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Dynamical X-Ray Diffraction Characterization of the Self-Organized Quantum Dot Formation In Imperfect Semiconductor Superlattices

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The self-organized quantum dot (QD) formation in InGaAs/GaAs superlattices grown by molecular beam epitaxy was investigated by the high-resolution X-ray diffraction technique. The investigated samples had the identical structure consisting of fifteen periods of {$In_xGa_{1-x}As$ (8 ML)/GaAs (26 ML)} with the nominal In concentration $x = 0.2$. The diffraction profiles and reciprocal lattice maps for these samples have been measured at symmetrical (004) reflection by using the triple-crystal X-ray diffractometer. The analysis of the measured data was performed by using the proposed diffraction model based on the statistical theory of dynamical X-ray scattering in imperfect single crystals and multilayer structures.

Keywords: quantum dots, superlattice, X-ray diffraction, diffuse-dynamical diffractometry.

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1. INTRODUCTION

Self-organized quantum dots (QDs) in superlattices grown by molecular beam epitaxy are of great interest due to their potential applications in modern optoelectronic devices, such as lasers, optical memories, infrared photodetectors, etc. [1, 2]. However, because of insufficiently good understanding the complicated self-organization process of nanosize islands formation in thin films, particularly, in the InGaAs/GaAs superlattice system, the production of QD-based devices has the unreliable yield.

The present work is devoted to the development of X-ray dynamical diffraction technique for reliable monitoring QD structures and, particularly, for its using in the investigation of InGaAs/GaAs superlattice systems.

2. THEORETICAL MODEL

2.1 Coherent scattering

According to results of the generalized dynamical theory of X-ray diffraction by imperfect single crystals [3–5], the coherent wave field in a crystal with homogeneously distributed defects can be represented in the two-wave approximation as the sum of transmitted and diffracted waves for each polarization state (σ and π):

$$D(\mathbf{r}) = D_{\mathrm{T}}(\mathbf{r}) + D_{\mathrm{S}}(\mathbf{r}),$$

$$D_{\mathrm{T}}(\mathbf{r}) = \sum_{\delta} D_0^{\delta}\, \mathrm{e}^{-i\mathbf{K}_0^{\delta}\mathbf{r}}, \quad D_{\mathrm{S}}(\mathbf{r}) = \sum_{\delta} D_{\mathbf{H}}^{\delta}\, \mathrm{e}^{-i\mathbf{K}_{\mathbf{H}}^{\delta}\mathbf{r}}, \qquad (1)$$

where $\delta = 1,2$, $\mathbf{r}$ is the space coordinate, D_0^{δ} and $D_{\mathbf{H}}^{\delta}$ are the amplitudes of transmitted and diffracted coherent plane waves, respectively, $\mathbf{H}$ is the reciprocal lattice vector, $K = 2\pi/\lambda$ is the module of the wave vector $\mathbf{K}$ of an incident plane wave, and λ is the wavelength.

To determine the amplitude reflectivity of the total multilayered system

$$R = b^{-1/2}\, E_{\mathbf{H}}/E_0 ,$$

where $b = \gamma_0/|\gamma_{\mathbf{H}}|$ is the parameter of diffraction asymmetry, γ_0 and $\gamma_{\mathbf{H}}$ are direction cosines of wave vectors of incident ($\mathbf{K}$) and scattered ($\mathbf{K}'$) plane waves, respectively; E_0 and $E_{\mathbf{H}}$ are amplitudes of the incident and coherently scattered plane waves in a vacuum, respectively, it is necessary to find amplitudes D_{0M}^{δ} and $D_{\mathbf{H}M}^{\delta}$ of a wave field in M th layer, which are connected as well to the amplitude of the wave reflected in ($M - 1$) th layer, etc.

The recurrence relation between amplitude reflection coefficients of adjacent layers can be determined as follows:

$$R_j = \frac{r_j + R_{j-1}(t_j^2 - \varsigma_j r_j^2)}{1 - \varsigma_j r_j R_{j-1}}, \qquad (2)$$

$$\zeta = \left(CE\chi_{\mathbf{H}} + \Delta\chi_{\mathbf{H}0}\right)\left(CE\chi_{-\mathbf{H}} + \Delta\chi_{0\mathbf{H}}\right)^{-1},$$

where r_j and t_j are amplitude reflectivity and transmissivity of j th layer, respectively, C is the polarization factor equal to 1 or $\cos 2\theta_{\mathrm{B}}$ for σ and π polarization, respectively, $\chi_{\mathbf{G}}$ are Fourier components of the perfect crystal polarizability ($\mathbf{G} = 0, \pm\mathbf{H}$).

2.2 Diffuse scattering

According to [3], the diffuse scattering (DS) amplitude from j th layer can be represented as

$$f_{\mathbf{H}j}\left(\mathbf{K}'_j, \mathbf{K}_j\right) = \sum_{\delta}\sum_{\mathbf{G}} D_{\mathbf{G}j}^{\delta} F_{\mathbf{HG}j}^{\delta} e^{-\gamma_j}, \qquad (3)$$

$$\gamma_j = \sum_{i=j+1}^{M} \mu_i d_i / |\gamma_{\mathbf{H}}|, \quad \mu_i = \mu_{0i} + \mu_{\mathrm{ds}}^{i}, \quad \delta = 1,2 .$$

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Partial amplitudes of scattering of strong Bragg waves into diffuse ones in the diffraction direction $\mathbf{K}+\mathbf{H}$ in Eq. (3) are described by:

$$F_{\mathbf{HG}}^{\delta}\left(\mathbf{K}',\mathbf{K}\right)=\frac{VK^2}{4\pi\left(B_1'-B_2'\right)}\sum_{\tau}\left(-1\right)^{\tau}M_{\mathbf{HG}}^{\delta\tau}\mathrm{e}^{-iK\Delta_{\tau}'t},$$

$$M_{\mathbf{HG}}^{\delta\tau}=c'^{(\delta)}M_{0\mathbf{G}}^{\delta\tau},$$

$$M_{0\mathbf{G}}^{\delta\tau}=\xi'\mathrm{X}_{0\mathbf{G}}\left(\mathbf{q}_{\delta\tau}\right)/c'^{(\tau)}+\mathrm{X}_{\mathbf{HG}}\left(\mathbf{q}_{\delta\tau}\right),$$

$$\mathrm{X}_{\mathbf{GG}'}\left(\mathbf{q}\right)=C_{\mathbf{GG}'}\delta\chi_{\mathbf{G}-\mathbf{G}'+\mathbf{q}},\quad B_{\tau}'=c'^{(\tau)}\mathrm{e}^{-iK\Delta_{\tau}'t}.$$

Here, $C_{\mathbf{GG}'}=1$ at $\mathbf{G}'=\mathbf{G}$, and $C_{\mathbf{GG}'}=C$ at $\mathbf{G}'\neq\mathbf{G}$, the quantities Δ_{τ}' are accommodations of wave vectors of diffusely scattered waves, μ_{ds}^{i} is the absorption coefficient due to DS from defects in i th type.

The diffuse component of differential reflectivity of multilayered system, which is measured by triple-crystal diffractometer (TCD), can be represented as

$$r_{\mathrm{diff}}(\boldsymbol{\kappa})=\sum_{j=0}^{M}F_{\mathrm{ext}}^{j}F_{\mathrm{abs}}^{j}r_{\mathrm{diff}}^{j}(\boldsymbol{\kappa}), \tag{4}$$

where F_{ext}^{j} and F_{abs}^{j} are extinction and absorption factors, respectively, and two-dimensional vector $\boldsymbol{\kappa}$ is the momentum transfer in scattering plane. The function $r_{\mathrm{diff}}^{j}(\boldsymbol{\kappa})$ represents the diffuse component of the differential reflection coefficient of j th layer integrated over a vertical divergence:

$$r_{\mathrm{diff}}^{j}(\boldsymbol{\kappa})=\frac{1}{K}\int dk_y R_{\mathrm{D}j}\left(\mathbf{k}\right) \tag{5}$$

$$R_{\mathrm{D}j}(\mathbf{k})=\left\langle\left|f_{\mathbf{H}j}\right|^2\right\rangle_j\Big/\left(\gamma_0 S\left|E_0\right|^2\right) \tag{6}$$

Here $\langle\ldots\rangle_j$ means averaging over chaotic distribution of defects in j th layer, and S is the entrance surface area of the crystal.

3. EXPERIMENT

The investigated samples had the same structure consisting of fifteen periods of {$In_xGa_{1-x}As$ (8 ML)/GaAs (26 ML)} with the nominal In concentration $x = 0.2$. The diffraction profiles and reciprocal lattice maps for these samples have been measured at symmetrical (004) reflection of Cu$K_{\alpha1}$ radiation by using the high-resolution X-ray diffractometer PANalytical X`Pert Pro MRD XL (Fig.1 and 2).

4. RESULTS AND DISCUSSION

The quantitative analysis and reliable interpretation of differential reflectivity from multilayered structures require using both adequate theoretical model of diffraction and realistic model of structural imperfections of these structures. The model of defect structure, which assumed the presence of randomly distributed microdefects in multilayered system, was created and applied to account for structural imperfections in multilayered system.

The analysis of the measured X-ray data within the framework of proposed diffraction model based on the statistical theory of dynamical X-ray scattering in imperfect single crystals and multilayer structures [3–5] has allowed to determine parameters of strain profiles and characteristics of Coulomb-type defects in the samples. The differences in both non-uniform In concentration profiles in the $In_xGa_{1-x}As$ layers (corresponding to self-organized QDs) and defect structures in samples have been determined for various growth temperatures.

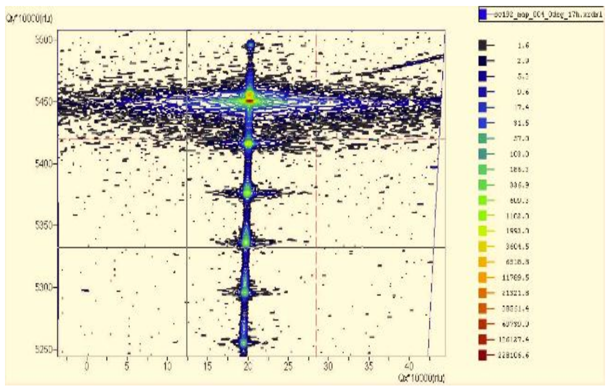


Fig. 1 – Measured reciprocal lattice map around GaAs 004 reflection for InGa/GaAs multilayer structure grown at substrate temperature of 510°C

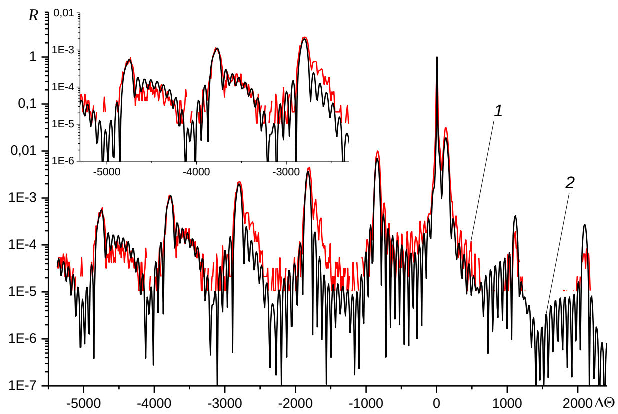
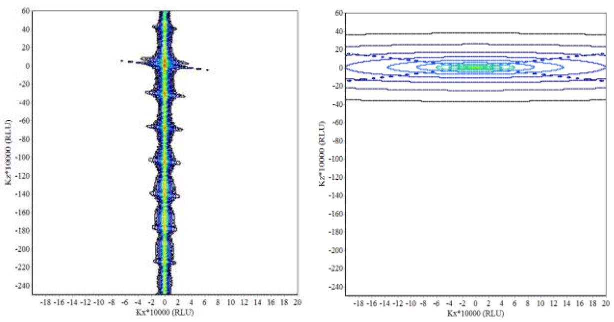


Fig. 2 – Measured (*1*) and calculated (*2*) profile for GaAs 004 reflection for $In_xGa_{1-x}As$/GaAs multilayer structure grown at substrate temperature of 510°C

Fitting the experimental diffraction profiles for GaAs 004 reflection for $In_xGa_{1-x}As$/GaAs multilayer structures grown at various temperatures (see Fig.2 for substrate temperature 510°C) and analysis of experimental and calculated reciprocal space maps has allowed to find more precisely thicknesses of layers and their chemical compositions, namely, fifteen periods of {$In_xGa_{1-x}As$ (8 ML)/GaAs (26 ML)} with $x = 0.195$.

Thus, analysis of diffraction profiles and reciprocal lattice maps from multilayered structure with InGaAs QW, which has been carried out on the basis of the developed model of dynamical X-ray diffraction by multilayered structures with defects, has allowed for obtaining much more information about crystal and defect structure of this system as compared with results of classical diffraction models. Substantially new diagnostic opportunities have appeared due to the introduction of DS effects from defects in the diffraction model, what has allowed to calculate correctly the diffuse component of measured diffraction profiles and reciprocal lattice maps and to find corresponding characteristics of defects.



a *b*

Fig. 3 – Calculated coherent (*a*) and diffuse (*b*) components of reciprocal lattice map for $In_xGa_{1-x}As/GaAs$ multilayer structure grown at substrate temperature of 510°C

5. RESUME AND CONCLUSION

The dynamical theory of X-ray scattering by multilayered structures with homogeneously distributed defects in each layer has been developed. The dynamical recurrence relation for amplitudes of coherent waves in such structures has been deduced. Expressions for the diffuse component of diffraction profiles and reciprocal lattice maps have been obtained with account for the redistribution of intensities of transmitted and diffracted coherent waves in each layer.

On the basis of the derived formulas, for the first time the full and adequate analysis of diffraction profiles and reciprocal lattice maps from multilayered structure with InGaAs QDs has been performed. As result of fitting the experimental diffraction profiles and reciprocal lattice maps, thicknesses of layers, and their chemical compositions have been found more precisely. Additionally, characteristics of microdefects have been determined.

The developed theoretical model of diffraction in multilayered structures with defects and the corresponding procedure of the self-consistent fitting the measured diffraction profiles and reciprocal lattice maps have created a basis for qualitatively new method of diffractometric diagnostics of structural characteristics of multilayered crystal systems with defects.

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