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Frequency comb-referenced measurements of self- and nitrogen-broadening in the $\nu_1 + \nu_3$ band of acetylene

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ABSTRACT

We report measurements of self- and nitrogen-pressure broadening of the P(11) line in the $\nu_1 + \nu_3$ combination band of acetylene at 195 739.649 5135(80) GHz by absorption of radiation emitted by an extended cavity diode laser referenced to a femtosecond frequency comb. Broadening, shift and narrowing parameters were determined at 296 K. For the most appropriate, hard collision, model in units of cm^{-1}/atm, we find 0.146317(27), 0.047271(104) and −0.0070819(22) for the acetylene self-broadening, narrowing and shift, and 0.081129(35), 0.022940(74) and −0.0088913(25) respectively, for the nitrogen-broadening parameters. The uncertainties are expressed as one standard deviation (in parenthesis) in units of the last digit reported. These parameters are 2–3 orders of magnitude more precise than those reported in previous measurements. Similar analyses of the experimental data using soft collision and simple Voigt lineshape models were made for comparison.

1. Introduction

The development of phase locked optical frequency combs in the near infrared during the past decade has revolutionized frequency metrology and optical clocks [1–4]. Applications to the measurement of molecular spectra have recently been reported [5–8] and spectroscopic techniques based on this technology will have a profound effect on the precision and accuracy of high resolution spectroscopy in the near future. One area where immediate advantage can be taken of these advances is in the measurement of spectral line shapes. Here, the determination of the subtle shifts and broadening caused by self- or foreign-gas collisions at increased pressures demands extremely good control of spectrometer frequency. Practical applications, such as the calibration of remote measurements, as well as our understanding of the molecular physics of the processes involved will benefit from spectrometers using the newly available technology.

In the present work, we report the first measurements of line broadening, collisional narrowing and shifts caused by self- and nitrogen-gas broadening in the $\nu_1 + \nu_3$ band of acetylene at 1.5 μm using an extended cavity diode laser locked to a femtosecond frequency comb. This is a strong combination band, situated in an atmospheric window, and it may be of interest in the study of the atmospheres of giant planets like Jupiter, Saturn, Uranus, Neptune and Titan (one of the moons of Saturn), where acetylene and polyacetylenes resulting from photochemistry in the upper atmosphere caused by solar radiation have been detected. For the present measurements, an absolute frequency accuracy of 8 kHz at 196 THz is shown to be possible, corresponding to a stability of 2.5 parts in 10^{12}. As a comparison, high resolution Fourier Transform Interferometers are usually controlled by helium neon (HeNe) lasers having a frequency stability of about 2 parts in 10^8, and tunable diode lasers locked to a stabilized Michelson interferometer exhibit similar performance [9,10]. The present work demonstrates how this increase in frequency stability translates into several orders of magnitude improvement in the measurement of pressure broadening and shift parameters of molecular absorption lines. We have studied self-broadening and pressure shift using cells with 1.085 cm or 16.551 cm pathlength. The cells may be cooled to 15 K, but for this work we stabilized the cell temperature to 296 K with a temperature stability of 0.01 K. Pressures range from a few mTorr up to 770 Torr.

2. Experiment

2.1. The spectrometer

The spectrometer at Stony Brook University uses a Menlo Systems FC-1500 Optical Frequency Synthesizer based on a

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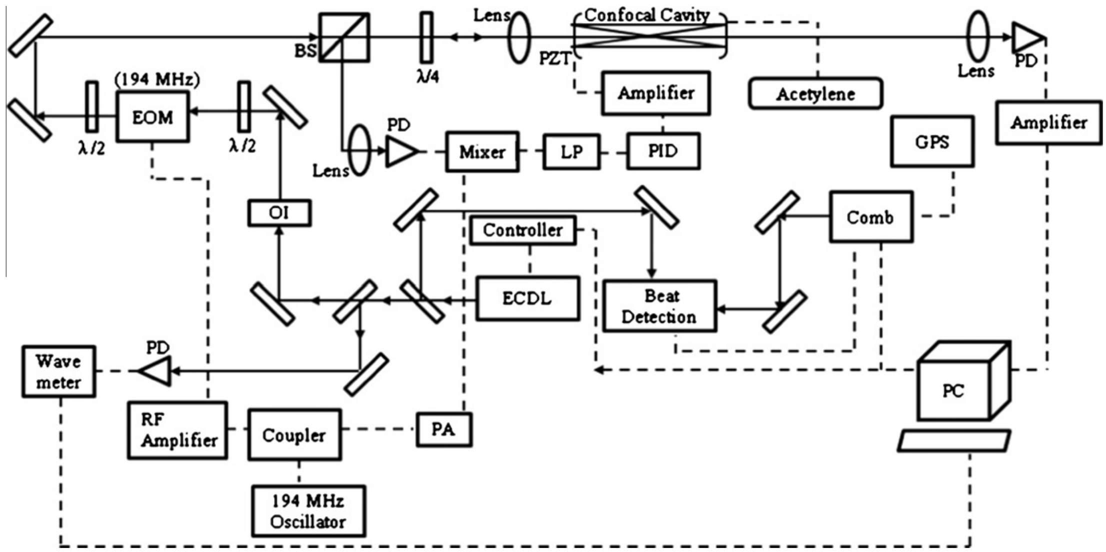

Fig. 1. Block diagram of the spectrometer used for sub-Doppler saturation dip measurements of the P(11) line in the $\nu_1 + \nu_3$ band of $^{12}C_2H_2$. ECDL = extended cavity diode laser, EOM = electro-optic modulator, OI = optical isolator, PD = photodiode detector, LP = Lowpass filter, BS = beam splitter, PA = phase adjustment, PZT = piezo electric translator and PID = Pound–Drever–Hall locking.

mode-locked Erbium fiber laser with a broadened output extending from 1050 nm to 2100 nm. The output of an extended cavity diode laser (ECDL) (Sacher TEC 520 with Sacher Pilot PC controller) was offset-locked to a single comb line and provided the spectroscopic source. Considerable effort was required to achieve stable locking of the diode laser to the comb frequencies. The table holding the experiment was air-suspended, and large amounts of acoustical and vibrational isolation were found to be necessary. The current optical stability is typically at the level of Allan variances of between 5 and 30 Hz relative to the comb frequency standard. Vibrational and acoustical noise in the building as well as temperature and humidity over longer time periods remain the limiting factors in the stability of the ECDL lock.

In order to characterize the absolute accuracy and precision attainable with this system, a sub-Doppler saturation dip measurement was made of the P(11) line in the $\nu_1 + \nu_3$ band of acetylene. A resonant cavity-based absorption cell was designed and constructed using a confocal mirror arrangement since the optical pumping rate attainable using the ECDL with powers of 2–3 mW is generally lower than the collisional dephasing rate in samples at pressures of more than a few mTorr. The cavity was locked to the laser frequency which was itself locked to the frequency comb as the spectrometer frequency was scanned by small adjustments of the nominal 250 MHz comb repetition rate. Cavity locking was achieved using the Pound–Drever–Hall scheme [11,12], which avoids the need to modulate the laser frequency to provide the error signal. Fig. 1 shows a schematic of the spectrometer as configured for the sub-Doppler experiments.

With reference to Fig 1, the comb system is located to the right of the figure. Part of the output from a 1550 nm (ECDL) laser (located toward the middle of the figure), is combined with the comb output in the beat detection unit. Another part of the stabilized output from the ECDL laser is directed to a wavemeter (Bristol Instruments, model 621), used to provide a convenient estimate of the laser emission frequency. The main beam is directed to an optical isolator, an electro-optic phase modulator (New Focus

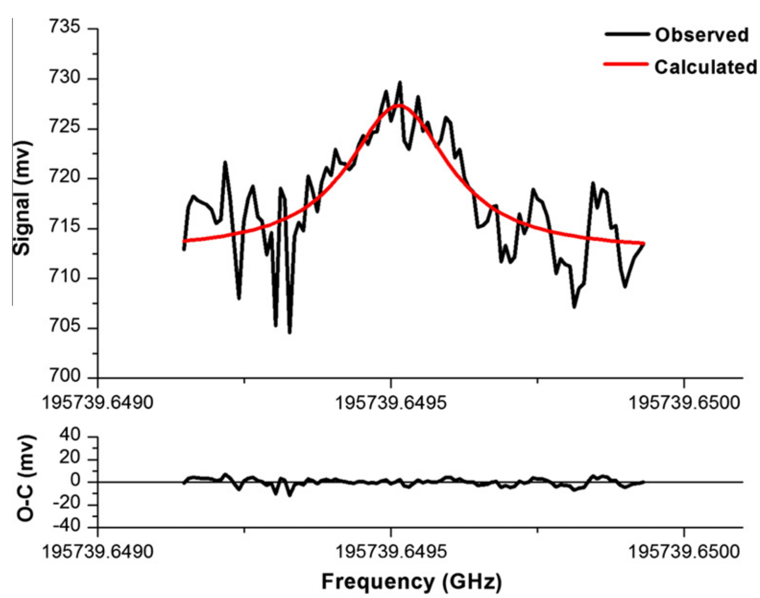

Fig. 2. Sub-Doppler saturation dip signal for the P(11) line in the $\nu_1 + \nu_3$ band of $^{12}C_2H_2$. 5 mTorr of C_2H_2 was used in a 1 m long confocal cavity with finesse measured to be 65. Incident laser power was 3 mW. O–C is the observed minus calculated residual. The fitted linewidth is 105(2) kHz. (HWHM).