SUB-DOPPLER SPECTROSCOPY OF MOLECULAR IONS IN THE MID-INFRARED

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High precision, high accuracy spectroscopy in the mid-IR plays an important role in many areas of study. It provides benchmarks for *ab initio* calculations, acts as a reference for astronomical searches, and enables the study of highly fluxional ions, such as CH_5^+ and H_5^+. In an effort to advance these interests, we built an instrument that uses Noise Immune Cavity Enhanced Optical Heterodyne Velocity Modulation Spectroscopy to achieve sensitive, high signal-to-noise spectra of molecular ions with sub-Doppler features.

There are four main components of the instrument: a tunable cw-OPO, an optical cavity with a finesse of ~ 200, a liquid nitrogen cooled AC positive column cell, and an optical frequency comb for absolute frequency calibration. The OPO is pumped by a fiber laser at 1064 nm with a signal cavity resonant between 1.5 and 1.6 μm, which corresponds to an idler wavelength tunable between 3.2 and 3.9 μm. The idler is actively locked to the cavity via a Pound-Drever-Hall locking scheme responsible for maintaining the resonance condition of the cavity. Inside the cavity is a AC driven positive column plasma, which produces a high ion density and modulates their velocity providing neutral discrimination. Since the cell can be cooled by liquid nitrogen, the instrument has an extra advantage due to signal enhancement of low J transitions. Saturation spectroscopy is possible because of enhancement of laser power in the cavity, which leads to improved precision with respect to line center determination due to narrow Lamb dips. To achieve sub-MHz accuracy, the signal and pump are roughly calibrated with a near-IR wavemeter and then referenced to a frequency comb. To demonstrate the advantages of this technique, measurements of the ν_2 fundamental band of H_3^+ will be presented.