

Absorptance and scattering losses measurements of the mid-infrared nonlinear crystals LiInSe₂ and LiInS₂ in the IR range

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Abstract

We report standardized absorption and scattering losses measurements of the nonlinear crystals LiInSe₂ and LiInS₂ in IR range by high average power 1064 nm radiation and tunable radiation of optical parametric oscillator (OPO) based on a periodically poled lithium niobate (PPLN) pumped by a diode-pumped, Q-switched TEM₀₀ mode Nd:YVO₄ laser operated at 1064 nm.

Keywords: optical characterization, absorption and scattering measurements, LiInS₂, LiInSe₂, PPLN, OPO.

Introduction

The optical absorptance of nonlinear crystals is an important factor in determining their utility for harmonic generation, optical parametric oscillation and electro-optic switching. Experiments indicate that these devices work well at average-power levels below 1 W. However, as the average-power level increases, absorption of one or more of the interacting waves in the nonlinear or electro-optical crystal can lead to an appreciable thermal load. As a result the crystal may develop a temperature distribution that depends on such factors as crystal shape, thermal conductivity, heat-removal geometry and beam properties. This temperature profile causes dephasing that decreases parametric gain and conversion efficiency. Stresses created by the thermal load cause birefringence and can also lead to fracture. Clearly one of these effects will limit the average-power scaling of these crystals.

For the absorptance measurements Lithium Indium Sulfide (LiInS₂; LIS) and Lithium Selenoindate (LiInSe₂ or LISe) were chosen. Non-linear characteristics of these crystals are close to AgGaS₂ and AgGaSe₂, but their crystal structures are different. Lithium indium sulfide belongs to the A¹B^{III}C₂^{VI} chalcogenide family. It is a biaxial crystal with *mm*2 symmetry, like the well-known KTP and its isotopes. It is optically transparent from 0.4 to 12 μm. The linear and nonlinear optical properties of LIS were briefly studied by Boyd *et al.* in the early 1970s [1]. Recently, renewed interest has been paid to LIS ([2,3]) because of its attractive optical properties, such as high damage threshold, large thermal conductivity, large energy gap, and relatively high nonlinear susceptibility.

The spectral transparency range and the high birefringence of the biaxial Lithium Selenoindate nonlinear optical crystals allow one to realize the SHG of all widely used mid-IR lasers. Their efficiency is identical to that of the AgGaS₂ crystals and twice as high as the efficiency of LiInS₂ crystals. The advantage of the LiInSe₂ crystals is the possibility of creating mid-IR parametric light oscillators pumped by radiation of near-IR solid-state

lasers, in particular, Nd:YAG lasers, with more than doubled efficiency as compared to that of AgGaS₂ and LiInS₂ crystals used [4]. The new physical parameters that are relevant for laser and nonlinear optics applications, such as thermal expansion and thermo-optic coefficients, lattice phonon vibrational spectra, piezoelectric and electro-optic coefficients, accurate optical linear dispersion and nonlinear second-order susceptibility coefficients have recently been reviewed [5].

In this paper we report the absorption and scattering losses measurements of the nonlinear crystals LiInSe₂ and LiInS₂ in the near infrared from 1.0 to 1.9 μm by the master oscillator radiation and high average power tunable radiation of optical parametric oscillator (OPO) based on a periodically poled lithium niobate (PPLN).

Set-up of the PPLN OPO

Singly resonant pulsed OPO's based on periodically poled lithium niobate are easily operated at low pulse energies [6-7] suitable for transmission, reflection and polarisation measurements. In the case of high repetition rate and high conversion efficiency such OPO's also are able to produce average power suitable for absorption measurements of optical components used in lasers and nonlinear optical devices. When such OPO's are used to perform the characterisation of crystals and optical components for lasers applications and nonlinear optical devices the most important parameters become high conversion efficiency and high output pulse stability at high repetition rate. As a master oscillator for the PPLN OPO is used diode-pumped Q-switched nanosecond Nd:YVO₄ laser (EKSPLA product NL640) with output power up to 7 W at 1064 nm and repetition rate up to 40 kHz. The highest energy per pulse is ~0.6 mJ getting lower at higher repetition rates. To avoid the laser-induced damage of the PPLN crystal, we have chosen the OPO pump energy to be ~0.5 mJ at the PPLN structure. For this experiment was optimally taken 5 kHz pump pulse repetition rate.

The multi-grating PPLN sample was fabricated by electric-field poling in a 1-mm-thick, 17-mm-long wafer. Seven poling regions with aperture of 1x1 mm² separated by 0.1-mm gaps and with poling periods between 29-32 μm stepped in 0.5- μm increments were produced on the wafer. The crystal was mounted in an oven held at an elevated temperature of 105 °C to avoid photorefractive distortion. Translating the crystal across the cavity allowed pump-beam interaction with the different grating regions and provided wavelength tuning in the range 1.5-3.7 μm with pulse energy of approximately 30 μJ at repetition rates up to 5 kHz. The radiation of the PPLN OPO and the master laser radiation were used for scattering and laser calorimetry measurements.

Absorptance measurements

Measurements of the linear absorption losses were performed according to the ISO11551 standard [8] by the “pulse” or “gradient” calorimetric method at different wavelengths. In these procedures the absorptance is determined calorimetrically as the ratio of energy absorbed by the crystal to the total energy impinging upon the crystal. This method is based on the measurement of the temperature of the sample during a heating and subsequent cooling cycle. There were available the 1064-nm pulsed (~7 ns) laser output at 5 kHz repetition rate and up to 3 W average power in a linearly polarized TEM₀₀ beam and PPLN OPO output at up to 0.1 W average power. The experimental setup of the laser calorimeter is illustrated in Fig. 1.

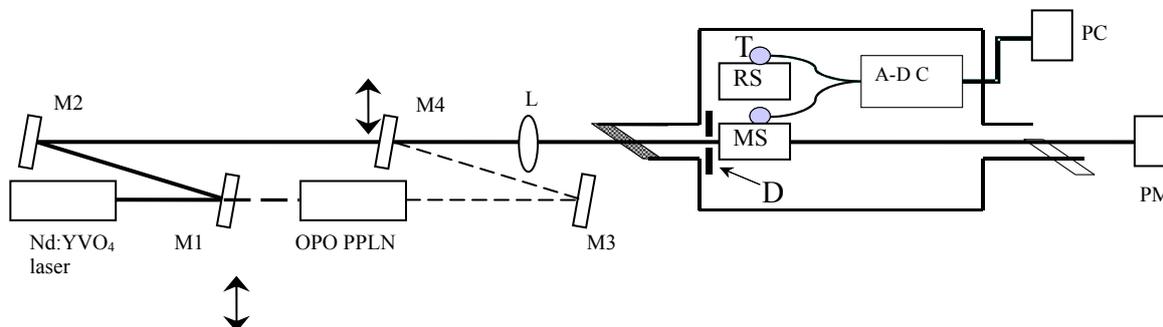


Fig. 1. Optical layout of the laser calorimeter: M1-M2- high- reflectivity mirrors at 1064 nm; M3-M4- high- reflectivity mirrors at 1500-1900 nm; PM-calibrated power meter; T- thermistors; L- lens; RS, MS- reference and measured samples; D- aperture; A-DC- analog digital converter; PC- computer.

Two high-reflectivity mirrors direct the beam into a thermally isolated chamber in which the measured (MS) and reference (RS) samples are mounted. The laser radiation is incident on only the measured sample; the reference sample is used to record temperature changes in the chamber. A good thermal contact between the thermistors and test or reference samples was achieved by placing the thermistors on special holders. The thermistor reading is recorded using an analog-to-digital converter (A-DC) and a computer (PC). A diaphragm is placed in the beam path in front of the test sample to ensure that the measuring beam irradiates only the central part of the test sample and that reflected or stray radiation will not strike the holder and thermistor. Prior to the examination of the component to be tested, a measurement with an empty

holder is performed to verify that the measuring arrangement is not influenced by reflected or stray radiation.

In order to speed up the measurements of the absorptance the software based on the programming package LabView was created. The programme saves the measurement data and performs the required extrapolations. After completion of measurement the programme automatically generates the measurement report document. The duration of irradiation was selected in range of 180 s, according to the requirements of the ISO standard. We calibrated the equipment by measuring materials with known absorptance (colour glass filters and blackened Al samples).

The absorption of LiInSe₂, LiInS₂ was measured in the range from 1064 to 1900 nm. The crystals were grown using the Bridgman-Stockbarger technique [5] in Institute of Mineralogy and Petrography Institute, Russia.

For the measurements used LiInSe₂ brownish crystal was 6x7 mm in face and 8.7 mm long parallelogram. The dimensions of also parallelogram shaped LiInS₂ crystal are 5x5 mm² area and 10 mm length.

Using PPLN OPO output radiation the absorptance values of the LiInSe₂ and LiInS₂ crystals were measured at different signal wavelengths for *e* and *o* polarizations (Fig.2). The absorption of LiInSe₂ is higher than of LiInS₂ in all spectral region of investigation. It has a maximum value (6.5 %cm⁻¹) reaching at 1064 nm (measured by master laser radiation).

The absorption of LiInS₂ is quite smaller at this wavelength exceeding 2.5 %cm⁻¹. The difference of the

absorption of radiation in *o* or *e* polarizations is more evidence for LiInS₂. It is important to note that the average output power of the PPLN OPO at the longer wavelengths is getting lower (from up to 50 mW at the 1700 nm to 8-10 mW at 1950 nm) and this could influence the accuracy of the measurements considerably.

Scattering measurements

The assessment of surface roughness is possible by scatter measurements. A standard procedure based on total integrated scattering has been developed for this purpose in the characterization of laser components. In this standard procedure, the power of radiation scattered by the specimen is collected and integrated by a Ulbricht-sphere or Coblentz-

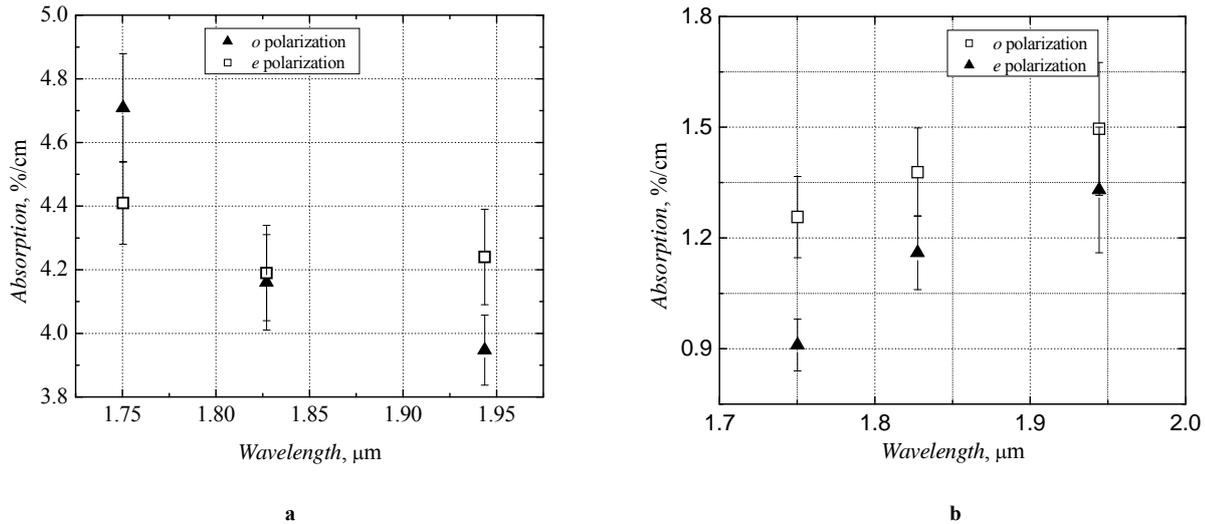


Fig. 2. The average linear absorption of the LiInSe₂ (a) and LiInS₂ (b) crystals at different wavelengths (for *e* and *o* polarization).

hemisphere [9]. Our total scattering measurement facility is equipped with an Ulbricht sphere (Fig. 3). The sensitivity of the facility was determined by measurements with the unloaded sphere. The background level, which arises due to Rayleigh scattering in the air and the diffraction on by the elements used in system, was 10^{-5} . For the determination of the total backward scatter values, the specimen is attached to the exit port of the sphere. For the measurement of forward scatter signal, the procedure is repeated with the specimen located at the entrance port. In the experiment 16-bit-resolution analogue-digital converter (ADC) was used. The experiment was controlled by the computer.

In order to control total scattering measurements a procedure based on the programming package LabView was created.

The scattering measurements were performed for the same LiInS₂ and LiInSe₂ crystals using 1064 nm laser radiation. The laser beam energy was 450 μJ per pulse at a beam diameter of 150 μm to avoid the damage of the optical components. The scattering maps of the crystals are presented in the Fig. 4. The bigger scattering losses on the side of the samples are due the diffraction from sample holder. Scattering losses for LiInSe₂ crystal is 6.8 %. LiInS₂ crystal has 2.5 % total scattering losses. From the measured scattering results it can be seen that there are no intense scattering centers on these samples what was proven by investigation of these samples by microscope. The influence of the wavelength of the radiation on the scattering was compatible with the transmission spectra of these samples measured by the spectrometer.

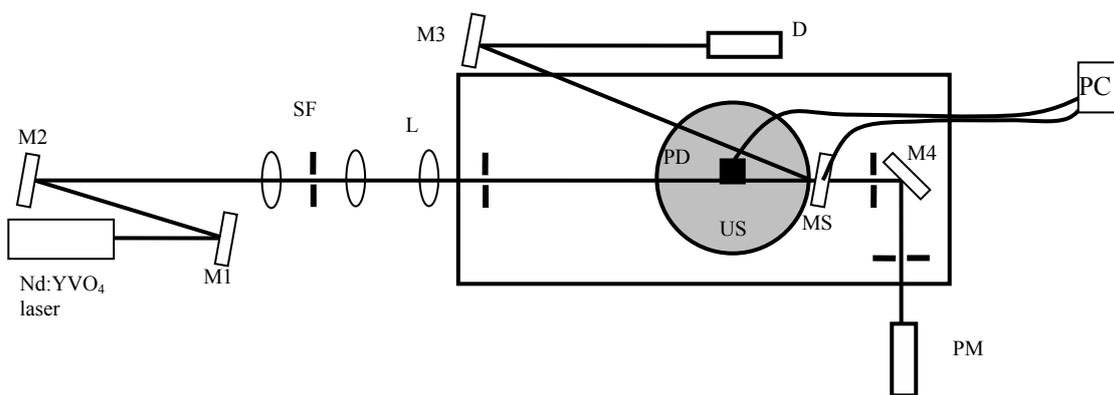


Fig. 3. Experimental set-up based on an Ulbricht sphere (US) for total scatter measurements. M1-M4 – mirrors; D –aperture, PD-photodiode, SF- spatial filter, L –lens, PC- computer, PM – power meter, D- dump

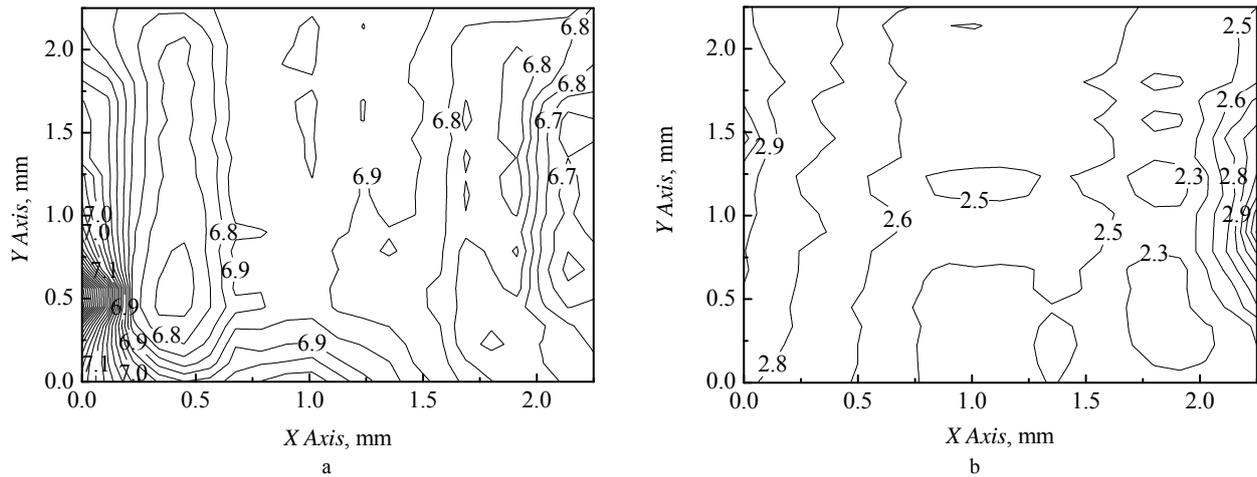


Fig. 4: Scattering losses (in %) of the LiInSe_2 (a) and LiInS_2 (b) crystals at various surface positions using $1.064 \mu\text{m}$ radiation.

Conclusions

In conclusion, we have performed standardized absorption and scattering losses measurements of the nonlinear crystals LiInSe_2 and LiInS_2 in the near infrared range. The absorbance was measured in the range 1.5-1.9 μm by high average power tunable radiation of optical parametric oscillator based on a periodically poled lithium niobate. Average linear absorption values were estimated for different polarization of the crystals. The scattering losses of the crystals was measured at 1064 nm by Nd:YVO_4 laser radiation. Estimated average scattering losses for LiInSe_2 and LiInS_2 were 6.8 and 2.5 % respectively.

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Netiesinių LiInSe_2 ir LiInS_2 kristalų sugerties koeficiento ir sklaidos nuostolių matavimai infraraudonojoje srityje

Reziumė

Pristatomi standartizuoti netiesinių LiInSe_2 ir LiInS_2 kristalų sugerties koeficiento ir sklaidos nuostolių matavimai infraraudonojoje srityje. Šie matavimai atlikti vidutinės galios 1064 nm lazeriu ir derinamos spinduliuotės optinio parametrinio osciliatoriaus ličio niobato lazeriu, kaupinamu moduluotos kokybės TEM_{00} modos Nd:YVO_4 lazeriniu 1064 nm diodu.

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