Characterizing of variation in the proton-to-electron mass ratio via precise measurements of molecular vibrational transition frequencies

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ARTICLE INFO

ABSTRACT

We propose measurements of $(v, N) = (0, 0) \rightarrow (1, 0)$ transition frequencies of alkali earth–alkali diatomic molecules and alkali earth-hydride molecular ions. We show that the vibrational transitions considered here are useful for detecting a possible variation in the proton–electron mass ratio, possibly at the 10^{-16} sensitivity level. In the experiments, cold alkali earth–alkali molecules, obtained by binding cold atoms or by molecular laser cooling, are trapped by laser light at a frequency, at which the Stark shift is eliminated. Alkali earth-hydride molecular ions are sympathetically cooled with another ion to form a string crystal at the position, at which the electric field is zero. The vibrational transitions are observed by the Raman transition induced by two lasers with frequencies, at which the Stark shift is eliminated.

1. Introduction

Since Dirac's 1937 formulation of the large number hypothesis that aimed to tie together the fundamental constants of physics [1], researchers have been interested in testing whether these constants are indeed fixed over time and space. Webb et al. and Berengut and Flambaum showed that the variation in the fine-structure constant α observed in quasars depends on not only the distance (time) but also the angle between the observer and the Australian dipole [2]. Rosenband et al. measured the ratio of the ^{27}Al$^+$ ^{1}S$_0$–^{3}P$_0$ and ^{199}Hg$^+$ ^{2}S$_{1/2}$–^{2}D$_{5/2}$ transition frequencies and determined the variation in α to be $(-1.6 \pm 2.3) \times 10^{-17}$/year [3].

It is also useful to test the variation in the proton-to-electron mass ratio m_p/m_e because the ratio of variations in α and m_p/m_e is important for a grand unification theory [4]. Molecular transition frequencies are useful for detecting the variation in m_p/m_e, because vibrational and rotational transition frequencies are approximately proportional to $(m_p/m_e)^{-1/2}$ and $(m_p/m_e)^{-1}$, respectively. Van Weerdenburg et al. observed the variation in m_p/m_e from the transition frequencies of H_2 molecules in the quasar source J2123-005 ($z = 2.059$, corresponding to 10.5 billion light-years) using ESO-VLT (Paranal, Chile) and the Keck telescope (Hawaii) and obtained the results of $\Delta(m_p/m_e)/(m_p/m_e) = (8.5 \pm 3.6_{stat} \pm 2.2_{syst}) \times 10^{-6}$

and $(5.6 \pm 5.5_{stat} \pm 2.9_{syst}) \times 10^{-6}$, respectively [5]. In addition, Bagdonaite et al. compared the transitions of CH_3OH molecules from PKS1830-211 ($z = 0.89$, corresponding to 7 billion light years) with those measured in a laboratory and obtained the result $\Delta(m_p/m_e)/(m_p/m_e) = (0 \pm 1) \times 10^{-7}$ [6].

To distinguish the spacial and temporal variations in fundamental constants, laboratory measurements are also required. The upper limit of 10^{-16}/years for the variation in a parameter that depends on both m_p/m_e and α has been obtained by measuring the ratio between the atomic transition frequencies in the optical region and Cs hyperfine frequency [7–10]. To measure the pure variation in m_p/m_e, it is preferable to compare frequencies of molecular transitions with those of atomic transitions, whose dependence on α and m_p/m_e are negligibly small (e. g., the ^{1}S$_0$–^{3}P$_0$ transition frequencies of the ^{27}Al$^+$ ion [3] or ^{87}Sr atom [11,12]). The molecular transition to be measured should be chosen after considering both sensitivity to (m_p/m_e) and accuracy of the measurement. Several authors have presented molecular transition frequencies with significantly higher sensitivities to m_p/m_e than to pure vibrational or rotational transition frequencies [13–15]. However, those transitions are difficult to measure with an uncertainty lower than 10^{-14}.

In this study, we discuss methods for characterizing the variation in (m_p/m_e) by measuring the vibrational transition frequencies of XY molecules in an optical lattice or XH$^+$ molecular ions in a string crystal with uncertainties lower than 10^{-16}. Here, X denotes even isotopes of alkali earth atoms (^{174}Yb, ^{88}Sr, and ^{40}Ca) and Y denotes alkali atoms (H, ^{6}Li, and ^{23}Na) [16–24].

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2. Possibility to test the variation in m_p/m_e

A test for the variation in (m_p/m_e) is possible when

$$\left|\frac{\Delta f_c}{f_c}\right| = \left|\lambda\frac{\Delta(m_p/m_e)}{(m_p/m_e)}\right| > \frac{\delta f_c}{f_c}, \tag{1}$$

is satisfied, where Δf_c is the variation in the transition frequency f_c induced by the variation in (m_p/m_e), $\Delta(m_p/m_e)$, and δf_c is the measurement uncertainty of f_c. The parameter λ demonstrates the sensitivity of f_c to (m_p/m_e), where $\lambda = -0.5$ for a pure harmonic vibration. This study shows that the $X^n\Sigma(v,N) = (0,0) \rightarrow (1,0)$ transition frequencies f_c of XY molecules ($n = 2$) or XH^+ molecular ions ($n = 1$) can be measured with an uncertainty below 10^{-16}, therefore, the variation in (m_p/m_e) on the order of 2×10^{-16} can be detected. Here v and N denote the quantum numbers for the vibration and rotation, respectively.

Table 1 lists values of f_c and λ for XY molecules and XH^+ molecular ions, obtained by ab initio calculations [25–28] (measurements for ^{40}CaH [29]). In addition, the natural linewidths f_N are listed for ^{40}CaH and XH^+ transitions, which are larger than 1 Hz. The natural linewidths of X^6Li and X^{23}Na molecules are less than 10 mHz, and spectral linewidths are obtained from probe laser linewidths, whose lower limit is currently 40 mHz [30]. The uncertainty in the frequency is given by the frequency shifts shown below.

2.1. Zeeman shift

In general, energy eigen-states $\Psi = X^{2S+1}\Sigma(v,N,J,F,M_F)$ $\left(\left(\vec{J} = \vec{S} + \vec{N}, \vec{F} = \vec{J} + \Sigma_i\vec{I_i}\right)\right)$ are given by

$$\Psi = \Sigma a_m \psi\big(M_S(m), M_N(m), M_{I_i}(m)\big),$$

$$M_F = M_S(m) + M_N(m) + \Sigma M_{I_i}(m),$$

$$-Q \leqslant M_Q \leqslant Q \quad Q = S,N,I_i,F \tag{2}$$

where S, and I_i denote the quantum numbers of electron spin and nuclear-spin of the i-th nucleus, respectively. M_Q denotes the component of $Q(= S,N,I_i,F)$ parallel to the magnetic field. The Zeeman energy shift induced by the magnetic field B at Ψ is given by

$$E_Z(\Psi) = h\mu_B B[g_S(\Psi)\langle M_S(\Psi)\rangle + g_N(\Psi)\langle M_N(\Psi)\rangle + \Sigma_i g_{Ii}(\Psi)\langle M_{Ii}(\Psi)\rangle],$$

$$\langle M_Q\rangle = \Sigma|a_m|^2 M_Q(m), \tag{3}$$

where $\mu_B(= 1.4 \text{ MHz/G})$ denotes the Bohr magneton and g_Q denotes the g-factor of Q ($g_S = 2.003$ and g_N and g_I are of the order

of 10^{-3}). In general, a_m depends on B, and the Zeeman shift is non-linear. For a low magnetic field, the Zeeman shift is approximately given by

$$E_Z(\Psi)/h = C_1 B + C_2 B^2, \tag{4}$$

where C_1 and C_2 are the first and second order Zeeman coefficients, respectively. C_1 is given by a_m with $B = 0$ and C_2 is given by the fine and hyperfine splittings, which depend on the vibrational–rotational states. However, for stretched states (defined by $F = S + N + I_{tot}$, $M = \pm F$, where $I_{tot} = \Sigma_i I_i$), $\langle M_Q\rangle$ are all determined to be $\pm Q$ and E_Z is strictly proportional to B. The Zeeman shift in the transition frequency between the a and b states is given by.

$$\delta f_Z(\Psi_a, \Psi_b) = \frac{E_Z(\Psi_a) - E_Z(\Psi_b)}{h}. \tag{5}$$

When both a and b states are stretched states with the same values of (S,N,I_i,F,M_F), the Zeeman shift in the transition frequency is given by

$$\delta f_Z(\Psi_a, \Psi_b) = \mu_B B[S(g_S(\Psi_a) - g_S(\Psi_b)) + N(g_N(\Psi_a) - g_N(\Psi_b)) \\ + \Sigma_i I_i(g_{Ii}(\Psi_a) - g_{Ii}(\Psi_b))]. \tag{6}$$

Here, we consider the molecules (neutral or ion) in the $^1\Sigma$ state. Then

$$E_Z(\Psi) = h\mu_B B\big(g_N(\Psi)\langle M_N(\Psi)\rangle + \Sigma_i g_{I_i}(\Psi)\langle M_{I_i}(\Psi)\rangle\big). \tag{7}$$

For the $(N,F,M_F) = (0,I_{tot},\pm I_{tot})$ state,

$$E_Z(\Psi) = \pm h\mu_B B\Sigma_i g_{I_i}(\Psi)I_i. \tag{8}$$

Actually, $|g_I(v = 1) - g_I(v = 0)|/g_I(v = 0)$ is estimated to be on the order of 10^{-7} [24], and δf_Z for the $(v,N,F,M) = (0,0,I_{tot},I_{tot}) \rightarrow (1,0,I_{tot},I_{tot})$ transition is proportional to B with a coefficient on the order of 0.1 mHz/G. Because g_N and $|g_N(v = 1) - g_N(v = 0)|/g_N(v = 0)$ are on the order of 10^{-3} and 10^{-2}, Zeeman coefficients for the $(0,0,I_{tot},I_{tot}) \rightarrow (1,1,I_{tot} + 1,I_{tot} + 1)$ and $(0,1,I_{tot} + 1,I_{tot} + 1) \rightarrow (1,1,I_{tot} + 1,I_{tot} + 1)$ transition frequencies are on the order of 1 kHz/G and 10 Hz/G, respectively.

This estimate for the Zeeman shift in the transition frequency between stretched states is valid also for molecules in the $^2\Sigma$ state, because g_S does not depend on the vibrational state. For molecules in the $^n\Sigma$ state ($n \geqslant 3$), the spin–spin interaction couples the $(N,M_N,M_S) = (0,0,S)$ state with $(2,0,S)$, $(2,1,S-1)$, and $(2,2,S-2)$ states and g_S has a slight dependence on the vibrational state. Therefore, $\delta f_Z/B$ in the $(v,N,F,M) = (0,0,S+I_{tot},S+I_{tot}) \rightarrow (1,0,S+I_{tot},S+I_{tot})$ transition frequency is significantly larger than those for molecules in the $^1\Sigma$ and $^2\Sigma$ states (> 1 Hz/G) [16].

The $(v,N,F,M_F) = (0,0,I_{tot}+S,\pm(I_{tot}+S)) \rightarrow (1,0,I_{tot}+S,\pm(I_{tot}+S))$ transition frequencies of molecules in the $^1\Sigma$ or $^2\Sigma$ states are highly advantageous for measurements without significant Zeeman shifts. Moreover, the $M_F = \pm F \rightarrow \pm F$ transitions are not resolved when $B < 10^4$ G and the overlapped spectrum has no Zeeman shift. The electronic ground states of alkali earth–alkali diatomic molecules and molecular ions are $^2\Sigma$ and $^1\Sigma$ states, respectively. Since X atoms (X: even isotope of alkali earth atoms) have no nuclear-spin, $I_{tot} = I_Y$ for XY molecules, where I_Y denotes the nuclear-spin of Y atoms (Y = H ($I_H = 1/2$), ^{6}Li ($I_{Li} = 1$), ^{23}Na ($I_{Na} = 3/2$), or ^{87}Rb ($I_{Rb} = 3/2$)). The alkali earth–alkali earth diatomic molecules in the $N = 0$ state have no Zeeman shift.

Table 2 lists the Zeeman coefficients for the $X^2\Sigma(v,N,F,M_F) = (0,0,I_Y+1/2,I_Y+1/2) \rightarrow (1,0,I_Y+1/2,I_Y+1/2)$ transition frequencies of ^{174}Yb^{6}Li, ^{88}Sr^{6}Li, ^{40}Ca^{6}Li, ^{88}Sr^{23}Na, ^{40}Ca^{23}Na, ^{88}Sr^{87}Rb, and ^{40}CaH molecules and $X^1\Sigma(v,N,F,M_F) = (0,0,1/2,1/2) \rightarrow (1,0,1/2,1/2)$ transition frequencies of ^{40}CaH$^+$, ^{88}SrH$^+$, and ^{174}YbH$^+$ molecular ions.

Table 1

$(v;N) = (0,0) \rightarrow (1,0)$ transition frequency f_c; ■ demonstrating the sensitivity of f_c on (m_p/m_e) expressed as $f_c \propto (m_p/m_e)^\blacksquare$; and natural linewidth f_N for ^{174}Yb^{6}Li, ^{88}Sr^{6}Li, ^{40}Ca^{6}Li, ^{88}Sr^{23}Na, ^{40}Ca^{23}Na, ^{88}Sr^{87}Rb, and ^{40}CaH molecules in the X^2■ state and ^{40}CaH$^+$, ^{88}SrH+, and ^{174}YbH$^+$ molecular ions in the X^1■ state.

	f_c (THz)	λ	f_N (Hz)
^{174}Yb^{6}Li[21]	4.17	−0.47	< 0.01
^{88}Sr^{6}Li[23]	5.06	−0.48	< 0.01
^{40}Ca^{6}Li[23]	5.77	−0.47	< 0.01
^{88}Sr^{23}Na	2.29	−0.56	< 0.01
^{40}Ca^{23}Na	2.57	−0.48	< 0.01
^{88}Sr^{87}Rb	0.802	−0.47	< 0.01
^{40}CaH	37.8	−0.49	7.5
^{40}CaH$^+$[19]	43.2	−0.49	2.5
^{88}SrH$^+$[19]	40.3	−0.49	4.0
^{174}YbH$^+$[19]	43.5	−0.48	1.5