Indirect THz Spectroscopy of Molecular Ions

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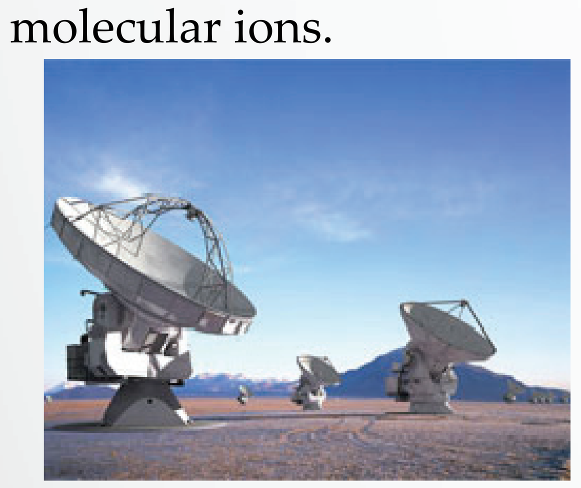
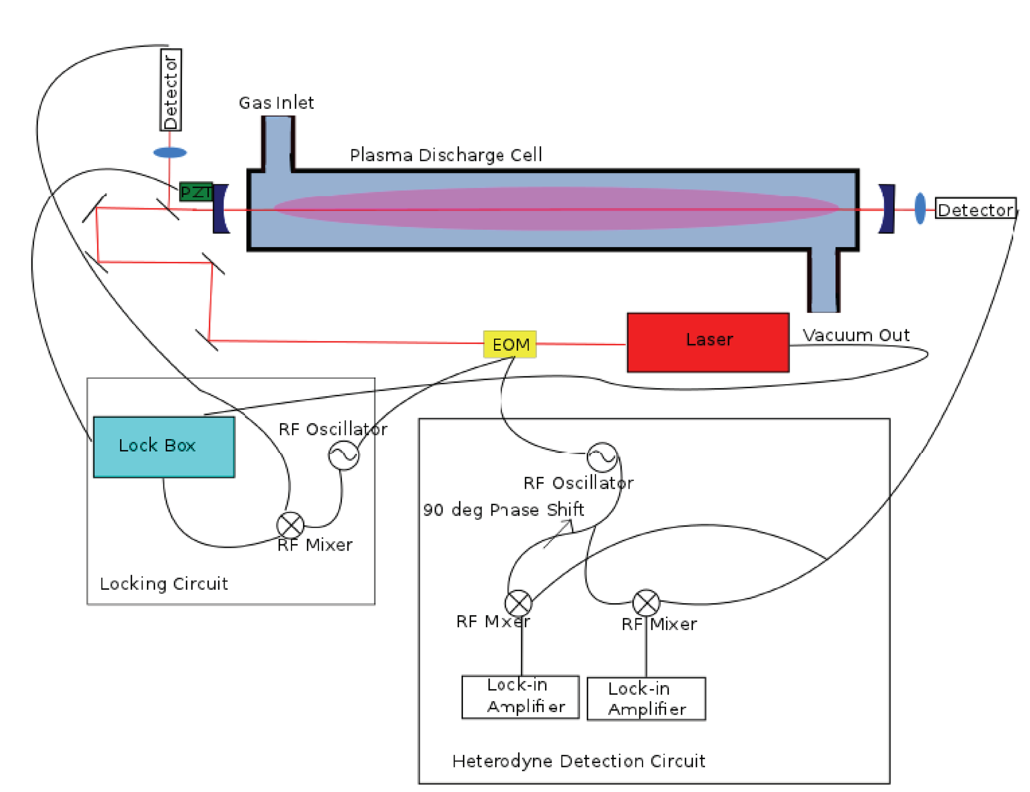
Introduction

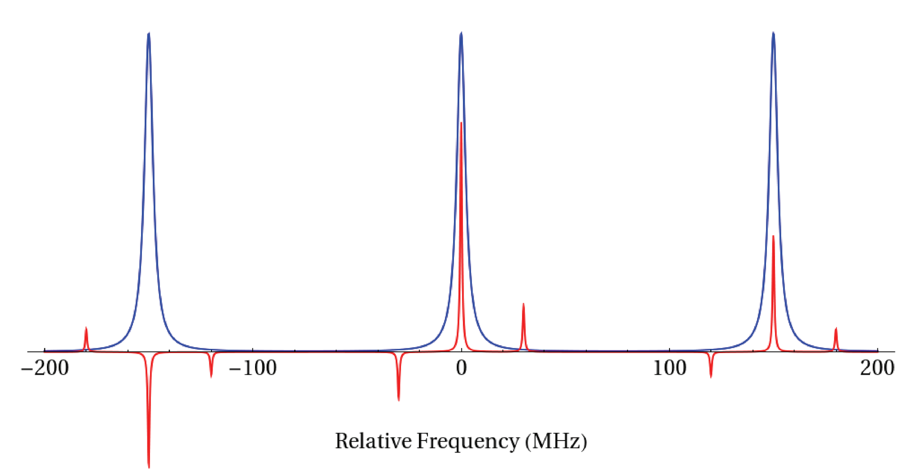
THz/sub-mm telescopes such as Herschel, ALMA, and SOFIA give data in a spectral region that plays a critical role to understanding the chemical composition in the interstellar medium. Within this spectral region, small molecules have rotational lines and large molecules have low lying torsional modes of vibration. Laboratory spectroscopy is essential to interpreting the wealth of data that comes from these instruments. However, laboratory work in the THz spectral region remains a relatively immature field. The infrared on the other hand is a well developed field. However, the accuracy and precision of rotationally-resolved vibrational spectroscopy in the infrared has traditionally been no better than ~150 MHz, and this is insufficient to guide THz astronomical observations. Here we present an instrument capable of improving the precision of infrared spectroscopy to <1 MHz by utilizing saturation spectroscopy, and better than 1 MHz accuracy using an optical frequency comb as a calibration source. By utilizing combination differences and effective Hamiltonian fitting, the frequencies of rotational transitions may be inferred. This represents a general instrument for indirect THz spectroscopy. Implementation of this instrument will allow us to aid astronomical searches for molecular ions.



NICE-OHVMS



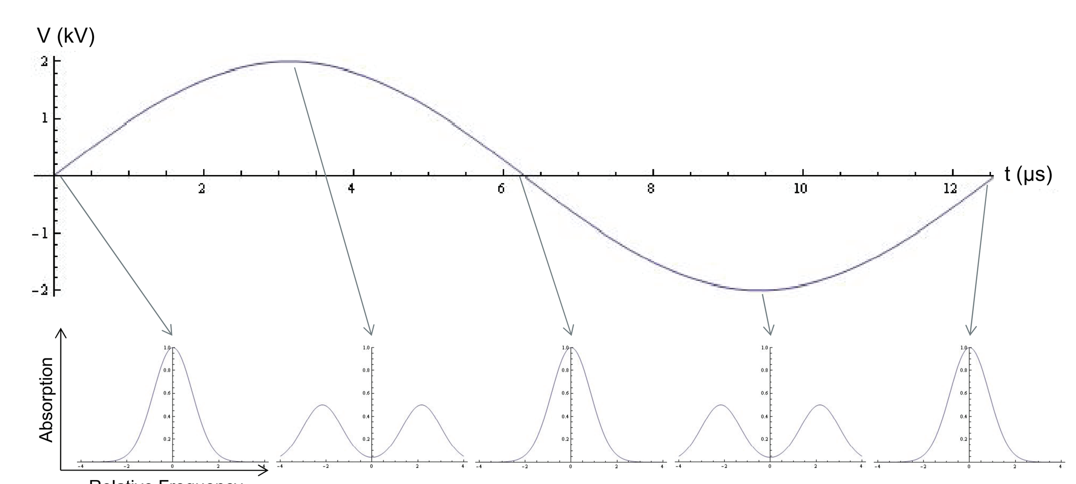
Block diagram of a NICE-OHVMS instrument.



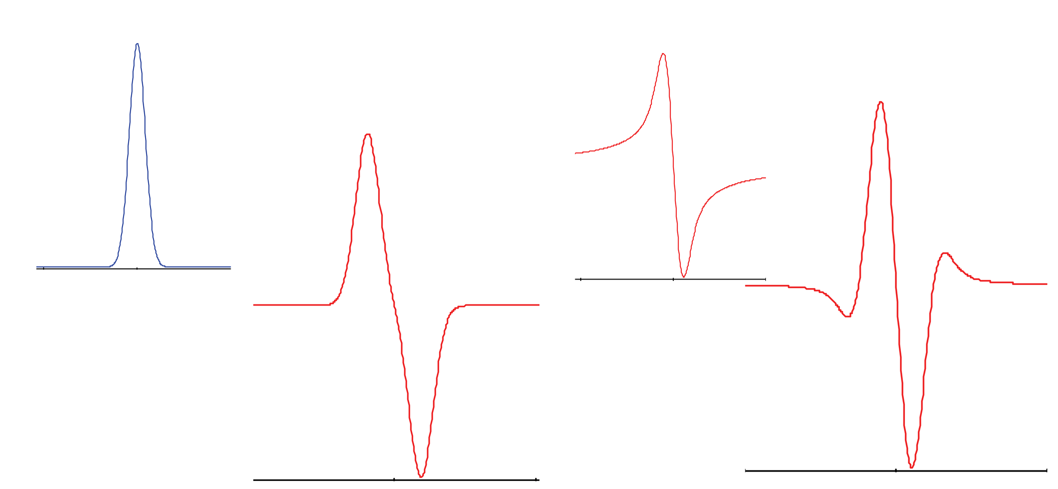
Cavity modes and heterodyne sidebands

NICE-OHVMS or noise immune cavity enhanced optical heterodyne velocity modulation spectroscopy, is a spectroscopic technique that combines optical heterodyne spectroscopy, velocity modulation spectroscopy, and the benefits of an optical cavity.

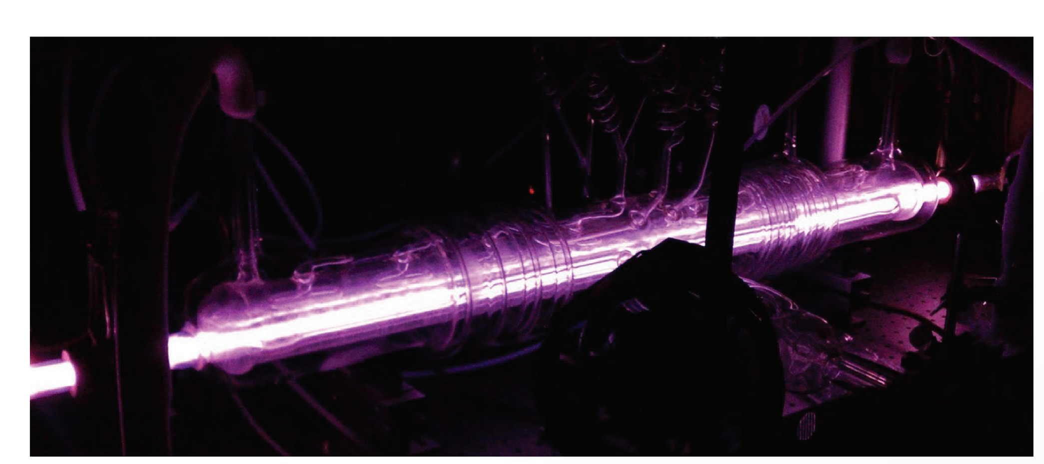
The figures below illustrate the effect of velocity modulation on absorption in a cavity, the way that heterodyne sidebands are transmitted through a cavity, and the line shapes expected from heterodyne absorption and dispersion.



Velocity modulation in a cavity and its effect on an ion's absorption profile



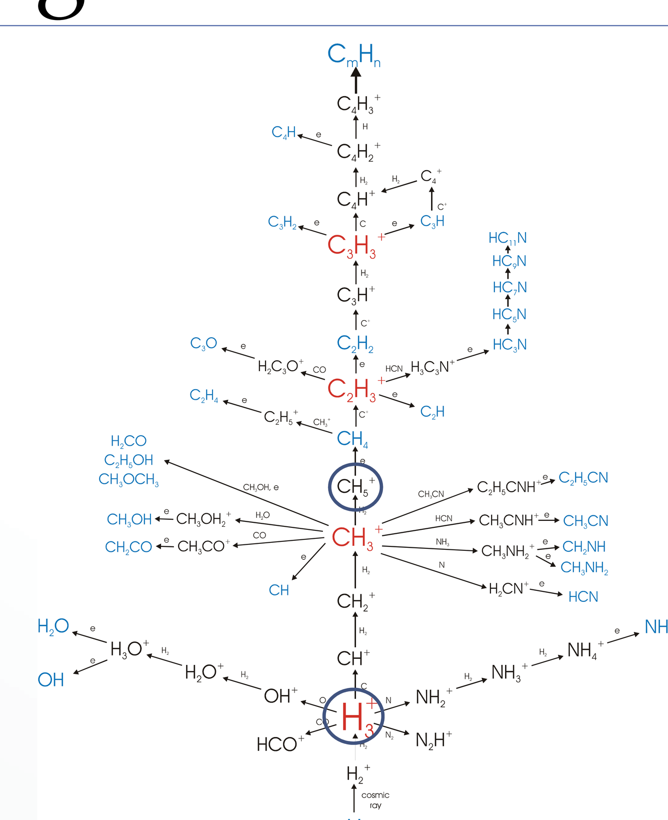
Heterodyne line shapes resulting from absorption (left) and dispersion (right).



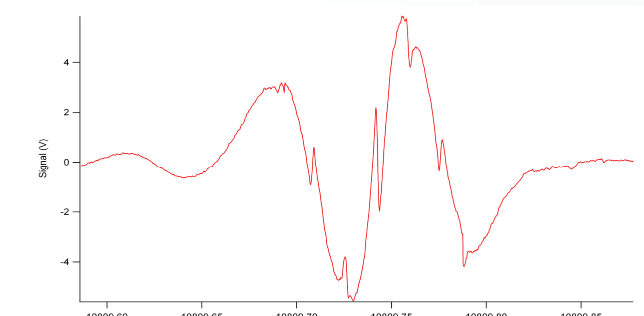
Liquid nitrogen cooled plasma in action.

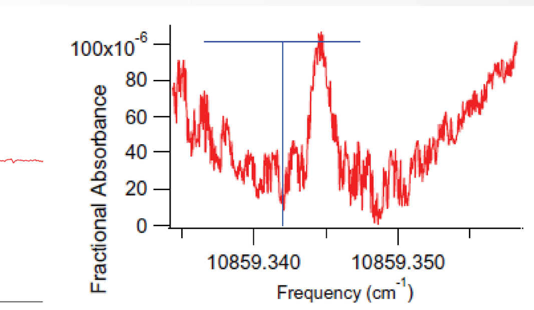
Future Work/Spectral Targets

We are beginning to constructed a cavity around a discharge cell to implement the NICE-OHVMS technique. In its final configuration, our instrument will be referenced to a frequency comb to calibrate our measurement s to sub-MHz accuracy. By using a cavity, and a high power laser, we should be able to saturate transitions from a variety of molecular ions including H_3^+ and CH_5. Gaining information on the rotational transitions we may enable the searches for CH_5^+. In the case of H_3^+ we seek to improve the accuracy and precision of the lines to better aid the determination of Doppler shifts in planetary atmospheres.



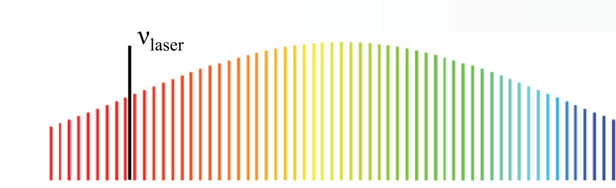
Near-IR Work





(Left)NICE-OHVMS spectrum of $Q_{11}(14)$ of N_2^+ acquired with 1 GHz heterodyne sideband spacing. (Right) Improved* precision and accuracy from Lamb dip.

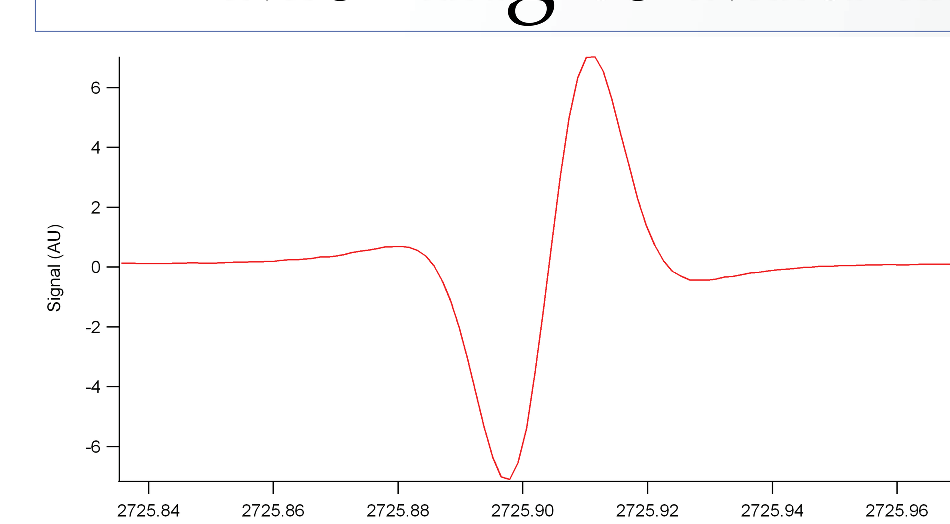
Our group discovered NICE-OHVMS with a Ti:Sapphire laser. Using this spectroscopic setup, Lamb dips have been observed in N_2^+ and their frequencies have been calibrated using a frequency comb.



Conceptual diagram of the frequency comb. Frequency calibration is a result of the beat note from the cavity.

*D.W. Ferguson, K.N. Rao, P. A. Martin, G. Guelachvili. J. Mol. Spec. 153, (1992) 599.

Moving to Mid-IR



R(1,0) transition of H_3^+. Double Pass heterodyne velocity modulation spectroscopy in liquid nitrogen cooled plasma S/N 1400.

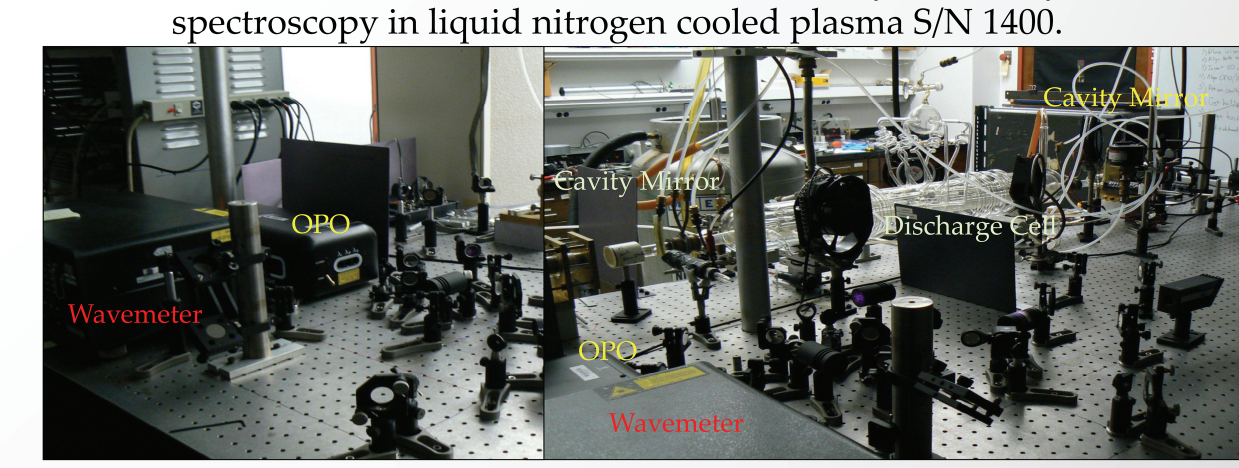


Figure 8: Photograph of the mid-IR instrument

We have scanned both the R(1,0) and R(1,1)^u lines in the ν_2 band of H_3^+. The spectra we observed had extremely high signal to noise ratios.