Ultrafast four-wave mixing strategies for the detection and quantification of reactive atomic species: hydrogen, oxygen, and fluorine

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The detection, quantification, and imaging of trace reactive atomic species in applications of low temperature plasma, such as during discharge of next generation power sources, plasma-assisted catalysis, and semiconductor etching applications, is of critical importance to the development of predictive kinetic plasma chemistry models. Such models in turn are paramount to the optimization of these critical processes. In many such applications, spectroscopic measurements are plagued with bright backgrounds and unknown collisional environments that hinder accurate measurement of reactive atomic species concentrations. We have recently explored the use of femtosecond nonlinear four-wave mixing approaches for the quantifiable detection and imaging of atomic hydrogen, atomic oxygen, and atomic fluorine during volumetric plasma operation and during high current breakdown discharges. In the case of hydrogen atom, we have demonstrated the capability of femtosecond two-photon-resonant (n=1 to n=2) parametric four-wave mixing detection for measurements in the extremely bright environment of a high current pulsed discharge. For the detection of atomic oxygen and fluorine, we have developed femtosecond coherent anti-Stokes Raman spectroscopy, another time-resolved four-wave-mixing variant, using the Raman transitions between spin–orbit coupled ground states. Through time-resolved measurement of the electronic coherence decays, we present a quantifiable determination of the concentration of these trace species.

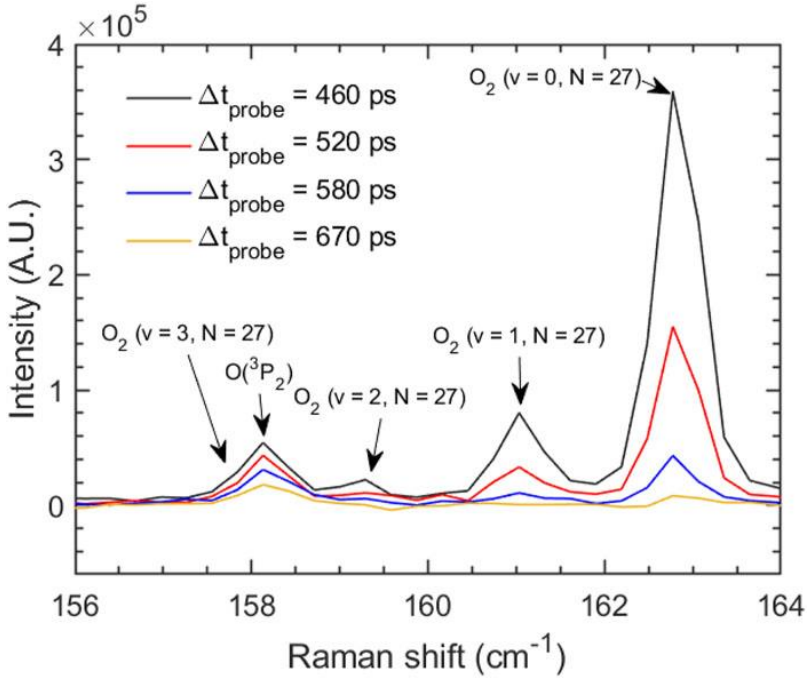

Figure 1. CARS spectrum of atomic oxygen in a H_2/O_2/Ar diffusion flame. Locations of O_2 (v = 0–3, N = 27) and $O(^3P_2)$ are indicated by the arrows.

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