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Have the Masses of Molecules Changed during the Lifetime of the Universe?

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Abstract—Based on an analysis of observations of molecular lines in the spectra of extragalactic objects, we have checked the nucleon masses for possible variability (with respect to the electron mass). This kind of variability is suggested by a number of many-dimensional cosmological models. No statistically significant variations in the masses of H_2 and CO molecules and, hence, in the masses of the proton m_p and neutron m_n at the epoch corresponding to cosmological redshifts $z \sim 0.2$–3, were found. Electronic-vibrational-rotational lines of the H_2 molecule at $z = 2.811$ were analyzed in the spectrum of the quasar PKS 0528-250 obtained by Lanzetta *et al.* (1995), and an upper limit of $|\Delta m_p/m_p| < 2 \times 10^{-4}$ was found. This constraint improves the previously reported result by one order of magnitude. Measurements of molecular CO radio lines in the spectra of the galaxy IRAS F10214+4724 at $z = 2.286$ and the quasars PKS 1157+014 at $z = 1.944$ and PKS 1413+135 at $z = 0.247$ were analyzed. The bounds $|\Delta M/M| < 8 \times 10^{-4}$, 3×10^{-4}, and 2×10^{-4} on possible changes (ΔM) in the mass of the CO molecule ($M \propto m_n + m_p$) were obtained for the above three values of z.

1. INTRODUCTION

Variability of the fundamental physical constants, suggested by a number of theoretical models for the unification of interactions [see, e.g., the review by Okun' (1991)], is of fundamental importance in cosmology. Spectroscopic studies of quasars provide a check on whether the fundamental constants have changed during the last 90% of the lifetime of the Universe. Temporal variations in the electromagnetic coupling constant can be directly derived from the relative magnitude of fine splitting of lines of lithium- and sodium-like ions (Varshalovich and Potekhin 1994). It is impossible to directly obtain such an estimate for variations in the parameters of strong and weak coupling, but their variability could be manifested in a change in the masses of the proton m_p and the neutron m_n with respect to the electron mass. The latter will be considered as a unit mass, as is commonly done in atomic physics.

Note that a decrease in m_n (or an increase in m_p) by only 0.08% would have catastrophic consequences: this would lead to a merger of protons and electrons to form neutrons and neutrinos, whereas the reverse process—neutron beta-decay—would become energetically unfavored. The Universe would become qualitatively different. Does such a catastrophe threaten the Universe in the future? Changes in m_n and m_p in the past, if detected, could be indicative of this. By comparing the predictions of the standard model of primordial nucleosynthesis with observational data on the relative ^{4}He abundance at the current epoch, Kolb *et al.* (1986) concluded that the mass difference ($m_n - m_p$) at the nucleosynthesis epoch corresponding to a cosmological redshift of $z \sim 10^9$ did not differ from the present value by more than a few percent. However, this consideration depends both on the assumptions underlying the cosmological and primordial-nucleosynthesis models and on the values of a number of physical parameters that are not known very well. It should also be noted that since the law of possible variations in the fundamental constants is not known in advance, and different relations are theoretically possible (Marciano 1984), it is desirable to obtain similar constraints for different z.

In this study, we checked the nucleon masses m_n and m_p at $z \leq 3$ for possible variability. Our test is based on observations of molecular lines in the spectra of four extragalactic objects: optical absorption lines of the H_2 molecule in the spectrum of the quasar PKS 0528–250 (Levshakov and Varshalovich 1985; Foltz *et al.* 1988; Lanzetta *et al.* 1995) and radio lines of the CO molecule in the spectra of the objects IRAS F10214+4724 (Brown and Vanden Bout 1991), PKS 1157+014 (Val'tts *et al.* 1993), and PKS 1413+135 (Wiklind and Combes 1994). We obtained upper limits on possible deviations of the masses of the H_2 and CO molecules at $z = 2.811$, 2.286, 1.944, and 0.247 from their current

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values, which allowed us to estimate possible changes in the nucleon masses.

2. AN UPPER LIMIT ON CHANGES IN THE PROTON MASS AT $z = 2.811$

The energies of electronic, vibrational, and rotational excitations of a diatomic molecule AB depend differently on its reduced mass $M = m_A m_B/(m_A + m_B)$. To a first approximation, these energies are proportional to M^0, $M^{-1/2}$, and M^{-1}, respectively. Therefore, by comparing the ratios of the wavelengths of various electronic-vibrational-rotational lines in the spectrum of a quasar corresponding to some redshift z and in a laboratory spectrum (i.e., at $z = 0$), one can detect a change in M. The potential of such measurements, first pointed out by Thompson (1975), has already been exploited by Foltz *et al.* (1988), Varshalovich and Levshakov (1993), and Varshalovich and Potekhin (1995). Recently, it has become possible to significantly improve these earlier results through high-resolution measurements of the spectrum of PKS 0528–250 made by Lanzetta *et al.* (1995).

Let $\lambda_j(z)$ be the wavelength of some electronic-vibrational-rotational line of a molecule at cosmological redshift z, and ΔM be the difference between the reduced mass at the epoch corresponding to z and its present value. Since the reduced mass of the H_2 molecule is $m_p/2$, $\Delta M/M = \Delta m_p/m_p$ for this molecule. To second order in $(\Delta M/M)$, we may write

$$\lambda_j(z) = \lambda_j(0)[1 + K_j \Delta M/M](1 + z), \qquad (1)$$

where the coefficients

$$K_j = (M/\lambda_j)\mathrm{d}\lambda_j/\mathrm{d}M \qquad (2)$$

determine the sensitivity of the wavelengths to a change in M. An important point is that the values of K_j differ for different lines. Thus, if the reduced mass M of the molecule at the epoch z differs from the present value, then $\lambda_j(z)$ and K_j must be linearly correlated. This correlation underlies the method.

The coefficients K_j have been previously calculated by Varshalovich and Levshakov (1993) from Dunham's spectroscopic constants for the H_2 molecule using theoretical ideas about the dependence of these constants on the mass of the molecule. Varshalovich and Potekhin (1995) employed another method for calculating the same coefficients that is not subject to uncertainties associated with these theoretical ideas, and that is based on experimental energies of electronic-vibrational-rotational terms of the H_2, HD, D_2, and T_2 molecules. It turns out that these two independent methods yield similar values of K_j, thus confirming their reliability.

In this study, we use the first method for calculating the coefficients K_j. Let us consider it in more detail. For each electronic-vibrational-rotational band, the wavelength of a transition between two states with vibrational and rotational quantum numbers (v, J) and (v', J') can be written as

$$\lambda = [\omega^u_{vJ} - \omega^l_{v'J'}]^{-1}, \qquad (3)$$

where the superscripts l and u refer to the lower and upper levels; for each of these levels,

$$\omega_{vJ} = \sum_{ik} Y_{ik}\left(v + \frac{1}{2}\right)^i (J(J+1))^k. \qquad (4)$$

The spectrum in question exhibits the Lyman $(X^1\Sigma^+_g - B^1\Sigma^+_u)$ and Werner $(X^1\Sigma^+_g - C^1\Pi_u)$ bands. The parameters Y_{ik} for the electronic states corresponding to the above transitions are given, for example, in Huber and Herzberg (1984). The coefficient Y_{00} was redefined in such a way that the energy of each rotational-vibrational band was measured from the ground level. For the $C^1\Pi_u$ states, the factor $(J(J + 1))$ in the terms with $k = 1$ of equation (1) was replaced by $(J(J + 1) - 1)$ to allow for the contribution from the projection $\Lambda = 1$ of the electronic orbital momentum on the axis of the molecule. It should be emphasized that the fit (4) with the currently known experimental parameters Y_{ik} is justified for not too large quantum numbers v and J.

The Born–Oppenheimer method shows that the parameters Y_{ik} are proportional to $M^{-k-i/2}$ with fairly high accuracy. Thus, $\partial Y_{ik}/\partial M \approx -(i/2 + k)Y_{ik}/M$. Using this approximation, we can readily derive the coefficients K_j from equations (2)–(4). Our calculated coefficients K_j are shown in the figure.

These values of K_j were used in the analysis of the spectrum of PKS 0528–250 performed by Lanzetta *et al.* (1995). As a result, they obtained the following estimate:

$$\Delta M/M = \left(8.3^{+6.6}_{-5.0}\right) \times 10^{-5} \quad \text{at } z = 2.8107998(24) \qquad (5)$$

from which it follows that the difference between the proton mass and its present value does not exceed (at the 2σ significance level)

$$|\Delta m_p/m_p| < 0.0002. \qquad (6)$$

This result improves the majorizing estimate of Varshalovich and Levshakov (1993) $|\Delta m_p/m_p| < 0.005$ and the estimate of Varshalovich and Potekhin (1995) $|\Delta m_p/m_p| < 0.002$ obtained previously from an analysis of the data by Foltz *et al.* (1988), having not so high resolution, by one order of magnitude.

The bound (6) places an upper limit on the mean rate of change of the proton mass during the $\sim 10^{10}$ years that have elapsed since the time when

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the H_2 spectrum under consideration was formed: on average, the rate of change of m_p could be no more than $2 \times 10^{-14} m_p\ \mathrm{yr}^{-1}$.

3. UPPER LIMITS ON CHANGES IN THE MASS OF THE CO MOLECULE

Just as the H_2 resonance lines provide information on the proton mass m_p, the neutron mass can be judged from the spectra of heavy elements. Presently, more and more new radio observations of rotational lines of the CO molecule at large redshifts continue to be reported. If there were shifts in the frequencies of a few rotational lines in a single spectral system, one could estimate the possible change in the reduced mass of this molecule by the method outlined in the preceding section. The necessary coefficients K_j are listed in the table. We used the data of George *et al.* (1994) to calculate them. Despite the small scatter in the quoted values of K_j, one could estimate the correlation coefficient between $\lambda_j(z)$ and K_j for the actually achievable accuracy of radio measurements of the order of a few kilometers per second.

Unfortunately, only single CO lines in different spectral systems are currently known. Nevertheless, we can estimate the possible change in the reduced mass by comparing the redshift in each measured CO line with the redshift in atomic and ionic lines in the same spectral system. To use this method, we also assume that the CO lines formed in the same region of interstellar gas as the accompanying atomic and ionic lines.

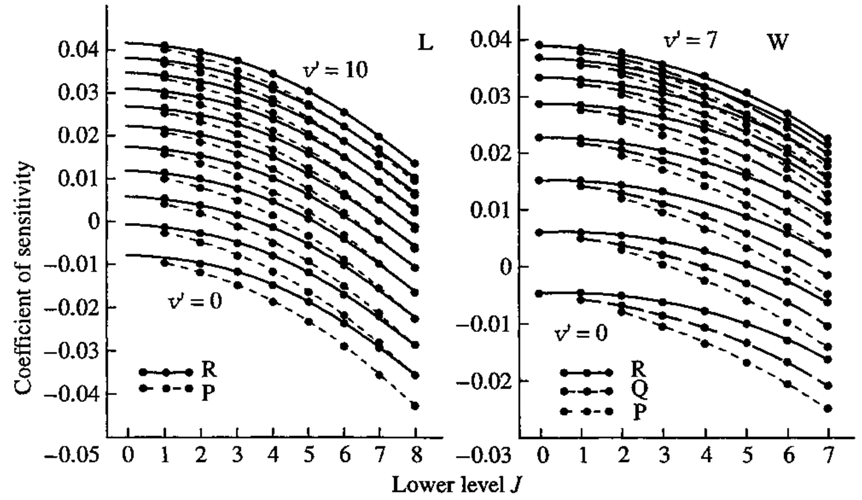
We analyzed three spectral systems in which fairly narrow radio lines of $^{12}C^{16}O$ molecules were observed along with optical spectral lines of ions.

One of these radio lines corresponding to the $J = 3 \longrightarrow 2$ (105233.1 MHz) transition was detected by Brown and Vanden Bout (1991) during their observations of the galaxy IRAS F10214+4724. Earlier, Rowan-Robertson *et al.* (1991) found the redshift of this galaxy from narrow emission lines of the C IV, C III, C II, [Ne IV], Mg II, N V, and Lyα ions in the optical and ultraviolet to be $z = 2.286 \pm 0.001$. These lines correspond to electronic radiative transitions and are essentially independent of the reduced mass M of the molecule. At the same time, the frequency of a CO rotational transition is proportional to M^{-1}. Hence, the relative difference $\Delta z/(1 + z)$ between the redshifts in the optical and radio lines originating from the same galaxy is, to within some small corrections, equal to $\Delta M/M$.

According to Brown and Vanden Bout (1991), the peak of the CO radio line occurred at $z = 2.2867 \pm 0.0003$. However, since the line can be composite and a statistically significant (4σ) excess of the signal above the background was observed in the channels corresponding to radial velocities between −115 and +250 km s^{-1}, we take the wider confidence interval, $z = 2.2862 \pm 0.0006$, as more reliable. Allowing for uncertainty in the optical redshift (~0.001), we obtain $\Delta z = (0.2 \pm 1.2) \times 10^{-3}$, and, consequently,

$$\Delta M/M = (0.6 \pm 3.7) \times 10^{-4} \text{ at } z = 2.286. \qquad (7)$$

Another observation pertains to the $z = 1.9438$ absorption system in the spectrum of the quasar PKS



Coefficients (K_j) of sensitivity of wavelengths (λ_j) to a change in the mass of the H_2 molecule [formula (2)] for transitions of the Lyman (L) $X^1\Sigma_g^+ - B^1\Sigma_u^+$ and Werner (W) $X^1\Sigma_g^+ - C^1\Pi_u$ bands. The solid lines connect the values of K_j for the transitions of the R branch $(0, J - v', J + 1)$; the short dashes are for the transitions of the P branch $(0, J \longrightarrow v', J - 1)$, and the long dashes are for the transitions of the Q branch $(0, J \longrightarrow v', J)$ (the latter are allowed only for the Werner band).

Frequencies ν (in GHz) and coefficients K_J of sensitivity to mass changes for transitions between the rotational levels of the ground $X^1\Sigma^+$, v = state of the $^{12}C^{16}O$ molecule

J	J'	$\nu(J \longleftrightarrow J')$	K_J
1	0	115.271 202 5	0.9931962
2	1	230.538 001	0.9931769
3	2	345.795 991	0.9931449
4	3	461.040 769	0.9931000
5	4	576.267 933	0.9930423
6	5	691.473 078	0.9929718
7	6	806.651 803	0.9928884
8	7	921.799 706	0.9927923
9	8	1036.912 386	0.9926832
10	9	1151.985 444	0.9925614

1157+014. In this system, Val'tts *et al.* (1993) measured the CO radio emission line corresponding to the $J = 2$–1 transition. The identification of this line is not too reliable (at the 2.5σ level), but, if confirmed by future observations, it would make it possible to significantly refine estimate (7). According to Val'tts *et al.* (1993), the detected CO radio line corresponds to radial velocities (20 ± 17) km s^{-1}, whereas the width of the H I absorption lines that determine the optical redshift is equal to 18 km s^{-1}. As a result, we obtain the estimate

$$\Delta M/M = \frac{(20 \pm 30)\ \text{km}\ \text{s}^{-1}}{3 \times 10^5\ \text{km}\ \text{s}^{-1}} \qquad (8)$$

$$= (0.7 \pm 1.0) \times 10^{-4} \quad \text{at} \quad z = 1.944.$$

Recently, Wiklind and Combes (1994) have measured a narrow CO absorption line ($J = 0$–1; 92460.3 MHz) in the spectrum of PKS 1413+135. The redshift of this line matches the redshift $z = 0.24671$ of the line-of-sight galaxy, in which the H I 21 cm line with a FWHM of 18 km s^{-1} was previously measured (Carilli *et al.* 1992). The CO line with a much smaller FWHM is displaced from the H I line center by –11 km s^{-1}. Hence,

$$\Delta M/M = (-4 \pm 6) \times 10^{-5} \text{ at } z = 0.247. \qquad (9)$$

Upper limits on the relative change in the mass of the CO molecule can be derived from estimates (7)–(9): at the 2σ level, we have $|\Delta M/M| < 8 \times 10^{-4}$, 3×10^{-4} and 2×10^{-4} at $z = 2.286$, 1.944, and 0.247, respectively.

4. CONCLUSION

By analyzing the wavelengths of the resonance spectral lines of diatomic molecules observed in the spectra of four objects with various cosmological redshifts z, we have established new upper limits on possible changes in the nucleon masses.

An analysis of H_2 absorption lines in the high-resolution spectrum of the quasar PKS 0528–250 obtained by Lanzetta *et al.* (1995) yielded constraint (6) on the possible difference of the proton mass at $z = 2.811$ from its current value. This constraint improves the estimates previously obtained by different methods from an analysis of lower-quality spectroscopic data (Varshalovich and Levshakov 1993; Varshalovich and Potekhin 1995) by one order of magnitude.

A comparison of the redshift of a CO emission line in the spectrum of the galaxy IRAS F10214+4724 exhibiting known optical emission lines gave estimate (7) for the possible change in the reduced mass of the molecule at $z = 2.286$. We found no statistically significant variation. The three times more stringent estimate (8) was obtained by comparing the optical redshift in the absorption system ($z = 1.944$) in the spectrum of the quasar PKS 1157+014 with that of a candidate for the CO emission radio line in the same system. Finally, a comparison of the redshifts in the H I and CO radio absorption lines in the spectrum of PKS 1413+135 gives estimate (9), which suggests a still more stringent constraint on the reduced mass of the molecule at a lower redshift of $z = 0.247$.

The fact that the variations in the reduced masses of the H_2 and CO molecules lie within a statistical uncertainty interval of ~0.02% suggests that in the ~10^{10} years that have elapsed since the time when the spectra under consideration were formed, the nucleon masses have not changed by more than a few hundredths of a percent and the binding energy of the ^{12}C and ^{16}O nuclei, by more than a few percent. The possibility that changes in the nucleon masses may conspire to cancel changes in the nuclear binding energy cannot in principle be ruled out, but such a conspiracy seems highly unlikely.

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