Phase-matching and optical properties of LiInS$_2$ nonlinear crystal

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ABSTRACT

Results on investigation of linear and non-linear optical properties, damage threshold and potential efficiencies of biaxial LiInS$_2$ crystal are represented. Transmission range is 0.4 – 12.5 $\mu$m at the 0.1 level and 0.5 – 11.0 $\mu$m at the 0.5 level. Typical absorption coefficients are as low as 0.1-0.25 cm$^{-1}$ at the maximum transmission range 1.0-8.0 $\mu$m and 1.1-2.3 cm$^{-1}$ at the CO$_2$ laser wavelengths. Coefficients of second order non-linear susceptibilities are $d_{31}$=6.2, $d_{32}$=5.4 and $d_{33}$=9.8 pm/V, but damage threshold is as high as 120-130 MW/sm$^2$ for 36 ns pulses at 9.55 $\mu$m. The phase-matching is estimated with using of determined Sellmeier coefficients. It is represented in graphic form so as fields of SHG efficiencies. It is shown that LiInS$_2$ can be used in middle IR OPO pumped by Cu-vapor laser, group-velocity phase matching take place in wide spectral range for sum- and difference-frequency generation of visible, near and middle IR lasers including SHG of 3 $\mu$m Er laser.

Keywords: frequency converters, LiInS$_2$, nonlinear crystal, optical parametric oscillator

1. INTRODUCTION

Despite of a wide range transparency spectrum, a rather high coefficient of second order nonlinear susceptibility and satisfactory birefrigence semiconductor nonlinear biaxial LiInS$_2$ crystals did not attracted attention of the specialists on nonlinear optics. Conceding to oxide crystals of visible and near IR ranges on damage thresholds, and to many known middle IR crystals on value of coefficient of second order nonlinear susceptibility, its can not pretend for a leading position in any part of the spectrum. The difficulties of growing of acceptable size high quality crystals were the additional constraining factor. These factor have not allowed anybody to estimate experimentally advantages or and disadvantages of LiInS$_2$ crystals, which can be given by presence of light Li cations, former time. However, the technological progress$^{4,5}$ stimulate and allow us to study physical properties of that crystals, determine a potential role and place of LiInS$_2$ crystals in nonlinear optics of visible, near and middle IR spectral ranges.

2. LINEAR AND NONLINEAR PROPERTIES

Biaxial negative LiInS$_2$ crystal belongs to mm2 space symmetry group (same as KTP), not gigoscopic, has 3.5 g/cm$^3$ density, 880°C melting temperature, 3-4 hardness on Moos.$^1$ In our researches transparent or slightly yellowish samples of about 4x4x4 mm in size crystals of rather high optical quality, which have been grown by the Bridgman-Stockbarger technique, were investigated. The transparency range was determined with Shimadzu UV 3101PC spectrophotometer and Specord 80M. For example, for 3.6 mm colorless crystal transparency range is determined as 0.4 – 12.5 $\mu$m at 0.1 level, and 0.5 – 11.0 $\mu$m at 0.5 one (Fig.1). At the maximal transparency range 1.0-8.0 $\mu$m coefficient of optical losses $\alpha$ is as high as $\geq0.1-0.25$ cm$^{-1}$, and 1.1-2.3 cm$^{-1}$ at the CO$_2$ laser wavelengths. The short wavelength end of the transmission spectrum of 3.5 mm crystal of comparable structure and quality, as it was previously described$^1$, is determined as 330-334 nm at 80K and 342-343 nm at 300K at $\alpha = 200$ cm$^{-1}$ level. It was at the same position for various polarizations. Long wavelength end can be determined as close to 13.2 $\mu$m at same level of optical losses.

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Fig. 1. Fresnel losses level (1), transparency (2) and absorption coefficient (3) spectra of 3.6 mm LiInS₂.

Dispersion dependence of refraction indices were determined (Fig. 2) by standard method with the help of 5 mm side prisms. Measurement data were fitted by least square method to equations of the Sellmeier type

\[ n^2 = A + \frac{B}{\lambda^2 - C} - D\lambda^2, \]

where \( \lambda \) is wavelength in \( \mu m \). The differences between measured and fitted refractive indices were ±10⁻³ at the final stage of fitting. Sellmeier coefficients estimated for spectral range 0.45-11.5 \( \mu m \) in optical coordinate system of the crystal are listed in Table 1.

Table 1. Sellmeier coefficients of LiInS₂ crystal

<table>
<thead>
<tr>
<th></th>
<th>X</th>
<th>Y</th>
<th>Z</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>4.559534</td>
<td>4.418222</td>
<td>4.59206</td>
</tr>
<tr>
<td>B</td>
<td>0.1403701</td>
<td>0.1254461</td>
<td>0.410887</td>
</tr>
<tr>
<td>C</td>
<td>0.069233</td>
<td>0.0657432</td>
<td>0.069287</td>
</tr>
<tr>
<td>D</td>
<td>0.0028731</td>
<td>0.0028850</td>
<td>0.0030589</td>
</tr>
</tbody>
</table>

Essential distinctions between our data on refraction indexes and measured and described in paper by [1] are not revealed.

Second order nonlinear susceptibility coefficients are determined as \( d_{31} = 6.2 \), \( d_{32} = 5.4 \) and \( d_{33} = 9.8 \) pm/V with accuracy about 15% from comparative measurements of SGH efficiencies in thin wedges (5°) of ZnGeP₂ and LiInS₂ on known procedure with use of pulse-periodic CO₂ laser. The coefficients \( d_{14} = d_{36} \) of basic ZnGeP₂ wedge were accounted as equal to 75 pm/V when estimation. Measured and settlemented phase-matching angle are coincided with accuracy not less than 0.3° at 9.55 \( \mu m \) pump.

Damage threshold was determined for 36 ns 9.55 \( \mu m \) TEM₀₀ mode TEA CO₂ pulses containing about 90% of total pulse energy as 120-130 MW/cm².

3. PHASE-MATCHING

It is comfortable to represent SHG phase-matching of LiInS₂ (so as in all biaxial crystals) in the form of diagrams of phase-matching direction. In Table 2 the number of diagrams of transitions between stereographic projections of phase-matching directions are submitted in accordance with the specified classification. These projections show the angle distribution of phase-matching directions for ssf- (solid curves) and ssf- (dotted curves) interactions. In the figures of this table oz axis is directed up, ox to the left, and oy orthogonal to figure plane.
Table 2. The transitions of SHG phase-matching diagrams

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Type</th>
<th>00-10</th>
<th>10</th>
<th>10-30</th>
<th>30</th>
<th>30-31</th>
<th>31</th>
<th>31-33</th>
<th>33</th>
<th>33-31</th>
<th>31</th>
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<th>30</th>
<th>30-10</th>
<th>10</th>
<th>10-00</th>
<th>00</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiInS$_2$ mm2</td>
<td>ssf</td>
<td>900</td>
<td>1573.5</td>
<td>1731.9</td>
<td>2294.8</td>
<td>2638.3</td>
<td>5104.7</td>
<td>5785.7</td>
<td>7945.2</td>
<td>8498.5</td>
<td>11500</td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>

The view of concrete angle distribution changes from one to other projections in dependence on difference between main indices of the refraction coefficients with laser wavelength variation and dispersive properties of the crystals. The numbers are correspond to transitions from one stereographic projection of a phase-matching to another, when the phase-matching coincides with one of the optical axis. Transition wavelengths are specified below. It is seen, that at no one wavelengths the phase-matching is in the direction of $x$ axis.

In Fig. 3 the dependences of $\lambda_1$ and $\lambda_2$ wavelengths versus $\lambda_3$ wavelength ($\lambda_3^{-1} = \lambda_1^{-1} + \lambda_2^{-1}$) are submitted for which in the direction of fsf- and sff-type phase-matching in the xy plane group velocity phase-matching take place also. Fig. 4 the dependence of phase-matching spectral bandwidth versus pump wavelength is shown at $\varphi=0^\circ$. It is seen that the spectral bandwidth is enough wide for problemless practical use of the LiInS$_2$.

For group velocity phase-matching fulfilling, the crystal length $L$ must be shorter than so called group length $L_g = \tau / |\Delta u^{-1}|$, where $\tau$ is pump pulse duration and

$$\Delta u^{-1} = (1/u_1 - 1/u_2) = 1/c \left[ (n_1 - \lambda_1 \frac{\partial n_1}{\partial \lambda_1}) - (n_2 - \lambda_2 \frac{\partial n_2}{\partial \lambda_2}) \right]$$

is group mismatch, $u_{1,2}$ and $\lambda_{1,2}$ are group velocities of pump and second harmonic emissions, respectively. Estimation result (Fig.5 and 6), show, that frequency conversion (SHG, SFG, DFG) of femtosecond pulses can be realized in a wide wavelength range. In Estimated OPO phase-matching diagrams in xy plane are shown in Fig. 7 and 8.
Both ssf- and fsf- type of interactions can be realized also under $\lambda_3=1.0642$ $\mu$m pump as Fig.9 is demonstrated. Last time SHG was obtained by use picosecond tunable radiation output of free electron laser as the pump in the range from 2.75 to 6.0 $\mu$m. Phase-matching angles determined are in good coincidence with our data. Let us note, in the 25 MHz repetition-rate mode for 0.5 ps pulses any optical damage for peak power density of more than 6 GW/cm$^2$ was not observed. It is very important to calculate the efficient nonlinearity coefficient for concrete type frequency converers, including harmonic generation. The ratio of values and marks of tensor of nonlinear susceptibility coefficients of LiInS$_2$ shows, that for ssf (slow-slow-fast) - type interaction effective nonlinear susceptibility coefficient is differ from zero in xz plane at $\theta < V_z$ ($V_z$ - angle to optical axis). For sff(slow-fast-fast) - and fsf (fast-slow-fast) - types of interactions effective nonlinear susceptibility coefficient is differ from zero in xz plane at $\theta > V_z$ and in xy and yz planes.

The maximal value of effective nonlinear susceptibility take place at second type of interaction in the direction of y axis. In particular, it was found that LiInS$_2$ is unique crystal of known ones for frequency conversion of femtosecond pulses of 3 $\mu$m spectral range erbium lasers with advantages in comparison with other known crystals if account a complete set of it parameters.
The above estimations were carried out for phase-matching directions. But, more information can be obtained by calculating of this value for all possible ranges of the angles $\varphi$ and $\theta$, that corresponds to the SHG efficient nonlinearity fields. Corresponded estimations are represented in Fig. 10 and 11.

This representation allows us to find directions of the maximal efficient nonlinearity. By temperature or by using of applied electrical cw-voltage it is possible to change phase-matching directions up to desirable direction.

It is interesting to note, for biaxial crystals the position of the maximal SHG efficient nonlinearity is determined not only by symmetry group (that is to say by view of the nonlinear tensor, as for single axis crystals), but also by relations between tensors component. Due to this reason, the angle distributions of the efficient nonlinearity will be different for crystals that belong to the one symmetry group.

4. CONCLUSIONS

In summary it is necessary to note, that transparency spectrum, birefrigence and nonlinear properties of single LiInS$_2$ crystals were investigated. It was found that these crystals allow anybody to consider its as perspective crystals for frequency
conversion of femtosecond lasers, including, as unique known for today, for frequency converters of femtosecond 3 μm range erbium laser pulses. Second order nonlinear susceptibility coefficients are about 80% less than known data. Other extraordinary potentialities were not found. All presented in this paper results are carried out with use of software complex LID-SHG (www.bmstu.ru/~lid).

REFERENCES