MASTER

EVALUATION OF FOCUSING
MONOCHROMATORS FOR AN EXAFS SPECTROMETER

S. M. Heald
Brookhaven National Laboratory
Upton, NY 11973

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EVALUATION OF FOCUSING MONOCHROMATORS FOR AN EXAFS SPECTROMETER*

S. M. Heald

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ABSTRACT

Various focusing monochromators are compared and evaluated for use in an EXAFS spectrometer. For a line focus the Johann and Johansson cases are found to be most suitable. Obtaining a point focus is more difficult, with the best case appearing to be a singly bent mirror used in conjunction with the Johann or Johansson crystals.

INTRODUCTION

A typical EXAFS experiment requires $10^6 - 10^8$ counts in each of ~ 200 channels. If laboratory experiments are to be completed in reasonable time spans efficient focusing monochromators are necessary. These must collect the maximum solid angle consistent with an energy resolution $\Delta E/E \lesssim 5 \times 10^{-4}$, and be easily tunable.

In this paper various focusing arrangements are examined and evaluated as to their suitability for EXAFS measurements. Most of the focusing arrangements present here have been reviewed previously,¹⁻³ but not specifically for EXAFS uses. Also, a Monte Carlo raytracing program originally developed for synchrotron x-ray optics has been applied to the focusing monochromators to estimate their resolution and relative intensities for specific cases. The program is particularly helpful in allowing various "non-ideal" geometries to be quickly evaluated.

GENERAL CONSIDERATIONS

In general, the x-rays striking a monochromator have a spread of angles $\Delta\theta_g$. The energy resolution is determined by the convolution of $\Delta\theta_g$ with the crystal rocking curve. For perfect crystals such as Si or Ge the rocking curve is nearly rectangular with width $\Delta\theta_c$ and peak reflectivities near one. For maximum efficiency $\Delta\theta_c$ should approximately equal $\Delta\theta_g$.

For perfect crystals $\Delta\theta_c$ is determined by the choice of crystal reflection. To change $\Delta\theta_c$ for a given reflection the crystal can be made imperfect by suitable surface treatment, or an asymmetric cut can be used. The angular acceptance of a perfect crystal with an asymmetric cut⁴ is

$$\Delta\theta_a = \Delta\theta_c \sqrt{\frac{\sin(\theta_B + \alpha)}{\sin(\theta_B - \alpha)}} \quad (1)$$

*Work performed under the auspices of the U.S. Department of Energy.

where α is positive when the incident beam makes an angle smaller than θ_B with respect to the surface. Both approaches have some disadvantages. Imperfect crystals do not have peak reflectivities as large as perfect crystals, reducing the efficiency of the monochromator over the case where a perfect crystal is used to match $\Delta\theta_g$. To significantly increase the acceptance of an asymmetric crystal α must be quite close to θ_B which is possible only for a limited energy range.

The geometrical angular spread $\Delta\theta_g$ has three major contributions. The first comes from aberrations in the focusing designs. This depends on the particular design used and will be discussed in detail in later sections. The other two are illustrated in Fig. 1 for a flat crystal. The finite source size in (a) means that each point on the crystal sees a spread S/l where $S = S_0 \sin(\beta)$. An important point here is that the take off angle β depends on the part of the crystal being considered. If the horizontal acceptance W_h is large than the extreme rays can see a source width significantly different from that seen by the central ray. In (b) the effect of a finite vertical acceptance W_v is illustrated. In this case the extreme rays striking the crystal out of the horizontal plane make a smaller angle with the surface than the central ray by an amount $W_v^2/8 \tan \theta$.

SINGLY BENT CRYSTALS

By far the best singly bent monochromators from the standpoint of easy tunability are the Rowland circle cases shown in Fig. 2, the Johann⁵ and Johansson^{6,7} crystals. Other designs such as the logarithmic spiral monochromator are difficult to scan in energy.

(a) HORIZONTAL

(b) VERTICAL

Fig. 1. Two sources of angular spread. a) Finite source size; note that s can vary if W_h is large. b) Finite vertical collection angle W_v .

Fig. 2. Two Rowland circle geometries. a) Johann in which the crystal is bent to a radius $2R$. b) Johansson in which the surface of the Johann crystal is ground to the Rowland circle radius R .

For a point source the Johansson case is an exact solution in the plane. For an actual case the only geometrical contribution to $\Delta\theta$ comes from a finite source width and the vertical collection angle as shown in Fig. 1. For the maximum intensity consistent with a given $\Delta\theta$ these two contributions should be approximately equal, although practical constraints on the size of the crystal and height of the focal line often limit the vertical collection. The horizontal collection angle can be as large as possible consistent with the practical constraints on crystal size, and the increase of the apparent source width shown in Fig. 1.

For the Johann case there is an additional geometrical contribution from the horizontal collection angle given by $W_h^2 \cot \theta / 8$. There is also an increase in the focal line width although for EXAFS measurements this is usually not important. Table I compares the Johann and Johansson monochromators with a flat crystal for various cases assuming $S/2R = 5 \times 10^{-5}$. For the case shown geometrical effects dominate and the monochromators reflect only about 10% of the photons incident within the 5 eV bandwidth. Improvement can be gained by increasing the rocking curve of the crystal. One way to do this is to use Ge (400) for which the diffraction width is $\sim 2 \frac{1}{2}$ times wider.

One might also ask if using an asymmetric cut crystal can improve the efficiency. This turns out to be not the case. For example, the source divergence can be reduced by making SC larger than CF in Fig. 2 which would increase the efficiency. However, the appropriate asymmetric crystal would have a reduced acceptance as seen in Eq. (1), and the two affects nearly cancel.

Table I Comparison of Johann and Johansson monochromators for Si (400) at 10 keV. $S/2R$ is assumed to be 5×10^{-5} and the numbers are for an energy resolution of 5 eV. $W_h = 4^\circ$ for the Johansson case assumes $R = 1$ m. and a maximum crystal length of 8 cm.

	W_h	W_v	I
Flat	.0093°	2.9°	1
Johann	1.5°	2.9°	158
Johansson	4°	3.2°	465

POINT FOCUS

In many cases it is desirable to concentrate the x-rays to a small area. For the cases in Table I if $R = 1$ m. and the source is 10 mm high, then the focal line is ~110 mm high. Often it is difficult to make a uniform sample of this height, and make use of all the available x-rays. To reduce the height R can be reduced, but then the source width must be reduced to maintain the resolution.

Doubly bent crystals can be used to focus a point source to a point focus. They are achieved by rotating the geometries in Fig. 2 about the line SF. Again the Johansson case is a perfect solution, and in this case the contribution from the vertical divergence is also eliminated. $\Delta\theta_g$ is determined completely by the finite source size.

There are, however, two problems associated with this approach. First it is difficult to bend suitable crystals to the two radii without breakage. This can be solved by using the approximate geometry of Berreman et al.⁸ with quartz. It consists of first grinding the crystal into a cylindrical shape, pressing it flat, and then bending it into a cylindrical shape in the orthogonal direction. The crystal planes are then in a doubly bent configuration with radii as small as 1000 times the crystal thickness.

The second problem is more serious. The radius of bend about SF changes with Bragg angle, and if the crystal radii are fixed the focusing condition is lost when the energy is scanned. To look at this problem a raytracing study was made of a Berreman monochromator as a function of energy. These results are summarized in Table II and Fig. 3. Figure 3 shows the change in the focal spot size and Table II also gives the characteristics of the doubly bent Johansson geometry. The crystal was assumed to be SiO_2 (203) which has nearly the same characteristics as the Si (400) used in Table I. The source spot was 0.1×0.1 mm (0.1×2 mm at 3° take off angle) which means $R = 1$ m for $S/2R = 5 \times 10^{-5}$. The Berreman crystal is about the same quality of approximation to a doubly bent Johansson as is the Johann for the singly bent case. The Berreman cylinder was oriented along SF which the raytracing showed to be significantly better than the opposite case. The limit of 4° was chosen from a consideration of maximum crystal sizes and $W_h = 0.6$ for the Berreman case was chosen to match the Johansson ΔE .

Table II Comparison of the Berreman and Johansson doubly bent monochromators at 10 keV for SiO_2 (203). Also, given is the Berreman 10 keV monochromator at 9.5 and 10.5 keV. The source size is 0.1×0.1 mm, and $R=1$ m. The 4° limit is not due to aberrations but is estimated to be a practical limit. See Fig. 3 for the actual shape of the focal spot for the Berreman cases.

	E (keV)	W_h	W_v	ΔE (eV)	Focal Spot (mm)
Johansson	10	4°	4°	3.0	0.1×0.1
Berreman	10	.6	4	3.0	0.1×0.1
Berreman	10.5	.6	4	4.8	0.4×12
Berreman	9.5	.6	4	4.7	0.5×14

As seen in Table II scanning the Berreman monochromator does not seriously change its energy resolution, but does greatly increase the size of the focus. This could cause difficulties if the EXAFS sample is not perfectly uniform, and also destroys the major advantage of a point focus. For a truly tunable point focus monochromator the radius must be adjusted as θ is changed.

Fig. 3. Focal spot of a Berreman 10 keV monochromator as a function of energy. For left to right the energies are 9.5, 10, and 10.5 keV. The heights are half those listed in Table II since $W_v = 2^\circ$ rather than the 4° used there.

TWO ELEMENT MONOCHROMATORS

A point focus can also be obtained using two singly bent elements^{1,9} as in Fig. 4. In this case the radii can remain fixed, but scanning in energy is very difficult since the two crystals must be kept accurately oriented with respect to each other. For $\Delta E = 5$ eV the tolerances in θ are a few seconds. The situation is simpler if one of the elements is a mirror. Then the focussing of one element is independent of energy and a Rowland circle type scan can be used for the crystal element.

Fig. 4. Use of two crystals to obtain a point focus.

X-ray mirrors require glancing angles; the critical angle above which reflection no longer occurs is $\sim 0.5^\circ$ for a Pt surface at 10 keV. Some examples of critical angles for other cases are given in Table III. This restricts the collection aperture of the mirror. For example, the cases in Table I have a source to crystal distance of 460 mm for $R = 500$ mm. If a 200 mm mirror were placed midway it could collect an angle of $\sim \theta_c$ or 0.5° for a 10 keV cutoff and 1° for a 5 keV cutoff (assuming the source is now a point). This is still respectable particularly since the full vertical acceptance in Table I will not always be usable.

Table III Examples of critical angles of reflection for various cases.

Element	4 keV	8 keV	16 keV
Si	0.45	0.22	0.11
Ni	0.86	0.22	0.11
Pt	1.20	0.60	0.30

For a point focus the mirror should have an elliptical profile. This is quite easy to obtain by bending an appropriately shaped flat mirror.¹⁰ There is still a difficulty in scanning energy. To maintain a point focus the mirror must be kept equidistant between the

source and crystal which means the height of the source above the plane of the Rowland circle ($h = SC \sin 2\theta \approx m/2$) must be varied. However, the motion is small and such a spectrometer should be much easier to build than the corresponding double crystal case.

If the smallest point focus is not needed then an even simpler arrangement would be to use a parabolic mirror. This would collect the vertical divergence and make the beam parallel. The height of the beam is just the height intercepted by the mirror, which if the mirror is close to the source can be substantially smaller than the height of an unfocused beam at the focus, as much as a factor of 5-10 for the same W_v . Since the final beam is parallel the mirror can be at a fixed distance from the source, and a scan carried out just as for a Rowland circle instrument with the source offset slightly above and inside the circle to account for the mirror reflection. This same scan can be used with the elliptical mirror, but the focus height will change as the scan progresses, although for the same source to mirror distance it will always be smaller than that produced by a parabolic mirror.

SUMMARY

The most crucial restriction on an EXAFS spectrometer is the necessity to easily scan wide ranges of energy. For a line focus a Rowland circle spectrometer using a Johann or Johansson crystal is most convenient. Large solid angles can be collected, and a scan is easily carried out. For maximum efficiency the crystal $\Delta\theta_c$ should be matched as much as possible to the geometrical $\Delta\theta_g$ of the spectrometer.

A point focus is more difficult to achieve. Doubly bent crystals cannot maintain a focus over a range of energy unless one radii is varied. This was demonstrated by a study of the Berreman monochromator geometry. Double crystal devices are another possibility, but are very difficult to scan since the rays must make the same angle with each crystal to within the crystal rocking curve. Probably the best candidate is a mirror-crystal combination used in a Rowland circle instrument. However, its use is probably limited to energies below about 10 keV by the extreme glancing angles required for the mirror.

REFERENCES

1. J. Witz, *Act. Cryst.* **A25**, 30 (1969).
2. B. W. Roberts and W. Parrish, *International Tables for X-Ray Crystallography*, Vol. III (Kynoch Press, Birmingham, 1962).
3. A. Sandström, *Handbuch der Physik*, **30** (Springer, Berlin, 1957), p. 94-124.
4. K. Kohra, M. Ando, T. Matsushita, and H. Hashizume, *Nucl. Instr. and Methods* **152**, 161 (1978).
5. H. H. Johann, *Z. Phys.* **69**, 185 (1931).
6. J. W. M. Dumond and H. A. Kirkpatrick, *Rev. Sci. Instr.* **1**, 88 (1930).
7. T. Johansson, *Z. Phys.* **82** 507 (1933).
8. D. W. Berreman, J. Stamatoff, and S. S. Kennedy, *Applied Optics*

- 16 2081 (1977).
9. T. C. Furnas, Rev. Sci. Inst. 28, 1042 (1957).
 10. J. H. Underwood and D. Turner, Proc. SPIE, 106, 125 (1977).