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OrientExpress: A new system for Laue neutron diffraction

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Abstract

A new automatic Laue neutron diffractometer has been developed at the ILL. The system is composed of a goniometer with two tilt stages mounted on an ω-rotation and a scintillator/CCD neutron detector which is mounted on a 2θ arm. All movements are computer controlled. The intensified neutron imaging system is unique and allows electronic capture of neutron Laue diffraction patterns in a much shorter time (few seconds) than conventional film-based methods. The detection system is based upon two high-performance image-intensified CCD cameras coupled to a large-area neutron scintillator. The system is also unique in permitting full back-reflection geometry. A gain of about 100 in efficiency is obtained compared to the conventional film method with comparable spatial resolution. Some examples for Laue patterns are presented and compared to those obtained by film. A quantitative analysis of the integrated intensity of the Laue spots is also made.

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Keywords: Laue diffraction; CCD detector; Rapid crystal alignment; Rapid crystallography

1. Introduction

The understanding of many physical properties in solid-state physics has been achieved by studies of oriented single crystals. These physical properties (electrical resistivity, magnetic susceptibility, etc.) are often anisotropic and their investigation requires accurate knowledge of the quality and the orientation of the crystal in order to perform the measurements along specific crystallographic directions. The Laue method, in which the single crystal is maintained fixed in an incident beam containing a wide spectral range of wavelengths, is the most convenient technique for the determination of the orientation and the symmetry of crystals. It is an easy and an efficient technique compared to monochromatic single-crystal methods. Because of the increasing power of computing and image software techniques, the Laue method is not only efficient in single-crystal characterization but also for quantitative structure determination. For a given position of the crystal the Laue technique allows exploration of a large volume of reciprocal space, while with the monochromatic method we can only explore reciprocal space plane-by-plane at best. In the usual Laue method, the patterns are recorded by flat detector films either in transmission or reflection geometry. In transmission geometry the zone lines are ellipses or parabolas, while in back-reflection geometry they are hyperbolas or straight lines [1]. For both X-rays and neutron radiation, back-reflection geometry is preferable because of the small absorption from the sample compared with transmission geometry. Also, the finite wavelength bandwidth reduces the number of spots at low scattering angles.

To date, only films or image plate detectors have been used for back-reflection geometry, since the incident beam must pass through the detector, a condition that is not possible with gas detectors. However, for certain applications, the exposure time can be very long ranging from one to several hours, and in extreme cases for a complete orientation one or more days. The usual effective exposure time for one setting is between 15 and 60 min for both film and image plate detectors. To reduce this measuring time,

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the use of electronic two-dimensional position-sensitive detectors becomes essential. Such detectors have been limited thus far to non-back-reflection geometry only, with $\alpha < 180^\circ$, where α is the angle between the normal to the detector plane and the incident beam.

A fast Laue diffractometer composed of two-dimensional scintillator detectors was developed for X-rays [2]. The detector works however only in transmission or in a narrow back-reflection region with $\alpha < 180^\circ$. To overcome this difficulty, we have developed a new Laue diffractometer, which allows Laue patterns to be taken at different positions of the detector with respect to the incident beam. Full back-reflection geometry ($\alpha = 180^\circ$) as in the case of the film detector can be achieved but in a much shorter time. The diffractometer is composed of a two-dimensional detector mounted on a 2θ arm, a two-stage tilt goniometer mounted on an ω-rotation, and a video system for the optical alignment of the crystal with respect to the incident neutron beam. The diffractometer is located at the end of the thermal neutron guide H24 at the ILL.

2. The detector

The detector was constructed by Photonic Sciences Limited. The Intensified Neutron Imaging system is a unique product allowing electronic capture of Laue diffraction patterns much more conveniently and in a much shorter time than with film-based methods. The system is based upon two high-performance thermoelectrically cooled image-intensified CCD cameras that view a large-area neutron scintillator via close-focus lenses (Figs. 1 and 2). The exposure of the two cameras is synchronized to take place simultaneously, but they transfer their images to the PC computer serially, where custom software reconstructs a stitched and undistorted image of the diffraction pattern. The active scintillator area of $252 \times 198\,\mathrm{mm}^2$ is thus rendered as a single image of 1680×1320 pixels, each pixel $150\,\mu\mathrm{m}$ on edge. The cameras view the scintillator inside a sealed light-tight box. The incident neutrons pass through this box and the scintillator via a small-diameter light-tight tube that itself passes through the center of the scintillator. The two cameras are positioned in such a way that their views overlap so the tube does not obscure any part of the scintillator.

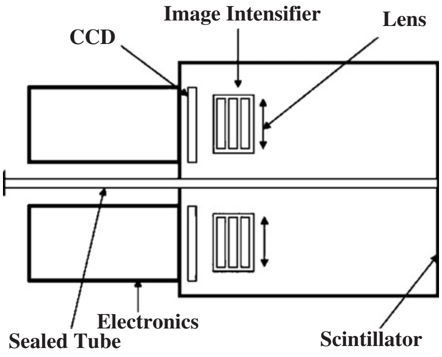

Fig. 1. Layout of the scintillator detectors of OrientExpress.

Fig. 2. OrientExpress: the new Laue diffractometer.

The cameras view the scintillator via f0.95 lenses to give maximum sensitivity. Inside each camera, coupling between the intensifier and the CCD is via a fiber-optic taper which is bonded directly onto the CCD to give maximum transmission and high sensitivity. The photocathode of the intensifier is gated and synchronized with the CCD integration period.

The neutron scintillator is an AST ND screen based on ^{6}LiF with ZnS:Ag added to provide high neutron capture efficiency [3]. The cameras feature digitization of the CCD signal to 12 bits at a choice of 10 or 20 MHz pixel frequency. Integration periods of 1 ms to many minutes are possible, and an on-chip binning option is available independently in both X and Y. The intensifier gain is fully adjustable via software. This allows optimization of the frame rate, resolution, sensitivity, image size and dynamic range.

3. Data acquisition and examples

The camera operation is controlled completely via the PC, from a dialog box integrated into the image-capture analysis software package, Image Pros Plus [4]. Features that can be controlled include image intensifier gain, integration period and on-chip binning. The interfacing with the PC is ensured by an IEEE1394A Fireware interface. The detector is mounted on a 2θ arm, which can sweep an angular range of 180°. The detector-to-sample distance is variable from 32 to 220 mm. All motors of the diffractomer are controlled through the standard ILL instrument-control program MAD running under LINUX.

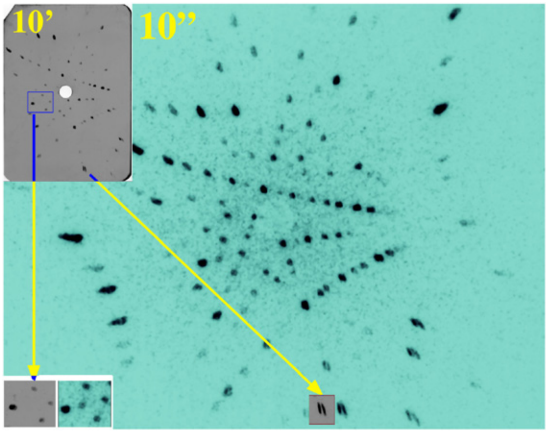

Fig. 3. Laue diffraction pattern obtained from a ruby crystal in 10 s with OrientExpress compared to that obtained from the same crystal in 10 min with Kodak film.

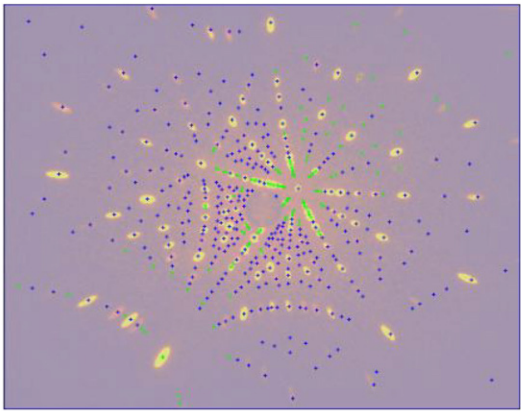

Fig. 4. Laue pattern of a ruby crystal obtained in 50 s, with the indexed pattern superimposed.

Fig. 3 compares the Laue patterns from a ruby crystal of 3 mm diameter obtained with the diffractometer OrientExpress in 10 s and with the old Kodak film camera in a 10 min exposure, both in back-reflection geometry. Some reflections observed in a 10 s acquisition with OrientExpress are not even visible in the 10 min exposure with film. These spots only start to appear in a 12 min film exposure. Another feature is that the well-known high spatial resolution of the film is comparable to that obtained by the scintillator detector as shown in the inset of Fig. 3. Indexing of the Laue diffraction patterns can be performed with either the program OrientExpress [5] running on the PC or the program lauegen [6] of the CCP4 Laue suite [7]; lauegen permits not only determination of the orientation matrix but also intensity integration for later structure refinement.

Fig. 4 shows the observed and indexed pattern for the same ruby crystal in a 50 s exposure as part of a full diffraction data collection to be used for structure refinement. Seven such Laue patterns were acquired, successive patterns separated by a 30° rotation in ω. These could be indexed, integrated and scaled to a common wavelength using programs of the CCP4 Laue suite [7]. Each pattern contains about 500 spots, of which about 250 are single well-resolved reflections. Because of the small beam divergence and small detector point-spread function, the resolution is determined principally by the crystal dimensions and the crystal-to-detector distance. Better resolution of the reflections could be achieved and a higher proportion of resolved reflections, at the expense of a smaller sampling of reciprocal space, by increasng the crystal-to-detector distance.

It is obvious that, in addition to the near hundredfold gain in efficiency in comparison with conventional film-based methods, new possibilities are now available with this new Laue diffractometer. In addition to the classical use of the Laue diffractometer which consists of orienting and characterizing crystals, a more quantitative analysis becomes possible, for example a rapid nuclear structure determination, since extensive diffraction data collection is possible in just a few minutes. Mosaic spread, twinning of crystals as well as preferred orientation in textured samples can also be investigated in a very short measuring time. The field of investigation would become larger if we included a sample environment like a cryostat. Nuclear and magnetic phase transitions, and short-range-order diffuse scattering and domain formation associated with these transitions, could then be studied.

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