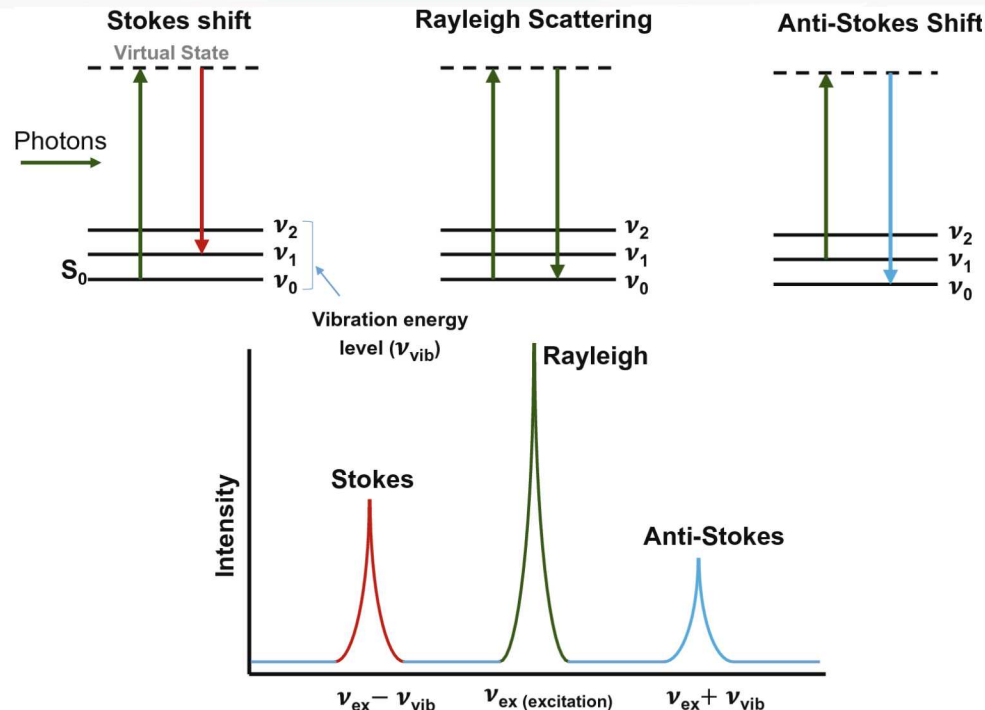# Hydrogen detection via Raman scattering

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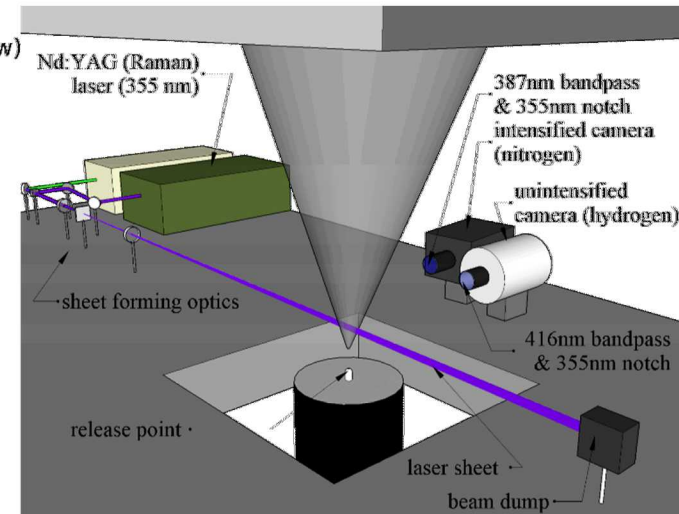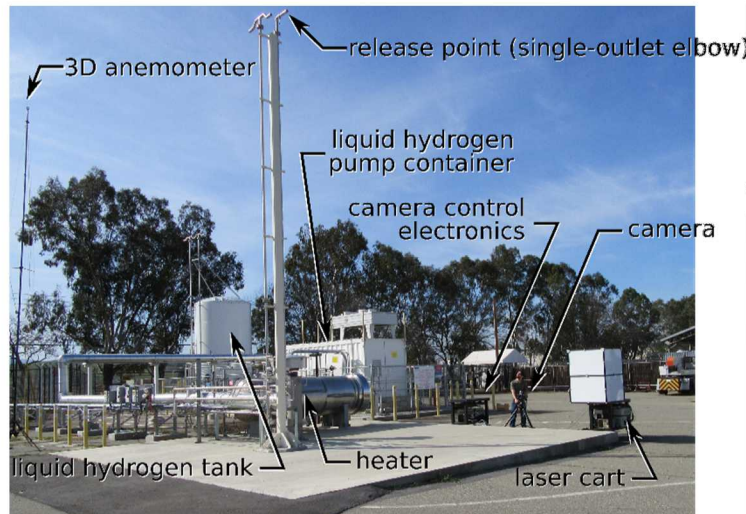
Raman scattering is inelastic light scatter – the color shifts depending on the gas molecule

- Rayleigh scattering is elastic
  - H_2 Rayleigh cross-section $\approx 10^{-27}$ cm^2
  - Scatter proportional to number density; variations in intensity caused by variations in both composition and temperature
- Entrained moisture in cryogenic plumes also produces inelastic Mie scattering
- Raman scattering is inelastic (therefore selective to a specific species)
  - H_2 Raman cross-section $\approx 10^{-30}$ cm^2
  - H_2: shift of 4161 cm^{-1} (532nm $\rightarrow$ 683 nm, 355nm $\rightarrow$ 416 nm)
  - N_2: shift of 2331 cm^{-1} (532nm $\rightarrow$ 607 nm, 355nm $\rightarrow$ 387 nm)

Image from: Cho, Y.C., Ahn, S.I. Fabricating a Raman spectrometer using an optical pickup unit and pulsed power. *Sci Rep* **10**, 11692 (2020). https://doi.org/10.1038/s41598-020-68650-7
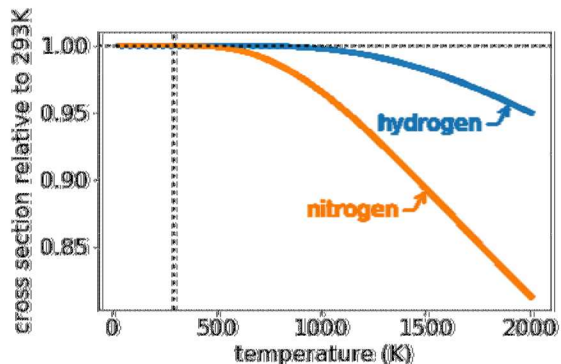
Planar Raman imaging has been used in a lab setting and Raman line-imaging has been used outdoors to detect cryogenic hydrogen plumes

- Signals are low
  - High powered light source required (e.g., ~400 mJ/pulse @ 355nm, 12mm tall sheet)
  - Fast optics needed for collection (f# ≤ 1.2)

- Large Raman shift enables use of high optical density filters to remove unwanted Mie and Rayleigh signals

- Challenges due to:
  - Background light
  - Polarization effects
  - Angular dependence of optical filters

Quantification of concentration from Raman signals requires calibration of detection system

- 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: $\dfrac{I_{H_2}}{I_0} = k_{H_2}\dfrac{x_{H_2}}{T}$ ← unknown 1
$\qquad\qquad\qquad\qquad$ ← unknown 2

measured values $\qquad\qquad$ calibration constants

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

based on the composition of air

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

$$
\begin{cases}
x_{H_2} = \dfrac{I_{H_2}}{k_{H_2}\left(\dfrac{I_{H_2}}{k_{H_2}} + \dfrac{1.28 I_{N_2}}{k_{N_2}}\right)} \\[3em]
x_{N_2} = \dfrac{I_{N_2}/I_0}{k_{N_2}\left(\dfrac{I_{H_2}}{k_{H_2}} + \dfrac{1.28 I_{N_2}}{k_{N_2}}\right)} \\[3em]
T = \dfrac{1}{\dfrac{I_{H_2}}{k_{H_2}} + \dfrac{1.28 I_{N_2}}{k_{N_2}}}
\end{cases}
$$