

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

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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. |

Schlieren imaging

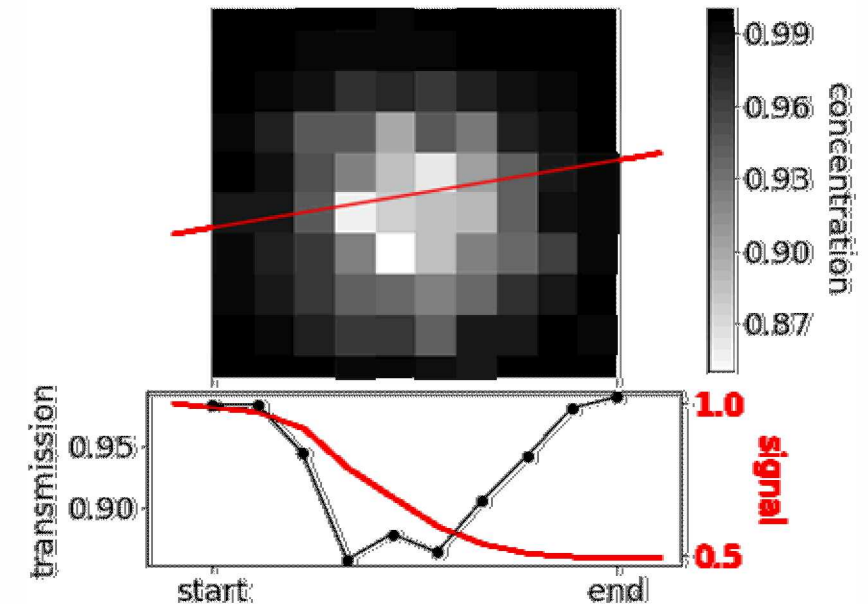
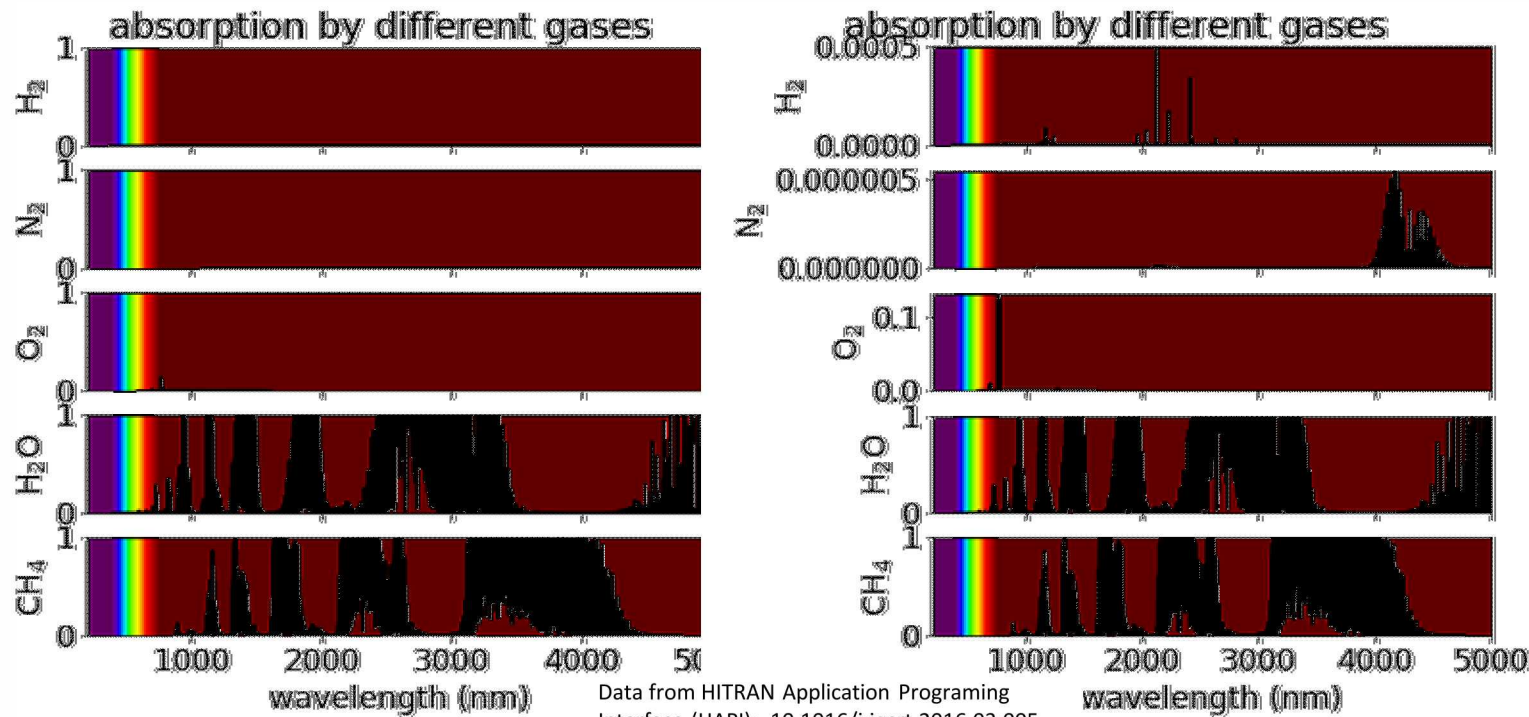
- Measures gradients in density (1st derivative)
- For quantitative measurements:
 - Calibrated schlieren – uniform light source, light intensity quantifies refraction angles
 - Rainbow schlieren – color cutoff filter in place of knife edge, color quantifies refraction angles
 - Diverging light background oriented schlieren (BOS) – pixel offset from original position determines refraction angle
- BOS (using sunlight) possible for H₂, 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

Fluorescence

- OH fluorescence possible, but only for flames, not unignited H₂
- Unignited concentration measurement would require seeding hydrogen with fluorescent tracer material (aliphatic ketones like acetone or 3-pentanone often used)
 - For cryogenic H₂, no gaseous or liquid options at LH₂ temperatures
 - Very challenging to get solid particles dispersed in liquid, and get them to follow gas flow during phase change

Absorption

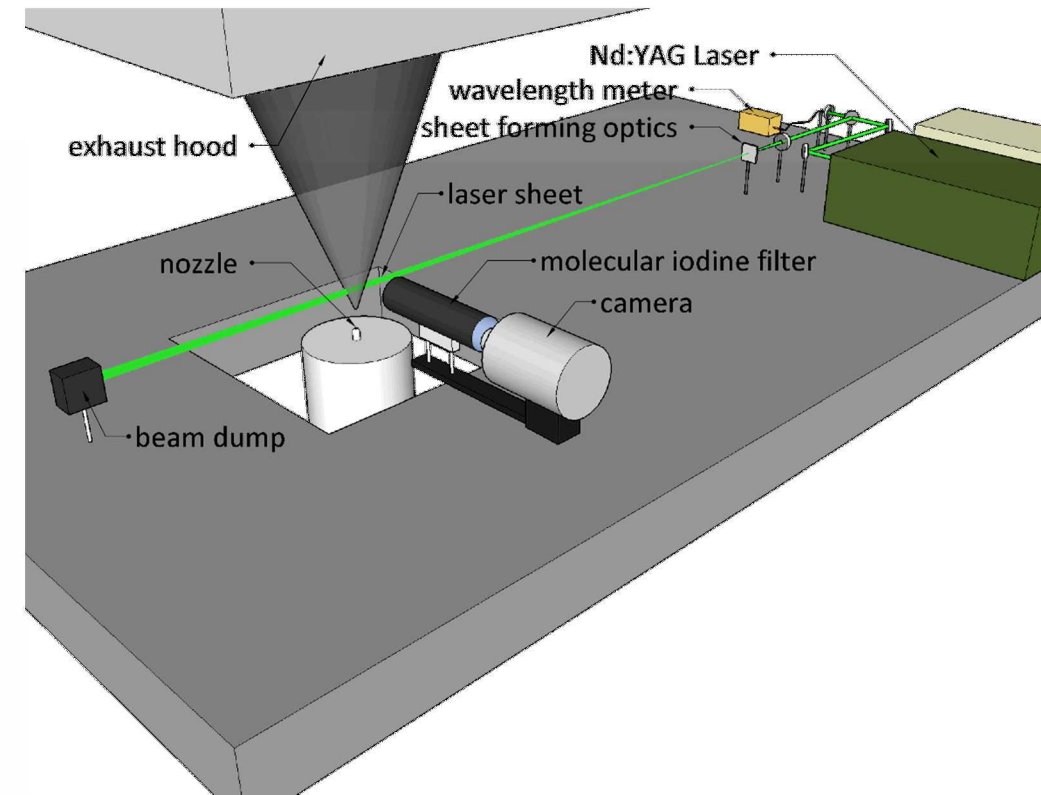
- H₂ lacks strong absorption features (unlike CH₄)
- Would require illumination and light collection on opposite sides of plume (or mirror to reflect light)
- Line-integrated absorption, to quantify, requires multiple angles, tomography



Rayleigh scattering

H₂ Rayleigh cross-section $\approx 10^{-27}$ cm²

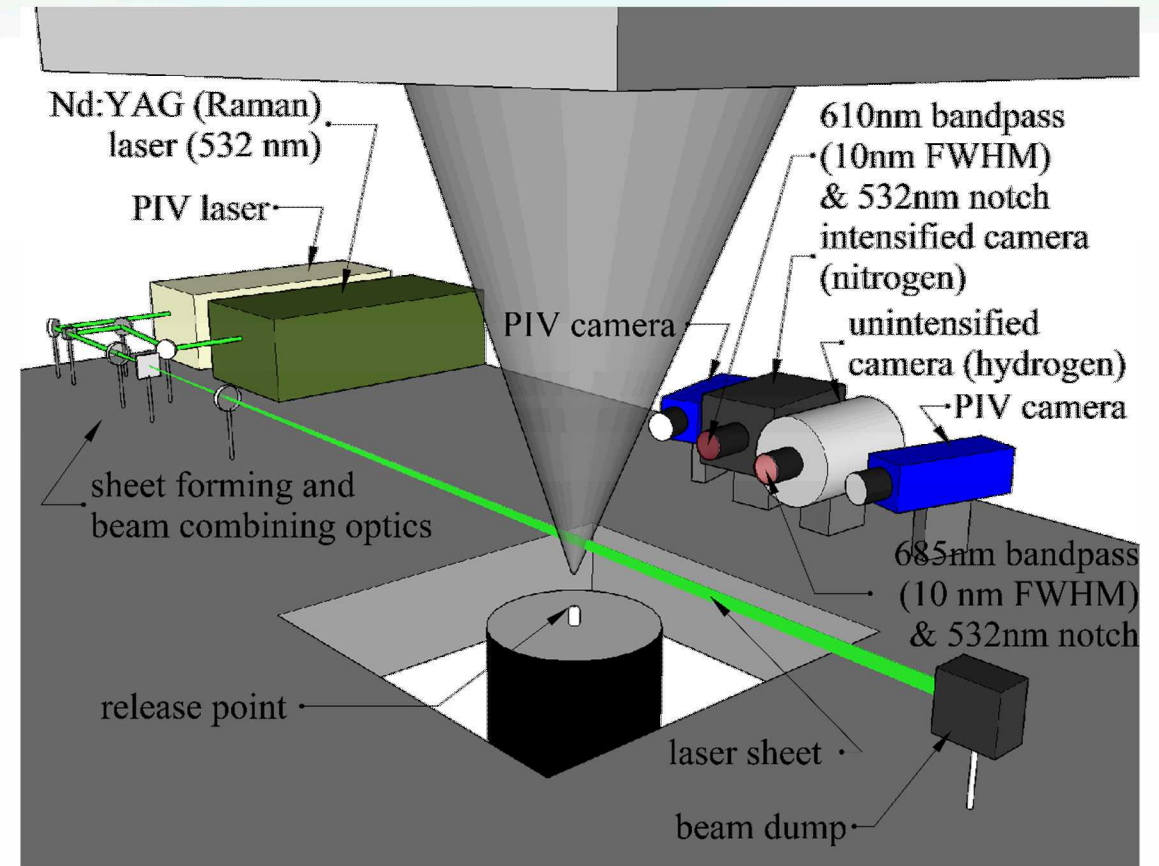
- Planar laser Rayleigh scattering used at Sandia for atmospheric temperature hydrogen releases
- Scatter proportional to number density; variations are caused by both composition and temperature
- For warm releases, always measured in atmospheric temperature region to eliminate this variable and enable 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 \approx 3)



Planar Raman imaging works in a lab setting

H₂ Raman cross-section $\approx 10^{-30}$ cm²

- 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

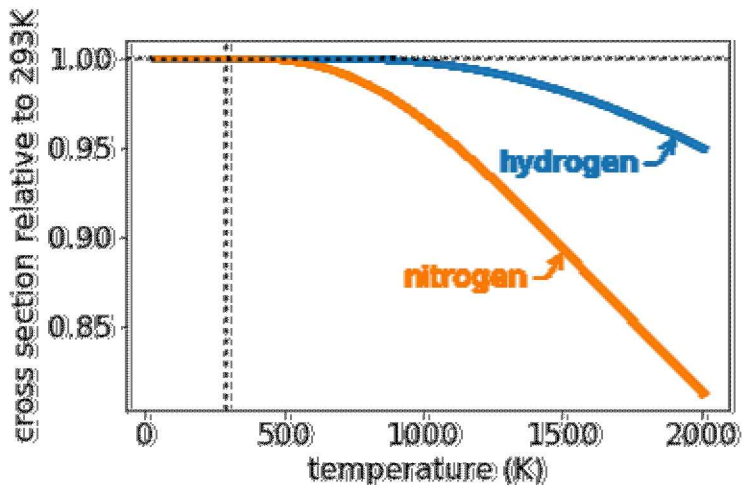


H₂: shift of 4161 cm⁻¹ (532nm → 683 nm, 355nm → 416 nm)

N₂: 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 $\left\{ \begin{array}{l} \frac{I_{H_2}}{I_0} \\ \frac{I_{N_2}}{I_0} \end{array} \right.$ 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

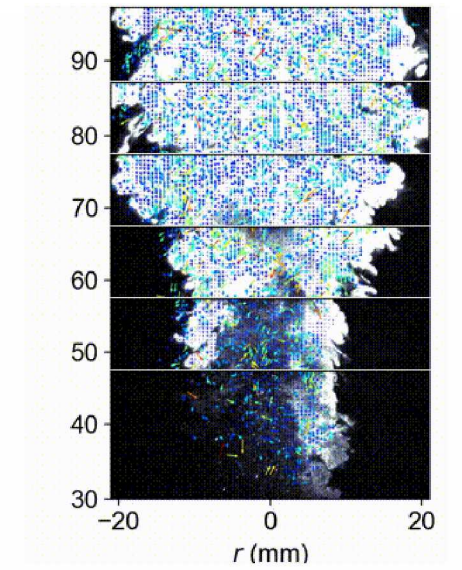
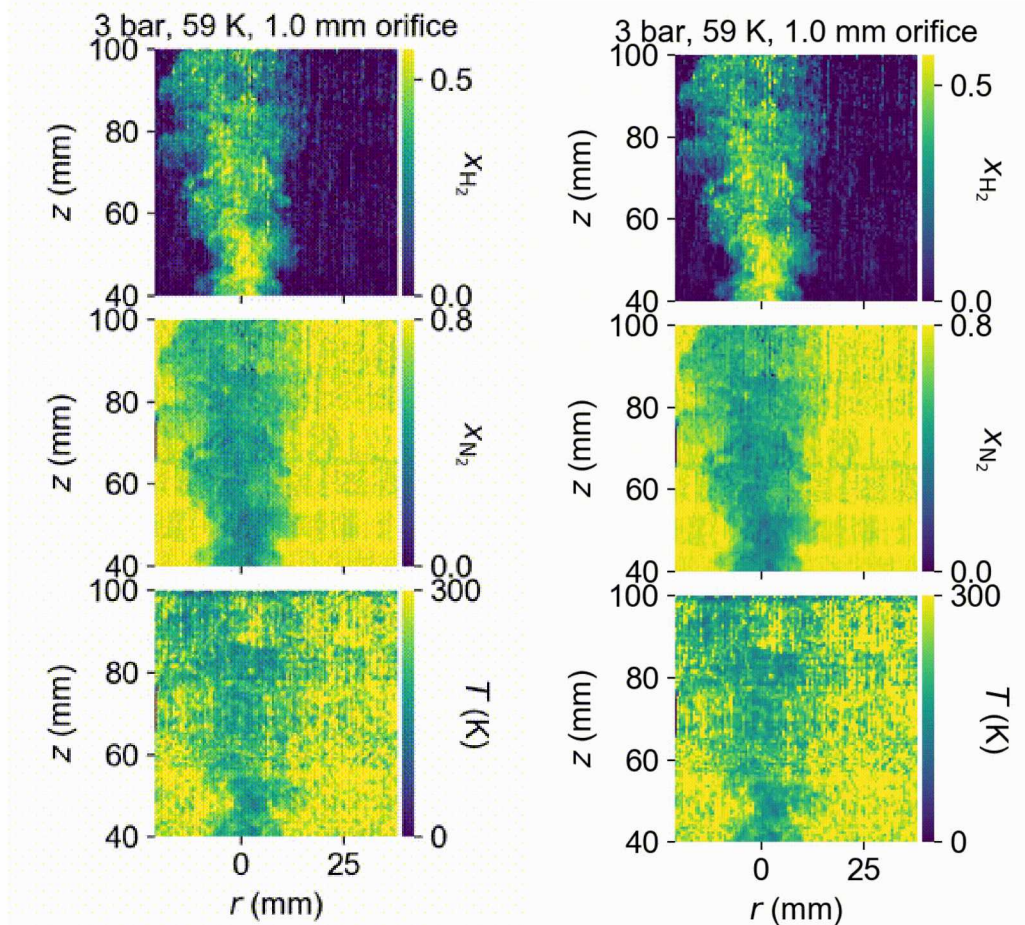
Eq. 3: $1 = x_{H_2} + 1.28x_{N_2}$

$$\left\{ \begin{array}{l} x_{H_2} = \frac{I_{H_2}}{k_{H_2} \left(\frac{I_{H_2}}{k_{H_2}} + \frac{1.28I_{N_2}}{k_{N_2}} \right)} \\ x_{N_2} = \frac{I_{N_2}/I_0}{k_{N_2} \left(\frac{I_{H_2}}{k_{H_2}} + \frac{1.28I_{N_2}}{k_{N_2}} \right)} \\ T = \frac{1}{\frac{I_{H_2}}{k_{H_2}} + \frac{1.28I_{N_2}}{k_{N_2}}} \end{array} \right.$$

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

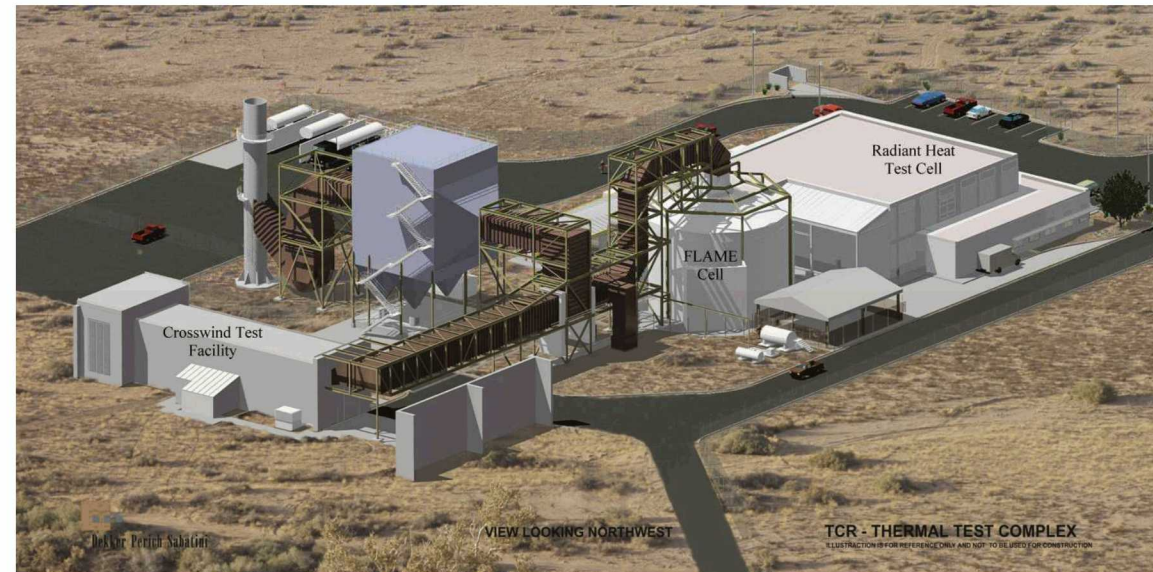
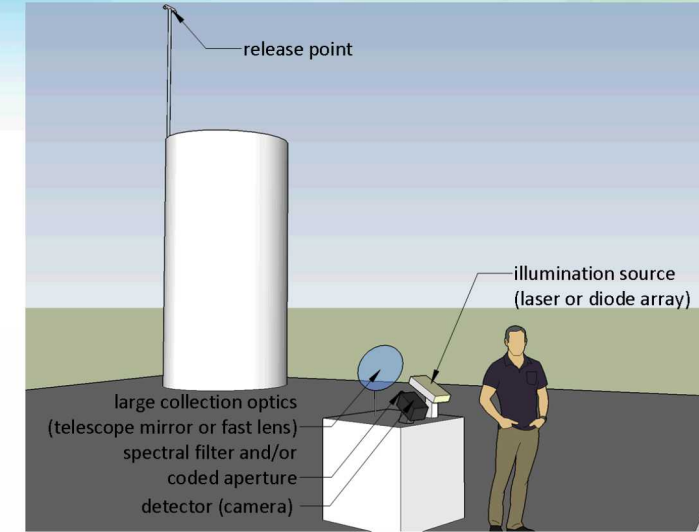
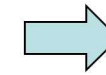
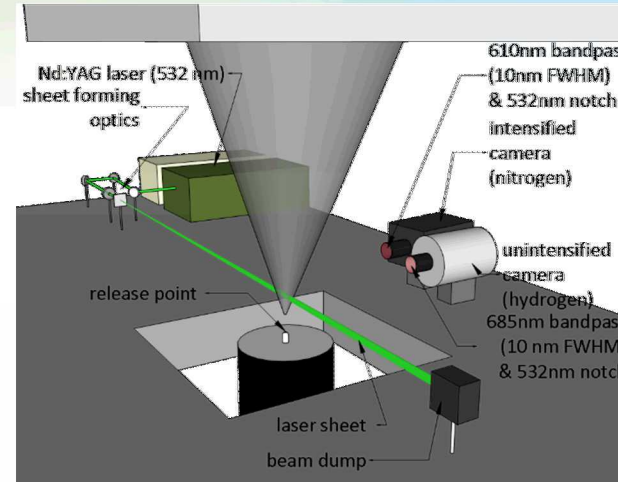
| T_{noz} [K] | P_{noz} [bar _{abs}] | d [mm] | T_{throat} [K] | n_{hts} |
|-------------------------|---|-------------|----------------------------|------------------|
| 58 | 2 | 1 | 43.5 | 4 |
| 56 | 3 | 1 | 41.9 | 4 |
| 53 | 4 | 1 | 39.6 | 4 |
| 50 | 5 | 1 | 37.4 | 5 |
| 61 | 2 | 1.25 | 45.7 | 6 |
| 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 LH₂ 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 LH₂ fill)
- Dedicated validation experiments at well-controlled facilities next fiscal year (FY19)
- Desire to collaborate with PreSLHy researchers, applying diagnostic to other experiments



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