Generation of Infrared Radiation by Stimulated Raman Scattering in Liquid and Solid Parahydrogen

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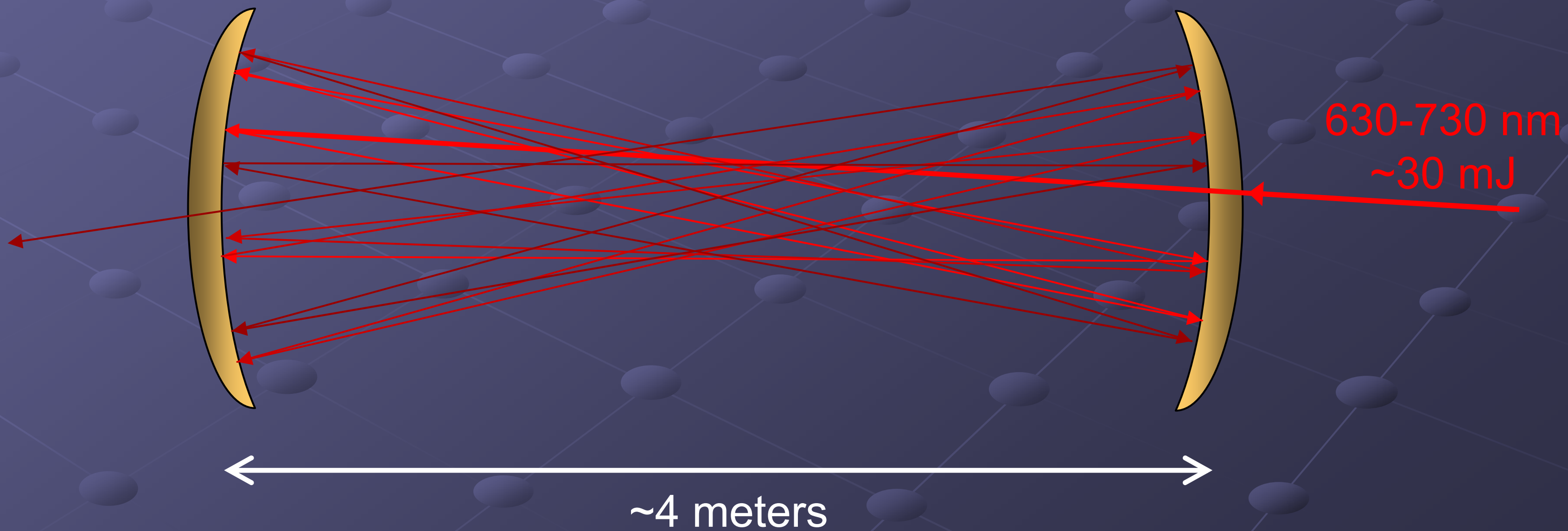
Stimulated Raman Scattering

- Many people want infrared lasers
- One of best options: Raman shifting a pulsed visible (e.g. dye) laser
- Widely tunable from near-UV to mid-IR
- Easily coupled with cavity ring-down
  - absorption sensitivity ~ 1 part in 10^6
  - ideal for pulsed (supersonic expansion) sources

SRS: The Method

- High pressure gaseous H_2 cell (4155 cm^{-1} shift)
- Third Stokes from dye laser → 3–8 μm
- Raman efficiency ∝ 1/λ
  - need multipass cell, high input power



SRS: The Problem

- Need high input power into Raman cell
- Shallow focus into long (~4 m) cell
- Focus can walk around in cell with multipasses
- Need high reflectivity, high damage threshold

new mirror

used mirror

The Promise of Solid H_2

- Gain $\propto n/\Gamma$
- Solid: higher n, smaller Γ
- Solid Raman gain is 1000x higher
  - Katsuragawa & Hakuta, *Opt. Lett.* **25**, 177 (2000)
- Eliminate multipass cell?

$\Gamma = 7$ MHz (0.0002 cm^{-1})
$n = 2.7 \times 10^{22}$ /cm^3

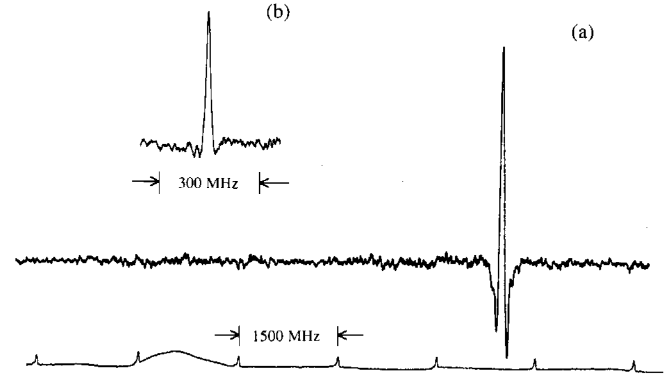
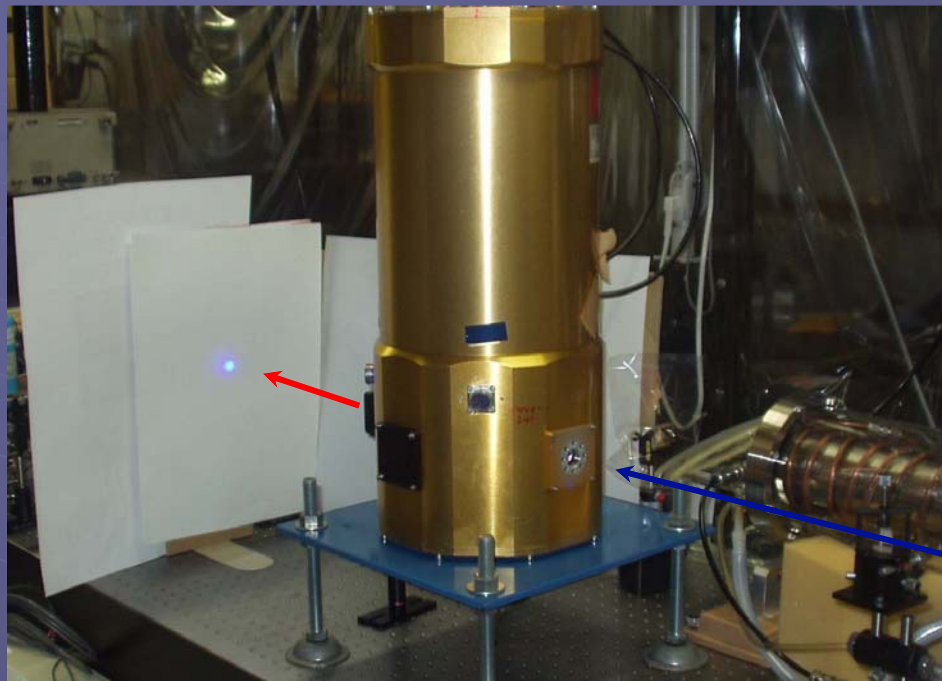


FIG. 1. Examples of the observed spectral line. (a) 80-MHz radiofrequency sidebands. The fine structure is due to overmodulated rf sidebands. The spacing of the spectrum analyzer markers is 1500 MHz. The feature on the left in the I_2 spectrum is at 16 831.5127 cm^{-1}. The detection time constant is 1 sec. (b) A high-resolution spectrum observed by using 8-MHz radiofrequency sidebands. The linewidth at half height at half maximum is ~7 MHz. The detection time constant is 1 sec.

Momose, Weliky, & Oka
J. Mol. Spectrosc. **153**, 760 (1992).

First Results (Kyoto)



crystal

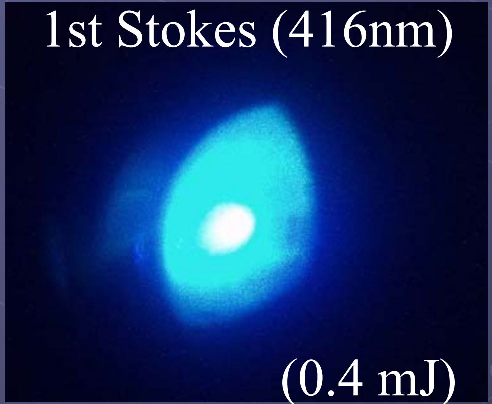
length: 10 cm

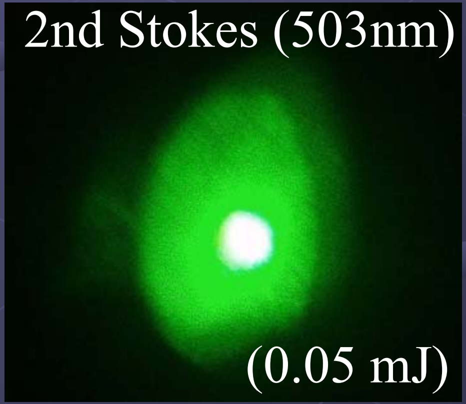
diam. : 4 mm

355nm

(8 mJ, unfocused

3ns, 250MHz width)

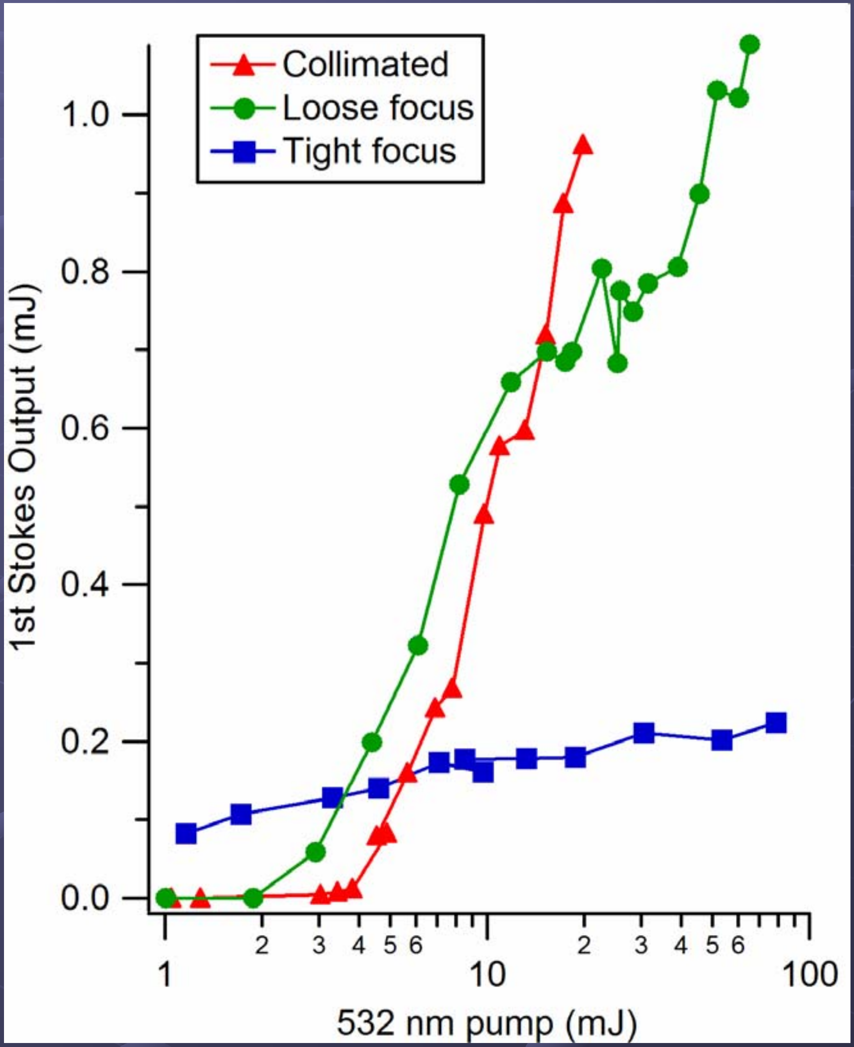




M. Fushitani, S. Kuma, Y. Miyamoto, H. Katsuki, T. Wakabayashi, T. Momose, & A.F. Vilesov, Opt .Lett. 28, 37, 2003

Chicago Experiments (June 02)

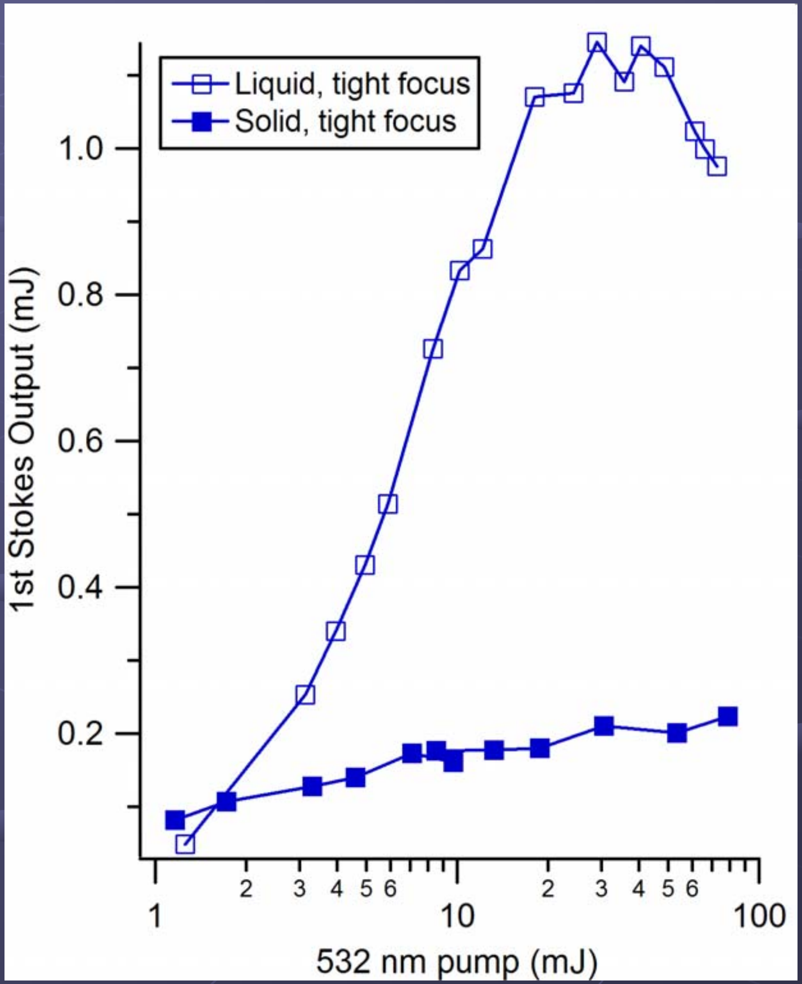
- Pump: broad linewidth doubled Nd:YAG
- Peculiar dependence on focal properties
- Poor higher-order conversion (70 mJ pump):
  - 2nd Stokes: 100 μJ
  - 3rd Stokes: 0.2 μJ



B. J. McCall, A. J. Huneycutt, R. J. Saykally,
C. M. Lindsay, T. Oka, M. Fushitani, Y. Miyamoto, & T. Momose, Appl. Phys. Lett. 82, 1350, 2003.

Liquid Parahydrogen

- Little studied since Stoicheff (1964)
- High "damage threshold"
- Free of defects
- Better than solid, with tight focus
- No output with collimated pump!



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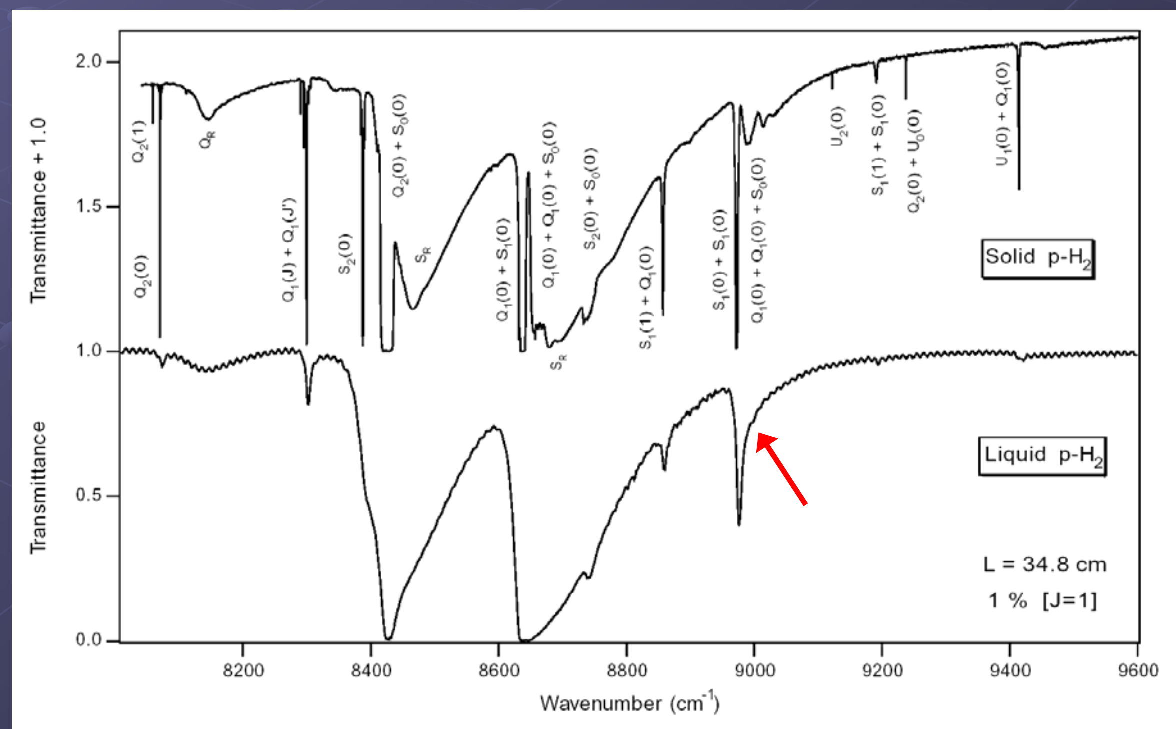
Alexandrite Laser (Berkeley)

- Light Age, Inc. PAL
  - Linewidth ~60 MHz
  - Pulse length ~100 ns
  - Wavelength 758 nm
- Directly into parahydrogen crystal:
  - Collimated: 62 mJ → 1.5 mJ (1st)
  - Focused: 39 mJ → 130 μJ (1st), 2 μJ (2nd)
- Using 1st Stokes output of D_2 shifter (978 nm)
  - 3 mJ → 50 μJ (1.65 μm)
  - 7 mJ → 150 μJ
  - 14 mJ → 250 μJ
  - lots of anti-Stokes light (up to 3rd order)

Alexandrite, ctd. (Berkeley)

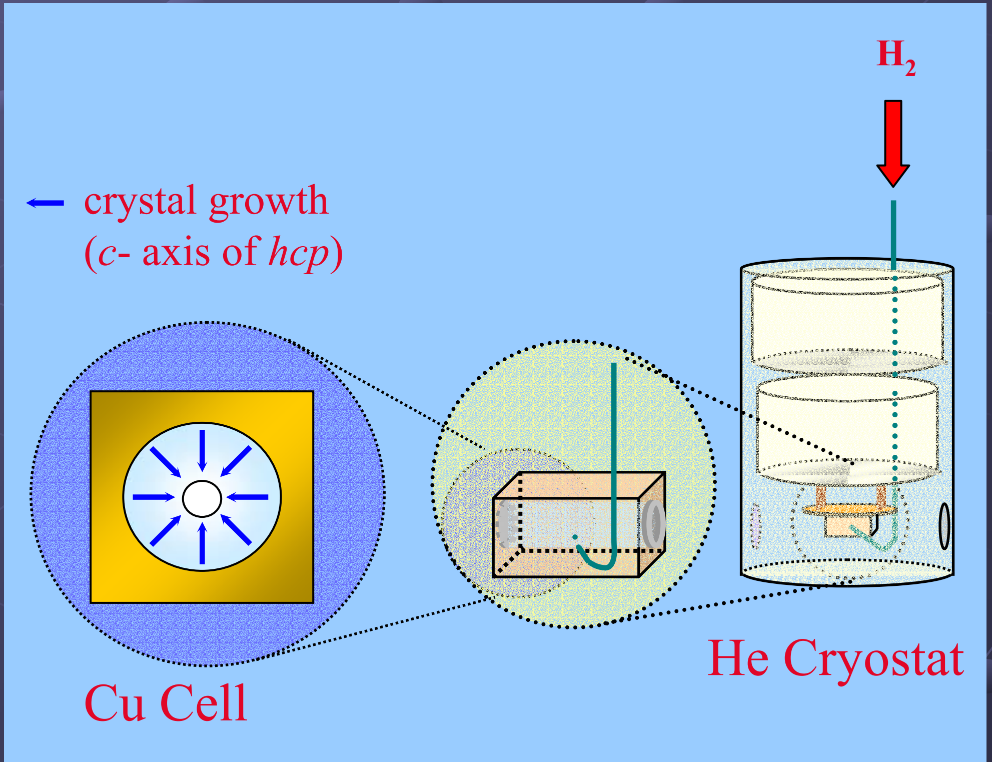
- Liquid parahydrogen
- "Boiling" observed with 1st-Stokes output
- Bad choice of wavelength

Mengel, Winnewisser, & Winnewisser
Can. J. Phys. 78, 317, 2000.



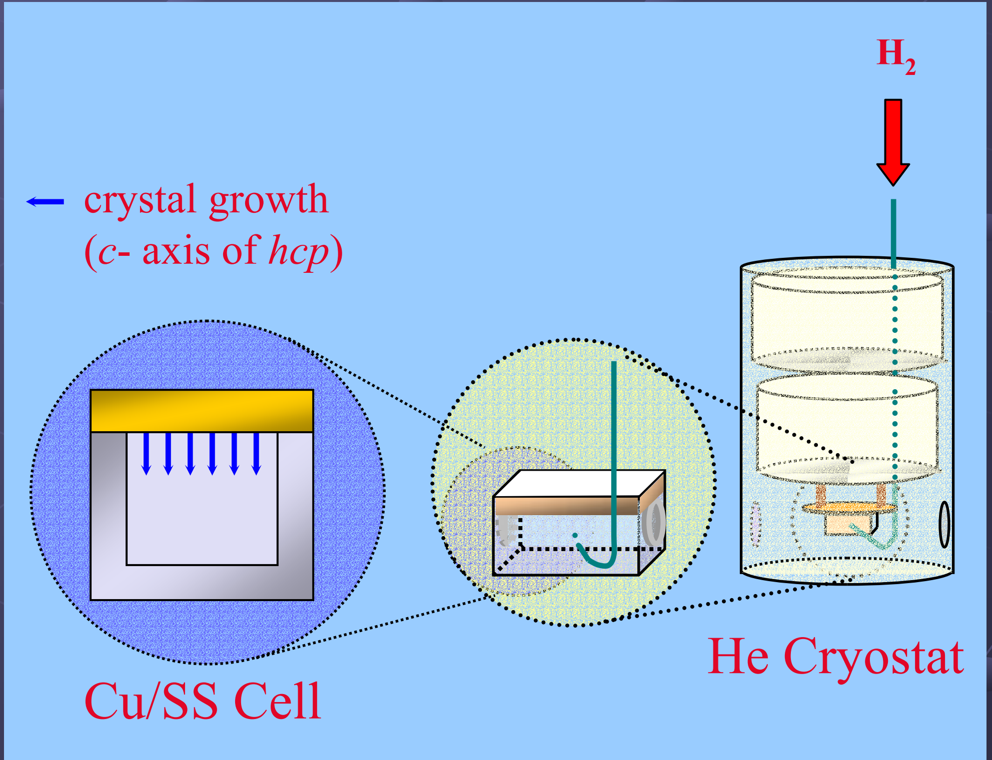
Problems

- Divergence of Stokes output beam
- Poor transparency of crystal, central void
- Damage frequently occurs at interface with window



Future Work

- So far, results are not very encouraging
- Not yet competitive with multipass cell
- One last idea: grow a transparent crystal from the liquid
  - no void in middle
  - fewer defects at interface with window



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