Highly Accurate H$_2$ Lyman and Werner Band Laboratory Measurements and an Improved Constraint on a Cosmological Variation of the Proton-to-Electron Mass Ratio

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Transition wavelengths on a large set of H$_2$ Lyman and Werner band spectral lines have been obtained at an accuracy of 5×10^{-8}, using a narrow band tunable extreme ultraviolet laser. The data are used to determine a constraint on a possible cosmological variation of the proton-to-electron mass ratio ($\mu = M_p/m_e$) from a comparison with highly redshifted spectral data of quasistellar objects, yielding a fractional change in the mass ratio of $\Delta\mu/\mu = -0.5 \pm 3.6 \times 10^{-5}$ (2σ), which would correspond to a temporal change of $d/dt(\Delta\mu/\mu) = -0.4 \pm 3.0 \times 10^{-15}$ per year (2σ) if a linear cosmological expansion model is assumed.

The search for a cosmological variability of fundamental physical constants has become an active field of research now that accurate spectroscopic data can be obtained from quasars that have emitted their radiation more than ten billion years ago. A comparison between laboratory data, obtained at the highest accuracy in the modern epoch, and analysis of corresponding redshifted spectra from distant objects allows for stringent constraints on variability of the physical constants underlying the spectra. One such issue is that of the fine structure constant α, which was claimed to vary based on observations of quasar spectra [1]. Another is that of the proton-to-electron mass ratio $\mu = M_p/m_e$, a dimensionless parameter equalling 1836.152 672 61(85) [2]. The idea that fundamental physical constants might not be constant on a cosmological time scale was pioneered by Dirac, Teller, Gamov, and others; the observational status and theoretical perspectives were recently discussed by Uzan [3], citing all relevant publications.

The spectrum of molecular hydrogen with the prominent Lyman and Werner band systems may be used to detect a change in μ over time, because each individual spectral line depends in a different way on the mass ratio, as was pointed out by Thompson [4]. In recent years, several observations have been made on the Lyman and Werner bands in quasars, taking advantage of the fact that Earth's atmosphere is transparent for these spectral regions at high z, and that ground-based echelle-grating spectrometers can be employed in high-resolution studies [5–10]. In comparisons, the laboratory data of the Lyman and Werner band tables of the Meudon group were used [11,12], which represent an extensive and accurate database. Nevertheless, it was emphasized that improved spectroscopic data are desired [8], because the statistical errors from astronomical and laboratory data contributed equally in the estimates of $\Delta\mu/\mu$, the fractional variation

of μ. This is illustrated by the fact that Ivanchik *et al.* [8] find rather different constraints on $\Delta\mu/\mu$, when comparing quasar data with different laboratory data sets [11–13]. Here we report on laser-based laboratory determinations of Lyman and Werner band spectral lines at an accuracy improved by more than an order of magnitude; the resulting laboratory wavelengths λ_i^0 can be considered *exact* for the purpose of comparison with quasar data.

Spectral lines of Lyman and Werner band transitions were investigated with the use of a tunable narrow band extreme ultraviolet (XUV) laser source in the range 91–108 nm. Data were recorded by performing 1 XUV + 1 UV resonance-enhanced photoionization under nearly Doppler-free conditions in a molecular beam. Since the experiment involves generation of the sixth harmonic, the absolute frequency metrology could be performed with respect to a reference standard of I$_2$ lines, measured in saturated absorption in the visible domain. An example of a very high-resolution molecular beam recording of a hydrogen resonance is shown in Fig. 1. It shows simultaneous recordings of the $R(0)$ line in the $B^1\Sigma_u^+$-$X^1\Sigma_g^+$ (4,0) Lyman band of H$_2$ at the sixth harmonic of the fundamental laser frequency (after pulse amplification of the output of a single-mode continuous wave ring laser, frequency doubling in a nonlinear crystal, and third-harmonic generation in a gas jet of xenon) and the marker fringes of an actively stabilized interferometer (FSR = 148.9567 MHz), as well as a recording of a saturated spectrum of the $R(80)$ line in the $B^3\Pi_u$-$X^1\Sigma_g^+$ (9,4) band of molecular iodine. The line marked with (*) is the t-hyperfine component, which was calibrated at 15 882.776 6 cm^{-1} [14]. The wavelength in the XUV domain, $\lambda = 104.936\,744 \pm 0.000\,004$ nm for this specific H$_2$ line, was deduced by fitting the spectral recordings and implementing scan linearization. Special care was taken to address possible sources of systematic
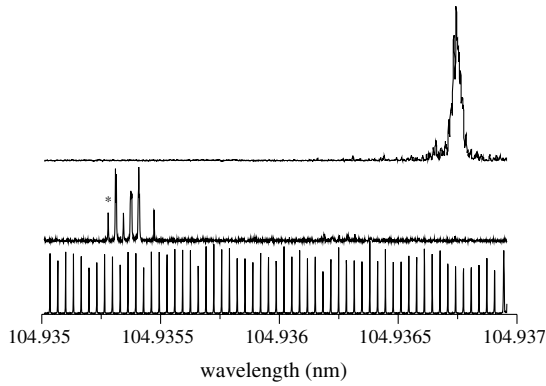
FIG. 1.   Recording of the $R(0)$ line in the B-X (4,0) Lyman band with the I_2 reference spectrum and etalon markers for the calibration. The line marked with * is the t-hyperfine component of the $R(80)$ line in the B-X (9,4) band of I_2.

uncertainties: residual Doppler shift, AC-Stark effects, and frequency chirp in the laser. The experimental setup and the analysis procedures are described in Eikema et al. [15]. The I_2 saturated absorption reference standard does unfortunately not extend far enough [14,16] to cover the range of the (0,0) and (1,0) Lyman bands. From an evaluation of the error budget, the uncertainty in the line positions is estimated at 0.000 005 nm. The values of the 40 accurately determined line positions with relevance for the present comparison with quasar data are listed in Table I; estimated uncertainties for individual lines are also included. Note that all values of wavelengths pertain to vacuum. From an analysis of combination differences between $R(J)$ and $P(J + 2)$ lines, it follows that the data are more consistent than estimated; hence, the stated uncertainty represents a conservative estimate. Further details on the experimental methods and a critical evaluation of the uncertainties are given by Philip et al. [17].

The present data set can be used to calculate wavelengths for additional nonmeasured lines via the known ground state rotational splittings [18]: $\Delta[(J = 2) - (J = 0)] = 354.3734(2)$ cm^{-1}, $\Delta[(3) - (1)] = 587.0325(2)$ cm^{-1}, and $\Delta[(4) - (2)] = 814.4246(3)$ cm^{-1}. This procedure provides wavelengths for ten lines, also listed in Table I, at an accuracy improved by an order of magnitude compared to the Meudon data set. For the remainder of the lines, not accessed in the present experimental study, we rely on the most accurate published values. We note that higher accuracy transition wavelengths can be derived from the excited state energies from the Meudon study [19], which are an average over many emission lines; these level energies can be converted into transition frequencies by adding highly accurate ground state rotational level energies of Jennings et al. [18].

Each spectral line in the B-X Lyman bands and the C-X Werner bands depends in a different, but specific way on the mass ratio μ. This can be quantified by a sensitivity coefficient K_i, and values for K_i were calculated by

TABLE I.   Measured wavelengths of the relevant Lyman (L) and Werner (W) transitions, and their standard uncertainties, used for the analysis of the quasar data as specified. The second set of lines are calculated from combination differences.

Line	λ_0 (nm)	$\Delta\lambda$	PKS	Q0347	Q1232
L 2-0 $R(0)$	107.713 874	0.000 004	*		
L 2-0 $R(1)$	107.769 894	0.000 004	*	*	
L 2-0 $R(2)$	107.922 491	0.000 004	*		
L 2-0 $P(1)$	107.892 547	0.000 004	*		
L 3-0 $R(1)$	106.346 014	0.000 004		*	
L 3-0 $R(2)$	106.499 481	0.000 004	*	*	
L 3-0 $R(3)$	106.747 855	0.000 004	*		*
L 3-0 $P(1)$	106.460 539	0.000 004	*	*	
L 3-0 $P(2)$	106.690 068	0.000 004	*		*
L 4-0 $R(0)$	104.936 744	0.000 004	*		
L 4-0 $R(1)$	104.995 976	0.000 004	*		
L 4-0 $R(2)$	105.149 858	0.000 004	*	*	
L 4-0 $R(3)$	105.397 610	0.000 004	*	*	
L 4-0 $P(1)$	105.103 253	0.000 004	*		
L 5-0 $R(2)$	103.869 027	0.000 004		*	
L 7-0 $R(1)$	101.343 701	0.000 005	*	*	
L 7-0 $R(2)$	101.497 685	0.000 005	*		
L 7-0 $P(1)$	101.432 724	0.000 005	*		
W 0-0 $R(2)$	100.902 492	0.000 006	*	*	
W 0-0 $Q(1)$	100.977 088	0.000 005	*		
W 0-0 $Q(2)$	101.093 844	0.000 006	*	*	
W 0-0 $P(3)$	101.450 423	0.000 006	*		
L 9-0 $R(0)$	99.137 891	0.000 005	*		
L 9-0 $R(1)$	99.201 637	0.000 005	*	*	
L 9-0 $R(2)$	99.355 062	0.000 009	*		
L 9-0 $P(1)$	99.280 968	0.000 005	*		
W 1-0 $R(2)$	98.624 406	0.000 006	*		
W 1-0 $R(3)$	98.744 863	0.000 009	*		
W 1-0 $Q(2)$	98.797 445	0.000 006		*	
L 10-0 $P(1)$	98.283 533	0.000 005		*	
L 10-0 $P(2)$	98.486 398	0.000 005	*		
L 11-0 $R(2)$	97.415 791	0.000 005	*		
L 11-0 $P(3)$	97.821 804	0.000 006	*		
L 12-0 $R(3)$	96.767 695	0.000 006	*	*	
W 2-0 $R(2)$	96.579 549	0.000 006	*		
W 2-0 $R(3)$	96.678 035	0.000 006	*		
W 2-0 $Q(1)$	96.609 608	0.000 005		*	
W 2-0 $Q(2)$	96.728 110	0.000 005		*	
L 13-0 $R(2)$	95.657 993	0.000 005	*		
W 3-0 $Q(1)$	94.742 188	0.000 005		*	
L 2-0 $P(2)$	108.126 603	0.000 004	*		
L 2-0 $P(3)$	108.456 034	0.000 005			*
L 3-0 $P(3)$	107.014 088	0.000 005			*
L 3-0 $P(4)$	107.431 296	0.000 005			*
L 4-0 $P(2)$	105.328 426	0.000 004	*		*
L 4-0 $P(4)$	106.058 104	0.000 005			*
L 5-0 $P(3)$	104.350 319	0.000 004	*		
L 6-0 $P(3)$	103.119 260	0.000 009	*		
L13-0 $P(3)$	96.045 063	0.000 005	*		
L15-0 $P(3)$	94.433 027	0.000 011	*		

Varshalovich and co-workers [5,6,8]. The concept underlying the analysis of quasar data for a possible change in μ is that all lines undergo a certain redshift z for an

absorbing cloud near a specific quasar, but slight variations between apparent values for z_i (redshift for a certain spectral line) may be caused by their differing sensitivity for μ via the K_i factors. It can be shown that a hypothetical change in μ [5,6,8] relates the values of z_i to the sensitivity coefficients via

$$z_i = \bar{z} + (1 + \bar{z})\frac{\Delta\mu}{\mu}K_i, \qquad (1)$$

with $z_i = \lambda_i/\lambda_i^0 - 1$. The values for λ_i and λ_i^0 represent the astronomically observed wavelength and the rest-frame laboratory wavelength for the ith spectral line.

The new data are implemented in the models for testing a possible variability of μ by comparing with available data on H_2 spectra from PKS 0528-250 [9], Q0347-382 [8,10], and Q1232+082 [8]. The quasistellar object PKS 0528-250 at redshift $z = 2.811$ has been investigated several times in the past for hydrogen features. From a spectrum by Morton $et\ al.$ [20], Levshakov and Varshalovich [21] identified molecular hydrogen absorption lines. Foltz $et\ al.$ [22] observed PKS 0528-250 and these data were further analyzed by Varshalovich and Levshakov [5] and Varshalovich and Potekhin [6] obtaining $|\Delta\mu/\mu| < 0.002$ at the 2σ level. Later Cowie and Songaila [7] obtained a spectrum with the Keck telescope and compared the 19 observed H_2 spectral lines with laboratory data of Dabrowski [23] yielding $-5.5 \times 10^{-4} < \Delta\mu/\mu < 7 \times 10^{-4}$ (2σ). Finally, Potekhin $et\ al.$ [9] obtained the most extensive spectrum of PKS 0528-250, recorded with the 4 m CTIO telescope, containing 50 lines in the Lyman and Werner bands, from which $-7.5 \times 10^{-5} < \Delta\mu/\mu < 2 \times 10^{-4}$ (2σ) was derived. Ivanchik $et\ al.$ [8] have investigated H_2 spectral lines and the possible variation of μ in the quasars Q 1232+082 at $z = 2.3377$ and in Q 0347-382 at $z = 3.0249$, using the VLT/UVES telescope. Combined analysis of these sets of 12 and 18 lines yielded a constraint of $\Delta\mu/\mu = 5.7 \pm 7.6 \times 10^{-5}$, in comparison with the data of Abgrall $et\ al.$ [11,12] and $\Delta\mu/\mu = 12.5 \pm 9.0 \times 10^{-5}$, when compared with the data of Morton and Dinerstein [13] (all at 2σ). In 2002, Levshakov $et\ al.$ [10] have obtained an alternative data set for Q 0347-382 again via the VLT/UVES, from which a 2σ constraint of $-3.0 \times 10^{-5} < \Delta\mu/\mu < 11.4 \times 10^{-5}$ was derived.

Of the set of 50 lines in PKS 0528-250, now 33 have been measured at high accuracy, and six additional ones were calculated (all listed in Table I). For Q 1232+082, seven were newly determined, while of the 18 lines analyzed in Q 0347-382, 17 were remeasured at high accuracy. The newly determined lines relevant for the three quasars are indicated in Table I. In weighted least-squares fitting procedures, where the weights were derived from accuracies of astronomically observed wavelengths, values for $\bar{z}$ and $\Delta\mu/\mu$ and their uncertainties at the 2σ level were derived.

In a first round of fits, it was found that the resulting χ^2 is dominated by only a few data points. Subsequently, a second round of fitting was performed after adaption of some of the error margins. For PKS 0528-250, 50% of χ^2 comes from three spectral lines, L2-0 $P(1)$, L10-0 $P(2)$, and L13-0 $P(2)$; for these lines and three additional ones the error margins were multiplied by a factor of 2.5. The same holds for Q1232+082, where 65% of χ^2 originates from L3-0 $R(3)$ and $P(3)$. For Q0347-382, we used a combination of data from Refs. [8,10]; the larger part of χ^2 stems from W1-0 $R(1)$, which is in fact blended with W1-0 $R(0)$ in the astrophysical observations; this line was rejected. For the PKS 0528-250 data, a negative value is found for $\Delta\mu/\mu$, for Q1232+082 a positive value, and for Q0347-382 an essentially zero value ($\ll 1\sigma$); however, within a 2σ error margin for all three data sets a null result is found. These fits also result in accurate values for the redshifts of the quasars under consideration: $\bar{z}_{PKS} = 2.810\,801(10)$, $\bar{z}_{Q0347} = 3.024\,890(3)$, and $\bar{z}_{Q1232} = 2.337\,708(3)$.

As was discussed by Ivanchik $et\ al.$ [8], spectral data of several quasars at varying redshifts can be included in a combined analysis by defining a reduced line redshift ζ_i:

$$\zeta_i = \frac{z_i - \bar{z}_q}{1 + \bar{z}_q} = \frac{\Delta\mu}{\mu}K_i, \qquad (2)$$

where $\bar{z}_q$ are the averaged redshifts, obtained via the analysis described above. The entire data set of 81 lines pertaining to the three quasars is plotted in Fig. 2 and a least-squares combined fit yields:

$$\Delta\mu/\mu = -0.5 \pm 3.6 \times 10^{-5} \qquad (3)$$

at the 2σ level. This zero constraint is more tight, by a factor of 3, than the best hitherto obtained value [10]. We have opted for using all available data in literature, although the accuracy in the PKS 0528 data is lower and the scatter higher (see also Fig. 2); if only the 31 data points of Q1232+082 and Q0347-382 are included in
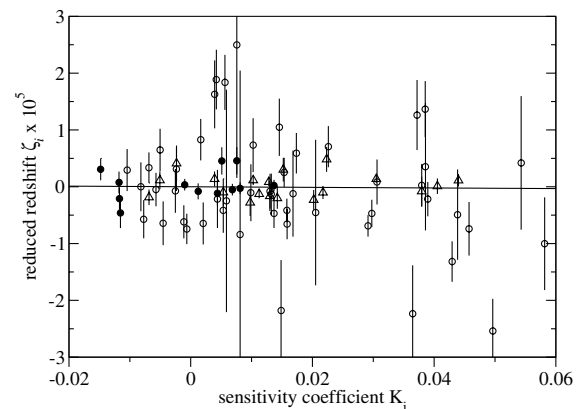

FIG. 2. Data of all three quasars compared with laboratory data in combined fit. Data with $\bigcirc$ for PKS 0528-250, $\bullet$ for Q1232, and $\triangle$ for Q0347. The line represents the result of the fit, with the slope equalling $\Delta\mu/\mu$.

the analysis one finds $\Delta\mu/\mu = 1.9 \pm 2.9 \times 10^{-5}$ (2σ), an even tighter constraint on the mass-ratio evolution.

The above values yielding a fractional constraint on the mass ratio μ can be used to derive an upper bound on the variability of the mass ratio itself. In a linear cosmological expansion model time evolution relates to the redshift parameter as $t = t_0[1 - (1 + z)^{-3/2}]$, where t_0 is the age of the universe, which may be taken as $\approx 14 \times 10^9$yr, and t is the backward time. Then $z = 2.75$, the average redshift for the data set used, corresponds to an epoch of 12 Gyr ago. The age difference between quasars, 600 Myr for the lowest and highest value of z, is neglected here. This yields a constraint on a temporal variation of the mass ratio of

$$\frac{d}{dt}\frac{\Delta\mu}{\mu} = -0.4 \pm 3.0 \times 10^{-15} \text{ yr}^{-1} \tag{4}$$

again at the 2σ level.

In the present laboratory study, transition wavelengths of Lyman and Werner band spectral lines have been determined at an accuracy over an order of magnitude better than in previous experiments; this accurateness of data allows for a comparison with the same spectral lines observed at high redshift in three quasars, which had produced these lines 12 billion years ago. Whereas in the previous analyses [5–10,20–22] the outcome was determined by both the uncertainties in the spectral observations on quasars and in laboratory data, in the present analysis the laboratory data can be considered exact, at least for the 50 presently determined wavelengths. The main result is a bound on a possible variability of the proton-to-electron mass ratio, which is lower than previously estimated. At the level of a few times 10^{-15} per year M_p/m_e does not vary. Calmet and Fritsch have argued that Grand Unification predicts, that a cosmological variation of nucleon masses should be 40 times larger than a variation of the fine structure constant α [24]; if so, the present result can be interpreted as a bound on $\Delta\alpha/\alpha$ on the scale of 10^{-16} per year, which is in contradiction with the findings of Ref. [1]. The new and accurate set of laboratory wavelengths may stimulate further astronomical observations of H_2 lines in quasistellar objects.

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