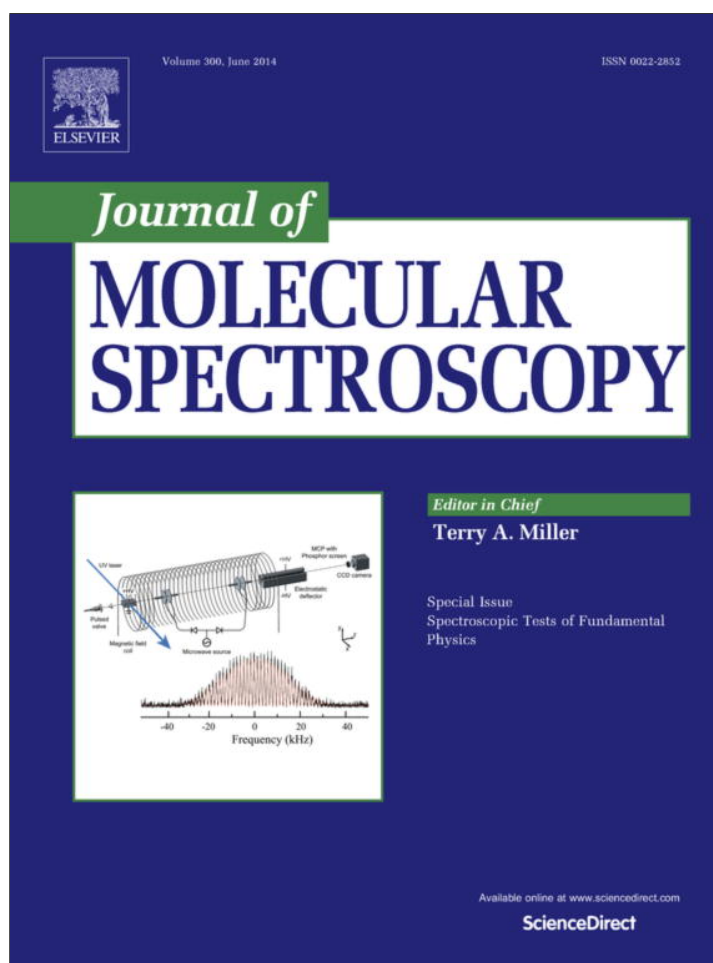


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## Journal of Molecular Spectroscopy

journal homepage: [www.elsevier.com/locate/jms](http://www.elsevier.com/locate/jms)Cavity ring-down spectroscopy of the electric quadrupole transitions of H<sub>2</sub> in the 784–852 nm region

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## ABSTRACT

Precision spectroscopy of the hydrogen molecule can be used to verify the quantum electrodynamics theory (QED) in a molecular system. The electric quadrupole transitions of the second overtone of H<sub>2</sub> have been recorded with a continuous-wave cavity ring-down spectrometer. In total, eight lines in the 784–852 nm range have been observed, including the extremely weak S<sub>3</sub>(5) line with a calculated intensity of  $9.2 \times 10^{-31}$  cm/molecule. The absolute line positions at the zero pressure limit have been determined with an accuracy of  $3 \times 10^{-4}$  cm<sup>-1</sup>, agreeing well with the high-level quantum chemical calculations including relativistic and QED corrections. The deviations between the experimental and theoretical frequencies are less than  $5 \times 10^{-4}$  cm<sup>-1</sup>, which is only 20% of the claimed theoretical uncertainty.

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## 1. Introduction

The hydrogen molecule is the smallest neutral molecule. It is particularly interesting in testing quantum chemistry theories taking into account high-order relativistic and quantum electrodynamics (QED) corrections. The ro-vibrational energy levels of H<sub>2</sub> (and its isotopologues) have been calculated with a relative accuracy at the 10<sup>-8</sup> level without using any adjustable parameters [1–5]. A comparable experimental accuracy has also been achieved in the last decade [6–10]. Direct comparison of the theoretical and experimental energy levels of the hydrogen molecule allows investigating the QED and relativistic contributions [11], and potentially to refine some fundamental physical constants like the proton-to-electron mass ratio [12].

The hydrogen molecule is also the most abundant neutral molecule in the universe. Its spectrum has been used in the studies of the atmosphere of planets, cold stars, and interstellar clouds [13–15]. The line positions of H<sub>2</sub> can also be applied to find the cosmological variation of the proton-to-electron mass ratio. A variation at the 3.5σ confidence level has been observed by Reinhold et al. [16] using the Lyman band lines of H<sub>2</sub> with a position accuracy of  $5 \times 10^{-8}$ .

As a homonuclear diatomic molecule, H<sub>2</sub> has only very weak electric quadrupole transitions in the infrared and visible region.

Since the pioneering work by Herzberg in 1949 [14], extensive laboratory studies have been carried out in last few decades. A review of these studies has been presented recently by Campargue et al. [17]. It is worth noting that the previous experimental results have less precision than the theoretical results. For the second overtone band of H<sub>2</sub> near 0.8 μm, positions of some lines have been measured with a relative precision of 10<sup>-7</sup> by Bragg et al. [18] using Fourier-transform spectroscopy, and with a relative precision of 10<sup>-6</sup> by Robie and Hodges [19] using pulsed cavity ring-down spectroscopy (CRDS). We have recently reported the line parameters of the S<sub>3</sub>(J) (J = 0–3) lines [20] using continuous-wave (cw) CRDS. The obtained positions of these lines agree with the calculated results within the experimental uncertainty of 0.002 cm<sup>-1</sup>. We also determined the frequency of the S<sub>3</sub>(3) line at 796 nm with an accuracy of  $5 \times 10^{-5}$  cm<sup>-1</sup> (relatively  $4 \times 10^{-9}$ ) [21] referenced to an accurate <sup>87</sup>Rb transition at 795 nm.

In this work, we extend the spectral range to cover altogether eight quadrupole transitions of H<sub>2</sub> in the 784–852 nm range. The extremely weak S<sub>3</sub>(4) and S<sub>3</sub>(5) lines were detected for the first time, and they are among the weakest molecular lines detected by absorption in the gas phase. Using the passive modes of a thermo-stabilized Fabry–Pérot interferometer referenced to a few precise atomic transitions in this range, we obtained the line positions with an accuracy of  $3 \times 10^{-4}$  cm<sup>-1</sup>. It allows us to examine the rotational energies in the vibrationally excited state. The experimental line positions agree with the theoretical values within a deviation of  $5 \times 10^{-4}$  cm<sup>-1</sup>, which implies that the theoretical uncertainty (0.0025 cm<sup>-1</sup>) might be over-estimated.

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## 2. Experimental details

The H<sub>2</sub> lines were recorded by a cavity ring-down spectrometer based on a continuous-wave Ti:Sapphire laser. The configuration of the spectrometer is close to that presented in our previous studies [21,22]. The Ti:Sapphire laser (Coherent 899–21) has been replaced with a new model, Coherent MBR-110, which offers a wider spectral range. But we also detected a considerable nonlinearity in its spectral scanning. The nonlinearity results in a 10 MHz limit on the spectral accuracy, which will be discussed later. The 1 m long ring-down cavity is composed of a pair of mirrors (Layertec GmbH) with a reflectivity of 99.995%. The mirrors are installed in a high-vacuum chamber and can be precisely adjusted by a set of step-motors (New Focus Picomotor) through a controller outside the chamber. The ring-down signal is detected by a photo-diode and recorded by an analog–digital converter (ADLink PCI 9228) installed in a personal computer. A nonlinear least-square fitting program is applied to fit the data to derive the ring-down time  $\tau$ . The sample absorption coefficient  $\alpha$  at the frequency  $\nu$  is obtained using the equation:

$$\alpha(\nu) = \frac{1}{c\tau(\nu)} - \frac{1}{c\tau_0} \quad (1)$$

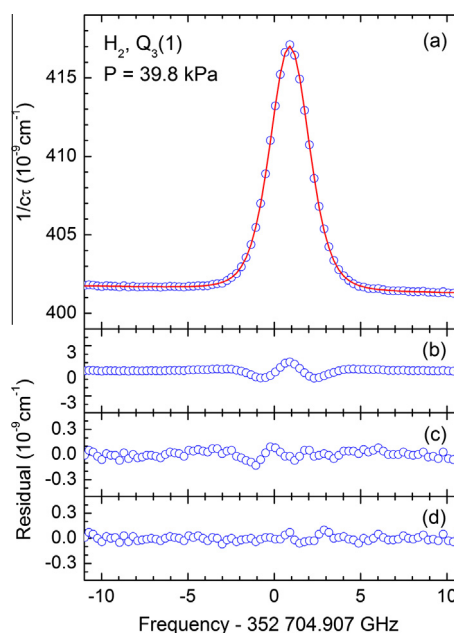
where  $c$  is the speed of light and  $\tau_0$  is the ring-down time of the empty cavity. The typical noise-equivalent absorption coefficient is  $1 \times 10^{-10} \text{ cm}^{-1}$ .

The spectrum is calibrated by using the passive modes of a Fabry–Pérot interferometer (FPI) made of ultra-low-expansion glass (ULE). The ULE-FPI (ATFilms Inc.) is used as relative frequency markers, and some atomic lines of Rb and Cs are used as absolute frequency standards. The ULE-FPI used in this study is a new one different from that used in previous studies [21,22]. It has a flatter reflectivity throughout the 750–850 nm region. The 10-cm-long ULE-FPI is installed in a vacuum chamber, thermo-stabilized with a temperature drift less than 10 mK at 302 K, the “magic-temperature” of the interferometer, where it has a near-zero expansion coefficient. The frequency of a given longitudinal mode of the ULE-FPI can be calculated from the equation:

$$\nu_N = \nu_0 + Nf_{\text{FSR}} \quad (2)$$

where  $N$  is the index of the mode, defined to be zero for the mode with a frequency ( $\nu_0$ ) of 377 104 082 MHz, and  $f_{\text{FSR}} = 1497.98645$  MHz, being the free-spectral-range (FSR) of the ULE-FPI. The values of  $f_{\text{FSR}}$  and  $\nu_0$  have been determined from saturation spectroscopy of a few atomic transitions of Rb and Cs. The method is the same as that presented in our previous study [22]. Three atomic lines have been used as frequency standards: the 795 nm line [23] and 780 nm line [24] of <sup>87</sup>Rb, and the 852 nm line [25] of <sup>133</sup>Cs. Taking into account the uncertainties from the saturation spectroscopy, the coating dispersion of the FPI, and the uncertainty in the atomic frequency standards, we estimate that the uncertainty in  $\nu_N$  should be below 3 MHz in the spectral range of 780–852 nm.

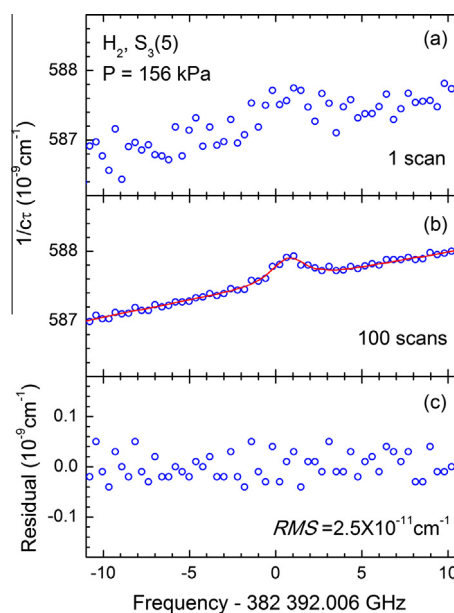
The electric quadrupole lines of H<sub>2</sub> in the 784–852 nm region were recorded at room temperature ( $296 \pm 1$  K). For each line, the spectrum was recorded at different sample pressures in the range of 40–160 kPa. The hydrogen gas was bought from Nanjing Special Gas Co., with a nominal purity of 99.99%. The sample pressure was measured by a capacitance gauge with an accuracy of 0.5%. An example of the obtained spectrum, the Q<sub>3</sub>(1) line at  $11765.0009 \text{ cm}^{-1}$  recorded at 39.8 kPa sample pressure, is presented in Fig. 1(a).



**Fig. 1.** The Q<sub>3</sub>(1) line of H<sub>2</sub> near 850 nm recorded at 39.8 kPa. (a) The observed spectrum (open circles), and the simulated spectrum (solid curve) using a Galatry profile. (b)–(d) are the fitting residuals using Voigt, Rautian and Galatry profiles, respectively.

## 3. Results and discussion

Altogether eight lines of the (3–0) band of H<sub>2</sub> were recorded, including six lines in the S branch and two lines in the Q branch. The S<sub>3</sub>(0), S<sub>3</sub>(1), S<sub>3</sub>(2) and S<sub>3</sub>(3) line positions have been reported in our previous study [20] with an accuracy of  $0.002 \text{ cm}^{-1}$ . Extended spectral coverage and improved calibration method allow us to detect more lines with a better precision. To the best of our knowledge, the S(5) line near 784 nm, with a calculated intensity of  $9.2 \times 10^{-31} \text{ cm}^2/\text{molecule}$ , is the weakest H<sub>2</sub> line observed by



**Fig. 2.** The S<sub>3</sub>(5) line of H<sub>2</sub> near 784 nm. Sample pressure: 156 kPa. (a) Spectrum obtained from 1 scan. (b) Averaged spectrum from 100 scans. Open circles and the solid line are for the observed and simulated spectrum, respectively. (c) Fitting residuals. The root-mean-square value of the residuals is  $2.5 \times 10^{-11} \text{ cm}^{-1}$ .

absorption. The spectrum of the  $S_3(5)$  line recorded with a sample pressure of 156 kPa is shown in Fig. 2.

Line parameters were derived from a fit of each line profile recorded at different sample pressures. As a result of the collision effects, the profile cannot be reproduced by the conventional Voigt line shape which is illustrated in Fig. 1(b). The “hard” collisional Rautian model [26] and the “soft” collisional Galatry model [27] are two of the most frequently applied line profiles which take into account the effects of Doppler broadening, Dicke narrowing and collision broadening. As shown in Fig. 1, the Galatry model gives the best fit of the observed spectrum. In the fitting, the Gaussian width was fixed at the calculated Doppler width value, and the narrowing parameter  $\beta$  was calculated by the diffusion theory from the equation [28]:

$$\beta = \frac{k_B T}{2\pi m c D} \quad (3)$$

where  $k_B$  is the Boltzmann constant,  $T$  is the temperature,  $m$  is the molecular mass,  $c$  is the speed of light, and  $D$  is the mass-diffusion coefficient of  $H_2$  which can be found in Ref. [29]. Other line parameters, including the line position, intensity, and Lorentzian width were derived from the fit.

A major source of the uncertainty on the line positions is the nonlinearity in the spectral scanning, which is illustrated in Fig. 3. We step-scanned the laser (Coherent MBR 110) by applying a voltage on its external-scan port. The frequency of the laser emission was determined from the beat signal with a second laser which was frequency-locked on a passive mode of the ULE-FPI. Fig. 3(a) shows the measured beat frequency changing with the applied voltage. Fig. 3(b) presents the residuals from a linear fit and a quadratic fit of the data. The curves show irregular fluctuations, which cannot be fitted unless a physically improper high-order polynomial function was used. Since the fluctuation amplitude can be as large as 10 MHz within an interval of 1.5 GHz (FSR of the ULE-FPI), it induces an uncertainty of 10 MHz in the derived line positions.

Fig. 4 shows the central frequencies relative to the frequency markers (ULE-FPI modes) of each  $H_2$  line obtained at different sample pressures. For each line, the line position at the zero pressure limit and the pressure induced shift coefficient are derived from a linear fit of the positions obtained at different sample pressures. The results for all the eight lines are collected in Table 1. The accuracy of the obtained line positions at the zero pressure limit is mainly limited by the nonlinearity in the laser scanning which is estimated to be 10 MHz, except that the accuracy for  $S_3(4)$  and

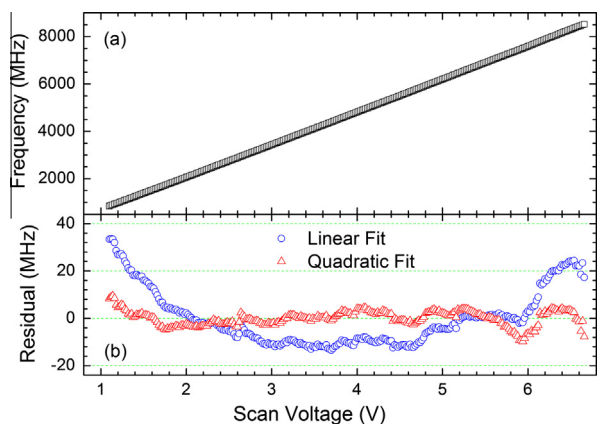


Fig. 3. Nonlinearity in the spectral scanning of the laser (Coherent MBR 110). (a) Laser frequency varying with the applied external voltage. The frequency was measured from the beat signal with a frequency-locked laser. (b) Residuals from a linear fit (circles) and from a quadratic fit (triangles).

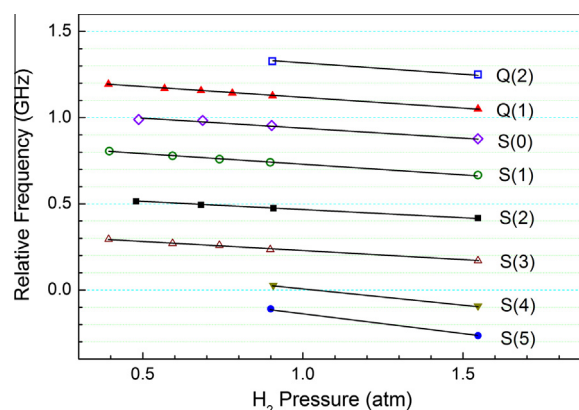


Fig. 4. Relative central frequencies of the  $H_2$  lines obtained at different sample pressures. Data have been shifted for a better visualization.

$S_3(5)$  is mainly limited by the signal-to-noise level of the observed spectrum. The  $S_3(3)$  frequency given here agrees with the value  $12559.74952(5) \text{ cm}^{-1}$  from our previous study [21], but the uncertainty obtained in this work is several times larger. The main reason is that the scan nonlinearity of the laser we used here (Coherent MBR 110) is considerably larger than the one used in previous work (Coherent 899-21).

A comparison of the experimental and theoretical [17] line positions and intensities is shown in Fig. 5. In general, the obtained line frequencies agree perfectly with the theoretically calculated values, where QED corrections to the order of  $\alpha^4$  ( $\alpha$  is the fine structure constant) have been included [5]. The differences between the experimental and calculated values are less than  $5 \times 10^{-4} \text{ cm}^{-1}$  (15 MHz) except for the very weak  $S(4)$  and  $S(5)$  lines. The difference is consistent with our experimental uncertainty but only about 20% of the claimed theoretical accuracy (75 MHz) [17]. Note that the  $Q_3(2)$  and  $S_3(0)$  lines share the same upper level of ( $\nu'' = 3, J'' = 2$ ). Since the energy of the ( $\nu'' = 0, J'' = 0$ ) level is defined as zero, the frequency difference between  $Q_3(2)$  and  $S_3(0)$  gives the energy of the ( $\nu'' = 0, J'' = 2$ ) level, which is  $354.3730(7) \text{ cm}^{-1}$ . It agrees well with the theoretical value [17] of  $354.37313(3) \text{ cm}^{-1}$ , and also the value  $354.37297(11) \text{ cm}^{-1}$  from Raman spectroscopy of the  $S_0(0)$  transition in the (0–0) band by Michaut et al. [30].

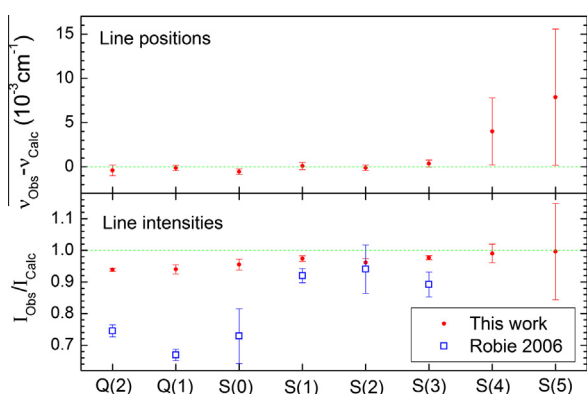
Using the rotational energy levels calculated by Komasa et al. [5], we can derive the energies of the upper levels in the  $\nu = 3$  vibrational state. The differences between the experimental and calculated energies are given in Table 2. For the  $J = 0 - 5$  levels, the differences are within  $5 \times 10^{-4} \text{ cm}^{-1}$ , consistent with the experimental uncertainties, but much smaller the stated theoretical uncertainties. Note that the stated theoretical uncertainties of the rotational energies in the vibrational ground state are below  $0.0001 \text{ cm}^{-1}$  for the  $J = 0 - 5$  levels [5], the uncertainties of the derived upper energies are mainly from the experimental uncertainties in this work. The analysis above confirms the experimental accuracy of this work, and also implies that the uncertainty on the calculated upper energy levels might have been over-estimated in Refs. [5,17].

The determined line intensities are also given in Table 1. For comparison, the experimental results presented in Refs. [18,19] are also given. The deviation between the present work and CRDS measurements by Robie and Hodges [19] is considerably larger than the combined uncertainties, particularly for the  $Q_3(1)$  and  $Q_3(2)$  lines. Our results agree well with the calculated values [17], which implies that the uncertainty given in Ref. [19] might be largely underestimated. As shown in Fig. 5, the relative deviations between the experimental and theoretical line intensities vary between  $-5\%$  and  $-2\%$  for all the observed lines except for



**Table 1**Line parameters of the observed quadrupole transitions of the (3–0) band of H<sub>2</sub>. The uncertainties given in parentheses are in unit of the last quoted digit.

Line	Position, cm <sup>-1</sup>			$\gamma \times 1000^c$ cm <sup>-1</sup> /atm	Intensity, 10 <sup>-30</sup> cm/molecule			
	Calc. <sup>a</sup>	This work	Ref. [18] <sup>b</sup>		Calc. <sup>a</sup>	This work	Ref. [18]	Ref. [19]
Q(2)	11730.3239	.3235(6)		-4.0(6)	20.95	19.6(2)		15.6(4)
Q(1)	11765.0009	.0008(3)		-4.0(1)	164.5	154.4(22)		110(3)
S(0)	12084.6970	.6965(3)	.6872(23)	-3.7(4)	127.7	121.9(21)	104(7)	93(11)
S(1)	12265.5949	.5950(4)	.5831(7)	-3.8(1)	494.0	480.7(41)	450(30)	454(11)
S(2)	12424.4421	.4420(3)	.4346(37)	-3.0(1)	91.50	87.9(10)	93(7)	86(7)
S(3)	12559.7492	.7496(4) <sup>d</sup>	.7424(59)	-3.5(2)	76.28	74.4(5)	67(11)	68(3)
S(4)	12670.3158	.3198(38)		-6(4)	3.76	3.7(1)		
S(5)	12755.2535	.2614(77)		-8(6)	0.92	0.9(1)		

<sup>a</sup> Calculated values from [17]. Stated theoretical uncertainty on line positions: 0.0025 cm<sup>-1</sup>, relative uncertainty on line intensities: 0.1%.<sup>b</sup> Line positions from Ref. [18], obtained at a sample pressure of 2.8 atm. Note that other positions given in the table are at the zero pressure limit.<sup>c</sup> Collision-induced shift coefficient (296 K) obtained in this work.<sup>d</sup> A more precise line position of 12559.74952(5) cm<sup>-1</sup> has been given in Ref. [21].**Fig. 5.** Comparison of the line positions and intensities between the experimental results and the calculated values (Ref. [17]). The line intensities given by Robie and Hodges [19] are also shown (open squares). See discussion in text.**Table 2**Ro-vibrational energy levels of the  $\nu = 3$  state of H<sub>2</sub>, in cm<sup>-1</sup>.

$J$	$E_{calc}, \nu = 0$	$E_{calc}, \nu = 3$	$E_{exp}, \nu = 3$	$\Delta E^a$
0	0	11782.3908(25)		
1	118.486812(9)	11883.4877(25)	11883.4876(3)	-1
2	354.37313(3)	12084.6970(25)	12084.6965(3)	-5
3	705.51883(5)	12384.0817(25)	12384.0818(4)	1
4	1168.79743(9)	12778.8152(25)	12778.8151(3)	-1
5	1740.1895(1)	13265.2681(25)	13265.2684(4)	3
6	2414.8970(2)	13839.1133(25)	13839.1172(38)	39
7	3187.4705(3)	14495.4431(25)	14495.4509(77)	78

<sup>a</sup>  $\Delta E = E_{exp} - E_{calc}$ , in 10<sup>-4</sup> cm<sup>-1</sup>. Calculated values are from Ref. [5] calculated energies in the vibrational ground state were used to derive the experimental upper state energies.

the very weak S<sub>3</sub>(5) line. Our typical experimental uncertainty is about 1% including the uncertainties from the sample pressures, temperatures, and linear fit of the results from different pressures. The estimated uncertainty on the calculated line intensities is 0.1% [17]. A possible reason of the discrepancy between the theoretical and experimental results is that the Galatry profile applied in the spectral fitting may be improper to describe accurately the collision induced spectral profile of H<sub>2</sub>. Studies using other more sophisticated profiles taking into account the speed-dependant effects in the collisions [31] may be helpful for further investigation on this topic.

#### 4. Conclusion

A total of eight electric-quadrupole lines of H<sub>2</sub> in the  $\nu = 3 \leftarrow 0$  band, Q<sub>3</sub>(1), Q<sub>3</sub>(2), and S<sub>3</sub>(0)–S<sub>3</sub>(5), have been recorded by a cavity

ring-down spectrometer. The line positions have been determined to an accuracy of  $3 \times 10^{-4}$  cm<sup>-1</sup> and the line intensities were determined with a relative accuracy of about 1%. The experimental line positions agree well with the *ab initio* calculations which have included the high-order QED corrections to the order of  $\alpha^4$ . For all the transitions except the very weak lines S<sub>3</sub>(4) and S<sub>3</sub>(5), the deviations are less than  $5 \times 10^{-4}$  cm<sup>-1</sup>, much smaller than the claimed theoretical uncertainty of 0.0025 cm<sup>-1</sup>, implying that the later one might be overestimated.

The experimental accuracy of the line positions obtained in this work is mainly limited by the nonlinearity in the laser scanning and the frequency calibration. It can be improved to the kHz level by using CRDS based on a laser locked to a set of frequency combs [32]. Therefore, the ro-vibrational energies of H<sub>2</sub> can be potentially determined with a relative accuracy of 10<sup>-10</sup>. Note that the present accuracies of the fine structure constant  $\alpha$  and the proton-to-electron mass ratio  $m_p/m_e$  are both at the 10<sup>-10</sup> level [33], precision spectroscopy of H<sub>2</sub> may provide a new opportunity to refine these fundamental constants [12], and to test the QED theory [34], in a molecular system.

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