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Tomographic imaging of fluid flows by the use of two-tone frequency-modulation spectroscopy

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Diode laser absorption measurements obtained with two-tone frequency-modulation spectroscopy (TTFMS) are combined with tomography to produce spatially resolved quantitative images of fluid flows. The high sensitivity, good accuracy, and high stability of TTFMS absorption measurements permit optical-absorption tomography to be performed with low noise on extremely weakly absorbing objects. The method is demonstrated on a small oxygen flow with a GaAlAs diode laser operating near 760 nm and with an absorption sensitivity of $1:10^6$.

Nonintrusive spatially resolved measurements of gas concentrations are important in fluid flow, combustion, and heat transfer research. Such measurements may be performed by planar imaging methods by the use of, e.g., laser-induced fluorescence.[1] The high temporal and spatial resolution in combination with high sensitivity makes laser-induced fluorescence ideally suited for studies of transient phenomena and of minor species. However, quantitative measurements of concentrations are difficult to perform because of quenching. Other imaging techniques, such as Rayleigh and Raman scattering,[2] are not species selective or have low sensitivity. Absorption measurements, on the other hand, provide quantitative results but are integrating, i.e., they do not yield spatial resolution. When multiangle absorption measurements are combined with tomographic reconstruction, quantitative and spatially resolved measurements may be performed. Because of the low absorbance of most gaseous species in the visible-wavelength region, previous attempts in optical-absorption tomography have been restricted to specially selected species in order to yield a sufficiently high signal-to-noise ratio (SNR) for the tomographic reconstruction.[3-5] In this Letter we combine tomography with diode laser two-tone frequency-modulation spectroscopy (TTFMS), a highly sensitive absorption technique that extends the possibilities for quantitative and spatially resolved measurements to a vast number of species. One demonstrates the method by mapping the concentration in a section of a weakly absorbing O_2 gas flow. Optical tomography has also been applied to quantitative measurements of other fluid-flow parameters by the use of interferometric,[6] deflection,[7] and emission[8] techniques.

In recent years, a variety of frequency-modulation (FM) methods implemented on diode lasers have been developed with the purpose of increasing absorption sensitivity. When the detection band is shifted to high frequencies, laser excess ($1/f$) noise is avoided. The carrier frequency of a diode laser is modulated by the addition of an ac component to the laser injection current. Different FM techniques are discussed in

Refs. 9 and 10. In TTFMS[11] the diode laser is modulated with two closely spaced frequencies ($v_m \pm \frac{1}{2}\Omega$), which generate sideband pairs in the laser spectrum. When the frequency-modulated laser beam interacts with an absorption line, a beat tone at the intermediate frequency Ω will occur because of the differential absorption of the sidebands and the carrier. With TTFMS, detector-limited sensitivities of the order of $10^{-7}-10^{-8}$ are reachable.[12]

Figure 1 schematically depicts the experimental arrangement. The experimental technique is described in detail in a previous paper.[13] In this Letter we describe the application of an optical dual-channel technique with balanced homodyne detection.[14] This technique can be employed to suppress optical interference fringes, residual amplitude modulation, and laser excess noise in order to reach an ultrasensitive detection limit. In our approach the balanced detection scheme effectively cancels the background absorption that is due to atmospheric O_2. The diode laser, which was temperature and current controlled by a precision diode laser driver (Melles-Griot 06DLD103), was a double-heterostructure GaAlAs laser (Mitsubishi ML4405) operating near 760 nm. The laser beam was divided by a nonpolarizing 50/50 beam splitter, and the signal and the reference beams, which were adjusted to have equivalent optical paths, were directed onto two photodiodes (EG&G FND-100). Over the entire absorbing object, the beam-waist diameter was <0.2 mm. The two photocurrents were combined out of phase in a 180° rf hybrid (Anzac H-1-4) and amplified, bandpass filtered, and demodulated with standard rf techniques. Our TTFMS experiment used laser modulation at 641.8 and 652.2 MHz ($v_m \pm \frac{1}{2}\Omega$) with signal demodulation at $\Omega = 10.4$ MHz. The two modulation frequencies were generated when the frequencies v_m and $\frac{1}{2}\Omega$ from two different signal generators were mixed in a frequency mixer. The linearity of the total registration system was carefully checked.

The absorption measurements were performed on the $R5Q6$ line at $\lambda = 761.140$ nm in the O_2 A band. The $R5Q6$ line was one of the strongest noninterfering O_2 lines within the wavelength range of the
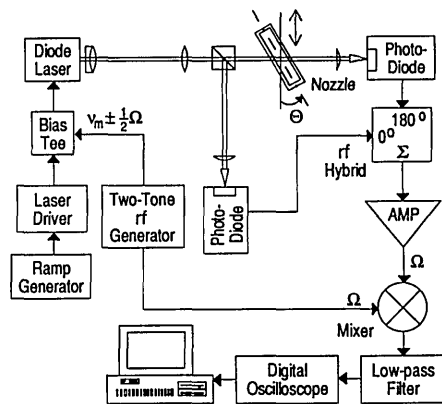
Fig. 1. Schematic diagram of the experimental arrangement.

particular diode laser we used. To measure the O_2 concentration quantitatively, we calibrated the TTFMS signal, shown as an inset in Fig. 2, using the atmospheric O_2 concentration (20.8%) in one of the detection channels that had a path length of 93 ± 0.5 cm. The signal associated with a TTFMS spectrum is the peak-to-peak value of the wave form. If the O_2 linewidth varies with the total pressure, the TTFMS signal exhibits nonlinear behavior with the O_2 concentration, as the sidebands will experience a different differential absorption. However, the influence on the line shape by variation of the O_2 partial pressure in a constant atmospheric pressure oxygen–air mixture is small, as the line-broadening coefficients are nearly equal for O_2 self-broadening and air broadening.[15] Figure 2 shows a measurement of the TTFMS signal as a function of the O_2 partial pressure (total pressure 751 Torr). The absorption path length in the cell was 90 cm. Because the response is highly linear, the TTFMS signal, calibrated to an absorption measured in direct transmission, can be extrapolated to measure weak absorptions with good accuracy. Similar results have also been presented by Kroll et al.,[16] who used derivative spectroscopy with a modulation frequency of 5 kHz. The difference in line-broadening coefficients[17] causes a signal increase of approximately 2.5% when pure O_2 is measured with a TTFMS signal calibrated to atmospheric O_2.

A 4 mm $\times$ 18 mm rectangular nozzle, constructed to give a laminar flow with a flat-topped velocity distribution, was used to produce a vertical asymmetric oxygen flow into the ambient atmosphere. When the laser beam is transmitted along the shortest path length through the asymmetric flow, the peak absorption corresponds to 3×10^{-4}. With a SNR of ~300, this indicates a detection sensitivity of 1×10^{-6}. The nozzle was translated across one of the detection channels, a step length of 0.5 mm, by use of a computer-controlled stepping motor. For each step the TTFMS spectrum, which was recorded in a 10-kHz bandwidth, was averaged 250 times in a personal computer, and a background spectrum was subtracted to remove the influence of a broad étalon fringe, causing a sloping baseline to appear in spite of the dual-channel detection scheme. Then the spectrum was searched for the maximum and minimum

wave-form values, and the peak-to-peak value was calculated.

To avoid long measurement times and complicated experimental arrangements, optical tomography often requires the use of few projections. In this Letter a modified multiplicative algebraic reconstruction algorithm[18] (MART) was used. Compared with convolution backprojection methods, iterative algorithms such as MART generally perform better when the number of projections is limited. The modified MART method behaves well for the reconstruction of smooth objects from as few as two or three projections. Many objects in combustion- or fluid-flow studies are intrinsically smooth because of diffusion, which makes the algorithm suitable for such studies. Naturally the use of few projection angles will result in a limited spatial resolution. The dependence of the spatial resolution on the number of parallel rays and projection angles is discussed in Ref. 18. Because of the small number of projection angles used in this study, major features are correctly determined, but high-spatial-frequency details may be lost.

The tomographic reconstructions were performed with six projection angles 30° apart. The rotation of the nozzle was performed manually with an accuracy of $\pm 1°$. In each projection, 81 consecutive measurements of the TTFMS signal were made with a data-acquisition time of 13 min. Figure 3 shows a 57 $\times$ 57 pixel tomographic reconstruction of the O_2 concentration in a section 3.2 mm above the nozzle orifice. Each pixel is 0.5 mm $\times$ 0.5 mm. The O_2 jet was flowing at 10 L/min from the nozzle. The expected flat-topped behavior of the laminar flow is clearly visible in the image, which shows that the O_2 flow fully displaces the ambient air. From the calibration procedure discussed above, the average O_2 concentration on the plateau was determined to be 104%, with a standard deviation of 4.7%. A recording, with 500 waveforms in the signal averaging, 9.5 mm above the nozzle, resulted in 103%, with a standard deviation of 2.5%. Images recorded higher above the nozzle showed smoother edges because of diffusion. The total measurement time may be reduced significantly by a decrease in the number of acquired waveforms, provided that the SNR is
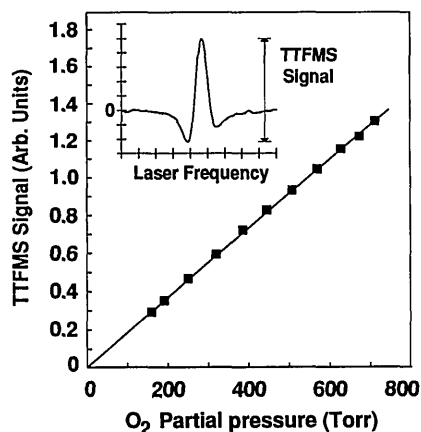

Fig. 2. TTFMS signal (peak-to-peak value) as a function of O_2 partial pressure. The inset shows the TTFMS signal.
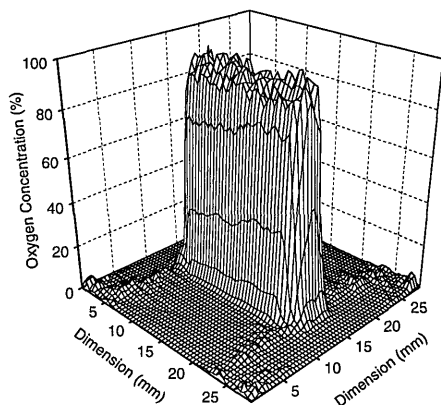
Fig. 3. Tomographic reconstruction of the concentration of O_2 in an expanding flow 3.2 mm above a rectangular (4 mm ×18 mm) nozzle.

sufficient and that a smaller spectral width in the signal averaging is used.

A systematic error in the measured data originates from a calibration error, which is caused mainly by the difference in the self-broadening and the air-broadening coefficients of O_2, as discussed above. This explains the slightly high concentrations at the plateau calculated above. We estimate the total error in the calibration to be less than 3.5%, including also an uncertainty in the absorption path-length measurement (0.5%) and signal variations between the reference spectra (0.5%). If the different line-broadening coefficients are taken into consideration, the accuracy can be increased considerably.

Two sources of error influence the tomographic reconstruction: statistical noise in the projection data and inconsistencies between the projections, e.g., small lateral displacements, angular deviations, and signal drifts. While the TTFMS SNR of 300 can be taken as an estimation of the statistical noise in the projection data, the inconsistencies between the projections are difficult to quantify briefly. Signal changes that are due to long-term drifts, e.g., temperature changes in the optical components that cause étalon fringes to wander about the TTFMS signal, were monitored to be less than 0.5% during a complete recording of a section of the O_2 flow. Small inconsistencies between projections result in errors, as seen in the nonabsorbing region of Fig. 3. When few projections are used, i.e., an underdetermined reconstruction, the pixel noise may be larger than the statistical noise in the projection data. From the obtained reconstruction data, shown in Fig. 3, we conclude that the total reconstruction error is approximately 5%.

The tomographic diode laser technique described above is an attractive method for performing quantitative and spatially resolved measurements of gas concentrations. Many species have weak overtone and combination bands within the wavelength range of conventional GaAs diode laser technology (0.7–1.7 μm) (e.g., H_2O, CH_4, NH_3, CO, CO_2, NO, NO_2). The high SNR in the measurements presented here indicates a significant potential for measurements on even more weakly absorbing species or smaller objects. Using PbS diode lasers in the 3–30-μm wavelength range, where most molecules have strong fundamental vibrational transitions, would permit a wide range of minor and major species in smaller samples to be probed. However, these lasers require cryogenic cooling, are expensive, and are not simple to operate. With the use of inexpensive GaAs diode lasers and standard rf components, multiarm arrangements for simultaneous data acquisition are conceivable. An alternative use is imaging ambient air pollution or water-vapor distributions of low concentration by the use of fan-beam tomography.[3] The fan-beam configuration would increase measurement speed considerably.

In conclusion, we have demonstrated that the high sensitivity of TTFMS permits quantitative tomographic imaging of weakly absorbing gaseous objects with high spatial resolution. In addition, the diode laser approach in optical tomography is applicable to a wide range of species and measurement problems. The low cost and simple operation of GaAs diode lasers make them attractive for field measurements and in multilaser configurations.

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