## MORE DETAILS ABOUT LITHIUM TRIBORATE $LIB_3O_5$ (LBO) CRYSTAL

### Part I. Crystallographic facts

LBO is an orthorhombic biaxial crystal with mm2 point group symmetry. Its principal crystallophysical axes x, y, z ($n_z > n_y > n_x$, $n_x > n_y > n_z$) are parallel to the crystallographic axes a, b, c. The calculated angle between the optical axes is equal to 109° at $\lambda$ = 532 nm and to 108° at 635 nm, which defines LBO as a negative biaxial crystal. Its transmission range ranges from 155 to 3200 nm. The linear absorption coefficient of LBO is $3.1\times10^{-3}$ $cm^{-1}$ in the spectral range 351 – 364 nm and $3.5\times10^{-4}$ $cm^{-1}$ at $\lambda$ = 1064 nm.

Dispersion relations - Sellmeier equations for $LiB_3O_5$ are the following:

$n_x^2 = 2.4542 + 0.01125 / (\lambda^2 - 0.01135) – 0.01388\, \lambda^2$

$n_y^2 = 2.5390 + 0.01277 / (\lambda^2 - 0.01189) – 0.01848\, \lambda^2$

$n_z^2 = 2.5865 + 0.01310 / (\lambda^2 - 0.01223) – 0.01861\, \lambda^2$

Typical values of LBO refraction indices are presented in the table below:

| Refractive indices: | $n_x$ | $n_y$ | $n_z$ |
|---|---|---|---|
| 1064 nm | 1.565 | 1.590 | 1.605 |
| 532 nm | 1.578 | 1.607 | 1.621 |
| 355 nm | 1.597 | 1.627 | 1.643 |
| 266 nm | 1.626 | 1.650 | 1.676 |

### Part II. Optical and nonlinear optical properties

The phase-matching directions for three-wave interactions in the LBO crystal are determined for the practical case of light propagation in principal planes of a biaxial crystal. The picture below demonstrates the first octant of the LBO crystal in the crystallophysical coordinate system (x, y, z) with the corresponding dependences of refraction indices on light propagation direction (index surfaces). It is seen that in the xy ($\theta$ = 90°) and xz ($\varphi$ = 0°, $\theta < V_z$) planes LBO is similar to the negative uniaxial crystal and in the yz ($\theta$ = 90°) and xz ($\varphi$ = 0°, $\theta > V_z$) planes to the positive uniaxial one.

Moreover, due to zero values of effective nonlinearity coefficients for some types of three-wave interactions only interactions of Type I are possible in xy and xz ($\theta > V_z$) planes ($o + o \rightarrow e$ and $e + e \rightarrow o$ interactions, correspondingly), whereas interactions of Type II occur only in yz and xz ($\theta < V_z$) planes ($o + e \rightarrow o$, $e + o \rightarrow o$ and $e + o \rightarrow e$, $o + e \rightarrow e$ interactions, correspondingly).

The calculated values of the birefringence - "walk-off" angles for waves with extraordinary polarization propagating inside the LBO crystal in the phase-matching direction corresponding to the generation of the different harmonics of Nd:YAG, Nd:YAP and Ti:Sapphire laser radiation are given in the next table:

| Interacting wavelengths [nm] | Phasematching angle [deg.] | Birefringence angle [deg.] | | |
|---|---|---|---|---|
| xy plane, $\theta = 90°$ $o + o \rightarrow e$ | $\varphi_{theor}$ | $\rho_1$ | $\rho_2$ | $\rho_3$ |
| 1079 → 539.5 | 10.68 | — | — | 0.37 |
| 1064.2 → 532.1 | 11.60 | — | — | 0.40 |
| 886 → 443 | 24.05 | — | — | 0.78 |
| 870 → 435 | 25.36 | — | — | 0.81 |
| 780 → 390 | 33.72 | — | — | 0.98 |
| 760 → 380 | 35.83 | — | — | 1.02 |
| 715 → 357.5 | 41.34 | — | — | 1.07 |

| | | | | |
|---|---|---|---|---|
| 1064 + 532 → 355 | 37.21 | — | — | 1.05 |
| 1064 + 355 → 266 | 60.63 | — | — | 1.01 |
| yz plane, φ = 90° *o* + *e* → *o* | $\theta_{theor}$ | $\rho_1$ | $\rho_2$ | $\rho_3$ |
| 1064.2 → 532.1 | 20.45 | — | 0.35 | — |
| 870 → 435 | 51.79 | — | 0.52 | — |
| 1064 + 532 → 355 | 42.19 | — | 0.53 | — |

The dependences of refraction indices on light propagation direction (index surfaces) in the first octant of LBO in crystallophysical coordinate system (x, y, z). Designations: θ is the polar angle, φ is the azimuthal angle, $V_z$ is the angle between one of the optical axes and the axis z. The similarity of LBO in its principal plane to a positive (+) or negative (-) uniaxial crystal is indicated.

The experimental phase-matching angular (internal $\Delta\varphi^i$, $\Delta\theta^i$ or external $\Delta\varphi^e$, $\Delta\theta^e$, FWHM) and temperature (ΔT, FWHM) bandwidth values in the case of SHG, THG and FHG processes induced in LBO at room temperature by Nd:YAG (λ = 1064 nm), Nd:YAP (λ = 1079 nm) and Ti:Sapphire (λ = 0.71 – 0.89 nm) laser radiation are shown in the next table below.

| Interacting wavelengths [nm] | Angular bandwidth [deg.] | | | Temperature bandwidth [°C] |
|---|---|---|---|---|
| xy plane, θ = 90° *o* + *o* → *e* | $\Delta\varphi^e$ | $\Delta\varphi^i$ | $\Delta\theta^e$ | ΔT |
| 1079 → 539.5 | 0.49 | | — | — |
| 1064.2 → 532.1 | 0.43 | 0.24 | 1.79<br>4.22 | 6.7 |
| 886 → 443 | | | | 7.8 |
| 870 → 435 | | 0.10 | | |
| 780 → 390 | | 0.07 | | |
| 760 → 380 | | | | 15.3 |
| 715 → 357.5 | | 0.06 | | |
| 1064 + 355 → 266 | | | | 3.8 |
| yz plane, φ = 90° *o* + *e* → *o* | $\Delta\theta^e$ | | $\Delta\varphi^e$ | ΔT |
| 1064.2 → 532.1 | 1.20 | | 4.70 | 6.2 |
| 1064 + 532 → 355 | 0.29 | | 4.90 | 3.7 |

The expressions for the effective nonlinearity for an arbitrary direction inside the LBO crystal (mm2 point group symmetry, x, y, z → a, b, c assignment between the crystallophysical and crystallographic coordinate systems) in the principal planes are the following:

xy plane: $$d_{eff}^{ooe} = d_{32}\cos\varphi$$

yz plane: $$d_{eff}^{oeo} = d_{eff}^{eoo} = d_{31}\cos\theta$$

zx plane, θ < $V_z$: $$d_{eff}^{eoe} = d_{eff}^{oee} = d_{32}\sin^2\theta + d_{31}\cos^2\theta$$

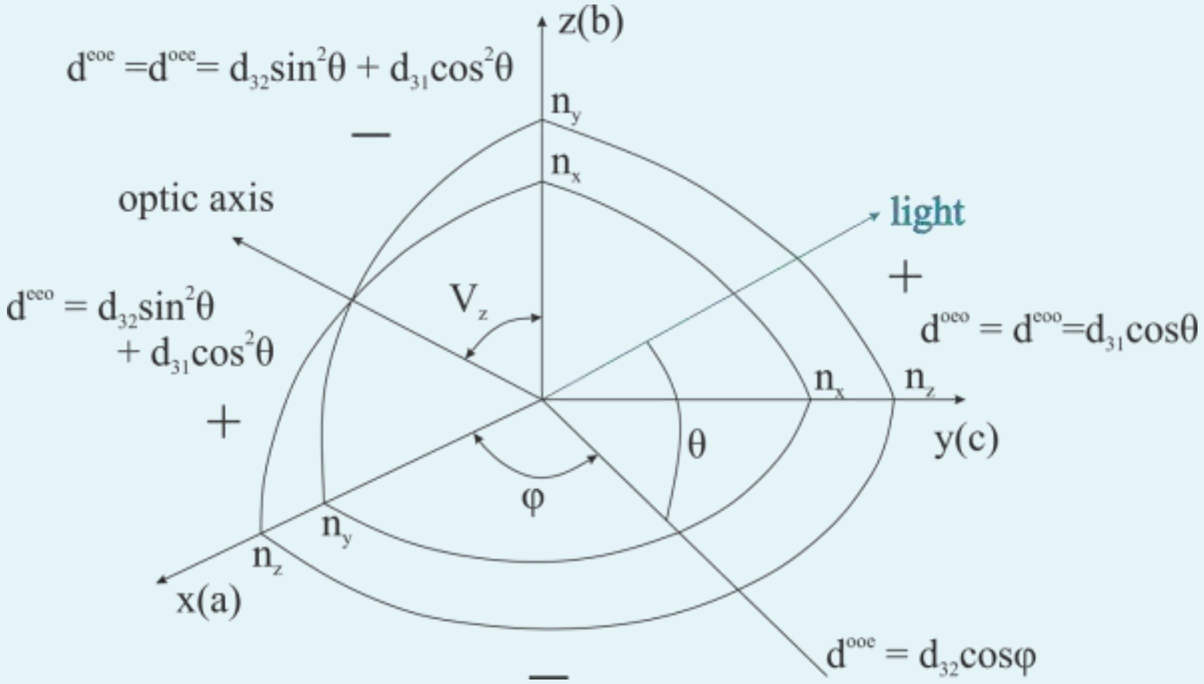
zx plane, θ > $V_z$: $$d_{eff}^{eeo} = d_{32}\sin^2\theta + d_{31}\cos^2\theta$$

where θ and φ are the polar and azimuthal angles in a polar coordinate system related to the crystallophisical coordinate system: θ is measured from z and φ from x (see the picture above).

The effective nonlinearity coefficients $d_{31}$ and $d_{32}$ for LBO with respect to $d_{36}$ (KDP) with the most accurate value of 0.39 pm/V were obtained:

$d_{31} = \pm(1.05 \pm 0.13)\times 10^{-12}$ m/V

$d_{32} = \pm(0.98 \pm 0.09)\times 10^{-12}$ m/V (note that $d_{31}$ and $d_{32}$ are of different sign, this is important for the calculation of $d_{eff}$ in the xz plane).




Calculated values of effective nonlinearity for SHG and SFG processes in the principal planes of LBO crystal:

| Interacting wavelengths [nm] | Phasematching angle [deg.] | Effective nonlinearity [pm/V] |
|---|---|---|
| xy plane, $\theta = 90°$ | $\varphi_{theor}$ | $d_{eff}^{ooe}$ |
| 1908 → 954 | 24.04 | 0.89 |
| 1500 → 750 | 7.03 | 0.97 |
| 1079 → 539.5 | 10.68 | 0.96 |
| 1064.2 → 532.1 | 11.60 | 0.96 |
| 886 → 443 | 24.05 | 0.89 |
| 870 → 435 | 25.36 | 0.88 |
| 780 → 390 | 33.72 | 0.81 |
| 760 → 380 | 35.83 | 0.79 |
| 715 → 357.5 | 41.34 | 0.73 |
| 1064 + 532 → 355 | 37.21 | 0.78 |
| 1064 + 355 → 266 | 60.63 | 0.48 |
| yz plane, $\varphi = 90°$ | $\theta_{theor}$ | $d_{eff}^{eoe}=d_{eff}^{oee}$ |
| 1908 → 954 | 49.00 | 0.69 |
| 1500 → 750 | 14.19 | 1.02 |
| 1908 → 954 | 24.04 | 0.89 |
| 1079 → 539.5 | 18.59 | 1.00 |
| 1064.2 → 532.1 | 20.45 | 0.99 |
| 870 → 435 | 51.79 | 0.65 |
| 1064 +532 → 355 | 42.19 | 0.78 |
| xz plane, $\varphi = 0°$ $\theta < V_z$ | $\theta_{theor}$ | $d_{eff}^{eoe}=d_{eff}^{oee}$ |
| 1318.8 → 659.4 | 5.10 | 1.04 |
| xz plane, $\varphi = 0°$ $\theta > V_z$ | $\theta_{theor}$ | $d_{eff}^{eeo}$ |
| 1318.8 → 659.4 | 86.26 | 0.96 |

LBO is a very useful nonlinear optical material, especially for SHG of high-intensity laser radiation, intracavity SHG, deep-UV SFG and OPO applications.

To compare the nonlinear optical properties of LBO with those of other nonlinear materials such as BBO, KDP and KTP, the attention should be given to the following values: the experimental phase-matching angles for the different interactions, the experimental values of angular and temperature bandwidths (FWHM), the calculated values of birefringence angle and effective nonlinearity in the phase-matching direction. Such a comparison is shown in the table below for SHG of Nd:YAG laser radiation, one of the nonlinear processes frequently used in quantum electronics.

From this table it follows that LBO has a relatively large angular acceptance bandwidth, which permits effective frequency doubling of multi-mode laser radiation. It possesses a rather small temperature acceptance bandwidth and a low birefringence. Concerning effective nonlinearity LBO has nearly the same nonlinearity as BBO but overcomes significantly KDP and compares unfavorably with KTP. On the other hand, LBO exhibits very high resistance to laser damage and its transparency range spreads deep into the UV.

Comparison of Nd:YAG laser frequency SHG made from LBO, BBO, KDP and KTP (1064.2 nm → 532.1 nm).

| Type of interaction | LBO | | BBO | | KDP | | KTP |
|---|---|---|---|---|---|---|---|
| | ooe | eoe | ooe | eoe | ooe | eoe | eoe |
| $\theta_{pm}$ [deg] | 90.0 | 20.5 | 22.8 | 32.7 | 41.0 | 58.0 | 90.0 |
| $\varphi_{pm}$ [deg] | 10.7 | 90.0 | 90.0 | 0.00 | 45.0 | 0.00 | 25.0 |
| $\Delta\theta^{e}_{exp}$ [ang. min] | 25.3 | 7.20 | 3.00 | 4.40 | 9.30 | 18.2 | 19.6 |
| $\Delta\varphi^{e}_{exp}$ [ang. min] | 25.8 | 30.0 | | | | | 63.0 |
| $\Delta T_{exp}$ [°C] | 5.80 | 6.20 | 50.6 | 37.1 | 11.0 | 13.2 | 24.0 |
| $\rho_1$ [deg] | | 0.4 | | 3.8 | | 1.2 | 0.2 |
| $\rho_3$ [deg] | 0.4 | | 3.2 | 4.0 | 1.6 | 1.4 | 0.3 |
| $d_{eff}$ [pm/V] | 0.96 | 0.99 | 2.00 | 1.60 | 0.26 | 0.35 | 3.30 |
| Cutoff of UV transmission | 155 | | 189 | | 177 | | 350 |