acta phys. slov. 24 (1974), No. 4

THE ULTRASONIC ORIENTATION OF CUBIC CRYSTALS IN THE VICINITY OF THE [001] AND [111] DIRECTIONS

PAVOL HEGEDÜŠ*, Žilina

In the present paper a method is suggested of the crystallographic orientation of a cubic crystal in the vicinity of the directions [001] and [111] using the velocity measurements of the longitudinal and shear ultrasonic waves. The method is based on the dependence of the ultrasonic velocity change on the change of the deviation angle of the investigated direction from the direction of the acoustic axis of the crystal. The theoretical discussion is specified to a NaCl single crystal and graphs and tables for the convenient application of this method are also presented.

I. INTRODUCTION

So far various methods of the crystallographic orientation of crystals have been developed. Generally the following methods are well known: the X-ray method [1], the optical method [2], the method of Schaefer and Bergman [3], and the ultrasonic method suggested for an aluminium crystal in paper [4]. In general the transmission and the diffraction optical methods require a more careful polishing and lapping procedure in preparing the crystal specimens than the X-ray method. The above methods applied to ionic crystals have a serious disadvantage as they may couse the formation of colour centres and various kinds of lattice defects due to radiation. This is a serious difficulty if perfect crystals are needed after the orientation has been performed. It is desirable in many cases to have such a method of orientation that does not change the number of defects in the crystal. From this point of view the ultrasonic method of orientation of crystals has a great advantage as the ultrasonic waves of a small amplitude have a slight influence on the concentration of the colour centres and other defects.

The method suggested in paper [4] allows to determine the orientation of a cubic crystal using the following procedure. The velocities of the longi-

* Katedra technickej fyziky VŠD, 010 88 ŽILINA, Marxa Engelsa 25.

tudinal wave $v^{(3)}$ and the two shear ultrasonic waves $v^{(1)}$ and $v^{(2)}$ spreading in the investigated direction are measured. The ratios $v^{(1)}/v^{(2)} = I_1$, $v^{(1)}/v^{(3)} = I_2$ and $v^{(2)}/v^{(3)} = I_3$ are calculated and from the graph constructed for the investigated crystal using the relation $v^{(i)}v^{(j)} = f(c_{klmn}, \varrho, \alpha, \beta, \gamma)$ (where c_{klmn} are the components of the elastic stiffness, ϱ is the mass density, α, β and γ are spherical angles) the curves corresponding to I_1, I_2 and I_3 are determined. The crossing point of these curves in the standard stereographic triangle given by the directions [001], [011] and [111] determines the spherical angles α, β and γ determining the unit vector $\boldsymbol{n}$ in the investigated direction. The change of the velocity of the ultrasonic wave with the change of its spreading direction is so small in the vicinity of the directions [001], [011] and [111] that the correct orientation of the crystal is particularly difficult and therefore this problem is solved in the present paper.

The directions perpendicular to the planes of symmetry in cubic crystals are pure mode axes of ultrasonic waves and crystals are usually investigated along these directions. The present ultrasonic techniques can give data of ultrasonic velocity with an accurasy from 0.1 % to 0.01 %. Using such accuracy of the velocity measurements the crystallographic orientation of the ionic crystals can be determined also in the vicinity of the directions [001] and [111].

II. THEORY

The dependence of the velocity of the ultrasonic waves on the angles of the misorientation Φ and Θ in cubic crystlas can be accordint to Fig. 1 expressed in the same form as in [5]:

$$v^{(i)} = v^{0(i)} + \Delta v^{(i)},$$

where $i = 1, 2, 3$, $v^{(i)}$ is the velocity of the i-mode of the ultrasonic wave spreading in the x_3-direction and $v^{0(i)}$ is the velocity of the i-mode spreading in the direction of y_3.

The y_3 axis of the coordinate system $\{y_1, y_2, y_3\}$ is directed along an axis in the vicinity of which the influence of the misorientation upon the velocity is investigated (see Fig. 1). In our case it is the direction [001] or [111]. The x_3 axis of the coordinate system $\{x_1, x_2, x_3\}$ is directed along the wave vector of the considered ultrasonic wave.

The velocity of the pure transverse wave of an arbitrary polarization spreading in the [001] direction is according to [6] given by

$$v^{0(1)} = v^{0(2)} = \left[\frac{c_{44}}{\varrho}\right]^{1/2} \tag{1}$$

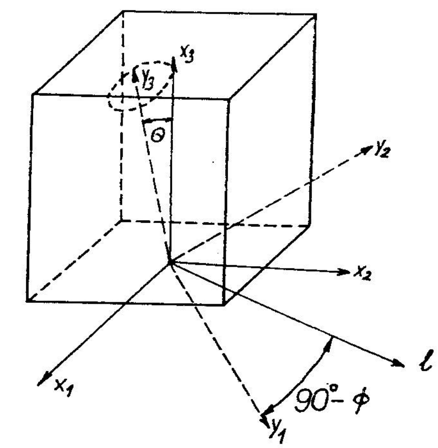


Fig. 1. The demonstration of the meaning of the angles Θ and Φ determining the misorientation of a crystal.

and the velocity of the longitudinal mode is

$$v^{0(3)} = \left[\frac{c_{11}}{\varrho}\right]^{1/2} \tag{1'}$$

where c_{11} and c_{44} are the corresponding elastic stiffness and ϱ is mass density.

For the change of the velocity of the ultrasonic waves with the change of the angles Θ and Φ in the vicinity of the direction [001] one can get [6] the following formulae:

The change of the velocity of the fast shear mode:

$$\Delta v^{(1)} = \frac{v^{0(1)}K_1\Theta^2}{4c_{44}} \{K_2 + [K_2^2 - (2K_2 - 1)\sin^2 2\Phi]^{1/2}\}. \tag{2}$$

The change of the velocity of the slow shear mode:

$$\Delta v^{(2)} = \frac{v^{0(2)}K_1\Theta^2}{4c_{44}} \{K_2 - [K_2^2 - (2K_2 - 1)\sin^2 2\Phi]^{1/2}\}. \tag{3}$$

The change of the velocity of the longitudinal mode:

$$\Delta v^{(3)} = -\frac{v^{0(3)}K_1K_2}{2c_{11}}\Theta^2, \tag{4}$$

where

$$K_1 = c_{11} - c_{12} - 2c_{44}$$

$$K_2 = \frac{K_1}{c_{44} - c_{12}} + 2$$

$$K_3 = \frac{K_1}{c_{12} + c_{44}} + \frac{3}{2}$$

The velocity of the pure transverse wave of an arbitrary polarization spreading in [111] is given by

$$v^{0(1)} = v^{0(2)} = \left[\frac{c_{11} - c_{12} + c_{44}}{3\varrho}\right]^{1/2} \qquad (5)$$

and the velocity of the longitudinal mode is

$$v^{0(3)} = \left[\frac{c_{11}}{\varrho}\right]^{1/2} \qquad (5')$$

For the change of the velocity of the ultrasonic waves with the change of the angle Θ in the vicinity of the direction [111] one can get following formulae:

The change of the velocity of the two shear modes is:

$$\Delta v^{(1)} = -\Delta v^{(2)} = -\frac{v^{0(1)}K_1}{2[c_{11} - c_{12} + c_{44}]}\Theta. \qquad (6)$$

The change of the velocity of the longitudinal mode is:

$$\Delta v^{(3)} = \frac{v^{0(3)}K_1[2K_3 + 3]}{3[c_{11} + 2c_{12} + 4c_{44}]}\Theta^2 \qquad (7)$$

The relations (2) to (7) have a straightforward utilization in the determina-ation of the misorientation of the investigated crystal in the vicinity of [001] and [111] directions if $\Delta v^{(i)}$, c_{11}, c_{12}, c_{44} and ϱ are known.

III. APPLICATION OF THE METHOD TO AN NaCl CRYSTAL

The room temperature values of the elastic stiffness constant in units $10^9\,\mathrm{Nm}^{-2}$ [5] are:

$$c_{11} = 49.00,\ c_{12} = 12.40,\ c_{44} = 12.60$$

Then

$$K_1 = 11.40 \times 10^9\,\mathrm{Nm}^{-2},\ K_2 = 1.68,\ K_3 = 1.98, \text{ and}$$

$$\varrho = 2.16 \times 10^3\,\mathrm{kgm}^{-3}.$$

The errors in measurement of c_{11}, c_{12}, c_{44} and ϱ exert an influence upon the estimation of $\Delta v^{(1)}$, $\Delta v^{(2)}$ and $\Delta v^{(3)}$ given by equations (2) to (7). It would be

necessary for the evaluation of these errors. We shall not do this analysis as we consider the estimation of all values used for calculation with an accuracy better than 0.01 %. This accuracy is reported in many papers so the above assumption can be taken as well checked.

Let us assume that the NaCl single crystal is slightly misoriented along the [001] direction. Then for the transverse waves of a different polarization different phase velocities are measured and the difference corresponds to the angles Θ and Φ according to relations (2), (3) and (4).

In practice, however, the misorientation with respect to Θ is most interesting and to get this the value $v^{(3)}$ is measured and using eq. (4) Θ is estimated from $\Delta v^{(3)}$. The meaning of Θ and Φ is clear from Fig. 1. The dependence $\Delta v^{(3)}$ on the angle Θ is plotted in Fig. 2 and accurate values of Θ as a function of $\Delta v^{(3)}$ are in Table 1. From Fig. 2 and Table 1 the angle Θ corresponding to the experimentally obtained value of $\Delta v^{(3)}$ can be estimated. One can see that the determination of the misorientation angle $\Theta \leq 1°$ requires an accuracy of measurement of $v^{(3)}$ better than 0.001 %. For Θ in the region $1° \leq \Theta \leq 3°$ the required accuracy is about 0.01 % and for $3° \leq \Theta \leq 5°$ the required accuracy is about 0.1 %.

In the case when the experimental method gives the accuracy in the velocity measurement of about 0.1 % the determination of the misorientation within 2° is not possible.

The dependence $\Delta v^{(1)}$ on Θ and Φ is in Table 2 and in graphs in Figs. 3a to

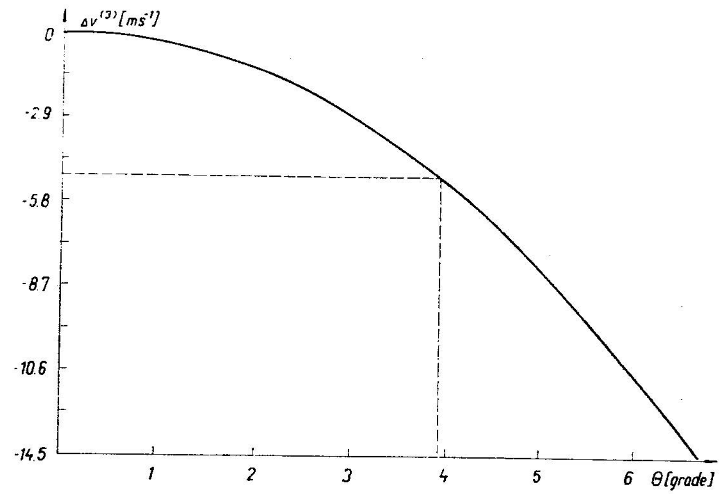


Fig. 2. The dependence of the velocity change of the quasi-longitudinal wave $\Delta v^{(3)}$ on the misorientation angle Θ in the vicinity of the [001] direction.

Table 1

The change of velocity $\Delta v^{(3)}$ with the angle Θ in the vicinity of the [001] direction

Θ [grade]	$\Delta v^{(3)}$ [ms^{-1}]
1	−0.29
2	−1.19
3	−2.70
4	−4.76
5	−7.44
6	−10.70
7	−14.58

3e. The particular graphs give $\Delta v^{(1)}$ versus Φ for different angles of Θ. The above mentioned graphs correspond to relations (1) to (7) and were calculated using a computer.

With regard to relation (2) the dependence $\Delta v^{(1)}$ on the angle Φ is considered for $0° \leq \Phi \leq 45°$ only.

Analysing Table 2 and Figs. 3a to 3e one can see that the determination of Φ at $\Theta = 1°$ requires the measurement of velocity with an accuracy better than 0.01 % for an arbitrary Φ. For $2° \leq \Theta \leq 4°$ the accuracy 0.01 % is sufficient and for $\Theta = 5°$ we need an accuracy of about 0.1 %.

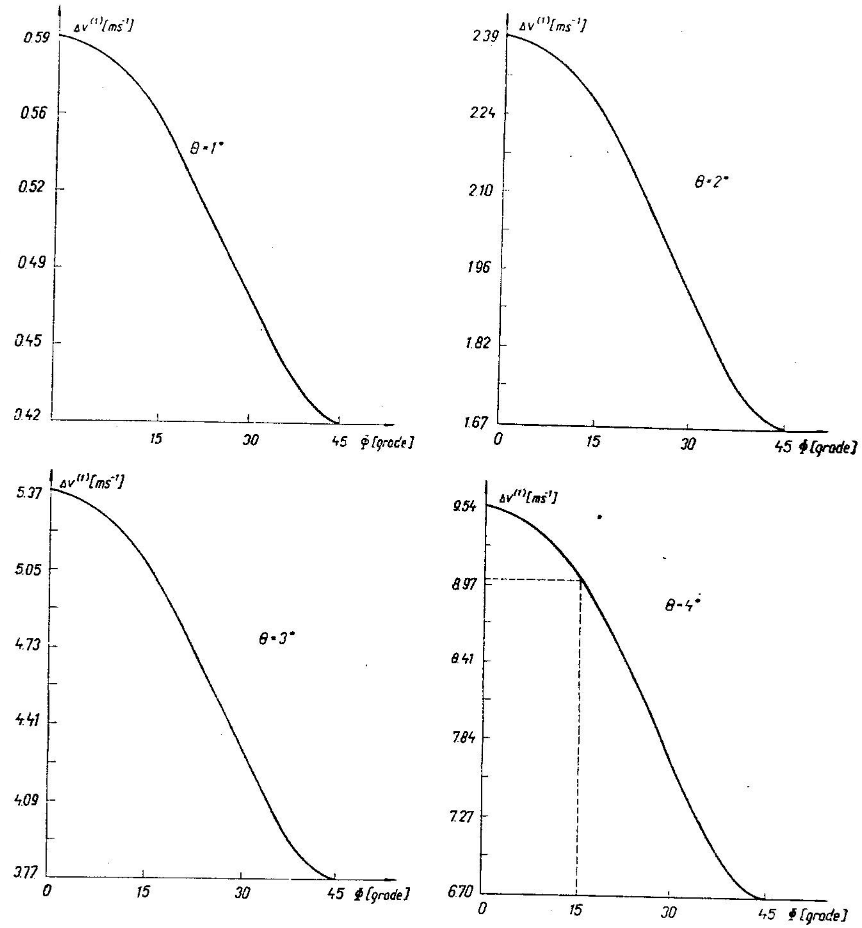
Table 3 and Figs. 4a to 4e give the dependence $\Delta v^{(2)}$ versus Θ and Φ. It can be shown that for angles $\Phi < 10°$ the required accuracy in velocity measurement must be better than 0.001 % for $\Theta \leq 5°$. For $10° \leq \Phi \leq 45°$ and for $\Theta \leq 2°$ the required accuracy is 0.001 %. For $10° \leq \Phi \leq 45°$ and $3° \leq \Theta \leq 5°$ the accuracy 0.01 % is sufficient.

If the single crystal of NaCl is oriented along the [111] direction the transverse wave velocity does not depend on its polarization and the first information

Table 2

The change of velocity $\Delta v^{(1)}$ [ms^{-1}] as a function of Φ and Θ in the vicinity of the [001] direction

	Θ [grade]				
φ	1	2	3	4	5
0	0.59	2.39	5.37	9.54	14.91
5	0.59	2.36	5.32	9.46	14.80
10	0.58	2.32	5.22	9.28	14.52
15	0.56	2.25	5.06	9.00	14.07
20	0.54	2.16	4.87	8.66	13.54
22.5	0.52	2.10	4.73	8.40	13.14
25	0.51	2.03	4.58	8.15	12.74
30	0.48	1.91	4.31	7.66	11.98
35	0.45	1.79	4.04	7.18	11.23
40	0.43	1.71	3.85	6.84	10.70
45	0.42	1.67	3.77	6.70	10.47



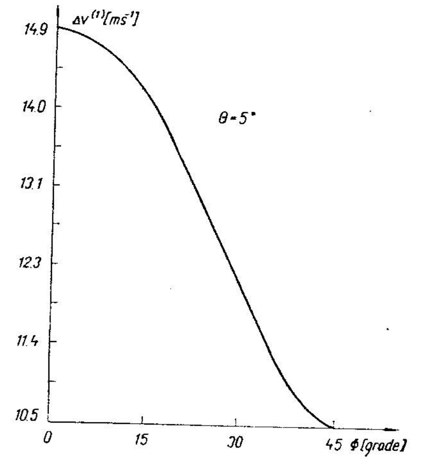


Fig. 3. The dependence of the velocity change of a fast quasi-shear wave $\Delta v^{(1)}$ in the vicinity of the [001] direction on the angles Θ and Φ: 3a for $\Theta = 1°$, 3b for $\Theta = 2°$, 3c for $\Theta = 3°$, 3d for $\Theta = 4°$, 3e for $\Theta = 5°$.

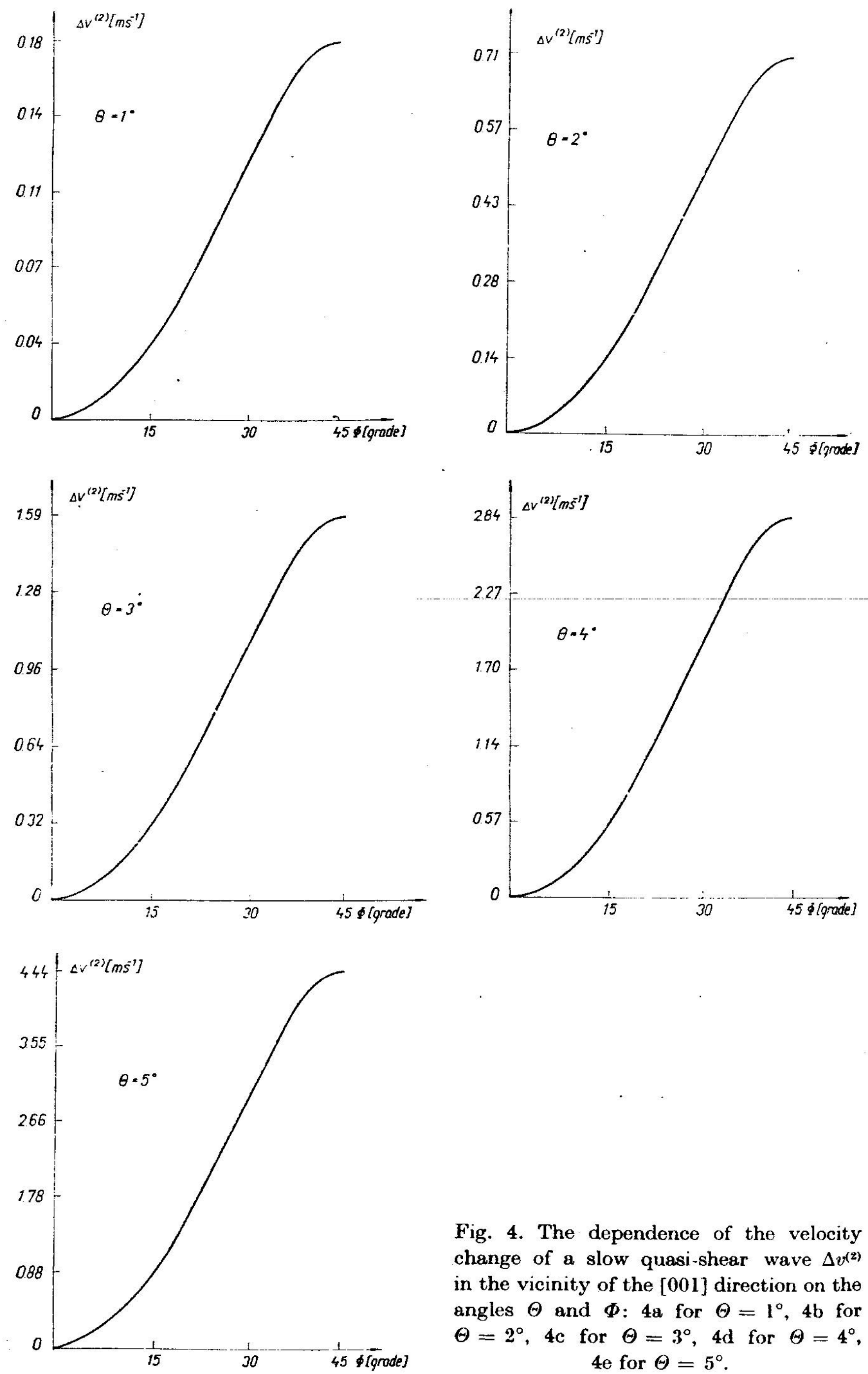


Fig. 4. The dependence of the velocity change of a slow quasi-shear wave $\Delta v^{(2)}$ in the vicinity of the [001] direction on the angles Θ and Φ: 4a for $\Theta = 1°$, 4b for $\Theta = 2°$, 4c for $\Theta = 3°$, 4d for $\Theta = 4°$, 4e for $\Theta = 5°$.

Table 3

The change of velocity $\Delta v^{(2)}$ [ms⁻¹] as a function of Φ and Θ in the vicinity of the [001] direction

φ [grade]	Θ [grade] 1	2	3	4	5
0	0	0	0	0	0
5	0.005	0.02	0,05	0.09	0.15
10	0.01	0.06	0.14	0.25	0.39
15	0.03	0.13	0.30	0.54	0.84
20	0.05	0.22	0.49	0.88	1.37
22.5	0.07	0.28	0.64	1.13	1.77
25	0.08	0.35	0,78	1.39	2.17
30	0.12	0.47	1.05	1.87	2.93
35	0.15	0.59	1.33	2.36	3.68
40	0,17	0.67	1.52	2.70	4.22
45	0.18	0.71	1.59	2.84	4.44

about a possible misorientation of the crystal can be obtained from the measurement of the ultrasonic velocity as follow: The velocity of the transverse ultrasonic wave generated by a Y-cut quartz transducer for different positions of the transducer with respect to the x_1 and x_2 axes is taken and if there is no difference in the obtained data the crystal is well oriented or the angle of misorientation is so small that it exerts no influence upon the velocity within the error of measurement.

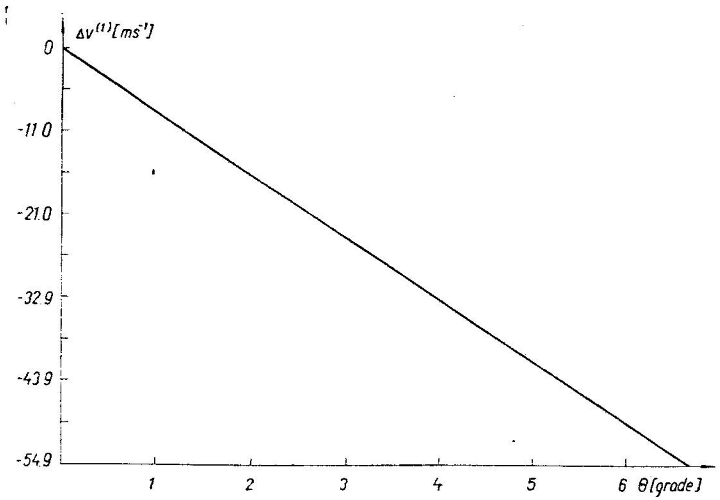
If the velocities measured for different positions of the transducer with respect to the x_1 and x_2 directions are different, then the misorientation angle Θ can be estimated.

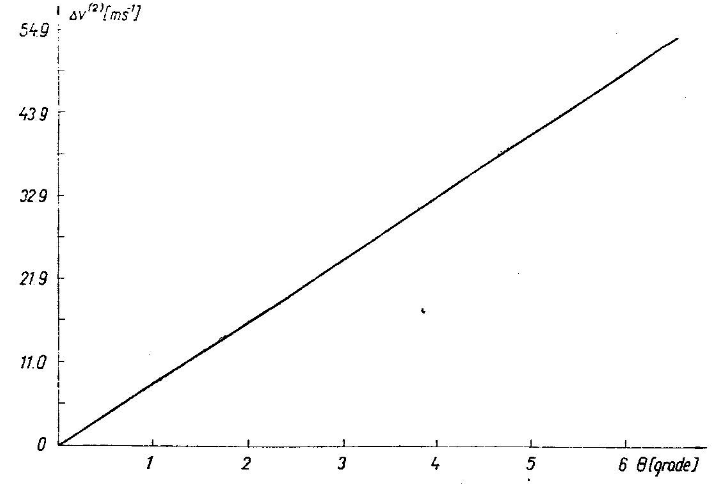
Table 4 and Figs. 5a to 5c show the change of $\Delta v^{(1)}$, $\Delta v^{(2)}$ and $\Delta v^{(3)}$ with the change of Θ. The analysis of reported graphs shows that the value of the misorientation angle Θ in the vicinity of the [111] direction can be determined

Table 4

The changes of velocities of all acoustic modes in the vicinity of the [111] direction as a function of the angle Θ

Θ [grade]	$\Delta v^{(1)}$ [ms⁻¹]	$\Delta v^{(2)}$ [ms⁻¹]	$\Delta v^{(3)}$ [ms⁻¹]
1	− 7.83	7.83	0.28
2	−15.67	15.67	1,13
3	−23.52	23.52	2.55
4	−31.34	31.34	4.53
5	−39.20	39.20	6.07
6	−47.00	47.00	10.05
7	−54.86	54.86	13.97





by the measurement of transverse wave velocities if the accuracy in the measurement of the phase velocity of the transverse ultrasonic wave is about 0.1 %. If the longitudinal ultrasonic wave is used to determine the crystallographic misorientation, at least an accuracy of 0.01 % is required. It follows that the transverse acoustic waves are more suitable for the orientation procedure than the longitudinal one.

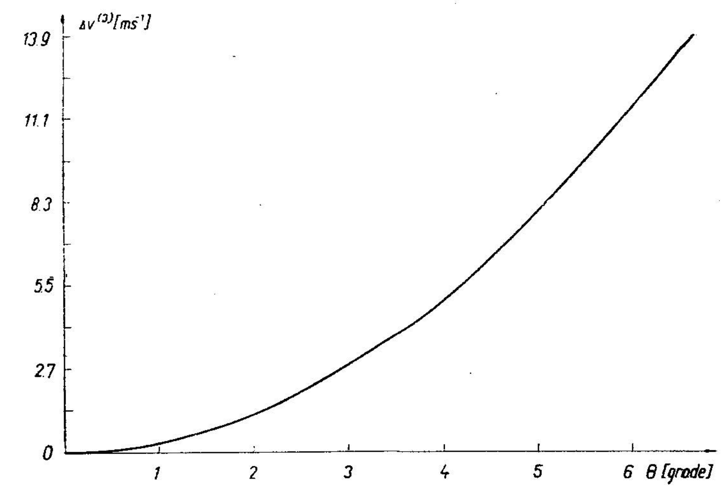


Fig. 5. The dependence of the velocity in the vicinity of the [111] direction as a function of the angle Θ. 5a for a slow quasi-shear wave $\Delta v^{(1)}$, 5b for a fast quasi-shear wave $\Delta v^{(2)}$, 5c for a quasi-longitudinal wave $\Delta v^{(3)}$.

The orientation procedure for the determination of the angles Θ and Φ in the vicinity of the [001] direction demonstrates the following example. The measurement of the ultrasonic velocities of the waves spreading in the vicinity of the [001] direction has given the following values: $\Delta v^{(3)} = -4.80\ \mathrm{ms}^{-1}$ and $\Delta v^{(1)} = 9.00\ \mathrm{ms}^{-1}$. We can see from Fig. 2 that the value $\Delta v^{(3)}$ gives $\Theta = 4°$. For the given value of Θ and the measured value $\Delta v^{(1)}$ from Fig. 3 we get $\Phi = 15°$. Corresponding values reported in the above example are marked in Figs. 2 and 3 with dashed lines.

IV. CONCLUSION

The above mentioned procedure can be used to construct the corresponding tables and graphs for an arbitrary crystal of cubic symmetry. A further utilization of this method requires the construction of appropriate graphs that would be usable for all cubic crystals. On the basis of the present results we can assume that this method is quite suitable in the vicinity of the [111] direction while the accurate crystallographic orientation of the crystal along the [101] direction remains an open problem.

The author wishes to express his thanks to RNDr. S. Kolník, CSc. and RNDr. I. Baják for stimulating discussions.

REFERENCES

[1] Бокий Г. Б., Порай-Кошиц М. А., *Практический курс рентгеноструктурного анализа*. Том I., Изд. МГУ, Москва 1952.

[2] Шубников А. В., *Оптическая кристаллография*. Изд. АН СССР, Москва 1950.

[3] Schaefer Cl., Bergmann L., Naturwiss. *22* (1943), 685.

[4] Green R. E. Jr., Henneke E. G. II., JASA *41* (1967), 84.

[5] Waterman P. C., Phys. Rev. *113* (1959), 1240.

[6] Truell R., Elbaum Ch., Chick B. B., *Ultrasonic methods in solid state physics*. Academic Press, London 1969.

Received March 27th, 1973