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X-ray studies of the distribution function of crystalline grains over orientation angles in mosaic crystals

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Summary. — The paper is devoted to theoretical and experimental studies of the new approach to X-ray diagnostics of mosaic crystals being proposed. The model of the scattering process on the crystals with the arbitrary distribution function of the crystalline grains over orientation angles is considered in detail. Experimental setup created on the base of this model is described as well as the first results of experimental studies conducted using highly oriented pyrolytic graphite with mosaicity 0.4 and 0.8 degrees.

PACS 78.70.-g – Interactions of particles and radiation with matter.
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1. – Introduction

Most part of real crystals consists of small grains with perfect structure oriented in space at small angles relative to each other. The function of the distribution of such grains over orientation angles is of interest in X-ray optics using the crystalline reflecting systems [1, 2], in high energy physics using the crystals for producing of X- and gamma-rays by means of relativistic electron beams [3-5] and so on.

Usually, the investigation is concerned with one parameter of the above distribution function only (a halfwidth as a rule). Angular measurements of scattered quasimonochromatic X-rays have a peak incidence to solve the task being discussed [6-8].

The new approach to mosaic crystal diagnostics on the base of broad-band X-ray beam scattering [9] is studied in this work theoretically and experimentally. It should be pointed out that the analogous approach has been used in [10] without any theoretical grounds. We show that the desired distribution function is proportional to the orientation dependence of strongly collimated X-ray flux scattered by the sample being studied.

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2. – Theoretical model

Let us consider X-ray scattering process basing on Maxwell equation for Fourier transform of the electric field $\mathbf{E}_{\omega\mathbf{k}}$

$$(k^2-\omega^2)\mathbf{E}_{\omega\mathbf{k}}-\mathbf{k}(\mathbf{k}\mathbf{E}_{\omega\mathbf{k}})=4\pi i\omega\mathbf{j}_{\omega\mathbf{k}}=\omega^2\int \mathrm{d}^3k G(\mathbf{k},\mathbf{k}')\mathbf{E}_{\omega\mathbf{k}'}, \tag{1}$$

where the induced current density of target electrons is determined within the frame of high energy limit $\omega \gg I$ (I is the average ionization potential of an atom). This approximation allows to consider atomic electrons as free in the scattering process. In the case being considered the function G in (1) is determined by the simple formula

$$G\approx -\frac{e^2}{2\pi^2 m}F(\mathbf{k}'-\mathbf{k})\sum_\beta e^{i(\mathbf{k}'-\mathbf{k})\mathbf{r}_\beta}. \tag{2}$$

Here $F(\mathbf{k}'-\mathbf{k})$ is the form factor of an atom, $\mathbf{r}_\beta$ is the coordinate of β-th atom in the target. The function G describes both reflecting and scattering properties of the target. To exclude possible singularities in the scattering cross-section one should separate from G the part averaged over atomic coordinates

$$G\equiv\langle G\rangle+\tilde G=\omega_p^2\delta(\mathbf{k}'-\mathbf{k})+\tilde G. \tag{3}$$

Substituting (3) in (1) we obtain the equation

$$(k^2-\omega^2\epsilon(\omega))\mathbf{E}_{\omega\mathbf{k}}=\omega^2\int \mathrm{d}^3k\tilde G(\mathbf{k},\mathbf{k}')\left(\mathbf{E}_{\omega\mathbf{k}'}-\frac{\mathbf{k}}{\omega^2\epsilon(\omega)}(\mathbf{k}\mathbf{E}_{\omega\mathbf{k}})\right), \tag{4}$$

very convenient to be solved by perturbations on the degree of $\tilde G(\mathbf{k},\mathbf{k}')$. Here $\epsilon(\omega)=1-\omega_p^2/\omega^2$ is the ordinary dielectric permeability in X-ray range, ω_p is the plasma frequency.

Reasoning $\mathbf{E}_{\omega\mathbf{k}}=\mathbf{E}^{(i)}_{\omega\mathbf{k}}+\mathbf{E}^{(S)}_{\omega\mathbf{k}}$, where the first item is the incident wave, following from (4) in zero order on $\tilde G(\mathbf{k},\mathbf{k}')$, and the second one is the scattered wave, following from (4) in the next order on $\tilde G(\mathbf{k},\mathbf{k}')$, one can obtain the following formulae:

$$\begin{aligned}\mathbf{E}^{(i)}_{\omega\mathbf{k}}&=\mathbf{e}_iE_\omega\delta(\mathbf{k}-\omega\sqrt{\epsilon}\mathbf{n_i}),\quad \mathbf{e}_i\mathbf{n}_i=0,\\ \mathbf{E}^{(S)}_{\omega\mathbf{k}}&=\frac{\omega^2E_\omega}{k^2-\omega^2\epsilon}\tilde G(\omega\sqrt{\epsilon}\mathbf{n_i},\mathbf{k})\left(\mathbf{e}_i-\frac{\mathbf{k}(\mathbf{k}\mathbf{e}_i)}{\omega^2\epsilon(\omega)}\right).\end{aligned} \tag{5}$$

Calculating integral Fourier by the stationary phase method one can obtain the following expression for the amplitude of scattered wave:

$$\begin{aligned}\mathbf{E}^{(S)}_{\omega}&=\int \mathrm{d}^3k\exp[i\mathbf{k}\mathbf{n}_Sr]\mathbf{E}^{(S)}_{\omega\mathbf{k}}\rightarrow\mathbf{A}_S\frac{\exp[i\omega\sqrt{\epsilon}r]}{r},\\ \mathbf{A}_S&=-2\pi^2(\mathbf{e}_i-\mathbf{n}_S(\mathbf{n}_S\mathbf{e}_i))\tilde G(\omega\sqrt{\epsilon}(\mathbf{n}_i-\mathbf{n}_S))E_\omega,\end{aligned} \tag{6}$$

where $\mathbf{n}_S$ is the unit vector to the direction of scattered wave propagation.

Let us estimate possibilities of the diagnostics of a crystal mosaicity on the base of the spectral-angular distribution of scattered quanta

$$\omega \frac{\mathrm{d}N}{\mathrm{d}\omega \mathrm{d}\Omega} = 4\pi^2 (1 - (\mathbf{n}_S \mathbf{e}_i)^2) |E_\omega|^2 \left\langle \left| \tilde{G} \left(\omega \sqrt{\epsilon} (\mathbf{n}_i - \mathbf{n}_S) \right) \right|^2 \right\rangle, \tag{7}$$

following from (6). Here brackets $\langle\rangle$ mean the averaging over atomic coordinates and grain orientations. The coherent Bragg scattering dominates in the process under study. Since the angular width of Bragg resonance is small as compared with characteristic mosaic angles, the contribution of different grains to scattering process may be considered as independent. Determining coordinates of atoms $\mathbf{r}_\beta$ by the formula

$$\mathbf{r}_\beta = \mathbf{R}_n + \mathbf{r}_{nl} + \mathbf{u}_{nl}, \tag{8}$$

where $\mathbf{R}_n$ is the coordinate of the n-th elementary cell in a grain, $\mathbf{r}_{nl}$ is the equilibrium coordinate of l-th atom in n-th elementary cell, $\mathbf{u}_{nl}$ is the thermal displacement of this atom, one can perform the averaging over $\mathbf{u}_{nl}$ in (7). The result of averaging is given by the simple formula

$$\begin{aligned} \omega \frac{\mathrm{d}N_{\mathrm{coh}}}{\mathrm{d}\omega \mathrm{d}\Omega} &= \sum_g \omega \left\langle \frac{\mathrm{d}N_g}{\mathrm{d}\omega \mathrm{d}\Omega} \right\rangle, \\ \omega \frac{\mathrm{d}N_g}{\mathrm{d}\omega \mathrm{d}\Omega} &= \frac{\pi}{2} V \omega_p^4 \frac{|S(\mathbf{g})|^2 e^{-g^2 u^2}}{(1 + g^2 R^2)^2} (1 - (\mathbf{n}_S \mathbf{e}_i)^2) \delta \left(\omega \sqrt{\epsilon(\omega)} (\mathbf{n}_i - \mathbf{n}_S) - \mathbf{g} \right), \end{aligned} \tag{9}$$

where V is the volume of the target, $S(g)$ is the structure factor of an elementary cell, u is the mean square amplitude of atom thermal vibrations, R is the screening radius in Fermi-Thomas atom model (a simplest model with exponential screening is used in the paper), brackets $\langle\rangle$ in (9) mean the averaging over grain orientations only, the summation in (9) is over all reciprocal lattice vectors $\mathbf{g}$. It should be noted that the contribution of each item in the sum (9) is independent, which is why one can consider the scattering by a single crystallographic plane separately.

It is convenient for further analysis to express all vectors in (9) in terms of angular variables in line with fig. 1 and formulae

$$\begin{aligned} \mathbf{n}_i = \mathbf{e}_y, \quad \mathbf{n}_S &= \mathbf{e}_2 \left(1 - \frac{1}{2} \Theta^2 \right) + \boldsymbol{\Theta}, \quad (\mathbf{e}_2 \boldsymbol{\Theta}) = 0, \\ \mathbf{g} &= g \left[\mathbf{e}_1 \left(1 - \frac{1}{2} \eta^2 \right) + \boldsymbol{\eta} \right], \quad (\mathbf{e}_1 \boldsymbol{\eta}) = 0. \end{aligned} \tag{10}$$

An average position of reflecting crystallographic plane will be considered to be close to exact Bragg resonance

$$\phi = \frac{\pi}{2} - \frac{\chi}{2} - \varphi', \quad \varphi' \ll 1. \tag{11}$$

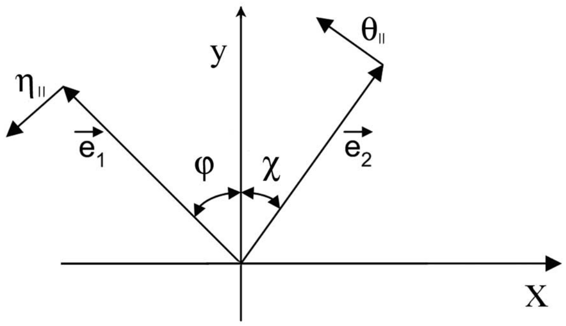

Fig. 1. – Geometry of scattering process. The axis y coincides with the axis of incident X-ray beam, χ is the scattering angle, φ is the total orientation angle.

Using (9)–(11), let us determine the total number of Bragg-scattered reflex quanta as a function of the orientation angle φ':

$$N_g(\varphi') = \int_{-\frac{\Delta\Theta}{2}}^{\frac{\Delta\Theta}{2}} \mathrm{d}\Theta_\perp \int_{-\frac{\Delta\Theta}{2}}^{\frac{\Delta\Theta}{2}} \mathrm{d}\Theta_\parallel \int \mathrm{d}^2\eta \Phi(\boldsymbol{\eta}) \int \mathrm{d}\omega \frac{\mathrm{d}N_g}{\mathrm{d}\omega \mathrm{d}^2\Theta}$$
$$= \pi \frac{\sin^2 \frac{\chi}{2}(1+\cos^2\chi)}{\cos\frac{\chi}{2}} V \frac{\omega_p^4}{g^3}$$
$$\times \frac{|S(g)|^2 e^{-g^2 u_T^2}}{(1+g^2R^2)^2} \int \mathrm{d}\eta_\parallel \Phi_\parallel(\eta_\parallel) \int_{\omega_-}^{\omega_+} \frac{\mathrm{d}\omega}{\omega} |E_\omega|^2 \delta$$
$$\times \left(\eta_\parallel - \varphi' - \left(\frac{\omega}{\omega_g} - 1\right) \tan\frac{\chi}{2}\right), \tag{12}$$

where $\Phi(\boldsymbol{\eta})$ is the desired two-dimensional distribution function, describing the distribution of crystal grains over orientation angles $\eta_\perp$ and $\eta_\parallel$, $\omega_g = g/2\sin(\chi/2)$ is the Bragg frequency, $\Delta\Theta \ll 1$ is the photon collimator angular size, other quantities in (12) are determined by the formulae

$$\Phi_\parallel(\eta_\parallel) = \int_{-\frac{\Delta\Theta}{4\sin(\chi/2)}}^{\frac{\Delta\Theta}{4\sin(\chi/2)}} \mathrm{d}\Theta_\perp \Phi(\eta_\parallel, \eta_\perp) \approx \frac{\Delta\Theta}{2\sin(\chi/2)} \Phi(\eta_\parallel, 0), \tag{13}$$
$$\omega_\pm = \omega_g \left(1 \pm \frac{\Delta\Theta}{4\tan(\chi/2)}\right).$$

Let us determine the function $\Phi(\eta_\parallel)$ from eq. (13). Under conditions of broad-band incident photon flux and small enough collimator size, so that the spectrum $|E_\omega|^2 \approx \mathrm{const}$ within the limits $\omega_- < \omega < \omega_+$, the task under consideration may be solved exactly by

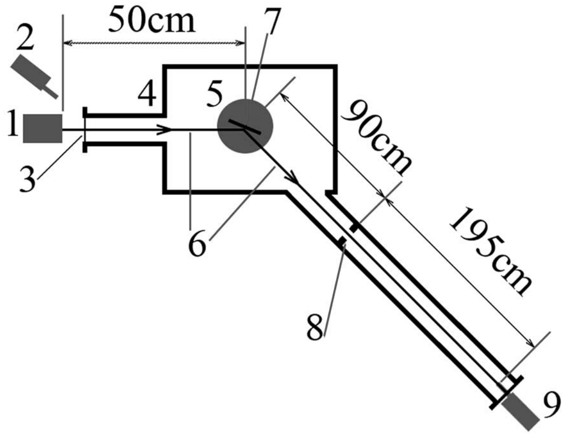

Fig. 2. – The experimental setup.

means of Fourier transformation. The result may be presented in the form

$$(14)\quad \Phi_{\|}(\eta_{\|}) = \frac{1}{A}\int_{-\infty}^{\infty} \mathrm{d}\xi e^{-i\xi\eta_{\|}} \frac{\xi}{\sin(\frac{\Delta\Theta}{4}\xi)} N_{g\xi} = -\frac{2}{A}\frac{d}{d\eta_{\|}}\left\{N_g\left(\eta_{\|}+\frac{\Delta\Theta}{4}\right)+N_g\left(\eta_{\|}+\frac{3\Delta\Theta}{4}\right)+N_g\left(\eta_{\|}+\frac{5\Delta\Theta}{4}\right)+\dots\right\},$$

$$A = 2\pi\sin(\chi/2)(1+\cos^2\chi)V\frac{\omega_0^4}{g^3}|E_{\omega_g}|^2\frac{|S(g)|^2 e^{-g^2u_T^2}}{(1+g^2R^2)^2}.$$

It is clear that a series of functions in the right side of (14) is defined as an integral sum with the proviso that the photo collimator size $\Delta\Theta$ is less than the characteristic angular scale of the orientation dependence $N_g(\eta_{\|})$, determined experimentally by one of the known methods [6-8]. Under the considered conditions formula (14) takes the simple form

$$(15)\qquad \Phi_{\|}(\eta_{\|}) = -\frac{4}{A\Delta\Theta}\frac{\mathrm{d}}{\mathrm{d}\eta_{\|}}\int_{\eta_{\|}+\Delta\Theta/4}^{\infty} N_g(t)\mathrm{d}t \approx \frac{4}{A\Delta\Theta}N_g(\eta_{\|}).$$

In accordance with (15), the desired distribution function $\Phi_{\|}(\eta_{\|})$ varies in direct proportion to the measured dependence $N_g(\eta_{\|})$. This property opens the possibility to measure $\Phi_{\|}(\eta_{\|})$ immediately experimentally. In order for the measurements of the distribution function $\Phi_{\perp}(\eta_{\perp})$ to be made, it is sufficient to turn the target by the angle $\pi/2$.

3. – Experimental setup and results of measuring

For the experimental verification of the above approach to X-ray diagnostics of mosaic crystals it is necessary to measure the orientation dependence of scattered by the target broad-band X-ray flux. The experimental setup was created on the base of broad-band X-ray radiation 6 produced by X-ray tube BS-11 with chromium anode 1 (fig. 2). The

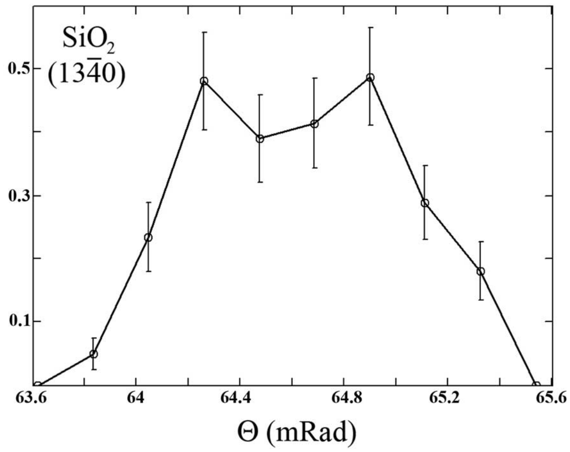

Fig. 3. – The orientation dependence of Bragg-scattered X-rays on SiO_2 crystal.

tube and vacuum chamber 4 of the setup were separated by 20 mkm mylar window 3. The vacuum of the system was better than 10^{-1} torr. The distance from the focal spot of the X-ray tube to the window was 50 mm. The target 7 was placed into goniometer 5 with 3 degrees of freedom rotation. The target was controlled in horizontal plane (incidence plane, the axis of plane's rotation coincides with the normal to the plane specified by the axes of both initial X-ray beam and detector 9) with the step of rotation $7.1 \cdot 10^{-5}$ rad and in vertical plane with the step of rotation $4 \cdot 10^{-5}$ rad (the axis of plane's rotation is given by both the surface of the target and the incidence plane). The scattered X-rays were collimated by the slit collimator 8 with horizontal size 400 mkm and vertical size 2 mm. The X-ray signal was detected by X-ray detector XR-100SDD 9 (shaped time 9.6 mks, Be window 7 mm^2, energy resolution 132 eV), placed into the vacuum chamber. The time of the statistics collection was controlled by X-ray p.i.n detector XR-100CR 2 (shaped time 9.6 mks, Be window 6 mm^2, energy resolution 152 eV). The detector 2 measured a background, that is proportional to the current of tube's emission. The distances of X-ray propagation between different parts of the setup are shown in fig. 2. The angle between axes of detector and initial X-ray flux was 135 degrees.

The desired distribution function proportional to the orientation dependence of Bragg-scattered broad-band X-rays is measured with the proviso that the angle between axes of detector and initial X-ray flux is constant. The angular resolution of the setup must be much less than the mosaicity of the target being studied. We have used two targets of the high-oriented pyrolytic graphite with mosaicity 0.4 (target 1) and 0.8 degrees (target 2). The angular resolution of the setup can be determined by means of the measuring of the orientation dependence of the yield of broad-band X-rays scattered by a perfect crystal. Such dependence for the crystal SiO_2 is presented in fig. 3. The intensity of X-ray tube 1 was not constant for the time of all statistics collection and the results were normalized by intensity of background measured by detector 2. It is shown that the resolution is less than 1 mrad (FWHM of the distribution contains the resolutions of the setup and the mosaicity of SiO_2). The initial position of the target before measuring of the orientation dependence must correspond to maximum in orientation dependence of the yield of scattered X-rays in vertical plane.

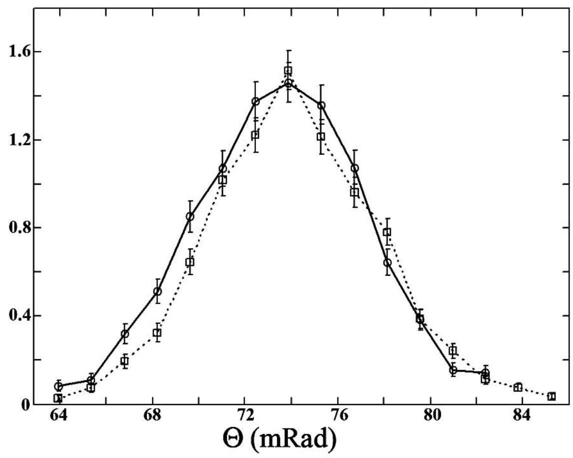

Fig. 4. – The orientation dependences of Bragg-scattered X-rays in the first order on highly oriented pyrolytic graphite with mosaicity 0.4 degrees. The dependences differ one from the other by rotation of the target by a 90° angle around its normal.

The crystals of high-oriented pyrolytic graphite with mosaicity 0.4 and 0.8 degrees were used in our experiments (lattice constant 3.35 angstroms). The angle between axes of detector and initial X-ray flux was chosen 135 degrees, the photoabsorption for this reflex was negligible in conditions of maximum intensity of the X-ray tube (the product of tube's intensity and coefficient of quanta transmission through Be window, air and mylar window was found to be maximum for our setup). The energy of Bragg-scattered photons for given geometry of the setup was about 4.9 keV for the first order of diffraction. The detector 9 registered reflexes of three orders of diffraction. The relation background/signal was better than 0.1%.

The orientation dependence for each target was measured in two orthogonal planes (the target was rotated around the normal to the plane of target's surface). The comparison of orientation dependences of Bragg-scattered initial quanta on target 1 in the first and second orders of diffraction for initial and rotated targets is presented in fig. 4 and fig. 5. The same but for target 2 is presented in fig. 6 and fig. 7.

Let us pay attention that the results of measuring in the first and second orders of diffraction are close together for each target. The difference in the positions of maxima in orientation curves measured in first and second orders of diffraction is caused by the shit of Bragg frequency. FWHM of the measured distribution function is in good agreement with the magnitude mentioned by manufacturer. The reflex of third order of diffraction was detected also, but its intensity was not enough for the statistics.

4. – Conclusions

It is shown that the distribution function of grains in a mosaic crystal over orientation angles can be determined by measuring the orientation dependence of the yield of Bragg-scattered reflex broad-band X-ray flux. For strongly collimated yield the desired distribution function becomes proportional to the measured orientation dependence.

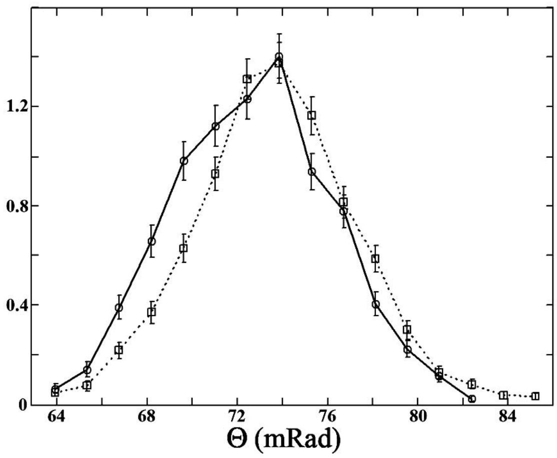

Fig. 5. – The same but for scattering in the second order.

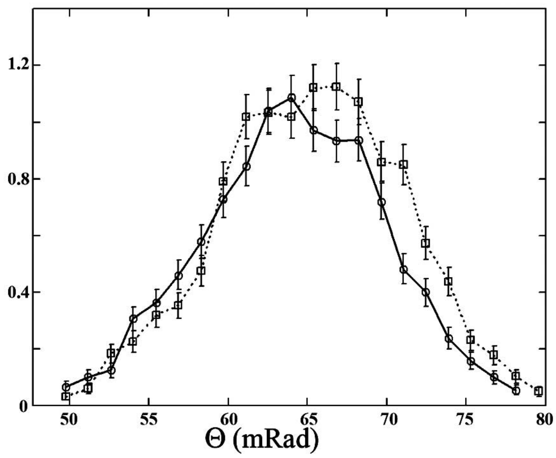

Fig. 6. – The orientation dependences of Bragg-scattered X-rays in the first order on highly oriented pyrolytic graphite with mosaicity 0.8 degrees. The dependences differ one from other by rotation of the target by a 90° angle around its normal.

The experimental setup for the measurement of the distribution function was created on the base of X-ray tube BS-11 and energy dispersive X-ray silicon drift detector XR-100SDD.

The distribution function was measured for two targets of highly oriented pyrolytic graphite with mosaicity 0.4 and 0.8 degrees in two orthogonal planes for each target.

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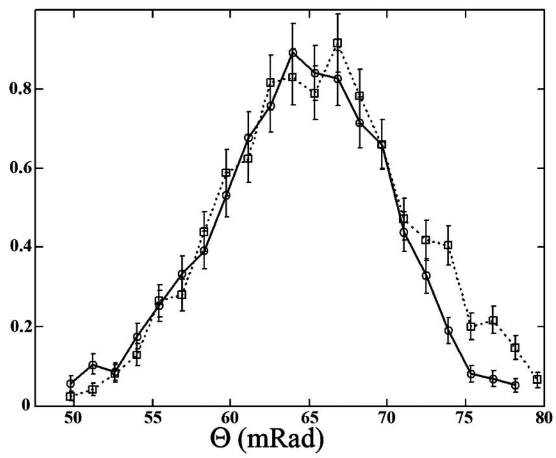

Fig. 7. – The same but for scattering in the second order.

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