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CRF, Stanford Researchers Develop Method for Trace Gas Chemical Detection in the 8-μm Region

Sandia scientists, in collaboration with Stanford University, have recently found a way to sensitively detect and speciate gases in the 8- to 12-μm spectral region, an important range for trace gas chemical detection. The new technique is based on nonlinear difference frequency generation (DFG) in a new material called quasi-phase-matched (QPM) orientation-patterned GaAs (OP-GaAs).

QPM light sources using periodically poled lithium niobate (PPLN) are widely used to access the ~400-nm to 4-μm spectral region for applications like blue-light generation, telecommunication sources, infrared generation, and terahertz and short-pulse systems. But use of PPLN is limited by intrinsic material absorption at wavelengths longer than 4 μm, precluding its use for investigating other important spectral regions. One such region is the 8- to 12-μm spectral band, which is important for gas-phase chemical detection and spectroscopic applications because many functional groups absorb there, and absorption strengths can be up to ten times larger than in the mid-infrared region (3 to 5 μm).

The difference frequency generation technique optically mixes light from two tunable telecom diode laser sources in an orientation-patterned GaAs crystal.

New QPM Material

OP-GaAs, developed at Stanford, has broad transparency (~1 to 16 μm) and a high nonlinear coefficient, resulting in high device efficiency with the possibility of exceptionally broad tunability. CRF researchers Scott Bisson and Tom Kulp have demonstrated over 350 nm of truly continuous, single-frequency tuning in the 8-μm region by incorporating OP-GaAs into a broadly tunable, continuous wave DFG source.

The DFG technique optically mixes light from two tunable telecom diode laser sources (1.3-μm O-band and 1.5-μm C-band) in an OP-GaAs crystal to produce their frequency difference in the 8- to 12-μm region. Figure 1 shows a schematic of the OP-GaAs-based DFG system. For maximum conversion efficiency into the infrared, both telecom sources were first amplified by rare earth-doped fiber amplifiers before being mixed in the OP-GaAs. Tuning in the 1.3-μm region extended from 1280 to 1320 nm, whereas tuning in the 1.5-μm region extended from 1532 to 1567 nm. By combining the

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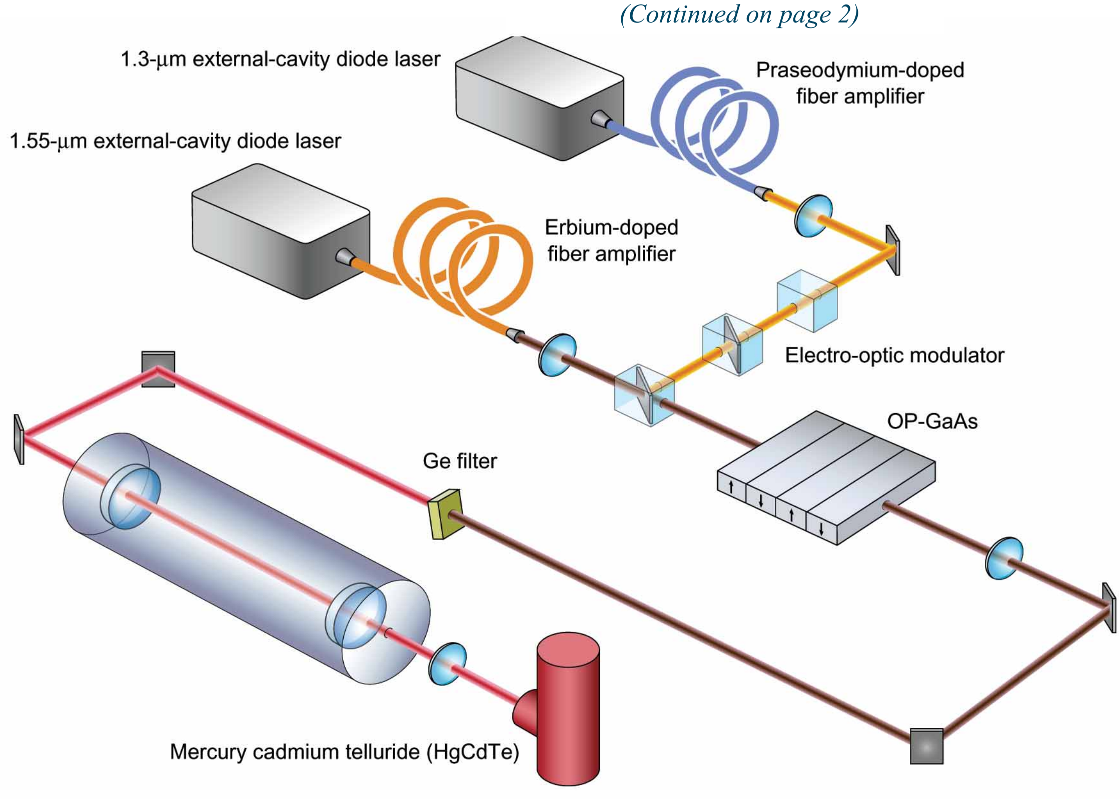

Figure 1. Schematic of the OP-GaAs-based DFG light source and cavity ringdown spectrometer.

OP-GaAs

(continued from page 1)

tunability of these two sources, over 350 nm of tuning was obtained.

The DFG source forms the basis of a long-wave (8 to 12 µm) infrared cavity ringdown spectrometer (CRDS) for trace gas detection. High-sensitivity CRDS, coupled with the broad tunability of the DFG source, will allow both speciation and quantification of a broad range of species.

CRDS operates by filling an optical cavity with light, then shuttering the light and recording the subsequent decay. The decay time yields a direct measure of the loss (optical absorption) and hence concentration of gas in the cavity. CRDS achieves its sensitivity by effectively amplifying the length of the cell (typically several km) through the use of ultra-high reflectivity (>99.96%) mirrors, increasing the effective absorption length.

Unlike modulation-based techniques, which are sensitive only to narrow line spectra, CRDS is sensitive to both broad and narrow absorbers, which greatly increases its versatility. CRDS also operates with low optical power (µW), making it amenable to portable field applications.

In the first demonstration of the system, Bisson and Kulp recorded a portion of the N_2O spectrum (see Figure 2). An overlaid theoretical spectrum shows excellent agreement.

The ability to tune continuously over broad spectral ranges with extremely narrow linewidth (~100 kHz) is unprecedented and will find many applications in laboratory spectroscopy.

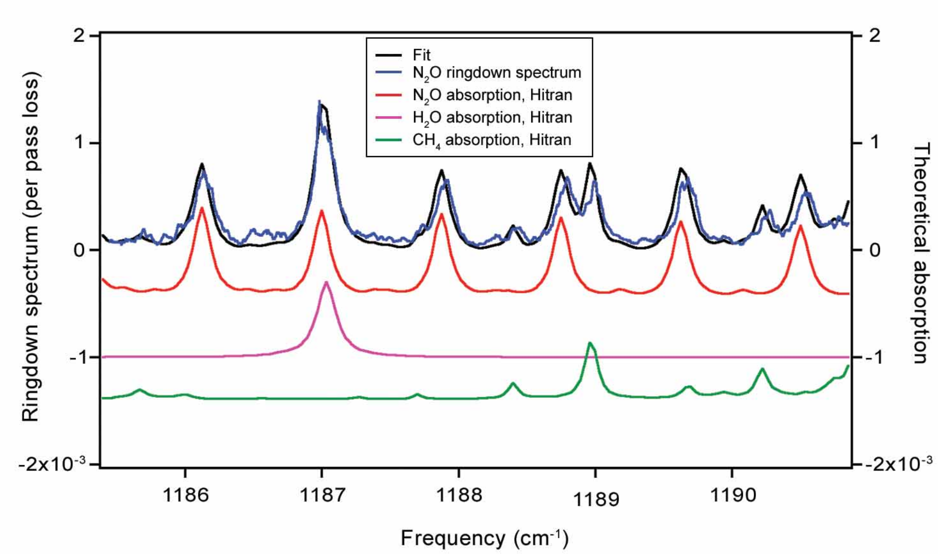

Figure 2. Portion of the first overtone spectrum of N_2O (shown in blue) acquired with the OP-GaAs DFG source. The theoretical spectrum is shown in red. The discrepancies are due to water vapor and methane impurities in the ringdown cell.

Greater Sensitivity, Smaller Sample Sizes are Features of Water Contamination Detection Method

Editor's note: The research featured in the following article takes place in Sandia's Microfluidics Department, which was created to further develop capabilities and applications arising from the CRF's chemical sciences and laser diagnostics expertise.

Growing concern over the vulnerability of the nation's water supply to naturally occurring pathogens and terrorist attacks has led Sandia scientists to look for better ways of testing for potential biological contamination. Because pathogens such as Cryptosporidium and anthrax can contaminate water at concentrations as low as one bacterium per liter, scientists face the challenge of developing sufficiently sensitive testing methods that eliminate the need for multiple, large samplings to ensure against false negative readings.

Sandia scientists Blanca Lapizco-Encinas, Blake Simmons, Eric Cummings, and Yolanda Fintschenko have recently found a way to selectively isolate and concentrate pathogenic bacteria using insulative (electrodeless) dielectrophoresis (iDEP). Compared to other methods, iDEP reduces the sample volume needed to successfully deliver a detectable amount of pathogenic material to an analytical device.

Polarizable particles can be moved in a nonuniform electric field by dielectrophoretic force, as reported by Pohl in 1951. Other investigators have demonstrated the ability to separate live and dead cells using pressure-driven flow with AC field in an electrode array. However, the drawbacks of this method include fabrication complexity and charging effects, fouling, and gas bubbles at the electrode surface.

iDEP avoids these problems by creating a heterogeneous electrical field between only two electrodes using the geometry of insulating posts etched in channels. Because no thin film electrodes are required, a much larger working area can be fabricated.

The flow is driven electrokinetically. Species trap when the dielectrophoretic mobility exceeds the electrokinetic mobility. The post geometry, spacing, and size affect the strength of the dielectrophoretic traps.

Figure 1 shows the separation and concentration of live and dead *E. coli*. The cells were observed to have negative dielectrophoretic mobility in a deionized water background, which is consistent with expectations. When particles are negatively dielectrophoretic, the field-induced dipole in the particles is less

(Continued on page 3)

iDEP

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...iDEP reduces the sample volume needed to successfully deliver a detectable amount of pathogenic material to an analytical device.

than that of the background electrolyte, which causes the particles to repel from regions of high electric field.

The dead bacteria have a leaky membrane, resulting in less negatively dielectrophoretic behavior. This difference in dielectrophoretic behavior causes live and dead cells to separate into distinct bands under iDEP trapping conditions.

To determine the behavior of *E. coli* in the presence of inert, noncellular particles that may be in the sample background, 1-µm polystyrene beads coated with rhodamine were added to a sample of live bacteria. The polystyrene beads exhibit positive dielectrophoresis, in contrast to the iDEP behavior for live cells (see Figure 2).

iDEP's ability to selectively concentrate bacteria could lead to potential applications such as a front-end device for large-volume applications like water analysis and small-volume applications including medical diagnostics.

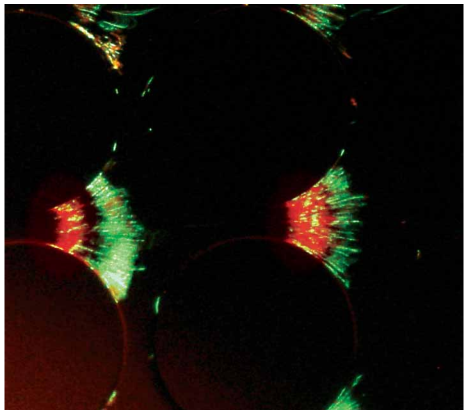

Figure 1. Image of iDEP simultaneous concentration and separation of live and dead *E. coli* at 10× magnification, inverted fluorescence microscope. Live cells are green, dead cells are red. A field of 130 V/mm was applied.

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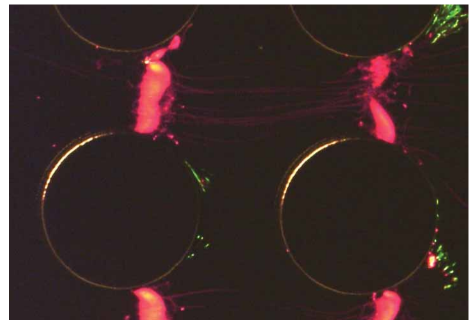


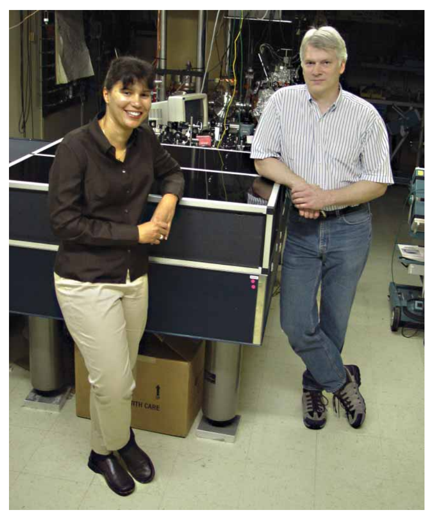
Figure 2. Image of simultaneous concentration and separation of live *E. coli* and inert 1-µm particles. Conditions were identical to those in Figure 1 except live cells were at a 1:100 dilution while the 1-µm rhodamine-labeled polystyrene beads were diluted 1:10. The applied voltage was 200 V/mm. The post offset is 0°, and the spacing is 200 µm.

People

Hope Michelsen Given Award of Excellence in Refereeing

Hope Michelsen will be honored with the Award for Excellence in Refereeing for the *Journal of Geophysical Research* at the December meeting of the American Geophysical Union. This award is given to only a few reviewers each year for their dedication to the review process as demonstrated by the number and quality of reviews, willingness to re-review, and timeliness.

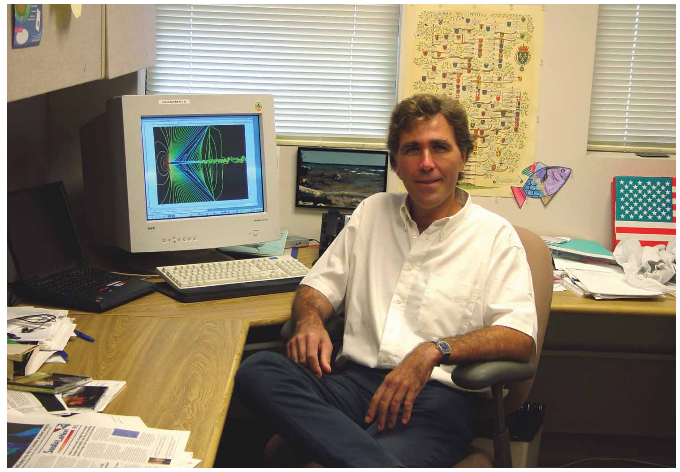
Michael Gershenzon, a postdoc with Hope Michelsen, was one of twenty-five invited speakers at the seventh biennial Atmospheric Chemistry Colloquium for Emerging Senior Scientists (ACCESS), which convened at Yellowstone National Park in Wyoming Sept. 4–7. The colloquium is jointly sponsored by DOE, NSF, NASA, and NOAA, and its purpose is to bring together—in scientific discussion and interaction—recent Ph.D. graduates and doctoral candidates in atmospheric chemistry, and representatives of the principal federal government agencies that fund atmospheric chemistry research.



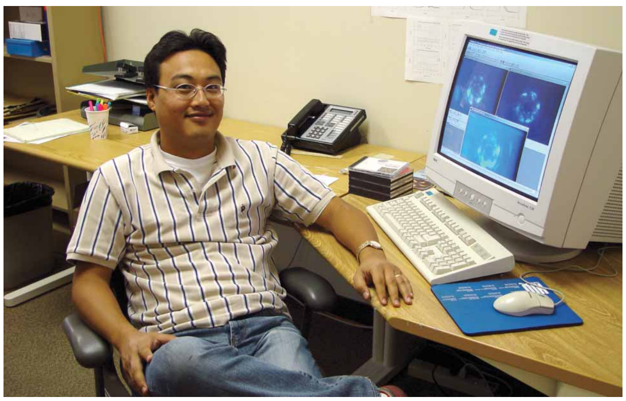
Engelene Chrysostom has spent the last two years working as a postdoc at the CRF. She first worked with Andy McIlroy using the cavity ringdown technique to measure the complex spectroscopy of CrO in a flame. She has also worked with Carl Hayden (right) doing femtosecond time-resolved coincidence imaging studies on the multiphoton dissociative ionization of CF_3I and the photodissociation of $(NO)_2$. Engelene will be leaving the CRF in December to continue her work in Europe.

People

Sean Moore (left), head of optical hybrid devices at the Naval Research Laboratory, Washington, D.C., visited Dahv Kliner in July to conduct experiments in the CRF's new fiber amplifier development laboratory.



Jerome Pousin is part of an ongoing collaboration with Philippe Pébay and Habib Najm to design advanced numerical schemes for simulating reactive flow behavior. Pousin, who visited the CRF Sept. 9–23, is a professor at the National Institute for the Applied Sciences in Lyon, France.



Hanho Yun, a mechanical engineering Ph.D. student from the University of Wisconsin at Madison, spent his summer working with Paul Miles on low-temperature combustion modes in small-bore diesel engines. Yun returned to Madison in September to complete his Ph.D.

Amélie Fayoux, an employee of PSA Peugeot-Citroen and a doctoral student at the École Centrale in Paris, recently spent eight months at the CRF working with Richard Steeper on homogeneous charge compression ignition (HCCI) engine combustion. Fayoux is returning to France to complete her Ph.D., which focuses on the application of optical diagnostics to HCCI combustion.

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