Simultaneous Imaging of Molecular Rovibrational Nonequilibrium, Reactive Species, and Electric Field for Plasma-Assisted Chemistry

Christopher J. Kliewer[(a)], Madeline Vorenkamp[(b)], Brian D. Patterson[(a)], Andrey Y. Starikovskiy[(b)], and Yiguang Ju[(b)]

(a) Sandia National Laboratories (cjkliew@sandia.gov)
(b) Princeton University (yju@princeton.edu)

Short Summary: The Sandia-PRF has built a new capability for the low-temperature plasma community for the simultaneous imaging of molecular rotation/vibration nonequilibrium, electric field, and the distribution of OH radical and formaldehyde in reactive low temperature plasma systems. The system is currently investigating the plasma-assisted deflagration to detonation transition in a micro-combustor channel.

The interaction between low temperature plasma and complex chemistry can yield enhanced rates of desired chemical reactions, shift reaction selectivity towards desired products, or even improve the control of otherwise unstable/chaotic chemical events. The combined kinetic and physical models describing the combination of low temperature plasma and complex chemical networks, however, are only beginning to emerge, and simplifying assumptions in the detailed kinetic models as well as the plasma physics are required. However, multicomponent experimental benchmark data is required to validate or improve such models. This is a required step for the rational optimization of plasma-assisted chemistry.

To answer this need, the Sandia-PRF has built an experimental capability for simultaneous and time-resolved measurement of several key physical and chemical states. The group has recently developed a new approach to directly measure the rotational and vibrational energy distribution of molecules through hybrid femtosecond/picosecond (fs/ps) coherent anti-Stokes Raman spectroscopy (CARS) of pure-rotational transitions [1]. This capability has been implemented in the present apparatus in a 1D imaging configuration such that a freeze-frame of the rotational and vibrational temperature is obtained across a one-dimensional 10 mm line. Overlapped with this probed region in the new apparatus, a separate nanosecond UV pulsed laser at 355 nm excites overlapping electronic transition of the formaldehyde molecule, which is a known marker of low-temperature oxidation chemical pathways. Formaldehyde fluorescence is isolated through spectral filters and imaged onto an intensified CCD camera. Formaldehyde is consumed under high temperature oxidation, where OH radical persists at high concentrations. Similarly, the laser is tuned to 308 nm for the excitation and fluorescence detection of OH radical. Finally, the electric field induced in the channel during DBD plasma initiation is measured through the electric field induced second harmonic generation signal (EFISH), which is obtained "for free" along the CARS fs beam line. The overall system thus enables the assessment of molecular thermal nonequilibrium, chemical markers of high temperature and low temperature oxidation chemistry, and electric field in a simultaneous and time-resolved manner with respect to the plasma pulse timing.

References [1] T.Y. Chen, B.M. Goldberg, B.D. Patterson, E. Kolemen, Y. Ju, C.J. Kliewer, Opt. Lett. **45**, 4252 (2020).

Customer: DOE/SC FES
POC: Christopher Kliewer, cjkliew@sandia.gov

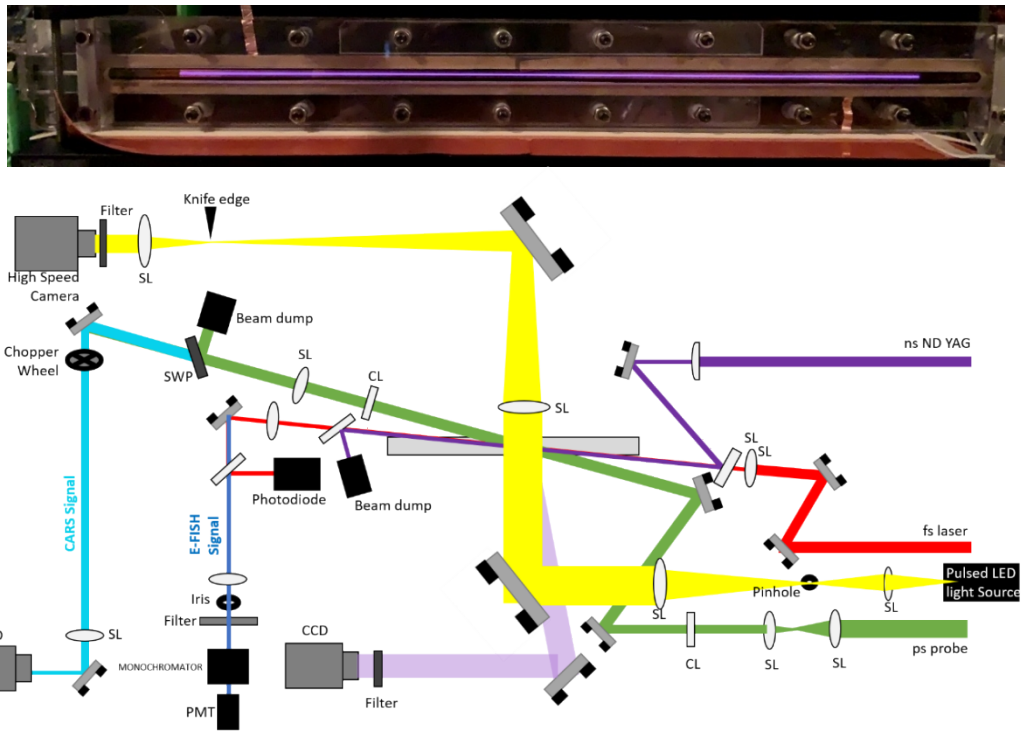

Fig. 1. (Top) Uniform plasma created in the reactive mixture (dimethyl ether and O_2) through dielectric barrier discharge prior to combustion initiation in a microchannel. Clear evidence for enhancement and stabilization of the deflagration to detonation has been demonstrated. (Bottom) Experimental layout for combined ultrafast 1D CARS (thermal nonequilibrium imaging), nanosecond LIF imaging of formaldehyde (low temperature oxidation chemistry), OH radical (high temperature oxidation chemistry), and EFISH (electric field).

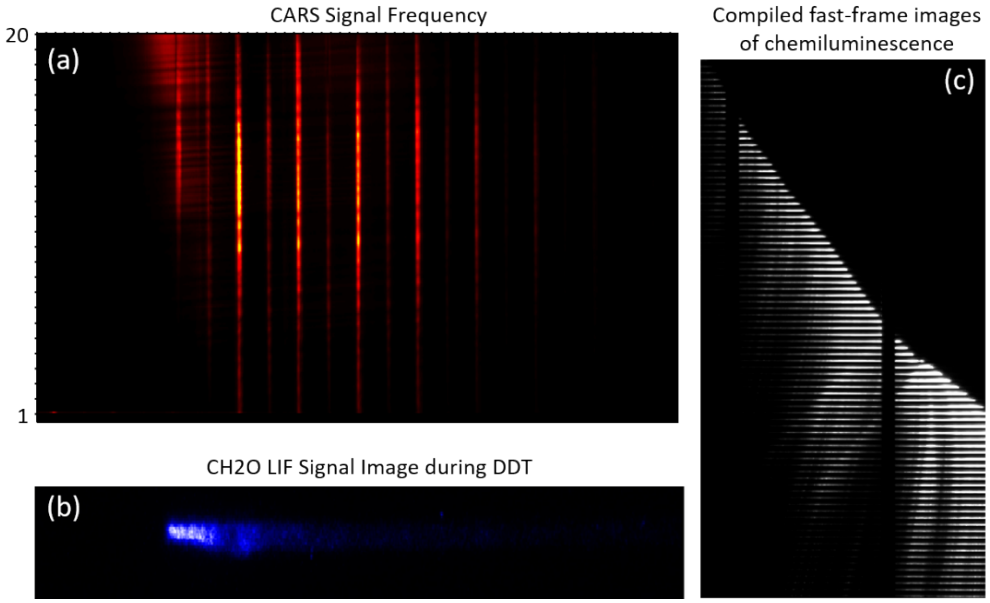

Fig. 2. (a) 1D Fs/ps CARS image taken within the microchannel during DBD discharge. Each horizontal slice through the data produces a pure-rotational CARS spectrum which is fit for rotational and vibrational temperature. (b) Formaldehyde laser-induced fluorescence image indicative of low-temperature chemistry in the plasma assisted combustion. (c) Compilation of fast-framing

chemiluminescence images evaluating the transition from deflagration flame velocities to detonation as a function of plasma DBD pulsing.