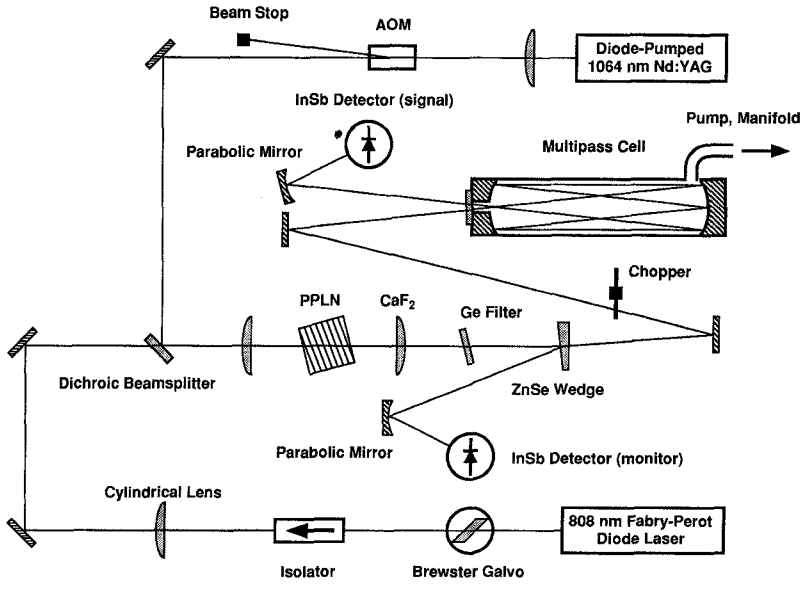
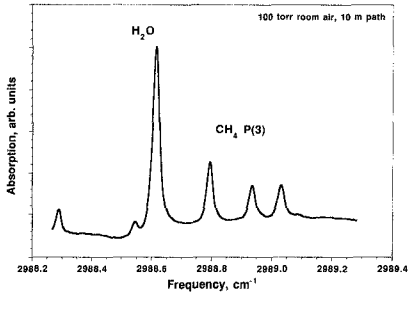


CFN4 Fig. 1 Schematic diagram of a compact 3.4 μm spectrometer based on bulk PPLN pumped by two solid-state lasers.



CFN4 Fig. 2 Absorption spectrum of room air near 3.35 μm at 100 torr in a 10-m multi-pass cell. The observed transitions belong to water (2988.6 cm^{-1} and lower) and the P(3) group of methane (2988.8 cm^{-1} and higher). The trace shown is a 1024-sweep average with baseline subtracted. The sweep rate was 100 Hz.

than 1 ppb, and methane mole fractions were assigned relative to the CMDL methane scale. Testing was performed in two parts. In the first part, two cylinders (identification numbers 30516 and 37057) were supplied with their methane mole fractions. The second part was a blind test; the two authors performing the spectroscopic measurements (KPP and SW) were not told the methane mole fractions in two other cylinders (64040 and 30482). A summary of data obtained for all samples analyzed in the experiment is given in Table 1. The combined uncertainty of the spectroscopic measurements over 1 minute was less than 1 ppb for a typical ambient methane mixing ratio of 1700–1900 ppb. The uncertainty was limited by residual interference fringes in the multi-pass cell. Without reference gas, the uncertainty of 20 ppb was limited by the long-term stability and reproducibility of the spectroscopic baseline.

We have demonstrated a precise spectroscopic application of diode-pumped difference-frequency generation. Diode-pumped DFG in PPLN[2,3] can effectively replace lead-salt diode lasers[4,5] and gas lasers in spectroscopic and trace gas monitoring applications requiring low-noise, single-frequency sources, continuously tunable in the 3–5 μm region, and eliminate the need for cryogenic cooling.

*Time and Frequency Division, National Institute of Standards and Technology, 325 Broadway, Boulder, Colorado 80303
**Climate Monitoring and Diagnostics Laboratory, National Oceanic and Atmospheric Administration, 325 Broadway, Boulder, Colorado 80303
†Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305-4085

CFN4 Table 1 Summary of methane mixing ratios measured using the 3.4 μm DFG spectrometer. Four samples were available in the experiment, and one of them (30516) was used as a reference in all measurements. The methane mixing ratio in this sample was assumed to be known exactly. Standard deviation shown for the DFG data therefore only includes errors of measurement of absorption, temperature, and pressure. Sample 37057 was measured without cancellation of interference in the multi-pass cell, hence the large error bar

CYLINDER IDENTIFICATION NUMBER	CMDL CH_4 ASSIGNMENT (ppb)	STANDARD DEVIATION (ppb)	3.4-μm DFG MEASUREMENT (ppb)	STANDARD DEVIATION (ppb)
64040	1753.8	0.2	1754.3	0.8
30516	1775.3	0.1	used as a reference	n/a
30482	1781.9	0.9	1781.1	0.8
37057	1896.7	1.3	1896.2	3.0

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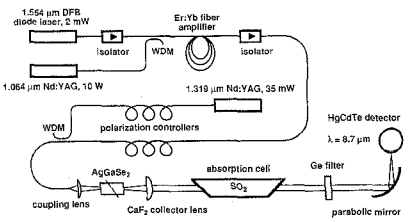
CFN5 2:15 pm

Tunable mid-infrared source pumped by fiber-coupled communications lasers

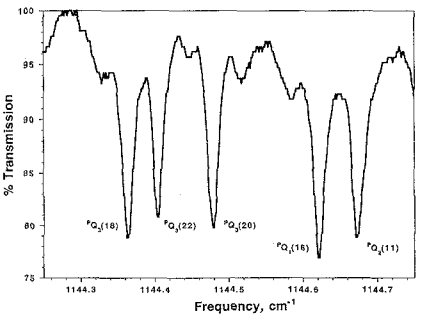
K. P. Petrov, R. F. Curl, F. K. Tittel, L. Goldberg,* *Rice Quantum Institute, Rice University, 6100 Main Street, Houston, Texas 77005-1892; E-mail: fkt@rice.edu*

The use of diode lasers operating at wavelengths of 1.3 μm and 1.5 μm as pump sources for difference-frequency generation (DFG) in $AgGaSe_2$ was proposed by Simon *et al.* in 1993.[1] The advantage of this scheme is the possibility of convenient generation of cw tunable narrowband light in the spectroscopic "fingerprint" region between 8 μm and 12 μm using readily available communications diode lasers. The recent development of Er/Yb co-doped fiber amplifiers[2] near 1.5 μm, and Pr^{3+}-doped fluoride fiber amplifiers[3] near 1.3 μm has made single-frequency output power in excess of 100 mW available. Such sources can be utilized for nonlinear optical conversion techniques, specifically for cw tunable low-noise DFG at the power level of a few microwatts. This radiation can be used for high-resolution mid-infrared molecular spectroscopy and, potentially, for spectroscopic detection and measurement of trace air contaminants such as ammonia, ethylene, sulfur dioxide, methane, nitrous oxide, and phosphine.

We report the operation of a compact all-solid-state room-temperature DFG source (Fig. 1) that employed a high-power Er/Yb co-doped fiber amplifier at 1.554 μm ("signal") and a 35 mW fiber-coupled diode-pumped monolithic ring Nd:YAG laser at 1.319 μm ("pump"). The amplifier was injection-seeded by an optically isolated 2 mW pigtailed distributed-feedback diode laser and operated near saturation, producing up to 0.5 W single-frequency power. The amplifier was



CFN5 Fig. 1 Schematic of an 8.7-μm tunable single-frequency CW DFG source pumped by fiber-coupled solid-state lasers operating at 1.319 μm and 1.554 μm.



CFN5 Fig. 2 Optical transmission of a 10-cm-long absorption cell filled with SO_2 at a pressure of 5 torr. Sweep time was 50 s, lock-in amplifier time constant was 0.1 s, chopper frequency was 2 kHz. The signal was detected without regard to phase (R component).

pumped by a Nd:YAG laser at 1.064 μm with a measured threshold of 87 mW, and slope efficiency of 12%. The pump and signal outputs were combined in a single-mode fiber by use of a fiber-optic wavelength-division multiplexer. This arrangement provided stable, alignment-free spatial and angular beam overlap required for optimal DFG conversion efficiency. The difference-frequency mixing was performed in a $4 \times 4 \times 10$ mm $AgGaSe_2$ crystal cut for type 1 phase matching at an internal angle of 65°. The effective crystal length of ~2.5 mm was limited by the birefringent beam walkoff at 1.319 μm. With 29 mW pump power and 370 mW signal power incident on the crystal, an output power of 0.1 μW was measured at 8.7 μm. This power level was sufficient for use in high-resolution spectroscopy. We used temperature-tuning of the pump laser at 1.319 μm at the rate of 50 seconds per sweep. The beam was chopped at a rate of 2 kIIz, and the idler power was detected with a lock-in amplifier. The observed infrared absorption lines of sulfur dioxide were assigned with use of a HITRAN database. Figure 2 shows the direct absorption spectrum of the ν_1 symmetric stretch band of sulfur dioxide near 1144 cm^{-1}.

We have demonstrated what to our knowledge is the first all-solid-state tunable cw single-frequency source operating at a wavelength above 8 μm at room temperature. It is based on difference-frequency mixing in $AgGaSe_2$ and uses commercial single-frequency pigtailed diode and diode-pumped lasers operating near the fiber communications wavelengths of 1.3 μm and 1.5 μm. The source was used for high-resolution spectroscopy of the ν_1 symmetric stretch band of SO_2. This technology may lead to the development of compact all-solid-state gas sensors for detection and measurement of air pollution and chemical analysis.[4]

Optical Sciences Division, Naval Research Laboratory, 4555 Overlook Avenue, Washington, DC 20375-5672; E-mail: goldber3@nrlfs.1.nrl.navy.mil

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CFN6 2:30 pm

Quantifying trace gas concentrations with use of digital signal processing

Chris Hovde, *Southwest Sciences Inc., Ohio Operations, 7969 Kimbee Drive, Cincinnati, Ohio 45244*

Diode lasers combine high sensitivity with high spectral resolution, resulting in excellent detection sensitivity and selectivity for trace gases by monitoring their spectral absorption features. Diode-laser based instruments have been used in atmospheric science to measure trace gas concentrations and emission rates. By sweeping the injection current of the diode laser, spectral scans over single absorption features can be collected in milliseconds. Rapid data processing algorithms are required to preserve this high data acquisition rate.

Multi-linear least squares is a well-known method to analyze the measured absorption spectra, including the optical baseline, but it requires knowledge of the absorption line position and width. These may not be precisely known in field measurements as a result of fluctuations in the pressure, temperature, gas composition, or laser frequency. The use of inaccurate values results in systematic errors in the estimated gas concentration. The measurement of gas emission rates by eddy correlation is especially susceptible to such systematic errors, even when they are less than 1%.

We have recently developed a series of digital filters that can eliminate these sources of systematic error in laser-based absorption measurements while preserving the high data rate of the multi-linear fitting algorithm. These digital filters can be very easily implemented on a commercial DSP platform, resulting in high processing speed at modest cost. The filters can also be used to stabilize the center frequency of the laser ramp. Simulations and experiments were used to demonstrate the effectiveness of this technique. A 1650-nm diode laser spectrometer was combined with a commercial DSP platform and used to measure the R(2) line of methane in dry nitrogen at pressures near one atmosphere. The laser ramp consisted of 64 discrete current steps that spanned approximately 1.4*fwhm of the absorption feature. The algorithm worked accurately (~0.1% measurement bias) despite pressure differences of ±10%. When the laser was locked by use of the frequency shift, the frequency error (measured inside the lock loop) was 20 MHZ rms, or less than 1% of the line width and 10% of the frequency step size. The data acquisition rate achieved in this demonstration, 300 scans per second, was limited only by the speed of the analog interface.

CFN7 2:45 pm

Photoacoustic spectroscopy with a frequency-modulated DFB diode laser

M. Wolff, H. Harde, *Universitaet der Bundeswehr Hamburg, Holstenhofweg 85, 22043 Hamburg, Germany*

Increasing perception of environmental problems has initiated the development of sensitive detectors for monitoring pollutants in the atmosphere. Particularly sensors based on the photoacoustic effect in conjunction with lasers have proven to monitor trace gases at concentrations of a few ppb or less.

Photoacoustic detection uses the fact that the absorption of light by a molecule is partially transformed into kinetic energy and transfered to the surrounding molecules. This causes a local pressure increase in the absorbing gas. If the excitation source is modulated, a sound wave is generated that can be detected by a microphone. Photoacoustic spectroscopy (PAS) has the advantage of producing a signal only when light is absorbed. Hence, contrary to transmission spectroscopy, PAS is an offset-free technique and it is possible to replace long absorption cells by much smaller ones. In addition, the potential exists of using acoustic cell resonances that enhance the signal and thereby increase the detection sensitivity.

The PAS technique has been almost exclusively used in conjunction with high power gas lasers as the photoacoustic signal is proportional to the intensity of the light.[1] The disadvantage of using these lasers is their nontunability and therefore the requirement of a coincidence with an absorption line. Usually the modulation of these lasers is performed with a mechanical chopper that generates coherent noise by the rotating blade and in the cell windows. This deteriorates the sensitivity of the detector.

We have developed a photoacoustic spectrometer using a diode laser for sensitive monitoring of trace gases. Experiments were performed on the rotational line P2 of the vibrational transition 2-0 (overtone) of hydrogen fluoride at 1304.53 nm. A room temperature distributed-feedback-laser was applied as the excitation source. Its emission wavelength (single mode) was continuously tuned over a frequency range of 700 GHz. A frequency modulation that was achieved by injection current modulation allowed both to generate a photoacoustic signal at the resonance frequency of the cell and to avoid coherent noise originating from a chopper. These advantages compensate for the low power of the device and the small absorption cross section of the