Difference-frequency generation in $AgGaS_2$ by use of single-mode diode-laser pump sources

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We explored the suitability of visible III–V single-mode cw diodes for difference-frequency generation of tunable infrared radiation by mixing a red single-mode cw III–V diode laser with a tunable single-frequency cw Ti:sapphire laser in $AgGaS_2$. More than 1 μW of cw tunable, infrared ($\lambda \approx 5$ μm), narrow-band coherent radiation was generated with type I noncritical phase matching. The wavelength and output-power characteristics of this novel tunable all-solid-state laser source are described, and we demonstrate the applicability of the source to high-resolution molecular spectroscopy by obtaining a test spectrum. The feasibility of a more compact solid-state cw laser spectrometer based on the mixing of two single-mode diode lasers (808 and 690 nm) as pump sources in $AgGaS_2$ is shown (infrared power generated $\approx$3 nW).

There is a need for continuous improvement of convenient laser-based spectroscopic sources in high-resolution spectroscopy. Because virtually all fundamental vibrational modes of molecules and molecular ions lie in the 2- to 20-μm wavelength region, tunable monochromatic probe lasers in this wavelength 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 type of laser suffers from practical drawbacks, such as the requirement of cryogenic cooling, operational wavelength ranges that do not reach regions of great interest, incomplete coverage of their nominal operational wavelength range, and lack of portability and ruggedness.

Instead of the source's being based on an infrared laser, tunable radiation can be generated by difference-frequency generation (DFG) in a suitable nonlinear medium. The spectroscopic source originally developed by Pine,[4] in which an Ar^+ laser is mixed with a cw dye laser in $LiNbO_3$, has proved useful for high-resolution spectroscopy but is limited to wavelengths <4 μm by the infrared transmission characteristics in $LiNbO_3$. Using $LiIO_3$ as the nonlinear medium, Oka and co-workers extended the long-wavelength limit for cw DFG spectroscopic sources to nearly 5 μm.[5] Recently we demonstrated the operation of continuously tunable cw DFG spectrometer in the 4- to 9-μm region based on type I noncritical phase matching in $AgGaS_2$[6,7] pumped by two single-frequency dye/Ti:sapphire lasers.

Recent advances in the development of high-power single-mode III–V diode-laser technology[8] offer the possibility of using fixed or tunable cw or pulsed diode lasers as pump sources in DFG. Because of the compact size and direct electric excitation of diode lasers, robust, portable spectrometers especially suitable for applications in sensitive and selective environmental monitoring of trace species can be constructed with a diode-laser-based DFG source. In this Letter we describe a new difference-frequency mixing spectrometer based on the use of single-mode III–V diode lasers. 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).[9]

The usefulness of single-mode diode lasers for nonlinear frequency-conversion experiments has been demonstrated. Efficient frequency doubling of AlGaAs diode-laser emission with a resonator for enhancement of the infrared light field within the nonlinear material has been reported in $KNbO_3$.[10,11] In addition, efficient sum-frequency generation of blue light has been accomplished by mixing a Nd:YAG laser with a single-mode AlGaAs diode laser in a monolithic $KNbO_3$ or KTP resonator.[12,13] Sum-frequency generation with a single-mode diode laser has also been obtained without an enhancement cavity.[12,14]

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

The experimental configuration used in the first step of this work is shown in Fig. 1. The outputs of the cw tunable Ti:sapphire ring laser (Coherent 899-29) operating in the wavelength range 690–840 nm and a single-mode diode laser polarized for 90° type I ($e \rightarrow o + o$) phase matching in $AgGaS_2$ were spatially overlapped with a polarization cube. The visible beams were focused into a 45-mm-long $AgGaS_2$ crystal to a beam waist of ~40 μm with a plano-convex lens with a focal length of

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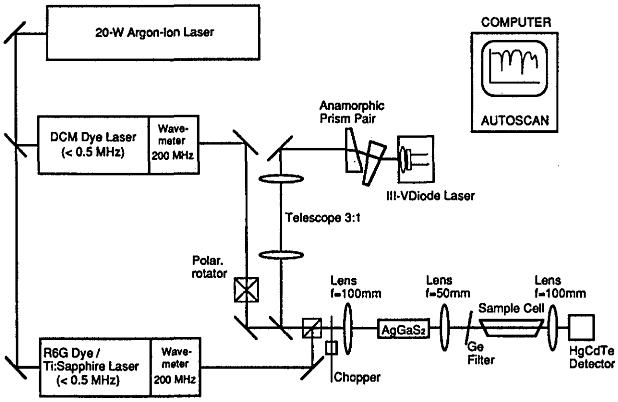

Fig. 1. Experimental setup of the Ti:sapphire/diode laser DFG mixing experiment. Optionally, the diode laser can be replaced by a DCM dye laser.

10 cm (because the diode-laser beam is not a Gaussian beam, we define its waist as the half-width where 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 and detected after the germanium filter with a liquid-N_2-cooled photoconductive 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 ~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 ~2 nm. This tuning range corresponds to an ~18-wave-number tuning range for the resulting difference frequency. The temperature control limits the frequency drift to 100 MHz.[15] 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 ~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:sapphire 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, ~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_{00} Gaussian beam would result in 95% transmission. The diode-laser output has a polarization ratio of ~100:1; the appropriate polarization 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 monochrometer. For signal and pump wavelengths of 808.3 nm (Ti:sapphire laser) and 690.3 nm [Toshiba TOLD 9140(s) diode laser], respectively, an idler wavelength of 4.73 μm was detected. Figure 2 shows the generated DFG power as a function of the input signal power. In this measurement, the diode-laser power was fixed at 12.1 mW. The Ti:sapphire laser power was varied by use of neutral-density filters. Laser powers were measured after the polarization cube with a photodiode calibrated against factory standards. Figure 2 shows the idler power increasing linearly with the signal power, as expected in the low-conversion regime. For 1 W of Ti:sapphire laser power and 12.1 mW of diode laser power, a DFG power of as much as 1.4 μW was measured. The fluctuations of the experimentally determined points in Fig. 2 resulted from changes in the relative spatial overlap of the visible beam waists inside the nonlinear crystal caused by the insertion of the neutral-density filters. The phase-matching bandwidth of the diode/Ti:sapphire pump laser configuration was observed to be as large as 20 cm^{-1}. This is much larger than the phase-matching bandwidth of ~1 cm^{-1} observed for the dye/Ti:sapphire pump laser configuration. Apparently, poor spatial coherence of the diode laser beam results in reduced power but an extended phase-matching range. We demonstrated the high-resolution capability of this novel spectroscopic source by obtaining a Doppler-limited CO absorption spectrum of ~2119 cm^{-1} with a 20-cm absorption cell and ~10 Torr of CO pressure, as depicted in Fig. 3. In this case, the diode-laser wavelength was fixed, and the infrared wavelength was tuned by tuning the Ti:sapphire laser. The detector's noise-equivalent power is 3.7×10^{-10} W. However, the dominant source of noise in the spectrum was fluctuations of the generated infrared power level resulting from changes in the relative spatial overlap of the visible beam waists

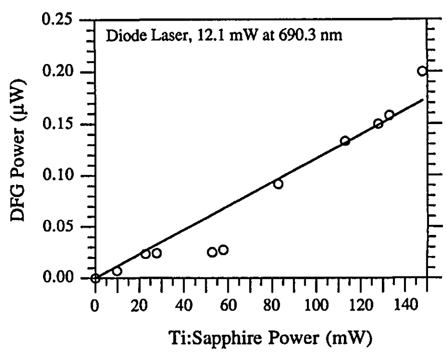

Fig. 2. Infrared DFG power as a function of the Ti:sapphire laser power. The diode laser emitted 12.1 mW of power in a single mode at 690.3 nm.

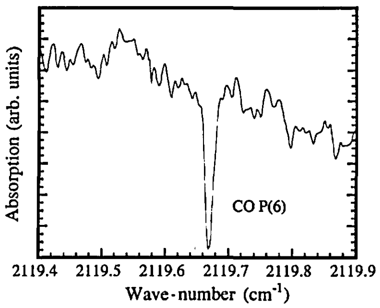

Fig. 3. CO absorption spectrum near 2119 cm^{-1} detected with the diode/Ti:sapphire laser pump configuration.

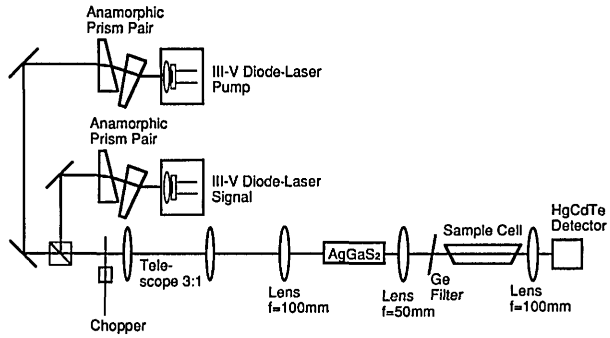

Fig. 4. All-solid-state laser-pumped compact DFG spectrometer with two single-mode diode lasers.

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:sapphire laser at 772.8 nm. For 5.2-mW diode-laser power and 1.15-W Ti:sapphire laser power, the infrared power at 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 the DCM dye laser set to the same wavelength and power level as the diode laser. The infrared output of the dye/Ti:sapphire laser combination, like that of the diode/Ti:sapphire 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 so effectively with the pure TEM_{00} Gaussian mode of the Ti:sapphire 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.

Finally, the feasibility of mixing two single-mode diode lasers in $AgGaS_2$ to generate tunable infrared radiation was also demonstrated. Figure 4 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 traversed 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 ~2115 cm^{-1}.

In conclusion, difference-frequency mixing in $AgGaS_2$ with diode/Ti:sapphire and a diode/diode pump laser configurations has been demonstrated for what is to our knowledge the first time to produce tunable infrared radiation at a wavelength of ~5 μm. DFG with diode lasers was not so efficient as that found with dye/Ti:sapphire lasers, presumably because of the poorer spatial mode quality of the diode lasers. The optical setup used in this research, to focus the visible light into the crystal and to collimate the generated infrared radiation, was optimized for the dye/Ti:sapphire pump laser configuration. By optimization of the spatial overlap of the diode-laser beams in terms of the size and location of the beam waists in the crystal and by use of broadband antireflection-coated optics, we expect much higher visible-to-infrared conversion efficiencies.

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