High pressure experiments at the XAFS Beamline, INDUS-2

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Abstract. The dispersive XAFS beamline BL-08 at the INDUS-2 synchrotron radiation source, RRCAT, Indore uses a bent Si (111) crystal as dispersive-cum-focusing element and a position sensitive CCD detector to enable instantaneous measurement of the whole XAFS spectrum around the absorption edge of interest. This beamline is ideal for characterisation of materials under high pressure using Diamond Anvil Cell with ~50µm spot size. For this setup, the theoretically determined spot size (Horizontal × Vertical) varies between 17 × 137 µm and 37 × 142 µm for the x-ray energy range 5 keV - 20 keV. To reduce the vertical spot size to <50 µm, we have designed an additional focusing mirror between the polychromator and sample position. The mirror, procured from SESO (France), will be installed shortly. Meanwhile, we have developed a dummy mirror bender setup at CDM (BARC) and have carried out feasibility tests to confirm reduction in spot size using the same. We have also conducted preliminary XAFS experiments (at BL-08) on SrRuO₃ at ~16 keV, under ambient conditions and inside diamond anvil cell, in order to assess the signal intensity and quality. We have obtained reasonably good signal.

1. Introduction:
X-ray Absorption Fine Structure (XAFS) beamline BL-08 (5-20 keV) has been recently commissioned at INDUS-2 Synchrotron Radiation Source, RRCAT, Indore uses a bent Si (111) crystal as dispersive-cum-focusing element and a position sensitive CCD detector to enable instantaneous measurement of the whole XAFS spectrum around the absorption edge of interest. This beamline is ideal for characterisation of materials under high pressure using Diamond Anvil Cell with ~50µm spot size. For this setup, the theoretically determined spot size (Horizontal × Vertical) varies between 17 × 137 µm and 37 × 142 µm for the x-ray energy range 5 keV - 20 keV. To reduce the vertical spot size to <50 µm, we have designed an additional focusing mirror between the polychromator and sample position. The mirror, procured from SESO (France), will be installed shortly. Meanwhile, we have developed a dummy mirror bender setup at CDM (BARC) and have carried out feasibility tests to confirm reduction in spot size using the same. We have also conducted preliminary XAFS experiments (at BL-08) on SrRuO₃ at ~16 keV, under ambient conditions and inside diamond anvil cell, in order to assess the signal intensity and quality. We have obtained reasonably good signal.
(iv) easy removal of diamond diffraction peaks in the energy range of interest (Bragg diffraction from the diamonds is inevitable, and leads to intense peaks superposed on the absorption spectrum, at energy values that match the Bragg condition on the intercepted crystal planes of the two diamond anvils)[3]. The energy-dispersive setup broadly caters to these requirements due to the sharp focusing by the elliptically bent crystal and absence of any mechanical movement of the crystal (during XAFS recording), leading to required focal spot stability.

For the present setup, the focal spot size at the sample position was calculated to be between [17 µm(H)×137 µm(V)] and [37 µm(H)×142 µm(V)] for the photon energy range 5-20 keV. The vertical spot size needs to be reduced to conform to DAC sample size. SHADOW calculations [4] show that inserting an additional focusing mirror prior to the sample position (Figure 1) reduces the spot size to [16 µm(H)×20 µm(V)] and [37 µm(H)×27 µm(V)] for the photon energy range 5-20 keV [5]. The radius of curvature of the mirror should vary between 277m and 401m correspondingly. For the above purpose, an elliptically bendable mirror has been procured from SESO, France and due to be installed in March 2012. The mirror is 150mm long, and has useful width and thickness 15mm and 15mm respectively. It has a U-shape and is Rhodium coated (coating thickness=500Å). Roughness of the mirror is less than 3 Å.rms. It has a meridional radius of 0.44197-0.401071 km and a bending resolution of 0.05%. An actuator is used to bend the mirror and is driven by a stepper motor. The actuator applies two equal forces on the legs of the U-mirror allowing it to apply the same moment on each leg of the mirror, causing it to bend.

2.1. Feasibility test to check the reduction in spot size:
To test the feasibility of the reduction in spot size, a dummy mirror bender was developed at the Centre for Design and Manufacture (CDM), Bhabha Atomic Research Centre (BARC) [6]. The focusing properties of the bendable mirror were simulated using divergent laser light. The dummy bender setup consists of a copper mirror plate supported on four support pins located at the two ends of the system. (Fig.3a) Two bender plates are supported on two pins and are rotated by the translating movements of two shafts. When the bender plate rotates, the copper plate is subjected to mechanical torques at its two ends, which results in the bending of the mirror plate. The translating movement to the shafts is realized by a special nut and worm wheel assembly fitted in the system. The worm wheel is rotated by a PC controlled Performax 2EX-SA two axis controller [7], the PC interface for which was developed by Integrated Electronics.

![Inclusion of an extra post mirror (without changing rest of the beamline setup)](image1)

**Figure 1.** Modified optical layout of the beamline.

![Focus spot at the image position for photon energies 5 keV, 10 keV and 20 keV](image2)

**Figure 2.** Focus spot at the image position for photon energies 5 keV, 10 keV and 20 keV

Figure 3b shows the schematic experimental arrangement for testing the focusing properties of the bendable mirror using laser light. He-Ne laser beam is focused at a pinhole by the microscope.
objective (MO). Light diverging from the pinhole is initially focused at the point $S_1$ by the focusing lens to form the primary image of the pinhole. Subsequently, the bent mirror focuses the laser light at the final image point $S_2$. There was a significant improvement of the focusing in the vertical direction due to reflection of laser light by the bendable mirror used in the grazing incidence configuration. The size of the focused laser spot also varied with the radius of curvature of the mirror substrate (varied by PC controlled stepper motor).

![Image](image.png)

**Figure 3:** a. Photograph of the mirror bender developed at CDM, BARC; b. Schematic Experimental arrangement for testing focusing properties of the bent mirror.

### 2.2. Feasibility test to check the transmission of synchrotron radiation through diamond:

For XAFS experiments at BL-08 (INDUS-2), $\text{SrRuO}_3$ sample was ground to powder of average particle size $\approx 5 \ \mu\text{m}$ and pasted uniformly on tape [8]. XAFS at $\text{Sr}$ K edge (~16 keV) was measured both within and without the diamond anvil cell (DAC) (2 diamonds, each having thickness = 2mm) at 60mA beam current and 2GeV ring energy. $\text{Sr}$ K edge was selected since at this energy (~16 keV), diamond absorption is less [9]. A Mao Bell type DAC, with anvil culet of diameter 400µm, was used for this experiment. The optimized spot size in this experiment was 200µm × 500µm (H×V). Note that the spot size is bigger than the calculated value (see Introduction) due to crystal imperfection and bigger source size from the design parameters. We have obtained a reasonable intensity signal for the DAC setup. The decrease in transmitted signal intensity is by $\approx 40\%$ when DAC is used (Figure 5). EXAFS spectra extraction and conversion to energy for this raw data is currently in progress. It may be noted that this feasibility test was conducted without the extra elliptical mirror. We expect the intensity to improve after the mirror is installed, due to reduced spot size ($<50 \ \mu\text{m}$ V). The theoretically calculated energy resolution, photon flux and spectral intensity at the final image point upon inclusion of the extra elliptical mirror are summarized in Table 1. Further, for high pressure studies at lower edges, perforated diamonds from Almax industries are in consideration.

![Graph](graph.png)
Figure 5: Transmitted intensity through the sample without DAC (black curve) and with DAC (red curve)

Table 1. Energy Resolution and Spectral Intensity

| E (keV) | \( \phi_0(\lambda) \) Photons/sec/mrad/0.1\% \( \Delta \lambda \) | \( \Delta E/E \) | \( \Delta E \) (eV) | \( \phi(\lambda) \) Photons/sec | \( I(\lambda) \) Photons/sec/mm² |
|--------|---------------------------------------------------------------|-----------------|-----------------|-------------------------------|-----------------------------|
| 5      | 1.21 \times 10^{11}                                          | 1.44 \times 10^4 | 0.72            | 5.37 \times 10^{11}           | 2.68 \times 10^{17}         |
| 10     | 5.62 \times 10^{12}                                          | 1.39 \times 10^4 | 1.39            | 3.42 \times 10^{11}           | 1.71 \times 10^{17}         |
| 20     | 9.98 \times 10^{11}                                          | 1.31 \times 10^4 | 2.62            | 0.87 \times 10^{11}           | 0.22 \times 10^{12}         |

3. Conclusion:
In summary, we have evaluated the possibility of high pressure experiments at the XAFS Beamline, RRCAT, Indore. Preliminary tests conducted at CDM (BARC), using an indigenously developed bender setup, confirm reduction in focal spot size on addition of an elliptically bent mirror prior to the sample position.

We have recorded XAFS of SrRuO₃ (powder tape sample) under ambient conditions, both within and without the DAC, in order to assess signal intensity and quality. We have obtained reasonable intensity signal. Following the installation of an elliptically bent mirror prior to the sample position, we expect the intensity to improve due to reduced spot size.

[1] Bhattacharyya D, Poswal A K, Jha S N, Sangeeta and Sabharwal S C 2009 Nuc. Insts. Meth. in Phys. Res. A 609 286.
[2] Zeng Q, Ding Y, Mao W L, Yang W, Sinogeikin S V, Shu J, Mao H K and Jiang J Z 2010, Phys Rev Lett, 104 105702; Giefers H, Porsch F and Wortmann G 2005 Physica Scripta T115 538; Miyauchi K, Qiu J, Shojiya M, Kawamoto Y, Kitamura N, Fukumi K, Katayama Y, Nishihata Y 2002 Sol. State Comm. 124 189; Jackson W E, Leon J M, Brown G E Jr, Waychunas G A, Conradson S D, Combes J M 1993, Science 262 29; Sadoc A, Itie J P, Polian A, Lefebvre S 1997 J Phys IV France 7 C2-991; Vaccari M, Aquilanti G, Pascarelli S, Mathon O 2009 J Phy: Cond Matt. 21 145403
[3] Pascarelli S, Mathon O, Aquilanti G 2004 J. Alloys and Comps 362 33
[4] http://www.esrf.eu/computing/scientific/raytracing/PDF/shadow_basics.pdf, http://www.esrf.eu/computing/scientific/raytracing/PDF/primer.pdf
[5] Das N C, Bhattacharyya D, Lahiri D, Sharma S M, “Studies on the possibilities of high pressure X-ray absorption experiments using XAFS beamline at INDUS-2 SRS”2007 BARC report E/001
[6] Ramanan N, Lahiri D, Sharma S M, Das N C, Udupa D V, Patil A D, “Development of a bendable mirror for studying materials under high pressure using EXAFS beamline at INDUS-2 SRS”2010 BARC report E/012
[7] http://www.arcus-technology.com/products/24.html
[8] Lahiri D 2006 Physica C 436 32
[9] Dadashev A, Pasternak M P, Rozenberg G K, Taylor R D 2001 Rev. Sci. Inst. 72 2633