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The high-resolution powder diffractometer at the high flux isotope reactor

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Abstract Neutron powder diffraction is increasingly recognized as one of the most powerful techniques for studying the structural and magnetic properties of advanced materials. Despite the growing demand to study an ever-increasing array of interesting materials, there is only a handful of neutron diffractometers available to serve the US neutron scattering community. This article describes the new high-resolution powder diffractometer that has recently been installed at the High Flux Isotope Reactor in Oak Ridge. The instrument is designed to provide an optimum balance between high neutron flux and high resolution. Due to its versatility the diffractometer can be employed for a large variety of experiments, but it is particularly adapted for refinements of structures with large interplanar spacings as well as of complex magnetic structures. In addition to traditional crystal and magnetic structural refinements, studies of phase transitions, thermal expansion, texture analysis, and ab initio structure solution from powder data can be undertaken.

1 Introduction

Neutron powder diffractometers installed at various neutron scattering facilities around the world stand out as the instruments most popular and widely used by experimentalists. With the growing complexity of materials comes the need for an improved understanding of their crystallographic and magnetic properties, and in this respect, neutron scattering has proven an invaluable experimental tool. Technologically important materials amenable to study by neutron diffraction include, but are not limited to, catalysts, ionic conductors, superconductors, alloys, ceramics, cements, colossal magnetoresistant perovskites, magnets, radioactive waste forms, zeolites and minerals.

The HB-2A High-Resolution Neutron Powder Diffractometer has recently been installed at the High Flux Isotope Reactor (HFIR) at Oak Ridge, and it began its commissioning process in December 2008. A picture of the instrument is shown in Fig. 1. The HB-2A was designed as an upgraded version of the old HFIR's powder diffractometer (HB4) that served the scientific community for a decade (1990–2000). Relocated to the HB-2 thermal beam tube, the powder diffractometer directly benefits from one of the highest steady-state neutron fluxes in the world, provided by the 85 MW HFIR research reactor. As such, it was redesigned to offer an optimal balance between intensity and resolution. This instrument is expected to be a workhorse for crystal and magnetic structural studies of powdered samples, particularly as a function of intensive conditions. This paper describes the main characteristics of the instrument and illustrates its current capabilities.

2 Instrument components

The diffractometer is positioned at the HFIR's largest beam tube (HB-2), which is situated radially relative to the reactor core. To reduce the high energy neutron component in the incident neutron flux, a liquid nitrogen cooled sapphire filter is installed upstream of the main shutter. The main shutter provides a means for turning the beam off for all the instruments located at this beam tube. After passing through

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Fig. 1 The HB-2A high-resolution neutron powder diffractometer located at the High Flux Isotope Reactor, at Oak Ridge National Laboratory. The distance from the sample to the detectors is 1 m

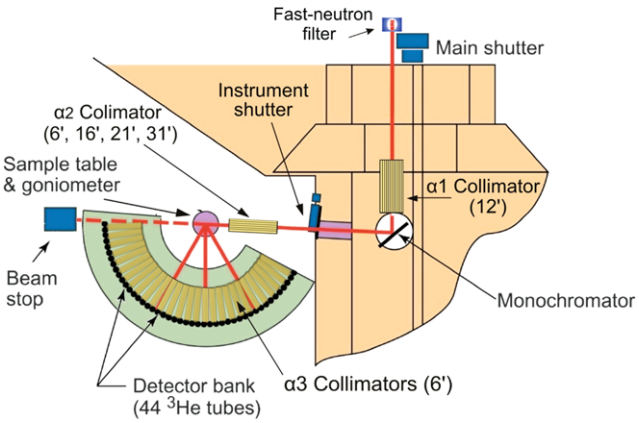

Fig. 2 Layout of the HB2A instrument

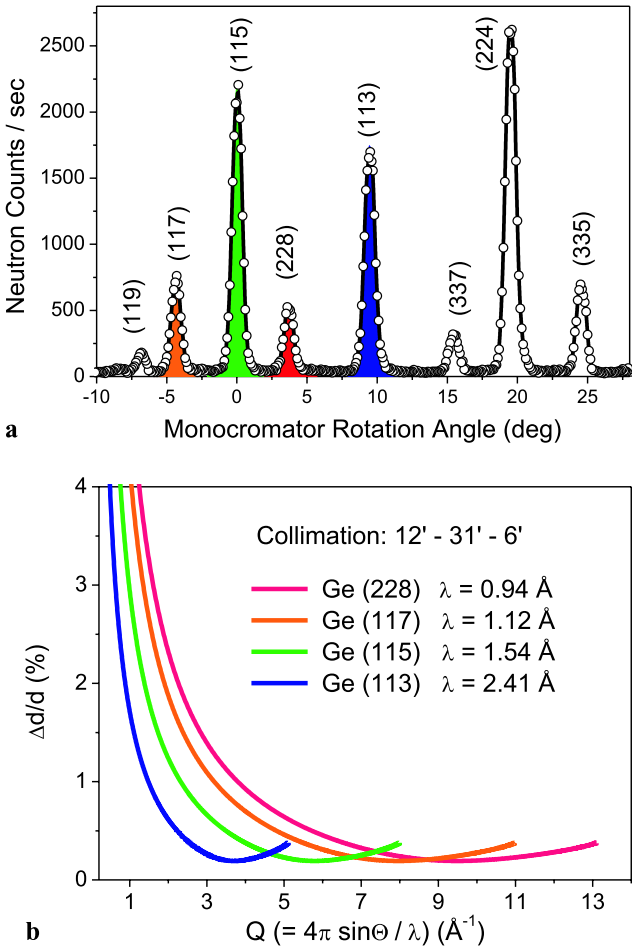

Fig. 3 (**a**) Rotation scan showing rocking curves for accessible Ge reflections, at a fixed take-off angle $2\theta_M = 90°$. (**b**) Instrument resolution profiles plotted as $\Delta d/d$ versus scattering vector **Q** (Å^{-1}) for different available wavelengths, which demonstrates the broad versatility of the HB-2A diffractometer. Color coding in (**a**) corresponds to that in (**b**)

the main shutter the neutron beam is incident on a monochromator crystal assembly which diffracts a pre-selected neutron wavelength determined by the monochromator d-spacing and the take-off angle ($2\Theta_M$). The monochromator take-off angle is fixed at 90°. Before reaching the monochromator, the incident beam may also pass through a collimator or a beam size reducing mask. The deflected monochromatic beam successively passes through the instrument shutter, a collimator and a beam-sizing diaphragm before illuminating the sample located at the center of a goniometer table. The neutrons scattered by the sample pass through another set of Söller collimators and are detected by 44 ^{3}He counters. The entire detector bank can rotate around the sample axis to give access to a scattering angle (2Θ) ranging from 2 to 154°. The main components of the HB-2A diffractometer are schematized in Fig. 2.

2.1 Monochromator

The monochromator system consists of 15 germanium (Ge) composite wafers segments, each measuring approximately $1 \times 1 \times 5$ cm^3. The wafers were cut from a Ge single-crystal ingot perpendicular to (115) crystal axis. Prior to forming the composite crystals, each wafer was successively deformed and reflattened at RISØ National Laboratory according to the procedure developed by Axe et al. [1]. The 15 Ge crystals are co-aligned on a vertically focussing unit with a convergence of approximately 21 mrad. The monochromator is placed on the rotation stage that allows the choice of several monochromatic wavelengths, ranging from 0.94 to 2.41 Å, depending upon which Ge (hhl) plane is brought into the diffraction condition (see Fig. 3(a)). Rocking curves measured at various positions show regular Gaussian peak shapes with a full width at half maximum (FWHM) of the mosaic distribution of about 48′. The maximum neutron flux at the sample position is of order of 10^7 n/cm^2 s and is ob-

Table 1 Monochromatic wavelengths available at the HB-2A, the accessible **Q** and d ranges, and the corresponding measured neutron flux at the sample position

(hkl)	λ (Å)	d_{max} (Å)	**Q** (Å^{-1})	$n°$ flux (n/cm^2 s)
(113)	2.41	27.6	0.2–5.1	5.2×10^6
(115)	1.54	17.6	0.35–7.9	10^7
(335)	1.21	13.8	0.45–10.1	5.9×10^6
(117)	1.12	12.8	0.5–10.9	5.6×10^6
(228)	0.94	10.7	0.6–13.1	4.8×10^6

tained from the Ge(115) reflection. For this configuration the wavelength is $\lambda = 1.54$ Å. Table 1 provides a list of the available wavelengths, the accessible **Q** ($= 4\pi \sin\theta/\lambda$) range, and the corresponding neutron flux at the sample position measured using a calibrated neutron beam monitor.

2.2 Collimation

To limit the horizontal divergence of the beam, collimators are inserted in the beam line at several positions: pre-monochromator (α_1), pre-sample (α_2), and pre-detector (α_3) [2, 3]. These are Söller collimators comprised of long, thin parallel sheets of neutron absorbing Gd_2O_3-coated mylar, separated by an appropriate distance to provide the desired beam divergence. In addition, diaphragms made of B_4C and having variable apertures are inserted in the beam to limit the horizontal and vertical beam dimensions. A 12′ Söller collimator is located in front of the monochromator system, inside the HB-2 tunnel. This collimator is mounted on a motorized rotation/translation positioning device. The unit has been carefully aligned to give optimum performance. The translation axis is perpendicular to the incident beam direction and allows driving the collimator out of the monochromator axis. This feature permits the operation of the instrument in high-resolution or high-intensity modes. A beam mask that fits a bracket mounted in front of the monochromator is used to redefine the size of incident beam and reduce the background scattering. When the 12′ collimator is positioned out of the monochromator axis the diaphragm acts like a 35′ collimator. There are several choices for the pre-sample collimation: $\alpha_2 = 6'$, 16′, 21′, or 31′. Each pre-sample collimator is a separate unit that is attached to a carriage mounted on an optical bench situated in front of the sample table. To further reduce the background scattering at small angles, a tube made of B_4C is set to redefine and shield the neutron beam. In addition several fixed aperture sizes are available. At the scattering area, the powder diffractometer is equipped with 44 separate 6′ Söller collimators. Each collimator has been independently aligned with a relative angular spacing of approximately 2.7°.

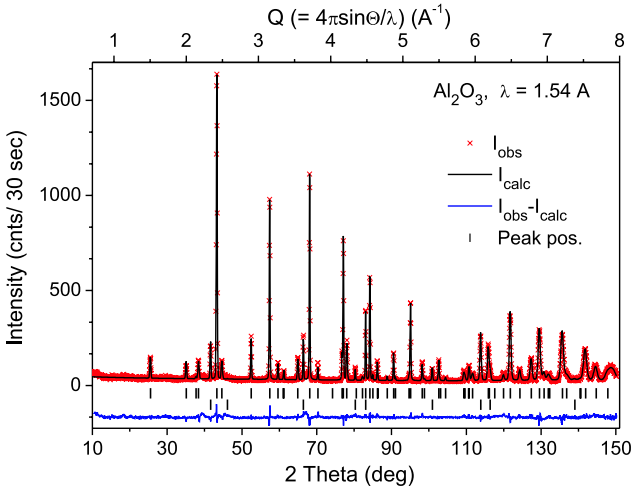

Fig. 4 Neutron diffraction pattern measured at the HB-2A using 12′-31′-6′ collimation and $\lambda = 1.54$ Å. Bragg peak positions corresponding to Al_2O_3 sample and Al holder are indicated by *vertical bars*, and the difference (observed-calculated) pattern by *the lower solid line*

2.3 Detector bank

The detector bank contains 44 ^{3}He tubes, each preceded by a 6′ Söller collimator. The detector tubes measure 3 cm in diameter and have a active length of approximately 12 cm. The He pressure in the tubes is 4.1 bar. The shield drum hosting the collimators and detectors tubes consists of three casings that are filled with borated, hydrogenated field-castable dry mix. This assembly is driven slowly on rails by an enclosed gear and its range of motion is controlled by mechanical hardware and software limits. In a typical experiment the detector assembly is moved to the desired starting position and counts are accumulated in the 44 detectors. A stepped drive (typically 0.05°) and count sequence is repeated until the total angle of rotation is at least 3°, so that data from each detector overlap with data from its neighbors. The assembly can be rotated through 35°, and measurements can be made with 2θ from 0 to 155°. A typical two-theta scan is exemplified in Fig. 4 that shows the diffraction pattern of alumina (Al_2O_3) powder collected using the wavelength $\lambda = 1.54$ Å.

2.4 Sample goniometer and environment

A sample goniometer is used to precisely control the physical location and orientation of the sample. It consists of a motor driven rotation, tilts and two orthogonal horizontal translations. The sample is generally centered in the beam using precise computer control of the horizontal motions of the sample goniometer, while a neutron camera is positioned in the path of the neutron beam behind the sample. In addition to high-resolution powder diffractometry, the instrument can be easily adapted to measure texture in materials, residual stresses or single-crystal diffraction.

A 2 ton full cantilever jib type crane was installed at the HB-2A experiment station to service both the instrument sample area and the detector shielding. The samples can be placed inside a number of different pieces of equipment providing various environments including: cryostats for low temperature, furnaces for high temperature, superconducting magnets and electromagnets for high magnetic field, and cells for high pressure.

3 Instrument performance

Operated at its fixed take-off angle of 90°, the resolution function of the diffractometer can be tailored to the specific requirements of each experiment by selecting the appropriate collimators (α_1, α_2). The estimated maximum resolution $\Delta d/d$ achievable at the HB-2A is approximately 1.9×10^{-3}. Figure 5 displays the FWHM of the diffraction peaks as a function of **Q** for different choices of the collimators. The instrument resolution function calculated by a simple Caglioti et al. formula [2] is plotted by solid lines. The calculations are compared with the peak widths extracted from fitting the experimental data measured for NIST standard reference material silicon powder 640b and Al_2O_3. An accurate description of the shapes of the peaks measured at HB-2A can be obtained using Thompson–Cox–Hastings pseudo-Voigt function [4] with asymmetry correction [5, 6].

The range of rotation of the detector bank (up to 35°), allows one to overlap the detector coverage which enables a reliable determination of the peak centers, and averages the deviation in detection efficiencies. An effective and well proven strategy for data acquisition at the HB-2A involves combining two separate scans that cover low-angle (2θ range $\approx$ 2–126°) and high-angle scattering ($\approx$ 30°–154°), respectively. Each of these scans consists of a stepped drive and count sequence for a total movement of the detector back of about 6°. As a consequence those data from each detector fully overlap with data from its neighbors, minimizing the systematic geometrical and detector efficiency errors.

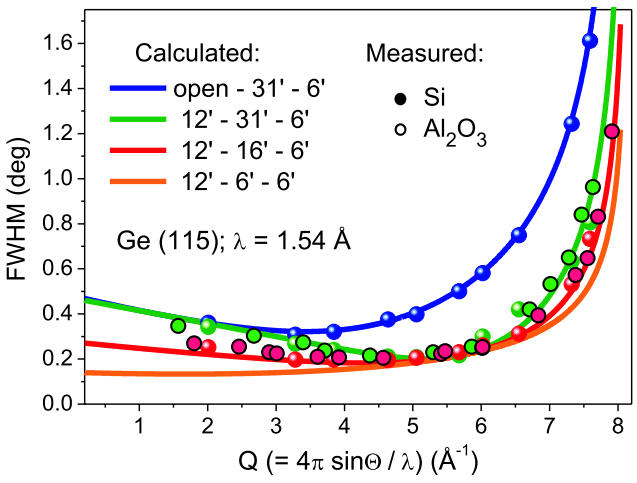

Fig. 5 HB-2A resolution function as a function of **Q** for four different combinations of the collimators. The calculated resolution profile is plotted by *solid lines* and the measured values are indicated by *symbols*

Another important flexibility in the instrument operation is the possibility of using different neutron wavelengths. It allows for optimization of the resolution function for specific **Q** ranges. As illustrated in Fig. 3(b), the larger available wavelength 2.41 Å from the Ge(113) reflection gives a good resolution in the small-angle region. This makes HB-2A a well adapted instruments for refinement and solution of structures with relatively large interplanar spacings ($d_{\max} = 27.6$ Å) as well as of complex magnetic structures. Additionally, the shorter wavelength 0.94 Å can be successfully used for the pair distribution function (PDF) analysis in disordered materials ($\mathbf{Q}_{\max} = 13.1$ Å^{-1}). Collecting data using distinct wavelengths may be considered also as an alternative strategy for data acquisition at the HB-2A. Such data can be used for combined refinements to take advantage of the extended number of observations.

4 Conclusion

In conclusion, the completely-rebuilt high-resolution neutron powder diffractometer HB-2A has been installed at the High Flux Isotope Reactor, and is now available to serve the neutron scattering user community. Due to its versatility the HB-2A diffractometer can be employed for a great variety of different experiments spanning many different disciplines. Rietveld refinements of powder samples with variable structural complexity yield high-quality structure data. In addition to traditional crystal and magnetic structural refinements, studies of phase transitions, thermal expansion, quantitative analysis, residual stress, and ab initio structure solution from powder data can be undertaken. As a part of the national User Program at the HFIR [7], the HB-2A diffractometer will select experiments and allocate beam time based upon a proposal driven peer review process.

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