A Vibrating Perfect Crystal Assumed to be a Real One

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A generalized formula for the integrated intensity of neutrons diffracted by a vibrating perfect crystal is derived on the basis of the lamella structure with the symbols of Zachariasen’s general theory. The formula is introduced together with new experimental data for diffraction from a longitudinally vibrating quartz single crystal.

1. Introduction

The theoretical and experimental investigations of neutron diffraction by a vibrating single crystal by Buras, Giebultowicz, Minor & Rajca (1972) and Michalec, Chalupa, Sedláková, Mikula & Petržilka (1974) (in these papers the references are given to other previous papers) gave results showing a manifold enhancement of the average value of integrated reflectivity. The average enhancement was reported to be a monotonically increasing function of the vibration amplitude.

If only the integrated reflectivity is assumed, an arbitrary real crystal may constitute an intermediate stage between two ideal cases; an ideally perfect crystal (dynamical theory) and an ideally imperfect crystal (kinematical theory).

According to Zachariasen (1967), the integrated intensity of a beam diffracted by the non-vibrating crystal, \( P_N \), can be expressed in the form

\[
P_N = P_{\text{kin}} \gamma_{\text{ext}},
\]

where \( P_{\text{kin}} \) is the intensity origin by the kinematical approximation and \( \gamma_{\text{ext}} \) the extinction factor. \( P_{\text{kin}} \) is given by the relation

\[
P_{\text{kin}} = P_0 v A(\mu) Q,
\]

where \( P_0 \) is the incident intensity, \( v \) is the irradiated crystal volume, \( A(\mu) \) is the transmission factor with linear absorption coefficient \( \mu \) and \( Q \) is the reflectivity per unit volume. Furthermore in this paper we shall assume \( A(\mu) = 1 \).

We are interested in the problem of determining the general extinction factor \( \gamma_{\text{ext}}^V \) of the vibrating perfect crystal, the finding of which enables the vibrating perfect crystal to be considered as a real one, being the intermediate case mentioned above.

The factor \( \gamma_{\text{ext}}^V \) will evidently be time dependent and responsible for the time modulation of the diffracted neutrons (Michalec, Sedláková, Chalupa, Galociová & Petržilka, 1971).

2. Integrated intensity of neutrons diffracted by a vibrating perfect crystal

Let us consider a lattice plane \((hkl)\) of a single-crystal bar parallel to the \(XZ\) plane (Fig. 1). The bar is cut in this way in order to be able to vibrate it longitudinally in the \(Y\) direction with the resonance frequency \( Kf = Kc_0/2\pi = Kc_Y/2L \), where \( K \) is the mode order, \( c_Y \) is the velocity of ultrasonic waves in the \(Y\) direction axis and \( L \) is the length of the bar parallel to the \(Y\) axis.

The displacement \( U_{yk} \) of the plane \((hkl)\) at an arbitrary point, \(y\), of the bar for the \(K\)th harmonic can be described by a simple sinusoidal function in space and time

\[
U_{yk} = U_{0k} \sin \frac{K\pi}{L} y \sin K\omega t,
\]

where \( U_{0k} \) is the maximum amplitude of the \(K\)th harmonic and \( t \) is the time.

The elastic deformation \( \partial U_{yk}/\partial y \) and the movement of the lattice plane with velocity \( \partial U_{yk}/\partial t \) (which induces Doppler and aberration effects) bring about a time-dependent alteration \( \phi(y,t) = \partial(y,t) - \theta_0 \) of the Bragg angle \( \theta_0 \) (Michalec, Sedláková, Čech & Petržilka, 1971)

\[
\phi(y,t) = - \left( \frac{\partial U_{yk}}{\partial y} + \frac{1}{|v_y|} \frac{\partial U_{yk}}{\partial t} \right) \tan \theta_0,
\]

where \( |v_y| = v_0 \sin \theta_0 \) and \( v_0 \) is the velocity of the incident neutrons. The relation (4) is valid for \( \theta_0 \ll \pi/2 \).

According Michalec et al. (1974) the integrated intensity is a monotonically increasing function of \( |\phi(y,t)| \), where to a good approximation for \( y \approx L/2K \)

\[
|\phi(y,t)| \approx U_{0k} \frac{K^2 c_0^2 D \sec \theta_0}{v_0^2} \sin \frac{K\pi}{L} y \sin K\omega t,
\]

in which the contribution of elastic deformation is neglected. \( \phi(y,t) \) corresponds to the change of the angle \( \phi(y,t) \) during the time \( \delta t = D \tan \theta_0/|v_y| \), which the neutrons spend in the bar of thickness \( D \). Formula (5) holds if \( D \tan \theta_0 \ll y \) and \( \delta t \ll T \), where \( T \) is the vibration period.
If a linear dependence of the integrated intensity on the value $|\delta \phi(y, t)|$ is assumed, the quantity $(2/\pi)|\delta \phi(y, t)|$ can be interpreted as the averaged angle of total reflectivity of the vibrating perfect crystal (Mikula, Michalec, Čech, Chalupa, Sedláková & Petržílka, 1974). Thus the integrated intensity of the vibrating perfect crystal $P_{\text{kin}}^v(y, t)$, in the sample position for symmetric transmission, can be simply expressed in the form

$$P_{\text{kin}}^v(y, t) = 2P_N \frac{|\delta \phi(y, t)|}{\pi S}, \quad (6)$$

where $P_N$ is the integrated intensity corresponding to the non-vibrating case and $2\theta_0$ is the angle interval of total reflectivity for a perfect non-vibrating crystal given by

$$2\theta_0 = \frac{2\lambda N c F_{\text{hkl}}}{\pi \sin 2\theta_0}, \quad (7)$$

in which the symbols have their usual meanings.

As in the paper of Buras et al. (1972) the ratio $|\delta \phi(y, t)|/(\pi S)$ can be assumed to be the number of independently diffracting perfect ‘crystalline layers’ parallel to the $Y$ axis and perpendicular to the $X$ axis [see Fig. 1(a)] in the position of symmetric Laue transmission. Mikula et al. (1974) prefer to deal with the ratio $|\delta \phi(y, t)|/(\pi S)$, which they interpret as the number of ideally perfect ‘layers’ perpendicular to the $Y$ axis, and they use the symbols defined for symmetric Bragg reflexion. Then $2P_N$ means the integrated intensity diffracted by one ‘layer’.

It is apparent that during the time interval $\Delta t$ when $|\delta \phi(y, t)| \leq \pi S$ the relation (6) is not valid, because the value $P_{\text{kin}}^v(y, t)$ cannot be lower than $P_N$. Therefore it is necessary to make $\Delta t \leq T/2$, which can be done by increasing the amplitude $U_0$. Then the deviation of the real value of the integrated intensity from the calculated one can be neglected.

If the amplitude $U_0$ is greatly increased, the integrated intensity $P_{\text{kin}}^v(y, t)$ approaches the limiting value $P_{\text{kin}}^0$ of an ideally imperfect crystal. This case occurs during the period $T$ whenever the ratio $|\delta \phi(y, t)|/(\pi S)$ is comparable with the value of the ratio $D \tan \theta_0/\theta_{\text{ext}}$, where $\theta_{\text{ext}}$ is the extinction length given by the expression

$$\theta_{\text{ext}} = \frac{2 \sin \theta_0}{\lambda N c F_{\text{hkl}}} \cdot \quad (8)$$

For an extension of the validity of formula (6) under these conditions, it is necessary to introduce the new relation for the integrated intensity $P_{\text{kin}}^v(y, t)$ in the form

$$P_{\text{kin}}^v(y, t) = 2P_N \frac{|\delta \phi(y, t)|}{\pi S} C(y, t), \quad (9)$$

where $C(y, t)$ is the correction (White, 1950; Michalec et al., 1974)

$$C(y, t) = 1 - \exp \left( - \frac{2D \tan \theta_0}{\theta_{\text{ext}}(\delta \phi(y, t))} \right). \quad (10)$$

When the time interval $\delta t$ is comparable to $T$, the integrated intensity $P_{\text{kin}}^v(y, t)$ of the primarily diffracted neutrons can be reduced as a result of possible secondary reflexions (Mikula et al., 1974). The reduction of $P_{\text{kin}}^v(y, t)$ to the value $P^v(y) = P^v(y, t)$ can be expressed by means of a factor $y^0(D)$ [for $C(y, t) = 1$] in the form

$$P^v(y) = 2P_N \frac{|\delta \phi(y, t)|}{\pi S} y^0(D) = P_{\text{kin}}^v(y, t) y^0(D), \quad (11)$$

in which (see Appendix)

$$y^0(D) = \frac{\sin K \omega D}{2v_0 \cos \theta_0} \cdot \quad (12)$$

$$2v_0 \cos \theta_0$$

The factor $y^0(D)$ can be interpreted as the ‘secondary extinction factor’ of the vibrating perfect crystal if $\delta t \leq T$ and $C(y, t) = 1$.

In a similar manner to definition (12), we may also introduce the coefficient $y^0_l = y^0_l(t)$ [for $C(y, t) = 1$]

$$y^0_l = \frac{2|\delta \phi(y, t)|}{\pi S}, \quad (13)$$

which characterizes the change due to primary extinction.

Hence from equations (1), (2), (11), (12) and (13) we obtain the following general expression for $P^v(y)$,

$$P^v(y) = P_N y^0_1 y^0_2 y^0_3 \cdot \quad (14)$$

The factor $y^0_3$ can be taken as a general extinction factor for the vibrating perfect crystal and
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$P'(y)$ as the integrated intensity of neutrons diffracted by a 'real mosaic' crystal. It is apparent that the extreme cases of this 'real' crystal are the perfect non-vibrating one ($U_{0K}=0$) and ideally imperfect one ($U_{0K} \rightarrow +\infty$).

For the amplitude $U_{0K}$ extremely large we must write the corrected extinction quantities $y_1$, $y_2(D)$ [instead of $y_1^0$, $y_2^0(D)$] in the following way (see Appendix)

\begin{equation}
y_1 = y_1(t) = y_1^0(t)C(y,t),
\end{equation}

\begin{equation}
y_2(D) = 1 - \frac{4K}{y_1(t)T} \int_0^{\delta t/2} y_1(t) \left(1 - \frac{K\omega t}{\beta_0}\right)C(y,t)dt,
\end{equation}

where

\begin{equation}
2\beta_0 = K\omega \delta t
\end{equation}

corresponds to the change of the vibration phase during the time interval $\delta t$.

It can be seen from formulae (15) and (16) that

\begin{equation}
l\lim_{U_{0K} \rightarrow +\infty} y_1 = \frac{1}{\beta_{\text{ext}}},
\end{equation}

\begin{equation}
l\lim_{U_{0K} \rightarrow +\infty} y_2(D) = 1.
\end{equation}

For the non-vibrating crystal ($U_{0K}=0$) we put $y_1 = y_1^0 = 1$, $y_2(D) = y_2^0(D) = 1$, which cannot be seen from the theory discussed.

3. Nearly perfect single crystal

The theoretical considerations in §2 can to a good approximation also be applied to nearly perfect crystals if we introduce a quantity $s_{\text{exp}}$ (instead of $s$, but with the same meaning) as the parameter which can be determined from the experimental value of the integrated reflectivity. Similarly the parameter $\delta_{1/2}$ - the half width of the diffraction pattern of a single domain - is introduced by Buras et al. (1972). If the real crystal is assumed to be an aggregate of perfect crystal domains, $\beta_{\text{ext}}$ is the 'Zachariasen' extinction factor of the non-vibrating sample, in which the effect of both primary and secondary extinctions is included.

From the dynamical theory of diffraction by perfect crystals, $P_N$ is a linear function of $s$. If $s_{\text{exp}}$ is introduced this linearity also holds for nearly perfect crystals. Then using either (6) or (9), we arrive at the following conclusion: nearly perfect crystals of different quality behave (for sufficiently large amplitude $U_{0K}$) in the same way as real crystals of nearly the same general extinction factor $\beta_{\text{ext}}^0$, if the same experimental conditions are applied to all the samples. The validity of this conclusion has been experimentally verified and reported by Chalupa, Michalec, Čech, Mikula, Sedláková, Petřžílková & Zelenka (1975).

4. Experimental results

To illustrate some of the results following from the theory we used a bar cut from a nearly perfect quartz

![Fig. 2. The integrated intensity $P'$ of neutrons diffracted by the planes (01.0) as a function of the resonant current $i$ for $K=1$, $\lambda=1.05$ Å and for two thicknesses $D=3$ mm [curve (a)] and $D=14$ mm [curve (b)].](image)

![Fig. 3. The integrated intensity $P'$ of neutrons diffracted by the planes (02.0) as a function of the resonant current $i$ for $K=1$, $\lambda=1.05$ Å and two thicknesses $D=3$ mm [curve (a)] and $D=14$ mm [curve (b)].](image)
The dimensions of the bar were: 3 mm in the X direction, 120 mm in the Y direction and 14 mm in the Z direction. The measurements were carried out by means of the crystal spectrometer TSKN-400 (Petřížilka, Michalec, Chalupa, Sedláková, Čech, Mikula & Vávra, 1972). The width of the incident beam was 10 mm. For the detection of the average intensity values a $^{10}$BF$_3$ proportional counter was used and for the investigation of the time modulation a thin glass scintillator NE 905.

The bar was piezoelectrically excited in the longitudinal mode in the Y direction at the fundamental resonance frequency ($K=1$) $f=22.6$ kHz by means of a precise sinusoidal generator and an amplifier. The amplitude $U_{01}$ for $K=1$ was measurable with a microscope and the linear dependence of $U_{01}$ on the resonant current $i$ in the interval from 0 to 10 mA was observed. The amplitude $U_{01}=10 \mu$m corresponds to a current $i=5$ mA.

The neutron beam axis crossed the bar at $y=5L/12$. Figs. 2 and 3 show the enhancement of the integrated intensity of neutrons diffracted by (01.0) and (02.0) planes versus the resonant current $i$ flowing through the vibrating crystal. The curves (a) correspond to the thickness $D=3$ mm and curves (b) to $D=14$ mm (using simple rotation round the Y axis). In both the cases the neutrons of wavelength $\lambda=1.05 \text{ Å}$ were used. In Michalec et al. (1974), the calculated theoretical values of $P_{\text{kin}}(y,t)/P_N$ are compared with the experimental results obtained using the same sample.

Fig. 4 shows the dependence of experimental value of the extinction coefficient $y_1$ on the resonant current $i$ for $\lambda=1.54 \text{ Å}$.

The time-modulation measurements were carried out by means of multichannel analyser with a channel width of 1 $\mu$s.

The time dependence of the extinction coefficient $y_1(t)$ throughout the period $T$ is shown in the Fig. 5 for $\lambda=1.54 \text{ Å}$ and $i=0.75$ mA, using diffraction by the plane (01.0).

Fig. 6 demonstrates the time dependence of $y_1(t)$ obtained using the plane (02.0) for $\lambda=1.54 \text{ Å}$ and $i=0.35$ mA.

5. Discussion

It can readily be seen from Fig. 2 that in the current interval from 1 to 2.5 mA, the integrated intensity $P^v(y)$ [see relation (11)] is dependent only on the irradiated crystal volume $v$ when the condition $y_1(D)\approx y_2(D)=1$ is accurately fulfilled. $P^v(y)$ can be simply expressed in the form $[C(y,t)=1]$

$$P^v(y) \approx P_{\text{kin}}^v(y,t) = 2P_N U_{0K} \frac{2K^2\omega^2 \rho g \theta_0}{\pi^2 sv_t^2} v \sin \frac{K\pi}{L} y,$$

(20)

in which $P_{\text{kin}}^v = P_N/S$ is the integrated intensity per unit area of the front face of the irradiated crystal volume; $v = S \cdot D$, where $S$ is the total area of the front face. Since in both positions of the vibrating bar, to which the curves (a) and (b) correspond, the irradiated crystal volume is the same, it follows from formula (20) that the corresponding integrated intensities will also be equal. The condition given above together with the
The neutrons from all parts of the crystal which are primarily reflected in the phase range \((\pi/2 < \omega t < \pi - \beta_0)\) will leave the bar without secondary reflexion and the contribution to the average intensity \(P^r(y) = \overline{P^r(y,t)}\) will be given by

\[
\frac{1}{\pi} \int_{\pi/2}^{\pi-\beta_0} P^r_{\text{kin}}(y,t) d(\omega t),
\]

(A1)

where \(P^r_{\text{kin}}(y,t)\) is defined by formula (9).

The neutrons primarily reflected in the range \((\pi - \beta_0 < \omega t < \pi)\) contribute

\[
\frac{1}{\pi} \int_{\pi-\beta_0}^{\pi} P^r_{\text{kin}}(y,t) \frac{\pi - \omega t}{\beta_0} d(\omega t).
\]

(A2)

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APPENDIX

Derivation of the general expression for \(y_2(D) (K=1)\)

Let us suppose that the time spent by neutrons in the crystal bar is \(\delta t\). During \(\delta t\) there also occurs the change \(\delta(\partial U_{yy}/\partial t)\) of the velocity \(\partial U_{yy}/\partial t\) and the phase \(\omega t\) is changed by the value \(\omega \delta t = 2\beta_0\). With the elastic deformation neglected, neutrons primarily reflected at the instant \(t_1\) by the planes moving with velocity \((\partial U_{yy}/\partial t)_{t_1}\) can be secondarily reflected back into the incident beam at the instant \(t_2\) by planes of the same type if the condition \((\partial U_{yy}/\partial t)_{t_1} = (\partial U_{yy}/\partial t)_{t_2}\) holds (Mikula et al., 1974). Apparently the relation \(t_2 - t_1 \leq \delta t\) must also be fulfilled.

It can be seen from Fig. 7 that in the secondary diffraction process at the instant \(t_2 = 2\pi/\omega - t_1\) only the neutrons primarily reflected at the instant \(t_1\) take part when \(\omega t_1\) lies in the phase interval from \(\pi - \beta_0\) to \(\pi\). An exactly similar situation occurs in the interval from \(2\pi - \beta_0\) to \(2\pi + \beta_0\). Further we restrict ourselves only to the phase interval from \(\pi/2\) to \(3\pi/2\) and omit the subscripts.

Fig. 6. Experimentally measured time dependence of \(y_2(t)\) obtained on diffraction by a vibrating crystal bar from the plane \((02.0), D=14\ \text{mm}, I=0.35\ \text{mA and } \lambda=1.54\ \text{Å.}\)

Fig. 7. Schematic diagram of sinusoidal dependence of the plane velocity \(\partial U_{yy}/\partial t\) on the phase \(\omega t\) used for the estimation of the secondary reflexion effect.
The part \( \frac{\pi - \omega t}{\beta_0} \) determines what proportion of neutrons primarily reflected at the arbitrary instant \( t(\pi - \beta_0 < \omega t < \pi) \) will not be engaged in the secondary diffraction process since they will leave the crystal before the instant \( 2\pi/\omega - (\pi < 2\pi - \omega t < \pi + \beta_0) \) when the diffraction conditions are fulfilled for the second time. The remaining part \( 1 - \frac{\pi - \omega t}{\beta_0} \) determines the proportion which are engaged in the secondary diffraction process. Formula (10) indicates that neutrons representing the intensity,

\[
P_{\text{kin}}(y, t) \left( 1 - \frac{\pi - \omega t}{\beta_0} \right) C(y, t), \quad (A3)
\]

are reflected to the direction of the incident beam and that neutrons representing

\[
P_{\text{kin}}(y, t) \left( 1 - \frac{\pi - \omega t}{\beta_0} \right) [1 - C(y, t)] \quad (A4)
\]

are flying in the direction of the primarily reflected beam. Hence the total contribution to the average intensity value is

\[
\frac{1}{\pi} \int_{\pi}^{\pi + \beta_0} P_{\text{kin}}(y, t) \left( 1 - \frac{\pi - \omega t}{\beta_0} \right) [1 - C(y, t)] d(\omega t). \quad (A5)
\]

In the range \( \pi < \omega t < \pi + \beta_0 \), it is necessary to estimate the proportion of neutrons, engaged only once in the diffraction process, which have entered the crystal during the time \( 2(t - \pi/\omega) \) since the equivalent diffraction conditions were last fulfilled. As in expression (A2), we obtain the intensity contribution of these neutrons in the form

\[
\frac{1}{\pi} \int_{\pi}^{\pi + \beta_0} P_{\text{kin}}(y, t) \left( 1 - \frac{\pi - \omega t}{\beta_0} \right) [1 - C(y, t)] d(\omega t). \quad (A6)
\]

In the range \( \pi < \omega t < \pi + \beta_0 \) we could also find the neutrons primarily engaged in the diffraction process in the range \( \pi - \beta_0 < \omega t < \pi \), but owing to the correction (10) they would remain in the incident beam. Thus, neutrons of intensity

\[
2P_N \left[ \frac{\phi(y, t)}{\pi^2} \right] (1 - C(y, t)) \left( 1 - \frac{\omega t - \pi}{\beta_0} \right) \quad (A7)
\]

from the incident beam are secondarily engaged at the diffraction process in the range \( \pi < \omega t < \pi + \beta_0 \) and intensity

\[
2P_N \left[ \frac{\phi(y, t)}{\pi^2} \right] (1 - C(y, t)) \left( 1 - \frac{\omega t - \pi}{\beta_0} \right) C(y, t) \quad (A8)
\]

is reflected to the direction of the primarily reflected beam. Then, the intensity contribution over the range is

\[
\frac{1}{\pi} \int_{\pi}^{\pi + \beta_0} P_{\text{kin}}(y, t) \left( 1 - \frac{\omega t - \pi}{\beta_0} \right) [1 - C(y, t)] d(\omega t). \quad (A9)
\]

In the range \( \pi + \beta_0 < \omega t < 3\pi/2 \), the intensity contribution is the same as in the range \( \pi/2 < \omega t < \pi - \beta_0 \) and we obtain

\[
\frac{1}{\pi} \int_{\pi}^{\pi + \beta_0} P_{\text{kin}}(y, t) d(\omega t). \quad (A10)
\]

When the contributions (A1), (A2), (A5), (A6), (A9) and (A10) are summed and substitution is made, the intensity \( P^v(y) \) can be expressed as

\[
P^v(y) = \frac{T}{4} \int_{\phi(y, t)}^{\pi/2} P_{\text{kin}}(y, t) dt - \frac{4}{T} \int_{\phi(y, t)}^{\pi/2} P_{\text{kin}}(y, t) \left( 1 - \frac{\omega t}{\beta_0} \right) C(y, t) dt, \quad (A11)
\]

in which \( P_{\text{kin}}(y, t) \) is determined by relation (9). Using formulae (9), (10), (13) and (15), we define the factor of 'secondary extinction' \( y_s(D) \) by the expression

\[
y_s(D) = 1 - \frac{4}{y_{s}(t)} \int_{\phi(y, t)}^{\pi/2} P_{\text{kin}}(y, t) \left( 1 - \frac{\omega t}{\beta_0} \right) C(y, t) dt. \quad (A12)
\]

Putting \( C(y, t) = 1 \) we obtain

\[
y_s(D) = 1 - \frac{4}{y_{s}(t)} \int_{\phi(y, t)}^{\pi/2} y_s(t) \left( 1 - \frac{\omega t}{\beta_0} \right) dt = \sin \frac{\beta_0}{\beta_0}. \quad (A13)
\]

For estimation of the time dependence of \( P^v(y, t) \) (time modulation of the neutron beam) we present the procedure in the range \( \pi/2 < \omega t < \pi \). In the range \( \pi/2 < \omega t < \pi - \beta_0 \), \( P^v(y, t) \) is, with good agreement, equal to the function \( P_{\text{kin}}(y, t) \). In the range \( \pi - \beta_0 < \omega t < \pi \), the theory presented yields \( P^v(y, t) \) in the form

\[
P^v(y, t) = P_{\text{kin}}(y, t) \left[ 1 - \left( 1 - \frac{\omega t}{\beta_0} \right) C(y, t) \right] \quad (A14)
\]

Comparing the relations (A12) and (A14) we obtain the following expression for the factor \( y_s(D) \).

\[
y_s(D) = \frac{P_{\text{kin}}(y, t) y_s(D)}{P_{\text{kin}}(y, t)}. \quad (A15)
\]

References


