Constraints on the variation of physical constants, equivalence principle violation, and a fifth force from atomic experiments

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The aim of this paper is to derive limits on various forms of "new physics" using atomic experimental data. Interactions with dark energy and dark matter fields can lead to space-time variations of fundamental constants, which can be detected through atomic spectroscopy. In this study, we examine the effects of a varying nuclear mass m_N and nuclear radius r_N on two transition ratios: the comparison of the two-photon transition in atomic hydrogen with the hyperfine transition in ^{133}Cs based clocks, and the ratio of optical clock frequencies in Al^+ and Hg^+. The sensitivity of these frequency ratios to changes in m_N and r_N enables us to derive new limits on the variations of the proton mass, quark mass, and the QCD parameter θ. Additionally, we consider the scalar field generated by the Yukawa-type interaction of feebly interacting hypothetical scalar particles with Standard Model particles in the presence of massive bodies such as the Sun and Moon. Using the data from the Al^+/Hg^+, Yb^+/Cs, and $Yb^+(E2)/Yb^+(E3)$ transition frequency ratios, we place constraints on the interaction of the scalar field with photons, nucleons, and electrons for a range of scalar particle masses. We also investigate limits on the Einstein equivalence principle (EEP) violating term (c_{00}) in the Standard Model extension (SME) Lagrangian and the dependence of fundamental constants on gravity.

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I. INTRODUCTION

The existence of life is consistent with a narrow possible range of values for fundamental physical constants, see e.g., the review [1]. This precise fitting of constants for life may be explained by the variation of these constants in space; life emerged in regions of the universe where the fundamental constants have suitable values. Potential evidence for such spatial variation in the fine structure constant α has been observed in quasar absorption spectra [2,3]. Based on this astrophysical data, it is suggested that we are moving in the direction of increasing α, which could lead to a slow drift of fundamental constants observable in laboratory experiments [4,5].

There are a number of models which predict the space-time variation of fundamental physical constants, which may be related to dynamical dark energy models, dark matter models, the variation of the unification scale and string theory models—see, e.g., the review [1]. For example, the space-time variation of fundamental constants may be due to an interaction with a slowly evolving scalar dark energy field or an oscillating dark matter field. The dark matter candidate particles in this class are the pseudoscalar axion (and axion like particles) and the dilatonlike scalar particle [6–8]. If the mass of the cold dark matter is very light ($m_{\rm DM} \ll 1$ eV), it may be considered to be a classical field oscillating harmonically at every particular point in space. For axions, we may write this as

$$a = a_0 \cos(\omega t + \varphi), \qquad \omega \approx m_a, \tag{1}$$

where φ is a (position-dependent) phase and m_a is the mass of the axion. Assuming that axions saturate the entire dark matter density, the amplitude a_0 may be expressed in terms of the local dark matter density $\rho_{\rm DM} \approx 0.4\ {\rm GeV/cm^3}$, see e.g., Ref. [9],

$$a_0 = \frac{\sqrt{2\rho_{\rm DM}}}{m_a}. \tag{2}$$

Similar expressions are used to describe the case of a scalar field dark matter ϕ.

The effects of the interaction between the scalar field and fermions may be presented as the apparent variation of fermion masses. This immediately follows from a comparison of the interaction of a fermion with the scalar field

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$-g_f M_f \phi^n \bar{\psi}\psi$ and the fermion mass term in the Lagrangian $-M_f \bar{\psi}\psi$. Adding these terms gives $M_f' = M_f(1 + g_f \phi^n)$, with $n = 1, 2$. Similarly, the interaction of the scalar field with the electromagnetic field may be interpreted as a variable fine structure constant $\alpha' = \alpha(1 + g_\gamma \phi^n)$. These variations may manifest themselves as a slow drift or oscillations of frequencies of atomic clocks [10–12]. If the interaction is quadratic in ϕ, the scalar field becomes interchangeable with the pseudoscalar (axion) field as ϕ^2 always has positive parity [11]. The corresponding theory has been developed in Ref. [13], in which limits on the axion interaction from atomic spectroscopy experiments were obtained (see also Refs. [14,15]).

In this paper we consider the slow drift of the physical constants, which may be due to an interaction with an evolving nonoscillating dark energy field or the variation of the density of the dark matter field ρ_{DM} in the case where $n = 2$, after averaging over fast oscillations of $\cos^2(\omega t + \varphi) = (1 + \cos 2(\omega t + \varphi))/2$. Such slow variation also appears in other models [1]. In then present paper we do not assume any specific model predicting the variation of the fundamental constants.

The dependence of atomic transition frequencies on α and the quark masses has been calculated in Refs. [16–21]. These calculations and results measuring the time dependence of atomic transition frequencies have previously been used to place improved limits on the interaction strength of the low mass scalar field dark matter ϕ with photons, electrons and quarks by up to 15 orders in magnitude [11,22]. The experimental results have been obtained by measuring the oscillating frequency ratios of electron transitions in a range of systems, including Dy/Cs [23], Rb/Cs [24], Yb/Cs [25], Sr/H/Si cavity [26], Cs/cavity [27], Cs/H [28], $\mathrm{Al}^+/\mathrm{Hg}^+$ [29], and $\mathrm{Yb}^+/\mathrm{Yb}^+/\mathrm{Sr}$ [30,31].

Scalar particles can mediate Yukawa-type interactions between Standard Model particles. A range of experimental methods have been used to constrain these interactions, some of which include equivalence principle tests via torsion pendulum experiments [32,33], lunar laser ranging [34] and atom interferometry [35]. Experimental limits on equivalence principle violation may be expressed in terms of the Eotvos parameter η

$$\eta \equiv 2\frac{|\vec{a}_A - \vec{a}_B|}{|\vec{a}_A + \vec{a}_B|}, \tag{3}$$

where $\vec{a}_A$ and $\vec{a}_B$ are the accelerations of two test bodies A and B (which are composed of different materials) toward a third body C. Most recently, direct fifth force measurements have placed constraints on η. Specifically, the MICROSCOPE mission monitored the difference in acceleration of two freely falling test masses (composed of Pt and Ti) as they orbited the Earth, constraining the Eotvos paramater to be $\eta = (-1 \pm 27) \times 10^{-15}$ at a 2-σ confidence level [36,37]. Constraints on this parameter may be repurposed into individual limits on the couplings of the scalar particle to standard model particles.

Another method of constraining these interactions is via atomic spectroscopy measurements. The scalar field is produced by massive bodies and causes the variation of fundamental constants, leading to a variation in the ratio of transition frequencies. Such calculations and measurements have been performed in Refs. [38,39].

This paper is divided into three parts. In the first part, we seek to obtain limits on the variation of physical constants. The variation of nuclear parameters affects electronic transitions, and the results of the measurements of the variation in the ratio of transition frequencies in Cs/H [28] and the ratio of optical clocks transitions in $\mathrm{Al}^+/\mathrm{Hg}^+$ [29] allow us to place constraints on the variation of the proton mass m_p, the variation of the average quark mass $m_q = (m_u + m_d)/2$ and the variation of the nuclear charge radius r_N and nuclear mass m_N, all of which may be used to place limits on the variation of the QCD parameter θ. The idea that the dependence of the electronic atomic transition frequencies on the nuclear radius (and subsequently on the hadronic parameters above) may be used in the search for dark matter fields in optical transitions was first proposed in Ref. [30], while the effect of the variation of the nuclear radius (and hadronic parameters) in hyperfine transitions was earlier calculated in Ref. [40]. In this paper, we employ a similar method to Ref. [41], in which the sensitivity of the optical clock transitions in Yb^+ to the variation in the nuclear radius was used to place constraints on the variation of the hadron and quark masses, and the QCD parameter θ.

In the second part we consider the beyond-standard-model effects of gravity, such as the Einstein equivalence principle (EEP) violating term (c_{00}) in the Standard Model extension (SME) Lagrangian [42] and the dependence of fundamental constants on the gravitational potential.

In the third part we consider the effects produced by the interaction between hypothetical scalar particles and standard model particles in the presence of massive bodies creating a Yukawa-type potential mediated by scalar particles. Such effects have previously been considered in Ref. [38]. The scalar field, produced by the Sun and the Moon, affects the fine structure constant α and the fermion mass m_f. We perform relevant calculations and use the measurement of the variation in the ratio of clock transition frequencies $\mathrm{Al}^+/\mathrm{Hg}^+$ in Ref. [29], $\mathrm{Yb}^+/\mathrm{Cs}$ in Ref. [43] and $\mathrm{Yb}^+(\mathrm{E2})/\mathrm{Yb}^+(\mathrm{E3})$ in Ref. [31] to determine limits on the interactions between this scalar field and standard model particles for a range of scalar particle masses. These constraints are compared to those obtained from the results of the MICROSCOPE mission [36,37].

Note that when discussing the variation of dimensionful parameters, we must be mindful of the units they are measured in, as these units may also vary. In other words, we should consider the variation of dimensionless parameters which have no dependence on any measurement units.

Nuclear properties depend on the quark mass and Λ_{QCD}. In this work, we keep Λ_{QCD} constant, meaning our calculations are related to the measurement of the variation of a dimensionless parameter $X_q \equiv m_q/\Lambda_{\mathrm{QCD}}$. As a result, we measure the quark mass in units of Λ_{QCD}—see Refs. [19,44,45]. A similar choice of units is assumed for the variation of hadron masses.

In this paper, we assume natural units $\hbar = c = 1$ if $\hbar$ and c are not explicitly presented.

II. ATOMIC TRANSITION FREQUENCY SHIFT DUE TO VARIATION OF NUCLEAR MASS AND RADIUS

The total electronic energy E_{tot} of an atomic state contains the energies associated with the finite mass of the nucleus m_N (mass shift, MS) and the nonzero nuclear charge radius r_N (field shift, FS). Both effects contribute to the isotope shifts of atomic transition frequencies. These are parametrized as [46]

$$E_{\mathrm{MS}} \approx K_{\mathrm{MS}} \frac{1}{m_A} \propto \frac{1}{A} \quad \text{and} \quad E_{\mathrm{FS}} \approx K_{\mathrm{FS}} r_N^2 \propto A^{2/3}, \tag{4}$$

where K_{MS} and K_{FS} are the mass and field shift coefficients respectively and m_A is the mass of an atom with atomic mass number A, which is largely determined by the nuclear mass m_N. The variation of the total electronic energy associated with the nuclear degrees of freedom can be written as [47]

$$\frac{\delta E_{\mathrm{tot}}}{E_{\mathrm{tot}}} = -\frac{E_{\mathrm{MS}}}{E_{\mathrm{tot}}} \frac{\delta m_N}{m_N} + \frac{E_{\mathrm{FS}}}{E_{\mathrm{tot}}} \frac{\delta r_N^2}{r_N^2}. \tag{5}$$

The mass shift term dominates for light nuclei while the field shift term dominates for heavy nuclei. In general, comparing the electronic transition frequencies ν_a and ν_b of two different atomic species, we obtain

$$\frac{\delta(\nu_a/\nu_b)}{(\nu_a/\nu_b)} = (K_1 - K_2) \frac{\delta r_N^2}{r_N^2} + (K_3 - K_4) \frac{\delta m_N}{m_N}, \tag{6}$$

where

$$K_1 = \frac{K_{\mathrm{FS}}^{\nu_a} r_{N,a}^2}{\nu_a}, \tag{7}$$

$$K_2 = \frac{K_{\mathrm{FS}}^{\nu_b} r_{N,b}^2}{\nu_b}, \tag{8}$$

$$K_3 = \frac{K_{\mathrm{MS}}^{\nu_a}}{\nu_a m_{N,a}}, \tag{9}$$

$$K_4 = \frac{K_{\mathrm{MS}}^{\nu_b}}{\nu_b m_{N,b}}. \tag{10}$$

III. LIMITS ON THE LINEAR DRIFT OF THE QCD PARAMETER θ AND PARTICLE MASSES

In this section, we use the above theory along with experimental observations of the drift in atomic transition frequencies to place limits on the variation of quark and hadronic parameters. The approach in this section is similar to that presented in Ref. [41].

Let us first consider the variation of fundamental constants due to the variation of the ratio of transition frequencies in Cs/H. Ref. [28] compared results of the variation in the $|1^2S_{1/2}, F = 1, m_F = \pm 1\rangle \to |2^2S_{1/2}, F' = 1, m'_F = \pm 1\rangle$ two-photon transition in atomic hydrogen to results from clocks based on ^{133}Cs in order to deduce limits on the fractional time variation of the fine structure constant α. The comparison of the transition in H against the ground state hyperfine transition in ^{133}Cs gives the following fractional time variation [28]

$$\frac{1}{(\nu_{\mathrm{Cs}}/\nu_{\mathrm{H}})} \frac{d(\nu_{\mathrm{Cs}}/\nu_{\mathrm{H}})}{dt} = 3.2(6.3) \times 10^{-15}\ \mathrm{yr}^{-1}. \tag{11}$$

The variation of this ratio may be related to the variation of fundamental constants—see Ref. [19]

$$\frac{\delta(\nu_{\mathrm{Cs}}/\nu_{\mathrm{H}})}{(\nu_{\mathrm{Cs}}/\nu_{\mathrm{H}})} = 2.83 \frac{\delta\alpha}{\alpha} + 0.009 \frac{\delta m_q}{m_q} + \frac{\delta(m_e/m_p)}{(m_e/m_p)}. \tag{12}$$

The relative variation of the electron to proton mass ratio can be described as [48]

$$\frac{\delta(m_e/m_p)}{(m_e/m_p)} = -0.037 \frac{\delta m_q}{m_q} - 0.011 \frac{\delta m_s}{m_s} + \frac{\delta m_e}{m_e}, \tag{13}$$

where m_s is the mass of the strange quark. For brevity, we may assume that $\delta m_s/m_s = \delta m_q/m_q$. Combining these expressions gives

$$\frac{\delta(\nu_{\mathrm{Cs}}/\nu_{\mathrm{H}})}{(\nu_{\mathrm{Cs}}/\nu_{\mathrm{H}})} = 2.83 \frac{\delta\alpha}{\alpha} - 0.039 \frac{\delta m_q}{m_q} + \frac{\delta m_e}{m_e}. \tag{14}$$

Therefore, using this expression along with the limit presented in Eq. (11), we may obtain a limit on the variation of the quark mass m_q assuming that there is no variation of α and m_e

$$\frac{1}{m_q} \frac{dm_q}{dt} = -8.2(16) \times 10^{-14}\ \mathrm{yr}^{-1}. \tag{15}$$

When considering the variation of the QCD parameter θ, it is convenient to consider the problem at the hadron level, rather than the quark level. Using the calculations presented in Ref. [19] and the limit (11) we obtain

$$\frac{\delta m_\pi}{m_\pi} = -4.1(8.0) \times 10^{-14}\ \mathrm{yr}^{-1}. \tag{16}$$

Using this value, along with the following result from Ref. [13]

$$\frac{\delta m_p}{m_p} = 0.13\frac{\delta m_\pi}{m_\pi}, \tag{17}$$

we may subsequently place limits on the drift of the proton mass due to the drift of the pion mass

$$\frac{1}{m_p}\frac{dm_p}{dt} = -5.3(10) \times 10^{-15}\ \mathrm{yr}^{-1}. \tag{18}$$

Finally, we may use the relation between the variation of the pion mass and the QCD parameter θ, presented in Ref. [49], to place limits on the linear drift of θ. The pion mass has dependence on θ, and the shift of the pion mass due to a small θ relative to the pion mass for $\theta = 0$ is given by [49]

$$\frac{\delta m_\pi}{m_\pi} = -0.05\theta^2 \tag{19}$$

Thus, substituting the value for the drift of the pion mass from Eq. (16) yields

$$\frac{d\theta^2}{dt} = 8.2(16) \times 10^{-13}\ \mathrm{yr}^{-1}. \tag{20}$$

We may also perform a similar calculation relevant to measurements of a different ratio of transition frequencies. The drift of the ratio of the optical clock transition frequencies of aluminium and mercury has been measured in Ref. [29]. This drift was used to place limits on the temporal variation of the fine structure constant. Using a similar method to that above, this result may be repurposed to extract limits on the variation of the nuclear radius and hadronic parameters. The rate of change in the ratio of the transition frequencies of ${}^1S_0 \to {}^3P_0$ transitions in Al^+ and ${}^2S_{1/2} \to {}^2D_{5/2}$ transitions in Hg^+ was found to be [29]

$$\frac{1}{\nu_{\mathrm{Al}^+}/\nu_{\mathrm{Hg}^+}}\frac{d(\nu_{\mathrm{Al}^+}/\nu_{\mathrm{Hg}^+})}{dt} = -5.3(7.9) \times 10^{-17}\ \mathrm{yr}^{-1}. \tag{21}$$

Once again we note the fact that mass shift dominates the isotope shift effects in light elements, while field (volume) shift dominates in heavy elements. As such, the isotopic shift in Al^+ is dominated by the mass shift component, while the isotopic shift in Hg^+ is dominated by the field shift component.

The mass shift is divided into two components: the normal mass shift (NMS) and the specific mass shift (SMS). The NMS results from a change in the reduced electron mass, and its parameter is easily calculated from the transition frequency using the nonrelativistic virial theorem stating that in the case of the Coulomb interaction, the electron's total energy change corresponds to the change in its kinetic energy, with a negative sign

$$K^{\nu}_{\mathrm{NMS}} = -\frac{\nu}{1822.888}\ \mathrm{amu}, \tag{22}$$

where the factor in the denominator refers to the ratio of the atomic mass unit to the electron mass. Using the transition frequency from the experimental observation of the ${}^1S_0 \to {}^3P_0$ transition in ${}^{27}\mathrm{Al}^+$ of Ref. [50], we calculate the normal mass shift factor in Al^+ to be $K^{\nu_{\mathrm{Al}^+}}_{\mathrm{NMS}} = -615$ GHz amu. Using the calculated ratio of the specific to normal mass shifts from Ref. [51], we yield a total mass shift parameter of $K^{\nu_{\mathrm{Al}^+}}_{\mathrm{MS}} = -1530$ GHz amu.

Now we must consider the effects from the field shift in Hg^+. We have performed relativistic many-body calculations of the shift of atomic transition frequencies due to the variation of the nuclear radius in Hg^+. The wave functions and energies in the zeroth-order approximation have been calculated using the Dirac-Hartree-Fock method, including Breit corrections. The correlation corrections were calculated using the CIPT method (configuration interaction with perturbation theory [52]), which allows us to deal with open electronic shells. This approach is similar to that described in Ref. [53]. We have found the field shift parameter for the ${}^2S_{1/2} \to {}^2D_{5/2}$ transition in Hg^+ to be 85.5 GHz/fm^2. Similar calculations are performed to find isotope shifts in atomic transitions. Our value for the isotope shift in the ${}^2S_{1/2} \to {}^2D_{5/2}$ transition of Hg^+ is in agreement with the calculations of the isotope shift in Hg^+ presented in Ref. [54]. Thus, using the MS and FS parameters along with Eq. (6), we obtain

$$\frac{\delta(\nu_{\mathrm{Al}^+}/\nu_{\mathrm{Hg}^+})}{\nu_{\mathrm{Al}^+}/\nu_{\mathrm{Hg}^+}} = -\left[\frac{K^{\nu_{\mathrm{Hg}^+}}_{\mathrm{FS}}\, r^2_{N,\mathrm{Hg}^+}}{\nu_{\mathrm{Hg}^+}}\frac{\delta r_0^2}{r_0^2} + \frac{K^{\nu_{\mathrm{Al}^+}}_{\mathrm{MS}}}{\nu_{\mathrm{Al}^+} m_{N,\mathrm{Al}^+}}\frac{\delta m_N}{m_N}\right]. \tag{23}$$

Here, we have used the fact that the nuclear radii in all nuclei may be quite accurately related to the internucleon distance r_0 by the universal formula $r_N = A^{1/3} r_0$. This implies that these quantities have equivalent fractional variations. Presenting limits on the variation of r_0 is more useful as it allows one to compare the results of measurements in different nuclei. Noting that $r^2_{N,\mathrm{Hg}^+} = 5.4474$ fm in ${}^{199}\mathrm{Hg}$ [55] and $m_{N,\mathrm{Al}^+} \approx 27 m_p$ in ${}^{27}\mathrm{Al}$, we yield the following limits

$$\frac{1}{r_0}\frac{dr_0}{dt} = -1.1(1.7) \times 10^{-14}\ \mathrm{yr}^{-1}, \tag{24}$$

$$\frac{1}{m_p}\frac{dm_p}{dt} = 1.0(1.6) \times 10^{-12}\ \mathrm{yr}^{-1}. \tag{25}$$

Considering these effects as separate sources of the variation allows us to derive independent limits on the variation of quark and hadronic parameters.

Firstly, using a similar method to the one presented in Ref. [41], we consider the effects arising from a variation in the internucleon distance r_0. Calculations of the dependence of nuclear energy levels and nuclear radii on fundamental constants were performed in Refs. [44,45,56]. Specifically, in Table VI of Ref. [45], the sensitivity coefficients of nuclear radii to the variation of hadron masses for several light nuclei have been presented. These results may be extended to all nuclei due to the relation $r_N = A^{1/3} r_0$. This relation follows from the constancy of nuclear density and reasonably describes the nuclear radius for nuclei with mass number $A > 2$. Therefore, by calculating the variation in r_0, a fundamental parameter, we can generalize the results for light nuclei to include all nuclei. The sensitivity coefficients are defined by the relation

$$\frac{\delta r_0}{r_0} = \sum_h K_h \frac{\delta m_h}{m_h}. \tag{26}$$

The sum over hadrons in Refs. [45,56] includes contributions from π, nucleon, Δ and vector mesons (these hadron masses are parameters of the kinetic energy and nucleon interaction operators used in Refs. [45,56]). The sensitivity to the pion mass is given by the coefficient $K_\pi = 1.8$ and the sensitivity to the nucleon mass is given by $K_n = -4.8$. We neglect contributions from Δ and vector mesons as they are of a similar magnitude with opposing sign, meaning their resulting contribution is small and unstable.

Note that the estimate in Eq. (26) is model dependent and may have an error O(1).

Subsequently, the variation of hadron masses may be related to variation of the quark mass, see, e.g., Ref. [57]:

$$\frac{\delta m_h}{m_h} = K_{h,q} \frac{\delta m_q}{m_q}, \tag{27}$$

where $m_q = (m_u + m_d)/2$ corresponds to the average light quark mass. The sensitivity coefficient for the pion mass is an order of magnitude bigger than that for other hadrons since the pion mass vanishes for zero quark mass ($m_\pi \propto m_q^{1/2}$) while other hadron masses remain finite. Indeed, according to Refs. [58,59] $K_{\pi,q} = 0.498$ for the pion, while $K_{n,q} = 0.06$ for nucleons. The sensitivity coefficients to the quark mass have been calculated for light nuclei in Ref. [45]. The average value is given by

$$\frac{\delta r_0}{r_0} = 0.3 \frac{\delta m_q}{m_q}. \tag{28}$$

We note that here there are partial cancellations of different contributions, so the sensitivity is smaller than that following from pion mass alone. References [45,56] have also presented calculations of the dependence of the nuclear energies and radii on the variation of the fine structure constant α. Applying Eq. (28) to the limit on the variation of the internucleon distance presented in (24), we determine limits on the variation of the quark mass to be

$$\frac{1}{m_q} \frac{dm_q}{dt} = -3.7(5.7) \times 10^{-14}\ \mathrm{yr}^{-1}. \tag{29}$$

Once again, in order to place limits on the variation of the proton mass and the QCD parameter θ, it is convenient to consider the problem at the hadron level, rather than the quark level. In Ref. [45], the sensitivity of the nuclear radius to the masses of the pion, nucleon, vector meson and delta has been calculated. In the following estimate, we do not include contributions from the vector meson and delta as their contributions are smaller. These contributions also have opposing signs, meaning they partially cancel each other out making their contribution less reliable. The variation of the nuclear radius may be written in terms of the pion and nucleon mass as

$$\frac{\delta r_0}{r_0} = 1.8 \frac{\delta m_\pi}{m_\pi} - 4.8 \frac{\delta m_n}{m_n} = 1.2 \frac{\delta m_\pi}{m_\pi}, \tag{30}$$

where in the last equality we have applied Eq. (17). Thus, once again applying Eq. (24), we obtain a limit on the drift of the pion mass

$$\frac{1}{m_\pi} \frac{dm_\pi}{dt} = -9.2(14) \times 10^{-15}\ \mathrm{yr}^{-1}. \tag{31}$$

Finally, we apply Eq. (19) in order to place constraints on the linear drift of the QCD parameter θ

$$\frac{d\theta^2}{dt} = 1.8(2.8) \times 10^{-13}\ \mathrm{yr}^{-1}. \tag{32}$$

These limits are approximately 44 times weaker than the limits imposed from the sensitivity of the optical clock transitions in Yb^+ obtained in Ref. [41]. This difference corresponds exactly to the difference in the accuracy of the experimental measurements of the variation in clock frequency ratios, showing that these systems have equivalent sensitivities to the variation of the internucleon distance. We however note that the limits from the Al^+/Hg^+ clock ratio are ∼2–4 times stronger than those calculated for the Cs/H system, despite the fact that the accuracy of the Al^+/Hg^+ measurements is ∼60 times better. This implies that the sensitivity of the Cs/H ratio to changes in the hadron constants is higher than that of the Al^+/Hg^+ ratio.

Let us now consider the effects arising from a variation of the nuclear mass m_N. The nuclear mass may be related to the proton mass m_p by the following relation $m_N \approx A m_p$.

As such, these parameters have equivalent fractional variations, meaning the limit from Eq. (25) applies. Using this result, along with the relation between the pion and the proton mass presented in Eq. (17), we may place limits on the drift of the pion mass

$$\frac{1}{m_\pi}\frac{dm_\pi}{dt} = 8.1(12) \times 10^{-12}\ \mathrm{yr}^{-1}. \quad (33)$$

Using the calculations presented in Ref. [19], we may use this result to place limits on the variation of the quark mass m_q

$$\frac{1}{m_q}\frac{dm_q}{dt} = 1.6(2.4) \times 10^{-11}\ \mathrm{yr}^{-1}. \quad (34)$$

We also use the relation between the variation of the pion mass and the QCD parameter θ presented in Eq. (19) to place constraints on the linear drift of θ^2

$$\frac{d\theta^2}{dt} = -1.6(2.4) \times 10^{-10}\ \mathrm{yr}^{-1}. \quad (35)$$

As expected, the limits obtained upon considering the variation in the ratio of frequencies (21) as being due to the drift of the nuclear (and hence nucleon) mass are weaker than the limits from the variation of the internucleon distance.

IV. GRAVITY RELATED VARIATION OF FUNDAMENTAL CONSTANTS

In some theoretical models, atomic transition frequencies and fundamental constants may depend on the gravitational potential. In Ref. [60] limits on the gravity related variation of fundamental constants are derived from measurements of the drift of atomic clock frequency ratios. In a similar way, we may obtain limits on the gravity related variation of the nuclear radius and nuclear mass using the measurement of the variation in the ratio of transition frequencies in Al^+/Hg^+ from Refs. [29,61]. We will also calculate limits on gravity related variation of these quantities for other systems of interest, Yb^+/Cs [43] and Yb^+/Yb^+ [31].

Noting the dependence of the frequency shift on the nuclear mass and radius from Eq. (6), we introduce the parameters $\kappa_{r,N}$ as follows: (see Ref. [60])

$$\frac{\delta r_N^2}{r_N^2} = \kappa_r \delta\left(\frac{GM}{rc^2}\right), \quad (36)$$

$$\frac{\delta m_N}{m_N} = \kappa_N \delta\left(\frac{GM}{rc^2}\right). \quad (37)$$

It is instructive to link these parameters κ_r and κ_N to the Einstein equivalence principle (EEP) violating term in the Standard Model extension Hamiltonian [42]. This term may be presented as a correction to the kinetic energy which in nonrelativistic form is equal to (see, e.g., [62])

$$\delta H = \frac{2}{3} c_{00} \frac{U}{c^2} \frac{p^2}{2m_e}, \quad (38)$$

where c_{00} is a parameter of the Standard Model extension (SME) Lagrangian [42], U is the gravitational potential, p is the electron momentum operator and m_e is the electron mass. Limits on c_{00} have been found by monitoring the drift of atomic clock frequencies for different transitions (see, e.g., [61,62]). Reference [61] subsequently used the limits on this parameter to determine a limit on the gravity-related variation of the fine structure constant κ_α

$$\frac{\delta\alpha}{\alpha} = \kappa_\alpha \delta\left(\frac{GM}{c^2 r}\right), \quad (39)$$

where in the case of a relative change of two transition frequencies in different atomic species $\delta\omega_a/\omega_a = K_{\alpha a}(\delta\alpha/\alpha)$ and $\delta\omega_b/\omega_b = K_{\alpha b}(\delta\alpha/\alpha)$, the parameter κ_α is equal to

$$\kappa_\alpha = \frac{2}{3} \frac{R_b - R_a}{K_{\alpha a} - K_{\alpha b}} c_{00}, \quad (40)$$

where R_a and R_b represent the relativistic factors of atoms a and b respectively, which describe the deviation from the expectation value of the kinetic energy of a relativistic atomic electron from the value given by the nonrelativistic virial theorem [61]

$$R = -\frac{\Delta E_i - \Delta E_j}{E_i - E_j}, \quad (41)$$

where ΔE_i is the energy shift of the state i due to the relativistic kinetic energy operator. In the nonrelativistic limit, $R = 1$. Thus, we employ a similar method and derive expressions for the parameters κ_r and κ_N describing the variation of the nuclear radius and nuclear mass

$$\kappa_r = \frac{2}{3} \frac{R_b - R_a}{K_1 - K_2} c_{00}, \quad (42)$$

$$\kappa_N = \frac{2}{3} \frac{R_b - R_a}{K_3 - K_4} c_{00}, \quad (43)$$

where $K_{1,2,3,4}$ are defined in Eq. (6). The value of R has been calculated for a number of clock transitions in Ref. [61], see Table I. Ref. [61] also calculated a limit on the value of the SME parameter c_{00} due to the change in the Sun's gravitational potential for the ratio of these transition frequencies ν_{Al^+}/ν_{Hg^+}. As such, we may substitute all known quantities into Eqs. (42), (43) and determine a limit on the gravity related variation of the

TABLE I. Relativistic factors (R) for optical clock transitions in atoms and ions [61].

Atom/Ion	Ground state	Clock state	$\hbar\omega$ [cm^{-1}]	R
Al^+	$3s^2\ ^1S_0$	$3s3p\ ^3P_0^o$	37393	1.00
Hg^+	$5d^{10}6s\ ^2S_{1/2}$	$5d^96s^2\ ^2D_{5/2}$	35515	0.2
Yb^+	$6s\ ^2S_{1/2}$	$5d\ ^2D_{3/2}$	22961	1.48
Yb^+	$6s\ ^2S_{1/2}$	$4f\ ^2F_{7/2}$	21419	−1.9

nuclear radius and nuclear mass. We also use the limits on the coupling to gravity of the fine structure constant α, presented in Refs. [43] (Yb^+/Cs) and [31] (Yb^+/Yb^+) in order to place improved limits on gravity's coupling to the nuclear radius κ_r. In performing these calculations, we make use of the following result for the difference in field shift parameters in the electric octupole (E3) and electric quadrupole (E2) transitions in Yb^+: $K_1 - K_2 = 2.4 \times 10^{-3}$ [30], and the sensitivity of variations in this ratio to the fine structure constant α, $k_\alpha(E3) - k_\alpha(E2) = -6.95$ [18]. The results are presented in Table II.

V. CONSTRAINTS ON THE YUKAWA-TYPE INTERACTION MEDIATED BY THE SCALAR FIELD PRODUCED BY MASSIVE BODIES

In this section, we consider the Yukawa-type interaction mediated by the scalar field produced by massive bodies. Let us first provide an introduction to the phenomenology of this scalar field, following Ref. [38]. A scalar field ϕ interacts with the standard model sector via the Yukawa-type Lagrangian

$$\mathcal{L}_{\text{int}} = -\sum_f \frac{\phi}{\Lambda_f} m_f \bar{f} f + \frac{\phi}{\Lambda_\gamma} \frac{F_{\mu\nu}F^{\mu\nu}}{4}. \tag{44}$$

Here, the first term represents the coupling to fermion fields f, with mass m_f and $\bar{f} = f^\dagger \gamma_0$, while the second term represents the coupling to the photon field. Λ_f and Λ_γ are new-physics energy scales which determine the strength of these couplings. Adding these interaction terms to the relevant terms in the standard model Lagrangian

$$\mathcal{L} \supset -\sum_f m_f \bar{f} f - \frac{F_{\mu\nu}F^{\mu\nu}}{4\alpha}, \tag{45}$$

we observe that we may present the effects of the interaction terms in the form of a variable fermion mass m_f and electromagnetic fine-structure constant α (see, e.g., [12])

$$m_f \to m_f \left(1 + \frac{\phi}{\Lambda_f}\right), \tag{46}$$

$$\alpha \to \frac{\alpha}{1 - \phi/\Lambda_\gamma} \approx \alpha \left(1 + \frac{\phi}{\Lambda_\gamma}\right). \tag{47}$$

Adding the kinetic term to the interaction Lagrangian (44) gives the following equations of motion for the field ϕ

$$(\partial_\mu \partial_\mu + m_\phi^2)\phi = -\sum_f \frac{m_f}{\Lambda_f} m_f \bar{f} f + \frac{1}{\Lambda_\gamma} \frac{F_{\mu\nu}F^{\mu\nu}}{4}, \tag{48}$$

where m_ϕ is the mass of the scalar particle. This implies that in the presence of the interaction (44), the standard model fermion and photon fields act as sources of the scalar field. Massive bodies such as stars or galaxies, which are composed of atoms, may act as these sources, producing a scalar field mediating Yukawa-type interactions. This field may produce a local variation of fundamental constants in the presence of a massive body with a varying distance to the laboratory. As such, we may investigate the influence of the scalar particle ϕ on atomic spectroscopy experiments, see Ref. [38]. In the following subsections we place constraints on the scalar field's interaction with the Standard Model particles. Specifically, we consider the effects produced by a variation in the scalar field due to both the semi-annual variation in the Sun-Earth distance, and the approximately semi-monthly variation in the Moon-Earth distance. These constraints are compared to those obtained from tests of the equivalence principle by the MICROSCOPE mission [36,37].

A. Effects produced by the variation in the Sun-Earth distance

Similar to the gravitational potential, the scalar Yukawa potential depends on the distance between Sun and Earth.

TABLE II. Limits on the Standard Model Extension parameter c_{00} and the parameters κ_α, κ_r, κ_n and κ_e describing the dependence of the fine structure constant α, the internucleon distance r_0 (derived from the nuclear charge radius r_N), the nucleon mass and the electron mass on the gravitational potential.

System	Source	c_{00}	κ_α	κ_r	κ_n	κ_e
Al^+/Hg^+ [29]	Sun	$-3.0(5.7) \times 10^{-7}$ [61]	$5.3(10) \times 10^{-8}$ [61]	$-6.7(13) \times 10^{-5}$	$-3.1(6.0) \times 10^{-3}$	⋯
	Moon	$-6.0(12) \times 10^{-3}$	$1.1(2.1) \times 10^{-3}$	−0.67 (1.3)	−63 (120)	⋯
Yb^+/Cs [43]	Sun	$4.2(3.3) \times 10^{-8}$	$14(11) \times 10^{-9}$ [43]	$4.0(3.1) \times 10^{-5}$	$7(45) \times 10^{-8}$ [43]	$-7(45) \times 10^{-8}$ [43]
Yb^+/Yb^+ [31]	Sun	$-7.4(9.3) \times 10^{-9}$	$-2.4(3.0) \times 10^{-9}$ [31]	$7.0(8.7) \times 10^{-6}$	$2.1(2.9) \times 10^{-4}$	⋯

Assuming the Sun's elemental composition to be 75% ^{1}H and 25% ^{4}He by mass, the resultant scalar field may be expressed as [38]

$$\phi_{\mathrm{Sun}} = -N_s m_n \left[\frac{0.15}{\Lambda_{n'}} + 1.1\left\{\frac{1}{\Lambda_p} + \frac{5\times 10^{-4}}{\Lambda_e}\right\} + \frac{8\times 10^{-4}}{\Lambda_\gamma}\right]\frac{e^{-m_\phi r}}{4\pi r} = -N_s \beta_s \frac{e^{-m_\phi r}}{4\pi r}, \tag{49}$$

where $m_n = (m_p + m_{n'})/2 = 0.94$ GeV is the average nucleon mass and N_s is the number of atoms inside the Sun. The number of nucleons inside the Sun may be determined as mass of the Sun divided by the proton mass, $N_s = M_s/m_p = 1.99\times 10^{30}\ \mathrm{kg}/1.67\times 10^{-27}\ \mathrm{kg} = 1.19\times 10^{57}$. The ratio of the number of neutrons and protons depends on the composition of the Sun, which is mainly composed of hydrogen and helium. According to Eq. (49), the average atom in the Sun contains 1.1 protons and 0.15 neutrons, i.e., 1.25 nucleons. This implies that the number of atoms in the Sun is $N_s = 0.95\times 10^{57}$. In obtaining limits on the nucleon constant Λ_n we consider the sum of the proton and neutron contributions assuming $\Lambda_{n'} = \Lambda_p$. In this case, the limits on Λ_n have no dependence on the composition of the Sun. The Earth's orbit is elliptical, with the Earth-Sun distance changing between 1.52×10^8 km and 1.47×10^8 km.

Using new and existing calculations of the variation of fundamental constants due to changes in the Yukawa potential (see the Appendix), we place constraints on the Yukawa-type interactions of the scalar field from the Sun with photons, nucleons and electrons for a range of scalar particle masses, using a similar method to Ref. [38]. Constraints on the parameters Λ_γ, Λ_n, and Λ_e are presented in Fig. 1 and summarized in Table III.

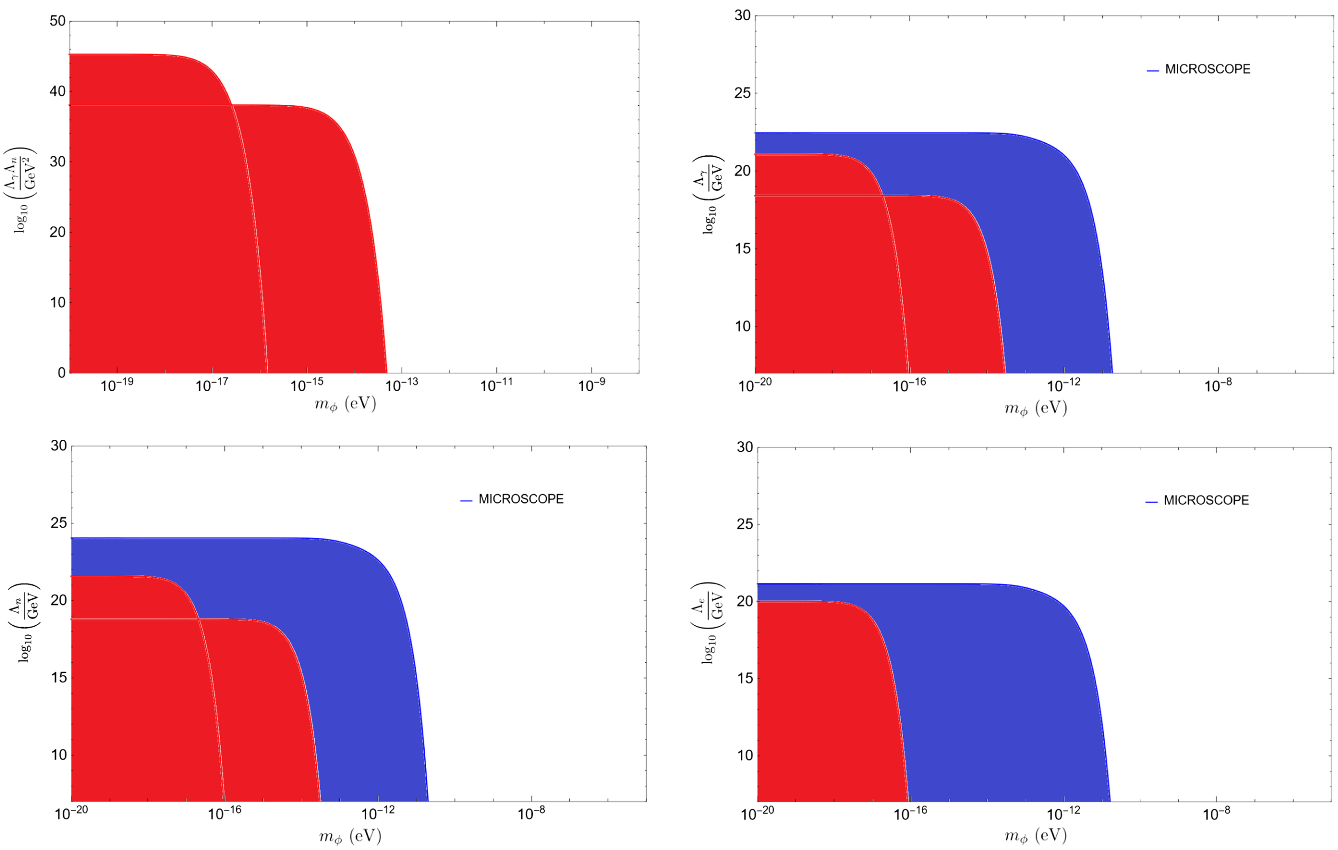

FIG. 1. Limits on the constants of the Yukawa-type interaction Eq. (44) of the scalar field ϕ_s with photons (Λ_γ), nucleons (Λ_n) and electrons (Λ_e). In obtaining limits on the nucleon constant Λ_n we sum the contributions from the proton and neutron coupling constants, assuming $\Lambda_{n'} = \Lambda_p$. The upper curve depicts the region constrained by the potential of the Sun, while the lower and longer curve depicts the region constrained by the Moon's potential. There is no data for the effect of the Moon's gravitational potential on the electron interaction constant (lower right tile). The region in blue shows the parameter space which is excluded by the MICROSCOPE experiment [36,37].

TABLE III. Lower limits on the constants of the Yukawa-type interaction Eq. (44) of the scalar field ϕ_s with photons (Λ_γ), nucleons (Λ_n) and electrons (Λ_e) in the interval of small scalar field masses determined by the condition that the scalar particle's Compton wave length $\hbar/m_\phi c$ is bigger than the distance to the source, $m_\phi < 1.0\times10^{-18}$ eV for the Sun-Earth distance and $m_\phi < 0.5\times10^{-15}$ eV for the Moon-Earth distance. In obtaining limits on the nucleon constant Λ_n we sum the contributions from the proton and neutron coupling constants, assuming $\Lambda_{n'} = \Lambda_p$. The full exclusion plots are presented in Fig. 1.

System	Source/Attractor	Λ_γ/β (GeV^2)	Λ_γ (GeV)	Λ_n (GeV)	Λ_e (GeV)
Al^+/Hg^+ [29]	Sun	7×10^{43}	2×10^{20}	1×10^{21}	⋯
	Moon	1×10^{38}	2×10^{18}	6×10^{18}	⋯
Yb^+/Cs [43]	Sun	4×10^{44}	6×10^{20}	5×10^{21}	1×10^{20}
Yb^+/Yb^+ [31]	Sun	2×10^{45}	1×10^{21}	4×10^{21}	⋯

B. Effects produced by the variation in the Moon-Earth distance

We may also extract limits on these parameters by considering the varying Yukawa potential of the Moon's orbit around Earth. Doing so allows one to investigate the coupling at larger values of the scalar particle mass m_ϕ. The average atom in the Moon contains 12 protons and 12 neutrons. Following Ref. [38] we obtain the Moon's scalar field to be

$$\phi_{\rm Moon} = -N_m m_n\left[\frac{12}{\Lambda_{n'}} + 12\left\{\frac{1}{\Lambda_p} + \frac{5\times10^{-4}}{\Lambda_e}\right\} + \frac{0.03}{\Lambda_\gamma}\right]\frac{e^{-m_\phi r}}{4\pi r} = -N_m\beta_m\frac{e^{-m_\phi r}}{4\pi r}. \quad (50)$$

The number of nucleons in the Moon may be determined as the mass of the Moon divided by the proton mass, $N_m = M_m/m_p = 7.34\times10^{22}$ kg$/1.67\times10^{-27}$ kg $= 4.40\times10^{49}$. The average atom in the Moon contains 24 nucleons. This implies that the number of atoms in the Moon to be $N_m = 1.8\times10^{48}$. In obtaining limits on the nucleon constant Λ_n we once again consider the sum of the proton and neutron contributions assuming $\Lambda_{n'} = \Lambda_p$. In this case, the limits on Λ_n have no dependence on the composition of the Moon.

The average Earth-Moon distance is 3.84×10^5 km, centre to centre, and this value varies between 3.69×10^5 km and 3.99×10^5 km with a period of approximately 27.3 days. Furthermore, due to the relatively large diameter of the Earth ($\sim1.27\times10^4$ km), we note that there is also a daily variation in the distance between the Moon and the laboratory. Despite this, the contributions from the monthly variations are more significant, and as such we consider a variation of $\sim3.0\times10^4$ km in the Earth-Moon distance, which corresponds to the minimal seasonal variation (see Ref. [38]).

Thus, using a similar method to that of the previous subsection, we may determine limits on the Yukawa-type interaction between the varying scalar field and photons/nucleons for a range of scalar particle masses. Constraints on these parameters are presented in Fig. 1, and summarized in Table III.

These limits may be compared to the limits from the atomic spectroscopy measurements presented in Refs. [38,39]. Our limits based on the Sun and Moon data are significantly stronger than that of those presented in Ref. [38]. Despite this, their results also contain limits on the scalar field produced by a 300 kg lead mass on a distance of 1 m. This area is sensitive to much bigger scalar masses and is not covered by our results. Further, our constraint on the photon coupling constant Λ_γ is stronger than the corresponding limit obtained from existing atomic clock experiments presented in Fig. 5 of Ref. [39]. This is due to the fact that the constraint presented in this reference uses the results obtained from Yb^+/Cs experiments [43] only, while our constraints are based on the more sensitive Yb^+/Yb^+ experiments [31].

Constraints yielded from the results of existing atomic clock experiments do not exceed the sensitivity provided by direct fifth force searches such as the MICROSCOPE mission [36,37] (see Fig. 1). In order for them to do so, we require an improvement of ~3 orders of magnitude in the fractional frequency uncertainty in optical clocks located on Earth, something which is predicted to occur over the next few decades [63]. Alternatively, there exist proposals for experiments with clocks based on nuclear transitions [64], which are very sensitive to the variation of fundamental constants [58], and optical clock experiments conducted in space [65,66]. Constraints yielded from such experiments are predicted to match/exceed the sensitivity of direct fifth force searches [39].

TABLE IV. Summary of the obtained constraints on the variation of fundamental constants due to changes in the distance to the Sun (interpreted in Refs. [29,31,43] as the effect of the variation in the Sun's gravitational potential).

System	$\delta\alpha/\alpha$	$\delta m_n/m_n$	$\delta m_e/m_e$
Al^+/Hg^+ [29]	$0.17(0.33)\times10^{-16}$	$1.2(2.3)\times10^{-15}$	...
Yb^+/Cs [43]	$4.6(3.6)\times10^{-18}$	$2.3(15)\times10^{-17}$	$-2.3(15)\times10^{-17}$
Yb^+/Yb^+ [31]	$-7.9(9.9)\times10^{-19}$	$1.3(1.6)\times10^{-16}$	...

C. Limits on combinations of coupling constants

Spectroscopy measurements may also be used to probe different combinations of the couplings constants $\Lambda_{\gamma,p,n,e}$ [38], some of which may not be otherwise probed using anomalous-force measurements. Such results may be useful in the case when the variation of the ratio of optical clock transition frequencies and the source-dependent functions $\beta_{s,m}$ are dominated by different terms. Thus we may use the results of the previous two subsections to provide constraints on the combination of parameters $\Lambda_\gamma\Lambda_n$, see Figure 1. If the scalar particle's Compton wave length $\hbar/m_\phi c$ is bigger than the distance to the source, we determine the following limits for the combination of parameters $\beta_{s,n}/\Lambda_\gamma$

$$\frac{\Lambda_\gamma}{\beta_s}\gtrsim 2\times10^{45}\ \mathrm{GeV}^2, \tag{51}$$

$$\frac{\Lambda_\gamma}{\beta_m}\gtrsim 1\times10^{38}\ \mathrm{GeV}^2. \tag{52}$$

Limits on the variation of α, the nucleon mass and the electron mass due to the variation of the Sun's distance to Earth are presented in Table IV. The details are presented in the Appendix.

VI. SUMMARY

Atomic spectroscopy measurements are used to search for the potential space-time variation of physical constants. In particular, we relate the proton mass m_p and the quark mass m_q variation to measurements in the variation of two frequency ratios: the comparison of the two-photon transition in atomic Hydrogen to the results from clocks based on ^{133}Cs in Ref. [28], and the variation in the ratio of the two optical clock frequencies in Al^+ and Hg^+ in Ref. [29]. We used this data to place new limits on the variation of the proton mass m_p, as well as independent limits on the variation of the quark mass m_q, both of which may be used to place limits on the variation of the QCD parameter θ.

In the second part of this paper we considered the beyond-standard-model effects of gravity, such as the Einstein equivalence principle (EEP) violating term (c_{00}) in the Standard Model extension (SME) Lagrangian [42] and the dependence of fundamental constants on the gravitational potential, based on the measurements of the dependence of the ratio of atomic transition frequencies Al^+/Hg^+ [29,61], Yb^+/Cs [43], and Yb^+/Yb^+ [31] to the Sun-Earth distance.

In the third part of this paper we considered the scalar field produced by massive bodies. We determine limits on the interactions of the scalar particle with photons, nucleons and electrons for a wide range of scalar particle masses, basing on the measurements of dependence of the ratio of atomic transition frequencies Al^+/Hg^+ [29,61], Yb^+/Cs [43], and Yb^+/Yb^+ [31] on the Sun-Earth and Moon-Earth distances. If the scalar particle's Compton wave length $\hbar/m_\phi c$ is bigger than the distance to the source, we place the following limits on the coupling constants of the interaction of the scalar field with photons, nucleons, and electrons: $\Lambda_\gamma\gtrsim 1\times10^{21}$ GeV, $\Lambda_n\gtrsim 5\times10^{21}$ GeV, and $\Lambda_e\gtrsim 1\times10^{20}$ GeV.

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APPENDIX: COUPLING OF FUNDAMENTAL CONSTANTS TO CHANGES IN THE YUKAWA POTENTIAL FROM THE SUN/MOON

In this appendix we detail the calculations of the variation of fundamental constants due to changes in the Yukawa potential. We will use experimental data on the variation of atomic transition frequencies as functions of Sun-Earth and Moon-Earth distances motivated by the search for dependence of the fundamental constants on the gravitational potential. Constraints on this dependence were obtained for all the systems of interest: Al^+/Hg^+, Yb^+/Cs, and Yb^+/Yb^+. The summary is presented in Table IV.

Let us start with the variation of the fine structure constant α. Equation (47) implies the following relation

$$\frac{\delta\alpha}{\alpha}=\frac{\delta\phi}{\Lambda_\gamma}, \tag{A1}$$

At $m_\phi\to0$ we have from (49) or (50)

$$\delta\phi=-\frac{\beta N_{S,M}}{4\pi}\delta\left(\frac{1}{r}\right)_{S,M}\equiv\beta D_{S,M}. \tag{A2}$$

Here $N_{S,M}$ is number of atoms in the Sun or the Moon ($N_S \approx 0.95 \times 10^{57}$ and $N_M \approx 1.8 \times 10^{48}$) and $\delta(1/r)_{S,M}$ is half-yearly variation of the inverted Sun-Earth or Moon-Earth distance. Then

$$|\Lambda_\gamma/\beta| = \left|\frac{D_{S,M}}{\delta\alpha/\alpha}\right|, \tag{A3}$$

and

$$\Lambda_\gamma = \sqrt{\left|\frac{m_n a_\gamma D_{S,M}}{\delta\alpha/\alpha}\right|}, \tag{A4}$$

where $a_\gamma = 8 \times 10^4$ for the Sun, and $a_\gamma = 0.03$ for the Moon [see (49) and (50)].

Now let us consider the variation of the fermionic masses, of which we consider the nucleon and electron mass, m_n and m_e respectively. From Eq. (30), we have $\frac{\delta r_0}{r_0} = 1.2 \frac{\delta m_\pi}{m_\pi}$, where r_0 is the internucleon distance and m_π is the pion mass. From Eq. (17) we see that $\frac{\delta m_p}{m_p} = 0.13 \frac{\delta m_\pi}{m_\pi}$, where m_p is the proton mass. Upon substitution into Eq. (30), we yield

$$\frac{\delta m_n}{m_n} = 0.11 \frac{\delta r_0}{r_0}. \tag{A5}$$

This relation indicates that the nucleon mass is less sensitive to variations in the pion mass (or quark mass) compared to the internucleon distance r_0. This is consistent with the fact that the quark contribution to nucleon mass is relatively small, while r_0 is very sensitive to the pion exchange potential. Thus, we yield

$$\frac{\delta m_n}{m_n} = \frac{\delta(\nu_a/\nu_b)}{(\nu_a/\nu_b)} \left(\frac{1}{18.5(K_1 - K_2) + (K_3 - K_4)}\right), \tag{A6}$$

where K_1, K_2, K_3, and K_4 are defined in Eq. (6). For the Al^+/Hg^+ and Yb^+/Yb^+ systems we use the calculated values of the field shift and mass shift constants (Ref. [67] for Yb^+ and the present work for Hg^+) and the experimental limits on the variation of the frequency ratios [29,31] to obtain the mass variation from (A6). For the Yb^+/Cs system, we use the limits on the coupling of the electron to proton mass ratio $\mu = m_p/m_e$ to gravity presented in Ref. [43] to place constraints on the fractional variation of the nucleon and electron mass due to changes in the distance to Sun. These calculations may once again be used to place constraints on the Yukawa-type interactions of the scalar field from the Sun/Moon with nucleons and electrons, noting the following relations which may be yielded from Eq. (46)

$$\frac{\delta m_n}{m_n} = \frac{\delta\phi_s}{\Lambda_n}, \tag{A7}$$

$$\frac{\delta m_e}{m_e} = \frac{\delta\phi_s}{\Lambda_e}. \tag{A8}$$

Then following the same steps as for Λ_γ we obtain

$$\Lambda_n = \sqrt{\left|\frac{m_n a_n D_{S,M}}{\delta m_n/m_n}\right|}, \tag{A9}$$

where $a_n = 1.25 m_n$ for the Sun and $a_n = 24 m_n$ for the Moon. To get an expression for Λ_e we replace a_n by a_e and $\delta m_n/m_n$ by $\delta m_e/m_e$ in (A9); $a_n = 5.5 \times 10^{-4} m_n$ for the Sun and $a_n = 6 \times 10^{-3} m_n$ for the Moon.

Note that in the case of the nucleon coupling constant Λ_n it is also possible to perform calculations using the values of κ_n, presented in Table II, however in this case the results provide weaker constraints.

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