Diode lasers, DFG and Molecules

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Abstract: The development and characteristics of compact diode laser pumped mid-infrared difference frequency gas sensors are reported. One such tunable cw sensor operates in the 3.3 to 4.4 μm spectral region and uses a diode-seeded fiber amplifier. Results show that fiber coupled sensors are capable of real-time, highly selective and sensitive measurements of numerous trace gas species in ambient air, presently CH_4, H_2CO, HCl, CO_2, N_2O, and H_2O
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Introduction

Significant developments in tunable diode laser technology and nonlinear optical materials have led to the demonstration of compact optical gas sensors operating in the 3-5 μm spectral region based on difference frequency generation (DFG). The characteristics of these sensors have been shown to be suitable for the sensitive, selective and real-time detection of numerous molecules with rovibrational spectra in the mid-infrared fingerprint region. Since CW DFG was first demonstrated using two low power single longitudinal mode laser diodes [1] (generating several nW), improvements in diode laser pump sources and nonlinear optical materials (eg. periodically poled $LiNbO_3$, PPLN) has increased the DFG power to tens of μW's, thereby yielding minimum detectable absorbances of $< 10^{-4}$ (or ppb concentrations) with the use of low noise thermo-electrically cooled infrared detectors (NEP ~ $pW(Hz)^{-1/2}$). Suitable DFG pump sources include distributed feedback (DFB) and distributed Bragg reflector (DBR) diode lasers, extended cavity diode lasers (ECDLs), semiconductor master oscillator power amplifiers (MOPAs), diode-pumped non-planar ring Nd: YAG lasers and more recently, diode laser seeded rare earth doped fiber amplifiers.

Portable and tunable DFG sensors have been demonstrated to be suitable for real-time trace gas sensing [2]. A sensor based on discrete optics (measuring 30x30x60 cm^3), generated mid-infrared radiation near 3.5 μm by difference frequency mixing a 100 mW Fabry-Perot laser and Nd: YAG laser. Such a sensor measured ppb levels of CO and H_2CO gas over a period of several weeks in a human rated simulation chamber located at NASA-Johnson Space Center for ground verification measurements for the life support system of the international space station program [3].

In recent DFG work, the emphasis has been on improved sensor robustness against vibrations and temperature drifts by using optical fiber for beam delivery, combining and amplification. Furthermore, attention has been paid to using diode laser pump sources that are fiber coupled ('pigtailed'), or can be fiber coupled. Fiber pump delivery also facilitates the efficient use of the wide gain bandwidth commercially available ytterbium and erbium doped fiber amplifiers. Diode laser

pump sources we have used include telecom DFB diode lasers near 1.55 μm (telecom ITU grid), DBR diodes available near 1 μm, α-DFB high power 1.064 μm diode lasers, and broadly tunable ECDLs. These diode lasers have characteristics that are desirable for high-resolution spectroscopy for field gas concentration measurements. Characteristics include inherent frequency stability, narrow linewidth, direct current modulation, and cw single frequency pump powers from 2-500 mW.

Sensor configuration

A portable DFG sensor that is continuously tunable from 3.25 - 4.40 μm is shown schematically in Fig 1. This device uses as pump sources an ECDL (P=20 mW, λ= 814 to 870 nm) and a distributed Bragg reflector (DBR, P=50mW, λ=1083 nm) diode laser seeded Ytterbium doped fiber amplifier pumped by a 975 nm, P=2 W diode laser [4,5]. The fiber amplifier boosts the seed pump power of 10 mW at 1083 nm to 540 mW. Both lasers are coupled into a single mode fiber and combined by a wavelength division multiplexer (WDM). The linear polarization output from the fiber for a **e**+**e**→**e** nonlinear mixing process in the PPLN crystal is maintained by using two polarization controllers in the fiber delivery system. Coarse tuning is achieved by rotation of the feedback mirror in the Littman style ECDL. Fine tuning and scanning of single or multi-component absorption lines of up to ~25 GHz is accomplished by current modulation of the DBR diode laser (~50 Hz). The sensor, including driver electronics, optics and an 18 m multi-pass cell, was mounted on a breadboard (45 x 45 x 12 cm^3).

An achromat lens (*f*=10 mm; 0.25 NA) was used for imaging the fiber output (terminated with a FC-APC connector) into the PPLN crystal. A 19 mm long, 0.5 mm thick PPLN crystal with a broadband AR-coating applied to both end faces contained 8 quasi-phasematching channels (0.5 mm wide) from 22.4 - 23.1 μm in 0.1 μm increments. In fig. 2 the mid-infrared power (DFG idler) as a function of wavelength is shown (right axis) with up to ~ 3 μW generated. The conversion efficiency is also plotted in fig. 2 (left axis) with a peak efficiency of 0.82 mW/W^2 (0.08%/W) at a wavelength of 3.5 μm. This value compares reasonably with a theoretically predicted DFG conversion efficiency of 1.4 $mW \cdot W^{-2}$.

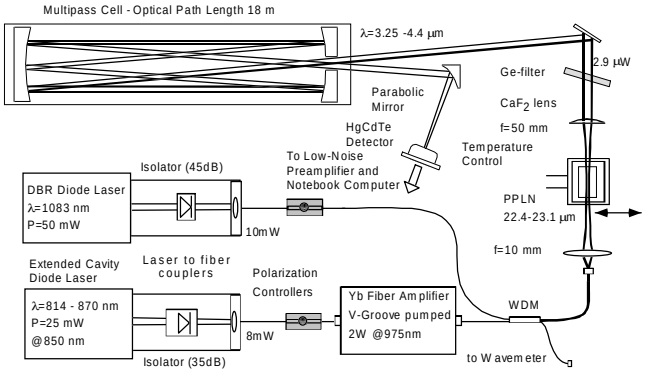

Fig. 1: Optical setup of a fiber coupled widely tunable DFG spectrometer.

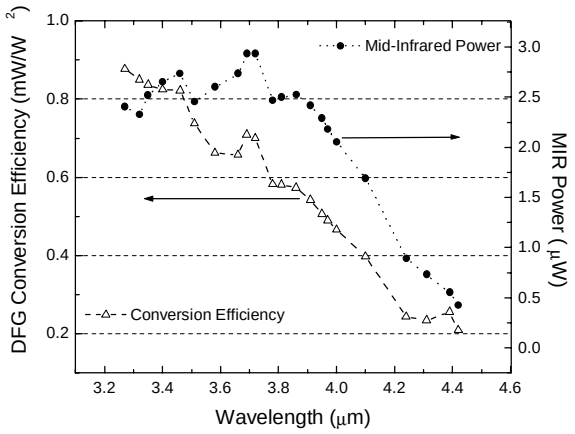

Fig. 3: DFG conversion efficiency and DFG power as a function of mid-IR wavelength

Spectroscopic performance

Shown in Fig. 3 is a Doppler broadened spectrum obtained from a 1 torr CH_4 sample (Doppler FWHM=270 MHz) in a 3 cm cell (20 averages over 0.2 s). The measured Gaussian absorption linewidth indicates a DFG linewidth of 42 ± 5 MHz (assuming a Gaussian DFG lineshape).

To characterize the detection sensitivity, prepared low concentration calibration mixtures of gases could be drawn through the multi-pass spectroscopic cell at reduced pressures (90-100 torr). For instance, using a 860 ± 43 ppb mixture of H_2CO in air (Scott Specialty gases) a concentration of 887 ± 19 ppb was measured (1 measurement every minute for 40 minutes, λ=3.53 μm). Using a 1773 ± 1 ppb CH_4 in air mixture a concentration of 1830 ± 15 ppb was measured, which is within 3.1 % of the calibrated concentration (1 measurement every 8.7 s for 19 minutes, λ=3.29 μm). These measurements

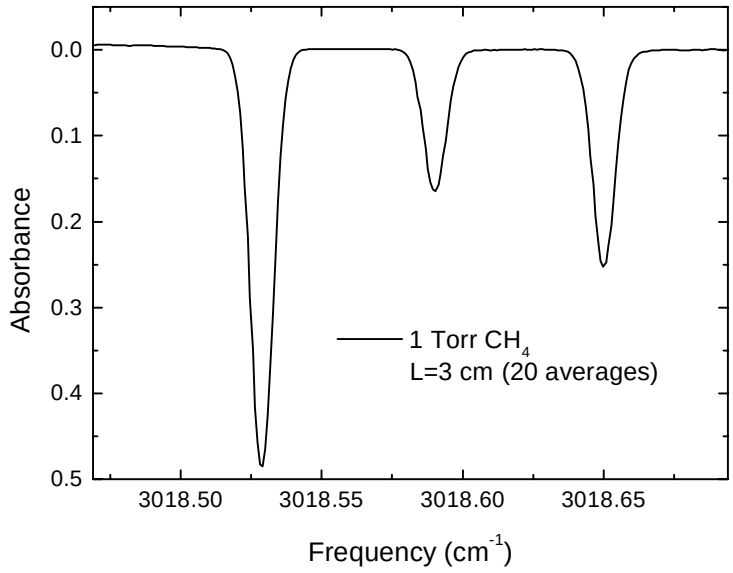

Fig.3: . Doppler broadened Q-branch CH_4 spectrum near 3.3 μm.

agree closely with the calibrated gas mixture concentrations if the ± 2 % uncertainty in the path length of the multi-pass gas cell is taken into account. Absorption cross sections used were from Ref [6] and Hitran96 respectively. Fig. 4 shows the detected methane concentration and the generated mid-IR power as a function of time. The ambient concentration was measured at ~2 ppm, with higher concentration spikes which are attributed to the intermittent operation of the central air-conditioning. This long-term experiment shows reliable operation of the fiber coupled spectroscopic source of both the generated mid-IR power as well as the inherent frequency stability for a time interval of 18 hours. In similar fashion, CO_2 (4.2 μm), N_2O (3.9 μm), NO_2 (3.5 μm), H_2CO (3.5 μm) and HCl (3.5 μm) have also been investigated.

In recent experiments using a new DFG sensor architecture, we have addressed the issues of increased power and sensitivity. The sensor, which is based on difference frequency mixing seeded 1μm Yb and 1.5 μm Er/ Yb fiber amplifiers in PPLN, has produced up to 0.7 mW [7]. Furthermore we have demonstrated the use of a dual beam detection configuration which allows optical noise to be

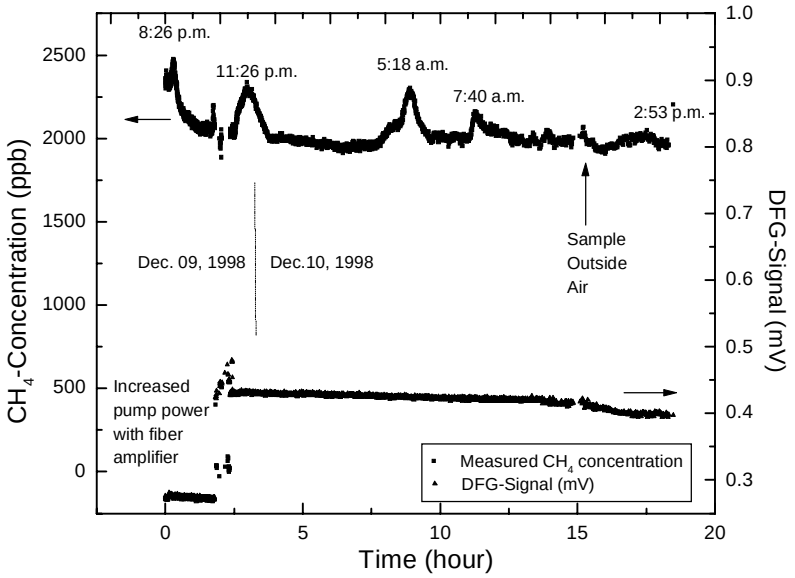

Fig.5: Long-term sampling of laboratory and outside air (upper trace) and mid-IR power for a 18 hour time interval

cancelled out by use of a reference beam, which is obtained by a beam splitter placed in front of the multi-pass cell, and directed to a second detector. Using this technique we have demonstrated an increased optical sensitivity of $\pm$ 2.8 x 10^{-5} (L= 100 m in a multi-pass cell) which implies, for instance, sub-ppb level CH_4 detection.

In conclusion, robust, compact and widely tunable fiber coupled diode laser pumped mid – IR sensors suitable for the real-time detection of various trace gases have been reported. The same sensor DFG architecture is also suitable for difference-frequency generation of longer IR wavelengths (> 5 μm) using quasi-phase matched GaAs with a transparency range from 2 μm to 16 μm [8].

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