

PHYSICAL PROPERTIES OF LITHIUM TETRABORATE SINGLE CRYSTALS AT HYDROSTATIC PRESSURE

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The lithium tetraborate $Li_2B_4O_7$ single crystals were produced using the Czochralski method by means of an HX-620 setup. The dielectric and dilatometric studies of the $Li_2B_4O_7$ single crystals were carried out within the 80-400 K temperature range under the high hydrostatic pressure (up to 600 MPa) conditions. The anomalies (at 100 KHz frequency) were observed at $T_1=248$ K being, probably, related to the lithium sublattice disordering. The increase of the hydrostatic pressure results in a linear shift of anomalies in the dependences $\epsilon(T)$ and $tg\delta$ towards the lower temperature region with -58 ± 2 K/GPa coefficient. For the $Li_2B_4O_7$ single crystals the values of the linear compressibility $\chi_{[100]}$ and $\chi_{[001]}$ and their temperature behaviour were determined.

Lithium tetraborate $Li_2B_4O_7$ (LBO) belongs to the 4mm tetragonal symmetry with a polar axis along the crystallographic c axis and is a promising piezoelectric material due to its elastic properties and high coefficient of electromechanical coupling. Besides, in these crystals in the temperature range of 248 K a phase transition to the superionic state is observed, associated with the disordering of lithium sublattice [1].

LBO crystals for our studies were obtained in platinum crucible at ambient air by Czochralski technique using a HX-620 setup with automatic diameter control. The crystal pulling rate was by 3 mm/day with the rotation of 5 rpm, the axial gradient above the melt being 2-3 K/mm. The crystal was grown in [001] direction from the composition with the excess of 0,5 mol.% B_2O_3 . The crystals were annealed for 24 h at 1130 K, the cooling rate was 15 K/h.

Dielectric and dilatometric studies of LBO single crystals in the temperature range from 80 to 400 K were measured under high hydrostatic pressure up to 600 Mpa. ϵ and $tg\delta$ measurements were performed using a conventional technique at 50 KHz using high hydrostatic pressure cell. The pressure in the cell was measured by a manganine manometer within ± 2 MPa. Dilatometric studies were carried out in a three-window optical high-pressure cell, the pressure being transferred by aviation petrol. The LBO compressibility was measured by the interference technique. With

the continuous variation of the pressure in the cell the changes in the laser light interference, passing through a Fabri-Perot interferometer. Between the interferometer mirrors, three pyramids cut of the crystal under investigation, were located, their size determining the distance between the mirrors. In order to eliminate the contribution of the pressure agent refractive index variation into the changes in the interference pattern, the measurements were carried out simultaneously for the sample and for the standard item, made of KBr, whose compressibility has been studied well. The relative error in the determination of LBO compressibility was 5%.

In fig.1,2 temperature dependencies of ϵ and $tg\delta$ of LBO crystal, measured along [001] crystallographic axis, are shown. In the range of temperatures near $T_1=248$ K an anomaly in the dielectric permeability ϵ is observed as a slight maximum as well as a rapid increase of the dielectric loss. No such anomalies of ϵ and $tg\delta$ were observed in [100] crystallographic direction. Such behaviour of ϵ and $tg\delta$ is, probably, related to the lithium sublattice disordering. The increase of the hydrostatic pressure linearly shifts the anomalies in $\epsilon(T)$ and $tg\delta$ plots towards lower temperatures with the coefficient of -58 ± 2 K/GPa, at $T > T_1$ ϵ and $tg\delta$ increase being observed.

In fig.3 pressure dependencies of linear compressibility for LBO crystal, measured along [100] and [001] crystallographic axis.

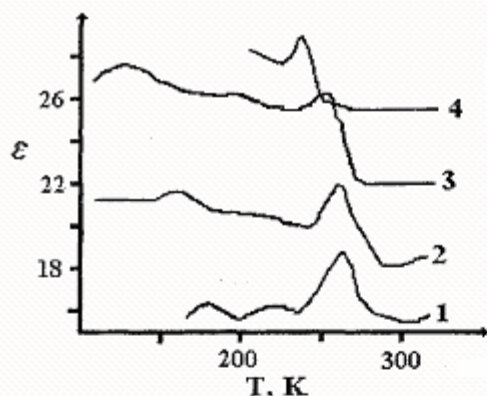


Fig.1. Temperature dependences of dielectric permittivity for *LBO* crystal at different meaning of hydrostatic pressure *P*, GPa: 1 - 0,05; 2 - 0,27; 3 - 0,33; 4 - 0,53

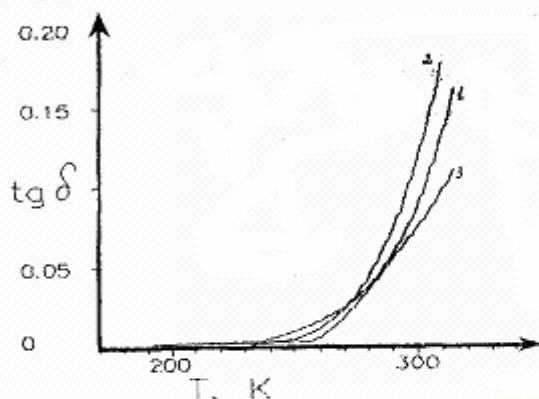


Fig 2. Temperature dependences of dielectric loss tangent for *LBO* crystal at different meaning of hydrostatic pressure *P*, GPa: 1 - 0,08; 2 - 0,33; 3 - 0,53

Linear isothermal compressibility at zero pressure $\chi_{[100]}=0,51 \cdot 10^{-11} \text{ Pa}^{-1}$, $\chi_{[001]}=1,15 \cdot 10^{-11} \text{ Pa}^{-1}$. The hydrostatic pressure increase

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Фізичні властивості монокристалу тетрабората літія в умовах гідростатичного тиску.

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Проведені діелектричні та ділактометричні дослідження кристалів $\text{Li}_2\text{B}_4\text{O}_7$, отриманих методом Чохральського, в широкому інтервалі температур та тисків (до 600 МПа). Виявлено аномалії діелектричної проникності $\epsilon(T)$ та $\text{tg}\delta$ в точці $T=248\text{K}$, які пов'язуються з розупорядкуванням підґратки літію. Зростання гідростатичного тиску приводить до лінійного зсуву аномалії $\epsilon(T)$ та $\text{tg}\delta$ в низькі температури з коефіцієнтом $-58 \pm 2 \text{ K/GPa}$. Для монокристалів $\text{Li}_2\text{B}_4\text{O}_7$ отримані значення лінійної стисливості $\chi_{[100]}$ і $\chi_{[001]}$.

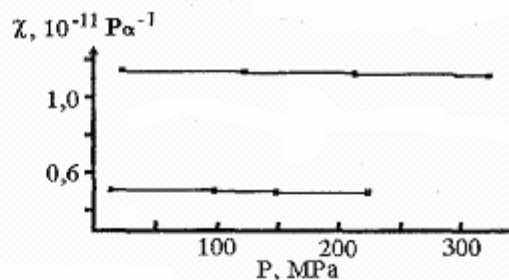


Fig.3. Pressure dependence of linear compressibility of *LBO* crystal along the crystallographic axis: 1 - [001], 2 - [100].

results in the decrease of this values by 1% Kbar^{-1} . *LBO* is seen to reveal a strong anisotropy of its elastic properties indicating the interatomic foreshin this crystal along the fourth-order axis to be much weaker than in the plane perpendicular to this axis. The linear thermal expansion coefficient α in *LBO* along [100] and [001] directions equals to $19,8 \cdot 10^{-6} \text{ K}^{-1}$ and $1,62 \cdot 10^{-6} \text{ K}^{-1}$ respectively [2]. The specific feature of lithium tetraborate is the absence of correlation between α and χ in various directions, since usually in crystals high χ values are observed along the directions with high α values. The measured linear compressibility values are in good agreement with those determined from the elastic compliance modules: $\chi_{[100]} = S_{11} + S_{12} + S_{13} = 0,53 \cdot 10^{-11} \text{ Pa}^{-1}$ and $\chi_{[001]} = S_{33} + 2S_{13} = 1,32 \cdot 10^{-11} \text{ Pa}^{-1}$ [2].