A Low-Temperature Weissenberg Camera for Neutrons

BY D. HOHLWEIN* AND A. F. WRIGHT

Institut Laue-Langevin, 156X, 38042 Grenoble CEDEX, France

(Received 14 September 1979; accepted 21 August 1980)

Abstract

The construction and performance of a neutron Weissenberg camera for temperatures down to 4 K is described. Exposure times are similar to the X-ray case. The background from the cryostat walls can be eliminated. Examples of applications in the fields of magnetic structures which are incommensurate with their chemical unit cells, crystallographic phase transitions and diffuse scattering are presented.

I. Introduction

The development of neutron film methods in recent years (Hohlwein, 1978) has shown that the classical X-ray film techniques like Weissenberg and precession are also possible with neutrons. Quality and exposure times are similar to those for X-rays in spite of a flux about 10^4 times lower at the sample position (Hohlwein & Joswig, 1976). The reason for this is a converter-film system which is about 10^3 times more efficient than normal X-ray films, combined with the possibility of using crystals with volumes of the order of 10 mm^3 without having absorption problems.

The advantage of a film compared to a counter lies in its multidetector properties. A 100 × 100 mm film with a resolution of 0.1 mm has 10^6 detector elements! The precision of intensity measurements is naturally lower than with a counter but the relatively good quality of film data has been proved with a structure refinement of histidine for which the parameters compared well with those from a refinement of conventional four-circle diffractometer data (Hohlwein, 1977).

A Weissenberg camera equipped with a helium cryostat is especially advantageous for the study of magnetic phases which appear at low temperatures, where, for example, the lattice of the new phase is not known beforehand, and is only revealed by the use of neutrons. Phase transitions within the chemical structure involving light atoms (particularly hydrogen) can also be more readily observed with the neutron camera than with X-rays as a result of the irregular variation of scattering length with the atomic weight throughout the periodic table of elements.

II. Principal features of the apparatus

The helium Weissenberg camera uses one of the two beams of the instrument D12 (Hohlwein, 1975) which is situated at the end of a thermal neutron guide of the ILL High-Flux Reactor.

The monochromatic beam (normally 1.6 Å) is reflected by a graphite monochromator and passes two collimators with circular holes before reaching the sample at a distance of 1 m. Vertical and horizontal divergences are about 0.4°, the wavelength spread is 2°, and the flux at the sample is 2 × 10^15 neutrons m^-2 s^-1. The geometrical arrangement corresponds to the normal-beam Weissenberg technique. Translation of the camera and the rotation axis of the sample are both vertical, whilst the monochromatic beam lies in the horizontal plane.

In the present arrangement (Fig. 1), the crystal is mounted in a fixed position and orientation inside the tail of a helium cryostat, type CT14, Cryogenic Associates of Indianapolis, USA. With the tails modified for this use, the temperature is variable over the range 5 to 300 K, using a Thor 3010 controller from Thor Cryogenics, Oxon, England. Helium consumption is 31 per day at 10 K. Rotation of the crystal as well as centering and orientation are achieved by operations on the whole cryostat. The cryostat is mounted on a goniometer comprising two arcs and two micrometric translation tables, which are supported by a turntable driven by a step motor. Inclinations of the arcs are limited to ±2° in this arrangement because of interference between the cryostat tail and the camera mechanism.

This limitation requires careful alignment of the crystal prior to its being mounted on the instrument. An optical bench mounted along the incident-beam axis serves to support the camera and collimation system, and is used for all alignment procedures. A vertical translation mechanism is fixed to the bench and carries two half-cylindrical cameras of radius 42 mm which can be easily positioned around the cryostat tail. The translation is by an independent stepping motor, thus allowing variable resolution and range on the film.

III. Camera mechanism

The neutron film converter has been described in detail elsewhere, and so will be only briefly discussed here. The

* Present address: Institut für Kristallographie der Universität, Charlottenstrasse 33, 74 Tübingen, Federal Republic of Germany.
converter material is a foil of 0.5 mm thickness made of finely ground $^{6}$LiF neutron absorber and silver-doped ZnS phosphor in a clear epoxy resin binder. In the half-cylinder film cassettes, the Kodak Regulix film is pressed between two converter foils. Close contact between the converter and the film is essential for good resolution of the neutron image. Consequently, the cassettes must be precisely and strongly made as well as being transparent to neutrons. In all cases where the incident or diffracted neutron beam must pass through a component of the apparatus, pure aluminium has been used in its construction. Diffraction from the aluminium used in the construction of the thin outer envelope of the cryostat tail and thermal screens does, however, give rise to a background powder pattern superimposed on the film after long exposure times. In order to suppress this effect we use a 'radial venetian blind' system of area collimation (Wright & Berneron, 1981) in conjunction with conventional layer-line screens, which occupy a position between the cryostat and the film cassette (see Fig. 1b). The area collimator consists of a cylindrical arrangement of Gd$_2$O$_3$-coated mylar lamellae glued under tension along the 150 mm long cylindrical axis to an aluminium support. The foils are 0.3 mm thick and 5 mm wide in the radial direction reaching a maximum radius of 35 mm, and the mean separation between foils is 1 mm. The collimator effectively defines a zone of radius 7 mm around the rotation axis, from which diffracted neutrons can reach the film. In practice, only neutrons diffracted from the crystal are recorded on the film. All neutrons diffracted from the cryostat walls and screens, or from any container of radius greater than ~10 mm used to contain the crystal (e.g. in a special atmosphere), are absorbed by the collimator.

Fig. 1. (a) Section of the Weissenberg camera through the sample position seen along the neutron beam. The turntable goniometer unit is only partly shown to indicate the optical bench supports before and after. A beam stop (not shown) is situated between the cryostat and the oscillating collimator. (b) The role of the oscillating collimator in reducing background scattering from the cryostat screens.

Fig. 2. First-layer \((k=1)\) Weissenberg photographs of \(N,N'\)-dimethylnitramine (Filhol et al., 1980). (a) Room temperature; space group \(P2_1/m, Z=2\). (b) Below the phase transition at 111 K; space group \(P2_1/c, Z=4\). Additional reflections indicate a doubling of the \(c\) parameter.
Uniformity of transmission is achieved by oscillation of the cylindrical collimator. The frequency of oscillation is not important, providing motion is much faster than the crystal rotation in order that any position on the film records the transmission integrated over several 'windows' between the foils for any crystal position. The angular velocity of oscillation must be constant, which is achieved through the operation of a continuously turning Archimedes spiral cam acting through a push rod tangential to the collimator axis. For simplicity, the layer-line screens made of aluminium coated with Gd$_2$O$_3$ are supported on the cylindrical collimator and may be moved independently along the rotation axis. Suppression of the background is therefore possible in Weissenberg and crystal-oscillation modes of operation.

IV. Applications

As already mentioned, one of the most important applications will be the study of magnetic phases. In particular, the reflections of incommensurate magnetic structures can easily be found. In the substance DyMn$_3$O$_5$ a propagation vector of the form $\mathbf{[\mathbf{1},0,0]}$ has been detected (Wilkinson & Forsyth, 1978). With a large ratio between the translation of the camera and the rotation of the crystal, a very small propagation vector $\mathbf{[0,0,0.02]}$ could be determined in NaMnCrF$_6$ (de Pape, Boucher, Courbion & Wright, 1978).

A crystallographic phase transition has been studied in $N,N'$-dimethyl nitramine (Filhol, Bravic, Rey-Lafon & Thomas, 1980). A doubling of the unit cell in an unexpected direction could be observed on Weissenberg photographs of upper layers (Fig. 2). Superlattice reflections readily observable on this photograph are of order $1/100$ (e.g. $3.5,1,-3.5$) to $1/1000$ (e.g. $4.5,1,-3.5$) less intense than principal reflections, which are considerably overexposed. The exposure time was 11 h.

An example for the study of the diffuse scattering near a phase transition is the work on biphenyl (Meinnel, Baudour, Cailleau, Toupet, 1978). Anisotropic diffuse spots (becoming Bragg spots in the low-temperature phase) can be observed above the phase transition indicating large correlations along the $b$ direction.

V. Conclusion

The good performance of the instrument has been proved in a number of experiments, some of which are mentioned above. The main fields of application are the study of magnetic structures which are incommensurate with their chemical unit cells, crystallographic phase transitions and different kinds of diffuse scattering.

The authors are indebted to the technical staff of the ILL for their valuable help, especially to Mr Berneron. We thank also Alain Filhol for making available photographs prior to publication.

References


