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HD as a Probe for Detecting Mass Variation on a Cosmological Time Scale

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The strong electronic absorption systems of the $B^1\Sigma_u^+ - X^1\Sigma_g^+$ Lyman and the $C^1\Pi_u - X^1\Sigma_g^+$ Werner bands can be used to probe possible mass-variation effects on a cosmological time scale from spectra observed at high redshift, not only in H_2 but also in the second most abundant hydrogen isotopomer HD. High resolution laboratory determination of the most prominent HD lines at extreme ultraviolet wavelengths is performed at an accuracy of $\Delta\lambda/\lambda \sim 5 \times 10^{-8}$, forming a database for comparison with astrophysical data. Sensitivity coefficients $K_i = d\ln\lambda_i/d\ln\mu$ are determined for HD from quantum *ab initio* calculations as a function of the proton-electron mass ratio μ . Strategies to deduce possible effects beyond first-order baryon/lepton mass ratio deviations are discussed.

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The observation of spectral features at high redshift ($z \sim 2-3$) provides an opportunity to probe minute variations of some fundamental constants over time intervals of 10^{10} years, corresponding to 80% of the lifetime of the Universe. For the fine structure constant α evidence has been reported for a temporal drift at 5σ significance [1]. Variation of another fundamental constant, the dimensionless proton-electron mass ratio $\mu = m_p/m_e$, may be probed through spectra of molecules. Recently, an indication for a possible decrease of μ was reported at $\Delta\mu/\mu = (2.45 \pm 0.59) \times 10^{-5}$ over a time interval of 12×10^9 years [2,3]. This result is derived from a set of 76 H_2 spectral lines in two absorption systems at $z = 2.59$ and $z = 3.02$ in the line of sight towards quasars Q0405 - 443 and Q0347 - 383 [4]. From observations of the NH_3 inversion splitting in the astrophysical object B0218 + 357 at $z = 0.68$, a tight constraint upon μ variation at $\Delta\mu/\mu = (0.6 \pm 1.9) \times 10^{-6}$ was deduced [5]. These seemingly contradictory results might be reconciled by invoking the concept of a phase transition having occurred at $z \sim 1$, transiting from a matter-dominated to a dark-energy-dominated Universe; variation of constants is hypothesized to occur only before the phase transition, hence for $z > 1$ [6].

The comparison of spectral lines over cosmological time scales depends on the availability of spectral transitions that can be observed at high accuracy and at high redshift. Molecular hydrogen is the most abundant molecule in the Universe by orders of magnitude. The abundance of the deuterated HD species competes with other abundant molecules such as CO and CH such that it is worthwhile to consider the opportunity of using HD absorption for probing mass-variation effects. HD lines in the Lyman bands have indeed been observed in the object Q1232 + 082 at $z = 2.34$ [7].

To facilitate this opportunity, a set of highly accurate zero-redshift (laboratory) transition wavelengths of HD

electronic absorption lines is required. Here we report on the spectral calibration of zero-redshift HD lines in the $B^1\Sigma_u^+ - X^1\Sigma_g^+$ Lyman and the $C^1\Pi_u - X^1\Sigma_g^+$ Werner bands, referred to as $L\nu$ and $W\nu$ bands with ν the vibrational quantum number, in the extreme ultraviolet range 100–110 nm at an accuracy of $\Delta\lambda/\lambda \sim 5 \times 10^{-8}$. At present-day large telescopes high-redshift spectra can be obtained at accuracies in the range $(0.2 - 1.0) \times 10^{-6}$ [4]; hence, these new HD laboratory results, which constitute a 2-orders-of-magnitude improvement over existing data [8], will then contribute only slightly in a comparison with astrophysical data. A second ingredient required for a comparison between laboratory and high-redshift data is a theory that relates possible changes in μ to observable shifts in spectra. For H_2 , sensitivity coefficients K_i were previously deduced both in a semiempirical fashion [3] and through quantum-chemical calculations [9]. Here we report on such *ab initio* calculations for K_i coefficients of HD spectral lines.

Laboratory measurements were performed to determine the zero-redshift wavelengths of the most prominent HD spectral absorption lines in the Lyman and Werner bands. The methods employed here for the HD molecule are similar to those described previously for the H_2 spectroscopic studies [10]. A narrow band extreme ultraviolet laser beam, wavelength tunable in the range 100–110 nm, perpendicularly intersects a collimated molecular hydrogen beam (enriched to 90% in HD). Upon resonant excitation a UV beam further ionizes the excited molecules to form HD^+ ions that are detected. The bandwidth of the laser system is better than 250 MHz, corresponding to $\Delta\lambda < 10^{-5}$ nm. The Doppler contribution to the linewidth is small in the perpendicular geometry, while it is further reduced (compared to previous measurements [10]) by selecting the lowest velocity components in the molecular beam. At observed linewidths of ~ 450 MHz the natural line broadening corresponding to an upper state lifetime of

0.5 ns is approached. A typical recording is shown in Fig. 1. The absolute calibration of the HD resonances relies on saturation spectroscopy in I_2 of the visible laser radiation that is pulse amplified and subsequently harmonically converted to extreme ultraviolet light for the spectroscopy. The uncertainty budget for the determination of the resonance wavelengths is similar to that in the H_2 studies, resulting in a relative uncertainty of $\Delta\lambda/\lambda = 5 \times 10^{-8}$ or in absolute terms $\Delta\lambda = 0.000\,005$ nm. The obtained HD line positions are collected in Table I, where a listing is given in units of wavelength (nm), for the purpose of comparison with astrophysical data. Values represent *vacuum* wavelengths. Values presented for the Lyman bands $L0$, $L1$, and $L2$ are taken from Ref. [12], while $L16$ values are from Ref. [13], after a correction for misassignment. The present data set in the range 100–110 nm will not cover the entire range of astrophysical observations of HD, which may well extend to 93 nm. For the moment the laser calibration data of Hinnen *et al.* [14], at an accuracy of 5×10^{-7} , can be included in comparisons with quasar data until more accurate data become available.

Combination differences between $P(J'' + 2)$ and $R(J'')$ lines can be compared with ground state splittings from far-infrared spectroscopy [11]; the average difference of -0.0004 cm^{-1} is well below the accuracy claimed in Table I and hence provides a consistency check on the present results. When comparing the present values with the classical data of Dabrowski and Herzberg [8], we find that these are on average higher by 0.29 cm^{-1} with respect to our values. The lines that present the larger discrepancies are marked as “blended” in Ref. [8].

Level energies in the HD molecule were calculated applying the same method as previously applied for the

D_2 molecule [15], using highly accurate *ab initio* data [16–18]. The four nearby-lying excited electronic states of *ungerade* symmetry, $B^1\Sigma_u^+$, $C^1\Pi_u$, $B^1\Sigma_u^+$, and $D^1\Pi_u$, are strongly coupled; hence, a system of four coupled radial Schrödinger equations was solved [19]. Given in

TABLE I. Measured wavelengths of lines, where $P(J'')$, $Q(J'')$, and $R(J'')$ have the usual meaning of a rotational transition, in the $B^1\Sigma_u^+ - X^1\Sigma_g^+$ Lyman bands (indicated as $L\nu$) and $C^1\Pi_u - X^1\Sigma_g^+$ Werner bands (indicated as $W\nu$) for HD with the Amsterdam narrow band extreme ultraviolet laser facility and the K_i coefficients as calculated. The typical uncertainty in the measured wavelengths is $\Delta\lambda = 0.000\,005$ nm, while that in the sensitivity coefficients is $\Delta K_i = 0.000\,15$.

Line	λ_0 (nm)	K_i	Line	λ_0 (nm)	K_i
L0P1	110.729 245	-0.007 89	L6R3	103.536 700	0.020 64
L0P2	110.911 618 ^a	-0.009 69	L6R4	103.790 376	0.018 31
L0P3	111.166 568 ^a	-0.012 11	L7P1	102.262 976	0.027 22
L0R0	110.584 055	-0.006 54	L7P2	102.425 462	0.025 51
L0R1	110.621 689	-0.006 96	L7P3	102.656 725	0.023 18
L0R2	110.732 767	-0.008 11	L7R0	102.146 045	0.028 31
L1P1	109.340 155	-0.001 73	L7R1	102.191 899	0.027 78
L1P2	109.519 532	-0.003 47	L7R2	102.307 083	0.026 44
L1P3	109.771 190	-0.005 89	L7R3	102.491 165	0.024 53
L1R0	109.200 126	-0.000 38	L7R4	102.743 486	0.022 16
L1R1	109.239 875	-0.000 84	L8P1	101.260 079	0.030 86
L1R2	109.352 696	-0.002 03	L8P2	101.420 145	0.029 15
L1R3	109.538 262	-0.003 84	L8P3	101.648 369	0.026 84
L2P2	108.194 838	0.002 24	L8R0	101.146 180	0.031 93
L2R0	107.883 104	0.005 28	L8R1	101.192 611	0.031 37
L2R1	107.924 459	0.004 83	L8R2	101.307 728	0.030 07
L3P1	106.758 691	0.009 39	L8R3	101.491 089	0.028 10
L3P2	106.932 076	0.007 62	L8R4	101.742 011	0.025 75
L3P3	107.176 737	0.005 26	L9P1	100.300 544	0.034 25
L3P4	107.491 936	0.002 51	L9P2	100.458 215 ^a	0.032 57
L3R0	106.627 568	0.010 61	L9P3	100.683 381	0.032 50
L3R1	106.670 180 ^a	0.010 14	L9R0	100.189 413	0.035 30
L3R2	106.784 752	0.008 89	L9R1	100.236 220	0.034 71
L3R3	106.970 884	0.007 02	L9R2	100.351 037	0.033 38
L3R4	107.227 954	0.004 74	L9R4	100.782 338	0.028 95
L4P1	105.556 544	0.014 35	L16P1	94.583 158	0.050 96
L4P2	105.727 058	0.012 63	L16P2	94.696 040	0.049 33
L4R0	105.429 354	0.015 57	L16P3	94.872 485	0.047 11
L4R2	105.587 950	0.013 79	L16R0	94.534 329	0.051 86
L5P1	104.408 545	0.018 98	L16R1	94.629 663	0.051 27
L5P2	104.576 264	0.017 24	L16R2	94.773 610	0.049 87
L5P3	104.814 022	0.014 86	L16R3	94.981 204	0.047 86
L5R0	104.285 005	0.020 13	W0P2	101.000 728	-0.006 76
L5R1	104.329 502	0.019 59	W0P3	101.176 906	-0.008 57
L5R2	104.444 653	0.018 32	W0P4	101.394 793	-0.010 50
L5R3	104.630 031	0.016 44	W0Q1	100.820 291	-0.004 86
L5R4	104.884 999	0.014 11	W0Q2	100.908 028	-0.005 90
L6P1	103.311 624	0.023 25	W0Q4	101.212 698	-0.008 80
L6P2	103.476 669	0.021 54	W0R0	100.729 020	-0.003 90
L6P3	103.711 120 ^a	0.019 23	W0R1	100.725 357	-0.003 88
L6R0	103.191 493	0.024 40	W0R2	100.765 326	-0.004 41
L6R1	103.236 721	0.023 89	W0R3	100.848 750	-0.005 28
L6R2	103.351 929	0.022 55			

^aObtained using the combination differences from Ref. [11].

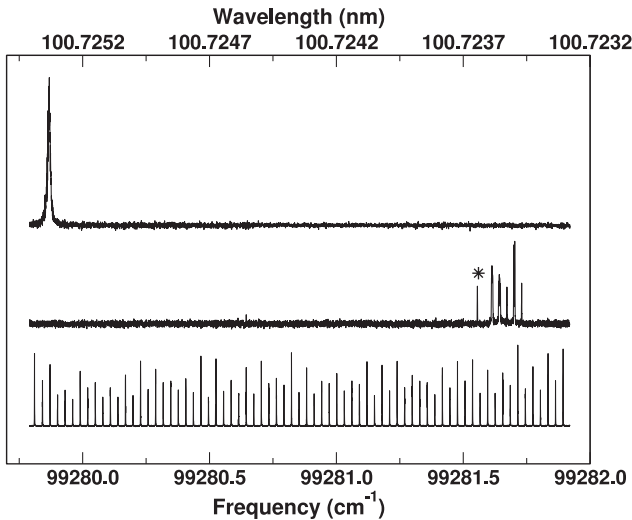


FIG. 1. Recording of the W0R1 line of HD (upper) with étalon markers (lower) and an I_2 -saturation spectrum (middle) for calibration. The line marked with an asterisk is the t -hyperfine component of the $B-X(11, 2) P(52)$ rotational line in I_2 at $16\,546.926\,62$ cm^{-1} used as an absolute reference.

matrix form, the system is written as

$$\left\{ -\frac{1}{2\mu_{\text{HD}}} \left[\mathbf{I} \frac{d^2}{dR^2} - \mathbf{I} \frac{J'(J'+1)}{R^2} + \mathbf{A}(R) + 2\mathbf{B}(R) \frac{d}{dR} \right] + \mathbf{U}(R) \right\} \varphi(R) = E\varphi(R) \quad (1)$$

with μ_{HD} the reduced mass of HD and $\mu_{\text{HD}'}$ its dimensionless form

$$\mu_{\text{HD}} = \frac{m_p m_d}{m_d + m_p}; \quad \mu_{\text{HD}'} = \mu_{\text{HD}}/m_e, \quad (2)$$

and m_p and m_d the mass of the proton and deuteron. \mathbf{I} is the identity matrix and $\mathbf{U}(R)$ is the diagonal matrix of adiabatic potential curves. The diagonal elements of the $\mathbf{A}(R)$ matrix are the adiabatic corrections obtained from Ref. [16] for the $^1\Sigma_u^+$ states and from Ref. [17] for the $^1\Pi_u$ states. The off-diagonal elements in \mathbf{A} involve both nonadiabatic couplings between states of the same symmetry ($\Sigma - \Sigma$ or $\Pi - \Pi$) and rotational couplings between $\Sigma - \Pi$ states, and finally $\mathbf{B}(R)$ is the radial coupling matrix. The nonadiabatic corrections are obtained from Ref. [18].

The Fourier Grid Hamiltonian method [20] was used to solve the coupled equations, where all the energy values and the coupled-channel wave functions are obtained in one single diagonalization of the Hamiltonian matrix expressed in a discrete variable representation. Solutions of nonadiabatic wave function are represented as a four-component vector:

$$\varphi_i(R) = \{\varphi_{n,i}(R), \varphi_{n',i}(R) \dots\}, \quad (3)$$

where the label n refers to the particular electronic state belonging to $\{B, B', C, D\}$.

The $X^1\Sigma_g^+$ ground state was treated separately. Its rovibrational energy levels were calculated by solving the single Schrödinger equation

$$\left\{ -\frac{1}{2\mu_{\text{HD}}} \frac{d^2}{dR^2} + \frac{J''(J''+1)}{2\mu_{\text{HD}}R^2} + U_X(R) \right\} \varphi_X(R) = E_X \varphi_X(R) \quad (4)$$

for each rotational quantum number J'' in the adiabatic approximation, adding the corresponding centrifugal term to the *ab initio* potential $U_X(R)$, which includes the adiabatic correction to the Born-Oppenheimer potential, computed by Wolniewicz [21]. The relativistic and radiative corrections [22] were also taken into account in the present calculations.

The $X^1\Sigma_g^+$ state in hydrogen undergoes a weak perturbative shift caused by nonadiabatic interactions with excited states of symmetries Σ_g and Π_g , leading to regular shifts ΔE_X . This was taken into account by means of semiempirical relations [23]

$$\Delta E_X = E_{\Sigma_g} + J''(J''+1)E_{\Pi_g}. \quad (5)$$

Furthermore, the HD molecule is subject to electronic inversion symmetry breaking ($g - u$), giving rise to inter-

actions between states of opposite g and u symmetries [24]. This causes a supplementary downward shift of the $X^1\Sigma_g^+$ levels of HD by

$$\Delta E_X^{gu} = E_{\Sigma_u} + J''(J''+1)E_{\Pi_u}. \quad (6)$$

The latter correction was included in the calculation of level energies of the $X^1\Sigma_g^+$ quantum states and the wavelengths of the $P(J'')$, $Q(J'')$, and $R(J'')$ transitions in the corresponding Lyman and Werner bands. The effects of symmetry-breaking interactions between $EF^1\Sigma_g^+$ levels and B and C levels [8,14] are not included here because only incomplete *ab initio* coupling operators are available [25]. Tentative calculations including these data showed that for the lines reported in Table I, the corresponding excited levels are almost unperturbed, resulting in wavelength shifts less than 10^{-4} nm and negligible changes in K_i coefficients.

The procedure to solve the coupled Schrödinger equations, derive the level energies for ground and excited states, and determine the transition wavelengths λ_i of the HD lines, was performed for various values of the relevant mass for the problem, i.e., the dimensionless $\mu_{\text{HD}'}$ as defined in Eq. (2). As a first, intuitive, and obvious approximation, the reduced mass of HD is considered to scale as the proton mass m_p , hence $\mu_{\text{HD}'} = a\mu$, with a a proportionality constant of no influence on the analysis ($a = 0.667$ for HD, while $a = 0.5$ for H_2). The physical background of this assumption is that baryonic mass is considered independent of quark composition. Then the sensitivity coefficient can be expressed in the usual form as

$$K_i = \frac{d \ln \lambda_i}{d \ln \mu_{\text{HD}'}} = \frac{d \ln \lambda_i}{d \ln \mu}. \quad (7)$$

The HD transition wavelengths were calculated for a range of values of the proton-electron mass ratio μ around the experimental value of $\mu_0 = 1836.152\,672\,61$ [26]. Since an indication for a possible variation was found at

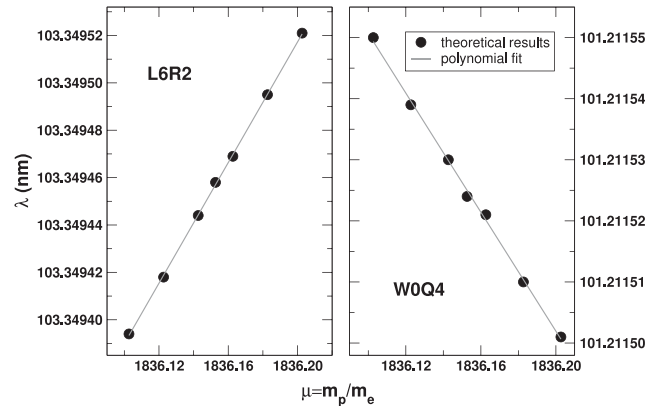


FIG. 2. Calculation of transition wavelengths as a function of μ for two lines in HD. It illustrates that some lines undergo an upward shift and others a downward shift upon a variation of μ .

a level of $\Delta\mu/\mu \sim 2.4 \times 10^{-5}$, the values were taken on a grid covering [1836.10–1836.21]. In Fig. 2 results for transition wavelengths as a function of μ are displayed for some HD lines. For each spectral line seven calculations were performed (at μ_0 and other points as represented in Fig. 2). Values for K_i for each individual spectral line i were then computed by performing a polynomial fit to the slopes, a procedure also yielding an uncertainty estimate on the results. The resulting values for K_i are included in Table I with the experimental wavelengths.

The package of present observations of accurate wavelength positions λ_i and the calculation of sensitivity factors K_i for the HD molecule can be utilized in estimating constraints on possible variations of the proton-electron mass ratio μ , or in obtaining proof of a variation of this fundamental constant. HD and H₂ lines can be included in a comprehensive analysis of $\Delta\mu/\mu$ through fitting of all molecular lines to

$$\frac{\lambda_i}{\lambda_0} = (1 + z_{abs}) \left(1 + \frac{\Delta\mu}{\mu} K_i \right) \quad (8)$$

with z_{abs} the overall redshift of the absorbing cloud.

As discussed in Ref. [5], the proton mass m_p is proportional to a fundamental parameter, the quantum chromodynamic scale Λ_{QCD} . A fundamental dimensionless parameter Λ_{QCD}/m_e can be defined in the standard model that is directly probed in the studies comparing molecular lines in the laboratory and at high redshift via

$$\frac{\Delta(\Lambda_{\text{QCD}}/m_e)}{\Lambda_{\text{QCD}}/m_e} = \frac{\Delta\mu}{\mu}. \quad (9)$$

Hence, through $\Delta\mu/\mu$ effectively the evolution of the strong force with respect to that of the electroweak scale is probed.

It is conceivable to go one step beyond this first-order evaluation; hypothetically, in second order masses of quark constituents may undergo a different time evolution. Such composition-dependent scenarios have been predicted in the framework of string theorie(s) [27], and they could be assessed by analyzing H₂ and HD spectra simultaneously. Under such conditions Eq. (7) would no longer hold and HD sensitivity coefficients could be calculated, adapted to the scenario of choice by invoking different effects on m_p/m_e and m_d/m_e . Probing of such second-order effects would require astrophysical spectra of extreme quality in terms of resolution and signal-to-noise ratio, beyond that of Ref. [7], that may be considered as first observations. A future generation of planned telescopes with 30–40 m dishes may provide such data.

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