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Influence of absorption level on mechanisms of Bragg-diffracted x-ray beam formation in real silicon crystals

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Abstract. The methods of numerical calculations based on the formulae of the X-ray dynamic scattering theory by real crystals and of the Takagi-Topin equations were used for investigation of the basic regularities of inherent to the Bragg diffraction in conditions of a strong and weak absorption. The mechanisms of profile formation of a spatial intensity distribution of diffracted beams depending on an energy of radiation and on structural perfection parameters of crystals are discussed. The formulae for an analytical description of spatial intensity distribution profiles which take into account the dynamical corrections (coefficients of extinction) for coherent and incoherent components of the total reflectivity were used.

Keywords: X-ray beams, Bragg-diffraction , reflectivity, extinction and absorption lengths, dynamical theory of scattering, structure defects, clusters.

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Introduction

The Bragg diffraction in well known as an important tool for developing effective nondestructuve techniques in investigations of structure defects intrinsic to thin near-surface layers of crystals widely used in microelectronics [1]. Despite existing opinion about weak sensitivity of methods based on the Bragg diffraction peaks intensity to structure defects [2], considerable attention was paid recently [3-6] for investigation of X-ray scattering in this case.

The revival of investigators interest to the Bragg geometry for the indicated aims is due probably to the results of papers [7-9], carried out with use of short-wave radiation, in which the high sensitivity of integral intensity to structure defects is, however, shown to take place. In particular, the total integrated reflectivity (TIR) was shown to increase essentially with growth of a distorsion level in crystals with chaotically distributed defects at the expense of a diffuse component [10]. The contribution of this component becomes noticeable already at $t \geq \Lambda$, ($t = 1/\mu_0$, μ_0 is normal photoelectric absorption coefficient, $\Lambda = \lambda \cdot \sin\vartheta / |\chi_h| \cdot C$ is length of an extinction). The series of experimental results [11,12] does not find, nevertheless, a convincing explanation within the framework of the existing theories of a scattering by real crystals, especially under the conditions of strong absorption of radiation, for example, near the absorption K-edges. It concerns, for example, to the excess of the X-ray dynamic scattering level in an ideal crystal in comparison with an ideal - mosaic crystal.

The aim of this work consisted in discovering of character and basic mechanisms of intensity losses of the Bragg diffraction peaks at variation of incident radiation energy by methods of numerical calculations using the formulae of the dynamical theory, developed for the TIR of crystals with homogeneously distributed defects [6] , as well as by the method of the Takagi-Taupin (TT) equations solution.

Object and methods of investigations

Convenient objects for solution of the posed problem are the silicon crystals containing so-called Coulomb centers of strains (clusters), which sizes and concentrations of which are suitable to change in wide reasonable limits. Besides, utilization of silicon also allows to vary a level of absorption for the known wavelengths of a characteristic spectrum over a wide range. The analytical calculations of the TIR and its components were carried out under the formulae of the Braggdiffraction theory [6] for homogeneously distributed clusters. The level of distorsions and its influence on a character of scattering was stimulated by introduction in to the formulae for an ideal crystal [2] the known parameters of structural perfection, i.e., exponent of the Debye - Waller static factor L, and coefficients of an extinction for coherent, μ_{ds}, and diffuse, μ^* components of intensity, too. TIR for a real crystal in case of the Bragg diffraction is known to be the sum of two parts, i.e., the Bragg (coherent) , R_{iB}, and diffuse (incoherent), R_{iD}, components :

$$R_i = R_{iB} + R_{iD} = R_{i0} \cdot E + (1 - E^2) \cdot R_{iK} , \tag{1}$$

where R_{i0}, R_{iK} are the integral reflectivity (IR) of an ideal and ideal - mosaic crystals, respectively, and $E = \exp(-L)$ is the static Debye - Waller factor. The values of the mentioned parameters of structural perfection (L, μ_{ds}, μ^*) were varied in a rather wide interval ($0 < L < 1$, μ_{ds}, μ^* up to $0.3\,\mu_0$). Except TIR, the change of its components, R_{iB} and R_{iD}, were also analyzed as those depending on an energy of incident radiation and level of distorsions for clearing up their contributions in R_i .

Theoretical [13,14] and the experimental [10] investigations have shown, that the analysis of a spatial distribution profiles of a diffracted beam $I(x)$ allows to gain the reliable information up on structural perfection degree of crystals. However, the approaches made in [7, 8], allow to handle experimental results only at low levels of distorsions, $L \ll 1$. Besides , the influence of coefficient of an extinction μ^* for diffuse scattered waves was not taken into account in the mentioned papers. The necessity of such an introduction for correct describing the TIR for a real crystal was justified much later.

Respective alterations in the formulae [7,8] were made taking into account the results of theoretical examinations of the TIR for the Bragg diffraction peak [6]. With this purpose, the expression for a spatial intensity distribution profile of a diffracted beam in an ideal absorbing crystal was used, following [14]:

$$\begin{aligned} I_B(x) &= (I_0 \cdot Q \cdot s) / \sin(2 \cdot \vartheta) \times \\ &\times \frac{J_1^2(\alpha \cdot x)}{(\alpha \cdot x / 2)^2} \cdot \exp(-\mu_0 \cdot x / \cos\vartheta) \cdot \end{aligned} \tag{2}$$

Here, I_0 is the intensity of a primary beam, $Q = \pi^2 \cdot d \cdot \cos\vartheta / \Lambda^2$, $\Lambda = \lambda \cdot \sin\vartheta / C \cdot |\chi_h|$ is an extinction length, $d = \lambda / 2 \cdot \sin\vartheta$, $J_1(\alpha \cdot x)$ the Bessel function of the first order, $\alpha = C \cdot K \cdot |\chi_h| / 2 \cdot \cos\vartheta$, $K = 2 \cdot \pi / \lambda$, ϑ is the Bragg angle, and s is a slit width at the detector, respectively.

In a real crystal, the diminution of the coherent component (2) happens at the expense of intensity transition in a diffuse background described by the factor $\exp(-\mu_{ds} \cdot x / \cos\vartheta)$. Besides, this diminution is possible to be taken into account also by renormalization of the Fourier components of a suspectibility χ_h on $\chi_h \cdot E$ according to [6]. Intensity of diffuse background can be submitted according to [5] as follows:

$$I_D(x) = I_o \cdot R_{iD}^{kin}(x) \cdot \Pi(x) , \tag{3}$$

where $R_{iD}^{kin}(x) = \dfrac{Q \cdot s}{\sin 2\vartheta} \cdot (1 - E^2) / E^2$ and

$\Pi(x) = \exp\left[-(\mu_0 + \mu^*) \cdot x\right]$.

Thus, distribution of a total intensity in the Bragg profile of a reflected beam in crystals with defects can be written as:

$$I(x) = I_B(x) + I_D(x) , \tag{4}$$

taking into account the results of [6]. For analysis of the profile of a possible intensity spatial distribution of a diffracted beam and its evolution the calculations were carried out by means of (2 - 4) and using the Takagi-Topin (TT) equations solution [15,16] for specific periodic (ultrasonic) strains. It was done for confirmation of an admissibility of the offered analytical approach for describing of regularities of scattering by a distorted crystal. The extend and the amplitude of distortions in the last case can be easily varied. The approach of the so-called short wavelengths ($\lambda_s \ll \Lambda$) was used. Such approach to modeling the structure distortions in relation to their influence on a X-ray scattering corresponds most closely to the case of homogeneously distributed lattice defects. The level of a lattice distortions was characterized by dimensionless parameter HW ($H = 1/d$ is a vector of reciprocal lattice, W is an amplitude). This value was varied in the interval from 0 up to 2. The physical sense of the parameter HW for a shortwave ultrasound is virtually close to concept of the Debye-Waller static factor [17] in real crystals with structure defects.

The calculations were carried out for the most strong Bragg reflection, 111, and the following wavelengths of X-rays: $WK_\alpha, AgK_\alpha, MoK_\alpha, CuK_\alpha$, as well as for soft radiations with $\lambda = 2$ Џ and $\lambda = 3$Џ.

Results of numerical simulations

Let us consider first the behavior of TIR for various levels of structure distortions in a crystal characterized by an exponent of the Debye-Waller static factor, L. With this purpose, the results of computation of the dependences TIR, R_i, as a function of a wavelength of diffracting radiation calculated using the formula (1) for an ideal and for an ideal - mosaic (the kinematic limit) crystals are given in the Table together with the TIR $R_{i1} \div R_{i3}$ for the samples with defects. The higher the level of parameter L, the more the TIR value of a distorted crystal. This does not contradict the majority of known experimental results obtained by different authors for the Bragg case of X-ray diffraction. Thus, the gaps between R_{iK} and R_{i0}, which make it possible to discriminate samples with various degrees of structural perfection, decrease with increasing wavelength, though the absolute values of the TIR increase under these conditions. Analysis of the R_i values for real crystals (with structure defects) shows that the level of the TIR with growing λ (for example for $\lambda > 2$ Џ) is caused by an essential enhancement of absorption, can be even smaller, than R_{is}. Such behaviour of intensity with increasing a level of distortions was known earlier for the Laue case of diffraction of X-rays in the approach of a thick crystal, when the Borrmann effect is realized. One should note that not only R_{i0}, but also $R_{i1} \div R_{i3}$ are less than the relevant value of a kinematic reflectivity, R_{iK}, in the wide interval of wavelengths ranging from $\lambda = 0.5593$ Џ to $\lambda = 1.930$ Џ.

The diagnostics of structural perfection degree of a sample becomes impossible near the point, where $R_{i1} \div R_{i3}$ are equal to value R_{i0}. It is important to note that the diminution of the TIR for a longwave (soft) radiation at some level of distortions characterized by parameter L can take place instead of increase of this characteristics. It means that the energy dependence $R_i = f(\lambda)$ in a wide interval of wavelengths is non-monotonic. This peculiarity of considered dependence behaviour is observed at large values of the static factor, when the known analytical expressions for the TIR require the extra analysis [4]. One of the most important result following from the performed calculations consists in weak influence of extinction parameter μ_{ds} for hard radiation, when the so-called one-parametric approach in describing of a TIR permissible, which was shown earlier in the case of Laue-diffraction [18]. The carried out calculations also show that the shortwave region of X-ray spectrum at the Bragg diffraction is the most sensitive to structure defects (because of greatest gap $\Delta r = R_{iK} - R_{i0}$). The important characteristics of a scattering for the analysis of the TIR variations in the Bragg case of diffraction is also the parameter $g = |\chi_{i0}|/|\chi_{rh}|$ [19], which characterizes a relation of contributions of a scattering and absorption. The analysis of character of energy dependence variations of this parameter shows, however, that more important for structural diagnostics is the decrement of R_i, i.e., $\Delta R = R_i - R_{i0}$. Really, the ΔR decreases with increasing of a wave length (see the Table) though the parameter g grows (Fig. 1).

The Bragg spatial intensity distributions $I(x)$ calculated for the first time using the formula (4) for various wavelengths are given for a set of structural perfection parameters (Fig. 2). As it follows from the analysis of the obtained results, enhacement of a lattice distortion degree results in noticeable growth of diffuse scattering intensity on large penetration depths of X-rays ($t >> \Lambda$). Thus, the dependences $\ln I(x) = f(x)$ in the thickness region ($t >> \Lambda$), which are not considered here for shortnese, are linear with a slope $(\mu_0 + \mu^*)$. It means that the role of an extinction coefficient μ^* at a diffuse scattering is essential. The magnitude of the Bragg maximum, i.e., the first Uragami peak, decreases in accord with the formula (3). As the slope of the indicated function for a kinematic limit of intensity is equal μ_0, with growth of a

Table. The TIR values and increments ΔR for various wavelengths (reflection 111) in silicon crystals with a various structure perfection degree.

λ, Å	$R_{i0} \times 10^5$	$R_{i1} \times 10^5$, $\Delta R = R_{i1} - R_{i0}$ $\times 10^5$	$R_{i2} \times 10^5$, $\Delta R = R_{i2} - R_{i0}$ $\times 10^5$	$R_{i3} \times 10^5$, $\Delta R = R_{i3} - R_{i0}$ $\times 10^5$	$R_{iK} \times 10^4$, $\Delta R = R_{iK} - R_{i0}$ $\times 10^4$
0.5593	1.49	1.74	1.95	1.94	7.49
		0.26	0.46	0.45	7.341
0.709	1.88	2.12	2.33	2.31	6.09
		0.24	0.45	0.43	5.902
1.54	3.88	4.01	4.11	3.985	2.8
		0.13	0.23	0.1	2.412
1.93	4.75	4.82	4.87	4.65	2.3
		0.07	0.12	-0.1	1.825

The note: reflectivities for ideal crystal; (R_{io}) and real crystals (R_{i1}, $L = 0.05$; R_{i2}, $L = 0,1$; R_{i3}, $L = 0,\ 1, \mu_{ds} = 0,3\,\mu_0$). Ideal-mosaic sample reflectivity is denoted by R_{ik}.

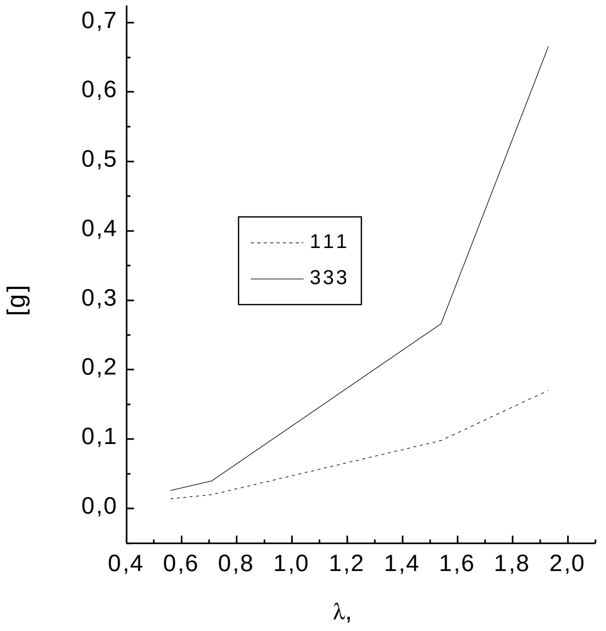

Fig. 1. Dependence of the absorption and scattering relation factor g for the reflections 111 and 333 in Si crystals on a wavelength of X-rays.

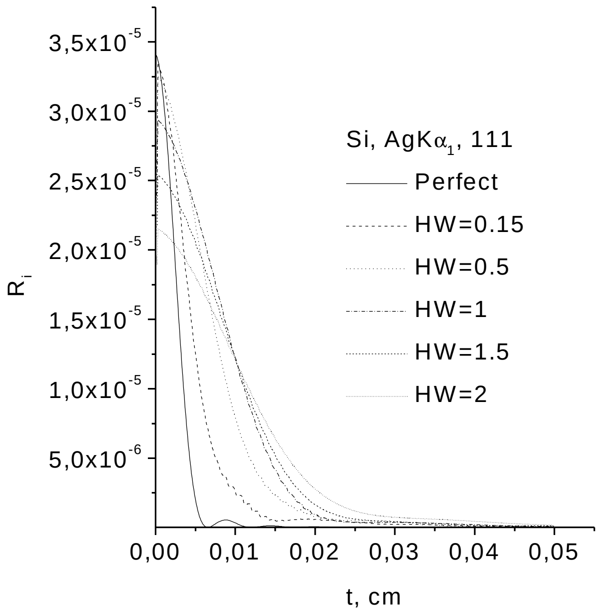

Fig. 2. Profiles of a spatial intensity distribution of Bragg diffracted beams, calculated with the help of the numerical solution of the Takagi-Topin equations for various levels of the parameter HW. $AgK_{\alpha 1}$ radiation, 111 reflection.

distortion degree in a crystal, the quantity μ^* tends, respectively, to zero, reducing the velocity of intensity diminution with a depth. The diminishing contribution of coherent component, observed at small values x (in the limits of Uragami peak) also favors this process. From the physical point of view, the drop of a level μ^* is possible to treat, as diminution of a primary extinction for a diffuse scattering at enhancement of lattice disordering degree.

Most of typical profiles of a spatial intensity distribution for a diffracted X-ray beam, obtained using the numerical solution of the TT equations are given in the Fig. 4 for the case of ultrasonic strains [16] considering series of wavelengths and the parameter HW levels. Some diminution of a coherent maximum and origin of a “diffuse” scattering near its “pedestal” are observed at small values of the HW in this figure. Its contribution increases with diminution of a wavelength of used radiation. At large HW values the diminution of maximum is more considerable for the wavelengths $\lambda > 2$ Џ, where the values of diffuse component reach a maximum in such a manner that its behaviour is described already by the laws of a kinematical scattering.

Influence of possible X-ray scattering mechanisms on the Bragg TIR

The fulfilled numerical experiments confirm that the TIR of a real crystal in the Bragg case of diffraction is formed as the sum of the coherent and incoherent components. Thus, the presence both Coulomb centres of strain and displacement caused by ultrasonic waves results in the following diffraction phenomena:

– Diffuse smearing out of the reciprocal lattice points (broadening of diffraction peaks);

– Appearance of interior sources of incoherent diffracted beams due to again scattered radiation observed in the case of ultrasonic strains [16];

– Drop of intensity of a coherent component, proportional to $\exp(-L)$, resulted in diminution of the coherent Uragami peak on curves of a spatial distribution (Fig. 3).

The intensity of diffuse component of a TIR is therefore determined by the competition of two factors: 1) by growth of the R_{iD} with enhancement of scattering volume and level of distortions; 2) by its exponential absorption at the expense of the total factor of absorption $\mu_0 + \mu^*$. Character of a relation of formation depths of a coherent maximum and appearence of the “diffuse” scattering “pedestal”, as a function of a wavelength, is shown in the Fig. 4 which exhibits an advantage of utilization just short wavelengths in an experiment follows.

Let us consider character of different TIR components variations with a changing wavelength. In the case of short wavelengths (weak absorption) the extinction length increases, while the absorption constant sharply drops ($\mu \sim \lambda^3$). The curve $I(x)$ is essentially expanded due to deep penetration of X-rays into a crystal. Defects, beginning from some particular level of distortions, created by them, result in diminution of coherent compo-

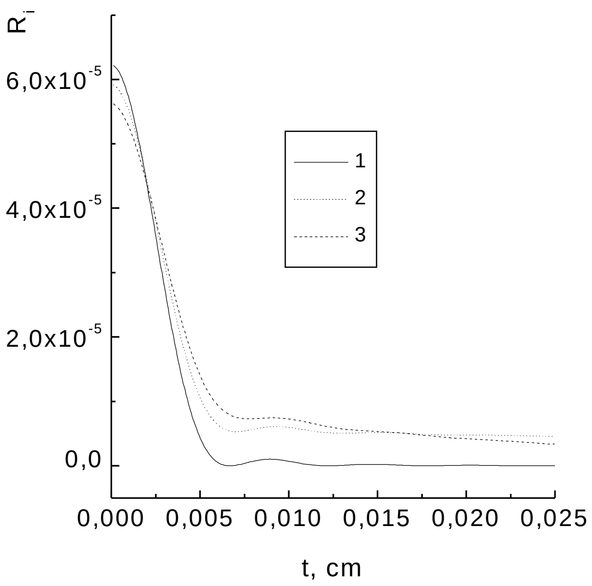

Fig. 3. Profiles of a spatial intensity distribution of the Bragg-diffracted beam of X-rays calculated under the formulae (2) – (4) for different values of structural perfection parameters: 1 – ideal crystal; 2 – real crystal (L = 0.05); 3 – real crystal (L = 0.1, $\mu_{ds} = 0.3\cdot\mu_0$). $AgK_{\alpha 1}$ radiation, 111 reflection .

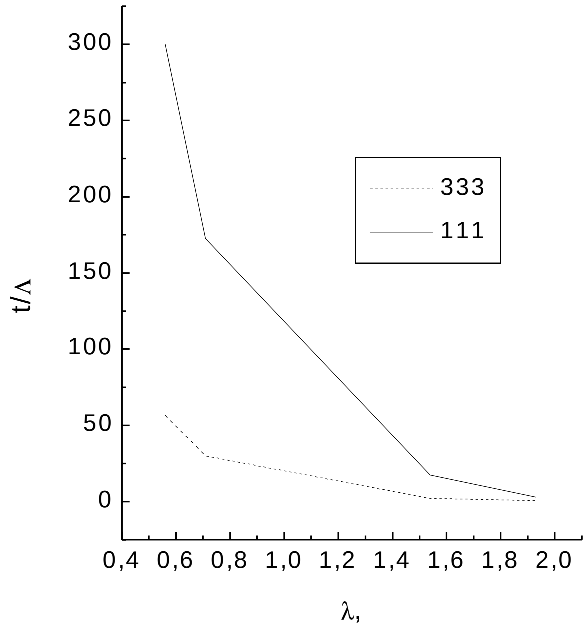

Fig. 4. A relation of absorption and extinction lengths as function of a wavelength for the reflections 111 and 333 in Si crystals.

nent, which can not be compensated by the growth of a incoherent part of the TIR registrated in our experiments, because of very high values of the photoelectric absorption coefficient μ_0 in the case of longwave radiation.

As it follows from results of the carried out calculations, the peak value of intensity in the region of the "pedestal" of a curve of spatial intensity distribution can be described by some enveloping curve (formula (3)). Thus, the behaviour of the TIR at high levels of absorption and lattice distortions is mainly determined by the contribution of coherent part of scattering, as far as the incoherent one reaches a maximum and then tends to the limit described by the kinematic theory, where the factor $2L$ defines a scattering volume of a crystal which is situated in a reflecting position.

The effects, discussed in the paper, are the basis of sensitivity of radiation with some wavelengths at Bragg diffraction to structural distortions of a lattice. In this connection utilization of diffuse scattering of soft radiation, which is strongly absorbed in a crystal and does not reach a surface, is not desirable for structural diagnostics of real crystals. It completely confirms conclusions made earlier [5, 6].

Conclusions

1. Character of the X-ray interferential interaction with a substance in geometry of Bragg diffraction is determined by a value of the relation of imaginary and real parts of the Fourier coefficient of a crystal succeptibility and essentially depends on a wavelength. The sensitivity of a X-ray scattering to structure defects increases with enhancement of this relation, though a difference of the TIR values for a real (measured) and for a perfect crystal is the most important for a structure diagnostics. The modeling of influence of structure defects of a various type (Coulomb centres, periodic strains) within the framework of the dynamic theory of a scattering by crystals with homogeneously distributed defects, or, as it has been shown using the Takagi-Topin equations solutions, the level of a kinematical scattering always exceeds the dynamical one. Using a hard radiation, one can reach rather high sensitivity of the intensity of the Bragg-reflections to lattice strain level described by the parameter L, or HW. With increasing their level the gradual transition $I_B \to I_{Kin}$ is observed.

2. The dependence of the TIR on a level of distortions has monotonous character for a Bragg diffraction of soft radiation. With an increase of a distortion level, the behaviour of the TIR in Bragg case of diffraction is determined by the contribution of a coherent component. Therefore, depending on a type, size and concentration of defects, it is possible to reach an enhancement (small values of μ_{ds}),as well as the diminution of the TIR. The last on was known earlier for the Laue case diffraction (Borrmann effect).

3. One can conclude, that particular caution in interpretation of experimental results should be taken into account by utilizing of soft radiation for structural diag-

nostics of boundary layers with the help of the TIR analysis.

4. Analytical calculations of curves of a spatial intensity distribution of the Bragg diffraction peaks by means of the dynamic theory of a scattering of radiations by real crystals, are in the whole adequate to data of the Takagi-Topin equations solution and describe satisfactorily the behaviour of measured reflectivity as well as the character of a spatial intensity distribution in a wide interval of wavelengths and absorption levels.

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