Acta Universitatis Carolinae. Mathematica et Physica

Václav Petržílka

Diffraction of neutrons by lattices of single crystals deformed statically or dynamically or by both ways

Acta Universitatis Carolinae. Mathematica et Physica, Vol. 17 (1976), No. 1, 3--44

Persistent URL: http://dml.cz/dmlcz/142376

1976 ACTA UNIVERSITATIS CAROLINAE - MATHEMATICA ET PHYSICA VOL. 17, NO. 1, Pag. 3-44

Diffraction of Neutrons by Lattices of Single Crystals Deformed Statically or Dynamically or by Both Ways

V. PETRŽÍLKA

Centre of Nuclear Physics, Faculty of Mathematics and Physics, Charles University, Prague*

Received 14 January 1975

In the course of investigations carried out on diffraction of neutrons by crystal lattices of quartz, germanium, silicon and kalium phosphate, which were set in acoustic or ultraacoustic vibrations piezoelectrically or electromechanically, two effects were observed: on one side the enhancement of the intensity of the diffracted slow neutron beam with respect to its intensity of neutrons diffracted by nonvibrating lattices, on the other side its modulation — at the satisfactory great amplitude of vibrations — mostly with the double frequency of vibrations, in which the crystal lattices of investigated samples were set. In the case of kalium phosphate the enhancement of the diffracted intensity of neutrons was achieved also by applying the static electric field of intensity of several kV/cm in a definite crystal direction. The aim of this paper was to compare these results mutually and to try to work out the obtained results from the uniform point of view analogous to the method, applied for the explanation of effects of diffraction of X-rays upon crystal lattices deformed statically or periodically.

В течение исследования диффракции нейтронов на кристаллических решетках кремня, германия и фосфата калия, которые были приводены в акустические или ультраакустические колебания пиэзоэлектрически или электромеханически, были обнаружены два эффекты: с одной стороны повышение интенсивности диффрактированого нейтронного пучка в отношении к интенсивности во время диффракции на не колебающихся решетках, с другой стороны ее модуляция, именно — при достаточно большой амплитуде колебаний — главным образом с двойной частотой колебаний, в которых быии возбуждены кристаллические решетки изученых образцов. В случае фосфата калия повышение диффрактированой интенсивности нейтронов было достигнуто тоже помощу статического электрического полья с интенсивностью несколько kV/cm возбужденного в данном направлении н изученном кристале. Цель этой работы — взаимное сравнение эффектов обнаруженых на упомянутых образцах и их обработка из общий точки зрения аналогичной способу, который быи использован для обяснения эффектов диффракции рентгеновских лучей на кристаллических решетках деформиронаных статически или периодически.

Při studiu difrakce neutronů na krystalových mřížkách křemene, křemíku, germania a fosforečňanu draselného, které byly uváděny do akustických nebo ultraakustických kmitů piezoelektricky nebo elektromechanicky, byly pozorovány dva jevy: jednak zvýšení intensity difraktovaného neutronového svazku vzhledem k jeho intensitě při difrakci na nekmitajících mřížkách, jednak jejı modulace, a to — při dostatečně velké amplitudě kmitů — převážně s dvojnásobnou frekvencí kmitů,

* Břehová 7, 115 19 Praha 1, Czechoslovakia

do nichž byly uváděny krystalové mříže studovaných vzorků. U fosforečňanu draselného bylo zvýšení difraktované intensity neutronů dosaženo také při vytvoření statického elektrického pole o intensitě několika kV/cm, vyvolaného v určitém krystalovém směru. Účelem této práce bylo tyto jevy pozorované na uvedených vzorcích vzájemně porovnat a pokusit se o jejich zpracování z jednotného hlediska obdobného ke způsobu, použitému pro vysvětlení jevů difrakce záření X na krystalových mřížkách deformovaných staticky nebo periodicky.

I. Introduction

In the year 1965, the experimental equipment of the team investigating in the Nuclear Physics Institute of the Czechoslovak Academy of Sciences in Řež the diffraction of slow neutrons [1], [2] reached such a stage, that it was possible to consider the successful realisation of neutron diffraction by a vibrating lattice of a single crystal in form of a bar, set in longitudinal vibrations piezoelectrically [3].

In that time, there were known experiments only which proved the intensity enhancement of the rocking curve of X-rays diffracted either in the Bragg case or in the Laue case by the lattice of a single crystal deformed either statically or dynamically (see [45] and further papers quoted on p. 331 and 332 of the vol. I of the book [92]). G. W. Fox and P. H. Carr proved in their paper [45] just in the year 1931 that the blackening of single points of a Laue diagram, produced upon a photographic film by a narrow beam of X-rays, diffracted on different lattice planes of the investigated single crystal, increased, if the Laue diagram was performed on a vibrating single crystal, i.e. on its vibrating — periodically deformed — crystal lattice. This fact shows that the intensity of X-rays diffracted by a vibrating lattice plane was increased with respect to the intensity diffracted by the same nonvibrating plane, in other words by the influence of crystal vibrations the reflectivity of its lattice planes increased too.

Later papers confirmed the observations of G. W. Fox and P. H. Carr and besides this they explained on the basis of the dynamical theory the connections between the changes of a diffracted intensity of X-rays and the deformations of the crystal lattice [46], [47], [48], [49] (see chap. 5).

With respect to the mentioned facts concerning the diffraction of X-rays, in the contribution [3] submitted to the "International Conference about Piezoelectricity" organised in September 1965 by the College of Mechanical and Textile Engineering in Liberec the attention was turned to the possibility to investigate the diffraction of slow neutrons by periodically deformed crystal lattices. In the same time it was stated that in the process of neutron diffraction by vibrating crystal lattices, e.g. of SiO_2, should — besides the neutron intensity enhancement — also its modulation occur (see [4] and [5]).

In the mean time, in the year 1966 T. F. Parkinson and M. W. Moyer from the University of Florida carried out experimentally the diffraction of neutrons by the piezoelectrically excited quartz plate vibrating with the frequency of 500 kHz [30], [31]. They proved in their first paper [30] not only the — approximately twofold —

intensity enhancement of neutrons diffracted by the vibrating quartz plate with respect to the intensity of neutrons diffracted by the nonvibrating plate, but even the modulation of the neutron beam with the frequency equal to the double multiple of the resonance frequency of the vibrating plate; they improved their results in the second paper [31].

One year later the Australian group from the University of Melbourne carried out experiments concerning also the enhancement and the modulation of the beam of neutrons diffracted by different lattice planes of a quartz plate vibrating in its fundamental frequency of 40 kHz of longitudinal vibrations [32]. The main results — the enhancement and the modulation of a beam of neutrons diffracted by a vibrating crystal lattice — have not been changed.

The Czechoslovak group of physisists carried out her preliminary experiments about the diffraction of neutrons by vibrating lattices using the double axis spectrometer SPN 100 of slow neutrons installed at VVR-S-reactor in the Nuclear Physics Institute in Řež. They applied for this purpose piezoelectrically excited plates or bars of SiO_2, produced by TESLA in Hradec Králové. Our measurements were performed with the collimated monochromatic beam of slow neutrons with the wave length $\lambda_n = 1.54$ Å and with the beam divergency of 15′ in the Bragg case on two quartz plates, the surface of which was perpendicular to the X_c-axis; one of them had the thickness of 2.84 mm and the fundamental resonance frequency of thickness (longitudinal) vibrations ~1 MHz, the other one the thickness 5.68 mm and the frequency of the same type of vibrations ~0.5 MHz. In the course of increasing of the exciting voltage, i.e. with the increasing amplitude of vibrations of the quartz plate, the intensity enhancement of the beam of neutrons diffracted by the lattice plane (11.0) of the vibrating quartz plate was observed too [6]. In the maximum of the rocking curve was in dependence upon the increasing voltage, exciting the quartz plate vibrations, even the 60-fold intensity enhancement achieved with respect to the intensity, measured in the course of diffraction upon the nonvibrating plate [11]. For two quartz bars having their lengths 70 and 90 mm and vibrating with resonance frequencies of ~30 kHz and ~40 kHz of longitudinal vibrations, the frequency modulation of the intensity of a neutron beam diffracted by one of the lattice planes of the vibrating quartz plate was by means of a time analyzer observed [7]; the modulation frequency was equal to the double multiple of the resonance frequency of the vibrating quartz bar as was expected according to the results of papers [4] and [5] (see also chap. 4).

For the excitation of different types of vibrations even for dielectric and semiconducting materials, which do not have the piezoelectric properties, an electromechanical method of excitation [18], [19], [15] was develloped, by which e.g. bars from single crystals of Si and Ge [41] were set in flexural vibrations. Analogous experiments were performed also with bars from single crystals of Fe [12], which were set to longitudinal vibrations by a transmitter of ultrasonic waves.

The process of diffracted neutrons was with a greater angular resolution and

precision more thoroughly investigated applying the tripple axis crystal spectrometer of slow neutrons TKSN 400 [22]. The obtained results of the Czechoslovak group were published in papers [16], [17], [24], [25], [41], [88], [89], [97], [98], [104], [105] and further experiments are going on.

Analogous experiments were started also using ferroelectric single crystals ADP [43], KDP [88] and KNT. There were found interesting connections between the intensity of diffracted neutron beam and the static and alternating voltage applied in the definite direction across the investigated bar from ferroelectric material.

In further chapters the summary review of the main results should be given, the observed effects should be classified, and an attempt of their explanation should be done. Real effort will be develloped to carry out corresponding analysis of the observed effects from the unit point of view, of course, as far it is possible to achieve this according to the contemporary knowledge of processes of neutron diffraction by statically or dynamically deformed lattices of single crystals.

2. Spectrometers Applied for Neutron Diffraction by Deformed Crystal Lattices

As in the year 1912 in which Max von Laue discovered the diffraction of X-rays by crystal lattices, in a similar way after the discovery of neutron in the year 1932 a new scientific discipline of neutron optics was develloped in which the first place has the diffraction of neutrons.

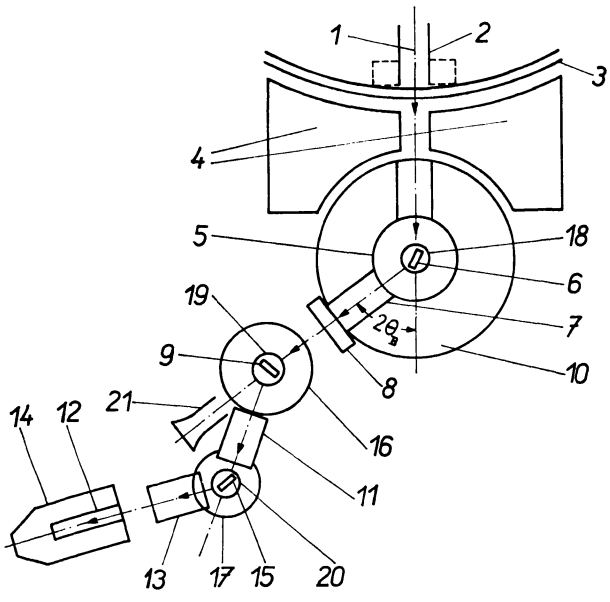


Fig. 1. Schematic ground plan of the tripple axis neutron crystal spectrometer TKSN 400

For neutron diffraction and for its application to the investigation of condensed matter, there were in all laboratories of neutron physics constructed instruments called "neutron spectrometers" [64].

In the first time it was a double axis neutron crystal spectrometer. The first Czechoslovak neutron spectrometer SPN 100 was also of this type and was built in Nuclear Physics Institute of Czechoslovak Academy of Sciences in Řež in the year 1965 [1], [2]. Using this spec-

trometer the first experiments concerning the diffraction of neutrons by vibrating crystal lattices were carried out.

B. N. Brockhouse develloped in the year 1961 the construction of a tripple axis neutron crystal spectrometer [67], [68]. Spectrometer of this type is also the Polish spectrometer TKSN 400 [69], [20], which was in the year 1971 provided by Czechoslovak Commission for Atomic Energy for the Department of Nuclear Physics of the Faculty of Mathematics and Physics of the Charles University. It is installed at the hole of the reactor VVR-S of the Nuclear Research Institute in Řež and used for the same purpose.

In this review the description of the spectrometer TKSN 400 will be given which includes also the characteristic features of the spectrometer SPN 100.

The schematic Fig. 1 shows the ground plan of the spectrometer TKSN 400. The beam (1) of slow neutrons goes out from the Soller collimator (2) of the reactor VVR-S (3) passes the hole in the supplementary shilding (4) and is monochromated in the course of diffraction by the lattice of the single crystal (6) (called monochromator) placed on the rotary table (5). Monochromatic beam entering under the angle $2\,\Theta_B$ the collimator (7) has the defined energy E_n, i.e. the defined wavelength λ_n. Across the monitor (8) it is flying to the investigated sample (9) and is absorbed by the catcher (21).

The beam of neutrons can be after the reflexion or diffraction by the investigated sample (9) and after passage through the Soller collimator (11) analysed in two ways:

1. It is possible to measure the intensity of the neutron beam by the proportional counter (12) filled with $^{10}BF_3$, before which the Soller collimator (13) is placed and which is provided by the shilding (14).

2. The energetic spetrum of the diffracted neutron beam can be analysed by means of neutron diffraction by a further single crystal (15). The rotary arm of the table (16) for the investigated sample and the rotary arm of the table (17) of the analyser are mutually independent; they are installed together on the rotary arm of the monochromator. The table of the monochromator and the table of the analysing crystal are coupled with the corresponding arms by means of special gears with the ratio 1 : 2.

The angular positions of the monochromator and analyser tables can be read off with an accuracy of $\pm 1'$ while the positions of the monochromator, of the sample arm and of the analyser can be recorded with an accuracy of $\pm 2'$ [69].

The spectrometer is controled by means of an electronic device, which permits — with respect to the possibility of using programme fed from a punch tape — even a quite large automatisation of measurements [69]. Single parts of this control unit are illustrated in the bloc schem of Fig. 2. The device is situated in a panel stage, in which the control and positioning panel for hand operation of goniometer heads is also placed; the heads can be driven electrically too.

The recording unit is a set of instruments of a trade mark "Optima", which is

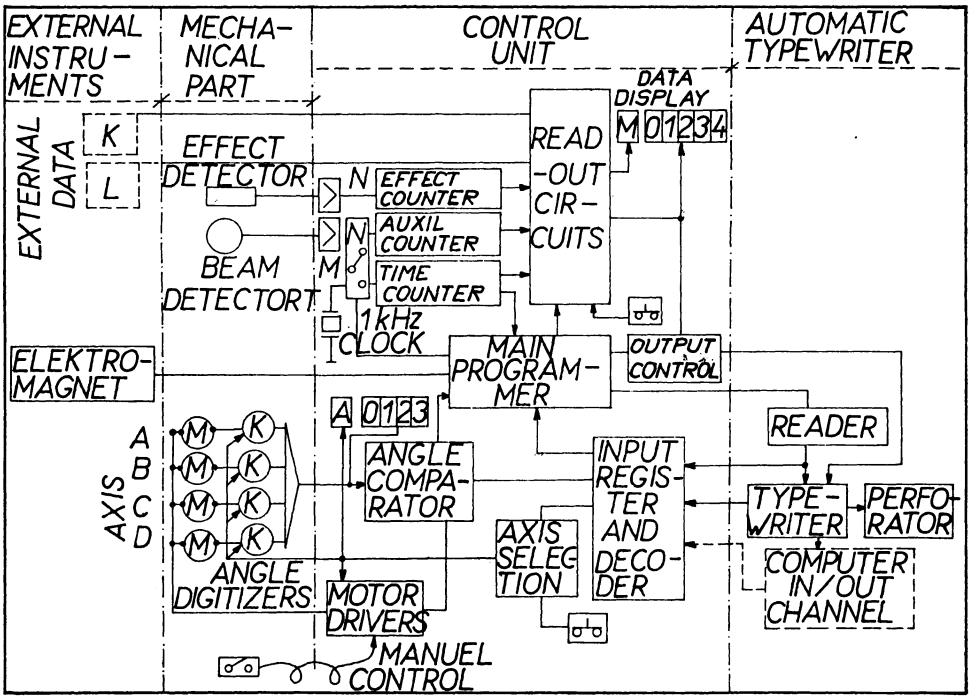


Fig. 2. Block schem of the control system of the TKSN 400 neutron spectrometer

combined from an electric typewritter, from a recorder of the punched tape and from a puncher.

Motion of single parts, e.g. of axes of the spectrometer, is possible to control from the keyboard of the typewritter, further from the 8-channel punched tape or by hand operation, i.e. by pressure-bottons upon the control panel, or by hand operation directly upon the spectrometer.

The main programmer controls the fixed cycle of the spectrometer according to the built — in programme, provides the following sequencing of operations such as reception of the input data from the keyboard of the typewritter, or from the punched type — positioning of parameters (angles) of the spectrometer — measurements from detectors — recording of measured data — respectively the repetition of measurements — conclusion of the cycle.

The construction of the electronic device gives the possibility of connection to the control computer.

3. Diffraction of Neutrons by Lattices of Single Crystals Deformed Statically

The process of diffraction by lattices deformed statically either by a direct mechanical pressure or by flexion was investigated by means of X-rays as well by means of neutrons. It is refered here about same of the obtained results from this reason, because this process has a series of characteristics which are common with the process of neutron diffraction by lattices deformed dynamically, e.g. by means of resonance vibrations either of bars or plates excited either piezoelectrically or electromechanically. Analogous deformations can be produced statically or dynamically also by the electric field — and probably even by the magnetic field — as will be shown in this chapter.

3.1 PROCESS OF NEUTRON DIFFRACTION BY LATTICE PLANES OF SINGLE CRYSTAL DEFORMED BY MECHANICAL STRESS

Several papers [71], [72], [73], [74] about these problems were carried out upon the suggestion of Professor H. Maier-Leibnitz [70] in the Institute Max von Laue — Paul Langevin in Grenoble, but also in other laboratories [75], [76], [77]. There were, however, used for this purpose also other principles, as a gradient of temperature [78] or a gradient of the interplanar lattice distance [79], [80] which are in their effects equivalent to the mechanical deformation.

T. Riste [75] investigated the diffraction properties of curved grafite single crystals ($5 \times 2.5 \times 0.1$ cm^3) with the radius $R_m = 79$ cm of curvature. The cylindrical area included the lattice plane (002). For the wave length $\lambda_n = 1.86$ Å of neutrons and for the focussing performed by decreasing the area of the reflected beam a gain of 3.8 for the neutron flux was achieved at the distance of $2R$ from the crystal.

G. Egert and H. Dachs [76] applied for neutron monochromators single crystals of germanium curved in the same way, which was used by Y. Cauchois [81] and T. Johansson [82] for diffraction of X-rays. On such neutron monochromators they carried out the measurements of the wave length spread, of the focussing properties and of the integrated reflecting power as a function of their curvature. On the basis of their results they come to the conclusion that germanium is a suitable material for monochromators.

P. Thomas [77] investigated the properties of a monochromator produced from a single crystal of copper in form of a plate ($5.8 \times 2.0 \times 0.3$ cm^3) the surface 5.8×2.0 cm^2 of which formed with the reflecting lattice plane (111) the angle $\alpha = 45°$. Its radius of curvature was $R_m = 89.5$ cm. The monochromator was develloped for the transmission method of diffracted neutrons with the wave length $\lambda_n = 1$ Å. The optimal thickness of the monochromator was $T_{opt} = 0.3$ cm. The experimental results confirmed the expected parameters of this monochromator.

M. Antonini, M. Corchia, E. Nicotera and F. Rustichelli [73] develloped for neutron monochromators curved silicon single crystals, the curvature of which they

reached by formation of a thin film of SiO_2 or Si_3N_4 on one surface of the single crystal in the form of a disk having radius of 40 mm a thickness either 0.5 mm or 1 mm. Both surfaces of this disk were parallel with the crystal lattice plane (111). The relative reflectivity of the lattice plane (111) of this monochromator, measured for the wavelength $\lambda_n = 1.2$ Å of neutrons, is increasing as a function of the decreasing radius of curvature and reaches almost the six multiple value.

Most of the neutron diffraction effects observed for curved single crystals was explained in the paper B. Klar and F. Rustichelli [74] by means of the conception of the dynamical theory of diffraction by perfect crystals, according to which the radius R_c of curvature is given by the expression

$$R_c = \frac{\Delta T}{\Delta\Theta_B}\operatorname{cotg}\Theta_B = \frac{V_c\tau}{2F_H}\frac{\Delta A}{\Delta\Theta_B}\operatorname{cotg}\Theta_B = \frac{1}{c}\frac{V_c\tau}{2F_H}\frac{\Delta y}{\Delta\Theta_B} = \frac{\pi}{c}\frac{V_c\tau^3}{4F_H^2}\operatorname{cotg}^2\Theta_B \tag{3.1}$$

in which $\Delta\Theta_B = \Theta - \Theta_B$ denotes the deviation of the angle Θ from the Bragg angle Θ_B, expressed in radians, the quantity A as a function of the thickness T of the diffracting single crystal is given by the relation

$$A = \frac{2F_H}{V_c\tau}T\,; \tag{3.2}$$

the quantity y is connected with A by the relation

$$y = y(0) + cA \tag{3.3}$$

in which c is defined by the expression

$$c = \frac{dy}{dA}\,. \tag{3.4}$$

In the expressions (3.1) and (3.2) denotes $\tau = \tau_{hkl} = 1/d_{hkl} = 1/d$ the reciprocal distance between the lattice planes, V_c the volume of the unit cell, F_H the structure factor, including the length of the coherent scattering of neutrons.

For the analysis of experimental data observed on single crystals curved in different ways, the calculation of their reflectivity according to the kinematical and dynamical theory of diffraction is necessary (see f.e. [62]).

From the results of all quoted papers [70] till [78], which were occupied by the investigation of curved monochromators, follows, that by the curvature of the single crystal and by the deformation of the lattice planes the increase of the crystal reflectivity takes place. For the silicon single crystal with thickness $T = 0.5$ cm a good agreement with the measured value $R_H^{(\Theta)}$ (exper.) $= 19.5''$ of the integrated reflectivity with the expected calculated value $R_H^{(\Theta)}$ (teor.) $= 17.2''$ for the radius $R_m = 35$ m of curvature was achieved; in this case the radius of curvature is very near to the optimal value $R_{opt} = 30{,}4$ m of the radius.

3.2 DIFFRACTION OF NEUTRONS BY LATTICE OF SINGLE CRYSTAL DEFORMED IN ELECTRIC FIELD

Even when the influence of the electric field upon the different features (properties) of crystals was studied in the series of papers [84], [85], [86], [87], the influence of the *static* electric field — at least as far as we know — was not till now investigated upon the value of the intensity of monochromatic neutron beam diffracted by the crystal lattice. We have carried out such experiment using KDP (KH_2PO_4) and obtained results described in the paper [88].

The bar of KDP ($a = 1.0$ cm, $b = 0.3$ cm, $l = 4.4$ cm) was cut from the single crystal of KDP and was oriented with respect to the crystallographic system X_c, Y_c, Z_c (see Fig. 3); its length $l = 4.4$ cm lies in the (X, Y)-plane and forms with the X_c – axis the angle 45°, while the shortest edge $b = 0.3$ cm was parallel to the axis $Z_c \equiv Z$. The static electrical voltage U_Z was applied in the direction of the $Z_c \equiv Z$ and produced the electric field with the intensity E_Z.

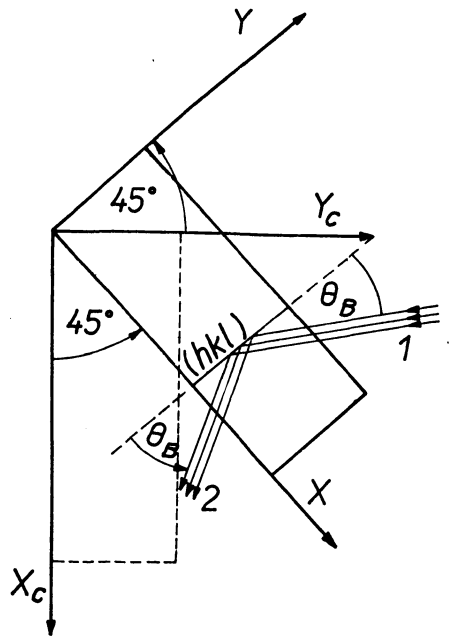


Fig. 3. Orientation of the bar of KH_2PO_4 with the laboratory system (X, Y, Z) with respect to its crystallographic system (X_c, Y_c, Z_c); 1 denotes the incident neutron beam, 2 the neutron beam diffracted by lattice planes $(h\,k\,l)$

Diffraction by neutrons by the lattice plane (220) of KDP was performed at the temperature of 18 °C, i.e. in the piezoelectric region, using the spectrometer TKSN 400 (see chap. 2). Rocking curves were measured for neutrons with $\lambda_n = 1.05$ Å in dependence upon U_Z resp. E_Z. Peak intensities $\mathcal{J}(\Theta_B)$ registered in the maximum of rocking curves for $\Theta = \Theta_B$ are plotted as a function of E_Z in Fig. 4. It was found that the intensity $\mathcal{J}(\Theta_B)$ increases with E_Z practically linearly and reaches for $E_Z = 10.000$ V/cm 1.55-times higher value with respect to the value $\mathcal{J}(\Theta_B)_{E_z=0}$ measured in the case without electric field.

The contraction or dilatation x_x in the direction of X-axis, i.e. in the direction l of the bar of KDP is directly proportional to the intensity $E_Z = U_Z/b$ of electric field and is — according to (4.17) — connected with it by the relation

$$\frac{\Delta d}{d} = x_x = \frac{\partial u(x)}{\partial x} = d_{36} E_Z = \frac{d_{36}}{b} U_Z \tag{3.5}$$

where d_{36} is the piezoelectric coefficient, b the shortest edge of the bar from KDP in the direction of the $Z_c \equiv Z$-axis (see Fig. 3).

The relative change $\Delta d/d$ of the interplanar lattice distance d causes the change of the Bragg angle Θ_B in the new angle

$$\Theta'_B = \Theta_B \pm \Delta\Theta_B . \tag{3.6}$$

The difference $\pm\Delta\Theta_B$ is given — according to (4.26) for the static case $(\partial u(x)/\partial t = 0)$ — by the relation

$$\pm\Delta\Theta_B = \pm \frac{\partial u(x,t)}{\partial x} \operatorname{tg}\Theta_B = \pm d_{36} \operatorname{tg}\Theta_B E_Z = \pm \frac{d_{36} \operatorname{tg}\Theta_B}{b} U_Z = DU_Z \tag{3.7}$$

if we use the relation (3.5) and denote the ratio $\pm d_{36}\operatorname{tg}\Theta_B/b$ by D.

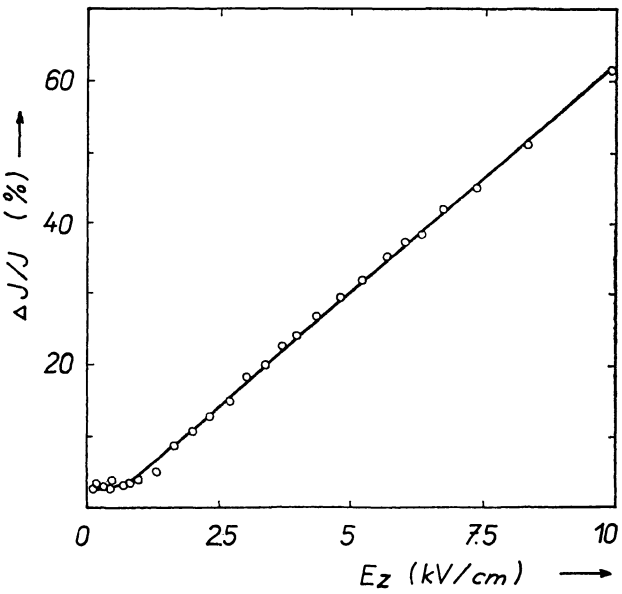


Fig. 4. Dependence of the relative variation $\Delta\mathcal{J}/\mathcal{J}$ (%) of the peak intensities of the neutron beam diffracted by the lattice plane (220) of KDP upon the intensity of the applied static electric field

For $U_Z = 0$ the rocking curve can be represented by the normal distribution of Gauss in the form as in the relation (4.23). When the voltage U_Z is applied, the shift (3.7) takes place and the intensity $\mathcal{J}$ of neutrons diffracted in the Laue case (Fig. 3) by the plane $(h,\ k = h,\ l = 0)$ is given by the expression

$$\mathcal{J}(\Theta) = \mathcal{J}(\Theta_B) \exp\left(-\frac{(\Theta - \Theta_B \pm \Delta\Theta_B)^2}{2\sigma^2}\right) \tag{3.8}$$

where σ denotes the half-width for the value $\mathcal{J}(\Theta)/\mathcal{J}(\Theta_B) = 0.607$.

If we writte the expression (3.8) in the following way

$$\frac{\mathcal{J}(\Theta)}{\mathcal{J}(\Theta_B)} = \exp\left(-\frac{(\Theta - \Theta_B)^2}{2\sigma^2}\right) . \exp\left(\pm\frac{(\Theta - \Theta_B)\Delta\Theta_B}{\sigma^2}\right) . \exp\left(-\frac{(\Delta\Theta_B)^2}{2\sigma^2}\right)$$

we obtain it like a product of three factors. (3.9)

The first of them is the Gauss' distribution for the experimental situation, when $E_Z = 0$, to which corresponds the rocking curve 1 of the Fig. 5.

The second factor of this product — according to the relation (3.7) — has its argument proportional to E_Z and causes the shift of the rocking curve 1 on the side of positive values of $\Delta\Theta_B$ in the case, that the sense of the intensity E_Z of the

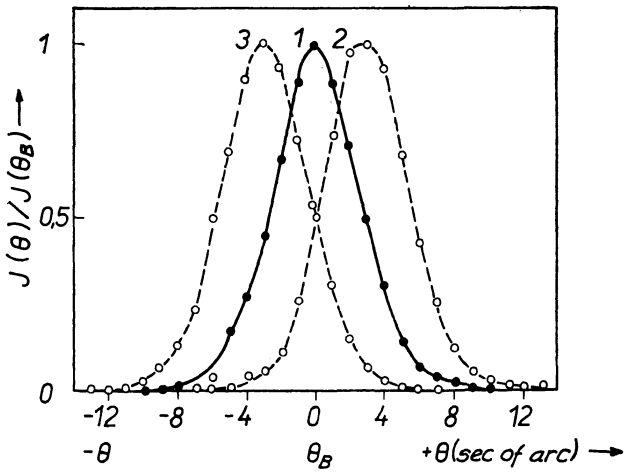


Fig. 5. Experimental shifts of the relative intensity measured on the lattice plane (440) of KDP for the applied static electric potencial difference $U_Z = 0$ (curve 1); $U_Z = -2.5$ kV (curve 2); $U_Z = +2.5$ kV (curve 3)

electric field is opposite to the sense of the crystallographic axis $Z_c \equiv Z$ (curve 2 in Fig. 5). The sense of the intensity E_Z being identical with the sense of the crystallographic Z_c-axis, the shift of the curve 1 towards negative values takes place, i.e. $\Delta\Theta_B < 0$ (curve 3 in Fig. 5).

The third factor of the product (3.9) is in its argument proportional to E_Z^2 and is, therefore, independent on the sense of the applied electric field. For small values of $\Delta\Theta_B$, this factor has the value very near to 1.

The shift of the maximum, using the double axis arrangement was not observed. The rocking curves were corresponding to the Gauss distribution and had — even when the voltage was applied — the same width equal to 18.5′ (see paper [88]).

As soon as in the spectrometer TKSN 400 the tripple axis arrangement was used, the shift of the maxima of rocking curves by the influence of the voltage U_Z was observed (Fig. 5) and the linear dependence of this shift upon the intensity E_Z of the electric field was found.

The change of the width of the rocking curves was not found, even not when double crystal (4, —4) setting was applied [89]; their width 12″ remained the same — in the limits of errors — untill the highest applied voltage $U_Z = \pm 2.5$ kV.

3.3 DIFFRACTION OF NEUTRONS BY LATTICE OF SINGLE CRYSTAL DEFORMED IN MAGNETIC FIELD

From the literature we do not know papers, which would describe effects caused by diffraction of neutrons by lattices of single crystals by the influence of a *static* magnetic field. With respect to the effects, which were described in the chap. 3.1 and 3.2, it is reasonable to suppose, that deformations caused by a magnetic field upon magnetic materials (and in a suitable arrangement even upon nonmagnetic materials) shall have the analogous influence upon the process of diffraction of neutrons as deformations caused by mechanical forces or by electric tension. It would be, therefore, very reasonable to complete the neutron diffraction in this sense and compare the results caused by the magnetic field with the results refered in chap. 3.1 and chap. 3.2.

4. Diffraction of Neutrons by Lattices of Single Crystals Deformed by Resonance Vibrations of Bars or Plates

From the existence of definite effects caused by the diffraction of X-rays by crystal lattices deformed statically or dynamically (see chap. 5), it is possible to expect, that analogous effects can arise in the course of diffraction of neutrons with respect to their wave properties; however, at the same time it can be concluded that these effects will have different characters with respect to distinct properties of X-rays and neutrons.

In this chapter we would like systematically to describe processes, which are caused by the diffraction of neutrons by periodically deformed lattices of single crystals and which manifest themselves by two basic characters: 1) by the increase of the intensity of the beam of diffracted neutrons and 2) by the modulation of this intensity in rythm of the double frequency of vibrations of crystal lattice.

Our first experiments, which were concerning the increase of the intensity of diffracted beam of neutrons with the wave length $\lambda_n = 1.54$ Å were carried out upon the lattice plane (11.0) of quartz plate with a surface perpedicular to the X_c-axis of quartz crystal and piezoelectrically excited in thickness (longitudinal) vibrations [6]; a 40-fold increase of intensity was achieved just in first experiments.

Further experiments the aim of which was the investigation of the modulation of the intensity of the diffracted neutron beam were carried out just on the lattice plane (200) of quartz bars excited piezoelectrically in longitudinal vibrations in direction of their length [7].

In further our papers there were applied to the realization of vibrations of crystal lattices like the longitudinal vibrations of bars [12], [13], [22], like the flexural ones [15], [41], or even the longitudinal — shear vibrations of plates [107]; they are described in chap. 4.1.

4.1 MODES OF VIBRATIONS OF BARS AND PLATES AND THEIR EXCITATION

4.1.1. *Fundamental Frequency and Overtones vf Longitudinal and Flexural Vibrations of Bars and Thickness Vibrations of Plates*

For diffraction of neutrons by vibrating crystal lattices the bars or plates prepared from single crystals of Si, Ge, Fe, natural and synthetic crystals of SiO_2, synthetic crystals of KH_2PO_4 (KDP), and of $KNaC_4H_4C_6 \cdot 4\,H_2O$ (KNT) were

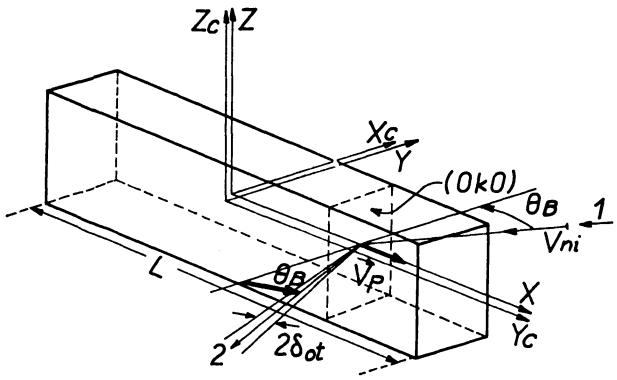


Fig. 6. Orientation of the bar of SiO_2 with its laboratory system ($X\,Y\,Z$) with respect to the crystallographic system (X_c, Y_c, Z_c) of the left-handed quartz crystal; 1 denotes the direction of the incident neutron beam, 2 the direction of the neutron beam diffracted by the lattice plane (0 k 0)

applied. According to the orientation of bars or plates in the single crystal, the elastic compliances s_{1k} given in the crystallographic system X_c, Y_c, Z_c of coordinates were transformed by means of the corresponding relations ([90], [91], [92], [93], [94], [95], [96]) in the laboratory system X, Y, Z of coordinates which is connected with the form of the bar as shown in Fig. 3 and Fig. 6.

Bars were excited in longitudinal or flexural vibrations, plates in thickness-longitudinal or thickness-shear vibrations. The equations of motion for these types of vibrations can be formulated in a unit form, as W. Voigt ([90] p. 792; see also [59]) showed. For the sake of easy survey they will be refered here separately.

For the excitation of longitudinal vibrations of bars in the direction of their length, i.e. in the direction of the X-axis all three possibilities described in chap. 4.1.2, 4.1.3 and 4.1.4 were applied. If we denote for these three cases the excitation force $S(x, t)$ we can writte the equation of motion for forced, damped, longitudinal vibrations of a bar free on both ends in the form [92], [95].

$$\varrho \frac{\partial^2 u}{\partial t^2} - \frac{1}{s'_{11}} \frac{\partial^2 u}{\partial x^2} - F \frac{\partial^3 u}{\partial x^2 \partial t} = S(x, t) = \xi(x)\, K U_g e^{j(\omega t + \varphi)} \qquad (4.1)$$

in which ϱ denotes the density, s'_{11} the elastic compliance in the direction of the X-axis (i.e. in the direction of the bar length L), F the coefficient of internal friction for the corresponding crystal, $\xi(x)$ and K are constants and $U_s e^{j(\omega t+\varphi)}$ the alternating voltage (Fig. 7).

The excitation force produces in the bar the elastic displacement $u(x, t) = u_0(x) + u_s(x, t)$ in the direction of the X-axis which is given by the sum of the

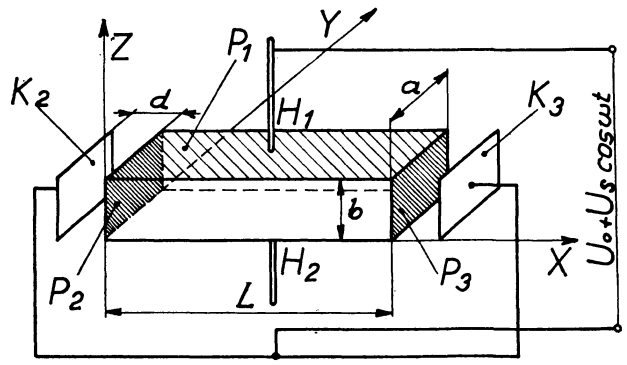


Fig. 7. Arrangement of the crystal bar for the electromechanical excitation of its longitudinal vibrations in the direction of the X-axis

static component $u_0(x)$ and of the periodical one $u_s(x, t)$; it represents the solution of the eq. (4.1) and can be written in the form

$$u(x,t) = u_0(x) + u_s(x,t) = \sum_h A_h \cos\gamma_h x + \sum_h B_h \cos\gamma_h x e^{j(\omega t+\varphi)} \qquad (4.2)$$

for $h = 1, 2, 3, 4 \ldots$ according to the fixation of the bar in its holder.

The quantities A_h and B_h can be written in general form

$$A_h = A_h(x, U), \quad B_h = B_h[x, U, (\omega - \omega_h)^2], \qquad (4.3)$$

where γ_h is connected with ω_h by the relation

$$\gamma_h^2 = \varrho s'_{11}\omega_h^2 = \pi^2 \frac{h^2}{L^2}, \qquad (4.4)$$

so that the resonance frequency f_h is given by the expression

$$f_h = \frac{h}{2L}\sqrt{\frac{1}{\varrho s'_{11}}}. \qquad (4.5)$$

By changing the frequency $f = \omega/2\pi$ of the exciting voltage $U_s e^{j(\omega t+\varphi)}$, we can for $f = f_h$ excite resonance vibrations and obtain the maximum value of the elastic displacement $u(x, t)$.

For the excitation of periodical displacements of lattice planes in the direction of the thickness a (i.e. in the direction of Y-axis) flexural vibrations in the plane XY

(Fig. 8) were applied and to their production the electromechanical excitation was used (chap. 4.1.3). According to [25], [59], [92], [95] we can writte

$$\frac{\partial^2 v}{\partial t^2} + \sigma^2 \left(1 + F \frac{\partial}{\partial t}\right) \frac{\partial^4 v}{dx^4} = S(x,t) = \xi(x)\, K U_s e^{j(\omega t+\varphi)} \tag{4.6}$$

where

$$\sigma^2 = \frac{\mathcal{J}_0}{\varrho a b s'_{11}}, \qquad \mathcal{J}_0 = \frac{1}{12} a^3 b\,. \tag{4.7}$$

Elastic displacement $v(x, t)$ is in the case of flexural vibrations of bars given by the expression

$$v(x,t) = v_0(x) + v_s(x,t) = \sum_{h=1}^{\infty} A_h \psi_h(x) + \sum_{h=1}^{\infty} B_h \psi_h(x) e^{j(\omega t+\varphi)} \tag{4.8}$$

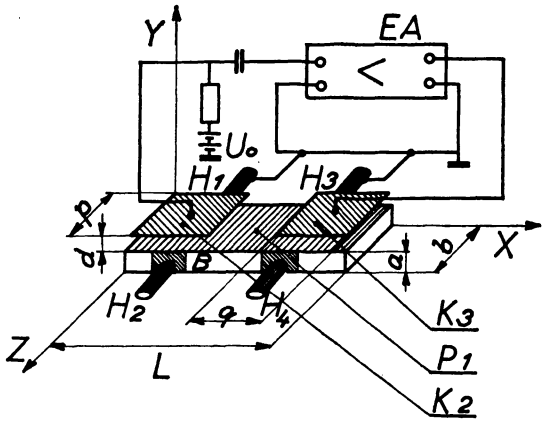


Fig. 8. Arrangement of the crystal bar for the electromechanical excitation of its flexural vibrations in the $(X\,Y)$-plane

in which the quantities A_h and B_h are characterized also by relations (4.3). The function $\psi_h(x)$ has for the on both ends free bar the form

$$\psi_h(x) = (\sinh k_h L + \sin k_h L)(\sinh k_h x + \sin k_h x) - \\ - (\cosh k_h L + \cos k_h L)(\cosh k_h x + \cos k_h x)\,. \tag{4.9}$$

The characteristic values $\varkappa_h = k_h L$ are roots of the transcendent equation

$$\cosh k_h L \,.\, \cos k_h L = 1 \tag{4.10}$$

as far as the ratio b/L (of the width to the length) of the bar is enough small. The first four roots have the values [59], [95]

$$\varkappa_1 = k_1 L = 0;\ \varkappa_2 = k_2 L = 4.7300;\ \varkappa_3 = k_3 L = 7.8532;\ \varkappa_4 = k_4 L = 10.9956\,. \tag{4.11}$$

The roots $\varkappa_n$ characterise the frequencies of the fundamental mode and of the overtones of flexural vibrations of bars and are mutually connected by the relation

$$f_h = \frac{\varkappa_h^2}{4\pi\sqrt{3}} \frac{a}{L^2} \sqrt{\frac{1}{\varrho s'_{11}}}\,. \tag{4.12}$$

In the case of quadratic plates thickness-longitudinal vibrations were excited, the displacement of which takes place in the direction of the plate thickness a. Their frequency f_h is given by the expression

$$f_h = \frac{h}{2a}\sqrt{\frac{c_{11}}{\varrho}} \tag{4.13}$$

in which c_{11} is the corresponding elastic coefficient in the direction of the plate thickness a.

In the case of circular plate the thickness — shear vibrations were excited, the frequency f_h of which is in the first approximation also given by the relation (4.13). However, the periodical course of the elastic displacement is much more complicated.

4.1.2 *Piezoelectric Excitation of Vibrations of Bars and Plates*

To bring bars of SiO_2, of KDP and of KNT into longitudinal vibrations for resonance frequencies we applied the piezoelectric excitation. We started from the known relations between the components of the elastic deformation x_{xc} till x_{yc} and components of the intensity E of the electric field produced by the voltage U, which are in the crystal system X_c, Y_c, Z_c of coordinates defined by the following equations [92] (vol. I., § 27)

$$\begin{aligned}
x_{x,c} &= d_{11}E_x + d_{21}E_y + d_{31}E_z \\
y_{y,c} &= d_{12}E_x + d_{22}E_y + d_{32}E_z \\
z_{z,c} &= d_{13}E_x + d_{23}E_y + d_{33}E_z \\
y_{z,c} &= d_{14}E_x + d_{24}E_y + d_{34}E_z \\
z_{x,c} &= d_{15}E_x + d_{25}E_y + d_{35}E_z \\
x_{y,c} &= d_{16}E_x + d_{26}E_y + d_{36}E_z
\end{aligned} \tag{4.14}$$

where d_{ik} are the piezoelectric coefficients and

$$E = \operatorname{grad} U\,. \tag{4.15}$$

Quarz has in the system X_c, Y_c, Z_c of coordinates the following piezoelectric coefficients different from zero:

$$d_{11};\quad d_{12} = -d_{11};\quad d_{14};\quad d_{25} = -d_{14}\quad\text{and}\quad d_{26} = -2d_{11}\,.$$

D.C. voltage U, connected to the electrodes or mettalized bar surfaces perpendicular to the X_c-axes produces the component E_x of the electric field and the deformation of the length unit in the direction of the Y_c-axis ($E_y = E_z = 0$)

$$\frac{\Delta L}{L} = y_{y,c} = d_{12}E_x = -d_{11}E_x = -d_{11}\frac{U}{a} = -d_{11}\operatorname{grad} U \tag{4.16}$$

according to the second of eq. (4.14) and using the relation (4.15).

If we instead of D.C. voltage U apply the A.C. voltage $U_s e^{j(\omega t+\varphi)}$ we set the quartz bar in longitudinal vibrations (see 4.1) in the resonance frequency f_h in the case that we choose the frequency $f = \omega/2\pi$ of the voltage $U_s e^{j(\omega t+\varphi)}$ equal to f_h.

For bars of KDP or KNT it is necessary to use the orientation as shown in Fig. 3. If we transform the equations (4.14) from the X_c, Y_c, Z_c system according to Cady [92] (vol. I, par. 135) into the XYZ system we obtain for the connection of the D.C. voltage U to electrodes perpendicular to the Z-axis the components x_x and y_y of deformation in the direction of the axes X and Y according to the relations

$$\frac{\partial u(x)}{\partial x} = x_x = d'_{31} E_Z = d_{36} E_Z = d_{36} \operatorname{grad} U , \tag{4.17}$$

$$\frac{\partial v(y)}{\partial y} = y_y = d'_{32} E_Z = -d_{36} E_Z = -d_{36} \operatorname{grad} U . \tag{4.18}$$

where E_Z is the intensity of the electric field produced by the voltage U in the direction of Z-axis.

Using the alternating voltage with a variable frequency we can set the bars of KDP and KNT in resonance longitudinal vibrations in one of the frequencies (4.5).

The excitation of thickness-longitudinal vibrations of quartz plates, the surface of which are perpendicular to the crystallographic X_c-axis, can be carried out in one of their resonance frequencies (4.13) by application of the alternating voltage $U_s e^{j(\omega t+\varphi)}$ with variable frequency connected to electrodes or metallized surfaces perpendicular to X_c-axis.

The excitation of the thickness — shear vibrations (chap. 4.1.1) can be done also piezoelectrically, however, it is more complicated and cannot be described by simple relations, mentioned above.

4.1.3 *Electromechanical Excitation of Vibrations of Bars and Plates*

For the purpose of electromechanical excitation of longitudinal vibrations of bars the electromechanical arrangement according to the Fig. 7 [18], [19] was applied. The bar was provided upon its end surfaces P_2 and P_3 and upon the surface P_1 by a silver layer. It was fixed in the place of the nodal line and situated between the electrodes K_2 and K_3 to which the D.C. voltage U_0 as well as the A.C. voltage $U_s e^{j(\omega t+\varphi)}$ is connected. This voltage produces between the metallized end P_2 of the bar B and the exciting electrode K_2 the electromechanical force $S_1(x,t)$ given by the relation

$$S_1(x,t) = \frac{\xi(x)\,\varepsilon}{2d^2\varrho}\left[\left(U_0^2 + \frac{U_s^2}{2}\right) + 2U_0 U_s e^{j(\omega t+\varphi)}\right] . \tag{4.19}$$

This force produces according to the eq. (4.1) of motion a periodic elastic displacement (4.2) of longitudinal vibrations of the bar in the direction of its length. We have used them for the determination of elastic constants [23].

The two electrodes arrangement can be in principle used for the excitation of flexural vibrations too. Fig. 8 shows, however, the arrangement, which we have most frequently used, and in which the bar excited, in its fundamental frequency with two nodal lines was used as a coupling element between the output and input

of the amplifier EA. The electromechanical force $S_f(x,t)$ for excitation of flexural vibrations produced by the electric field between the electrode K_3 and the metall layer P_1 has the half value of the force $S_1(x,t)$ given by (4.19). This force is exciting the elastic displacement (4.8) in the direction of the Y-axis.

4.1.4 *Excitation of Vibrations of Bars by a Power Transfer*

The excitation of longitudinal vibrations of a single crystal bar can be done by coupling the bar with a magnetostrictive resonator [12] supplied by a power generator. The relative strain amplitude of the resonator vibrations was measured by means of a detector made from an electrostrictive ceramic of $BaTiO_3$ type.

4.2 INCREASE OF THE INTENSITY OF THE NEUTRON BEAM DIFFRACTED BY A VIBRATING CRYSTAL LATTICE

4.2.1 *Experimental facts*

As soon as the existence of the increase and of the modulation of the intensity of the neutron beam diffracted by vibrating crystal lattices — expected in the abstract [3] and in the papers [4], [5] — was experimentally proved and described in papers [31], [32], [6], [7], [8], [11], [12], thorough investigation of these effects was started. The experiments of neutron diffraction were carried out by means of the double axis crystal neutron spectrometer SPN 100 [1], [2] and later also by means of the triple axis crystal neutron spectrometer TKSN 400 [20], [69] (see chap. 2) on single crystals of SiO_2, Si, Ge, Fe and later also on KDP and KNT [13], [15], [16], [17], [21], [22], [24], [41], [42].

If longitudinal vibrations and piezoelectric excitation were used, the samples of SiO_2 single crystal were bar shaped and oriented for the purpose of neutron diffraction — as shown in Fig. 6. The X_c-axis is the electric axis in the direction of which the voltage from a finely tunable oscillator (RFT typ PDG-1) of sufficient stability and amplified by a selective amplifier was applied. In the paper [13], [22] and [24] measurements of neutron diffraction on quartz bars excited in longitudinal vibrations in the direction of their length were performed in the Nuclear Physics Institute of the Czechoslovak Academy of Sciences by means of the above mentioned spectrometers. A monochromatic neutron beam with the wavelengths of $\lambda_1 = 1.05$ Å or $\lambda_2 = 1.54$ Å having the angular divergency from 2′ till 15′, at the VVR-S reactor of the Nuclear Research Institute in Řež was used. The orientation of all quartz samples was chosen in such a way that diffraction from lattice planes of the (0 k 0) type (the YZ-plane, Fig. 6) took place in the symmetric Laue case.

The experiments offered in all cases a similar picture, as regards the intensities of neutrons diffracted by vibrating crystal lattice (see Fig. 9). The rocking curve taken off on the vibrating single crystal bar has in corresponding points of the curve higher values of diffracted intensity than the rocking curve of the nonvibrating single

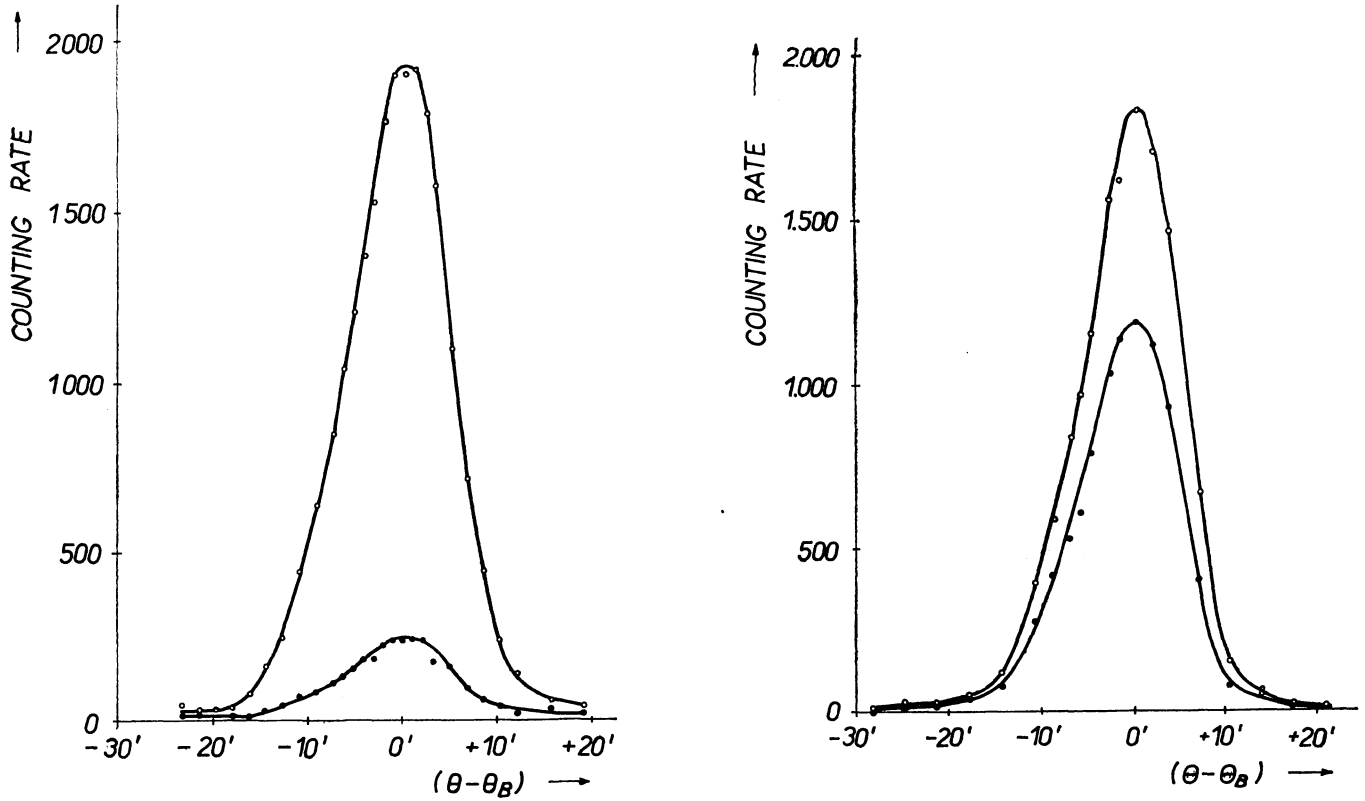


Fig. 9. Rocking curves of neutrons ($\lambda_n = 1.54$ Å) measured in the Laue case of symmetric transmission upon the lattice plane (200) of vibrating bars (curves with open circles): (*a*) from the natural quartz, (*b*) from the synthetic quartz; by closed circles are illustrated rocking corves for nonvibrating bars

crystal bar. In the double axis arrangement, their width was in both cases the same, which fact is caused by the instrumental feature of the spectrometer, i.e. by the great angle of divergency of its neutron beam (0° 2′) in the double axis arrangement.

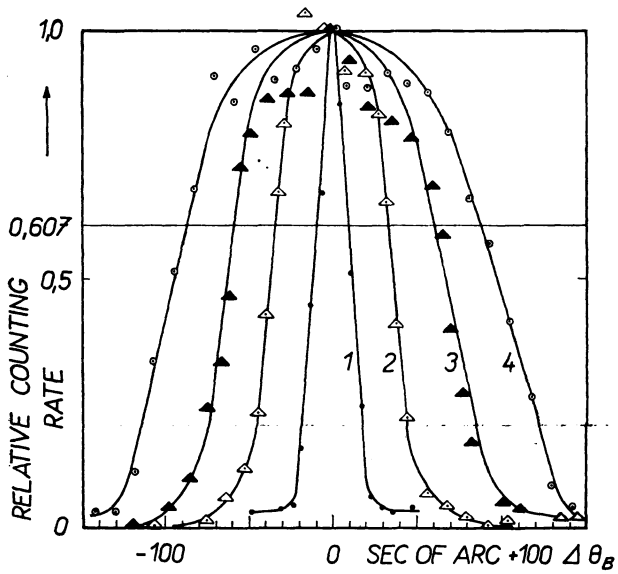


Fig. 10. Increase of rocking curve widths with growing vibration amplitude in double (1, −1) crystal setting: 1 nonvibrating Si bar; 2, 3 and 4 Si bar vibrating with the amplitude of 0.75 μm, 1.5 μm, and 3 μm at its end

However, by using the triple axis arrangement of the spectrometer TKSN 400 the increase of the rocking curve width 2σ on the amplitude u_0 of the periodic changes of the elastic displacement

$$u_h = u_0 \sin\left(\frac{h\pi}{L}\right) x \sin \omega t \tag{4.22}$$

was proved and is shown in Fig. 10 [22].

In this case all rocking curves can be represented by the normal distribution of Gauss in the form [22],

$$\mathcal{J}_{u_0}(\Theta) = \mathcal{J}_{u_0}(\Theta_B) \exp\left(-\frac{(\Theta - \Theta_B)^2}{2\sigma_{u_0}^2}\right) \tag{4.23}$$

where $\mathcal{J}_{u_0}(\Theta_B)$ is the neutron intensity in the maximum of the rocking curve, $(\Theta - \Theta_B)$ is the deviation from the Bragg angle Θ_B and σ_{u_0} is connected with the full width H_{u_0} of the rocking curve for a crystal vibrating with an amplitude u_0 by the relation.

$$H_{u_0} = 2\sigma_{u_0}\sqrt{2 \ln 2}\ . \tag{4.24}$$

For σ_u. it is possible to writte the approximate relation

$$\sigma_{u_0} \approx \sqrt{\overline{(\Delta\Theta_B)^2}} \approx u_0 \tag{4.25}$$

because $\Delta\Theta_B \sim u_0$ according to [44]. This dependence was proved experimentally [22].

Using the quartz bar of length $L = 120$ mm, the ratio of peak intensities of diffracted neutrons $\overline{R} = \overline{\mathcal{J}}_V/\overline{\mathcal{J}}_S = \overline{R}(i)$ as a function of the excitation current i [24], where $\overline{\mathcal{J}}_V$ and $\overline{\mathcal{J}}_S$ are the intensities of neutrons for a vibrating and a nonvibrating

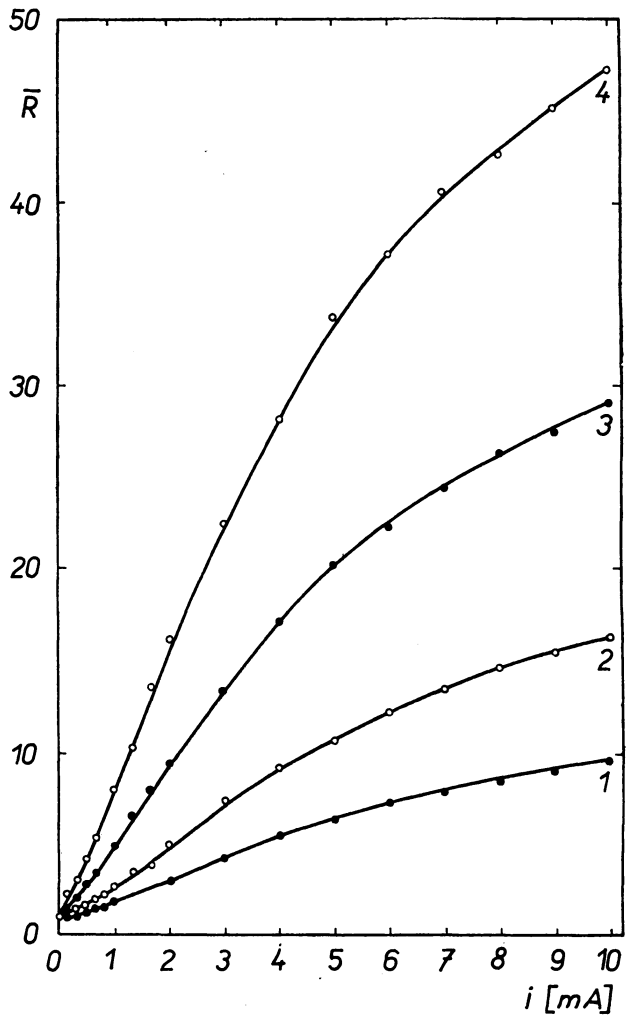


Fig. 11. $\overline{R}$ versus excitation current i for $\lambda_1 = 1.05$ Å, $T_X = 3$ mm (1); $T_Z = 14$ mm (3), and for $\lambda_2 = 1.54$ Å, $T_X = 3$ mm (2); $T_Z = 14$ mm (4), for the quartz bar with the length $L = 120$ mm

single crystal respectively, was measured for two neutron wavelengths and for two crystal thicknesses. The obtained experimental data for the dependance of $\overline{R} = \overline{R}(i)$ are shown in Fig. 11 [24].

The bars from single crystals of SiO_2 and also from Si and Ge can be excited in flexural vibrations too. It means that the elastic strains in the XYZ-system of

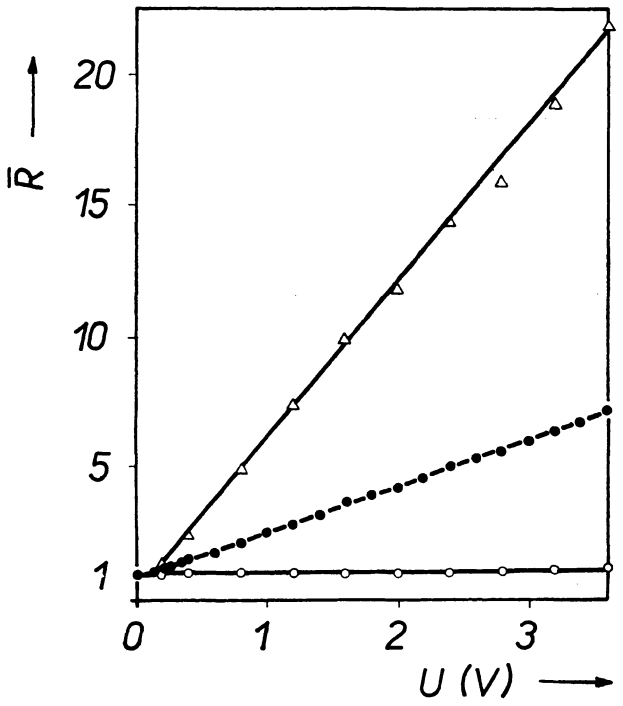


Fig. 12. Dependence of $\overline{R} = \overline{R}(U)$ for the single crystal of Si; $\lambda_n = 1.05$ Å; ○ (111) transmission; ● (224) reflexion; △ (112) reflexion; U is the voltage drop R_{Si}, where i is the excitation current flowing through the sample

the bar (see Fig. 8 occur in the XY-plane and the bending of the bar takes place in the direction of the Y-axis. For this purpose the corresponding bar is used in the arrangement shown in Fig. 8 and electromechanically excited [15], [18], [25], [41].

In Fig. 12 the experimental values of $\overline{R}$ obtained for neutrons with the wave length $\lambda_n = 1.05$ Å and for lattice planes (111), $(11\overline{2})$ and $(22\overline{4})$ of a bar shaped single crystal of Si are given. The bar having dimensions $5 \times 12 \times 105$ mm^3 was oriented with its length parallel to the crystal direction [111]. It was excited in flexural vibrations with the fundamental resonance frequency $f = 4300$ Hz. The maximum value of the displacement in the bar centre ($x = 0$) was $u_{max} = 4{,}5$ μm for the control voltage $U = 3$ V.

For the lattice plane (111) one has $\overline{R} \approx 1$ which fact corresponds to the con-

dition $(\vec{\tau} \cdot \vec{u}) = 0$. In such a case the vibrations of the lattice plane cannot have any influence on the intensity of diffracted neutrons. The largest values of $\bar{R}$ were observed for the forbidden neutron reflexion on the plane $(11\bar{2})$. The function $\bar{R} = \bar{R}(U)$ for the planes $(11\bar{2})$ and $(22\bar{4})$ differ very little from the linear dependence (see chap. 4.2.2).

Similar results were obtained for bar shaped single crystals of Ge and SiO_2 (see chapter 4.2.2).

It is still interesting to mention that in the year 1974 three papers [43], [66], [88] were published which describe experiments on ferroelectrics ADP and KDP, similar to that, which were performed on Si, Ge and SiO_2.

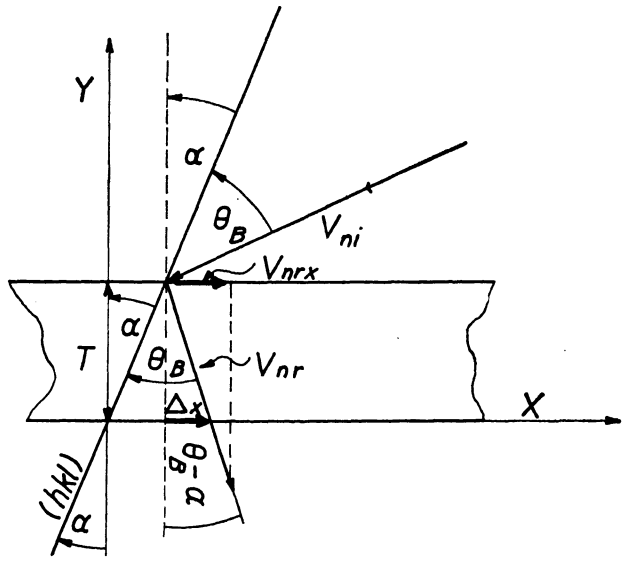


Fig. 13. Assymmetric Laue case in which the lattice plane (hkl) reflecting neutrons forms with the plane (YZ) the angle $(+\alpha)$

Professor T. F. Parkinsson and his collaborators have investigated the enhancement of intensities of rocking curves taken off on vibrating resonators cut from ADP [43], [66] in the form of plates excited piezoelectrically in thickness-shear modes coupled with flexure modes. They writte:

"With the ADP crystal stationary, rocking curves were run for several $(h\,k\,l)$ planes to determine precise values for Θ_B. The rocking curves were then repeated with the crystal vibrating ...

The enhancement ratio was found by integrating the area under each curve:

$$R = \left(\int_{\Theta_1}^{\Theta_2} \Phi_v \, d\Theta\right)\left(\int_{\Theta_1}^{\Theta_2} \Phi_s \, d\Theta\right)^{-1}$$

where Φ_V and Φ_S are respectively, the intensities with the crystal vibrating and stationary".

Two typical results of the enhancement measurements for two $(h\,k\,l)$ planes are given in Fig. 5 of the paper [43], [66] and the maximum 2.4-times enhancement was found.

Similar results on KDP were published in the "Short Note" [88], in which in the maximum of rocking curves on the vibrating and stationary bar of KDP the

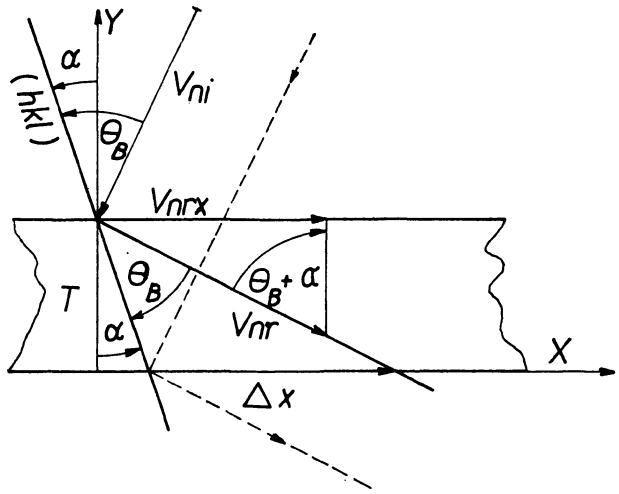


Fig. 14. Assymetric Laue case in which the lattice plane (hkl) reflecting neutrons forms with the plane (YZ) the angle $(-\alpha)$

1.9-times enhancement of the intensity of diffracted neutrons was achieved. If to the alternating voltage the static field of $E_Z = 10$ kV/cm was applied, the enhancement of the intensity of diffracted neutrons reached the value 2.4.

4.2.2 *An attempt of an approximate explanation of observed results*

The piezoelectric excitation of longitudinal vibrations of bars with the frequency (4.5) causes the periodic displacement (see chapter 4.1.1) of crystal planes and therefore also of lattice planes in the direction of the bar length. The displacement u in the defined point (x, y, z) of the bar can be described by the expression (4.22).

First we shall suppose that the neutrons are incident at the Bragg angle Θ_B upon the nonvibrating single crystal in the assymmetric Laue case shown in Fig. 13 and Fig. 14; later for the sake of simplicity we shall consider the symmetric Laue orientation (Fig. 6).

As soon as the single crystal bar will be set in longitudinal vibrations — than due to the strain $(\partial u/\partial x)$ and to the velocity $(\partial u/\partial t)$ of lattice planes — there arises

a deviation $\Delta\Theta_B = \Theta - \Theta_B$ from the Bragg angle Θ_B [17], [44] which can be expressed in the form

$$\Delta\Theta_B = -\operatorname{tg}\Theta_B \frac{\partial u(x,t)}{\partial x} + \frac{\cos\alpha}{v_{nr}\cos\Theta_B}\frac{\partial u(x,t)}{\partial t}\,, \tag{4.26}$$

in which v_{nr} denotes the velocity of the diffracted neutrons. From the physical point of view the deviation $\Delta\Theta_B$ is caused by the aberration and Doppler effects, due to the mouvement of lattice planes, and by their deformation.

The change $\delta\Delta\Theta_B$ during the time of flight of neutrons across the vibrating sample with thickness T is given for the case shown in Fig. 13 by the formula

$$\delta\Delta\Theta_B = \frac{T}{\cos\Theta_B\cos(\Theta_B-\alpha)}\left\{-\sin\Theta_B\sin(\Theta_B-\alpha)\frac{\partial^2 u}{\partial x^2} + \right.$$
$$\left. + \frac{\cos\alpha}{v_{nr}^2}\frac{\partial^2 u}{\partial t^2} - \frac{\sin\alpha\cos(\Theta_B-\alpha)}{v_{nr}}\frac{\partial^2 u}{\partial x\partial t}\right\} \tag{4.27}$$

and for the case shown in Fig. 14 by the expression

$$\delta\Delta\Theta_B = \frac{T}{\cos\Theta_B\cos(\Theta_B-\alpha)}\left\{-\sin\Theta_B\sin(\Theta_B+\alpha)\frac{\partial^2 u}{\partial x^2} + \right.$$
$$\left. + \frac{\cos\alpha}{v_{nr}^2}\frac{\partial^2 u}{\partial t^2} - \frac{\sin\alpha\cos(\Theta_B+\alpha)}{v_{nr}}\frac{\partial^2 u}{\partial x\partial t}\right\} \tag{4.28}$$

Inserting in (4.27) or (4.28) the angle $\alpha = 0$, we obtain the expression

$$\delta\Delta\Theta_B = \frac{T}{\cos^2\Theta_B}\left(\frac{1}{v_{nr}^2}\frac{\partial^2 u}{\partial t^2} - \sin^2\Theta_B\frac{\partial^2 u}{\partial x^2}\right) \tag{4.29}$$

for the symmetric Laue case illustrated in Fig. 6. Using in (4.29) the relation (4.22) we obtain it in the form

$$\delta\Delta\Theta_B = \frac{u_0\omega^2 T}{v_{px}^2\cos^2\Theta_B}\left(\frac{v_{px}^2}{v_{nr}^2} - \sin^2\Theta_B\right)\sin\frac{\pi x}{L}\sin\omega t \tag{4.30}$$

in which v_{px} denotes the velocity of propagation of ultrasonic vibrations along the direction of X-axis.

For calculating the integrated intensity of diffracted neutrons we shall proceed in a similar way as for the diffraction of X-rays [49] or for the diffraction of neutrons on statically bent single crystals [73].

First we shall suppose that for a nonvibrating single crystal perfect neutron reflexion occurs over a range H and that the area under the rocking curve is according to the Darwin [108] treatment equal to $H = 2.66s$, where s is given by the formula

$$s = \frac{N_c\lambda^2 F}{\pi\sin 2\Theta_B} \tag{4.31}$$

in which N_c is the number of unit cells per unit volume and F the structure factor of neutrons.

The second assumption is that the extinction coefficient ϵ, which governs the depth of penetration of the neutron beam before it is reflected, has the constant value ϵ_{ave} over the whole range w and is zero elsewehre. The value is the linear average of Darwin's ϵ over the angular range w, and is given for neutrons by

$$\epsilon_{ave} = \frac{3}{8}\pi N_c \lambda F \tag{4.32}$$

similar to [49].

Using these assumptions we can state how the integrated intensity diffracted by a vibrating single crystal should depend on the displacement u. For this purpose we shall divide the vibrating single crystal in n layers, each from which is diffracting neutrons independently as a thick perfect crystal. In the case that the time of flight of neutrons across the crystal is much lower than the period of vibrations of a single crystal bar, i.e. for $T/v \cos \Theta_B \ll 2\pi/\omega$ it is possible in each time to writte

$$n(t) = \frac{|\delta\Delta\Theta_B|}{w} = p|\sin \omega t| \tag{4.33}$$

$$p = \frac{u_0\omega^2 T}{w v_{px}^2} \frac{1}{\cos^2 \Theta_B} \left(\frac{v_{px}^2}{v_{nr}^2} - \sin^2 \Theta_B \right) \sin \frac{\pi x}{L} . \tag{4.34}$$

In the case of negligible absorption of neutrons in the sample we shall obtain for the integrated intensity $\mathcal{J}_{VO}$ of neutrons diffracted by a vibrating crystal the relation:

$$\mathcal{J}_{VO} = \mathcal{J}_S n(t) = \mathcal{J}_S \frac{|\delta\Delta\Theta_B|}{w} \tag{4.35}$$

where $\mathcal{J}_S$ is the integrated intensity of neutrons diffracted by the lattice planes of a nonvibrating thick single crystal given by the Darwin's formula:

$$\mathcal{J}_S = \frac{8}{3} \frac{N_c \lambda^2 F}{\pi \sin 2\Theta_B} \mathcal{J}_0 \tag{4.36}$$

in which $\mathcal{J}_0 = \mathcal{J}_0(\lambda)$ denotes the intensity of the incident neutron beam.

It is apparent that as amplitude u_0 of vibrations is increased, the layers described above become thinner, whereas the distance required for almost complete reflection remains fixed. Applying the corresponding correction C (see [49]) of the intensity $\mathcal{J}_{VO}$ of diffracted neutrons for the "thin crystal" effect, we obtain:

$$\mathcal{J}_V = \mathcal{J}_{VO} C = \mathcal{J}_S \frac{|\delta\Delta\Theta_B|}{w} [1 - \exp(-\epsilon_{ave} w T / |\delta\Delta\Theta_B| \cos \Theta_B)] . \tag{4.37}$$

The mean value $\mathcal{J}_V$ of the intensity of neutrons diffracted on the vibrating single crystal is given by the relation:

$$\overline{\mathcal{J}_V} = \mathcal{J}_S p \overline{|\sin \omega t| \left[1 - \exp\left(- \frac{1}{|\sin \omega t|} \frac{\epsilon_{ave} T}{p \cos \Theta_B} \right) \right]} \tag{4.38}$$

which is impossible to express by simple functions.

The ratio $R = \mathcal{J}_V/\mathcal{J}_S$ of the intensity of neutrons diffracted on a vibrating ($\mathcal{J}_V$) and on the nonvibrating ($\mathcal{J}_S$) single crystal can be obtained directly from the relation (4.37). In the case when the correction for the "thin crystal" effect does not take place, i.e. $C \approx 1$, the mean value is given by the simple expression

$$\overline{R} = \overline{\mathcal{J}}_V / \overline{\mathcal{J}}_S = \frac{2}{\pi} p \tag{4.39}$$

which is valid for $n \gg 1$.

For testing the theoretical suppositions about the influence of ultrasonic vibrations on the process of neutron diffraction the measurements of the value $\overline{R} = \overline{R}(i)$ and of the time modulation of the neutron beam were carried out.

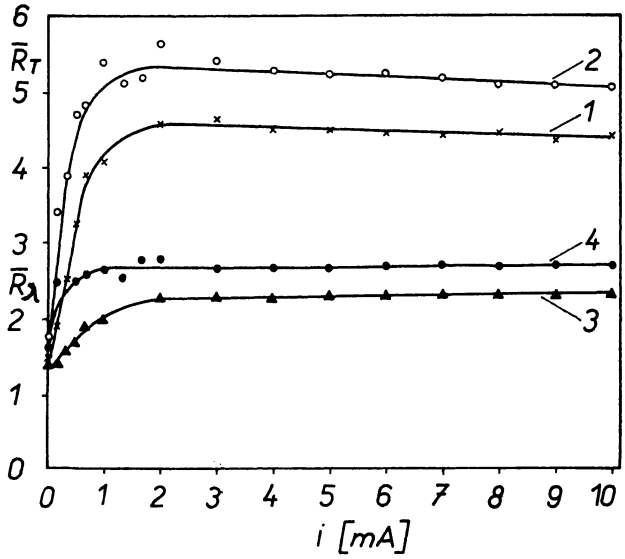


Fig. 15. $\overline{R}_T$, $\overline{R}_\lambda$ versus excitation current i for $\lambda_1 = 1.05$ Å (1), $\lambda_2 = 1.54$ Å (2), $T_X = 3$ mm (3) and $T_Z = 14$ mm (4) of a quartz bar with the length $L = 120$ mm

The experimental data of Fig. 11 show the nearly linear dependence of $\overline{R} = \overline{R}(i)$ versus the current i on the beginning of the curves 1 to 4. However, it can be seen clearly from Fig. 11 that there exists the nonlinearity for higher currents of i, which is most probably caused by two reasons: the first one is connected with the nonlinearity of $u_0 = u_0(i)$ for high amplitudes of the displacement, the second one is due to the "thin crystal" effect.

For testing the dependence of values p from relation (4.34) on the bar thickness T, the relation $\overline{R}_T = \overline{R}(i, T)$ for two wavelengths of diffracted neutrons was investigated. In Fig. 15 the curves 1 and 2 represent the experimental dependence on i of the relation

$$\overline{R}_T = (S_X/S_Z) \cdot \overline{\mathcal{J}}_V(i, T_Z)/\overline{\mathcal{J}}_V(i, T_X) \tag{4.10}$$

where (S_X/S_Z) is the ration of irradiated areas, $\bar{\mathcal{J}}_V(i, T)$ is the mean value of the peak intensity of diffracted neutrons. For the values $p \gg 1$ we shall get from the relation (4.38) that $\bar{R}_T = T_Z/T_X$. In the case of our synthetic single crystal of SiO_2 the calculated value $\bar{R}_T = T_Z/T_X = 4{,}7$ agrees quite well with the experimental value for $i > 2$ mA ($p \gg 1$) for both wavelengths of diffracted neutrons.

The dependence of the values p on the wave length of neutrons was investigated experimentally for two thickness es ($T_x = 3$ mm and $T_z = 14$ mm) of the bar cut from the single crystal of SiO_2. In Fig. 15 the experimental values corresponding to the relation

$$\bar{R}_\lambda = (H_2/H_1) \cdot \mathcal{J}_0(\lambda_1)/\mathcal{J}_0(\lambda_2)\,[\bar{\mathcal{J}}_V(i, \lambda_2)/\bar{\mathcal{J}}_V(i, \lambda_1)] \tag{4.41}$$

are shown as curves 3 and 4. In the relation (4.41) H_2 and H_1 denotes the half-width of the rocking curve for the wave lengths λ_2 and λ_1 respcetively, and $\bar{\mathcal{J}}_V(i, \lambda)$ is the mean value of the peak intensity of neutrons diffracted by the vibrating bar cut from the single crystal of SiO_2. For the values $p \gg 1$ the relation $\bar{R}_\lambda \approx$ $\approx (\lambda_2/\lambda_1)^2$ results from the expression (4.38) under supposition that $\cos \Theta_{B1}/$ $/ \cos \Theta_{B2} \approx 1$. In our case the calculated value of $\bar{R}_\lambda$ is equal to $\bar{R}_\lambda = 2.2$. For $i > 2$ mA the experimental values of $\bar{R}_\lambda$ are independent of the resonator current i and are in quite a good agreement with the calculated value 2.2 for both thicknesses of the single crystal bar of SiO_2.

Considering for the neutron diffraction the possibility of secondary reflexions during their flight across the sample, the integrated intensity $\mathcal{J}^\nu$ was calculated in the paper [97] supposing — like in the paper [73] — that this intensity is proportional to the number

$$\overline{n(t)} = \frac{|\delta\Delta\Theta_B(t)|}{2.66s} = \frac{4}{\pi} u_{0k} \frac{k\omega \operatorname{tg} \Theta_B}{|v_{nx}|} \sin \frac{k\omega T \operatorname{tg} \Theta_B}{2|v_{nx}|} \sin \frac{k\pi}{L} x \tag{4.42}$$

of perfect "crystalline layers" normal to the length L of the bar; the quantity s is given by the expression (4.31).

Hence $\mathcal{J}^\nu$ can be written in the form

$$\mathcal{J}^\nu = \mathcal{J}_1 \frac{\overline{n(t)}}{T} \nu \tag{4.43}$$

where $\nu = S_0 T$ is the irradiated crystal volume, S_0 the area of the face of the irradiated volume element and $\mathcal{J}_1$ the integrated intensity of a beam diffracted by one "crystalline layer" with a unit area of the face.

The quartz single crystal bar was excited in the 1st, 3rd and 5th harmonic frequencies (i.e. for $k = 1$, 3 and 5) and the diffraction of neutrons by the lattice plane (01.0) in the position of symmetric Laue transmission was investigated for both thicknesses T_x and T_z. Fig. 16 illustrates the dependence of the integrated intensity $\mathcal{J}^\nu$ by a vibrating quartz single crystal bar on the resonator current i to which the vibration amplitude u_{01} is considered to be proportional. A vibration amplitude $u_{01} = 4$ μm corresponds to the resonator current $i = 5$ mA.

The results shown in Fig. 16 can be assumed as a very good agreement of the experimental results with the theoretical considerations mentioned above for $k = 1$. The linear dependence of $\mathcal{J}^v$ on the excitation current i proves the relations (4.42) and (4.43) in the case of $T_X = 3$ mm, resp. $T_Z = 13$ mm for $i > 1$ mA, resp. $i < 0.3$ mA, considering u_{01} as a linear function of i.

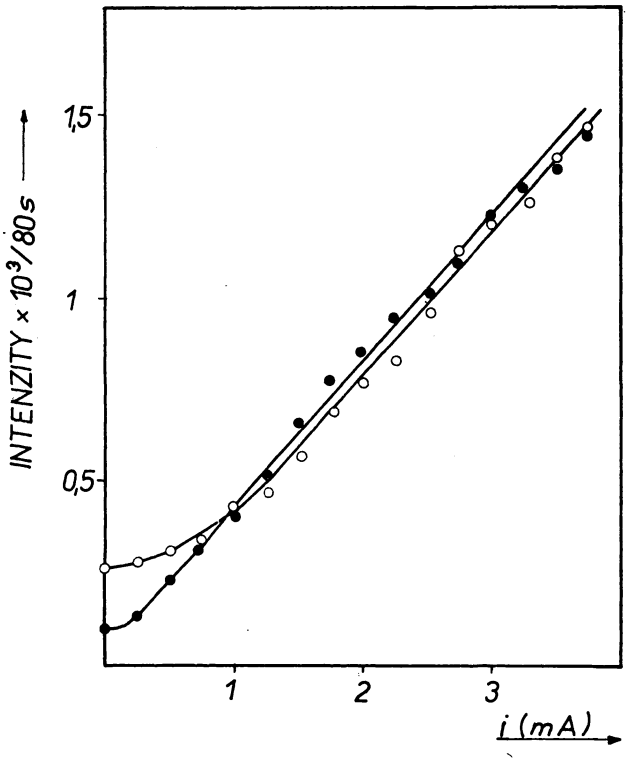
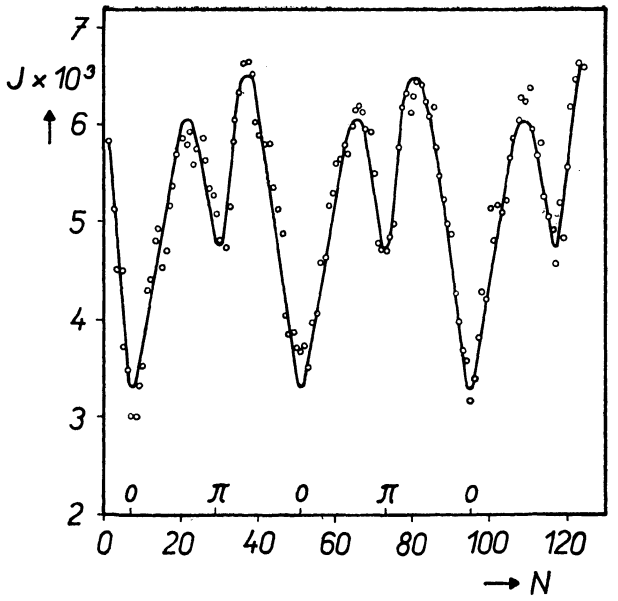


Fig. 16. The integrated intensity $\mathcal{J}^v$ as a function of the current i flowing through the quartz resonator for longitudinal vibrations at the fundamental frequency $f = 38.948$ kHz for two thicknesses $T_X = 3$ mm [curve ○] and $T_Z = 13$ mm [curve ●]; the bar length $L = 77$ mm

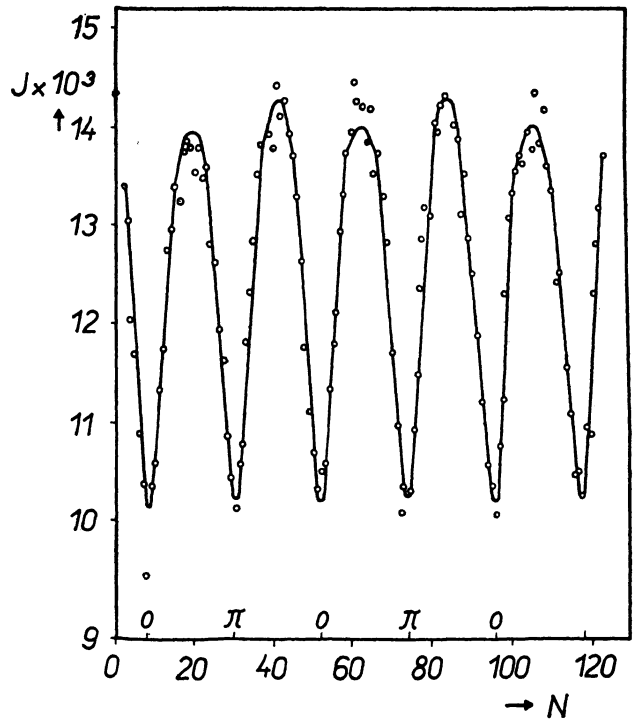
As u_{0k} becomes still larger, $\mathcal{J}^v$ in equ. (4.43) increases indefinitely. From the physical point of view, this is impossible; for large values of u_{0k} the curve is expected to level off. A further factor that should be considered is the fact that for larger amplitudes u_{0k} is no longer a linear function of the crystal current i.

Therefore, the deviation from linearity in the intensity dependence for $k = 3$ and $k = 5$ may be due to either one or both of the reasons mentioned above.

The analysis of the experimental results — showing on the contrary to the Fig. 16. — great differences for $T_X = 3$ mm and for $T_Z = 13$ mm in cases of


J x 10³
7
6
5
4
3
2
0
π
0
π
0
0
20
40
60
80
100
120
N

a


J x 10³
15
14
13
12
11
10
9
0
π
0
π
0
0
20
40
60
80
100
120
N

b

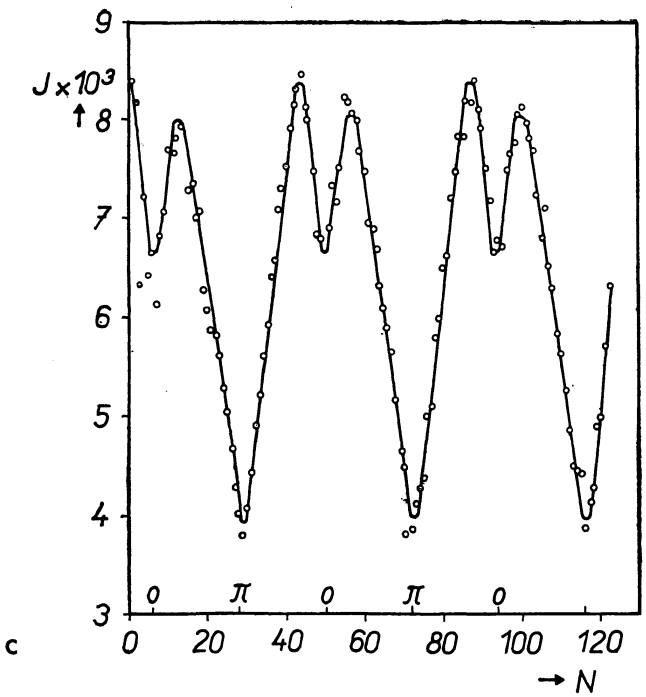


Fig. 17. Time modulation of neutrons diffracted by vibrating quartz single crystal in the position with (*a*) $\Delta\Theta_B = \Theta - \Theta_B = -5.4'$, (*b*) $\Delta\Theta_B = 0$, (*c*) $\Delta\Theta_B = \Theta - \Theta_B = +6.7'$. The smooth curves were obtained from experimental values of neutron intensities by the method of minimum squares

$k = 3$ and $k = 5$, requires relations in which the presence of the term

$$\sin \frac{k\omega T_Z \operatorname{tg} \Theta_B}{2|v_{ny}|}$$

in $\mathcal{J}_Z^v$ enables us to estimate the magnitude of secondary reflexions presuming that each of the $\overline{n(t)}$ "crystalline layers" diffracts totally in the Braggs case.

Theoretical values of $(\mathcal{J}_Z^v/\mathcal{J}_X^v)_c$ for $k = 1, 3$ and 5 are 1.00, 0.76 and 0.41. The averige experimental values of $(\mathcal{J}_Z^v/\mathcal{J}_X^v)_{exp}$ for $k = 1$ and 3 are 1.03 (for $i \geqq 0.75$ mA) and 0.61 (for $i \geqq 0.25$ mA).

For $k = 5$ the comparison of the calculated value with the average experimental one is not possible, because the experimental value of $(\mathcal{J}_Z^v/\mathcal{J}_X^v)_{exp}$ depends on the high-frequency exciting current i, which dependence was observed only for $k = 1$ and $k = 3$. Thus in the case of $k = 5$ it is only possible to compare individual experimental quantities at low values of i.

4.3 MODULATION OF THE INTENSITY OF THE NEUTRON BEAM DIFFRACTED BY A VIBRATING CRYSTAL LATTICE

4.3.1 *Experimental facts*

At the International conference concerning investigations on piezoelectric resonators in Liberec in September 1965 the attention was brought to the possibility of intensity modulation of neutrons diffracted by a vibrating crystal lattice [3] which was in more detailed way described in further publications [4] and [5]. Practically in the same time T. F. Parkinson and M. W. Moyer [30], [31] and A. G. Klein et al. [32] have realised the beam modulation of a neutron beam by vibrating lattices of quartz resonators. The observed results were confirmed and further develloped on quartz resonator in papers [7], [8], [9], [14], [15], [33], [16], [34], on Ge and Si in [41], on ADP in paper [43] and on KDP in the diplom work [98].

On a bar shaped quartz single crystal, having dimensions 3 mm $\times$ 14 mm $\times$ $\times$ 120 mm the time modulation of neutrons with wavelength $\lambda_n = 1.54$ Å by the lattice plane (020) was investigated in paper [16]. The quartz bar was piezoelectrically excited in the series resonance of the fundamental mode of vibration having the resonance frequency $f = 22.6$ kHz. The centre of the neutron beam impinging the lattice plane was at a distance $y = 3\,l/8$ from the centre of the vibrating quartz bar. The time modulation of diffracted neutrons was measured by a multichannel analyzer by applying the time digital converter with the channel width of 1 μs. The modulation takes places with the repetition frequency of 2ω, where ω is the circular frequency of longitudinal vibrations in the direction of the bar length (see chapter 4.3.2). The variations of the neutron intensity with time shows Fig. 17. In this figure the measurements of the time modulation of neutrons diffracted by a vibrating single crystal in positions $\Delta\Theta_B = -5.4'$, $\Delta\Theta_B = 0$ and $\Delta\Theta_B = +6.7'$ are illustrated. The intensity of neutrons for a non-vibrating single crystal is time independent in all positions and about four times smaller than the mean intensity of neutrons diffracted by a vibrating single crystal.

Similar results were obtained for the time modulation of the neutron beam ($\lambda_n = 1.05$ Å) by the lattice plane of a single crystal quartz bar, having dimensions $3 \times 14 \times 120$ mm^3 and oriented in the quartz crystal in such a way that the surface 14×120 mm^2 was identic with the crystallographic plane (01.1). The bar was excited electromechanically in its fundamental frequency $f = 1160$ Hz of flexural vibrations in the direction of the 3 mm long edge.

The results in the dependence on the control voltage are given in Fig. 18, in which the curve 2 shows that for small excitation voltage the fundamental modulation component has the frequency ω. With increasing amplitude of vibrations the components with the frequency 2ω, 3ω, 4ω ... (see curves 3 and 4 of Fig. 18) are gradually arising. The observed phenomena can be explained by the assumption of the existence of the static bending of the investigated bar, characterized by the radius R_0 of curvature (see paper [41]).

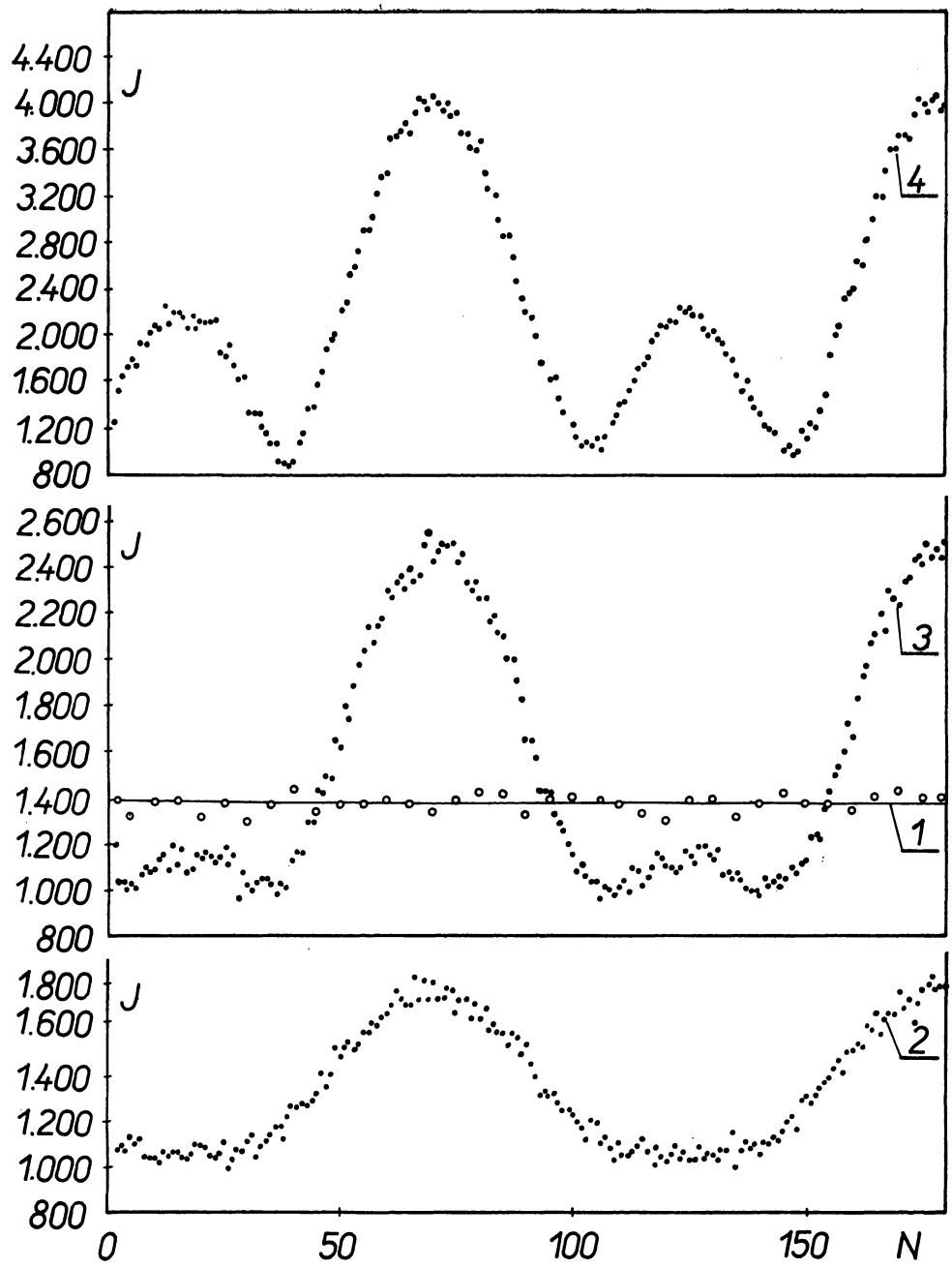


Fig. 18. Time modulation of the neutron beam diffracted by the plane (01.1) of the single crystal SiO_2; curve 1 for $U = 0$ mV; curve 2 for $U = 200$ mV; curve 3 for $U = 500$ mV; curve 4 for $U = 1000$ mV. J denotes the counts per channel of the analyzer, and N the channel number

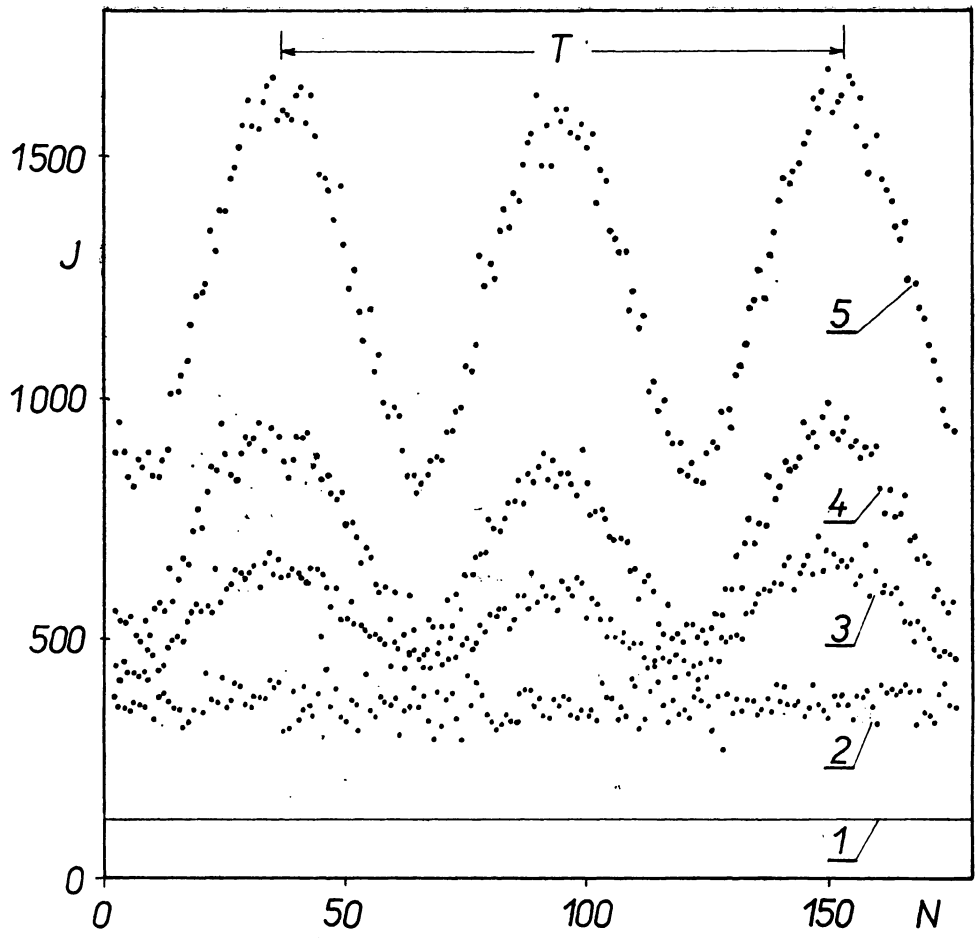


Fig. 19. Time modulation of the neutron beam diffracted by the plane $(22\bar{4})$ of the vibrating single crystal of Si. Curve 1 background for $U = 0$ mV; curve 2 for $U = 0$ mV; curve 3 for $U = 500$ mV; curve 4 for $U = 1000$ mV; curve 5 for $U = 2000$ mV. J denotes counts per channel, N the number of the time channel of the analyzer, and T the period of bar vibrations

Measurements of the time modulation of the diffracted neutron beam were performed as a function of the amplitude of vibration of the bar also for the diffracting lattice plane $(22\bar{4})$ of the silicon single crystal. The obtained experimental data are shown in Fig. 19 from which it can be seen that the neutron beam is modulated by the fundamental component with frequency of 2ω. For the explanation of this fact see again the chapter 4.3.2.

With the same experimental arrangement the time modulation of a neutron beam was carried out for the diffraction by the lattice plane (111) and by the lattice plane (333) of single crystal of germanium. The experimental data are shown in Fig.

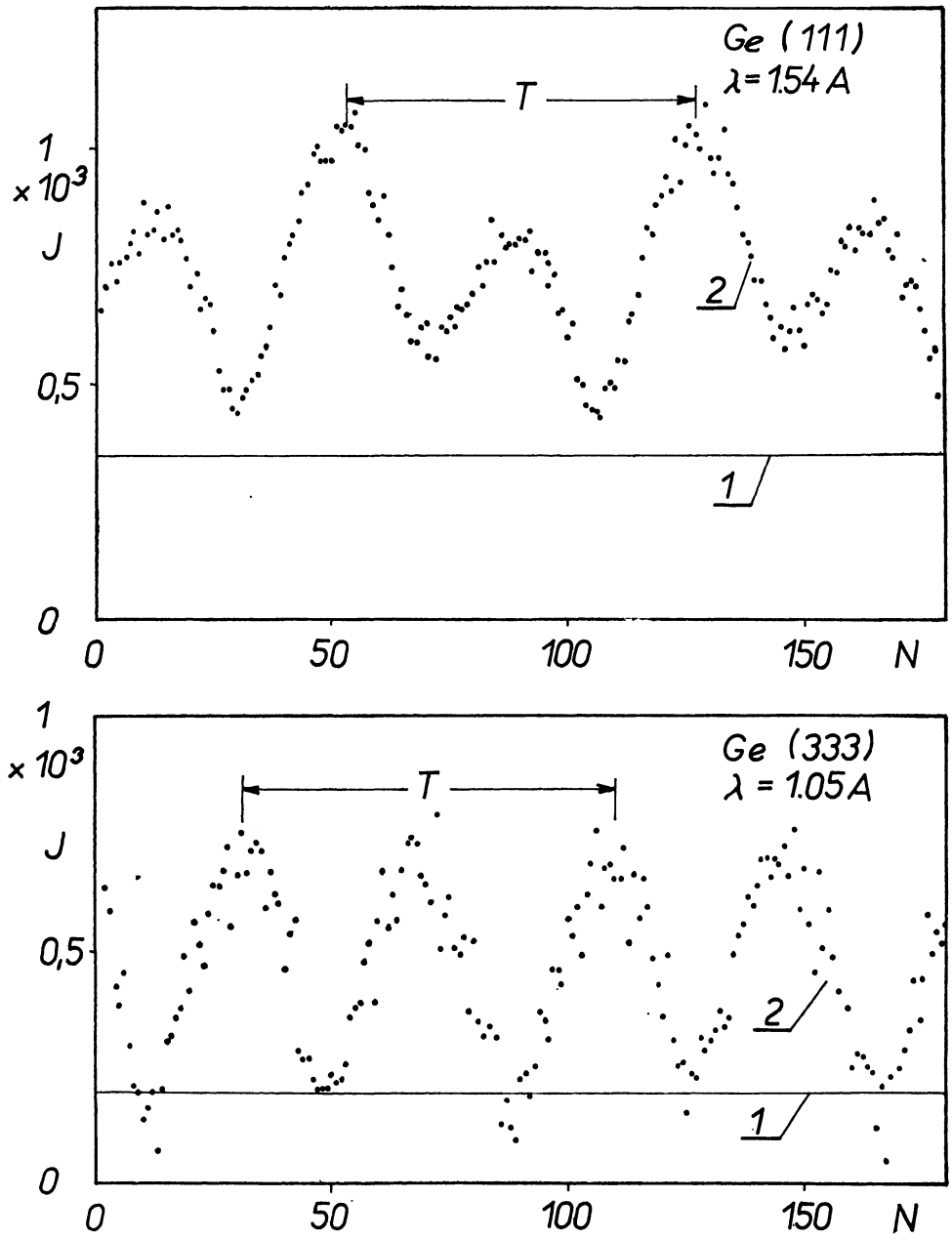


Fig. 20. Time modulation of the neutron beam diffracted by a vibrating single crystal of Ge. *a*) (111); $\lambda_2 = 1.54$ Å; *b*) 333; $\lambda_1 = 1.05$ Å. Curve 1 for $U = 0$ mV; curve 2 for $U = 800$ mV. $\mathcal{J}$ denotes counts per channel, N the number of the time channel of the analyzer, and T the period of bar vibrations

20. For neutrons with $\lambda_n = 1.05$ Å diffracted on the lattice plane (333) it can be seen in the lower part of Fig. 20, that the fundamental modulation component has the frequency 2ω. This diagram shows also, that there exists some influence of Doppler and aberration effects causing different counting rates of neutrons in odd and even minima of the time spectrum as a consequence of the assymmetry of the rocking curve of neutrons diffracted by the plane (333). The time modulation of neutrons with the wavelength of $\lambda_n = 1.54$ Å diffracted by the plane (111) may be explained like in the case of SiO_2 (see Fig. 18) assuming a certain degree of static deformation existing in the non-vibrating single crystal.

Experiments on time modulation of a neutron beam diffracted on lattice planes of plates cut from ADP [43], [66] excited in thickness-shear vibrations, and of bars cut from KDP [98] excited in longitudinal vibrations, offered similar results.

4.3.2 *An attempt of an approximate explanation of observed results*

It is difficult to describe and explain the physical process of the time modulation of a neutron beam by vibrations of lattice plane, however it can be illustrated in the following way.

If we consider a single crystal bar vibrating longitudinally in the direction of its length L (Fig. 6), we can use for the displacement u the expression (4.22). The velocity of the motion of crystallographic lattice planes of the bar in the direction of the X-axis is then given by the relation,

$$v = \frac{\partial u}{\partial t} = v_p \cos \omega t\,, \quad v_p = u_0 \omega \sin \frac{\pi x}{L}\,. \tag{4.44}$$

If the diffraction lattice plane of the single crystal is moving with this velocity a change of the Bragg angle Θ_B takes place. In the case when the direction of the velocity amplitude v_p is collinear with the reciprocal lattice vector $\vec{\tau}$, $v_p/v_n \ll 1$, δt expresses the change of the diffraction angle Θ_B in the form

$$\delta t = \delta_{0t} \cos \omega t \tag{4.45}$$

where

$$\delta_{0t} = \frac{v_p}{v_n} \cos \Theta_B + \frac{v_p}{v_n} \sin \Theta_B \tan \Theta_B = \frac{v_p}{v_n} \frac{1}{\cos \Theta_B} \tag{4.46}$$

which conforms with equation (1) of Shull et al. [36]. The first term of the eq. (4.46) is due to aberration the second one to the Doppler effect.

The rocking curve of the single crystal can be expressed in the form (see also (3.9) and (4.23))

$$\mathcal{J}(\delta) = \mathcal{J}(0) \exp\left(-\frac{\delta^2}{2\sigma^2}\right) \tag{4.47}$$

where $\mathcal{J}(0)$ is the neutron intensity in the maximum of the rocking curve, $\delta = \Theta - \Theta_B$ is the deviation from the Bragg angle Θ_B and σ is connected with the full width H_{u_0} of the rocking curve at half maximum intensity by the relation (4.24).

The influence of the periodic displacement of the vibrating bar on the deviation δ can be described by the relation

$$\delta = \delta_0 + \delta_t = \delta_0 + \delta_{0t} \cos \omega t \tag{4.48}$$

in which δ_0 is a time — independent deviation from the Bragg angle given by the position of the diffracting lattice plane and δ_{0t} is given by eq. (4.46). The influence of the periodic time variation δ, i.e. of the time modulation on the neutron intensity $\mathcal{J}(\delta, t)$ can be described in analogy to (4.47) by the corresponding expression

$$\mathcal{J}(\delta, t) = \mathcal{J}(0, t) \exp\left[-\frac{(\delta_0 + \delta_t)^2}{2\sigma^2}\right] . \tag{4.49}$$

The value $\mathcal{J}(0, t)$ represents the time modulated maximum intensity of the rocking curve in the position $\delta_0 = 0$.

Applying the following relations [99], [100]

$$\exp(ix \sin \Theta) = \sum_{n=-\infty}^{+\infty} \mathcal{J}_n(x) \exp(in\Theta) \tag{4.50}$$

$$\mathcal{J}_n(iz) = \exp\left(n\frac{\pi}{2} i\right) I_n(z) \tag{4.51}$$

where $\mathcal{J}_n(x)$ and $I_n(z)$ are Bessel functions of the n-th order of the real and imaginary argument. Substituting for δ_t from equation (4.45), equation (4.49) becomes

$$\mathcal{J}(\delta, t) = \mathcal{J}(\delta_t, t) \exp\left(-\frac{\delta_0^2}{2\sigma^2}\right) \times \sum_{n=-\infty}^{+\infty} I_n\left(\frac{\delta_0 \cdot \delta_{0t}}{\sigma^2}\right) \exp[in(\omega t + \pi)] \tag{4.52}$$

where

$$\mathcal{J}(\delta_t, t) = \mathcal{J}(0, t) \exp\left(-\frac{\delta_{ot}^2}{4\sigma^2}\right) \times \sum_{m=-\infty}^{+\infty} I_m\left(\frac{\delta_{ot}^2}{4\sigma^2}\right) \exp[im(2\omega t + \pi)] . \tag{4.53}$$

As can be seen from equations (4.52) and (4.53), supplementary modulations occur in addition to the time modulation of diffracted neutrons. At the maximum of the rocking curve ($\delta_0 = 0$) the supplementary modulation components are $a_2 \cos 2\omega t$, $a_4 \cos 4\omega t$, $a_6 \cos 6\omega t$, etc. In positions where $\delta_0 \neq 0$ still further modulation components occur, $b_1 \cos \omega t$, $b_2 \cos 2\omega t$, $b_3 \cos 3\omega t$, etc., which are due to the influence of abberation and the Doppler effect on the time modulation of neutrons diffracted by a vibrating single crystal.

Due to these facts the integrated intensity $\mathcal{J}^v$ given by the formula (4.43) changes periodically with time — according to the given experimental conditions — as described in chapter 4.3.1.

5. Conclusions

Finally we would like to draw the attention to the fact that the phenomena observed in the course of investigations upon the neutron diffraction by vibrating lattice planes are interesting also by comparing them with analogous results obtained

in experiments of X-rays diffraction. They have, however, useful applications too, several examples of which we would to mention here.

As concerns the diffraction of X-rays by lattice planes of single crystals deformed statically or periodically, there exist similar observations of the enhancement of the diffracted intensity [45] as in the case of neutron diffraction (especially in the publications from the last time [38], [49], [107], [27], [28], [29], [42], [48], [83], [108] till [113]) what is brought about by the fact, that in principle the causes of this effect are in both cases the same. For the excitation of lattice vibration of nonpiezoelectric crystals an acustoelectrical method [115] equivalent to our electromechanical method [18], [25] was applied too.

It was, therefore, possible to apply in our paper [24] for the explanation of the phenomena observed in our investigations of neutron diffraction the procedure of the paper of White [49]. The main difference between the process of X-rays and neutron diffraction can be seen in the different absorption of X-rays and neutrons in the majority of single crystals and their different extinction features.

Comparing both cases of diffraction, it is interesting to mention that we do not know any paper in which the observation of the modulation of the intensity of X-rays by the frequency of the vibrating geometrical body prepared from the investigating single crystal could be observed. Observations of this effect could be probably performed in a similar way as was done in the statement of the acustic modulation of gamma-rays from ^{57}Fe by S. L. Ruby an D. L. Bolef. [116].

It is also interesting to state, that the neutron diffraction by vibrating lattice planes has important applications not only for solution of physical problems but especially in practical investigations [117]. The periodically modulated neutron beam is much more useful for different investigations than the normal beam of neutrons with constant intensity. In many cases, e.g. for the investigations of metastable states of atomic nuclei, it would be much more effective, to apply the modulated neutron beam than to use the normal one. Applying the lattice vibrations for monochromators, it is possible to increase their effectivness many times (even 100-times). From changeable intensity of neutrons diffracted by vibrating piezoelectric resonators, their quality can be determined. By neutron diffraction topography [21], [106] the different frequency modes of vibrating plates from single crystals can be identified in a similar way as in the paper [118]. For special cases even a pulse neutron beam can be formed by means of vibrating single crystal [11].

Acknowledgments

To the work outlined in this paper made in the course of investigations many iniciative contributions Ing. R. Michalec CSc., Ing. B. Chalupa, L. Sedláková, RNDr. P. Mikula and Ing. J. Čech from the Nuclear Physics Institute in Řež, Ing. J. Vrzal from the Department of Nuclear Physics of the Charles University and Doc. Ing. J. Zelenka CSc. from the College of Mechanical and Textile Engineering in Liberec; they offered also valuable comments while reading

the manuscript. Indispensable technical assistance provided Mr. J. Vávra in carrying out the experiments and Miss B. Hašková in working out the obtained data. To all of them I wish to express my sincere thanks for their help.

References

[1] MICHALEC R., VAVŘÍN J., CHALUPA B., VÁVRA J.: Report ÚJV ČSAV, Nr. 1562 (1966).
[2] MICHALEC R., VAVŘÍN J., CHALUPA B., VÁVRA J.: Jaderná energie *12* (1966), 421.
[3] PETRŽÍLKA V.: CZECH. J. PHYS. *B 16* (1966), 458.
[4] PETRŽÍLKA V.: NATURE *218* (1968), 80.
[5] PETRŽÍLKA V.: CZECH. J. PHYS. *B 18* (1968), 1111.
[6] CHALUPA B., MICHALEC R., PETRŽÍLKA V., TICHÝ J., ZELENKA J.: phys. stat. sol. *29* (1968), K 51.
[7] MICHALEC R., CHALUPA B., PETRŽÍLKOVÁ V., GALOCIOVÁ D., ZELENKA J., TICHÝ J.: phys. stat. sol. *31* (1969), K 95.
[8] MICHALEC R., CHALUPA B., GALOCIOVÁ D., MIKULA P.: Phys. Lett. *28A* (1969), 546.
[9] CHALUPA B., MICHALEC R., GALOCIOVÁ D., ČECH J.: Report ÚJV ČSAV, Nr. 2240 F (1969).
[10] ZELENKA J., TICHÝ J., CHALUPA B., MICHALEC R., PETRŽÍLKA V.: BRIT. J. Appl. Phys. *2* (1969), 1041.
[11] CHALUPA B., MICHALEC R., GALOCIOVÁ D.: Nucl. Instr. and Meth. *67* (1969), 357.
[12] MICHALEC R., CHALUPA B., ČECH J., PETRŽÍLKA V., KADEČKOVÁ S., TARABA O.: Phys. Lett. *29A* (1969), 679.
[13] GALOCIOVÁ D., TICHÝ J., ZELENKA J., MICHALEC R., CHALUPA B.: phys. stat. sol. *2* (1970), 211.
[14] CHALUPA B., MICHALEC R., VÁVRA J.: Atomnaja energija *5* (1970), 413.
[15] PETRŽÍLKA V., VRZAL J., MICHALEC R., CHALUPA B., MIKULA P., ZELENKA J.: phys. stat. sol. *42* (1970), 895.
[16] MICHALEC R., SEDLÁKOVÁ L., CHALUPA B., GALOCIOVÁ D., PETRŽÍLKA V.: Acta Cryst. *A27* (1971), 410.
[17] MICHALEC R., SEDLÁKOVÁ L., ČECH J., PETRŽÍLKA V.: Phys. Lett. *37A* (1971), 403.
[18] ZELENKA J., PETRŽÍLKA V., MICHALEC R., CHALUPA B., SEDLÁKOVÁ L.: Acustica *27* (1972), 159.
[19] PETRŽÍLKA V., ZELENKA J., VRZAL J., MICHALEC R., CHALUPA B.: Autorské osvědčení čís. 153 607, ČSSR (1973), Patent AP 92 092 DDR (1972), Patent 2,143.103 BRD (1975).
[20] PETRŽÍLKA V., MICHALEC R., CHALUPA B., ČECH J., SEDLÁKOVÁ L., MIKULA P.: Jaderná energie *11* (1972), 367.
[21] HRDLIČKA J., SEDLÁKOVÁ L., PETRŽÍLKA V.: Nature, Phys. Sc. *242* (1973), 109.
[22] MIKULA P., MICHALEC R., SEDLÁKOVÁ L., ČECH J., CHALUPA B., PETRŽÍLKA V.: phys. stat. sol. (a) *17* (1973), 163.
[23] PETRŽÍLKA V., ZELENKA J., ČECH J., CHALUPA B., MICHALEC R., MIKULA P., SEDLÁKOVÁ L.: phys. stat. sol. (1) *17* (1973), 691.
[24] MICHALEC R., CHALUPA B., MIKULA P., SEDLÁKOVÁ L., PETRŽÍLKA V.: J. of Appl. Cryst. *7* (1974), 588.
[25] PETRŽÍLKA V., ZELENKA J., VRZAL J., HOLAN P., MICHALEC R., CHALUPA B., SEDLÁKOVÁ L.: Acustica (to be published).
[26] KATO N.: Acta Crystal. *13* (1960), 349.
[27] KATO N.: J. of Phys. Soc. Japan *18* (1963), 1785.
[28] KATO N.: J. of Phys. Soc. Japan *19* (1964), 67.
[29] KATO N.: J. of Phys. Soc. Japan *19* (1964), 971.
[30] PARKINSON T. F., MOYER M. W.: Nature *211* (1966), 400.

[31] MOYER M. W., PARKINSON T. F.: Nucl. Instr. and Meth. *53* (1967), 299.
[32] KLEIN A. G., PRAGER P., WAGENFELD H., ELLIS P. J., SABINE T. M.: Appl. Phys. Lett. *10* (1967), 293.
[33] ENGELHART R. W., JACOBS A. M.: Nature *229* (1971), 111.
[34] JACOBS A. M., KENNEY E. S., JEFFRIES J. D. E.:
[35] SHULL C. G., GINGRICH N. S.: J. Appl. Phys. *35* (1964), 678.
[36] SHULL C. G., MORASH K. R., ROGERS J. G.: Acta Cryst. *A 24* (1968), 160.
[37] NÉMET A.: Helv. phys. acta *98* (1935), 117.
[38] HARUTA K.: J. Appl. Phys. *38* (1967), 3312.
[39] PETRŽÍLKA V., KOTLER A.: Věstník Král. české spol. nauk (1948), čís. IX.
[40] BECHMANN P., AYERS S.: Piezoelectricity, Post Office Research Station, London (1957).
[41] MICHALEC R., SEDLÁKOVÁ L., CHALUPA B., ČECH J., PETRŽÍLKA V., ZELENKA J.: phys. stat. sol. *23* (1974), 667.
[42] PENNING P., POLDER D.: Philips Res. Repts. *16* (1961), 419.
[43] PARKINSON T. F., GÜRMEN E., LOYALKA S. K., MUHLENSTEIN L. D.: Trans. Am. Nucl. Soc. *15* (1972), No. 1.
[44] BURAS B., GIEBULTOWITZ, MINOR W., RAJCA A.: Report H. C. Oersted Institute, University of Copenhagen (1971), phys. stat. sol. (a) *9* (1972), 423.
[45] FOX G. W., CARR P. H.: Phys. Rev. *37* (1931), 1622.
[46] DARWIN C. G.: Phil Mag. *27* (1914), 315 and 675.
[47] KATO N., ANDO Y.: J. Phys. Soc. Japan *21* (1966), 964.
[48] BONSE U.: Ztschr. f. Phys. *177* (1964), 385.
[49] WHITE J. E.: J. Appl. Phys. *21* (1950), 855.
[50] KLAR B., RUSTICHELLI R.: Nuovo Cimento *13B* (1973), 249.
[51] SHULL C. G.: Phys. Rev. Lett. *21* (1968), 1585.
[52] SHULL C. G., OBERTEUFFER J. A.: Phys. Rev. Lett. *29* (1972), 871.
[53] SHULL C. G., SHAW W. M.: Ztschr. f. Naturforschung *28* (1973), 657.
[54] SHULL C. G.: Phys. Rev. *179* (1969), 752.
[55] SHULL C. G.: J. Appl. Cryst. *6* (1973), 257.
[56] CHALOUPKA P.: Čs. čas. fys. *4* (1954), 562.
[57] CHALOUPKA P., TICHÝ J.: Slaboproudý obzor *16* (1955), 33.
[58] TICHÝ J.: Čs. čas. fys. *6* (1956), 250.
[59] TICHÝ J.: Ann. d. Phys. *7* (1958), 219.
[60] ZACHARIASEN W. H.: Phys. Rev. Lett. *18* (1967), Z 524 1—2.
[61] ZACHARIASEN W. H.: Acta Cryst. *23* (1967), 558.
[62] ZACHARIASEN W. H.: Theory of X-Ray Diffraction in Crystals, John Wiley, New York (1945).
[63] JAMES R. W.: The Optical Principles of the Diffraction of X-Rays, G. Bell and Sons Ltd., London (1967).
[64] BACON G. E.: Neutron Diffraction, At the Clarendon Press, Oxford (1955).
[65] BACON G. E., LOWDE R. D.: Acta Cryst. *1* (1948), 303.
[66] PARKINSON T. F., GÜRMEN E., LOYALKA S. K., MUHLESTEIN L. D.: J. of Appl. Phys. *45* (1974), 2021.
[67] BROCKHOUSE B. N.: Methods for Neutron Spectrometry, Atomic energy of Canada Ltd., Chalk River, Ontario 1961; AECL — 1183; CRNP — 947.
[68] BROCKHOUSE B. N.: Inelastic Scattering of Neutrons in Solids and Liquids, Atomic Energy Agency, Vienna (1961).
[69] Triple Axis Neutron Crystal Spectrometer TKSN-400; Technical Instruction, Institut badań jądrowych, Swierk K/Otwocka, PLR (1971).
[70] MAIER-LEIBNITZ H.: Ann. Acad. Sci. Fennicae, Phys. Ser. VI. (1967), 267; Nukleonik *8* (1966), 61.

[71] RUSTICHELLI F.: Nucl. Instr. and Meth. *74* (1969), 219.
[72] RUSTICHELLI F.: Report EUR 4414 (1970).
[73] ANTONINI M., CORCHIA M., NICOTERA E., RUSTICHELLI F.: Nucl. Instr. and Meth. *104* (1972), 147.
[74] FREY F.: Nucl. Instr. and Meth. *96* (1971), 471.
[75] RISTE T.: Nucl. Instr. and Meth. *86* (1970), 1.
[76] EGERT G., DACHS H.: J. Appl. Cryst. *3* (1970), 214.
[77] THOMAS P.: J. Appl. Cryst. *5* (1972), 78.
[78] ALEFELD B.: Ztschr. f. Phys. *228* (1969), 454.
[79] KENNEY S., JEFFERIES D. E., JACOBS M.: Nucl. Instr. and Meth. *117* (1974), 391.
[80] RUSTICHELLI F.: Nucl. Instr. and Meth. *83* (1970), 124.
[81] CAUCHOIS Y.: J. de Phys. et Rad. *3* (1932), 320.
[82] JOHANSSON T.: Ztschr. f. Phys. *82* (1933), 507.
[83] PERTMER G. A., HANSEN D. A., PARKINSON T. F.: J. Appl. Phys. *45* (1974), 3258.
[84] OKKERSE B., PENNING P.: Philips. Res. Repts *18* (1963), 82.
[85] RÖNTGEN W. C.: Wied. Ann. d. Phys. *18* (1883), 213; 534.
[86] NY TSI ZE: J. de Phys. et Radium *9* (1928), 13.
[87] YOSHINOBU KAKIUCHI: Phys. Rev. 54 (1938), 772.
[88] SEDLÁKOVÁ L., PETRŽÍLKA V., KAWALEC M., ČECH J., MICHALEC R., MIKULA P., CHALUPA B., ZELENKA J.: phys. stat. sol. (a) *22* (1974), K 223.
[89] SEDLÁKOVÁ L., PETRŽÍLKA V., MIKULA P., CHALUPA B., MICHALEC R.: phys. stat. sol. (a) *27*, 309 (1975).
[90] VOIGT W.: Lehrbuch der Kristallphysik, Verlag Teubner B. G., Leipzig—Berlin (1910).
[91] SCHEIBE E.: Piezoelektrizität des Quarzes, Verlag Steinkopff Th., Dresden und Leipzig (1938).
[92] CADY W. G.: Piezoelectricity, Mc Graw-Hill Book Co., New York and London; 1. Ed. (1946); 2. Ed. (1964).
[93] MASON W. P.: Piezoelectric Crystals and their Application to Ultrasonics, D. van Nostrand, New York and London (1950).
[94] PETRŽÍLKA V., SLAVÍK J. B., ŠOLC I., TARABA O., TICHÝ J., ZELENKA J.: Piezoelektřina, Nakl. ČSAV, Praha 1961.
[95] HEISING R. A.: Quartz Crystals for Electrical Circuits, D. van Nostrand, New York and London (1952).
[96] WIEN W., HARMS F.: Handbuch der Experimentalphysik, Teil 1, Bd. 17, Akad. Verlagsges., Leipzig 1934.
[97] MIKULA P., MICHALEC R., ČECH J., CHALUPA B., SEDLÁKOVÁ L., PETRŽÍLKA V.: Acta Cryst. *A 30* (1974), 560.
[98] KAWALEC M.: Diplom Work, Charles University, Prague (1973).
[99] JAHNKE E., EMDE F., LÖSCH F.: Tafeln höherer Funktionen, Verlagsgesellschaft Teubner B. G., Leipzig (1960).
[100] JEFFREYS H., JEFFREYS B. S.: Methods of Mathematical Physics, Cambridge University Press, Cambridge 1948.
[101] JUNA J.: Report ÚJV ČSAV 2249 F, Řež u Prahy (1969).
[102] TAUPIN D.: Thèses Université de Paris (1964).
[103] TAUPIN D.: Bull. Soc. Franc. Minér. Cryst., *87* (1964), 469.
[104] MIKULA P., MICHALEC R., SEDLÁKOVÁ L., CHALUPA B., ČECH J.: phys. stat. sol. (a) *26* (1974), 317.
[105] CHALUPA B., MICHALEC R., ČECH J., MIKULA P., SEDLÁKOVÁ L., PETRŽÍLKA V., ZELENKA J.: phys. stat. sol. (a) *32* (1975), 541.
[106] MICHALEC R., MIKULA P., SEDLÁKOVÁ L., CHALUPA B., ZELENKA J., PETRŽÍLKA V., HRDLIČKA Z.: J. Appl. Cryst. *8* (1975), 345.

[107] HUNTER L. P.: J. Appl. Phys. *30* (1959), 874.
[108] KÖHLER P., MÖHLING W., PEIBST H.: phys. stat. sol. *41* (1970), 75.
[109] LEMKE H., MÜLLER G. O., SCHNÜRER E.: phys. stat. sol. *41* (1970), 539.
[110] MÖHLING W., MÜLLER G. O., PEIBST H., SCHNÜRER E.: phys. stat. sol. (a) *2* (1970), 725.
[111] KIES J., KÖHLER R., MÖHLING W., PEIBST H.: Ztschr. f. Maturforsch. *28a* (1973), 610.
[112] KÖHLER R., MÖHLING W., PEIBST H.: phys. stat. sol. (b) *61* (1974), 173.
[113] KÖHLER R., MÖHLING W., PEIBST H.: phys. stat. sol (b) *61* (1974), 439.
[114] KENNEY E. S., JEFFERIES D. E., JACOBS A. M.: Nucl. Instr. Meth. *117* (1974), 391.
[115] HUTSON A. R., MC FEE J. H., WHITE D. L., Phys. Rev. Letters *7* (1961), 273.
[116] RUBY S. L., BOLEF D. L.: Phys. Rev. Letters *5* (1960), 5.
[117] PETRŽÍLKA V., MICHALEC R., CHALUPA P., SEDLÁKOVÁ L., MIKULA P., ČECH J.: Nucl. Instr. and Meth. *123* (1975), 353.
[118] SPENCER W. J.: Appl. Phys. Letters *2* (1963) 133; Phys. Acoustics., vol. 5., Ed. by W. P. Mason, Academic Press, New York and London 1968.