

Femtosecond mid-IR difference-frequency generation in LiInSe₂

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Abstract: We employ LiInSe₂ nonlinear crystal for difference-frequency generation between signal and idler of a synchronously pumped femtosecond optical parametric oscillator at 80 MHz achieving continuous tuning from 4 μm (16.7 mW) to 11.5 μm (2.3 mW).

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References and links

1. A. Lohner, P. Kruck, and W. W. Rühle, "Generation of 200 femtosecond pulses tunable between 2.5 and 5.5 μm ," *Appl. Phys. B* **59**(1), 211–213 (1994).
2. J. D. Kafka, M. L. Watts, J. W. Pieterse, and R. L. Herbst, "Mid-infrared pulse generation using a sub-picosecond OPO," *Appl. Phys. B* **60**(5), 449–452 (1995).
3. S. Ehret and H. Schneider, "Generation of subpicosecond infrared pulses tunable between 5.2 μm and 18 μm at a repetition rate of 76 MHz," *Appl. Phys. B* **66**(1), 27–30 (1998).
4. M. Beutler, I. Rimke, E. Büttner, V. Panyutin, and V. Petrov, "80-MHz difference-frequency generation of femtosecond pulses in the mid-infrared using GaS_{0.4}Se_{0.6}," *Laser Phys. Lett.* **10**(7), 075406 (2013).
5. J. M. Fraser, D. Wang, A. Haché, G. R. Allan, and H. M. van Driel, "Generation of high-repetition-rate femtosecond pulses from 8 to 18 microm," *Appl. Opt.* **36**(21), 5044–5047 (1997).
6. V. Petrov, J.-J. Zondy, O. Bidault, L. Isaenko, V. Vedenyapin, A. Yelisseyev, W. Chen, A. Tyazhev, S. Lobanov, G. Marchev, and D. Kolker, "Optical, thermal, electrical, damage, and phase-matching properties of lithium selenoindate," *J. Opt. Soc. Am. B* **27**(9), 1902–1927 (2010).
7. G. D. Guseinov and A. I. Rasulov, "Heat conductivity study of GaSe monocrystals," *Phys. Status Solidi* **18**(2), 911–922 (1966).
8. V. Petrov, V. L. Panyutin, A. Tyazhev, G. Marchev, A. I. Zagumennyi, F. Rotermund, F. Noack, K. Miyata, L. D. Iskhakova, and A. F. Zerrouk, "GaS_{0.4}Se_{0.6}: relevant properties and potential for 1064 nm pumped mid-IR OPOs and OPGs operating above 5 μm ," *Laser Phys.* **21**(4), 774–781 (2011).
9. S. Fossier, S. Salaün, J. Mangin, O. Bidault, I. Thenot, J.-J. Zondy, W. Chen, F. Rotermund, V. Petrov, P. Petrov, J. Henningsen, A. Yelisseyev, L. Isaenko, S. Lobanov, O. Balachninaite, G. Sleky, and V. Sirutkaitis, "Optical, vibrational, thermal, electrical, damage and phase-matching properties of lithium thioindate," *J. Opt. Soc. Am. B* **21**(11), 1981–2007 (2004).

1. Introduction

With the development of mode-locked Ti:Sapphire lasers in the 1990-ies synchronously pumped optical parametric oscillators (SPOPOs) became operational in the steady-state femtosecond regime. Using oxide nonlinear materials they cover nowadays mainly the near-IR part of the spectrum. Their wavelength coverage can be extended to the mid-IR spectral range by difference-frequency generation (DFG) with convenient tuning capability possible due to the simultaneously changing signal and idler wavelengths. For wavelengths exceeding 4-5 μm , non-oxide nonlinear crystals, transparent in the mid-IR, have to be employed in the DFG stage. Non-oxide nonlinear crystals exhibit smaller band-gap and thus two-photon absorption (TPA) limitations will come into play at tight focusing. Moreover, at tight focusing and high repetition rates also the damage resistivity with respect to the peak (axial) average power (the same as for cw radiation) plays a role and this is a critical parameter for all non-oxide nonlinear crystals.

The most popular nonlinear crystal for DFG with 800-nm pumped SPOPOs is AgGaS₂ (AGS) [1–3]. The highest DFG pulse energies previously demonstrated were obtained using GaSe crystals (26.3 pJ against 17.1 pJ with AGS at 8.5 μm) at a repetition rate of 84 MHz [3]. GaSe offers much higher nonlinearity and extension to longer DFG wavelengths but it is soft, can only be cleaved normal to the optic axes, and it is impossible to apply antireflection coatings. Recently we demonstrated that at least the mechanical stability problem can be solved with S-doping which simultaneously increases the band-gap, making GaS_{0.4}Se_{0.6} also suitable candidate for such DFG [4]. The most essential restriction with tight focusing in both crystals remains, however, the relatively large spatial walk-off. AgGaSe₂ (AGSe) also exhibits higher nonlinearity and wider transparency compared to AGS but is limited to long wavelength DFG due to birefringence and TPA restrictions. Energies of only 12 fJ at 84 MHz have been demonstrated with AGSe in the 10–18 μm range [5].

In this work, we employ for the first time nonlinear crystals of LiInSe₂ (LISE) for DFG of femtosecond pulses at 80 MHz achieving, to the best of our knowledge, highest single pulse energies (130 pJ at 7.2 μm) in the mid-IR above 5 μm.

2. Relevant properties of LISe

LISE (lithium selenoindate) belongs to the family of Li-chalcogenides with wurtzite-type structure and is a biaxial (orthorhombic) nonlinear crystal. Out of the four members of this family, it exhibits the smallest band-gap and the highest nonlinearity. While nonlinearity and transparency window of LISe are similar to AGS it is more damage resistant than all of the materials mentioned in the introduction. The band-gaps of these crystals follow the sequence LISe>AGS>GaS_{0.4}Se_{0.6}>GaSe>AGSe. The thermal conductivity of LISe is 3 to 5 times higher than that of AGS and AGSe [6] and ~2.5 higher than the thermal conductivity of layered GaSe along its optic axis [7]. We measured cw damage threshold at 1064 nm of 6 MW/cm² for LISe compared to 0.6 and 1 MW/cm² for GaSe and GaS_{0.4}Se_{0.6}, respectively [6,8]. Comparative data on AGS and AGSe does not exist but it is widely accepted that damage thresholds in such chalcopyrites are very low and estimations are in the 20–40 kW/cm² range for AGS and 5–22 kW/cm² for AGSe [9]. The high damage resistivity of LISe under tight focusing has been evidenced by successful cw DFG at shorter wavelengths (Ti:Sapphire lasers) [6].

As can be seen from Fig. 1(a), the phase matching-angle φ varies from 50° to 28° for the 4–12 μm DFG spectral range. However, it is known that the effective nonlinearity of LISe changes little in this plane [6], in this case from 9.7 to 11 pm/V. Note that the clear transparency range of LISe extends from 1 to 8 μm but samples thinner than 1 cm can be still used up to ~12 μm. The spatial walk-off for the pump and idler beams is almost constant, ~14 mrad (~2/3 of that in AGS) and should be taken into account. For sample thickness of 4 mm it is equivalent to ~56 μm.

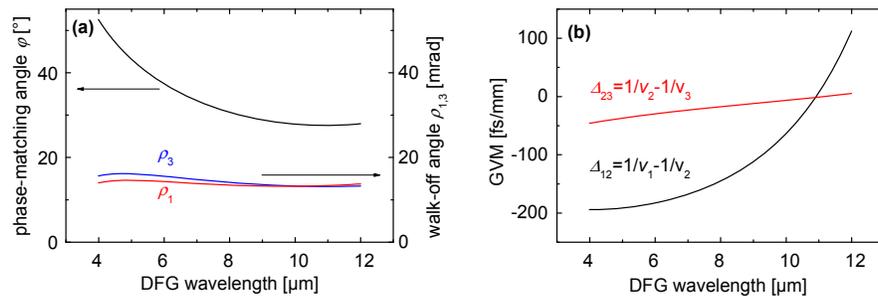


Fig. 1. Internal phase-matching angle φ and spatial walk-off angles $\rho_{1,3}$ for type-II DFG in LISe valid for mixing signal and idler pulses from an 800-nm pumped SPOPO (a) and corresponding GVM parameters (b). The indices 1,2,3 denote DFG, idler, and signal pulses.

The LISe crystal employed in the present experiment was with an aperture of $3 \times 5 \text{ mm}^2$ and thickness of 4 mm. It was cut at $\theta = 90^\circ$ for type-II eo-e interaction in the x-y plane which exhibits highest effective nonlinearity [6], at azimuthal angle $\varphi = 35^\circ$. The entrance surface was AR-coated for 1.3-2 μm and the exit surface for 5-10 μm . From Fig. 1(b) it can be concluded that the temporal walk-off between signal and idler is small in the entire tuning range and will not affect the conversion efficiency for pulse durations of the order of 200 fs. However, the temporal walk-off of the DFG pulse is relatively large (larger than in AGS) which shall lead to longer mid-IR pulse durations.

3. Experimental set-up and results

The DFG scheme shown in Fig. 2 is pumped and seeded by the separate outputs (signal and idler) of a commercial SPOPO (OPO PP Auto FAN, APE, Germany) in turn pumped by a Coherent Chameleon Vision Ti:Sapphire laser (80 MHz, 150 fs, max. 4 W at 800 nm). The signal (~ 1330 -1510 nm) and idler (~ 2000 -1700 nm) pulses from the SPOPO have durations of ~ 200 fs and ~ 190 fs, respectively, and spectral extension corresponding to ~ 1.5 times the Fourier limit. Typical average output powers in these tuning ranges are 650-800 mW for the signal and 500-650 mW for the idler. After temporally overlapping them through a delay line, the two beams are spatially recombined with a dichroic 45° mirror (reflecting for the signal in s-polarization and transmitting for the idler in p-polarization) and focused with a 80-mm CaF₂ lens to a beam diameter of $\sim 85 \mu\text{m}/110 \mu\text{m}$ for the signal and idler, respectively, in the position of the nonlinear crystal.

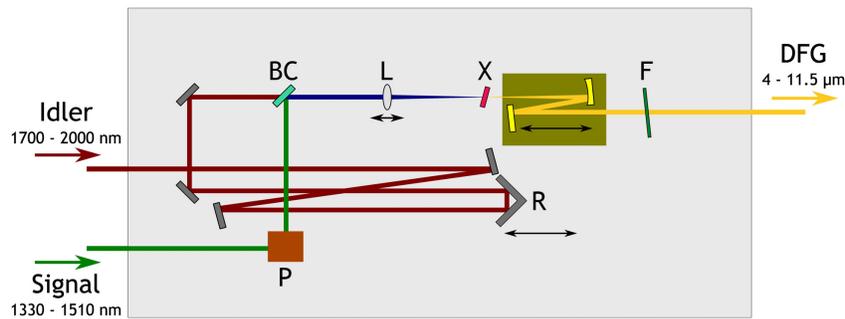


Fig. 2. Schematic of the DFG set-up. BC: beam combiner (dichroic mirror), P: periscope for polarization rotation, L: focusing lens, R: retro-reflector in delay line (also adjusting the beam height), X: LISe crystal, F: 3.6- μm cut-on Ge-filter.

Figure 3 shows the average output power obtained in the mid-IR and the external quantum conversion efficiency calculated from the incident signal and output DFG powers. The dip around 6 μm in Fig. 3(a) is due to declining SPOPO performance in a spectral range of air absorption for the resonated wave and competing sum-frequency and second-harmonic generation processes in the SPOPO periodically-poled crystal. At 7.2 μm , the single pulse energy reached 130 pJ, roughly 5 times higher than the best of the previous demonstrations with GaSe [3] and about 2 times higher than with GaS_{0.4}Se_{0.6} in which we obtained 70 pJ under similar conditions [4]. While the improvement in comparison to previous work is partially related to the more powerful pump source available for the present experiment and the thicker, AR-coated nonlinear crystal, we note that we were able to cover a very broad tuning range (4-11.5 μm) with one and the same LISe sample. The last two factors obviously contribute to the much higher powers compared to a 2-mm thick uncoated reference AGS sample (cut at $\theta = 42^\circ$ for type-II eo-e interaction) which has a similar and almost constant over the tuning range effective nonlinearity. The quantum conversion efficiency from signal to DFG photons in LISe exceeded 9% at 7.2 μm , see Fig. 3(b).

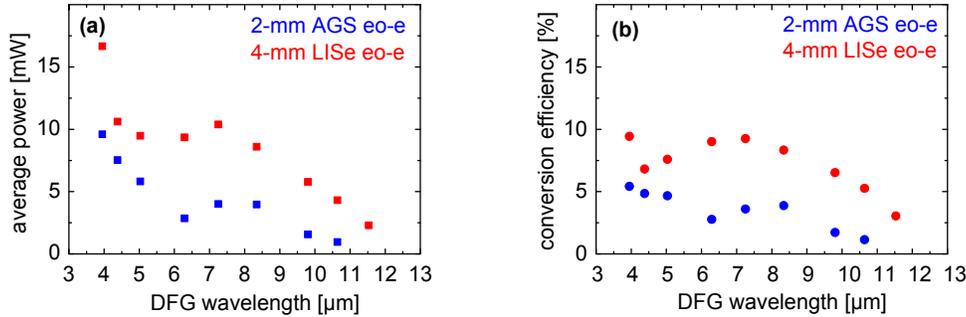


Fig. 3. Average power (corrected for the 3.6 μm cut-on Ge-filter) (a) and external quantum conversion efficiency (b) versus DFG wavelength obtained with the 4-mm AR-coated LISe crystal in comparison to a 2-mm thick uncoated AGS reference sample.

The DFG spectra shown in Fig. 4(a) were recorded with a $f = 50$ cm monochromator, 150 g/mm grating and a N_2 -cooled HgCdTe detector. Their bandwidths correspond to DFG pulse durations of 250-450 fs if a time-bandwidth product of ~ 0.5 is assumed, similar to the one measured for the input pulses. To measure the pulse duration of the DFG pulses we built a home-made autocorrelator using a 1-mm thick type-I AGSe crystal ($\theta = 45^\circ$) for which the upper limit (around 8 μm) was determined by the beam splitter (on CaF_2 substrate) transmission and the second harmonic detector (PbSe photoresistor).

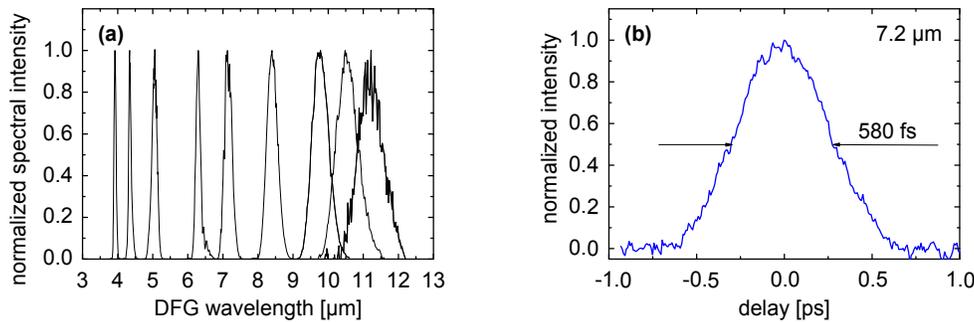


Fig. 4. Recorded DFG spectra demonstrating the achieved spectral tunability (a) and recorded autocorrelation function at 7.2 μm (b) with LISe.

From Fig. 4(b) one can conclude that the DFG pulses at 7.2 μm have a FWHM of 410 fs assuming Gaussian pulse shape (or ~ 380 fs assuming sech^2 shape). With the corresponding spectral width of ~ 250 nm this gives a Fourier product of ~ 0.6 for Gaussian pulses. Obviously some reshaping of the DFG pulses takes place in the nonlinear process with large GVM towards more square shaped pulses which have larger time-bandwidth product. The pulses obtained with AGS at the same wavelength are shorter (~ 215 or ~ 200 fs depending on the assumed shape) with a Fourier product of ~ 0.5 for Gaussian pulses. Note that the pulse durations in [3] where the input pulses were of similar width were not directly measured and thus a comparison is impossible. In general we observe that the SPOPO and DFG stage with crystal length ~ 2 mm maintain the Ti:Sapphire pulse duration while thicker crystals can lengthen the DFG pulse duration at improved efficiency. From Fig. 4(a) one can conclude that this is advantageous in the sense of better (by a factor of ~ 1.5) spectral resolution in potential spectroscopic applications.

4. Conclusion

The nonlinear crystal LISe showed excellent performance in DFG of femtosecond pulses at high (80 MHz) repetition rate by mixing the signal and idler output of an 800 nm pumped

SPOPO. Tuning from 4 to 11.5 μm was achieved with a single crystal cut, with single pulse energies exceeding any previous results obtained with such chalcogenide crystals. In contrast to nanosecond OPO experiments, no surface, bulk, or AR-coating damage was observed. The high damage resistivity to the average intensity in this regime means that yet higher average powers could be possible by up-scaling the input beams using 1- μm pumped SPOPOs based on diode pumping.