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A high resolution powder diffractometer using focusing optics

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Abstract. In this paper, we describe the design, construction and performance of a new high resolution neutron powder diffractometer that has been installed at the Dhruva reactor, Trombay, India. The instrument employs novel design concepts like the use of bent, perfect crystal monochromator and open beam geometry, enabling the use of smaller samples. The resolution curve of the instrument was found to have little variation over a wide angular region and a $\Delta d/d \sim 0.3\%$ has been achieved. The instrument provides sample environment of very low temperatures and high magnetic fields using a 7 Tesla cryogen-free superconducting magnet with a VTI having a temperature range of 1.5–320 K. The special sample environment and high resolution make this neutron powder diffractometer a very powerful facility for studying magnetic properties of materials.

Keywords. Neutron diffraction; Bragg diffraction optics.

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1. Introduction

The design and construction of a neutron powder diffractometer is mainly governed by the scientific purpose, the constraint that the available neutron flux imposes on the resolution and intensity, space and hardware considerations. The scientific case for the diffractometer was mainly driven by the demand from several users from universities for a neutron diffraction beamline with high resolution and special sample environment for determination of magnetic and chemical crystal structures. Phenomena like magnetic field-driven charge order melting and insulator–metal transitions in manganites, and magnetic transitions in low-dimensional magnetic systems are being widely studied in light of their applications and novelty. Very often, such studies require neutron spectrometers with special capabilities. The present diffractometer has been designed keeping in mind the requirements for such frontline research problems. In this paper, we describe the design and construction of the powder diffractometer, including the rationale behind the particular design choices made.

2. Design calculations

The powder diffractometer is installed on an off-centred through tube TT-1015 at the Dhruva reactor, Trombay. This beam tube consists of a D_2O moderator at its centre and has an in-pile collimator measuring 70 mm×70 mm. The monochromator crystal is placed at a distance of 9.05 m from the source. Bent perfect crystal monochromators with open beam geometry exploit the angular correlation between the incident neutrons to achieve focusing at the sample position with the same momentum transfer [1,2]. It has been known from experimental studies that focusing in both horizontal – the dispersion plane – and the vertical plane can be used to increase the intensity of the monochromatic beam without distorting the lineshape [3–5]. To achieve high resolution without too much loss in intensity, it was decided to adopt an open beam geometry, without Soller collimators, and use a focusing monochromator that is bent both horizontally and vertically. In order to optimize the various parameters of the beamline-like collimator openings, horizontal and vertical radii of curvature, distances between different optical components and sample size, and to work out various tolerances in these parameters, Monte Carlo simulation technique was used. Our calculations showed that the use of open beam geometry and a doubly bent, asymmetrically-cut perfect crystal monochromator resulted in a substantial increase of intensity and an improved resolution over a wide angular range [6]. The advantages of using the above combinations are: (i) gain in intensity due to focusing of neutrons in real and momentum space and beam condensation by asymmetric diffraction and (ii) improvement in resolution due to focusing of neutrons in momentum space.

3. Mechanical design, beam characteristics and data acquisition

Figure 1 shows a schematic lay-out of the diffractometer. The doubly bent perfect Si monochromator consists of nine asymmetrically cut Si crystals covering an area of 17 cm×13 cm, which are mechanically bent in the horizontal plane and stacked over barrel-shaped posts to get a vertical curvature. The horizontal and vertical radii of curvature are 12.6 m and 1.8 m, respectively and the take-off angle is 90°. The monochromator can be aligned to give incident wavelengths of 1.17, 1.48, 1.76 and 2.3 Å. The flexibility in the choice of different wavelengths would enable the determination of a wide variety of chemical and magnetic structures. The monochromatized beam is led out through a nose cone collimator and then an adjustable boron carbide slit defines the final incident beam size. The focused neutron beam at the sample position was 15 mm×25 mm in size. A low efficiency BF_3 detector is used as a beam monitor. The optimum sample diameter is about 6 mm to achieve good resolution. The complete diffraction pattern is obtained over four overlapping banks of three linear ^{3}He PSDs in each bank, covering an angular range up to 123°. Each PSD (LND make) has an active length of 1 m and diameter of 1 inch. The fill gas is ^{3}He at 10 atm pressure and the spatial resolution is about 3 mm. The overlapping detector banks are arranged tangentially above and below the horizontal scattering plane at a distance of 1.5 m from the sample. The detector banks are enclosed in a shielding box made of boronated wood enclosed in

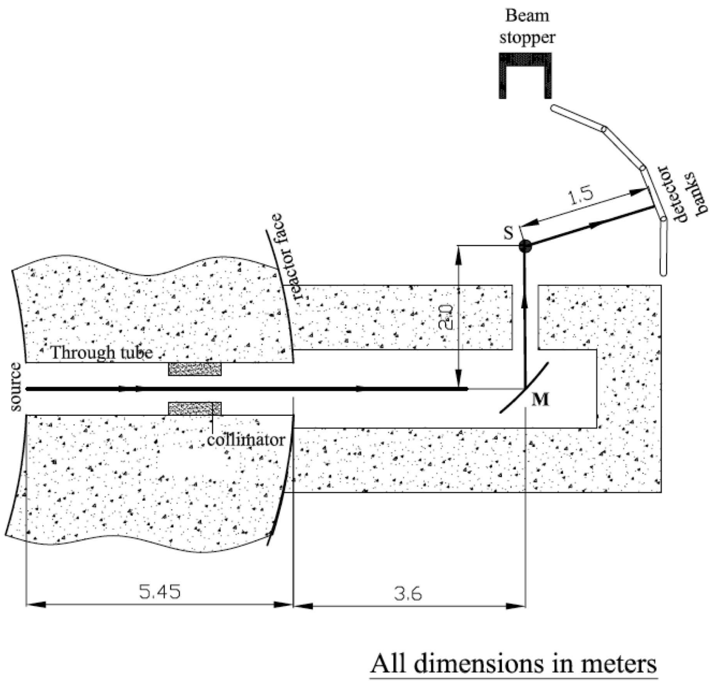

Figure 1. Schematic lay-out of the neutron diffractometer based on focusing monochromator.

steel containers and lined with cadmium on the inside. Vertical stacking of PSDs in each bank increases the overall throughput. The position of an incident neutron falling on the detector is determined by the charge division method [7]. When a neutron induces ionization in a gas, the charge collected on the anode wire is divided between two amplifiers placed at either end of the wire in a proportion that is related to the position of interaction. Data acquisition electronics and software were developed locally [8]. The raw data acquired from the PSDs is converted into equiangular data, using simple geometrical considerations and data interpolation routines. Equiangular data from different PSDs is appropriately combined and displayed on the screen for visualization.

4. Performance of diffractometer

Figure 2 shows the rocking curve of the bent perfect Si monochromator aligned for (5 1 1) reflection at a take-off angle of 90°. It is seen that the rocking curve is symmetric and the half-width is about 0.5°. The horizontal and vertical beam profiles of the focused neutron beam at the sample position are shown on the right of the figure. The widths of these curves confirm the effect of focusing from the monochromator. To test the performance of the powder diffractometer, diffraction patterns of a series of samples, usually considered as standards, were measured. As an example, figure 3 shows a pattern of sintered Al_2O_3 (99.99% pure) rod

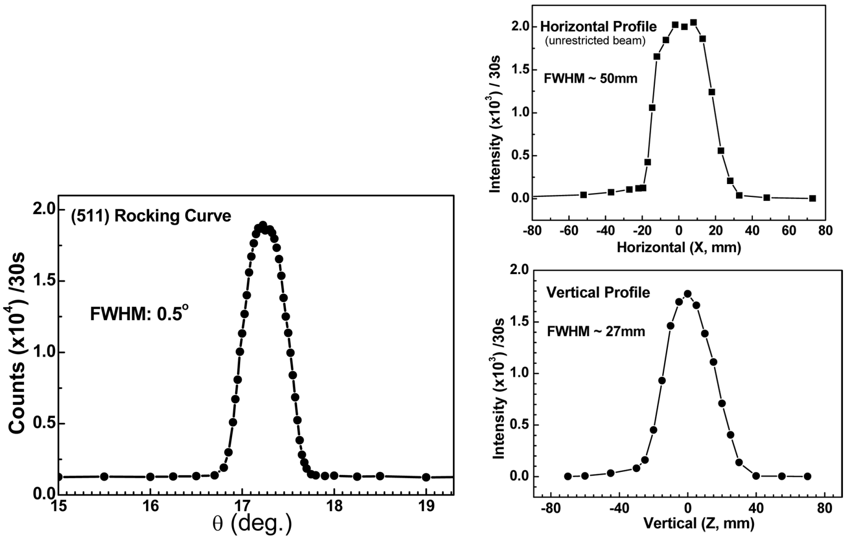

Figure 2. Rocking curve of bent perfect Si monochromator at a take-off angle of 90° and $\lambda = 1.48$ Å. Figures on right show the horizontal and vertical beam profiles measured at sample position.

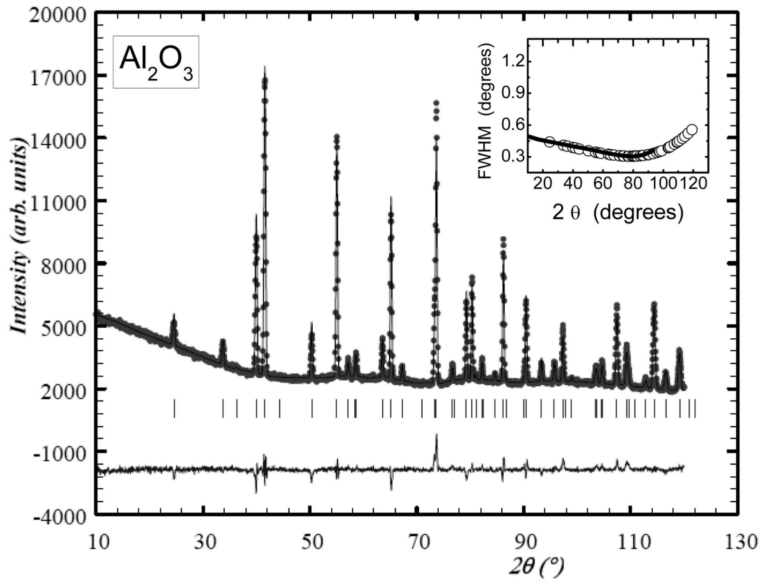

Figure 3. Rietveld refinement of sintered Al_2O_3 rod. Inset shows the 2θ dependence of FWHM obtained from Rietveld analysis (open circles) and Monte Carlo simulation (solid line) (ref. [6]).

and measured for a period of 3 h. The diffraction pattern was analysed using Rietveld method. It is seen that there is good agreement between the observed and calculated data and the R-factors obtained were: $R_{\mathrm{p}} = 2.49\%$, $R_{\mathrm{wp}} = 3.38\%$ and $R_{\mathrm{exp}} = 1.77\%$. It was found that there was a preferred orientation in the [1 0 0] direction. The cell parameters obtained from the fit were $a = 4.7584$ Å and $c = 12.9927$ Å, which compare very well with the NIST SRM-1976 values of $a = 4.758877$ Å and $c = 12.992877$ Å. It is also observed that focusing of neutrons has resulted in reducing the data acquisition time, showing a gain in the intensity. Inset of figure 3 shows the 2θ dependence of FWHM of each of the refined Bragg reflections and the $\Delta d/d$ calculated from this data is about 0.3%, indicating very good resolution over a wide angular range. The measured resolution is in excellent agreement (solid line in inset) with the value calculated from our earlier Monte Carlo simulations [6]. Another advantage of focusing that naturally follows is that smaller sample sizes ($\sim$0.5 cc by volume) could be used for diffraction experiments.

5. Sample environment

This diffractometer offers sample environment of low temperatures and high magnetic fields. Subjects like magnetic field-induced charge order melting, metal insulator transitions and phase coexistence in manganites and field-induced transitions in low-dimensional systems have attracted intense research activity among the academic community in the country. Since the study of such phenomena require special sample environment, a cryogen-free superconducting magnet has been installed and can be used on the diffractometer. This system provides very low temperatures (down to 1.5 K) and high magnetic fields (up to 7 Tesla). The system is automated so that an individual experiment can be planned for a series of magnetic field and temperature situations. Preliminary experiments have shown that very good quality data under low temperature–high magnetic field conditions can be acquired. It is planned to use an oscillating radial collimator to cut off extraneous scattering from walls of cryostats and also to reduce the high background observed at low scattering angles.

6. Conclusion

We have described the design, construction and performance of a high resolution neutron powder diffractometer at Dhruva reactor. The resolution curve has been found to be very good over a wide angular range and $\Delta d/d \sim 0.3\%$ has been achieved. The diffractometer has a special sample environment of very low temperatures and high magnetic fields and is expected to be widely used in the study of magnetic and structural properties of novel materials.

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