Modeling and Parametric Studies on Chirped Probe Pulse Femtosecond CARS Spectra Applied to Hydrogen

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The growing demand for energy and reliance on fossil fuels contribute to the increase in greenhouse gas emissions and resource depletion. In response, hydrogen fuel is emerging as a sustainable alternative to conventional fuels. However, its efficient use in propulsion engines requires a deeper understanding of combustion mechanisms. Laser-based diagnostics provide a valuable solution for in-situ combustion analysis, as they are non-intrusive and enable quantitative measurements of local temperature and chemical species. Temperature plays a key role in characterizing and understanding the physico-chemical processes involved in combustion. Moreover, due to hydrogen's high diffusivity, high-speed laser diagnostics are required for characterizing flame dynamics. In the current study, temperature is measured by Chirped Probe Pulse Femtosecond Coherent Anti-Stokes Raman Spectroscopy (CPP-fs-CARS) [1]. This measurement strategy uses a Legend Elite Laser system combined to a TOPAS Prime Plus to probe H_2 rovibrational Raman transitions with two 100 fs laser pulses (Pump at 603 nm and Stokes at 800 nm). A delayed 603 nm probe chirped pulse, temporally stretched in a 30 cm glass rod, probes coherence to perform single-shot measurements of gas temperature with a folded BOXCARS configuration at repetition rates up to 10 kHz. Thanks to energy and wave-vector conservation, a coherent CARS signal is generated and recorded by a spectrograph/EMCCD camera, then processed by a multi-parameter least-squares minimization procedure via a genetic algorithm to compare experimental spectra with theoretical ones. This post-processing method has proven its effectiveness in analyzing CARS spectra for temperature measurement by probing the rovibrational energy states of nitrogen (N_2), from academic to practical kerosene/air flames encountered in aero-engine operating conditions [2]. Although this laser-based diagnostic has been revealed large potential when probing N_2, there are rare demonstrations regarding the feasibility of probing H_2 by CPP fs-CARS [3]. To perform further investigations, a home-made CARS simulation code was developed to calculate H_2 CPP fs-CARS spectra. This modeling includes the spectroscopic constants and thermodynamic properties of H_2. The simulation code was validated against data from literature and this reliability enables a parametric analysis of the effects of temperature and probe pulse characteristics, serving as an initial step in the definition of the future experimental setup. Figure 1 shows the spectral intensity of theoretical H_2 CARS spectra as a function of temperature, ranging from 500 K to 2500 K with a probe delay of 7 ps. The simulations, based on Gaussian pulses for the pump, Stokes, and probe pulses, show how the number of modulations in the H_2 CARS signal increases with increasing temperature. This spectral evolution follows the Boltzmann distribution, where higher temperatures lead to larger populations in the higher rotational levels of the first vibrational band. Additionally, compared to N_2, the significantly larger energy gap between H_2 rotational energy levels leads to an increase in spectral width as the temperature increases. For the post-processing operation, this strong spectral dependence has the benefit of improving the efficiency of the least-squares minimization procedure. These simulations, along with additional parametric studies, will be presented to illustrate the impact of various laser parameters and arrangements on the spectral response of H_2. Experimental investigations will complement this study.

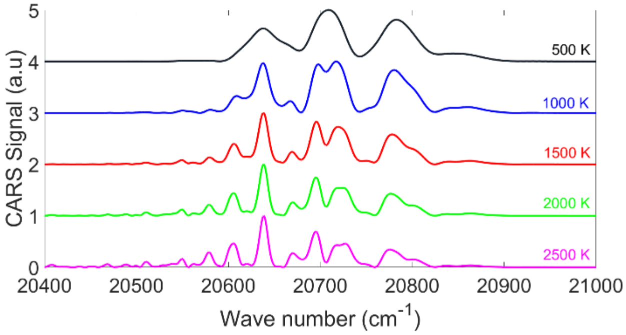

Figure 1 : H_2 CPP fs-CARS spectra with temperature

1. D.R. Richardson et al., Applied Physics B, Vol. 104, 2011, 699-714.
2. S. Legros et al., Combustion and Flame, Vol. 224, 2021, pp. 273–294.
3. Z. Chang et al., Opt. Express 32, 13701-13719 (2024)