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Influence of Polarization of the Incident Beam on Integrated Intensities in X-ray Energy-Dispersive Diffractometry

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Polarization measurements of the primary X-ray beam produced by thick copper and tungsten anodes are reported and formulas derived for integrated intensities of Bragg reflections in energy-dispersive diffract-ometry with the polarization of the primary beam taken into account. It was found that for an angle of 45° between the scattering plane and the plane containing the electron beam and the primary beam, the influence of polarization vanishes, while it increases as the angle changes from 45° to either 0 or 90°. For the latter values, the influence of polarization is considerable at high photon energies and at scattering angles close to 90°.

1. Introduction

In conventional X-ray crystallographic studies, mono-chromatic characteristic radiation from the anode is used. It is unpolarized (in case of characteristic K X-radiation) and the polarization factors appearing in the formulas for integrated intensities – both for powders and single crystals – have the well known form arising from averaging the polarization factors for a linearly polarized beam over all possible directions of polarization (Zachariasen, 1945; Laue, 1960). In the energy-dispersive method, and sometimes also in double-crystal spectroscopy, the 'white' radiation

(*Bremsstrahlung*) is used. It is well known that in the case of a thin target the white radiation is strongly polarized in the direction parallel to the electron beam incident on the target (Kuckuck & Ebert, 1973; Tseng & Pratt, 1973; and references therein). Our knowledge of polarization in the case of thick targets stems mainly from early measurements (Siegbahn, 1925; Ross, 1928; and references therein). Recently, Slivinsky (1971) measured the polarization of X-radiation emitted by commercial X-ray tubes with thick targets and found that it is strongly polarized in the direction parallel to the electron beam at the high-energy limit of the *Bremsstrahlung* spectrum, while the lower-energy X-rays exhibit a low but constant polarization in the same direction. However, to the best knowledge of the

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present authors, no quantitative data have been published. This finding is in general agreement with most of the previous measurements and may be qualitatively explained (Siegbahn, 1925) by the fact that the highest-energy photons are only emitted from the surface of a thick target, while low-energy photons are also emitted from the internal part of a target where the electrons are deflected from the original path as they traverse the material of the target.

Until very recently the polarization of the primary X-ray beam was not taken into account in crystallographic studies using the energy-dispersive white-beam method (see, for example, Laine, Lähteenmäki & Hämäläinen, 1974). For this reason, we studied the influence of the polarization of the primary X-ray beam on integrated intensities of Bragg reflections obtained by means of energy-dispersive diffractometry. Some preliminary results were previously presented at the Tenth International Congress of Crystallography in Amsterdam (Alstrup, Gerward, Selsmark, Buras & Staun Olsen, 1975). At the same time in a study of liquid mercury by means of the energy-dispersive method, Prober & Schultz (1975) introduced a polarization correction; however, their treatment is mainly limited to the problem they were investigating.

This paper describes direct measurements of the polarization of *Bremsstrahlung* from thick copper and tungsten targets as a function of photon energy. We should like to stress that very high accuracy of polarization measurements was not our aim, because we chiefly wished to illustrate the importance of the polarization of the primary beam for energy-dispersive diffractometry and to estimate roughly the error if the polarization effects were not accounted for.

§ 2 contains the definition, the outline of the principles of the measurements and the derivation of the basic formulas. The experimental details are briefly described in § 3, while § 4 contains the results of measurements followed in § 5 by a detailed discussion of the influence of polarization on the integrated intensities of Bragg reflections in the energy-dispersive method.

2. Definitions, principles of measurements, basic formulas

We introduce two coordinate systems: one (x,y,z) – called the scattering frame – used for describing the scattering experiment, and the other (x',y',z') – called the source frame – used for describing the primary beam.

In the case of the source frame (Fig. 1), the z' axis is oriented along the direction of the electron beam of the X-ray tube and the y' axis along the direction of the primary beam. The polarization $P(E)$ of the primary beam is defined in the usual way:

$$P(E) = [i'_{0,n}(E) - i'_{0,p}(E)]/[i'_{0,n}(E) + i'_{0,p}(E)], \quad (1)$$

where $i'_{0,n}(E)$ and $i'_{0,p}(E)$ are the intensities per unit interval of energy E of the normal and parallel polarization components, respectively, with respect to the $(y'z')$ plane.

We performed a scattering experiment based on Barkla's (1905) original method of polarization measurements using an amorphous scatter and a $2\theta_0 = 90°$ scattering angle (Fig. 1). From Thomson's formula (Zachariasen, 1945) it follows that the intensity $I(E)$ scattered in the direction z (see Fig. 1) is equal to

$$I(E) = [i'_{0,p}(E) \sin^2\alpha + i'_{0,n}(E) \cos^2\alpha]k(E), \quad (2)$$

where $k(E)$ depends only on the sample and the photon energy E, and α is the angle between the z and z' axes. If we measure $I(E)$ for two different angles α_1 and α_2, the polarization $P(E)$ of photons of energy E is easily obtained from (1) and (2):

$$P(E) = \frac{I_2(E) - I_1(E)}{I_1(E) \cos 2\alpha_2 - I_2(E) \cos 2\alpha_1}. \quad (3)$$

We now derive the formulas for integrated intensities for powdered crystals and mosaic single crystals using the kinematical approximation, and for single crystals using the dynamical theory in all three cases, taking into account the polarization $P(E)$ of the incident beam. These are used in the final discussion in § 5.

For a fixed scattering angle and the energy-dispersive method, it follows from the formulas of integrated intensities (see, for example, Buras & Gerward, 1975) and Thomson's formula that the integrated intensity of the *HKL* reflection measured in the scattering frame,
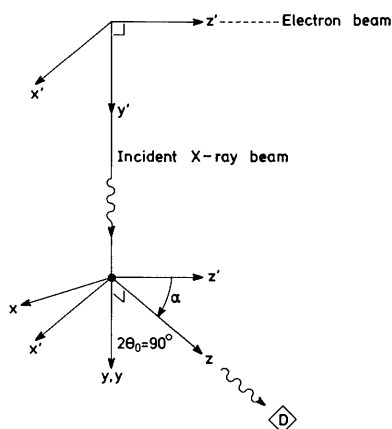

Fig. 1. Experimental arrangement showing the scattering on an amorphous material with a fixed scattering angle $2\theta_0 = 90°$. (x',y',z') and (x,y,z) are the source frame and the scattering frame respectively. D is the detector.

both for a powdered crystal and for a mosaic single crystal,* is

$$I_H(\text{kin}) = \text{const.} \times \{jd^2|F|^2[i_{0,n}(E) + i_{0,p}(E)\cos^2 2\theta_0]\}_H \qquad (4a)$$

and, in the case of a perfect single crystal, is

$$I_H(\text{dyn}) = \text{const.} \times \{d|F|[i_{0,n}(E) + i_{0,p}(E)|\cos 2\theta_0|]\}_H, \qquad (4b)$$

where j is the multiplicity factor, d the lattice spacing, F the structure factor, H indicates the reflection indices HKL, and $i_{0,n}(E)$ and $i_{0,p}(E)$ are the intensities per unit interval of energy of the normal and parallel polarization components with respect to the scattering plane (yz).

Performing a transformation of the intensity components $i'_{0,n}$ and $i'_{0,p}$ from the source frame to the corresponding unprimed intensities in the scattering frame, and taking into account that

$$i_0(E) = i_{0,n}(E) + i_{0,p}(E) = i'_{0,n}(E) + i'_{0,p}(E), \qquad (5)$$

one obtains

$$I_H(\text{kin}) = \text{const.} \times \{jd^2|F|^2 i_0(E)\, C_p(\text{kin})\,(E, \theta_0, \alpha)\}_H \qquad (6a)$$

$$I_H(\text{dyn}) = \text{const.} \times \{d^2|F|i_0(E)C_p(\text{dyn})(E, \theta_0, \alpha)\}_H, \qquad (6b)$$

where $C_p(\text{kin})(E, \theta_0, \alpha)$ and $C_p(\text{dyn})(E, \theta_0, \alpha)$ are the polarization factors for powdered or mosaic single crystals and perfect single crystals, respectively:

$$C_p(\text{kin})\,(E, \alpha, \theta_0) = \tfrac{1}{2}[1 + \cos^2 2\theta_0 + P(E)\cos 2\alpha \sin^2 2\theta_0] \qquad (7a)$$

$$C_p(\text{dyn})\,(E, \alpha, \theta_0) = \tfrac{1}{2}[1 + |\cos 2\theta_0| + P(E)\cos 2\alpha(1 - |\cos 2\theta_0|)]. \qquad (7b)$$

3. Experiment

Both copper and tungsten X-ray tubes were used to produce X-rays. The Philips tube was provided with stabilized (0·01%) current and voltage from a Philips generator, type 1130. The detector system was a Princeton Gamma Tech ultra-pure germanium detector (150 eV FWHM at 5·9 keV).

A special holder was constructed for the X-ray tube so that it could be moved around two axes, one along the electron beam direction and one perpendicular to the beam direction. The electron beam was almost perpendicular to the X-ray beam (the take-off angle was about 3·5°).

As already mentioned, in the experiment the vertical X-ray beam was scattered ($2\theta_0 = 90°$) by an amorphous

material (perspex), rotating around a vertical axis to avoid absorption corrections when the angle α was changed.

4. Experimental results

In Fig. 2 are shown the results of measurements of P as a functions of E/T, where T is the maximum tube voltage, in the case of a tungsten anode. The measurements were made for values of T equal to 25, 30, 40 and 45 keV, and within the experimental errors shown in the figure we found that the results for different values of T lie on one curve if P is plotted versus E/T. The results for the copper anode also lie on one curve if plotted versus E/T; however, this curve is different from that for tungsten, as can be seen from Fig. 3.

The accuracy of our measurements is not very good. The accuracy at low energies is influenced by the absorption edge of the germanium detector at 11·1 keV, and near the high-energy limit by the low intensity. In this situation we made no corrections for X-ray incoherent scattering because they would be much smaller than our estimated errors.

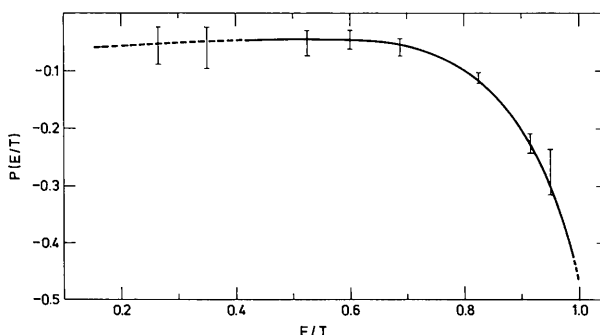The measured polarization is in agreement with the

Fig. 2. Polarization of primary X-ray beam (tungsten tube) as function of E/T (E photon energy, T incident electron energy). The curve is a smoothed result from scattering on an amorphous material ($T = 25$–45 keV).
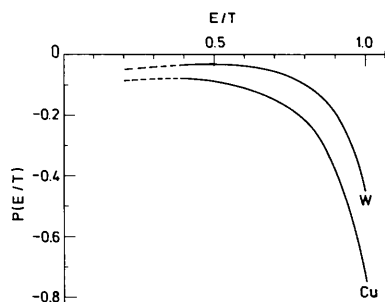
Fig. 3. Polarization as function of E/T measured for a copper tube and a tungsten tube (smoothed curves).

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* The same formula (but with different constants) is valid both for a mosaic single crystal and for a powdered crystal within the kinematical approximation.

qualitative statement of Slivinsky (1971) and provides the necessary data for calculations of the influence of polarization on integrated intensities of Bragg reflections in energy-dispersive diffractometry.

5. Influence of polarization on integrated Bragg reflections

From formulas (7a) and (7b) we see that for unpolarized radiation $[P(E) = 0]$, the polarization factors are given by the known expressions

$$C_p(\text{kin}) = \tfrac{1}{2}(1 + \cos^2 2\theta_0) = C_0(\text{kin})$$

$$C_p(\text{dyn}) = \tfrac{1}{2}(1 + |\cos^2 2\theta_0|) = C_0(\text{dyn}).$$

An interesting feature also appearing from (7a) and (7b) is that *the influence of polarization can be cancelled completely by choosing* $\alpha = 45°$. At this setting the incident beam can be treated as unpolarized for all photon energies, irrespective of the value of P, the polarization of the primary beam. On the other hand, the influence of the polarization is greatest for $|\cos 2\alpha| = 1$, *i.e.* when the plane containing the electron beam and the incident X-ray beam is either parallel or perpendicular to the scattering plane.

The influence of the polarization on the integrated intensities in a given experiment with a tungsten tube is presented in Fig. 4, which shows the relative change of the polarization factor $(C_p - C_0)/C_0$, as a function of the photon energy and the scattering angle. Because of the previously mentioned poor resolution at low values of E/T, the curves at low E/T are dashed. The curves are drawn for $|\cos 2\alpha| = 1$, and it can be seen from Fig. 4 that the influence of polarization on the integrated intensities is considerable at high energies and at a scattering angle of around 90°. At energies below one half of the high-energy limit, the influence of polarization is negligible for all scattering angles, expecially if one is concerned with relative values of integrated intensities only. The integrated intensities of the powder sample are more affected than those of the large perfect crystal. For a scattering angle equal to 20°, the influence of polarization is less than 2·5% for the powder over the whole energy range, whereas for the large perfect crystal it is less than 1·5%.

Formulas (6) and (7) are of a general nature for X-ray energy-dispersive diffractometry and can also be used if synchrotron radiation is the source of X-radiation.
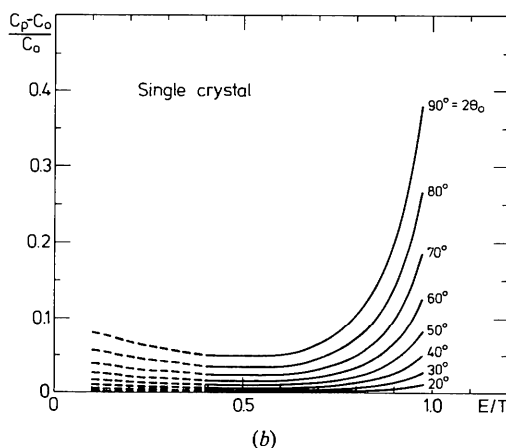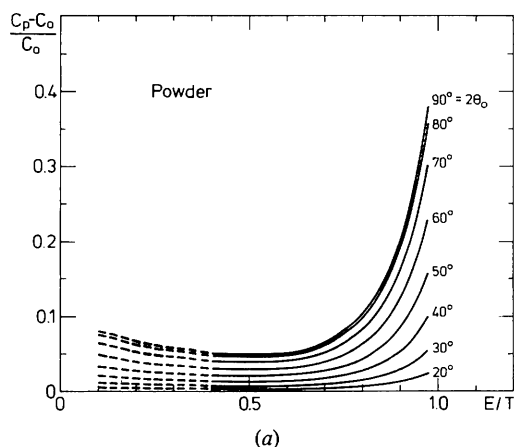
Fig. 4. The relative change of the polarization factor due to the polarization of the X-ray beam from a tungsten tube. The curves are drawn for $|\cos 2\alpha| = 1$; (a) powder or mosaic single crystal, (b) large perfect crystal.

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