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BENT CRYSTAL ANALYZER WITHOUT GROOVES FOR INELASTIC SCATTERING -- FIRST EXPERIMENTAL RESULTS*

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Bent crystal analyzer without grooves for inelastic scattering -- first experimental results

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ABSTRACT

A new design of a bent crystal analyzer for high energy resolution inelastic X-ray scattering has been recently proposed (Kushnir and Popovici, Rev. Sci. Instr., in press). It has been theoretically predicted that an analyzer with reflecting planes at a certain angle with respect to a crystal surface, bent with two different radii of curvature, will have the same energy resolution as a perfect crystal. The first experimental measurement obtained at the Advanced Photon Source of a bandwidth of such an analyzer is presented. The overall energy resolution of the analyzer and monochromator observed with a narrow beam is equal to 16.4 meV (FWHM) at 13.84 KeV.

Keywords: inelastic X-ray scattering, backscattering, bent crystals, focusing, synchrotron radiation instrumentation.

1. Introduction

An analyzer for x-ray inelastic scattering has to provide milli-eV energy resolution while dealing with a divergent spherical wave scattered by a finite size specimen (on the order of 1 mm) at a distance of 1-3 m from the analyzer. Thus the analyzer must have a 0.3-1 milliradian angular acceptance. Such a large angular acceptance can be achieved by using X-ray diffraction in the backscattering regime — i.e., at a scattering angle close to 180°. [1-4] This means that the atomic planes of the crystal have to be perpendicular to the incoming rays. Since a wave scattered by a specimen is a spherically divergent wave, the analyzer crystal therefore has to be bent. The problem is that the bending stress broadens the energy resolution curve of the crystal (compare Fig. 1a and b). The known solution for this problem is to make the analyzer with grooves thus eliminating the stress on the top surface of the crystal.[5] An analyzer made of separate Si crystals has been also demonstrated.[6]

Recently a new design of a bent crystal analyzer without grooves has been proposed.[7] The idea is to find a direction in a bent crystal in which the interplanar spacing stays constant, and thus bending does not broaden the energy resolution curve. The main features of this new design are:

a) diffraction planes are not parallel to the crystal surface;
b) the crystal is bent with two different radii of curvature.

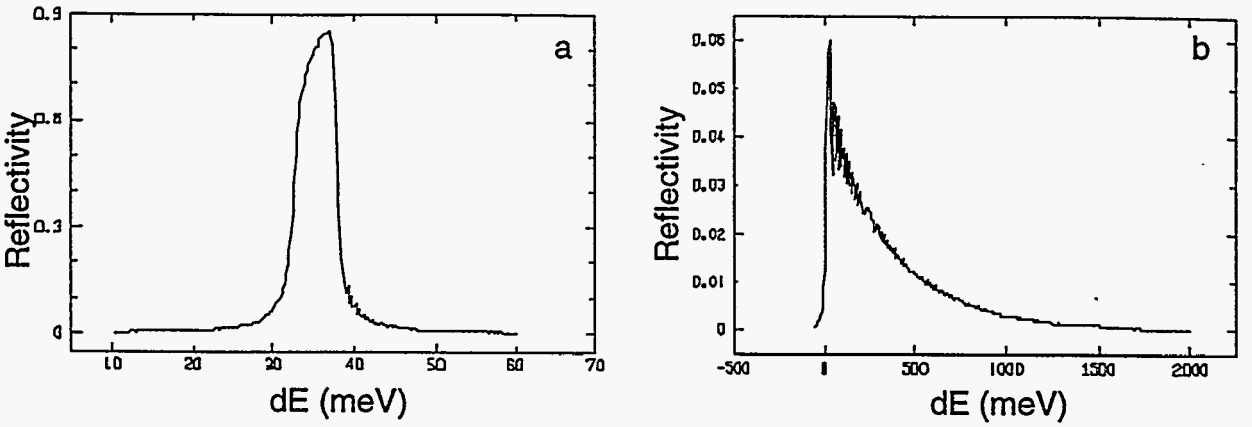

Fig. 1. Theoretical reflectivity curves of a perfect silicon crystal in exact backscattering, reflection Si(777) at photon energy E=13.84 KeV: a) perfect crystal -- FWHM = 5.04 meV, b)spherically bent crystal (R=3 m) -- FWHM = 200 meV.

The theory and concrete parameters of such analyzers are discussed in detail in Ref. 7. Here we will outline briefly the main idea of such analyzer.

2. Principle of a high energy resolution bent analyzer

Let us consider diffraction of an X-ray beam on a crystal having the shape of a thin plate and uniformly bent with two different radii of curvature (Fig. 2).

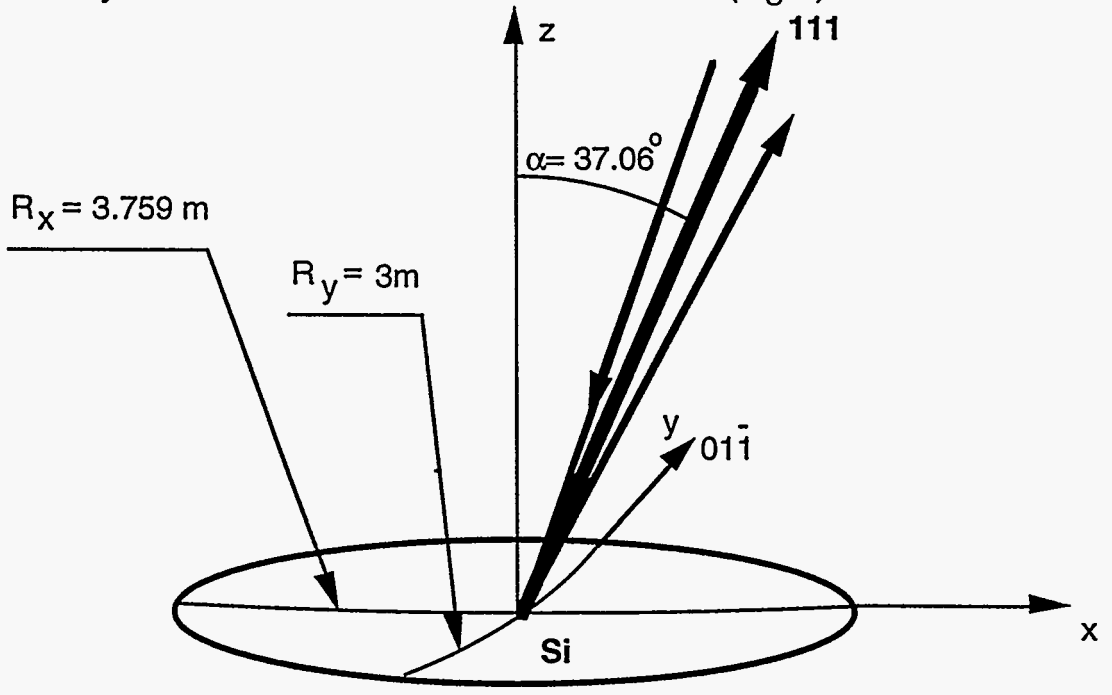

Fig. 2. Orientation and radii of curvature of the proposed analyzer (from reference 7).

It will be clear from the discussion below that it is impossible to build such an analyzer by using diffraction from the atomic planes parallel to the crystal surface. Therefore, we shall consider a more general case of diffraction from atomic planes making an angle α with the crystal surface. Let us also suppose that the crystal is bent with two different radii

of curvature R_x and R_y. In order to insure that all beams reflected by the crystal will be focused back to the source, it is necessary to satisfy the focusing condition:

$$R_y = R_x \cos(\alpha). \qquad (1)$$

This means that the focusing length of the analyzer is the same in the two perpendicular planes containing the diffraction vector [in Fig. 2 these planes are: a) the x-z plane, and b) a plane containing the vector <111> and the y-axis].

Since Bragg's law for backscattering diffraction with a Bragg angle $\theta = \pi/2$ reduces to:

$$\lambda = 2d_{hkl}, \qquad (2)$$

the only parameter that influences X-ray diffraction is the local interplanar distance d_{hkl}. It is easy to see from Fig. 3 that, due to the non-zero Poisson ratio of the material, the d-spacing *decreases* in the z direction normal to the crystal surface ($\alpha \approx 0$), while, for a beam coming at a grazing angle ($\alpha \approx \pi/2$) close to x axis, the d-spacing *increases*.

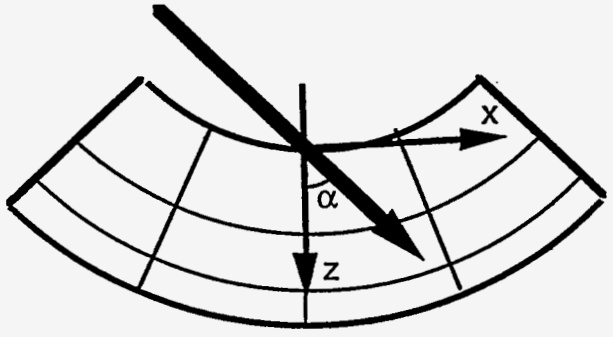

Fig. 3. The *d*-spacing gradients in a bent crystal in different directions: *d*-spacing decreases along the direction z, *d*-spacing increases along x, *d*-spacing is constant along the direction making angle α with z in x-z plane (planes normal to this direction are not shown).

Therefore, an angle of asymmetry α exists for which the interplanar distance between atomic planes normal to that direction is constant.

In an isotropic approximation, the effective deformation in the direction making angle α with z-axis in x-z plane is equal to:

$$\varepsilon_\alpha = \left[\sin(\alpha)^2 \frac{1}{R_x} - \cos(\alpha)^2 \frac{\nu}{1-\nu} \left(\frac{1}{R_x} + \frac{1}{R_y} \right) \right] z. \qquad (3)$$

Here ν is the Poisson ratio of the crystal; for Si, ν=0.278. [8] There is an angle

$$\alpha = \operatorname{atan}\left(\sqrt{\frac{\nu}{1-\nu}\left(1+\frac{R_x}{R_y}\right)} \right) \qquad (4)$$

giving zero deformation and therefore constant *d*-spacing along the beam path inside the crystal. Combining this equation with the focusing condition (1), one gets for Si, α=43.8°.

For the general case of an anisotropic crystal, instead of expression (3), we use a previously defined value of a *strain gradient* : [7,9]

$$B = \frac{\lambda}{2\pi\chi_{hkl}^{\,2}} \left[\frac{C_x(\theta,\alpha)}{R_x} + \frac{C_y(\theta,\alpha)}{R_y} \right], \qquad (5)$$

here λ is the radiation wavelength, χ_{hkl} is the Fourier component of crystal polarizability, $C_x(\theta,\alpha)$ and $C_y(\theta,\alpha)$ are dimensionless coefficients depending on the geometry of diffraction and, for an anisotropic crystal, on crystal orientation.
B is a dimensionless parameter originally introduced by Petrashen' and Chukhovskii; [10-11] it is proportional to the slope $d\varepsilon_\alpha/dz$ in expression (3) and must be equal or close to zero. We have shown previously,[7] that it is enough to satisfy the condition $|B| < 0.1$ in order to have the energy resolution of a bent crystal equal (or close) to that of a perfect crystal.

Taking into account the fact that silicon is highly anisotropic, the value of α calculated in eq. (4) above now depends on the orientation of the crystal. The real geometry of the analyzer proposed previously[7] is shown in Fig. 2, and the asymmetry angle α is equal to 37.06°. For this particular orientation, the expression for the strain gradient becomes:

$$B = \frac{\lambda}{2\pi\chi_{hkl}^{\,2}} \left[\frac{-0.1224}{R_x} + \frac{0.0897}{R_y} \right]. \qquad (6)$$

The following radii of curvature:

$$\begin{aligned} R_x &= 3.759m \\ R_y &= 3m \end{aligned} \qquad (7)$$

satisfy both the focusing condition (1) and the condition of the strain compensation B=0. Here one of the radii can be chosen arbitrarily, and we have chosen $R_y = 3m$ as a specimen-to-analyzer distance based on the length of the Huber goniometer 2θ arm.

3. Analyzer Design

A schematic of the analyzer is shown in Fig. 2. A thin silicon plate has to be bent to a concave shape with two main radii of curvature given by expression (7). In order to keep the strain gradient reasonably small ($|B|<0.1$), the orientation of the crystal of $\alpha=37.06°$ has to be kept within an accuracy of $0.03°$.

The crystal has been cut with a diamond blade from a boule with a <110> axis and then oriented within 0.01° accuracy while polishing. The distorted layer produced by polishing has been removed by etching in a solution of 5%HF + 95%HNO_3 for 3 minutes at room temperature. The quality of removal of the distorted layer was monitored by measuring rocking curves Si(220) with MoK_α radiation. After polishing, the final dimensions of the crystal were: diameter = 100 mm, and thickness = 0.55 mm.

A concave pyrex glass substrate with two different radii of curvature has been produced using a computer-operated diamond saw. Two main radii of curvature were as given in equations (7). The crystal has been glued onto a substrate with epoxy under mechanical pressure. The resulting radii of curvature of the crystal were measured with X-rays by scanning the crystal with respect to an incoming X-ray beam and measuring the angular displacement of the rocking curve. The experimentally measured radii are:

$$R_x = 3.36m$$
$$R_y = 3.05m \qquad (8)$$

The differences between the curvatures of the substrate (7) and the curvatures of the crystal (8) are probably due to the fact that, during the process of gluing, we did not apply enough pressure onto the crystal.

4. Experimental results

The energy resolution of the analyzer has been measured at the undulator beamline of the sector 3 of the Advanced Photon Source. The optical scheme of the experiment is shown in Fig. 4.

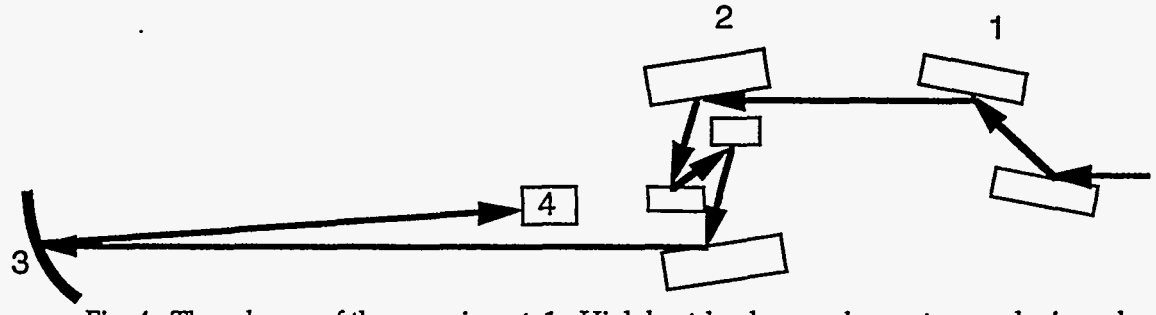

Fig. 4. The scheme of the experiment: 1 - High heat-load monochromator producing a beam with a photon energy E=13.84 KeV and a bandwidth of approx. 3 eV; 2 - high-resolution monochromator consisting of two nested asymmetric channel-cut crystals Si(422) and Si(884), it reduces bandwidth to approx. 5 meV; 3 - analyzer, 4 - detector.

The nested monochromator produces monochromatic radiation with a theoretically calculated bandwidth of 5.04 meV FWHM at the photon energy E=13.840 KeV. The experimentally measured transmission function of the monochromator convoluted with the reflectivity curve of a plain Si(777) crystal as an analyzer has been found to have a width of 7.0 meV (FWHM).

The experimentally measured reflectivity curve of the analyzer is shown in Fig. 5. Its width is 16.4 meV (FWHM). The minimal theoretically possible width of this curve in the case of ideal stress compensation should be the same as for a perfect crystal (7 meV). However, we do know that the experimentally measured radii of curvature of the analyzer (8) were different from the ones required for complete stress compensation (7).

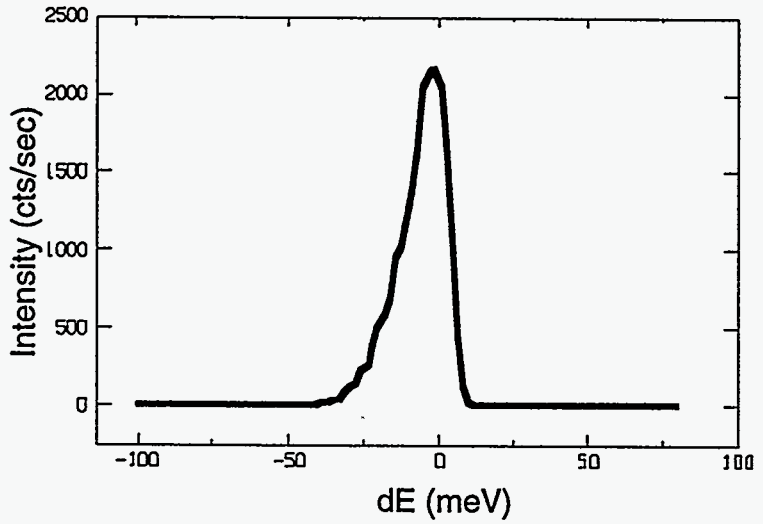

Fig. 5. The experimental energy resolution curve of the scheme shown in Fig. 4, obtained by tuning the inner crystal of the high resolution monochromator.

Based on the measured radii of curvature (8), we have calculated a value of the strain gradient using expression (6) :

$$B = -0.45, \qquad (9)$$

which corresponds to the reflectivity curve shown in Fig. 6a. Fig. 6b shows the same curve convoluted with a theoretical energy profile of the monochromator. The width of this resulting curve is 11.4 meV (FWHM). A discrepancy between this number and the experimentally obtained width of 16.4 meV can be ascribed to a number of reasons, among them a deviation of the diffraction in the real experiment from exact backscattering. Another possible reason is the presence of some residual stress at the back surface of the crystal because of incomplete removal of the distorted layer, and penetration of this stress through the thin crystal to the front surface. Nevertheless, if one tries to bend a silicon plate with (111) orientation to a curvature of R=3 m and use the planes parallel to the surface, one would obtain the resolution curve shown in Fig. 1b with a FWHM of 200 meV. Therefore, the results of this experiment confirm the principle of the proposed design.

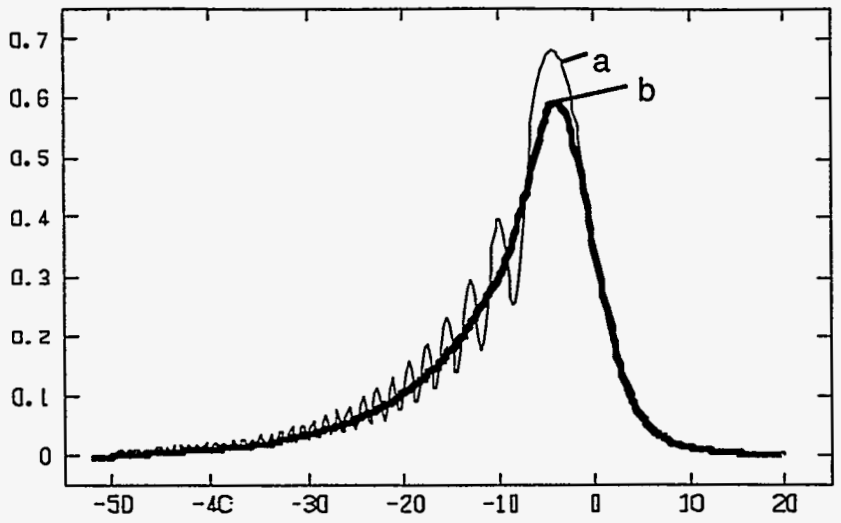

Fig. 6. Theoretical reflectivity curves of the crystal with radii of curvature as given in eq. (8) corresponding to a finite value of the strain gradient B=-0.45: a) reflectivity of the crystal itself, b) the curve *a* convoluted with the theoretical energy profile of the monochromator (FWHM=5 meV). The FWHM of curve *b* is 11.4 meV.

5. Conclusion

We have presented the results of the first experimental test of a new bent asymmetric aspherical analyzer. This new design has certain advantages and disadvantages with respect to the more traditional design of a diced analyzer:

The advantages are:

a) there are no technical difficulties associated with the production of a diced crystal;
b) no area is lost for grooves;
c) there is no limitation on resolution caused by the finite element size of a diced analyzer.

The disadvantages are:

a) it is very difficult technically to achieve the exact shape of a bent crystal, especially in the case of the special nonsymmetric orientation of an anisotropic crystal;
b) the orientation of the crystal has to be very precise - up to 0.1 mrad;
c) the design of analyzer requires very precise knowledge of the elastic constants of silicon;
d) some effective area is lost because of the fact that the analyzer is at an angle with respect to the incoming beam (by a factor of $\cos(37.06°) = 0.8$).

Even though the crystal had radii of curvature slightly different from the values required by theory, the first measurement shows that the underlying idea of stress compensation works: the experimentally measured energy resolution is equal to 16.4 meV on a crystal spherically bent to 3 m. This first result looks very promising, and we will

continue to explore new ways to make the analyzer shape more precise and thus achieve better energy resolution.

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7. References

1. K. Kohra and T. Matsushita, Z. Naturf., **27a**, 484, 1972.
2. O. Brummer and H.R. Hoche, J. Nieber, Phys. Stat. Sol. **a 53**, 565, 1979.
3. A. Caticha and S. Caticha-Ellis, Phys. Rev. **B 25**, 971, 1982.
4. V.I. Kushnir and E.V. Suvorov, Sov. ZhETPh. Lett., **44**, 205, 1986.
5. E. Burkel, *Inelastic Scattering of X-rays with Very High Energy Resolution*, Springer-Verlag, Vol. **125** , 1991.
6. F. Sette, G. Ruocco, M. Krisch, U. Bergmann, G. Masciovecchio, V. Mazzacurati, G. Signorelli, R. Verbrni, Phys. Rev. Lett. 75, 850-853, 1995.
7. V.I. Kushnir, M. Popovici, *Bent Crystal Analyzer without Grooves for Inelastic X-Ray Scattering*, Proc. of the SRI-95 conference at Argonne, Oct. 1995, Rev. Sci. Instr., in press.
8. J.J. Wortman and R.A. Evans, J. Appl. Phys., **36**, 153-156, 1965.
9. V.I. Kushnir and A.T. Macrander, Nucl. Instr. and Meth. **A347**,331-337, 1994.
10 P.V. Petrashen', F.N. Chukhovskii, Sov. Phys. JETP, **42**, 243 , 1976.
11. F.N. Chukhovskii, K.T. Gabrielyan and P.V. Petrashen, Acta Cryst, **A34**, 610-621, 1978.