Semiconductor Physics, Quantum Electronics & Optoelectronics, 2005. V. 8, N 1. P. 36-45.

PACS: 61.72.Dd, 78.67.Lt

Anisotropy of elastic deformations in multilayer (In,Ga)As/GaAs structures with quantum wires: X-ray diffractometry study

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Abstract. Using the method of high-resolution X-ray diffraction (HRXRD), we have studied 17-period $In_{0.3}Ga_{0.7}As$/GaAs multilayer structure with self-assembled quantum wires (QWRs) grown by the MBE and subjected to postgrowth rapid thermal annealing (RTA) at temperatures (T_{ann}) from 550 to 850 °C for 30 s. It has been shown that the spatial arrangement of QWRs (lateral and vertical) causes the quasi-periodical strain distribution, the strains being essentially anisotropic relatively to crystallographic directions of $\langle 011 \rangle$ type. At $T_{ann} \le 750$ ° , the driving mechanism of structural transformations is relaxation of residual strains due to thermally-activated and strain-enhanced processes of In/Ga atom interdiffusion at the interface QWRs-2D layer, which does not result in considerable changes of the In concentration in (In,Ga)As QWRs. The presence of two superlattice vertical periods in the samples under study and their changes during RTA we explained by an anisotropic character of elastic strain distribution and lowered structure symmetry. The revealed increase in the (In,Ga)As QWRs lateral period caused by RTA is a direct evidence of running lateral mass-transfer processes and can be explained using the model “nucleation plus strain-enhanced In/Ga atom lateral interdiffusion”. At low annealing temperatures, there takes place dissolution of intermediate QWRs as a result of interdiffusion enhanced by residual anisotropic strains. At high RTA temperatures, the interdiffusion process is mainly determined by the composition gradient existing between QWRs and 2D layer.

Keywords: multilayer structures, quantum wires, elastic straines, X-ray diffraction.

Manuscript received 14.01.05; accepted for publication 18.05.05.

1. Introduction

Unique electrophysical and optical properties of semiconductor nanostructures with quantum dots (QDs) and quantum wires (QWRs) obtained in Stranski – Krastanow mode using epitaxy of strained heterosystems open new perspectives to create modern devices for nano- and optoelectronics, for instance, laser diodes with a low threshold current and power consumption [1]. From the viewpoint of practical applying of these nanostructures, it is very important to reduce QDs sizes, to increase the density of their surface distribution and the ordering. Multilayer structures with spatially ordered QDs and QWRs are characterized by high homogeneity of their sizes, shapes as well as distances between QDs and QWRs. As it was recently shown, growing the (In,Ga)As/GaAs multilayer structures provides obtaining of laterally ordered chains of (In,Ga)As QDs with the length up to 5 μm [2] and (In,Ga)As QWRs [3, 4]. The process of thermal annealing enables one to improve homogeneity of QDs and QWRs size distribution. For example, as a result of the thermal annealing of single-layer (In,Ga)As/GaAs QDs [5] there was improved QDs size homogeneity and was shown the possibility of gradual changing the emission wave length. It was found that strain-enhanced atom interdiffusion [6] plays an important role in the process of InAs QDs annealing. However, up to date, the mechanism of In/Ga interdiffusion and structural transformations in (In,Ga)As QDs and, moreover, in structures with (In,Ga)As QWRs has not been ascertained yet. The investigation of structural and optical properties of spatially ordered multilayer structures as well as their changes in the course of the thermal annealing is of great interest to understand mechanisms of their epitaxial growth and formation, to study the features of elastic strain relaxation and generation of structural defects.

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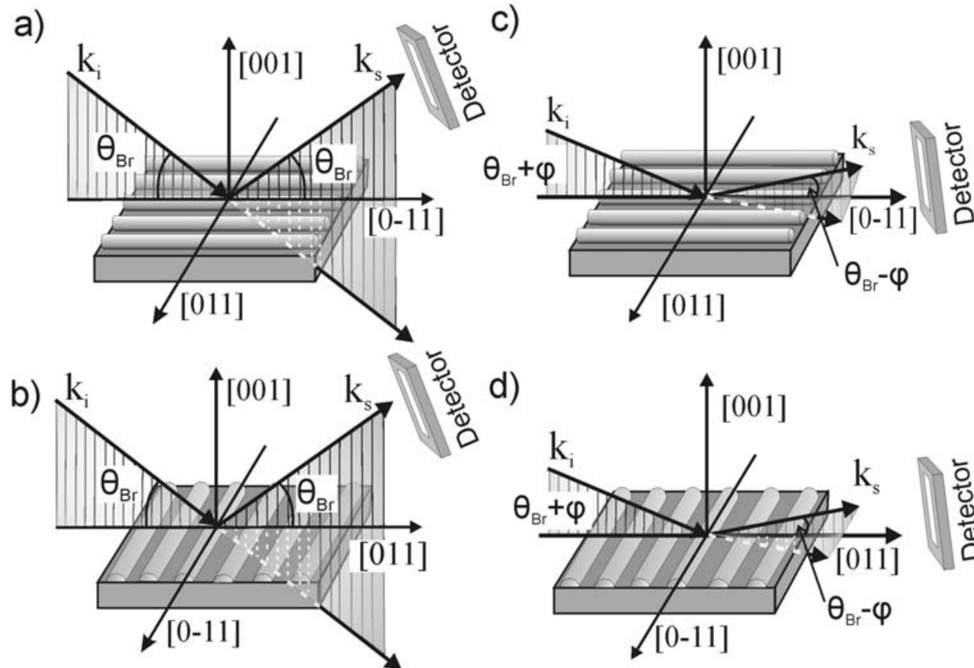

Fig. 1. Schematic representation of experimental geometry for studying quantum wires: a, b – for 004 symmetrical reflection; c, d – 311 asymmetrical reflection, where φ is the angle between the reflection plane and surface.

It is known that 2D-3D transition and the process of three-dimensional QDs (or QWRs) formation in the course of epitaxial deposition of InGaAs/GaAs layers essentially changes the character of the elastic strain distribution as compared with that in planar layers. In multilayer structures, relaxation of elastic strains induced by QDs or QWRs [7] is realized through their spatial arrangement. In this case, there takes place an anisotropic character of the strain distribution relatively to crystallographic directions of ⟨011⟩ type [8], which determines an anisotropy of structural parameters (for example, lowering the structural symmetry [9]) and was observed in planar heterostructures [10, 11]. This structural anisotropy influences on X-ray diffraction, which allows to use high-resolution X-ray diffractometry in investigations of elastic strain anisotropy in nanostructures with QDs and QWRs.

In this work, adduced are the results of structural investigations of multilayer (In,Ga)As/GaAs (100) structures with self-assembled (In,Ga)As QWRs after RTA by HRXRD. The obtained results allows to ascertain regularities of structural changes in the course of the thermal annealing as well as to study peculiarities of elastic strain relaxation in the spatially ordered (In,Ga)As QWRs, which is of great importance for understanding physical properties of quantum-sized objects.

2. Sample

The samples used in this investigation were grown on semi-insulating GaAs (100) substrates with a miscut smaller than 0.05° by conventional solid-source molecular beam epitaxy (MBE). After the native oxide was desorbed from the GaAs substrate surface at 650 °C for 10 min, the GaAs buffer layer of 500 nm thickness was grown at T = 600 °C and the growth rate about one monolayer (ML) per second. After that, the substrate temperature was reduced down 540 ° , and 17 periods of (11.5ML)$In_{0.3}Ga_{0.7}As$/(67ML)GaAs were deposited. The growth rates of GaAs and $In_{0.3}Ga_{0.7}As$ were equal to 0.4 and 0.8 ML/s, respectively. As_4 beam equivalent pressure was 10 μTorr for the whole structure growth. The growth interruptions were introduced during the deposition of the GaAs spacer [12].

Rapid thermal annealing was performed in argon atmosphere at temperatures 550 to 850 °C for 30 s. The time of setting the operation temperature was 10 s.

The structural morphology of the sample surface was investigated using atomic force microscopy (AFM) in the tapping mode.

3. Experimental details

The X-ray scattering experiments were performed using double-crystal diffractometer (GaAs(100) crystal-monochromator, CuK radiation, the fourth reflection order) for symmetrical 400 and asymmetrical 311 reflections. The sample was scanned in the vicinity of the exact Bragg position close to 3° using the so-called /2θ-mode. The measurements were performed in the discrete angular mode with the step 2″. The registration was realized in the accumulation regime for the pulse quantity measured at the signal-to-noise ratio 10^{-4}. In

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[19]. In essence, the diffusion may provide a means of relaxing strain.

Structure parameters obtained using the experimental data of HRXRD and the method of numeric modelling the experimental rocking curves.

Sample T_{ann}, °	GaAs 004 reflection							GaAs 311 reflection	
	d_{GaAs}, nm	d_{InGaAs}, nm	$\Lambda^{\perp}$, [011], nm	$\Lambda^{\perp}$, [011], nm	av	$\varepsilon_{\perp}$ (GaAs)	$\varepsilon_{\perp}$ (InGaAs)	Λ , nm	ε (InGaAs)
Initial	19.7	3.09	22.79	22.05	0.27	0.00245	–0.02	83.9	0.0047
550	18.4	3.24	21.64	21.17	0.30	0.0025	–0.022	97.8	0.0052
600	19.7	3.27	22.97	22.93	0.30	0.0025	–0.02	106.5	0.0056
650	18.3	1.46	19.76	20.7	0.30	0.0025	–0.022	-	
750	18.2	1.3	19.5	20.36	0.30	0.0025	–0.021	-	
850	18.4	3.3	21.7	22.5				-	

Thus, we can assume that there is re-distribution (relaxation) of strains in the regions close to the interface QWR – 2D layer. This relaxation of residual strains, as a consequence of thermally-activated and strain-enhanced processes of In/Ga atomic interdiffusion, is a driven mechanism of structural transformations caused by RTA of (In,Ga)As QWRs.

The samples under investigation showed good coincidence between calculated and experimental data at the level 2 – 5 % both for the position and intensities over the whole RC. Analyzing the experimental data obtained, we determined main structural parameters of (In,Ga)As/GaAs structure.

The multilayer structure period, Λ, was determined through angular positions of any two superlattice satellites by using the relation:

$$\Lambda = \frac{(m-n)\lambda/2}{\sin\theta_m - \sin\theta_n}. \qquad (2)$$

where λ is the wavelength of X-ray radiation $CuK_{\alpha 1}$ (λ = 1.54051 Å), $m(n)$ are the orders of X-ray wave λ reflections, $\theta_m(\theta_n)$ are the reflection angles for these orders.

Stemming from the angular position of zero satellite, θ_0, the average content of InAs, x_{av}, in the period of the multilayer structure can be determined using the following relation:

$$x_{av} = \frac{a_{GaAs}}{a_{InAs} - a_{GaAs}} \frac{1-\nu}{1+\nu} \cot\theta_{Br} \left(\theta_{Br} - \theta_0\right), \qquad (3)$$

where $a_{GaAs} = 5.63375$ ($a_{InAs} = 6.0584$) is the lattice parameter of GaAs (InAs), $\nu = 0.333$ is the Poisson coefficient, $\theta_{Br} = 33.02$ is the Bragg angle inherent to the GaAs substrate. Having determined the change of zero satellite angular position ($\Delta\theta_0$) relatively to the GaAs peak from experimental reflection spectra and using the relation [20]:

$$\varepsilon_{\perp} = [\sin\theta_{Br} / \sin(\theta_{Br} - \Delta\theta_0)] - 1 \qquad (4)$$

we estimated the strain mean value ($\varepsilon_{\perp}$) inside the layers along the direction of the structure growth. The obtained data are summarized in Table.

Note: d_{GaAs} (d_{InGaAs}) is the layer thickness for GaAs (InGaAs), $\Lambda^{\perp}$ (Λ) is the period of the structure along the growth direction (lateral), av is the average In concentration determined from zero satellite position, $\varepsilon_{\perp}$(GaAs) ($\varepsilon_{\perp}$(InGaAs)) is the strain along the structure growth direction for GaAs (InGaAs) layer, ε (InGaAs) is the strain in the layer plane.

Shown in Fig. 5 are diffraction RCs of 004 reflection for the initial (a) and annealled at ann = 750 ° (b) samples obtained in two mutually orthogonal azimuthal directions of incident radiation: parallel (a) and perpendicular (b) relatively to orientations of (In,Ga)As QWRs. It is easy to seen that asymmetric broadening the diffraction peaks observed in the initial sample (Fig. 5a) is increased after RTA (Fig. 5b) and especially clear pronounced with growing the reflection order. As mentioned above, the main reasons for broadening these peaks can be: planar roughness of interfaces, In gradient distribution along the growth direction, mosaic character of the film structure and presence of two vertical periods in the multilayer structure. Roughness of interfaces is not a dominant effect, as we observed experimentally the change in peak angular positions [21], but this effect contribution into broadening the reflection peaks grows in the samples treated by RTA (Fig. 5b). Performed modelling the RCs with taking into account the In concentration gradient along the structure growth direction, as it was made in [22], showed that the thickness of layers containing In is too small to reach respective asymmetry of experimental reflection peaks. We suppose that the main and dominant effect providing this asymmetrical broadening the peaks is the dependence of peak angular positions on the

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QWRs lateral periodicity. The latter results in decreasing the diffracted scattering intensity to a level lower than the sensitivity of our experimental facility. Also interesting is the fact that, with increasing the RTA temperature, the lateral period grows up to 106.5 nm ($_{ann} = 600$ °C). It is a direct evidence of running thermally-stimulated processes of the lateral mass transfer in the form of In/Ga atomic interdiffusion.

5. Conclusions

HRXRD data obtained for symmetrical and asymmetrical reflections in two mutually orthogonal directions of incident radiation (parallel and perpendicular to QWRs orientation direction) enabled to reveal anisotropy of elastic strain distribution in QWRs structure. It has been shown that, at the temperatures $_{ann} \leq 750$ ° , the driven mechanism of structural transformations is relaxation of residual strains as a consequence of thermally-activated and strain-enhanced processes of In/Ga atomic interdiffusion at the interface QWR – 2D layer, which does not result in a considerable change of In concentration in (In,Ga)As QWRs. The presence of two superlattice vertical periods and their changes after RTA in the samples under study are explained using the data about elastic strain anisotropic distribution and reduction in the structure symmetry. Also shown is that the increase in the lateral period of (In,Ga)As QWRs after RTA is caused by the processes of anisotropic strain-enhanced In/Ga atomic interdiffusion.

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