Vibrational and Pure-Rotational Hybrid fs/ps CARS for High-Speed Thermometry

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Summary
The need for accurate and precise measurements of temperature and species concentration with high spatial and temporal resolution in turbulent combusting flows has motivated the development of several new laser-based probes. We are currently studying the use of a hybrid fs/ps coherent anti-Stokes Raman scattering technique that addresses several challenges related to practical combustion systems:

- Determination of temperature from 300 - 2500 K, and concentration of multiple species simultaneously (N₂, O₂, CO, CO₂)
- Measurement acquisition of 1-10 KHz
- Suppression of interferences such as nonresonant background
- Insensitivity to high pressure effects (energy transfer effects)

Time- and Frequency-Resolved Model

The pump and Stokes pulses (100 fs) impulsively excite the Raman polarization. The probe pulse is spectrally narrowed and delayed in time by τ. For VCARS, the pump and CARS pulses are degenerate, while RCARS is fully degenerate. Neglecting the nonresonant contributions, the CARS intensity is given as:

\[ I_{CARS}(\omega, \tau) \propto \int \frac{N_c}{n_e} E_1(t) \int d\Omega [ R(t) \left( E_2^* \left( t+\tau_2 - \tau_1 \right) E_2(t+\tau_2) e^{i\omega_{21} \tau_1} \right)]^2 \]

Suppression of NR Background
Both time-symmetric (sin² in time) and time-asymmetric (step-exponential in time) probe pulses were studied. These resulted from probes with flat-top and Lorentzian spectral profiles, respectively. Both suppress NR by 1000×, but the Lorentzian probe allows shorter probe delays for greater signal/NR.

Single-Shot fs/ps Vibrational CARS
High temperatures (1200 – 3000 K) can be measured by probing the ground and excited vibrational states of the molecule using VCARS.

Single-Shot fs/ps Rotational CARS
Low temperatures (300 – 1200 K) can be measured by probing the rotational states of the molecule using RCARS. Both N₂ and O₂ have been studied with single-shot thermometry accuracy of ~1%. Simultaneous measurements of both are possible.

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