

On the reflection properties of germanium and quartz focusing crystals

E O Baronova¹, I A Subbotin¹ and A A Lomov²

¹ National Research Center “Kurchatov Institute”, Kurchatov Square 1, Moscow 123182, Russia

² Physical Technological Institute of the Russian Academy of Sciences, Nahimovskii Prospekt 36/1, Moscow 117218, Russia

E-mail: baronova04@mail.ru

Abstract. A diffraction component of angular resolution of spherical Q2131 and Ge422 crystals was calculated, measured in experiment and the good agreement was found. Simulation of geometrical component of angular resolution of bent crystals was also performed and the code is designed to simulate reflectivity of the concave crystals. Such code is also aimed to estimate the resolution of focusing x-ray spectrometers.

1. Introduction

Concave x-ray crystals are used to focus x-radiation from synchrotrons, they are key elements of Johann and Johansson spectrometers [1, 2] as well as x-ray microscopes [3–5] implemented in plasma diagnostics. Resolution, spectral range, dispersion, light power of various focusing schemes based on concave crystals depend on the optical aberrations [6, 7], diffraction properties of the crystals [8, 9], geometrical parameters of the experiment. Reflection of x-rays from the crystals happens according to the Bragg’s law:

$$2d \sin \theta = n\lambda, \quad (1)$$

where d is an interplanar crystal distance, θ is diffraction (Bragg) angle, λ is wavelength of radiation, $n = 1, 2, \dots$ is reflection order. It is also known that reflection is realized within angle interval $\delta\theta$ and the corresponding wavelength interval is $\delta\lambda$. The relation between $\delta\theta$ and $\delta\lambda$ follows from the equation (1):

$$\frac{\delta\lambda}{\lambda} = \cot \theta \delta\theta. \quad (2)$$

Equation (2) is universal, it works for arbitrary origin of $\delta\theta$ or $\delta\lambda$. $\delta\theta$ and $\delta\lambda/\lambda$ are angular and spectral resolutions correspondingly. The angular resolution is usually associated with two values: diffraction broadening (DB), $\Delta\theta_d$ that is the interval of Bragg angle where the reflectivity of a crystal differs from zero, and the geometrical one $\Delta\theta_g$, which depends on the geometry of instrument. Diffraction broadening is associated with the nature of x-ray reflection from crystals and characterized by the rocking curve (RC) of the crystal, while the geometrical (or optical) broadening (GB) in case of concave crystal is determined by optical aberrations. As will be shown below in this paper the diffraction broadening $\Delta\theta_d$ becomes larger for bent crystal compared to the flat one.



Implementation of any equipment assumes that it should be calibrated and characterized, where the latter means that both DB and GB are measured and/or calculated. In this paper we simulated the GB component of angular resolution of spherical crystals. We likewise studied the DB, namely we calculated the shape of RC numerically along Takagi–Taupin equations [10,11] and compared with experimental data taken for quartz and Ge crystals. Our codes make it possible to estimate resolution of focusing crystals and let to choose the adequate x-ray dispersive elements for the variety of experimental conditions.

2. Calculation of optical broadening

We assume that the crystal is a part of a spherical surface of radius R . If such crystal is used in focusing spectrometer the scheme is called Johann scheme. In this scheme if the point-like source is placed on the circle $r = R/2$ (Rowland circle), reflected radiation is well enough focused in conjugate point D situated on the same circle if Bragg condition is satisfied. The width of line situated on the Rowland circle is characterized by diffraction and geometrical defocusing. First we consider solely geometrical component $\delta\theta_{\text{geom}}$ which depends on geometrical features of the instrument.

Generally discussing, the geometrical part of spectral resolution of focusing schemes depend on the crystal geometrical parameters (length, width and curvature radius), detector space resolution, source size, optical distance, *etc.* Geometrical (or optical) aberrations are defined by the size of crystal surface which reflects the lines. The size of this surface might be equal or less than crystal size, depending on source size, optical distance, sizes of slits.

The reliable approximate expressions were suggested in [6] to estimate aberrations of cylindrical, spherical and toroidal crystals both Johann and Johansson types. To calculate optical aberrations we follow the analytics suggested in [6] where the Cartesian coordinates of arbitrary point C on a crystal are expressed in spherical coordinates by the formulas (γ and φ are polar and azimuthal angles, respectively):

$$x_c = R \sin \gamma \sin \varphi; \quad y_c = R \sin \gamma \cos \varphi; \quad z_c = R \cos \gamma. \quad (3)$$

The detector coordinates of point D for both spherical schemes are:

$$x_d; \quad y_d = R \sin \theta_0 \cos \theta_0; \quad z_d = R \cos^2 \theta_0. \quad (4)$$

The unit vector $\vec{n}$, normal to a crystalline plane in the point C (in this case to a surface of a crystal) is as follows:

$$\vec{n} = \{\sin \gamma \sin \varphi; \sin \gamma \cos \varphi; \cos \gamma\}, \quad (5)$$

and the vector $\vec{DC}$ for Johann spherical crystal is:

$$\vec{DC} = \vec{r} = \{R \sin \gamma \sin \varphi; R(\sin \gamma \cos \varphi - \sin \theta_0 \cos \theta_0); R(\cos \gamma - \cos^2 \theta_0)\}. \quad (6)$$

Now Bragg angle θ at the point C is determined from the relation:

$$\sin \theta = \cos(\vec{n}, \vec{r}) = \frac{(\vec{n} \cdot \vec{r})}{|\vec{r}|}, \quad (7)$$

where $(\vec{n} \cdot \vec{r})$ is a scalar product of vectors $\vec{n}$ and $\vec{r}$. Using (3)–(7) the dependence between the position of arbitrary point C and $\delta\theta = \theta - \theta_0$ is found and the code OptBroad is constructed, which generates the distribution of intensity within particular spectral line assuming that width of line is broadened by optical aberrations. The result is shown in figure 1 for the crystal with the length 50 mm, height 10 mm and the grazing angle 45° . Figure 1 testifies that red wing of spectral line in Johann type device with spherical crystal is much more broadened, compared to

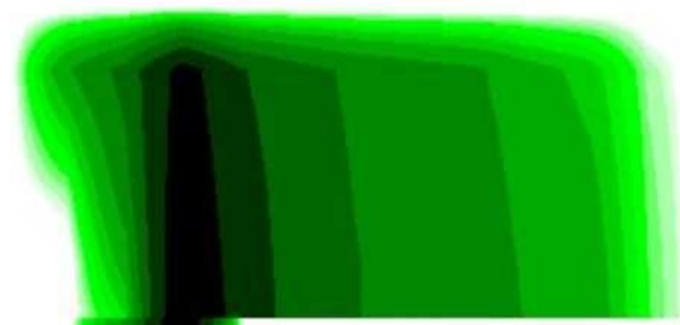
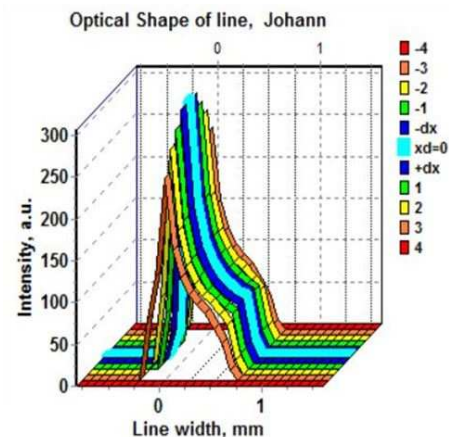


Figure 1. The image of spectral line and its densogram, crystal length 50 mm, crystal height 10 mm, Bragg angle 45° .



the blue wing. Our simulation has shown that the broadening decreases with decreasing crystal length while it does not depend on crystal height in a compliance with the results, presented in [6]. It means that light power of spectrometer with spherical crystal can be increased by using crystal with larger height. Obviously the angular resolution, considered in this section, does not depend on crystal type (germanium, mica, quartz, *etc*), but depends on crystal sizes and on the angle at which radiation is reflected.

3. Calculation of diffraction broadening

The diffraction component of angular resolution $\delta\theta_{\text{dif}}$ of bent crystal is determined by the width of RC, which is not yet known in detail contrary to much better studied RCs of flat crystals. It obviously comes from experiment [8,12] and calculations [10–12] that RC of thick flat crystals becomes much wider after bending and the corresponding integrated reflectivity increases [12]. It should however be noted that reliable simulations of the shape of RC is not a trivial task, partially because of their results should be verified and matched with experimental one while fabrication of bent crystals is unique and expensive procedure. Anyway to make a right choice of bent crystal for someone's experimental geometry it would be ideal to have good theoretical data on the RC of wide list of bent crystals.

There are two main approaches to calculate the shape of RC of bent crystals: multi lamellar approach or ML [13,14] and the T–T approach, both are based on the dynamical theory of x-ray diffraction. In ML approach the crystal is represented by the set of flat layers disoriented with respect to each other, transportation of radiation through the layers are counted by boundary conditions. The “deformation gradient” is represented by parameter c , which is a reduced curvature.

Takagi [10] derived from Maxwell equations and finally suggested to solve the system of two differential equations for incoming and reflected x-ray beams with the goal to calculate reflection–rocking curves of bent crystals. The further development was made by Taupin [11] who managed to find the single differential equation for shape of rocking curve of stressed bent crystals. Both ML and T–T methods are approximate and their results differ especially for relatively small curvature radiuses [13].

We computed the shape of RC of bent germanium and quartz crystals in the Bragg case using the dynamical theory of x-ray diffraction in the frame of T–T theory [10,11]. The gradient of deformation due to bending and difference in the Bragg angles along the crystal surface due to crystal curvature are counted. For the thick crystal (thickness essentially exceeds the penetration

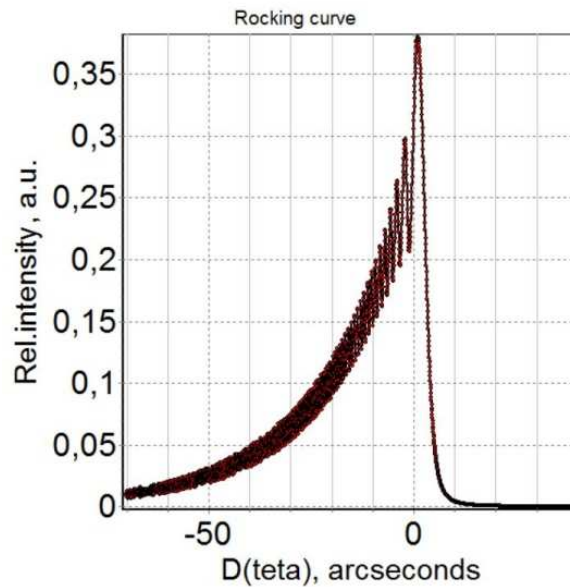


Figure 2. Shape of RC for Q2131 (where 2131 are Miller indexes of quartz wafer), $R = 672$ mm, $\lambda = 1.54$ Å.

depth) the reflection curve $R(y) = XX^*$ of elastically bent crystal was determined by solving the following differential equation:

$$iC \frac{dX}{dy} = X^2(1 + ik) - 2X(u + ig) + (1 + ik), \quad (8)$$

X is the amplitude of the diffracted radiation at the crystal surface (it is in general complex and normalized to unity for the incoming radiation), y is the normalized angular variable of the reflection curve, g represents the absorption of incoming radiation while k represents the absorption of the scattered one. Exactly the same formalism is given by Taupin in [11]. The deformation C which arises due to elastic bending is expressed as:

$$C = \frac{2\lambda \sin \theta_B}{\pi(\chi_h \chi_{\bar{h}})} \left[\frac{1}{R_x} \cos^2 \theta_B - \left(\frac{1}{R_x} + \frac{1}{R_y} \right) \frac{\sigma}{1 - \sigma} \sin^2 \theta_B \right]. \quad (9)$$

R_x is the bending radius in the diffraction plane while R_y is the one in perpendicular to the diffraction plane, θ_B is the Bragg angle, $\chi_h, \chi_{\bar{h}}$ are the Fourier components of the electric susceptibility, λ is the x-ray wavelength and σ is the Poisson ratio. The first term in equation (9) takes into account the pure geometrical bending, the second one takes into account the change of the lattice plane distance due to bending and depends on the elastic properties of the crystal. Rocking curve for Q2131 (where 2131 are Miller indexes of quartz wafer), and curvature radius $R = 672$ mm, is given in figure 2. Figure 2 shows strong asymmetry and oscillations on the left wing of the rocking curve. It was found in [11] and then confirmed in [12,13] that those effects are necessary features of the rocking curves of bent crystals. We note, that oscillations on the left wing of the curve cannot be resolved in the experiments.

4. Results of experiment and comparison with simulations

Figure 3, 4 show the shapes of RC obtained in experiment for quartz2131 (figure 3) and Ge422 (figure 4) crystals. The shape of RC of Ge crystal, $R = 216$ mm, size 15×50 mm², was measured

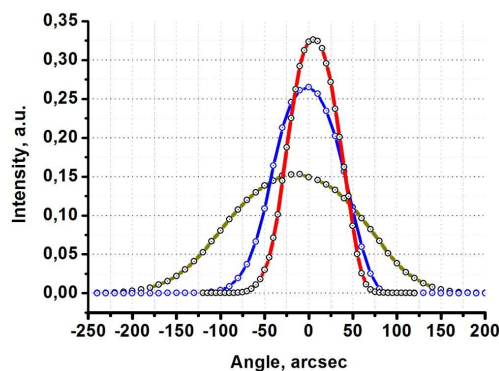


Figure 3. Shape of RC for Q2131, $R = 250$ – 672 mm, Bragg angle $\theta_m = 32.996^\circ$, angular divergency $7''$.

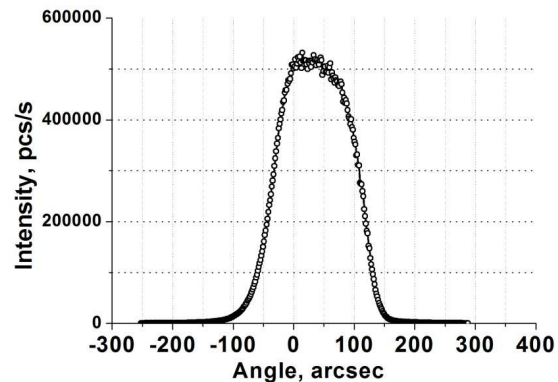


Figure 4. Shape of RC for Ge422, $R = 216$ mm, Bragg angle $\theta_m = 41.834^\circ$, angular divergency $12''$.

with K_α of Cu radiation by Rigaku Smart Lab diffractometer, at high voltage $U = 45$ kV, equipped by Bartels four crystal Ge220 monochromator with divergency $12''$. Diameter of focal spot was equal to 400μ , slit size was equal to 100μ . Angle with monochromator was equal to 22.648° , angle with the concave sample was equal to 41.834° while source to crystal distance was equal to $L = 0.56$ m. Measured half a width of RC (HWRC) is equal to $150''$. Shape of RC of quartz crystals ($R = 250$ mm, $R = 500$ mm, $R = 672$ mm), was measured by Triple Crystal Spectrometer with conventional copper x-ray tube at $U = 30$ kV equipped by slit Ge 400 3-fold monochromator (Bragg angle $\theta_m = 32.996^\circ$, angular divergency $7''$). Bragg angle with concave samples was 30.017° . Source to crystal distance was 0.55 m, source size was equal to 50μ m, slit size was equal to 100μ m. The error in measured angle was about 2 arcseconds, the error in measured diffraction amplitude was below 5%.

The measured shapes of RC of Ge and quartz are the convolutions of the following values:

$$\beta^2 = \beta_m^2 + \beta_0^2 + \phi^2 + D^2, \quad (10)$$

where β_m is the widening due to monochromator, β_0 is the widening due to bent crystal sample, $\phi = (L\Delta\lambda \tan \theta_m)/(R\lambda \sin \theta_B)$ is the widening due to radiative width of line, D is the widening due to dispersion, which in case of bent crystal is [12]:

$$D = \frac{\Delta\lambda}{\lambda} (-(k-1)/2k \tan \theta_m + \tan \theta_0), \quad (11)$$

where $k = 1/(1 - 2 \tan \theta_0 / \tan \theta_m)$, while θ_m , θ_0 are Bragg angles with monochromator and bent crystal respectively [11, 13].

The widths of the rocking curves of bent crystals, extracted from the convoluted curves, taken in experiment, are given in table 1. Same widths, calculated by our code, based on T–T model, and the widths of bent crystals, calculated by ML approach, realized in code XOP [15] are also listed in the table 1. Widths for flat crystals, taken from [16] are presented for the comparison.

It is seen, that narrow rocking curves of flat crystals ($1.8''$ and 6 – $7''$ for Q2131 and Ge422, correspondingly) become wider after bending, and the less the curvature radius the wider are the rocking curves. A good agreement between the simulated and experimental values testifies that T–T code works enough adequately and can be used for the estimation of rocking curve shapes of bent crystals. Table 1 likewise shows the resolution of focusing type spectrometers, based on the studied elements. We note that slit size was equal to 100μ , hence optical aberrations are

Table 1. Half a width of RC in arcseconds for bent and flat crystals.

Bent sample	Experiment	T–T code	ML, code XOP	flat sample	$R = \delta\lambda/\lambda$
Q2131, R215	34	37	36	1.8	3×10^{-4}
Q2131, R250	18	19	19		1.5×10^{-4}
Q2131, R672	12.8	14	14		1×10^{-4}
Ge422, R216	24	20	21	6–7	1.3×10^{-4}

negligibly small ($\delta\theta_{\text{geom}} < 0.1''$) and $\delta\theta_{\text{dif}}$ was key value to estimate the resolution provided by those elements when used at $\lambda = 1.54 \text{ \AA}$. Such the optimal resolutions might be achieved in the experiments with point like sources, where the condition $\delta\theta_{\text{dif}} \gg \delta\theta_{\text{geom}}$ is easily met, since size of the crystal reflecting spectral line is small enough (except the situation when source is placed on the Rowland circle). For the extended sources at arbitrary distances and for the point-like sources placed on the Rowland circle $\delta\theta_{\text{geom}}$ should be counted because the widening due to optical aberrations cannot be neglected.

5. Summary

The code is designed to predict reflectivity of bent crystals in various experimental geometries. A shape of RC of concave crystals (diffraction component of angular resolution) was simulated within Takagi–Taupin equations. It was shown that rocking curve becomes wider for smaller crystal radii. To check this statement the rocking curves of bent crystals were also measured for spherically bent quartz 2131, $R = 250 \text{ mm}$, $R = 500 \text{ mm}$, $R = 672 \text{ mm}$ and for spherically bent Ge422, $R = 216 \text{ mm}$. The results of calculations are well matched with experimental one. The satisfactory coincidence of our results was found with those, generated by code XOP.

Our code also includes calculations of geometrical component of angular resolution. The important practical application of the code is demonstrated: analysis is done for resolving properties of list of bent crystals used in focusing x-ray spectrometers, imaging experiments, etc.

References

- [1] Johann H H 1931 *Z. Phys.* **69** 185
- [2] Johansson T 1932 *Naturwissenschaften* **3** 320
- [3] Aglitskiy Y, Karasik M, Velikovich A L, Serlin V, Weaver J L, Kessler T J, Nikitin S P, Schmitt A J, Obenschain S P, Metzler N and Oh J 2012 *Phys. Plasmas* **19** 102707
- [4] Koch J A, Landen O L, Barbeea T W, Celliers P, Silva L B D, Glendinning S G, Hammel B A, Kalantar D H, Brown C and Seely J 1998 *Appl. Opt.* **37** 1784–1795
- [5] Baronova E O, Garanin P V, Gidkov N V, Stepanenko M M and Suslov N A 2004 *Instrum. Exp. Tech.* **2** 139–142
- [6] Stepanenko M M 2009 *Plasma Devices Oper.* **17** 191–200
- [7] Baronova E O and Stepanenko M M 2001 *Nucleonika* **46**
- [8] Lider V V, Baronova E O and Stepanenko M M 2001 *Crystallogr. Rep.* **46** 341
- [9] Pereira N R, Macrander A T, Hill K W, Baronova E O, George K M and Kotick J 2015 *Rev. Sci. Instrum.* **86** 103704
- [10] Takagi S 1962 *Acta Crystallogr.* **15** 1311–1315
- [11] Taupin D 1964 Ph.D. thesis Univ. Paris, France
- [12] Uschman I, Foerster E, Gabel K and Holzer G 1993 *J. Appl. Crystallogr.* **26** 405–412
- [13] Caciuffo R, Melone S, Rustichelli F and Boeuf A 1987 *Phys. Rep.* **152** 1–71
- [14] Gronkowski J and Malgrange C 1984 *Acta Crystallogr. A* **40** 507
- [15] URL <http://www.esrf.eu/Instrumentation/software/data-analysis/xop2.4>
- [16] URL <http://sergey.gmca.aps.anl.gov/>