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A Neutron TOF Investigation of Dynamical Reflectivity of Elastically Deformed Crystals

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Abstract

Neutron diffraction in both an absorbing InSb crystal and a non-absorbing Si crystal deformed by a temperature gradient has been investigated experimentally. The observed variation of diffracted intensities with the magnitude and direction of the deformation gradient was compared with results of calculations based on quasi-classical formulas recently reported. A good quantitative agreement was observed in the whole range between the dynamical (perfect crystal) and the kinematical (an ideally imperfect one) limits. At the same time, the possibility of accurate intensity measurements by the TOF method in the wavelength range 0·5–3·0 Å was demonstrated.

1. Introduction

Propagation of thermal neutrons in distorted crystals can be treated in terms of quantum mechanics which enables one to employ numerous approximate methods. A quasi-classical approach to the derivation of the reflectivity was introduced in the paper of Kulda (1984), where the interbranch scattering effect taking place at stronger deformations is interpreted as a quantum transition of a neutron between two branches of a dispersion surface (in the sense of quantum states). The reflectivity formula obtained in such a way has a simple form suitable for a practical integrated reflectivity evaluation. The accompanying experimental results obtained with an elastically bent Si crystal fully confirmed the predicted saturation of the observed integrated intensities when approaching the kinematical limit. In a series of subsequent papers, the quasi-classical approach has been extended to absorbing crystals (Lukáš & Kulda, 1987) and to cases of small deformation gradient (Kulda & Lukáš, 1989). As a result, quite simple analytical formulas are now available for the rocking curves of both absorbing and nonabsorbing crystals valid for an arbitrary magnitude of the deformation gradient. The only limitation being the crystal thickness which has to exceed the extinction length.

The present paper reports results of experiments aimed at a quantitative test of the above-mentioned theoretical approach. In order to cover a range of deformation gradients and absorption coefficients as large as possible we have chosen for the experiments Si ($\mu t \simeq 0$) and InSb ($\mu t \simeq 1$–4) crystals and the TOF method permitting simultaneous work at various wavelengths and reflection orders.

2. Theoretical background

The reflectivity of a weakly deformed crystal has been derived with the help of the adiabatic approximation and reported in a recent paper by Kulda & Lukáš (1989). The resulting formula can be written in the form

$$\begin{aligned} r = {} & \tfrac{1}{2}\exp(-2V_{0i}\Delta t/\hbar) \\ & \times\Bigg(\tfrac{1}{2}\{1 - Y(0)/[1+Y^2(0)]^{1/2}\} \\ & \times\{1 + Y(A)/[1+Y^2(A)]^{1/2}\} \\ & \times\exp\left\{-\eta\int_0^A 1/[1+Y^2(a)]^{1/2}\,\mathrm{d}a\right\} \\ & +\tfrac{1}{2}\{1 + Y(0)/[1+Y^2(0)]^{1/2}\} \\ & \times\{1 - Y(A)/[1+Y^2(A)]^{1/2}\} \\ & \times\exp\left\{+\eta\int_0^A 1/[1+Y^2(a)]^{1/2}\,\mathrm{d}a\right\} \\ & -\cos\left\{\int_0^A [1+Y^2(a)\,\mathrm{d}a]^{1/2}\right\}\Big/[1+Y^2(0)]^{1/2} \\ & \times[1+Y^2(A)]^{1/2}\Bigg), \end{aligned} \tag{1}$$

where $\eta = V_{Gi}/V_{Gr}$ is the ratio of the imaginary to the real part of the matrix element V_G of the interaction potential $V(r)$. The dimensionless parameter Y is related to the local Bragg-angle deviation $\Delta\theta$ due to deformation (Kulda, 1984) by

$$Y = (2F_G d_G^2 \tan\theta_B/\pi\Omega)\Delta\theta. \tag{2}$$

In an elastically deformed crystal, the parameter Y can be considered as a function of the space and time

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variables. Similarly, the beam path in the sample is normalized to the extinction length $\Delta_G = \Omega/\lambda F_G$, so that the dimensionless parameter A may be introduced as (Kulda & Lukáš, 1989)

$$A = 2V_G t/\hbar = (s_0 + s_G)/\Delta_G, \quad (3)$$

where $s_{0,G}$ are the beam paths along the directions of wavevectors $\mathbf{K}_{0,G}$. In terms of this parameter, $Y(0)$ and $Y(A)$ in (1) mean the angular deviations at the entrance and exit surface, respectively.

For higher deformation gradient, the reflectivity has to cover the interbranch scattering effect which results in a drop in the peak reflectivity and in a saturation of the integrated reflectivity up to the kinematical limit of an ideally imperfect crystal. For the probability w of the transition between two branches of the dispersion surface we shall use a quasi-classical formula derived by Kulda (1984) which was later shown by Lukáš & Kulda (1987) to remain valid also for absorbing crystals. Therefore, the reflectivity r of (1) has to be multiplied by a factor $(1-w)$, given by

$$1 - w = \left\{1 - \exp\left[-\int_{-1}^{1} (1 - Y^2)^{1/2} |Y'|^{-1} \, dY\right]\right\}, \quad (4)$$

where $Y' = \partial Y/\partial A$.

The integrated reflecting power $\rho(\Delta Y)$ of a deformed absorbing crystal depending on the direction and magnitude of the deformation gradient can be obtained by numerical quadrature of the product of r and $(1-w)$ given by (1) and (4), respectively. For the presently used TOF method, the intensity reflected by an elastically deformed perfect single crystal fixed in a polychromatic neutron beam is given by (Buras & Gerward, 1975)

$$I = i_0(\lambda) S_0 |b|^{-1/2} N |F_G| \lambda^3 (2\pi \sin^2 \theta_B)^{-1} \rho(\Delta Y), \quad (5)$$

where $\rho(\Delta Y)$ is the integrated reflecting power on the Y scale and is dependent on the deformation degree ΔY, $i_0(\lambda)$ is the incident-beam intensity per unit wavelength range, S_0 is the cross section of the incident beam, b is the ratio of the direction cosines of the incident and reflected beams with respect to the surface normal, N is the number of crystal unit cells per unit volume and F_G is the structure factor.

3. Experimental

The experiment was performed on the DIFRAN diffractometer (Alexandrov, Chalupa, Eichhorn, Kulda, Lukáš, Machekhina, Michalec, Mikula, Sedláková & Vrána, 1988) at the tangential tube of the pulsed IBR-2 reactor at JINR, Dubna, USSR. The reactor produces neutron pulses 270 μs wide with the repetition rate of 5 pulses s^{-1}. Mean reactor power is 2 MW. The flight path of the neutrons in the vacuum tube is 28·42 m. The spectral distribution of the incident flux $i_0(\lambda)$ was calibrated by intensities scattered from a vanadium cylinder and a thin perfect Si plate.

For the experimental verification of the theoretical prediction of the reflectivity behaviour, it was necessary to use a deformation method enabling a reproducible transition through the zero-deformation point when changing the direction of the deformation gradient. For this purpose a temperature gradient appeared to be most suitable. A sketch of the deformation holder is drawn in Fig. 1. The temperature gradient was evoked by heating one end of a crystal bar, the opposite end was cooled by surrounding air only. The magnitude of the temperature gradient was estimated by a chromel–constantan thermocouple. When the temperature gradient is increased in such a way, the average temperature of the whole crystal is also increased. This effect is important when the TDS correction has to be considered, therefore we used another thermocouple to record the temperature at the centre of the crystal bar, too.

With the assumption of a constant dilatation coefficient α and constant temperature gradient Δ_t the angular deviation of lattice planes $\Delta\theta(\Delta x, \Delta y)$ between two points in the crystal may be expressed by two components. The first one is due to the longitudinal dilatation of the crystal lattice (*cf.* Fig. 2)

$$\Delta\theta_1(\Delta x) = (\Delta d/d) \tan\theta_B = \alpha\Delta_t \tan\theta_B \Delta x. \quad (6)$$

The second contribution corresponds to the rotational deviation of the lattice planes caused by a transverse dilatation of the crystal lattice

$$\Delta\theta_2(\Delta y) = \alpha\Delta_t \Delta y. \quad (7)$$

The total angular deviation $\Delta\theta$ along the beam path

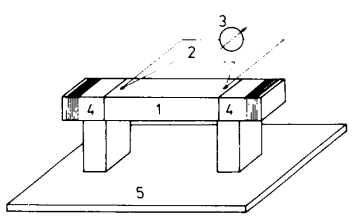

Fig. 1. A schematic view of the deformation device: 1 crystal; 2 thermocouple; 3 microvoltmeter; 4 Al blocks with heating coils; 5 base.

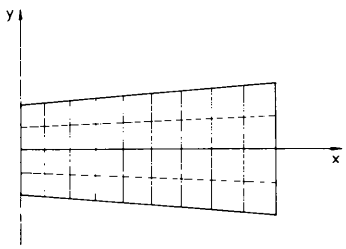

Fig. 2. A scheme of crystal-lattice deformation by a temperature gradient.

inside the crystal with thickness D can finally be expressed as

$$\Delta\theta = \Delta\theta_1 + \Delta\theta_2 = \alpha\Delta_t D(1+\tan^2\theta_B). \quad (8)$$

Then the parameter Y [according to (2)] is given by the formula

$$Y = (\pi\Omega/2F_G d_G^2)\alpha\Delta_t D(\tan\theta_B + \cot\theta_B), \quad (9)$$

which was used for the data processing.

For the experiments we employed two different samples: an absorbing InSb and a nonabsorbing Si perfect single-crystal bar having dimensions $4\times8\times40$ mm and $5\times11{\cdot}5\times53{\cdot}5$ mm, respectively. They were both cut with the longest edge parallel to the scattering vector [111]. The 10 mm wide neutron beam crossed the crystals in the central part, where the most homogeneous deformation field was expected. The TOF spectra for different values of the temperature gradient were collected in symmetric Laue geometry at two different scattering angles $\theta = 7{\cdot}89$ and $24{\cdot}04°$ for the InSb and $\theta = 29{\cdot}89°$ for the Si crystal. Commonly, up to eight reflection orders of the 111 reflection could be observed; in what follows, however, only the five lowest orders were used for evaluation, as for the higher ones the contribution from TDS and multiple reflections became too strong.

4. Data evaluation

The wide separation of the observed reflections enabled a straightforward procedure of background subtraction and integrated intensity determination. In a further step the observed $I_n(U)$ curves - here n is the index of the reflection order and U represents

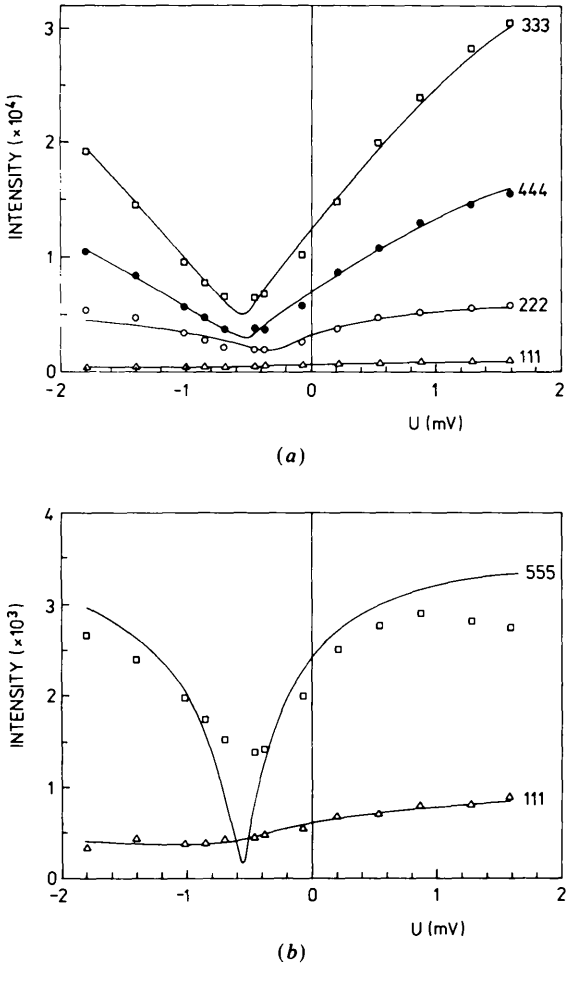

Fig. 3. (*a*), (*b*) Observed reflected intensity variation with temperature gradient for various orders of the 111 reflection for the InSb crystal at $\theta = 24{\cdot}04°$. Full lines represent the best fit according to (10).

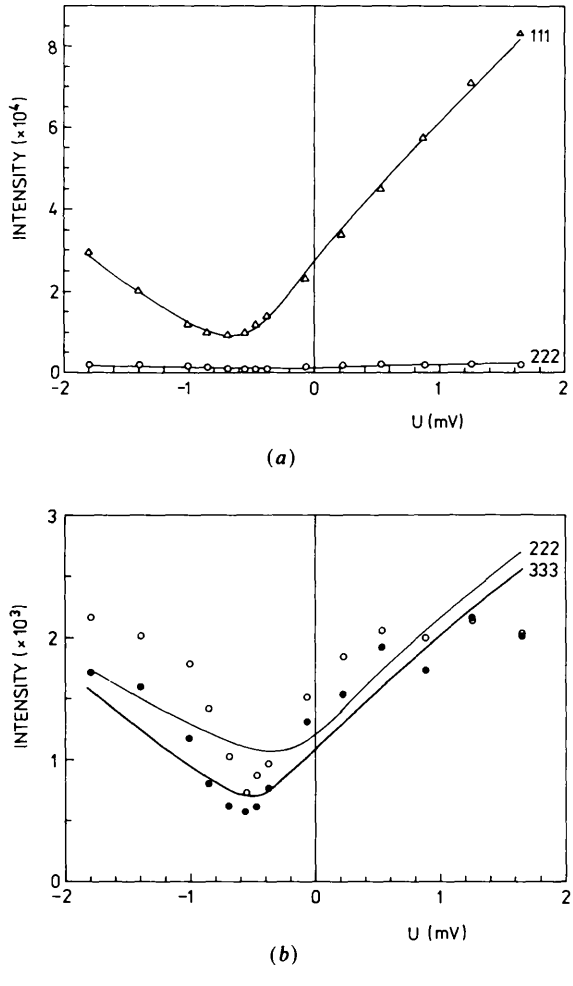

Fig. 4. (*a*), (*b*) Observed reflected intensity variation with temperature gradient for various orders of the 111 reflection for the InSb crystal at $\theta = 7{\cdot}89°$. Full lines represent the best fit according to (10).

the measured thermovoltage - were jointly fitted by a model

$$I_n = a_1 i_0(\lambda)\rho_n(a_{3n-1}U + a_{3n}) + a_{3n+1}, \quad (10)$$

with $i_0(\lambda)$ denoting the spectral distribution of the incident flux determined from independent vanadium runs. Besides the common scale factor a_1 there were three free parameters included for each reflection: a_{3n-1}, a conversion factor from the thermovoltage to the Y scale; a_{3n}, a zero shift of the Y scale representing effects of possible residual strains in the crystals; a_{3n+1}, accounting for additional intensity independent of U such as TDS. To speed up the computation procedure, the dynamical integrated reflecting power ρ_n was tabulated in advance for λ, b_c and σ_{tot} values appropriate for each reflection order and a three-point interpolation formula was used to determine its value for each particular Y. No attempt has been made to refine the Debye-Waller (DW) parameters and absorption coefficients. The coherent scattering lengths were taken from Sears (1984). The DW parameter of silicon was put at $B_{Si} = 0{\cdot}459$ Å^2. For InSb some ambiguity exists in this respect; our choice was $B_{InSb} = 0{\cdot}59$ Å^2 originating from the paper of Kjutt (1978). The absorption factors of the silicon crystal were calculated from the formulas given by Freund, Friedrich, Nistler & Scherm (1985); for InSb a sum of total cross sections of In and Sb, as compiled by Mughabhab (1984), was employed. The results of the fits are presented in Figs. 3-5.

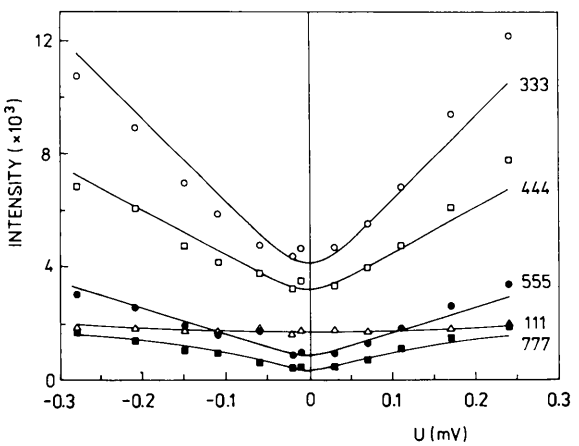

Fig. 5. Observed reflected intensity variation with temperature gradient for various orders of the 111 reflection for the Si crystal at $\theta = 29{\cdot}89°$. Full lines represent the best fit according to (10).

5. Discussion

For all the data good qualitative agreement of the observed behaviour with the theoretical prediction of $\rho_n(\Delta Y)$ is obvious. From the quantitative point of view, the most striking feature is the depth of the minimum for the low-order reflections of InSb. The reflected intensity dropped to about 30% of the value for a nonheated crystal instead of 70% predicted by theory. The only plausible explanation seems to be the presence of a quite homogeneous residual strain field in the sample which can be compensated by the applied deformation. As a result, the origin on the horizontal scale in Figs. 3 and 4 is shifted to the left by about 0·5 mV. For the silicon sample, no such effect was observed and thus the a_{3n} parameters were fixed to zero during the final fits. The a_{3n-1} parameters (conversion from U to Y) refined to values which do not significantly differ from those expected on the basis of (9). The vertical shift a_{3n+1} can most probably be atributed to the thermal difuse scattering (TDS). The corresponding TDS correction factor

$$\alpha_n = a_{3n+1}/[a_1 \pi \Delta A i_0(\lambda)] \quad (11)$$

then acquires reasonable values (ranging for Si from $\alpha_{111} = 0{\cdot}0002$ to $\alpha_{555} = 0{\cdot}058$). However, to our knowledge there does not exist a reliable algorithm for TDS calculation applicable to the present case of total scattering to make a quantitative check. Some disagreement observed at shorter wavelengths and higher reflection orders (InSb at $\theta = 7{\cdot}89°$, *cf.* Fig. 4*b*) is most probably caused by the presence of a simultaneous reflection effect which is difficult to be assessed quantitatively.

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