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X-ray Specular Reflection under Conditions of Extremely Asymmetric Noncoplanar Diffraction from a Bicrystal

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Abstract—The angular dependence of the intensities of X-ray specular reflection has been rigorously analyzed under conditions of noncoplanar grazing Bragg diffraction in a crystal coated with a crystalline film (bicrystal). It is shown that the anomalous angular dependence of the specular-reflection intensity is extremely sensitive to the thickness (from fractions of a nanometer up to several nanometers), deformation, and the amorphization degree of the crystalline films. The optimum conditions for recording intensities are attained at grazing angles equal to 1.5–4.0 of the critical angle of the total external reflection. *© 2003 MAIK "Nauka/Interperiodica".*

INTRODUCTION

Recently, extremely asymmetric X-ray diffraction has been widely used in the diagnostics of the subsurface layers of semiconductor crystals [1–3]. For the first time, an extremely asymmetric diffraction scheme was used in the case where the reflecting atomic planes formed with the crystal surface an angle approximately equal to the Bragg angle, whereas an incident or a diffracted beam forms with the surface a small angle close to the angle of the total external reflection [4, 5]. In this case, X-ray specular reflection starts playing an important role, which considerably reduces the penetration depth of the field in the crystal and allows one to study ultrathin layers with thicknesses of the order of 10 nm. The shortcoming of the extremely asymmetric coplanar system is the requirement that the specimen surface have a special orientation, which hinders the use of these schemes in surface diagnostics.

In [6], a new scheme of noncoplanar diffraction from the reflecting planes normal to the crystal surface was suggested (the tilt angle with respect to the normal is $\psi = 0$). In this case, both incident and diffracted beams may simultaneously form small angles φ_0 and φ_h with the surface and experience strong specular reflection. Experimentally, this diffraction geometry was used in [7] for studying 7.5 to 200.0-nm-thick crystalline aluminum films grown on a GaAs substrate.

Unlike the conventionally used two-wave approximation [1–3], the analysis of diffraction in the grazing geometry requires a rigorous solution of the equations of the dynamical theory. This theory (in the case $\psi = 0$, based on the solution of a biquadratic dispersion equation) has been constructed for both ideal single crystals [8, 9] and crystals coated with an amorphous [10] or crystalline [11] film. It was shown that the diffraction reflection (rocking) curves are very sensitive to the degree of perfection of the subsurface layers with the thicknesses of several nanometers or higher. The results of the corresponding experiments are considered elsewhere [1, 3].

In practice, cutting and the subsequent treatment of crystals do not allow one to obtain surfaces that are strictly parallel to the atomic planes. Therefore, the theory of the grazing X-ray diffraction from an ideal crystal whose planes form a small tilt angle $\psi \neq 0$ with the surface normal was developed [12, 13], and it was shown that even small tilt angles (several angular minutes) can considerably change the shape of the diffraction reflection curves.

In the most general case $\psi \neq 0$, one has to solve the fourth-degree dispersion equation, which can be solved only numerically. The problem is simplified at the grazing angles φ_0 or φ_h exceeding the critical angle of total external reflection, where the effect of the specularly reflected wave on diffraction drastically decreases. In this connection, the approximate modified dynamical theory of diffraction was developed [14–16], which allows one to solve the problem analytically in the whole range of the angles φ_0 and φ_h except for a narrow interval in the vicinity of the critical angle of the total external reflection for both ideal crystals [14, 15] and crystals coated with homogeneous amorphous films [16], and also for crystals with imperfect crystal structures in thin subsurface layers [14].

The theory of grazing diffraction was further developed in [17, 18], where the method for studying the curves of grazing X-ray diffraction from multilayer crystal structures and superlattices was considered on the basis of the solution of the problem of the dynamical diffraction in each layer. It was shown that the curves of the diffraction reflection are very sensitive to deformation $\Delta a/a \sim 10^{-3}$ of the 10-nm-thick layer of

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crystalline germanium on the surface of a perfect Ge crystal [18].

In all the above studies, attention was focused on the analysis of the diffraction reflection, whereas the angular dependence of the intensities of specular reflection was ignored. At the same time, as was first indicated in [3] and then considered in detail theoretically in [19] and observed experimentally in [20], the angular behavior of grazing reflection in the diffraction region at fixed grazing angles is essentially dependent on the presence of ultrathin amorphous films on the surface and their thicknesses.

This study continues the investigation of the specular reflection of X-rays under the simultaneous fulfillment of the conditions of extremely asymmetric noncoplanar Bragg diffraction begun in [19]. Based on a rigorous solution of the fourth-degree dispersion equation, we performed a detailed analysis of the angular dependences of the specular and diffraction reflection from a bicrystal in the whole range of the grazing and tilt angles of the reflecting planes. It is shown that the specular-reflection curves are extremely sensitive to the parameters of homogeneous crystalline films on the crystal surface.

THEORY OF SPECULAR REFLECTION FROM A BICRYSTAL

Consider the formation of the curves of diffraction and specular reflection from a homogeneous plane-parallel film of arbitrary thickness d with interplanar spacings $a = a_0 + \Delta a$, the Fourier components of polarizability χ_{01} and χ_{h1}, and the reciprocal-lattice vector $\mathbf{h}_1$. The substrate is a single crystal with the reflecting planes forming an angle $\psi \ll 1$ with the surface normal $\mathbf{n}$ directed into the crystal along the z axis and characterized by the Fourier components of polarizability χ_0 and χ_h, the reciprocal-lattice vector $\mathbf{h}$, and the interplanar spacings a_0. The rigorous solution of the problem of dynamical diffraction can be obtained under the condition of equality of the tangential (along the crystal surface) components of the reciprocal-lattice vector, $h_{1t} = h_t$. In this case, the tilt angles of the film ψ_1 are determined from the condition $\cos\psi_1 = (1 + \delta)\cos\psi$, where $\delta = \Delta a/a_0$ is deformation. In the opposite case, one has to analyze the interference of the multiply scattered radiation which, in the film, consists of an infinite set of plane waves [21].

A plane monochromatic X-ray wave $\mathbf{E}_0\exp(i\mathbf{k}_0\mathbf{r})$ is incident from vacuum onto a bicrystal at a grazing angle φ_0 to the surface, so that, simultaneously, the diffraction reflection from the atomic planes of the substrate takes place. The fields in vacuum above the bicrystal surface and in the substrate have the form

$$\mathbf{E}_{\text{vac}}(\mathbf{r}) = \mathbf{E}_0\exp(i\mathbf{k}_0\mathbf{r}) + \mathbf{E}_s\exp(i\mathbf{k}_s\mathbf{r}) + \mathbf{E}_h\exp(i\mathbf{k}_h\mathbf{r}), \quad (1)$$

$$\mathbf{E}_{\text{cr}}(\mathbf{r}) = \sum_j \mathbf{D}_{0j}\exp(i\mathbf{q}_{0j}\mathbf{r}) + \sum_j \mathbf{D}_{hj}\exp(i\mathbf{q}_{hj}\mathbf{r}), \quad (2)$$

where E_0, E_s, and E_h are the amplitudes of the incident, specularly reflected, and diffracted waves, respectively; $|\mathbf{k}_0| = |\mathbf{k}_s| = |\mathbf{k}_h| = k_0 = 2\pi/\lambda$ is the length of the wave vector in vacuum and λ is the wavelength; $k_{sz} = -k_{0z}$; $\mathbf{q}_{0j} = \mathbf{k}_0 + k_0\varepsilon_{\text{cr}j}\mathbf{n}$ and $\mathbf{q}_{hj} = \mathbf{q}_{0j} + \mathbf{h}$ are the wave vectors; and D_{0j} and D_{hj} are the amplitudes of the transmitted and diffracted waves in the substrate ($j = 1, 2$). The values of $\varepsilon_{\text{cr}j}$ are determined from the solution of the general dispersion equation of the dynamical theory [1, 3]

$$(\varepsilon^2 + 2\gamma_0\varepsilon - \chi_0)(\varepsilon^2 + 2\gamma_{h0}\varepsilon - \chi_0 - \alpha) - C^2\chi_h\chi_{\bar{h}} = 0, \quad (3)$$

where $\gamma_0 = \cos(\mathbf{k}_0, \mathbf{n})$ and $\gamma_{h0} = \cos(\mathbf{k}_0 + \mathbf{h}, \mathbf{n})$ are the directional cosines of the incident and diffracted waves, respectively; $C = 1$ and $C = \cos 2\theta$ for the σ- and π-states of the radiation polarization and θ is the angle formed by the incident radiation and the reflecting planes of the substrate; and the parameter $\alpha = 2(\theta - \theta_B)\sin 2\theta_B$ characterizes the deviation of the diffraction angle $\Delta\theta = \theta - \theta_B$ from the exact Bragg angle of the substrate θ_B, which is determined by the relationship $h = 2k_0\sin\theta_B$. If φ_0 is the grazing incidence angle, then

$$\gamma_0 = \sin\varphi_0, \quad \gamma_{h0} = \gamma_0 - \psi_B, \quad (4)$$

where $\psi_B = 2\sin\psi\sin\theta_B$ is the effective parameter of the tilt angle of the reflecting planes in the substrate. The diffraction reflection into the region $z < 0$ (Bragg geometry) is observed at such grazing angles φ_0 that $\gamma_0 < \psi_B$, i.e., $\gamma_{h0} < 0$ in (4).

In the general case, Eq. (3) is a fourth-degree equation with respect to ε and, therefore, has four roots ε_j. If the substrate is thick, the solutions should be chosen based on the condition $\text{Im}\varepsilon_j > 0$. In the Bragg geometry, this condition is met only by two roots (see [12]) denoted here as $\varepsilon_{\text{cr}1}$ and $\varepsilon_{\text{cr}2}$.

In the case of a crystalline film of a finite thickness, one has to take into account four roots in dispersion equation (3); therefore, the field in the film consists of four transmitted and four diffracted waves,

$$\mathbf{E}_f(\mathbf{r}) = \sum_j \mathbf{A}_{0j}\exp(i\mathbf{a}_{0j}\mathbf{r}) + \sum_j \mathbf{A}_{hj}\exp(i\mathbf{a}_{hj}\mathbf{r}), \quad (5)$$

where A_{0j} and A_{hj} are the amplitudes and $\mathbf{a}_{0j} = \mathbf{k}_0 + k_0\varepsilon_{fj}\mathbf{n}$ and $\mathbf{a}_{hj} = \mathbf{a}_{0j} + \mathbf{h}_1$ are the wave vectors of the transmitted and diffracted waves in the crystal film ($j = 1, 2, 3, 4$). The ε_{fj} values are determined from the solution of dispersion equations (3) in which the following replacements are made:

$$\chi_0 \longrightarrow \chi_{01}, \quad \chi_h \longrightarrow \chi_{h1}, \quad \chi_{\bar{h}} \longrightarrow \chi_{\bar{h}1},$$

$$\alpha \longrightarrow \alpha_1 = 2(\theta - \theta_B - \Delta\theta_f)\sin 2\theta_B,$$

$$\gamma_{h0} \longrightarrow \gamma_{h01} = \gamma_0 - \psi_{B1},$$

where $\Delta\theta_f = -2\delta\tan\theta_B + \delta\sin\varphi_0/(\sin\psi\cos\theta_B)$ is the displacement of the maximum of the diffraction reflection curve of the film from the maximum of the diffraction reflection curve of the substrate, $\psi_{B1} = 2\sin\psi_1\sin\theta_{B1}$ is the effective parameter of the tilt angle of the reflecting planes of the film, and θ_{B1} is the Bragg angle of the film.

It follows from the basic system of dynamical equations [1] that the amplitudes of the diffracted and transmitted waves in the film and the substrate are related as $A_{hj} = R_{aj}A_{0j}$, $D_{hl} = R_{0l}D_{0l}$ (j = 1–4, l = 1, 2) where

$$R_{aj} = (\varepsilon_{fj}^2 + 2\gamma_0\varepsilon_{fj} - \chi_{01})/C\chi_{\bar{h}1},$$
$$R_{0l} = (\varepsilon_{crl}^2 + 2\gamma_0\varepsilon_{crl} - \chi_0)/C\chi_{\bar{h}}. \quad (6)$$

In order to determine the field amplitudes in Eqs. (1), (2), and (5), we write the continuity condition for the tangential components of the electric and magnetic fields at the upper and lower boundaries of the film. We also have to take into account that at grazing angles, the continuity of the magnetic field is equivalent to the continuity of the first derivative of the electric field with respect to the coordinate z. As a result, we arrive at the following system of eight equations. At the vacuum–film boundary, we have

$$E_0 + E_s = \sum_j A_{0j}, \quad \gamma_0(E_0 - E_s) = \sum_j \Gamma_{f0j}A_{0j},$$
$$E_h = \sum_j R_{aj}A_{0j}, \quad -\gamma_h E_h = \sum_j \Gamma_{fhj}R_{aj}A_{0j}. \quad (7.1)$$

At the film–substrate boundary, we have

$$\sum_j \Gamma_{f0j}A_{0j}g_{fj} = \sum_l \Gamma_{cr0l}D_{0l}g_{crl},$$
$$\sum_j R_{aj}A_{0j}g_{fj}\tau_f = \sum_l R_{0l}D_{0l}g_{crl}\tau_{cr}, \quad (7.2)$$
$$\sum_j \Gamma_{fhj}R_{aj}A_{0j}g_{fj}\tau_f = \sum_l \Gamma_{crhl}R_{0l}D_{0l}g_{crl}\tau_{cr},$$

where j = 1–4, l = 1, 2, $\gamma_h = \sin\varphi_h$ ($\varphi_h > 0$), and φ_h is the angle of the diffracted-radiation exit into vacuum with respect to the surface; at the given angles φ_0 and ψ, the exit angle φ_h is determined by equation $\gamma_h = (\gamma_{h0}^2 + \alpha)^{1/2}$ [12] and the condition $\alpha > -\gamma_{h0}^2$ sets the admissible deviations $\Delta\theta$ from the Bragg angles; and $g_{fj} = \exp(ik_0\varepsilon_{fj}d)$, $g_{crl} = \exp(ik_0\varepsilon_{crl}d)$, $\tau_f = \exp(-ik_0\psi_{B1}d)$, and $\tau_{cr} = \exp(-ik_0\psi_B d)$ are the coefficients that take into account the change in the phases of the waves and their absorption during their propagation in the film. We used the following notation:

$$\Gamma_{f0j} = \gamma_0 + \varepsilon_{fj}, \quad \Gamma_{fhj} = \Gamma_{f0j} - \psi_{B1},$$
$$\Gamma_{cr0l} = \gamma_0 + \varepsilon_{crl}, \quad \Gamma_{cr0l} - \psi_B. \quad (8)$$

The solution of system (7) for the amplitude coefficients $R_S = E_s/E_0$ of the specular reflection and $R_h = E_h/E_0$ of the Bragg reflection have the following form:

$$R_S = \sum_j (\gamma_0 - \Gamma_{f0j})Q_j \Big/ \sum_j (\gamma_0 + \Gamma_{f0j})Q_j,$$
$$R_h = (\gamma_0/\gamma_h)\sum_j R_{aj}(\gamma_h - \Gamma_{fhj})Q_j \Big/ \sum_j (\gamma_0 + \Gamma_{f0j})Q_j. \quad (9)$$

Here, Q_j are the coefficients relating the amplitudes of the transmitted waves in the field: $A_{0j} = Q_jA_{01}$. For a crystalline film, the coefficients take the form

$$Q_1 = 1,$$
$$Q_2 = -\sum_j R_{aj}(\gamma_h + \Gamma_{fhj})U_{1j} \Big/ \sum_j R_{aj}(\gamma_h + \Gamma_{fhj})U_{2j}, \quad (10)$$
$$Q_k = (-1)^{k+1}(U_{1k} + U_{2k}Q_2) \quad (k = 3, 4),$$

where the following notation was used

$$U_{j3,\,j4} = (g_{fj}/g_{f3,\,f4})(T_{1j}T_{24,\,23} - T_{2j}T_{14,\,13})/(T_{14}T_{23} - T_{24}T_{13}),$$
$$T_{1j,\,2j} = R_{02,\,01}(\Gamma_{cr01,\,2} - \Gamma_{f0j})\tau_{cr} - R_{aj}(\Gamma_{crh1,\,2} - \Gamma_{fhj})\tau_f. \quad (11)$$

Relationships (9)–(11) are the rigorous solution of the problem of the specular and diffraction reflection of X-rays from single crystals coated with homogeneous crystalline films. These relationships are valid for all the grazing angles φ_0 and the exit angles φ_h at $\gamma_0 < \psi_B$ and any admissible deviations $\Delta\theta$ from the exact Bragg angle.

Consider some limiting cases. If $d = 0$ (there is no film), then $g_{fj} = g_{crl} = \tau_f = \tau_{cr} = 1$ (j = 1–4, l = 1, 2), and general formulas (9) are reduced to the formulas that describe the specular and diffraction reflection from an ideal single crystal [19]. For a thick film, one has to select the solutions of the diffraction equation in the film proceeding from the condition $\mathrm{Im}\varepsilon_{fj} > 0$. The absorption factor $g_{fj} \longrightarrow 0$ and $g_{crj} \longrightarrow 0$ and formulas (9) coincide with the corresponding formulas for a medium that has film parameters.

Now, consider a homogeneous amorphous film on the surface of a single crystal. Two waves (transmitted and specularly reflected) excited by the incident radiation, A_{01} and A_{02}, and two waves excited by the Bragg wave that enter the film from the crystal, A_{h2} and A_{h1}, propagate in the film. In this case, $\Gamma_{f01} = -\Gamma_{f02} = (\gamma_0^2 + \chi_{01})^{1/2}$ and $\Gamma_{fh1} = -\Gamma_{fh2} = (\gamma_h^2 + \chi_{01})^{1/2}$, $Q_{3,\,4} = 0$, $R_{a3,\,4} = 0$, and $R_{a1,\,2}$ are the coefficients relating the amplitudes of the waves in the film; i.e., $A_{h1,\,2} = R_{a1,\,2}A_{01,\,2}$. The coefficients $R_{a1,\,2}$ are not determined by Eqs. (6) but from the solution of the system of the boundary equations. Thus, formulas (9) are reduced to the expressions that describe the specular and diffraction reflection from a single crystal coated with an amorphous film [19].

In the range where the grazing angle φ_0 is much larger than the critical angle of the total external reflection $\varphi_c = \arcsin(|\chi_0|^{1/2})$, the roots of the dispersion equation (3) have considerably different values. With due regard for the smallness of polarizabilities χ_h and χ_{h1}, one can show that $\varepsilon_{f1,f2} \approx -\gamma_0 \pm (\gamma_0^2 + \chi_{01})^{1/2}$ and $\varepsilon_{f3,f4} \approx |\gamma_{h0}| \pm (\gamma_{h0}^2 + \alpha_1 + \chi_{01})^{1/2}$, and $\varepsilon_{cr1} \approx \chi_0/2\gamma_0$, $\varepsilon_{cr2} \approx 2|\gamma_{h0}| + (\alpha + \chi_0)/2|\gamma_{h0}|$. A rigorous numerical solution of Eq. (3) gives the same results, whence it follows that $R_{a1,2} \ll R_{a3,4}$ and $R_{01} \ll R_{02}$, i.e.,

$$U_{13} \approx 0, \quad Q_2 \approx 0, \quad Q_3 \approx 0,$$

$$Q_4 \approx -U_{14} \approx -(R_{a1}\tau_f - R_{01}\tau_{cr})g_{f1}/(R_{a4}\tau_f - R_{01}\tau_{cr})g_{f4}.$$

Since $\varepsilon_{f1}, \varepsilon_{f4}, \varepsilon_{cr1} \ll \gamma_0$, then $R_S \ll 1$; therefore, one can ignore the effect of specular reflection on the diffraction process. At the same time, the specific behavior of the total wave field in a crystal in the region of strong diffraction reflection from the substrate dramatically affects the angular dependence of the specular reflection. As a result, Eqs. (9) for the amplitude coefficients of the diffraction and specular reflection yield the following expressions:

$$R_h = \frac{R_{a1} + Q_4 R_{a4}}{1 + Q_4},$$
$$R_S = \frac{(\gamma_0 - \Gamma_{f01}) + Q_4(\gamma_0 - \Gamma_{f04})}{(\gamma_0 + \Gamma_{f01}) + Q_4(\gamma_0 + \Gamma_{f04})}, \quad (12)$$

which coincide with the corresponding expression obtained for a bicrystal in the two-wave approximation of large grazing angles in [22].

RESULTS AND DISCUSSION

Figure 1 and 2 show the curves of the diffraction $P_h = (\gamma_h/\gamma_0)|R_h|^2$ and specular $I_S = |R_S|^2 I_0$ reflection, where I_0 is the intensity of the X-ray beam incident onto a silicon single crystal coated with a film of crystalline silicon at different film thicknesses and grazing angles. As is seen from Fig. 1, the diffraction reflection curves are sensitive to the thicknesses of coating crystalline films, which is seen from the thickness oscillations. At large grazing angles and deviations from the exact Bragg condition, it follows from Eq. (12) that the oscillation period is determined by the expression $\Delta\theta = -4\pi\gamma_{h01}/(k_0 d \sin 2\vartheta_B)$; for the parameters that were used in the calculation of curve *5* in Fig. 2c, $\Delta\theta \approx 600''$. With an increase in the grazing angle, the oscillation period and the intensity of the reflected signal drastically decrease (cf. Figs. 1a, 1c). At the same time, the situation for the specular reflection curves (Fig. 2) is quite different. The angular dependences of specular reflection have extremely high sensitivity to the presence and the thicknesses of the crystalline films: with an increase in the grazing angle, the sensitivity increases (cf. Figs. 2a, 2c), whereas the intensity of the useful signal increases by two to three orders of magnitude.

The most interesting situation is observed at the grazing angles $\varphi_{0,h} > \varphi_c$ for silicon $\varphi_c = 13.38'$. In this case, the specular reflection curves show very high sensitivity to the presence of a very thin disturbed layer on the surface, whose thickness can be of several nanometers (Figs. 2b, 2c).

As was first noted in [3] and then considered in detail for an ideal crystal and a crystal coated with an amorphous film in [19], the characteristic feature of specular reflection under the diffraction conditions is a pronounced anomaly in the angular dependence $I_S(\Delta\theta)$, which is of the dispersion type with the minimum and maximum in the vicinity of the diffraction angles $\Delta\theta_{1,2} = \Delta\theta_0 \mp \Delta\theta_B$ corresponding to the boundaries of the region of the total diffraction reflection:

$$\Delta\theta_0 = -\chi_0(1+b)/(2b\sin 2\theta_B),$$

$$\Delta\theta_B = C|\chi_h|/(b^{1/2}\sin 2\theta_B),$$

where $b = -\gamma_0/\gamma_{h0}$ is the asymmetry coefficient of the Bragg reflection ($b > 0$).

It should be noted that the curves of the secondary-radiation yield $I_{SP} \sim 1 + |R_h|^2 + 2\sigma \mathrm{Re} R_h$ with a yield depth that is small in comparison with the extinction length $L_{ex} = \lambda(\gamma_0|\gamma_{h0}|)^{1/2}/\pi C|\chi_h|$, where $\sigma = C|\chi_{hi}|/\chi_{0i}$, $\chi_{gi} = \mathrm{Im}\chi_g$ [1, 3, 23], have approximately the same shape. The analogy becomes more obvious if the quantity Q_4 in (12) is expressed in terms of the amplitude coefficient of the diffraction reflection R_h. Then the amplitude of the specular reflection is

$$R_S \approx -(\chi_0/4\gamma_0^2)(1 + \sigma_s R_h), \quad (13)$$

where $\sigma_s = C b_f^{1/2}(\chi_{h1}\chi_{\bar{h}1})^{1/2}/\chi_{01}$. Similar to the method of X-ray standing waves (XRSW) [1, 23], the second factor in (13) characterizes the amplitude of the total field on the bicrystal surface. However, the value of σ_s in (13) is not determined by the relative ratio of the imaginary parts of the Fourier components of the polarizabilities χ_h and χ_0 any more. Despite the fact that, at the grazing angles $\varphi_0 > \varphi_c$, the coefficient of specular reflections is very small, the intensity of this reflection can considerably (by several orders of magnitude, all other conditions being the same [20]) exceed the photoelectron or fluorescent quantum yield in the XRSW method.

The presence of the minimum and maximum on the specular-reflection curve $I_S(\Delta\theta)$ (13) is explained by the fact that, in the region of diffraction reflection, $P_h \approx 1$, and the phase R_h changes almost linearly from π at $\Delta\theta = \Delta\theta_1$ to zero at $\Delta\theta = \Delta\theta_2$. In this case, $R_h(\Delta\theta_{1,2}) \approx \mp b_f^{1/2}$, i.e., has different signs, which results in the formation

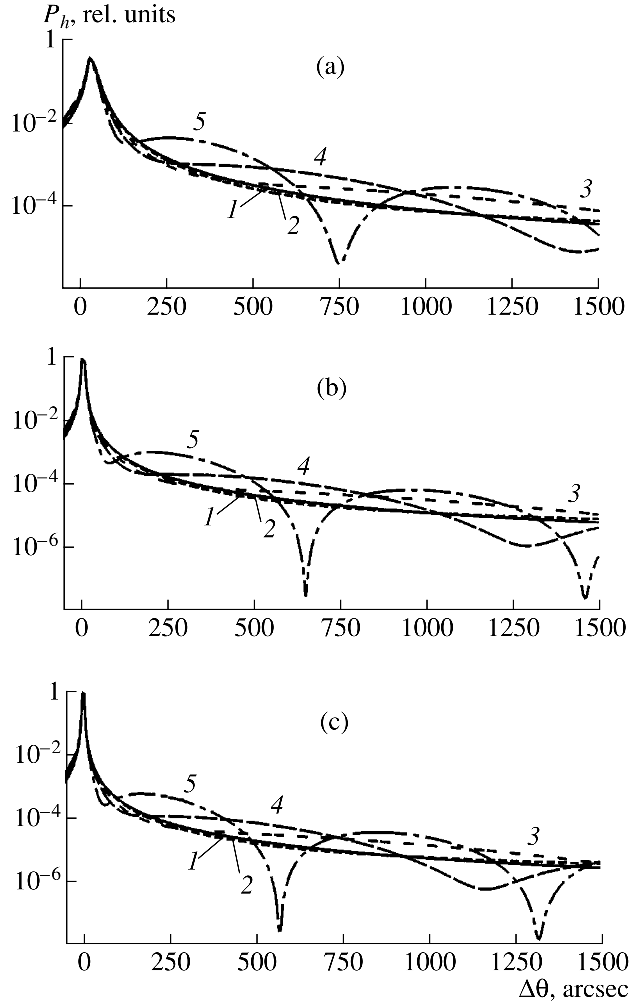

Fig. 1. Effect of the thickness of the surface crystalline silicon film on the shape of the diffraction-reflection curves depending on the angular deviation $\Delta\theta$ from the Bragg angle of the substrate (Si) at the grazing angles φ_0 = (a) 13′, (b) 25′, and (c) 45′. The film thickness d (nm) is (*1*) 0 (ideal crystal), (*2*) 1, (*3*) 2, (*4*) 3, (*5*) 5. CuK_α radiation, Si(220) reflection, $\psi = 3°$, the amorphization factor $F_{am} = 1$, deformation $\delta = -4 \times 10^{-4}$.

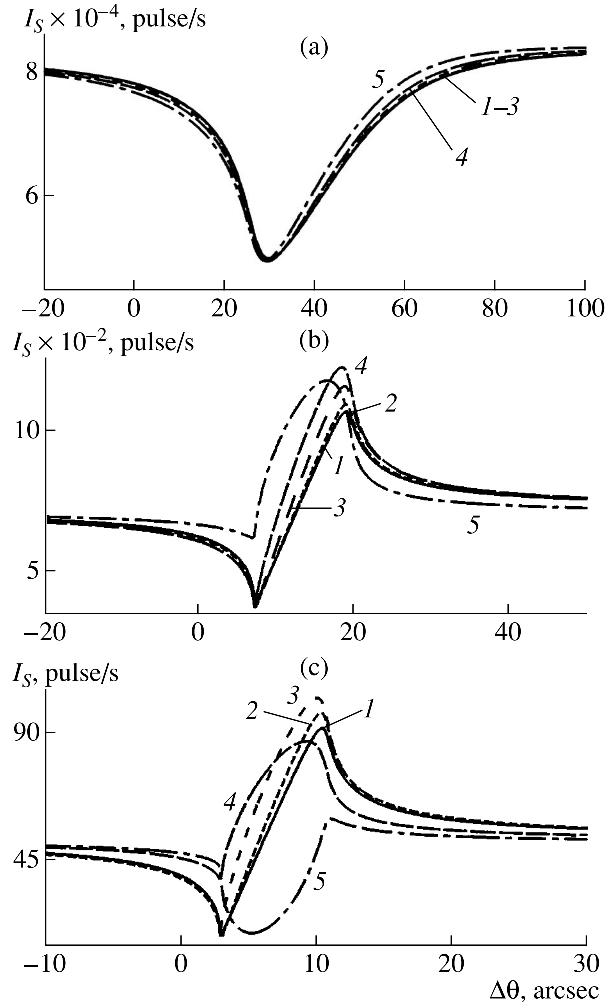

Fig. 2. Effect of the thickness of the surface crystalline silicon film on the shape of the angular curves of the specular-reflection intensities depending on the angular deviation $\Delta\theta$ from the Bragg angle at the grazing angles φ_0 = (a) 13′, (b) 25′, and (c) 45′. The film thickness d (nm): (*1*) 0 (ideal crystal), (*2*) 1, (*3*) 2, (*4*) 3, (*5*) 5, amorphization factor $F_{am} = 1$, deformation $\delta = -4 \times 10^{-4}$. The intensity of incident radiation I_0^5 pulse/s.

of the minimum and maximum on the specular-reflection curve I_S. At small grazing angles ($\gamma_0 \ll \psi_B$), the asymmetry coefficient of reflection $b_f \ll 1$. With an increase in the angle φ_0, at $\gamma_0 \approx \psi_B$, we have $b_f \gg 1$, and the contrast of the specular-reflection curve I_S increases.

The penetration depth of the field under conditions of specular reflection and large grazing angles obeys the inequality $L_s \gg L_{ex}$, where $L_s = \lambda/(2\pi \mathrm{Im}\gamma_s)$, and $\gamma_s = (\gamma_0^2 + \chi_0)^{1/2}$. Therefore, the formation of the refracted wave is determined by the coherent superposition of the transmitted and diffracted waves. Unlike this situation, in the region of small angles $\varphi_0 \le \varphi_c$, the penetration depth of the field $L_s \le L_{ex}$; i.e., it is determined mainly by the total external reflection. Diffraction reflection occurs in a thinner layer, which results in a decrease in the amplitude of the maximum of the diffraction-reflection curve and, simultaneously, in its broadening (Fig. 1a). The specular-reflection curve in this case is pronouncedly smoothened and acquires the shape of a shallow minimum (Fig. 2a). For the parameters used in the calculation of the curves in Figs. 1 and 2, the penetration depths L_s = 1.9 μm, L_{ex} = 0.5 μm, and b_f = 0.69; L_s = 0.6 μm, L_{ex} = 0.2 μm, and b_f = 0.19; and L_s = 0.03 μm, L_{ex} = 0.13 μm, and b_f = 0.12 at the grazing angles φ_0 = 50″, 20″, and 13″, respectively.

At large grazing angles and pronounced angular deviation from the exact Bragg condition for the crystalline film, the amplitude coefficient of specular reflec-

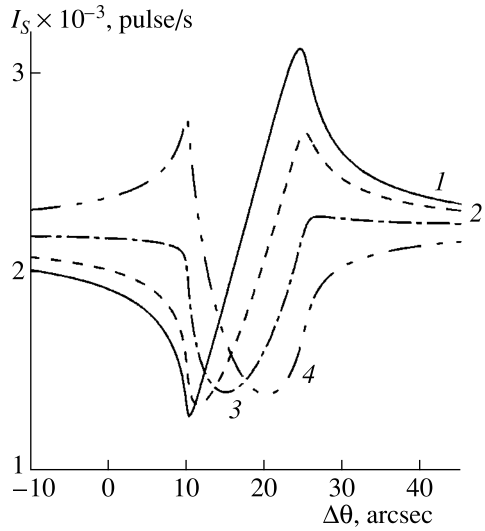

Fig. 3. Effect of deformation of the crystal film on the shape of the angular dependences of the intensities of specular reflection. Deformation $\delta \times 10^{-4}$; (*1*) 0 (ideal crystal), (*2*) 2, (*3*) 4, (*4*) 6. Grazing angle $\varphi_0 = 20'$. Thickness of the Si film $d = 5$ nm. Amorphization factor $F_{am} = 1$.

tion given by Eqs. (12) can be written in the convenient form

$$R_S \approx -(\chi_{01}/4\gamma_0^2)\{1 - R_{01}C_f(\tau_{cr}/\tau_f)\exp(i\omega)/\chi_{01}\}, \quad (14)$$

where, in accordance to [23], the following notation is introduced:

$$y_1 = [\alpha_1 b_f + \chi_{01}(1+b_f)]/2C_f, \quad y_{12} = (y_1^2 - 1)^{1/2},$$

$$C_f = Cb_f^{1/2}(\chi_{h1}\chi_{\bar{h}1})^{1/2}, \quad \omega = k_0 C_f d y_{12}/\gamma_0.$$

The quantity y_1 characterizes the deviation from the exact Bragg condition for the crystalline film.

The first term in (14) describes the behavior of the specular-reflection curve from the film far from the diffraction condition. The second term describes the dispersion behavior and the thickness oscillations (caused by the presence of the film) on the angular dependence of the specular reflection. A further increase in the film thickness results in a lower contrast of the specular-reflection curve in the region of diffraction reflection from the substrate and an increase in the contrast in the region of diffraction reflection from the film.

Figures 3 and 4 show the angular dependences of specular reflection from a bicrystal with various deformations δ and amorphization factors F_{am} of the film ($\chi_{h1} = F_{am}\chi_h$), respectively. As was indicated above, the diffraction-reflection curves of thin films are almost the same as those of the substrate. At the same time, even insignificant changes in the deformation and amorphization factor of the film lead to considerable changes in the shape of the specular-reflection curves in the region of diffraction reflection from the substrate.

Figure 5 shows the specular-reflection curves from a bicrystal at various tilt angles ψ of the reflecting planes of the substrate. It is seen that the sensitivity of the

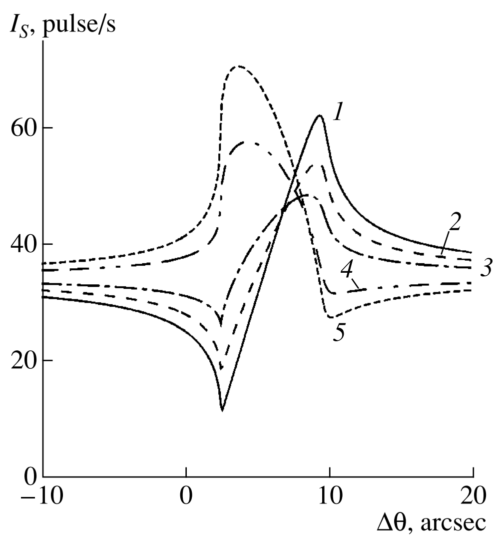

Fig. 4. Effect of the amorphization factor of the crystalline film on the shape of the angular dependences of the specular-reflection intensity. Amorphization factor F_{am}: (*1*) 1 (ideal crystal, (*2*) 0.8, (*3*) 0.6, (*4*) 0.2, (*5*) 0 (amorphous film). Grazing angle $\varphi_0 = 50'$. Thickness of the Si film $d = 2$ nm. Deformation $\delta = 0$.

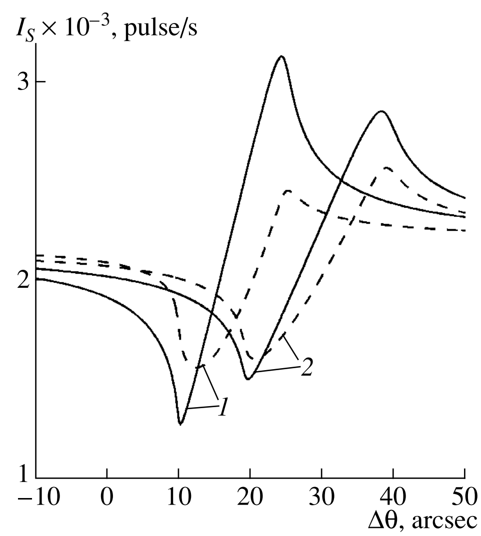

Fig. 5. Effect of the tilt angle of the atomic planes ψ on the angular dependence of the specular-reflection intensity from a bicrystal (dash lines) and an ideal crystal (solid lines). The tilt angle (*1*) $\psi = 3°$ and (*2*) $\psi = 5°$. Grazing angles $\varphi_0 = 20'$. Thickness of the Si film $d = 4$ nm. Amorphization factor $F_{am} = 1$, deformation $\delta = 4 \times 10^{-4}$.

angular dependence of specular reflection increases with a decrease in the tilt angle.

CONCLUSIONS

The rigorous dynamical theory of specular reflection of X-rays from a bicrystal under conditions of extremely asymmetric diffraction and specular reflection are solved in the general form so that the results obtained are valid in the whole range of grazing angles of an incident beam and exit angles of diffracted radiation.

It is shown that the angular dependence of the specular-reflection intensity is very sensitive to the presence, thickness, deformation, and degree of amorphization of a thin (from fractions of a nanometer to several nanometers) crystalline film in the crystal surface. The problem can readily be generalized to the case of grazing and diffraction reflection from an arbitrary multilayer structure with any profiles of the variations in polarizability, deformation, and the amorphization factor.

The intensity of the specular reflection is sufficiently high and allows one to perform rapid analysis of thin subsurface and transient layers. The sensitivity of the method to the film thickness is about 0.5 nm and increases with an increase in the grazing angle; however, the intensity of the reflected signal simultaneously decreases. The optimum grazing angles range from one and a half to three to four critical angles of the total external reflection. At smaller grazing angles, the intensity of specular reflection increases; however, the sensitivity considerably decreases.

Thus, the results obtained show that it is possible to perform rapid nondestructive analysis of the structure of superthin subsurface layers and the interfaces using the specular-reflection data obtained under conditions of grazing Bragg diffraction.

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