Towards Acquisition of A High Resolution Gas Phase Spectrum of C_{60}

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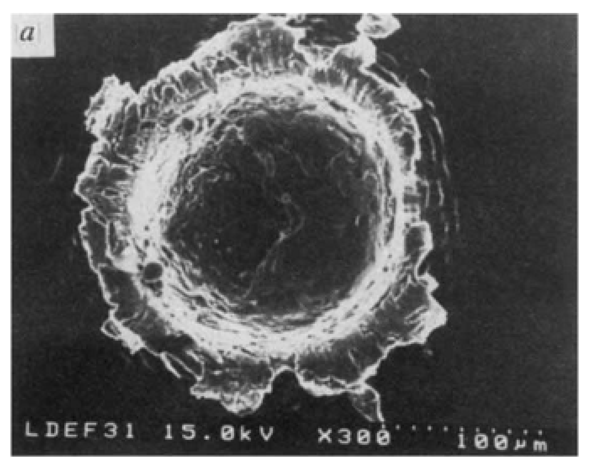
Background

➢C_{60} was discovered in 1985 during experiments trying to understand the chemistry that could lead to long carbon chain molecules in carbon-rich stellar outflows[1]

➢Since its discovery, C_{60} has been found:

*Embedded in impact craters on the Long Duration Exposure Facility (an earth orbiting satellite)[2]



*Discovered in sediments related to meteorite impacts[3]

➢Potential evidence for an extraterrestrial source of C_{60}, coupled to its stability to photofragment[4], provide an argument for its presence in circumstellar shells of carbon-rich stars and in the ISM.

➢Unfortunately, no rotationally resolved cold gas phase absorption spectrum of C_{60} is available for comparison to interstellar observations.

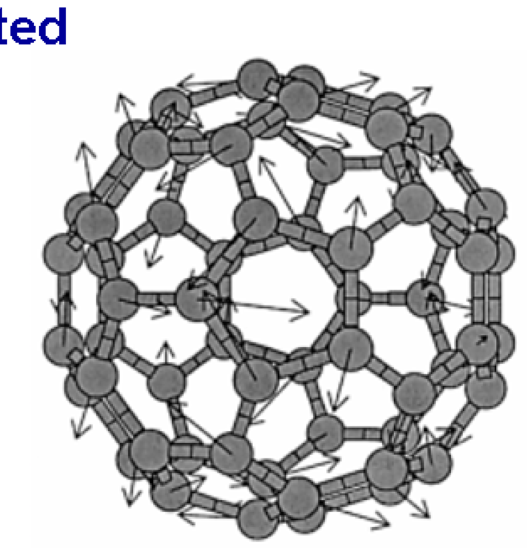
Where To Look?

➢Electronic bands are broad and/or forbidden (weak).

➢Rotational spectroscopy is forbidden due to lack of a permanent dipole moment.

➢Vibrational spectroscopy of C_{60} is the only reasonable remaining choice. C_{60} has 174 vibrational degrees of freedom, but symmetry restrictions confine an experiment to looking at only 4 F_{1U} IR active modes near 1432, 1185, 577, and 528 cm^{-1}.

➢The 1185 cm^{-1} band has been selected as a target due to two primary constraints:

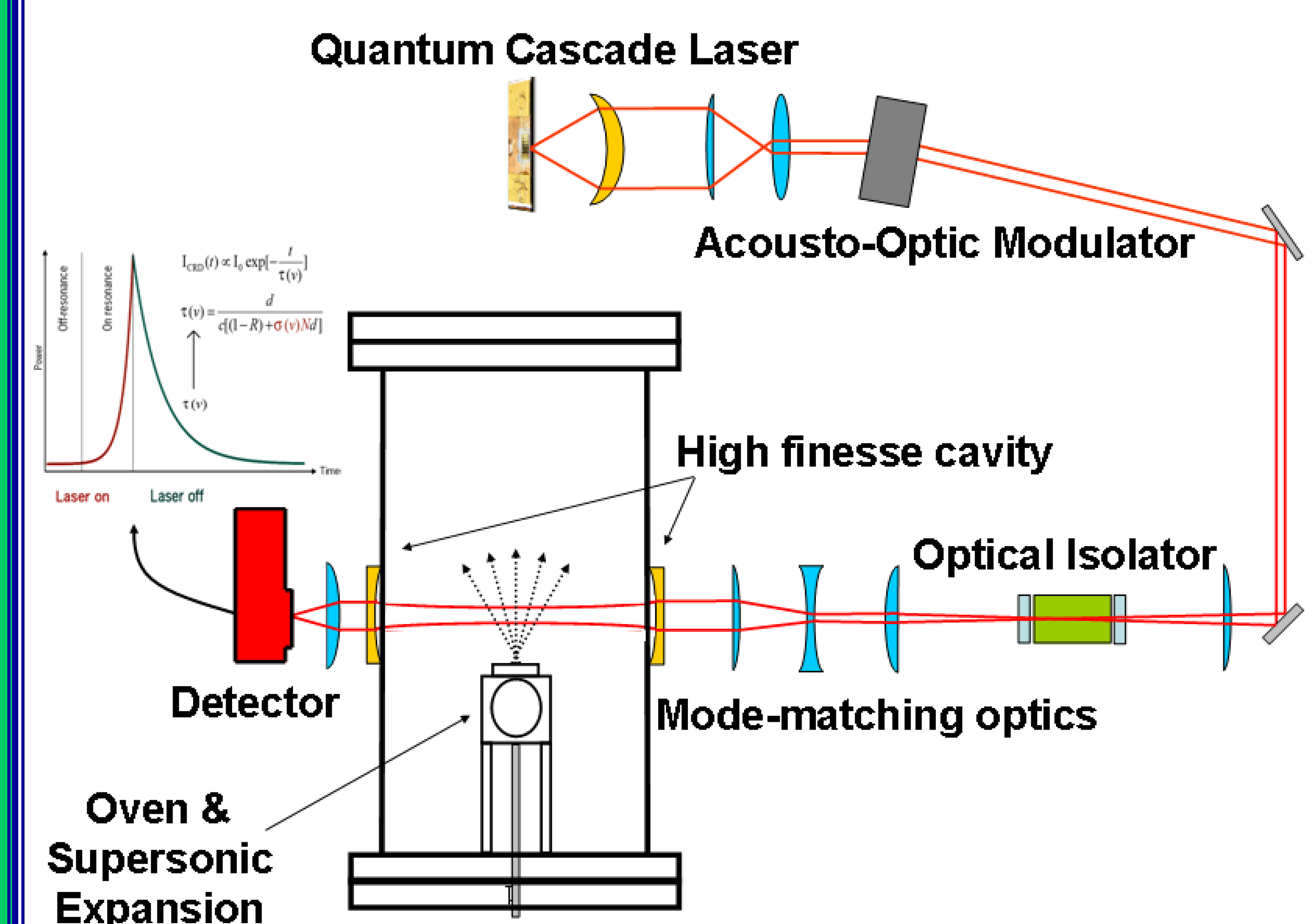


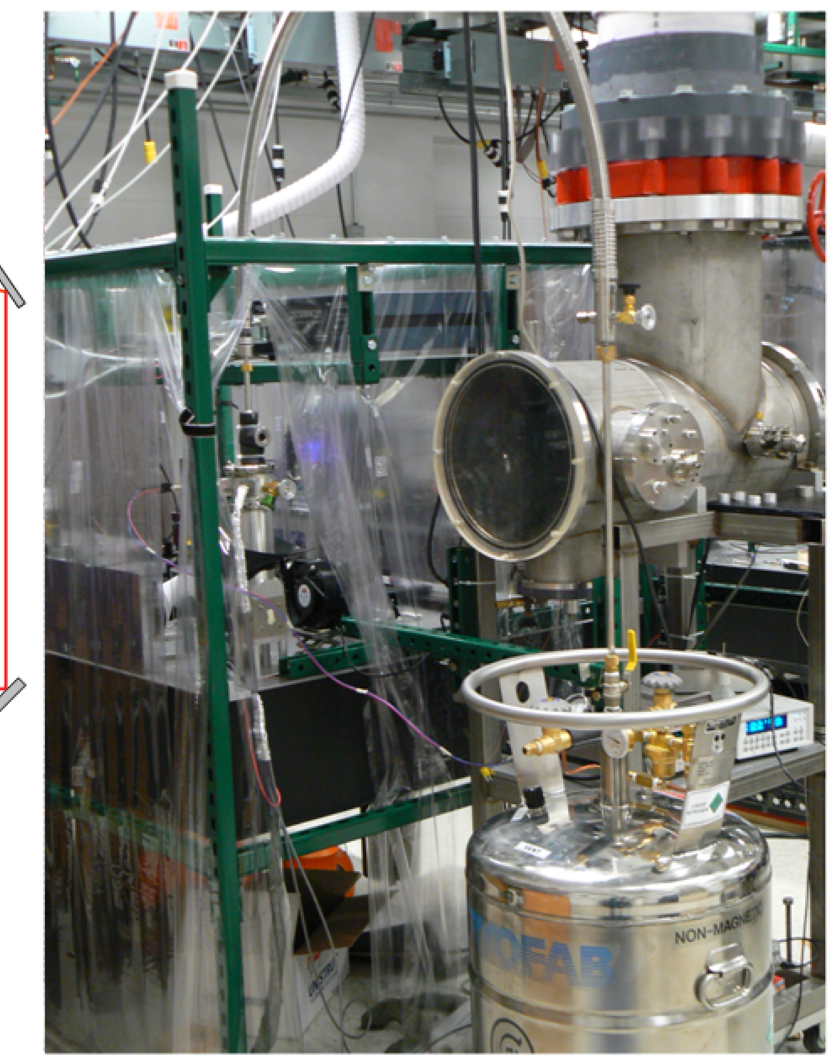
*Availability of laser source through our collaboration with the Gmachl group at Princeton.

*Suitable atmospheric transmission ~1185cm^{-1} that would allow using laboratory results in ground based astronomical searches.

Experiment

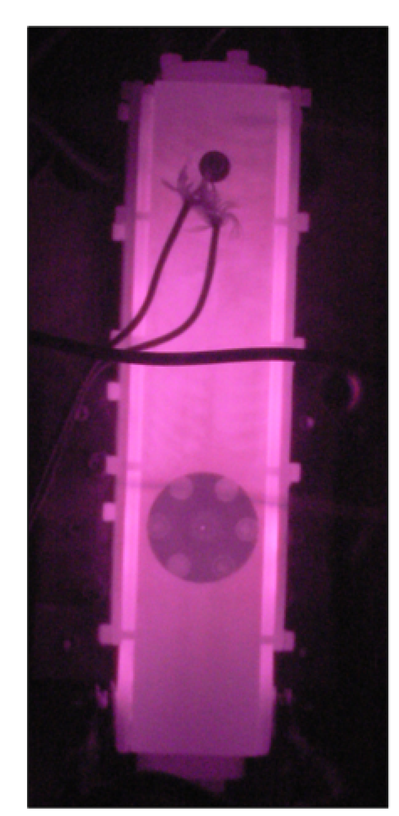
➢We have constructed a spectrometer for continuous-wave cavity ringdown spectroscopy (cw-CRDS) ~8.5 μm for the study of rotationally cold gas phase C_{60}. The output of a continuous-wave quantum cascade laser (cw-QCL) is matched to the focal properties of a high finesse cavity. A piezo changes the length of the cavity, and the light couples to the cavity when in resonance. The light is diverted when resonance is achieved, and the intensity exponentially decays with a rate proportional to the cavity absorption. Gas phase C_{60} is produced in a high temperature oven and supersonically expanded into the cavity.





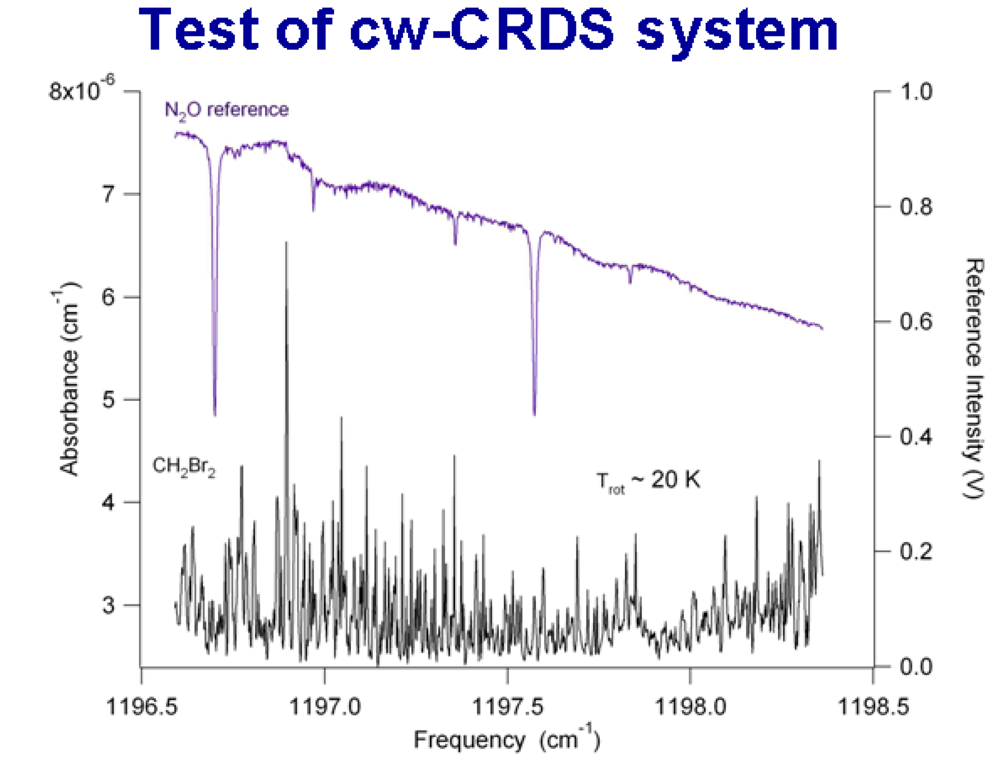
High Temperature Source

➢Room temperature vapor pressure of C_{60} is negligible. To get any significant number density in the supersonic expansion it is necessary to build a cw source capable of maintaining a temp >600°C for many hours without failing.



Deposition of gas phase C_{60} on flange with source pinhole

Test of cw-CRDS system



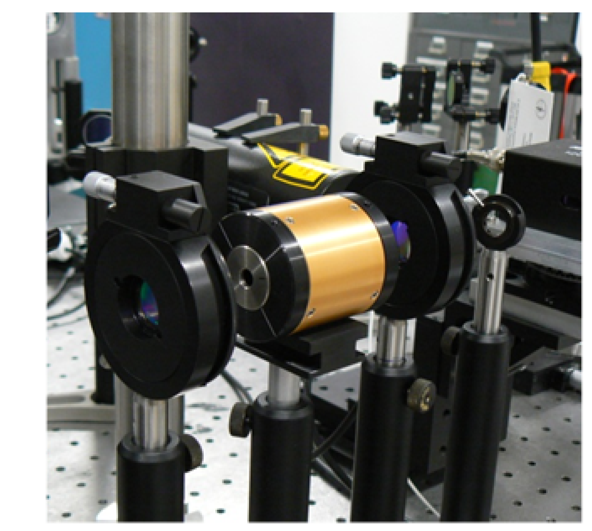
➢Methylene bromide (CH_2Br_2) was chosen as a test molecule for the spectrometer set-up without heating the source.

➢Calibrated frequency window is from 1195.81 to 1196.96 cm^{-1}. Estimated FWHM of 50 MHz for unblended lines.

➢P and Q branch were acquired, but R branch didn't fall within frequency coverage of laser.

Optical Isolator

➢Fabry-Perot quantum cascade lasers are especially vulnerable to optical feedback. Optical feedback destabilizes the laser during scanning make it virtually impossible to avoid frequency jumps that can potentially ruin any spectroscopy.



➢To eliminate feedback from our high reflectivity mirrors in the experiment we are working with Optics For Research (OFR) developing an optical isolator at ~8.5μm

Future Work

➢Continue colloborative effort with OFR to develop an improved optical isolator

➢Revisit CH_2Br_2 work and complete coverage of vibrational band

➢Receive and characterize a FP-QCL that can scan from 1183 – 1186 cm^{-1}

➢Begin process of scanning over proposed C_{60} vibrational band center ~1185 cm^{-1}

References

1. Kroto et al. 1985, *Nature* **318**, 162
2. Brozolo et al. 1994, *Nature*, **369**, 37
3. Becker et al. 2001, *Science*, **291**, 1530
4. O'Brien et al. 1988, *J Chem Phys*, **88**, 220

Acknowledgements

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