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INTENSITY PROFILES IN A POLYCHROMATIC FOCUS OF ENERGY-DISPERSIVE *X*-RAY SPECTROMETER

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Интенсивность в полихроматическом фокусе
энергодисперсионного рентгеновского спектрометра

В работе проведены вычисления величины интенсивности излучения в полихроматическом фокусе рентгеновского спектрометра, работающего в энергодисперсионном режиме. Рассчитаны пространственное и спектральное распределения интенсивности в фокусе, оценены величины соответствующих разрешений.

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Intensity Profiles in a Polychromatic Focus
of Energy-Dispersive X-Ray Spectrometer

The calculations of the value of radiation intensity in a polychromatic focus of X-ray spectrometer have been accomplished in the paper. The spatial and spectral intensity distributions in the focus were determined and the corresponding resolutions were estimated.

The investigation has been performed at the Laboratory of Particle Physics, JINR.

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Introduction

The wide interest to X- ray spectrometers with bent crystals is caused by the possibility for a XAS-experiment in energy-dispersive scheme to be carried out by them with the use of synchrotron radiation sources. The basic element of such spectrometers is a bent crystal which extracts from the incident beam a radiation of X-ray spectrum. The energy range (E_0-ΔE, E_0+ΔE) of extracted radiation makes up a bandpass of the spectrometer [1]. The rays of different energies from the bandpass are subject to a Bragg reflection from the crystal and gather at a small area to be called a polychromatic focus (p. f.). In a XAS-experiment with such a spectrometer the radiation passes through the sample situated in the p. f. region simultaneously for all energies from the bandpass. Since the propagation directions of radiation of different energies are different the intensity of transmitted radiation can be registered by a position-sensitive detector (D) situated behind the sample (fig. 2).

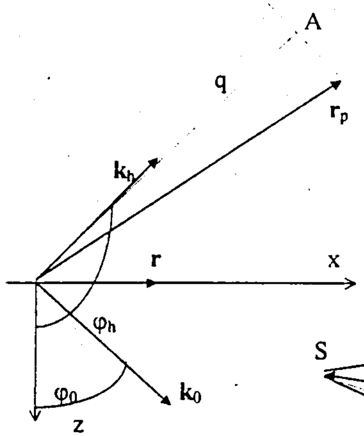

Fig. 1

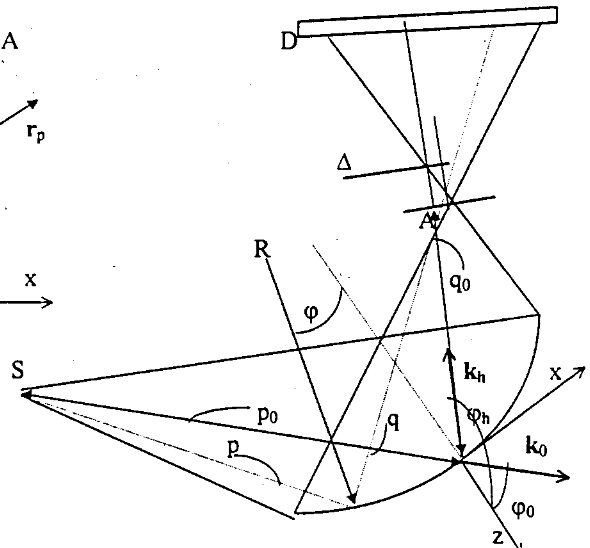

Fig. 2

Describing the properties of X-ray spectrometers with bent crystal a great deal of calculations can be carried out by a kinematics consideration of X-ray propagation from crystal to the sample. Thus there can be determined the distance q_0 from crystal to the position of p. f. region, the value of the chromatic aberration Δ (fig. 2) (the dependence of the image position Δ on the value of X-ray energy E). An angular resolution of position-sensitive detector of the spectrometer can be also determined and minimized [2].

However the matter of interest is also an absolute value of intensity of radiation in the p. f. region. To calculate the value of intensity it is necessary to take into account that the mechanism of reflection of incident waves from crystal is of a diffraction kind. In the paper on the basis of the principles of X-ray dynamic diffraction theory the calculation of spatial and spectral intensity distributions in the p. f. region in case of symmetrical reflection is fulfilled.

The kinematics

The principal peculiarities of X-rays movement in the spectrometers with bent crystals is shown on fig. 2 for the case of symmetrical reflection. Normally the distance p_0 from a radiation source S and illumination length l of the crystal are fixed values of the scheme. The purpose of a given experiment allows one to determine the value of energy domain $2\cdot\Delta E$ chosen for the energy scanning. Making use of these data one can define with the help of fig. 2 the value of a bending radius R:

$$\frac{1}{R}=\frac{\sin\theta_0}{p_0}+\frac{2tg\theta_0}{l}\frac{\Delta E}{E_0}. \qquad (1)$$

Here θ_0 is the reflection angle. This gives R=9.4 m (for the experimental set: p_0=20 m, l=20 cm, ΔE=300 eV, E_0=9 keV, Si(111)-symmetric reflection). Using the relation of cylindrical optics

$$\frac{1}{p_0}+\frac{1}{q_0}=\frac{2}{R\sin\theta_0} \qquad (2)$$

the distance q_0 to the p. f. region for the central ray can be obtained (in our case q_0=1.3 m). Since the reflecting crystal has a constant curvature radius the chromatic aberration in the vicinity of the point A (fig. 2) will exist [3]. These aberrations are displayed as a dependence of the deviation Δ (from point A) of a ray on its energy. The deviations of all rays picked out by the crystal have one and the same sign. Two rays which are symmetrical with regard to the central one (their energies $E_2>E_0>E_1$ are so that E_2-E_0=E_0-E_1) have equal deviations. From the construction of fig. 2 one can obtain the dependence of deviation on energy (aberration rate)

$$\Delta=\frac{3}{2}tg\theta_0 q_0\frac{(p_0+q_0)(2p_0-q_0)}{(p_0-q_0)^2}\left(\frac{E-E_0}{E_0}\right)^2. \qquad (3)$$

This gives the maximum value of the deviation Δ=1.43 mm.

To define the value of radiation intensity one ought to get out of the frames of geometrical optics. Then for any plane perpendicular to the ray the intensity has a spatial distribution. This means that for every energy from the bandpass the value of intensity in the p. f. region is defined by the deviation ξ from point A. The intensity maximum for a given energy is situated near the position

$$\xi=\Delta. \qquad (4)$$

As far as the spatial intensity distribution for a given energy does exist the energy distribution or spectral intensity for a given value of deviation ξ from point A does exist also. The domain of the function of intensity is a phase space (ξ, E). As is seen from (4) and (3) the intensity has its maximum value on parabola.

Let us fix a single energy value from the bandpass. When the diffraction on a flat crystal takes place the intensity depends on the variation of registration angle near the exact Bragg angle. When the ray of a given energy reflects from the bent crystal the intensity in the p. f. region will also have an angular distribution. Since the measurements in a spectrometer are fulfilled by a position-sensitive detector the registration direction once changed will correspond to detecting another value of energy. From the point of view of choosing the values to be measured it is convenient to consider not an angular distribution but a spectral one (since the angle to energy connection in the theory of diffraction does exist). The spectral intensity just introduced will further be treated as a spectral intensity in a monochromatic regime.

Thus the kinematics considerations lead to the determination of the position and the phase shape of the polychromatic focus formed by spectrometer. Further in the paper the relations (2), (3) are applied to calculate the following items : a) spatial distribution of intensity for the given energy from the bandpass, b) spectral intensity in a monochromatic regime, c) spectral intensity in a polychromatic regime, d) the dependence of intensity on the source size.

Intensity of reflection

Because of the diffraction process in a bent crystal the reflected X-wave field $E_h(\vec{r})$ is built up on the crystal surface. A further propagation of a reflected wave takes place in agreement with the Huygens-Fresnel principle. Owing to this principle the field amplitude at a given point can be determined once the amplitude distribution on a fixed wave surface is known. Following the principle the wave amplitude at an observation point $\vec{r}_p$ (fig. 1) is given by expression

$$E_h(\vec{r}_p) = 2ik\gamma_h \int E_h(\vec{r}) G_0(\vec{r}_p - \vec{r}) d^2r, \tag{5}$$

where $G_0(\vec{r}) = e^{ikr}/(4\pi r)$, k is the wave vector, $\gamma_h = \cos\varphi_h$ and integration spans the surface z=0. To make the value of the amplitude on the surface z=0 to be known it is necessary to use the solution of a diffraction problem. In this problem the wave with a wave vector $\vec{k}_0$ and amplitude $E_0(x,y,z)$ is subject to a Bragg reflection on a deformed crystal.

To treat the problem properly one should apply the methods of the theory of X-ray diffraction on nonhomogeneous matter. In this theory the Bloch amplitudes $\varepsilon_{0,h}$ of transmitted and diffracted waves are submitted to Takagi differential equations [4]. In the scattering geometry under consideration these equations have the form [5]

$$\begin{cases} i(a_0 \dfrac{\partial}{\partial x} + \dfrac{\partial}{\partial z})\varepsilon_0 + \dfrac{k\chi_0}{2\gamma_0}\varepsilon_0 + \dfrac{k\chi_{-h}}{2\gamma_0} e^{ih\vec{u}}\varepsilon_h = 0 \\ -i(a_h \dfrac{\partial}{\partial x} + \dfrac{\partial}{\partial z})\varepsilon_h + \dfrac{k(\chi_0 - \alpha)}{2|\gamma_h|}\varepsilon_h + \dfrac{k\chi_h}{2|\gamma_h|} e^{-ih\vec{u}}\varepsilon_0 = 0 \end{cases}, \tag{6}$$

where $a_{0,h} = tg\varphi_{0,h}$, $\gamma_0 = \cos\varphi_0$, $\alpha = 2\sin 2\theta \cdot \Delta\theta$, $\Delta\theta$ is a rocking angle, $\vec{h}$ is the reflection vector, χ are the structural constants, $\vec{u} = \vec{u}(x,z)$ - the field of deformations of a crystal. Since the scattering plane is perpendicular to the crystal plane the field of deformation $\vec{u}$ with regard to x,z is a function of two variables x,z. The deformation field of a thin plate bent with a constant radius is described by a quadric displacement function $\vec{u}$ in a coordinate system where the middle plane of the crystal plate coincides with the plane z=0 and x-axis belongs to this plane. For the coordinate system of the given problem one will have for the scalar product $\vec{h}\vec{u}$ [6]

$$\vec{h}\vec{u} = \frac{k}{2R}(\mu(z - t/2)^2 + 2(s_h - s_0)x(z - t/2) + (\gamma_0 + |\gamma_h|)x^2), \tag{7}$$

$$\mu = \frac{(s_h - s_0)a_{15} - (\gamma_0 + |\gamma_h|)a_{13}}{a_{11}},$$

where $s_{0,h} = \sin\phi_{0,h}$, a_{ik} are the components of a reverse tensor of elasticity modules, R is the bending radius, t - the thickness of the crystal plate. Let us assume X-rays to be emitted by a point source allocated at the distance p_0 (fig. 2). Then for the amplitude of incident wave in the vicinity of origin one has

$$E_0(x,y) = \frac{1}{p}\exp(ik(p + \frac{\gamma_0^2 x^2 + y^2}{2p})). \tag{8}$$

Here the p value corresponds to a given energy from the bandpass. Since the bending radius is large ($l/R \ll 1$) the crystal appears to be deformed weakly. In an approach of weakly deformed crystal one can obtain the following approximate solution of equations (6) for the diffracted wave on the interface z=0 (with incident wave from (8))[7]:

$$\varepsilon_h(x,y) = \exp i(bx - \frac{k|\gamma_h|}{2R}x^2 + \frac{ky^2}{2R} + \frac{k\mu t^2}{8R})\sqrt{\frac{\gamma_0}{|\gamma_h|}} \int_{-\infty}^{\infty} L(\frac{\tau}{\sigma}) G(\tau) e^{i\tau x} \frac{d\tau}{2\pi}. \tag{9}$$

$$\frac{1}{L(y)} = \begin{cases} y - i\sqrt{1 - y^2}, |y| \le 1 \\ y + sign(y)\sqrt{y^2 - 1}, |y| \ge 1, \end{cases} \tag{10}$$

$$b = \frac{k}{4a}\left(\frac{1}{\gamma_0} + \frac{1}{|\gamma_h|}\right)\chi_0 - k\gamma_0\Delta\theta + \frac{k}{4a}[a_0(s_h - s_0) + \mu]\frac{t}{R} = b_1 - k\gamma_0\Delta\theta,$$

$$\sigma = \frac{k\sqrt{\chi_h\chi_{-h}}}{2a\sqrt{\gamma_0|\gamma_h|}},\ 2a = a_0 - a_h,\ \lambda = \frac{kt}{2R}(s_h - s_0).$$

Here $G(\tau)$ is a Fourier transformation of function (8). In order to elaborate a final approximate solution let us apply a stationary phase method to estimate the value of integral in (9). We will have then for the Bloch amplitude ε_h

$$\varepsilon_h(x,y) = \frac{1}{p}\sqrt{\frac{\gamma_0}{|\gamma_h|}}\exp ik(p + \frac{\mu t^2}{8R} + \frac{y^2}{2p} + \frac{\lambda x}{k} - \frac{k}{2}((\gamma_0 + \gamma_h)/R - \gamma_0^2/p)x^2) L[(\lambda - b - k\gamma_0(1/R - \gamma_0/p)x)/\sigma]. \tag{11}$$

To find the wave amplitude in the p. f. region let us make use of expression (5) where for the function G_0 we admit the following approximate form:

$$G_0(\vec{r}_p - \vec{r}) = G_0(\xi, x) = \frac{1}{4\pi q}\exp ik(q + s_h(\xi/|\gamma_h| - x) + \frac{y^2}{2q} + \frac{\gamma_h^2}{2q}(\xi/|\gamma_h| - x)^2 - \frac{\gamma_h^2 s_h}{2q^2}(\xi/|\gamma_h| - x)^3). \tag{12}$$

Similarly to the p value from (8) the q value (figs. 1, 2) also corresponds to the individual energy from the bandpass. Thus for the amplitude in the polychromatic focus for the case of symmetrical reflection we have

$$\mathrm{E}(\xi)=\sin\theta\sqrt{\frac{k}{2\pi pq(p+q)}}\int_{-\infty}^{\infty}L[(b_1-k\sin\theta\Delta\theta-k\sin\theta(1/R-\sin\theta/p)x)/\sigma]\cdot$$

$$\cdot\exp ik[-x\xi\frac{\sin\theta}{q}-\frac{k}{2}\sin^2\theta(2/R\sin\theta-1/p-1/q)x^2+\frac{\cos\theta\sin^2\theta}{2q^2}(x-\xi/|\gamma_h|)^3]\,dx. \tag{13}$$

Let us admit in a further consideration a small wavelength variation in the vicinity of a given energy

$$\frac{\Delta\lambda}{\lambda}=ctg\theta\Delta\theta. \tag{14}$$

As a function of the argument (14) the squared modulus of amplitude (13) exposes a spectral intensity in a monochromatic regime of the spectrometer. Owing to the dispersive properties of X-rays in crystals this kind of spectral distribution of intensity does exist for any given reflection angle (or X-ray energy). Let us express the quantities p,q,θ in (13) by the parameters of the central ray p_0,q_0,θ_0 and energy. The relation (13) aside from the phase factor becomes

$$\mathrm{E}(\xi,\Delta\lambda/\lambda,E)=\sqrt{\frac{\sin\theta}{2dp_0q_0(p_0+q_0)}}\int_{-\infty}^{\infty}L[(b_1-\frac{\pi\cdot tg\theta}{d}\frac{\Delta\lambda}{\lambda}-\frac{\pi\sin\theta}{2d}(\frac{1}{q_0}-\frac{1}{p_0}-\frac{2(p_0+q_0)}{p_0(p_0-q_0)}\cdot$$

$$\cdot\frac{E_0-E}{E_0})x)/\sigma]\exp\frac{i\pi}{d}[-(1+\frac{2p_0}{p_0-q_0}\frac{E_0-E}{E_0})\frac{\xi x}{q_0}+\frac{3}{R}\frac{E_0-E}{E_0}x^2+$$

$$+\frac{\cos\theta_0\sin\theta_0}{2q_0^2}(x-\xi/|\gamma_h|)^3]\cdot dx, \tag{15}$$

where the relation (2) and the Bragg-Wolf formula

$$k\sin\theta=\pi/d,\ d=4.7A$$

were used. Expression (15) describes all principal peculiarities of spatial and spectral structures of a wave in the p. f. region. To obtain the formula (15) the approximation of a weakly bent crystal was used. This approximation corresponds to the joint coexistence of two processes: a) the wave of each energy from the bandpass is subject to a diffraction in an area assumed to be an ideal crystal; b) a region is forming within which all rays of the outgoing flux go. By means of relation (15) a great deal of intensity properties can be described.

Intensity in a monochromatic regime

Let us consider a monochromatic regime. In this case a single wavelength drops at the crystal. The reflection rate will be of a significant value if the deviation of registration angle from a given Bragg angle is small. Since this deviation (rocking angle) is connected to the relative wavelength deviation (14) the spectral profile versus the wavelength variation around a given energy can be obtained from (15). Let the energy of the central ray be envisaged. Retaining in an exponent's phase in (14) only the term linear with x we will have for the amplitude [7]

$$\mathrm{E}(\xi,\Delta\lambda/\lambda,E_0)=\sqrt{\frac{2dp_0q_0}{\sin\theta_0(p_0+q_0)}\frac{2\sigma}{p_0-q_0}}\frac{J_1(\sigma X)}{\sigma X}\theta(X), \tag{16}$$

$$X=\frac{2p_0}{\sin\theta_0(p_0-q_0)}\left\{\xi-\frac{3ctg\theta_0}{2q_0}(\xi+\frac{2p_0q_0}{p_0-q_0}(tg\theta_0\Delta\lambda/\lambda-\frac{db_1}{\pi}))^2\right\}, \tag{17}$$

where $J_1(y)$ - Bessel function, $\theta(y)$ - Heavyside function. From (16), (15) one can see that the spatial intensity distribution possesses a width

$$\Delta\xi=\frac{\sin\theta_0(p_0-q_0)}{2\sigma p_0}. \tag{18}$$

In our case this value is $6.9\cdot10^{-5}$ cm. The spatial intensity profiles calculated with the use of relation (14) for several different energies are shown on fig. 3. Since the relation (15) has a θ-function the intensity will have a nonzero value in a phase space $(\xi,\Delta\lambda/\lambda)$ region comprised by the parabola

$$\xi_0=\frac{6p_0^2q_0ctg\theta_0}{(p_0-q_0)^2}(db_1/\pi-tg\theta_0\Delta\lambda/\lambda)^2. \tag{19}$$

The spectral intensity calculated with formula (14) for three deviation rates is shown on fig. 4. Scrutinizing these spectral shapes one can find a minimal spectral resolution

$$\frac{\Delta\lambda}{\lambda}=6.15\cdot10^{-5}. \tag{20}$$

The value (20) adds up into the rate of an error of intensity determination in a curved crystal spectrometer. This value is not similar to the resolution of a flat monochromator as far as the outgoing wavefield in a curved crystal is focused in addition.

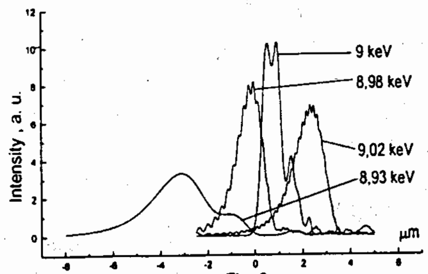

Fig. 3

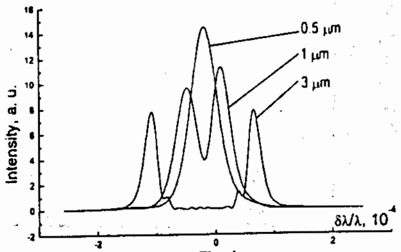

Fig. 4

Polychromatic focus

From fig. 2 one can infer that the intensity of a wave with energy E is distributed over a small area in the vicinity of a point described by deviation Δ from (3). Thus altering the expression (15) we can define an intensity in the polychromatic focus as follows

$$I=|\mathrm{E}(\xi-\Delta,0,E)|^2. \tag{21}$$

For a given deviation value ξ the quantity (21) determines a spectral intensity in the polychromatic focus. The intensity (21) is a characteristic to be measured at the distance q_0 from the crystal with the $\vec{k}_h$-direction normal to a detector (fig. 2). The spectral intensity calculated for several deviation rates by formulas (15), (21) is depicted on fig. 5. The characteristic curve (3) in the phase space (ξ, E) is a set of points in which the intensity (21) takes its maximum.

The intensity on a detector. Source-size effect

The wavefront of a single energy is subject to focusing once the reflecting crystal is curved. The wavefield of each energy moves convergently until the focus region is reached and divergently when the focus region is passed through. Thus the intensity measured by detector (D) will not coincide with the intensity in a focus. In this case one

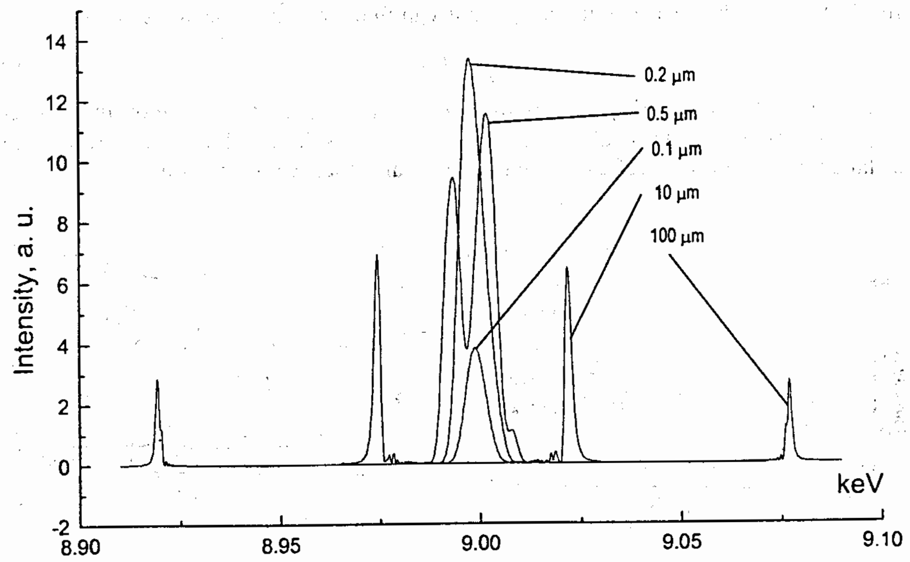

Fig. 5

can evaluate the integral (13) by means of the stationary phase method. The amplitude on the detector will obey the proportionality (the central ray is envisaged again)

$$E(\xi) \propto L[(b_1 - k\sin\theta_0\Delta\theta + \frac{k\sin\theta_0}{2}(1/q_0 - 1/p_0)(1 + q_0/d_0)\xi)/\sigma], \qquad (22)$$

where d_0 is the distance from the focus point to the detector. We have then for a spatial width and a spectral resolution on the detector $(d_0 = q_0)$

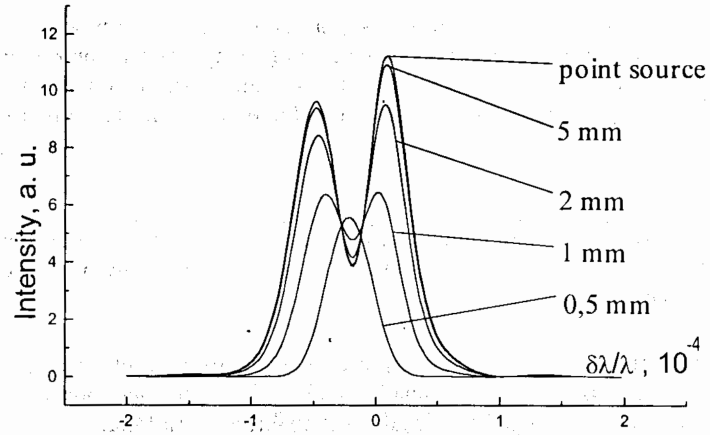

Fig. 6

$$\Delta\xi = \frac{p_0 q_0}{p_0 - q_0}\frac{\sigma}{k\sin\theta_0} = 23.8\mu m, \quad \frac{\Delta\lambda}{\lambda} = \frac{\sigma\cos\theta_0}{k\sin^2\theta_0} = 6.2\cdot 10^{-5}. \qquad (23)$$

Spatial and spectral intensity distributions keep similarity and have the shape of a rocking curve of ideal crystal. The spectral resolution retains its value for every distance d_0 where intensity is measured.

Let us consider an influence of the source size on the intensity structure. One may show that after this influence is taken into account the integrand in (13) acquires an additional factor

$$\exp[-\frac{x^2}{4w^2}],$$

where w is a linear size of X-ray source. The source-size influence is demonstrated on fig. 6 with the use of a spectral intensity ($1\mu m$ curve from fig. 4) in a monochromatic regime. The damped intensities from sources of several kinds are exhibited.

Inferences

The calculations of the value of X-ray radiation intensity created by a spectrometer in an energy-dispersive working regime are carried out in the paper. Such spectrometer provides an illumination of a space region of size ~1 mm with X-ray of all energies from some fixed interval $(E_0 - \Delta E, E_0 + \Delta E)$. Selection of a given energy interval and creation of illuminated area (polychromatic focus point) are realized by means of a spectrometer's curved crystal plate. The present consideration remains restricted by the case of symmetrical reflection. The value of intensity in the focus is considered to be a function of three variables. The first variable allows this function to define a spectral intensity when the spectrometer does work in a monochromatic regime. The spectral structure is concentrated in a range of relative wavelength variation of the order of 10^{-4} in the proximity of a given value λ_0 and is given rise by the diffraction mechanism of X-ray reflection. The second variable stipulates a spatial distribution of intensity. The third one makes the intensity vary when the energy ranges within the bandpass.

Proceeding from the quality of distributions of these three types obtained one can deduct the following inferences. The maximal value of a spectral resolution registered by detector is

$$\frac{\Delta\lambda}{\lambda} = 6.2 \cdot 10^{-5}.$$

This quantity is presumed to be built up by diffractive and focusing reflective properties of the crystal. The minimal spatial resolution is attained for energy of the central ray and has the value

$$\Delta\xi = 1\mu m. \tag{24}$$

The kinematics criteria of the focusing (2) in case of symmetrical reflection gives rise to formation of a focus spot with a very small size (24). Since for every noncentral ray the criterum (2) appears to be slightly violated the spatial resolution for corresponding energies increases (fig. 3). Because the focused intensity is always of different peculiarities than the intensity anywhere off the focus the intensity registered by detector has a worse spatial resolution. The data obtained in the paper conform with results based on the kinematics approach [2], [9].

References

[1] V. Aksenov et al. JINR preprint R14-96-502 (1996) (in Russian).
[2] H. Tolentino et al. J. Appl. Cryst., **21**, 15 (1988).
[3] M. Born, E. Wolf, Principles of optics, Nauka, M., 1970 (in Russian).
[4] S. Takagi, Acta. Cryst., **15**, 1131 (1962).
[5] A. Afanasiev, V. Kohn, Acta. Cryst., **A27**, 421 (1971);
Z. Pinsker, Dynamic scattering of X-rays in ideal crystals, Nauka, M., 1974 (in Russian).
[6] S. Lekhnitsky, Elasticity theory of anisotropic body, Gostekhizdat, M., 1950 (in Russian).
[7] F. Chukhovskii, Metallofizika, **3**, 3 (1981) (in Russian).
[8] K. Gabrielian, F. Chukhovskii, Z. Pinsker, ZhETF, **50**, 1641 (1980) (in Russian).
[9] H. Tolentino et al. NIM, **A289**, 307 (1990).

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