vartheta_0 = k_1\langle\varepsilon_\perp\rangle + k_2\langle\varepsilon_\parallel\rangle,$$

where

$$k_1 = \cos^2\Psi \mathrm{tg}\vartheta_B \pm \sin\Psi\cos\Psi,$$

$$k_2 = \sin^2\Psi \mathrm{tg}\vartheta_B \pm \sin\Psi\cos\Psi. \quad (10)$$

Brackets for deformations mean the averaging over the period of SL.

The amplitude of n-order satellites is proportional to the value of the structural factor of SL in view of condition (8):

$$F_{Sn} = \frac{\sin(A_aY_{an})}{A_aY_{an}}\left(A_a + \frac{A_b}{1 - \frac{n\pi}{A_aY_{an}}}\right). \quad (11)$$

Because the angle parameter of a satellite is connected with Y_{an} through Y_{a0} [formulas (9) and (2)], it depends only on A_a, A_b, and Y_{a0}. At $n = 0$, relation (11) acquires a simple form:

$$F_{S0} = \frac{\sin(A_aY_{an})}{A_aY_{an}}(A_a + A_b) \approx A_a + A_b. \quad (12)$$

By measuring the ratio of the scattering amplitudes of satellites of n-order and zero order, we can calculate the thickness of one of the SL layers and the mean stresses in it. But the most suitable in this case are the so-called quasiforbidden 200-type reflexes, as the analysis of relations (11) and (12) shows. The latter are more sensitive to deformations, because the parameter A_a related to them is sharply decreased relative to structural reflexes. Their application in the study of multilayered structures will be considered elsewhere.

Experimental Procedure and the Object of Studies

The measurement of CDR for symmetric 400- and 200-reflexes and asymmetric 311-reflex was performed at the Institute of Physics of PAS on the 3-crystal spectrometer of the "Philips" firm. The intensities of reflected rays were registered by a detector with the system of slits. The specimen was scanned near the Bragg exact position near 4° in the so-called $\omega/2\vartheta$-mode. The measurements were carried out in a discrete angular mode with a step of $2''$. For the given parameters of accumulation of the number of impulses, the signal-to-noise ratio reached 10^6. While analyzing the experimental data, we compared the experimental and theoretical CDR by fitting with the use of the so-called χ^2 method, which allows one to find the average values of parameters and their errors. We normalized the experimental spectra by maxima of the reflectivity of SL.

The technology of derivation of SL consisted in the following. First, a buffer layer of GaAs of 20 nm in thickness was grown on the substrate GaAs(001). Then the growth was terminated in 60 s, and further 50 periods of SL (AlAs of 1.13 nm in thickness and GaAs of 2.26 nm in thickness) was added. This whole structure was covered by one layer of AlAs (30 nm) and 30 layers of GaAs (6 nm). Thus, we got specimen 1. The next specimen 2 differed from the previous one by only the thicknesses of layers of SL (1.413 nm for AlAs and 2.83 nm for GaAs). The last specimen 3 had the following structure: the buffer layer of 200 nm in thickness was covered with a layer of $Al_{0.5}Ga_{0.5}As$ of 25 nm in thickness and then 200 periods of SL of the total thickness of 500 nm with layers which consisted of 3 monolayers of AlAs and 6 monolayers of GaAs.

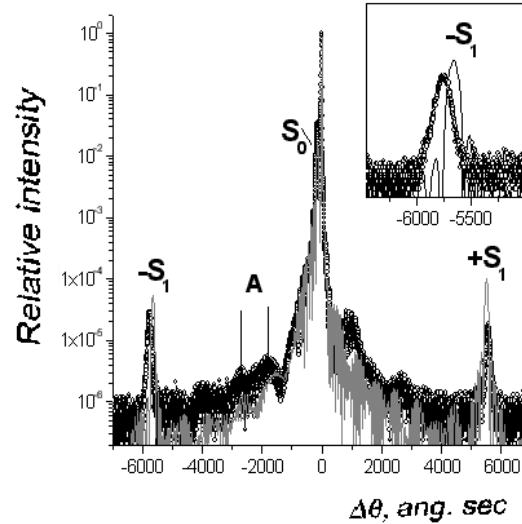
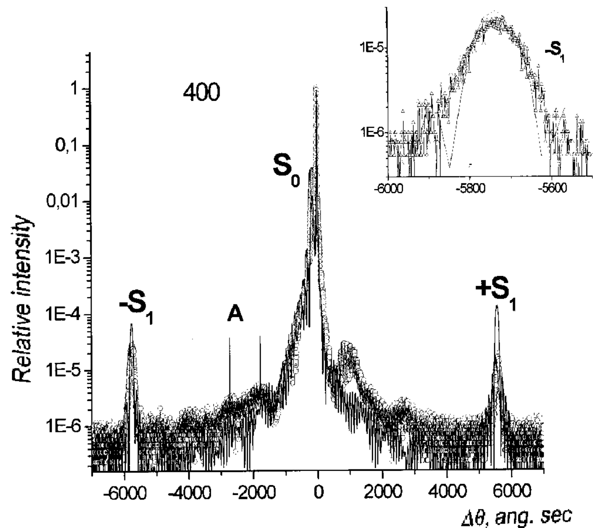


Fig. 1. CDR for 004 reflection from SL in GaAs/AlAs: points — experiment, solid line — the result of calculations of the diffraction pattern with the use of technological parameters. The insert shows the position of the first-order satellites ($-S_1$)

Results and their Discussion

Fig. 1 shows the diffraction spectra from superlattice structures for 004 reflection. Their analysis indicates the presence of clearly pronounced satellites $\pm S_1$ caused by the SL period and a number of other peaks from pendular oscillations of the intensity. Some peaks located near the zero-order satellite S_0 can be referred to the so-called fast oscillations whose periodicity is described by (1). The other peaks marked by A in Fig. 1 are caused by the presence of the buffer layer and define its thickness. As seen, the zero satellite possesses the highest intensity, which is a sign of short-periodic SL [5]. The technological parameters of the SL structure were used as the first approximation in the quantitative analysis of experimental curves during a theoretical fitting. The behavior of the calculated curve indicates that these parameters do not correspond to the true data for both the SL period thickness and the ratio of the thicknesses of these layers (see the insert). For such a choice of the initial conditions of the fitting, we have got a large value of the parameter $\chi^2 = 2300$. Fig. 2 demonstrates experimental and calculated curves, 004 reflection, for specimen 1. In the insert of Fig. 2, we give the positions of the experimental and calculated satellites $-S_1$. As seen the experimental and calculated CDR coincide with rather high accuracy ($\chi^2 = 4.67$). The structural characteristics of layers and the lattice parameters derived by fitting are presented in Table 1.



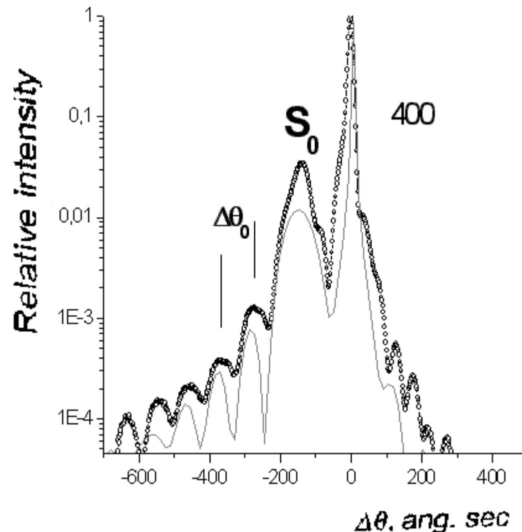


Fig. 2. CDR for 004-reflection from SL: points — experimental data, solid line — the result of fitting. Symbols $-S_1$, $+S_1$, and S_0 stand for the position of satellites, the distance A is defined by the buffer layer thickness (20 nm). The insert shows the calculated and experimental positions of satellite ($-S_1$)

Fig. 3. Fragment of CDR for 004-reflection from SL presented in Fig. 2 on a larger scale: points — experiment, solid line — the theoretical result with fitted parameters. The position of zero-order satellite S_0 and the distance between maxima of fast oscillations $\Delta\vartheta_0$ which define the thickness of the whole SL are shown

These data indicate a certain inconsistency of experimental and technologically set values of the layer thicknesses for SL. Using the angle position of the zero satellite, the average value of deformation in the SL period given in Table 2 was calculated by means of formula (2). Knowing the layer thicknesses, we also determined the lattice period of each layer according to [1]. The period thickness can be calculated from the period of the sinusoidal denominator in (6). Despite the ratio of the thicknesses of SL layers set by technology is the same for all SL, the average deformations are different. This fact confirms the inconsistency of set and real parameters of SL once again.

T a b l e 1. Parameters of SL layers

Number of a specimen	Parameters of SL set by technology, nm	Experimental values of parameters of SL from 004 reflection, nm	Error of parameters, %
1	1.13 (AlAs) 2.26 (GaAs)	1.1 (AlAs) 0.003 (AlGaAs) 2.24 (GaAs)	0.2
2	1.413 (AlAs) 2.83 (GaAs)	1.414 (AlAs) 0.004 (AlGaAs) 2.863 (GaAs)	0.4
3	3ML (AlAs) 6ML (GaAs)	0.835 (AlAs) 1.736 (GaAs)	0.134

The values of the total thicknesses of SL and the whole structure were determined by utilization of the distance between the above-mentioned oscillations of the intensity presented in Fig. 3 (the section of CDR in Fig. 2 which is shown on a larger scale). A further decrease in χ^2 can be apparently attained with regard for the

T a b l e 2. Parameters of deformation in the period and separate layers of SL

Number of a specimen	Position of zero-satellite, ang. sec	Mean deformation in the period, %	Deformation in layers of AlAs and GaAs, %	Interplanar distances (004) in AlAs and GaAs, nm	
				Exp.	Theor.
1	–139	0.1038	0.175 –0.081	0.14330 0.14053	0.14155 0.14133
2	–143	0.1068	0.178 –0.073	0.14341 0.14060	—
3	–123	0.0925	0.140 –0.039	0.14295 0.14094	—

presence of the diffuse scattering component of X-rays according to [9]. The presence of fast oscillations of the intensity on CDR testifies to the pseudomorphic growth of SL, because the dislocations of the incommensurability of lattices smear out the zero satellite and others as well [10].

In view of the rather narrow half-widths of satellites $\pm S_1$ (Fig. 2), we may assert to a certain extent that the structure of SL has a high structural perfection, crystalline coherence, and planarity of layers [8]. For example, the estimates of mean deviations for the SL period and the thickness of separate layers gave the values, respectively, of 0.0003 and 0.0001 nm. Now we briefly discuss the form of experimental satellites. The analysis of CDR shape (Fig. 2, the insert) shows that they are asymmetric relative to the intensity maximum. This fact can be interpreted as a breaking of the symmetry of the distribution of stresses in layers. With the use of the ratio of the intensities of zero- and first-order satellites, we determined the layer thicknesses in the period and the value of deformation in a layer.

The experimental and theoretical curves for the asymmetric 113-diffraction are given in Fig. 4. As seen, a rather satisfactory coincidence of both curves by the angle positions of the maxima of oscillations takes place for the considered angle interval. However, the amplitudes of these pendular oscillations are somewhat decreased relative to the theoretical curve, which can be related to the presence of defects of the unknown nature in a real SL.

Conclusions

The performed X-ray diffraction study of short-periodic superlattices have shown the possibility to obtain the quantitative information on their structural parameters (the true period of SL, the degree of stress in layers, and the lattice constant for them). We demonstrated a certain difference of the experimentally determined period of SL and technologically set parameters. This difference goes far beyond the errors of determination of these parameters. The simultaneous analysis of symmetric 400- and asymmetric 113-reflexes allows us to get the information on the deformations normal or parallel to the structure surface. The results of the studies carried out indicate that the SL possess a rather high structural perfection with clearly pronounced boundaries despite the large values of deformation, which is supported by the small widths of satellites and the presence of a fine structure of the spectrum (fast oscillations).

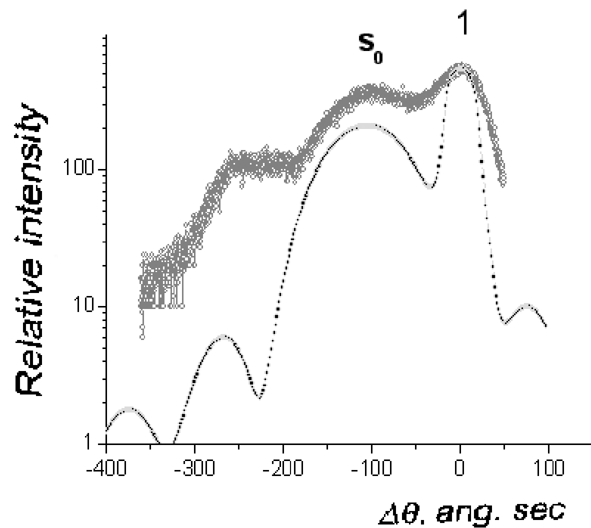


Fig. 4. CDR for asymmetric 113 reflection from SL in GaAs/AlAs: points — experiment, solid line — the result of numerical fitting of the experimental data. The position of satellite S_0 is shown

Specimens were produced at P. Drude Institute of Solid-State Electronics (Berlin).

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ДОСЛІДЖЕННЯ КОРОТКОПЕРІОДНИХ НАДГРАТОК GaAs/AlAs ЗА ДОПОМОГОЮ ВИСОКОРОЗДІЛЬНОЇ РЕНТГЕНІВСЬКОЇ ДИФРАКТОМЕТРІЇ

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Р е з ю м е

Методом високороздільної рентгенівської дифрактометрії досліджено розсіяння рентгенівських променів в короткоперіодних надгратках (НГ) GaAs/AlAs. Шляхом підгонки теоретичних кривих дифракційного відбиття до експериментальних отримано як параметри дифракції, так і структурні параметри НГ (товщини субшарів, рівень деформації в шарах). Встановлено, що, незважаючи на значні величини деформацій в шарах НГ, їх когерентність та структурна досконалість досить висока.