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MID-INFRARED DIFFERENCE FREQUENCY GENERATION USING OPTICALLY AMPLIFIED DIODE LASERS AS PUMP SOURCES

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Abstract

Recent advances in the development of new nonlinear optical materials, such as $AgGaS_2$ and $AgGaSe_2$, 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). The use of semiconductor diode lasers as pump sources in the nonlinear DFG process is particularly attractive as their compact size and ease of operation allow the construction of a portable and robust infrared laser spectrometer for sensitive environmental monitoring. Approximately 3 nW of cw tunable infrared radiation around 5 μm have been generated by mixing two single-mode diode lasers (808 and 690 nm; total power $\sim$10 mW) as pump sources in $AgGaS_2$. Higher infrared ouput power levels can be achieved by using optical amplifiers to boost the power output of the low-power diode lasers. We report the generation of 47 μW of cw infrared radiation and 89 μW of pulsed infrared radiation, tunable around 4.3 μm by mixing the outputs of a high-power GaAlAs tapered traveling-wave amplifier at 858 nm and a $Ti:Al_2O_3$ laser at 715 nm in $AgGaS_2$. The GaAlAs amplifier delivered up to 1.5 W of diffraction-limited cw power into the nonlinear crystal while the $Ti:Al_2O_3$ laser power was set to 330 mW.

Introduction

There is a need for continuous improvement of convenient laser-based spectroscopic sources in high resolution spectroscopy. Since virtually all fundamental vibrational modes of molecules, molecular ions and free radicals lie in the 2–20 μm wavelength region, tunable monochromatic probe lasers in this spectral region are particularly useful for high-sensitivity, high-resolution ($<$ 100 MHz), and time resolved molecular spectroscopy. In particular, cw laser sources have the greatest potential for providing an optimum combination of spectral control and frequency stability. Although cw tunable laser sources, such as color center lasers,[1] lead salt diode lasers,[2] and CO and CO_2 sideband lasers,[3] exist in this spectral region, each of these lasers suffers from practical drawbacks, such as the requirement of cryogenic cooling, operational wavelength ranges that do not cover regions of interest, and lack of portability and ruggedness.

Instead of the source being based on an infrared laser, tunable radiation can be generated by difference frequency generation (DFG) in a suitable nonlinear medium.[4,5] We demonstrated the operation of a continuously tunable cw DFG spectrometer in the 4 to 9 μm region based on type I noncritical phasematching in the nonlinear material $AgGaS_2$[6,7] pumped by two single-frequency dye/$Ti:Al_2O_3$ lasers.

Recent advances in the development of single-mode III–V diode laser technology[8] offer the possibility of using fixed or tunable diode lasers as pump sources in DFG. Because of the compact size and direct electric excitation of diode lasers, robust, portable spectrometers suitable for wide range of applications, including chemical analysis, pollution detection, and medical research, can be constructed with a diode laser-based DFG source. However, the infrared DFG output power of all-diode laser pumped DFG sources has been limited in the past by the relatively low power output of commercially available single-mode III–V diode lasers. The DFG conversion efficiency can be increased, however, by either using optical semiconductor amplifiers to boost the output power of the single-mode diode lasers or by placing the nonlinear crystal in an external enhancement cavity or in the laser cavity[9] of one of the pump lasers. Significant progress has been made in obtaining diffraction limited coherent radiation from high-power broad-area and array laser diodes,[10,11] and more recently from traveling wave amplifiers.[12] When seeded by a single stripe low power master diode laser, the semiconductor amplifier has been demonstrated to generate near diffraction-limited, single-longitudinal mode emission required for applications such as nonlinear frequency conversion.[13,14] Under pulsed operation 11.6 W of peak power were generated by a broad area amplifier seeded by a laser diode.[12] High-power cw amplifier operation, required for many applications, has been considerably more difficult to achieve. Recently however, 5.25 W of cw emission[15] was obtained from a tapered stripe GaAlAs amplifier[12,16] seeded with a $Ti:Al_2O_3$ laser.

We describe a new difference-frequency mixing spectrometer based on the use of single-mode III–V diode lasers.[17] We report on DFG using a high-power GaAlAs tapered traveling wave semiconductor amplifier and a $Ti:Al_2O_3$ laser in order to obtain improved infrared output power.[18] Potentially, infrared radiation from 3 to 6 μm by DFG in $AgGaS_2$ with type I noncritical phase matching can be generated by mixing III–V diode lasers (AlGaInP, AlGaAs, InGaAs, and InGaAsP).[19]

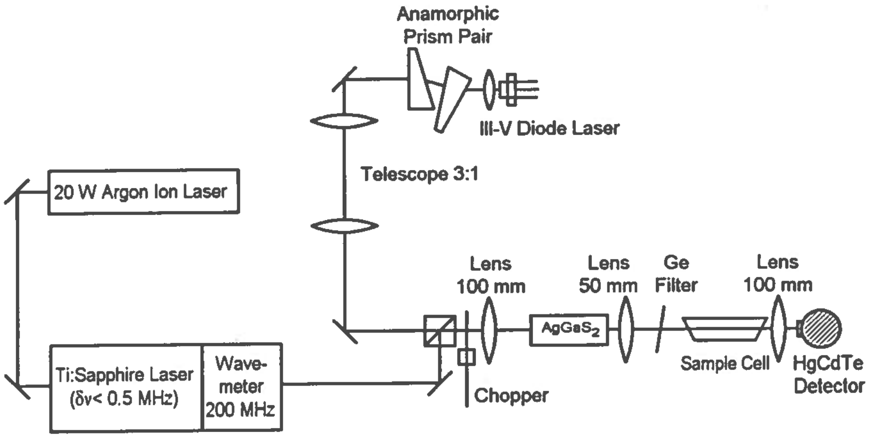

FIG. 1. Experimental setup of the Ti:Al_2O_3/diode laser DFG mixing experiment.

In the development of a compact DFG spectrometer based on two single-mode diode lasers, we made use of an already existing DFG spectrometer pumped by two single-frequency dye/Ti:Al_2O_3 lasers.[6,7] In the first step, the dye laser was replaced by a single-mode diode laser, which was then mixed with the Ti:Al_2O_3 laser. Finally two single-mode diode lasers were mixed in the $AgGaS_2$ crystal.[17]

Diode/Ti:Al_2O_3 laser based DFG

The experimental configuration used in the first step of this work is shown in Fig. 1. The outputs of the cw tunable Ti:Al_2O_3 ring laser (Coherent 899-29) operating in the wavelength range 690-840 nm and a single-mode diode laser polarized for 90° type I ($e \rightarrow o + o$) phasematching in $AgGaS_2$ were spatially overlapped with a polarization cube. The visible beams were focused into a 45-mm-long $AgGaS_2$ crystal down to a beam waist of $\sim$40 μm with a plano-convex lens with a focal length of 10 cm (because the diode laser beam is not a Gaussian beam, we define its waist as the halfwidth were the beam intensity is decreased by $1/e^2$). The infrared radiation generated in the crystal was collimated with a 5-cm focal-length plano-convex CaF_2 lens; an antireflection-coated germanium filter was used to block the pump and signal waves and pass the difference-frequency radiation. The generated radiation was detected using a liquid-N_2-cooled photo conductive HgCdTe detector followed by a lock-in amplifier. The detector has a square active area of 1 mm^2. The responsivity of the detector-preamplifier combination could be toggled between 1.2×10^5 and 1.2×10^6 V/W (as measured by the manufacturer at $\lambda_{\max} \approx$ 10–11 μm; the relative responsivity at the actual infrared wavelength of 5 μm is expected to be $\sim$42% of this value).

The three diode lasers used were unmodified commercial devices operating at 671, 690, and 808 nm. Their spectral linewidths were measured to be 120, 30, and 90 MHz, respectively. Each was operated in a single longitudinal mode, so that both spontaneous background and extraneous modes were down by >25 dB from the dominant spectral mode. Varying the temperature and the current of the diodes permitted their emission wavelength to be tuned over $\sim$2 nm. This tuning range corresponds to an $\sim$18-wavenumber tuning range for the resulting difference frequency. The temperature control limits the frequency drift to 100 MHz.[20] The amplitude stability was measured with a fast photodiode to be better than ±0.1% on time scales down to a few microseconds. The collimated diode laser beam (Optima 336-1027 collimating lens), which had a rectangular beam shape of 1 mm × 5 mm cross section, was converted to a square beam with a beam dimension of $\sim$5 mm with an anamorphic prism pair. A 3:1 telescope transformed the diode laser beam size to a dimension comparable with that of the Ti:Al_2O_3 laser. We characterized the spatial mode quality of each diode laser by passing it through an aperture and measuring the transmitted power. For each diode $\sim$20% of the light was found to be transmitted through an aperture whose diameter was equal twice the focused laser beam waist. A TEM_{00} Gaussian beam would result in 95% transmission. The diode laser output has a polarization ratio of $\sim$100:1; the appropriate direction for 90° type I phase matching was chosen by proper mounting of the diode lasers.

The diode laser wavelengths were determined to within 0.5 nm with an optical multichannel analyzer consisting of a 0.32-m Czerny-Turner configuration monochromator (Instruments SA, HR-320) with a photodiode array (EG&G, Reticon R2512G) attached to the output slit plane of the monochromator. For signal and pump wavelengths of 808.3 nm (Ti:Al_2O_3 laser) and 690.3 nm (Toshiba TOLD 9140(s) diode laser), respectively, an idler wavelength of 4.73 μm (2115cm^{-1}) was detected. For 1 W of Ti:Al_2O_3 laser power and 12.1 mW of diode laser power, a DFG power of as much as 1.4 μW was measured.

We demonstrated the high resolution capability of this novel spectroscopic source by obtaining a Doppler-limited CO absorption spectrum of $\sim$2119 cm^{-1} with a 20-cm absorption cell and $\sim$10 Torr of CO pressure, as depicted in Fig. 2. In this case the diode laser wavelength was fixed, and the infrared wavelength was varied by tuning the Ti:Al_2O_3 laser.

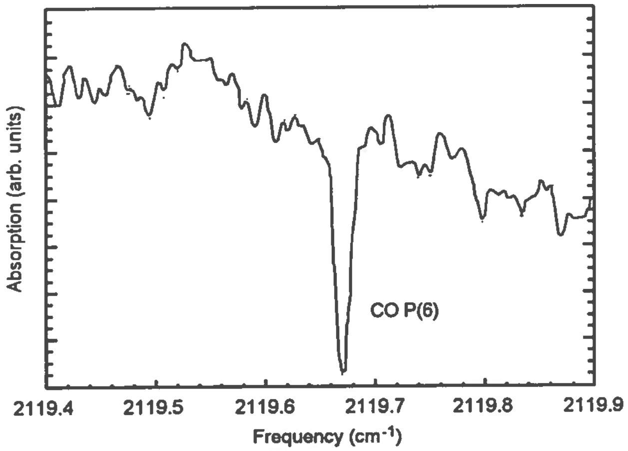

FIG. 2. CO absorption spectrum near 2120 cm^{-1} detected with the diode/Ti:Al$_2$O$_3$ laser configuration.

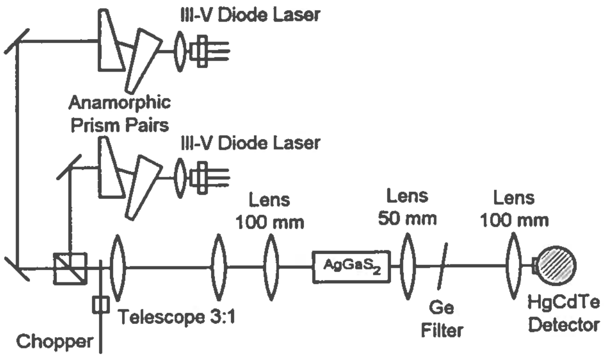

FIG. 3. Experimental setup of the diode/diode laser DFG mixing experiment.

The noise-equivalent power of the detector is 3.7×10^{-10} W. However, the dominant source of noise in the spectrum was fluctuations in the generated infrared power level resulting from changes in the spatial overlap of the visible beam waists in the AgGaS$_2$ crystal resulting from mechanical instabilities of the experimental setup.

Another laser diode (Toshiba TOLD 9215(s)), emitting up to 9 mW of power in a single mode at 671.4 nm was phase matched with the Ti:Al$_2$O$_3$ laser at 772.8 nm. For 5.2 mW diode laser power and 1.15 W Ti:Al$_2$O$_3$ laser power, the infrared power at 5.12 μm (1954.3 cm^{-1}) was ~1.2 μW. To investigate the effect of the non-Gaussian diode laser beam on the DFG conversion efficiency, we repeated the experiment with a DCM dye laser set to same wavelength and power level as the diode laser. The infrared output of the dye/Ti:Al$_2$O$_3$ laser combination, like that of the diode/Ti:Al$_2$O$_3$ laser combination showed a linear dependence of infrared power on the input signal power, but the slope was a factor of 3 greater. Thus, as might be expected, the non-Gaussian diode laser mode does not mix as effectively with the pure TEM$_{00}$ Gaussian mode of the Ti:Al$_2$O$_3$ signal source as does the pure TEM$_{00}$ Gaussian mode from the dye laser. In future research, better characterization of the diode laser beam spatial modes will help to clarify the consequences of using non-Gaussian beams.

Diode/diode laser based DFG

Subsequently, the feasibility of mixing two single-mode diode lasers in AgGaS$_2$ to generate tunable infrared radiation was also demonstrated.[17] Figure 3 shows a scheme of the experimental setup used in an all-diode-laser DFG experiment. The radiation from each diode laser was collimated and then converted to a square beam with anamorphic prism pairs. After being spatially overlapped by a polarization cube, the beams transversed a 3:1 telescope and were focused into the AgGaS$_2$ nonlinear crystal. Using a Toshiba TOLD 9140(s) diode laser and a Sharp LT010MD diode laser emitting at 690 and 808 nm with power levels of 10.1 and 1.93 mW respectively, we were able to generate as much as 3.3 nW of infrared radiation at 4.7 μm (~2115 cm^{-1}).

With this system, the first major step towards our goal of an diode-laser based DFG spectrometer was accomplished. However, due to the relatively low output power of commercially available single-mode diode lasers the DFG power dropped almost to the noise-equivalent-power level of standard IR detectors, which is too small for practical applications. Therefore, the next step was to improve the DFG power to a spectroscopically useful level.

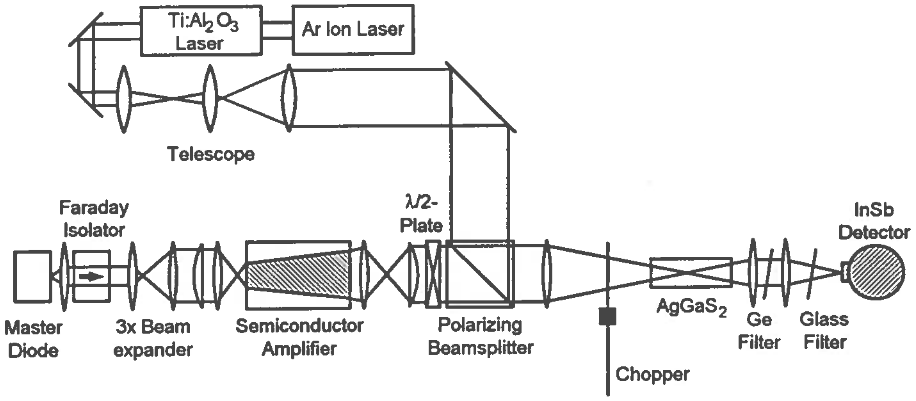

FIG. 4. Experimental setup of the GaAlAs tapered amplifier/Ti:Al_2O_3 laser DFG experiment. The amplifier was seeded by a cw single-mode diode laser.

DFG using optical semiconductor amplifier

We describe difference-frequency mixing of a high power GaAlAs tapered traveling-wave semiconductor amplifier with a cw Ti:Al_2O_3 laser in a 45-mm-long $AgGaS_2$ crystal cut for type I noncritical phasematching at room temperature.[18] The amplifier was injection seeded by a single-mode index-guided master diode laser. The amplifier was operated in cw and pulsed mode to permit an investigation of the DFG conversion efficiency in $AgGaS_2$ at high power levels.

The experimental setup used in this research is shown in Fig. 4. The master laser was an index guided diode laser (SDL Inc., Model SDL5410C) emitting as much as 130 mW of power in a single longitudinal mode around 860 nm with a linewidth of less than 20 MHz. By changing the temperature, we could tune the laser wavelength over ~1–2 nm. The amplifier chip was bonded active side down on a heat sink, which in turn was attached to a water-cooled fixture and permitted unobstructed optical access to both facets. After collimation by an $f = 2.0$ mm lens (Corning, Model 350150, 0.5 N.A.), the master laser beam passed through a Faraday isolator and a 3× beam expander composed of an $f = 8.6$ mm lens and $f = 25.6$ mm lens. Final coupling of the injected signal into the amplifier was accomplished by a combination of a closely (3 cm) spaced $f = 15$ cm cylindrical lens and a high numerical aperture $f = 7.7$ mm lens. The cylindrical lens served to move the focus in front of the facet in the junction plane.[12] This highly astigmatic input resulted in Gaussian input beam widths of approximately 150 μm in the junction plane and 1 μm in the perpendicular plane. With 100 mW of master laser power incident upon the amplifier, 38 mW of power was coupled into the amplifier. The coupling efficiency was low compared to the 60–70% typically measured with a Ti:Al_2O_3 laser, a fact attributed to underfilling of the 7.7-mm focusing lens in the plane perpendicular to the junction by the non circular laser diode beam. The GaAlAs tapered amplifier used in this research is characterized by a 250 μm input width, a 500 μm output width, a 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, positioned one focal length away from the facet, was used to collimate the amplifier emission perpendicular to the junction. In the junction plane, a focused spot approximately 8 cm to the right of the lens was formed, corresponding to a magnified image of the virtual input point source in front of the amplifier. Finally the output beam was collimated by an $f = 10$ cm cylindrical lens positioned one focal length from the focused spot. This optical arrangement resulted in a collimated output beam with nearly square cross section.

The pump wave was provided by a Ti:Al_2O_3 ring laser (Coherent, Model 899) operated at 715 nm, close to its short wavelength operation limit. In order to maximize the power output, we operated the laser multi-longitudinal mode without intracavity étalons, resulting in a linewidth of ~1 GHz. Pump and signal wave polarizations were chosen perpendicular to each other for 90° type I phase matching in $AgGaS_2$. Both beams were overlapped by a polarizing beam splitter and focused into the 45-mm-long $AgGaS_2$ crystal with a 30-cm focal-length lens. A scanning slit beam profiler (Merchantek Inc., Beamscope) was used to measure and optimize the beam-waist widths as well as the lateral and longitudinal beam overlap. A three-lens telescope design in the pump laser beam path permitted matching of beam-waist widths and focal points for the two beams at the location corresponding to the center of the crystal. The beam waists (half-width at $1/e^2$ maximum) were set to ~33 μm in both vertical and horizontal planes, close to optimum focusing.[6] As measured with the beam profiler, the spatial distributions of pump and signal beams were 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.

We obtained a maximum signal wave power of 1.5 W, incident upon the nonlinear crystal, under cw operation and 3.2 W under pulsed operation with 50-μs-long pulses and a 0.1% duty cycle. After correcting for the 75% capture efficiency of the $f = 7.7$ mm lens and transmission losses of the beam splitter and other optical components, we measured the actual

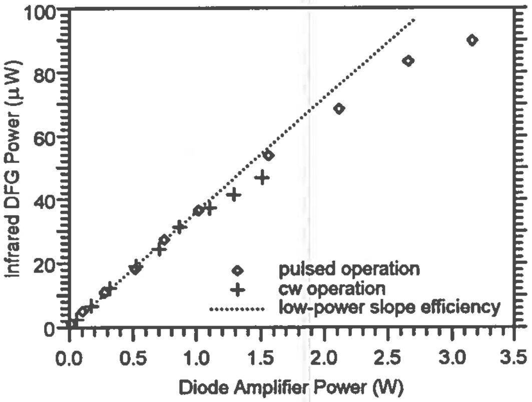

FIG. 5. Generated infrared DFG power at $\lambda \approx 4.3\,\mu$m as a function of the amplifier power incident upon the crystal for cw and pulsed modes of operation. The Ti:Al_2O_3 laser cw power was fixed at 330 mW.

amplifier output at the maximum cw current of 6 A to be 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 onto a liquid-N_2-cooled photovoltaic InSb detector with a sensitive area element of 4-mm diameter (Graseby Infrared IS-4). To avoid saturation of the detector-preamplifier combination at the highest powers obtained under pulsed amplifier operation, we placed a glass filter ($T = 2.8\%$ at $4.3\,\mu$m) in front of the detector to attenuate the detector signal. Phase matching was found to occur at wavelengths of 714.6 and 858.6 nm for the pump and signal wavelength, respectively, corresponding to a generated difference-frequency wavelength of $\sim 4.26\,\mu$m ($2350\,\text{cm}^{-1}$). The laser wavelengths were determined with a spectral resolution of 0.1 nm and an absolute spectral accuracy of ~0.1 nm with a multimode probe fiber connected to an optical spectrum analyzer (HP70951A 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 $\sim 25\,\text{cm}^{-1}$ by the limited temperature tuning range of the master diode laser.

Figure 5 shows the generated infrared DFG power as a function of the diode amplifier power incident upon the nonlinear crystal for cw and pulsed operation. Values shown are corrected for the $4.3\,\mu$m 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_2O_3 laser output was fixed at a cw power of ~330 mW and we changed the diode amplifier output by varying the pump current. The dotted line indicates the low-power external slope efficiency of $\sim 35\,\mu$W/W for the DFG process. Maximum difference-frequency powers of 47 and $89\,\mu$W were obtained for cw and pulsed operation, respectively. The drop in experimental slope efficiency at high amplifier power levels is attributed to degradation in the diode laser beam quality occurring at the 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, we determined the absorption loss in the $AgGaS_2$ crystal at the signal wavelength. From the ratio of transmitted to reflected signal power we found the absorption in $AgGaS_2$ at 858 nm to be as low as ~1 to $1.5\,\%\text{cm}^{-1}$. At high signal power levels, no decay of the infrared pulse amplitude during the 50-μs 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 $AgGaS_2$ crystal, the germanium filter, and the CaF_2 lenses results in an internal slope efficiency of $\sim 65\,\mu$W/W at 330 mW of cw Ti:Al_2O_3 pump power.

Conclusions

Difference-frequency mixing in $AgGaS_2$ with diode/Ti:Al_2O_3 and diode/diode pump laser configurations has been demonstrated to produce tunable infrared radiation at a wavelength of $\sim 5\,\mu$m.[17] Output power levels of $\sim 1.2\,\mu$W and 3.3 nW have been obtained in the case of diode/Ti:Al_2O_3 and diode/diode pump laser configurations, respectively. DFG with diode lasers was not as efficient as with dye/Ti:Al_2O_3 lasers, presumably because of the poorer spatial mode quality of the diode lasers.

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 (the signal source) and a Ti:Al_2O_3 ring laser (the pump source) in $AgGaS_2$.[18] An internal slope efficiency of $65\,\mu$W/W has been achieved at 330 mW of cw Ti:Al_2O_3 pump power.

Further improvements in system efficiency and compactness should be possible through the use of an all-diode system, for which combinations of GaAlAs amplifiers in the range of 750–860 nm and InGaAsP amplifiers below 700 nm could be used to achieve the specific midinfrared wavelengths. The midinfrared source presented is promising for a wide range of applications, including chemical analysis, pollution detection and medical research.

Acknowledgments

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