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# Temporal correlation scheme for spectroscopic gas analysis using multimode diode lasers

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The reliability of diode lasers used in spectroscopic applications is limited by their intrinsic multimode and mode-jump behavior when wavelength-tuned by current or temperature. We report on a scheme for gas analysis based on temporal correlation between absorption signals from an unknown external and a known reference gas concentration, simultaneously recorded when the diode laser wavelength is temperature-tuned across absorption features of the gas of interest. This procedure, which does not require any knowledge of the exact spectrum, also eliminates light intensity fluctuations due to mode competition. The method is illustrated for atmospheric oxygen absorption applied to diffusion measurements.

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Gas concentrations can be conveniently measured by high-resolution absorption spectroscopy employing single-mode tunable laser sources, especially diode lasers. The narrow-band laser radiation is scanned over an isolated absorption line, specific for the species to be studied, and the concentration is retrieved from the measured absorbance according to Beer–Lambert’s law. The technique, commonly referred to as tunable diode laser absorption spectroscopy (TDLAS), has applications ranging from, e.g., long-path absorption measurements of environmental pollutants<sup>1</sup> to probing of gas confined in porous materials.<sup>2</sup>

However, semiconductor diode lasers have intrinsic properties that often impair their spectroscopic applicability.<sup>3</sup> For example, several longitudinal modes can oscillate simultaneously, since the width of the spectral gain profile is much broader than the separation between adjacent longitudinal modes. The mode competition leads to interfering gas absorption signals from the submodes. Additionally, the tuning range of diode lasers is often not continuous and mode-hops occur, which causes light intensity fluctuations that can be misinterpreted as absorption. To cope with these problems, essentially single-mode operation can be achieved by employing distributed feedback, distributed Bragg reflector, or external cavity arrangements. All these technologies increase the degree of complexity and the cost of the systems. Additionally, the wavelength characteristics of the laser can change over time due to aging. Diode laser system realizations are frequently subject to thermal drift and active stabilization may be needed. These inconveniences as well as the need of expert operators have been major obstacles for the widespread application of TDLAS.

Another approach is used in the gas filter correlation (GFC) technique,<sup>4</sup> in which, frequently passive, broadband radiation is either passed directly through the external gas that is analyzed, or is additionally filtered through an optically thick reference cell filled with the target gas. The direct recording is strongly dependent upon the presence of the target gas in the external path, whereas the signal from the

reference cell is essentially the same, since the light at all characteristic wavelengths is anyway absorbed. The difference in spectral transmittance between the two paths is thus a sensitive indicator of the target gas concentration. Due to the inherent “holistic” property of the GFC technique, which matches out the unique spectral signature of the target gas, the presence of interfering gases does not affect the analysis. Notably, no knowledge of the gas spectrum is needed, which has been effectively used in, e.g., lidar gas monitoring<sup>5</sup> and gas imaging.<sup>6,7</sup>

In this letter a scheme for gas analysis, denoted temporal gas correlation spectroscopy (TEGACOS), is proposed. The technique combines the superior sensitivity and selectivity of the high-resolution TDLAS technique with the simplicity and robustness of a GFC spectrometer. The technique is particularly suited for analysis of gas with sharp absorption characteristics, and allows employment of multimode diode lasers. Similarly to the GFC technique, the radiation is split into one beam transmitted through the external gas, and another beam passed directly through a reference cell with a well-calibrated concentration of the target gas, as illustrated in the schematic setup shown in Fig. 1. Generally, the diode laser wavelength can be tuned across absorption structures of the target gas by changing the diode temperature or current. The simultaneously recorded signals from the two optical paths may contain gas absorption imprints, as well as intensity variations due to mode competition and mode hops, background fluctuations, and interference fringes. If the target gas is present in the external path, the gas signatures will correlate in time. Oppositely, mode-instability-induced fluctuations of the laser output will correlate differently, since they are independent of the presence of the gas. Thus, in order to discriminate between gas-related absorption and

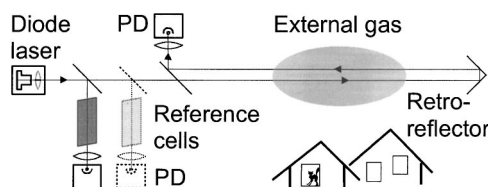


FIG. 1. Experimental setup for temporal gas correlation spectroscopy.

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mode instability, at each point in time both the external and the reference signals are normalized to the laser output power, which is conveniently monitored using the integrated photodiode in the diode laser case.

In conclusion, if the diode laser operates in a single longitudinal mode when scanned through the gas absorption structures, the temporal recording obtained constitutes the actual absorption spectrum of the gas. This is the ideal case used in conventional TDLAS. Oppositely, if the laser mode is unstable, the detected signal no longer corresponds to the actual spectrum. By using the temporal correlation scheme, information about the gas absorption can still be retrieved. It should be noted that the influence of laser frequency fluctuations due to thermal drift are automatically eliminated with the temporal correlation scheme, due to the simultaneousness of the measurement. Note also that external background and internal optical variations, including changes in the optical system alignment, and interference fringes generated by external feedback from optical surfaces, should be minimized, as in conventional TDLAS. If the perturbation widths are comparable to the linewidth of the absorbing species under investigation, they can obscure the detection and reduce system performance. Increased sensitivity is achieved with the suggested method, as in conventional GFC, in wavelength regions with strong differential absorption, i.e., where the gas spectrum has high contrast.

To demonstrate the TEGACOS concept, preliminary proof-of-principle measurements were performed on atmospheric oxygen, which has close-lying and well-resolved absorption lines in the A-band around 760 nm. An AlGaAs Fabry-Pérot-type diode laser (Sharp LT031MDO) with a nominal wavelength of 757 nm at 25 °C and a free running output power of 7 mW was scanned across a wide wavelength range including several oxygen absorption lines, by changing the laser temperature about 12 °C. Although temperature scanning by a Peltier element in a standard laser mount was slow ( $\sim 20$  s per scan), a faster method based on photothermal heating of the diode laser by pulsed radiation from an external laser<sup>8</sup> was also investigated. Many commercially available diode lasers are notoriously emitting light at multiple longitudinal modes. Enhanced multimode behavior of the diode laser used in our experiments was obtained by operating it at an injection current close to the lasing threshold. The widely tunable but spectrally noisy diode laser source, not suitable to use in conventional TDLAS, was diagnosed using a spectrometer. The output spectrum obtained had discontinuities and mode hops, as illustrated in Fig. 2(b). The experimental setup, shown schematically in Fig. 1, consisted of an external path of variable length and several reference paths, corresponding to different absorbancies, utilized for calibration. Figure 2(a) shows the simultaneously recorded direct-absorption signals normalized to the monitor current and divided by a fitted third order polynomial for display purposes. The signals displayed correspond to 138, 304, and 830 g/m<sup>2</sup> path-integrated concentration of oxygen, respectively. The absorption features observed (counted from left to right) correspond to the P3P3, P3Q2, R13Q14, R15R15, R15Q16, and R17R17 lines of the oxygen A band. However, it should be stressed that such a detailed spectral knowledge is not required using the present technique, which functions for any combination of gas absorption signals obtained when the diode laser source exhibits arbitrary frequency shifts or mode competition.

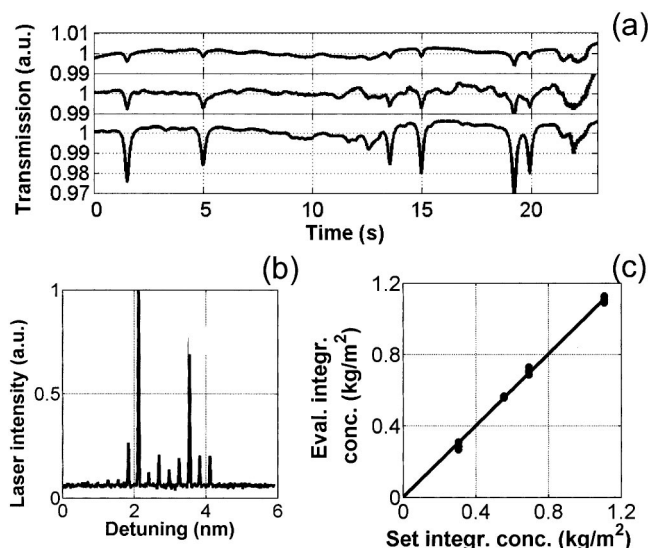


FIG. 2. (a) Simultaneously recorded single-scan time-correlated oxygen signals. The curves correspond to 138, 304, and 830 g/m<sup>2</sup> path-integrated concentrations of oxygen, equivalent to path lengths in air of 0.5, 1.1, and 3 m, respectively; (b) typical multimode diode laser spectrum around 760 nm; (c) evaluated vs set path-integrated oxygen concentrations in air, plotted with a linear fit.

Similarly to the broadband GFC technique, the reference cell provides an ultrasharp matching signature of the target gas. However, owing to the spectral sharpness of the laser source used in the temporal correlation scheme, the absorbed radiation is detected in a narrow wavelength interval at the time, and not integrated over all wavelengths as in the conventional case. Thus, in this scheme the reference cell is preferably not totally absorbing, and can also be used for calibration purposes. Since the reference gas concentration is well known, the external gas concentration can be related to it in a way that resembles the standard addition method. An important difference is that the standard addition calibration is usually not performed simultaneously, but in a time multiplexing procedure. To avoid line shape discrepancies of the absorption lines that would impair the correlation, the temperature and pressure in the reference path are kept as similar as possible to the conditions in the external gas. This can be achieved, e.g., by long-term temperature stabilization and pressure equilibrium in nonsealed reference cells.

The simple data processing consisted of initial subtraction of the background level, assessed by temporarily switching of the laser, followed by normalization of the signals to the laser output power. In order to remove the influence on the signals caused by path-differential variations due to, e.g., slowly wavelength-dependent transmission, a sliding pointwise-symmetric division was performed. The logarithms of the signals obtained were subject to a sliding correlation interval with a width corresponding to the expected width of the gas absorption features, resulting in a pointwise time-interval-integrated ratio between the signals. In a calibration procedure, a pair of reference signals of known concentration was used to single out points in time having the expected time-interval-integrated ratio, thus corresponding to the gas signature. Subsequently, the pointwise time-interval-integrated ratio between one of the reference signals and the external signal was used to determine the unknown gas concentration in the external path. The evaluated path-integrated concentrations obtained by the TEGACOS method versus

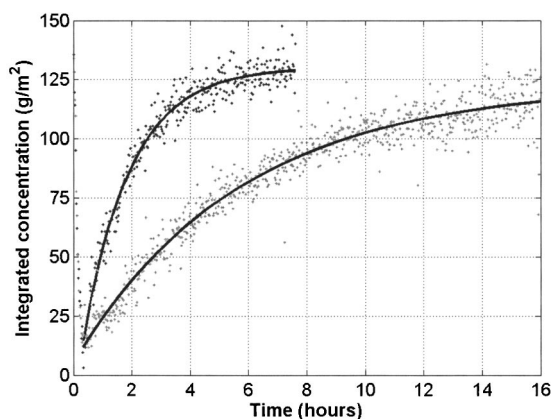


FIG. 3. Recording of the reinvasion of ambient air into a previously nitrogen-flushed glass container covered by polystyrene foam slabs of (a) 20 mm and (b) 10 mm thickness, respectively. An offset path-integrated concentration of 73 g/m<sup>2</sup>, due to the air outside the container, was subtracted from the signals.

the corresponding set values from a multipath experiment are displayed in Fig. 2(c). The detection scheme was used to monitor gas exchange through differently thick slabs of polystyrene foam covering a glass container placed in ambient air after previously being flushed with pure nitrogen gas. The temporal variation of the path-integrated oxygen concentration in the container is shown in Fig. 3. Although the measurement lasted several hours, thermal drifts do not affect the measurement.

Improved signal-to-noise ratios can be achieved by averaging the concentrations evaluated from several wavelength scans. With the proposed temporal correlation scheme, as in conventional TDLAS, several orders of magnitude higher sensitivity compared to direct absorption could be obtained by employing modulation techniques. A large modulated signal proportional to the species concentration is generated, which can be temporally correlated in a similar

way as in the case presented. Multivariate statistical methods, such as the powerful partial least-squares (PLS) technique,<sup>9</sup> are expected to further improve the accuracy and speed of data processing.

Although demonstrated for oxygen, it is evident that the TEGACOS technique works in any wavelength region and for any gas with close-lying and fairly sharp absorption lines. For example, in the mid-IR region, molecular band heads of several environmentally and biologically important gases could be monitored using the proposed scheme in combination with quantum cascade lasers. The requirements on the system are less stringent, facilitating use of cheaper, high-power multimode diode lasers without need for frequency stabilization. Although the detection sensitivity is lower than for conventional TDLAS, its robustness and relaxed stabilization requirements promise to make the technique very versatile.

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