Efficiency Assessment of Single Cell Raman Gas Mixture for DIAL Ozone Lidar

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[05].[Greenhouse gases, tracers, and transport in the free troposphere and above]

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Aim of the study

To generate coaxial beam of multiple SRS with a single Raman cell.

To identify best suitable gas mixing ratio for optimum conversion efficiency.

To figure out the SRS mechanisms (steady-state gain, four-wave mixing, transient gain reduction)

Advantage: Simple optical configuration and make the DIAL system compact for mobile operation.

Experimental Setup

Procedure: 1. Change the Gas pressure @ constant input energy & focal point
2. Change the input laser energy @ constant gas pressure and focal point.
3. Change the focal point @ constant gas pressure and laser energy.

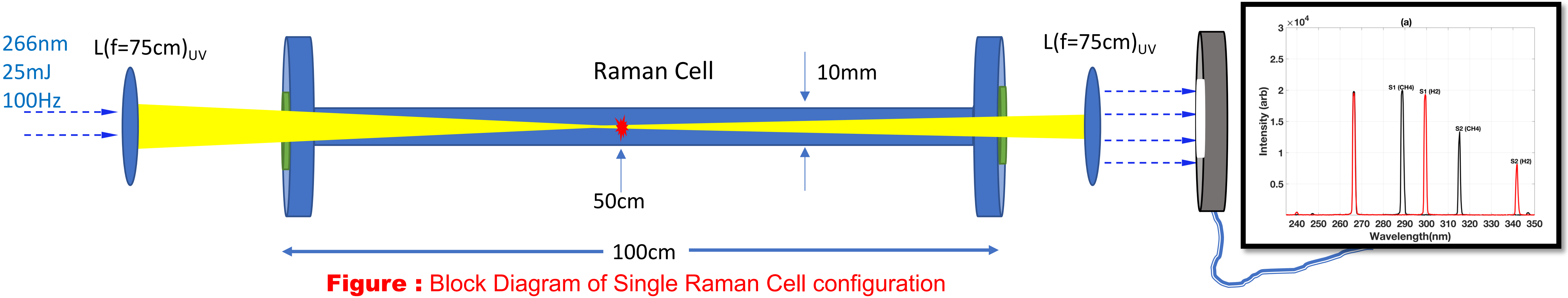
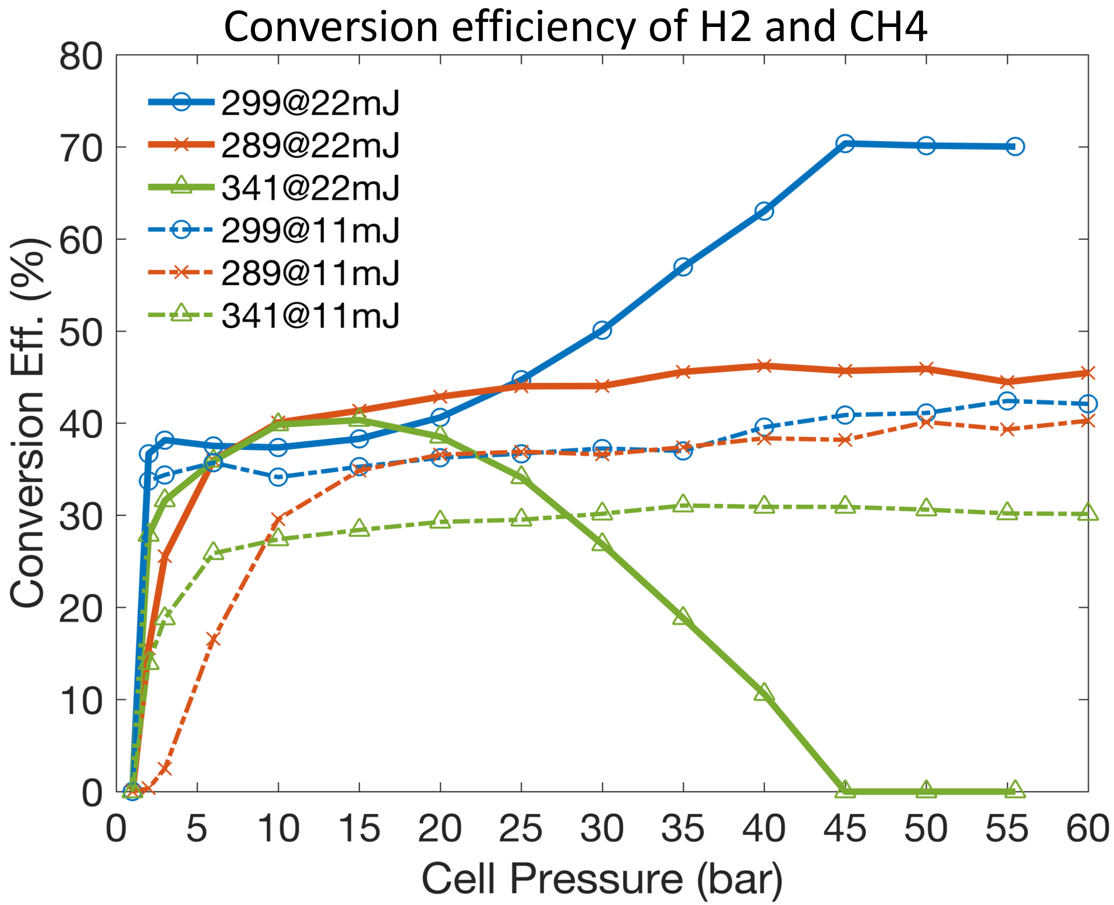


Figure : Block Diagram of Single Raman Cell configuration

- Stimulated Raman Scattering (SRS) is a coherent nonlinear optical mixing process that mainly dependent on the laser spectral linewidth, input laser intensity and the interaction length of the Raman active medium (Jones et al., 2019).
- Several researchers used longer (~2m) and two Raman cells to generate wavelength pair while indoor regular operations are required (Nakazato et al., 2019; Sullivan et al., 2014;Haner et al., 1990).
- The first Stokes line is produced by SRS. The second and third Stokes may be produced stimulated Raman gain (SRG) or FWM due to the mixing of original pump and the generated Stokes (Chu et al., 1991)..
- We examined the effect of steady-state Raman gain, transient reduction, and four-wave mixing on conversion efficiency of H_2 and CH_4.

Individual Gases and Four Wave Mixing (FWM)



- When high power laser is passed through low pressure H2, Four-wave mixing (FWM) processes will generate Second stoke (S2).
- When high power laser is passed through low pressure CH4, Steady state Raman gain(SRG) will generate Second stoke (S2) and FWM will generate Second stoke (S3).
- At low input laser energy, Second stokes in both H2 and CH4 is generated by SRG

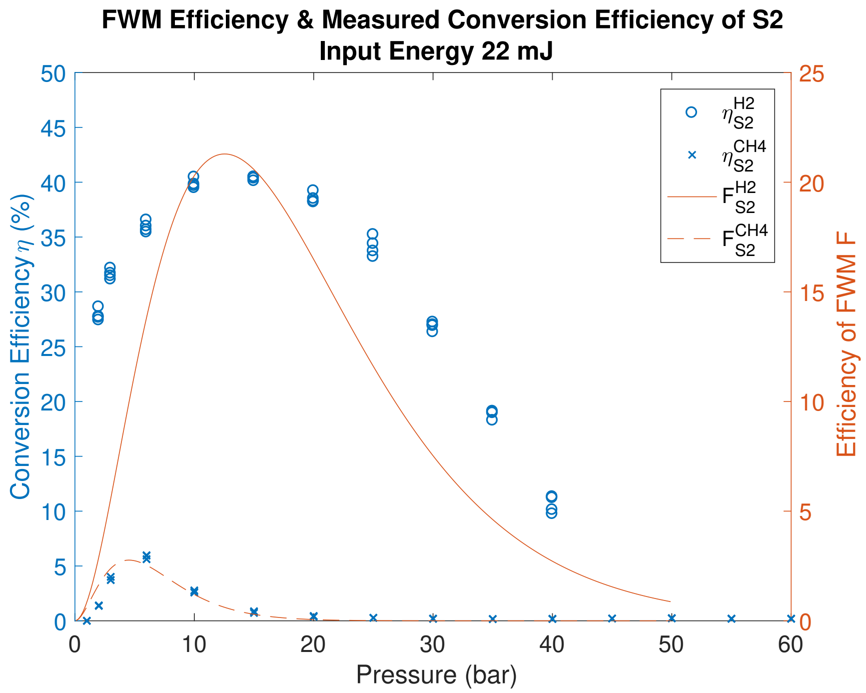
Raman Process	H_2	CH_4	FWM Process	H_2	CH_4
$\nu_{S_1}=\nu_{S_0} - \nu_R$	299	289	$\nu_{F_2}=\nu_{S_1}+\nu_{S_1} - \nu_{S_0}$	341.48	314.86
$\nu_{S_2}=\nu_{S_0} - 2\nu_R$	341.48	314.86	$\nu_{F_2}=\nu_{S_0}+\nu_{S_1} - \nu_{AS_1}$	341.48	314.86
$\nu_{S_3}=\nu_{S_0} - 3\nu_R$	397.95	346.70	$\nu_{F_3}=\nu_{S_1}+\nu_{S_2} - \nu_{S_0}$	397.95	346.70
$\nu_{S_4}=\nu_{S_0} - 4\nu_R$	476.78	385.71	$\nu_{F_4}=\nu_{S_1}+\nu_{S_3} - \nu_{S_0}$	476.78	385.71

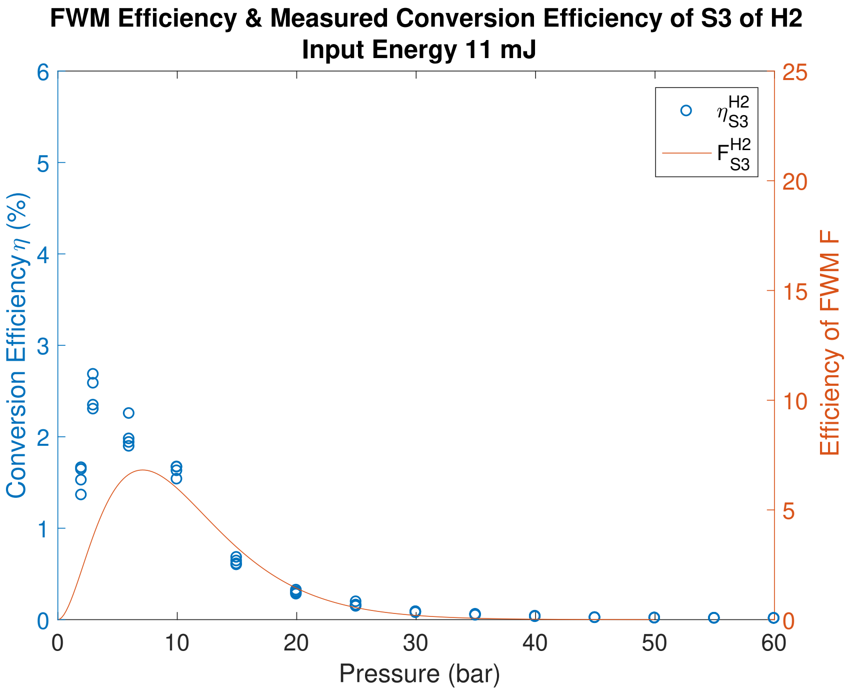
H2 (299.1nm) : Maximum conversion efficiency of 70% is achieved at 45bar with higher stokes completely suppressed.
CH4 (288.4nm) : Maximum conversion efficiency of 42% is achieved at 25bar.

The process of generation of second and third stokes highly depends on laser input energy, type of molecule and pressure.

Use of individual gases requires two cells that makes complex optical configuration for DIAL system.

FWM & Conversion efficiency





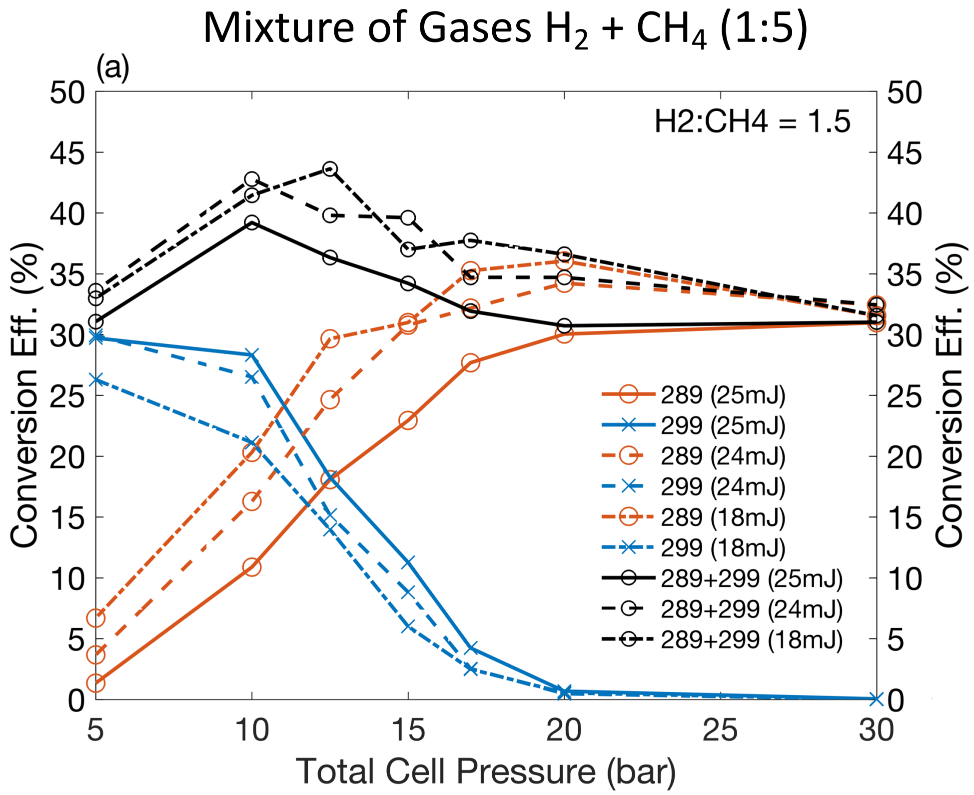
Four Wave mixing is a third order non-linear process that coexists with SRS. The efficiency of FWM can be expressed as

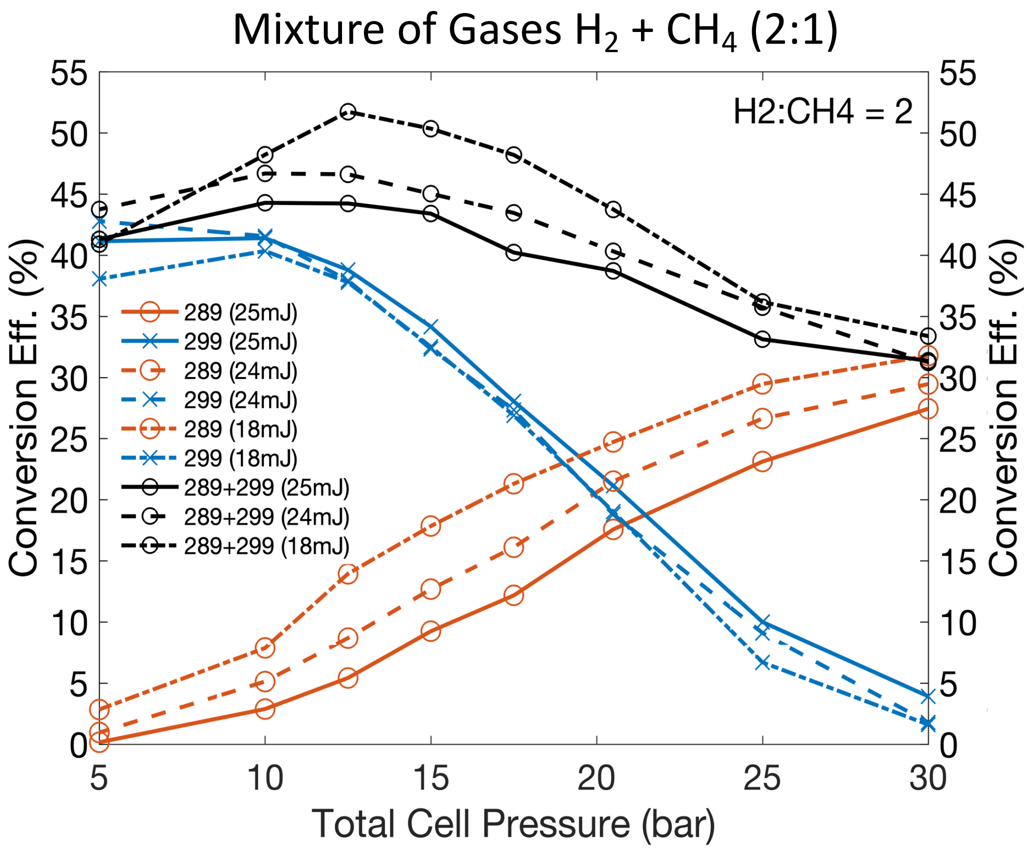
$$F = Bp^2 \exp(-b|\Delta k|)$$

where b=2z_R is the confocal beam parameter, p is the active gas pressure, and k is the wavevector mismatch caused by the dispersion of the medium. The parameter B depends on the nonlinear coefficient of the media, the beam-focusing parameters, and the wave-vector values.

- Conversion efficiency of S2 and S3 of Raman active medium H_2 and CH_4 is directly proportional to the Four wave mixing process.
- Four Wave Mixing (FWM) may generate higher order stokes however wave vector mismatch reduces its energy.

Conversion Efficiency of Mixture of Gases



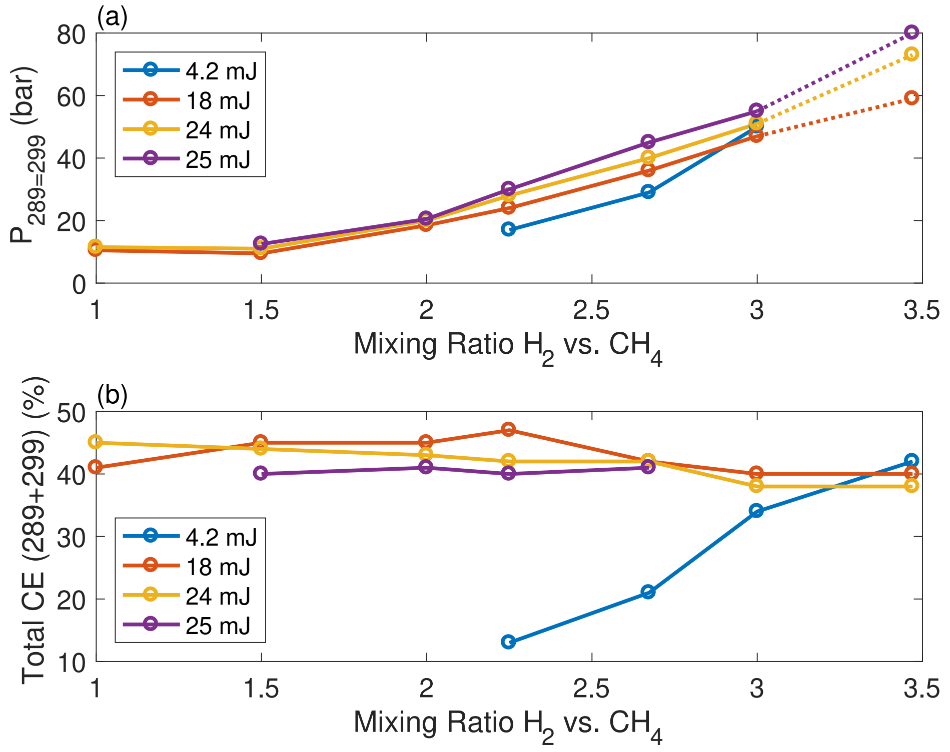


- ✓ Efficiency of different mixing ratios (H2+CH4) with varying energy is measured.
- ✓ A mixing ratio (H_2:CH_4) of 2:1 could give a total conversion efficiency (288.4+299.1) of about 42% with 18mJ input laser energy.

Coaxial beam of On and OFF wavelengths is generated from single cell. Provides simple optical configuration for mobile DIAL system for ozone measurements.

Summary

- ✓ **Pure H2 and CH4 :** Conversion efficiencies of first stokes lines in general monotonically increase with pressure or laser power.
- ✓ **Mixture of gases**: Efficiency of first stokes line of H2 is inversely proportional to pressure while that of CH4 directly proportional to pressure at a given input energy.
- ✓ Single Raman cell filled with mixture of gases and full laser power can have higher CE than using two Raman cells filled with pure gases with split input laser power (half).
- ✓ The total output energy of coaxial beam of 288.4+299.1nm is about 12mJ with an input laser energy of 18mJ entering the cell.
- ✓ Four wave Mixing is responsible for generation of higher order stokes and wave vector mismatch contributes to reduce higher order stokes energy.



References:

1. Jones, R.R., Hooper, D.C., Zhang, L., Wolverson, D., Valev, V.K. (2019): Raman techniques: fundamentals and frontiers. *Nanoscale research letters* 14(1), 1-34.
2. Sullivan, J.T., McGee, T.J., Sumnicht, G.K., Twigg, L.W., Hoff, R.M.: A mobile differential absorption lidar to measure sub-hourly fluctuation of tropospheric ozone profiles in the Baltimore-Washington, D.C. region. *Atm. Meas. Tech.* 7(10), 3529-3548 (2014)
3. Chu, Z., Singh, U. N., & Wilkerson, T. D. (1991). Multiple Stokes wavelength generation in H_2, D_2, and CH_4 for lidar aerosol measurements. *Applied Optics*, 30(30), 4350.
4. Nakazato, M., Nagai, T., Sakai, T., & Hirose, Y. (2019). Differential Absorption Lidar For Tropospheric Ozone Measurement Using Stimulated Raman Scattering In Co2, *IEEE J. of Quantum Electronics*, 26, 7, 1292-1298.
5. Haner, D. A., & Mcdermid, I. S. (1990). Stimulated Raman Shifting of the Nd: YAG Fourth Harmonic (266 nm) in H2, HD, and D2. IEEE Journal of Quantum Electronics, 26(7), 1292-1298.

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