Femtosecond coherent anti-Stokes Raman scattering (fs-CARS) for temperature and concentration measurements on combustion species using a dual output OPCPA

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Abstract. Using two beam femtosecond coherent anti-Stokes Raman scattering (fs-CARS), temperature and concentration measurements are performed on relevant combustion species such as H_2, N_2, CO, and CO_2.

1 Introduction

Simultaneous temperature and concentration determination is of great importance for applications in gasification [1] and combustion [2]. In this regards, femtosecond coherent anti-Stokes Raman scattering (fs-CARS) is a suitable approach, in particular for high pressure and high temperature conditions [3]. Furthermore, two beam fs-CARS, using a few-cycle pulse for Raman excitation, in principle, allows determining gas temperatures and concentrations of multiple species from a single spectrum [4, 5]. In this study, we use an optical parametric chirped pulse amplifier (OPCPA) to provide both a broadband ~ 7 fs pump/Stokes pulse and a ~ 1 ps probe pulse.

2 Experimental Setup

The experimental setup is shown in Fig. 1. The two outputs of an OPCPA (venteon OPCPA, Laser Quantum GmbH, Hannover, Germany) are used for the pump/Stokes pulse (650 nm - 1100 nm, 10 µJ, 200 kHz), and the probe pulse (515 nm, 20 µJ, 200 kHz). These two beams are overlapped using a long pass dichroic mirror (LPDM) and then focused into the gas reactor using a concave mirror (CM). This reactor is designed for high temperature and high pressure environment with optical ports made of sapphire [6]. Afterwards, the CARS signal is separated from the residual pump/Stokes and probe pulses with a LPDM and is focused into the spectrometer. The modelling for determining the gas temperature and concentration

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from the CARS spectra is described in detail elsewhere [5, 7]. In this study, experiments were performed on the combustion relevant species H_2, N_2, CO, and CO_2.

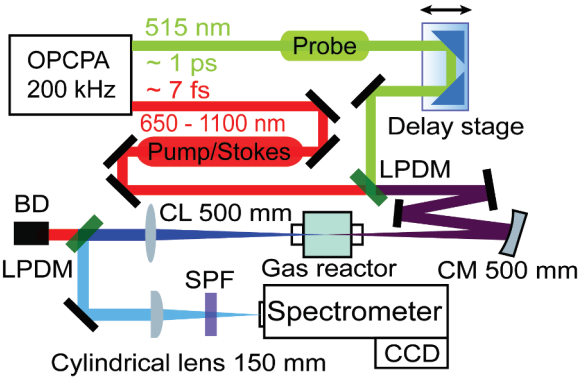

Fig. 1. Fs-CARS experimental setup. LPDM: long pass dichroic mirror; CM: concave mirror; CL: collimating lens; SPF: short pass filter.

3 Temperature and concentration measurements

Fig. 2 shows experimental and simulated fs-CARS spectra for concentration determination. Fig. 2(a) shows the fs-CARS spectrum of 2 % CO in a CO and CO_2 gas mixture at a temperature of 1073 K and a pressure of 1.4 bar. The best fit of the simulation yields a CO concentration of 1.8 %. Fig. 2(b) shows the fs-CARS spectrum of a gas mixture of multiple species with 7 % H_2, 55 % N_2, and 38 % CO_2 at 478 K and 1.4 bar, which covers Raman shifts from ~ 1200 cm^{-1} to ~ 4180 cm^{-1}. The corresponding best fits are: 9 % H_2, 45 % N_2, and 46 % CO_2. The deviation between experimental concentration and simulated results is probably mainly influenced by the inhomogeneity of the pump/Stokes spectrum, and will be analysed in future work.

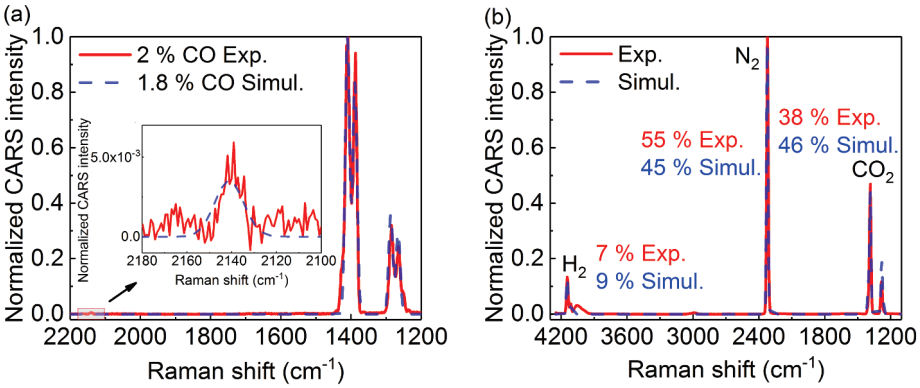

Fig. 2. Experimental and simulated fs-CARS spectra for concentration determination for (a) 2 % CO (simulated 1.8 %) in a CO-CO_2 gas mixture at 1073 K and 1.4 bar (b) gas mixture of 7 % H_2, 55% N_2, and 38% CO_2 (simulated 9 %, 45 % and 46 %, respectively, for H_2, N_2 and CO_2) at 478 K and 1.4 bar.

For CO_2, we have shown recently that the gas temperature can be determined from a single spectrum by means of the temperature dependent population of the vibrational levels [7]. Here, we show that a similar approach is feasible for H_2. Due to the high values of the spectroscopic rotational and vibrational constants of H_2, the Q-branch CARS lines are widely spaced [8] and are therefore at least partly resolved by our ps probe pulse.

Fig. 3 shows the corresponding fs-CARS spectra of H_2 at 296 K/478 K with simulated temperature of 323 K/528 K. The Q-branch Q(0), Q(1) and Q(2) lines are apparently hidden within the first large peak at ~ 4151 cm^{-1}, whereas the Q(3) peak can be clearly identified. The pronounced background covering 3950 cm^{-1} to 4100 cm^{-1} range comes from the pump/Stokes beam of the OPCPA. Future work will try to filter out this background and optimize the probe pulse duration to improve the accuracy of the temperature measurement.

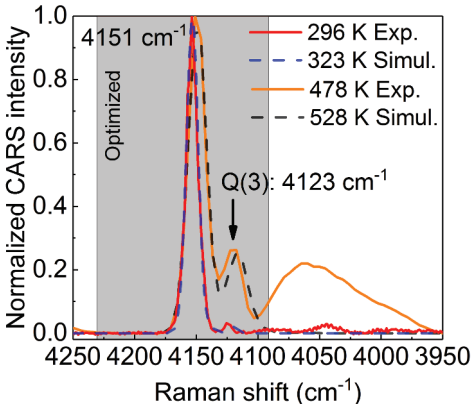

Fig. 3. Experimental and simulated fs-CARS spectrum for temperature determination based on H_2 at 296 K (simulated 323 K) and 478 K (simulated 528 K) of 1.4 bar. The shadowed grey area shows the optimization range for temperature determination.

4 Conclusions

To summarize, two beam fs-CARS using a dual output OPCPA allows concentration measurements in a wide temperature range for the Raman shifts of the most relevant combustion species. For temperature measurements, we detected the ro-vibrational fs-CARS spectrum of H_2, and proved its feasibility for temperature determination based on a single spectrum.

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