# Toward a Determination of the Proton-Electron Mass Ratio from the Lamb-Dip Measurement of HD

L.-G. Tao,[1] A.-W. Liu,[1,2] K. Pachucki,[3] J. Komasa,[4] Y. R. Sun,[1,2] J. Wang,[1] and S.-M. Hu[1,2,*]

[1]*Hefei National Laboratory for Physical Sciences at Microscale, iChem center, University of Science and Technology of China, Hefei, 230026 China*
[2]*CAS Center for Excellence in Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei, 230026 China*
[3]*Faculty of Physics, University of Warsaw, Pasteura 5, 02-093, Warsaw, Poland*
[4]*Faculty of Chemistry, Adam Mickiewicz University, Umultowska 89b, 61-614 Poznań, Poland*

Precision spectroscopy of the hydrogen molecule is a test ground of quantum electrodynamics (QED), and it may serve for the determination of fundamental constants. Using a comb-locked cavity ring-down spectrometer, for the first time, we observed the Lamb-dip spectrum of the $R(1)$ line in the overtone of hydrogen deuteride (HD). The line position was determined to be $217\ 105\ 182.79 \pm 0.03_{\text{stat}} \pm 0.08_{\text{syst}}$ MHz ($\delta\nu/\nu = 4 \times 10^{-10}$), which is the most accurate rovibrational transition ever measured in the ground electronic state of molecular hydrogen. Moreover, from calculations including QED effects up to the order $m_e\alpha^6$, we obtained predictions for this $R(1)$ line as well as for the HD dissociation energy, which are less accurate but signaling the importance of the complete treatment of nonadiabatic effects. Provided that the theoretical calculation reaches the same accuracy, the present measurement will lead to a determination of the proton-to-electron mass ratio with a precision of 1.3 parts per billion.

$H_2$, $H_2^+$, and their deuterated isotopologues are the simplest molecules whose energy levels can be derived from the quantum electrodynamics (QED) theory using a few fundamental physical constants: the Rydberg constant, the fine structure constant, the proton(deuteron)-to-electron mass ratio, and the proton(deuteron) charge radius. The precision spectroscopy of molecular hydrogen has long been a test ground of the molecular theory [1–4] and QED [5,6]. A comparison of the experimental and theoretical energy levels of molecular hydrogen also sets constraints on some hypotheses beyond the Standard Model, such as the long-distance fifth force between two hadrons [7]. Having many long-lived rovibrational energy levels in the ground electronic states, the molecular hydrogen ion has been considered as a candidate for an optical clock [8]. Recently, an agreement at 1 ppb (part per billion) accuracy between the experimental measurements and theoretical calculations has been demonstrated for $HD^+$, which allows for a determination of the proton-to-electron mass ratio with an accuracy of 2.9 ppb [9].

It is more challenging to precisely calculate the energy levels of the four-body neutral hydrogen molecule than the three-body molecular hydrogen ion. In the last half century, the accuracy of calculations of $H_2$ (and its isotopologues) in the ground electronic state has been continuously improved [10–14], and a precision of $10^{-6}$ cm$^{-1}$ will be achievable in the near future [15,16]. If the rovibrational transition

frequencies of the hydrogen molecule are also measured with corresponding accuracy, it will lead to an improved determination of the proton-to-electron mass ratio $\mu_p \equiv m_p/m_e$. The present $\mu_p$ value recommended by 2014 CODATA [17] has an uncertainty of 0.095 ppb. However, a deviation of $3\sigma$ was observed by a recent measurement of the atomic mass of the proton [18], indicating that more measurements from various methods with comparable accuracies are needed for a consistency check of the constant.

In the electronic ground state, symmetric $H_2$ and $D_2$ molecules have only extremely weak quadrupole ($E2$) transitions, while HD exhibits weak dipole ($E1$) transitions due to nonadiabatic effects. Although extensive spectroscopy of molecular hydrogen has been carried out ([19] and references therein) since the pioneering work by Herzberg in 1949 [20], only Doppler-broadened spectra of the hydrogen molecule have been reported. Attempts to improve the accuracy using the Doppler-limited spectra have been carried out for a few lines [21,22], but the ambiguity in the line profile model may result in an uncertainty of several MHz [23]. Sub-MHz accuracy is only possible when the line shape has been carefully investigated. Doppler-free spectroscopy of the rovibrational transitions of molecular hydrogen has been hindered by the very small transition rates.

Here we present the first Lamb-dip measurement of a rovibrational transition of molecular hydrogen. The $R(1)$ line in the $v = 2$–0 band of HD has an Einstein coefficient

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of $2.1 \times 10^{-5}$ s$^{-1}$ [24], corresponding to a typical saturation intensity [25] of $10^7$ W cm$^{-2}$ at room temperature. Taking advantage of a high-finesse resonant cavity, we carried out saturation spectroscopy measurements using a continuous-wave diode laser with a power of only several tens of milliwatts. A sub-MHz linewidth was observed, and the line center was determined with a fractional uncertainty of $4 \times 10^{-10}$. Compared with the previous value obtained from Doppler-limited spectra [26], the accuracy has been improved by a factor of 300. This accuracy is, so far, the best among the rovibrational transitions of the hydrogen molecule in the electronic ground state including molecular hydrogen ions [9].

The experimental setup is close to the one used in our previous study [27], and the configuration is shown in Fig. 1. A brief introduction of the experimental method is given here, and more details are given in the Supplemental Material [28]. An external-cavity diode laser is used as the probe laser, being locked to a ring-down (RD) cavity using the Pound-Drever-Hall (PDH) method. The RD cavity is composed of a pair of high-reflectivity (HR) mirrors ($R = 99.998\%$), corresponding to a finesse of $1.2 \times 10^5$. The 80 cm-long RD cavity is temperature stabilized at 25 °C and the fluctuation is below 10 mK. The cavity length is stabilized through a piezo actuator (PZT) by a locking circuit driven by the beat signal between the probe laser and an optical frequency comb. The frequency comb is synthesized by an Er-fiber oscillator operated at 1.56 $\mu$m. Its repetition frequency ($f_R \approx 198$ MHz) and carrier offset frequency ($f_0$) are both referenced to a GPS-disciplined rubidium clock (SRS FS725). A separated beam from the probe laser, frequency shifted by an acousto-optic modulator (AOM) and a fiber electro-optic modulator (EOM), is coupled into the RD cavity from another side of the cavity and used for cavity ring-down spectroscopy (CRDS)

measurements. The AOM (AOM2 on Fig. 1) also serves as a beam chopper. Triggered by an external rectangle wave, it shuts off the CRDS-probing beam to initial a ring-down event. The light emitted from the cavity is detected and the signal is recorded by a data acquisition card. The ring-down curve is fit by an exponential decay function, and the sample absorption coefficient $\alpha$ is determined from the change of the cavity loss rate: $\alpha = (c\tau)^{-1} - (c\tau_0)^{-1}$, where $c$ is the speed of light, and $\tau$ and $\tau_0$ are decay times of the cavity with and without sample, respectively.

The $R(1)$ line in the 2-0 overtone band of HD is located at 7241.85 cm$^{-1}$, and the line intensity is $3.6 \times 10^{-25}$ cm molecule$^{-1}$ [26]. The HD sample (Sigma-Aldrich Co.) was purified by a liquid-N$_2$ trap before use. The Doppler-broadened spectrum were recorded at sample pressures of 125 Pa and 244 Pa and are shown in Fig. 2. By fitting the spectrum with a Gaussian function, we derived a line center of 217 105 181(2) MHz and a Gaussian width (half width at half maximum, HWHM) of 771 MHz, which agrees well with the calculated Doppler half width of 775 MHz. The uncertainty of the line position mainly comes from the parasitic optical interference ("fringes"), the collision effect [23], and the influence due to a few lines of water vapor ($\sim$0.1 Pa) that emitted from the walls of the RD cavity during the measurement. Since the fitting residual shown in Fig. 2 is below 0.1% of the height of the HD line, we can estimate that the deviation on the line center should be below 2 MHz, about 0.2% of the half linewidth.

Sample pressures below 30 Pa were used for Lamb-dip measurements. The laser power used for spectral probing was about 15 mW and the intracavity laser power was estimated [29,30] to be about 200 W, leading to a maximum
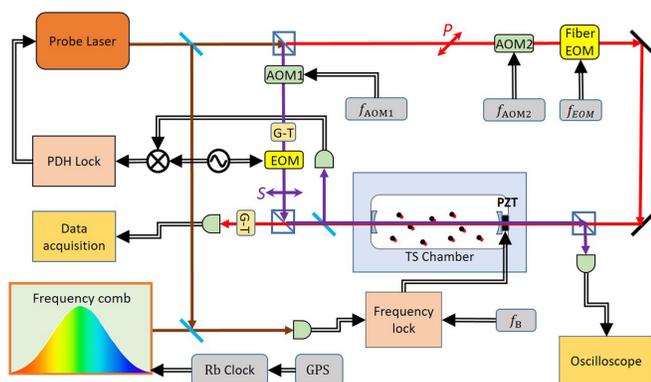


FIG. 1. Configuration of the experimental setup. The probe laser frequency is PDH-locked with the cavity. A beam from the probe laser is frequency shifted and used for CRDS measurements. The ring-down cavity length is locked by the beat signal between the probe and a frequency comb. Abbreviations: AOM: acousto-optic modulator; EOM: electro-optic modulator; G-T: Glan-Taylor prism; PZT: piezo actuator.
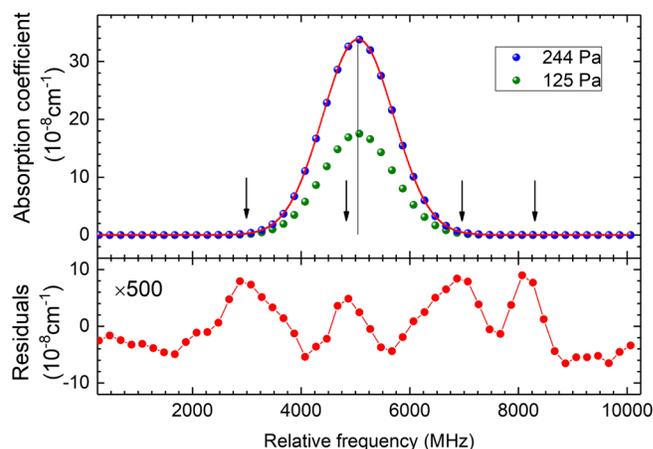


FIG. 2. Doppler-broadened cavity ring-down spectra of the $R(1)$ line in the 2-0 band of HD. The vertical line indicates the HD line center. The positions of the nearby weak water lines (see the Supplemental Material [28]) are marked with arrows on the figure. The lower panel shows fitting residuals of the spectrum recorded at 244 Pa using a Gaussian function. Note that the water lines were not included in the fit.
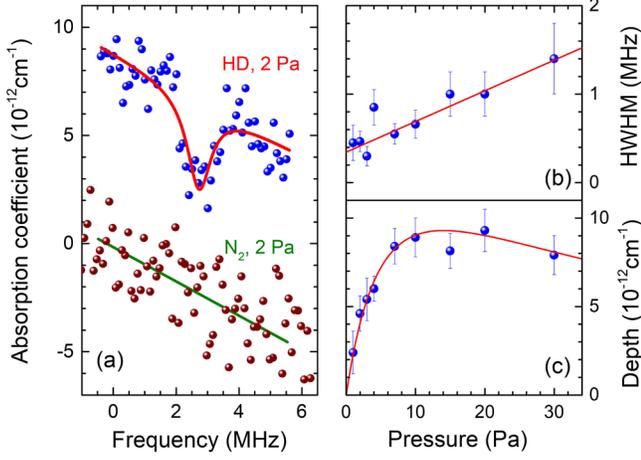
FIG. 3. Cavity ring-down spectra at 7241.8494 cm$^{-1}$ recorded with HD sample (a, upper) and pure nitrogen (a, lower). The spectra were shifted for better illustration. The Lamb dip of the $R(1)$ line was fit with a Lorentzian function. The width (HWHM, half width at half maximum) (b) and depth (c) of the Lamb dip vary with the sample pressure. For more discussions, see the Supplemental Material [28].

saturation parameter of about 0.2% with a laser beam waist radius of 0.46 mm. The spectrum recorded at a pressure of 2 Pa is shown in Fig. 3(a). It is an average of about 400 scans taken from a continuous measurement of about 12 hours. The Lamb dip of the $R(1)$ line has a width (HWHM) of about 0.4 MHz and a depth of about $5 \times 10^{-12}$ cm$^{-1}$. For comparison, the same experimental procedure was applied in a measurement with a nitrogen sample, and the recorded spectrum is also given in Fig. 3(a).

The Lamb-dip central position, width, and depth were derived from a fit of the spectrum using a Lorentzian function. A linear fit of the line widths obtained at different sample pressures yields a collision-induced broadening coefficient of 0.035(9) MHz Pa$^{-1}$. The linewidth at the zero pressure limit is 0.34(7) MHz, comparable to the calculated transit-time broadening width of 0.52 MHz at 298 K. The natural width, the laser line width (below 1 kHz), and power broadening are negligible. The depth of the Lamb-dip is proportional to the coefficient: $D \propto (1 + S)^{-1/2} - (1 + 2S)^{-1/2}$, where $S$ is the saturation parameter (see the Supplemental Material [28]).

In order to reduce the influence due to the collision-induced shift, the line center was determined from the spectra recorded with HD sample pressures of 1–4 Pa, as shown in Fig. 4. In this pressure region, no evidence of the pressure-induced shift has been observed. A statistical uncertainty of 0.03 MHz was obtained from 2600 scans recorded in 87 hours. A major systematic uncertainty arises from the possible asymmetry in the line profile that would lead to a bias on the line center derived from the fit. The asymmetry may come from the hyperfine structure, which was not resolved here but has been reported by Quinn *et al.* from the radio-frequency spectra of HD [31]. We have
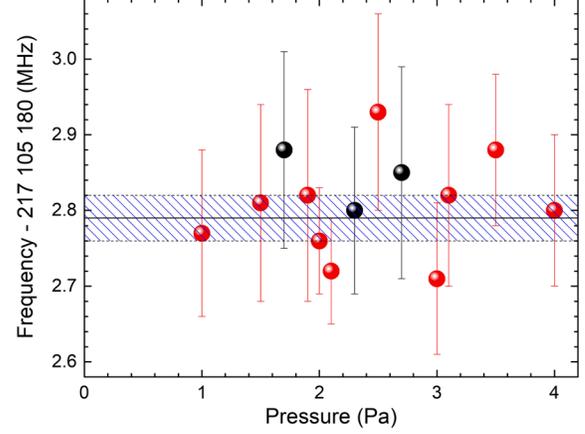


FIG. 4. $R(1)$ positions determined from spectra recorded at HD sample pressures of 1–4 Pa. Black and red points indicate measurements by switching the laser beams used for frequency locking and spectral probing. The region of shadow indicates the average value with $1\sigma$ uncertainty.

examined the low-pressure spectra and concluded that such asymmetry, if any, should be below the present noise level. Taking a signal-to-noise ratio of $5:1$ and a line width (HWHM) of 0.4 MHz, we give an uncertainty of 0.08 MHz due to the line profile model. Other contributions to the uncertainty budget are much smaller and negligible in this study. The laser frequency is calibrated by the frequency comb and, eventually, by the GPS-disciplined rubidium clock that has a fractional uncertainty of $2 \times 10^{-12}$ (0.4 kHz at 1.4 $\mu$m). The radio frequencies used to drive AOM(s) and EOM have a drift below 50 Hz. The second-order Doppler shift is 3 kHz. Note that beside the spectral laser beam scanning over the transition, another beam used to lock the laser, with a frequency shift of one free-spectral-range of the cavity, is also present in the cavity. We purposely switched between the two beams and repeated the measurement, but we found no difference (black and red points in Fig. 4) within the experimental uncertainty. The final value of the line position determined in this work is:

$$\nu_0 = 217105182.79(3)_{\text{stat}}(8)_{\text{syst}} \text{ MHz}$$
$$= 7241.849386(1)_{\text{stat}}(3)_{\text{syst}} \text{ cm}^{-1}. \quad (1)$$

The $R(1)$ line frequency determined in this study agrees with the value 7241.8497(10) cm$^{-1}$ derived from Doppler-limited spectra reported by Kassi *et al.* [26], while the accuracy has been improved by a factor of 300.

Our theoretical value, as given in Table I, amounts to 7241.84912(6) cm$^{-1}$. It was obtained as follows. The energy of a rovibrational level is expanded in powers of the fine-structure constant $\alpha$:

$$E = \sum_{n=2}^{\infty} E^{(n)}, \quad (2)$$

TABLE I. Calculated and experimental energies of HD.

|  | $D_0$, (0,0), cm$^{-1}$ | 2-0, $R(1)$, cm$^{-1}$ |
|---|---|---|
| $E^{(2)}$ | 36406.510839(1) | 7241.846169(1) |
| $E^{(4)}$ | −0.531325(1)(425) [a] | 0.040719(0)(32) [a] |
| $E^{(5)}$ | −0.1964(2)(2) [a] | −0.03743(4)(3) [a] |
| $E^{(6)}$ | −0.002080(6) | −0.000339 |
| $E^{(7)}$ | 0.00012(6) | 0.000021 |
| $E_{\rm FS}$ [b] | −0.000117 | −0.000021 |
| Total | 36405.7810(5) | 7241.84912(6) |
| Expt. | 36405.78366(36) [c] | 7241.849386(3) |
| Diff. | 0.0026 | 0.00027 |

[a]Additional uncertainty in terms $E^{(4)}$ and $E^{(5)}$ due to nonadiabatic corrections is estimated to be the value multiplied with a factor of $8 \times 10^{-4}$.
[b]$E_{\rm FS}$ is the finite nuclear size correction.
[c]From Ref. [33].

where each $E^{(n)}$ is proportional to $\alpha^n$ (and may contain $\ln \alpha$). The leading term of this expansion, $E^{(2)}$ is the nonrelativistic energy. It was calculated without any approximations, using a nonadiabatic explicitly correlated wave function, with a numerical uncertainty of $10^{-6}$ cm$^{-1}$. This is the part that has been significantly improved with respect to previous studies [13]. Other expansion terms in Eq. (2) were calculated within the adiabatic approximation. The next term $E^{(3)}$ is absent, $E^{(4)}$ is the relativistic correction [16], $E^{(5)}$ is the QED correction [32], and the terms with $n \geq 6$ constitute higher-order relativistic and QED corrections. The recent accurate calculation of $E^{(6)}$ [14] was a significant step in achieving high-precision theoretical predictions. Although numerical uncertainties in $E^{(n)}$ are at the order of $10^{-5}$ cm$^{-1}$ or less, as shown in Table I, the discrepancies with the experiment for the dissociation energy [33] and the $R(1)$ transition frequency are 0.0026 cm$^{-1}$ and 0.00027 cm$^{-1}$, respectively. They are both about five times the estimated theoretical uncertainty given here. One of the most probable reasons is the underestimation of relativistic nonadiabatic effects. A preliminary estimate of these effects is $E^{(4)}$ times the electron-nuclear mass ratio, which is about 10 times smaller than the discrepancy.

In the calculation, we used the CODATA recommended values [17] of the following constants: the Rydberg constant $R_y = 109737.31568508(65)$ cm$^{-1}$, the fine-structure constant $\alpha = 0.0072973525664(17)$, and the proton- or deuteron-to-electron mass ratios $\mu_p = 1836.15267389(17)$, $\mu_d \equiv m_d/m_e = 3670.48296785(13)$. For the proton and deuteron charge radii, we used the values from the muonic hydrogen measurements [34]: $r_p = 0.840087(39)$ fm and $r_d = 2.12771(22)$ fm. The deviation in the HD transition frequency $\nu$ can be translated to deviations of the physical constants:

$$\frac{d\nu}{\nu} = \beta_{R_y}\frac{dR_y}{R_y} + \beta_\alpha\frac{d\alpha}{\alpha} + \beta_{\mu_p}\frac{d\mu_p}{\mu_p} + \beta_{\mu_d}\frac{d\mu_d}{\mu_d} + \beta_{r^2}\frac{dr^2}{r^2}, \quad (3)$$

where $r^2 = r_p^2 + r_d^2$. For the 2-0 $R(1)$ transition of HD, the $\beta$ coefficients are: $\beta_{R_y} = 1$, $\beta_\alpha = -4.3 \times 10^{-6}$, $\beta_{\mu_p} = -0.31$, $\beta_{\mu_d} = -0.060$, and $\beta_{r^2} = -2.9 \times 10^{-9}$. Taking into account the relative uncertainties of these constants, the most significant term in Eq. (3) is $\beta_{\mu_p}(d\mu_p/\mu_p)$. Therefore, the transition frequency measured in this work could lead to a determination of the $\mu_p$ value with an uncertainty of 1.3 ppb if the theoretical calculation reaches the corresponding precision.

Note that the current experimental accuracy is mainly limited by the line width due to transit-time broadening. The accuracy could be considerably improved by conducting cavity-enhanced saturation spectroscopy with the sample gas cooled to a few Kelvin by buffer-gas cooling [35]. In this case, the width of the Lamb dip would decrease by an order of magnitude. Moreover, the reduced line width will also reduce the saturation power of the transition and result in an improved signal-to-noise ratio in the Lamb-dip measurement. Therefore, a fractional accuracy of $10^{-12}$ of the HD transition frequency can be expected. On the theoretical side, it has been recently demonstrated that the numerical solution of the Schrödinger equation of molecular hydrogen can be as accurate as $10^{-12}$, which paves the way for using the energy levels of molecular hydrogen to determine other physical constants in Eq. (3), such as the Rydberg constant and the proton charge radius [14,16], similar to their determination from the spectroscopy of atomic hydrogen [36].

*Note added.*—In parallel to the present study, the noise-immune cavity-enhanced optical heterodyne molecular spectroscopy (NICE-OHMS) method was applied at Amsterdam to search the Lamb-dip spectrum of HD in the same spectral region. Recently, we were aware of the results reported from both groups. There is a deviation of 0.90 MHz between the $R(1)$ position obtained from NICE-OHMS [37] and that given here, about ten times the combined experimental uncertainty. In order to check the experimental consistency, both groups recorded the saturation spectra of a $C_2H_2$ line at 7239.79 cm$^{-1}$, which is close to the HD $R(1)$ line but stronger by a factor of 360 [38]. The $C_2H_2$ line position determined by CRDS is 217 043 458.146(8) MHz (see the Supplemental Material [28]), and the NICE-OHMS value is 217 043 458.139(6) MHz. The agreement implies that the discrepancy in the HD $R(1)$ positions is not from the metrology applied in both studies. The origin of the

discrepancy is still unknown and further investigation of the spectroscopy of the hydrogen molecule is needed.

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*Corresponding author.
 smhu@ustc.edu.cn

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