Cryogenic hydrogen flow measurements

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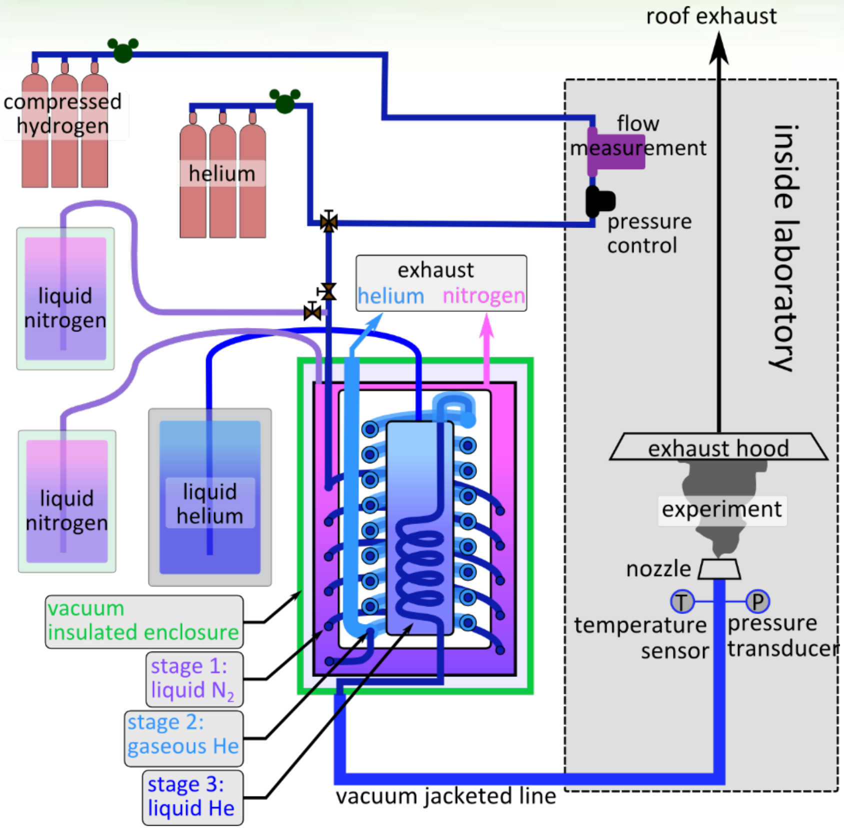
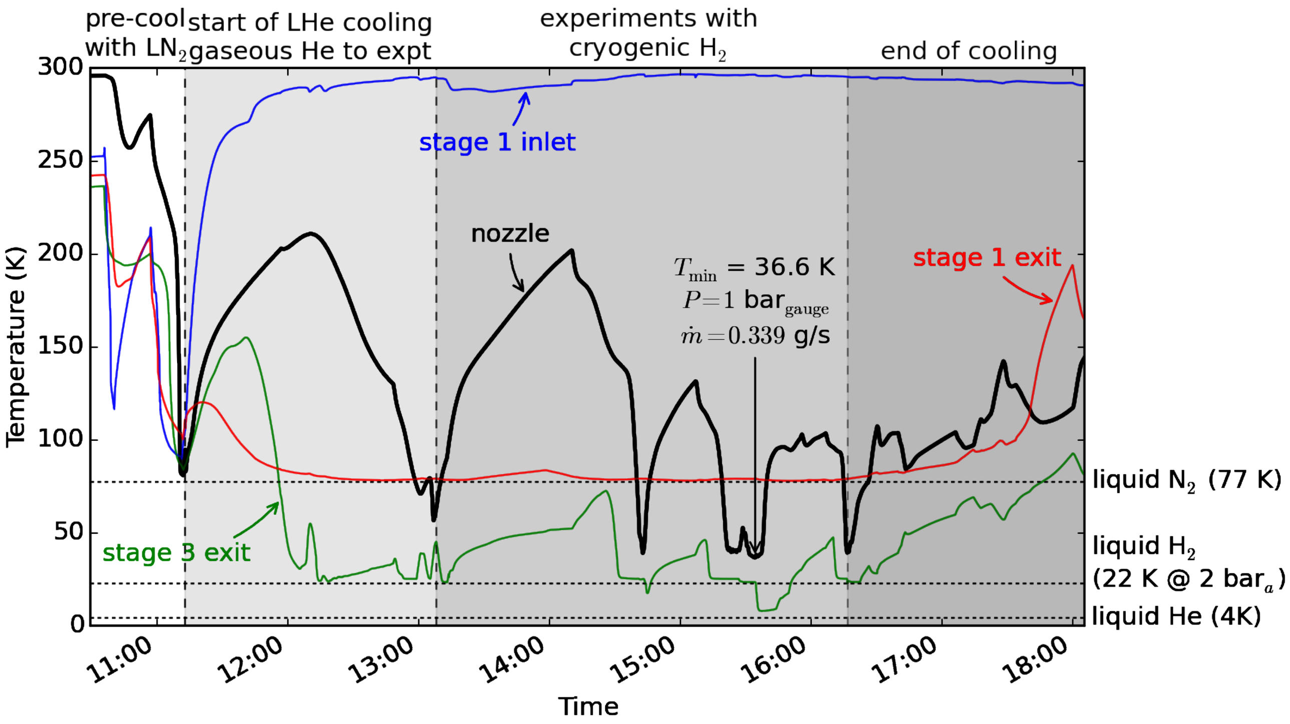
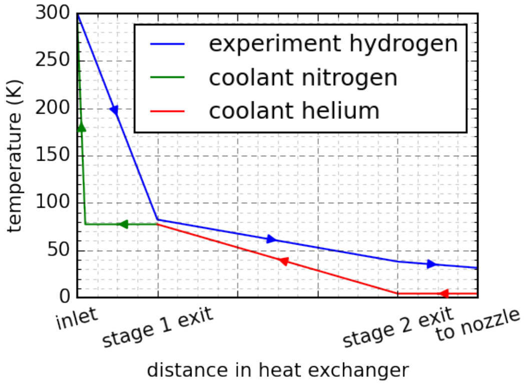
Our laboratory experiment uses a heat exchanger to liquefy hydrogen



- Gaseous hydrogen is liquefied using liquid nitrogen and liquid helium
- Flow rate is measured as a gas using a thermal mass flow meter
- Nozzle pressure is controlled upstream of heat exchanger
- Silicon diode temperature sensors

Silicon diode temperature sensors in the heat exchanger and near the nozzle give accurate measurements of temperature





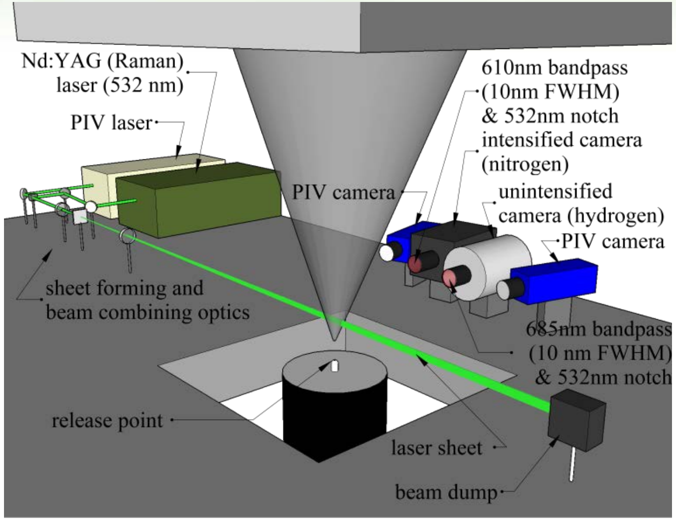
Moisture and air freeze on the nozzle as the temperature drops

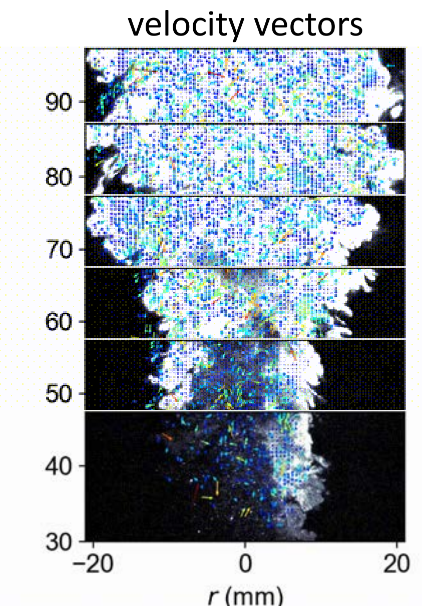
Air and moisture icing around liquid H_2 jet column – improves dispersion and reduces hazard distance

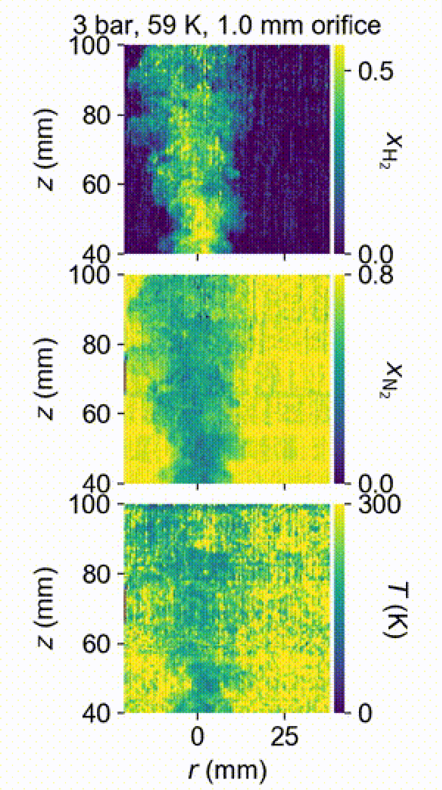
Strength at Sandia is using optical and laser diagnostics to measure external gas flows

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.
Particle Imaging Velocimetry	Seed particle motion between two illumination events (usually laser pulses) is measured. Displacement and time used to calculate velocity.
Laser Doppler Velocimetry	Particles pass through interference fringes and reflect light with intensity fluctuations at the Doppler shift frequency.
Raman scattering	Inelastic scattering off of different molecules gives each component a spectral fingerprint.

H_2-N_2 Raman imaging and particle imaging velocimetry are used to measure concentration, temperature, and velocity of cryogenic H_2



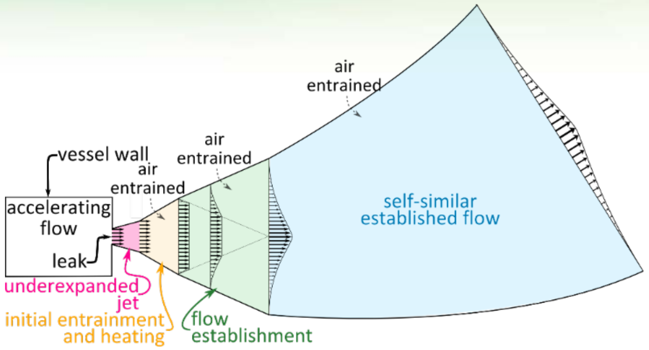


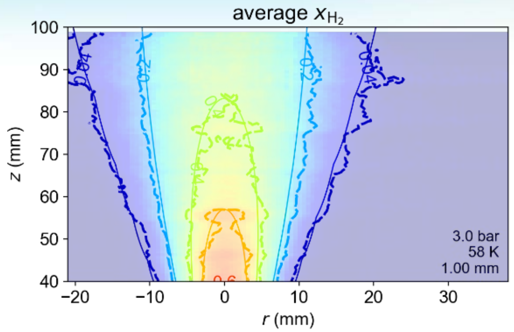


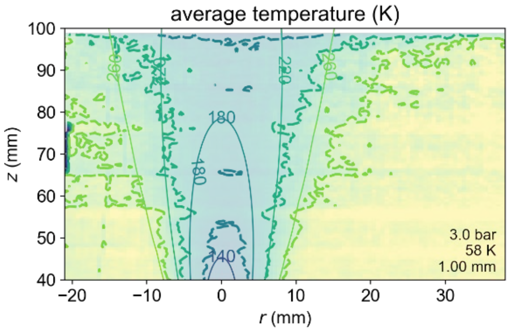
Independent model parameters:

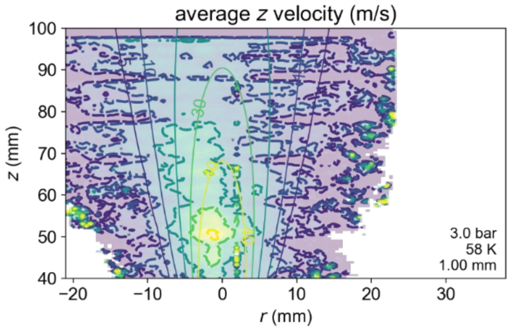
- ✓ T - temperature
- ✓ x - mole fraction
- ✓ v - velocity
- ✓ B - halfwidth (both velocity and concentration)

ColdPLUME model shows good agreement with the data









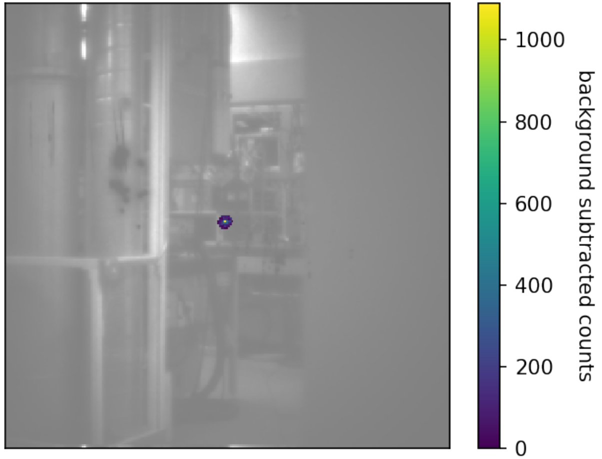
- Experimental results shown by shading and thick, dashed lines
- ColdPLUME model results are thin, solid lines

➢ Model accurately simulates mole fraction, temperature, and velocity -- can be used as a predictive tool

We are developing a modified Raman diagnostic to study LH_2 vents and large-scale experiments

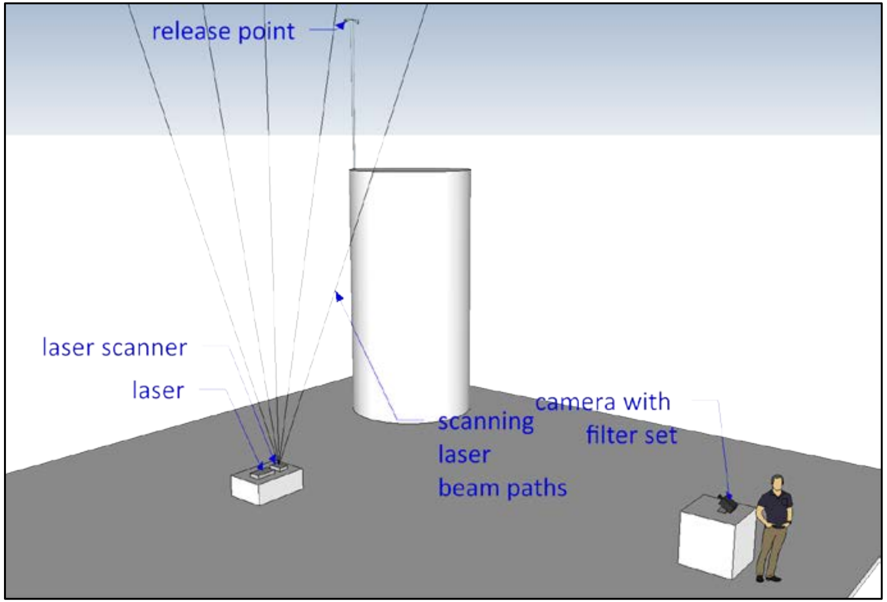
- Demonstrated acceptable signal to noise for large-scale diagnostic
- Uniquely fast optics enable collection of small Raman signal
- Imaged hydrogen from 40 foot standoff distance in the laboratory
- Observed nearly 30 degree field of view (20 ft scene from 40 ft distance)

Raman signal overlaid on laboratory scene



We have strategies for illumination of large-scale scene

- On-camera accumulation will provide a complete snapshot of the plume with reasonable resolution
- Effective background light suppression is key (both sunlight and illumination source that reflects off of condensed water vapor)
  - Time gating
  - Spectral gating
- High-powered light source required to excite as many molecules as possible
  - High-power laser scanning in space
  - Concentrations measured along a series of lines
  - 1st generation: low speed galvanometer using 10 Hz laser
  - 2nd generation: high speed polygonal scanning using pulse-burst laser



We are working with our colleagues at LLNL to perform LH_2 vent stack releases

- Additional temperature sensors along vent stack to validate internal flow model
- May require additional plumbing changes
- Replacing bull-horn with single outlet to enable model comparisons
- Variations in temperature, flow-rate, and external conditions (e.g. wind) in experiments
- Comparison to NREL sensor approach for some tests

Petitpas & Aceves, IJHE 43: 18403-18420: https://doi.org/10.1016/j.ijhydene.2018.08.097

- ➢ Heaters and pump enable a wide range of flow rates and temperatures at vent stack
- ➢ Proximity to SNL enables experiments to be run on short notice (when weather is right)

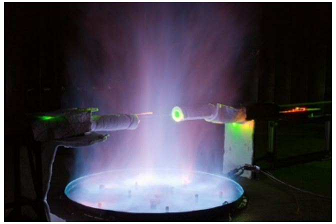
Remaining challenges: Executing outdoor experiments and planning additional large-scale experiments

Ensure safety when operating laser outdoors

- follow ANSI Z136 standard
- Non-visible (UV light) helps

Perform experiments during a range of weather conditions

- High- and low-wind conditions
- Humidity differences (potentially with precipitation)



Need experiments to characterize:

- Pooling
- Evaporation from LH_2 pools
- Interactions of plumes with ambient

Solution:

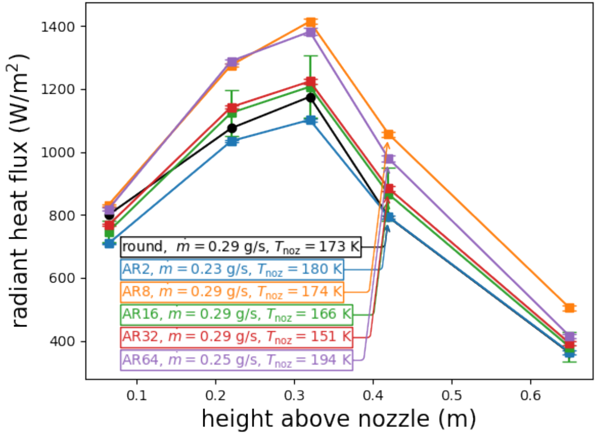
- Well-controlled experiments at Sandia facilities
- Partner with others, applying diagnostic at remote locations (European colleagues)

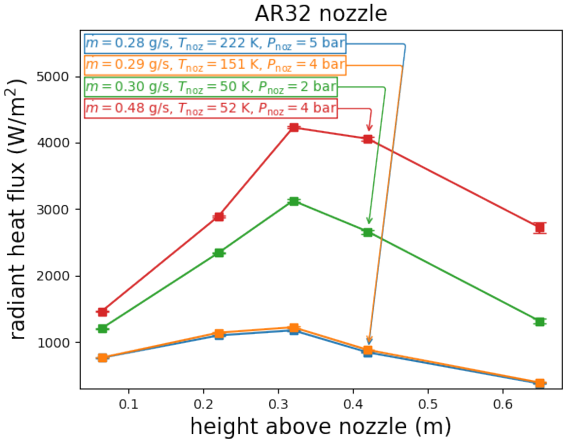
Ignited measurements (heat flux) are also important for calculating safety

Measuring whether the round nozzle is worst-case scenario as assumed

- Aspect Ratios: 2-64
- Nozzle pressures: 1.5-6 bar
- Nozzle temperatures: 48-295K

- For an equivalent mass flux, heat flux increases at cryogenic temperatures



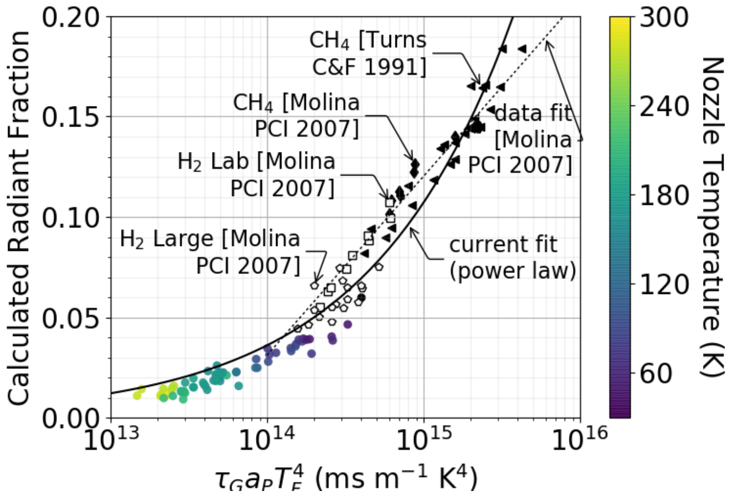


Hydrogen flames have less visible emission than other fuels

These experiments were conducted at SRI International in collaboration with Sandia National Laboratories Combustion Research Facility

We are supporting the CGA G-5.5 testing task force measurements of LH_2 vent stack flames

- Calculation of heat flux from vent stacks in CGA G-5.5 assumes high radiant fraction
- Radiant fraction for hydrogen much lower than other gases (no carbon that makes soot)
- Making measurements of vent stack flames to improve heat flux calculations



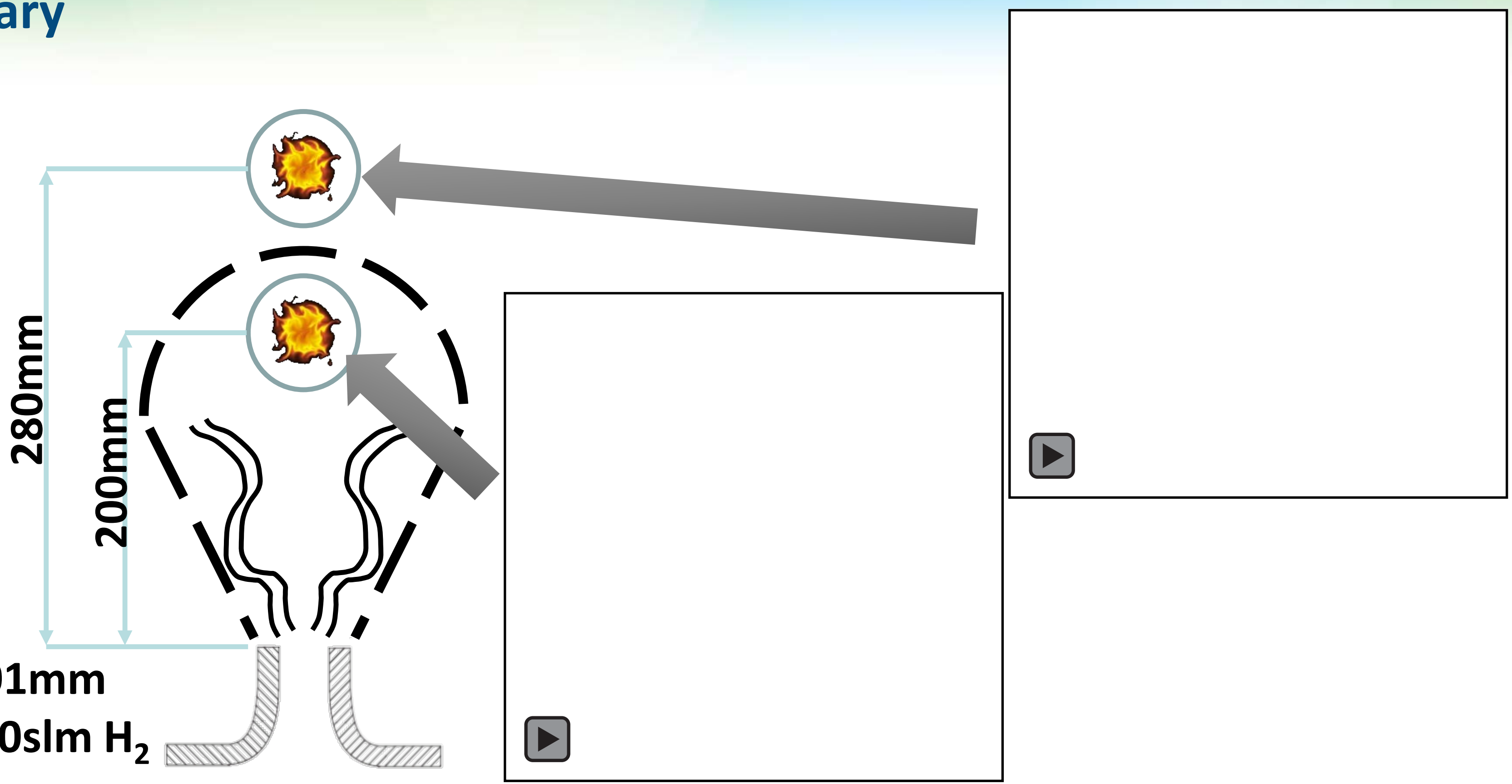
$$radiant\ heat\ flux \propto (radiant\ fraction)(mass\ flow)(heat\ of\ combustion)(transmissivity)$$

The ignition distance/light-up boundary is an important aspect of hydrogen flows for calculating safety

P = 1 bar, T = 290 K, distance = 85 mm

P = 1 bar, T = 37 K, distance = 325 mm

A laser spark ignition is used to precisely determine the light-up boundary



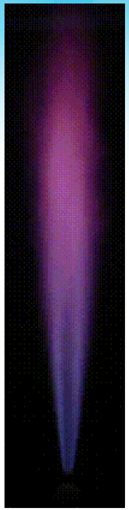
D = Ø1.901mm

Flow = 100slm H_2

Summary

- Lab-scale experiment liquefies gaseous hydrogen with liquid nitrogen and liquid helium
  - Silicon diode sensors for temperature
  - Mass-flow measured as a gas
- Cryogenic hydrogen dispersion measured using simultaneous Raman scattering and particle imaging velocimetry
- Raman diagnostic being scaled for larger experiments
  - Camera remains fixed
  - Laser illumination scanned in space to create 3-D measurements of concentration
- Measurements of reacting hydrogen also made at lab and larger scale
  - Ignition distance using laser spark
  - Radiant fraction and heat flux
    - Round and high aspect ratio nozzles at lab scale
    - Liquid hydrogen vent stacks with CGA G-5.5 testing task force

QUESTIONS?



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- United States Department of Energy, Energy Efficiency &Renewable Energy, Fuel Cell Technologies Office, Safety, Codes, and Standards subprogram managed by Laura Hill
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- Previous researchers: Pratikash Panda, Joe Pratt, Katrina Groth, Isaac Ekoto, Adam Ruggles, Bob Schefer, Bill Houf, Greg Evans, Bill Winters

Schlieren imaging

- Measures gradients in density (1^{st} 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

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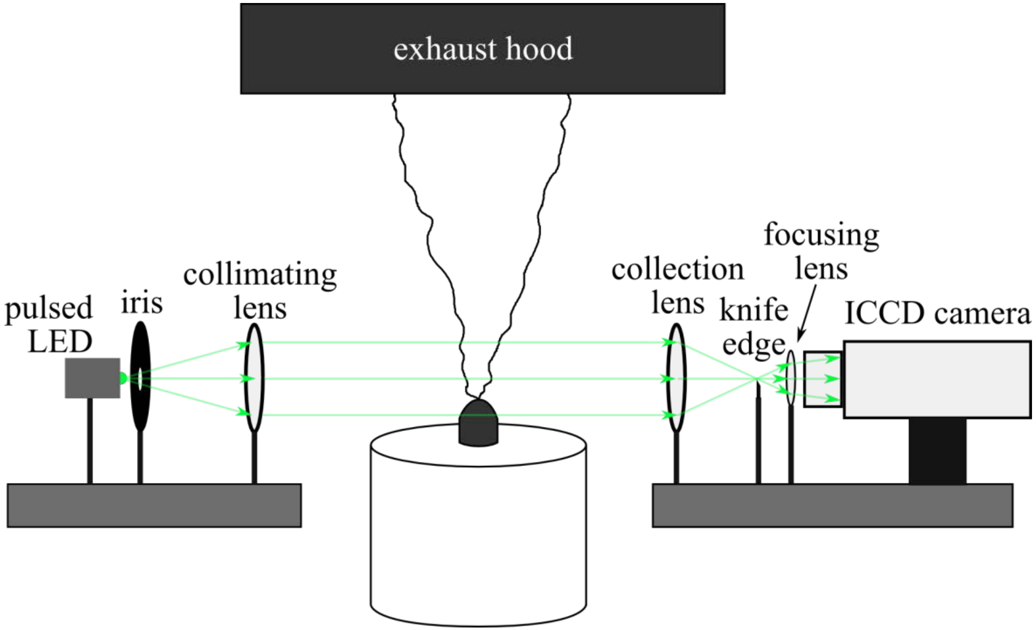
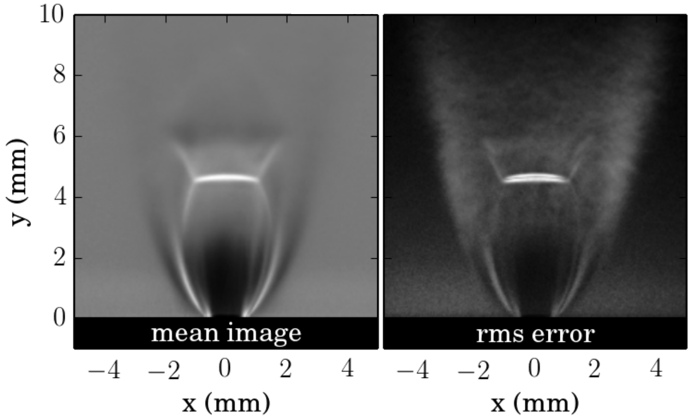
H2FC Hydrogen and Fuel Cells Program

Schlieren imaging is used to characterize near-nozzle region and other regions with high density gradients



50 bar room temperature H_2 release

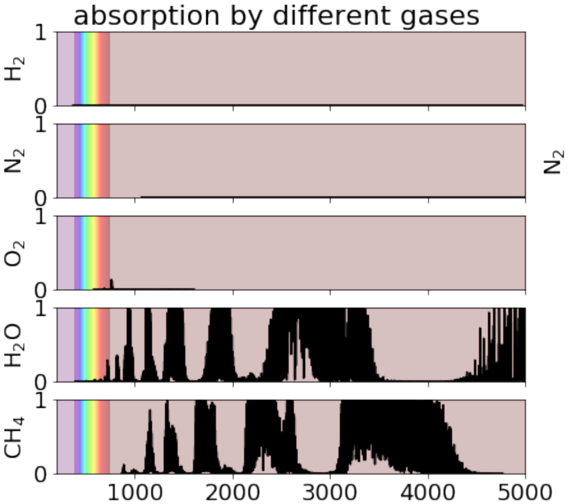


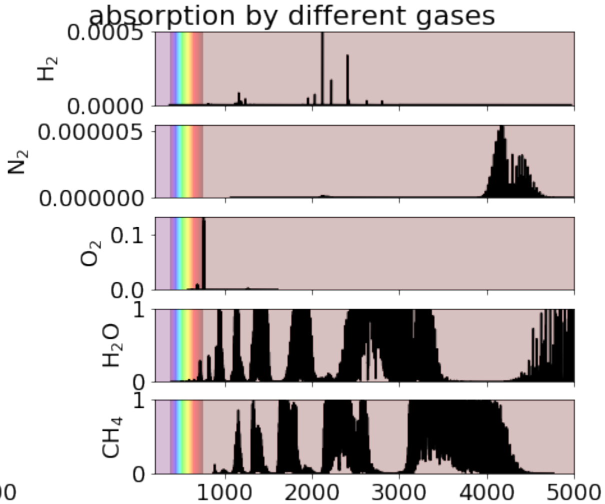
Fluorescence

- 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

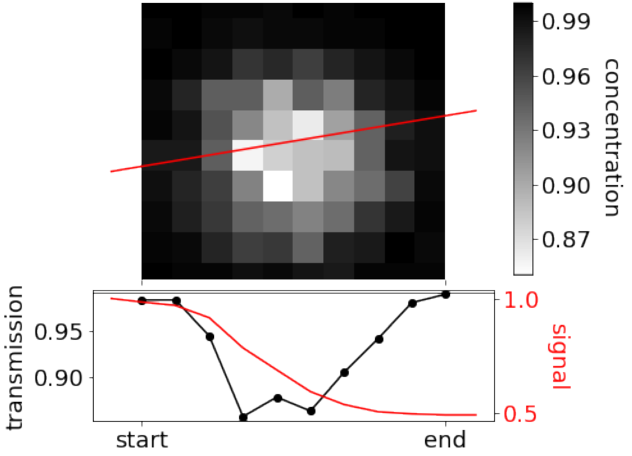
Absorption

- 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





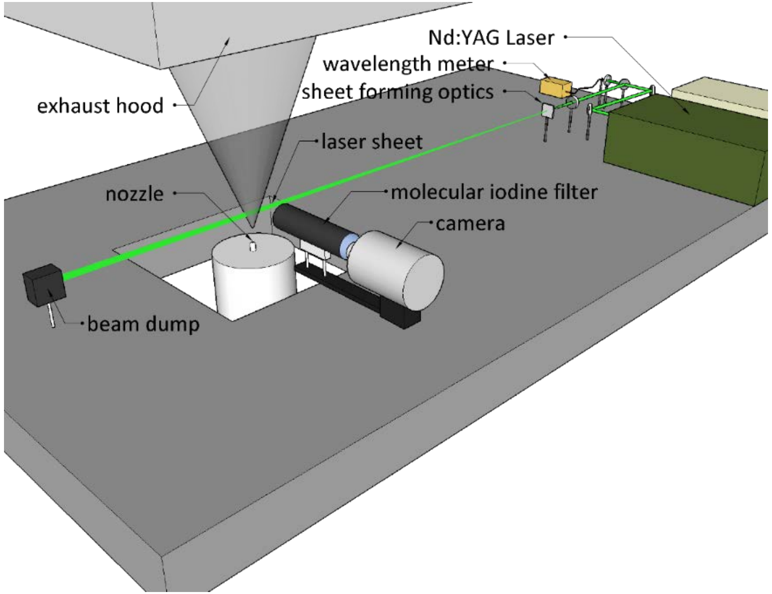
Data from HITRAN Application Programing Interface (HAPI): 10.1016/j.jqsrt.2016.03.005



Rayleigh scattering

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

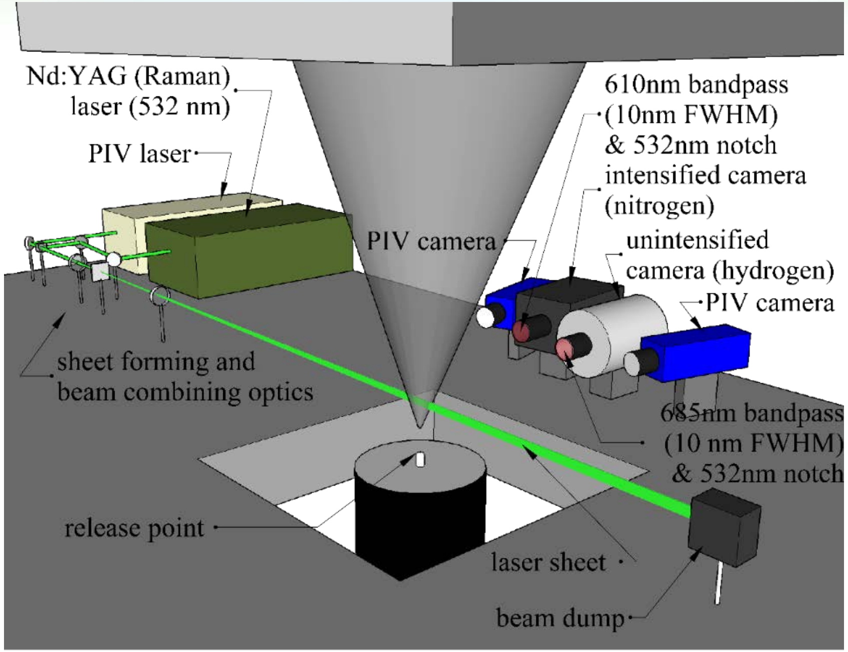
- Planar laser Rayleigh scattering used at Sandia for atmospheric temperature hydrogen releases
- Scatter proportional to number density; variations are caused by both composition and temperature
- For warm releases, always measured in atmospheric temperature region to eliminate this variable and enable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

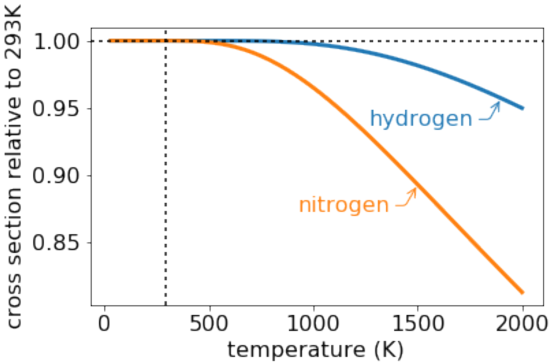


H_2: shift of 4161 cm^{-1} (532nm → 683 nm, 355nm → 416 nm)

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: $1 = x_{H_2} + 1.28x_{N_2}$

$$\begin{cases} x_{H_2} = \dfrac{\mathrm{I}_{\mathrm{H}_2}}{k_{H_2}\left(\dfrac{\mathrm{I}_{\mathrm{H}_2}}{k_{H_2}} + \dfrac{1.28\mathrm{I}_{\mathrm{N}_2}}{k_{N_2}}\right)} \\[2ex] x_{N_2} = \dfrac{\mathrm{I}_{\mathrm{N}_2}/\mathrm{I}_0}{k_{N_2}\left(\dfrac{\mathrm{I}_{\mathrm{H}_2}}{k_{H_2}} + \dfrac{1.28\mathrm{I}_{\mathrm{N}_2}}{k_{N_2}}\right)} \\[2ex] T = \dfrac{1}{\dfrac{\mathrm{I}_{\mathrm{H}_2}}{k_{H_2}} + \dfrac{1.28\mathrm{I}_{\mathrm{N}_2}}{k_{N_2}}} \end{cases}$$

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