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Letter

$LiGaSe_2$ optical parametric oscillator pumped by a Q-switched Nd:YAG laser

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Abstract
Optical parametric oscillation is demonstrated for the first time with the chalcogenide nonlinear crystal $LiGaSe_2$ pumped by a nanosecond Nd:YAG laser. Angle tuning provides coverage of the 4.8–9.9 μm spectral range in the mid-IR by idler pulses.

Keywords: optical parametric oscillator, mid-infrared, lithium gallium selenide

(Some figures may appear in colour only in the online journal)

1. Introduction

$LiGaSe_2$ (LGSe) belongs to the group of the orthorhombic lithium ternary chalcogenides with the chemical formula $LiBC_2$, where B = In or Ga and C = S or Se. These compounds crystallize in the β-$NaFeO_2$ structure with space group $Pna2_1$ (point group mm2). Although the essential crystallographic properties were reported early on [1], the linear and nonlinear optical properties of LGSe could only be characterized after the technology for growing large-size optical-quality single crystals had been developed [2, 3]. Compared with $LiGaS_2$ (LGS) and $LiInSe_2$ (LISe), the bandgap of LGSe (3.34 eV at room temperature [2]), has an intermediate position. Thus, it can be expected that not only the nonlinearity [3] but also the damage threshold will have a similar intermediate position between the isomorphs of LGSe, in which Se is substituted by S or Ga by In. The transparency range of LGSe at a 5 cm^{-1} absorption level extends from 0.37 to 13.2 μm [2].

From the bandgap value it is clear that LGSe, like all the other $LiBC_2$ isomorphs, can be employed in frequency down-conversion schemes pumped at 1.064 μm (Nd:YAG laser systems) without detrimental two-photon absorption. Optical parametric oscillators (OPO) pumped at this wavelength have already been demonstrated with LGS and LISe [4, 5]. The main problem for power scaling of LISe OPO has been identified as the low surface damage threshold [5] (LISe exhibits the highest nonlinearity but the smallest bandgap of all the $LiBC_2$ compounds [6].) The use of LGS (the compound with largest bandgap and highest damage threshold but the lowest nonlinearity [6]) on the other hand is rather limited because of the high OPO threshold, leading to damage of other optical cavity elements [5]. LGSe has never been employed in an OPO for frequency down-conversion [7]; so far it has been used only in optical parametric amplification schemes employing femtosecond pump pulses [3, 8] in which regime the damage issue has a substantially different

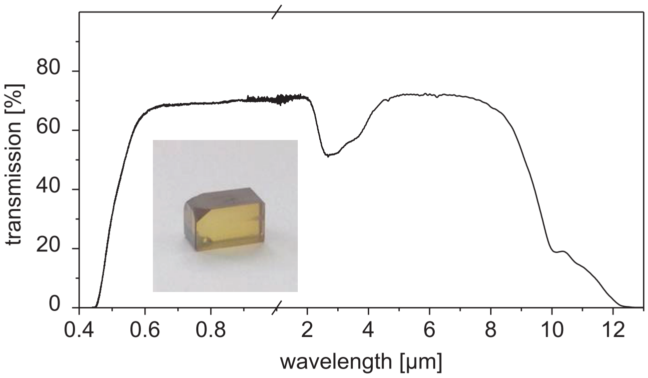

Figure 1. Transmission spectrum of the $LiGaSe_2$ (LGSe) nonlinear optical element recorded with unpolarized light along the 12.5 mm dimension. The sample itself is shown in the inset.

character (normally a result of self-focusing) allowing for some control.

This intermediate position with respect to the optical and thermo-mechanical properties motivated us to study the OPO performance of LGSe. In this work, for the first time to our knowledge, we report on successful operation of a 1.064 μm pumped LGSe OPO at a repetition rate of 20 Hz.

2. Experimental set-up

Elemental Li with 99.99% purity, and Se and Ga with 99.999% purity were used to synthesize LGSe. The starting components were loaded into a glass–graphite crucible which was placed inside a silica ampoule. The synthesis ampoule was connected to a vacuum system, evacuated to a residual pressure of 10^{-3} Torr and then sealed off. The synthesis was carried out in a vertical double-zone resistance furnace. The temperature in the hot zone was 100 °C above the melting point of LGSe, and the temperature in the cold zone was 500 °C. The fine-grained charge obtained was transferred to the crystal growth ampoule. Single-crystal growth was carried out by the vertical Bridgman technique. The ampoule was moved from the hot to the cold zone of the furnace at a speed of 0.02 mm h^{-1}. The temperature gradient in the growth zone was 2–5 °C cm^{-1}. In this way single LGSe crystals up to 50 mm long and 15 mm in diameter were obtained. The as-grown single crystals are normally milky or translucent due to uncontrollable deviations from stoichiometry. This requires post-growth annealing in an evacuated ampoule with the deficient component [9].

The LGSe OPO element was cut at $\theta = 90°$, $\varphi = 34°$ for eo-e type-II phase-matching in the x–y principal plane, which ensures higher and relatively constant effective nonlinearity [3]. The crystal was 12.5 mm long, with an aperture of 7 × 6 (//z) mm^2. The unpolarized transmission spectrum recorded prior to coating is shown in figure 1. The good transmission range extends up to ~8 μm in the mid-IR but one can expect that LGSe will still be useful for such a thickness up to ~10 μm. This sample was single-layer anti-reflection coated for high transmission in the pump (1.064 μm) and signal (1.15–1.6 μm) spectral ranges.

The pump source was a lamp-pumped, electro-optically Q-switched Nd:YAG laser (Model LQ215, Solar Laser Systems, Belarus), optimized for a repetition rate of 20 Hz. The spectral linewidth of this laser was 1.5 cm^{-1}, $M^2 < 2$, with a divergence of <2.5 mrad. The laser generated pulses with an energy of up to 180 mJ and 6 ns duration (FWHM) resulting in a maximum average power of 3.6 W. The measured energy stability was ±2.5%.

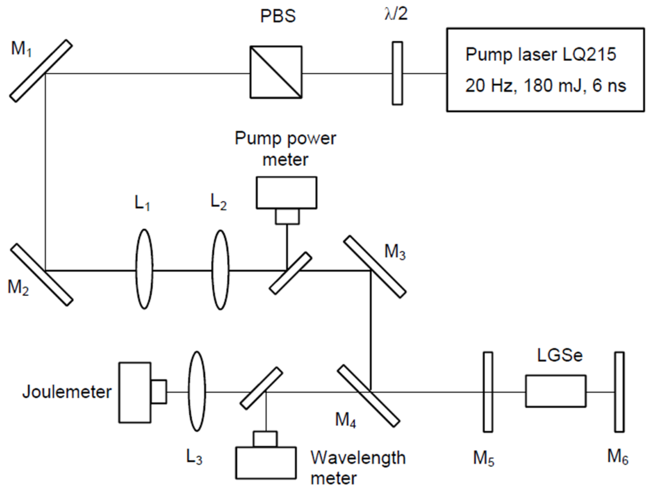

Figure 2. Experimental set-up of the LGSe OPO: M_1–M_5, dielectric mirrors; M_6, Au total reflector; L_1–L_3, lenses; PBS, polarizing beam splitter; $\lambda/2$, half-wave plate.

Figure 2 shows the entire experimental set-up. A combination of a half-wave plate ($\lambda/2$) and a polarizing beam splitter (PBS) was used to adjust the pump energy. The pump laser was protected by an internal Faraday isolator and the separation from the OPO was large enough to avoid feedback during the Q-switching process.

A telescope comprising two lenses, L_1 ($f = 200$ mm) and L_2 ($f = -50$ mm), was used to reduce the size of the pump beam in the position of the nonlinear element to a waist diameter of 2.23 mm in the vertical direction (along the z-axis of the LGSe crystal) and 2.98 mm in the horizontal direction. This slightly elliptical shape of the pump beam was defined by the initial pump beam from the Nd:YAG laser. The dielectric mirrors M_1, M_2, M_3 were used to steer the pump beam into the OPO cavity. Pumping via a 45° ZnSe bending mirror (M_4), which was highly reflecting for the pump and highly transmitting for the idler, provided separation of the input and output waves.

The plane input–output coupler (IOC) ZnSe mirror M_5 was highly transmitting for the pump and idler (85–90% at 2–12 μm) waves with ~90% reflectivity at 1.15–1.6 μm for the signal. The flat Au rear mirror M_6 was highly reflecting for all three waves: signal, idler and pump. Thus double-pass pumping of the singly resonant OPO was realized and the idler was extracted after a double pass through the LGSe crystal. The LGSe sample was mounted in a copper block attached to a rotation stage with precise φ-angle adjustment. The cavity length was 24 mm.

3. Results and discussion

The idler output was collected by a 20 cm CaF_2 lens, L_3, behind the bending mirror M_4, and the idler energy was measured by a

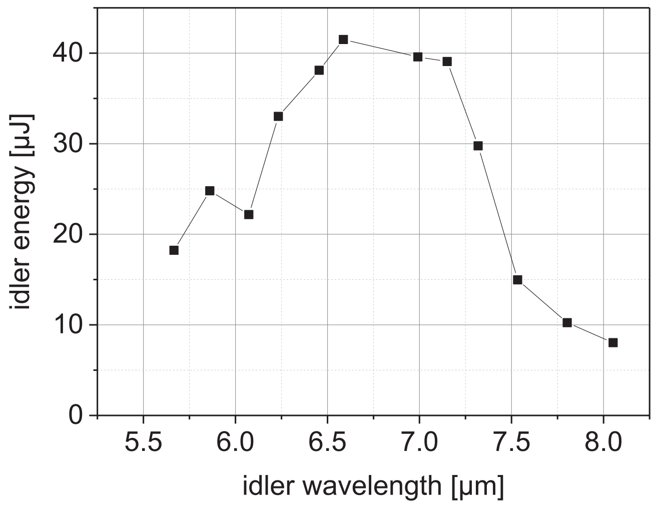

Figure 3. Idler energy of the LGSe OPO recorded versus idler wavelength.

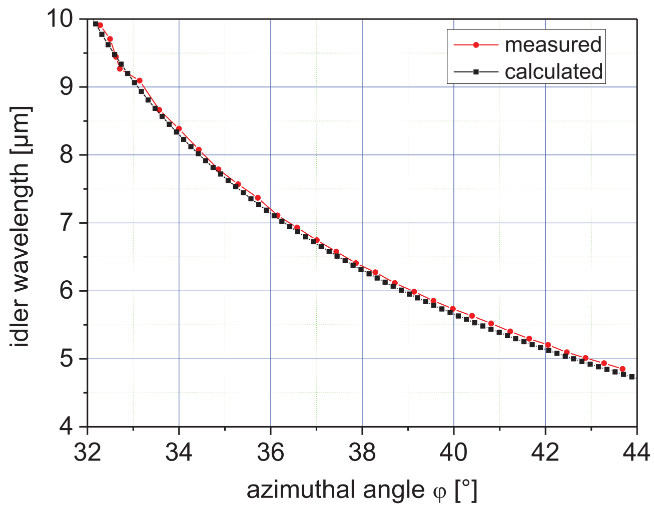

Figure 4. Calculated and experimental idler wavelength versus azimuthal angle φ.

pyroelectric detector using suitable filters to suppress the residual pump and signal radiation. The signal wavelength was measured using a commercial wavelength meter (Model LSA IR-I, HighFinesse, Germany) with a resolution of $\lambda/\Delta\lambda = 2000$. The idler wavelength was calculated from the obtained signal wavelength. The idler energies specified below were corrected for the filters and L_3 transmission, while the pump energy is specified as the one incident on the LGSe crystal.

The LGSe OPO threshold was 4 mJ for an idler wavelength of 6.57 μm when the crystal was rotated away from normal incidence. This threshold value corresponds to an average fluence of ~0.08 J cm^{-2} or an average pump intensity of 13 MW cm^{-2}. To study the tuning potential of the OPO we increased the pump energy to 12.5 mJ, i.e. roughly three times threshold, which is equivalent to an average pump fluence of 0.24 J cm^{-2} (40 MW cm^{-2}). These values correspond roughly to 60% of the damage threshold for LGSe which we measured on separate test plates.

Figure 3 shows the measured idler energy versus wavelength for the LGSe OPO. The maximum idler energy reached 41.5 μJ at 6.57 μm. The entire tuning range evaluated from the measured signal wavelength is broader, extending from 4.8 to 9.9 μm (figure 4). Note that normal incidence corresponds to an idler wavelength of 8.4 μm. The agreement of the derived idler wavelength with calculations based on the Sellmeier equations published for LGSe [2, 6] is excellent.

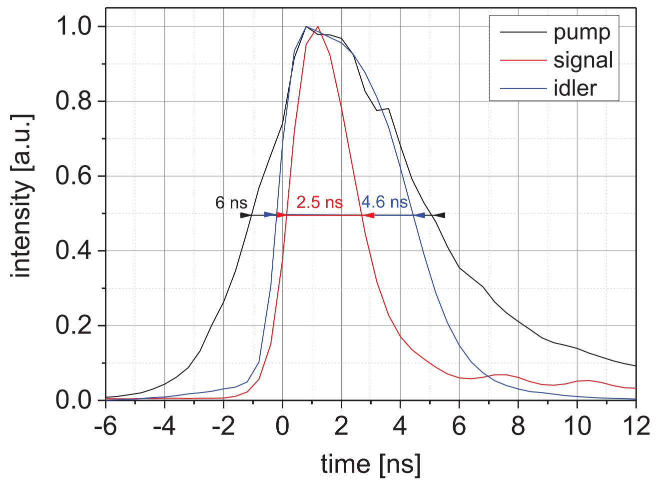

Figure 5. Pump, signal and idler pulse shapes.

The pulse duration of the pump, signal and idler pulses was measured using a fast InGaAs photodiode in the near-IR and a HgCdTe photodetector (Model PMV-10.6, Vigo Systems, Poland) in the mid-IR. Figure 5 shows the recorded pulse profiles: the FWHM amounts to 6 ns (pump), 2.5 ns (signal) and 4.6 ns (idler). The idler pulse duration is affected by the finite response (<2 ns rise time) of the HgCdTe photodetector, and from energy conservation considerations the actual FWHM should be close to that of the signal pulses. The fact that the signal pulses are more than two times shorter than the pump indicates a relatively long build-up time, which is one of the reasons for the relatively low conversion efficiency.

4. Conclusions

In summary, we have demonstrated nanosecond OPO operation with an LGSe crystal in the mid-IR wavelength range from 4.8 to 9.9 μm for the idler wave by angle tuning. A maximum idler pulse energy of 41.5 μJ was obtained at ~6.57 μm at a repetition rate of 20 Hz. The OPO threshold in terms of fluence was roughly three times lower compared with LGS [5], which is attributed to the higher nonlinear coefficients of LGSe [3] and to the availability of larger single crystals permitting the sample length to be increased. This resulted in a quantum conversion efficiency about two times higher with LGSe. Compared with LISe [4], the LGSe OPO showed very similar tuning range and threshold although the available sample was shorter. Similarly high idler energies (>150 μJ) can be expected by utilizing the full aperture of the available LGSe sample.

The position of the $LiBC_2$ isomorphs among other non-oxide crystals applicable for down-conversion into the mid-IR, and in particular in 1.064 μm pumped OPOs, has been widely discussed in [7]. There is no ideal crystal that provides the highest energy and broadest spectral tenability but at the same time is damage resistant and can be operated at higher repetition rates. The specific potential of LGSe, in OPOs or other nonlinear frequency conversion devices with ultrashort pulses, will depend on its susceptibility to optical damage under different conditions and its thermo-mechanical properties, which are still under investigation. Comparative damage results obtained with different laser sources emitting in the 1 μm region are currently being analyzed and will be published separately in a forthcoming paper.

Acknowledgments

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