

## ON A VARIATION OF THE PROTON-ELECTRON MASS RATIO

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Recently indication for a possible variation of the proton-to-electron mass ratio  $\mu=m_p/m_e$  was found from a comparison between laboratory  $H_2$  spectroscopic data and the same lines in quasar spectra. This result will be put in perspective of other spectroscopic activities aiming at detection of variation of fundamental constants, on a cosmological as well as on a laboratory time scale. Furthermore the opportunities for obtaining improved laboratory wavelength positions of the relevant  $H_2$  absorption lines, as well as the prospects for obtaining a larger data set of  $H_2$  absorptions at high redshift will be presented. Also an experiment to detect  $\Delta\mu$  on a laboratory time scale will be discussed.

### 1. Introduction

Recently the finding of an *indication* for a decrease of the proton-to-electron mass ratio  $\mu=m_p/m_e$  by 0.002% in the past 12 billion years was reported [1]. Laser spectroscopy on molecular hydrogen, using a narrow-band and tunable extreme ultraviolet laser system resulted in transition wavelengths of spectral lines in the B-X Lyman and C-X Werner band systems at an accuracy of  $5 \times 10^{-8}$  for the best lines. This corresponds to an absolute accuracy of 0.000005 nm. A database of 233 accurately calibrated  $H_2$  lines is produced for future reference and comparison with astronomical observations. Recent observations of the same spectroscopic features in cold hydrogen clouds at redshifts  $z=2.5947325$  and  $z=3.0248970$  in the line of sight of two quasar light sources (Q 0405-443 and Q 0347-383) resulted in 76 reliably determined transition wavelengths of  $H_2$  lines at accuracies in the range  $2 \times 10^{-7}$  to  $10^{-6}$ . Those observations were performed with the Ultraviolet and Visible Echelle Spectrograph at the Very Large Telescope of the European Southern Observatory at Paranal, Chile [2]. A third ingredient in the analysis is the calculation of an improved set of sensitivity coefficients  $K_i$ , a parameter

associated with each spectral line, representing the dependence of the transition wavelength on a possible variation of the proton-to-electron mass ratio. Details of the methods are reported in Ref. [3].

A statistical analysis of the data yields an indication for a variation of the proton-to-electron mass ratio of  $\Delta\mu/\mu = (2.45 \pm 0.59) \times 10^{-5}$  for a weighted fit and  $\Delta\mu/\mu = (1.98 \pm 0.58) \times 10^{-5}$  for an unweighted fit. This result has a statistical significance of  $3.5\sigma$ . The redshifts of the hydrogen absorbing clouds can be converted into look-back times of 11.7 and 12 billion years with the age of the universe set to 13.7 billion years. Mass-variations as discussed relate to inertial or kinematic masses, rather than gravitational masses. The observed decrease in  $\mu$  corresponds to a rate of change of  $d\ln\mu/dt = -2 \times 10^{-15}$  per year, if a linear variation with time is assumed. This remarkable result should be considered as no more than an *indication* for a possible variation of  $\mu$ . Only a very limited data set is available: two quasar systems with a total of 76 spectral lines.

In the following we put these results in perspective of other spectroscopic activities concerning variation of fundamental constants, and present possibilities to obtain confirmation of the findings in the near future by producing improved laboratory data for  $H_2$  and extend the data set of  $H_2$  astronomical observations.

## 2. Variation of dimensionless fundamental constants: $\alpha$ and $\mu$

Renewed interest in the possibility of temporal variation of fundamental constants arose through the findings of Webb et al. [4]. Based on highly accurate spectroscopic observations of atomic and ionic resonance lines at high redshift (from the HIRES-Keck telescope at Hawaii) a variation of the fine structure constant  $\alpha$  was deduced. This breakthrough could be made through implementation of the so-called Many-Multiplet method, which allows for using many transition wavelengths in the analysis [5], rather than just separations between fine structure lines, as in the alkali-doublet method. By this means the sensitivity to detect  $\Delta\alpha$  is improved. These findings on a lower value of  $\alpha$  in the past were disputed by competing teams who found essentially a null result on  $\Delta\alpha$  from data obtained with the UVES-VLT on the southern hemisphere [6,7]. Meanwhile the Webb-Murphy-Flambaum team extended their data set to some 150 quasar systems, obtaining a more than  $5\sigma$  effect with  $\Delta\alpha/\alpha = (-0.574 \pm 0.102) \times 10^{-5}$  [8]. The discrepancy in the findings by different teams on  $\Delta\alpha/\alpha$  were resolved by the recent reanalysis of the UVES-VLT data set by Murphy et al. [9]; flaws in the fitting procedures were uncovered and a reanalysis yields a revised value of  $\Delta\alpha/\alpha = (-0.44 \pm 0.16) \times 10^{-5}$ , in agreement with the values of [8].

The invention of frequency comb lasers has immensely increased the accuracies in atomic spectroscopy, to the extent that absolute precision at the level of  $10^{-15}$  can be obtained, in fact limited by the Cs time and frequency standard. From atomic precision experiments on various systems boundaries to the rate of change in the fine structure constant  $d\ln \alpha/dt$  were set to the level of  $2 \times 10^{-15}$  per year by performing laboratory laser spectroscopic studies with time intervals of one or a few years. The NIST-Boulder group set a limit of  $1.2 \times 10^{-15}$  per year from measurements on a singly trapped  $^{199}\text{Hg}^+$  ion [10]. The Munich group deduced a similarly small rate from calibrating the H-atom (1s-2s) transition against the Paris portable Cs fountain clock [11], as did the PTB-Braunschweig team from  $^{171}\text{Yb}^+$  ions [12]. Very recently the NIST-Boulder group pushed the boundary on  $d\ln \mu/dt$  to  $1.3 \times 10^{-16}$  per year by comparing  $\text{Hg}^+$  against Cs [13]; at the ICOLS 07 even a tighter limit was presented at the  $2 \times 10^{-17}$  level from a comparison of  $\text{Hg}^+$  and  $\text{Al}^+$  clock transitions (see this book).

It has been hypothesized that the changes in the proton-electron mass ratio  $\mu$  and the fine structure constant  $\alpha$  are linked and that  $\mu$  would change faster by an order of magnitude or more; this hypothesis [14] is based on Grand Unification Theories. From a recent analysis of microwave spectra from the astrophysical object B0218+357 at redshift  $z = 0.68$  Flambaum and Kozlov [15] put a limit to the variation of the mass-ratio at  $\Delta\mu/\mu = (0.6 \pm 1.9) \times 10^{-6}$ . Data on the inversion motion of ammonia (23 GHz) were compared to microwave transitions in other molecules.

Hence there is evidence for a variation of  $\alpha$ , and some indication for a variation of  $\mu$  at high redshifts ( $z > 1$ ), while the laboratory studies seem to put strict boundaries on  $\Delta\alpha$ . At the same time the recent findings of high redshift ammonia put a strict boundary to  $\Delta\mu$  at a redshift of  $z = 0.68$ . In this context the hypothesis of a phase transition occurring in the history of the universe, going from a matter-dominated (dust era) to a dark energy dominated (curvature era) universe may play a role [16]. Barrow hypothesized that only before this transition, which may have occurred near  $z=0.5$ , the fundamental constants may have changed.

### 3. Extension of the database of molecular hydrogen at high redshift

There exists only a limited data set of  $\text{H}_2$  absorptions at high redshift. Of the tens of thousands identified quasar sources some 600 are known to be associated with a damped Lyman- $\alpha$  system; such systems are characterized by a fully saturated and broad L- $\alpha$  absorption feature from a relatively dense cloud of atomic hydrogen with a column density of  $N(\text{H}) > 2 \times 10^{20} \text{ cm}^{-2}$ . Such systems display metal absorptions and in some cases also  $\text{H}_2$

absorptions. For the investigations probing  $\Delta\alpha$  some 200 systems have been spectroscopically analyzed (from metal lines Mg, Si, Zn, etc), but in only 14 of them  $H_2$  has been detected [3].

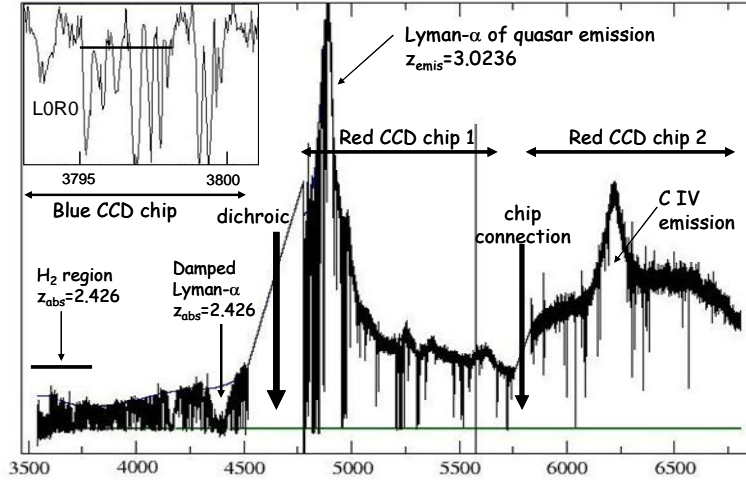


Fig. 1. Typical spectrum of a damped Lyman- $\alpha$  quasar system, in this case Q2348-011, as recorded by Noterdaeme et al. with the Ultraviolet-Visible Echelle Spectrometer at the Very Large Telescope at Paranal, Chile [18]. The large emission peaks can be assigned to H-L $\alpha$  and to C IV, and give the redshift of the quasar emission ( $z=3.0236$ ). Spectra are recorded with a certain setting of a dichroic, detecting blue light on a single CCD, and the red part on two distinct CCDs. A damped L $\alpha$  absorption line is found at  $z = 2.426$  and also  $H_2$  absorption is found at this redshift. Hence the Lyman and Werner absorption lines (in the laboratory at 90-112 nm) are shifted to below 380 nm. Part of the  $H_2$  window is enlarged in the left upper corner, displaying the complicated velocity structure of the  $H_2$  cloud: at least 7 velocity sub-components are visible for each absorption line (LORO is shown).

Obtained spectra in existing databases have been surveyed and besides the two systems used in our previous analysis (Q0347 and Q0405) three others have a potential to play a role in detecting  $\Delta\mu$  if spectra at sufficient SNR and resolution, with optimum wavelength calibrations were to be obtained. The system Q2348-011 at  $z = 2.426$  (an archived spectrum shown in Fig. 1) will be observed under such conditions at UVES-VLT in August 2007. Other appropriate systems would be Q0528-250 at  $z=2.81$  and Q1443+272 at  $z=4.22$ ; the latter is the system with  $H_2$  detected at the highest redshift [17]. Of course there should be many damped L- $\alpha$  systems with  $H_2$  at high redshift

in the universe that can be implemented in  $\Delta\mu$  analyses. They ‘just’ need to be found and subjected to high resolution observation; as a figure of merit, at current dish sizes of 8 m typical observation times in excess of 20 h on target (depending on brightness of the quasar background source) are needed to obtain spectra at resolutions of  $R = 60000$  and SNR of 50. In view of the importance of the subject the data set will be extended in coming years; currently a number of observation stations, HIRES-Keck (Hawaii), UVES-VLT (Chile), and HDS-Subaru (Japan), are suitable for the purpose. In 2009 the PEPSI-LBT system in Arizona, equipped with two 8 m dishes, will become available for detection of  $H_2$  at high redshift.

#### 4. Improving the laboratory accuracy of the Lyman and Werner lines

The prominent electronic absorption systems, also detected in quasars, are the  $B^1\Sigma_u^+ - X^1\Sigma_g^+$  or Lyman and the  $C^1\Pi_u - X^1\Sigma_g^+$  Werner band systems. At zero redshift these lie in the difficult to access wavelength range of 90-112 nm. With the use of a narrowband and tunable extreme ultraviolet (XUV) laser source the lines could be calibrated to an accuracy of  $5 \times 10^{-8}$  [19].

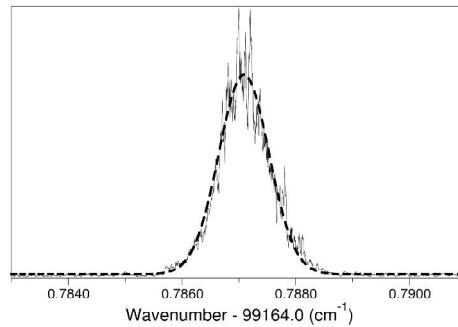


Fig. 2. Recording of the  $Q_0$  two-photon line in the EF-X (0,0) band of  $H_2$  at  $99164.78691(11) \text{ cm}^{-1}$ . Note that the resonance width is 36 MHz, determined by the linewidth of the laser at its 8<sup>th</sup> harmonic.

We have devised an alternative spectroscopic scheme to derive the wavelengths in the B-X and C-X systems via combination differences. This method is based on two independent spectroscopic measurements. First the lowest energy levels in the  $EF^1\Sigma_g^+$ ,  $v=0$  state are determined via a Doppler-free two-photon-excitation scheme in the deep-UV at  $\lambda=202 \text{ nm}$ , that was previously described [20]. Using various advanced techniques, such as calibration against a frequency comb laser, a Sagnac configuration to avoid Doppler shifts, and on-line frequency chirp evaluation for each of the laser

pulses an absolute accuracy of 3.5 MHz on the resonances (see Fig. 2 for a spectrum of the  $Q_0$  line) was obtained, which translates to a relative accuracy of  $\Delta\lambda/\lambda = 1 \times 10^{-9}$ .

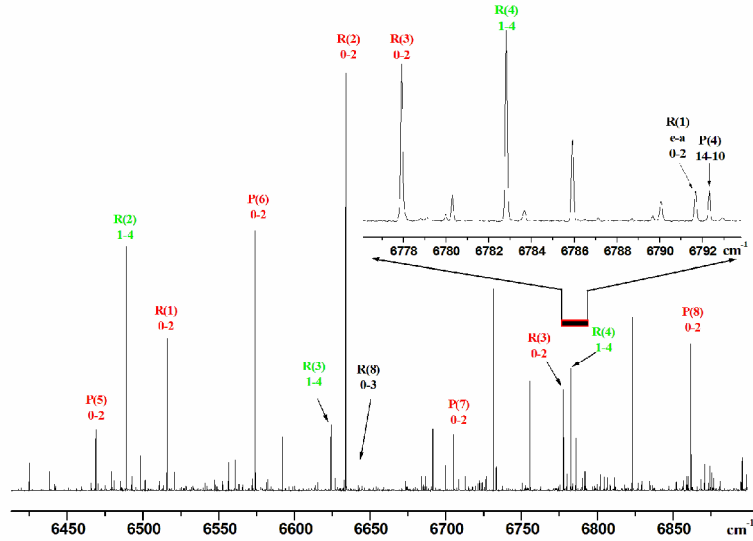


Fig. 3. A portion of the FT infrared emission spectrum of  $H_2$  in the range 6400-6900  $cm^{-1}$  displaying lines in the EF-B (0,2) and (1,4) bands as indicated.

A second experiment entails Fourier-Transform infrared and visible emission spectroscopy performed on a low pressure electrodeless discharge in  $H_2$ . In the near infrared domain ranging between 0.5-4  $\mu m$  many lines in the EF-B ( $v',v''$ ) are observed (a portion of the spectrum is shown in Fig. 3). Although the spectral lines are Doppler-broadened (0.02 - 0.2  $cm^{-1}$ ), the high SNR and the fact that each energy level is connected to 10 or more other quantum levels produces a consistent framework of energy levels at accuracies in the  $10^{-3} - 10^{-4} cm^{-1}$  range. Level energies in the C state are determined, somewhat less accurate, through transitions in the  $1^1\Pi_g-C$ ,  $J^1\Delta_g-C$ ,  $H^1\Sigma_g^+-C$  and  $GK^1\Sigma_g^+-C$  systems. Systematic effects are addressed by absolute wavelength calibration in a wide range using Ar and CO lines. This work in progress will yield relative level energies that can be combined with the level energies of the lowest EF,  $v=0$  levels; from the combined set transition wavelengths of most of the relevant Lyman and Werner lines can be calculated at accuracies in the range  $\Delta\lambda/\lambda = 1-5 \times 10^{-9}$ . Bearing in mind that the uncertainties in the current quasar absorption data are at the level of  $\Delta\lambda/\lambda = 2 \times 10^{-7}$ , these

studies provide a laboratory or zero-redshift data set for  $\text{H}_2$  which is exact for the purpose of comparison.

### 5. A molecular fountain for precision studies and detection of $\Delta\mu$

Variation of the proton-electron mass ratio may be detected from comparisons on a cosmological time scale, but also on a laboratory time scale. Since the intervals are reduced to years the required spectroscopic precision has to be much higher in the latter case. In view of the fact that quantum tunneling phenomena scale exponentially with mass, the inversion splitting in ammonia is extremely sensitive for  $\Delta\mu$ . This also underlies the tight constraint to  $\Delta\mu$  from ammonia spectra at  $z=0.68$  discussed above [15].

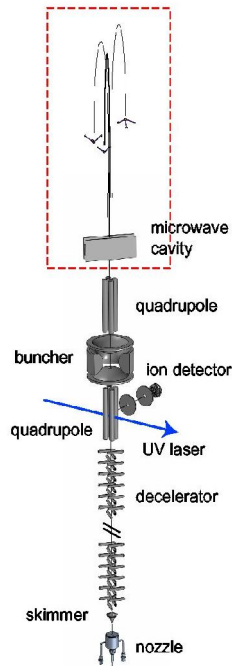


Fig. 4. Design of the molecular fountain under construction in Amsterdam.

In order to obtain long effective measurement times in a Ramsey-type scheme a molecular fountain is under construction, in which  $\text{NH}_3$  molecules will be launched, after deceleration by the Stark technique [21].

### Acknowledgement

We thank Prof. G. Meijer (Fritz Haber Institut, Berlin, Germany) for the collaboration on the molecular fountain project.

### References

1. E. Reinhold, R. Buning, U. Hollenstein, A. Ivanchik, P. Petitjean, W. Ubachs, *Phys. Rev. Lett.* **96**, 151101 (2006).
2. A. Ivanchik, P. Petitjean, D. Varshalovich, B. Aracil, R. Srianand, H. Chand, C. Ledoux, P. Boisseé, *Astron. Astroph.* **440**, 45 (2005).
3. W. Ubachs, R. Buning, K. S. E. Eikema, E. Reinhold, *J. Mol. Spectr.* **241**, 155 (2007).
4. J. K. Webb, V. V. Flambaum, C. W. Churchill, M. J. Drinkwater, J. D. Barrow, *Phys. Rev. Lett.* **82**, 884 (1999).
5. V. A. Dzuba, V. V. Flambaum, J. K. Webb, *Phys. Rev. Lett.* **82**, 888 (1999).
6. R. Srianand, H. Chand, P. Petitjean, B. Aracil, *Phys. Rev. Lett.* **92**, 121302 (2004).
7. R. Quast, D. Reimers, S. Levshakov, *Astron. Astroph.* **415**, L7 (2004).
8. M. T. Murphy, J. K. Webb, V. V. Flambaum, *Mon. Not. Roy. Astr. Soc.* **345**, 609 (2003).
9. M.T. Murphy, J. K. Webb, V. V. Flambaum, arXiv:astro-ph/0612407v1.
10. S. Bize, et al. *Phys. Rev. Lett.* **90**, 150802 (2003).
11. M. Fischer et al. *Phys. Rev. Lett.* **92**, 230802 (2004).
12. E. Peik, B. Lipphardt, H. Schnatz, T. Schneider, C. Tamm, S. G. Karshenboim, *Phys. Rev. Lett.* **93**, 170801 (2004).
13. T. M. Fortier et al. *Phys. Rev. Lett.* **98**, 070801 (2007).
14. X. Calmet, H. Fritsch, *Eur. J. Phys. C* **24**, 639 (2002).
15. V. V. Flambaum, M. G. Kozlov, *Phys. Rev. Lett.* **98**, 240801 (2007).
16. J. D. Barrow, H. B. Sandvik, J. Magueijo, *Phys. Rev. D.* **65**, 063504 (2002).
17. C. Ledoux, P. Petitjean, R. Srianand, *Astroph. J.* **640**, L25 (2006).
18. P. Noterdaeme, P. Petitjean, R. Srianand, C. Ledoux, F. Le Petit, *Astron. Astroph.* **469**, 425 (2007).
19. J. Philip, J. P. Sprengers, P. Cacciani, C. A. De Lange, W. Ubachs, E. Reinhold, *Can. J. Chem.* **82**, 713 (2004).
20. S. Hannemann, E.J. Salumbides, S. Witte, R. T. Zinkstok, E.-J. Van Duijn, K. S. E. Eikema, W. Ubachs, *Phys. Rev. A* **74**, 062514 (2006).
21. H.L. Bethlem, G. Berden, G. Meijer, *Phys. Rev. Lett.* **83**, (1999).