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Tunable middle infrared downconversion in GaSe and $AgGaS_2$

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Abstract

We report on the generation of middle infrared radiation, tunable over the 3.5–8.5 μm spectral range. This was achieved by difference frequency generation (DFG) in GaSe and $AgGaS_2$ crystals, using the new combination of two solid state lasers; the alexandrite (0.73–0.75 μm) laser and the alexandrite-pumped tunable LiF:F_2^{+**} (0.8–1.2 μm) color center laser.

Keywords: Nonlinear optics; LiF:F_2^{+**} color center lasers; Difference frequency generation; GaSe; $AgGaS_2$

1. Introduction

Middle-infrared solid state laser sources operating in the range of atmospheric transparency (3–5 and 7.5–9 μm) are of great importance for molecular spectroscopy, eye-safe medical laser sources, eye-safe laser, radar and remote sensing of atmospheric constituents, and for numerous military applications such as target designation, obstacle avoidance and infrared countermeasures, etc.

The alexandrite-pumped color center laser has many positive features of a solid state dye-like laser system [1–4]. Advantages include high gain coefficients, broad homogeneous gain profile, low threshold and highly efficient TEM_{00} mode operation with narrow spectral outputs, wide wavelength tunability, compactness, long operational lifetime, rigidity, ease of handling, and insensitivity to the quality of the cavity optical elements, and to the spatial angular and spectral characteristics of the pump source. The system also exhibits virtually no temporal delay between the pump and output pulses, which facilitates nonlinear optical interactions.

The LiF:F_2^{+**} crystals used in the experiments do not exhibit any photodegradation when pumped with radiation at energy densities of up to 5–10 J/cm^2, the limit being set mainly by the optical damage of the LiF crystal [1,2]. The estimated lifetime of these color center crystals exceeds 10 years at room temperature [3]. It was shown in Refs. [1–4] that LiF:F_2^{+**} yields very high pump radiation conversion efficiencies of up to 60%. The LiF color center lasers have narrow spectral widths, achievable, virtually without loss, while preserving a wide region of continuous tuning (up to 2500 cm^{-1}), due to quasi-homogeneous broadening of their absorption and fluorescence bands [1]. The extension of the LiF:F_2^{+**} laser tuning region into the mid infrared region was made possible using GaSe and $AgGaS_2$ crystals. Below, we briefly describe some of the salient properties of these crystals that made them suitable for use in our experiments.

GaSe crystal, first explored as a non-linear optical material in 1972 [5] is a promising crystal for mid-IR frequency conversion [6]. Amongst its notable properties

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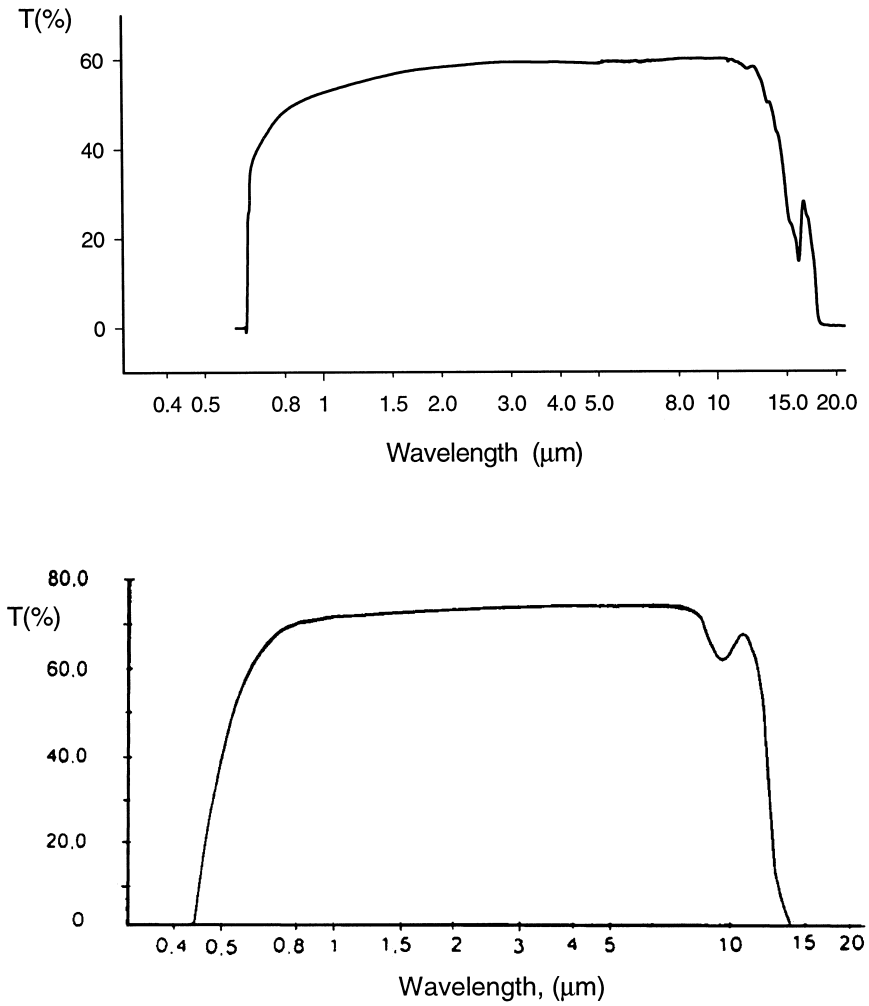

Fig. 1. (a) The transmission curve of GaSe. (b) Transmission curve of $AgGaS_2$.

are its extreme transparency range (0.62–18 μm, Fig. 1a) and its high second order nonlinearity (d_{22} (10.6 μm) = 54 pm/V) [7], which is among the top five measured for birefringent crystals. In addition, due to a very large birefringence of GaSe ($\Delta n \sim 0.35$ at 1 μm) it can satisfy phase matching conditions for a variety of non-linear optical interactions within the medium.

$AgGaS_2$ crystals on the other hand, are widely used for frequency doubling, frequency mixing, and in parametric oscillators, which can produce continuously tunable radiation from 1.0 to 12 μm with suitable pump laser. $AgGaS_2$ is transparent in the region 0.47–13 μm [7] (see Fig. 1b) and has a nonlinear coefficient, d_{36} (10.6 μm) = 11–13 pm/V, and $\Delta n \sim 0.053$ at 1 μm. DFG phase-matching conditions are available for this crystal in a wide spectral range of mixed radiations (0.73–0.75 and 0.8–0.95 μm).

Recent advances in crystal growth, have made available, large single GaSe and $AgGaS_2$ crystals with improved transparency. In this paper, we demonstrate a generation of mid-IR light via DFG in these crystals using an efficient combination of room temperature solid state lasers: the alexandrite laser and the alexandrite-pumped tunable $LiF:F_2^{+**}$ (0.8–1.2 μm) color center laser.

2. Experiment

GaSe is a layered material, which can be cleaved only along the 001 plane (*z*-cut orientation). So, we utilized a GaSe crystal with dimensions,14 mm × 14 mm × 8.3 mm (8.3 mm, along the principal axis), *z*-cut ($\theta_c = 0$) and with no antireflection coatings. The linear absorption (Fig. 1a) was ~ 0.3 cm^{-1} in the visible (700–800 nm) and < 0.03 cm^{-1} in the IR (1.5–12 μm), and the measured damage threshold at 750 nm was 20 MW/cm^2 for 50 ns pulses.

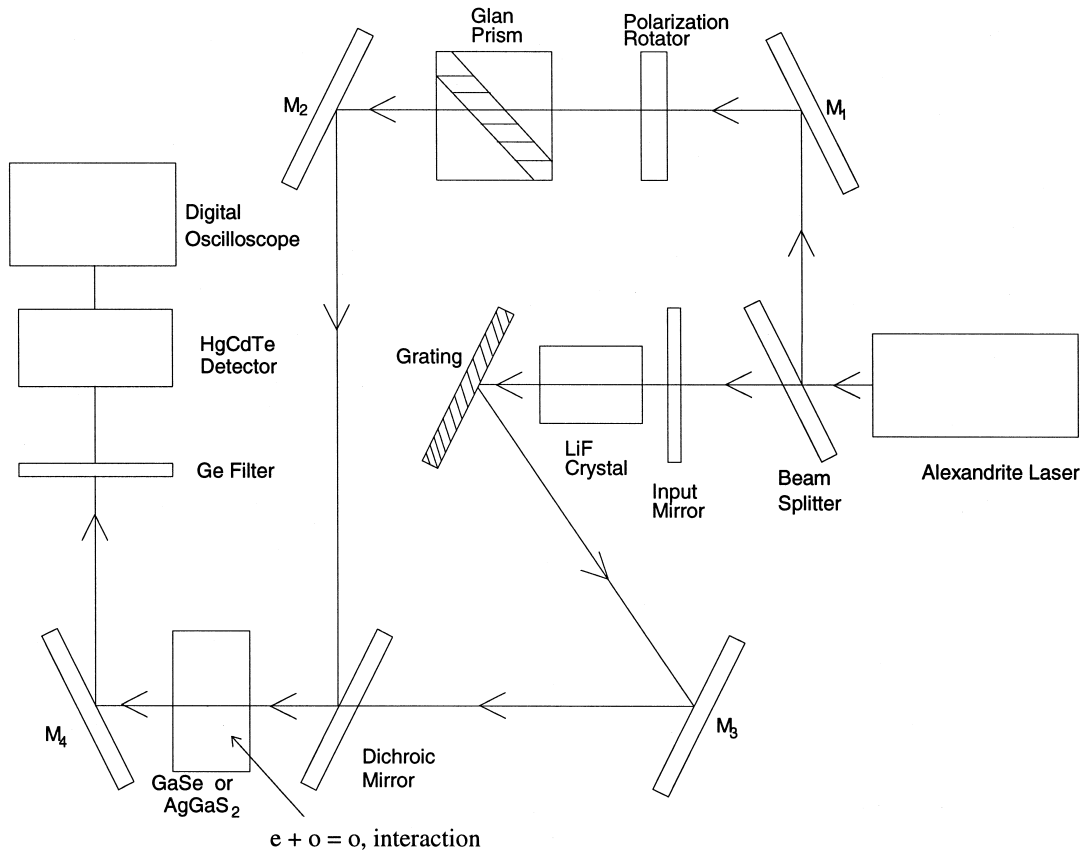

Fig. 2. Schematic for the experimental setup of collinear DFG in GaSe or $AgGaS_2$ crystals. M_{1-4} are mirrors. In the e + o = o interaction, e represents the polarization of the alexandrite laser; o, that of F_2^+ and the resultant o, the polarization of the generated middle IR radiation.

The 16 mm × 14 mm × 38 mm $AgGaS_2$ crystal was cut at $\theta_c = 65°$ for type I phase matching. No anti-reflection coatings were applied to this crystal. It is a research grade crystal with a linear absorption in the 0.7–9 μm range of about 0.01 cm^{-1} (Fig. 1b) and a measured damage threshold at 750 nm of about 25 MW/cm^2 for 50 ns pulses.

Fig. 2 depicts the setup used for the collinear DFG in GaSe and $AgGaS_2$, between the outputs of the alexandrite and the alexandrite-pumped LiF:F_2^{+**} lasers. The LiF:F_2^{+**} laser is pumped with the fundamental output of the alexandrite laser PAL 101 (Light Age Inc.) with an energy of 70–100 mJ, a pulsewidth of 50 ns and a repetition rate of 20 Hz. The LiF:F_2^{+**} laser output energies of up to 20 mJ (measured, using a Molectron, PM10V1 power meter) were obtained in the 0.8–1.2 μm spectral region. The rest of the alexandrite laser radiation was directed into the nonlinear crystal (GaSe or $AgGaS_2$), where it is mixed with the tunable near IR LiF:F_2^{+**} laser radiation.

The polarization of the alexandrite laser beam (diameter ~ 3 mm) was rotated (from horizontal to vertical), and the output power, simultaneously attenuated, using a polarization rotator-Glan prism combination. The alexandrite laser had a power density of about 3 MW/cm^2.

The generated middle IR radiation was detected by means of a cryogenically cooled HgCdTe detector or Joulemeter (J3S-10) combined with a digital oscilloscope. A Ge plate was used to filter out near IR input radiations. The IR wavelength verification was performed with the use of LiF, sapphire, MgF_2 and CaF_2 plates as transmission filters.

3. Discussion

Fig. 3 shows the theoretical (solid lines, calculated for type I DFG, ooe, using the dispersion relations from Ref. [7]), and the experimental angular tuning curves in GaSe, for different wavelengths of the pump radiation (dots, for 740, 745, and 750 nm, respectively), versus the spectral output of the alexandrite-pumped LiF:F_2^{+**} laser. For the 'ooe' interaction, the first symbol denotes the wave with the lower frequency in the interacting waves. The inset, depicts the wavelengths of the infrared DFG signal, λ_1 versus the wavelengths of tunable LiF:F_2^{+**} radiation, λ_2.

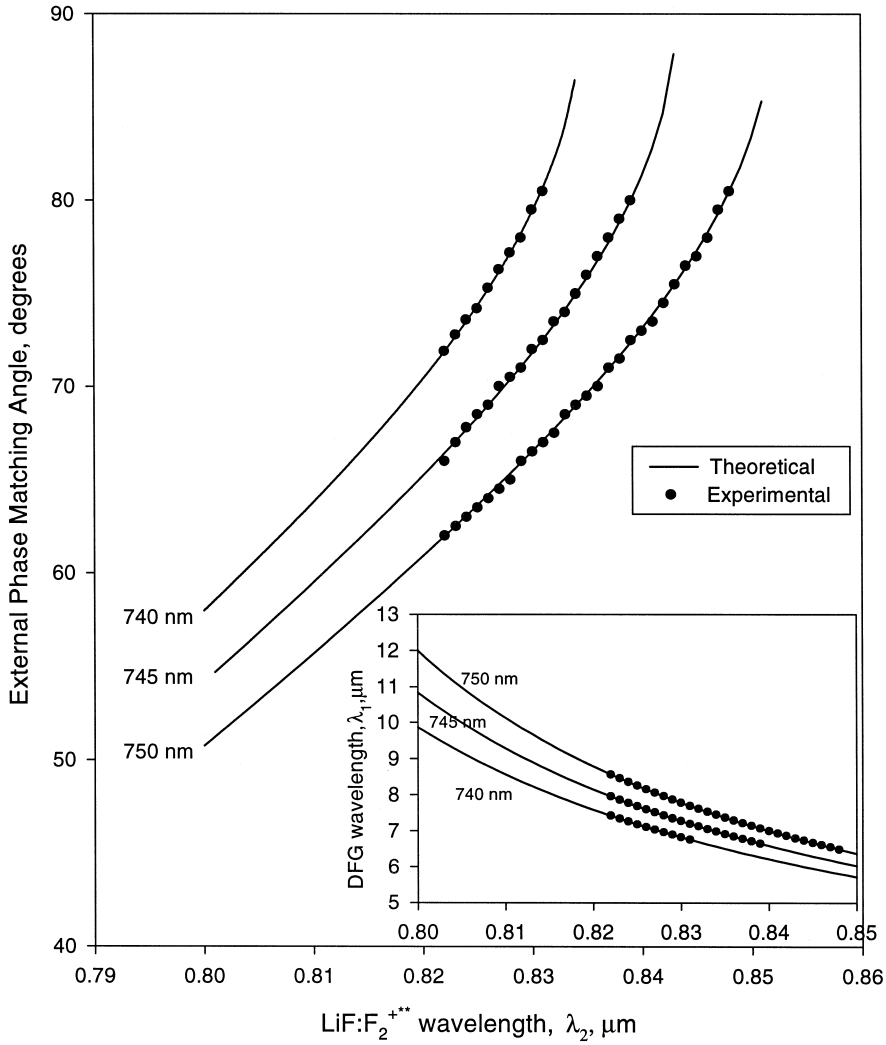

Fig. 3. The external phase matching angle for GaSe. Inset: DFG wavelengths as a function of F_2^{+**} wavelength, at different pump wavelengths.

The theoretical and experimental phase matching curves are in very good agreement over the entire range of the middle IR output. The inset also shows that with the use of GaSe crystal, we experimentally obtained DFG signals over the spectral region 6.5–8.5 μm. Output energies of about 30 μJ and efficiency of 1% were obtained with a very good pulse-to-pulse stability. Using the equations for nonlinear conversion efficiency published in Ref. [7], the maximum DFG conversion efficiency for GaSe was calculated to be about 4%.

Keeping in mind that GaSe cannot be cut at an arbitrary angle, DFG phase matching conditions cannot be realized for the whole range of LiF frequencies. For LiF:F_2^{+**} wavelengths larger than 0.85 μm, the external phase matching angle approaches 90° (Fig. 3). Thus, IR wavelengths shorter than 5.5 μm (Inset, Fig. 3) will be impossible to obtain. In addition, IR wavelengths longer than 9 μm will be difficult to obtain because they correspond to the short wavelength tail ($\lambda < 820$ nm) of the LiF:F_2^{+**} gain profile.

Again, in Fig. 4, using the dispersion relations as given in Ref. [7], we illustrate the theoretically calculated (solid line) and experimental (dots) tuning curves for the pump radiation at a wavelength, λ_3, of 750 nm for $AgGaS_2$ crystal. The inset shows the dependence of the infrared DFG signal, λ_1 on the wavelength, λ_2 of the tunable LiF:F_2^{+**} laser radiation. Here, as in the case of GaSe, there is good agreement between the theoretical and experimental phase matching curves. The slight discrepancy between experimental and theoretical curves may be attributed to the necessity to use improved Sellmeier's equations [7].

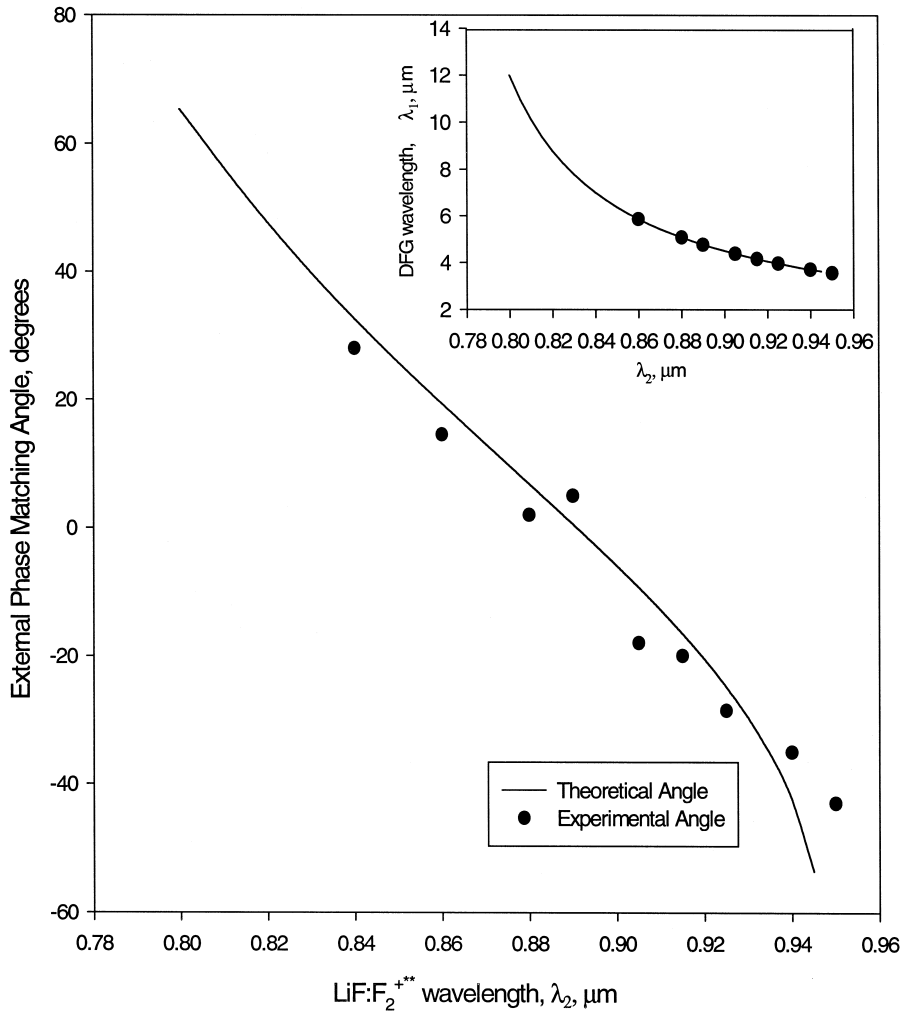

Fig. 4. The external phase matching angle for $AgGaS_2$. Inset: DFG wavelengths as a function of $LiF:F_2^{+**}$ wavelengths.

Similar to the GaSe case, it is difficult to obtain a DFG, longer than 9 μm, because the emission corresponds to the short wavelength tail of the $LiF:F_2^{+**}$ gain profile. In addition to this, the available beam combiner, did not transmit $LiF:F_2^{+**}$ radiation with wavelength shorter than 0.82 μm, which corresponds to an IR wavelength of about 9 μm. We obtained DFG signals (inset, Fig. 4) in $AgGaS_2$ over a spectral region of about 3.6–7 μm, with output powers of about 10 μJ and efficiency $\sim 0.5\%$. As in the case of GaSe, the calculated maximum conversion efficiency for $AgGaS_2$ was about 4%. In principle, by means of mixing the radiations of the alexandrite and $LiF:F_2^{+**}$ lasers, one can cover the spectral range, 3–12 μm.

The discrepancy between the experimental and theoretical conversion efficiencies may be attributed to a number of things, amongst which are: the quality of the bulk crystal used, and the ability to obtain exact spatial overlapping of the interacting waves over the whole length of the nonlinear crystal. Moreover, the interacting beams used, did not have pure planar wavefronts. They had appropriate divergences that were not in accordance with the assumptions of planar wavefronts adopted by Ref. [7] in calculating conversion efficiencies.

4. Conclusion

We have demonstrated tunable middle infrared down-conversion in GaSe and $AgGaS_2$ with up to a 10% quantum conversion efficiency, using the convenient combination of two solid state laser sources, the alexandrite laser and the alexandrite-pumped $LiF:F_2^{+**}$ color center laser. We have shown that both GaSe and $AgGaS_2$, pumped by this laser combination, have potential for use as mid IR sources in the range 3–12 μm for $AgGaS_2$ and 5.5–12 μm for GaSe.

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