

Hybrid fs/ps rotational CARS temperature/species detection in flames at kHz rate

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Abstract: A hybrid rotational CARS scheme with femtosecond pump/Stokes preparation and a high-energy probe, generated by second-harmonic bandwidth compression (SHBC), is described. The instrument is demonstrated for collision- and background-free, kHz-rate temperature/oxygen probing in the product gases of C₂H₄/air flat flames for both lean and rich conditions..

OCIS codes: (120.1740) combustion diagnostics; (300.6230) spectroscopy, coherent anti-Stokes Raman scattering

1. Introduction

Coherent anti-Stokes Raman scattering (CARS) is a powerful tool for temperature and species detection in combustion. Recent work [1-5], has focused on the utility of femtosecond laser sources for CARS combustion diagnostics. These new approaches enable collision- and background-free detection, with dramatic improvements in data rate and precision. For CARS of pure-rotational Raman transitions at kHz rates, hybrid schemes utilizing broadband femtosecond pump/Stokes preparation and a frequency narrow probe for resolution of the Raman spectrum have been preferred [2,4]. These earlier studies utilized filtering of the femtosecond bandwidth for generation of the picosecond probe beam—a convenient, but inefficient, approach that discards most of the available pulse energy and limits the technique to low-temperature measurements. More recently, we have applied sum-frequency generation of two linearly chirped, phase-conjugate pumps to generate high-energy (1 mJ or more) picosecond pulses at the second harmonic of the pump frequency [5]. This approach was demonstrated in H₂/air combustion products up to 2000 K. Here, we report new data in which the new Second Harmonic Bandwidth Compressed (SHBC) probe is applied to fuel-rich C₂H₄/air flat flames stabilized on the McKenna burner.

2. Experiment

A Ti:sapphire amplifier provided nearly Fourier-transform-limited, ~100-fs duration, 3.1-mJ pulses at 1-kHz repetition rate. The amplifier output spectrum was centered near 800 nm, with a nominal bandwidth of ~180 cm⁻¹ (FWHM). Eighty percent of the amplifier output energy was used to generate a frequency-narrow probe pulse at 400 nm, using the SHBC device illustrated in Figure 1. Pump and Stokes energies were 100 μ J/pulse each. The linear pump and SHBC probe polarizations were rotated to be orthogonal to the Stokes beam, so that the resulting CARS signal beam was orthogonally polarized to the high-energy, spectrally proximal SHBC probe, allowing for efficient background rejection with an analyzer placed in the CARS collection optics. The resulting CARS signal beam near 400 nm was dispersed onto a back-illuminated electron-multiplying CCD detector at a dispersion of 0.5 cm⁻¹/pixel using a 1-m grating spectrograph.

For an SHBC-generated picosecond probe, the broadband ~100-fs pump beam was split into two equal-energy legs, which were then linearly chirped. Equal-and-opposite conjugate phase was imparted to each pump by two carefully matched stretchers that utilize a common diffraction grating. Sum-frequency generation of the two pumps in beta-barium-borate (BBO) results in annihilation of the linear chirps in the picosecond output beam centered at $\omega_{SFG} = \omega_1 + \omega_2 = 2\omega_0$. In these experiments, the SHBC probe pulse-duration was 5.8 ps FWHM with a bandwidth of 4.1 cm⁻¹, and an energy in excess of 1 mJ.

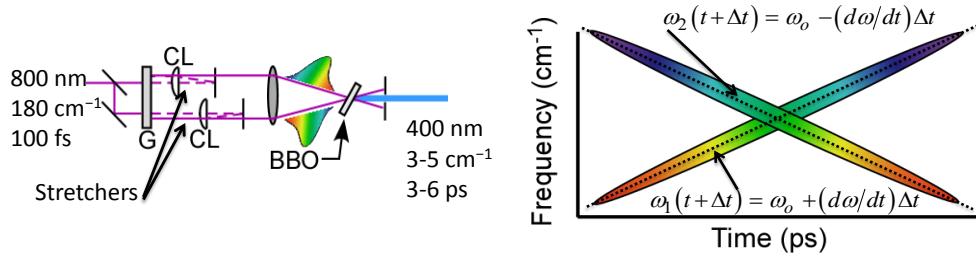


Fig. 1. Second Harmonic Bandwidth Compression Scheme.

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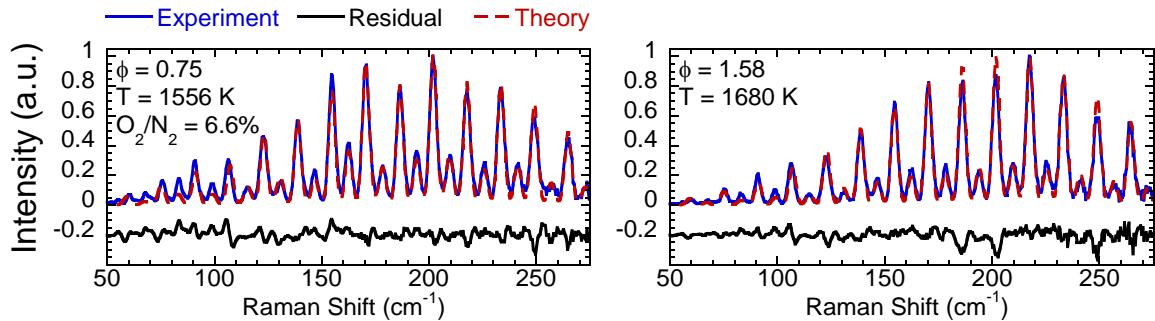


Fig. 2. Single-laser-shot rotational CARS spectra from C_2H_4 /air flames on the McKenna burner.

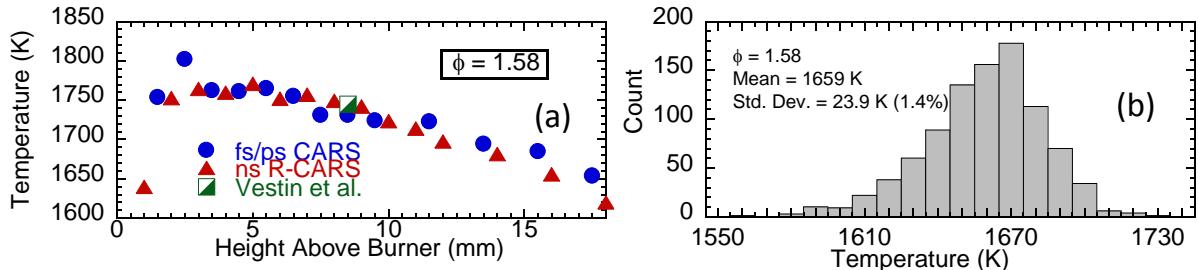


Fig. 3. Vertical temperature profiles monitored with fs/ps rotational CARS (spectra averaged for 100 laser shots) and traditional ns rotational CARS (a). Histogram constructed from 1000 single-shot fs/ps CARS temperature measurements (b).

3. Results and Discussion

Single-laser-shot spectra were acquired at 1-kHz in the product gases of C_2H_4 /air flat flames stabilized on the McKenna burner, with a delay of 16 ps between the 400-nm probe beam and 800-nm femtosecond preparation pulses. This delay was chosen to minimize complications from CO_2 on the spectra, and to eliminate nonresonant background. Collisional effects influence the evaluated temperature by only $\sim 0.7\%$ at this probe delay.

Representative spectra for lean ($\phi = 0.75$) and rich flames ($\phi = 1.58$), and the associated theoretical fits, are shown in Figure 2. Good agreement between the measured and calculated spectra is achieved for fuel-lean conditions. In rich flames, there is some systematic deviation between the fitted and measured spectral envelopes. Interaction between the CO and N_2 Raman polarizations not captured in the spectral modeling code may be the cause of this discrepancy, as CO production increases markedly under fuel-rich conditions. Accuracy of the measured temperatures at $\phi = 1.58$ was checked by comparing the fs/ps CARS data to nanosecond rotational CARS results recorded in our lab and by Vestrin *et al.* [6]. A profile of the measured temperatures as a function of height above the center of the burner is shown in Figure 3a, where the fs/ps CARS are generally within 1% of the nanosecond CARS measurements in the 5-15 mm region where the flame is most stable. The level of agreement illustrates the robustness of temperature measurement to the overall spectral envelope, where some localized discrepancy in the fits does not appear to have a great impact on the measured temperatures. The precision of the fs/ps CARS temperatures is excellent. A histogram constructed from 1000 single-shot temperature measurements is shown in Figure 3b for the $\phi = 1.58$ flame. The standard deviation of the measured temperatures is 1.4% of the mean, illustrating the outstanding precision achieved when nearly Fourier-transform limited femtosecond pump/Stokes preparation is employed.

References

- [1] D.R. Richardson, D. R., Bangar, and R.P. Lucht, "Polarization Suppression of the Nonresonant Background in Femtosecond Coherent Anti-Stokes Raman Scattering for Flame Thermometry at 5 kHz," *Opt. Express* **20**, 21495-21504 (2012).
- [2] H. Stauffer, J.D. Miller, S. Roy, J.R. Gord, and T.R. Meyer, "Hybrid femtosecond/picosecond rotational coherent anti-Stokes Raman scattering thermometry using a narrowband time-asymmetric probe pulse," *J. Chem. Phys.* **136**, 111101 (2012).
- [3] B.D. Patterson, Y. Gao, T. Seeger, C.J. Kliewer, "Split-probe hybrid femtosecond/picosecond rotational CARS for time-domain measurement of S-branch Raman linewidths within a single laser shot," *Opt. Lett.* **38**, 4566-4569 (2013).
- [4] S.P. Kearney, D.J. Sciglietti, and C.J. Kliewer, "Hybrid femtosecond/picosecond rotational coherent anti-Stokes Raman scattering temperature and concentration measurements using two different picosecond-duration probes," *Opt. Express* **21**, 12327-12339 (2013).
- [5] S.P. Kearney and D.J. Sciglietti, "Hybrid Femtosecond/Picosecond Rotational Coherent Anti-Stokes Raman Scattering at Flame Temperatures Using a Second-Harmonic Bandwidth-Compressed Probe," *Opt. Lett.* **38**, 833-855 (2013).
- [6] F. Vestrin, M. Afzelius, C. Brackmann, and P.E. Bengtsson, "Dual-Broadband Rotational CARS Thermometry in the Product Gas of Hydrocarbon Flames," *Proc. Combustion Institute* **30**, 1673-1680 (2005).