Observation of New Induced Sharp Transitions in *p*-H_2 Crystals doped with CH_4

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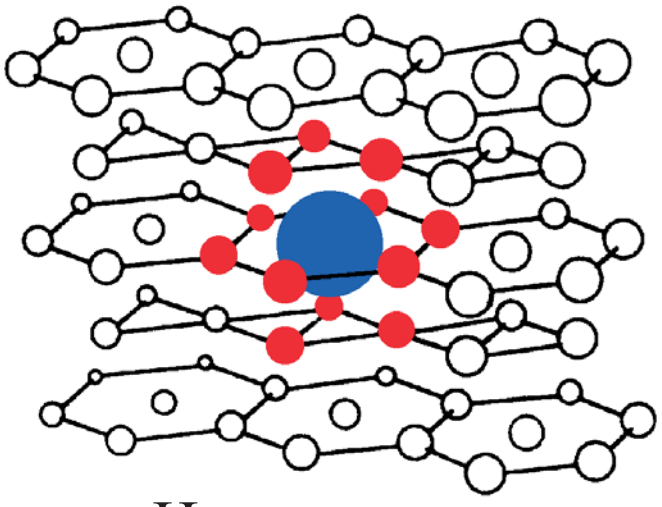
Solid Parahydrogen

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

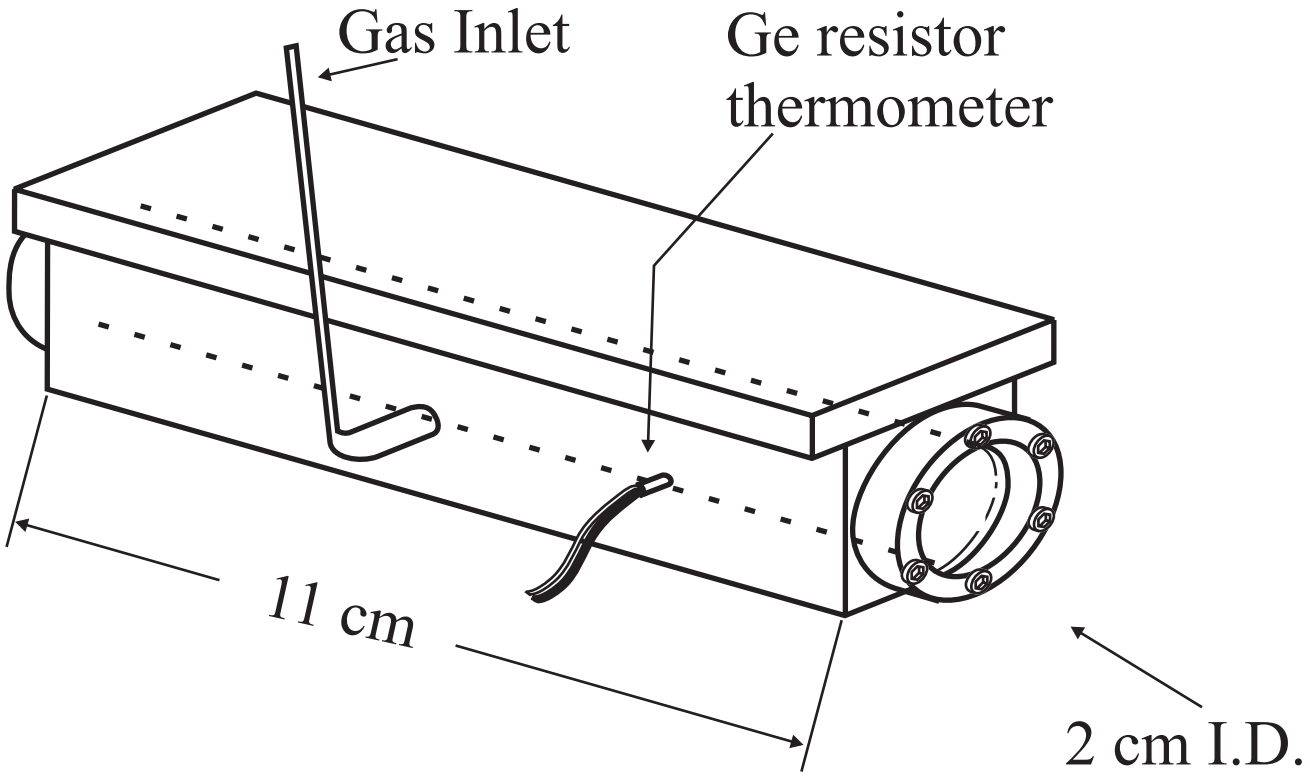


○ *p*-H_2
● nearest neighbor *p*-H_2
● methane

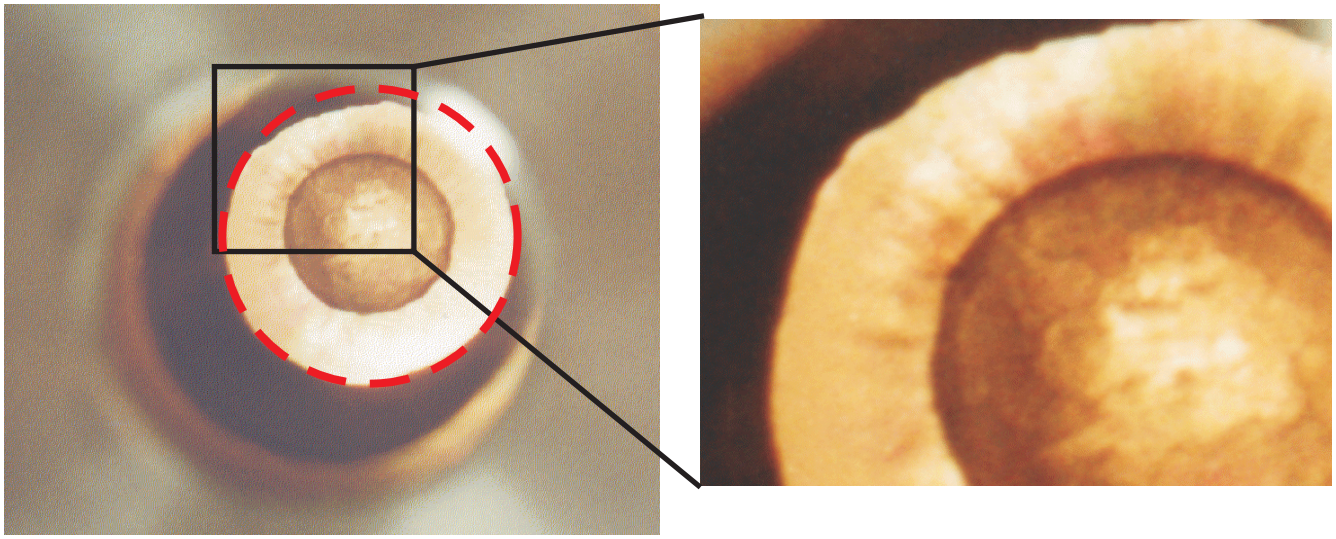
p-H_2 as a Matrix Medium

- 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

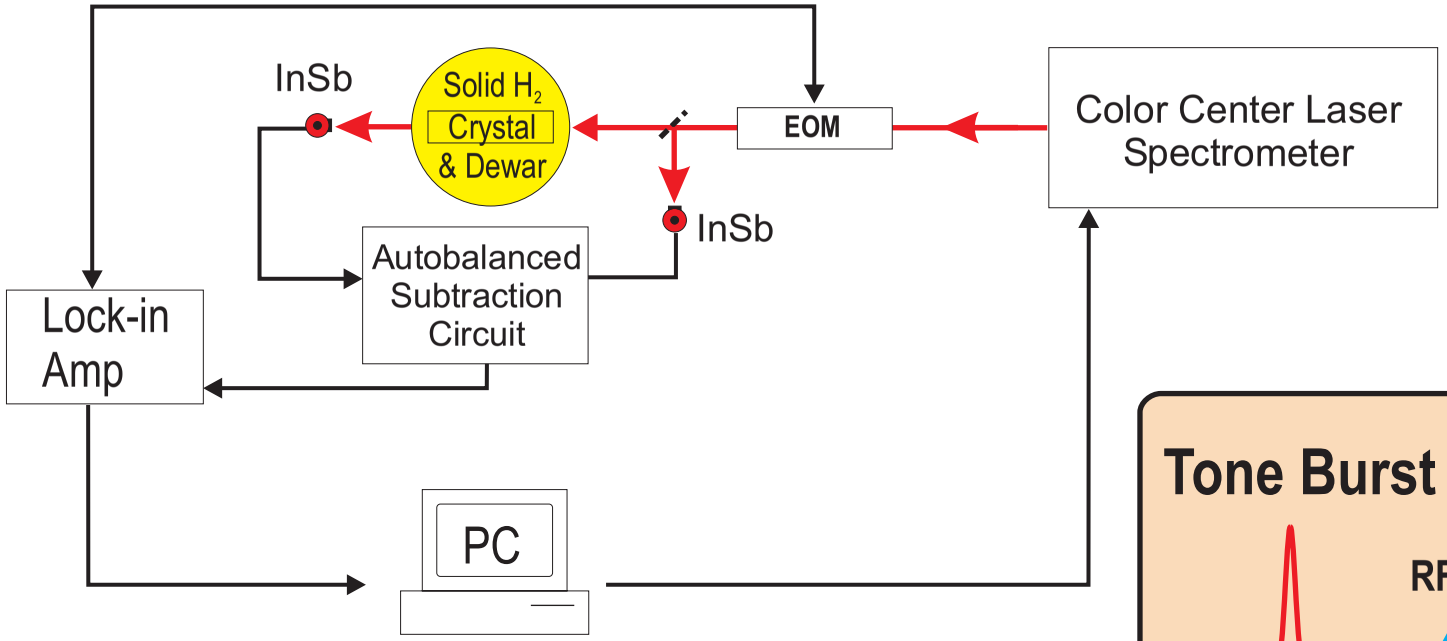
Production of *p*-H_2 Crystals



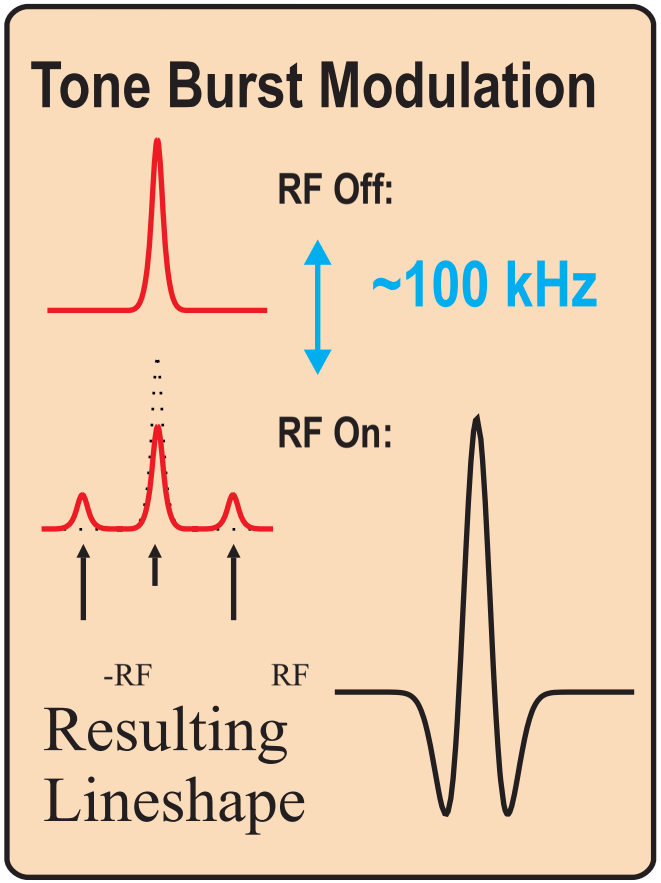
- Ultra high purity H_2 converted to ~99.9% *p*-H_2
- Cooled in a liquid He cryostat
- Lattice grown radially inward-- exclusively HCP



Absorption Spectroscopy



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

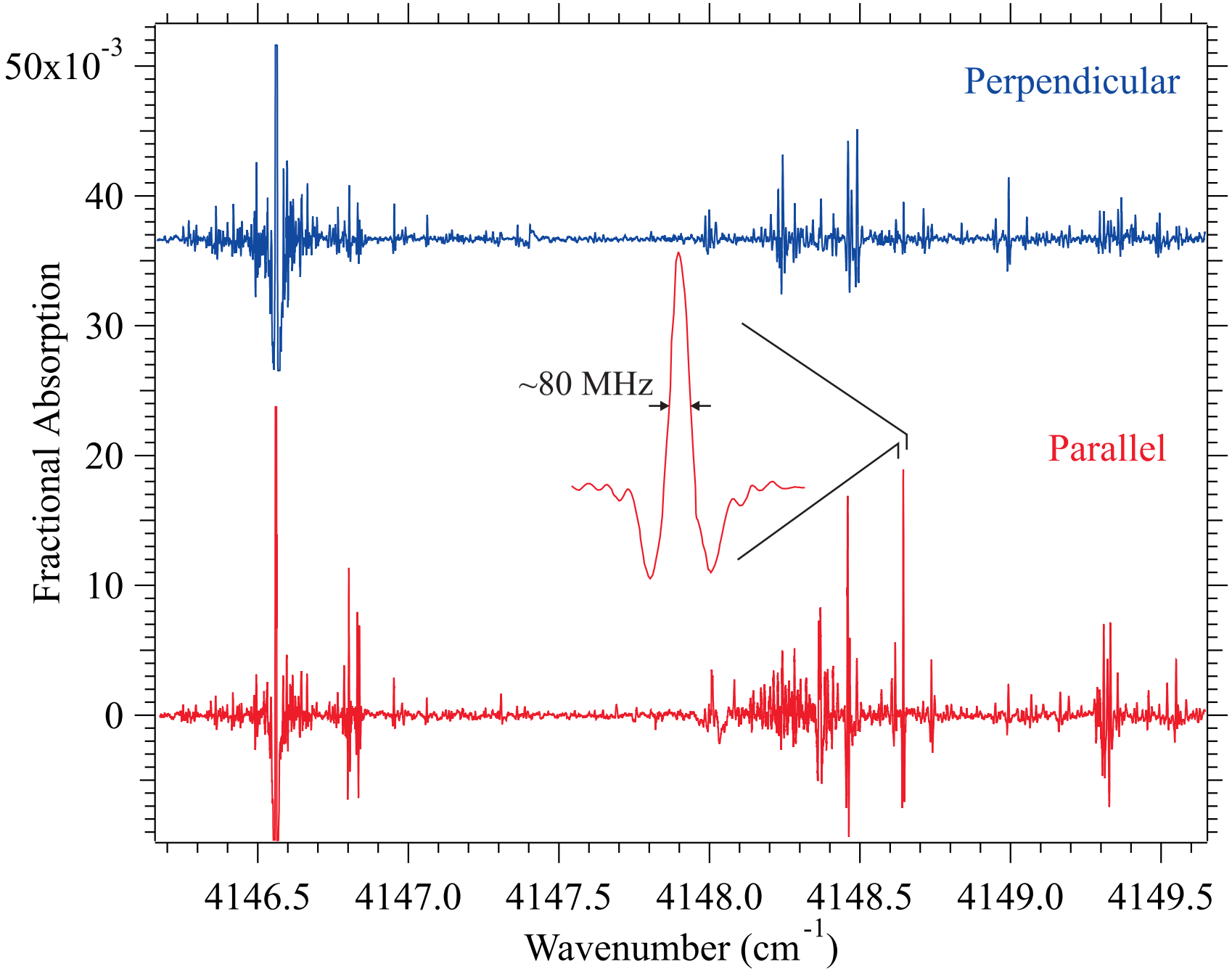


p-H_2 Crystals as a Matrix Medium

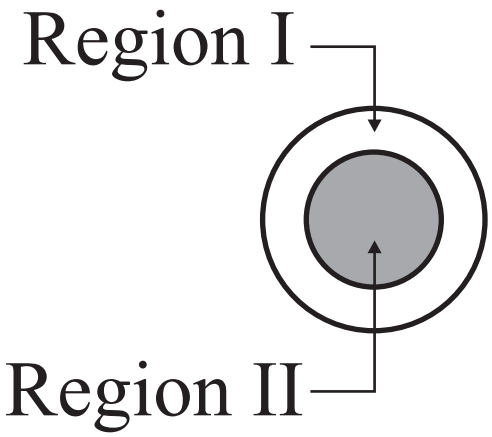
- *p*-H_2, *o*-H_2, HD, D_2
- CH_4, CD_4, CD_3H
- CH_3, CD_3
- $H_3^+(H_2)_n$, $H^-(H_2)_n$ → Momose *et al.* Phys. Rev. Lett. 86, 4795 (2001).
- *Ions of imbedded impurities*???

Let's try to irradiate *p*-H_2 doped with CH_4!

Unexpected Spectrum! *(Before irradiation)*



Unusual Crystal: Two "Regions"



Region I:

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

Region I:

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

New spectrum appears in BOTH regions of crystal!

IR Activity in Solid H_2

Isolated Molecular Transitions

- p-H_2 ✗ No electrostatic moment
- o-H_2 ✗ *Extremely* weak quadrupole
- CH_4 ✗ No bands in this region

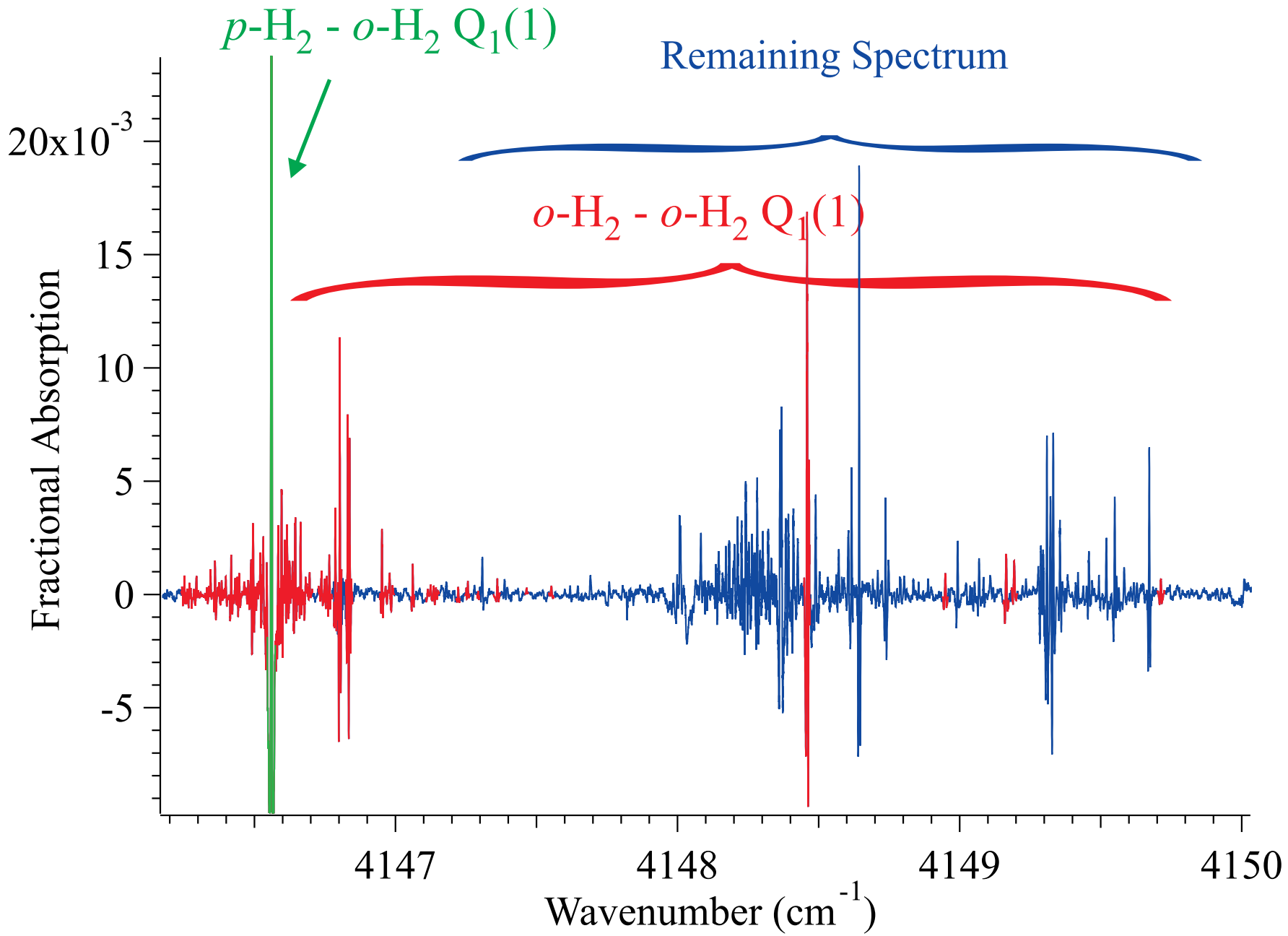
Pair Transitions

- p-H_2 - p-H_2 ✗ Double transition at ~8300 cm^{-1}
- p-H_2 - o-H_2 ✓ Quadrupole induced $Q_1(1)$ @ 4146.56 cm^{-1}
- p-H_2 - CH_4 ?

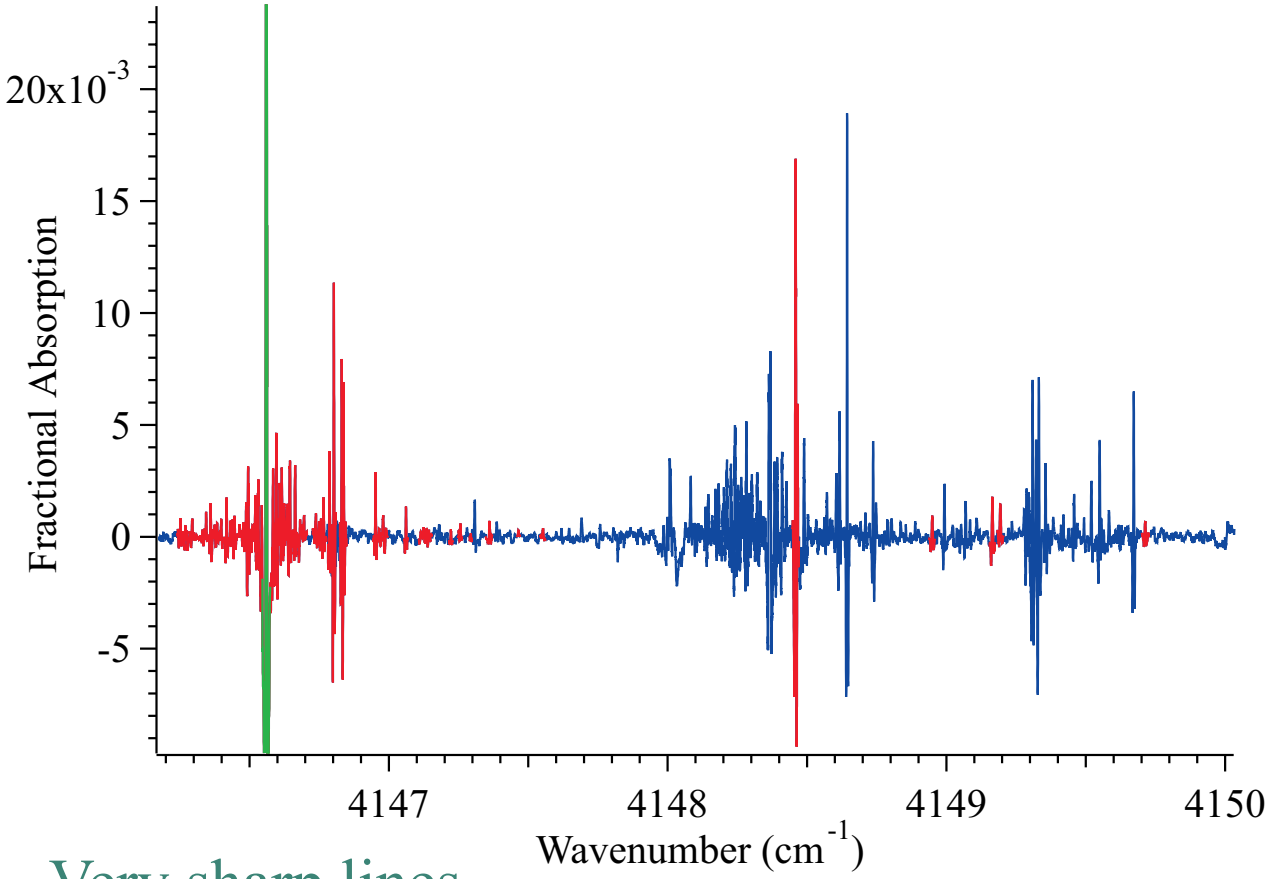
- o-H_2 - o-H_2 ✓ Pair spectrum in this region
- o-H_2 - CH_4 ?

- CH_4 - CH_4 ✗ Cluster bands should be very broad

Discussion of the Spectrum



Characteristics of the Spectrum



- Very sharp lines
- H_2 stretch region between $Q_1(1)$ and $Q_1(0)$
- Spectrum "similar" to *o*-H_2 pair spectrum
- Both perpendicular and parallel type transitions present
- Spectrum weaker in "methane" region
- Spectrum did not change upon -ray irradiation

And The Carrier is...

- *p*-H_2 – CH_4
- *o*-H_2 – CH_4
- Different crystal stucture
- 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?

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