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Rotational CARS for simultaneous measurements of temperature and concentrations of N₂, O₂, CO, and CO₂ demonstrated in a CO/air diffusion flame

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Abstract

Rotational coherent anti-Stokes Raman spectroscopy (CARS) has over the years demonstrated its strong potential to measure temperature and relative concentrations of major species in combustion. A recent work is the development and experimental validation of a CO₂ model for thermometry, in addition to our previous rotational CARS models for other molecules. In the present work, additional calibration measurements for relative CO₂/N₂ concentrations have been made in the temperature range 294–1246 K in standardized CO₂/N₂ mixtures. Following these calibration measurements, rotational CARS measurements were performed in a laminar CO/air diffusion flame stabilized on a Wolfhard–Parker burner. High-quality spectra were recorded from the fuel-rich region to the surrounding hot air in a lateral cross section of the flame. The spectra were evaluated to obtain simultaneous profiles of temperature and concentrations of all major species; N₂, O₂, CO, and CO₂. The potential for rotational CARS as a multi-species detection technique is discussed in relation to corresponding strategies for vibrational CARS.

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1. Introduction

Today there is a strong development of laser diagnostics for application to combustion processes [1,2] and such techniques are regularly used for validation of combustion modeling results. Laser techniques offer unique features such as non-intrusiveness, remote detection capability, and high spatial as well as temporal resolution. Another unique feature for many laser techniques is species specificity. On the other hand, this

pronounced selectivity may be a limitation, in that only a limited number of parameters related to that species can be measured. Multiple species detection is therefore valuable as more information can be extracted from an experiment. Combinations of different techniques or extensions of existing techniques are, however, then usually required.

One of the most important species as a result of combustion is CO₂, and it is of global importance to control the CO₂ emissions from human activities, since CO₂ is considered the main contributor to the greenhouse effect. In this perspective, combustion processes must be developed for higher efficiency and directed from the use of fossil fuels to renewable fuels. Regarding

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quantitative non-intrusive diagnostics of CO₂, it should be noted that its strongest electronic transitions are in the vacuum-ultraviolet (UV) spectral region, thus laser-induced fluorescence (LIF) on CO₂ is therefore of limited use in practical applications. Although CO₂-LIF was recently demonstrated in the infrared (IR) spectral region [3–5] its potential as a quantitative diagnostic for concentration measurements in combustion seems still to be limited. Raman techniques [1] have promising potential for CO₂ monitoring, although spontaneous Raman scattering is subject to interferences due to very weak signals.

One of the most important techniques for quantitative measurements of major species in combustion is coherent anti-Stokes Raman spectroscopy (CARS) [1,2]. This technique has been used since the beginning of the 1970s for combustion diagnostics [6] and mainly for thermometry using nitrogen as the detected species. There are two main approaches of the technique, vibrational CARS and pure rotational CARS. Normally one species can be detected at a time for a vibrational CARS experimental setup, however, simultaneous detection of several species can be made using multi-color vibrational CARS approaches [1]. The experimental setup then becomes more complex with at least an additional laser in comparison with the standard vibrational CARS setup.

The other CARS approach, pure rotational CARS had a scientific breakthrough in 1986 when the dual-broadband approach was introduced [7,8]. In addition to the development of the method for nitrogen thermometry, see for example, Ref. [9], theoretical models have been developed for other diatomics (oxygen [10,11] and carbon monoxide [12]), and also for larger linear molecules (acetylene [13,14] and carbon dioxide [15]). The experimental investigations have been limited so far for the larger molecules and several assumptions have been made in the theoretical codes. Thus more investigations are needed in a wider range of conditions to understand the limitations and further extend the technique for wider applicability.

The first development of a rotational CARS model for CO₂ was done by Schenk et al. [15]. Experimental validation of their model was performed at low temperatures (300–773 K) for pressures up to 5 MPa. In another paper by the same group [16], relative CO₂ concentration measurements in the same temperature and pressure range were demonstrated. In these thorough investigations different linewidth models were tested and the precision was evaluated for both temperatures and concentrations.

In the development of the rotational CARS technique for CO₂ in Lund, the aim is to do multi-species flame diagnostics including CO₂, and recently an investigation on pure CO₂ at higher temperatures and with improved spectral

resolution was presented [17]. The developed rotational CARS model for CO₂ was tested in the temperature range from 294 to 1143 K, and very good agreement with thermocouple data was found. In the present paper, an investigation is presented where the rotational CARS model for relative N₂/CO₂ concentration measurements has been further tested. Calibration measurements were performed in two standardized mixtures of CO₂ and N₂ in the temperature range 294–1246 K. Based on these results, the pure rotational CARS technique was applied to a laminar CO/air diffusion flame stabilized on a Wolfhard–Parker burner. This flame has only four major species; CO, O₂, N₂, and CO₂ (since dissociation is low even at the highest temperature). The rotational CARS setup can without modification probe all four major species since their spectral lines all show up in the same spectral region and our developed theoretical codes can be used for evaluation of all concentrations. Temperature and concentration profiles of CO, O₂, N₂, and CO₂ extracted from the same experimental spectra are presented and discussed.

2. Theory

The general CARS theory has been extensively discussed in the past, see for instance, Ref. [1] and references cited therein. In the dual-broadband rotational CARS approach used in this work, two broadband dye laser beams from a single-dye laser source are used for coherent excitation at the Raman frequencies ω_R , where the photon pairs (ω_1 and ω_2 from the broadband spectral profile) fulfill the criterion $\omega_R = \omega_1 - \omega_2$. The CARS signal is generated at the anti-Stokes signal frequency ω_4 after sum-frequency generation of the Raman frequency ω_R and the narrowband laser frequency ω_3 , i.e. $\omega_4 = \omega_1 - \omega_2 + \omega_3 = \omega_R + \omega_3$.

The CARS signal is modeled through the squared modulus of the third-order susceptibility $\chi^{(3)}$ that in general, when the photon frequencies are far from electronic resonances, has resonant contributions from active rotational and vibrational Raman modes, $\chi_R^{(3)}$, and electronic non-resonant contributions from all molecules in the gas, $\chi_{NR}^{(3)}$.

$$\chi^{(3)}(\omega_1 - \omega_2) = \chi_{NR}^{(3)} + \chi_R^{(3)}(\omega_1 - \omega_2) = \chi_{NR}^{(3)} + \sum_J \frac{a_{JJ+2}}{\omega_{JJ+2} - \omega_1 + \omega_2 - i\text{p}\gamma_{JJ+2}} \quad (1)$$

The resonant third-order susceptibility can, in the isolated-line approximation, be written as a sum of Lorentzian functions representing the different Raman transitions. For a single species, the rotational CARS susceptibility can be written as (for multiple species there is also a sum over species),

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