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## Title

High Resolution Spectroscopy of Vibrational Overtone Transitions of Methane and Methyl Iodide Molecules with a Fabry-Perot Cavity Absorption Cell

## Abstract

From early days, spectroscopy has contributed extensively in various fields of science such as physics chemistry and astronomy In particular after the advent of lasers a frequency resolution of spectrometers has been drastically improved using a strong interaction between light and matter High resolution laser spectroscopy however, has not been carried out for molecular vibrational overtone bands because their transition dipole moments are small, and only weak radiation sources of diode lasers are available in the near infrared region where molecular overtone bands lie

In this thesis we present a novel laser spectroscopy using a Fabry<sub> $\overline{4}$ </sub>Perot cavity as an absorption cell in order to enhance the optical field strength This feature has allowed us to carry out saturation spectroscopy of molecular overtone bands with a high resolution

To attain higher resolution we have improved a sensitivity of the spectrometer As the sample gas pressure and the laser power get higher not only signal intensities increase more but also spectral widths become broader obstructing measurements with high resolution. To reduce both the gas pressure and the laser power we employed a frequency modulation technique called NICE-OHMS, hence improved the sensitivity by a factor of 10. Consequently we have successfully observed a saturated spectral line of the

 $2v_3$  band of methane in the 1.6 µm region with a spectral width of 320 kHz, which corresponds to a relative frequency resolution of  $5.6 \times 10^8$ .

We applied the spectrometer to precise measurements of absolute frequencies of the methane transitions in the  $2v_3$  vibrational band. Each frequency of the 1.53- and 1.67-µm lasers was stabilized to the narrow saturated spectral lines of acetylene and methane, and the frequency differences between them were precisely measured. Using the known absolute frequency of acetylene, the absolute frequencies of the R(0) and Q(1) transitions of methane were determined to be 180.345 065 08(37) and 180.021 253 10(61) THz, respectively. The accuracy of these values are two orders better than the previous measurement.

Saturation spectroscopy was performed for the  $2v_4$  band of methyl iodide. The spectral lines overlapped in a Doppler-limited resolution were resolved into individual hyperfine components, the pattern of which makes unambiguous assignments possible. The hyperfine coupling constant in the vibrationally excited state was determined to be -1.943 GHz from the hyperfine splittings of the *Q*-branch transitions. These results were obtained because of wide tunability as well as the high resolution of the present spectrometer.

Near-infrared radio-frequency double-resonance spectroscopy was carried out for methyl iodide. Dressed molecules excited by the radio-frequency radiation resonant with a hyperfine transition were observed by high-resolution near-infrared spectroscopy. All the observed spectra including quantum interference agreed well with the calculation based on the dressed-state theory, proving that the spectrometer is a powerful tool for precise investigation of dressed molecules.