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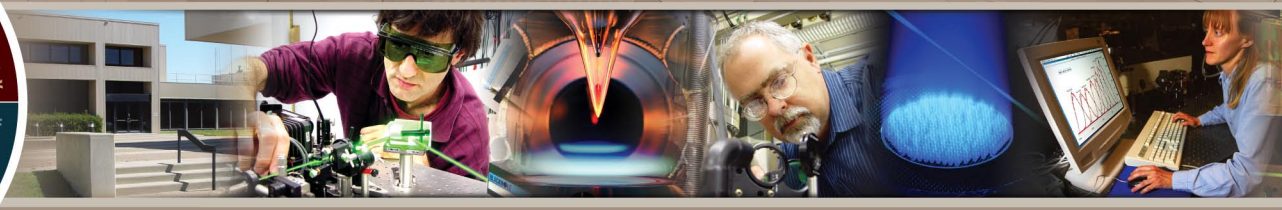

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Laser-based detection of elemental mercury emissions

Increasingly stringent regulations regarding the pollutants released from coal-fired power plants have triggered the development of next-generation sensors that can monitor trace gas emissions with high specificity, selectivity, and with a fast time response. Sandia researchers have been developing a real-time, laser-based mercury emissions monitor that can detect and differentiate between the two forms of gas-phase mercury emitted from utility coal boilers, Hg0 and $HgCl_2$.

(Continued on page 2)

Improving Laser Diagnostics

Picosecond lasers produce interference-free two-dimensional measurements of atomic hydrogen in flames

Atomic hydrogen is a central radical in combustion chemistry, and quantitative measurements of H-atom concentrations are important for understanding the coupling between chemistry and transport in flames. The hydrogen atom is highly reactive and diffusive, and preferential diffusion of hydrogen atoms can affect both flame structure and the dynamics of ignition and extinction.

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Stabilization of lifted turbulent jet flames in a heated coflow

When a jet flame is lifted from the burner by increasing the fuel or surrounding air co-flow velocity, the flame can stabilize without a physical element to use for stabilization, and a lifted jet flame is generated. The flame can exist over a range of jet velocities until a critical velocity is reached and the flame blows out globally. Lifted flames are found in practical applications like industrial burners for power generation, where a lifted jet flame is utilized to reduce damaging thermal stresses to the nozzle material by minimizing contact between the flame and the nozzle. They are also found in stratified combustion – for example, in direct injection gasoline engines, diesel engines, and gas turbines – where the fuel and oxidizer streams are partially premixed prior to combustion. The position downstream of a fuel injector where a diesel fuel jet establishes a flame, the so-called "stabilization point", influences the degree of premixing between the cold fuel and heated air prior to combustion which, in turn, affects the combustion and soot formation processes downstream. For example, soot levels decrease as the fuel and air streams are better mixed upstream of the stabilization point and can even be eliminated if sufficient premixing has occurred upstream. Therefore, fundamental knowledge of the mechanism by which a lifted flame stabilizes in a hot environment of air or combustion products will lead to the development of predictive models used in engineering CFD for the design and optimization of fuel efficient, clean burning combustion devices.

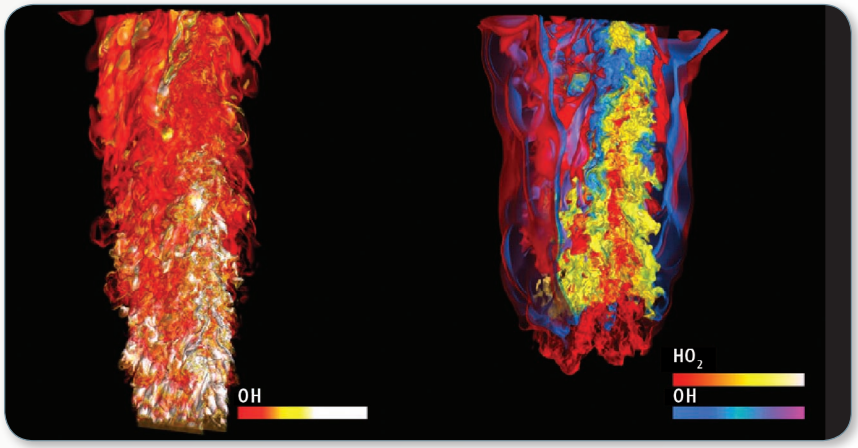

Figure 1. *3D volume rendering of scalar dissipation rate (left) and the mass fraction of OH and HO_2. OH varies from blue (low) to violet (high) and HO_2 varies from red (low) to white (high).*

Despite the practical importance of lifted flame base stabilization, however, there has thus far been little consensus among researchers regarding the dominant mechanism which stabilizes a lifted flame base, not only because of the complex structure and propagation characteristics of turbulent lifted flames, but also because of the difficulty in obtaining simultaneous time-resolved time-series measurements of multi-scalar and velocity fields.

Recently, well-resolved terascale 3D direct numerical simulation (DNS) of the near

(Continued on page 4)

Laser-based detection (cont.)

(Continued from page 1)

With funding from the National Energy Technology Laboratory, Alex Hoops, Tom Reichardt, and Dahv Kliner have characterized and quantified the detection approach for $HgCl_2$ (*CRF News*, Vol. 27, No. 3) and demonstrated the ability to detect ppb levels of $HgCl_2$ using the frequency-quintupled output of a fiber laser (*CRF News*, Vol. 29, No. 1). Recent efforts have been directed towards developing a compact, tunable, ultraviolet (UV) laser and filtering approach suitable for detecting Hg0.

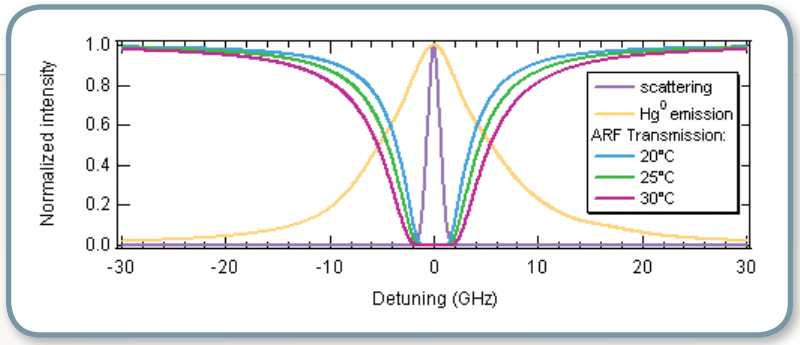

Figure 1. *Spectral modeling of the elastic scattering, Hg0 emission from the probe volume, and atomic resonance filter transmission for several filter temperatures.*

Hg0 will be monitored using resonant laser-induced fluorescence; ground state Hg0 is excited at 254 nm and the resulting resonant emission is detected. Given that the excitation and detection wavelengths are the same, filtering is required to remove the large background signal that is caused by elastic scattering from the probe volume. An atomic resonance filter is used that absorbs the narrow linewidth elastically scattered light and transmits a portion of the broader emission from Hg0 (see Figure 1).

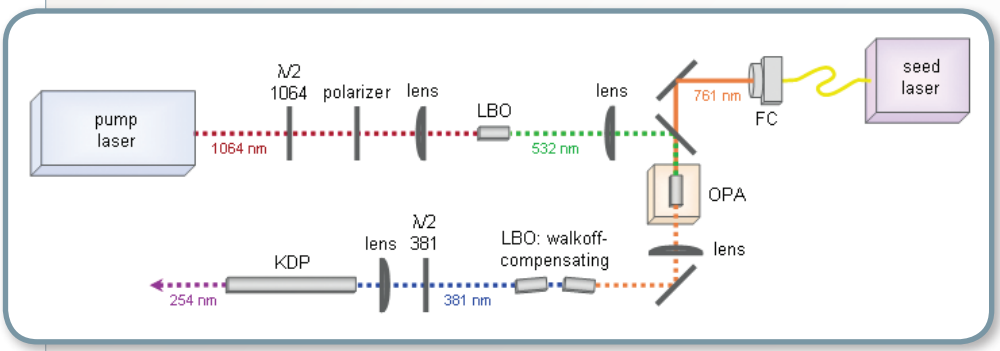

Figure 2. *Diagram of laser for Hg0 sensor showing nonlinear frequency conversion stages. FC, fiber coupler; λ/2, half-wave plate (at the designated wavelength in nm). Straight dashed and solid lines denote free-space pulsed and cw beams, respectively. The curved yellow line represents a fiber.*

Performance modeling of the detection approach indicates that a UV pulse energy of ≥ 0.1 μJ is required to meet the target sensitivity of ≤ 0.1 ppb Hg0. In addition, the laser must operate at the peak of the Hg0 absorption feature at 254 nm with a linewidth ≤ 5 GHz to allow efficient filtering of the elastic scatter. For field deployment, the laser must have the physical characteristics required for use in a coal-fired power plant: it must be compact, robust, and require no consumables other than electricity. To develop the laser, Hoops, Reichardt, and Kliner teamed with CRF researchers Roger Farrow, Ray Bambha, visiting student Paul Schulz, and Sandia Albuquerque's Randy Schmitt. The laser architecture that they are pursuing is a multistage nonlinear frequency-conversion design involving a fiber-amplifier-pumped optical parametric amplifier (OPA). Specifically, the OPA is pumped by the 532 nm frequency-doubled output of the fiber amplifier, and the seed source is a fiber-coupled, cw laser operating at 761 nm. The resultant amplified signal beam is frequency tripled to yield 254 nm light. The laser is depicted schematically in Figure 2.

Figure 3. *Photograph of laser for Hg0 sensor mounted on a 12″ × 18″ breadboard. The beam path has been added for clarity.*

The work has involved initial modeling of the laser performance using the Sandia-developed nonlinear optics package SNLO, construction of a breadboard device (see Figure 3), and comparison of the measured performance to SNLO simulations. While the Sandia-built fiber laser pump source is being constructed, a passively Q-switched, monolithic Nd:YAG microlaser developed by Schmitt is used to assess the performance of the OPA and subsequent frequency conversion stages. The output pulse energy of the microlaser is comparable to that expected from the fiber laser, but at a repetition rate several orders of magnitude below that of the fiber laser. With this laser design, 254-nm pulse energies of 1.8 μJ have been achieved with a linewidth of 1.4 GHz, exceeding the required specifications.

In the near future, the researchers will improve the temperature stability of the laser, which will increase the output wavelength stability. They will also replace the microlaser pump source with a high-repetition-rate fiber laser to increase the average power out of the laser.

Visiting student Paul Schulz returns to Germany

Visiting student Paul Schulz left the CRF in February. He will return to the University of Applied Sciences in Berlin, Germany, where he will defend his thesis for an Engineering Degree in Applied Physics (roughly between a bachelor's and master's degree in the U.S.). Paul spent one year at the CRF working with Alexandra Hoops, Roger Farrow, Tom Reichardt, and Dahv Kliner on a sensor to detect atomic mercury emitted from coal-fired power plants.

Paul was involved in development of a compact, UV laser based on a pulsed, frequency-converted optical parametric amplifier. He characterized the performance of the laser system and helped develop a wavelength calibration and control system.

CRF Researchers receive 2008 INCITE allocations

2008 supercomputing allocations were announced under DOE's Innovative and Novel Computational Impact on Theory and Experiment (INCITE) program, which supports computationally intensive, large-scale research projects. Among the new projects awarded is a project led by Jacqueline Chen (Top) and Joe Oefelein (middle) from Sandia and Ramanan Sankaran (bottom) from Oak Ridge National Laboratory on "High-Fidelity Simulations of Clean and Efficient Combustion of Alternative Fuels," awarded 18 million cpu-hours on the Cray-XT4 at the National Center for Computational Sciences at Oak Ridge National Laboratory. The project will use Direct Numerical Simulation (DNS) and Large-Eddy Simulation (LES) approaches in high-fidelity simulations to capture complex turbulence-chemistry interactions, and in particular, effects of fuel variability, interactions that are currently poorly understood even at a fundamental level. These studies will provide the foundational science necessary to develop a validated, predictive combustion modeling capability to optimize the design and operation of evolving fuels in advanced engines for transportation applications.

Stabilization (cont.)

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field of a spatially developing turbulent hydrogen/air slot jet flame in a heated coflow was performed by Chunsang Yoo and Jacqueline Chen of SNL as part of a DOE INCITE grant on the CrayXT3/XT4 at Oak Ridge National Laboratory. The DNS was performed with a detailed chemical mechanism at a jet Reynolds number of 11,000 with approximately 1 billion grid points, requiring 3.5 million cpu-hrs running for 15 days on ~10,000 processors. Given the detailed spatial and temporal resolution of the simulation, the data can uniquely provide instantaneous data and stationary statistics regarding the velocity and reactive scalar fields necessary for fundamental understanding and modeling of flame stabilization.

Figure 1 shows a 3D volume rendering of scalar dissipation rate (left) and the mass fractions of OH and HO_2 (right) from the DNS of the turbulent lifted jet flame. Fuel issues from a central jet, which consists of 65% hydrogen and 35% nitrogen by volume at 400K. The central jet is surrounded on either side by co-flowing heated air streams at 1,100K and atmospheric pressure. The fuel jet width, H, is approximately 2mm and the inlet jet velocity is 347m/s. The domain size is 12.5H × 16.7H × 3.3H and one jet time, τ_j, is approximately 0.07ms. In this study, we investigate the role of auto-ignition resulting from the heated coflow and the role of the near-field coherent jet motion in the stabilization mechanism by Lagrangian tracking of the flame base together with relevant scalar and velocity fields.

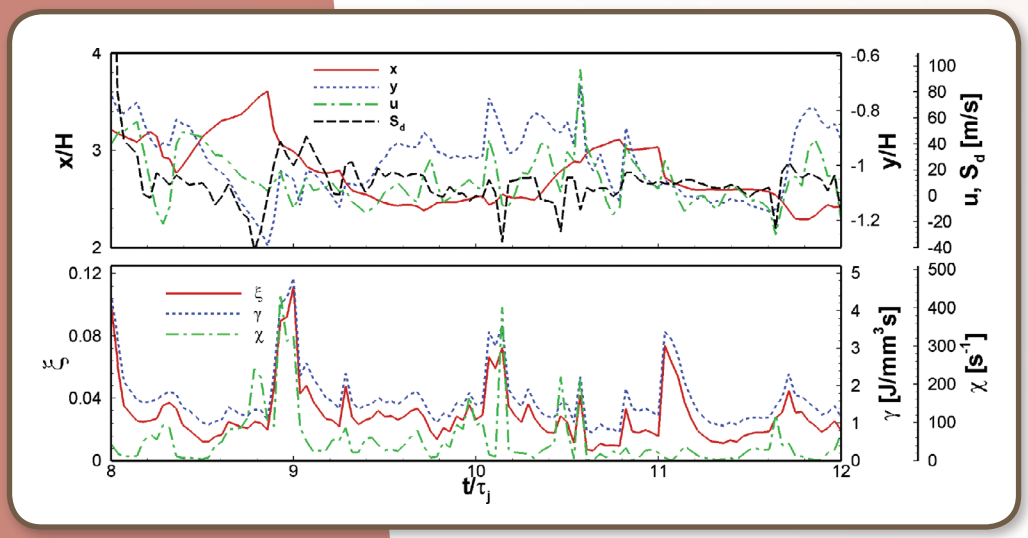

Figure 2. *DTemporal evolution of key scalar variables at the stabilization points at z = 0: axial and transverse location, axial velocity, and displacement speed (top), and mixture fraction, heat release rate and scalar dissipation rate (bottom).*

From Figure 1, it is readily observed that HO_2 radical accumulates upstream of OH and other high-temperature radicals such as H and O that are not shown here. HO_2 intermediate is an important precursor of auto-ignition in hydrogen-air chemistry such that its existence upstream of other intermediate species indicates the importance of auto-ignition in the stabilization of the lifted flame base. Several other independent measures also confirm the role of autoignition in stabilizing the flame. However, the flame base is also observed to fluctuate both spatially and temporally around a statistically stationary mean position. The flame base movement is, thus, further investigated by correlating it with velocity and key scalar variables. Figure 2 shows the temporal evolution of the flame base with axial velocity (u1), displacement speed (Sd) which represent the flame base speed relative to the local gas velocity, mixture fraction (ξ), heat release rate(γ), and scalar dissipation rate (χ) in a given spanwise plane.

During the time period t/τ_j = 8.8 ~ 9.6, the stabilization point moves upstream and transversely inward. The corresponding mixture fraction is fuel-lean and the corresponding χ is relatively small. Therefore, the flame base is situated far from the core jet and the local conditions are favorable for autoignition. Consequently, the corresponding Sd is greater than the laminar flame speed by an order of magnitude and also greater than the local axial velocity. Hence, the stabilization point moves upstream and transversely inward following a large jet structure. By subsequently migrating radially inward towards relatively fuel-rich regions, the stabilization point encounters a jet structure with

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high axial and transverse velocities and high χ. Under these conditions, auto-ignition is easily retarded and the stabilization point has low and, in some instances, even negative Sd. This results in a rapid movement of the stabilization point downstream and transversely outward. Subsequently, the stabilization point returns to its original point and forms a complete cycle. From these observations, we postulate a stabilization mechanism such that the flame base fluctuates with the passage of a series of large scale organized jet motion and auto-ignition in fuel-lean mixtures as depicted in Figure 3.

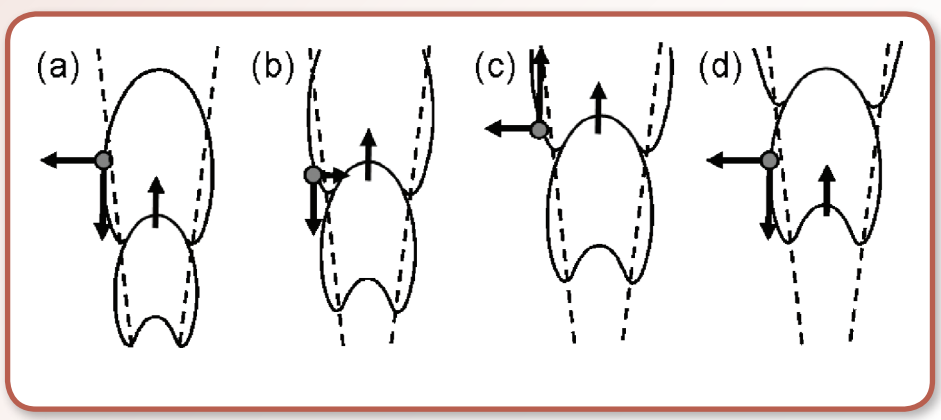

Figure 3. *Schematic of the flame base movement (adopted from Su et al. 2006, Combustion and Flame 144 (2006)): (a) ignition occurs in lean mixtures with low χ, (b) the stabilization point propagates following coherent jet structure, (c) local extinction occurs by high χ and the stabilization point moves downstream with high axial velocity, and (d) ignition occurs in another coherent jet structure. Dashed line represents auto-ignition limit due to low temperature.*

In summary, detailed investigation of the DNS data revealed that auto-ignition occurring in hot, fuel-lean mixtures is the key mechanism responsible for flame base stabilization. It is also found that the motion of the lifted flame base is cyclic with the passage of organized jet motion, and the flame stabilization is determined by the balance between the local axial velocity and auto-ignition which favors hot environments with low scalar dissipation rate.

In addition to the fundamental insights obtained, SNL researchers Ed Richardson, Chunsang Yoo and Jackie Chen have been using the DNS data to study and validate advanced engineering models, such as the Conditional Moment Closure (CMC), in the auto-ignitive regime. The timing and location of auto-ignition can be highly sensitive to turbulent fluctuations of composition. Second-order CMC provides transport equations for conditional fluctuations in turbulent reacting flows. CMC equations accounting for compressibility and differential diffusion were analyzed using the DNS data. The researchers found that second-order moments are required to accurately model the conditional reaction rates; however, it was demonstrated that the majority of the second-order reaction rate component was obtainable with a small subset of the species-temperature covariances. The balance of the second-order CMC equation reveals that transport due to turbulent flux between fluid with differing histories is responsible for the initial generation of conditional scalar variances. These variances give rise to the range of ignition delays observed, which in turn modulate the spatial location of the flame base.

In the future, the researchers are planning to investigate the effect of fuel ignition chemistry (relevant to bio-diesel and renewable fuels) and coflow temperature on the stabilization of lifted jet flames at ambient and elevated pressure.

Improving Laser Diagnostics **Picosecond lasers (cont.)**

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Laser-induced-fluorescence is a promising diagnostic technique for quantitative imaging of atomic hydrogen in flames. Over the last 3 decades, several multi-photon excitation schemes have been used to excite fluorescence in hydrogen. The most straightforward approach employs a single laser to excite 656-nm fluorescence via two-photon-resonant absorption at 205 nm. To produce sufficient excitation, this approach requires high-intensity laser pulses, which can also photodissociate other flame constituents, most notably H_2O and CH_3. The photolytically produced hydrogen atoms can also be excited by the same laser pulse, producing additional fluorescence that interferes with the measurement of the naturally occurring atomic hydrogen in the flame. This process is well documented in the literature and represents significant limitation for quantitative imaging in many systems.

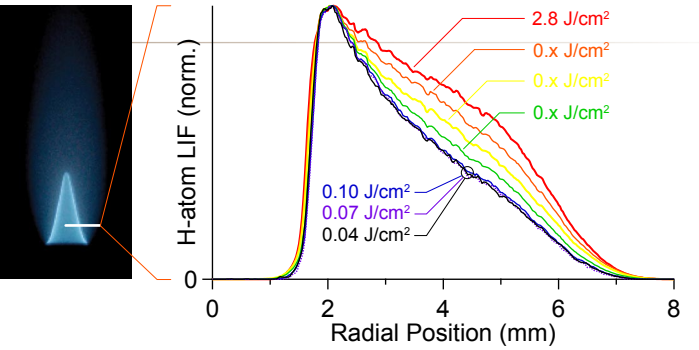

Figure 1. *Radial profiles of H-atom LIF in a slightly rich (Φ=1.14) premixed $CH_4/O_2/N_2$ flame at several ns-laser fluences.*

Figure 1 demonstrates the significance of photolytic interference to one-dimensional line images produced in a laminar premixed $CH_4/O_2/N_2$ flame. In the figure, normalized LIF signals are shown as function of radial position in the flame for a range of laser fluences. The profiles produced with fluences of 0.12 J/cm² or less represent "interference-free" measurements because the photolytic interference is not detectable and the shape of the LIF profile does not depend on the laser fluence. At higher fluences, however, distortion in the radial profile between 3 and 6 mm is clearly evident. This increase in the relative signal is consistent with the photolytic production of atomic hydrogen that results from photodissociation of H_2O in the flame products.

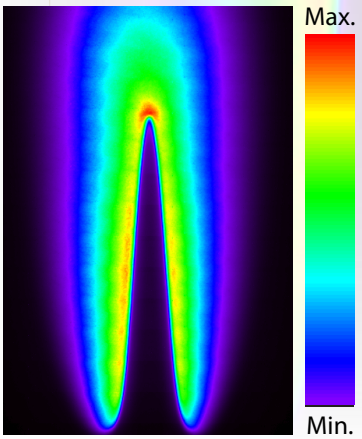

Figure 2. *Interference-free composite H-atom LIF image produced with ps excitation in a Φ=1.2 premixed $CH_4/O_2/N_2$ flame. Enhanced signal is evident at the highly strained tip of the flame.*

Generally, previous H-atom LIF measurements used nanosecond (ns) pulsed lasers for excitation. CRF researchers, Waruna Kulatilaka, Tom Settersten, Brian Patterson, and Jonathan Frank, investigated the potential for reducing the impact of photolytic interference by exciting H-atom fluorescence with a custom-built picosecond laser that has a pulse duration that is two orders of magnitude shorter than conventional ns pulsed lasers. For the flame pictured in Figure 1, ps excitation produced interference-free signals that were approximately 40x larger than those produced with the conventional ns pulsed laser. Similar results in other CH_4 and H_2/O_2 flames also demonstrated approximately an order of magnitude improvement in the achievable signal levels by using the ps laser.

The researchers exploited ps excitation to produce high-quality, interference-free, two-dimensional images of H-atoms in methane flames by forming the laser beam into a 2.5-mm sheet. Figure 2 presents a composite H-atom LIF image in a premixed $CH_4/O_2/N_2$ flame that was formed by compiling multiple images at different axial positions in the flame. The notable increase in signal level near the strained tip of the flame is consistent with diffusional focusing of H. The demonstrated ability to observe this effect is stimulating a new collaboration between Settersten and Frank to use LIF imaging of H to study the effects of preferential diffusion on ignition and flame propagation.

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