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Short Communication

Singly-resonant optical parametric oscillation based on the wide band-gap mid-IR nonlinear optical crystal $LiGaS_2$

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ABSTRACT

We demonstrate optical parametric oscillators based on the wide band-gap (3.76 eV) $LiGaS_2$ (LGS), pumped by 100-Hz/8-ns and 1-kHz/1-ns laser sources at 1064 nm. Notwithstanding the modest second order nonlinearity of this material singly resonant operation was possible due to the high optical damage resistivity of LGS. Idler pulses near 5.46 μm were generated with an energy of 134 μJ and duration of 5.4 ns in the first case and energy of 1.1 μJ and sub-nanosecond duration in the second case. We present also optical surface damage measurements at 1064 nm using 14-ns pulses.

1. Introduction

Among the non-oxide nonlinear optical crystals transparent beyond ~5 μm in the mid-IR [1], the orthorhombic lithium ternary chalcogenides with the chemical formula $LiBC_2$, where B=In or Ga and C=S or Se, are characterized by the widest band-gaps [2], which enable pumping at relatively short wavelengths, e.g. Nd:YAG lasers at 1064 nm, for down-conversion into the mid-IR without two-photon absorption. In comparison with their Ag-chalcopyrite analogs, $AgBC_2$, the orthorhombic $LiBC_2$ exhibit more than four times better thermal conductivity and positive thermal expansion for all directions, higher damage thresholds and lower refractive indices. The $LiInC_2$ compounds show higher nonlinear coefficients than their $LiGaC_2$ analogs while the selenides have higher nonlinearity than the corresponding sulfides, all in accordance with the relative band-gap values [1,2].

Due to limited interaction lengths when using ultrashort pulses and the low parametric gain in the continuous-wave (cw) regime, highest down-conversion efficiency of a nonlinear process can be expected in the intermediate temporal regime covered by short-pulse optical parametric oscillators (OPOs). In our previous research we were able to demonstrate doubly-resonant and singly-resonant OPOs with $LiInSe_2$ (LISe) at first near 3.5 μm where the parametric gain is higher [3] and later in a broader tuning range from 4.7 to 8.7 μm [4] because this compound exhibits the highest effective nonlinearity in this family of crystals. However, it turned out that LISe exhibits also the lowest chemical stability while the only compound in which absolutely no surface degradation occurred (even for a very long period of >7 years) was $LiGaS_2$ (LGS). Though with lowest effective nonlinearity, $d_{eff} \sim 5.5$ pm/V for conversion of 1064 nm radiation to an idler wavelength of 6450 nm [1], LGS shows the widest band-gap (3.76 eV from transmission cut-off) among the $LiBC_2$ compounds. Since from our OPO experience with various LISe samples we suspected a correlation between the surface chemical stability and the quality and reproducibility of the antireflection (AR) coatings, we decided to focus our further efforts on LGS. Here we demonstrate for the first time to our knowledge singly-resonant operation of 1064-nm pumped OPOs based on LGS.

2. Surface damage threshold of LGS

It is obvious that a material with relatively low nonlinearity will require much higher pump intensities for OPO operation and that is why we first measured the surface damage threshold of LGS test samples annealed in S vapor. Three colorless plates of LGS, 1.25-mm thick, with unknown orientation but good optical quality were prepared for the damage tests. The plates were mounted in metallic holders. The first plate was uncoated, the second one had an AR-coating on one side and the third one was AR-coated on both sides. The single layer AR-coating was designed for the

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pump and signal (OPO resonated wave) only, in order to obtain higher resistivity. However, the material chosen (YF_3) ensures that there is no absorption loss at the idler wavelength. The AR-coating was centered at 1250 nm since in an OPO the signal losses are more detrimental than those for the pump. Also an older uncoated LGS element was tested. This sample was with an aperture of 5×6 mm^2, thickness of 3 mm and orientation $\varphi = 41°$, $\theta = 90°$ but its bulk optical quality was low. It was the same sample used for optical parametric amplification studies in 2003 [5].

The intensity of the unfocused pump beam from the 1064 nm multimode (but quasi-Gaussian) Nd:YAG Q-switched laser (14 ns, 100 mJ, 100 Hz, Innolas GmbH) was insufficient for this experiment and a 57 cm focal length lens was used with the samples positioned in front of its focus. At this position the measured beam diameter ($2w$) was 1.55 mm in the horizontal and 1.47 mm in the vertical direction. The pump level was increased in small steps (between 0.07 and 0.17 J/cm^2) and the reported damage values are average of the fluence before and after damage occurred for exposure time of 1 min (6000 pulses). Scattering centers (turbidities) were seen in visible light as a first sign of surface damage but the quality of the old sample did not allow this to be properly observed. They occurred not necessarily in the center of the pump spot and the actual damage developed from them at higher intensity. The cracks occurred in fact on the next day after the experiments and developed from the damaged front surface. The results in terms of peak on-axis fluence are summarized in Table 1 and the damages are illustrated in Fig. 1.

The main conclusion is that the damage thresholds are roughly 5 times higher than for LISe. In addition:

- The damage threshold values show good reproducibility and no drops like in the case of LISe where damage (turbidity) was observed also at 0.5 J/cm^2 [6]. Bulk damage does not occur and there is no dependence on the bulk optical quality or aging.
- There is no systematic difference between uncoated and AR-coated surface damage thresholds. Since similar coatings were applied also to LISe, one can conclude that the AR-coating problems in LISe are related to the surface chemical stability and not necessarily to the coating itself [6].

3. LGS OPO operating at 1 kHz with 1-ns pump pulses

LGS single crystals are grown using the Bridgman-Stockbarger technique, in a double-zone vertical furnace. The upper and lower zones are separated by a diaphragm from heat-insulating material. The temperature of the melting zone is maintained at about 1150 °C (roughly 100 °C above the melting temperature) with an accuracy of ±0.1°C. The axial temperature gradient is ~2°/mm. Typical rates for the ampoule sinking are 2–10 mm/day. The starting reagents are with 99.999% (Ga, S) and 99.9% (Li) purity. The charge is placed into a glass–graphite crucible and the latter is located in a silica ampoule. Such construction allows one to avoid the chemical reaction between lithium and the walls of the silica ampoule. The sample available for the present OPO studies was initially only 8.2 mm long, with an aperture of 5 mm (along Z-axis) $\times$ 7 mm, see Fig. 2a. It was cut at $\theta = 90°$, $\varphi = 40.6°$ for eo-e type-II phase-matching in the X–Y plane and AR-coated with a single layer of YF_3 centered at 1250 nm (the same one as for the damage test plates in the previous section).

Table 1
Summary of the surface damage thresholds obtained with the 4 LGS plates.

Sample	Damage (turbidity) (J/cm^2)	Full damage (J/cm^2)
#1 (Uncoated)	3.33	4.70 (crack)
#2 (Single side AR-coated)	3.44	3.71 (crater)
#3 (Double side AR-coated)	3.84	4.45 (crack)
Old (uncoated, thicker)	Could not be observed	3.52 (crater)

The growth of $LiBC_2$ crystals is rather complex and deviation from stoichiometry is typical for this family because of the high chemical activity of lithium and the chalcogen volatility. Post-growth annealing in appropriate atmosphere is usually used to achieve the desired crystal composition and get closer to the stoichiometric formula. Unfortunately, of the four $LiBC_2$ compounds, LGS turns out also to be the most difficult to grow in larger sizes. That is why we decided to perform the first OPO tests using a special pump laser system delivering pulses of only 1-ns duration at 1064-nm. It consisted of an electro-optically Q-switched, 1 ns $Nd:YVO_4$ microlaser (ECR Model PML 1000), a cw pumped $Nd:YVO_4$ regenerative amplifier, and a double pass Nd:YAG post amplifier with pulsed pumping (both HighQLaser), optimized for a repetition rate of 1 kHz. The maximum available pump energy was about 1.4 mJ, of which ~0.85 mJ were incident on the LGS crystal. A combination of a half-wave plate, $\lambda/2$, and a polarizer, P, served to adjust the pump energy, see Fig. 2b. The pump beam had a Gaussian diameter of $2w \sim 0.61$ mm in the position of the OPO. Only the idler energy was measured behind the pump bending mirror BM, the residual pump radiation and the signal were blocked by a 2.5 μm cut-on filter, F.

Using a relatively short cavity such a pump source enables sufficiently high pump intensity to reach the OPO threshold with fluence that can be still below the damage limit of the optical components. The 10-mm long linear OPO cavity was formed by a 10-cm radius of curvature (RC) input–output coupler (OC) on a 3-mm thick YAG substrate, highly reflecting ($R > 99\%$) the signal and highly transmitting ($R < 5\%$ front side, AR-coated rear side for $R < 1\%$) the pump and idler waves, and a 5-cm RC Ag total ($R > 97\%$) end reflector (TR). The dichroic pump BM on ZnSe substrate was highly reflecting for the pump and highly transmitting both for the signal ($T = 82\%$) and the idler ($T = 84\%$).

The input–output OPO characteristics at normal incidence are shown in Fig. 3. Although the threshold was ~80 times higher compared to the highly nonlinear, but otherwise limited in tunability $CdSiP_2$ crystal used in the same set-up [7], it was possible to operate the LGS OPO at pump levels >2 times above threshold without optical damage. Angle tuning was studied in a slightly lengthened (15 mm) cavity, covering the 4.046–6.014 μm spectral range for the idler. Unfortunately such wavelengths were absorbed by the YAG substrate of the output coupling mirror which transmitted only ~55% at ~5.46 μm, the idler wavelength measured at normal incidence to the LGS crystal. Taking this into account the maximum quantum conversion efficiency reached in Fig. 3 amounts to 3.5%. Note that the maximum applied peak on-axis pump intensity in Fig. 3 exceeded ~550 MW/cm^2, values which are far above the damage limit of most other non-oxide nonlinear optical crystals [7], however, according to our damage threshold measurements we had a safety reserve with LGS of about a factor of 6 in terms of pump intensity.

4. LGS OPO operating at 100 Hz with 8-ns pump pulses

In order to utilize the full aperture of the same LGS OPO element (after repolishing and recoating its length was ~8 mm; this was necessary as a result of accidental secondary surface damage indirectly caused by a damage occurring in a cavity mirror during initial experiments) we decided to pump with a higher energy nanosecond laser source at 1064 nm. In fact this was the same diode-pumped system used in the damage tests described earlier which, after modification of the oscillator and addition of a double pass Nd:YAG amplification stage, provided an output energy of up to 250 mJ at 100 Hz with a pulse duration of ~8 ns and beam M^2

Fig. 1. Surface damage in sample #1 (left), sample #2 (middle) and old LGS sample (right).

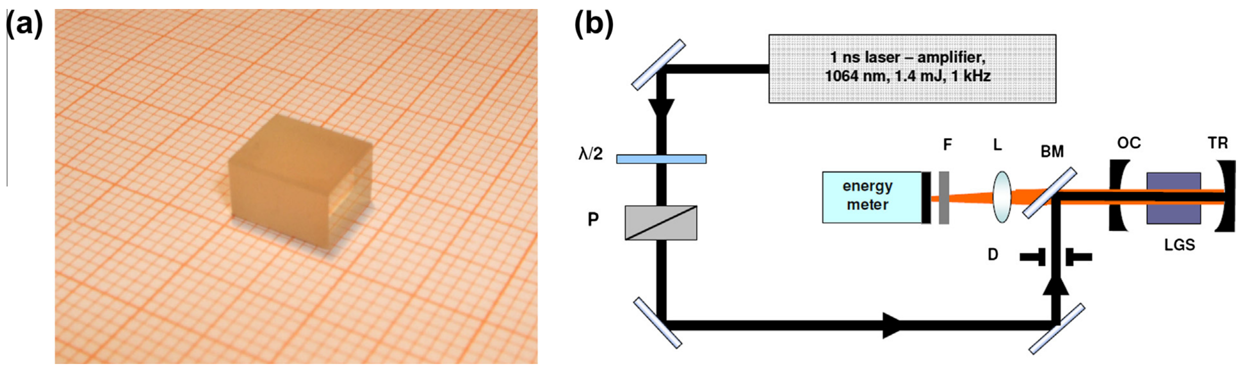

Fig. 2. Oriented AR-coated LGS element prepared for the OPO experiments (a) and experimental set-up of the 1-kHz LGS OPO (b): L – lens collecting the output onto the pyroelectric detector, D – diaphragm used for alignment of the OPO.

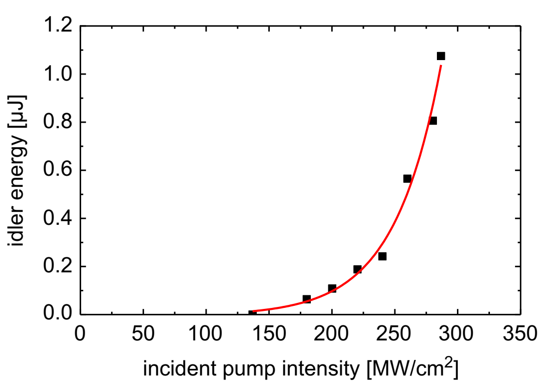

Fig. 3. Idler pulse energy of the 1-kHz LGS OPO at 5.457 μm versus average pump intensity (approx. ½ of the peak on-axis intensity) at 1064 nm. The output coupler transmits about 55% at this wavelength due to the YAG substrate absorption.

factor of 1.4. The set-up was very similar to the one shown in Fig. 2b, with a telescope added to increase the cross section of the pump beam to a diameter 2*w* of ~5 mm (along *Z*-axis) × ~7 mm (LGS critical plane). The singly resonant cavity was 12-mm long, the OC was substituted by a RC = 2 m mirror on ZnS substrate which was highly transmitting the pump (~90%) and idler (~94%) waves (with AR-coatings on the rear side) and reflecting ~70% at the signal wavelength, and the curved Ag TR was substituted by a plane one. Again, double pass was realized for the pump and idler waves to reduce the OPO threshold.

The input–output OPO characteristics at normal incidence are shown in Fig. 4a. The maximum idler energy at ~5.46 μm amounted to 134 μJ. As expected, the OPO threshold was still quite high, ~60 mJ. Thus, it was possible to reach OPO operation only up to ~1.4-times above threshold and the maximum quantum conversion efficiency was only ~0.8%. Still the net result in terms of idler energy exceeded the value reported in the previous section more than 100 times while the average power at 100 Hz reached 13.4 mW.

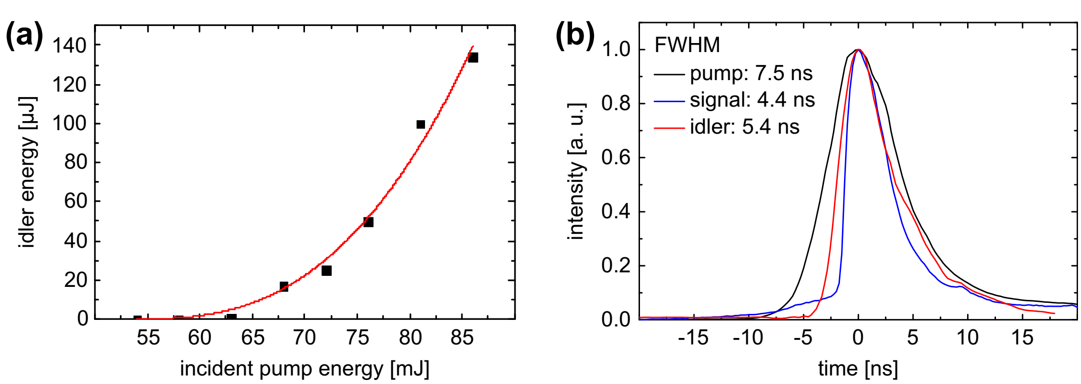

Fig. 4. Idler pulse energy of the 100 Hz LGS OPO at 5.457 μm versus pump energy at 1064 nm (a), and measured pump, signal and idler pulse shapes (b).

The maximum applied on-axis pump fluence in Fig. 4a was $\sim$0.62 J/cm^2 (or $\sim$78 MW/cm^2). This is much less than the LGS surface damage threshold measured but the present limit in this setup is set by optical damage of the cavity mirrors. Once this problem is resolved, the obtained slope in Fig. 4a indicates that idler energies exceeding 1 mJ are realistic at the maximum available pump energy (roughly 4 times pump threshold).

With the help of a 1-mm thick Ag-coated CaF_2 Fabry–Perot etalon and a CCD camera we estimated a signal ($\sim$1.32 μm) bandwidth of 56 GHz (1.9 cm^{-1} or 0.32 nm). The temporal characteristics of the LGS OPO were measured at maximum pump level using fast photodiodes and 2 GHz oscilloscope. The (HgCdZn)Te detector used for the idler (Vigo systems model PCI-9) had a time constant of <2 ns. As can be seen from Fig. 4b, both signal and idler exhibit shorter durations compared to the pump which is typical for the intensity-dependent nonlinear process in the OPO. The actual idler pulse duration should be <5.4 ns due to the finite detector response and thus comparable to that of the signal. Similarly, this means that in the 1-kHz pumped LGS OPO described in the previous section, sub-nanosecond output pulse durations should be expected but detectors with such temporal resolution do not exist for wavelengths exceeding $\sim$2 μm.

5. Conclusions

Using 14 ns pulses we established that the surface damage threshold of LGS is very high, $\sim$3.5 J/cm^2 (peak on-axial fluence) at 1064 nm and 100 Hz. This is more than 5 times higher in comparison with LISe, the Li-compound with highest d_{eff}. Moreover, LGS exhibits no surface aging effects as the other $LiBC_2$ compounds.

Thus, singly-resonant OPO operation in the mid-IR was possible with LGS although the pump threshold was quite high. Idler pulses near 5.46 μm were generated with an energy of 134 μJ and duration of 5.4 ns pumping by a 100-Hz/8-ns laser system at 1064 nm, and energy of 1.1 μJ and sub-nanosecond duration pumping by a 1-kHz/1-ns laser system at the same wavelength. We are convinced that much higher output energies and reasonable conversion efficiencies can be obtained with this new nonlinear material but to this aim, samples at least 2 cm in length and with good optical quality will be necessary.

Acknowledgments

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