Picosecond mid-IR optical parametric amplifier based on $GaS_{0.4}Se_{0.6}$ pumped by a Nd:YAG laser system at 1064 nm

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Abstract: Operation of a $GaS_{0.4}Se_{0.6}$ optical parametric amplifier is demonstrated with 5-11 µm idler tuning range and maximum energies ~10 µJ for sub-30-ps pulse durations, and performance ~3 times better than with pure GaSe.

OCIS codes: (190.4970) Parametric oscillators and amplifiers; (190.4400) Nonlinear optics, materials

Recently, substantial progress in the characterization of mixed GaS_xSe_{1-x} nonlinear crystals was achieved, measuring their transparency, refractive index dispersion, nonlinear coefficient, damage threshold, and two-photon absorption [1]. Two essential advantages are associated with the S-doping of the well known nonlinear crystal GaSe: increase of the band-gap value or the short wave cut-off limit and improved hardness [1] which is one of the basic limitations of GaSe. In [1] we concluded that $GaS_{0.4}Se_{0.6}$ (shortly GaSSe, the composition with maximum S content before the transition to a centrosymmetric phase) is a promising nonlinear material for down-conversion of pulsed 1.064 µm radiation to the mid-IR above 5 µm without significant two-photon absorption (TPA), possessing a nonlinear coefficient d_{22}(GaSSe)= 0.76 d_{22}(GaSe). Unfortunately, the problems with cutting, polishing and coating of such crystals are still not resolved and application in optical parametric oscillators (OPOs) depends on the stability and damage resistivity of antireflection (AR) coatings. However, in optical parametric amplifiers (OPAs), pumped by high-energy ultrashort pulses, cleaved uncoated crystals can be employed and in this work we present such results with GaSSe using a pump system operating at 1.064 µm. This is the first real application of GaSSe in nonlinear optics and we compare the results with pure GaSe under identical experimental conditions. GaSe has been used in the past in similar schemes (pump at 1.053 µm and tunable dye laser as a seed signal) to cover the 6-18 µm idler range [2] but saturation was observed already at pump intensities beyond 100 MW/cm^2. Although improved results were reported later with an optical parametric generator (OPG) providing the seed signal [3], as we demonstrate here, the use of GaSe is in general limited by TPA when pumped near 1 µm which is not the case with GaSSe.

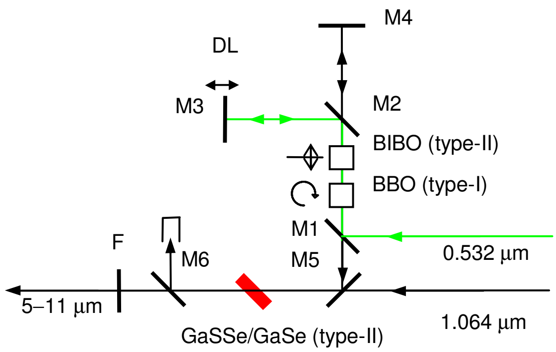

Fig. 1. Schematic of the experimental set-up for OPA in GaSSe and GaSe. M1–M3: 0.532-µm HR dichroic mirrors, M4: metallic mirror, M5: 1.15–1.4-µm HR dichroic mirror, M6: 1.064-µm HR dichroic ZnSe mirror, F: 2.5-µm cut-on filter, DL: Delay line.

As a pump source for the OPA we employed a mode-locked Nd:YAG laser-amplifier system at 1.064 µm with a pulse duration of 58 ps (FWHM) at a repetition rate of 10 Hz. The seed source was derived from another OPA system pumped by the second-harmonic (SH) beam at 0.532 µm (horizontal polarization), consisting of a 6 mm-long BBO and 5-mm long BIBO crystals used in tandem for type-I (ooe) and type-II (oeo) interactions, respectively, in a double-pass configuration. In this OPA system, the BBO serves as an OPG in the first pass and as a broadband amplifier in the second pass, which is seeded by the narrow signal spectrum from type-II OPA in BIBO with an appropriate pump delay realized by translation of the M3 mirror in Fig. 1. The vertically polarized idler beam from this OPA system, with energy in the 1–10 µJ range, depending on the wavelength which could be varied between 1.175 and 1.4 µm, is then collinearly mixed in GaSSe or GaSe with the collimated fundamental beam at 1.064 µm (horizontal polarization) through the bending mirror M5. In the present OPA experiment, type-II (eoe) process is chosen rather than type-1 (ooe) process, considering the fact that it provides higher idler output in the absence of AR coating because of the much lower surface reflection for p-polarization (this was confirmed by comparing the idler output for both processes under the same experimental conditions).

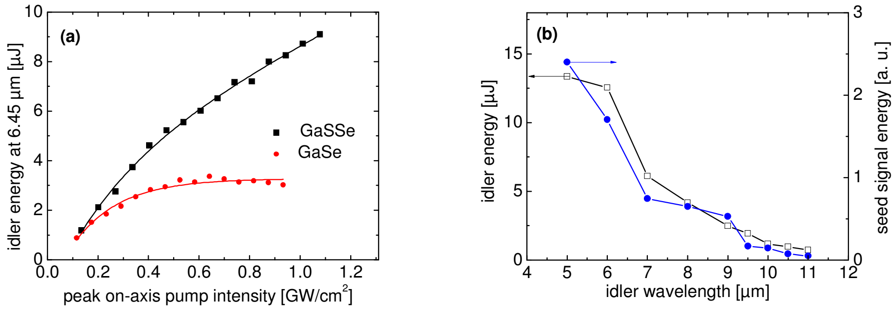

Fig. 2. (a) Idler energy at 6.45 µm versus pump intensity of the GaSSe and GaSe OPAs and (b) idler tuning achieved with GaSSe at maximum pump level. The peak on-axis pump intensity inside the crystal is calculated using the actual value of the pump pulse duration of 76 ps (as a result of SH depletion), the beam size inside the crystal and correcting by the Fresnel reflection at the input surface.

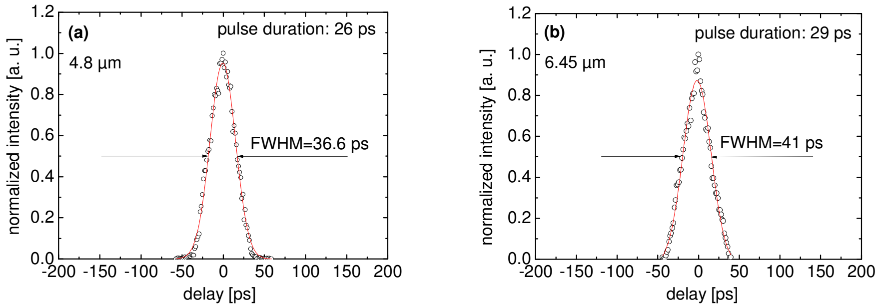

Fig. 3. Autocorrelation functions recorded for the GaSSe OPA at idler wavelengths of (a) 4.8 µm and (b) 6.45 µm by noncollinear SH generation in a 3-mm thick GaSe crystal.

Figure 2a shows the idler energy measured at a wavelength of 6.45 µm in dependence on the pump intensity at 1.064 µm at a fixed SH energy of 1.6 mJ (after the bending mirror M1) for pumping the BBO–BIBO seeder. For the actual phase-matching angles, effective nonlinearities of 38 and 49 pm/V are estimated for GaSSe and GaSe, respectively. Taking into account the 20% lower figure of merit of GaSSe, a 4.7-mm-long *c*-cut sample was prepared for fair comparison with a 3.9-mm-long GaSe sample. For the beam diameters and pulse durations in the present experiment, spatial and temporal walk-off effects can be neglected. At low pump intensity (<0.2 GW/cm^2), the idler energies with GaSSe and GaSe were very similar as expected from the calculated parametric gain factors. However, when the pump intensity was further increased, GaSSe showed much better performance. At a pump intensity of 1.08 GW/cm^2 (~2.8 mJ incident pump energy at 1.064 µm), the idler energy reached 9.1 µJ, about 3 times higher than with GaSe. The behavior of GaSe was similar to the one observed in [2], however, we attribute the ”saturation” not to pump depletion but to nonlinear pump losses, e.g. TPA at 1.064 µm, which we also confirmed experimentally. The tuning performance of GaSSe (5-11 µm) is determined in the present experiment to a great extend by the available seed energy at the signal wavelength (Fig. 2b). The estimated idler pulse duration was 26 ps at 4.8 µm and 29 ps at 6.45 µm (Fig. 3a,b), shorter than the pump pulse duration as could be expected.

In conclusion, we demonstrated that the mixed GaSSe can be used for down-conversion to the mid-IR with powerful ~1 µm pump sources. In comparison to the few other nonlinear crystals that exhibit no TPA at this wavelength (e.g. $AgGaS_2$), GaSSe possesses the best potential to cover deep mid-IR wavelengths up to ~14 µm [1].

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