

Powder Bragg backscattering spectroscopy

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Motivation

Powder diffraction is a powerful tool in x-ray crystallography. In the present paper we study potentialities of this technique in extreme backscattering geometry.

Backscattering means that in contrast to standard powder diffraction the scattering angle is kept fixed at 180° and the x-ray energy is tuned. One expects several advantages in comparison to the standard technique. The simple geometrical arrangement with only one path for the incoming and outgoing beams allows access to samples with very small angular aperture. This is important if the samples are placed in a pressure cell, cryostat, or furnace, etc. As the scattering geometry does not change the extinction corrections are not required. The highest possible momentum transfer and thus the highest possible spatial resolution is achieved in backscattering.

Another important point is that Bragg's law $2d_{hkl} \sin \theta = \lambda$ for x rays with energy $E = hc/\lambda$ and for crystal planes (hkl) with interplanar distance d_{hkl} reduces in case of backscattering ($\theta \simeq \pi/2$) to

$$2d_{hkl} \left[1 - \frac{(\delta\theta)^2}{2} \right] = \lambda, \quad (1)$$

where $\delta\theta = \pi/2 - \theta$. If $\delta\theta \ll 1$ then the direct relation $2d_{hkl} = \lambda$ may be used for simple and accurate measure-

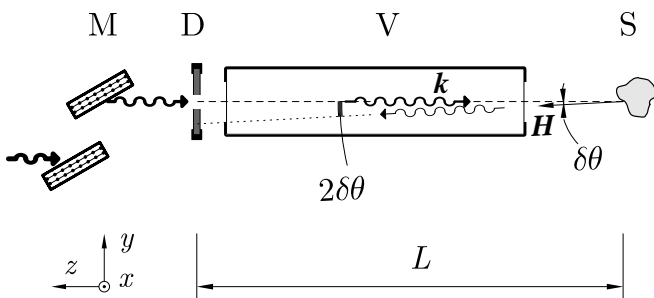


FIG. 1. The scheme of the experiment. M: tunable high heat load diamond C(111) monochromator used as the λ -meter; D: x-ray detector with a time resolution $\simeq 1$ ns; V: vacuum tube; S: powder sample; $\delta\theta$: angle between the vector \mathbf{k} of the incident radiation and the scattering vector \mathbf{H} ; $L \simeq 6.2$ m.

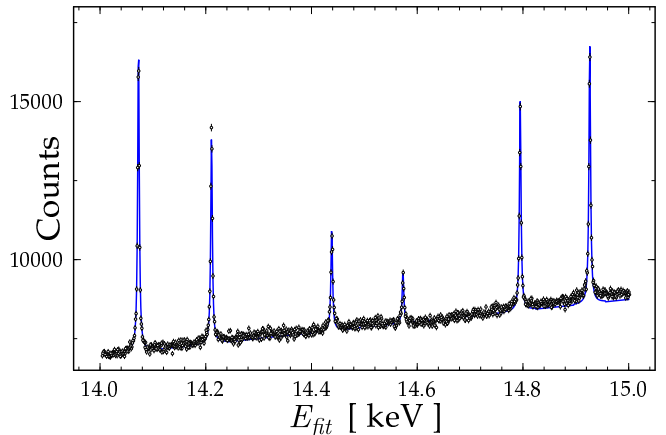


FIG. 2. Bragg backscattering spectrum from Si powder sample (circles). As the lattice constant of silicon $a = 5.4310196$ Å is precisely known a fit of the theoretical backscattering spectrum (solid line) to the experimental points determines the energy scale E_{fit} of the λ -meter.

ments of crystal structure parameters as well as of x-ray wavelengths.

The accuracy of measurements are limited:

1. by the angular deviation $\delta\theta$ from normal incidence. $\delta\theta$ can be easily made less than 10^{-3} , so that the accuracy of the relation $2d_{hkl} = \lambda$ can be made better than 1 ppm.
2. by the energy resolution of the λ -meter, of the instrument, which selects and tunes the wavelength (energy) of the incident x-rays. Presently it can be chosen in the range from $\simeq 1$ eV down to $\simeq 1$ meV [1].
3. by the energy width of the backscattering reflections. For thick single crystals it is in the meV range. However, for small ($\simeq 1$ μm) powder particles the energy width may increase to about 1 eV.

Method

The detector should be put as close to the incident beam as possible (Fig. 1) to achieve $\delta\theta < 10^{-3}$. However, this leads to an enormous background and hinders performance of experiments in backscattering geometry.

One can overcome the problem by exploiting the pulse structure of the synchrotron radiation and using a detector with good time resolution. Because the background arises from the incident pulse going to the sample and

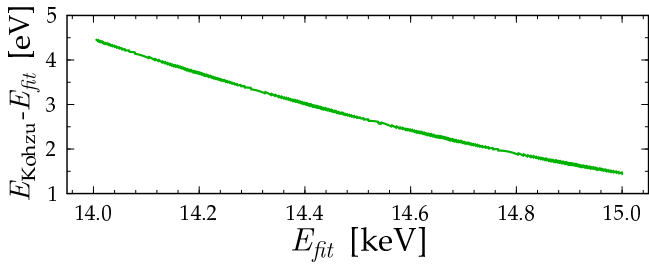


FIG. 3. Difference in values of the x-ray energy of the Kohzu high-heat-load monochromator read-back E_{Kohzu} and the x-ray energy E_{fit} obtained from the fit of the Si backscattering spectrum in Fig.2.

the signal consists of the pulse reflected from the sample, a time delay $\tau = 2L/c$ exists between the background and the signal. This allows the background to be distinguished from the signal and rejected as shown in [2,3].

The experiments were performed at the undulator beamline 3-ID at the APS. Figure 1 shows the scheme of the experimental set-up. A diamond C(111) high-heat-load monochromator, used as the λ -meter, transmits x-rays with a bandwidth of 1 eV and is tuned in the range from 14 to 15 keV.

The detector consists of two avalanche photo-diodes, with a time resolution of $\simeq 1$ ns [4], placed 2 mm above and below the incident beam, at a distance of $L = 6.2$ m upstream from the sample. Under these conditions $\delta\theta < 0.4$ mrad and $\tau \simeq 40$ ns.

Results and discussion

Figures 2 and 4 show results of measurements of the standard NIST powder samples of Si and Al_2O_3 respectively. The radiation flux was 10^{12} photons/s. The data at each point were collected for about 0.5 s in case of the Si and 3 s in case of the Al_2O_3 sample and normalized to the incident flux. The samples were rotated about an axis perpendicular to the beam. The spectra recorded without rotating the sample show the spectral lines at the same positions but with changed strengths.

The line form is very close to Lorentzian. This is typical for Bragg backscattering spectra in non perfect crystals [2,3]. The linewidth is typically 4 eV. This is more than the 1 eV width of the incident radiation. Presumably it is determined by the size of the particles. However, to cause such a broadening the particles should have a diameter of $\simeq 100$ nm.

The evaluation of the spectra were performed with the program package BASS [5]. The lattice constant of silicon is well known. This allows to define the energy scale of the λ -meter from the fit of the experimental spectrum - Fig. 2. Fig. 3 shows the difference between the energy values E_{fit} obtained from the fit and the internal calibration of the Kohzu high-heat-load monochromator

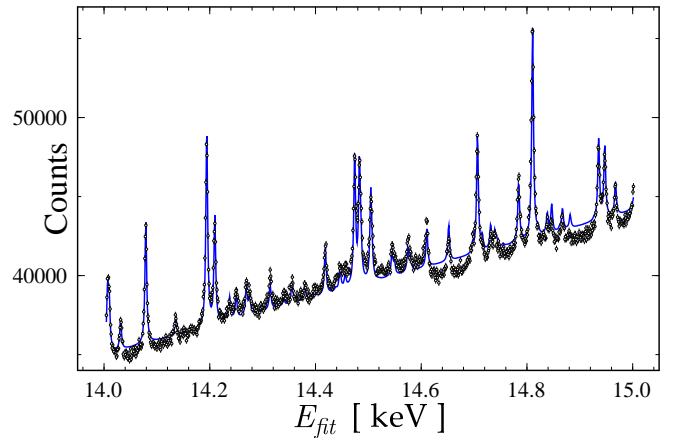


FIG. 4. Bragg backscattering spectrum from Al_2O_3 powder sample. The solid line was calculated with BASS [5].

E_{Kohzu} . This result demonstrates how one can check in a simple way the performance of an x-ray monochromator by using powder Bragg backscattering spectroscopy.

The best fit of the Al_2O_3 Bragg backscattering spectrum is achieved for the lattice parameters $a = 4.75931(4)$, $c = 12.9920(1)$. The accuracy is 8 ppm, which is high for powder diffraction technique. The other crystallographic parameters: position and anisotropic thermal parameters are in agreement with Ref. [6].

Acknowledgements

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- [1] T. Toellner, *Hyperfine Interactions*, **125** (2000) 3.
 - [2] Yu.V. Shvyd'ko, E. Gerdau, J. Jäschke, O. Leupold, M. Lucht, and H.D. Rüter, *Phys. Rev. B* **57**, 4968 (1998).
 - [3] Yu.V. Shvyd'ko, and E. Gerdau, *Hyperfine Interactions*, **123/124** (2000) 741.
 - [4] A.Q.R. Baron, *NIM A* **352**, 665 (1995).
 - [5] BASS calculates energy Bragg backscattering spectra of x rays in kinematical approximation and provides fits to experimental ones. Written by Yu.V. Shvyd'ko.
 - [6] A.S. Brown, M.A. Spackman, and R.J. Hill, *Acta Cryst.* **A49** (1993) 513