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# Calculation of class-$b$ mosaic crystals reflectivity by Monte Carlo technique

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**Summary.** — Mosaic crystals of $b$-type, for example, pyrolytic graphite, is widely used in experimental physics as neutron and X-ray monochromators and provide a higher yield of monochromatic radiation than perfect ones. The technique is proposed and implemented to calculate the reflectivity of such crystals by Monte Carlo modeling, correctly considering the multiple reflections of photons inside the crystal and the geometry of experiment for random distribution of the mosaic.

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PACS 61.50.-f – Structure of bulk crystals.

## 1. – Introduction

The diffraction of electromagnetic radiation with continuous or linear spectrum in crystals is used to obtain the beams of monochromatic radiation in many areas of science and technology. It is well known (see, for example, [1]) that the integral reflectivity of the mosaic crystals is significantly higher than that of perfect ones. By the degree of perfection crystals can be classified according to two criteria: the dimensions of the regular blocks or regions in the crystal, and the degree of their mutual misalignment [1]. According to the first criterion all crystals can be divided into two types, $a$ and $b$. In $a$-type crystals separate regions are large enough for the considerable influence of the primary extinction to be manifested, *i.e.* their linear dimension is comparable with the length of the primary extinction $l_{\mathrm{ex}}$. In $b$-type crystals the dimensions of the regular block are small; therefore, the effect of primary extinction is practically not observed. According to the second criterion, crystals can also be divided into two groups, $\alpha$ and $\beta$. In $\alpha$-group crystals the blocks are almost parallel to each other, their mutual disorientation is small;

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therefore, the contribution of the secondary extinction is high. In $\beta$-group crystals the blocks are distributed irregularly; therefore, the contribution of this effect is small.

The ability of the crystals to reflect X-ray radiation is related to the perfection of their structure. The crystals of $a\alpha$-group provide the narrowest rocking curve (FWHM $\sim$ 20–30$''$), and their integral reflectivity is low. The mosaic crystals of $b\beta$-group have the maximum integral reflectivity. The most famous mosaic crystal of $b$-type, used in the applied physics, is pyrolytic graphite. Such crystals are used to obtain quasi-monochromatic X-ray beam [2] and neutrons [3].

As noted in a number of experimental works, see, for example, [2] and references therein, the theory of X-ray diffraction in mosaic crystals [4,5] does not always accurately describe measurement results even with the crystals of pyrolytic graphite. The simulation allows to take into account more accurately all the experimental factors: the dimension and geometry of the crystals, the actual distribution of sample mosaic, the presence of multiple reflections inside the crystal and the corresponding change of the absorption and a number of others.

## 2. – Simulation

Our approach is also based on the theory of diffraction of X-ray radiation in mosaic crystals [1,4,6] and has already been partially used in [7]. The calculation procedure of the reflectivity of $b$-type mosaic crystals [4,6] is obtained for one-dimensional distribution of mosaic and monodirectional beam of the external radiation with a fixed energy. In the general case a divergent photon beam with a continuous spectrum is incident on the crystal or is born in it. For the reflection of monodirectional and monoenergetic photon beam from the element of mosaic crystal with volume $\Delta V$ it can be written [1]

$$\int P(\theta)\mathrm{d}\theta = Q\Delta V, \tag{1}$$

where $P(\theta)$ is the reflectivity of the crystal element at the angle $\Theta$, that is proportional to the distribution of mosaic blocks in the crystal [6]. $Q\Delta V$ is the integral reflection from the element $\Delta V$, where $Q$ is the integral reflectivity depending on the parameters of the crystal and the radiation energy as follows:

$$Q = \left(\frac{e^2}{mc^2}\right)^2 \frac{N^2\lambda^3}{\sin(2\Theta)} \left|F_p\right| \left|F(\vec{g}\,)\right|^2. \tag{2}$$

Here, $N$ is the concentration of dispersing centers, $\lambda$ is the radiation wavelength, $\Theta$ is the rotation angle of the crystal plane relative to the direction of photon beam. $|F(\vec{g}\,)|^2 = |S(\vec{g}\,)|^2 \cdot |f(\vec{g}\,)|^2 \cdot \exp[-2W]$, where $S|(\vec{g}\,)|^2$ is the structural factor, $\exp[-2W]$ is the Debye-Waller factor, $f(\vec{g}\,)$ is the Fourier component of the spatial distribution of electrons in the crystal atom ($f(0) = z$, where $z$ is the number of electrons in the atom). $|F_p|$ is the polarization factor, which depends on the polarization of the radiation incident on the crystal.

For the analysis of the reflection process, an approach with the introduction of several systems of coordinates related to the direction of the primary photon beam (laboratory system) and the direction of crystal plane (crystal system) is used, which was proposed in [8]. Suppose that in a mosaic crystal with the distribution of inverse lattice vectors $P(\vec{g}\,)$ a photon beam with the spectral-angular distribution of $N_\gamma(\omega, \vec{n}\,)$ spreads, where

$\omega$ and $\vec{n}$ are the energy and the unit vector along the photon pulse, respectively. Here $\vec{g} = |\vec{g}\,|\vec{\alpha}$, where $\vec{\alpha}$ is the unit vector describing the deviation of bulks from the middle direction $\vec{g}_0 = \langle\vec{g}\rangle$. The vector $\vec{g}_0$ is perpendicular to the plane of the crystal and rotated by an angle $\pi/2 - \Theta_B$ relating to the $z$-axis. Plane diffraction is determined by the vectors $\vec{n}_0$ and $\vec{g}$. On the basis of Bragg's law for the photon energy of $\omega$ and direction of $\vec{n}$ one can write the condition for the direction of the vector $\vec{g}$ of the bulk, on which this photon may diffract:

$$\omega = \frac{\vec{g}\,\vec{n}}{\sqrt{\varepsilon_0}(1 - \vec{n}'\,\vec{n})} = \frac{|\vec{g}\,|\sin\Theta}{\sqrt{\varepsilon_0}(1 - \cos 2\Theta)}\,, \tag{3}$$

where $\vec{n}'$ is the vector describing the moving direction of the diffracted photon and $\varepsilon_0$ is the dielectric permittivity of the medium. Here and further, the system of units $\hbar = m_e = c = 1$ is used. Hence, the angle between the vectors $\vec{n}$ and $\vec{g}$ must meet the condition

$$\sin\Theta = \frac{|\vec{g}\,|}{2\omega\sqrt{\varepsilon_0}}\,. \tag{4}$$

In a mosaic crystal, this condition is conformed with a set of mosaic blocks satisfying the equation

$$\sin\Theta = \frac{(\vec{g}\,\vec{n}\,)}{|\vec{g}\,|} = \frac{[n_x g_x + n_y g_y + n_z g_z]}{|\vec{g}\,|}\,. \tag{5}$$

By this equation one can determine the mosaic blocks, on which this photon can diffract, and determine the moving direction of the reflected photon $\vec{n}'$. Then the reflection probability density of the photon with fixed $\omega$ and $\vec{n}$ in a mosaic crystal with thickness $\Delta t$ can be written as

$$f(\omega, \vec{n}\,) = q(\omega, \vec{n}\,)Q(\omega)\Delta t, \tag{6}$$

where $q(\omega, \vec{n}\,)$ is the coefficient considering the crystal mosaic

$$q(\omega, \vec{n}\,) = \int P_m(\alpha_x(\omega, \vec{n}, \alpha_y), \alpha_y)\mathrm{d}\alpha_y. \tag{7}$$

Here, $P_m$ $(\alpha_x,\, \alpha_y)$ is the crystal mosaic distribution, expressed in $\omega, \vec{n}, \alpha_y$ according to expressions (2)–(5). In accordance with the approach [7], the diffracted photons yield in the aperture of the collimator for each order of reflection is determined by $i$ folding of the spectral-angular distribution of radiation intensity with reflection probability density for all variables, including energy and angles of the start of photons and the crystal thickness. Secondary diffraction of the reflected photons in the direction of the primary beam on the path from the diffraction region to the outlet from the crystal is taken into account similarly. The absorption of photons is determined by the length of the way passed by them in the crystal, and their energy.

This technique allows taking into account the spectral and angular distribution of radiation and actual distribution of the sample mosaic. The main disadvantage is that it is difficult to consider the real geometry of measurements (the rotation of the crystal, the

possible differences of the mosaic distribution of the sample from a Gaussian one, etc.) and uncontrolled change of the photon path length in the crystal and the absorption in it due to multiple reflections. Considering (6), (7), the changes in the number of photons due to the diffraction in the crystal after passing through a layer of thickness $\Delta t$ can be written as

$$\Delta N_\gamma(\omega, \vec{n}\,) = -N_\gamma \cdot q(\omega, \vec{n}\,)Q(\omega)\Delta t = -N_\gamma \cdot \mu_{\text{dif}}(\omega, \vec{n}, \vec{g}\,)\Delta t, \tag{8}$$

where $\mu_{\text{dif}}(\omega, \vec{n}, \vec{g}\,)$ is the linear radiation absorption coefficient with the energy $\omega$ and the moving direction $\vec{n}$ due to the diffraction in a mosaic crystal. In view of (8) the dependence of the photon number of the thickness of the traversed layer of the crystal $T$ can be written in the traditional form

$$N_\gamma(\omega, \vec{n}, t) = N_0(\omega, \vec{n}\,) \exp[-\mu_{\text{tot}} t], \tag{9}$$

where $N_0(\omega, \vec{n}\,)$ is the spectral and angular distribution of radiation incident on the crystal, and $\mu_{\text{tot}} = \mu_{\text{dif}}(\omega, \vec{n}, \vec{g}\,) + \mu_{\text{ph}}(\omega) + \mu_{\text{inc}}(\omega) + \mu_{\text{coh}}(\omega) + \mu_{\text{pair}}(\omega)$ is the complete linear factor of the absorption of primary radiation resulting from the process of diffraction, $(\mu_{\text{dif}}(\omega, \vec{n}, \vec{g}\,))$ photo absorption $(\mu_{\text{ph}})$, incoherent (Compton) dispersion $(\mu_{\text{inc}})$, coherent dispersion $(\mu_{\text{coh}})$ and electron-positron pairs initiation $(\mu_{\text{pair}})$. Such form of writing allows using a well-known method of statistical modeling (Monte Carlo method) to describe the passing of photons through the mosaic crystal, see, for example, [9].

We present the main steps and the approximations used in the simulation by the example of determining the energy resolution and efficiency of crystal diffraction spectrometers based on mosaic crystals of pyrolytic graphite used in the experiment [10]. A beam of bremsstrahlung from a disoriented tungsten target is incident on the crystal mounted in a goniometer and rotated at the angle $\Theta_B = \Theta_D/2$. Here $\Theta_D$ is the angle at which the detector for the diffracted radiation is located. Calculation method for the spectral-angular distribution of bremsstrahlung, the characteristics of the used crystals and the scheme of the experiments location are similar to those given in [7].

For the photon with energy $\omega$ and a wave vector $\vec{k} = \omega\vec{n}\sqrt{\varepsilon_0}$ the point of hitting the crystal was defined. Taking into account the measured distribution of crystal mosaic the bulk disorientation angle was simulated with respect to the $y$-axis in the crystal systems of coordinates $\alpha_y$. Based on the values of $\omega$, $\vec{k}$, $\Theta_B$ in accordance with expressions (4), (5) the angle of bulks disorientation was determined, on which the photon with $\omega$ and $\vec{k}$ can diffract, relating to the axis $x$, $\alpha_x$. Further, based on the measured distribution of mosaic relating to the $x$-axis we determined the probability of the existence of such block in the crystal, $w(\alpha_x)$ and the linear coefficient of the primary photons absorption due to diffraction, $\mu_{\text{dif}} = wQ$. For Gaussian distribution of bulks the desired probability is equal to

$$w = \frac{1}{\sqrt{2\pi}\sigma} \exp[-\alpha_x^2/2\sigma^2], \tag{10}$$

where $\sigma$ is the characteristic angle of the mosaic of the crystal used in a horizontal plane. Then, in accordance with the traditional approach of simulation the passing of photons through the matter [9] we simulated the photon passing to the point of interaction $l = \ln\xi/\mu_{\text{tot}}$, where $\xi$ is the random number between zero and one, indicated a point of interaction and simulated what process occurred: diffraction, photoabsorption, Compton

(incoherent) dispersion or coherent dispersion on a separate atom. For simulation the values of the cross sections of interaction of low energy photons given in [11] were used. If the point of interaction did not belong to the crystal the photon hitting the detector was verified and the drawing started again.

Further simulation depended on the type of interaction occurred. If there was a process of photoabsorption, the simulation began anew, and if photon was dispersed, then using the known methods of modeling of interaction of photons with matter [9] the energy of the dispersed photon $\omega'$ and its moving direction $\vec{n}'$ were determined. Further, again the disorientation angle of bulks $\alpha_y$ was simulated, angle $\alpha_x$ was determined, the free path length and the type of interaction were simulated. If the diffraction occured, in accordance with the law of pulse conservation it can be written

$$\vec{k} = \vec{k}' + \vec{g}. \tag{11}$$

Here $\vec{k}'$ is the wave vector of the diffracted photon, $\vec{g}$ is the vector of the inverse lattice of crystal bulks, on which the diffraction occurred. All three vectors are defined in the laboratory system of coordinates. Therefore, from the crystal system of coordinates, where the vector $\vec{g}$ is described by the angles of bulks disorientation $\alpha_x$ and $\alpha_y$, we should move to axis laboratory frame by the method given in [8]. Then starting from $\omega$ and vectors $\vec{g}$, $\vec{n}$ the direction of the diffracted photon was determined

$$\vec{n}' = \vec{n} - \frac{\vec{g}}{\omega\sqrt{\varepsilon_0}}\,. \tag{12}$$

For the photon with energy $\omega$ and moving direction $\vec{n}'$ the bulks disorientation angle $\alpha_y$ was simulated again, the angle $\alpha_x$ was determined and the whole process of drawing the free path length, determination of the point of interaction coordinates, testing the condition of starting from the crystal and interaction process repeated. The main difference of the second and all subsequent even-numbered reflections is that the photon is reflected from the opposite side of the plane. Therefore, for such reflection the crystal system of coordinates was centered around the axis $y$ to 180°. The history of each photon was traced until its absorption in the crystal, or starting from it with a check on hitting the detector.

The proposed method has no restriction to the thickness and geometry of the used crystal, which may consist of several samples, the angular distribution of mosaic blocks and so forth. The main conditions for the applicability of the technique: the used crystal must be a crystal of *b*-type, and the reliable information on the two-dimensional angular distribution of mosaic blocks in it must be available.

The diffraction of low energy neutrons in crystals is described by analogy with the X-ray diffraction in mosaic and perfect crystals [12]. In accordance with the cited work the integral reflection of neutrons from a small volume element $\Delta V$ is equal to $Q_n \Delta V$, where the value of $Q_n$ can be written as follows:

$$Q_n = \frac{\sigma_{\text{Bragg}}}{4\pi} \frac{N^2 \lambda_n^3}{\sin(2\Theta)} |S(\vec{g})|^2 \cdot \exp[-2W], \tag{13}$$

where $\lambda_n$ is the de Broglie wavelength, $\sigma_{\text{Bragg}}$ is the cross-section of neutron dispersion on the atoms of the crystal. The other symbols are the same as mentioned above. Such analogy allows to use the proposed method for calculating the reflectivity of mosaic

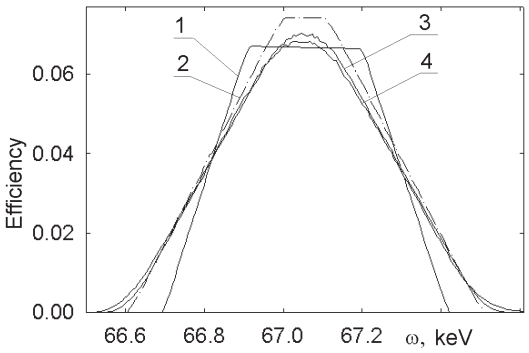


Fig. 1. – Calculated efficiency of the spectrometer for $\Theta_B = 1.58°$.

crystals of *b*-type for neutron beams. This will allow to consider more accurately the absorption of neutrons in a crystal, to increase the energy range in which one can calculate the reflectivity, and to get rid of the correction factor $\sim 0.8$, used in most studies on neutron diffraction to account for the absorption contribution and "residual" elastic dispersion, see, for example, [3].

## 3. – Simulation results

As noted above, the development of the proposed method of calculation of the *b*-type mosaic crystals reflectivity of X-rays was initiated by the need for processing of data on research of PXR characteristics at small angles to the direction of fast electrons in the tungsten crystal, obtained in the experiment [10]. Because of the narrow spectral range of the effect manifestation the most important parameters are the energy resolution and the efficiency crystal diffraction spectrometers used in the experiment. The calculation results of the absolute values of radiation yield using the proposed method of calculating the characteristics of crystal diffraction spectrometer for the experimental conditions [10] coincide with the measurement results with an error less than 5% [13].

To isolate the radiation with a fixed energy in the cited work one used a two-crystal diffraction spectrometer based on mosaic crystals of pyrolytic graphite with dimensions $2.5 \times 6.5 \times 22.5\,\mathrm{mm}^3$ and $3.5 \times 5.5 \times 20\,\mathrm{mm}^3$. Distribution of graphite crystals mosaic was determined during measuring of the diffraction curve and a peak of diffraction for each of the detector angles in the experiment [7]. In a thinner crystal it can be represented as a sum of two Gaussian distributions with parameters $\sigma_m^1 = 4.2 \pm 0.1\,\mathrm{mrad}$, $S_1 \sim 0.67 \pm .05$ and $\sigma_m^2 = 9.0 \pm 0.5\,\mathrm{mrad}$ $S_2 \sim 0.33 \pm 0.05$, where $\sigma$ and $S$ are the standard deviation and the weighting factor, respectively.

Figure 1 shows the calculation results of the efficiency of the spectrometer for the energy of photons of the first-order reflection $\omega \approx 67\,\mathrm{keV}$ and for the following conditions: the angle of collimation of the reflected radiation in the diffraction plane is $\Delta\Theta_x = 0.42\,\mathrm{mrad}$, the angular capture in the diffraction plane is $\Delta\theta_x = \pm 0.092\,\mathrm{mrad}$, the acceptance is $\Delta\theta_x \Delta\theta_y = 1.84 \cdot 10^{-7}\,\mathrm{sr}$. A crystal with dimensions $2.5 \times 6.5 \times 22\,\mathrm{mm}^3$ was used. Primary spectrum was generated by the electrons with energy of 500 MeV in the amorphous target of 0.5 mm thick.

Dependences 1,2 were calculated by the method [7] for a point electron beam. Curve 1 is obtained using only the angular dimensions of the primary radiation beam and the

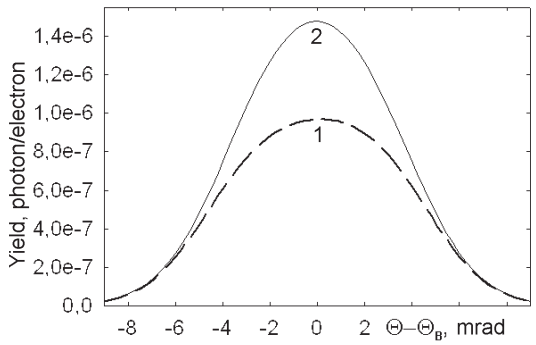


Fig. 2. – Orientation dependence of the radiation yield for $\Theta_D = 7.49°$.

angular capture of the detector of the diffracted radiation, that is, without taking into account the coordinates of the photon hitting the analyzing crystal. For the photon moving not along the axis of the experimental setup, the detector is located at an angle different from $\Theta_D = 2 \cdot \Theta_B$, that leads to changes of spectral dependence of the reflection efficiency (see curve 2). The simulation results, taking into account all known experimental factors (curves 3 and 4), differ slightly from those obtained by the method of [7]. Consideration of the crystal rotation, the point of diffracted photons starting from the crystal and multiple reflections leads to some deterioration of the resolution and decrease of the efficiency of reflection. However, the width (FWHM) remains virtually unchanged. The dependence 3, as well as the 1, 2, is calculated for a point beam. Consideration of the spatial distribution of the electron beam hitting the inner target of synchrotron [14], (curve 4) showed that this factor has little effect on the spectrometer characteristics.

As can be seen from the figure, the difference between the dependences obtained using different approaches is not very large. In accordance with (2) the decrease of the photon energy leads to the increase of reflectivity, which increases the probability of multiple reflections. For example, if for the photons energy $\omega = 67\,\text{keV}$ the fraction of photons that have undergone single, double and triple reflections in the crystal is equal to 0.075, 0.004 and $6 \cdot 10^{-5}$, then for $\omega = 28.3\,\text{keV}$, these values increased to 0.29, 0.08 and 0.007. The maximum number of reflections changed from 5 to 6. For $\omega = 28.3\,\text{keV}$ the distribution width (FWHM), obtained by simulation, is approximately 20% higher than that obtained by [7].

This effect influences the results of measuring the characteristics of mosaic crystals and, in some cases, can lead to uncontrollable errors. Figure 2 shows the calculated orientation dependences of the diffracted radiation yield received during the rotation of the graphite crystal for a fixed angle of the detector $\Theta_D = 7.49°$ ($\omega = 28.3\,\text{keV}$). The conditions coincide with the experimental conditions [10] for this photons energy, except for a crystal mosaic. The calculation was performed for Gaussian distribution of mosaic with $\sigma_m = 3\,\text{mrad}$, instead of the sum of two distributions with different $\sigma_m$.

Curve 1 with $\sigma = 3.38\,\text{mrad}$ and $\Delta\Theta = 9.37\,\text{mrad}$ is the result of simulation for the conditions [10] and the selected value $\sigma_m$. Here $\sigma$ is the standard deviation, and $\Delta\Theta$ is the width at half height. The dependence 2 with $\sigma = 3.15\,\text{mrad}$ and $\Delta\Theta = 8.65\,\text{mrad}$ was obtained at "programmed" break of the second and subsequent reflections. The difference between $\sigma$ and $\sigma_m$ is caused, apparently, by the finiteness of the angular dimensions of radiation beam incident on the crystal and the collimation angle of the radiation. From

the figure it is seen that multiple reflections greatly distort the observed dependence in comparison with the model one and with that calculated without taking into account the subsequent reflections. It is not like a Gaussian, and its width is approximately 10% larger. Consequently, in the case of crystals with high reflectivity the same error can also occur during measurements of the mosaic distribution. This effect is important for using of mosaic crystals for neutron diffraction, where the formula for the reflectivity contains the mosaic distribution width at half height, see, for example, [3].

## 4. – Conclusion

The research results can be formulated as follows:

1) A method for calculating the reflectivity of the mosaic crystal of *b*-type by Monte Carlo modeling was proposed and implemented, that allows to correctly account for the multiple reflections of photons inside the crystal and the geometry of the experiment for any distribution of the mosaic.
2) With slight modifications it can also be used to calculate the neutron reflection by such crystals that can increase the energy range in which one can calculate the reflectivity, and get rid of correction factors.
3) In studying of the characteristics of the crystals with high reflectivity for a selected range of photon energies the multiple reflections can significantly distort the measured dependence and lead to errors in measurement parameters.

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