Title: Direct Coherent Raman Temperature Imaging and Wideband Chemical Detection in a Hydrocarbon Flat Flame

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Supporting Information:

Synthetic temperature profile and spatial-filtering functions

The synthetic temperature profile describing the combustion time-history of a premixed-flame was simulated as a variant of an exponential-growth process, and implemented with the logistic function, $T(k)$, progressed over discrete indices k according to:

$$T(k) = (T_{end} - T_{start})\frac{1}{1+e^{-(k-\Delta k)/b}} + T_{start}, k = \{1,2,...,100\} \qquad \text{Eqn (1)}$$

with boundaries T_{end} = 2000 K, T_{start}= 300 K, inflexion point Δk=50 and rate-constant b={5,10}. The domain of the argument is arbitrary, however, with the range set to span ~2mm the choices of b representing the two flames with thicknesses ~360 μm and ~720 μm, respectively.

The spatial-filtering Gaussian functions simulating the imaging system blur were generated according to:

$$g(k) = C^{-1}e^{-\left(2\sqrt{ln(2)}(k-\Delta k)/w\right)^2}, w = \{2,5,7.5\} \qquad \text{Eqn (2)}$$

where C is the integral normalization constant, and the employed width (w) representing the current filters with 40 μm, 100 μm and 150 μm FWHM, respectively.

CARS signal simulation and spectral fitting

The temperature was evaluated by fitting the extracted spectra to a theoretical library of pre-calculated spectra for the prevailing conditions. The calculated CARS spectrum is recovered via the Fourier transform of the time-dependent CARS response (E_{CARS}), which is the product of the impulsively excited N_2 transients and the delayed (τ) probe-electric-field time-varying amplitude (E_{probe}), according to;

$$E_{CARS}(t,\tau) = E_{probe}(t-\tau)\sum_{J=0}^{J\,max} \Lambda_J e^{\left((i\Delta E_J - \Gamma_J)t/\hbar\right)} \qquad \text{Eqn (3)}$$

where ΔE_J denote the N_2 transition energy, Γ_J the reciprocal dephasing time, and Λ_J the oscillation amplitude of the *J*-specific S (Δv=0,ΔJ=2) transition involved. The specific contributions from major perturbing species-colliders on the total dephasing time were included in the calculation and weighted according to chemical equilibrium calculations of the specific flame-conditions involved. The oscillation amplitude depends on the isotropic- and/or anisotropic polarizability tensor invariant, and the normalized Boltzmann population difference factor. The CARS response is calculated here as an unmodulated signal, cancelling the probe-electric-field carrier-frequency, improving the speed of calculation. The probe-electric-field amplitude is approximated with a Gaussian shape (70 ps, FWHM), as verified through autocorrelation, measuring the CARS signal in a series of probe-delay-scans performed in a non-resonant gas of argon.