

In-situ Combustion Diagnostics Using Diode Laser Absorption Sensors

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Abstract: The availability of extended wavelength DFB diode lasers makes *in-situ* flame measurements of important combustion species such as CO₂ possible. This work presents recent measurements near 2 μm of CO₂ concentration above flat-flame burners.

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In the past, multiplexed diode laser sensor systems have been applied for *in-situ* measurements of important combustion parameters such as gas temperature and water concentration [1, 2]. These *in-situ* measurements traditionally have been restricted to the near-infrared (NIR), where single-mode distributed feedback (DFB) diode lasers have been available. Conveniently, major combustion products H₂O and CO₂ both have absorption bands in the NIR at 1.4 and 1.5 μm respectively (See Figure 1). However, the large population (10–18%) of hot H₂O in the products of premixed natural gas combustion combined with H₂O's strong absorption band at 1.4 μm introduces significant spectral interference with the much weaker CO₂ band at 1.5 μm and inhibits accurate *in-situ* measurements of CO₂ at that wavelength. Previous work with this CO₂ band used fast extractive sampling techniques to make combustion measurements of CO₂ populations in order to eliminate the water interference and increase sensitivity [3]. The availability of new fiber-coupled DFB lasers at longer wavelengths, including 2.0 μm , enables interference-free *in-situ* flame measurements of CO₂. The 2.0 μm band of CO₂ is advantageous because its relative band strength is much greater than at 1.5 μm (see Figure 1) and because isolated CO₂ lines are available for *in-situ* monitoring (see Figure 2).

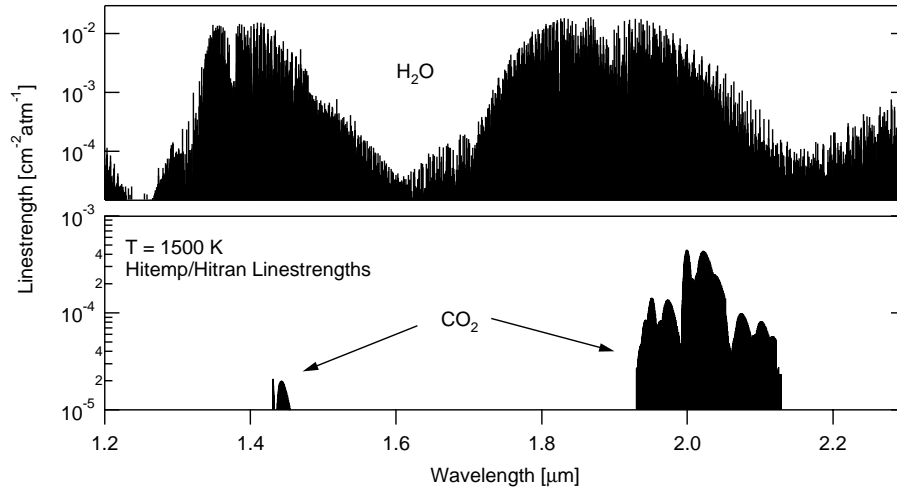


Fig. 1. Linestrengths of CO₂ and H₂O at $T = 1500\text{K}$ in the near-infrared.

Previous measurements of CO₂ at 2.0 μm employed external-cavity diode lasers (ECDL) whose convoluted power curves and low-bandwidth wavelength-tuning (less than 50 Hz) made the combustion sensors susceptible to baseline fitting errors and beam-steering related noise [4]. New DFB lasers can be tuned up to 10 kHz, thereby lessening beam-steering related noise, and have smooth power curves that can be estimated with polynomial fits for approximating zero-absorption baselines. Thus, the new work presented here offers improved results for fundamental spectroscopic measurements and *in-situ* flame measurements of CO₂.

The fundamental theory governing absorption spectroscopy is embodied in the Beer-Lambert law, Equation 1. The ratio of the transmitted intensity I_t and initial (reference) intensity I_o of laser radiation through

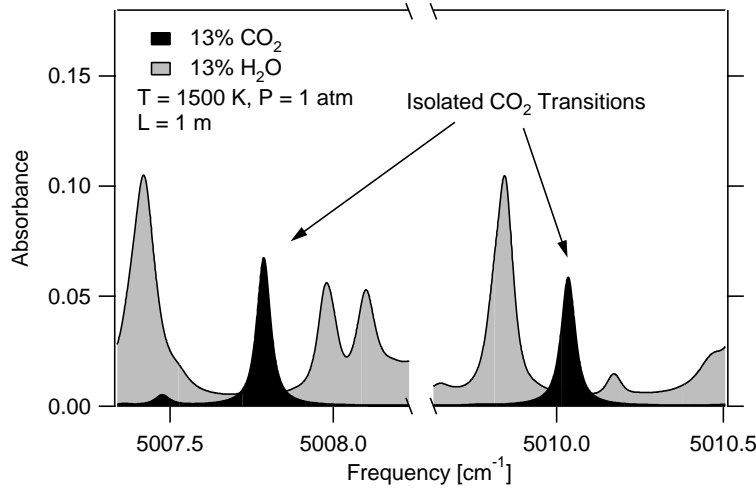


Fig. 2. Two isolated CO₂ transitions suitable for *in-situ* combustion monitoring based on HITRAN96 and HITEMP calculated spectra.

an absorbing medium at a particular frequency is exponentially related to the transition linestrength S_i [$\text{cm}^{-2}\text{atm}^{-1}$], lineshape function ϕ [cm], total pressure P [atm], molefraction of the absorbing species x_j , and the pathlength L [cm].

$$\frac{I_t}{I_o} = \exp(-S_i \phi P x_j L) \quad (1)$$

The linestrength for an absorption transition can be measured using controlled partial pressures of pure CO₂ gas in a static cell that has a known pathlength. For premixed combustion environments, adiabatic temperatures are often greater than 1500 K. Thus, CO₂'s absorption linestrengths at these elevated temperatures must be understood. The linestrength as a function of temperature for a particular CO₂ transition i is governed by its linestrength S_i at a reference temperature T_o ; CO₂'s partition function $Q(T)$; the frequency of the transition, $\nu_{o,i}$; and the lower-state energy of the transition, E''_i . This relationship is given by

$$S_i(T) = S_i(T_o) \frac{Q(T_o)}{Q(T)} \left(\frac{T_o}{T} \right) \exp \left[-\frac{hcE''_i}{k} \left(\frac{1}{T} - \frac{1}{T_o} \right) \right] \times \left[1 - \exp \left(\frac{-hc\nu_{o,i}}{kT} \right) \right] \left[1 - \exp \left(\frac{-hc\nu_{o,i}}{kT_o} \right) \right]^{-1} \quad (2)$$

The measured variation of linestrength as a function of temperature for a particular transition and Equation 2 can be used to determine the lower-state energy of the transition. Once the temperature-dependent linestrength is known, laser absorption can be used to monitor the molefraction x_j of the target species.

Experimental room-temperature linestrength measurements for the two isolated transitions in Figure 2 are listed in Table 1. The linestrength measurements over a range of elevated temperatures are plotted in Figure 3 and overlaid with HITRAN96 predictions for CO₂. The measured lower-state energies for both transitions are also listed in Table 1, along with values from HITRAN96 for comparison.

Position [cm^{-1}]	HITRAN96		Measured	
	S_o [$\text{cm}^{-2}\text{atm}^{-1}$]	E'' [cm^{-1}]	S_o [$\text{cm}^{-2}\text{atm}^{-1}$]	E'' [cm^{-1}]
5007.787	0.00129	994.2	0.00143	983
5010.035	0.000424	1244.2	0.000427	1224

Table 1. Measured room-temperature linestrengths and lower-state energies compared with values from HITRAN96 for the two isolated CO₂ transitions shown in Figure 2.

The measured room-temperature linestrengths disagree with HITRAN96 by 11% for the transition at 5007.787

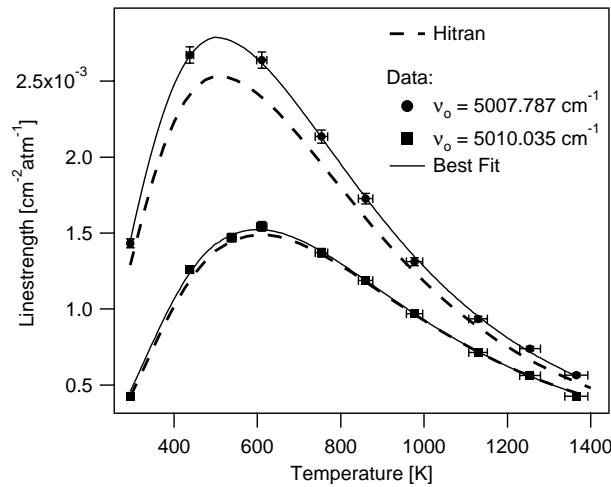


Fig. 3. Linestrengths vs. temperature for two isolated CO₂ lines.

cm⁻¹ and agree within 1% for the transition at 5010.035 cm⁻¹. The measured lower-state energies for both transitions agree within 2% with HITRAN96.

With the development of real-time *in-situ* CO₂ sensors, diagnostics are now available that offer a more complete sensing capacity for combustion environments. Future multiplexing opportunities of CO₂ with H₂O and temperature diagnostics along with novel extended-wavelength room-temperature lasers at 2.3 μm for *in-situ* CO monitoring will be discussed.

Acknowledgments

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