Single-Beam CARS Imaging for Reacting Flow Diagnostics

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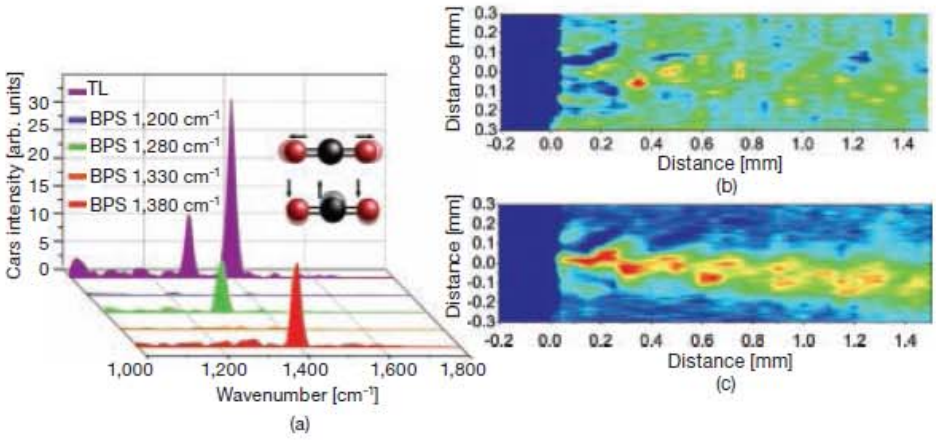
Coherent anti-Stokes Raman Scattering (CARS) is a long-standing diagnostic technique of choice in the combustion community for measuring temperature and major species concentration in gas-phase-reacting flows. Additionally, the coherent excitation of Raman-active molecular vibrations offers intrinsic chemical specificity and high spatial resolution.

The application of traditional nanosecond and picosecond CARS is hindered by its cumbersome experimental implementation, requiring multiple laser beams that are spatially and temporally overlapped in a complex beam geometry. However, advances in laser and pulse-shaping technologies have made it possible to measure the CARS signal using a single laser beam.[1] The use of supercontinuum sources allows for acquisition of an entire Raman spectrum without wavelength scanning or changes to experimental parameters.

In single-beam CARS, researchers use spectrally broad, ultrashort pulses that contain the pump, Stokes, and probe components. In our work, sub-10 fs amplified pulses are capable of impulsively exciting Raman transitions with wavenumbers up to 2,500 cm^{-1} range.[2,3] A narrow region on the high-frequency side of the spectrum is selected as the probe. Its polarization is rotated by 90° and a π-phase step is applied to minimize the nonresonant contribution due to the instantaneous electronic response. Additional phase-modulation techniques can be used to selectively excite a single vibrational mode. More importantly, mode-selective phase modulation allows for the elimination of multichannel detection. Single-channel acquisition is desirable for imaging applications that require high-speed beam scanning.

One proven method for mode-selective excitation is the use of pseudo-random binary sequences.[4] We have applied binary phase shaping (BPS) for mode-selective CARS in liquids and, more recently, in gas-phase samples.[2,5] We have demonstrated independent excitation of CO_2 Fermi dyads and nearly complete suppression of the nonresonant four-wave mixing, as illustrated in (a).



Selective single-beam CARS: (a) CARS spectra of pure CO_2 gas in a cell for a set of phase functions targeting Raman transitions at various wave numbers. Excitation selectivity is accomplished using BPS. (b) Image of a CO_2 jet taken without mode-selective excitation. The CARS signal is integrated over 300-2,500 cm^{-1} range. (c) Image of the same CO_2 jet with the selective excitation of the symmetric stretch (1,280 cm^{-1} band).

To highlight the advantages of selective Raman excitation for species imaging, we have focused the beam of shaped pulses into a jet of CO_2 flowing from a rectangular nozzle mounted on a motorized 2-D translation stage. The spectrally integrated CARS signal is collected at each *XY* position of the stage. Two different pulse shaping methods are used: (i) impulsive excitation for all Raman-active vibrational modes using transform limited pulses and (ii) selective excitation of the symmetric stretch of CO_2 at 1280 cm^{-1} using BPS.

The image obtained using impulsive excitation (b) exhibits poor contrast due to the resonant contribution from N_2 and O_2 molecules, abundant in ambient air, and the nonresonant response that dominates the low-frequency part of the integrated CARS spectrum. The second phase shaping approach (c) has the advantage of suppressing excitation at Raman frequencies other than the target frequency as well as nearly complete elimination of the nonresonant background. It provides high imaging contrast for the jet of CO_2 gas relative to ambient air. Furthermore, static turbulent modulation of the CO_2 flow is clearly visible. ▲

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References

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