Supplemental Material for
Toward a determination of the proton-to-electron mass ratio from a Lamb-dip measurement of HD

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I. FREQUENCY CALIBRATION

Two beams from the external cavity diode laser (ECDL), separated by a polarizing beam splitter, were used for frequency locking and spectroscopy detecting, denoted as the "locking" beam and the "spectral" beam, respectively. The "locking" beam was frequency shifted (f_{AOM1}) by an acousto-optic modulator (AOM1, AA MGAS110-A1) and then sidebands of 20 MHz were added by an electro-optic modulator (EOM, Thorlabs). Finally, a power of about 0.3 mW of the "locking" beam was coupled to the ring-down cavity. A Pound-Drever-Hall locking servo was used to lock the beam frequency f_L with the $n-$th longitudinal mode of the cavity. The "spectral" beam was also frequency shifted (f_{AOM2}) by another acousto-optic modulator (AOM2, AA MGAS80-A1) and then sidebands were added by a fiber electro-optic modulator (EOM, EOSPACE). The fiber EOM was driven by a tunable radio-frequency (f_{EOM}) source (R&S SMB100A). The "spectral" beam was coupled into the ring-down cavity from the other side of the cavity and the light transmitted from the cavity was detected by an avalanche photodiode. A Glan-Taylor prism was used to block the "locking" beam reflected from the cavity.

The frequency difference between the "locking" and "spectral" beam is set exactly equal to the difference between two longitudinal modes of the cavity:

$$f_S - f_L = f_{AOM2} + f_{EOM} - f_{AOM1} = f_{n+i} - f_n. \quad (1)$$

The absolute frequency of the ECDL laser was determined from the beat signal with an optical frequency comb. The repetition frequency (f_R) and carrier offset frequency (f_0) of the comb are locked to precise radio-frequency sources (Rigol DG4202) referenced to a GPS-disciplined rubidium clock (SRS FS725). The beat frequency between the probe laser and a tooth of the comb ($f_N = f_0 + Nf_R$) was locked to a preset frequency f_B by sending the feedback control to the piezo actuator attached on one of the high-reflective cavity mirrors. Therefore, the "spectral" beam frequency can be determined as:

$$f_S = f_0 + Nf_R + f_B + f_{AOM2} + f_{EOM}. \quad (2)$$

II. DOPPLER-BROADENED ABSORPTION SPECTRUM

Broad spectral scan was accomplished by tuning f_{EOM} used to drive the fiber EOM. AOM1 was bypassed during the broad scan. The frequencies used to drive AOM2 and the beat locking servo were fixed during the scan: $f_{AOM2} = 79.999$ MHz, and $f_B = 50.000$ MHz. The scan step is fixed equal to the free-spectral-range of the cavity to fulfill the requirement given in Eq. 1. The comb repetition frequency and carrier offset frequency were $f_R \approx 198.163$ MHz and $f_0 \approx 250$ MHz, respectively. The Doppler broadened spectrum of the R(1) line of HD at 7241.85 cm^{-1} was shown in Fig.2 of the *Main Text*.

In order to investigate the influence of the water lines, spectrum of wet air sample ($P = 3.5$ kPa) was recorded in the same method and the result is shown in Fig. 1. Four peaks are evident and a fit of the spectrum gives the line centers which are collected in Table I. The water line positions agree well with those given in the HITRAN database [1]. The frequency differences between each of the water lines and the HD R(1) line are also given in Table I. Note that the shortest distance between the water line to the HD R(1) line is 0.0145(7) cm^{-1} (435(21) MHz). Although we confirmed that very little (< 0.1 Pa) water vapor presented in the HD sample, there could be influence on the Doppler-broadened HD R(1) spectrum due to weak water absorption lines.

TABLE I. Water lines around 7241.8 cm^{-1} (unit: cm^{-1}).

	HITRAN [a]		This work	
Molecule	ν'	Intensity	ν	$\nu - \nu_{\mathrm{HD}}$
$H_2^{17}O$	7241.7749	1.476E-24	7241.7766(20)	−0.0728
$HD^{16}O$	7241.8355	7.746E-25	7241.8349(7)	−0.0145
$H_2^{16}O$	7241.9126	2.288E-26	7241.9131(9)	+0.0637
$H_2^{16}O$	7241.9529	2.784E-24	7241.9580(4)	+0.1086
HD	7241.8493	1.098E-28	7241.8494	

[a] Line positions and line intensities (in cm/molecule) under natural abundance are taken from the HITRAN database [1]. Note that for pure HD gas, the line intensity should be multiplied with a factor of 3200.

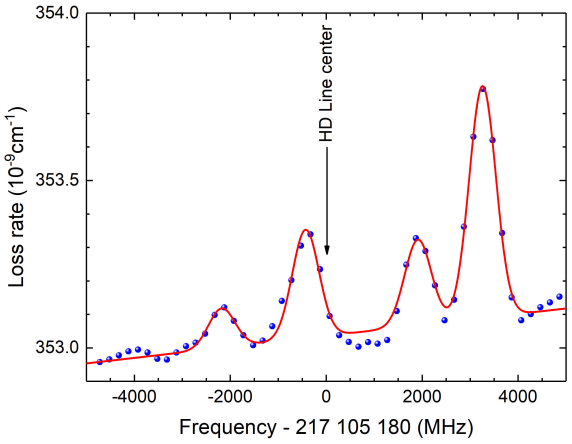

FIG. 1. The water lines recorded with a wet air sample. The dots present the experimental CRDS data while the continuous curve shows the simulated spectrum. The position of the HD R(1) line is indicated on the figure.

III. SATURATION ABSORPTION SPECTRUM OF HD

Fine spectral scan was accomplished by tuning the beat frequency between the probe laser and the comb. In order to reduce the power loss from the fiber EOM, in the Lamb-dip measurement of the HD R(1) line, the fiber EOM was removed. The "spectral" beam power coupled to the cavity was 15 mW. We have measured that the transmittance of an empty cavity was 10 %, and the ring-down time of the empty cavity was 95 μs. Using the equations given in Refs. [2, 3], we estimated that the intra-cavity laser power was about 200 W. The saturation intensity of the R(1) line can be calculated as [4]:

$$I_s = \frac{32\pi^3 hc}{3}\frac{\Gamma^2}{A\lambda^3}, \tag{3}$$

where h is the Planck constant, c is the speed of light, A is the Einstein coefficient, Γ is the width of the Lamb dip (HWHM), and λ is the wavelength of the transition. Taking the laser beam waist radius of $w = 0.46$ mm, we obtain a saturation power ($P_s = I_s \times \pi w^2/2$) of 99 kW at a Lamb-dip width of $\Gamma = 0.5$ MHz.

Note that the laser power is less than 1 % of the saturation power, which leads to a very small saturation parameter $S = I/I_s$, therefore the saturation broadening is negligible here, and the width of the Lamb dip has a linear dependence on the sample pressure:

$$\Gamma = \Gamma_0 + \gamma P. \tag{4}$$

A linear fit of the observed Lamb-dip width obtained at different pressures yields the γ coefficient of 0.035(9) MHz/Pa and the Γ_0 value of 0.34(7) MHz. The Lamb-dip width at the zero pressure limit is consistent with the transit-time broadening width of HD which is 0.52 MHz at 298 K. The depth of the Lamb dip (D) is dependent on the sample pressure and also the saturation parameter [2]:

$$D = \alpha_0\left(\frac{1}{\sqrt{1+S}} - \frac{1}{\sqrt{1+2S}}\right), \tag{5}$$

where α_0 is the linear absorption coefficient at the line center which can be derived from the Doppler-broadened spectrum. Using the parameters given above, we obtained the calculated Lamb-dip depth under different sample pressures which agree well with the experimental values.

IV. SATURATION ABSORPTION SPECTRUM OF C2H2

In order to check the consistency of the CRDS measurements, the saturation absorption spectroscopy of a nearby $^{12}C_2H_2$ line was measured under similar experimental conditions except that the HD sample was replaced by an acetylene sample of 0.2 Pa. The R(4) line in the $\nu_1 + \nu_2 + (2\nu_4 + \nu_5)^1$ band of $^{12}C_2H_2$ at 7239.79045 cm^{-1} [5] was selected. As given in the HITRAN database [1], the line intensity is 4.43×10^{-24} cm/molecule, and the Einstein coefficient is 7.55×10^{-3} s^{-1} (360 times of the HD 2-0 R(1) line). Fig. 2 shows the Lamb dip spectrum of the acetylene line recorded with an intra-cavity laser power of 50 W.

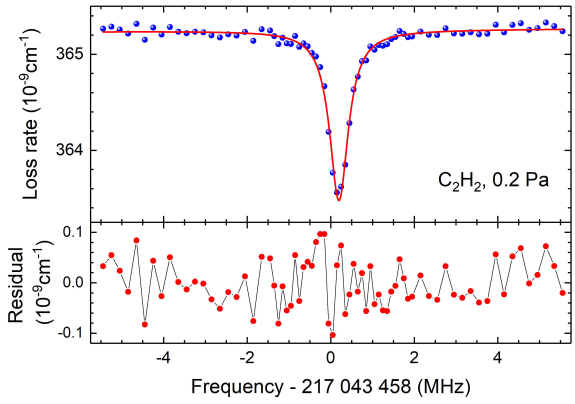

FIG. 2. Lamb-dip spectrum of the R(4) line of $^{12}C_2H_2$ recorded at a sample pressure of 0.2 Pa and an intra-cavity power of 50 W. A Lorentzian function was used to fit the spectrum. The lower panel shows the residual of the fit.

A Lorentzian function was used to fit the Lamb-dip spectrum and the simulated spectrum was also shown in the figure. The half width of the observed Lamb dip is 0.25 MHz, agreeing with the calculated transit-time broadening width of 0.18 MHz. Combined with the statistical uncertainty from a series of measurements, the position of the $^{12}C_2H_2$ line was determined to be 217 043 458.146(8) MHz (7239.7904735(3) cm^{-1}).

[1] I. E. Gordon, L. S. Rothman, C. Hill, R. V. Kochanov, Y. Tan, P. F. Bernath, M. Birk, V. Boudon, A. Campargue, K. V. Chance, B. J. Drouin, J. M. Flaud, R. R. Gamache, J. T. Hodges, D. Jacquemart, V. I. Perevalov, A. Perrin, K. P. Shine, M. A. H. Smith, J. Tennyson, G. C. Toon, H. Tran, V. G. Tyuterev, A. Barbe, A. G. Császár, V. M. Devi, T. Furtenbacher, J. J. Harrison, J. M. Hartmann, A. Jolly, T. J. Johnson, T. Karman, I. Kleiner, A. A. Kyuberis, J. Loos, O. M. Lyulin, S. T. Massie, S. N. Mikhailenko, N. Moazzen-Ahmadi, H. S. P. Müller, O. V. Naumenko, A. V. Nikitin, O. L. Polyansky, M. Rey, M. Rotger, S. W. Sharpe, K. Sung, E. Starikova, S. A. Tashkun, J. V. Auwera, G. Wagner, J. Wilzewski, P. Wcisło, S. Yu, and E. J. Zak, J. Quant. Spectrosc. Radiat. Transf. **203**, 3 (2017).

[2] L. S. Ma, J. Ye, P. Dube, and J. L. Hall, J. Opt. Soc. Am. B **16**, 2255 (1999).

[3] J. Wang, Y. R. Sun, L.-G. Tao, A.-W. Liu, T.-P. Hua, F. Meng, and S.-M. Hu, Rev. Sci. Instrum. **88**, 043108 (2017).

[4] G. Giusfredi, S. Bartalini, S. Borri, P. Cancio, I. Galli, D. Mazzotti, and P. De Natale, Phys. Rev. Lett. **104**, 110801 (2010).

[5] D. Jacquemart, N. Lacome, J. Y. Mandin, V. Dana, H. Tran, F. K. Gueye, O. M. Lyulin, V. I. Perevalov, and L. Régalia-Jarlot, J. Quant. Spectrosc. Radiat. Transf. **110**, 717 (2009).