Acoustooptic figure of merit for $Sn_2P_2S_6$ crystals and the temperature dependences of acoustic wave velocities

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Received: 25.09.2011

Abstract. We report on the studies for temperature dependences of both longitudinal and transverse acoustic wave velocities in $Sn_2P_2S_6$ crystals. We show that the above temperature dependences, in particular those concerned with the slowest waves, should not impose notable temperature variation of their acoustooptic figure of merit.

Keywords: acoustooptic figure of merit, acoustic wave velocity, ferroelectric phase transition.

PACS: 78.20.hb, 62.65.+k, 77.84.-s
UDC: 535.4, 534.22

1. Introduction

Tin thiohypodiphosphate crystals ($Sn_2P_2S_6$) are wide-gap semiconductors that possess a proper, second-order paraelectric-to-ferroelectric phase transition, with the change of point symmetry $2m \leftrightarrow m$ at $T_C = 337$ K [1]. These crystals are widely known as an excellent photorefractive and electrooptic material [2–5]. In our recent works [6–9] we have studied and analysed acoustic properties of $Sn_2P_2S_6$ crystals at the room temperature, thus demonstrating that the material has great potential for acoustooptic (AO) applications. The acoustooptic figure of merit (AOFM) of $Sn_2P_2S_6$ calculated on the basis of our experimental AO data can reach very high values ($M_2 = (1.7 \pm 0.4) \times 10^{-12}$ s^3/kg [8]). We have also obtained the diffraction efficiency as high as $\eta = (16.0 \pm 3.2)\%$ at the acoustic powers as low as $P_a = 6.7 \times 10^{-4}$ W [8]. Hence, the crystals of $Sn_2P_2S_6$ reveal one of the highest AOFMs known for the AO materials that operate in the visible spectral range (see, e.g., the study of AOFM for Hg_2Br_2 crystals [9]).

It is necessary to note that the AOFM value mentioned above has been obtained under the conditions of AO interaction taking place with the acoustic wave which does not represent the slowest one. Being more precise, we have dealt with the waves having the velocities $v_{21} = 2100$ m/s and $v_{23} = 2610$ m/s (here the first and second indices indicate respectively the directions of the wave vector **q** and the polarisation vector **e** of the acoustic wave) [8].

Let us remind in this respect that the AOFM value is inversely proportional to the cube of acoustic wave velocity and so becomes extremely large with lowering velocity: $M_2 = n^6 p_{ef}^2 / \rho v^3$, where n is the refractive index, p_{ef} the effective elastooptic coefficient, and ρ the crystal density. We have shown in our recent work [10] that it is the transverse wave with the velocity 1550 ± 120 m/s which is the slowest for the $Sn_2P_2S_6$ crystals at the room temperature. The propagation direction for this wave coincides with neither of principal crystallographic directions

but is determined by the directional cosines $\cos\alpha = 0.6678$, $\cos\beta = 0.7193$ and $\cos\gamma = -0.1915$ in crystallographic system, while the polarization is characterised by $\cos\alpha = -0.6914$, $\cos\beta = 0.6947$ and $\cos\gamma = 0.1983$.

One can easily estimate that, in case of AO interaction with this wave, the AOFM can increase four times, when compared with the value obtained in the work [8], if only the effective elastooptic coefficient remained approximately the same. On the other hand, reduction of the acoustic wave velocities occurring with approaching the phase transition temperature T_C, as revealed for some of the longitudinal waves propagating in $Sn_2P_2S_6$ [11], could lead to further increase in the AOFM. As far as we know, temperature dependences of the transverse acoustic wave velocities for $Sn_2P_2S_6$ have not yet been studied at all. That is why the aim of the present work is to study experimentally the temperature behaviour of all of the principal acoustic wave velocities in the $Sn_2P_2S_6$ crystals in the vicinity of the Curie temperature. Here the subject of a primary importance is the parameters of those transverse waves which are responsible for the highly efficient AO interaction, i.e. the velocities v_{21} and v_{23}.

2. Experimental procedures

$Sn_2P_2S_6$ crystals were grown with a vapour-transport technique at the Uzhgorod National University (Ukraine). Orientation of samples with respect to the crystallographic axes was carried out using an X-ray diffraction technique. The lattice parameters of $Sn_2P_2S_6$ are equal to $a = 0.9378\,\text{nm}$, $b = 0.7448\,\text{nm}$, $c = 0.6513\,\text{nm}$, and $\beta = 91.15°$ (at the room temperature and the atmospheric pressure), where the crystallographic axis b is perpendicular to the symmetry mirror plane [12].

Temperature dependences of the velocities of longitudinal and transverse ultrasonic waves were measured with a standard pulse-echo overlap method. The acoustic waves were excited in our samples using $LiNbO_3$ transducers. The resonant frequency of the transducer was $f = 10\,\text{MHz}$, the bandwidth $\Delta f = 0.1\,\text{MHz}$, and the acoustic power $P_a = 1 \div 2\,\text{W}$. When measuring the acoustic wave velocities, we used four single-domain samples with different crystallographic orientations. One of them represented a cube with its faces parallel to [100], [001] and [010] axes, while the other three samples had their faces parallel to the following sets of directions: [110], [110] and [001], [011], [011] and [100], and [101], [101] and [010].

3. Results and discussion

The temperature dependences of the velocities for longitudinal acoustic waves measured by us are shown in Fig. 1. As seen from Fig. 1a, the dependence obtained for the longitudinal wave propagating along the crystallographic axis [001] manifests a weak anomaly in the region of ferroelectric phase transition. At the same time, the velocity of the longitudinal wave propagating along the [010] axis weakly decreases when approaching the Curie temperature from below, then rapidly falls down from 2880 to 2200 m/s just at the Curie temperature, and after that increases up to 3100 m/s in the paraelectric phase (see Fig. 1a). In our recent studies [13, 14], we have shown that such a temperature behaviour of the acoustic wave velocities in $Sn_2P_2S_6$ is caused by a closeness of those crystals to the tricritical point on their (x, T)- and (p, T)-phase diagrams.

The velocities of the longitudinal waves propagating along the [110], [011] and [101] directions have a step-like behaviour at T_C (see Fig. 1b). It is worthwhile to notice that the experimental measurements for the longitudinal waves propagating along the [011] direction become rather complicated in the vicinity of the phase transition point, since the reflected signal

gradually weakens and then disappears. As a consequence, our experiments have not been successful in this temperature region.

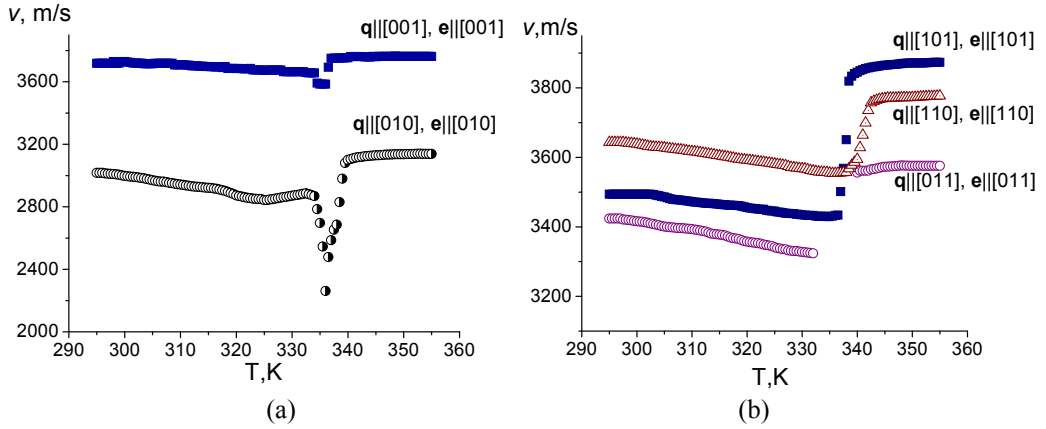

Fig. 1. Temperature dependences of longitudinal acoustic wave velocities for $Sn_2P_2S_6$ crystals.

Let us consider the temperature behaviour typical for the transverse acoustic waves. As seen from Fig. 2a, the transverse waves propagating along the crystallographic direction [010] do not reveal essential changes in their velocities (v_{21} and v_{23}) in the vicinity of the phase transition point. For example, the velocity v_{23} decreases approximately by ~ 150 m/s at T_C with respect to its initial value at the room temperature, while v_{21} reveals only a weak step-like behaviour at T_C. Furthermore, the velocities v_{31} and v_{13} do not change with temperature at all, while the v_{32} parameter shows up only a small (~ 100 m/s) anomaly at T_C (see Fig. 2b).

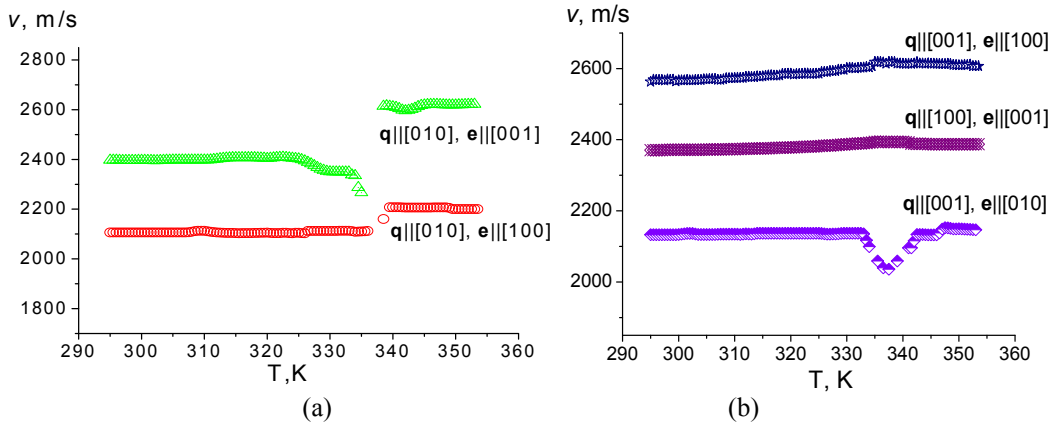

Fig. 2. Temperature dependences of transverse acoustic velocities for the waves propagating along the principal crystallographic directions in $Sn_2P_2S_6$ crystals.

As one can see from Fig. 3, the velocities of the transverse waves propagating along the bisector of principal crystallographic directions are almost temperature independent. Only some of them ($v_{4\bar{4}}$ and $v_{5\bar{5}}$) reveal a kind of step-like behaviour at T_C. What is the most important, the waves which are the slowest one (the velocities $v_{63} \approx v_{4\bar{4}} \approx 1860$ m/s) are temperature independent or reveal a step-like behaviour at T_C. Thus, the temperature dependences of the acoustic velocities (in particular, those typical of the slowest waves, together with v_{23} and v_{21}) should not make any notable effect on the temperature dependences of the corresponding AOFM values for the $Sn_2P_2S_6$ crystals.

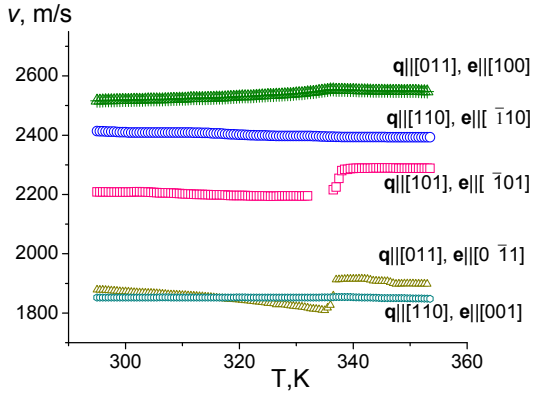

Fig. 3. Temperature dependences of transverse acoustic velocities for the waves propagating along the bisector of principal crystallographic directions in $Sn_2P_2S_6$ crystals.

Nonetheless, the velocities of the longitudinal waves show some anomalies at the Curie temperature and a corresponding increase in the AOFM can indeed occur. However, the mentioned velocities are higher than those peculiar of the transverse waves, and so the relevant AOFMs will be much lower than the values measured in the work [8].

4. Conclusion

In the present work we have studied the temperature dependences of the velocities of both longitudinal and transverse acoustic waves propagating in the $Sn_2P_2S_6$ crystals. It has been shown that the temperature dependences of the acoustic velocities, in particular those of the slowest waves, should not give rise to notable temperature variations of the AOFM values. The velocities v_{21} and v_{23} do not essentially change at T_C, too. Hence, the AOFMs characteristic of AO interaction with these waves should also be almost independent of temperature.

It is also known that the elastooptic coefficients in the ferroelectric materials of such a type as studied here are usually almost temperature-independent in the region of the phase transition. As a matter of fact, only temperature dependences of the refractive indices in $Sn_2P_2S_6$ can be a reason of some temperature variations in the AOFM.

References

1. Vysochanskii Yu M, Janssen T, Currat R, Folk R, Banys J, Grigas J and Samulionis V. Phase transitions in ferroelectric phosphorous chalcogenide crystals: Vilnius University Publishing House (2006).
2. Odoulov S G, Shumelyuk A N, Hellwig U, Rupp R, Grabar A A and Stoyka I M, 1996. Photorefraction in tin hypothiodiphosphate in the near infrared. J. Opt. Soc. Amer. B. **13:** 2352–2360.
3. Jazbinsek M, Montemezzani G, Gunter P, Grabar A A, Stoika I M and Vysochanskii Y M, 2003. Fast near-infrared self-pumped phase conjugation with photorefractive $Sn_2P_2S_6$. J. Opt. Soc. Amer. B. **20:** 1241–1246.
4. Vlokh R O, Vysochanskii Yu M, Grabar A A, Kityk A V and Slivka V Yu, 1991. Electrooptic effect in $Sn_2P_2S_6$ ferroelectrics. Izv. Akad. Nauk USSR, Ser. Neorg. Mater. **27:** 689–692.
5. Haertle D, Caimi G, Haldi A, Montemezzani G, Günter P, Grabar A A, Stoika I M and Vysochanskii Yu M, 2003. Electrooptical properties of $Sn_2P_2S_6$. Opt. Commun. **215:** 333–343.

6. Mys O, Martynyuk-Lototska I, Grabar A, Vysochanskii Yu and Vlokh R, 2006. Piezooptic coefficients and acoustic wave velocities in $Sn_2P_2S_6$ crystals. Ukr. J. Phys. Opt. **7:** 124–128.
7. Mys O G, Martynyuk-Lototska I Yu, Grabar A A, Vysochanskii Yu M and Vlokh R O, 2007. Piezooptic coefficients of $Sn_2P_2S_6$ crystals. Ferroelectrics. **352:** 171–175.
8. Martynyuk-Lototska I Yu, Mys O G, Grabar A A, Stoika I M, Vysochanskii Yu M and Vlokh R O, 2008. Highly efficient acousto-optic diffraction in $Sn_2P_2S_6$ crystals. Appl. Opt. **47**: 52–55.
9. Gottlieb M, Isaacs T J, Feichtner J D and Roland G W, 1974. Acousto-optic properties of some chalcogenide crystals. J. Appl. Phys. **45**: 5145–5151.
10. Mys O, Martynyuk-Lototska I, Grabar A and Vlokh R, 2009. Acoustic and elastic properties of $Sn_2P_2S_6$ crystals. J. Phys.: Condens. Matter. **21:** 265401–265407.
11. Samulionis V, 2002. Acoustic and piezoelectric properties of new polar semiconductor compounds of $Sn_2P_2S_6$ family. Ultragarsas. **4(45)**: 7–12.
12. Carpentier C D and Nitsche R, 1974. Vapour growth and crystal data of the thio(seleno) hypodiphosphates $Sn_2P_2S_6$, $Sn_2P_2Se_6$, $Pb_2P_2S_6$, $Pb_2P_2Se_6$ and their mixed crystals. Mater. Res. Bull. **9:** 401–410.
13. Martynyuk-Lototska I, Mys O, Zapeka B and Vlokh R, 2011. About the existence of a Lifshitz point on the phase diagram of $Sn_2P_2(Se_xS_{1-x})_6$ solid solutions: acoustic and optical studies. Phil. Mag. **91**: 3519–3546.
14. Martynyuk-Lototska I, Say A, Mys O and Vlokh R, 2011. On the tricritical point on the (p, T)-phase diagram of $Sn_2P_2S_6$ crystals. Phil. Mag. doi: 10.1080/14786435.2011.613413.

Mys O., Martynyuk-Lototska I. and Vlokh R., 2011. Acoustooptic figure of merit for $Sn_2P_2S_6$ crystals and the temperature dependences of acoustic wave velocities. Ukr.J.Phys.Opt. **12**: 166-170.

Анотація. *В роботі наведені результати дослідження температурних залежностей швидкостей поздовжніх і поперечних акустичних хвиль в кристалах $Sn_2P_2S_6$. Показано, що температурна поведінка швидкостей акустичних хвиль, зокрема - найповільніших, не суттєво впливатиме на температурну поведінку коефіцієнта акустооптичної якості даних кристалів.*