Further Studies on Sharp Transitions Apparently Not Induced in Solid Parahydrogen by Methane

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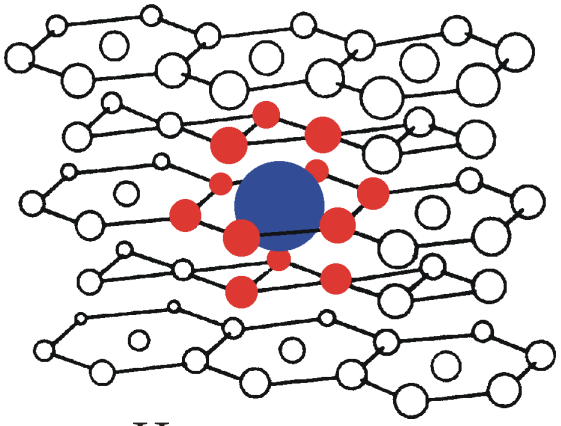
Solid parahydrogen

Crystal Features:

- Weak intermolecular interactions
- Slow relaxation times
- Quantum crystal-self repairing lattice via tunneling
- ~10 MHz FWHM routinely observed
- Large intermolecular distance, 3.783 Å

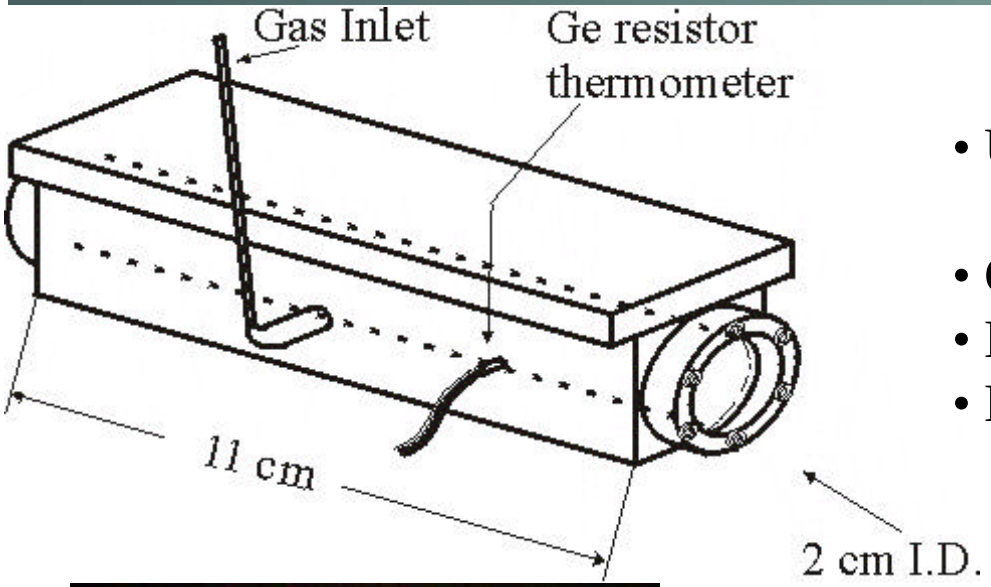
Methane in solid *p*-H_2



***p*-H_2 as a Matrix Medium**

- Larger intermolecular distance than noble gas matrices
- Very small matrix shifts
- Nearly free rotation
- High impurity mobility
- Spectroscopy of *p*-H_2 near impurity
- Narrow impurity spectral linewidths

Copper cell production of solid *p*-H_2



- Ultra high purity H_2 converted to ~99.9% *p*-H_2
- Cooled in a liquid He cryostat
- Lattice grown radially inward
- Exclusively HCP

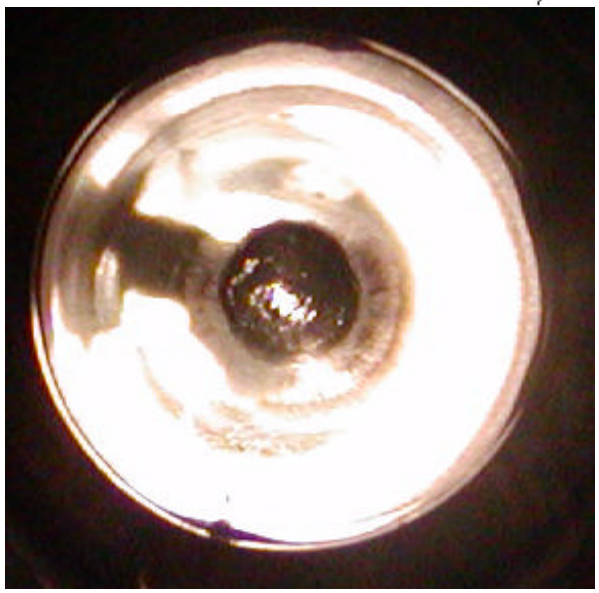
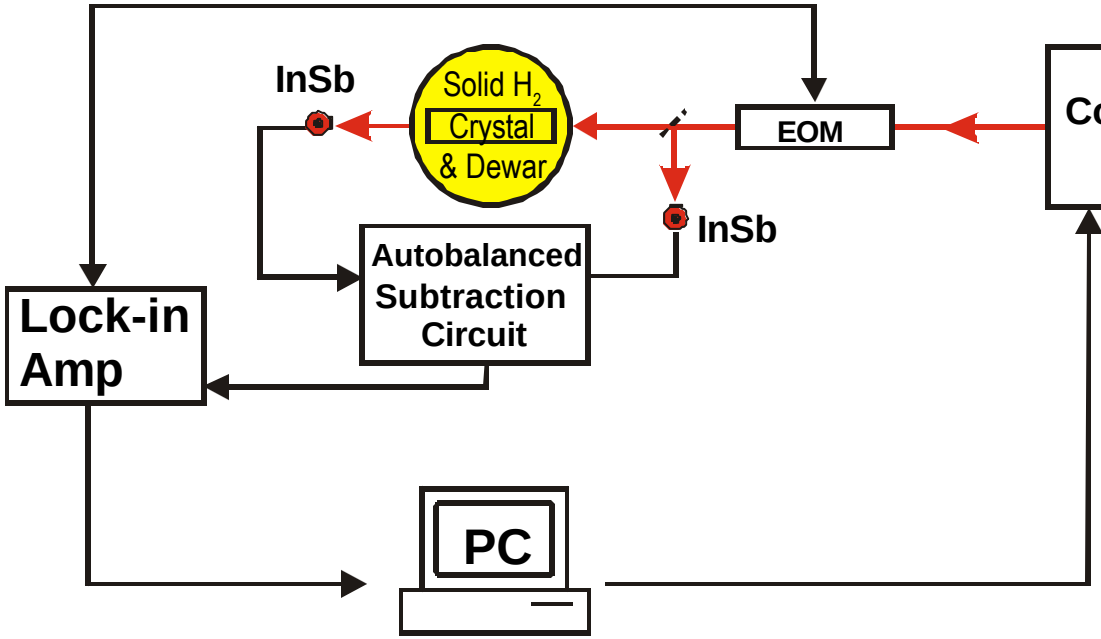
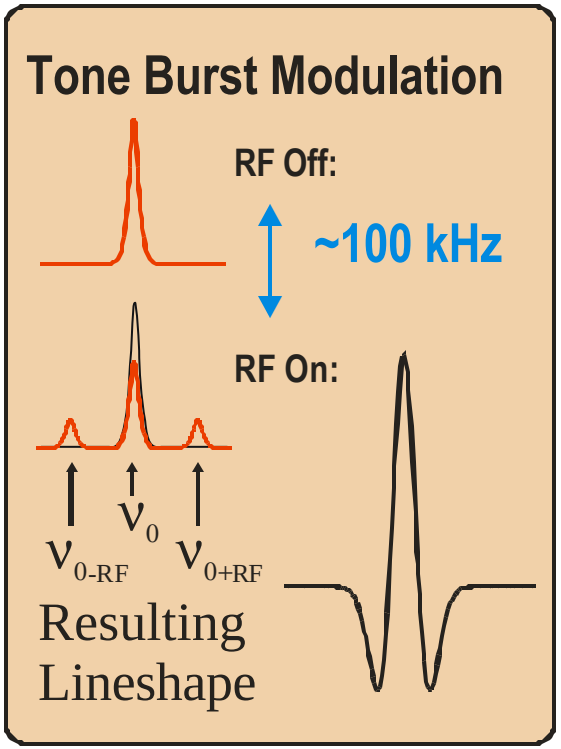


Photo From One End

Copper cell cryostat

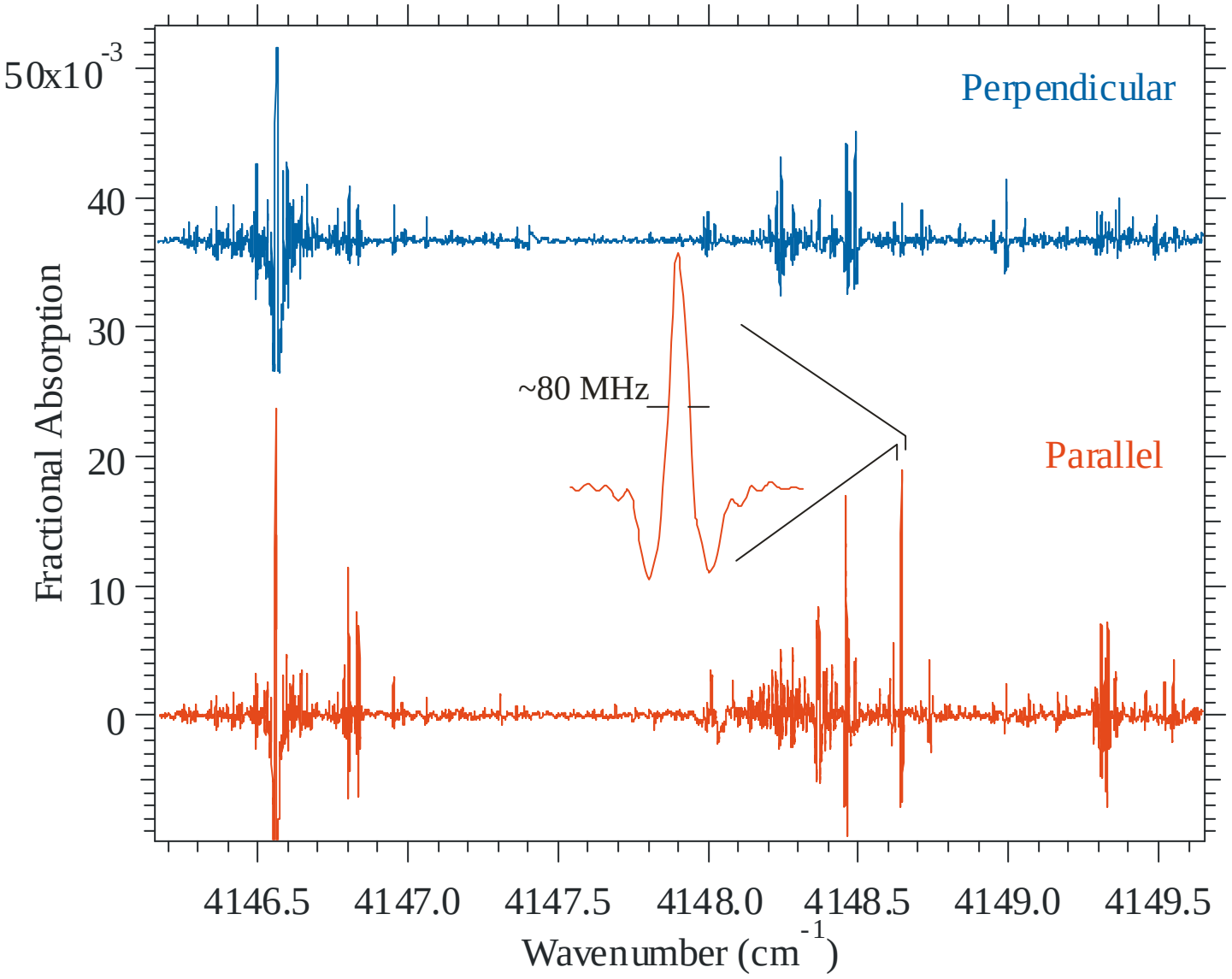
Laser spectroscopy of solid *p*-H_2



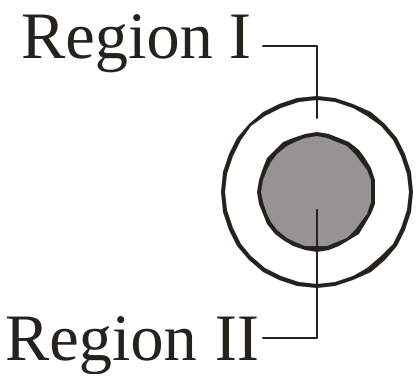


- Color Center Laser: ~1 MHz bandwidth
- Tone-burst modulation
- Autobalanced subtraction
- Sensitivity ~1×10^{-4}

Last year's unexpected spectrum…



Unusual crystal: 2 regions



Region I:

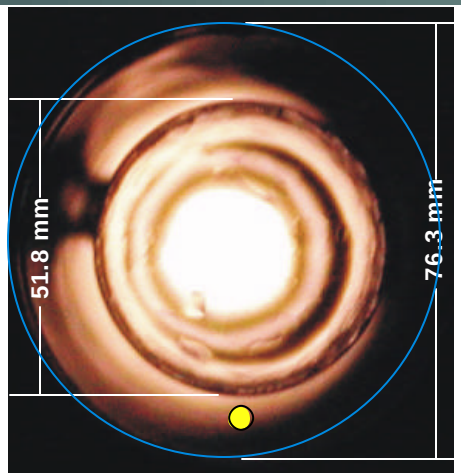
- Outermost region of cell
- ~2 mm thick
- No methane added
- 0.06 % ortho-H_2

Region II:

- Innermost region of cell
- ~16 mm diameter
- 80 ppm methane added
- 0.1 % ortho-H_2

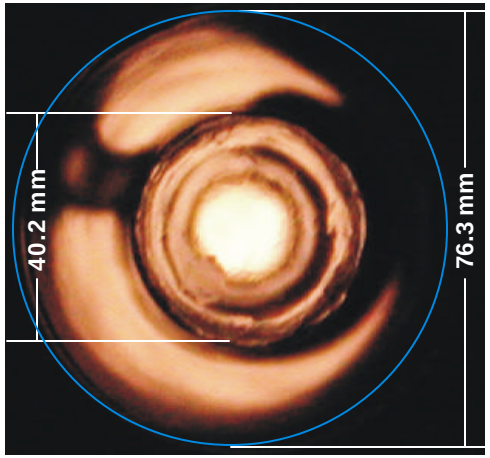
New spectrum appears in BOTH regions of crystal!

Trying to reproduce the spectrum…



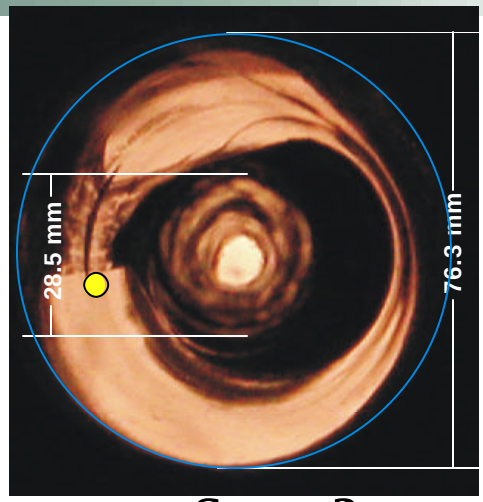
Stage 1:

p-H_2 + o-H_2 (0.03 %)



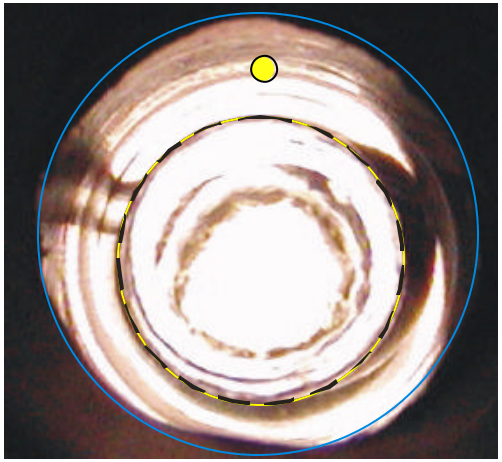
Stage 2:

p-H_2 + CH_4 (0.002 %) + o-H_2 (0.09%)



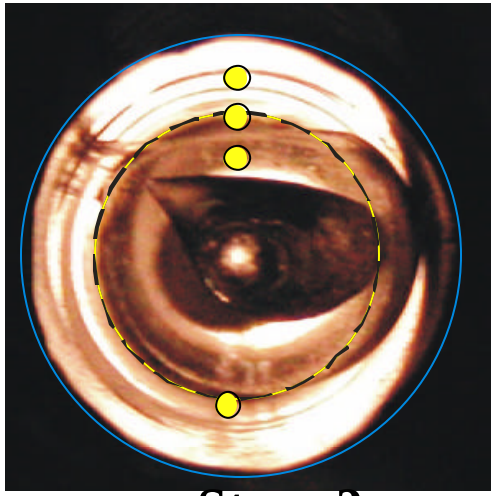
Stage 3:

p-H_2 + CH_4 (0.002 %) + o-H_2 (0.09%)



Stage 1:

p-H_2 + o-H_2 (0.2 %)

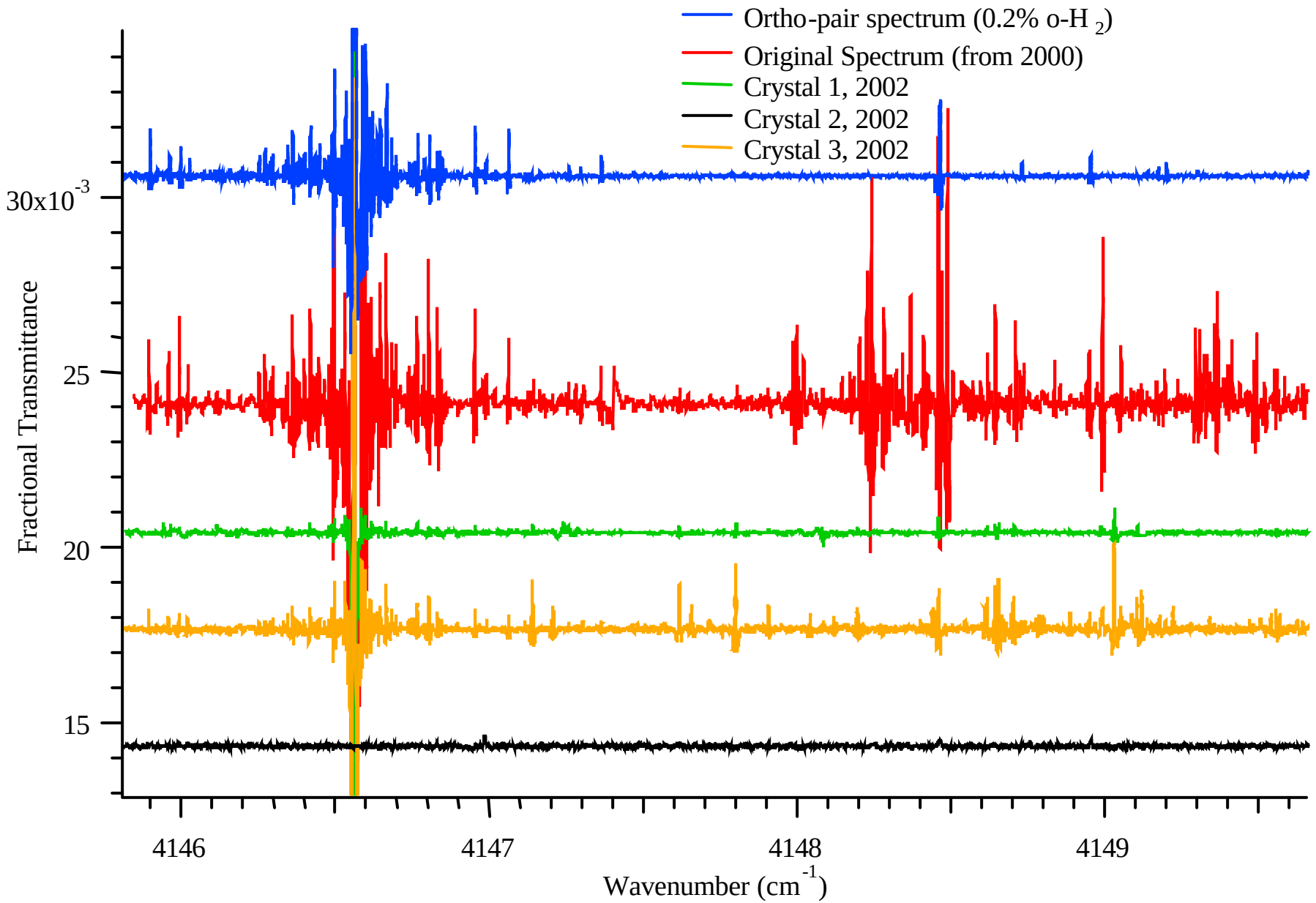


Stage 2:

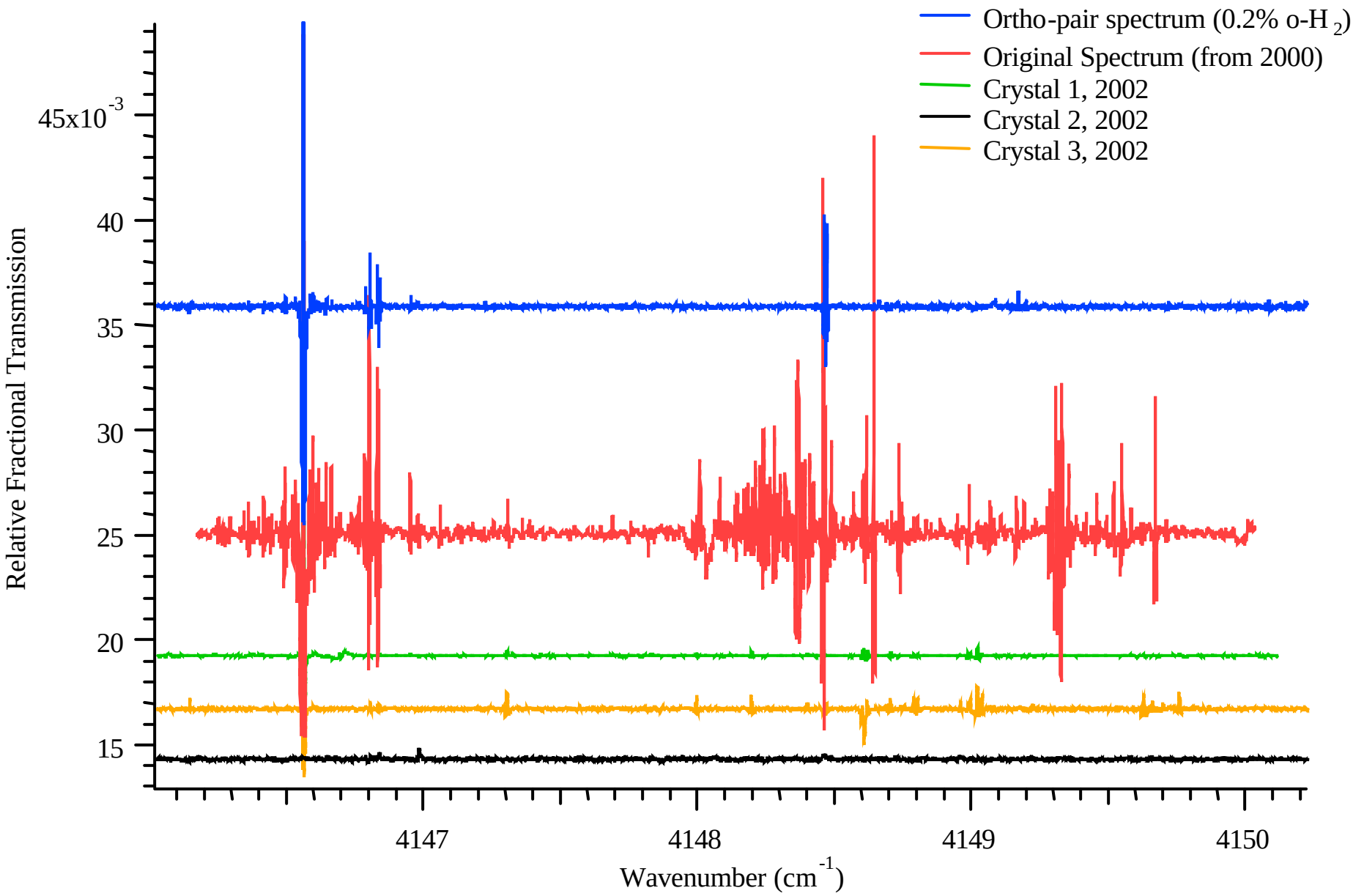
p-H_2 + CH_4 (0.02%) + o-H_2 (0.2 %)

Region probed by laser

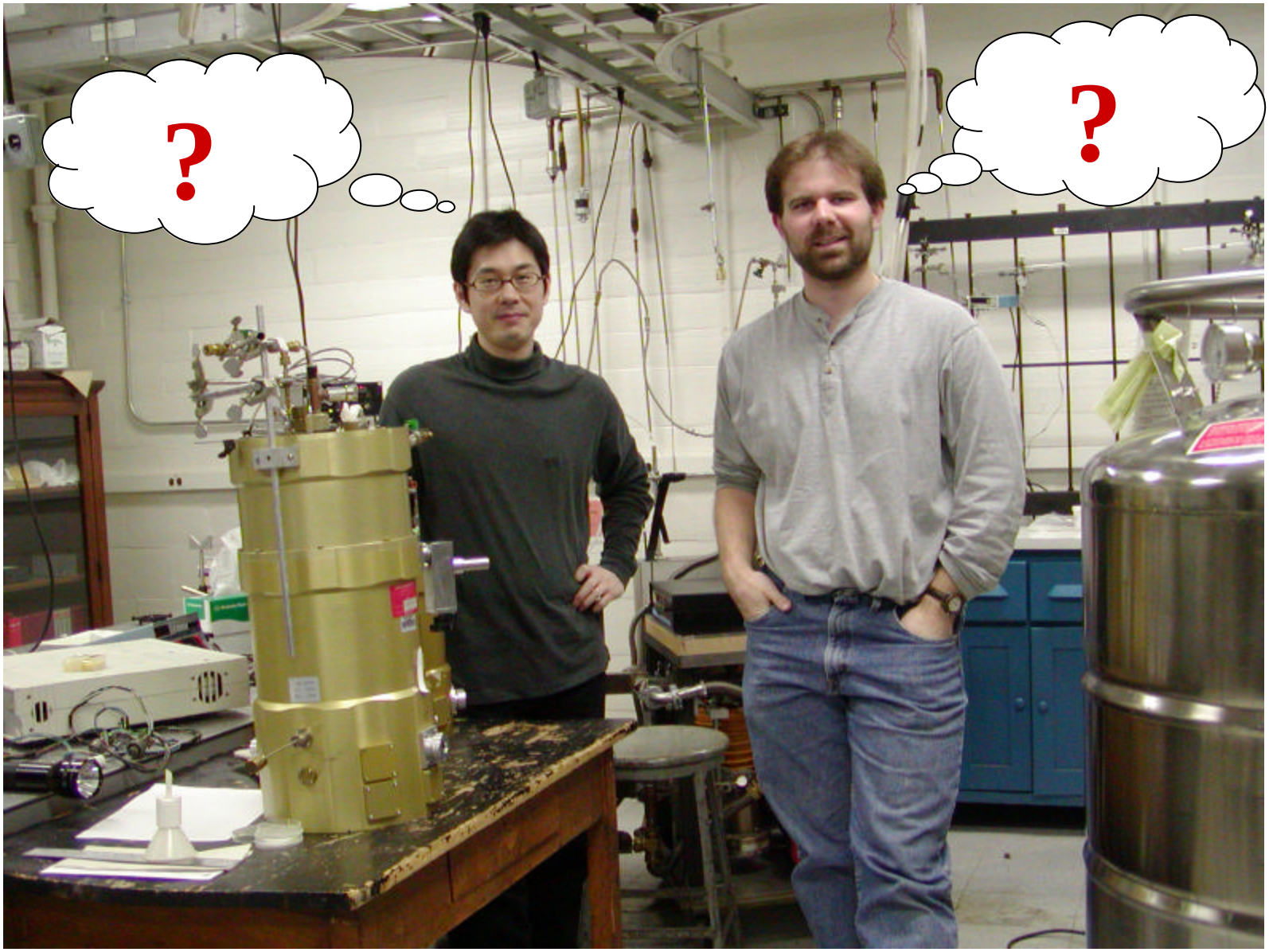
Summary of ‘best’ laser scans (perpendicular transitions)



Summary of ‘best’ laser scans (parallel transitions)



So, what is going on?!?!



So, what is going on?!?!

Last year's summary

?

- *p*-H_2 – CH_4
- *o*-H_2 – CH_4
- Different crystal structure
- Perturbed *o*-H_2-*o*-H_2 pairs
- ~~Band of CH_4~~

NEED MORE DATA!!!

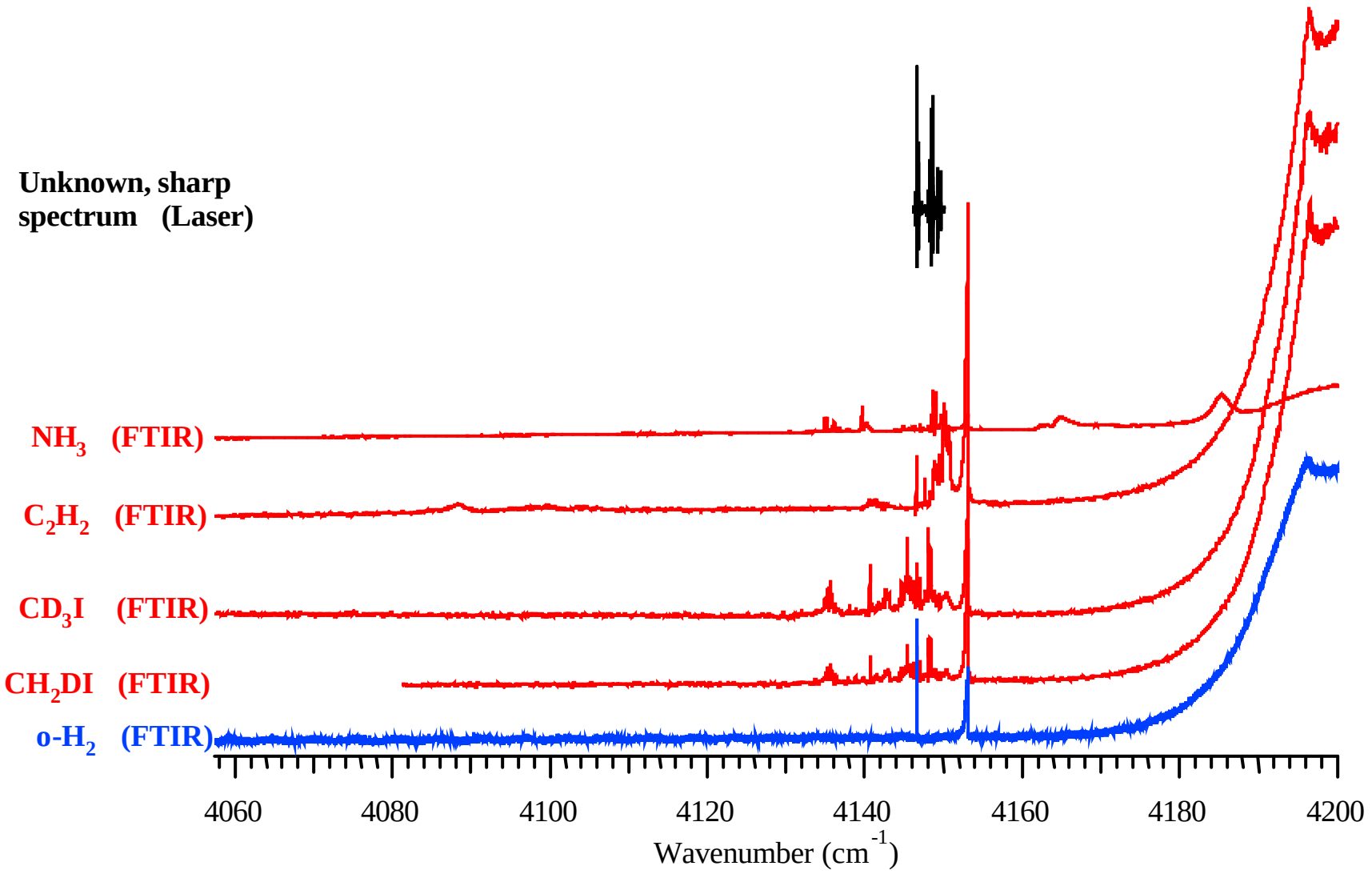
- **Does the spectrum depend on the "double crystal"?**
- **Is it due to only CH_4 or both CH_4/*o*-H_2?**
- **If it is due to CH_4, why are the lines larger in the region without CH_4?**

Summary

- **The sharp lines are due to a perturbation to the crystal by an impurity.**
  - **Shift in Q1(1) lines**
  - **Induced and shifted Q1(0) line**
- **We don't know what impurity is inducing the spectra in our data (but it is clearly not CH_4)**

  … Perhaps H_2O or air?
- **Lines are reproducible from crystal to crystal**
- **Crystal around the impurity is homogenous**

Induced infrared activity is a frequent occurrence



Induced infrared activity is a frequent occurrence