INFLUENCE OF THE CURVATURE RADIUS OF MULTILAYER STRUCTURES ON X-RAY DIFFRACTION SPECTRA

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By the method of high-resolution X-ray diffraction, we have established the effect of a change of the period of a multilayer periodic structure on a change of its curvature radius. We have explained this phenomenon observed as a variation of the positions of peaks of the satellite structure on the diffraction spectra of X-rays under the influence of a change of the deformation along the growth direction of the structure caused by the diffusion of components in the process of thermoannealing.

1. Introduction

In recent years, the phenomenon of self-ordering in semiconducting heterostructures InGaAs/GaAs with quantum dots or chains of quantum dots is the object of intense studies. To attain the optimum conditions of their growth, it is necessary to establish the mechanisms of interaction between the elastic relaxation of deformations and the degree of ordering of the formed islets. This requires the information about a number of structural and deformational characteristics of the structure, in particular, about the thickness of layers, the structure period, and the values of deformation of the lattice in layers and on boundaries. The question of a change of the period of a multilayer periodic structure, a superlattice (SL), on the disorientation of the substrate was studied in [1, 2].

However, there exist the differences in the interpretation of values of the periods of the SLs of structures under the action of changes in the deformation fields induced by the processes of self-ordering of quantum dots and various external factors [3–5]. We also indicate certain ambiguities in the interpretation of factors that influence the process of change of the position of the zero satellites of an SL (the mean deformation over a period). The search for the information about the effect of the elastic bending of a multilayer system on the behavior of diffraction patterns gave no required result. At the same time, it was shown in [6] that, on the elastic bending of a single crystal, there appears a linear change Δd in the interplane distance d: $\Delta d/d \approx -\mathrm{ctg}\theta_{\mathrm{B}}\Delta\theta^{(\mathrm{bend})}$, which together with refraction causes an additional change $\Delta\theta^{(\mathrm{bend})}$ in the Bragg angle. Moreover, this decrement depends on the depth of penetration of the radiation into a crystal, the curvature radii of the crystal in the plane of diffraction and in the normal plane, and the crystal thickness.

A number of works were devoted to theoretical studies of the influence of the curvature of a crystal on the angle position and the expansion of diffraction peaks [7–9]. The problem of relationship of a deformation and the curvature for multilayer structures was considered in [10]. However, a number of diffraction effects remain to be unexplained and have no proper interpretation.

Therefore, a goal of this study was both the establishment of the relationship between the mean deformation in layers of the structure and its curvature and the effect of such a curvature on the SL periodicity determined in X-ray measurements.

Our second important task is the establishment of the causes of a change in the period of the SL as the annealing temperature varies, as well as the simulation of the influence of a change in both the curvature and the level of variations in the SL period on the distance between satellites, on the basis of the obtained experimental data.

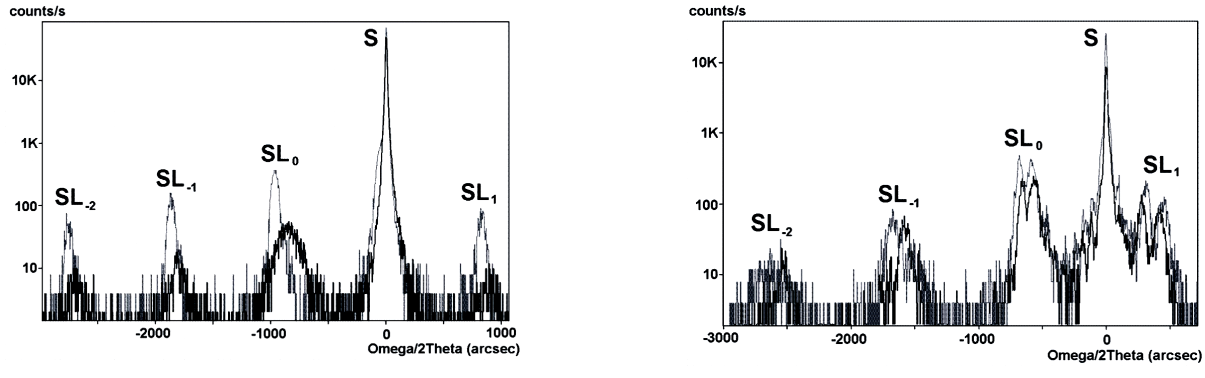

Fig. 1. Symmetric 004 reflection curves *a*) sample *1*: grey curve – initial (annealed at 630 °C), black curve – annealing at 700 °C; *b*) sample *2*: grey curve – initial, black curve – annealing at 600 °C

2. Experiment

We grew 17-period superlattices in $In_xGa_{1-x}As/GaAs$ with chains of quantum dots on the semiinsulating substrate of gallium arsenide (100) on the MPE setup. The buffer layer of GaAs of 0.5 μm in thickness was grown at a substrate temperature of 580 °C. Then the substrate temperature was reduced to 500 °C in order to apply the epitaxial layers with parameters given in Table 1. The samples underwent the rapid thermal annealing in the argon atmosphere for 30 s (sample *1*) and 75 s (sample *2*) at different temperatures.

The measurement of structural characteristics of samples was performed on a high-resolution X-ray diffractometer PANalytical X'Pert Pro MRD XL with the use of a 4-fold (220) Ge monochromator and a 3-fold analyzer of the same type. In order to determine the curvature, SL period, lattice constant, and deformations, we carried out the registration of the diffraction rocking curves (DRC) of CuK_α radiation for symmetric 004 reflection.

3. Results and discussion

In Fig. 1, we present the results of diffraction measurements. The DRCs show the lateral averaged SL structure of specimens. On these curves, we see SL satellites of high orders which serve a qualitative criterion of the structural perfection of specimens in the initial state. The peak marked as S corresponds to the diffraction from the substrate, and SL_n stands for the n-th satellite peak of the laterally averaged SL. The distance between satellites is $2\pi/T$ (T is the SL period), and the distance between SL_0 and the peak from the substrate is proportional to the relative difference of the vertical lattice constant of the substrate and the mean parameter of the SL. It is worth noting that, for the samples under study, the period does not depend on their azimuthal position relative to the crystallographical direction of the diffraction plane.

It is seen from the plots in Fig. 1 that the SL period (the distance between satellites of the SL) changes as the annealing temperature varies. Values of the SL periods obtained experimentally and their change with the temperature are given in Fig. 2.

The position of zero-order satellites relative to the peak from the substrate indicates the deformation level averaged over the period and the concentration of In in a layer of $In_xGa_{1-x}As/GaAs$. It changes its position as the annealing temperature varies (Fig. 1). Mainly, these shifts of zero satellites relative to the GaAs substrate with increase in the annealing temperature can be caused by the relaxation of stresses under the action of interdiffusion. However, this cannot explain a change in the period, i.e. in the distance between satellites on the thermal influence.

For all the samples, we observed a change in the vertical period of the SL as the annealing temperature was varied (Figs. 1 and 2). An analogous effect was seen in [11], though without any explanation. It was shown in [12] that a change in the sizes and the composition of quantum dots after a fast thermal annealing is related to the interdiffusion of atoms of In and Ga on the interface between layers. For temperatures of a fast annealing of the order of 600–700 °C, no significant changes in

T a b l e 1. Technological parameters of samples under study

Sample number	t_1, InGaAs	t_2, GaAs	Content of In, x
1	5.7 ML	67 ML	0.5
2	7.6 ML	67 ML	0.4

structural and optical properties was observed, despite an increase in the interdiffusion rate of atoms of In and Ga between quantum dots and the wetting layer [3]. Despite the fact that these processes are dominant during the annealing, they have no such effect on the periodicity of the SL. The mean SL remains invariable, because the material does not leave the crystal. As a single real reason for the change in the SL period, we can indicate the effect of curvature which is inherent to all epitaxial structures with elastic stresses between layers.

The results of measurements demonstrate that the curvature radius in the process of fast thermal annealing increases for sample *1* and, on the contrary, decreases for sample *2*. This can be explained by the relaxation of stresses between layers of the SL.

A linear dependence of the mean parameter of the SL on the shift in the two-layer approximation is given by the relation [6]

$$\Delta a_{\rm av}/a_{\rm av} \approx -{\rm ctg}\theta_{\rm B}\Delta\theta, \qquad (1)$$

where $a_{\rm av}$ is the mean parameter of the SL, and $\Delta\theta$ is the shift of the zero satellite.

According to [10], we can express a deformation in terms of the curvature radius of the crystal as

$$\frac{\Delta a}{a} = \frac{t_0^2}{6t_1}\frac{1+6(t_1/t_0)}{R}\frac{1+\nu}{1-\nu}, \qquad (2)$$

where t_0 is the substrate thickness, t_1 is the SL thickness, R is the measured curvature radius, and ν is the Poisson's ratio.

The results of calculations of deformations by formulas (1) and (2) are presented in Table 2.

As seen, the results of calculations of deformations for sample *1* correlate with one another, but not for sample *2*. Probably, this is related to the complicated structure of the sample itself. The results given in our previous work [13] showed the presence of two individual regions with different periods in this sample. This is seen also on the spectrum presented in Fig. 1*b* (the splitting of coherent satellites). That is, it is wrong to use the two-layer approximation in the calculation of deformations for such a structure.

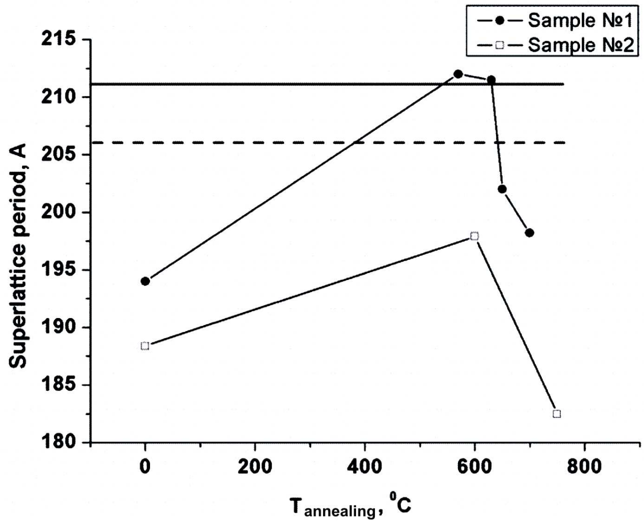

Fig. 2. Vertical period of the SL versus the annealing temperature. Dashed and solid lines give the values of technologically prescribed periods, respectively, for samples *1* and *2*

As seen from the results of measurements given in Table 2, the increase in the curvature radius of the crystal leads to a decrease in the SL period.

Both samples are characterized by the same behavior of the dependence of the vertical SL period on the annealing temperature (Fig. 2). As seen from this figure, the periods of initial samples are significantly less than those prescribed technologically. This is related to the fact that, in the process of growth of epitaxial structures, a sample acquires a certain curvature induced by elastic deformations between layers of the SL due to a difference of their lattice constants. Up to annealing temperatures of 600÷630 °C, the period increases, whereas the curvature radius decreases

T a b l e 2. Dependences of the SL periods, curvature of the system, and variations in the SL period on the annealing temperature for various samples

Sample	$t_{\rm anneal}$, °C	$T_{\rm exp}$, Å	$T_{\rm theor}$, Å	R, m	$(\Delta a/a)\times10^3$ (2)	$(\Delta a/a)\times10^3$ (1)
1	Nonannealed	194.0	–	–	–	7.52±0.04
	570	212.0	209.58	11.95	6.76±0.07	7.27±0.08
	630	211.5	209.38	12.98	6.61±0.02	7.20±0.07
	650	202.0	202.69	13.54	6.21±0.04	7.02±0.07
	700	198.2	197.31	15.67	5.31±0.05	6.33±0.02
2	Nonannealed	188.4	188.29	8.91	10.11±0.02	4.74±0.03
	600	197.9	200.82	7.26	11.82±0.04	4.52±0.04
	750	182.5	–	–	–	4.77±0.04

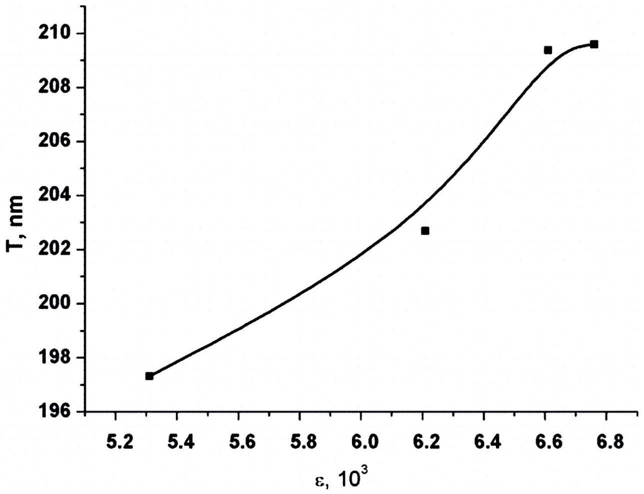

Fig. 3. SL period versus the deformations induced by the structure curvature

(sample *2*). For temperatures of 600÷700 °C, the SL period decreases, and the curvature radius increases. As the annealing temperature increases (above 750 °C), the period tends to that prescribed technologically. At these temperatures, there occurs the full dissolution of quantum dots [3].

In Table 2, we present the theoretically calculated and experimental values of the periods of SLs under study. Their differences do not exceed 2 %. The SL period was calculated with the use of the angle positions of satellites in the spectrum of scattered X-rays with regard for the deformation induced by the crystal curvature as

$$T = \frac{\lambda - 2l_2 K x \varepsilon \sin\theta_{\rm B}}{2\cos\theta_{\rm B}\Delta\theta}, \tag{3}$$

where l_2 is the thickness of an $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layer, x is the concentration of In in the layer, ε is the deformation, $K = (\mathrm{C}_{11} + 2\mathrm{C}_{12})/\mathrm{C}_{11}$ is a factor that takes the tetragonal distortion of the crystal lattice into account; and C_{11} and C_{12} are the elastic constants. In Fig. 3, we display the SL period versus the deformation of the system (the curvature radius). As the initial period, we took a period prescribed technologically for sample *1*.

4. Conclusions

Thus, on the basis of the results presented in this work, we may draw a main conclusion that the period of a superlattice system measured with the use of X-ray diffraction methods depends on the curvature radius of a sample. Therefore, one should take the correction concerning this curvature in the determination of the period of a multilayer structure on the basis of experimental DRCs.

It is clear that the disagreement of the technologically prescribed periods and those obtained experimentally directly after the growing can be explained by just this effect in some cases.

A change of the curvature radius of a multilayer structure in the process of thermoannealing correlates with the level of macrodeformations obtained with the use of various methods.

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ВПЛИВ РАДІУСА КРИВИЗНИ БАГАТОШАРОВИХ СТРУКТУР НА СПЕКТРИ ДИФРАКЦІЇ X-ПРОМЕНІВ

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Р е з ю м е

Методом високороздільної рентгенівської дифрактометрії встановлено ефект зміни величини періоду багатошарової періодичної структури при зміні її радіуса кривизни. Спостережене явище проявляється як зміна положень піків сателітної структури у спектрах дифракції X-променів. Ефект пояснюється впливом зміни деформації вздовж напрямку росту структури, викликаної дифузією компонент в процесі термовідпалу.