$GaS_{0.4}Se_{0.6}$: relevant properties and potential for 1064 nm pumped mid-IR OPOs and OPGs operating above 5 μm

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The nonlinear optical properties of mixed GaS_xSe_{1-x} crystals with x=0.2 and 0.4 were studied as early as 1982 [1]. Relative SHG measurements gave d_{22}(x=0.2)=0.525 d_{22}(GaSe) and d_{22}(x=0.4)= 0.31 d_{22}(GaSe) for the nonlinear coefficients. Two essential advantages can be expected from adding S to the well known nonlinear crystal GaSe: increase of the band-gap value or the short wave cut-off limit (which is not the case using In as a dopant) and improved hardness which is one of the basic limitations of GaSe.

For our studies, GaS_xSe_{1-x} crystals were grown by the Bridgman-Stockbarger method in quartz ampoules with a diameter of 14 mm for compositions x = 0.0, 0.05, 0.10, 0.40 in the charge. The whole growth process took 20-25 days and uniform single crystals up to 60 mm in length were obtained. Interestingly, for the composition with x=0.4 the crystals grow without any "cap" at the top of the boule. This means that the charge and crystal compositions are identical, which is normally not the case even for pure GaSe. This observation could possibly mean congruent melting character which is equivalent to the existence of a separate compound in the system of solid solutions.

Characteristic unpolarized spectra corresponding to the o-wave were measured using thin (50-600 μm) cleaved plates and the direct band-gap was obtained from the linear fit to the $(\alpha h\nu)^2$ dependence on $h\nu$. The band-gap increases from 1.976 eV (628 nm) to 2.304 eV (538 nm) from x=0 to x=0.4. The nonlinear coefficient of the mixed compounds was re-measured by comparing the SHG conversion efficiency to that of pure GaSe. A $KNbO_3$ femtosecond optical parametric amplifier was used as a laser source, operating at 4.65 μm at a repetition rate of 1 kHz. The results obtained, $d_{22}(GaS_{0.05}Se_{0.95})$=0.9 d_{22}(GaSe), $d_{22}(GaS_{0.1}Se_{0.9})$=0.88 d_{22}(GaSe), and $d_{22}(GaS_{0.4}Se_{0.6})$= 0.83 d_{22}(GaSe) indicate much weaker dependence on the doping level. The measured dependence on the azimuthal angle suggests that either the symmetry of GaSe ($\bar{6}m2$) is preserved or the d–coefficient which is independent of the azimuthal angle (in 3m symmetry) has a negligible contribution. The index of refraction of $GaS_{0.4}Se_{0.6}$ was measured in the 0.633 – 10.0 μm spectral range using the auto-collimation technique on a prism with an aperture of 15x15 mm^2 and apex angle of 13.74°. Sellmeier equations with two-poles were then constructed and the SHG phase-matching curve was compared with experimental results at 2.79, 9.59 and 10.6 μm from the literature [2]. The damage threshold at 1064 nm was measured for the same composition both with 14 ns pulses at 100 Hz and CW radiation; it is roughly 50% higher than in pure GaSe. The two-photon absorption, TPA, (measured with ps pulses at 1064 nm) is lower than for GaSe.

The high nonlinearity of $GaS_{0.4}Se_{0.6}$, together with the improved thermo-mechanical properties, the increased band-gap and damage threshold, and the lower TPA make this crystal attractive for mid-IR optical parametric oscillators and generators operating above 5 μm for the idler and pumped by 1064 nm radiation (nanosecond and picosecond pulses, respectively).

References

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