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A dynamical theory for the X-ray diffraction from the partially relaxed layers

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The dynamical theory for X-ray diffraction from the bilayer crystal structure with different lateral periods of the crystal unit cells (lateral mismatch) is considered in the present paper. The amplitudes of the principal diffraction waves and all harmonics conditioned by the lateral mismatch are calculated. The formation of the Bragg peaks is analysed taking into account the sphericity of the incident beam wave front set. The connection between the parameters of the coherent diffraction potential in the partially relaxed (epitaxial) crystals and the microscopic characteristics of the dislocations is also discussed.

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1 Introduction It is well known that the standard boundary conditions of the dynamical diffraction theory [1] in the framework of the two wave approximations are unapplicable when the interface between the crystal films with lateral mismatch of the unit cells is considered. This problem arises when simulation of the X-ray diffraction profiles from the epitaxial multilayered structures both on the basis of Takagi–Taupin equations [2] or the recurrent matrix methods is held [3].

Let us consider, for example, the boundary between two crystals with the tetragonal unit cells in case of the coplanar diffraction, z-axis is directed along the normal $\boldsymbol{N}$, which lies in the same plane as the wave vector $\boldsymbol{k}_0$ of the incident beam, the reciprocal lattice vectors $\boldsymbol{h}_\mathrm{S}$ of the substrate and $\boldsymbol{h}_\mathrm{L}$ of the epitaxial layer. It is supposed that the unit cell parameters a_L, c_L for the layer and a_S, c_S for the substrate do not coincide and define the lateral $\xi_\parallel$ and normal $\xi_\perp$ mismatches

$$\xi_\parallel{}^{\mathcal{R}} = \frac{a_\mathrm{L}^{\mathcal{R}} - a_\mathrm{S}}{a_\mathrm{S}}; \qquad \xi_\perp{}^{\mathcal{R}} = \frac{c_\mathrm{L}^{\mathcal{R}} - c_\mathrm{S}}{c_\mathrm{S}}. \tag{1}$$

Here the values $a_\mathrm{L}^{\mathcal{R}}$, $c_\mathrm{L}^{\mathcal{R}}$ are referred to the *relaxed* layer corresponding to the crystal in the free state. The unit cell of the epitaxial layer is deformed because of the interaction with the substrate and the only parameter (relaxation $\mathcal{R}$) can be effectively used for simulation of the diffraction profiles for all intermediate cases [4, 5]

$$\mathcal{R} = \frac{a_\mathrm{L} - a_\mathrm{S}}{a_\mathrm{L}^{\mathcal{R}} - a_\mathrm{S}}; \qquad \xi_\parallel = \frac{a_\mathrm{L} - a_\mathrm{S}}{a_\mathrm{S}}; \qquad \xi_\perp = \frac{c_\mathrm{L} - c_\mathrm{S}}{c_\mathrm{S}}. \tag{2}$$

The relaxation parameter changes between the value $\mathcal{R} = 1$ (a fully relaxed layer) and $\mathcal{R} = 0$ (a fully strained or 'pseudomorphic' layer). The values $\xi_\parallel$ and $\xi_\perp$ are connected because of the Poisson ratio ν (e.g. for the cubic unit cell [6])

$$\frac{c_\mathrm{L} - c_\mathrm{L}^{\mathcal{R}}}{c_\mathrm{L}^{\mathcal{R}}} = -\frac{2\nu}{1-\nu}\,\frac{a_\mathrm{L} - a_\mathrm{L}^{\mathcal{R}}}{a_\mathrm{L}^{\mathcal{R}}}. \tag{3}$$

In the case of the partially relaxed layer ($0<\mathcal{R}<1$) $a_\mathrm{L} \neq a_\mathrm{S}$; $\xi_\parallel \neq 0$ the lateral components of the vectors $k_{x\mathrm{L}} = k_{0x} + h_{x\mathrm{L}}$ and $k_{x\mathrm{S}} = k_{0x} + h_{x\mathrm{S}}$ for the waves diffracted from the substrate and the corresponding layer are not equal. Therefore one should take into account the additional harmonics $\sim \exp[\mathrm{i}\, l(h_{\mathrm{S}x} - h_{\mathrm{L}x})x]$, $l = 0, \pm 1, \pm 2, \ldots$ in order to provide the wave field continuity at the boundary between the layer and the substrate (Fig. 1). The amplitudes of these harmonics were calculated numerically in the paper [7] for the case $\xi_\perp = 0$ which is not realized in real structures

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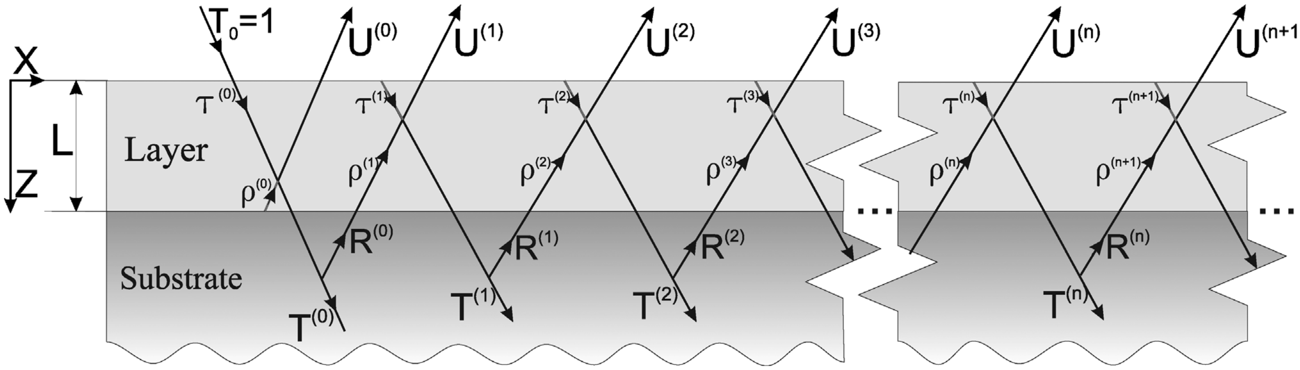

Figure 1 Sketch of the additional harmonics formation for diffraction from the partially relaxed layer.

because of the condition (3). The general solution of this problem is considered analytically in the present paper (Section 2). It is also shown that the sphericity of the incident beam wave front set and the relaxation transition layer near the interface should be taken into account when mapping the diffracted profile in a reciprocal space.

In spite of the characterization of the epitaxial layer by means of the phenomenological parameter $\mathcal{R}$ it is rather useful for applications in high-resolution diffraction to express this value through the microscopic parameters of the dislocations that actually define the real deformation of the crystal structure in such films [2]. The kinematical theory of the diffraction from the non-ideal crystals was developed in detail by Krivoglaz [8] taking into account the statistical distribution of the dislocations. The conditions for the formation of the coherent diffraction peak were described. These results were essentially advanced recently for the epitaxial films [9, 10]. In the present paper (Section 3) analogous ideas are used in order to estimate the value $\mathcal{R}$ in terms of the microscopic deformation parameters.

2 Solution of the boundary problem Let us consider the coplanar Bragg diffraction from the bilayer structure consisting of the substrate and the layer with the unit cell parameters a_S, c_S and a_L, c_L correspondingly. It is supposed that the incident plane wave $T_0\,\mathrm{e}^{ik_0r}$ excites the reflections $\boldsymbol{h}_\mathrm{S}$ and $\boldsymbol{h}_\mathrm{L}$ in the substrate and the layer

$$2(\boldsymbol{k}_0\boldsymbol{h}_\mathrm{L}) + h_\mathrm{L}^2 = 2(\boldsymbol{k}_0\boldsymbol{h}_\mathrm{S}) + h_\mathrm{S}^2 = 0. \tag{4}$$

In the framework of the two wave approximations of the dynamical diffraction theory the transmitted waves $\tau_{1,2}^{(0)}$ and $T_{1,2}^{(0)}$ as well as the diffracted waves $\rho_{1,2}^{(0)}$ and $R_{1,2}^{(0)}$ should be taken into account in the layer and the substrate. Lateral and normal components of the substrate and the layer wave vectors $\boldsymbol{k}_i^{\mathrm{S,L}}$ can be written as $k_{ix}^{\mathrm{S,L}} = k_{0x} \equiv k_x$; $k_{iz}^{\mathrm{S,L}} \equiv k_0 u_i^{\mathrm{S,L}}$ [3], dimensionless values $u_i^{\mathrm{S,L}}$ being the roots of the dispersion equation

$$\left[\left(u_i^{(\mathrm{S,L})} + \psi^{(\mathrm{S,L})}\right)^2 + \left(\frac{k_x + h_x^{(\mathrm{S,L})}}{k_0}\right)^2 - \left(1 + \chi_0^{(\mathrm{S,L})}\right)\right]$$
$$\times \left[u_i^{(\mathrm{S,L})^2} + \left(\frac{k_x}{k_0}\right)^2 - \left(1 + \chi_0^{\mathrm{S,L}}\right)\right] = C\chi_{\boldsymbol{h}}^{(\mathrm{S,L})}\chi_{-\boldsymbol{h}}^{(\mathrm{S,L})}, \tag{5}$$

here $\chi_0^{(\mathrm{S,L})}$, $\chi_{\boldsymbol{h}}^{(\mathrm{S,L})}$ are the components of the X-ray polarizability; C is a polarization factor [1]; the parameter $\psi^{(\mathrm{S,L})} = h_z^{(\mathrm{S,L})}/k_0$. The amplitudes of the transmitted and the diffracted waves are connected as follows:

$$\rho_i^{(0)} = v_i^{(\mathrm{L})}\tau_i^{(0)}; \qquad R^{(0)} = v^{(\mathrm{S})}T^{(0)};$$
$$v_i^{(\mathrm{S,L})} = \frac{\chi_{\boldsymbol{h}}^{(\mathrm{S,L})}}{\left(u_i^{(\mathrm{S,L})} + \psi^{(\mathrm{S,L})}\right)^2 + \left(\frac{k_x + h_x^{(\mathrm{S,L})}}{k_0}\right)^2 - \left(1 + \chi_0^{(\mathrm{S,L})}\right)}. \tag{6}$$

The only solution of Eq. (5) with Re $u^{(\mathrm{S})}>0$; Im $u^{(\mathrm{S})}>0$ should be taken into account when the wave fields in the substrate are considered [3]. Other wave field harmonics are excited in the considered approximation of dynamical diffraction theory in accordance with the iteration sequence shown in Fig. 1. For example, the diffracted wave $R^{(\mathrm{S})}$ going out of the substrate excites in the layer the field $\rho^{(1)}$ with the wave vector $\boldsymbol{k}^\mathrm{S} + \boldsymbol{h}^\mathrm{S} \approx \boldsymbol{k}_0 + \boldsymbol{h}^\mathrm{S}$. It satisfies the Bragg condition with the reciprocal lattice vector $(-\boldsymbol{h}^\mathrm{L})$ and its diffraction leads to the field with the amplitude $\tau^{(1)}$ and the wave vector $\boldsymbol{k}^{(1)} \approx \boldsymbol{k}_0 + \boldsymbol{h}^\mathrm{S} - \boldsymbol{h}^\mathrm{L}$ and so on. So, the set of the wave field harmonics arises and the wave vectors and the amplitudes of the mth harmonics satisfy the equations analogous to Eqs. (5) and (6) with the substitution $k_x + m(h_x^\mathrm{S} - h_x^\mathrm{L})$ instead of k_x.

The recurrent equations for the amplitudes of neighbouring harmonics can be found using the boundary conditions and the conservation of the wave vectors lateral components

$$\tau_1^{(n)} + \tau_2^{(n)} = 0, \quad v_1^{(n)}\tau_1^{(n)} + v_2^{(n)}\tau_2^{(n)} = U^{(n)},$$
$$\tau_1^{(n)}\,\mathrm{e}^{iu_1^{(n)}L} + \tau_2^{(n)}\,\mathrm{e}^{iu_2^{(n)}L} = T^{(n)}, \tag{7}$$

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where L is the layer thickness; $U^{(m)}$ is the amplitude of the field which appears in the vacuum due to the mth harmonic. Equation (7) leads to the ratio $\kappa^{(n)} \equiv U^{(n+1)}/U^{(n)}$, and the amplitude $U^{(m)}$ can be found as

$$U^{(n)} = U^{(1)} \prod_{i=1}^{n-1} \kappa^{(i)},$$

$$\kappa^{(n)} = v^{(n)} \frac{v_1^{(n+1)} - v_2^{(n+1)}}{v_1^{(n)} - v_2^{(n)}} \frac{e^{iu_1^{(n)} L} - e^{iu_2^{(n)} L}}{v_1^{(n+1)} e^{iu_1^{(n+1)} L} - v_2^{(n+1)} e^{iu_2^{(n+1)} L}}. \quad (8)$$

The principal amplitudes $U^{(0)}$, $U^{(1)}$ can be found from the boundary conditions (the amplitude of the incident wave $T_0 = 1$)

$$\begin{aligned} &\tau_1^{(L)} + \tau_2^{(L)} = 1, \quad v_1^{(0)} \tau_1^{(L)} + v_2^{(0)} \tau_2^{(L)} = U^{(0)}, \\ &\tau_1^{(L)} e^{iu_1^{(L)} L} + \tau_2^{(L)} e^{iu_2^{(L)} L} = T^{(0)}, \\ &v_1^{(L)} \tau_1^{(L)} e^{iu_1^{(L)} L} + v_2^{(L)} \tau_2^{(L)} e^{iu_2^{(L)} L} = 0, \quad (9) \\ &v_1^{(1)} \tau_1^{(1)} e^{iu_1^{(1)} L} + v_2^{(1)} \tau_2^{(1)} e^{iu_2^{(1)} L} = v^{(0)} T^{(S)}, \\ &\tau_1^{(1)} + \tau_2^{(1)} = 0, \quad v_1^{(1)} \tau_1^{(1)} + v_2^{(1)} \tau_2^{(1)} = U^{(1)}. \end{aligned}$$

The expressions (7)–(9) define completely all amplitudes. As distinct from the result of the paper [7] these solutions satisfy correctly the limit cases $\xi_\parallel = 1$ (when the diffraction waves from the layer and the substrate are independent) and $\xi_\parallel = 0$ (when the standard form of the dynamical diffraction theory is applicable). Figure 2 shows the simulated results for the reflection (224) with the Cu Kα X-rays diffracted by the sample consisting of $Si_{0.82}Ge_{0.18}$ layer with $L = 100$ nm and the Si substrate and for the various relaxation parameters. The profiles $I^{(n)}(q_z) = |U^{(n)}/T_0|^2$ for several harmonics are shown in the logarithmic scale. Actually the interference between the amplitudes $U^{(0)}$ (diffracted from the layer) and $U^{(1)}$ (from the substrate) is essential only for the pseudomorphic case ($\mathcal{R} = 0$). For rather small but non-zero relaxation ($\mathcal{R} = 0.1$ in Fig. 2) the diffraction from the layer and the substrate can be considered independently and the amplitudes of the higher harmonics do not affect the total intensity profile.

The results above show that the diffraction profiles for the partially relaxed structures can be calculated independently for each layer if the incident wave is considered as the monochromatic one and the interface between a layer and substrate is described by the ideal plane $z = z_0 =$ const. In these approximations the reciprocal space maps (RSM) from the substrate and the layer are proportional to the δ-functions corresponding to the different values $q_x = h_{xL}$ and $q_x = h_{xS}$ and can be represented as the thin lines. In order to simulate RSM the strain transition layer and the sphericity of the incident wave is taken into account in the present paper. The variation of the layer lattice parameter $a_L(z)$ was considered as the result of the relaxation parameter alteration $\mathcal{R}(z)$ in accordance with the model $\mathcal{R}(z) = \mathcal{R}_0 \tanh[(L - z)/a_d]$, with $\mathcal{R}_0$ as the relaxation parameter in the point being far from the interface $z = L$, and a_d as the width of the transition layer.

Another reason for spreading of RSM from the layer and substrate in the q_x direction is conditioned by the limit value of the coherent length L_{coh} of the incident beam connected with the dispersion of the lateral component $\Delta k_x \approx L_{coh}^{-1}$. It was shown that the main limitation for L_{coh} was defined by the

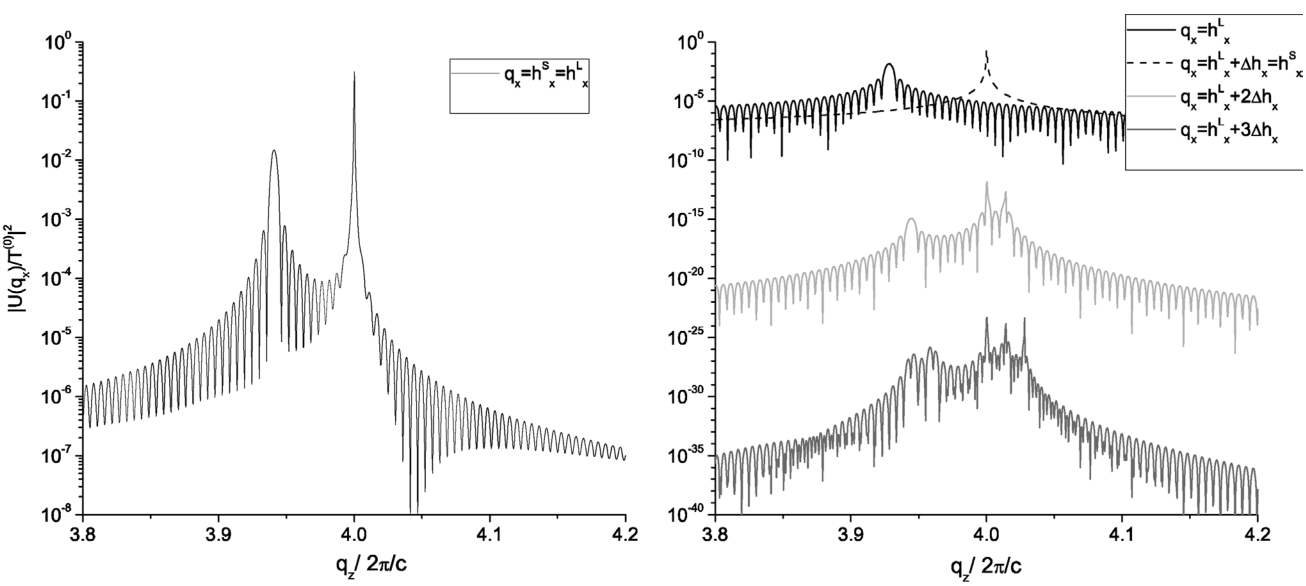

Figure 2 Simulated profiles of the intensities for the various harmonics in case of the fully strained $\mathcal{R} = 0$ (left) and the partially relaxed $\mathcal{R} = 0.1$ (right) structure described in the text.

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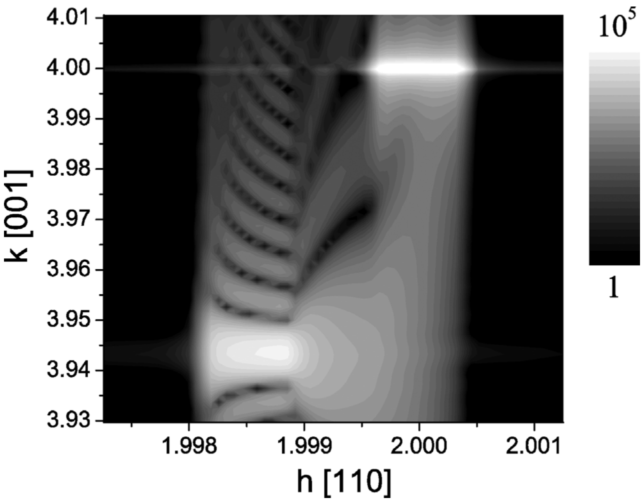

Figure 3 Simulated RSM for the sample with the strain transition layer as described in Section 2.

sphericity of the wave front set. The following expression for the scattering amplitude $T(\boldsymbol{q}) \equiv T(q_x, q_z)$ can be obtained

$$\begin{aligned} T(\boldsymbol{q}) &= T_0(\boldsymbol{q}) + T_1(\boldsymbol{q}); \\ T_0(\boldsymbol{q}) &= 2if(q_x, h_x^{\mathrm{S}}) \left(\sqrt{k_0^2 - k_{ix}{}^2} + h_z^{\mathrm{S}}\right) U^{(1)}(k_{ix}), \\ T_1(\boldsymbol{q}) &= \int_0^{t_{\mathrm{L}}} \mathrm{d}z f(q_x, h^{\mathrm{L}}(z)_x) k_0^2 \chi^{\mathrm{L}}_{h(z)}\, \mathrm{e}^{\mathrm{i}(\phi_z - q_z z)}, \\ k_{ix} &= \frac{1}{2}\left(\sqrt{\frac{q_z^2[4k_0^2 - (h_x^{\mathrm{S}2} + q_z^2)]}{h_x^{\mathrm{S}2} + q_z^2}} - h_x^{\mathrm{S}}\right), \end{aligned} \quad (10)$$

here

$$f(q_x, h_x^{(\mathrm{L,S})}) = \frac{1}{2\pi}\int_0^{L_\parallel} \mathrm{d}\xi\, \mathrm{e}^{\mathrm{i}\left(q_x - h_x^{(\mathrm{L,S})} - \frac{k_0 \sin^2\theta\xi}{2D}\right)\xi},$$

$L_\parallel$ is the lateral size of the sample; D is the distance between the X-ray source and the sample; the intensity RSM $I(\boldsymbol{q})$ is calculated as $|T(\boldsymbol{q})|^2$.

Figure 3 shows the simulated RSM for the layer $Si_{0.82}Ge_{0.18}$ with $L = 100$ nm, $a_{\mathrm{d}} = 30$ nm, $\mathcal{R}_0 = 0.1$ far from the Si substrate. The following parameters were used $D = 3$ m, $L_\parallel = 1$ mm. One can see the clear peak from the substrate (right) and the layer peak being spread because of the strain transition layer. In spite of the strain gradient the thickness oscillations with a low intensity amplitude still appeared at RSM.

3 Coherent diffraction potential for the partially relaxed layer

The above considered model RSM illustrates mainly the possibilities of the dynamical theory for simulation of maps. In order to simulate the experimental RSM the misfit dislocations in the partially relaxed layer and the diffuse scattering should be taken into account [8]. Let us consider briefly the connection between the relaxation parameter $\mathcal{R}$ used in dynamical diffraction theory and the microscopic characteristics of the dislocations in the epitaxial layer.

Both the scattering potential and the wave fields in the Maxwell equations for the crystal should be averaged over the statistical deformation field. Then the value $V_c(\boldsymbol{r})$ averaged over the random distribution of the dislocations defines the coherent ('optical') potential. Analogous approach was described in Ref. [11] for the formation of the coherent potential in the reflectometry from the rough surfaces. This problem is differed a little bit from the approach considered in Refs. [8, 10] where the kinematical scattering intensity was averaged. However, the analogous averaging method can be used and it leads to the following estimation for the relaxation parameter

$$\mathcal{R} = \boldsymbol{a}^{\mathrm{L}}\boldsymbol{A}(\boldsymbol{h}); \quad A_i(\boldsymbol{h}) = h_l \int \mathrm{d}\boldsymbol{\eta}\rho(\boldsymbol{\eta})\frac{\partial u_l(\boldsymbol{\eta})}{\partial \eta_i}, \quad (11)$$

where $\boldsymbol{u}(\boldsymbol{\eta})$ is the displacement field from the dislocation in the point $\boldsymbol{\eta}$ and $\rho(\boldsymbol{\eta})$ is the volume density of the dislocations in the crystal.

Besides, the diffuse scattering from the dislocations diminishes the coherent potential amplitude with the static Debye–Waller factor $\mathrm{e}^{-W_{\mathrm{s}}}$ [8] and leads to the additional spread of the peaks [9]. These effects in the framework of the dynamical theory will be described in detail separately.

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