Continuous wave cavity ringdown spectroscopy of C_{60} at 8.5 μm using a quantum cascade laser and a supersonic expansion source

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Introduction

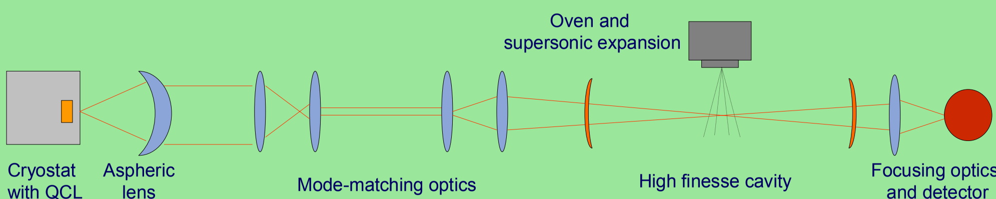
C_{60} was discovered during experiments simulating carbon star outflows[1], and has been found in craters on the Long Duration Exposure Facility (an earth orbiting satellite)[2], and in sediments related to meteorite impacts[3].

C_{60} has 174 vibrational degrees of freedom, 60 quantum-mechanically indistinguishable (spin 0) bosons, and icosahedral (I_h) symmetry: 6 five-fold axes, 10 three-fold axes, and 15 two-fold axes. Symmetry restrictions lead to only 4 F_{1U} IR active modes near 1432, 1183, 577, and 528 cm^{-1}.

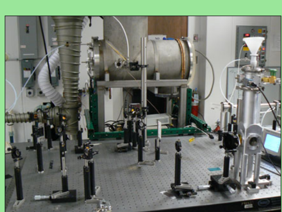
C_{60} should be readily formed and stable to dissociation in space, but no rotationally cold gas phase absorption spectrum of C_{60} is available for comparison to interstellar observations.

Experiment

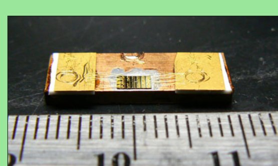
We have constructed a spectrometer for continuous-wave cavity ringdown spectroscopy (cw-CRDS) at 1184 cm^{-1} (8.5 μm) for the study of rotationally cold gas phase C_{60}.



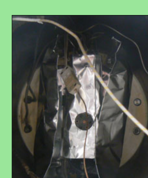
The output of a continuous-wave quantum cascade laser (cw-QCL) is matched to the focal properties of a high finesse cavity. A piezo changes the length of the cavity, and the light couples to the cavity when in resonance. The light is diverted when resonance is achieved, and the intensity exponentially decays with a rate proportional to the cavity absorption. Gas phase C_{60} is produced with a high temperature oven and supersonically expanded into the cavity.



The experimental setup



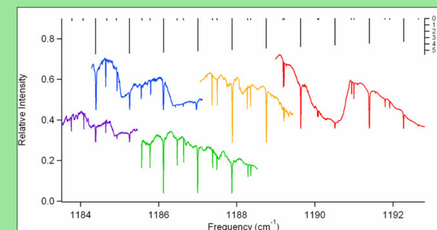
8.5 μm quantum cascade laser (QCL)



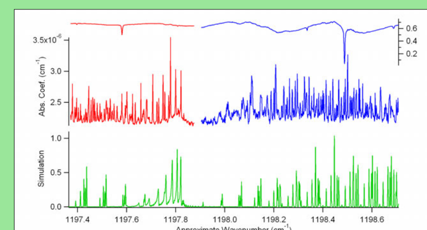
> 600 ºC oven and supersonic expansion for rotationally cold gas phase C_{60}

Progress

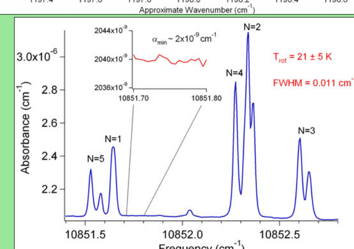
We have completed the optics setup and achieved ringdown with a minimum detectable absorbance of ~2x10^{-9} cm^{-1}.



Direct absorption spectra of N_2O have been obtained from 1183 – 1193 cm^{-1} to test the tuning of the QCL over the expected C_{60} frequency range.

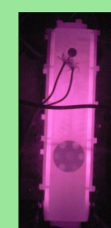


CRD spectra of CH_2Br_2 have been obtained and the linewidth is 60 MHz. We found that electronically switching the QCL rather than diverting the beam during ringdown acquisition leads to mode hops.



A rotationally cold CRD spectrum of N_2^+ has been obtained with the pinhole source and 650 °C N_2 gas using a 950 nm tunable external cavity diode laser.

The oven reaches and maintains temperatures > 600 ºC, and deposition of C_{60} vapor has been observed on the source and in the vacuum chamber.



Ongoing Work

We are installing an acousto-optical modulator (AOM) for diversion of the light from the cavity upon resonance so as to eliminate the mode hops induced by the electronic switching. Once installed, we will confirm mode-hop free laser tuning through continued cw-CRDS investigation of CH_2Br_2. We will then obtain the C_{60} spectrum with this experimental setup.

References

1. Kroto et al. 1984, *Nature* 318, 162
2. Radicati di Brozolo et al. 1994, *Nature*, 369, 37
3. Becker et al. 2001, *Science*, 291, 1530

Acknowledgements

NASA Laboratory Astrophysics, American Chemical Society, Dreyfus Foundation

We thank Richard Saykally for the loan of equipment.