

# Precision Spectroscopy of H<sub>2</sub> and a Possible Variation of $m_p/m_e$ over Cosmological Time

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**Abstract.** A novel and highly accurate (at the level of  $\Delta\lambda/\lambda = 5 \times 10^{-8}$ ) database of 233 spectral lines in the Lyman and Werner bands of H<sub>2</sub> is determined via laboratory spectroscopy employing a narrowband and tunable extreme ultraviolet laser system. Furthermore an updated set of so-called  $K_i$  coefficients, representing the sensitivity of each spectral line on a variation of the proton-to-electron mass ratio  $\mu = m_p/m_e$  are derived for all lines in the H<sub>2</sub> spectrum. The laboratory wavelengths and  $K_i$ 's are used in a comparison with a recent set of highly accurate H<sub>2</sub> spectral lines observed in the Q 0347-383 and Q 0405-443 quasars yielding a fractional change in the mass-ratio of  $\Delta\mu/\mu = 2.4 \pm 0.6 \times 10^{-5}$  ( $1\sigma$ ). This result indicates, at a  $4\sigma$  confidence level, that  $\mu$  may have decreased in the past 12 billion years.

**Keywords:** Molecular Hydrogen, Precision spectroscopy, Quasar absorption, Fundamental constants

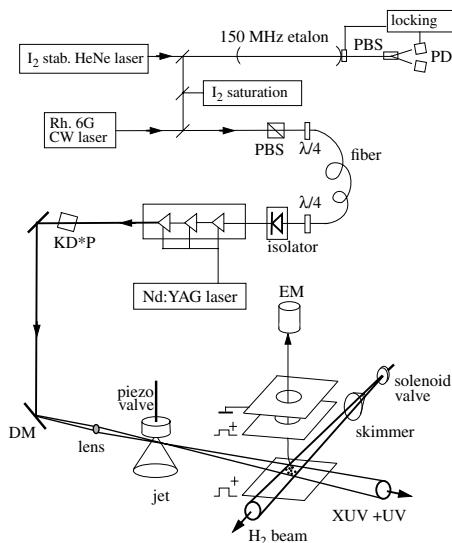
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## INTRODUCTION

The possibility of a possible variation of fundamental constants has re-emerged at the forefront of modern physics with the recent findings of the Sydney group of a variation of the fine structure constant  $\alpha$  [1], in the most recent reports even at the  $5\sigma$  level of statistical significance. The result is based on a comparison of wavelengths of spectral absorption lines of atomic and ionic species in cold interstellar clouds at high redshift in the line-of-sight of quasar light sources and the same lines observed in the laboratory at zero redshift in the modern epoch. Important for the comparison are the so-called sensitivity factors  $q$ , which are different for each spectral line and that can be derived from *ab initio* relativistic calculations of atomic structure [2]. As was first noted by Thompson [3] similar comparisons between spectra of molecules can be employed to detect possible variations of the proton-electron mass ratio  $\mu$  over cosmological time. From a theoretical perspective an important connection can be made between a possible variation of  $\alpha$  and of  $\mu$ . Within the framework of Grand Unification theories, in which the coupling constants  $g_i$  for the strong, electromagnetic and weak fundamental forces converge at a certain energy scale, the rates of change of the couplings must vary alongside, thus yielding a relationship (see e.g. Ref. [4] and various other papers):

$$\frac{\Delta\mu}{\mu} = R \frac{\Delta\alpha}{\alpha} \quad (1)$$

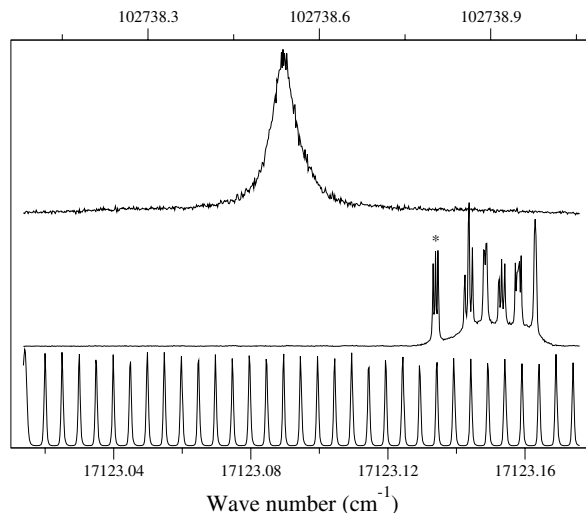
The values of  $R$  derived are usually in the range 20 – 40, implying that  $\mu$  is a more sensitive test ground to detect a variation of a constant *per se*. Since the value for the



**FIGURE 1.** Schematic drawing of the experimental setup. In the top part a continuous wave ring-dye laser produces a stable carrier frequency of 1 MHz bandwidth; its absolute frequency is calibrated against an  $I_2$  reference standard, while relative frequency spacings are measured with respect to an etalon, that is stabilized to an HeNe laser. The CW output is pulse-amplified in a three-stage pulsed dye amplifier (PDA), pumped by a pulsed Nd:YAG laser, and the generated pulses of 5 ns duration are subsequently frequency doubled in a KD\*P crystal, and thereupon frequency tripled in a gas jet (xenon, krypton or nitrogen). The generated XUV beam, of 250 MHz bandwidth, is perpendicularly crossed with a pulsed and skimmed  $H_2$  molecular beam to suppress Doppler broadening. On resonance  $H_2^+$  ions are produced via 1 XUV + 1 UV photo-ionization, which are then accelerated by a pulsed electric field (switched on after the laser pulse) into a time-of-flight drift tube for mass separation. Signal is registered via detection of the ions on an electron multiplier (EM). This is the generic setup used for the majority of the measurements [9]. Variations on this scheme are used to perform measurements in specific wavelength regions, involving mixing with the 532 nm output of a Nd:YAG laser, addition of a short wavelength ionization laser [10], and using a sum-frequency mixing scheme for the production of XUV light.

proportionality factor  $R$  depends on the exact details of a specific GU-theory, Eq. (1) opens a future perspective for atomic and molecular physics: discovery of details of GUT from accurate determinations of variations of  $\alpha$  and  $\mu$ .

Highly redshifted absorption spectra of  $H_2$  molecules have been observed in the light of sight of some 10 quasar systems. Based on spectral data from three systems (Q 0528-250, Q 1232+082 and Q 0347-382) we previously reported a constraint on a possible proton-electron mass ratio variation of  $\Delta\mu/\mu = -0.5 \pm 1.8 \times 10^{-5}$  ( $1\sigma$ ) [5]. Recently a novel set of more accurately calibrated spectral lines (at  $\Delta\lambda/\lambda = 0.2 - 1.0 \times 10^{-6}$ ) in two quasar systems (Q 0405-443 and Q 0347-383) was obtained [6]. A comparison was made between this set of lines and the novel database of  $H_2$  laboratory lines, also based on a recalculated set of sensitivity coefficients  $K_i$ , resulting in an indication that the proton-electron mass ratio has decreased by 0.002% over the past 12 billions years at a significance of  $4\sigma$  [7]. The fit procedures as well as the assessment of possible systematic effects that might mimic the result of a  $\mu$  variation are analyzed.



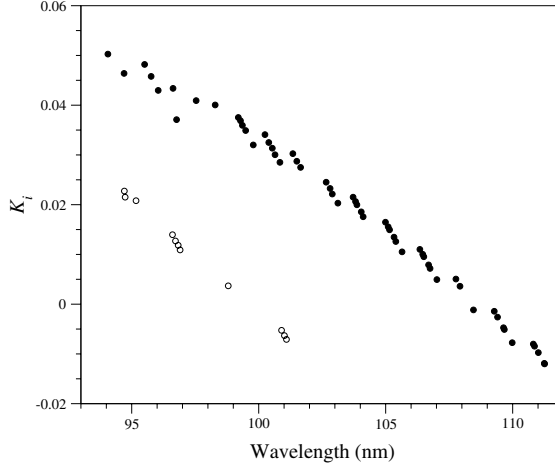
**FIGURE 2.** Recording of the P(1) line in the B  $^1\Sigma_u^+$ -X  $^1\Sigma_g^+$  (11,0) Lyman band (upper spectrum) with the I<sub>2</sub> reference spectrum (middle spectrum) and étalon markers (bottom spectrum) for the calibration. The line marked with an asterisk is the *t*-hyperfine component of P(83) line in the B-X (15,1) band of I<sub>2</sub> at 17123.13404 cm<sup>-1</sup> [8]. The scale of XUV frequencies (top) is exactly a factor 6 times that of the fundamental (bottom); the center of the H<sub>2</sub> resonance is determined by fitting a Voigt profile. The frequency scale of the fundamental is linearized by fitting a spline through the étalon fringe positions and turned into an absolute scale by taking the I<sub>2</sub> *t*-component as a reference and by the measured Free-Spectral-Range of 148.96 MHz.

## XUV-LABORATORY SPECTROSCOPY ON H<sub>2</sub>

During the course of the past years the absorption spectrum of the H<sub>2</sub> molecule has been re-investigated by means of XUV-laser spectroscopy. The setup of the coherent radiation source with tunability in the range 90-110 nm is displayed in Fig. 1 and relevant details of the measurements procedures are described in the caption. Accurate transition wavelengths of 162 spectral lines in the B  $^1\Sigma_u^+$ -X  $^1\Sigma_g^+$  Lyman and C  $^1\Pi_u$ -X  $^1\Sigma_g^+$  Werner band systems were determined at an accuracy of  $5 \times 10^{-8}$ . A typical spectrum, of the P(1) line in the B  $^1\Sigma_u^+$ -X  $^1\Sigma_g^+$  (15,0) Lyman band, is reproduced in Fig. 2; in the caption some details of the calibration procedures are described. The database was extended by using the accurately know combination differences between P(*J*+2) and R(*J*) transitions from far-infrared spectroscopy, therewith extending the H<sub>2</sub>-database to 233 lines. For more details on the measurement procedures we refer to Refs. [9, 10].

## SENSITIVITY COEFFICIENTS *K*

The transition wavelengths  $\lambda_i$  of spectra of cold hydrogen clouds at high redshift were obtained with the UVES-spectrometer at the Very Large Telescope of the European Southern Observatory in Paranal Chile [6]. After a first analysis 37 lines in Q 0347 and



**FIGURE 3.** Calculated sensitivity coefficients for those Lyman and Werner lines observed in Q 04050 and Q 0347 quasar systems as a function of wavelength. Black symbols: Lyman lines and Open symbols: Werner lines.

39 lines in Q 0405 were found to be unblended by the Lyman- $\alpha$  forest, and these can be compared to the same lines contained in the highly accurate database of laboratory wavelengths, via:

$$\frac{\lambda_i}{\lambda_i^0} = (1 + z_Q) \left( 1 + \frac{\Delta\mu}{\mu} K_i \right) \quad (2)$$

where  $\Delta\mu$  is defined as  $\Delta\mu = \mu_z - \mu_0$ . The first factor on the right-hand side of Eq. (2) represents the usual redshift-dependence of a line, while the second factor represents a possible additional dependence on a variation of the mass ratio  $\mu$ . The  $K$ -coefficient for each line relates to the sensitivity for each line to such a mass variation:

$$K_i = \frac{d \ln \lambda_i}{d \ln \mu} = \frac{\mu}{\lambda_i} \frac{d \lambda_i}{d \mu} \quad (3)$$

The  $K$ -sensitivity coefficients are derived in a stepwise procedure. First a Dunham representation is fitted to the level energies, obtained from the new database combined with the accurately known  $X^1\Sigma_g^+$  ground state levels [11]. Then Dunham representations of the form:

$$E(v, J) = \sum_{k,l} Y_{kl} \left( v + \frac{1}{2} \right)^k [J(J+1) - \Lambda^2]^l \quad (4)$$

were fitted to the level energies resulting in sets of  $Y_{kl}$  coefficients for all four involved electronic states (both  $C^1\Pi_u^+$  and  $C^1\Pi_u^-$  parity states were treated separately). The mass-dependence of the Dunham expansion coefficients is known to yield (in first but sufficient approximation):

$$\frac{dY_{kl}}{d\mu} \approx -\frac{Y_{kl}}{\mu} \left( l + \frac{k}{2} \right) \quad (5)$$

Through Eqs. (3) to (5) the  $K$ -coefficients were calculated. The resulting values range from  $-0.01$  to  $0.05$  for the set of 76 lines of relevance for the present comparison. The values are much smaller than unity, because the electronic part of the energy in the transition is independent of the mass ratio (at least in the Born-Oppenheimer approximation). Thereafter the values for  $K$  were corrected for some non-Born-Oppenheimer effects. Firstly the effect of the adiabatic correction on the values of  $K_i$  was calculated, and then the mixing, as a result of electronic-rotational angular momentum coupling in the B  $^1\Sigma_u^+$  and C  $^1\Pi_u$  states, was addressed. That resulted in changes of five  $K_i$  values of about 10%. While the resulting values of this semi-empirical analysis are estimated to be accurate with  $1 - 2\%$  they also are found to be in agreement with a calculation based on mass-dependent *ab initio* calculations of H<sub>2</sub> within 1% [12].

The final  $K$ -values are plotted in Fig. 3 as a function of wavelength. Note that the values for each band system vary rather smoothly with wavelength (or photon energy), but that the values pertaining to the Lyman and Werner lines, even in the same wavelength interval differ strongly.

## INDICATION FOR A $\mu$ VARIATION

In order to include the full set of 76 data pertaining to two quasar systems at differing redshifts, Eq. (2) is transformed into:

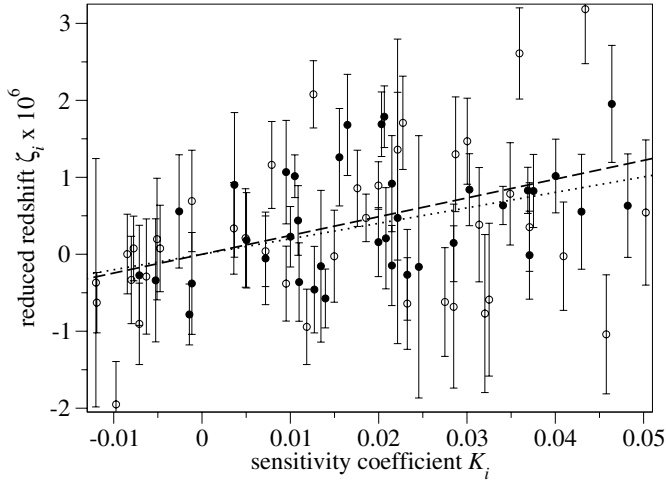
$$\zeta_i = \frac{z_i - z_Q}{1 + z_Q} = \frac{\Delta\mu}{\mu} K_i \quad (6)$$

where  $\zeta_i$  represent the reduced redshifts. When all data are plotted in a comparative study following Eq. (6), such as in Fig. 4, then the slope of the fitted line directly gives  $\Delta\mu/\mu$ . In the Figure the result of two least squares fits are drawn: one for a weighted fit yielding  $\Delta\mu/\mu = (2.44 \pm 0.59) \times 10^{-5}$  and one for an unweighted fit yielding  $\Delta\mu/\mu = (1.98 \pm 0.58) \times 10^{-5}$ . These results are significant at the  $4\sigma$  and the  $3.5\sigma$  level of significance.

Analysis on the sets of data for each quasar system separately yields  $\Delta\mu/\mu = (2.06 \pm 0.70) \times 10^{-5}$  for Q 0347-383 and  $\Delta\mu/\mu = (2.78 \pm 0.88) \times 10^{-5}$  for Q 0405-443. The robustness of the fitting procedure was tested by replacing the usual least-squares criterion by a least-absolute value criterion yielding  $\Delta\mu/\mu = 2.15 \times 10^{-5}$  for the combined data set. This criterion makes the analysis less dependent on outliers. Similarly outliers producing large  $\chi^2$  contributions were left out of the fit in a stepwise fashion for the 9 most severe cases; the fit converges to  $\Delta\mu/\mu = (1.98 \pm 0.43) \times 10^{-5}$ . All these tests on the statistics lead to similar results.

As for the systematics it is imperative to search for mechanisms that can mimic a variation of  $\mu$ . One such possibility is the division of the absorbing H<sub>2</sub> clouds in warmer and colder parts, that might be located at differing locations, and hence  $z$ -values. Separate fits to subclasses in the data set for rotational states  $J = 0 - 1$  (colder part of the clouds) and for states  $J = 2 - 4$  (warmer parts. Such a procedure yields  $\Delta\mu/\mu = (2.94 \pm 1.12) \times 10^{-5}$  for the cold clouds and  $\Delta\mu/\mu = (1.96 \pm 0.78) \times 10^{-5}$ .

An underlying energy dependence of the reduced redshift data could mimic a  $\mu$  variation. This is possible due to the rather strong energy or wavelength dependence



**FIGURE 4.** Linear fit to reduced redshift of quasar absorption lines  $\zeta$  as defined by Eq. (6). Filled circles: Q 0347-383,  $z = 3.0248970$  (5); open circles: Q 0405-443,  $z = 2.5947325$  (4). The error-weighted linear fit is shown by a dashed line, the unweighted fit by a dotted line.

of the  $K$ -coefficients (*cf.* Fig. 3). Indeed a fit of the  $\zeta_i$  values with respect to energy  $\nu_i$  of the form:

$$\zeta_i = z_Q + A\nu_i \quad (7)$$

yields a proportionality factor  $A = (0.14 \pm 0.09) \times 10^{-9}$  per  $\text{cm}^{-1}$ . This signifies a correlation of  $1.6 \sigma$  between  $\zeta_i$  and  $\nu_i$ . This correlation, which is markedly smaller than the  $3.5 \sigma$  correlation between  $\zeta_i$  and  $K_i$  indicating the  $\mu$ -variation effect, is a consequence from the strong correlation between the  $K_i$  values with photon energy  $\nu_i$  as shown in Fig. 3 and is expected. The enhanced correlation on the true  $\mu$  variation effect is in fact produced by the fact that both Lyman end Werner lines are included in the data set. This analysis demonstrates that inclusion of Werner lines ( $\text{C}^1\Pi_u\text{-X}^1\Sigma_g^+$  band system) is a necessary requisite for making a distinction between an energy-dependent artifact and a  $\zeta_i - K_i$  representing the mass variation effect.

## CONCLUSION

Based on a comparison of 76  $\text{H}_2$  spectral absorption lines observed at high redshift in the line-of-sight of the quasar systems Q 0347 and Q 0405 with the same highly accurate lines observed in the laboratory at zero redshift an *indication* for a variation of the proton-electron mass ratio is found. This ratio  $\mu$  may have decreased by 0.002% in the past 12 billion years at a  $3.5 \sigma$  confidence level. Future observations of  $\text{H}_2$  spectral lines in additional cold interstellar clouds at high redshift may provide more definite evidence on the presently found indication. Assuming a linear rate of change over time, and within a cosmological model of linear expansion the present variation can be converted into a rate of change of  $2 \times 10^{-15}$  per year.

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