douze plans *hkl* du Tableau 1:

$$d_{hkl}(160\,^\circ\mathrm{C}) = d_{hkl}(22\,^\circ\mathrm{C}) + \Delta T \,.\, (\alpha_{obs}) \,.$$

Ensuite, à l'aide du programme des moindres carrés utilisé dans l'article de Bouvaist & Weigel (1970), on peut trouver les paramètres de la maille à 160°C:

$$T = 160\,^\circ\mathrm{C};\ a = 7{,}826 \pm 0{,}003;\ b = 5{,}644\ \text{Å} \pm 0{,}003;$$
$$c = 8{,}477 \pm 0{,}003\ \text{Å};\ \beta = 124^\circ\ 44' \pm 5' \,.$$

Il est alors possible de calculer les coefficients de dilatation α_{hkl} pour tous les plans (*hkl*)

$$\alpha_{hkl}(\mathrm{calc}) = \Delta d_{hkl}/d_{hkl}\,\Delta T \,.$$

Nous avons trouvé un accord entre α_{calc} et α_{obs} aussi bon que par la première méthode. Cependant cette deuxième méthode à le désavantage de ne pas fournir les coefficients principaux de dilatation et leurs écarts-types, et aussi d'utiliser des mesures relatives $\Delta\theta$ pour calculer d_{hkl} (160°C). C'est pour ces raisons que nous avons préféré la première méthode et que nous conseillons vivement son utilisation.

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The Determination of the Atomic Scattering Factor of Germanium by a Pendellösung Method

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(*Received* 15 *September* 1969 *and in revised form* 17 *November* 1969)

The atomic scattering factor of germanium has been obtained from measurements of the Pendellösung period in wedge-shaped single crystals with a three-crystal X-ray spectrometer. For the 220 reflexion the value is 23·78 ± 0·35, in good agreement with the theoretical value of 23·76. Fine structure in the profile of the rocking curves has been observed in thickness regions close to Pendellösung minima.

Introduction

Several authors have used the Pendellösung technique to measure the atomic scattering factor of quartz (Kato & Lang, 1959), of silicon (Hattori, Kuriyama, Katagawa & Kato, 1965; Hart, 1966; Hart & Milne, 1969; Hattori & Kato, 1966; Kato & Tanemura, 1967; Yamamoto, Homma & Kato, 1968) and of germanium (Batterman & Patel, 1968).

In this work Pendellösung measurements of the atomic scattering factor of wedge-shaped germanium single crystals were performed with a three-crystal X-ray spectrometer. In order to obtain the depth periodicity of the diffracted intensity, measurements of rocking curves were made for various crystal thicknesses.

The appearance of a fine structure in the profile of the rocking curves has been observed for thin silicon crystals (Kohra & Kikuta, 1968; Kikuta & Kohra, 1968; Lefeld-Sosnowska & Malgrange, 1968). We have observed a similar phenomenon for a thin germanium crystal.

Theory

The plane wave dynamical theory of X-ray diffraction in perfect crystals which has been developed by von Laue (1960) gives the following formula for the intensity of the reflected beam for the symmetric Laue case and for a centrosymmetric crystal:

$$R = \frac{\exp(-\mu_0 D/\gamma_0)}{4\cosh^2 v_r}\{\exp(\sigma'' D) + \exp(-\sigma'' D) - 2\cos[2\pi k D(\delta_{1r} - \delta_{2r})]\} \quad (1)$$

where, according to von Laue's notation and under the condition that $\chi_{rh} \gg \chi_{ih}$,

$$\sigma'' = \frac{4\pi k C \chi_{rh}\chi_{ih}}{\gamma_0\left\{\left(\frac{\beta_r}{C}\right)^2 + 4\chi_{rh}^2\right\}^{1/2}},$$

$$\delta_{1r} - \delta_{2r} = \frac{C}{2\gamma_0}\left\{\left(\frac{\beta_r}{C}\right)^2 + 4\chi_{rh}^2\right\}^{1/2},$$

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$$\sinh v_r = \frac{\beta_r}{2C|\chi_{rh}|},$$

$$\beta_r = 2(\theta_B - \theta)\sin 2\theta_B,$$

where

λ wavelength of the incident beam in vacuum ($1/\lambda = k$),
θ glancing angle,
θ_B Bragg angle,
μ_0 linear absorption coefficient,
D crystal thickness,
C polarization factor,
γ_0 direction cosine of the incident beam relative to the surface,
χ_{rh}, χ_{ih} real and imaginary parts of the Fourier coefficient of order h of the dielectric susceptibility,

$$\chi_{r220} = -\frac{e^2\lambda^2}{\pi mc^2}\frac{8(f_0+\Delta f')}{V},$$

$$\chi_{i220} = -\frac{e^2\lambda^2}{\pi mc^2}\frac{8\Delta f''}{V},$$

$\frac{e^2}{mc^2}$ classical electron radius,

f_0 atomic scattering factor neglecting anomalous scattering,
$\Delta f'$, $\Delta f''$ real and imaginary parts of the anomalous contribution and
V volume of the unit cell.

The spacing ΔD between the Pendellösung fringes which corresponds to one oscillation period can easily be found from (1):

$$\Delta D = \frac{2\gamma_0}{kC\left\{\left(\frac{\beta_r}{C}\right)^2 + 4\chi_{rh}^2\right\}^{1/2}}. \quad (2)$$

When $\theta = \theta_B$ one has

$$\Delta D_0 = \frac{\gamma_0}{kC|\chi_{rh}|}. \quad (3)$$

By using equation (3) the scattering factor f_0 can be determined from the measurement of ΔD_0.

Numerical calculations of the reflexion curves were carried out using equation (1) for the 220 reflexion of germanium, Mo $K\alpha_1$ radiation and the thickness interval $0 \le D \le 200$ μm. The following values of the input parameters were used:

$\mu_0 = 318$ cm^{-1} (Grimvall & Persson, 1969), $\chi_{r220} = -462\times10^{-8}$, $\chi^{\sigma}_{i220} = -35\times10^{-8}$ and $\chi^{\pi}_{i220} = -32\times10^{-8}$.

χ_{r220} was calculated from the expression given in connexion with equation (1) and using $f_0 = 23{\cdot}76$ from *International Tables for X-ray Crystallography* (1962) and $\Delta f' = +0{\cdot}24$ from Hönl (1933) and Eisenlohr & Müller (1954). Thermal motion is included through the Debye–Waller factor $\exp(-M) = 0{\cdot}965$ (Batterman, 1964). σ and π refer to the two polarization states. Calculation of the quantities χ^{σ}_{i220} and χ^{π}_{i220} was carried out using formulae given by Wagenfeld (1966). The value

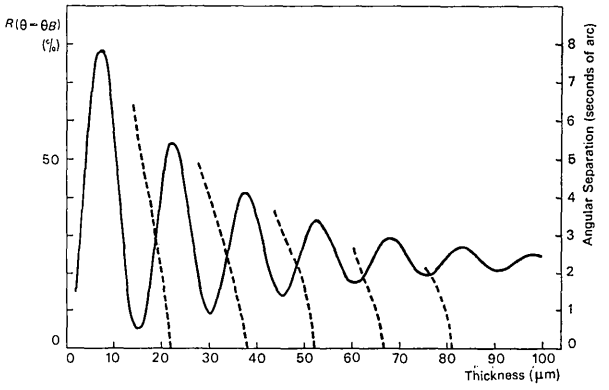

Fig. 1. The solid curve is the theoretical thickness dependence of the Laue diffracted intensity R (for $\theta = \theta_B$). The dashed curves show the theoretical thickness dependence of the angular separation of the two peaks nearest to θ_B (see text). The germanium 220 reflexion and Mo $K\alpha_1$ radiation were used for the calculations.

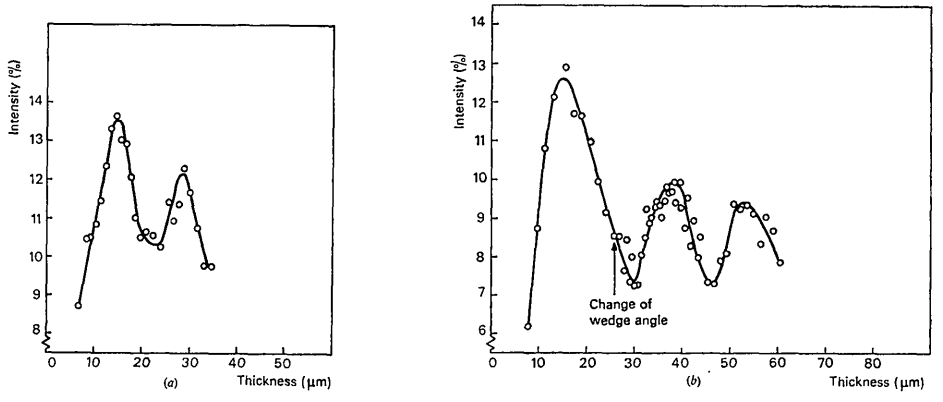

Fig. 2. Maximum values of experimental rocking curves *versus* crystal thickness for (*a*) crystal *A* and (*b*) crystal *B*. The germanium 220 reflexion and Mo $K\alpha_1$ radiation were used.

of the diffracted intensity for $\theta=\theta_B$, $R(\theta=\theta_B)$, *versus* crystal thickness is plotted in Fig. 1. This curve gives the depth periodicity of the Pendellösung interference.

For a thickness corresponding to a Pendellösung maximum the reflexion curve has a main peak at $\theta=\theta_B$ and subsidiary maxima. However, at a Pendellösung minimum, the main peak has disappeared and only subsidiary maxima remain. In Fig. 1 we have also plotted the angular separation of the two peaks nearest to θ_B starting from zero separation at a thickness corresponding to a Pendellösung maximum.

With increasing thickness the first term in equation (1) will dominate. For thicknesses greater than 83 μm the calculations still show a weak fine structure but the character is changed so that the maximum value of the reflexion curve always occurs for $\theta=\theta_B$.

Experimental technique

(*a*) *Sample preparation*

The wedge-shaped crystals were prepared from germanium single-crystal wafers by mechanical grinding. The dislocation density was less than 100 cm^{-2}. The edges were parallel to the (220) planes. To remove the damaged surface layer the samples were chemically polished in two steps with iodine etch B (about 5 min) and iodine etch A (about 2 min) (Wang, 1958). To check the crystal perfection Lang topographs were made using Mo $K\alpha_1$ radiation. Six crystals were prepared but only two crystals were found to be good enough for spectrometric measurements, mainly because of damage to the edges caused by the grinding.

(*b*) *Spectrometric arrangement*

The measurements were performed with a three-crystal X-ray spectrometer which has been described previously (Bengtsson, Brogren, Raunio & Svensson, 1964). Mo $K\alpha_1$ radiation was used. The crystal holder could be moved perpendicular to the beam by a micrometer screw with an accuracy of about 1 μm. The experiments were made using an X-ray fine-focus tube (focal dimensions 0·4 × 0·4 mm). The X-ray intensities were recorded with a scintillation detector connected to a linear amplifier and a pulse height analyzer. Measurements of the incident and transmitted intensities were performed with an aluminum foil of a thickness of about 0·8 mm to reduce the counting rate.

A double-crystal monochromator was first used in which germanium single crystals were arranged in anti-parallel positions (111, +333). The two crystals were oriented with the (111) planes parallel to the surface. The dimensions of the monochromatic beam were reduced by the collimator slits to give a width of 0·09 mm and a height of 2 mm. The measurements on the crystal denoted as crystal *A* were performed with this setting. The measurements on crystal *B* were made with a monochromator setting (111, +111). The beam dimensions in this case were 0·04 × 1 mm.

(*c*) *Wedge angle measurements*

Measurements of the wedge angle α were made with the three-crystal spectrometer. The intensity of the transmitted beam far away from the Bragg angle was measured for every thickness at which a rocking curve was recorded. The value of tg α was deduced from the slope of the plot of the logarithm of the transmission coefficient *versus* the crystal shift by the least-squares method. Thus the depth periodicity of the diffracted intensity and the wedge angle could be measured simultaneously.

Results and discussion

The experimental depth periodicity of the Pendellösung interference was obtained by plotting the maximum

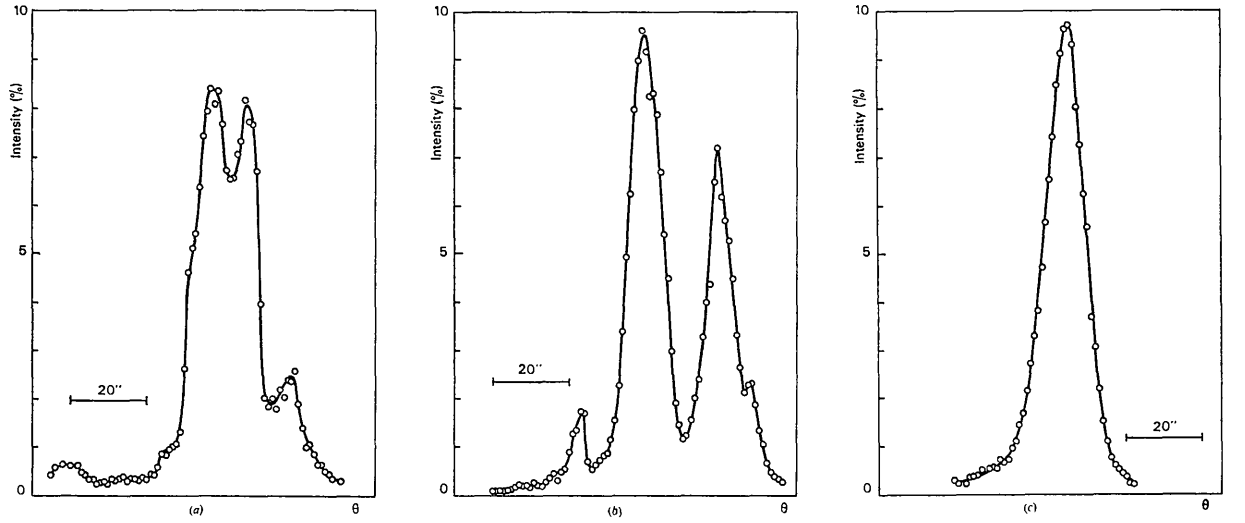

Fig. 3. Experimental rocking curves from crystal *B* in a thickness region from 30 μm to 40 μm. (*a*) to (*c*) show the influence of an increase in thickness. The germanium 220 reflexion and Mo $K\alpha_1$ radiation were used.

values of the rocking curves *versus* crystal shift (Godwood, Lefeld-Sosnowska & Zielińska–Rohozińska, 1964). By multiplying the distance between two maxima by the measured value of tg α the Pendellösung period ΔD_0 could be calculated. The scattering factor was then obtained from equation (3). In Fig. 2(*a*) and (*b*) the maximum values of the rocking curves have been plotted directly *versus* crystal thickness for crystals *A* and *B* respectively. The measured intensities are lower than the theoretical ones as seen by comparing Figs. 2 and 1. This is due to the fact that the measured rocking curve corresponds to a convolution of the theoretical reflexion curve and the monochromator smearing function.

One difficulty that arose in the experimental determination of R (for $\theta=\theta_B$) was the appearance of the fine structure in the rocking curves in thickness regions close to Pendellösung minima. Some examples of rocking curves which show the variation in the profile with thickness are given in Fig. 3. The curves were measured for crystal *B* in the thickness region from 30 μm to 40 μm. The divergence of the incident beam was as large as about 10 seconds. Because of this fact the fine structure cannot be explained by the plane wave theory used in this work. Experimentally it was impossible to determine the intensity value for $\theta=\theta_B$ when fine structure was present. The use of the maximum intensity value instead did not affect the value of the fringe spacing ΔD_0 because this was determined from the distance between two successive maxima of the depth periodicity and in these thickness regions the rocking curves have a main peak at $\theta=\theta_B$ [*cf.* Fig. 3(*c*)].

Fig. 4. Lang topograph of crystal *A*. The edge is parallel to the (220) reflecting planes. The germanium 220 reflexion and Mo $K\alpha_1$ radiation were used.

The experimental results are summarized in Table 1. Several independent measurements were performed for each crystal. The values in Table 1 are the weighted mean values. The errors quoted there are weighted mean deviations from the weighted mean. The experimental values of f_0 were corrected for the real part of the dispersion correction $\Delta f'=+0{\cdot}24$ and for the Debye–Waller factor $\exp(-M)=0{\cdot}965$. The calculation was made for the σ polarization state ($C=1$).

In order to determine the interference order of the observed fringes the theoretical and experimental positions of the Pendellösung maxima and minima are given in Table 2. The experimental data are the usual arithmetic mean values. The agreement between theory and experiment is good for crystal *B* except for the second maximum. The first maximum was not observed. It was also verified by Lang's traverse topography technique that crystal *B* did not contain any dislocations and other defects. The distance between the third and fourth maxima was used to calculate the Pendellösung period. There is a change of the wedge angle (from 3°27′ to 2°1′) between the second and the third maximum. The position of the change of the wedge angle is indicated by an arrow in Fig. 2(*b*).

There is no coincidence between the theoretical positions of maxima and minima and the measured posi-

Table 1. *Experimental and theoretical values of the Pendellösung period ΔD_0 in μm, of the scattering factor f_0, and the experimental values of the wedge angle α*

			ΔD_0		f_0	
Crystal	tg α	α	Theor.	Exp.	Theor.	Exp.
A	$0{\cdot}05356\pm0{\cdot}00036$	3°4′	15·11	$14{\cdot}80\pm0{\cdot}31$	23·76	$24{\cdot}29\pm0{\cdot}51$
B	$0{\cdot}035293\pm0{\cdot}000091$	2°1′	15·11	$15{\cdot}10\pm0{\cdot}23$	23·76	$23{\cdot}78\pm0{\cdot}35$

Table 2. *The theoretical and experimental positions of the Pendellösung minima and maxima in μm*

Interference order	Theory		Experiment	
	σ-Polarization	π-Polarization	Crystal *A*	Crystal *B*
First max.	7·6	8·1		
First min.	15·1	16·1		
Second max.	22·7	24·2	13·7	15·2
Second min.	30·2	32·2	23·8	30·0
Third max.	37·8	40·3	28·5	38·7
Third min.	45·3	48·3		46·7
Fourth max.	52·9	56·4		53·8

tions for crystal *A*. The fringes are displaced about half a period from the theoretical positions towards the thin end of the wedge. Although the fourth maximum was not observed in the spectrometer measurements it is seen on the X-ray topograph in Fig. 4. Microdensitometer measurements on the topograph indicate that the fourth maximum is also shifted in the same manner.

The presence of a defect which is possibly a surface scratch is apparent on the Lang topograph of crystal *A* (Fig. 4). The strain field from this defect could be responsible for the shift of the Pendellösung fringes. Hart (1966) has shown that the presence of elastic strain due to a thermal gradient will cause the fringes to move towards the thin end of a wedge-shaped crystal and the fringe spacing to decrease. The decrease in fringe spacing will cause an increase in the value of f_0. This effect can be seen in Table 1 where $f_{0\ \text{Crystal}\ A} > f_{0\ \text{Crystal}\ B}$.

The experimental value of f_0 for crystal *B* is about 0·1 % higher than the theoretical value. Thus, this value lies well within the experimental deviation of about ±1·5%. The experimental value of f_0 for crystal *A* is about 2·2 % higher than the theoretical value. The experimental deviation is about ±2·1 % and the agreement with theory cannot be considered satisfactory.

The present measurements of the atomic scattering factor for crystal *B* give a value which lies approximately 2·1 % below the value of 24·3 given by Batterman & Patel (1968) who also used a Pendellösung method. Inspection of Table 2 shows that the observed positions of the Pendellösung maxima and minima are closer to the theoretical values for the σ polarization than to the values for the π polarization. Although the fringe pattern is a combination of the two polarization components of the radiation, the contribution from the σ state is more important due to stronger absorption of the π state. The contribution from the π-polarized radiation would give an increase of the Pendellösung period and hence a decrease of the scattering factor. Thus, the discrepancy between our value and that of Batterman & Patel could not be explained by our neglect of the π polarization.

Summary

The atomic scattering factor has been measured for two germanium crystals by the determination of the Pendellösung period. For one crystal, which was found to be effectively perfect, the agreement between the measured value of the scattering factor and the theoretical value is remarkably good while for the other crystal the agreement is not satisfactory. The presence of a defect could be responsible for this result.

A fine structure of the rocking curves of the Laue-reflected beam has been observed though it has not been possible to make a quantitative comparison between the experimental and the calculated curves.

We would like to thank Professor G. Brogren for his interest in our work and for many helpful discussions. We are also indebted to Dr K.-G. Strid for his assistance in the computer calculations. A grant from Statens Naturvetenskapliga Forskningsråd (EP & LG) and a scholarship from the Swedish Institute (EZ-R) are gratefully acknowledged.

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