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LETTER TO THE EDITOR

Absolute Frequency Measurement of an HDO Absorption Line near 1480 cm^{-1}

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A carbon monoxide laser operating on the P(15) 25–24 transition at 1480 cm^{-1} was used as an intermediate transfer frequency in our cesium-clock-based measurement of an optical frequency standard at 193 THz (1.556 μm). The CO laser frequency was locked to the saturated absorption dip of a nearby HDO line and was simultaneously measured against the NRC H4 hydrogen maser (a cesium clock transfer standard) with help of the NRC infrared frequency chain (*1*). Thus, as a by-product, the absolute frequency of the HDO line was obtained. The absorption line measured belongs to the ν_2 band of HDO and is identified as $J', K'_A, K'_C–J'', K''_A, K''_C = 5, 2, 4–5, 1, 5$ according to reference (*2*).

A small fraction (20 mW) of the available CO laser power (300 mW) was sent through a quartz crystal controlled acousto-optic modulator (AOM) which served as an optical isolator and frequency shifter. The AOM shifted the laser frequency by +52.000 MHz into resonance with the HDO line. A telescope was used to locate the laser beam waist ($w_0 = 3$ mm) on the reflecting mirror at the end (and inside) of a 2.7-m-long absorber cell. The cell contained the stochiometric mixture of H_2O, D_2O, and HDO vapors resulting from a liquid mixture of 50% H_2O and 50% D_2O. The laser was frequency modulated at 1.3 kHz with a peak-to-peak deviation of 500 kHz. The retroreflected beam from the absorber cell was sent to a HgCdTe detector followed by phase-sensitive third harmonic demodulation of the signal.

Figure 1 shows a fast (20 s) frequency scan of the saturation dip using the free running laser. The dip is slightly asymmetric and it is the only absorption feature within the Doppler tuning profile of the laser. The scan was done using a 1 ms time constant for the integrator of the demodulated signal. At a total pressure of 1 mTorr, the HWHM is ~200 kHz. This width was estimated from the amplitude ratio of the central peak to the smaller side peak in the third derivative scan which was taken with the known peak-to-peak deviation of 500 kHz. The transit time broadening (~25 kHz) is small compared to the observed broadening which is dominated by the laser frequency noise. From heterodyne

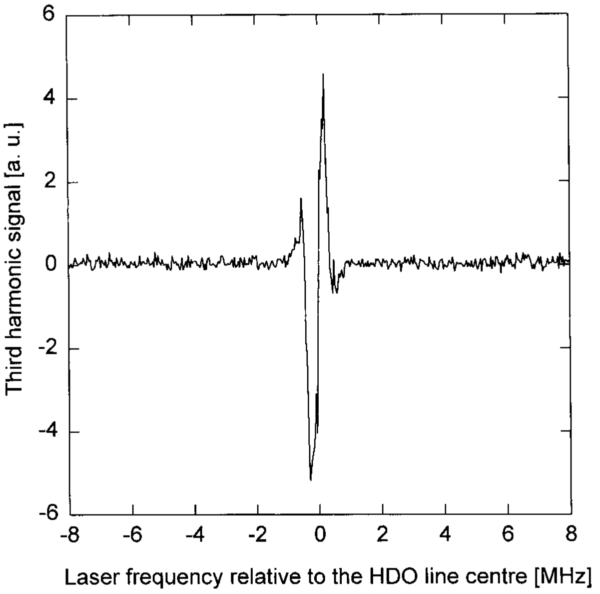

FIG. 1. Saturated absorption dip of the 5, 2, 4–5, 1, 5 transition of HDO recorded with a lock-in amplifier with third-harmonic detection at a time constant of 1 ms. Laser power 10 mW, pressure 1 mTorr, scan time 20 s.

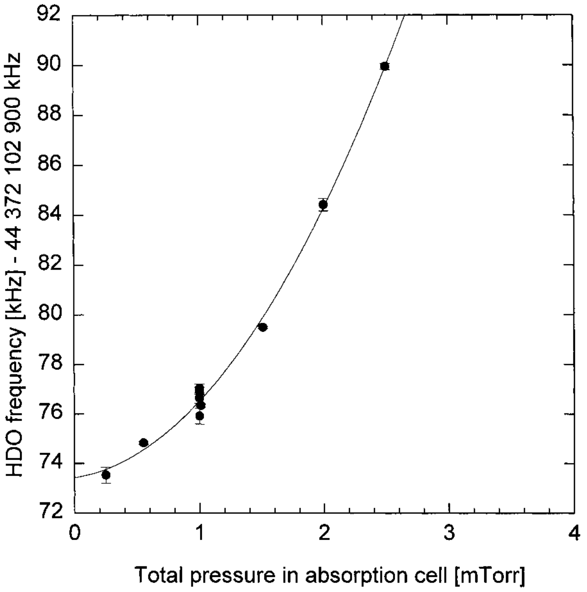

FIG. 2. Pressure shift of the saturated absorption dip of HDO: The pressure was measured with a calibrated membrane-type capsule gauge (MKS type 627) with a total uncertainty of 6% of each reading. At each pressure, the laser was locked to the center of the dip and heterodyne measurements were taken over time periods of 300 s.

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TABLE 1

Reference [number]	Wavenumber cm^{-1}	Uncertainty cm^{-1}	Frequency MHz	Uncertainty MHz
[2]	1480.094 376	0.000 064	44 372 113.1	1.9
[3] and [2]	1480.094 035	0.000 064	44 372 102.9	1.9
[4]	1480.094 048	0.000 030	44 372 103.3	0.9
present result	1480.094 038 033	0.000 000 067	44 372 102.973	0.002

Note. Row 1 lists the wavenumbers and their uncertainties for the HDO line as given by (*2*), these numbers are then converted to frequencies and shown in the last two columns. Row 2 uses a scaling factor taken from (*3*) and applied to the data from (*2*). Row 3 uses the data from (*4*) and their conversion to frequencies.

measurements against the NRC frequency chain, the frequency jitter of the free running laser measured ~50 kHz for 1 s averaging periods which accounts for some of the structure seen in the narrow saturation dip of Fig. 1. The CO laser frequency was servo locked to the center of this dip and then measured by the NRC frequency chain. The servo lock reduced the laser frequency noise from 50 to 4 kHz for 1 s counting periods.

Most of the data were taken at a pressure of 1 mTorr but, in a separate experiment, the pressure shift of the center of the dip was also measured. Figure 2 shows that the center of the absorption line shifts to the blue with increasing HDO pressure. The shift is linear with pressure at higher pressures. The deviation from linear behavior at the lower pressures may be due to the domination of power and transit time broadening. From three measurements done on separate days at a pressure of 1 mTorr, a mean value of 44 372 102 976.5 kHz with a (1σ) standard uncertainty of 0.6 kHz for the HDO frequency is obtained. From a quadratic fit of the pressure dependent data of Fig. 2 the HDO center frequency can be extrapolated to zero pressure: The line center value then becomes 44 372 102 973.4 kHz with an increased uncertainty due to the fit of 1 kHz. Measurement of the line center frequency at 1 mTorr showed no dependence on the applied laser power within the range 6–20 mW.

Table 1 shows a comparison of our value for the HDO line frequency (at zero pressure) with recent literature values. The wavenumbers and uncertainties from references (*2–4*) were converted into frequencies using the internationally defined speed of light of 299 792 458 m/s. Our frequency uncertainty is very small compared to any of the previous measurements and we believe that this is the first cesium-based frequency measurement of an HDO absorption line. Given the scarcity of accurate cesium-based absorption measurements in this region of the infrared spectrum it is anticipated that the HDO absorption line may be of use as a reference in molecular spectroscopy.

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