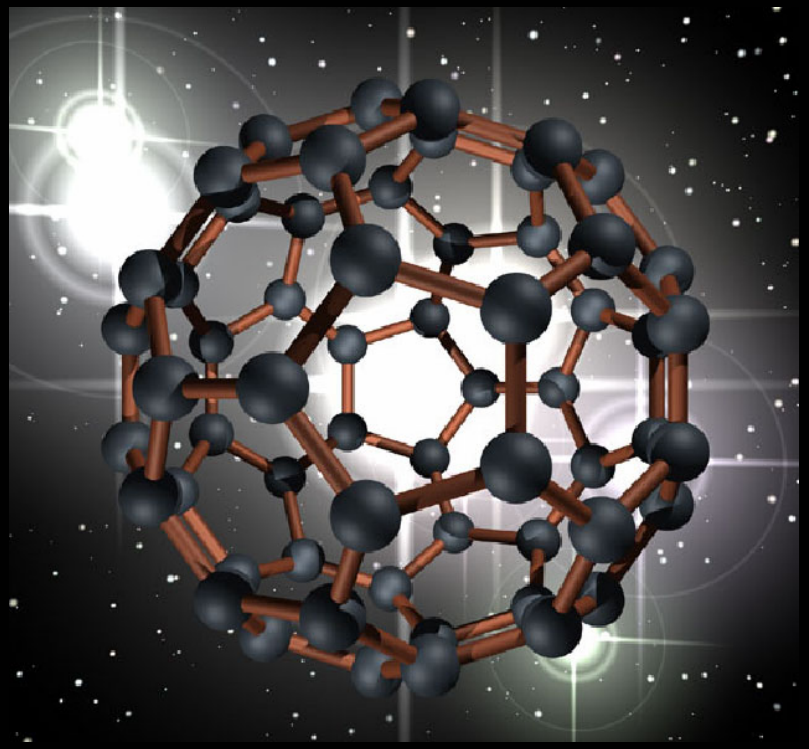
A Search for the 8.5 μm Vibrational Spectrum of C_{60} in the Laboratory and Space



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The discovery of C_{60}

C_{60}: Buckminsterfullerene

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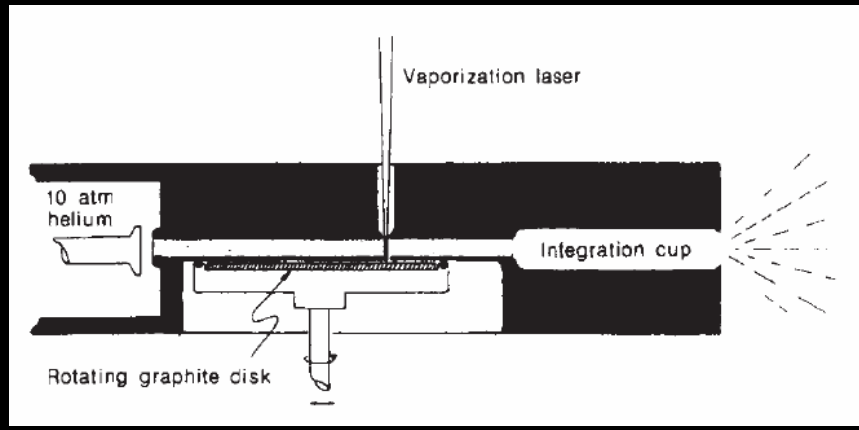
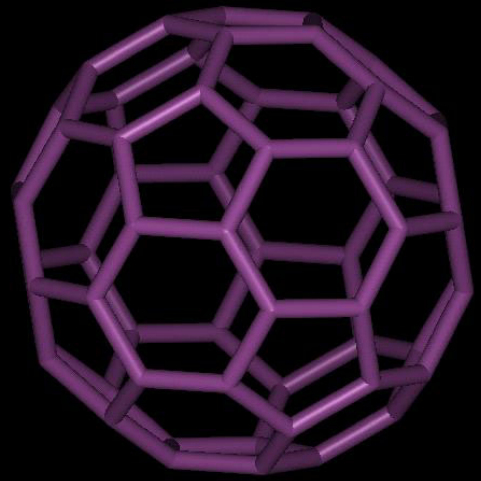
Rice Quantum Institute and Departments of Chemistry and Electrical Engineering, Rice University, Houston, Texas 77251, USA

During experiments aimed at understanding the mechanisms by which long-chain carbon molecules are formed in interstellar space and circumstellar shells[1], graphite has been vaporized by laser irradiation, producing a remarkably stable cluster consisting of 60 carbon atoms. Concerning the question of what kind of 60-carbon atom structure might give rise to a superstable species, we suggest a truncated icosahedron, a polygon with 60 vertices and 32 faces, 12 of which are pentagonal and 20 hexagonal. This object is commonly encountered as the football shown in Fig. 1. The C_{60} molecule which results when a carbon atom is placed at each vertex of this structure has all valences satisfied by two single bonds and one double bond, has many resonance structures, and appears to be aromatic.

Kroto et al., Nature 318, 162 (1985)

Laboratory experiments designed to simulate carbon star outflows

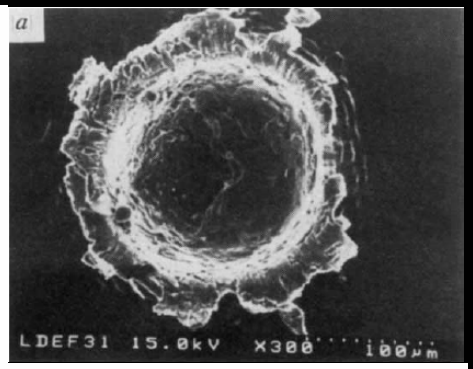


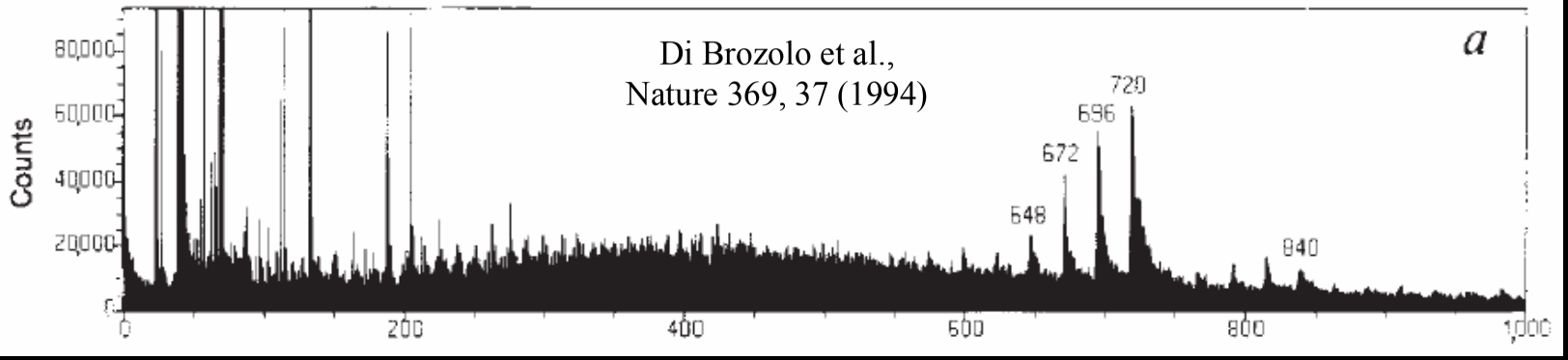


C_{60} in space?

Fullerenes in an impact crater on the LDEF spacecraft

Filippo Radicati di Brozolo*, Theodore E. Bunch†, Ronald H. Fleming* & John Macklin‡

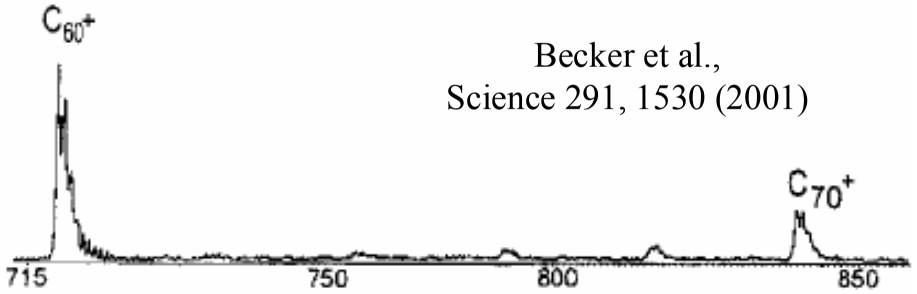




Di Brozolo et al.,
Nature 369, 37 (1994)

Impact Event at the Permian-Triassic Boundary: Evidence from Extraterrestrial Noble Gases in Fullerenes

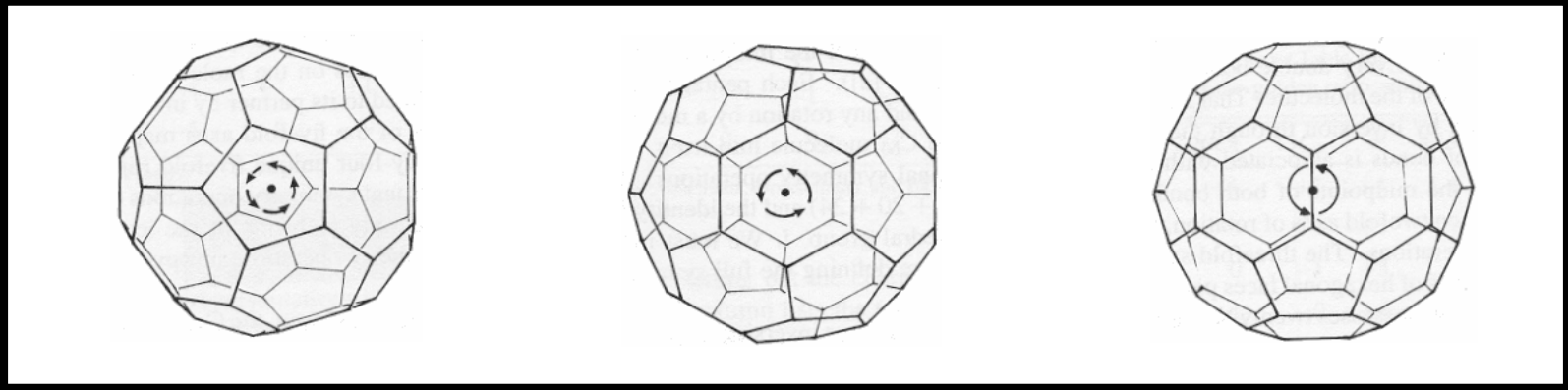
Luann Becker,[1]* Robert J. Poreda,[2] Andrew G. Hunt,[2]
Theodore E. Bunch,[3] Michael Rampino[4]



Becker et al.,
Science 291, 1530 (2001)

About C_{60}

- 3(60)-6 = 174 vibrational degrees of freedom
- Sixty quantum-mechanically indistinguishable (spin 0) bosons
- Icosahedral (I_h) Symmetry: 6 five-fold axes, 10 three-fold axes, 15 two-fold axes

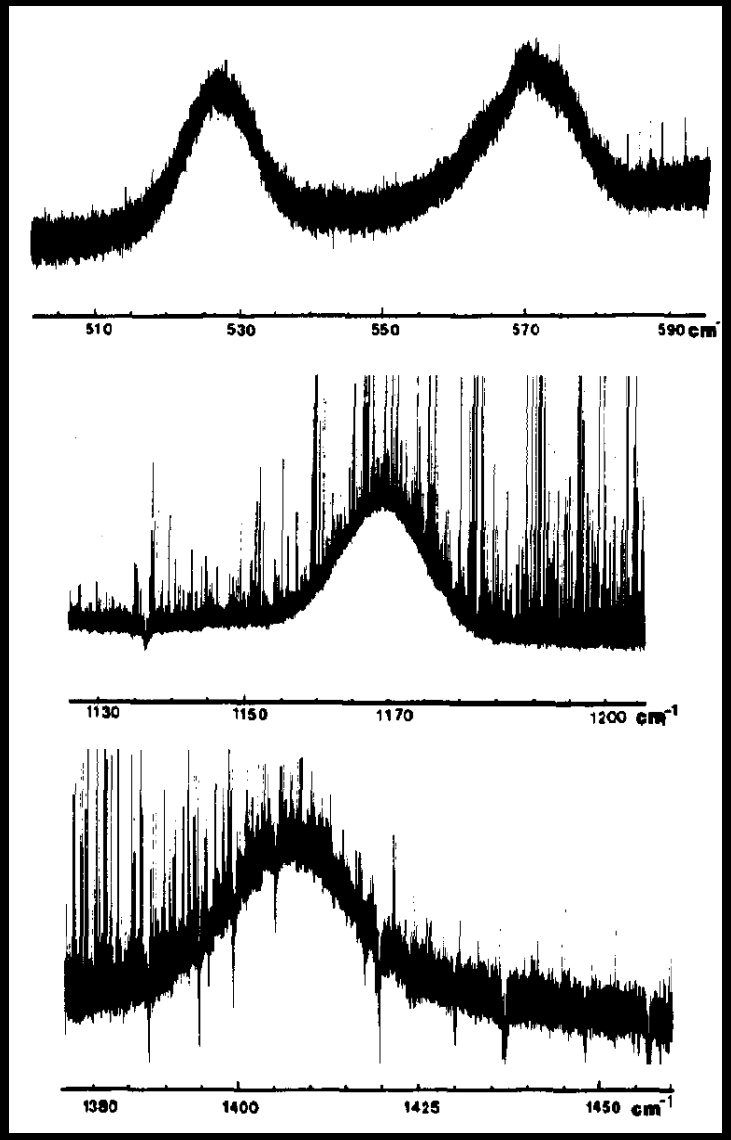


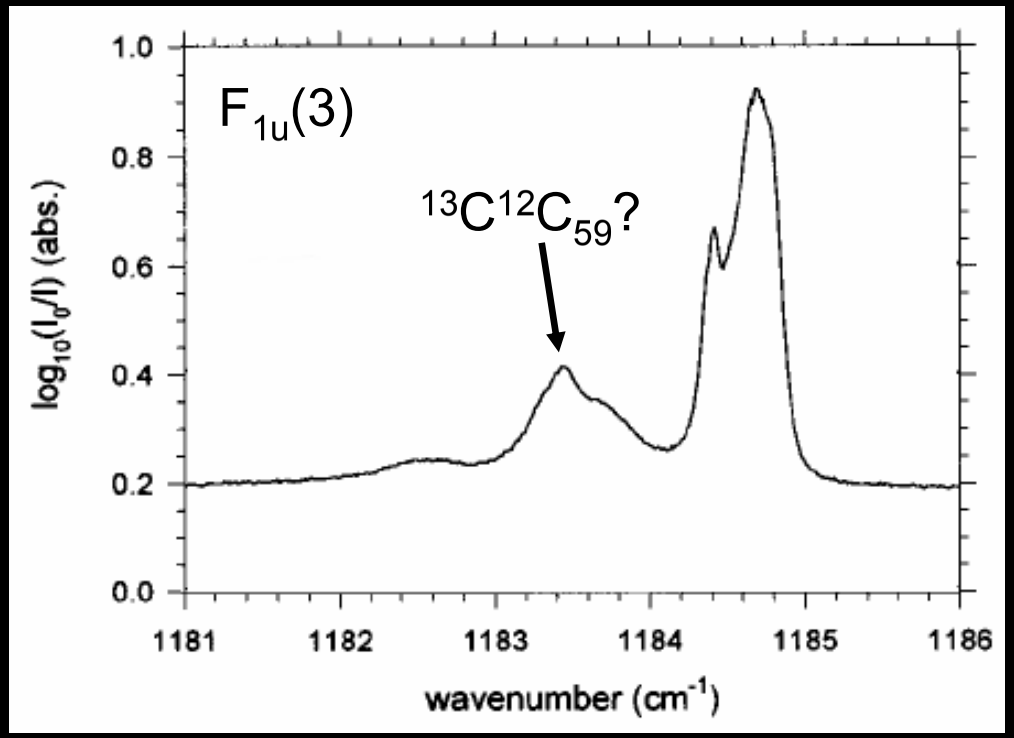
- Symmetry restrictions on total wavefunction
- 4 F_{1u} IR active modes

Previous laboratory studies of C_{60}

Gas phase IR emission spectrum observed at 1065 K; no rotational structure resolved

Frum et al. Chem. Phys. Lett. 176, 1991





IR spectrum observed in *p*-H_2 matrix

Sogoshi et al. J. Phys. Chem. 104, 2000

A rotationally cold, resolved, gas phase C_{60} spectrum is needed to guide observational searches!

What do we need?

- Gas phase C_{60}

  High temperature oven (>600 ºC)

- Vibrationally and rotationally cold C_{60}

  Supersonic expansion source

- Rotational resolution

  Supersonic source

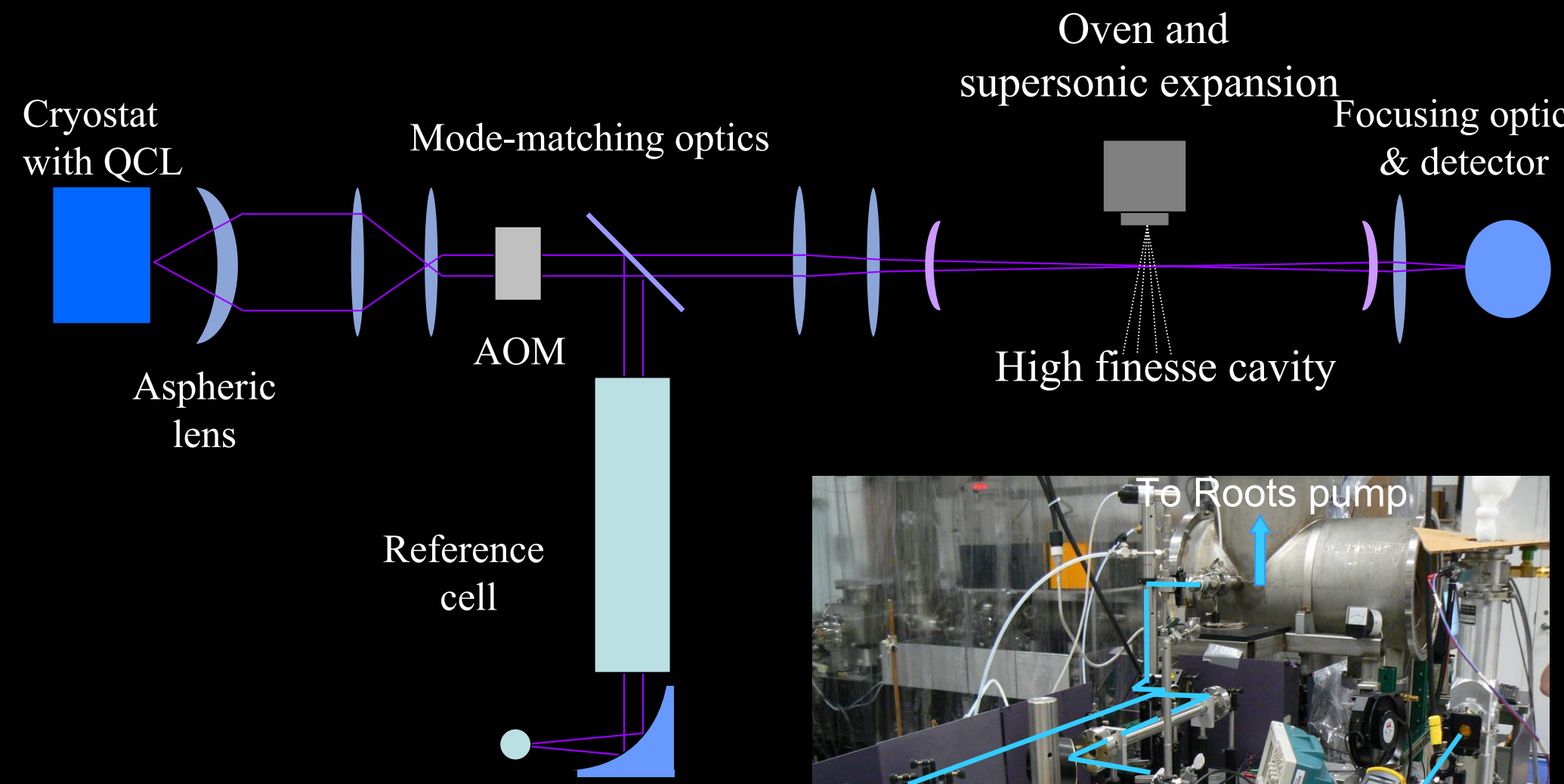
- Sensitivity

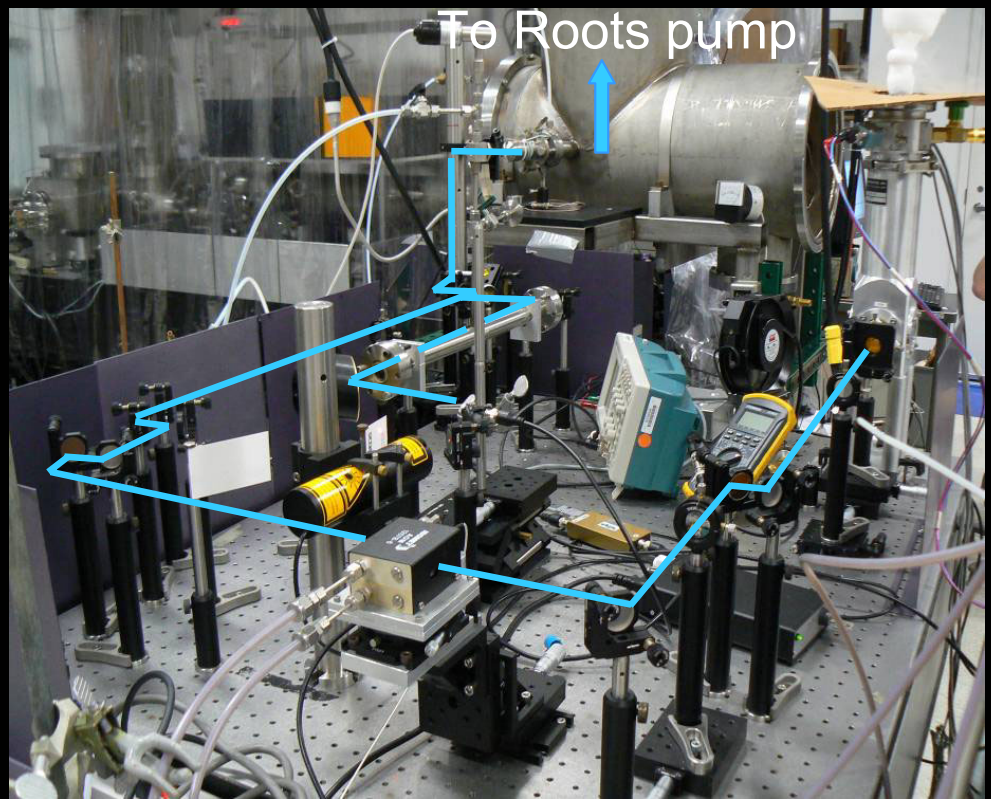
  Continuous-wave cavity ringdown spectroscopy (cw-CRDS)

- Tunability at 1184 cm^{-1}

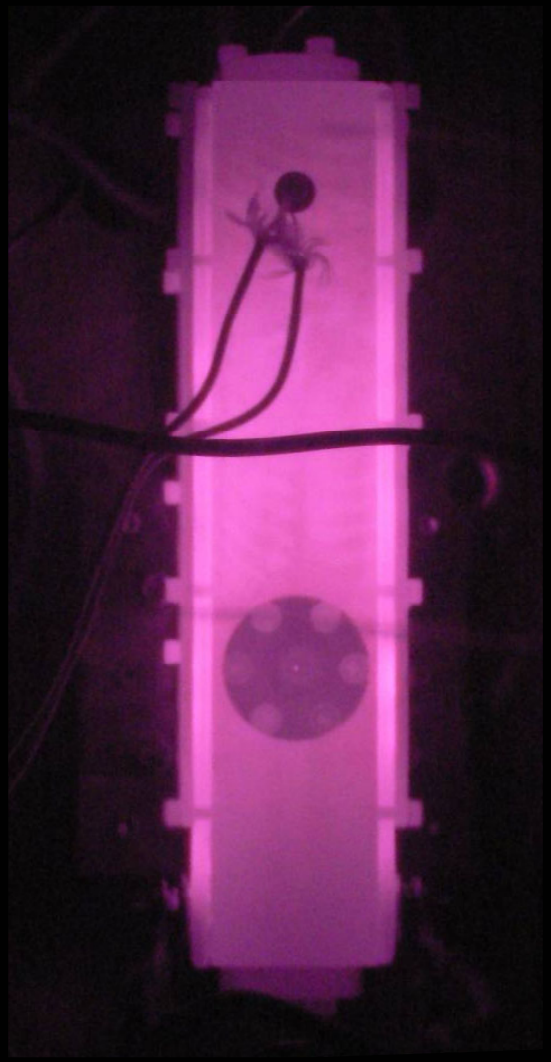
  Continuous-wave quantum cascade laser (cw-QCL)

Experimental Setup

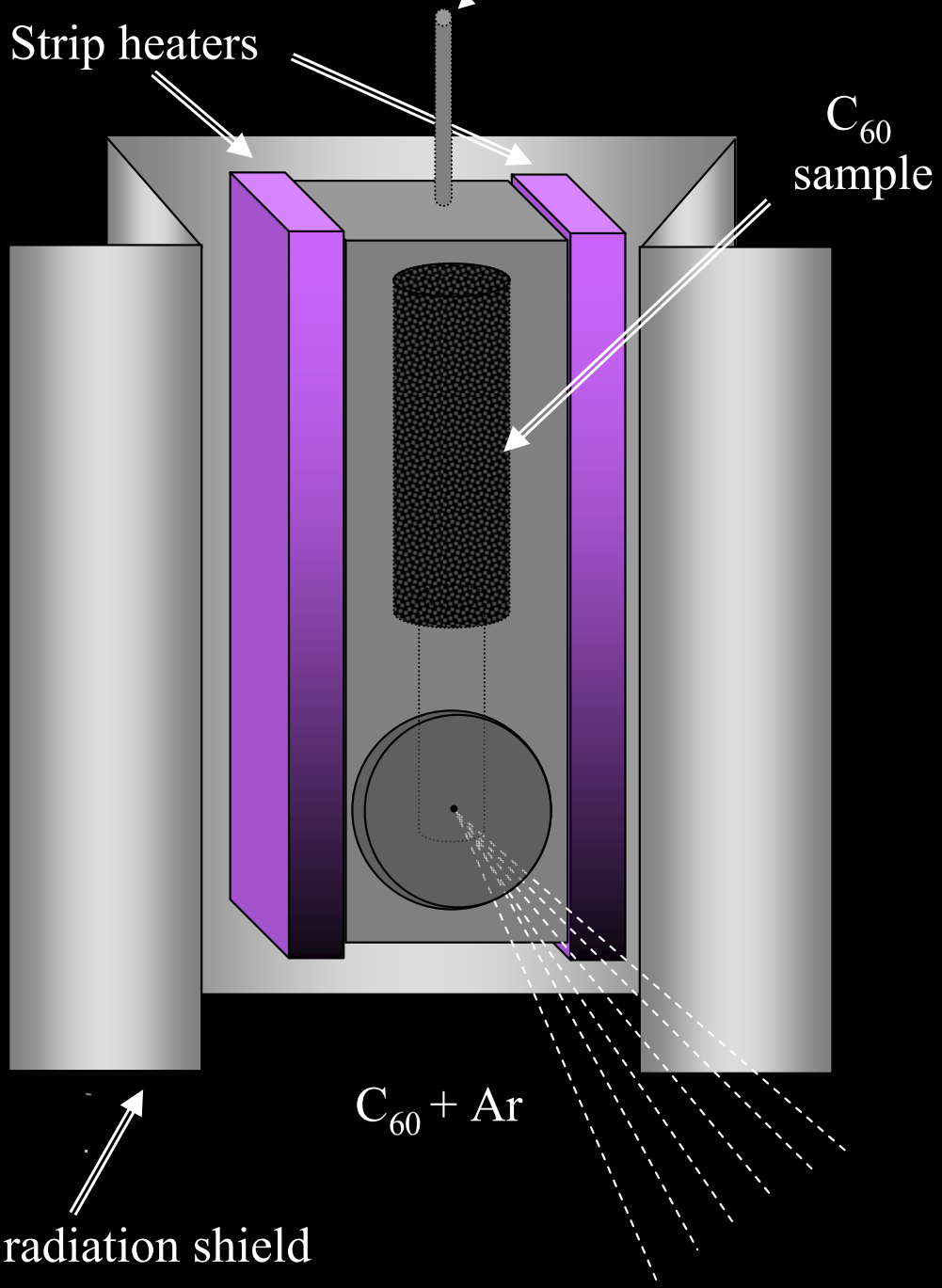




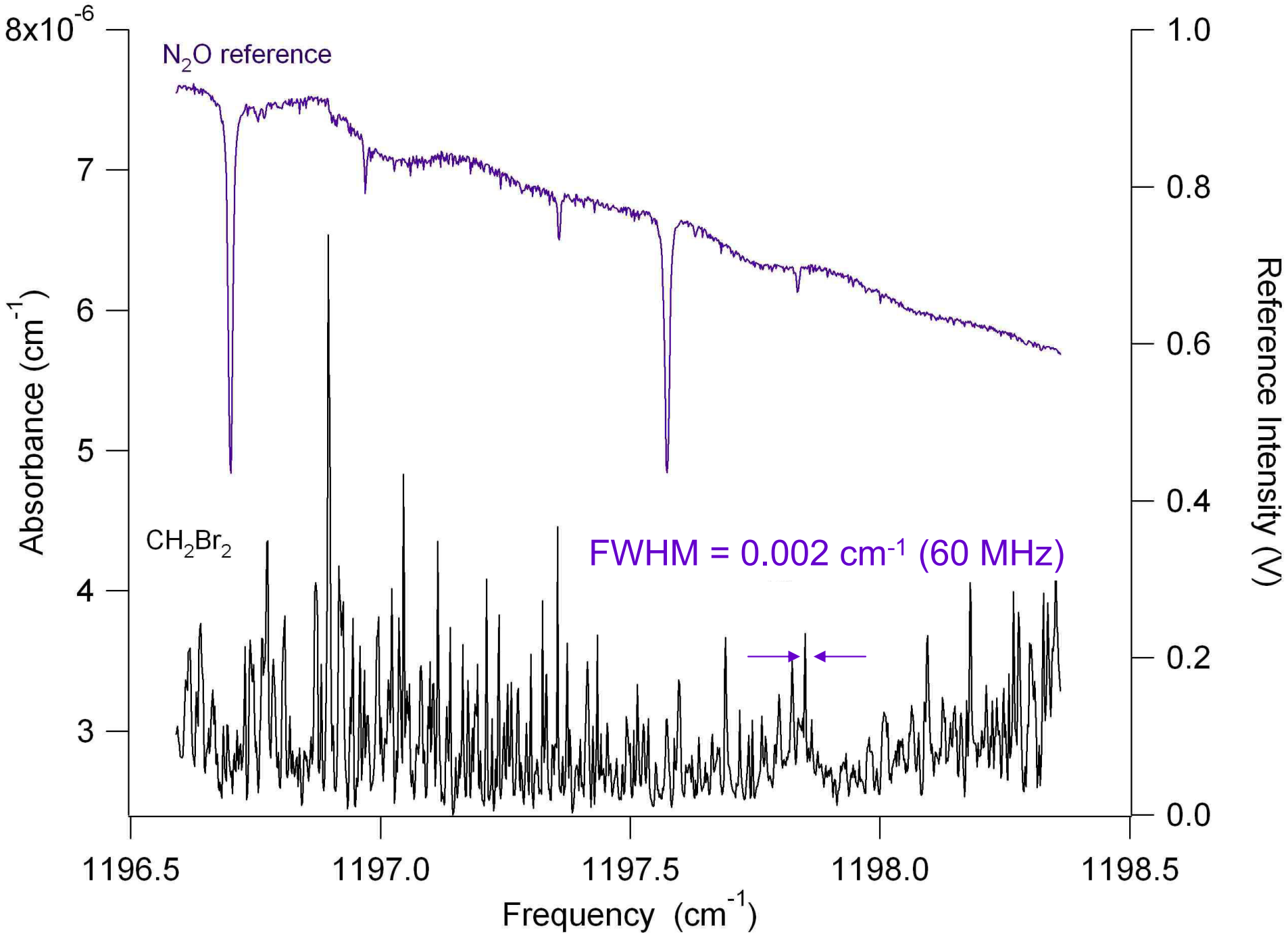
C_{60} Oven



T > 600 ºC!

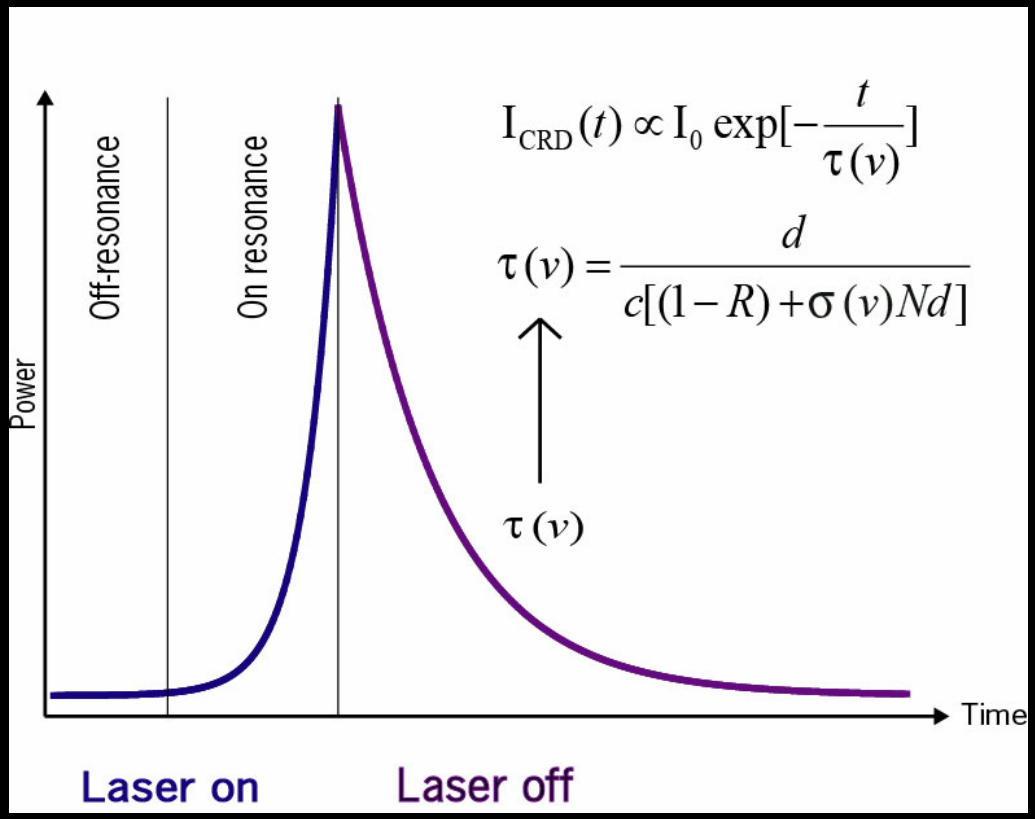


Supersonic Expansion



CW Cavity Ringdown Spectroscopy (cw-CRDS)

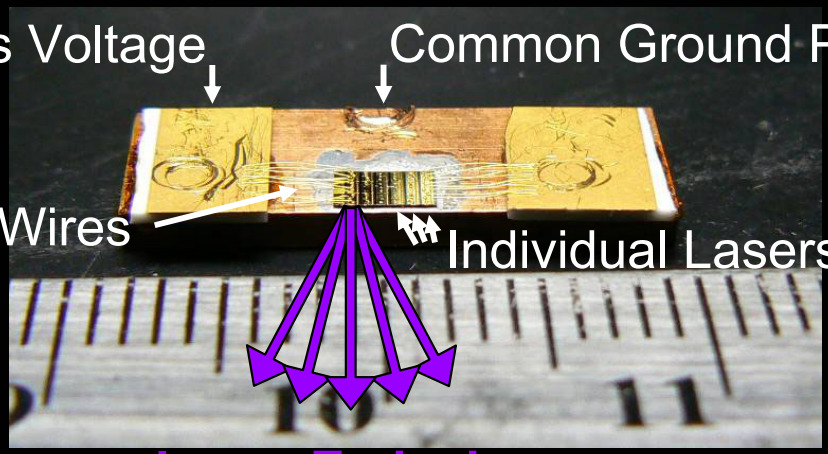
- A high finesse cavity is placed around the supersonic expansion.
- Laser light is coupled into the cavity, which is cycled in and out of resonance.

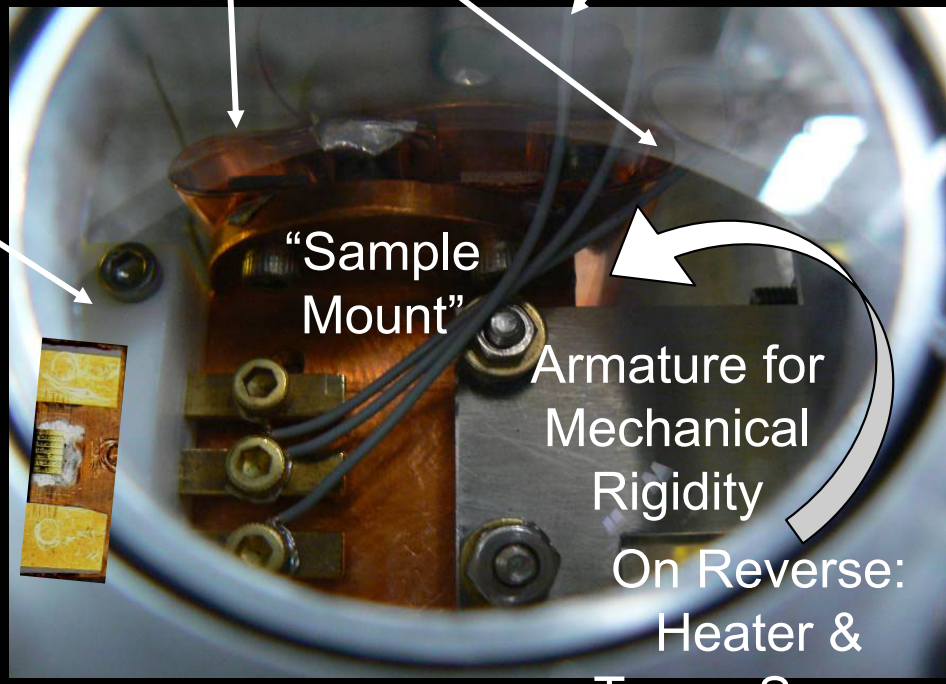


- When the cavity is on resonance the laser light is diverted or switched off.
- The exponential decay rate is a direct measurement of absorption.

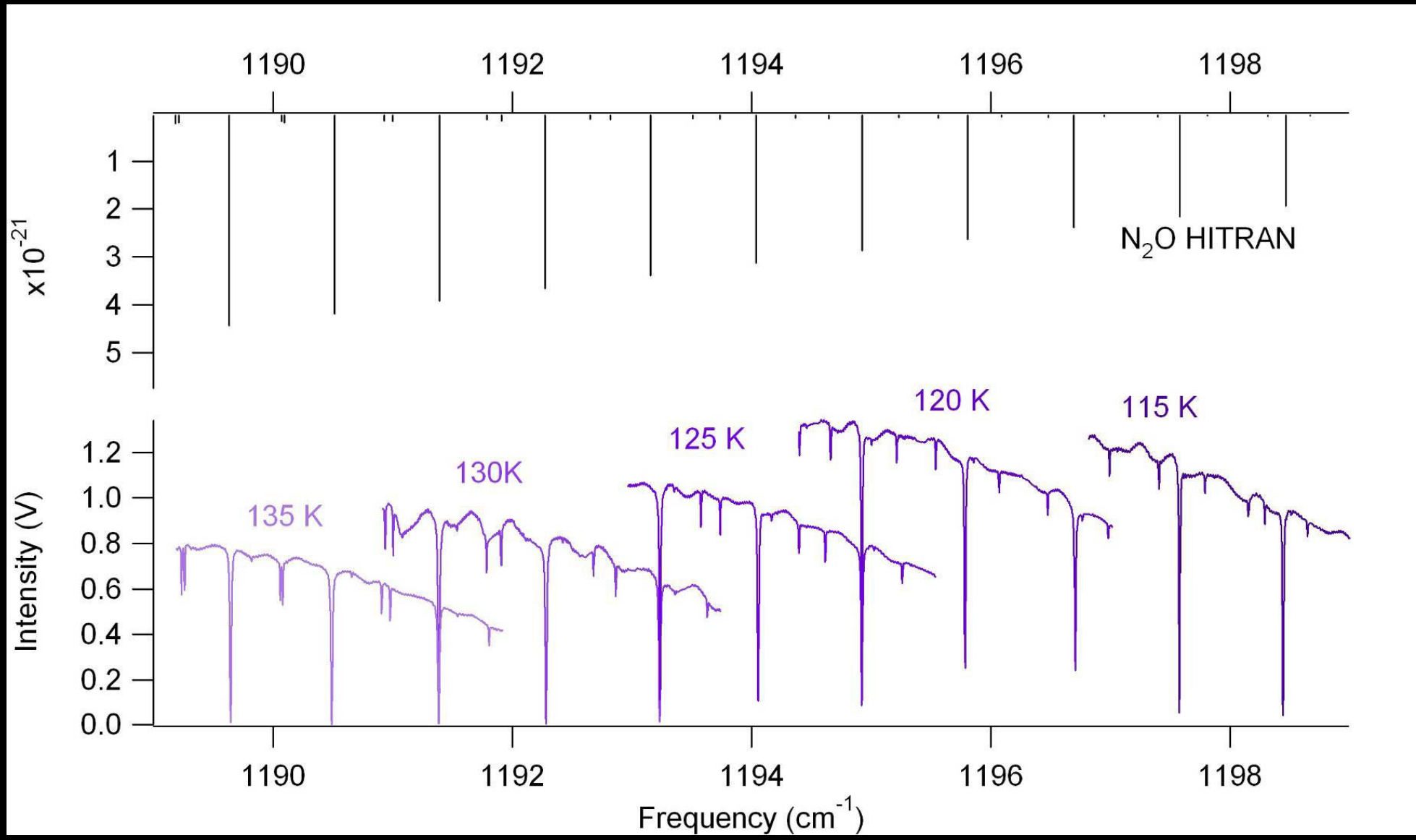
QCLs from the Gmachl Group

Janis VPF-100





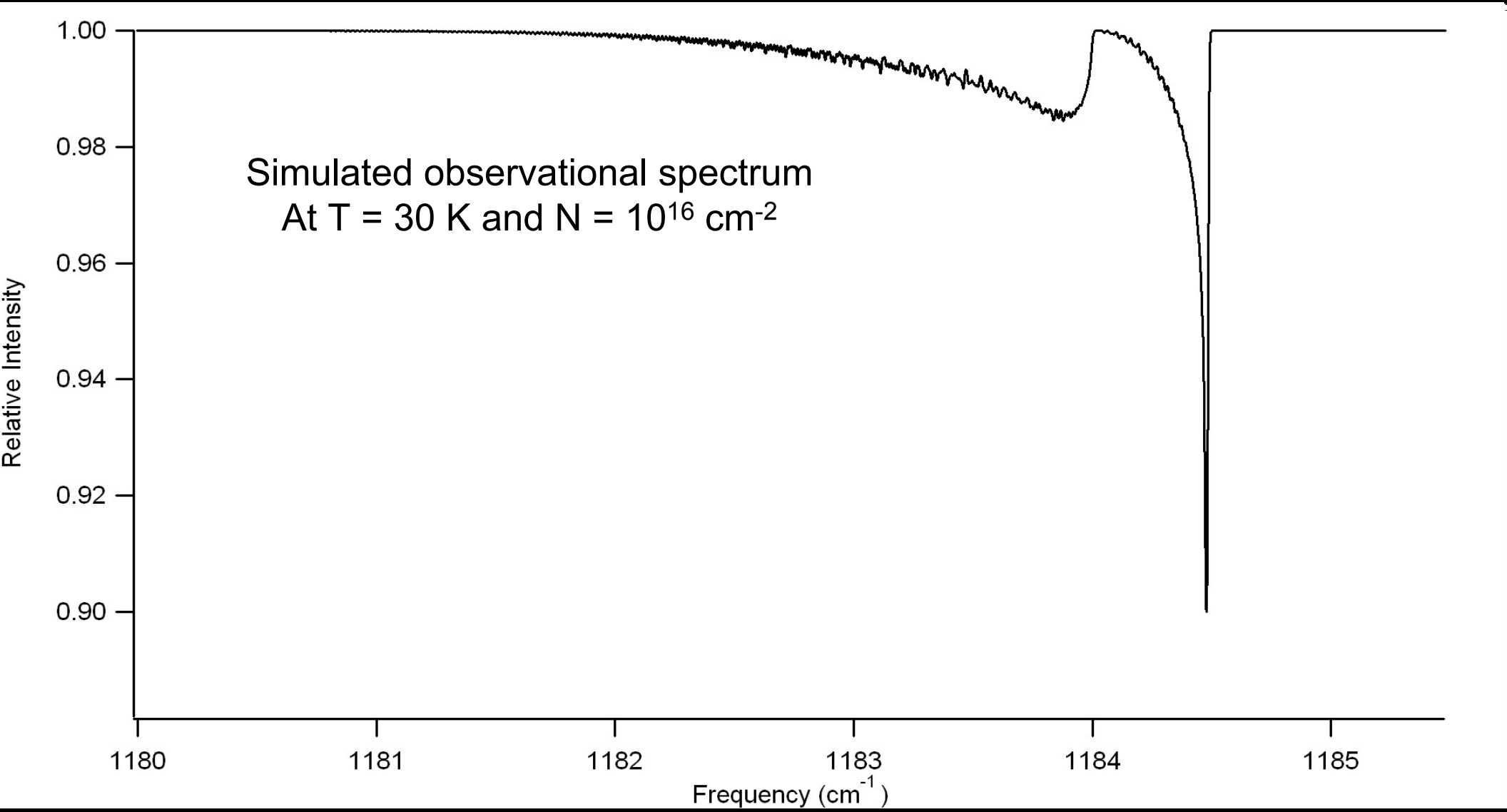
QCL Scanning



Fine tuning with current ~ 2 cm^{-1}

Coarse tuning with temperature ~10 cm^{-1}

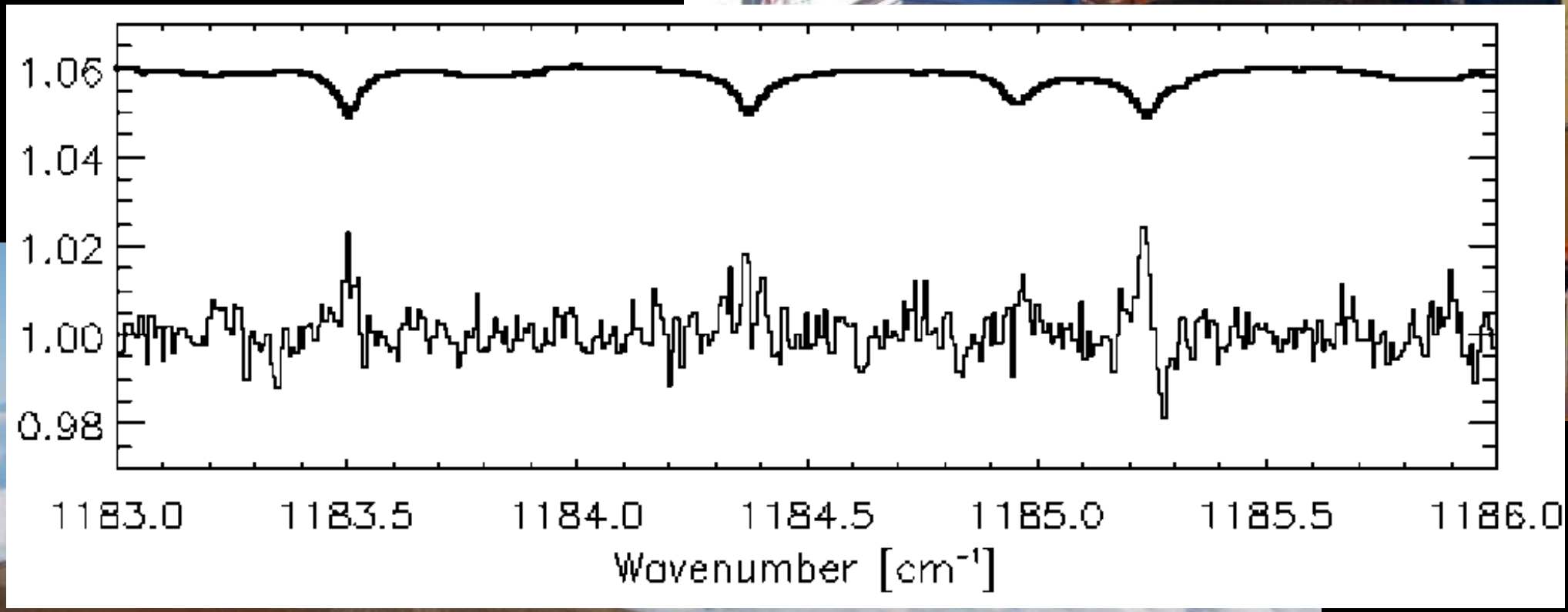
What will the C_{60} band look like?



Astronomical Search

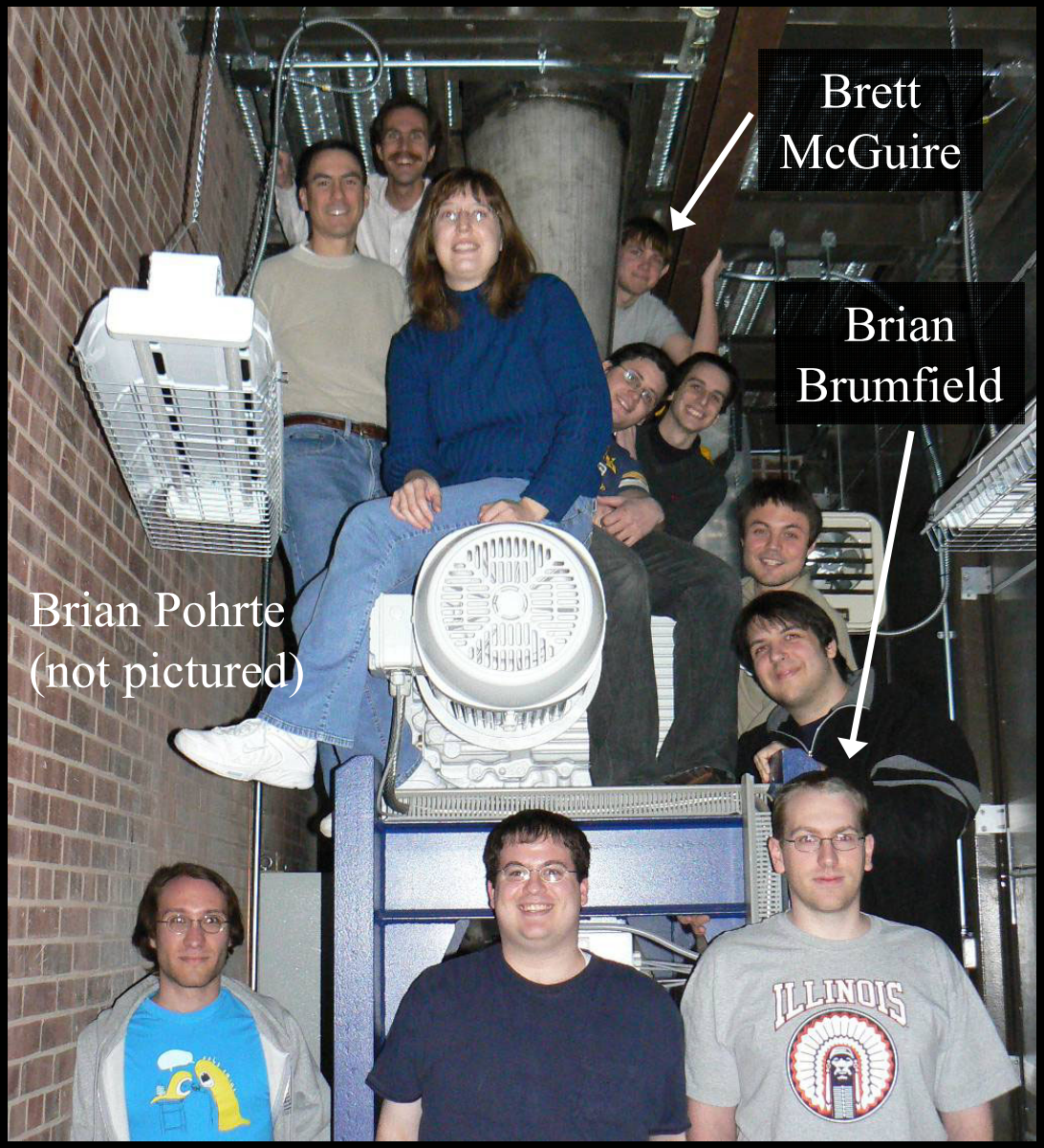
"Blind" upper limit

- ~3×10^{15} cm^{-2}
- < 0.6% of carbon



NASA's 3-meter IRTF (InfraRed Telescope Facility), Mauna Kea, Hawaii

Acknowledgments



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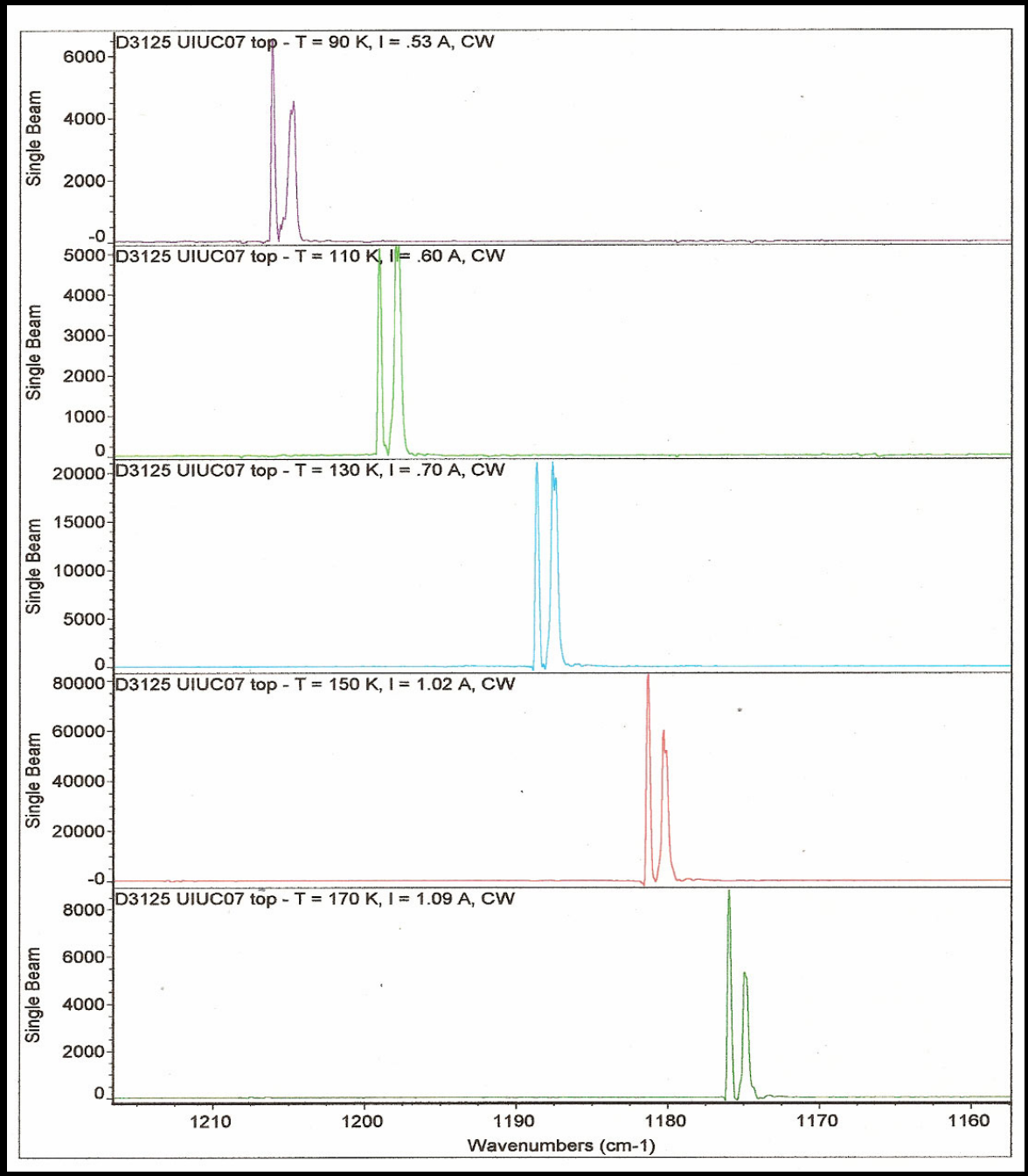
Laboratory
Astrophysics

QCL Scanning Difficulties

Some QCLs are inherently multi-mode.

Electronic chopping and back-reflection cause mode hops.

Solutions:

- Single-mode laser
- Acousto-optical modulator (AOM)
- Optical isolator