# Compact tunable difference-frequency sources in the mid-infrared pumped by single-mode diode lasers

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#### **ABSTRACT**

The suitability of III-V single-mode cw diode lasers for difference-frequency generation of tunable infrared radiation has been explored by mixing a red single-mode diode laser with a tunable single-mode cw Ti:Al<sub>2</sub>O<sub>3</sub> laser in AgGaS<sub>2</sub>. More than 1  $\mu$ W of cw tunable, infrared ( $\lambda \approx 5 \mu$ m), narrow band coherent radiation has been generated by using type I noncritical phase-matching. The feasibility of a more compact, solid-state cw laser source based on the mixing of two single-mode diode lasers (808 and 690 nm) as pump sources in AgGaS<sub>2</sub> has also been demonstrated (infrared power generated  $\approx 3$  nW).

Techniques to increase the infrared difference-frequency output power level such as the use of a high-power optical semiconductor amplifier or an external buildup cavity for the nonlinear mixing crystal have been investigated. As much as 47  $\mu$ W of cw infrared radiation and 89  $\mu$ W of pulsed infrared radiation, tunable around 4.3  $\mu$ m have been generated by mixing the outputs of a high-power tapered semiconductor amplifier at 858 nm and a Ti:Al<sub>2</sub>O<sub>3</sub> laser at 715 nm in AgGaS<sub>2</sub>. The GaAlAs tapered traveling-wave amplifier delivered up to 1.5 W of diffraction-limited cw power into the nonlinear crystal. Recent progress in generating cw infrared radiation near 3.2  $\mu$ m by mixing the outputs of an extended cavity diode laser near 800 nm (pump wave) and a compact diode-pumped Nd:YAG laser at 1064 nm (signal wave) in AgGaS<sub>2</sub> with an external enhancement cavity to resonate the signal wave inside the nonlinear mixing crystal is also described.

# 1. INTRODUCTION

Recent advances in the development of nonlinear optical materials, such as AgGaS<sub>2</sub> and AgGaSe<sub>2</sub>, now offer a convenient technique of generating cw tunable infrared narrow-band coherent radiation over a wide wavelength range (3 to 18 µm) by means of difference-frequency generation (DFG) at room temperature<sup>1-2</sup>. The use of semiconductor diode lasers as pump sources in the nonlinear DFG mixing process is particularly attractive as their compact size and ease of operation allow the construction of a portable and robust mid-infrared laser source especially suitable for environmental remote sensing, pollution detection, chemical analysis, and medical research.

In this paper we describe our recent progress towards the development of such a compact diode-laser based widely tunable cw DFG source with AgGaS<sub>2</sub> as the nonlinear optical material.

# 2. DFG WITH SINGLE-MODE LOW-POWER DIODE LASERS

In a first step<sup>3</sup> a cw tunable Ti:Al<sub>2</sub>O<sub>3</sub> ring laser (Coherent 899-29) operating in the wavelength range from 690 to 840 nm and a single-mode diode laser polarized for  $90^{\circ}$  type I ( $e\rightarrow o+o$ ) phase-matching in AgGaS<sub>2</sub> were spatially overlapped with a polarizing beam splitter (figure 1). The visible beams were focused into the 45 mm long AgGaS<sub>2</sub> crystal to a beam waist of about 40  $\mu$ m by using a 10-cm-focal-length lens (as the diode laser beam is not a Gaussian beam, we define its "waist" as the half-width where the beam intensity is down by  $1/e^2$ ). The infrared radiation generated in the mixing crystal was collimated with a 5-cm-focal-length CaF<sub>2</sub> lens and detected after the germanium filter with a liquid-N<sub>2</sub> cooled photoconductive HgCdTe detector followed by a lock-in amplifier.

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The three diode lasers used in this experiment were unmodified commercial devices operating at 671 nm, 690 nm, and 808 nm. Their spectral linewidths were measured to be 120 MHz, 30 MHz, and 90 MHz, respectively. Each was operated in a single longitudinal mode, so that both spontaneous background and extraneous modes were down by more than 25 dB from the dominant spectral mode. By varying the temperature and the current of the diodes, their emission wavelength could be tuned over about 2 nm. The collimated diode laser beam (Optima 336-1027 collimating lens) with a rectangular beam shape of 1 x 5 mm<sup>2</sup> cross-section was converted to a square-shaped beam with a beam dimension of approximately 5 mm by using an anamorphic prism pair. A 3:1 telescope transformed the diode laser beam size to a dimension comparable to that of the Ti:Al<sub>2</sub>O<sub>3</sub> laser. The spatial mode quality of each diode laser was characterized by passing it through an aperture and measuring the transmitted power. For each diode, approximately ~20% of the light was found to be transmitted through an aperture whose diameter was equal to twice the focused laser beam waist. A TEM<sub>00</sub> Gaussian beam would result in 95% transmission. The diode laser output had a polarization ratio of ~100:1; the appropriate polarization direction for type I phase-matching was chosen by proper mounting of the diodes.

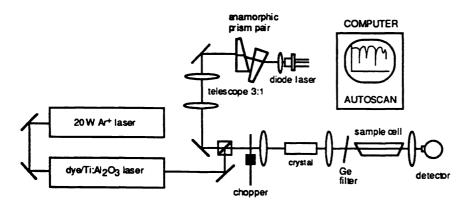


Fig.1: DFG setup using a low-power single-mode diode laser and a Ti:Al<sub>2</sub>O<sub>3</sub> laser as the pump sources and AgGaS<sub>2</sub> as the nonlinear material.

The diode laser wavelengths were determined to within 0.5 nm by using an optical multi-channel analyzer consisting of a 0.32 m Czerny-Turner configuration monochromator with a photodiode array (EG&G Reticon R2512G) attached to the output slit plane of the monochromator. For a signal and pump wavelength of 808.3 nm (Ti:Al<sub>2</sub>O<sub>3</sub> laser) and 690.3 nm (Toshiba TOLD 9140(s) diode laser), respectively, an idler wavelength of 4.73 µm was detected. For 1 W of Ti:Al<sub>2</sub>O<sub>3</sub> laser power and 12.1 mW of diode laser power a DFG power of up to 1.4 µW were measured. The phase-matching bandwidth of the diode/Ti:Al<sub>2</sub>O<sub>3</sub> pump laser configuration was observed to be as large as 600 GHz. This is much larger than the phase-matching bandwidth of about 30 GHz observed for the dye/Ti:Al<sub>2</sub>O<sub>3</sub> pump laser configuration<sup>1-2</sup>. Apparently poor spatial coherence of the diode laser beam results in reduced power, but extended phase-matching range. The high resolution capability of this novel spectroscopic source was demonstrated by obtaining a Doppler-limited CO absorption spectrum around 2119 cm<sup>-1</sup> using a 20 cm absorption cell and about 10 Torr of CO pressure (figure 2). In this case the diode laser wavelength was fixed and the infrared wavelength was tuned by tuning the Ti:Al<sub>2</sub>O<sub>3</sub> laser. The detector's NEP is 3.7·10<sup>-10</sup> W. However, the dominant source of noise in the spectrum were fluctuations of the generated infrared power level resulting from changes of the relative spatial overlap of the visible beam waists in the AgGaS<sub>2</sub> crystal due to mechanical instabilities of the experimental set-up.

A laser diode (Toshiba TOLD 9215(s)) emitting up to 9 mW in a single mode at 671.4 nm was phase-matched with the Ti:Al<sub>2</sub>O<sub>3</sub> laser at 772.8 nm. For 5.2 mW diode laser power and 1.15 W Ti:Al<sub>2</sub>O<sub>3</sub> laser power the infrared power at 1963 cm<sup>-1</sup> was ~ 1.2 μW. In order to investigate the effect of the non-Gaussian diode laser beam on the DFG conversion efficiency, the experiment was repeated using the DCM dye laser set to the same wavelength and power level as the diode laser. The infrared output of the dye/Ti:Al<sub>2</sub>O<sub>3</sub> laser combination, like the diode/Ti:Al<sub>2</sub>O<sub>3</sub> laser combination, showed a linear dependence of IR power upon the input signal power but the slope was a factor of three greater. Thus, as might be expected, the non-Gaussian diode laser mode does not mix as effectively with the pure TEM<sub>00</sub> Gaussian mode of the Ti:Al<sub>2</sub>O<sub>3</sub> signal source as does the pure TEM<sub>00</sub> Gaussian mode from the dye laser.

The feasibility of mixing two single-mode diode lasers in AgGaS<sub>2</sub> to generate tunable infrared radiation was also demonstrated. The radiation from each diode laser was collimated and then converted to a square-shaped beam using anamorphic prism pairs. After being spatially overlapped with a polarizing beam splitter the beams traversed a 3:1 telescope and were focused into the AgGaS<sub>2</sub> nonlinear crystal. Using a Toshiba TOLD 9140(s) diode laser and a Sharp LT010MD diode lasers emitting at 690 nm and 808 nm with power levels of 10.1 mW and 1.93 mW, respectively, we were able to generate up to 3.3 nW of infrared radiation around 2115 cm<sup>-1</sup>.

Using KTP as the nonlinear optical DFG material, Wang and Ohtsu<sup>4</sup> recently generated as much as 300 nW near 1.6 µm from two diode lasers (50 mW each). Accounting for the different experimental conditions such as input power levels, input wavelengths, crystal size, and effective nonlinear coefficient the nonlinear conversion efficiencies of both all-diode-DFG experiments is on the same order of magnitude.

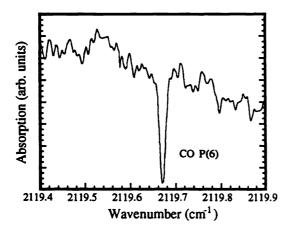


Fig. 2: CO absorption spectrum near 2119 cm<sup>-1</sup> using a 20 cm absorption cell and about 10 Torr of CO pressure.

# 3. DFG WITH HIGH-POWER SEMICONDUCTOR OPTICAL AMPLIFIERS

One way to increase the DFG output power to a level that is useful for spectroscopic applications is the use of optical semiconductor amplifiers to boost the power output of the single-mode diode lasers. Significant progress has been made in obtaining diffraction-limited coherent radiation from high-power broad-area and array laser diodes<sup>5, 6</sup>, and more recently, traveling-wave (TW) amplifiers<sup>7</sup>. When seeded by a single-stripe low-power master diode laser<sup>7</sup>, the semiconductor amplifier has been demonstrated to generate near diffraction-limited, single-longitudinal mode emission required for applications such as nonlinear frequency conversion through second harmonic generation<sup>8</sup> or sum-frequency mixing<sup>9</sup>. Under pulsed operation 11.6 W of peak power was generated by a broad-area amplifier seeded by a laser diode<sup>7</sup>. High-power cw amplifier operation, required for many applications has been considerably more difficult to achieve. Recently, however, 5.25 W of cw emission<sup>10</sup> was obtained from a tapered stripe<sup>7, 11</sup> GaAlAs amplifier seeded with a Ti:Al<sub>2</sub>O<sub>3</sub> laser.

We demonstrated difference-frequency mixing of a high-power GaAlAs tapered traveling-wave semiconductor amplifier with a cw Ti:Al<sub>2</sub>O<sub>3</sub> laser in a 45 mm long AgGaS<sub>2</sub> crystal cut for type I noncritical phase-matching at room temperature. The experimental setup used in this work is shown in figure 3. The master laser was an indexguided diode laser (SDL Inc. Model SDL5410C) emitting up to 130 mW in a single-longitudinal mode around 860 nm with a less than 20 MHz linewidth. By changing the temperature, the laser wavelength was tunable over ~1-2 nm. The amplifier chip was bonded active side down on a heatsink which in turn was attached to a water cooled fixture and allowed unobstructed optical access to both facets. After collimation by a f=2.0 mm lens (0.5 NA), the master laser power passed through a Faraday isolator, and a 3x beam expander. Final coupling of the injected signal into the amplifier was accomplished by a combination of a closely spaced f=15 cm cylindrical lens and a high

numerical aperture f=7.7 mm lens. The cylindrical lens served to move the focused point of the input beam to several hundred  $\mu$ m in front of the facet in the junction plane, while allowing it to coincide with the facet in the perpendicular plane. This highly astigmatic input resulted in a Gaussian input beam width of approximately 150  $\mu$ m in the junction plane and 1  $\mu$ m in the perpendicular plane. With 100 mW of master laser power incident on the amplifier, 38 mW was coupled into the amplifier. The GaAlAs tapered amplifier used in this work was characterized by a 250  $\mu$ m input width, 500  $\mu$ m output width, 1.5 mm length, and a single quantum well separate confinement heterostructure active region. At high currents the amplifier had a peak gain near 860 nm. The facets of the amplifier were antireflection coated ( $R \approx 10^{-3}$ ) at 860 nm for single-pass traveling-wave operation. A second f=7.7 mm lens was used to collimate the amplifier emission, perpendicular to the junction. In the junction plane, the emission was brought to a focus by the f=7.7 mm lens, and collimated by a f=10 cm cylindrical lens.

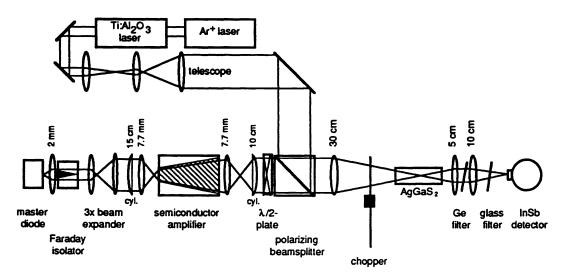


Fig. 3: DFG using of a high-power GaAlAs tapered traveling-wave semiconductor amplifier with a cw Ti:Al<sub>2</sub>O<sub>3</sub> laser in a 45 mm long AgGaS<sub>2</sub> crystal cut for type I noncritical phase-matching at room temperature.

The pump wave was provided by a Ti:Al<sub>2</sub>O<sub>3</sub> ring laser operated at 715 nm, close to its short-wavelength operation limit. In order to maximize the power output, the laser was operated multi-longitudinal-mode without intracavity etalons, resulting in a linewidth of ~1 GHz. Pump and signal wave polarizations were chosen perpendicular to each other for  $90^{\circ}$  type I phase-matching in AgGaS<sub>2</sub>. Both beams were overlapped using a polarizing beam splitter and focused into the 45 mm long AgGaS<sub>2</sub> crystal using a 30-cm-focal-length lens. A three-lens telescope design in the pump laser beam path allowed matching of beam waist widths and focal points for the two beams at the location corresponding to the center of the crystal. The beamwaists were set to ~ 33  $\mu$ m in both vertical and horizontal planes, close to optimum focusing<sup>1</sup>. As measured with a scanning slit beam profiler, the spatial distribution of pump and signal beams was near-Gaussian, where in the case of the signal wave (amplifier) approximately 20% of the focused light was in sidelobes outside the main Gaussian envelope.

The amplifier output power incident on the nonlinear crystal was measured with an integrating sphere detector as a function of the amplifier current. A maximum signal wave power of 1.5 W was obtained under cw operation, and 3.2 W under pulsed operation with 50 ms long pulses and 0.1% duty cycle. After correcting for the 75% capture efficiency of the f=7.7 mm lens and transmission losses of the beamsplitter and other optical components, the actual amplifier output at the maximum cw current of 6 A was 2.5 W. The maximum operating currents for the amplifier were arbitrarily chosen and do not represent a fundamental operating limit of the amplifier.

The difference-frequency long wavelength radiation generated in the  $AgGaS_2$  crystal was collimated with a 5-cm-focal-length  $CaF_2$  lens and focused on a liquid- $N_2$ -cooled photovoltaic InSb detector with a sensitive area element of 4 mm diameter (Graseby Infrared IS-4). An antireflection coated germanium filter was used to block pump and signal waves and pass the difference-frequency radiation. To avoid saturation of the detector-preamplifier combination at the highest powers obtained under pulsed amplifier operation, a glass filter (T = 2.8% at  $4.3 \mu m$ ) in front of the detector

was used to attenuate the detector signal. Phase-matching was found to occur at a wavelength of 714.60 nm and 858.60 nm for pump and signal wave, respectively, corresponding to a generated difference-frequency wavelength of  $\sim$  4.26  $\mu$ m (2350 cm<sup>-1</sup>). The laser wavelengths were determined with a spectral resolution of  $\sim$  0.1 nm and an absolute spectral accuracy of  $\sim$  0.1 nm using a multi-mode probe fiber connected to an optical spectrum analyzer (HP 70951 A Optical Spectrum Analyzer). The phase-matching bandwidth was observed to be on the order of the spectral resolution of the optical spectrum analyzer. Tuning of the infrared wavelength was limited to  $\sim$  25 cm<sup>-1</sup> by the limited temperature tuning range of the master diode laser.

Figure 4 shows the generated infrared DFG power as a function of the diode amplifier output power for cw and pulsed amplifier operation. Values shown are corrected for the 4.3 µm transmission loss of the optical components between the crystal and the detector, but not for the 17% Fresnel reflection at the crystal output facet. For all measurements the Ti:Al<sub>2</sub>O<sub>3</sub> laser output was fixed at a cw power of ~330 mW, and the diode amplifier output power was changed by varying the pump current. The doted line indicates the low-power external slope efficiency of ~ 35 μW/W for the DFG process. Maximum difference-frequency powers of 47 μW and 89 μW were obtained for the cw and pulsed operation, respectively. The drop in experimental slope efficiency at high amplifier power levels is attributed to the degradation in the diode laser beam quality occurring at highest amplifier currents. To evaluate the contribution of thermal effects in the mixing crystal to the drop in slope efficiency at high amplifier power levels the absorption loss in the AgGaS2 crystal was determined at the signal wavelength. From the ratio of transmitted to reflected signal power we found the absorption in AgGaS<sub>2</sub> at 858 nm to be as low as ~0.01 to 0.015 cm<sup>-1</sup>. At high signal power levels, no decay of the infrared pulse amplitude during the 50 ms pulse was observed, indicating that thermal lensing did not contribute to the drop in experimental slope efficiency occurring at the highest pulsed powers. Correction for the Fresnel reflection losses for pump, signal and idler waves at the surfaces of the uncoated AgGaS<sub>2</sub> crystal, the germanium filter and the CaF<sub>2</sub> lenses, results in an internal slope efficiency of  $\sim 65 \mu \text{W/W}$  at 330 mW cw Ti:Al<sub>2</sub>O<sub>3</sub> pump power. No significant improvement in the slope efficiency was observed when changing the Ti:Al<sub>2</sub>O<sub>3</sub> laser from multi-mode to single-mode operation by inserting the intracavity etalons into the ring laser cavity.

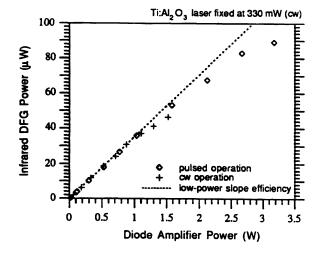


Fig. 4: Generated infrared DFG power as a function of the diode amplifier output power for cw and pulsed amplifier operation. The doted line indicates the low-power external slope efficiency of  $\sim 35 \,\mu\text{W/W}$  for the DFG process.

# 4. RESONANT ENHANCEMENT CAVITY SCHEME FOR DFG

Alternatively, the DFG conversion efficiency can be increased by making use of the high circulating fields present inside optical cavities by placing the nonlinear crystal either in an external (passive) enhancement cavity or in one of the pump laser cavities 13.

We are currently investigating the use of an external enhancement cavity built around the nonlinear optical AgGaS<sub>2</sub> crystal to resonate the signal wave thereby increasing the signal power present inside the mixing crystal. As

the infrared power generated in the nonlinear three-wave mixing process scales with the product of the signal and pump powers, enhancement of the signal power equally increases the nonlinear conversion efficiency. An extended cavity diode laser near 800 nm and a compact diode-pumped ring Nd:YAG laser at 1064 nm are used as the pump and signal sources of the difference-frequency generation (DFG) process, respectively.

Our passive DFG buildup cavity (figure 5) used to resonate the 1064 nm signal wave is a modification of the four-mirror cavity used by Polzik and  $Kimble^{14}$ . It consists of three flat mirrors ( $M_1$ - $M_3$ ) and two lenses ( $f_{L_1}=25$  mm,  $f_{L_2}=50$  mm) together with a 5 mm long AgGaS<sub>2</sub> crystal cut for type I noncritical phase-matching. The linear dimension of the cavity are ~25 cm with the total angle  $\Theta$  on mirror  $M_1$  being  $3^{\circ}$ . Both facets of the nonlinear optical mixing crystal are coated with a three-layer antireflection-coating for low loss at 1064 nm ( $R=4\cdot10^{-5}$ ). In spite of the quality of coatings, passive losses from the lenses (R<0.2%/surface) reduce the cavity buildup relative to bow-tie cavities with curved mirrors<sup>15</sup>. However, the cavity setup shown in figure can be realized almost exclusively with standard Nd:YAG laser optics which was available in our lab. Mirrors  $M_2$  and  $M_3$  are HR-coated (R>99.7%) at 1064 nm. The infrared radiation generated inside the crystal is coupled out through  $M_2$  which was an Al<sub>2</sub>O<sub>3</sub>-substrate HR-coated (R>99.7%) at 1064 with a transmission T=70% at the generated difference-frequency of ~3  $\mu$ m.  $M_1$  is used as the input coupler with R=96% at 1064 nm chosen to impedance-match<sup>16</sup> the buildup cavity to the driving signal power. Approximately 70% of the diode laser power at 795 nm incident on the cavity could be coupled in through  $M_1$ . Using the theoretical approach given in Ref. 16 the enhancement factor for the signal wave power by the passive cavity was calculated to be ~30.

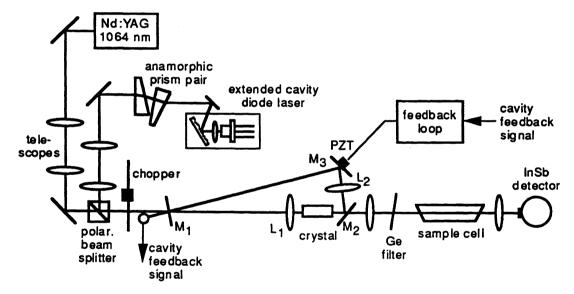


Fig. 5: Use of an 3-mirror external buildup cavity for the nonlinear crystal to increase the infrared DFG output power

So far, a cavity enhancement factor of ~20 was measured for the 1064 nm light. However, difficulties in locking the buildup cavity to the Nd:YAG laser (due to optical feedback and tendency to multi-longitudinal mode operation of the Nd:YAG laser) and in focusing the diode laser beam to a small spot are yet to be solved.

#### 5. CONCLUSION

In conclusion, DFG in AgGaS<sub>2</sub> utilizing diode/Ti:Al<sub>2</sub>O<sub>3</sub> and diode/diode laser input configurations has been demonstrated producing tunable infrared radiation at a wavelength around 5 µm. DFG with diode lasers was not as efficient as that found with dye/Ti:Al<sub>2</sub>O<sub>3</sub> lasers presumably because of the poorer spatial mode quality of the diode lasers. A maximum infrared DFG power of ~3 nW was generated with diode laser input powers of 10 mW plus 2 mW.

As much as 47  $\mu$ W of cw and 89  $\mu$ W of pulsed infrared radiation around 4.3  $\mu$ m have been generated by difference-frequency mixing the outputs of an injection-seeded GaAlAs tapered semiconductor amplifier and a Ti:Al<sub>2</sub>O<sub>3</sub> laser in AgGaS<sub>2</sub> using type I non-critical phase-matching. It is anticipated that the use of an external enhancement cavity for the nonlinear mixing crystal will also result in significantly improved DFG performance.

The developed compact mid-infrared source is promising for wide range of applications including chemical analysis, remote sensing, pollution detection, and medical research.

#### 6. ACKNOWLEDGMENT

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