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DIFFRACTION AND SCATTERING OF IONIZING RADIATIONS

On the Theory of Diffraction of Limited Synchrotron Radiation Beams in Single Crystal in the Laue Case

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Abstract—The features of the Bragg diffraction of coherent synchrotron radiation from the atomic lattice of a single crystal in the Laue geometry have been studied theoretically, provided that the radiation beam is limited by a relatively large slit placed in front of the crystal. The method of numerical simulation is used, and dependences of the intensity distribution are obtained for different crystal thicknesses. It is shown that the slit edges introduce inhomogeneous intensity distortions inside the Borrmann fan with an angle of $2\theta_B$, where θ_B is the Bragg angle. In the area where the triangles intersect the intensity distribution is similar to that for the diffraction from a slit in air at a certain (large) distance. An equation for the correspondence between the distance and crystal thickness is derived, which describes well the numerical calculation results.

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INTRODUCTION

The diffraction of a limited beam of visible light was observed by even primeval people, when sunlight entered a cave through a hole in a pile of stones. At short distances the light propagates in a straight line, and the beam takes the transverse shape of the hole. Later the light propagation theory was developed, known as geometric optics. However, this theory becomes invalid with a decrease in the slit size, and wave properties of light begin to manifest themselves. Currently it is generally accepted to select three ranges of distance, in which light beam transformation with an increase in distance z after a slit of width d occurs in radically different ways [1].

At short distances (near field) the geometric optics is approximately valid. At long distances (far field), when a plane wave is incident on a slit, the transverse beam size is approximately equal to $\lambda z/d$ and increases proportionally to the distance (λ is the monochromatic light wavelength). In this case the slit is a kind of a secondary source with a limited angular divergence $\alpha = \lambda/d$. There is also a transition region, referred to as the Fresnel diffraction region with a center at the distance $z_d = d^2/2\lambda$, where the increase in the beam size due to the angular divergence αz is half of slit width d.

For synchrotron radiation (SR) with a photon energy E in the range from 5 to 50 keV, the situation is similar to that for visible light with the only difference: at reasonable distances the slit size should be much smaller than for visible light, and the beam should be coherent on the slit width, which is not easy to implement in laboratory X-ray sources. For example, z_d = 25 m at d = 50 μm and λ = 0.05 nm (E = 25 keV). In other words, for relatively large slits the wave properties of radiation manifest themselves at very long distances. Nevertheless, the wave properties of third-generation SR sources have been experimentally observed at diffraction from slits with sizes from 10 to 100 μm and used to measure the degree of SR coherence [2–4].

Divergent beams from two very narrow slits, located close to each other, overlap and form a simple interference pattern: an array of bright and dark fringes. In the visible light region this experiment was performed for the first time by Thomas Young in the beginning of the XIX century; the first such experiment with coherent SR was carried out in 2001 [5].

The fact that the interference of hard SR during its propagation in air is observed at relatively long distances is a certain inconvenience for experiments. At the same time, SR undergoes diffraction in single crystals, the lattice period of which only slightly exceeds the SR wavelength. Initially (in the first half of the XX century), the theory of SR diffraction in single crystals was developed for plane waves and crystals with planar input and output surfaces. Then, in 1961, the theory for a spherical wave was constructed using the Fourier transform. Later on, a theory was developed for the general case based on the solution of Takagi equations [6, 7].

A specific feature of two-wave X-ray diffraction of a spherical wave from a lattice is that the radiation from a secondary source (for example, a narrow slit) is distributed within a Borrmann fan (we will call it triangle) having an angle $2\theta_B$, where θ_B is the Bragg angle, found from the condition $2a\sin\theta_B = n\lambda$. Here, a is the interplanar spacing for the system of planes on which diffraction occurs and n is an integer (reflection order). The Bragg angle is relatively large for typical values of diffraction parameters: it may be 10° or even larger.

For this reason, two beams from small secondary sources overlap very rapidly and can form an interference pattern in a crystal not worse than when propagating in air. This fact was used when developing the theory of two-slit interferometer for the reflected beam in the case of diffraction in a single crystal. The calculation was performed using the influence function (propagator) of the crystal for the Takagi equations. The main result was the equation for the period of interference fringes [8].

It was shown later that an interference pattern of the same type can be obtained with one slit and two crystals separated by a thin air layer [9], as well as using a bilens interferometer [10] based on a compound refractive lens [11]. In this case, the interference fringe period is the same. It was shown in those studies that the operation of a crystal under conditions of diffraction from the lattice and in reflected beam is similar to that of air. The role of the distance after the scattering object is played by the crystalline plate thickness. In other words, an interference pattern can be obtained almost at a zero distance.

Coherent diffraction from a slit of finite sizes under conditions of diffraction from the atomic lattice in a single crystal was studied theoretically more than 50 years ago, both solving the Takagi equations [12] and using a propagator crystal [13, 14]. The solutions were formal, and the analogy with the diffraction from a slit in air was not analyzed in detail; in other words, it was only demonstrated that the problem has a solution. Experiments for such a system have not been performed until now for the following reasons. On the one hand, there are no coherent beams of necessary size. On the other hand, it is difficult to state an experiment without understanding properly its purpose and motivation. It is of interest that a method of numerical calculation of the Takagi equations was formulated for the first time in [12]; then this method was widely applied in numerous publications devoted to the analysis of images of various lattice defects. Recently this method was further developed to adopt it for crystals of arbitrary shape [15].

The main result of this study is a detailed analysis of the analogy between two media—air and crystal lattice (case of diffraction from a lattice in reflected beam)—for the relatively simple case of plane-wave diffraction from a slit of finite sizes. This analogy is of great practical importance, because many interference effects that are relatively difficult to observe when detecting in air can be used under plane-wave diffraction conditions in a crystal in reflected beam.

Nevertheless, the two aforementioned media do not coincide completely. The most significant differences between them are as follows. First, in the case of diffraction in a crystal, there arise two fields with different refractive indices, which leads to small extinction oscillations of intensity. We will not be interesting in this effect. It is absent in the case of diffraction from a slit in air, and is not interesting in a crystal in the case under consideration. The second difference leads to the fact that we will be interested in only the Fresnel diffraction region. The near-field region is trivial (although different), whereas in the far-field zone the crystal differs from air by absorption of radiation. Here, the key role is played by the Borrmann effect [6, 7], due to which radiation remains only in the region where the absorption is minimal.

All numerical calculations were performed using the XRWP program [16], developed by one of the authors to solve a wide circle of problems of X-ray optics, both with crystals and without them. The program is freely distributed on the Internet and has a detailed description for self-learning to work with it. All one has to do is correctly understand the problem conditions and be able to understand the calculation results.

STATEMENT OF THE PROBLEM AND METHOD FOR ITS SOLUTION

The possible experimental scheme considered in this study is partially presented in Fig. 1. The radiation from an SR source with small transverse sizes, located at a fairly long distance (is not shown), passes through monochromator (1), which does not change the spatial properties of the SR beam. Then it is limited by slit (2), after which crystal (3) is installed in the position of diffraction reflection. A symmetric case of Laue diffraction is assumed to take place. The surfaces of the crystalline plate make an angle θ_B with the direction of the SR beam incident on the slit. Detector (4) records the reflected radiation.

In the case of diffraction from a slit in air, a crystal is absent, and the detector records the beam transmitted through the slit at a relative long distance from the slit. It is assumed that the monochromator selects a fairly narrow line in the SR spectrum, and consideration of this spectrum does not change the calculation results for monochromatic radiation. However, this is not always true, because crystals are characterized by a fairly high sensitivity of SR diffraction to a change in the radiation wavelength. Nevertheless, it makes sense to take into account the spectrum when carrying out a detailed comparison of the calculation and experimental results.

Formally, high monochromaticity can be achieved using higher reflection orders in the monochromator as compared with the sample crystal. A record was the monochromatization based on diffraction in crystals to a spectral width of 5×10^{-7} keV, obtained at energy of 14.4 keV of the Mössbauer nuclear transition of the Fe^{57} isotope [17]. Monochromatization at the same level can be achieved for any photon energy.

The solution to the Maxwell equations for SR propagation in air in the paraxial approximation has the form of a convolution of the SR wave function with a Fresnel propagator. The convolution can be most rapidly and easily calculated by the method of double Fourier transform with application of the fast Fourier transform [18]. In the case under consideration diffraction occurs in the (x, z) plane, and the result is independent of the coordinate y. The z axis coincides with the beam propagation direction, and the x axis is perpendicular to the z axis.

The calculation was performed in three steps. First the Fourier transform of the wave function after the slit was calculated. Then the result was multiplied by the Fresnel propagator's Fourier transform (PFT):

$$P(q,z) = \exp(-i(\lambda z/4\pi)q^2), \tag{1}$$

where $z = z_2 - z_1$, $z_{1,2}$ are the initial and final distances in empty space. In our case $z_1 = 0$. Then the inverse Fourier transform was calculated.

The diffraction in crystal was calculated according to the same scheme, only the Fresnel PFT was replaced with the solution to the diffraction problem for plane waves at an arbitrary deviation of the wave vector direction from the exact direction, satisfying the Bragg condition. This solution was obtained even in the beginning of the XX century; it is described in detail in textbooks [6, 7]. As was shown in [9, 10], it is reasonable to consider at once the (2×2) matrix, because the wave function in a crystal has two components (for transmitted and reflected beams).

In this study we will consider only the transition from incident to reflected beam. Correspondingly, the PFT of the crystal for this process has the form

$$\begin{aligned} P_c(q,t_c) &= F(q)(X_h/2g) \\ &\times [\exp(i(A+G)) - \exp(i(A-G))], \end{aligned} \tag{2}$$

where

$$A = (X_0 + \alpha_q)t_c/2\gamma_0, \quad g = (\alpha_q^2 + X^2)^{1/2}, \tag{3}$$

$$\begin{aligned} G &= gt_c/2\gamma_0, \quad X = (X_h X_{-h})^{1/2}, \\ \alpha_q &= (q - q_0)\sin(2\theta_B), \end{aligned} \tag{4}$$

$$F(q) = \exp(-iqt_c \sin\theta_B). \tag{5}$$

Here, t_c is the crystal thickness; $X_{0,h,-h} = K\chi_{0,h,-h}$, $X_{0,h,-h}$ are the diffraction parameters, i.e., the Fourier components of the crystal polarizability on the reciprocal lattice vectors $\mathbf{0}$, $\mathbf{h}$, $-\mathbf{h}$; $\gamma_0 = \cos\theta_B$; $q_0 = K\theta_0$; θ_0 is the deviation angle of the crystal from the exact Bragg position; and $K = 2\pi/\lambda$ is the wave number. The function $F(q)$ is introduced to shift the origin of coordinates to the midpoint of the Borrmann triangle base. The equations are given for the axis oriented perpendicular to the reflected beam direction. For convenience of further presentation, we assume that $\mathrm{Re}(X) > 0$.

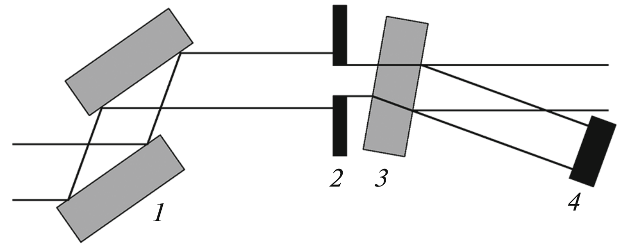

Fig. 1. Part of the experimental scheme (without a source): (*1*) monochromator, (*2*) slit, (*3*) single crystal, and (*4*) detector.

The crystal propagator is calculated through the inverse Fourier transform of the function $P_c(q, t_c)$ (2). It has an analytical form, being expressed in terms of a zero-order Bessel function [19, 20]. However, for the purposes stated, it is more convenient to analyze the analogy between the Fresnel and crystal PFTs. The calculation procedure is the same in both cases, the only distinction is the difference between Eqs. (1) and (2)–(5). Equation (1) contains an exponential, the argument of which is proportional to q^2. In Eqs. (2)–(5) this argument is also present (being symmetric at $q_0 = 0$) but in a limited range of q values, when $|\alpha_q| < \mathrm{Re}(X)$. If a plane wave is incident on a slit, $q = 0$ before the slit. After the slit a narrow range of integration over q arises in the integral; the wider the slit, the smaller this region is. In other words, small q values have priority for a wide slit.

It is of interest to compare the factors before q^2 in the argument of the exponentials in order to determine which parameter in the crystal PFT is analogous to the distance in the Fresnel PFT. A calculation shows that the role of distance in the crystal is played by the parameter

$$z_c = 2\sin^2\theta_B \cos\theta_B t_c/\mathrm{Re}(\chi), \quad \chi = X/K. \tag{6}$$

This equation is obtained from comparison of the factor at q^2 in the argument of the exponential in (1) and the factor at q^2 in the expansion of parameter G in (4) in a power-law series in q^2 at $q_0 = 0$. There is still a certain difference between the media, because the propagator in the crystal has two exponentials rather than one, as in air. Correspondingly, for a plane wave (a very wide slit) the squared modulus of the function $P_c(q, t_c)$ (2) is proportional to the factor $\sin^2(G)$, which oscillates with an increase in thickness t_c with a period $\Lambda_c = \lambda\cos\theta_B/\mathrm{Re}(\chi)$.

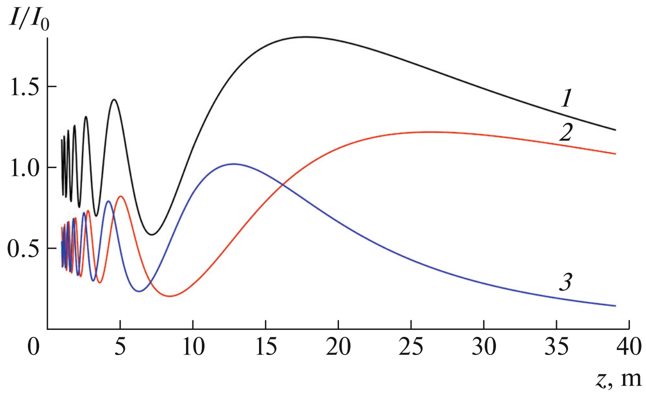

Fig. 2. Relative intensity of the SR beam at the slit center in dependence of the distance to the detector (*1*) and the contributions to this intensity from the cosine (*2*) and sine (*3*) parts for $d = 50$ μm and photon energy of 25 keV.

CALCULATION RESULTS AND THEIR ANALYSIS

The equations of the previous section were written for a plane incident wave. It is known that, for a spherical wave emitted by a point source located at a distance z_0 from the slit, the result of calculating the phase contrast [21] will be the same as for a plane wave at the distance $z = z_0z_1/z_t$, where $z_t = z_0 + z_1$, z_1 is the distance from the slit to the detector; the only difference is that the pattern will have a width enlarged by the factor z_t/z_0. It can be seen that, under the condition $z_0 >> z_1$, the parameter z differs only slightly from z_1. The same situation occurs in the case of diffraction from a slit in air, because the aforementioned equations correspond to geometric optics. For this reason it is sufficient to perform a calculation for an incident plane wave.

The diffraction in crystals will be analyzed based on the calculation results for an incident plane wave, specifically, provided that $z_0 >> z_1$, where the distance z_1 is calculated from the crystal thickness according to formula (6). Practically the same case can be implemented using a compound refractive lens [22]. The diffraction focusing effect [23], which has no analogues in air, is implemented for a spherical wave in a crystal. At some combination of parameters this effect may spoil the analogy between crystal and air in the case of diffraction from a relatively large slit.

The effect of diffraction of a plane light wave from a slit is considered in detail in all textbooks on optics. Nevertheless, the full-scale numerical simulation has not been discussed until now, whereas it must be done to perform a detailed comparison. The equations for SR are similar to those for visible light. The dependence of the SR wave function $\psi(x,z)$ on x is found as the integral of the Fresnel propagator $P(x - x_1, z)$ over a finite range of the variable x_1. This dependence is symmetric, and the most interesting result is the intensity at the point $x = 0$ in the dependence on z. In this case the SR wave function has the form

$$y(0,z) = (2/i)^{1/2}[C(r) + iS(r)],$$
$$r = (z_d/z)^{1/2}, \quad z_d = d^2/2\lambda, \tag{7}$$

where $C(r)$ and $S(r)$ are the Fresnel cosine and sine integrals, i.e., the integrals over the variable s from 0 to r for the functions $\cos(\pi s^2/2)$ and $\sin(\pi s^2/2)$.

The relative intensity $I_R(0, z)/I_0$ is the sum of the squares of C and S, multiplied by 2. At $z = 0$ it is unity; with an increase in z it first oscillates with small step and amplitude. Gradually the oscillation step increases; the amplitude increases to maximum and then monotonically tends to zero. The parameter z_d is referred to as the diffraction length; it corresponds to the point of maximum for the contribution from the Fresnel cosine integral; the maximum intensity is obtained approximately at $z_m = 0.7z_d$.

The function $I_R(0, z)/I_0$ is presented in Fig. 2 (curve *1*). Curves *2* and *3* show the contributions to this function from the Fresnel cosine and sine integrals. The calculation was performed for the slit width $d = 50$ μm and photon energy 25 keV ($\lambda = 0.0496$ nm). In this case $z_d = 25.2$ m and $z_m = 17.6$ m. Since the integral intensity over x is independent of z, the increase in the maximum is accompanied by compression of the SR beam. This fact is demonstrated in Fig. 3, where the total dependence $I_R(x, z)/I_0$ in the range of distances from 0 to 40 m is shown. One can see in this figure that the beam begins to expand almost immediately, i.e., at small distances from the slit, but the intensity beyond the slit is very weak.

The near-field zone, where the beam changes weakly (according to the geometric optics), has relatively small sizes. Strong interference occurs in the Fresnel diffraction region; it is caused by the slit edges, where space homogeneity modes change sharply. The most interesting is the effect of beam compression with an increase in intensity at $z = z_m$. The slit works as a weakly focusing optical device. The computer calculation presented in Figs. 3–5, was performed using the XRWP program. All calculation details can be found on the program website [16], including theoretical equations and examples of solving various problems. The calculation was performed using a point grid with a step of 0.1 μm and the number of points $2^{15} = 32\,768$. Figure 3 was plotted based on a 401 × 401 matrix, and the calculation time was several seconds.

Figure 4 presents a similar intensity distribution but for the case where directly after the slit the SR beam enters a silicon single crystal installed in the exact Bragg position for the 220 reflection. Therefore, instead of the distance z over the vertical crystal axis, the thickness t_c is shown. The diffraction parameters were calculated using the on-line program [24]. In this case, $\theta_B = 7.42°$, $\mathrm{Re}(\chi) = 9.34 \times 10^{-7}$, and $\Lambda_c = 52.7$ μm. Because of the extinction effect the pattern is

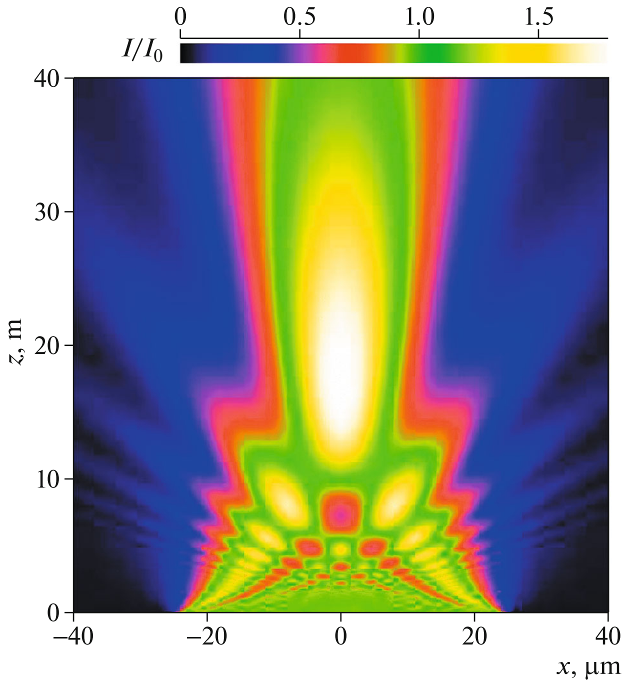

Fig. 3. Dependence on x for the relative SR beam intensity in the region of distances corresponding to the Fresnel diffraction at $d = 50$ μm and photon energy of 25 keV.

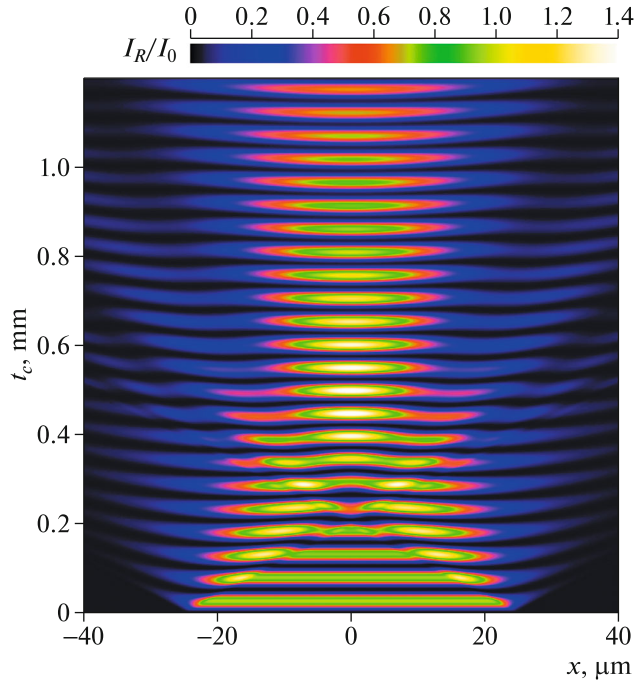

Fig. 4. Dependence on x for the relative intensity of reflected (220) SR beam at diffraction in a silicon single crystal for the range of thicknesses corresponding to the Fresnel diffraction at $d = 50$ μm and photon energy of 25 keV.

a set of fringes. However, tracing the intensity distribution in the maxima of extinction beatings, one can note some analogy with the diffraction from a slit in air, which is shown in Fig. 3. The figure is based on a matrix with a number of points 801 × 801. The calculation time, correspondingly, increased to several minutes.

The most complete correspondence between the single crystal thickness t_c and the distance in air z is observed in the region of the main maximum. According to (6), for the given case, this correspondence is determined by the formula $t_c = 2.82 \times 10^{-5} z$. For $z = 17.6$ m we obtain the value $t_c = 0.496$ mm, which is in complete correspondence with the calculation result. The maximum t_c value is, however, somewhat smaller because of the SR absorption in the single crystal. In the region of small thicknesses the analogy is incomplete because, in the case of single crystal, the changes related to the space homogeneity jump at the slit edges propagate in the Borrmann triangle with the angle $2\theta_B$. While these triangles do not intersect, there is no interference, and the slit influence manifests itself in no way. In the case considered above they intersect specifically before the main maximum.

Figure 5 shows the calculation result for a slit with a size $d = 100$ μm. The parameter z_m is proportional to d^2; it increased by a factor of 4 in comparison with the previous version. The single crystal thickness at which Borrmann triangles intersect only doubled. Therefore, one can observe not only the main maximum but also several maxima preceding it in Fig. 3. Here, the pattern is based on a 901 × 901 matrix. Note that the central maximum does not correspond to the highest relative intensity, which is observed at smaller thicknesses. The reason is the same: SR absorption in the crystal. The reduction of intensity caused by normal absorption for $t_c = 2$ mm is $\exp(-\mu_0 t_c) = 0.367$. In fact, the value in the maximum is somewhat larger, because some part of radiation is absorbed weakly due to the Borrmann effect.

For a slit with a size of 25 μm or smaller, vice versa, even the main maximum does not fall in the overlap region of Borrmann triangles with vertices at the slit edges, and the beginning of the far-field zone and SR beam expansion are observed immediately in the interference region. In other words, the slit operates as a secondary source, and the calculation results are close to those obtained by Kato even in 1961 [25].

The calculation results for other photon energies show distributions fairly close to those presented in Figs. 4 and 5 but for other crystal thicknesses. The point is that the proportionality factor between the distance z and crystal thickness t_c in formula (6) depend weakly on energy, because $\sin\theta_B$ is proportional to λ and $\mathrm{Re}(\chi)$ is proportional to λ^2. However, the diffraction length, i.e., the distance corresponding to the center of the Fresnel diffraction region, is

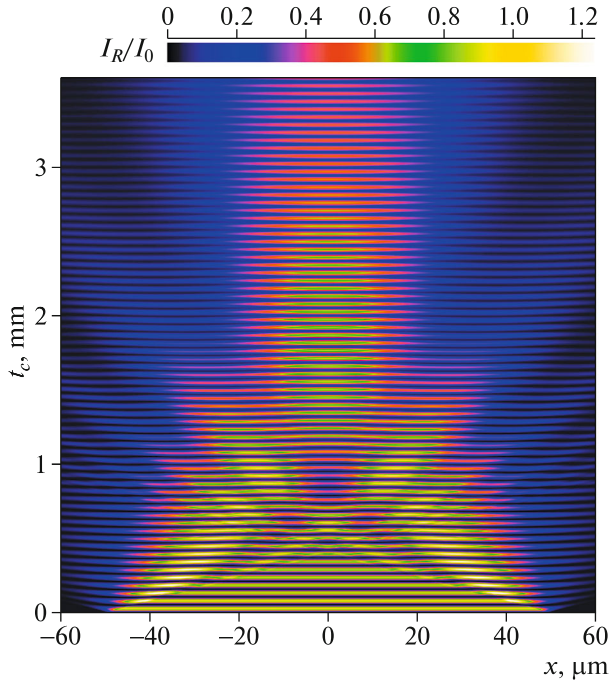

Fig. 5. Dependence on x for the relative intensity of reflected (220) SR beam at diffraction in a silicon single crystal for the range of thicknesses corresponding to the Fresnel diffraction at $d = 100$ μm and photon energy of 25 keV.

inversely proportional to λ and decreases with decreasing photon energy. The absorption in crystal, vice versa, increases under these conditions. However, its role is not very important because of the Borrmann effect.

Until now, it was assumed that the crystal is installed in the exact angular position for the diffraction from the lattice. Naturally, a question arises about the accuracy with which this angular position must be specified in order not to violate the correspondence under consideration. An analytical answer to this question has not yet been obtained, but the answer can be found by numerical simulation. Figure 6 shows three curves for a photon energy of 25 keV, silicon crystal thickness of 500 μm, and (220) reflection. Curve *1* corresponds to Fig. 4 for the aforementioned thickness, and curves *2*, *3* were obtained using crystal rotation by an angle 5×10^{-7} rad relative to the exact angle, satisfying the Bragg condition, to different sides. The curves become asymmetric and are specularly reflected with a change in the rotation angle sign.

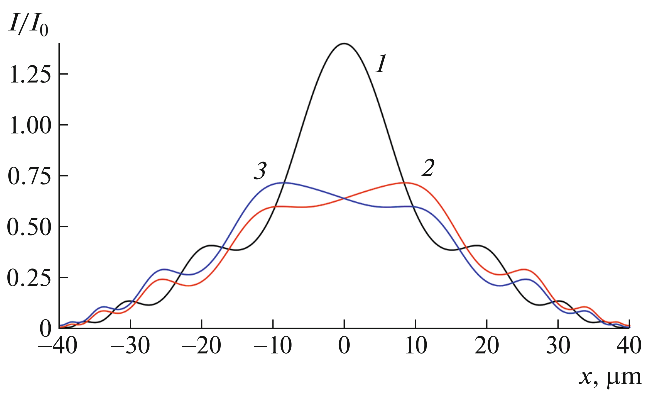

Fig. 6. Dependence on x for the relative intensity of reflected (220) SR beam at diffraction in a 500-μm thick silicon single crystal at $d = 50$ μm and photon energy of 25 keV; (*1*) exact angular position, (*2*, *3*) crystal rotated from the exact position by an angle of 5×10^{-7} rad to different sides.

As follows from the calculations, correct diffraction occurs in a very narrow angular range, which decreases with an increase in the photon energy. This high sensitivity to the angular position of crystal is generally characteristic of the Laue diffraction in relatively thick crystals.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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