Sandia National Laboratories

Combustion Research Facility

H2FC Hydrogen and Fuel Cells Program

How to see and quantify hydrogen concentration (and cryogenic hydrogen) using optical diagnostics

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PreSLHy kick-off meeting

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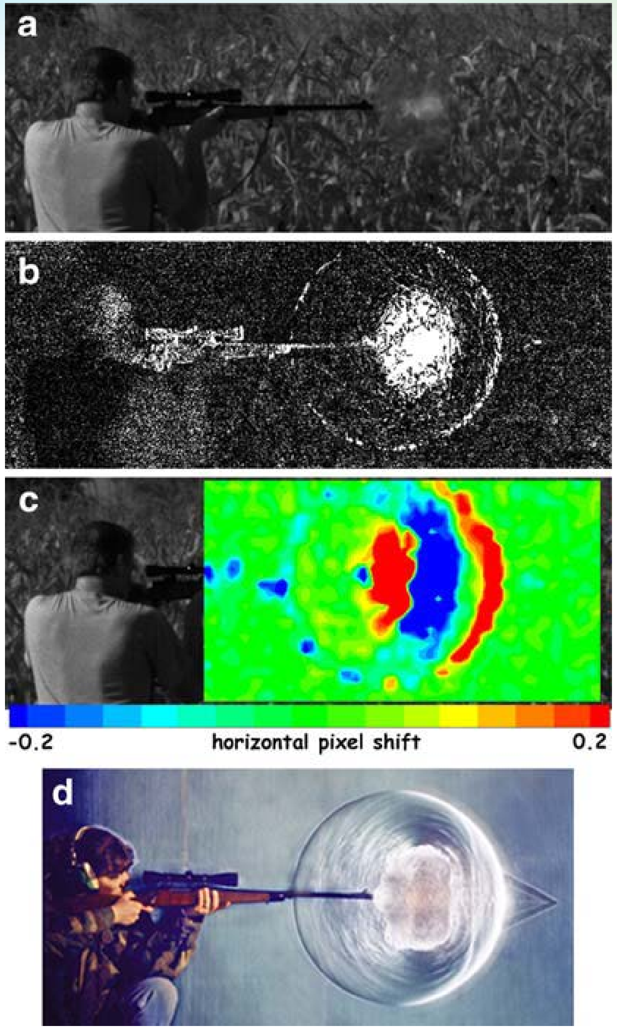
Common optical techniques to visualize gas flows

Technique	Principle
Shadowgraphy	Refractive index gradients bend light rays as they pass through density variations.
Schlieren	Same as shadowgraphy. Knife edge enables focused image to form rather than simply shadow.
Fluorescence	Photons are absorbed by molecules at a resonant transition and light is reemitted at a shifted wavelength
Absorption	Gases have absorption features for certain wavelengths of light.
Rayleigh scattering	Elastic scattering off of different molecules is proportional to their cross-sections and number density.
Raman scattering	Inelastic scattering off of different molecules gives each component a spectral fingerprint proportional to cross-section and number density.

Schlieren imaging

- Measures gradients in density (1st derivative)
- For quantitative measurements:
  - Calibrated schlieren – uniform light source, light intensity quantifies refraction angles
  - Rainbow schlieren – color cutoff filter in place of knife edge, color quantifies refraction angles
  - Diverging light background oriented schlieren (BOS) – pixel offset from original position determines refraction angle
- BOS (using sunlight) possible for H_2, however:
  - Need semi-ordered background
  - Density gradients caused by both temperature and composition
  - Line-integrated, total refraction measured, extremely complex to quantify, even with tomography
  - No symmetries for an open plume



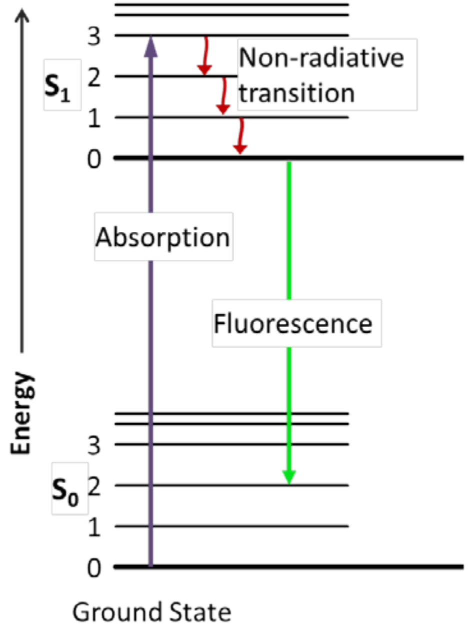
BOS

Conventional schlieren →

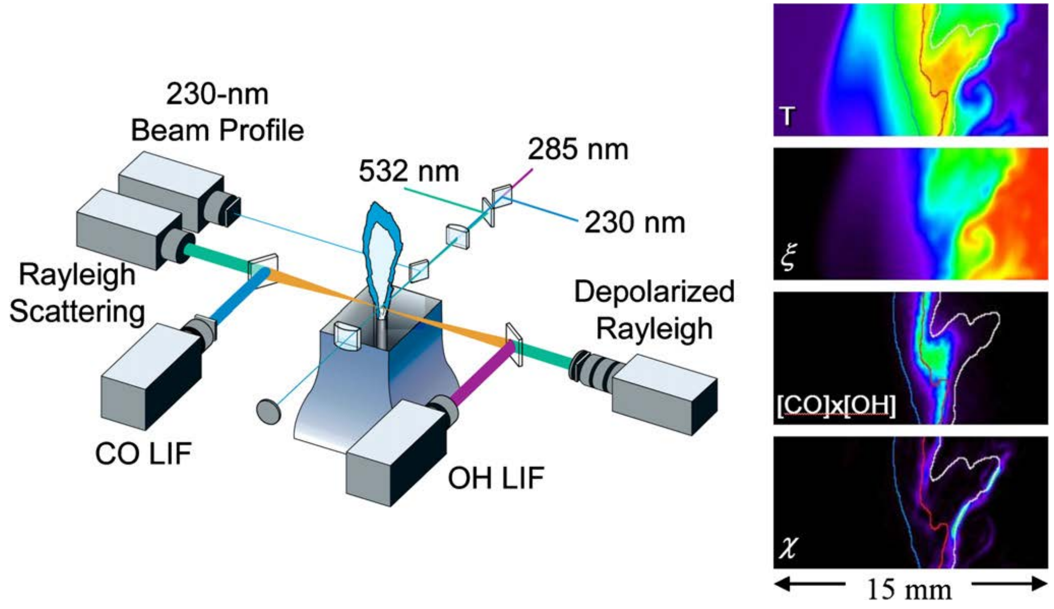
Images from Hargather & Settles, Exp. In Fluids, 2010

Fluorescence

- OH fluorescence possible, but only for flames, not unignited H_2
- Unignited concentration measurement would require seeding hydrogen with fluorescent tracer material (aliphatic ketones like acetone or 3-pentanone often used)
  - For cryogenic H_2, no gaseous or liquid options at LH_2 temperatures
  - Very challenging to get solid particles dispersed in liquid, and get them to follow gas flow during phase change



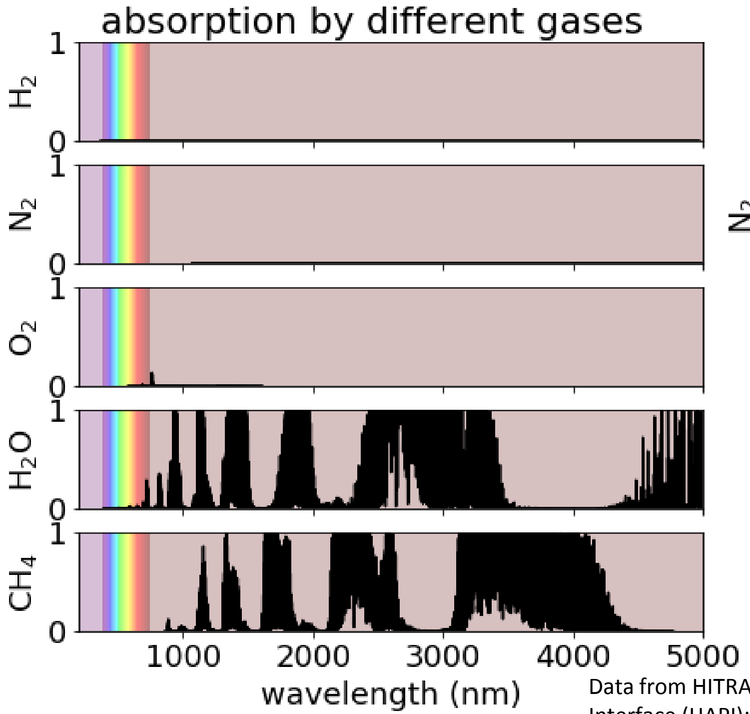
By Jacobkhed - Own work, CC0, https://commons.wikimedia.org/w/index.php?curid=19180813

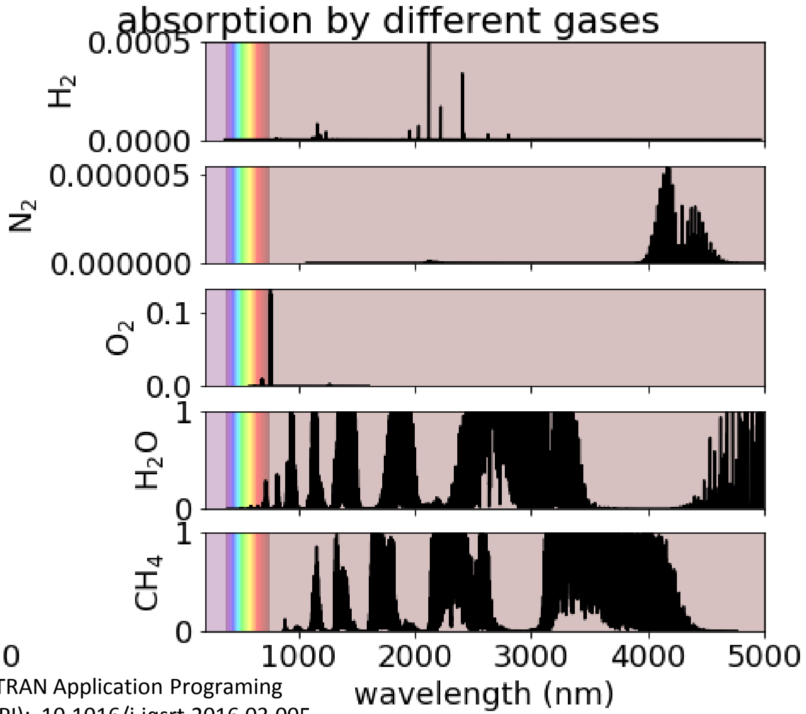


Frank, Kaiser, Long, Combust. Flame, 143 (2005) 507-523

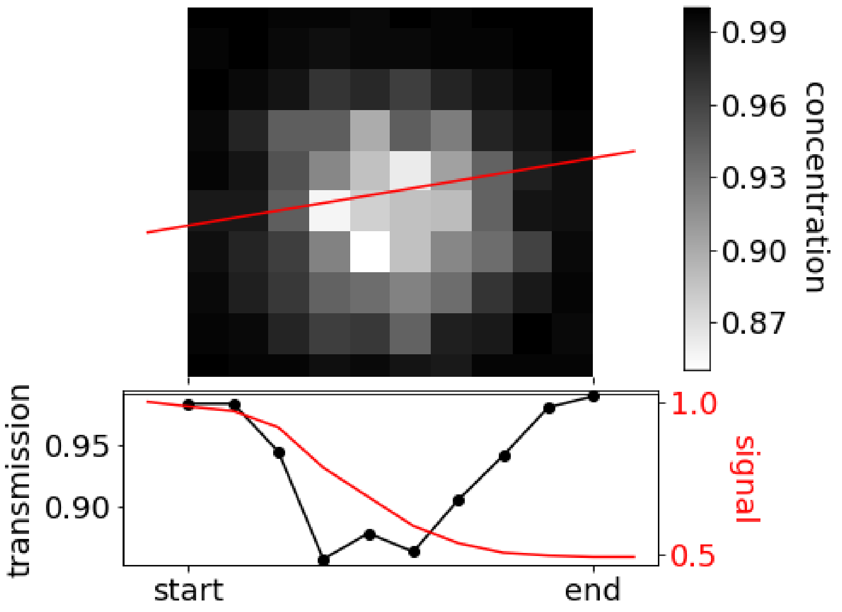
Absorption

- H_2 lacks strong absorption features (unlike CH_4)
- Would require illumination and light collection on opposite sides of plume (or mirror to reflect light)
- Line-integrated absorption, to quantify, requires multiple angles, tomography





Data from HITRAN Application Programing Interface (HAPI): 10.1016/j.jqsrt.2016.03.005



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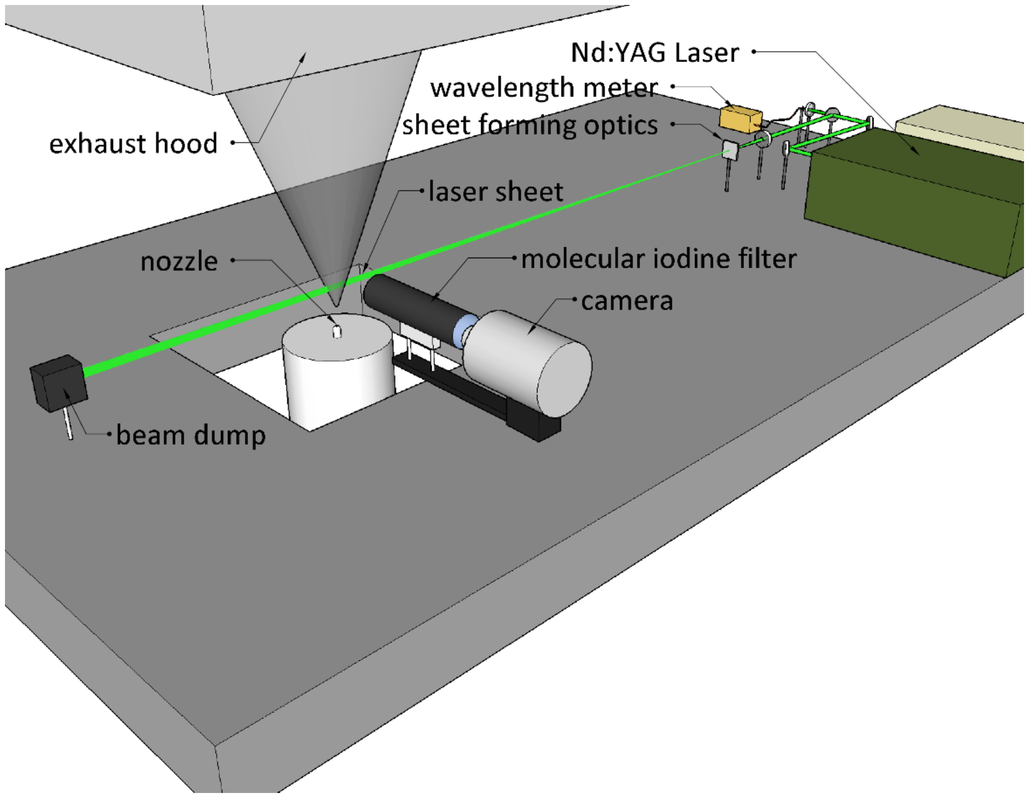
Rayleigh scattering

H_2 Rayleigh cross-section ≈ 10^{-27} cm^2

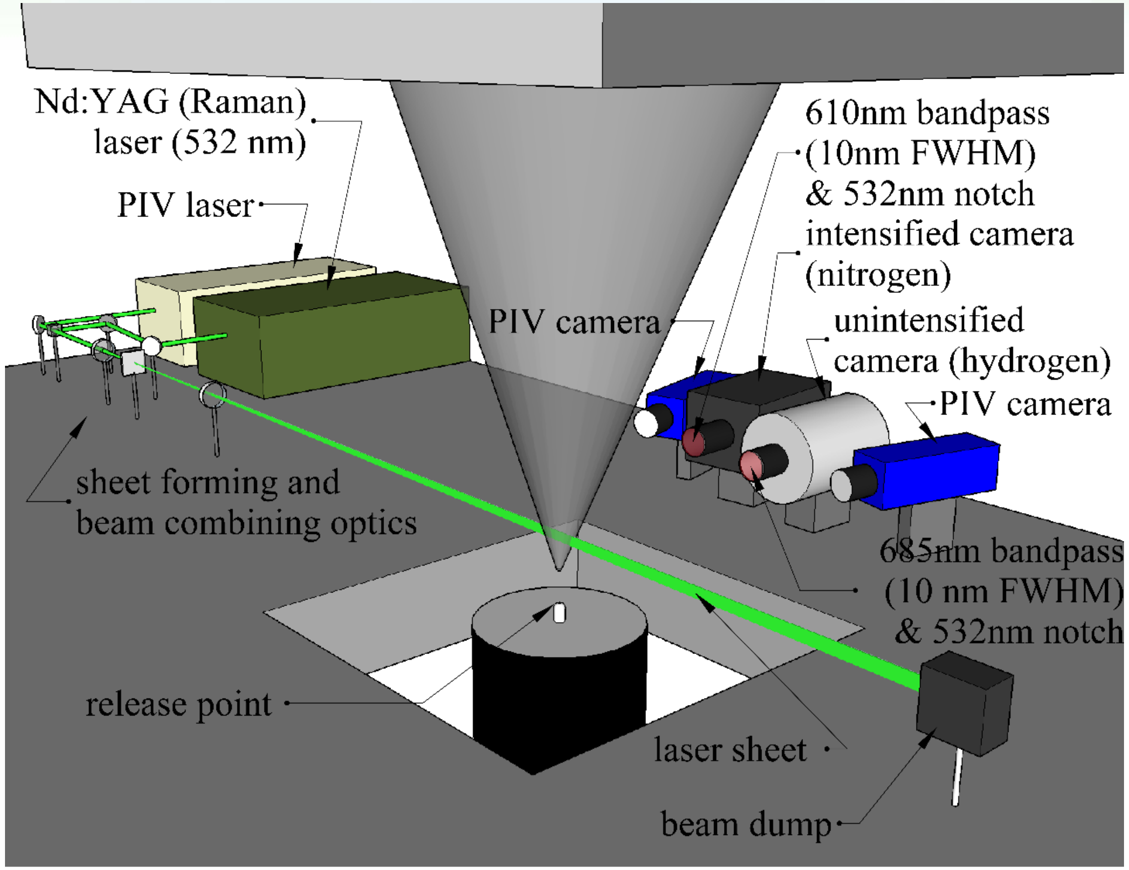
- Planar laser Rayleigh scattering used at Sandia for atmospheric temperature hydrogen releases
- Scatter proportional to number density; variations are caused by both composition and temperature
- For warm releases, always measured in atmospheric temperature region to eliminate this variable and enable composition quantification
- Not feasible to wait until cryogenic plume has warmed back to atmospheric temperature
- Rayleigh imaging will have signal overwhelmed by Mie scattering off of condensed entrained moisture in cryogenic plume
- Filtered Rayleigh has insufficient Mie scattering (condensed, entrained moisture) light suppression (OD≈3)



Planar Raman imaging works in a lab setting

H_2 Raman cross-section ≈ 10^{-30} cm^2

- Signals are low
  - High powered light source required (~700 mJ/pulse @ 532nm, 12mm tall sheet)
  - Fast optics for collection (F/1.2)
- Large Raman shift enables higher optical density filters to remove unwanted Mie scatter
  - 10 nm FWHM bandpass filters at wavelengths of interest
  - OD of 12 @ all wavelengths
  - OD of 18 @ 532 nm
- Signals for other Raman lines (rotational, etc.) low at cryogenic temperatures

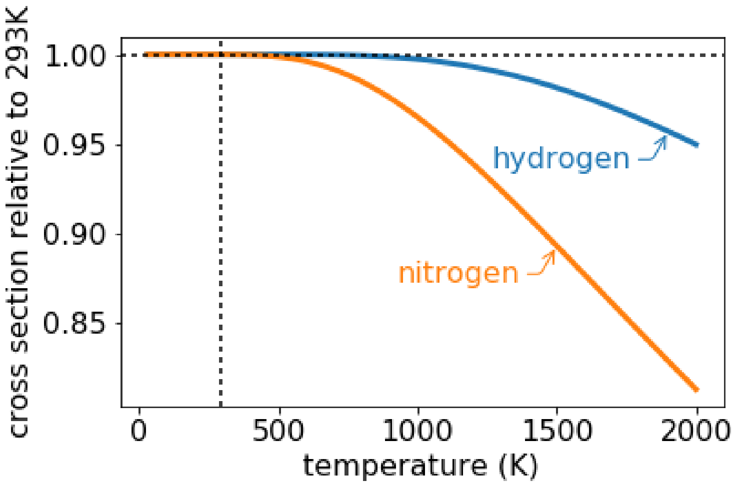


H_2: shift of 4161 cm^{-1} (532nm → 683 nm, 355nm → 416 nm)

N_2: shift of 2331 cm^{-1} (532nm → 607 nm, 355nm → 387 nm)

Quantification of Raman signals

- Signal is proportional to number density of molecules
- We use the ideal gas law to relate temperature and mole fraction to number density
  - $\frac{n_{total}\Sigma x}{V} = \frac{P_{total}\Sigma x}{RT}$
  - other equation of state could be used but may not have analytical solution
- Cross-section dependence matters for high-T (flames), but not low-T (cryogenic)



Eq. 1: $\frac{I_{H_2}}{I_0} = k_{H_2}\frac{x_{H_2}}{T}$ ← unknown 1 ← unknown 2

measured values

calibration constants

Eq. 2: $\frac{I_{N_2}}{I_0} = k_{N_2}\frac{x_{N_2}}{T}$ ← unknown 3

based on the composition of air

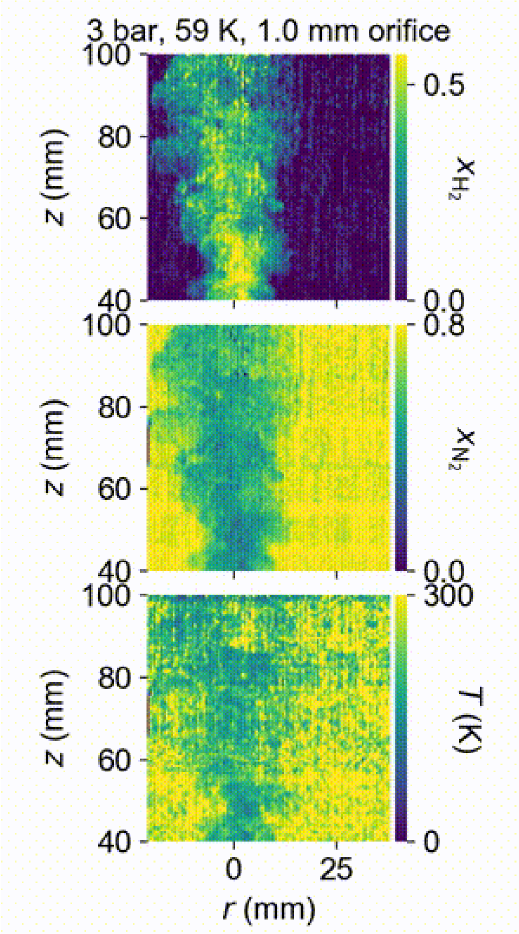
Eq. 3: $1 = x_{H_2} + 1.28x_{N_2}$

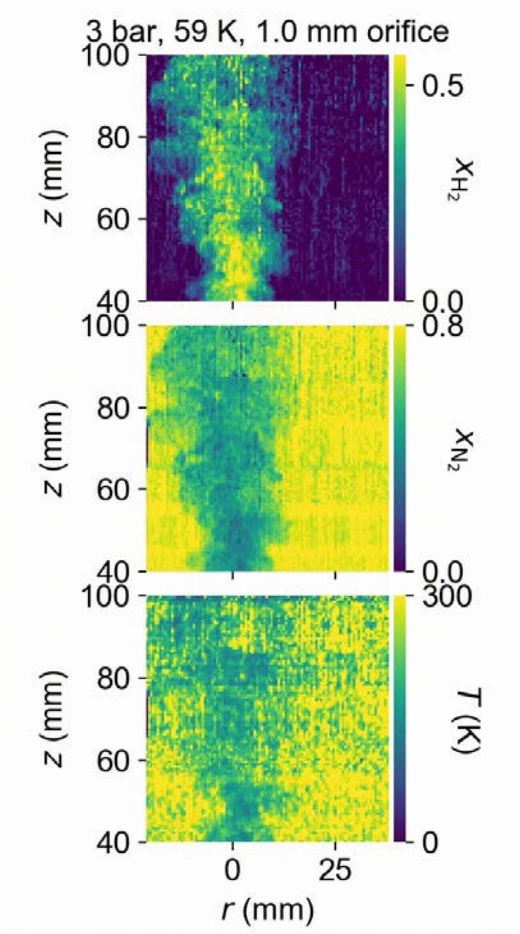
$$\begin{cases} x_{H_2} = \dfrac{\mathrm{I_{H_2}}}{k_{H_2}\left(\dfrac{\mathrm{I_{H_2}}}{k_{H_2}} + \dfrac{1.28\mathrm{I_{N_2}}}{k_{N_2}}\right)} \\ x_{N_2} = \dfrac{\mathrm{I_{N_2}}/\mathrm{I_0}}{k_{N_2}\left(\dfrac{\mathrm{I_{H_2}}}{k_{H_2}} + \dfrac{1.28\mathrm{I_{N_2}}}{k_{N_2}}\right)} \\ T = \dfrac{1}{\dfrac{\mathrm{I_{H_2}}}{k_{H_2}} + \dfrac{1.28\mathrm{I_{N_2}}}{k_{N_2}}} \end{cases}$$

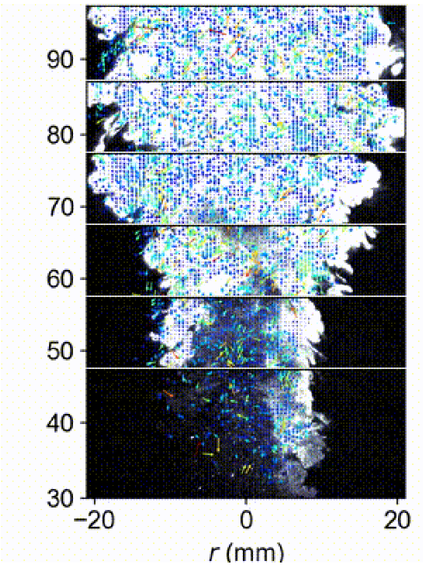
Raman has been used in a lab-scale campaign to measure releases from ≈1 mm orifices

T_{noz} [K]	P_{noz} [bar_{abs}]	d [mm]	T_{throat} [K]	n_{hts}
58	2	1	43.5	4
56	3	1	41.9	4
53	4	1	39.6	4
50	5	1	37.4	5
61	2	1.25	45.7	6
51	2.5	1.25	38.2	2
51	3	1.25	38.2	6
55	3.5	1.25	41.2	3
54	4	1.25	40.4	2
43	4	1	32.1	2
59	3	1	44.2	6
56	3.5	1	41.9	1
80	3	1	60.3	5

With PIV

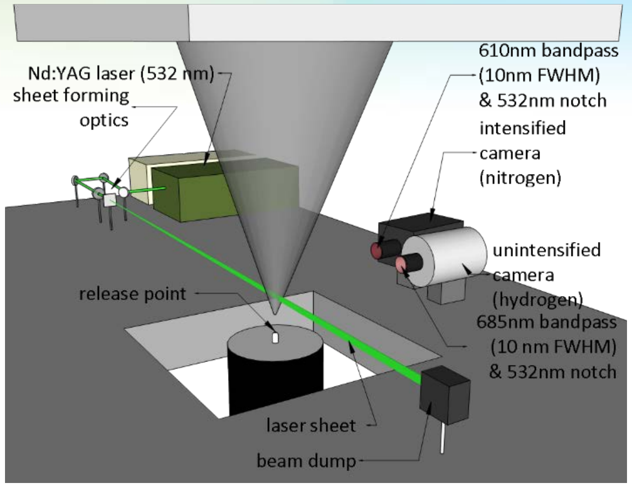


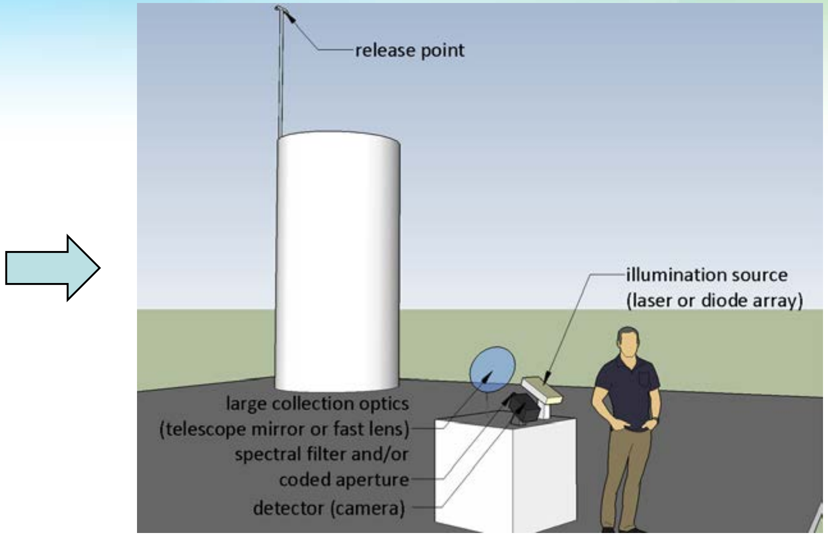


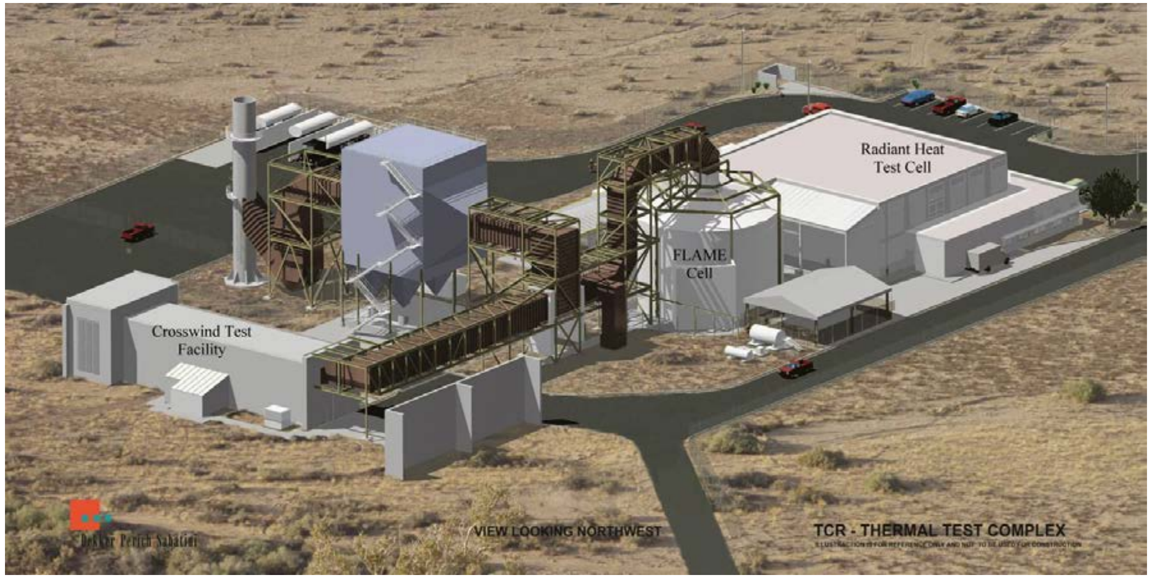


We are currently working to scale-up our Raman imaging techniques

- Enable characterization and modeling of
  - Interactions with ambient (i.e. wind)
  - Pooling
  - Evaporation from LH_2 pools
- **Currently developing an imaging diagnostic** for outdoor and large-scale experiments
  - Quantitative concentration measurements
  - 2- or 3-dimensions
  - Video frame rates
  - Portable
- Will apply diagnostic to normally occurring outdoor releases (e.g., venting after LH_2 fill)
- Dedicated validation experiments at well-controlled facilities next fiscal year (FY19)
- Desire to collaborate with PreSLHy researchers, applying diagnostic to other experiments







Summary of optical techniques

- Schlieren – cannot distinguish between temperature and concentration caused density variations (not quantitative)
- Fluorescence – no fluorescing species in the flow or species that could be seeded into the flow
- Absorption – no strong absorption features, and complex detector/illumination scheme
- Rayleigh – cannot distinguish between temperature and concentration caused density differences, entrained moisture scatters too much light
- Raman – shown to work in a laboratory setting, enables quantification of temperature and composition in multiple dimensions

> ➢ Sandia is developing a portable Raman imaging setup for larger releases that can be applied to a range of experiments

Acknowledgements

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  - Previous researchers including: Pratikash Panda, Katrina Groth, Isaac Ekoto, Adam Ruggles, Bob Schefer, Bill Houf, Greg Evans, Bill Winters

Currently identifying technical hardware solutions to enable the large-scale diagnostic

- Challenge: need large field of view and large aperture to collect small number of photons emitted
  - Reflective optics (large telescope mirror)
  - Refractive optics (Fresnel lens)
- Challenge: reasonable cost illumination system with high-power, low-wavelength, pulsed system
  - High-power laser with volumetric illumination
  - High-repetition rate laser scanned across the area quickly
  - High-power diodes/diode arrays
- Challenge: Effective background light suppression (both sunlight and reflected illumination light from condensed water vapor)
  - Time gating
  - Spectral gating
- Challenge: Improved temporal, spectral, and/or spatial resolution
  - Coded aperture sensing
  - Tomography