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by

B. Buras

Physics Laboratory II, H. C. Ørsted Institute,
University of Copenhagen, DK-2100 Copenhagen, Denmark
and
Risø National Laboratory, DK-4000 Roskilde, Denmark

L. Gerward

Laboratory of Applied Physics III,
Technical University of Denmark, DK-2800 Lyngby, Denmark

J. Staun Olsen and S. Steenstrup

Physics Laboratory II, H. C. Ørsted Institute,
University of Copenhagen, DK-2100 Copenhagen, Denmark

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A spectrometer for x-ray energy-dispersive diffraction using synchrotron radiation

J. Staun Olsen[a], B. Buras[a,c], L. Gerward[b] and S. Steenstrup[a]

a) Physics Laboratory II, H.C. Ørsted Institute, University of Copenhagen, DK-2100 Copenhagen, Denmark

b) Laboratory of Applied Physics III, Technical University of Denmark, DK-2800 Lyngby, Denmark

c) Also Risø National Laboratory, DK-4000 Roskilde, Denmark

Abstract

The paper describes a white-beam x-ray energy-dispersive diffractometer built for HASYLAB in Hamburg, FRG, using the synchrotron radiation from the electron storage ring DORIS. The following features of the instrument are discussed:

Horizontal or vertical scattering plane, collimators, sample environment, remote control of goniometer, data acquisition, energy-sensitive detectors using small-area and large-area detector crystals, modes of operation, powder and single crystal diffraction.

An example is given from a high-pressure study of YbH_2 using a diamond anvil cell.

1. Introduction

The availability of x-ray synchrotron radiation sources and the constructions under way of electron storage rings dedicated to synchrotron radiation research (Buras, Farge and Thompson 1980) have called for the development of instruments making full use of the specific features of synchrotron radiation (European Synchrotron Radiation Facility, Suppl. III, 1979). It is the purpose of this paper to describe a white-beam x-ray energy-dispersive diffractometer, newly built for the HAmburg SYnchrotron radiation LABoratory (HASYLAB), utilizing synchrotron radiation from the electron storage ring DORIS at DESY. The diffractometer has been tested and used during two runs in 1980 at a temporary beamline and will be permanently installed in 1981. The construction is a joint project of Physics Laboratory II, University of Copenhagen, Laboratory of Applied Physics III, Technical University of Denmark and HASYLAB, in collaboration with Risø National Laboratory.

Section 2 of the present paper recalls briefly the principles of white-beam x-ray energy-dispersive diffraction. Section 3 is devoted to a discussion of the pros and cons for a horizontal scattering plane*) and

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*) The scattering plane is defined by the incident and scattered beam directions.

stresses the necessity of remote control. The mechanical construction is described in Section 4 and the electronic equipment in Section 5. Section 6 describes the modes of operation, and gives an example from a high-pressure study of YbH_2.*) This section also serves as a summary.

2. The white-beam x-ray energy-dispersive method

In energy-dispersive diffraction (see, for example, Laine and Lähteenmäki 1980) a beam of polychromatic (white) x-rays is incident on the sample. The wavelength (photon energy) spectrum of the scattered x-rays is measured by a semiconductor detector connected to a multichannel pulse-height analyser. The relation between the modulus of the scattering vector $\vec{\kappa}$, the scattering angle 2θ and the energy E of the scattered photon is given by

$$\kappa = \frac{4\pi}{\lambda} \sin\theta = \frac{4\pi}{hc} E \sin\theta = 1.014 \left(\frac{\text{Å}^{-1}}{\text{keV}}\right) E \cdot \sin\theta \ , \qquad (1)$$

where h is Planck's constant and c the velocity of light. In crystal diffraction the intensity of the scattered beam is peaked at discrete values of $\kappa = 2\pi/d$, where d is the interplanar spacing, characterized by indices HKL. The Bragg equation then reads as follows:

$$E \cdot d \cdot \sin\theta = hc/2 = 6.199 \ (\text{keV} \cdot \text{Å}) \qquad (2)$$

*) Work under way in cooperation with B. Johansson, B. Lebech and H. Skriver.

The scattering angle is defined by slits in the incident and scattered beams when the sample is a powdered crystal, a liquid, an amorphous substance or a rotating single crystal. In the case of a fixed single crystal the scattering angle of a reflection from a particular set of lattice planes is defined by the incident beam and the crystal orientation. A slit is then necessary in the incident beam only.

The reflections from a fixed single crystal can be measured using a small-area detector or a large-area detector. In the former case reflections along a certain line in the reciprocal lattice are recorded, in the latter case the reflections in a certain plane or volume of reciprocal space (Buras, Staun Olsen and Gerward 1980).

The diffractometer described in the present paper is equipped with a small-area detector and the scattering angle can be set remotely at will. If available, a large-area detector can easily be mounted. It is also possible to make an angular scan in the horizontal plane with the small-area detector, thus simulating a large one-dimensional detector along an arc of a circle. This type of measurement has been described by Buras, Staun Olsen and Gerward (1980).

3. Horizontal versus vertical scattering planes; remote control

Synchrotron radiation is inherently collimated in the vertical direction and almost fully polarized in the horizontal plane. Thus a vertical scattering plane would be preferable. However, as will be shown below, the disadvantage of a horizontal scattering plane does not seem to be very great. Moreover, there are many practical difficulties connected with a vertical scattering plane: The mechanical construction (gravitational forces lead to undesired torques), the placing of ovens, cryogenic equipment, high pressure cells, etc. at the sample table and the tilting of the detector dewar. As concerns collimation of the incident beam, the small size of the source (σ_x < 2 mm), its large distance (30 m - 40 m) from the sample and its high spectral brightness enable - by means of slits - the production of an intense beam, with a collimation in the horizontal plane close to the inherent collimation in the vertical plane.

As concerns the loss of reflected power due to polarization, one looses only a factor of 2 for a scattering angle as large as $2\vartheta = 45^{\circ}$. Table 1 shows for this angle the maximum value of the modulus of the scattering vector, which can be measured for a given electron energy of

the storage ring DORIS. Here the general assumption is made that useful intensity is available up to 5 times the critical energy (1/5 of the critical wavelength). The table shows that most crystallographic requirements can be satisfied when the electron energy is about 4 GeV or higher.

In this situation it was decided to construct the diffractometer mainly for use in the horizontal scattering plane. However, by providing the diffractometer with a special detector model (see below) a vertical scattering plane is also possible. In the latter case the scattering angle must be set manually when the x-ray beam is off. In the standard horizontal mode of operation the scattering angle can be set remotely with the beam on.

All movements of the spectrometer with the beam on are remotely controlled because of radiation hazard. Limit switches prevent driving the moving part out of the range permitted by the mechanical construction and driving the detector into the direct beam. Thus the diffractometer is "fool proof" and able to serve as a users' facility.

4. Mechanical construction

A photograph of the mechanical part of the instrument is shown in figure 1. A side and top view of the apparatus is shown in figure 2.

Table 2 shows the precision of all movements described below.

A large table (1)[x] carries the diffractometer. It is a steel construction with a manually adjustable height. On the top of the table a perfectly plane surface of araldit has been cast and covered with a plastic laminat in order to enable the use of air cushions.

The whole instrument can be moved perpendicular to the direction of the incident beam by means of a stepping motor, permitting high precision alignment. For this movement the apparatus is lifted by five air cushions (required pressure about 4 atm).

The bottom of the diffractometer consists of a Huber turn-table (3) connected to the detector arm (2). The scattering angle can be changed with a high precision by a stepping motor, the detector arm being supported by two air cushions. Another Huber turn-table (4) for rotation of the sample is placed on the table (3). It is provided with a table (5) (called z) of variable height, on top of which is placed a table (6) (called x,y) enabling horizontal xy movements of the sample. This arrangement

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x) The numbers within paranthesis refer to the corresponding numbers in figure 2.

allows a very precise placing of samples with dimensions of the order of micrometers in the direct beam. Finally the diffractometer is equipped with a large Huber goniometer head (7). The whole construction is sufficiently rigid to take a load of 100 kg without decreasing the precision of any setting.

The detector (9) is placed on a table of variable height controlled by a stepping motor (8). The support of the detector table can slide on the detector arm (2), enabling a remote control of the sample-detector distance. As can be seen from the top view (figure 2) the detector arm (2) is "tangential" to the turn-table (3). A line through the centre of the detector crystal and the centre of the turn-table (3) is parallel to the detector arm (2). This construction was chosen because it enables the use of a vertical scattering plane with a special detector model as shown in the side view of figure 2. Scattering angles between -10° and $+110^{\circ}$ with respect to the horizontal plane are possible. The detector crystal cannot be rotated by a remote control in present-day detectors. Thus a continuous angular scan in the vertical plane is not possible.

The incident and the scattered beams are defined by slits (no slits are required in the scattered beam for a fixed single crystal as discussed at the end of Section 2). The slit in the incident beam is

in width,
made of tantalum and continously variable. The position of the slit in the incident beam can be moved manually ±5 mm in steps of 1 µm in both vertical and horizontal direction. For a small sample the scattered beam is defined by a tantalum slit placed directly onto the detector. For a large sample an additional slit has to be placed between the sample and the detector. The width of the slits can be changed in steps from 100 µm to 800 µm.

5. Electronic equipment

The electronic system is composed of two parts, one for data acquisition, and one for motor control.

The data handling system is the Canberra/Jupiter system. A block diagram is shown in figure 3. The system is centred around a PDP 11/03 computer with two RX02 floppy disk stations, a lineprinter, a Tektronix console and a Canberra series 80 multichannel analyser. The multichannel analyser can be operated manually or via the computer. Data collected by the multichannel analyser are transferred to the PDP 11/03 and stored on floppy disks. The programs, written in Fortran, controlling the diffractometer and the multichannel analyser are also located on the floppy disks.

The size of the PDP 11/03 permits only simple data analysis. More advanced spectral analysis, for instance by the Rietveld method, requires a large computer.

A Camac system shown schematically in figure 4 is used for the motor control. Stepping motor drivers trigger the SLO-SYN (ST 103 AX) translators and control the SLO-SYN stepping motors. Three motors at a time can be driven, either manually or by the computer. The system can easily be extended to more motors. Displays for motor positions are situated in the Camac crate. Figure 4 also shows the controls for the air cushions and the limit switches.

6. Modes of operation - Summary

The diffractometer is designed to work in two basic modes, numbered a and b below. However, several other modes of operation can be devised, for example c and d mentioned below.

a. White-beam x-ray energy-dispersive powder, amorphous and liquid sample diffraction with either horizontal or vertical scattering plane.

The powdered sample can be rotated either by rotating the turntable (4) or by an additional mechanism placed on top of the goniometer head (7).

The precision of all movements enables the use of very small samples and scans across the sample in two directions. This is of great value, for example when studying a sample in a diamond anvil high-pressure cell with a pressure gradient.

The remote control of the scattering angle in the horizontal plane is especially important when the diffraction spectrum contains additional fluorescence lines and absorption edges.

Because the technique involves a fixed geometry it can be particularly useful where the sample environment is to be changed. Facilities are provided for mounting cryostats, ovens, magnets, etc. on top of the goniometer head. As an example we show in figure 5 an x-ray energy-dispersive diffraction spectrum of YbH_2. The spectrum has been obtained using a diamond anvil high-pressure cell. The applied pressure is about 28 G.Pa.

b. White-beam x-ray energy-dispersive single crystal diffraction

In this mode of operation it is possible to carry out measurements at any point of the reciprocal lattice that can be reached by the scattering vector (see Table 1). If required an Euler cradle can be placed on the sample table. In particular, one can measure reflections either along a certain direction in the reciprocal lattice (e.g. forbidden reflections,

satellites, etc.), or in a certain plane of the reciprocal lattice as discussed in Section 2. By rotating the sample and simultaneously changing the scattering angle, a reflection can be moved across an absorption edge. This operation is equivalent to a tuning of the incident beam in a monochromatic-beam diffractometer (Fukamachi, Hosoya and Okunuki 1976).

c. White-beam x-ray energy-dispersive small angle diffraction

In this mode of operation, the detector has to be placed outside the diffractometer table (1) in order to achieve a sufficiently long sample-detector distance. An x-ray guide tube, evacuated or helium filled, might be necessary.

d. Monochromatic-beam diffraction

This mode of operation can be achieved in two ways:

(i) The diffractometer is placed at a monochromatic x-ray beamline. The multichannel pulse-height analyser can now be used as a single-channel pulse-height analyser in order to increase the signal-to-background ratio and to eliminate the harmonics from the monochromator.

(ii) A white incident beam is used. The scattered beam is "monochromatized" using a single-channel pulse-height analyser with a narrow energy window. The resolution is mainly determined

by the detector.

Neither of these two modes of operation seems to be superior to an optimized monochromatic-beam diffractometer. However, these possibilities illustrate the great versatility of the instrument at no extra cost.

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Table 1. Maximum modulus of the scattering vector and minimum interplanar spacing for $2\theta = 45^{\circ}$ at several electron energies at DORIS

E(GeV)	κ_{max} ($\AA^{-1}$)	d_{min} ($\AA$)
3.0	9.6	0.66
3.3	12.8	0.49
3.7	18.0	0.35
4.0	22.7	0.28
5.0	44.4	0.14

Table 2. Precision of the spectrometer movements

Motor number	Movement	Movement per step of the stepping motor
1	Detector up and down	0.15 µm
2	Detector slide	16 µm
3	Traverse movement of the whole diffractometer	1.3 µm
4	Detector arm (2θ)	0.5 millidegree
5	Sample turn-table (θ)	0.5 millidegree
6	Sample up and down	0.025 µm
7	Sample y-movement	1/6 µm
8	Goniometer head, upper tilt	0.25 millidegree
9	Goniometer head, lower tilt	0.25 millidegree
10	Sample x-movement	1/6 µm

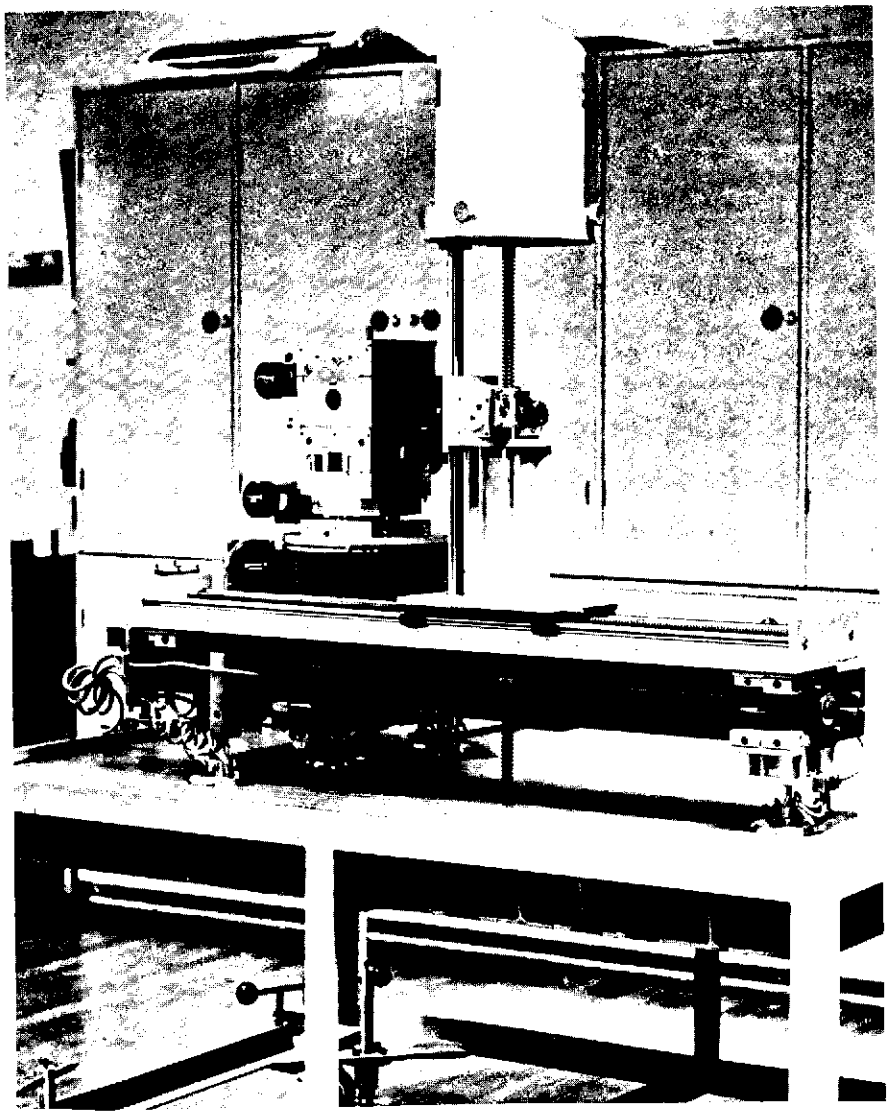

Fig. 1. Photograph of the mechanical part of the instrument.

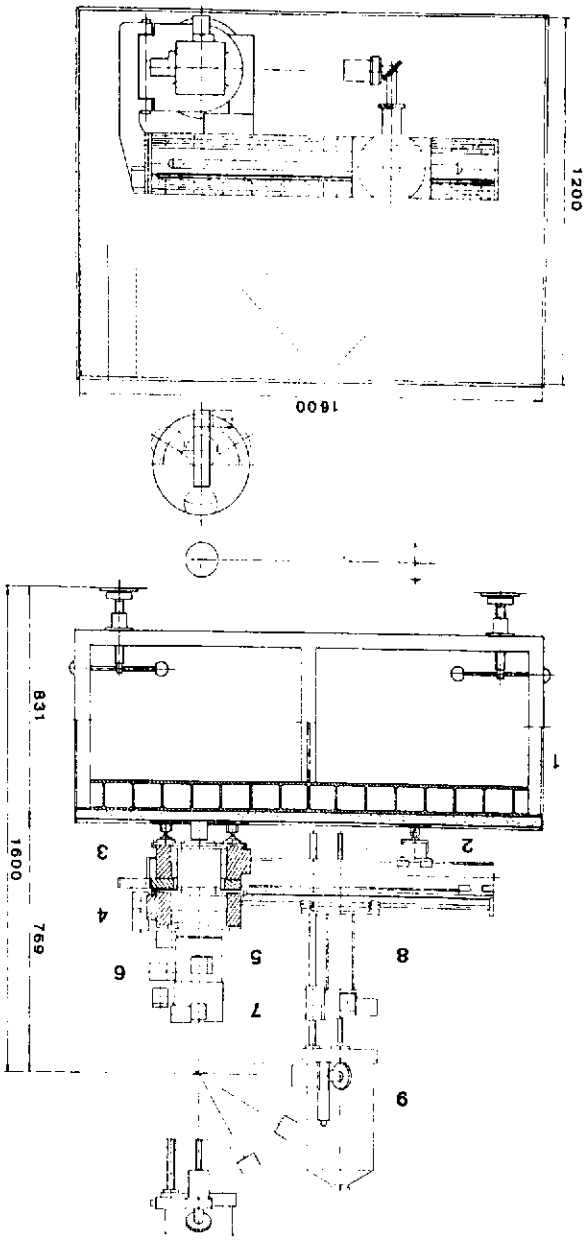

Fig. 2. Side and top view of the instrument. (1) Table, (2) detector arm, (3) turn-table for the detector arm, (4) turn-table for rotation of the sample, (5) z-table, (6) xy-table, (7) goniometer head, (8) detector table with variable height and (9) ultra-pure germanium detector.

Fig. 3. System for data acquisition and display. The Jupiter system is contained within the dashed line. Outside are the connections to the proces control system.

Fig. 4. Proces control system according to the Camac system. Only one stepping motor is shown in the figure. However, in practice there are ten stepping motors (numbered 1-10 in Table 2). Three motors at a time can be connected to the system using the connectors I, II and III of the resistor box.

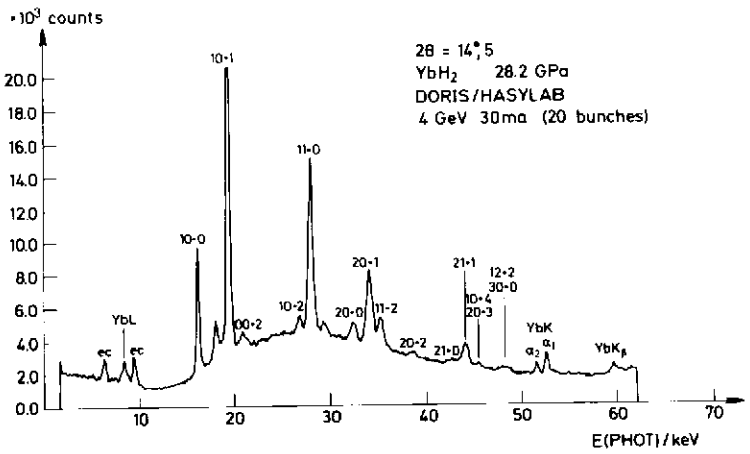

Fig. 5. Diffraction spectrum of YbH_2 in diamond anvil Pressure 28,2 GPa. Scattering angle $2\theta = 14,6^{O}$, counting time 1000 s.