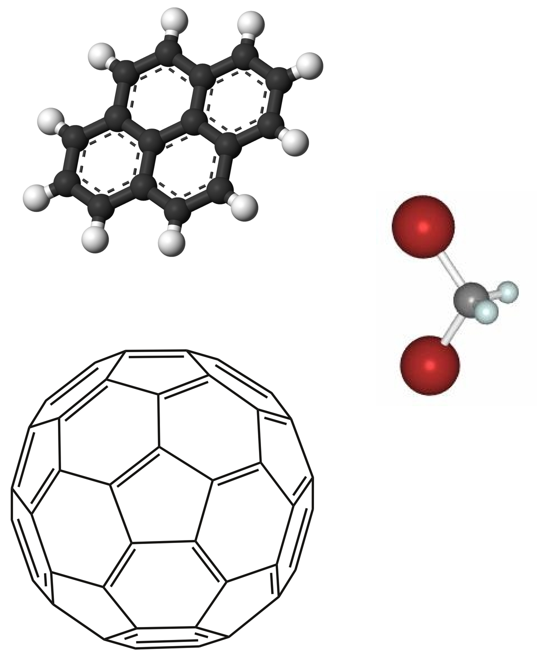

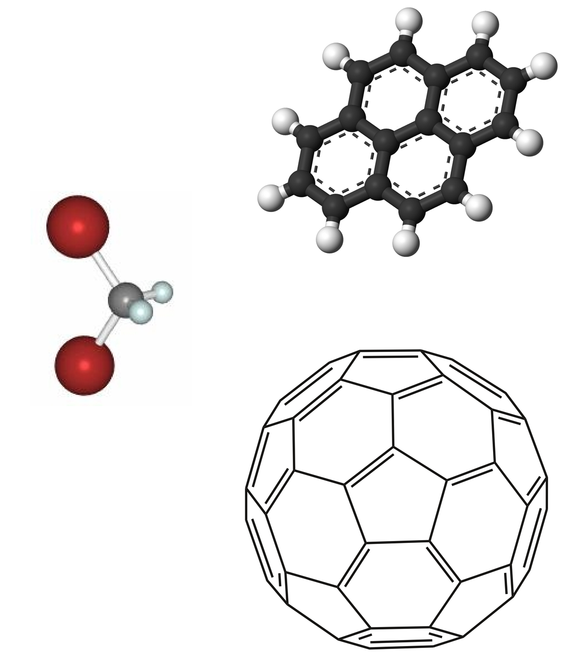

Continued Development of a Sensitive Mid-IR Cavity Ring-down Spectrometer Using a Quantum Cascade Laser

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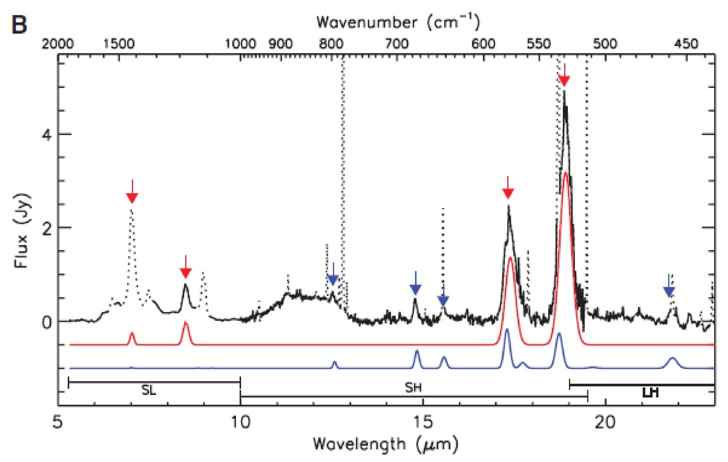
Background

•C_{60} was originally discovered in experiments that attempted to recreate carbon star outflow chemistry[1]

•C_{60} has been detected in emission from a planetary nebula[2] and two reflection nebulae[3]

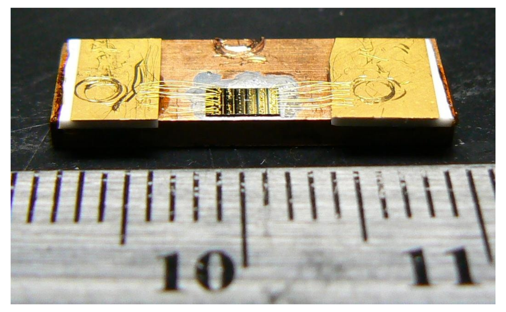


Infrared emission from C_{60} (red arrows) and C_{70} (blue arrows) in planetary nebula Tc1[2]

•Our goal is to record the cold, high resolution, gas phase spectrum at ~1185 cm^{-1} using a sensitive mid-IR spectrometer, which will aid a search for C_{60} in absorption

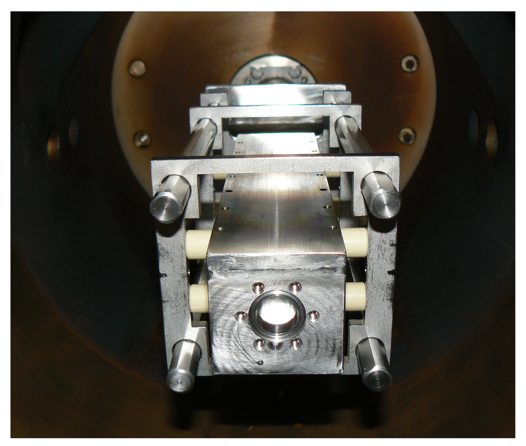
Quantum Cascade Laser

We use quantum cascade lasers (QCLs) provided by Claire Gmachl's group at Princeton. QCLs are composed of repeating sequences of quantum wells which can be changed to produce different laser frequencies.



High Temperature Oven

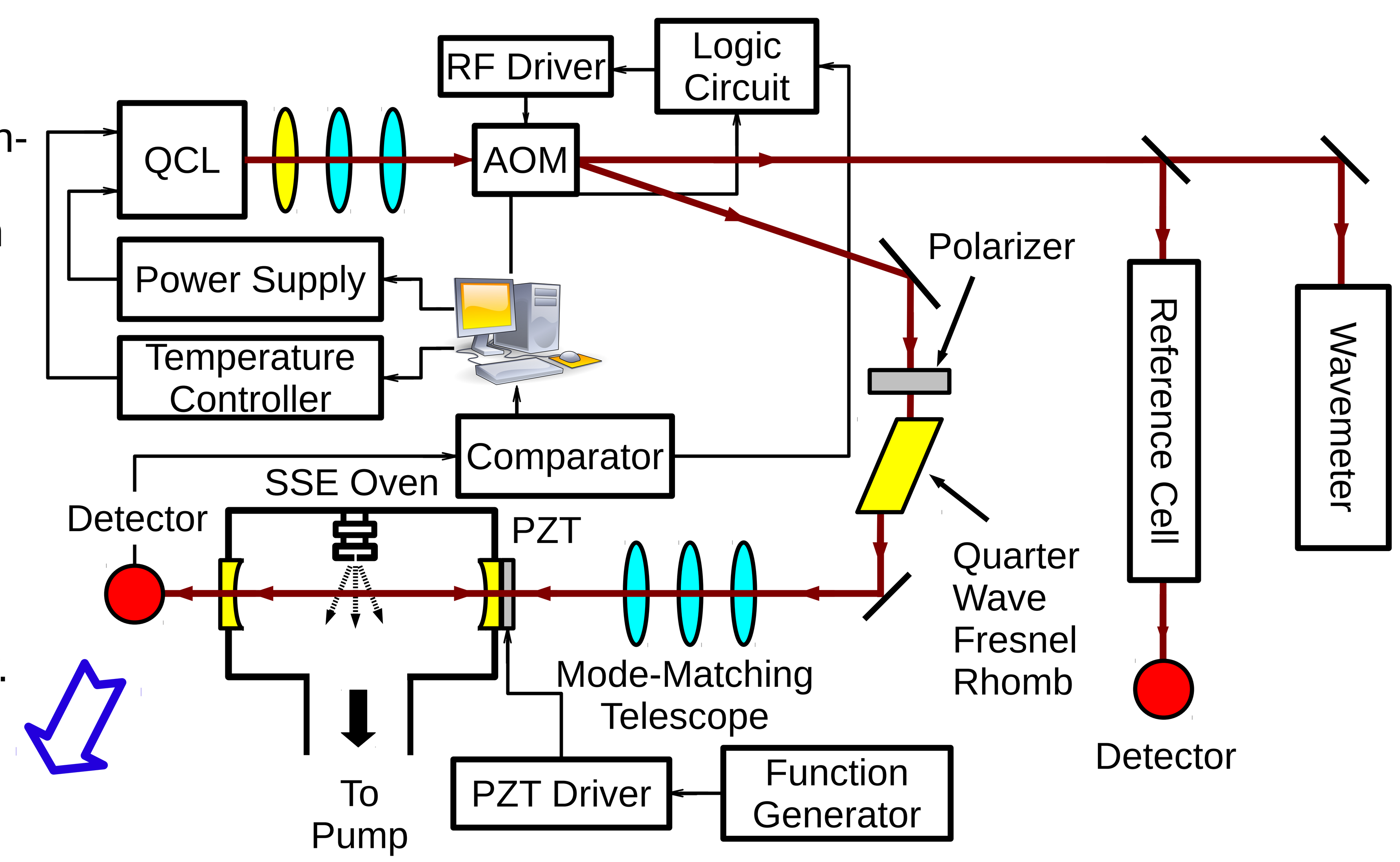
We have designed a high temperature oven capable of operating at 700°C for many hours. The oven also acts as a supersonic expansion source to cool the sample before it is measured.

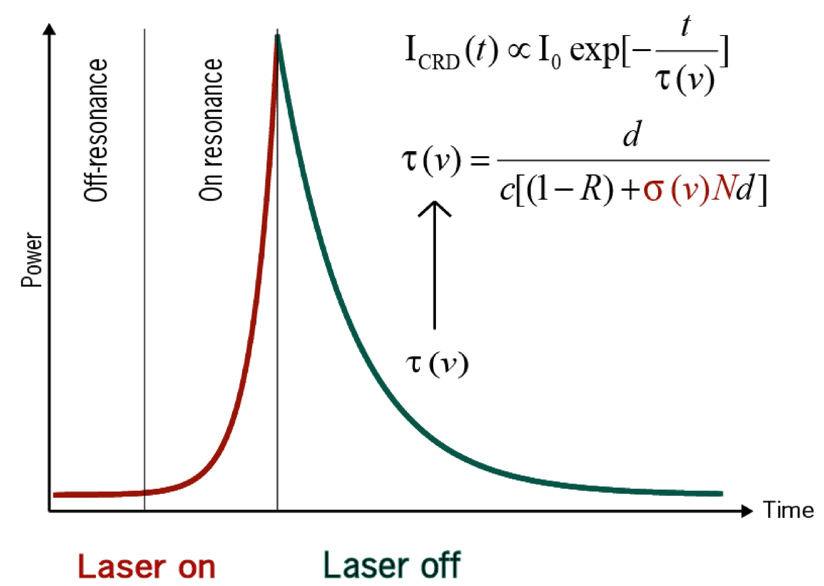


The oven was recently redesigned to hold more sample and work with samples that melt, such as PAHs. We want to work with PAHs to understand cooling of large molecules.

Mid-IR Spectrometer

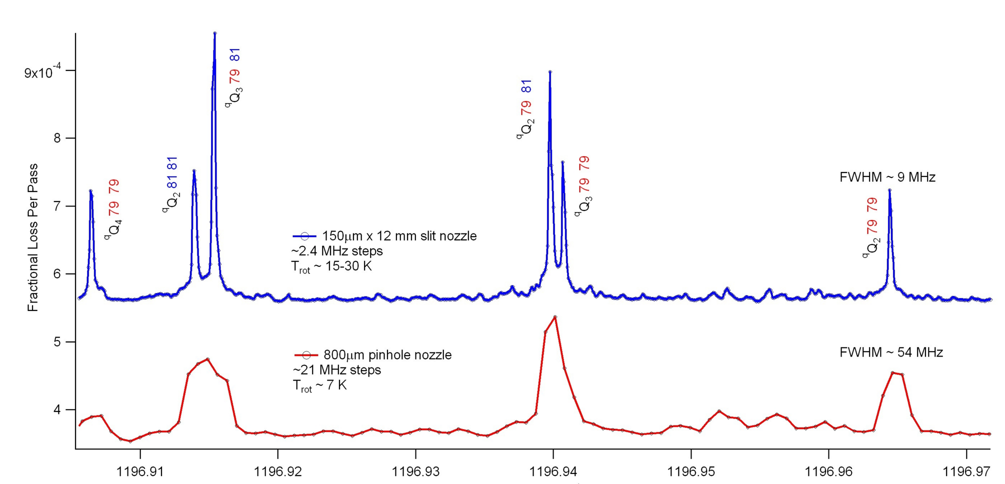
We have constructed a mid-infrared cavity ringdown spectrometer to record the high-resolution vibrational spectrum of C_{60}. Light from our quantum cascade laser is sent through an acousto optic modulator (AOM) and then into a high-finesse optical cavity formed from two high-reflectivity mirrors. One of the mirrors is mounted to a piezoelectric transducer which allows us to change the length of the cavity.





As the cavity length changes, the laser comes into resonance with the cavity, leading to a buildup of signal on the detector. When the signal reaches a predefined threshold, the AOM is turned off, and the decay of light from the cavity is measured. The time constant of the decay is proportional to the amount of light absorbed by molecules within the optical cavity. To prevent back-reflection from the input mirror reaching and destabilizing the QCL, we have implemented a Fresnel rhomb-based optical isolator.

Methylene Bromide Spectroscopy



•For initial testing of our spectrometer we have observed the ν_8 vibrational band of methylene bromide (CH_2Br_2)[4]

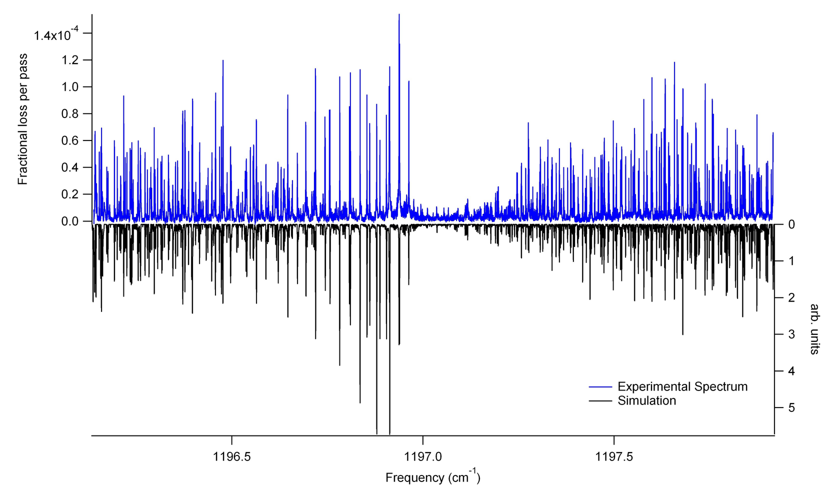
•Our first measurements were done using a pinhole expansion—moving to a slit led to improved resolution (left)

•Use of the Fresnel rhomb has given us continuous frequency coverage over multiple wavenumbers (below)

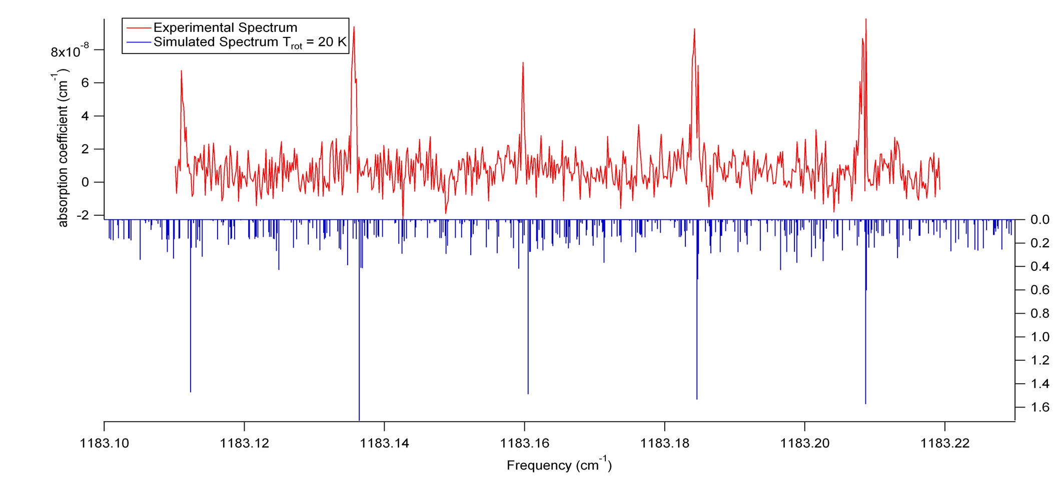
•We observed three isotopologues of methylene bromide: $CH_2{}^{79}Br_2$, $CH_2{}^{79}Br^{81}Br$, and $CH_2{}^{81}Br_2$

•We used PGOPHER[5] to assign 240 rovibrational lines with an average observed - calculated of 0.00037 cm^{-1} (11 MHz)

•From our assignment we obtained values for ν_0, A', B', and C' for all three isotopologues



Pyrene Spectroscopy



•We need molecules of intermediate difficulty between methylene bromide and C_{60} such as polycyclic aromatic hydrocarbons (PAHs)

•We have obtained preliminary spectra of pyrene ($C_{16}H_{10}$) heated to 120°C

•A simulation of this pyrene vibrational band from known constants indicates we are able to cool this large molecule to ~20 K

•Largest PAH to be observed with rotational resolution in the infrared

Future Work

•Finish observing the ν_{19} band of pyrene

•Continue from pyrene to larger PAHs (such as coronene) to test cooling of large molecules in supersonic expansions

•Measure the gas phase vibrational spectrum of C_{60}

References and Acknowledgments

[1] Kroto et al. *Nature* **318**, 162 (1985).
[2] Cami et al. *Science* **329**, 1180 (2010).
[3] Sellgren et al. *ApJ Lett.* **291**, 1530 (2010).
[4] Brumfield et al. *Rev. Sci. Instrum.* **81**, 063102 (2010).
[5] Western, http://pgopher.chm.bris.ac.uk

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