acta phys. slov. 40 (1990), No. 1

LOCAL CHARACTERISTICS OF SAW PROPAGATION IN TGS AND ROCHELLE SALT[1)]

HAŁACIŃSKI, B.,[2)] KOTLICKA, E.,[2)] LATUSZEK, A.,[2)] Warsaw

The angular dependence of the surface acoustic waves (SAW) velocity has been measured for the planes (010), $(\bar{1}\bar{1}0)$ and (001) for triglycine sulfate (TGS) and for the (100) and (001) planes for the Rochelle Salt at some temperatures. The objective was to explain the disagreement between calculation and experimental results. Also reported is the observation of different kinds of modes in these monocrystals.

I. INTRODUCTION

The knowledge of the SAW velocity in materials used for devices utilizing surface waves is the main information on those materials. Even if all the elastic, dielectric and piezoelectric constants are known, and there is the possibility to calculate the velocity by computer, the agreement between the calculation and measurement results very often is not satisfactory. Besides, for the crystals with domain structure, or materials such as the ceramics, the velocity of SAW may change from point on the same line on the plane.

II. THE MEASUREMENT EQUIPMENT

Measurements were done using the standard ultrasonic defectoscope, the adaptation of which for the SAW velocity measurements with high accuracy (of about 10^{-4}) was presented previously [1]. It was proposed to use the microscope countershaft fixed on the $X - Y$ shift table with micrometer. The position of the pulse-signal on a CRT screen could be determined by the order of the magnitude, more precisely due to that adaptation, with an accuracy defined by the mean-root-square error of a single measurement, that is about 20 μm.

Oriented samples were fixed on the rotary protractor which could be turned in the range of the round angle relative to mounted transducers.

The shape of the detected signals affects the measurement accuracy. The best shape of the impulse would be the δ-shape. Such shape of the impulse has been received for the interdigital transducers with two or three electrodes.

The velocity of SAW was determined from the measurement of the delay time of the electrical signal between the generating and the detecting transducers. The delay time was calibrated by means of a Y cut quartz. The velocity of the Rayleigh wave for the Y cut quartz in the X direction is equal to 3159 m/s [2].

III. EXPERIMENTAL RESULTS

During the preliminary investigations of about twenty different crystals of TGS it has been ascertained that the piezoelectric effect connected with natural unipolarity allows to generate SAW by interdigital transducers directly. Becauses of this possibility the studies have been carried our for crystalls which had not been thermal or electrically treated previously.

The crystal of the Rochelle Salt is piezoelectric in all phases.

In Fig. 1 and Fig. 2 there are presented the results obtained with two-electrodes transducers for the planes $(\bar{1}\bar{1}0)$ and (001), respectively, in crystals grown

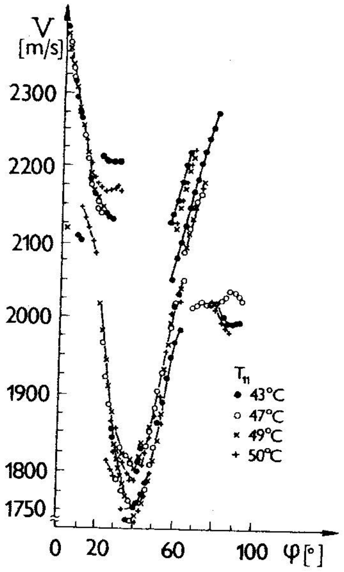

Fig. 1. The SAW velocity as a function of the wave propagation direction for the sector of the $(\bar{1}\bar{1}0)$ plane for the crystals grown at temperatures 43 °C (●), 47 °C (o), 49 °C (×) and 54 °C (+).

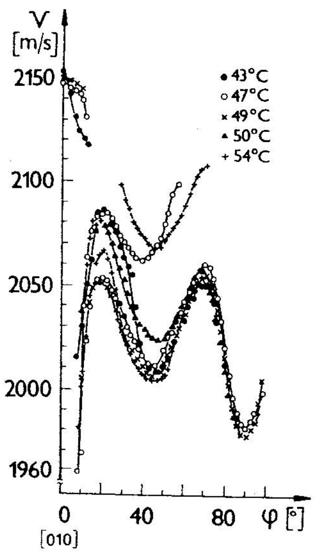

Fig. 2. The SAW velocity versus the propagation direction for the segment of the (001) plane for the TGS crystals grown at different temperatures: 43 °C (●), 47 °C (o), 49 °C (×) and 54 °C (+).

[1)] Contribution presented at the 11th Conference of Ultrasonic Methods in ŽILINA, August 31—September 2, 1988

[2)] Institute of Physics, Technical University of Warsaw, ul. Chodkiewicza 8, 02 525 WARSZAW, Poland in the temperature range 43 °C to 54 °C. These two planes were chosen as a natural plane of the crystal, very well shaped and big enough to carry out investigations. The crystal grown in the ferroelectric phase had biger dimensions than those which had been obtained in the paraelectric phase.

The used method allows to determine the wave velocity with an accuracy of 1 m/s with respect to quartz. An analysis of the measuring accuracy was done from the repeatibility of measurements for the same plane sector. However, for the given direction of wave propagation for different plane sectors, there are observed differences in the velocity value of the order of 50 m/s. The plotted results are the mean value of the results obtained for different plane sectors.

The assumed zero direction on the $(\bar{1}\bar{1}0)$ plane was the edge of the crystal, parallel to the *c*-axis (according to the system of crystallographic axes of the Hoshino et al. [3]), and the crystal had been rotated clockwise.

For the (001) plane the *b*-axis was chosen as a reference direction, and angles were measured to the left and to the right of it.

From the results it can be stated that the SAW velocity considerably changes with the change of the propagation direction. The greatest differences are observed for the $(\bar{1}\bar{1}0)$ plane. They are up to 600 m/s, and for the (001) plane — up to 200 m/s. For the same propagation direction there were observed some waves propagating with velocities ranging from 30 m/s to 300 m/s. In an anisotropic medium, when acoustic (volume) waves are propagating, even in the plane of symmetry of the sample, to each direction of the wave propagation there correspond three different rays, but in the direction of one ray there can propagate from three to five waves differing in the velocity and in the direction of the normal to the front of the wave. In the case of five existing waves, one of them is quasilongitudinal, the second is a pure shear wave, and the last three are quasishear waves [4].

From the plotted results it can be seen that the curves of the angular dependence of SAW velocity in TGS crystals have a consistent character but are not the same (in value). Both the maximum and the minimum values of velocity are different for the crystals grown at different temperatures. This dependence is observed in all investigated planes [5].

In Fig. 3 there are plotted the measurements and calculated results for the (010) plane [6]. The plane (010) is the cleavage plane and the *b*-axis is the polar axis. For the numerical analysis of the SAW propagation in TGS there were used the values of material constants obtained by Konstantinova et al. [7]. The samples for which the stiffness modulus c_{IK}, the coefficients of the piezoelectric strains d_{iJ} and the piezoelectric stress modulus e_{iK} were measured, were cut from unpolarized crystals and have not been polarized during the experiments.

In the angle range from 80° to 145°, i.e. in the vicinity of the [100] and the [101] directions, one can say that theoretical values are in line with measurements results with respect to the material constants determination. For the other directions it is possible to speak only about a qualitative agreement between those results. The reason for this disagreement between experimental and numerical results may be that for the calculation numerical constants measured for other crystals were taken than the crystals for which the wave velocity had been measured. The unpolarized TGS crystals have a domain structure varying from crystal to crystal.

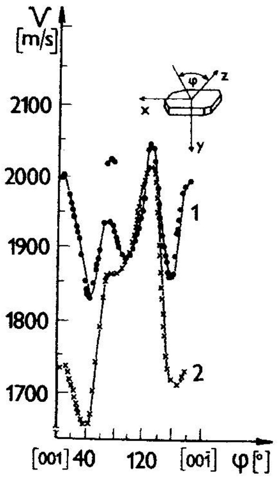

Fig. 3. Comparison of the measured (curve 1●) and the calculated (curve 2×) values of the phasse velocity of SAW for the (010) plane in TGS crystals.

The character of the domain structure in ferroelectrics very strongly depends on the number of possible orientations of a spontaneous polarization in crystallographic structure. In unipolar ferroelectrics, the direction of spontaneous polarization in neighbouring domains may differ by 180° only. In TGS the domains are elongated in the ferroelectric axis direction and in the plane perpendicular to it they have an oval (lense) shape. Many domains run through the whole crystal. Some, little needle-shaped domains terminate inside the crystal. There are also kind of domains, cigar-shaped, located inside the crystal, which do not run to the external planes. The shape and dimensions of the domain depend on the growth pyramid and the conditions of growing, especially on the temperature of growing. The largest domains are in the growth pyramid (110). They are irregular, lense-shaped, elongated perpendicularly to the natural edge of the crystal, which is the *c*-axis. In the growth pyramid (111) and (010) the domains have the same shape around the axes parallel to the (102) wall. In the pyramid (001) there are domains around the direction parallel to this wall, and the domains in the shape of a block forming a 60° angle with the edge of the crystal. These domains run through the crystal. The dimensions of domains in the TGS crystals are from 100 μm to a dozen or so millimeters [8].

It seems that similarly to the case of one-domain and multi-domain samples, the values of the moduli c_{iK} and d_{iK} are different, such differences in values of these moduli can be observed from point to point on the surface of the crystal, which depends on the domain dimensions. This heterogeneity of the sample may explain the differences noticed in the values of the SAW velocity at various points of the surface for the same direction on the wave propagation. In non-linear piezoelectrics the sound velocity depends on the applied electric field [9]. The different values of the critical field E_k (E_k are obtained from the displacement of the hysteresis loop with respect to the centre of the system) being the results of various states of the polarization stabilization, it may be treated as a different applied field.

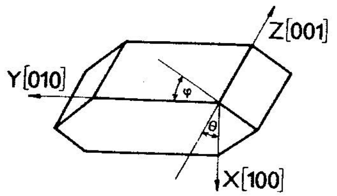

Fig. 4. The system of coordinates for the velocity measurements for the Rochell Salt crystal.

The dependence of the SAW velocity on the domain structure was verified on the measurements in the Rochelle Salt (R. S.). The most characteristic singularity of the R. S. is the presence of two Curie temperatures, one at −18 °C and the other at +24 °C, and between them this material reveals ferroelectrical properties. Outside these limits crystals are piezoelectric belonging to the rhombohedrical 222 class of symmetry [9]. The ferroelectric phase belongs to the monoclinic class 2 with the polar axis parallel to the rhombohedrical axis [100]. The crystal on which the velocities of SAW were measured was of the shape pictured in Fig. 4 in which the directions of the axes and the measured angles of SAW propagation are indicated.

In Fig. 5 and Fig. 6 there are presented the results for the (100) and (001) planes, respectively. The measurements carried out above the Curie point (+24 °C), at the temperature of 26 °C are plotted by points and crosses. The results achieved at the ferroelectric phases, at the temperature of 22.5 °C, are denoted by the other crosses.

At the temperature of 26 °C the crystal of the R. S. is not in the ferroelectric phase. For the (100) plane (Fig. 5) there are observed five modes with different

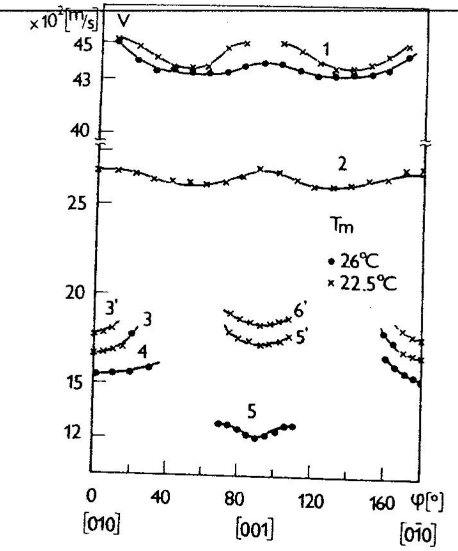

Fig. 5. The SAW velocity for the Rochelle Salt crystal versus an angle of propagation for the (100) plane.

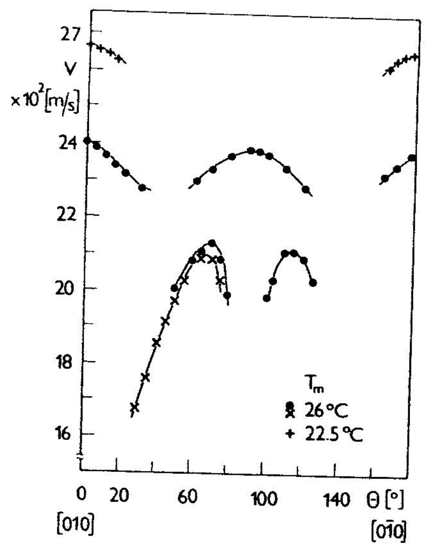

Fig. 6 The SAW velocity for the R. S. crystal as a function of the propagation direction for the (001) plane.

velocities, but only one (mode 2) is observed in full angle range. The modes 3 and 4 were observed only in the vicinity of the $+Y$ and the $-Y$ directions, and the slowest mode 5 was observed in the neighborhood of the Z direction. These three modes are supposed to be Bluestein—Gulayev modes, the mode 2 is supposed to be a pure surface wave of the Rayleigh mode. The quickest mode 1 is supposed to be near the surface volume wave. In the ferroelectric phase, with domain structure, (the measurements were done at the temperature 22.5 °C) the curve 5 disappears and there appear two modes 5′ and 6′ in the vicinity of the Z axis, and all the velocities are higher by about 30—50 m/s.

In the case of the (001) plane, it was experimentally established that all observable modes are comparatively lower in amplitude in comparison with those measured in the plane (100). There was observed one asymmetrical, quickly changing velocity mode, depicted by small crosses. It is supposed that this mode is the pseudo-surface mode. The most striking fact in Fig. 6 is the absence of modes covering continuously the whole range of angles. During the ferroelectric phase, there have been observed modes in the vicinity of the Y axis only, and the velocity was higher by about 250 m/s.

IV. CONCLUSIONS

Comparing the obtained results it is possible to say that the velocities of the SAW change with the change of the domain structure. In the crystal of the R. S., at the ferroelectric phase with domain structure, the velocities of the modes are higher than those at the piezoelectric phase.

In all the investigated planes there are directions in which more than one mode propagates.

From the findings for the R. S. crystals it may be stated that the symmetry of the velocity curves makes it comparatively easy to calculate the elastic and other constants for that material from the experimental data.

This work was supported by the Government Project CPBP 01 08, D 1.4.

REFERENCES

[1] Kotlicka, E., Latuszek, A.: Prace Inst. Fizyki, *31, 32* (1986), 51.

[2] Slobodnik, A. J., Conway, D. E.: *Microwave Acoustic Handbook*, AFCRL-70-0164, 1970.

[3] Hoshino, S., Okaya, Y., Pepinsky, R.: Phys. Rev. *115* (1959), 323.

[4] Aleksandrov, K. S.: *Problemy Sovriemiennoy Kristallografii*, Moskva 1975, 327.

[5] Kotlicka, E.: *Proc. of Surface Physics Conf.*, Łódź, Vol. II, (1987), 217.

[6] Kotlicka, E.: *Proc. of Surface Physics Conf.*, Wrocław Vol. IV, (1988), 106.

[7] Konstantinova, W. P., Silvestrova, J. M., Aleksandrov, K. S.: Kristallografia *69* (1959), 4.

[8] Stankowska, J.: *Badanie Korelacji Pomiędzy Strukturą Domenową a Własnościami Dielekrycznymi Kryształów Grupy TGS*, Poznań 1975.

[9] Pluznikov, W. M., Siemienov, W. S.: *Piezoelektriczeskiye Tviordyie Schemy*, Moskwa 1975.

[10] Jona, F., Shirane, G.: *Ferroelectric Crystals*, Oxford—London—N. Y.—Paris 1962.

Received November 1st, 1988
Accepted for publication March 17th, 1989

ЛОКАЛЬНЫЕ ХАРАКТЕРИСТИКИ РАСПРОСТРАНЕНИЯ ПАВ В TGS И СЕГНЕТОВОЙ СОЛИ

Была измерена угловая зависимость поверхностных акустических волн (ПАВ) для поверхностей (010), (110) и (001) для и для плоскостей (100) и (001) сегнетовой соли для некоторых температур. Сделана попытка объяснить несоответствие эксперименальных и теоретических результатов. Наряду с этим в работе докладывается о наблюдении различных мод в используемых монокристаллах.