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THE ANOMALOUS ELASTIC ANISOTROPY OF $Li_2B_4O_7$ AND ITS INFLUENCE ON SAW PROPERTIES

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Abstract

The anomalous elastic anisotropy of $Li_2B_4O_7$ caused by the "incorrect" relation between two elastic modula, c_{33} and c_{44}, is proved to be responsible for the existence of earlier found undamped longitudinal horizontally polarized (LH) type leaky waves. A simple provement is presented to show that "ray-polarized" quasilongitudinal bulk wave propagating in the plane of reflectional symmetry satisfies the stress-free conditions when the boundary plane is parallel to polarization vector and orthogonal to the plane of reflectional symmetry. The close relation between the particular directions, such as acoustic axes, and the existence of "non-leaky" waves is analyzed using the "exceptional wave line" method. LH type leaky waves exist in crystal cuts with Euler angles (φ,θ,90), where φ is arbitrary when θ=40...46°. If φ=45° the permitted interval of θ angle is the largest: θ=35-68°.

1.Introduction

The acoustic anisotropy of any crystal which means the anisotropy of bulk acoustic wave (BAW) velocities in infinite medium plays a very important role in the investigation of surface acoustic waves (SAW). The geometrical image of acoustic anisotropy is the refraction (or slowness) surface. While analyzing this characteristic surface one can predict the behavior of SAW in selected crystal cuts, for example, the splitting of general SAW solution into two separate waves with different velocities and mechanical displacement (polarization) vectors or the existence of SAW with "supersonic" velocity in relation to the slowest bulk mode.

The analysis of acoustic anisotropy should include the search for particular directions [1]: longitudinal and transverse normals and acoustic axes. While propagating along two first particular directions one of three bulk modes is pure longitudinal or pure transverse respectively. If propagation direction is parallel to one of acoustic axes, at least two bulk modes have the same velocity and thus degenerate. The last type of particular directions in crystals of middle symmetry systems - trigonal, tetragonal and hexagonal - is usually localized in the planes of reflectional symmetry or the planes orthogonal to even-fold symmetry axes. The bulk waves, propagating along 3-,4- and 6-fold symmetry axes also degenerate. However in some crystals the acoustic axes of "general position" can exist. The example is quartz, a crystal with very strong acoustic anisotropy.

An infinite number of bulk modes with different polarization vectors can propagate parallel to the acoustic axis and if the last one is of conical type [2], the Poynting vector strikes a cone while propagation direction rotates around the axis. It is obvious that all acoustic properties change rapidly in the vicinity of this particular direction. The refraction surface is characterized by the negative curvature in this special area. Thus if the acoustic axis occurs to be in the sagittal plane for the specified SAW orientation it leads to the appearance of additional "limiting bulk waves"[3], propagating parallel to the surface. The corresponding surface skimming bulk waves (SSBW) can be observed experimentally.

Usually only slowest and middle bulk modes degenerate but some exclusions are known, so called "longitudinal acoustic axes". For example, in parathellurite TeO_2 (symmetry 422) in the plane (100) the velocities of the fast and the middle bulk waves coinside. As a result there is a strong deviation of power flow vector from the propagation direction for quasilongitudinal wave. In (001) plane the angle between these two vectors exceeds 73°. The other important result is the existence of crystallographic orientations in TeO_2, where the quasilongitudinal bulk wave satisfies the stress-free mechanical boundary conditions. This phenomenon was discovered first in [4]. The other example of crystal with anomalous elastic anisotropy is $Li_2B_4O_7$.

2.Elastic anisotropy of $Li_2B_4O_7$ and ray-polarized waves

The crystal of lithium tetraborate $Li_2B_4O_7$ (symmetry 4mm) is one of the most promising new piezoelectric materials for SAW devices due to the high piezoelectric coupling and the existence of temperature compensated cuts for SAW [5-6]. The crystal is already used in SAW devices [7]. Though currently there are many publications devoted to theoretical and experimental investigation of $Li_2B_4O_7$ one important subject, to author's best knowledge, was not discussed. It is the anomalous elastic anisotropy of this material and its influence on SAW and leaky wave properties.

Despite of some difference in the values of material constants of lithium tetraborate obtained by different researchers [8] the elastic stiffness module c_{33} is usually smaller than c_{44}. It is known that if piezoelectric coupling is ignored the longitudinal BAW propagates along Z axis of tetragonal crystal with velocity $V_l = \sqrt{c_{33}/\rho}$ and all degenerated shear waves polarized in XY-plane have velocity values $V_s = \sqrt{c_{44}/\rho}$, where ρ is a mass density. Usually c_{33} is larger than c_{44} and the longitudinal wave is faster than shear ones. The anomalous relation between these constants leads to the degeneration of the bulk acoustic mode with the highest velocity.

Fig.1a,b shows the cross section of the refraction surface by the plane (100) calculated without (fig.1a) and with (fig.1b) piezoelectric effect. Because of symmetry one bulk mode, pure shear, is polarized orthogonal to (100) plane. Two other bulk modes - quasishear and quasilongitudinal - have polarization vectors within this plane. There is an acoustic axis at an angle 69° to Z-axis. It is of ordinary type: the slowest and the middle bulk modes degenerate. One more acoustic axis is parallel to [001] direction (4-fold symmetry axis). If piezoelectric effect is ignored (fig.1a) then two internal (fast and middle) branches degenerate. Since both degenerated modes have polarization vectors in the plane (001), they should be pure shear, while one of them should be pure longitudinal when propagation direction is parallel to [010]. The third non-degenerated mode is pure longitudinal for [001] and pure shear for [010] direction. It is obvious that these bulk modes must have polarization angles varying in the interval 0...90°.

If piezoelectric effect is taken into account the situation changes (fig.1b). The anomalous acoustic axis is transformed to the ordinary one. However the strong anisotropy can be observed for the quasilongitudinal wave and the power flow angle exceeds 32°. The material constants of $Li_2B_4O_7$ from [6] were used for calculations.

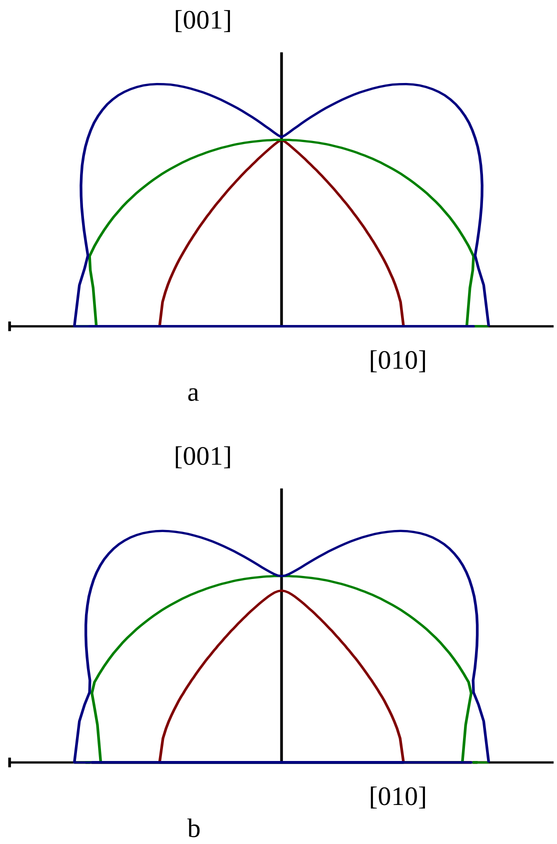

Figure 1. Cross-section of the refraction surface for $Li_2B_4O_7$ without (a) and with (b) piezoelectric effect.

Such behavior is typical for any crystallographic plane parallel to [001] direction. Fig.2 shows the polarization angle β and the power flow angle ϕ for the bulk mode with the highest velocity in (110) plane of $Li_2B_4O_7$ as functions of propagation angle φ to [110] direction. Piezoelectric effect is ignored. The angle ϕ reaches the value 36.7° when the propagation direction is close to Z axis ($\varphi \approx 87°$) being zero for φ=0, 46 and 90°. Zero values of ϕ correspond to longitudinal normals (one of bulk modes has β=0).

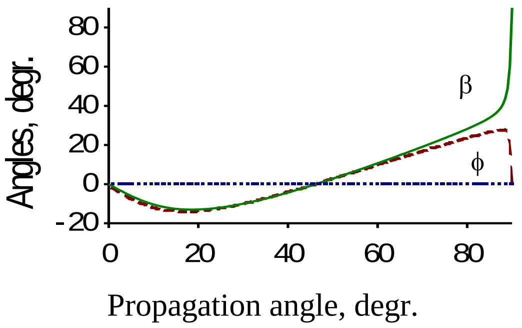

Figure 2. Polarization angle β and power flow angle ϕ of quasilongitudinal wave as functions of propagation angle related to [110] direction in the plane (110).

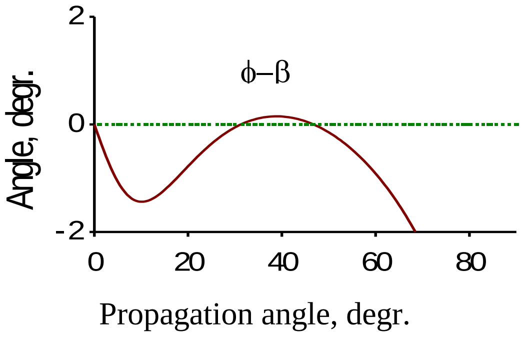

Figure 3. The difference (ϕ–β) as function of propagation directionof quasilongitudinal bulk wave in (110) plane.

In fig.3 the difference between two angles (ϕ–β) is shown for the same wave. It is important that in (110) plane the directions could be found where these two angles are equal and the power flow vector is parallel to polarization.

If piezoelectric effect is ignored, the wave equation for bulk acoustic waves is [1]

$$c_{ijkl}\, n_j\, n_k\, u_l = \rho V^2 u_i \qquad (1)$$

where **n** is a unit wave normal, **u** is a polarization vector, ρ is a mass density and c_{ijkl} is an elastic stiffness tensor. Equation (1) multiplied by vector **m** which is orthogonal to **u** and parallel to the plane (100) yields

$$c_{ijkl}\, n_j\, n_k\, u_l\, m_i = 0 \qquad (2)$$

For quasilongitudinal waves polarized in (110) plane with polarization angle β one can write

$$\mathbf{n} = \cos\beta\, \mathbf{u} + \sin\beta\, \mathbf{m} \qquad (3)$$

After substitution of (3) the equation (2) changes to

$$\cos\beta\, c_{ijkl} n_k u_l u_i m_j + \sin\beta\, c_{ijkl} n_k u_l m_i m_j = 0 \qquad (4)$$

The first term in (4) with account of expression for group velocity [1]

$$V^g_i = c_{ijkl}\, u_j\, u_k\, n_l \qquad (5)$$

and the orthogonality relation for $\mathbf{V^g}$ and **m** gives

$$\mathbf{V^g}\, \mathbf{m} = 0 \qquad (6)$$

Then the equation (4) is satisfied if sinβ=0 or

$$c_{ijkl} n_k u_l m_i m_j = 0, \qquad (7)$$

which can be written as

$$\mathbf{T}\, \mathbf{m} = 0, \qquad (8)$$

where

$$T_i = c_{ijkl} n_k u_l m_j \qquad (9)$$

are the normal stresses caused by the propagation of the bulk wave with unit wave vector **n** along the boundary surface of half-infinite medium normal to **m**. Since the analyzed quasilongitudinal wave is polarized in the plane of reflectional symmetry (110), vector **T** is localized in the same plane. It can be easily shown that this vector is orthogonal to **u**, since the equation (9), multiplied by **u** with account of (5) yields

$$\mathbf{T}\, \mathbf{u} = \mathbf{V^g}\, \mathbf{m} = 0. \qquad (10)$$

Then vector **T** should be parallel to **m** and from (8) follows that **T**=0.

It means that the quasilongitudinal (but not pure longitudinal) wave, propagating in the plane of reflectional symmetry and polarized in the same plane does not disturb the free surface with normal **m** lying within the same crystal plane, if its power flow (group velocity) vector is parallel to polarization direction. This theoreme was proved for general case in [9] and such waves were called “ray-polarized”.

It is important to mention that, though the wave can be quasishear as well as quasilongitudinal, the power flow angle is usually smaller than 45° and thus the “ray-polarized” waves shoud be quasilongitudinal rather than quasishear.

Since both the polarization and the power flow vectors are normal to **m** and localized in the boundary plane, the corresponding SAW is pure longitudinal and horizontally polarized (LH) wave.

In addition to Z axis there are two points in fig.3, where the difference function vanishes: φ=31 and 46° with Euler angles (45, 41, 90) and (45, 46, 90). While the second point corresponds to the longitudinal normal (β=0), the first one allows the propagation of LH wave if the crystal is non-piezoelectric.

The piezoelectric effect while taken into account usually changes the structure of LH wave, making it quasi-LH, with small contribution of other partial modes. It can be transformed to leaky wave or dissappear as non-physical solution, which was theoretically shown for SH waves in transversely isotropic media [10]. However nearly undamped SSBW with quasilongitudinal polarization usually can be found in these specified crystal cuts.

3.EXCEPTIONAL WAVE LINE ANALYSIS

The collinearity of power flow and polarization vectors is a sufficiant but not necessary condition for bulk wave to satisfy the mechanical boundary conditions. Similar bulk waves, called “exceptional waves”(EW)[3] can be found in non-symmetric crystallographic orientations. The general criterion was established in [2]: the bulk wave is exceptional if:

$$\det(\mu_{ij})=0, \qquad (11)$$

where

$$\mu_{ij} = c_{ijkl}\, n_k\, u_l \qquad (12)$$

Such wave satisfies stress-free boundary conditions only on the selected boundary plane with normal **m** from

$$\mu_{ij} m_j = 0 \qquad (13)$$

EW is always horizontally polarized [2]. Thus to answer the question whether a bulk wave propagating in arbitrary crystallographic direction **n** is exceptional or not one must solve the wave equation (1) to find polarization vectors **u** of three bulk modes and substitute them into equations (11)-(13). If (11) is satisfied for any of bulk waves this one is exceptional. While vector **n** scans the three-dimensional space in crystallographic basis, the one-dimensional space of permitted directions for EW can be found. These directions form the continuous “exceptional wave lines”(EWL) on the stereographic projection of unit wave normal sphere $\mathbf{n}^2=1$ [2]. Their configuration depends on the crystal symmetry and the position of acoustic axes. The analysis of this dependence as well as some numerical results for crystals of different symmetry can be found in [11]. Only three of six analyzed materials - parathellurite TeO_2, lithium tetraborate and quartz - allow the propagation of quasilongitudinal EW, while quasishear one exist in all crystals. In TeO_2 the occurrence of EWL for quasilongitudinal mode is caused by the presence of “longitudinal acoustic axis”, since according to general theory [2,11] EWL must go from one degenerated branch to another in this particular point.

Fig.4a,b shows the stereographic projection of unit wave normal sphere with EWL for $Li_2B_4O_7$ without (fig.4a) and with (fig.4b) piezoelectric effect. Only the part of sphere is plotted with account of crystal symmetry. The acoustic axes are marked by bold points. For the trivial pure SH waves both unit wave vectors **n** and plane normals **m** belong to the planes of reflectional symmetry. These waves are polarized normally to symmetry planes and in the points of conical acoustic axes EWL goes from one degenerated branch to another.

In addition to these “trivial” lines the “non-trivial” EWL have been found for all three bulk modes. Their configuration depends on piezoelectric effect. The permitted unit wave vectors for quasi-longitudinal EW in non-piezoelectric crystal form a cone (fig.4a) with the rotation axis parallel to Z direction. The last direction being the acoustic axis of “tangent” type allows the propagation of infinite number of pure shear EW [12], one of these waves belongs to the branch with the highest velocity. Thus it may be considered as one more cone, degenerated. While piezoelectric effect becomes appreciable both cones are coupled and for the real values of piezoelectric constants in $Li_2B_4O_7$ there are four small cones with the axes in (110) planes (fig.4b). If piezoelectric coupling grows, these four cones disappear. It means that the existence of LH type waves is the result of the remaining elastic anomaly.

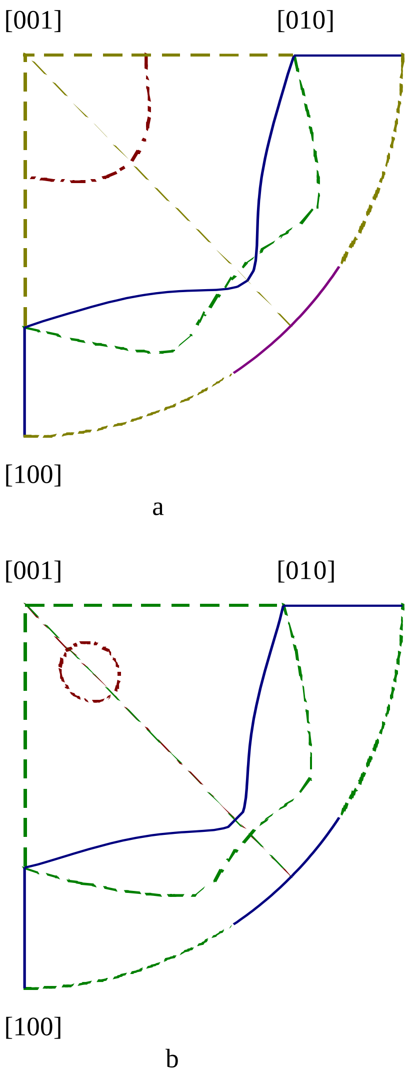

Figure 4. Exceptional wave lines in lithium tetraborate without (a) and with (b) piezoelectric effect: quasilongitudinal waves (dashed-dotted line), fast shear (dashed line) and slow shear (solid line) waves

The intersections of quasilongitudinal EWL with the plane (110) in fig.4b give two cuts with Euler angles (45, 41, 90) and (45, 46, 90).

To analyze the structure of modified EW with account of electric boundary conditions it is necessary to solve the boundary problem by means of some numerical numerical techniques.

4. LH type leaky waves in $Li_2B_4O_7$

The rigorous solution of boundary problem with both mechanical and electric boundary conditions reveales that in lithium tetraborate longitudinal horizontally polarized (LH) type leaky waves with decay coefficients smaller than 0.001 dB/wavelength can propagate in orientations with Euler angles (φ,θ,90), where φ is arbitrary when θ=40...46°.

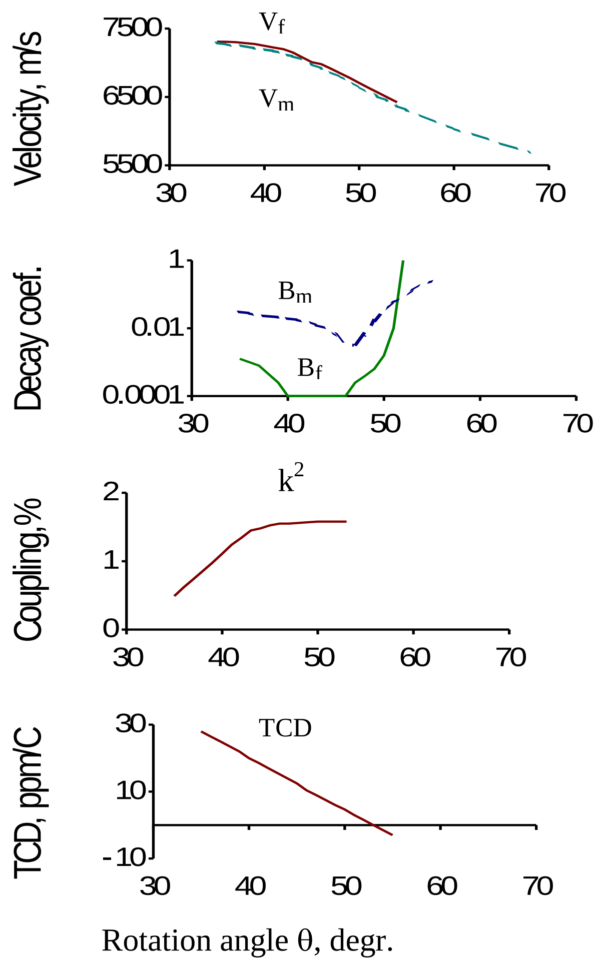

Figure 5. LH type leaky wave characteristics in (45,θ,90) cuts of $Li_2B_4O_7$

However nearly undamped quasi-longitudinal SSBW can be found outside this area. When φ=45° the permitted interval of angle θ for leaky waves is the largest: θ=35...54 for free surface and 35...68° for metallized one.

Leaky waves propagating in orientations with Euler angles (45,θ,90) were analyzed earlier in [13-15]. The velocities are slightly smaller than cut-off velocities of quasi-longitudinal SSBW for the same sagittal planes. The leaky waves are three-partial and include only one homogeneous mode, shear vertical one. SH wave is uncoupled. The predominate quasilongitudinal partial wave is tilted in relation to the boundary surface, the tilt angle is large and reaches 25°.

Fig.5 shows the calculated values of leaky wave velocities V, decay coefficients B, electromechanical coupling coefficients k^2 and temperature coefficients of decay (TKD) for crystal cuts with Euler angles (45,θ,90). The temperature coefficients from [5] were used. Coupling coefficients k^2 were calculated through the velocity difference between free and shorted surfaces. Maximum coupling for LH type leaky waves k^2=2.1%, the velocity V varies within the interval 6424-7308 m/s for free surface and there is a cut, where TCD=0.

It is important to mention here that leaky wave becomes "non-leaky" with zero decay coefficient for two cuts: (45, 41, 90) and (45, 46, 90), where exceptional waves exist. In the interval between these two cuts the wave degenerates into SSBW if the surface is unmetallized. For metallized surface the minimum value of decay coefficient B=0.002 dB/λ is observed for θ=46.5°. The orientation with minimal decay for application in SAW devices. (45, 46, 90), has the following parameters: V_f=6980.2, V_m=6926.1 m/s, k^2=1.6%; B_f=0, B_m=0.003 dB/λ, TCD=10 ppm/°C. For SAW in the same cut V_f=3095.3, V_m=3081.0 m/s, k^2=0.92%.

The effective dielectric permittivity function [16] versus slowness is shown for this cut in fig.6. One can see that only two waves can be radiated: LH leaky wave and SAW. Since both real and imaginary parts of function vanish in the same cut-off point for LH-wave, it means that this wave has zero attenuation on the free surface.

The longitudinal character of leaky wave is proved by the depth dependence of polarization components in (45, 46, 90) cut, shown in fig.6a,b. There is practically one longitudinal polarization, when the surface is free. The vertical shear component is very small, and horizontal shear one is zero because of symmetry. The metallization (fig.6b) causes an energy concentration within 4-5 wavelengths from the surface.

5.Conclusion

The anomalous elastic anisotropy is shown to be responsible for the existence of LH type leaky waves with small and zero decay coefficients. The exceptional wave line analysis can be successfully used to find orientations in any crystal, allowing the propagation of bulk acoustic waves along the free surface. In contrast to numerical techniques widely used for SAW analysis in the specified crystallographic orientation this method reveals the decisive role of crystal symmetry in the existence of SAW solutions with "transonic" velocities and the close relation between particular directions, first of all acoustic axes, and the permitted orientations for leaky and non-leaky waves.

However it should be notices that such anomaly is not necessary. For example, LH type leaky waves have been recently found existing in quartz.

6.References

[1] F.I.Fedorov, The theory of elastic waves in crystals, New York: Plenum Press, 1968.

[2] V.I.Alshits, J.Lothe "Elastic waves in triclinic crystals. III. The problem of existence and some general properties of exceptional surface waves," Sov.Phys. Crystallography, vol.24, pp. 1122-1130.

[3] P.Chadwick, G.D.Smith, "Foundations of the theory of surface waves in anisotropic elastic materials", in Advances in Applied Mechanics, vol.17, New York: Academic Press, p.303,1977.

[4] N.F.Naumenko, N.V.Perelomova, V.S. Bondarenko, "Some peculiarities of bulk and surface acoustic wave propagation in the crystal of parathellurite", Sov.Phys.Solid. Stat., vol.25, pp.2628-2630, 1983.

[5] N.M.Shorrocks,R.W.Whatmore,F.M.Ainger, I.M.Young "Lithium tetraborate - a new temperature compensated piezoelectric material for surface acoustic wave devices", in Proc.IEEE Ultrasonics Symp., 1981, pp.337-340.

[6] M.Adachi, T.Shiozaki, H.Kobayashi, O.Ohnishi, A.Kawabata "Temperature compensated piezoelectric lithium tetraborate crystal for high frequency surface acoustic wave and bulk wave device applications, " in Proc.IEEE Ultrasonics Symp., 1985, pp.228-232.

[7] H.Abe, M.Ohmara, H.Saitou "SAW devices on lithium tetraborate ($Li_2B_4O_7$)", in Proc.1994 IEEE International Frequency Control Symp., 1994, pp.289-295.

[8] J.A.Kosinski, A.Ballato, Lu Yicheng "Pure-mode measurements of dilithium tetraborate material properties," in Proc. 1993 IEEE International Frequency Control Symp., 1993, pp.359-370.

[9] P.Chadwick "A general analysis of transonic states in an anisotropic elastic body", Proc. Roy. Soc. Lond., vol. A401, pp.203-223, 1985.

[10] V.N.Lyubimov, V.I.Alshits, J.Lothe "On the bulk and surface quasibulk waves in half-infinite piezoelectric medium", Sov.Phys. Crystallography, vol.25, pp.33-42, 1980.

[11] V.I.Alshits, V.N.Lyubimov, N.F.Naumenko, N.V.Perelomova, A.L.Shuvalov, 'Exceptional bulk elastic waves in crystals of different symmetry", Sov.Phys. Crystallography, vol.37, pp. 213-219, 1985.

[12] V.I.Alshits, V.N.Lyubimov, A.L.Shuvalov "Exceptional bulk waves in the vicinity of acoustic axes of different type", Sov.Phys. Crystallography, vol.32, pp.828-835, 1987.

[13] N.F.Naumenko "Quasi-horizontally polarized quasi-bulk acoustic waves in piezoelectric crystals", Sov.Phys.Crystallography, vol.37, pp.427-432, 1992.

[14] N.F.Naumenko "Leaky surface acoustic waves with quasi-longitudinal polarization in the crystal of lithim tetraborate", Sov.Phys. Crystallography, vol.32, pp.979-982, 1992.

[15] N.F.Naumenko "Longitudinal horizontally polarized leaky and non-leaky SAW in lithium tetraborate", Phys.Lett., vol.A195, pp.258-262, 1994.

[16] R.F.Milsom, N.C.H.Reilly, M.Redwood "Analysis of generation and detection of surface and bulk acoustic waves by interdigital transducers", IEEE Trans.Sonics Ultrason., vol.SU-24, pp.147-166, 1977.