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A bent Laue analyzer for fluorescence EXAFS detection

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An analyzer for detection of fluorescence radiation was developed for fluorescence XAFS of dilute samples at energies above 13 keV. The analyzer is a bent Laue crystal of logarithmic spiral shape. The K fluorescence lines can be collected with a large solid angle and moderate reflectivity using the asymmetric [111] reflection of silicon crystals of 200 micron thick. Tuning of the analyzer diffraction energies for different samples can be made with one adjustment. Experiment was performed at the APS BioCAT ID beamline using a silicon crystal bent to an average radius of 200 mm to diffract silver fluorescence x-rays. The crystal (35 mm by 120 mm active area) covers a solid angle of 0.1 SR of the sample fluorescence. The measured reflectivity was 10%.

Keywords: crystal analyzer, fluorescence detector, bent crystal

1. Introduction

XAFS spectra can be measured in several experimental modes: transmission mode; fluorescence mode, which is sensitive to intensity of x-ray fluorescence produced as a consequence of x-ray absorption; and electron yield. Fluorescence mode is generally preferred for dilute systems, because the detected signal originates only from the species of interest. One difficulty facing XAFS detectors at the third generation sources is the greatly increased flux of the undulator beam relative to bend magnet beamlines at second generation sources by more than two orders of magnitude. Since solid state detectors already are operated at maximum count rate, they have serious limitations at the new sources.

The increase in flux at third generation sources is accompanied by reduction in beam size, due to the low angular divergence of the beam (if no focusing is done) and the small source size (if demagnifying optics are used). At the APS it is routine to focus beams down to less than 100 micron. The small spot size incident on the sample provides an ideal source of fluorescence x-rays for a crystal analyzer. A practical analyzer should be able to discriminate between fluorescence and elastically scattered background; accept the fluorescence from a source less than 0.2 mm in one dimension; be adjustable to cover a broad range of elements and have a solid angle of data collection comparable to or greater than existing detectors.

Such an XAFS analyzer was developed by adapting a bent-Laue crystal monochromator developed at the NSLS for medical imaging (Zhong et al.). The NSLS monochromator diffracts an area beam of characteristic x-rays having a solid angle on the order of 0.01 SR using dynamical bending of a thin crystal by a four-bar bender. This work describes the design, construction and testing of the XAFS analyzer system to diffract beams with a solid angle of around 0.1 SR for XAFS.

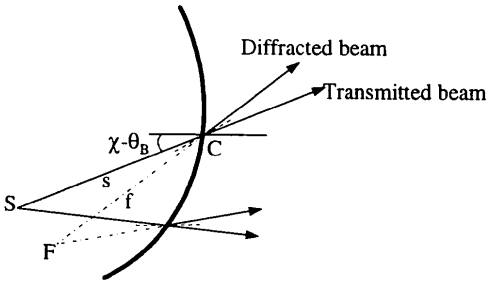

Figure 1
A cylindrically bent Laue analyzer

2. Geometrical Considerations

Bending the crystal solves the mismatch between the narrow angular bandwidth of perfect crystal diffraction and the large divergence of the fluorescence beam from a sample. It enables the diffraction planes to make the same Bragg angle with the incident K fluorescence beam. Bending also increases the angle width of the crystal diffraction to around one milli-radians, so that the device can accept a large fluorescence source size and is not sensitive to bending inhomogeniety (typically on the order of 0.2 mradians).

The geometry for a bent Laue monochromator is shown in Fig.1. The x-rays from a point source at S are reflected by the Bragg planes in the bent crystal and focused at the virtual source F. The asymmetry angle χ is the angle between the crystal surface normal and the Bragg planes used for the reflection of x-rays. The Bragg angle θ_B is the angle between the incident x-rays and the Bragg planes. The distance between the source point and the centre of crystal (C) is s and the distance between the virtual focal point and C is f (negative). If there is no variation of the angle of incidence along the crystal surface, then the whole crystal surface satisfies the diffraction condition for single energy x-rays originating from the source. The condition for diffracting a single energy is (the Rowland condition)

$$s=\rho\cos(\chi-\theta_B) \quad (1)$$

$$f=-\rho\cos(\chi+\theta_B) \quad (2)$$

where ρ is the bending radius of the bent crystal.

The Rowland condition applies for small crystal size accepting a small solid angle of the fluorescence x-rays. A crystal shaped in the plane of diffraction as a logarithmic spiral has the property that any ray from the pole makes the same angle with the crystal surface and thus the underlying diffraction planes (Sakayanagi 1982, Wittry et al. 1993). The logarithmic spiral is represented in polar coordinates by

$$r=\rho_0\cos(\chi-\theta_B)\exp[\tan(\chi-\theta_B)\theta] \quad (3)$$

where ρ_0 is the average bending radius of the logarithmic spiral defined as bending radius at the centre of the crystal ($\theta = 0$).

The NSLS monochromator used differential four-bar bending with dynamical adjustment to approximate the logarithmic spiral for intermediate opening angle of the incident beam. For large incident beam divergence of around 1 radians, true logarithmic spiral shape is necessary. This can not be produced reliably with dynamical bending. Thus a fixed bender has to be used. Since the shape of the logarithmic spiral is dependent on energy through the Bragg angle θ_B in Eq. 3, there is error at the edges of the crystal if energies other than the energy for which the bender is designed is used. If this error is smaller than the large bandwidth of the bent-crystal, the crystal will still satisfy the diffraction condition. For example, with the bender designed for 20 keV using [111] reflection, the errors at the edge of the crystal is lower than 1 mradians for energies of 20±3 keV.

For crystals bent into a logarithmic spiral, there is variation of local bending radius from one end to the other. The local bending radius range from 150 to 250 mm from the bottom to the top of the crystal assuming the mean bending radius is 200 mm. Since the reflectivity and angular bandwidth of the crystal are not a strong function of the bending radius, this variation of local bending radius does not strongly affect the performance of the crystal.

3. Simulations

Simulations were performed using an analytical solution of a lamellar model (Erola et al., 1990) on bent crystals and an IDL code that takes into account the shape of the source spot and the spectral distribution of the source. The purpose of the simulation is to find the appropriate configuration that gives the optimum reflectivity and collection solid angle.

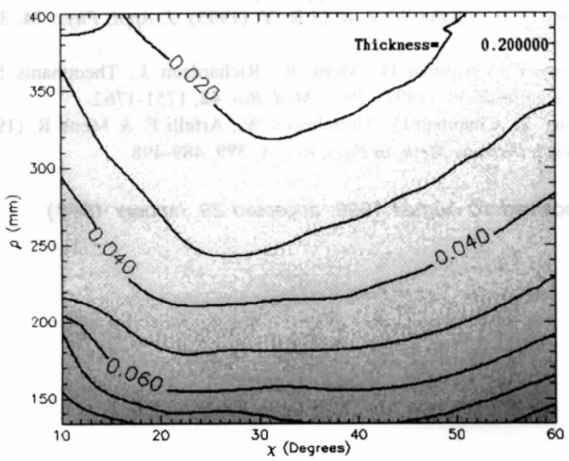

Figure 2
Figure of merit (FOM) for a 0.2 mm thick crystal of different bending radius and asymmetry angle. The energy was assumed to be 22.163 keV and 21.990 for silver Ka lines. The crystal size was assumed to be 100 mm by 100 mm. The source was assumed to be of 0.1 mm diameter.

The source is assumed to consist of $K\alpha_1$ and $K\alpha_2$ lines with the $K\alpha_2$ line having half the intensity of $K\alpha_1$ line. A cylindrical incident beam of 100 micron diameter (representing a focused beam through a dilute sample) is assumed. The figure of merit (FOM) is defined as the reflectivity times the solid angle subtended by a 100 mm × 100 mm analyzer. The FOM is calculated for various crystal thickness, bending radii, and asymmetry angles. The reflectivity, angular FWHM of the reflectivity curve and the resolution (dE/E) are also calculated.

Fig. 2 shows an example of the FOM simulation for 0.2 mm crystal thickness. As the crystal thickness and bending radius is reduced, the FOM increases, mainly as a result of increase in solid angle subtended by the crystal. Overall, a bending radius of 200 mm for a 0.2 mm thick crystal appears to be the a comfortable configuration since it offers a moderate FOM of 0.05, a reflectivity of 15%, and a spacing of 170 mm between the source and the analyzer. This allows more than ten crystals to be arranged to complete a circle around the source if desired, with freedom for design of benders and other hardware. The FOM is not sensitive to asymmetry angle around this area, so an asymmetry angle of 35.3 that offers a reflection angular bandwidth of 1.2 mradians is chosen. Since this is the angle between the (111) planes and [100] direction, commercially available silicon [100] wafers can be used. The large angular bandwidth enables the crystal to satisfy the Bragg condition across the area of the crystal with modest effort in controlling the bending.

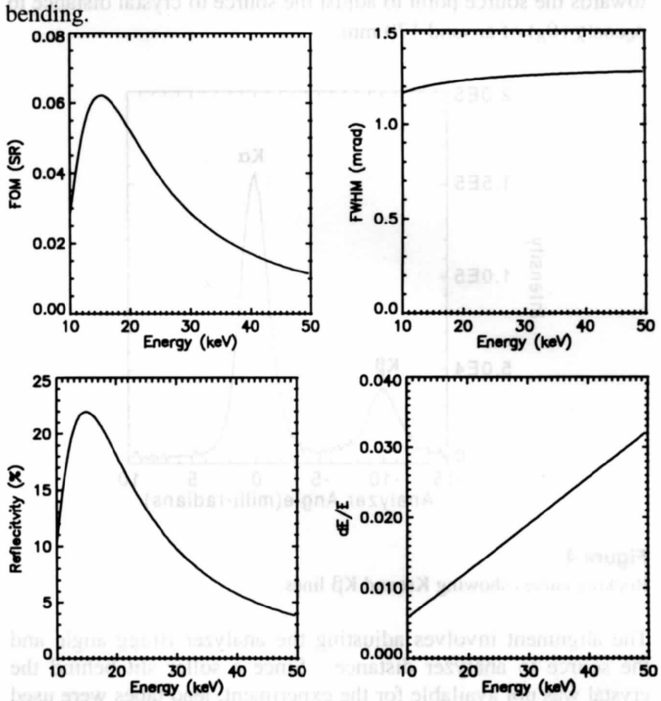

Figure 3
The FOM, angular FWHM, reflectivity and dE/E of the analyzer system vs. the energy of the fluorescence. The calculation assumes that a crystal of 0.2 mm thick is bent to an average radius of 200 mm. The asymmetry angle of the analyzer is 35.3 degrees

The energy dependence of the chosen configuration is also studied. At energies below 10 keV, the device suffers from excessive absorption by the analyzer crystal. At higher energies, the FOM drops slowly. As seen from Fig.3, the FOM is between 0.01 to 0.06 for energies between 10 and 50 keV. The angular bandwidth of reflection is dominated by the large asymmetry angle and is not sensitive to the energy. The energy resolution is approximately proportional to the energy.

4. Experiment

A fixed logarithmic spiral bender with an average bending radius of 200 mm was constructed by machining the surface of two aluminium blocks (50×160 mm surface, 25 mm thick) into the desired shape with the logarithmic spiral along the long edge of the bender. A rectangular window of 35 mm by 120 mm was machined in centre of the bender. This defines the active area of the crystal. A square piece of 50 mm by 135 mm silicon crystal was cut from 200 mm diameter commercial wafer of 0.2 mm thick and clamped between the convex and concave surfaces of the bender.

The silicon [333] monochromator at the BioCAT undulator beamline was tuned to an energy slightly above the silver K edge. Silver foil was used as target to generate the fluorescence x-rays for testing. The incident beam on the silver foil was focused to a horizontal width of around 0.2 mm at the time of the experiment. The analyzer diffracts in the horizontal plane. The bender was mounted on the theta arm of a diffractometer and an ion chamber filled with Krypton gas was mounted on the 2theta arm. The whole diffractometer was on a linear slide to move away and towards the source point to adjust the source to crystal distance to $\rho_0\cos(\chi+\theta_B)$ of around 170 mm.

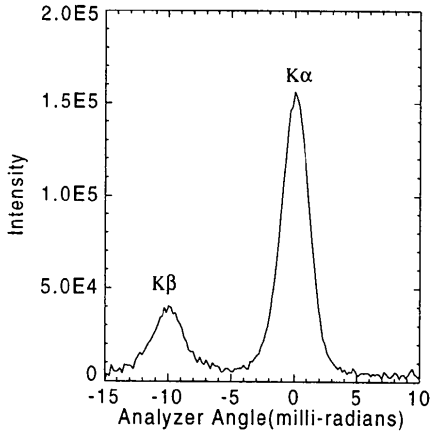

Figure 4
Rocking curves showing Kα and Kβ lines.

The alignment involves adjusting the analyzer Bragg angle and the source to analyzer distance. Since a soller slit behind the crystal was not available for the experiment, lead tapes were used in front of the crystal to break the incident fluorescence beam into 10 mm wide sections, so that the diffracted beam and the beam transmitted through the Laue analyzer are separated at the detector position. The detector aperture was adjusted to allow only the diffracted beam to enter the ion chamber. The Bragg angle of the analyzer was then scanned. Fig.4 shows the analyzer rocking curve obtained this way. The FWHM of the silver Kα (including both the $K\alpha_1$ and $K\alpha_2$ lines) peak is 2.6 mradians. This is wider than the intrinsic crystal reflection angular width of 1.2 mradians due to the relatively large source size.

The analyzer angle was then tuned to the peak of the Kα line. Image plate was positioned behind the crystal to produce an image of the diffracted beam. The distance between the crystal and the fluorescence target was adjusted till uniform diffracted beam appears across the crystal. The intensity uniformity of the diffracted beam across the analyzer crystal measured by the image plate was found to be sensitive to the change in source to crystal distance of around 2 mm. The average reflectivity of the Kα fluorescence across the crystal was around 10%, as measured by the image plate intensity level. This will approach the theoretical level of 15% if the source point size is reduced to around 0.1 mm.

5. Discussions

Using a thin (200 micron) silicon [100] wafer, bent into a logarithmic spiral shape of 200 mm average bending radius by a fixed bender and [111] reflection, a medium reflectivity of around 10% was achieved with silver K fluorescence. The analyzer has achieved an energy selectivity of 1-2%, large solid angle and uniformity of diffracted beam.

Future plans include decreasing the source size so that the $K\alpha_1$ and $K\alpha_2$ lines can be separated, and incorporating Soller slits behind the crystal to block away the beam transmitted through the Laue analyzer, so that only the diffracted fluorescence beam enters the detector. This allows testing of the analyzer with relevant dilute samples. The tunability of the analyzer for different edges will also be studied.

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