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Mid-infrared difference-frequency generation source pumped by a 1.1–1.5-μm dual-wavelength fiber amplifier for trace-gas detection

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Continuous-wave mid-infrared radiation near 3.5 μ m is generated by difference-frequency mixing of the output of a compact 1.1–1.5- μ m dual-wavelength fiber amplifier in periodically poled LiNbO₃. The diode side-pumped amplifier is constructed with double-cladding Yb-doped fiber followed by single-mode Er/Yb codoped fiber. Output powers of as much as 11 μ W at 3.4 μ m are obtained, and spectroscopic detection of CH₄ and H₂CO is demonstrated. © 1998 Optical Society of America

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Various approaches to difference-frequency generation (DFG) of $3-5-\mu m$ radiation for use in compact optical gas sensors were recently described.¹ Typically these approaches use a periodically poled LiNbO₃ (PPLN) crystal, a low-power tunable laser diode, and a higher-power laser such as a diode-pumped Nd:YAG or a large-aperture semiconductor amplifier as pump sources. It is necessary to use the latter to achieve the microwatt DFG power levels typically required in practical gas sensors. To decrease the overall system cost and sensitivity to environmental disturbances that can cause misalignment of bulk optical components, the use of fiber beam delivery and fiber amplifiers as one of the pump sources was recently implemented.^{2,3}

Here we describe a compact DFG source based on mixing of 1.1- and 1.5- μ m outputs of a dual-wavelength fiber amplifier. The amplifier uses a high-power Ybdoped fiber to produce optical gain near 1.1 μ m and to pump an Er/Yb codoped fiber for amplification at 1.5 μ m. With a 19-mm-long PPLN crystal, a maximum DFG power of 11 μ W is measured at 3.4 μ m, and wavelength tunability is demonstrated from 3.3 to $3.5 \ \mu m$ (pumping at 1064 nm). The source offers the advantages of compactness, low cost, a robust all-fiber design, and a large tuning range made possible by wide gain bandwidth of the fiber amplifier. In addition, the large optical gains provided by the amplifiers make it possible to use low-power diode seed lasers, and spatial overlap between the two pump beams in the crystal is ensured by the single fiber output of the pump source. Using this DFG source, we demonstrated spectroscopic detection of CH_4 and H_2CO .

As shown in Fig. 1, the key elements of the DFG source include a $1.5 \ \mu m$ tunable external-cavity laser diode (Photonetics), a fiber pigtailed 1083-nm distributed Bragg reflector (DBR) laser diode (SDL, Inc.), a $1064 \ -1550 \ nm$ fiber wavelength-division multiplexer combiner (WDM), the dual-wavelength fiber amplifier, and the PPLN crystal. DFG was also carried out with a 1064-nm diode-pumped Nd:YAG seed laser. Powers coupled into the fiber were 20 and

2.7 mW from the 1083-nm and $1.5-\mu$ m seed lasers, respectively. A 10-dBm fiber preamplifier was used to increase the $1.5-\mu$ m seed power. Slow wavelength tuning could be obtained with the external-cavity $1.5-\mu$ m laser, whereas the rapid tuning required for spectroscopic measurements was obtained by application for a 100-Hz sawtooth modulation current to the 1083-nm distributed Bragg reflector laser.

The dual-wavelength fiber amplifier used a 7.2-mlong Yb-doped double-cladding fiber (Lucent Technologies) with a 0.45-N.A. polymer outer cladding, a nearly hexagonal 131- μ m inner cladding, a 6- μ m-diameter core with a N.A. of 0.16, and an absorption of 2.2 dB/m at 975 nm. The fiber was side-pumped with a 975-nm, 2-W (at 3.0 A) broad stripe diode (SLI, Inc.) by a Vgroove pump coupling technique,⁴ with pump light counterpropagating relative to the signal. The double-cladding fiber's ends were fusion spliced to conventional single-mode fiber (Flexcore 1060) pigtails. When it was seeded with 20 mW of power at 1083 nm or 40 mW at 1064 nm, the saturated amplifier emitted 780 mW. Under maximum pump power conditions the amplifier exhibited⁵ a small-signal gain of 47 dB



Fig. 1. Schematic of the dual-wavelength amplifierpumped DFG source-based gas sensor: FI's, Faraday isolators.

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at 1064 nm and a gain of more than 40 dB from 1040 to 1100 nm. The Yb-doped fiber had a negligible propagation loss at $1.5 \ \mu$ m.

To achieve gain at 1.5 μ m we fusion spliced a 5-mlong section of single-mode Er/Yb codoped fiber⁶ (Lucent Technologies), with a N.A. of 0.16 and a core diameter of 5.1 μ m, to the counterpropagating output pigtail of the Yb-doped fiber amplifier. Because of efficient Yb-to-Er energy transfer, pumping at the long-wavelength tail of the 975-nm Yb absorption peak yielded an Er population inversion (the respective fiber absorption coefficients are $\alpha_{975 \text{ nm}} = 560 \text{ dB/m}$ and $\alpha_{1535 \text{ nm}} = 20 \text{ dB/m}$). The small-signal Yb/Er fiber absorption coefficient versus wavelength, shown as an inset in Fig. 2, was 2.3 dB/m at 1064 nm and 0.6 dB/m at 1083 nm. Under conditions of high pump power, the fiber transmission increases because of depletion of the ground-level population and the increase in the ${}^2F_{5/2}$ excited-level Yb population. Complete transparency is achieved with sufficiently large pump intensity and population inversion. For fixed values of pump power and wavelength, and $1.5 \cdot \mu m$ seed power and wavelength, the Er/Yb fiber length can be adjusted to maximize the $P_{1.1\,\mu\text{m}} \times P_{1.5\,\mu\text{m}}$ power product of the amplifier and therefore the DFG power. For our experimental conditions we found the optimum fiber length to be approximately 5 m for 1064-nm DFG. The amplifier output at 1064 and 1550 nm is shown in Fig. 2 as a function of the pump laser diode current, where seed powers were 10 mW at 1550 nm and 40 mW at 1064 nm. A maximum of 280 and 125 mW was obtained at 1064 and 1550 nm, respectively. When it was seeded with 20 mW of power at 1083 nm, the amplifier output was 460 mW at 1083 nm and 25 mW at 1550 nm.

To maximize DFG power we adjusted the polarization controllers between the seed sources and the amplifier to achieve linear and vertical polarizations for both wavelengths at the amplifier output. A single antireflection- (AR-) coated achromatic 10-mm focal-length lens was used to image the beam spot from the output fiber end (cleaved at $\sim 8^{\circ}$) onto the 19-mm long AR-coated PPLN crystal (Crystal Technology). For optimum conversion a magnification of $11 \times$ was experimentally determined. For calculated mode field diameters in the Er/Yb codoped fiber of 7.5 μ m at 1550 nm and 5.7 μ m at 1064 nm the $1/e^2$ beam diameters were 89 and 63 μ m inside the PPLN crystal, corresponding to confocal parameters of 8 and 6 mm, respectively. The 19-mm-length crystal contained eight gratings with periods of $28.5-29.9 \ \mu m$ in increments of 0.2 μ m. Its temperature was controlled to extend the spectral quasi-phase-matching range. The crystal was AR coated with a single SiO_2 layer with transmissions of 0.87 and 0.98 at 1064 and 1550 nm, respectively.

The mid-infrared DFG output was collimated by an uncoated $f = 50 \text{ mm CaF}_2$ lens, and the residual pump beams were blocked by an AR-coated Ge filter and focused onto a calibrated thermoelectrically cooled HgCdTe detector (1-mm² active area) by a 5-cm focallength off-axis parabolic mirror. The 3.4- μ m power generated for pump wavelengths of 1064 and 1548 nm is shown in Fig. 3. A maximum power of 11 μ W was achieved, and the slope efficiency was 0.31 mW/W^2 . When the 1.5- μ m fiber preamplifier was removed, the 1.5- μ m amplifier output and the DFG power decreased by approximately 30%. For the 1083-nm DFG pump case the 1.5- μ m output decreased to 25 mW because of the nonoptimum fiber length, whereas the 1083-nm power increased to 460 mW, resulting in a maximum DFG power of 4.1 μ W at 3.5 μ m.

A comparison with the calculated theoretical DFG conversion efficiency¹ of 1.36 mW/W^2 indicates that we are achieving 23% of the expected conversion. We attribute our lower than expected conversion to nonoptimum overlap of the mixing beams in the PPLN crystal. In particular, the different mode field diameters in the fiber lead to different confocal parameters in the crystal. Moreover, increased conversion is expected by ensuring better spatial pump beam overlap in the crystal by careful alignment of the fiber-output and



Fig. 2. Amplifier output powers as a function of the 3-W (at 3 A) pump diode current. (a) Pump wavelengths of 1064 and 1550 nm with seed powers of 40 and 10 mW, respectively (b). Pump wavelengths of 1083 and 1550 nm with seed powers of 20 and 10 mW, respectively. Inset, measured absorption coefficient of Er/Yb fiber as a function of wavelength.



Fig. 3. DFG power at 3.4 μ m versus pump power mixing product $P_{1064 \text{ nm}} \times P_{1550 \text{ nm}}$ for a 2-cm PPLN crystal.



Fig. 4. Absorption spectra near 2772.3 cm⁻¹ of (a) a CH₄ and H₂CO mixture and (b) CH₄. The H₂CO peak were predicted in Ref. 7.



Fig. 5. Absorption spectrum of the ν_5 6_{0,6} \leftarrow 6_{1,5} transition at 2833.2 cm⁻¹ for a calibrated 5.78-ppm H₂CO-in-N₂ mixture. A Hitran 96 spectrum of 5.78-ppm H₂CO is shown for comparison.

imaging lens to achieve paraxial propagation through this lens.

A high-resolution spectral scan over 0.5 cm⁻¹ near 2772 cm^{-1} (3.61 μm) of a dual-component mixture of H_2CO and CH_4 in a 5-cm-long cell filled with 5 Torr of CH₄ and a room-temperature equilibrium concentration of *para*-formaldehyde (1-s averaging time) is shown in Fig. 4. The formaldehyde line positions indicated by the diamonds are from Ref. 7 and are in close agreement with the experimentally measured line positions. We used a methane spectrum obtained from a 20-Torr CH₄ reference cell (L = 3 cm) to determine the linewidth of the DFG source, which we determined to be <30 MHz by comparing the measured methane absorption peak width and the theoretically predicted Doppler and pressure-broadened linewidths. The apparent discrepancy in the 2772.1-cm⁻¹ CH₄ line position between the two experimental spectra is due to the presence of an unresolved lower-frequency H₂CO line at 2772.098 cm^{-1} in the dual-component mixture, which displaces the peak to a lower frequency.

The formaldehyde ν_5 band near 3.5 μ m was investigated because of its significance for atmospheric monitoring and urban air pollution. The spectrum of a 5.78-parts-in-10⁶ (ppm) calibrated formaldehydein-nitrogen mixture (Scott specialty gases) near 2833.2 cm^{-1} is shown in Fig. 5 at a pressure of 95 Torr for a path length of 18 m in a multipass cell (1-s averaging time). To ensure that the formaldehyde mixture was not affected by adsorbing to the surfaces of the gas handling equipment, we continuously passed the gas mixture through the cell. For comparison, a Hitran96 H₂CO 5.78-ppm spectrum is superimposed. A 600-MHz Gaussian line shape was convolved with the Hitran predicted line shape to account for the broad linewidth displayed by the $1.5-\mu m$ external-cavity diode laser, which operated somewhat unstably at this frequency, as is evident from the lower-frequency H₂CO line-shape shoulder. Accidental etalons in the baseline limit the detection sensitivity to $\sim 0.6\%$, (1.1-ppm H₂CO). We expect to reduce the magnitude of the etalon effects by more than an order of magnitude by judicious attention to the fiber cleave angles and the incident beam angles on the optical surfaces in the sensor. Moreover, the comparatively high DFG power produced will permit the possible implementation of a balanced detection scheme to increase further the minimum trace-gas detection sensitivity.

In conclusion, a compact and robust fiber-coupled DFG-based mid-infrared spectroscopic source pumped by a $1.1-1.5-\mu$ m dual-wavelength fiber amplifier has been developed. High-resolution spectroscopic detection of CH₄ and H₂CO was demonstrated. Since large gain is exhibited by the Yb-doped amplifier from 1030 to 1100 nm, and by the Er/Yb amplifier from 1530 to 1570 nm, with appropriate seed lasers, the DFG source can generate tunable narrow-linewidth mid-IR radiation in the $3.0-3.9-\mu$ m spectral range.

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