Estimation of grains size in mosaic crystals by means of radiation of fast electrons in these samples

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Abstract. There were analyzed the possibilities of the previously suggested method for determining the characteristic dimensions of grains in mosaic crystals of diffraction suppression of yield of the bremsstrahlung with the fixed energy and calculated results. There were defined the bounds of the method applicability and also there were taken into account more accurately the effect of the absorption process of X-rays on the reflectivity.

1. Introduction
Ordered arrangement of atoms in condensed matter leads to the appearance of orientation and interference effects in the yield of the secondary processes that occur while passing through it fast charged particles. The existence of connection between the internal structure of the target and the yield of the secondary processes allows us to raise a question about the analysis of the internal structure of the target in accordance with the results of the measurements. For example, by output of backscattered channeled ions we could see the location of impurities in the crystal lattice, and by emission spectra of fast channeled electrons we could specify the form of potential, electron density, the amplitude of thermal vibrations of lattice atoms and the like, see, for example, [1, 2] and references there in.

In the same row there is the task of analyzing the quality of the internal structure of crystalline samples, that is, determining the presence of outside inclusions and the mosaic blocks in them, also the distribution of the grains along the angle of misalignment regarding to the basic direction and the sizes according to the characteristics of hard electromagnetic radiation generated by passing through them fast electrons. The advantages of this approach include the high penetration ability of such radiation and clarity of interpretation. In the clearest form these benefits are realized in the case of analysis of the microstructure of the thick samples and the task of estimation of the characteristic dimensions of the blocks in which the use of traditional X-ray analysis can not be provided the quality control of internal structure.

It is well known (see [3] and references therein) that according to the degree of perfection, crystals can be classified by two factors: by the size of regular blocks or sections in the crystal, and by the extent of their mutual disorientation. According to the first factor all crystals can be divided into two classes $a$ and $b$. In crystals of class $a$ the separate sections are large enough to demonstrated a marked influence of the effect of primary extinction, that is, their linear size is comparable to the primary extinction length $l_{ex}$. In crystals of class $b$ the size of right units is small, so the effect of primary extinction is almost not observed. According to the second factor the crystals can also be divided into two classes - $\alpha$ and $\beta$. In crystals of class $\alpha$ the blocs...
are almost parallel to each other, their mutual disorientation is small, so the contribution of a secondary extinction effect is large. In crystals of class $\beta$ blocks are distributed irregularly, so that the contribution of the secondary extinction effect is small. The limit of the class $a\alpha$, in case when disorientation of grains is less than the region of the total reflection of X-rays $\Delta \Theta$ (Darwin “table”), is the perfect crystal, when the class $b\beta$ is the ideal mosaic crystal.

The most challenging of the above mentioned is the identification of common block sizes, as from them it often depends the possibilities of use of crystals for applied purposes. In particular, to extract beams from accelerators of high energy we need perfect crystals or, in extreme cases, the crystals of class $a\alpha$, and as for the use in medicine, also for X-ray and gamma-ray astronomy and for the obtaining of intensive quasi-monochromatic neutron beams we are required mosaic crystals of class $b\alpha$. It should be noted that belonging of crystal to a particular class is not given once and for all, because it is determined by $l_{ex}$, which, in turn, depends on the order of reflection and the photon energy [3]. That is, for different orders of reflection or significantly different photon energies one and the same sample may relate to different classes [4].

The direct measurement of the size of grains using X-ray radiation beams is a difficult experimental problem, and it can be only implemented for the analysis of surface layers [5]. Methods of electron microscopy can resolve this problem as applied to thin polycrystalline and crystalline samples, in the case when the angles of disorientation of the nearby blocks and their sizes are more than divergence and linear dimensions of electron beam on the target. If you can’t get thin crystalline structure without disrupting the target as, for example, in case with ductile metallic crystals, the use of electronic microscopy cannot provide the required information.

When using fast electrons by varying the angle of observation [4] and the energy of the detected photons [6] we can get more qualitative information about the microstructure of crystals of large thickness, in particular, we can estimate the characteristic dimensions of grains instead of using X-rays with fixed-wavelength or the method of electron microscopy. Therefore, the problem of estimation of the quality of the internal structure of crystals and the identification of common grains sizes by radiation generated while passing through them fast electrons, and achievable parameters is important and relevant.

2. Formulation of the problem

Our method of estimating of the typical sizes of grains in crystals of class $a\alpha$ is based on the results of an experiment [7] in searching and researching of parametric X-ray radiation at small angles to the velocity of a particle in a tungsten crystal. From the point of view of our problem the most important outcome of the experiment was the detection of diffraction suppression of the bremsstrahlung yield i.e. the presence of minima of the orientational dependences (OD) of yield of high-energy photons with $\omega \approx \gamma \omega_p$, where $\gamma$ - is Lorentz factor, $\omega_p$ - is plasma frequency of the medium (see Figure 1), which position is consistent with kinematic conditions of the diffraction of photons with this energy, the error of which is not worse than 1%.

While calculating the orientational dependences of x-ray yield for the conditions of an experiment [7] we took into account the efficiency of the spectrometers and contribution of transition radiation from the output face of the crystal fulfilled in the work[6] and used the approximate method that took into account the diffraction of the bremsstrahlung in perfect crystals [9]. The calculation showed that the position of confidently manifested minima coordinate greatly with the calculation including those for the weaker reflecting planes (112) and (220). But depth of the dips in the calculated orientation dependences for the type of planes (110) and the photon energies 67 keV and 96 keV, respectively, $\sim 2.5\%$ and $\sim 1.5\%$ was almost five times smaller than the experimental ones $\sim 15\%$ and $\sim 10\%$.

Based on these results and the fact that in the experiment [7] it was firstly observed the dynamic effects in the emission of fast electrons in crystals - so to say parametric X-rays along the particle velocity (forward PXR), which supposes the perfection of the structure by omission,
the authors [6] classified the crystal used in this experiment as a crystal of class $a\alpha$ and suggested two methods of estimation of the sizes of the blocks in such crystals: the first method is estimated by the degree of manifestation of forward PXR in the region of the photon energy $\omega < \gamma \omega_p$ and diffraction of bremsstrahlung in the region of energies $\omega > \gamma \omega_p$; the second one is estimated by ratio of experimentally measured values of the diffraction suppression of the yield of photons with fixed energy in a mosaic crystal with the calculation or the result of measurements for the perfect crystal. The first method requires the accelerator with the energy $\sim 1$ GeV and above for its implementation but it is not always justified. Therefore, more interesting and promising is the second one, because such measurements can be made to by means of less energy accelerators.

3. Estimation of the size of blocks according to the diffraction suppression yield of bremsstrahlung

Method of estimating the typical sizes of grains in mosaic crystals according to the diffraction suppression of the yield of bremsstrahlung [6] is based on two assumptions. The first is that the investigated crystal is a mosaic crystal of class $a\alpha$. That is the sizes of grains in it is significantly longer than the primary extinction $l_{ex}$ for a given order of reflection and the photon energy $\omega$. The second one assumes that the probability of a repeated reflection of the diffracted photons in other grains is negligible.

For the crystal used in the experiment [7], validity of the first assumption is justified by the fact that in the crystal it was for the first time registered forward PXR. The presence of minimum in yield of high-energy photons, the depth of which significantly exceeds the contribution of the diffraction suppression of release of hard photons in a perfect crystal, is a clear indication of blocking structure along the direction of the photon beam, because otherwise we can’t provide the experimentally observed magnitude of suppression. The same is with the width of the minimum which is larger than it is predicted by the calculation for the perfect crystal. For any other crystal, the implementation of this conditions must necessarily be checked.

The surface mosaicity of tungsten crystal, used in the experiments [7], is $\sigma_m \leq 0.2$ mrad. In fact, this value corresponds to the sensitivity of the methodology used certification of crystal surface, that is the true meaning of the interior mosaicity can be either smaller of the this value or bigger. This value is comparable with the width of the Darvin Table for photon energy $\omega=67$ keV $\Delta \Theta \sim 0.03$ mrad. Therefore, to determine the bounds of applicability of techniques [6] we are required additional study of the effect multiple reflection on the recorded values of diffraction suppression.

For the exact solution of the contribution of repeated reflections we need unknown information about the distribution of grains along the angle of misalignment and the sizes which, strictly speaking, we need to find by the results of the measurements. So let’s analyze the influence of this factor on the results of measurements of the photons yield with fixed energy in the case of the crystal belonging to the class $b\alpha$ and by using techniques [10].

Figure 1 shows results in comparison of the measured photon OD with $\omega = 67$ keV of a tungsten crystal in the spectral range, clipping crystal-diffraction spectrometer in the experiment [7] and calculation results for three crystals of tungsten with a thickness of 0.41 mm and different microstructure. They are as follows: a perfect crystal by the method of [9]; mosaic crystal with $\sigma_m=0.2$ mrad and $\sigma_m=0.5$ mrad by the method [10], that is there is the assumption that the characteristic length of the blocks in it is smaller than the primary extinction length of the photons of this energy $l_{ex} \approx 3.1 \mu m$, and the number of blocks on the absorption length $l_a \approx 183 \mu m \geq 50-100$, depending respectively 1-3. The calculation takes into account the efficiency and angular capture of crystal-diffraction spectrometer. The experimental errors are statistical and do not include normalization errors and definitions efficiency of the NaI(Tl) detector.

The figure shows that the presence of a mosaic leads to a broadening of a dip in yield of photons in comparison with the case of a perfect crystal and of its much more depth. As we
expected, none of the dependencies describes the results of measurements, because the used in the experiment [7] tungsten crystal is a crystal of class $aa$, and not $bo$. Nevertheless, its mosaicity value is close to the estimation of $\sigma_m \approx 0.2$ mrad since the dependence of 2 with $\sigma_m=0.2$ mrad is closer to the experimental one than the dependence of 3 for $\sigma_m = 0.5$ mrad.

As can be seen from the figure, the difference in the magnitude of $\sigma_m$ didn't practically count to the magnitude of the diffraction suppression. In both cases it is close to 45%. For mosaic crystals of class $b$ the total reflectivity of X-ray radiation is $Q \sim z^2$, see, for example, [3], and the probability of the reflection is $W \sim Q/\sigma_m^2$ [10], so for small values of $\sigma_m$ multipath probability increases dramatically. In order to improve abovementioned in Figure 2 we found the estimated value of $W_n$ which is probability for a photon to experience at least $n$ reflections before it absorption in the crystal, or departure from it, averaged according to the energy resolution of the spectrometer. The value of $n$ varies from 1 to 17-18, depending on the value of $\sigma_m$. The calculation is made for the experimental conditions [7] and for four values of $\sigma_m$.

![Figure 1. OD yield of photons with $\omega=67$ keV. Points - the experiment [7]; Curves: 1 - a perfect crystal, 2 - $\sigma_m=0.2$ mrad, 3 - $\sigma_m=0.5$ mrad.](image1)

The figure shows that in the crystal with $\sigma_m=0.2$ mrad the probability of one or more reflections of $\sim 65\%$ is more than in the crystal with $\sigma_m=0.5$ mrad ($\sim 46\%$) and it exceeds the value of diffraction suppression of the yield of photons ($\sim 45\%$). The resulting suppression of the bremsstrahlung yield is determined by the ratio of a number of odd and even reflections. The odd reflections decrease the recorded yield, while the even ones partially compensate this loss (see Figure 3).

As can be seen from Figure 3, for $\sigma_m=0.2$ mrad the fraction of photons that have experienced

![Figure 2. The dependence of $W_n$ on the crystal mosaicity. ○ - $\sigma_m=0.2$ mrad; ● - $\sigma_m=0.5$ mrad; △ - $\sigma_m=2$ mrad and + - $\sigma_m=10$ mrad.](image2)
only a single reflection, is responsible for the observed value of the diffraction suppression, \(\sim 23\%\) and is significantly smaller than the diffraction suppression of \(\sim 45\%\) and slightly less than for a crystal with \(\sigma_m = 0.5\) mrad (\(\sim 26\%\)). While the probability of all subsequent reflections in a crystal with lower value of \(\sigma_m\) is significantly higher. As a result, the net suppression of yield for these two crystals is close enough (See Fig. 1, curves 2 and 3). With the increase of the characteristic angle of mosaicity the probability of multiple reflections decreases sharply. For photon energy \(\omega=67\) keV, the probability of re-reflection is reduced by 10 times in comparison with the probability of a single reflection, but for a crystal with \(\sigma_m \sim 3\) mrad. That is, for large values of \(\sigma_m\) and for the photon energy we may not take into account the effect of multiple reflections. It is necessary to emphasize that the reflectivity for crystals of class \(b\) is \(Q \sim \lambda^2\), so that when the photon energy changes the boundary value of \(\sigma_m\), in which the effect of re-reflections may be ignored, should be determined again.

The analysis prove that multiple reflections significantly change the observed dependence of the diffraction suppression of the radiation yield in comparison with the case in which this effect can be neglected. For crystals of class \(a\alpha\) the situation is significantly more complicated, since there is no information about the distribution of blocks in size, so that the accurate statistical analysis is impossible without it. This problem seems to be solved by successive approximations using an additional information about the distribution of blocks in the corners of misalignment and size. If this information is missing, the methodology of estimating the number and characteristic sizes of blocks by means of the magnitude of the diffraction suppression of photon yield, proposed in [6], can only be used if value of registered suppression is much smaller than for the crystal class of \(b\alpha\) with the same value \(\sigma_m\). If it is wrong and they are comparable, the estimated number of grains obtained in this way, would be understated, but their characteristic size, on the contrary, would be significantly overstated.

This condition is satisfied for the tungsten crystal used in the experiment [7], and for photon energy \(\omega=67\) keV. The recorded magnitude of diffraction suppression of radiation yield \(\sim 17\%\) (see Fig. 1) is significantly less than calculated magnitude for the crystal class of \(b\alpha\) with the same thickness and characteristic mosaicity angle \(\sim 45\%\), and therefore we can use the ratio of the depths of experimental and calculated minima in the radiation yield as an estimate of the number of blocks along the path of the photons in the crystal \(N_{bl} \approx 5.4\).

As we noted above, the magnitude of the diffraction suppression for a perfect crystal (Fig. 1, curve 1) is calculated by using the approximation about absence of influence of the photon absorption on the process of diffraction [9]. While comparing this approach with a more precise
one [11] we found out that for unidirectional and monochromatic beam of photons the difference in the diffraction suppression, calculated by means of these approaches for blocks with thickness up to 50 μm is not more than 10%. That’s why the use of results obtained with the help of a simple technique [9], to estimate the number of blocks is justified. The value $\sigma_m \approx 0.2$ mrad is almost an order larger than the characteristic angle of total reflection $\Delta \Theta \sim 3 \cdot 10^{-5}$ mrad for photon energies $\omega=67$ keV. Therefore, if there is a photon on the way of $\sim 5$ blocks, the probability of re-reflection of the once diffracted photon in the first approximation cannot be taken into account.

The formation of recorded yield of photons with the fixed energy in the present experiment depends not only on their decreasing because of diffraction processes and absorption, but also because of the photons generation by electrons passing through the investigated crystal. Therefore, as a characteristic distance over which the recorded radiation yield is formed it is better to take not the length of photon absorption $l_a$, as it is done in [6], but the effective path length of photons in crystal, that is the average distance traveled by the photons which were emitted from crystal into the solid angle, which was overlain by the spectrometer.

$$< l_{ph} > = \int \int (T-t)dt \int d\omega \int \frac{d^2I^*}{d\omega d\Omega} S(\omega, \vec{n}, t) d\Omega,$$

where $d^2I^*$ is a spectral and angular distribution of bremsstrahlung intensity, if we take into account multiple scattering of electrons in the target [9], and $S(\omega, \vec{n}, t)$ is a coefficient, which takes into account the absorption of photons with the direction of $\vec{n}$ and energy $\omega$ in the target and the efficiency of crystal-diffraction spectrometer, as for $T$ it means thickness of crystal. The integration takes place all over the angles of emission and photon energies taking into account angular and energy capture of the spectrometer. For photon energies $\omega=67$ keV and the crystal thickness of tungsten is 0.41 mm, $< l_{ph} > = 236$ μm $> l_a \approx 184$ μm. Therefore, the characteristic size of the blocks in the crystal of tungsten, which was used in the experiment [7], is $l_{bd} = < l_{ph} > / N_{l_a} \sim 40$ μm instead of 30 μm [6].

In order to measure by assess of characteristic dimensions of grains in crystals of class αα it is not necessary to use an accelerator with energy of $\sim 1$ GeV as in the experiment [7]. The measurements may be conducted with the help of relatively cheap and more common electron accelerators with energies of 30-50 MeV and with a sufficiently large database that would give an opportunity to implement the method of allocation of emission with the fixed energy by means of crystal diffraction spectrometers.

The additional advantage of low-energy electrons is the lack of forward PXR in X-ray frequencies, which can mask diffraction suppression of radiation yield [7, 6]. The analyzed method for estimating the characteristic dimensions of grains is based on its registration. This allows scientists to conduct measurements with less energy photons thus limiting the size of the viewing area of the crystal and the number of blocks contributing to the suppression of the recorded yield of photons.

From the viewpoint of the testing method for assessing the quality of the internal structure of crystals and the characteristic dimensions of grains, one of the important differences of medium-energy accelerators from accelerators with higher energies is significantly larger size of the electron beam on the target $\sim 5 \times 10$ mm² and more, see, for example, [12]. Resolution and efficiency of the crystal-diffraction spectrometer depends on the angular distribution of the radiation hitting on the analyzer crystal, and on the distances between the target, where investigated radiation is generated, and the crystal-analyzer together with the detector, registering the diffracted radiation. The change of the size of the emitting section leads to a change of the angular distribution of radiation on crystal-analyzer and it can also lead to a deterioration of monochromatic of the detected radiation and, consequently, to a deterioration
of sensitivity of the proposed method.

In order to test the influence of this factor on the resolution of the spectrometer by means of the method [10] it was carried out the simulation of efficiency of crystal diffraction spectrometer based on a mosaic crystal of pyrolytic graphite with the width of 2.5 mm for different transverse dimensions of electron beam hitting on a crystalline sample under study. Figure 4 shows the simulation results for the electron and photons energies of 50 MeV and 67 keV, respectively. The remaining conditions are identical with experimental conditions [7] for the photon energy. In the first case (○) the size of the electron beam were not taken into account (approaching a point of the beam), while the second (●) were 5 × 16 mm² horizontally and vertically, respectively.

Figure 4. The efficiency of crystal diffraction spectrometer. \( E_e = 50 \) MeV, \( \omega = 67 \) keV. ○ - spot beam; ● - with a beam size of 5 × 16 mm².

The figure shows that increasing of the size of the electron beam on the target practically did not affect the characteristics of the spectrometer. The area under the two dependencies (total efficiency of the spectrometer) has changed less than 2%, while resolution deteriorated by \(~ 30\%\). Therefore we can expect that approximately the same may take place with the change of depth and width of a minimum in OD caused by diffraction suppression of the yield of bremsstrahlung in a perfect crystal. However, it should be noted that for the experimental conditions [7] OD of the yield of radiation calculated for the same size of an electron beam on the tungsten crystal, were completely identical.

Another feature of such common electron accelerators of medium-energy as microtron or a linear accelerator is a short time of acceleration cycle \(~ 6-10 \mu s\), which is comparable with a typical duration of event for traditional X-ray detectors as NaI(Tl) or silicon detector \(~ 1-10 \mu s\), depending on the type of a detector. Therefore, to avoid overlap and separation of the photons only one order of reflection with a help of a differential discriminator as it was done in the experiment [7], we should use faster and more expensive detectors as \( \text{CeF}_3 \) for example or the current of the accelerator must be maintained in the way that it would be recorded no more than 0.2-0.4 event during a single cycle of acceleration [13].

For an NaI detector in order to obtain the statistical error of 1-2% even when the frequency of the accelerator is 50 Hz we need at least 200 seconds for each crystal orientation. In order to identify without doubts the effects of diffraction suppression it is necessary to make measurement of the yield of radiation for at least 200-500 points in increments of \(~ 0.1 \text{ mrad}\) that requires 10-20 hours of non-stop operation of the accelerator. We need a comparable time, first to adjust the orientation of the crystal-analyzer spectrometer, and second to search the area of diffraction suppression for each plane of interest in the investigated crystal.

The inclusion of the detector may become the way out of the situation. The detector will record the diffracted radiation in the integral mode. As it is shown in [14] such mode of an NaI detector located in an accelerator hall, allows us to carry out measurement of radiation yield
from the OD number points of $\sim 500$ for 30-40 minutes with an accuracy of $\sim 1-2\%$. When the detector is well protected from the background, the contribution of photons, which are not related with the process of diffraction, doesn’t exceed 3-5\%, see for example, [6]. Simultaneous recording of several orders of reflection, but not one as in [7], can be accounted during data processing, the more so as for the crystal pyrolytic graphite reflectivity for the second one allowed reflection order is of at least twenty times smaller than the first one [8]. If necessary, the contribution of second and higher orders of reflection can be further reduced by choosing a detector with low detection efficiency for these photon energies.

One of the advantages of the proposed approach over traditional ones is in the fact that this method is useful for the thickness of crystals, which is much higher than absorption length of photons of the characteristic X-rays material of the anode x-ray tubes used in X-ray analysis. With the help of this approach you can see the mutual misalignment of blocks in the volume sample at an angle $\sim 10^{-5}$ rad, which is practically impossible to make using traditional methods.

4. Conclusion
The results of the analysis of the method of estimation of the internal microstructure of crystals of large thickness according to the diffraction suppression of the bremsstrahlung yield with the fixed energy can be briefly summarized as follows:
1) Belonging of the crystal to the class $b\alpha$ drastically changes the appearance of orientation dependence of the diffraction suppression in comparison with the measurements [7].
2) Measurement of the diffraction suppression of the X-ray radiation yield with the fixed energy of fast electrons in mosaic crystals and its comparison with the calculated or measured ones for a perfect crystal allows us to determine the characteristic dimensions of grains in these crystals under condition that the magnitude of the recorded suppression is significantly less than for the crystal of class $b\alpha$ with the same value of the characteristic mosaicity angle.
3) Methodology to estimate the characteristic dimensions of grains in such crystals according to diffraction suppression of the yield of the X-ray radiation with the fixed energy possesses great capacity for work not only on high-energy accelerators, but also on medium energy electron accelerators, since the size of an electron beam target does not affect on the resolution of crystal diffraction spectrometers based.

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