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Limits on gravitational Einstein equivalence principle violation from monitoring atomic clock frequencies during a year

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The Sun's gravitational potential at Earth varies during a year due to varying Earth-Sun distance. Comparing the results of very accurate measurements of atomic clock transitions performed at different times in the year allows us to study the dependence of the atomic frequencies on the gravitational potential. We examine the measurement data for the ratio of the frequencies in Hg^+ and Al^+ clock transitions and absolute frequency measurements (with respect to the caesium frequency standard) for Dy, Sr, H, hyperfine transitions in Rb and H and obtain significantly improved limits on the values of the gravity-related parameter of the Einstein equivalence principle violating term in the electron sector of the Standard Model extension Hamiltonian $c_{00} = (-3.0 \pm 5.7) \times 10^{-7}$ and the parameter for the gravity-related variation of the fine structure constant $\kappa_\alpha = (-5.3 \pm 10) \times 10^{-8}$.

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Theories unifying gravity with other interactions suggest that local Lorentz invariance (LLI) and the Einstein equivalence principle (EEP) might not be exact at very high energy [1]. This can manifest itself at low energy via a tiny change of atomic frequencies. Obtaining sensitivities to LLI and EEP violation requires that the frequencies being compared depend differently on the LLI and EEP violating parameters. One can search for the LLI or EEP violation in the electron sector, taking advantage of the extremely high accuracy of atomic clocks or using a system where the effect is strongly enhanced. The strongest constraint on the gravity-related EEP violation has been obtained with the use of atomic dysprosium [2]. The results of the measurements are interpreted in terms of the Standard Model extension (SME) Hamiltonian [3]. The corresponding term for the electron can be presented in the form (see, e.g. [4])

$$\delta H = c_{00}\frac{2}{3}\frac{U}{c^2}\frac{p^2}{2m}, \tag{1}$$

where c_{00} is one of the parameters in the SME characterizing the magnitude of the EEP violation, U is the gravitational potential, c is the speed of light, and p is the operator of the electron momentum ($\mathbf{p} = -i\hbar\nabla$).

The Hamiltonian (1) is written in the laboratory reference frame. The constant corresponding to c_{00} in the Sun reference frame is called c_{TT}. The difference between c_{00} and c_{TT} is small and usually neglected (see, e.g. Ref. [2]). There are also anisotropic gravity-dependent terms in the SME Hamiltonian which we do not consider in the present paper. We should note that the transformation between the Sun and laboratory reference frames produces sensitivity to c_{00} in the SME nongravitational term describing a correction to the kinetic energy of particles. The nongravitational limits on c_{TT} are presented in the regularly updated review [5]. For example, the limit $c_{TT} > -5 \times 10^{-21}$ has been obtained from astrophysical observations [5,6].

The change of the frequency of atomic transition between states a and b between two dates in the year is

$$\Delta\omega_{ab} = c_{00}\frac{2}{3}\frac{\Delta U}{c^2}\left[\left\langle\frac{p^2}{2m}\right\rangle_a - \left\langle\frac{p^2}{2m}\right\rangle_b\right] \equiv c_{00}\frac{2}{3}\frac{\Delta U}{c^2}\delta K_{ab}. \tag{2}$$

To avoid any confusion with the sign, let us assume that state a is always above state b on the energy scale so that $\hbar\omega_{ab} = E_a - E_b > 0$. ΔU in (2) is the change of the Sun's gravitational potential due to changing of the Earth-Sun distance, $\langle\frac{p^2}{2m}\rangle_a$ is the expectation value of the kinetic energy of electrons in state a, and δK_{ab} is the difference between the kinetic energies of the states a and b. The maximal change of the gravitational potential is between January and July, $\Delta U/c^2 \approx 3.3 \times 10^{-10}$ [7,8]. Therefore, comparing accurate frequency measurements performed in January and July, or fitting several measurements with a cosine function with the zero phase in the beginning of January and with period of one year, one can put constrains on the parameter c_{00},

$$c_{00} = \frac{3}{2}\frac{\Delta\omega_{ab}}{(\Delta U/c^2)\delta K_{ab}}. \tag{3}$$

Measuring atomic frequency means comparing it to some reference frequency, e.g. caesium primary frequency standard or another microwave or optical reference frequency. Therefore, we need to consider a ratio of two frequencies. In the nonrelativistic limit, one can use the virial theorem ($\langle p^2/2m\rangle = -E_{\mathrm{total}}$) and obtain from Eq. (2)

$$\frac{\Delta\omega}{\omega} = -c_{00}\frac{2}{3}\frac{\Delta U}{c^2}, \tag{4}$$

i.e. $\Delta\omega/\omega$ is the same for all electron transitions (except for the hyperfine transitions where the splitting is due to the "relativistic" magnetic interaction). This means that, in the nonrelativistic limit, the effect in the ratio of optical frequencies is unobservable. Therefore, we should perform relativistic calculations. It is convenient to introduce relativistic factors R which describe deviation of the expectation value of the kinetic energy from the value, given by the virial theorem,

$$R = -\frac{\Delta E_a - \Delta E_b}{E_a - E_b}. \tag{5}$$

Here ΔE_a is the energy shift of the state a due to the kinetic energy operator. In the nonrelativistic limit, $R = 1$. In the relativistic case, R can be larger or smaller than one and can even be negative. For the relative change of two frequencies, we now have

$$\frac{\Delta\omega_1}{\omega_1} - \frac{\Delta\omega_2}{\omega_2} = (R_2 - R_1)\frac{2}{3}c_{00}\frac{\Delta U}{c^2} \equiv (\beta_1 - \beta_2)\frac{\Delta U}{c^2}, \tag{6}$$

where $\beta = \frac{2}{3}Rc_{00}$. It is clear from (6) that, for higher sensitivity, one should compare the frequencies of atomic transitions with the largest possible difference in the values of relativistic factors R. It is convenient to rewrite (6) in a form

$$c_{00} = \frac{3}{2}\frac{\Delta\omega_1/\omega_1 - \Delta\omega_2/\omega_2}{(R_2 - R_1)\Delta U/c^2}. \tag{7}$$

If we fit the change of atomic frequencies to a cosine function

$$\frac{\partial}{\partial t}\ln\frac{\omega_1(t)}{\omega_2(t)} = A\cos(2\pi t/1\ \mathrm{yr}) \tag{8}$$

and note that maximum change of the frequency ratio is $2A$ then we can obtain from (7)

$$c_{00} = \frac{3A}{(R_2 - R_1)(\Delta U/c^2)}. \tag{9}$$

The change of an atomic frequency can also be attributed to the variation of the fine structure constant α ($\alpha = e^2/\hbar c$)

$$\frac{\Delta\omega}{\omega} = K_\alpha\frac{\Delta\alpha}{\alpha} \equiv \frac{2q}{\omega}\frac{\Delta\alpha}{\alpha}. \tag{10}$$

Here K_α and q are the electron structure factors ($K_\alpha = 2q/\omega$) which come from atomic calculations. Assuming that α can vary with the gravitational potential, one can write [7]

$$\frac{\Delta\alpha}{\alpha} = \kappa_\alpha\frac{\Delta U}{c^2}, \tag{11}$$

where κ_α is an unknown parameter. Using (10) and (11) we find

$$\kappa_\alpha = \frac{(\Delta\omega/\omega)}{K_\alpha(\Delta U/c^2)}. \tag{12}$$

For the case of relative change of two frequencies one can write

$$\kappa_\alpha = \frac{\Delta\omega_1/\omega_1 - \Delta\omega_2/\omega_2}{(K_{\alpha1} - K_{\alpha2})(\Delta U/c^2)}. \tag{13}$$

Comparing (3) and (12) we see that the same experimental data on the change of atomic frequencies between January and July can be used to put constrains on the gravity-related parameter c_{00} of the EEP violating Hamiltonian, and variation of the fine structure constant due to the change of the gravitational potential (κ_α). In principle, the parameters c_{00} and κ_α may have different physical origin and have different dependence on the reference frame (see e.g. Refs. [1,3,7]). However, for convenience in interpretation of different laboratory experiments, we can relate two parameters to each other using Eqs. (3), (10), and (11),

$$c_{00} = \frac{3q\kappa_\alpha}{\delta K_{ab}}, \tag{14}$$

or for the case of two frequencies, using (6) and (13), we get

$$\kappa_\alpha = \frac{R_2 - R_1}{K_{\alpha1} - K_{\alpha2}}\frac{2}{3}c_{00}. \tag{15}$$

The study of the variation of the fine structure constant α due to the change of the gravitational potential was a subject of previous works [7–10]. In this paper, we mostly focus on the EEP violating term (1).

It was shown in Ref. [4] that the values of the matrix elements of the kinetic energy operator are very sensitive to the many-body effects. Therefore, it is convenient to reduce the calculations to the calculation of the energies where we have accurate methods for the relativistic many-body calculations. The relativistic form of the EEP violating operator can be written as $2E_K = c\gamma_0\gamma^j p_j$, where E_K is kinetic energy, c is speed of light, γ^j are Dirac matrixes, and p is an operator of electron momentum (see, e.g [11]). The inclusion of the EEP violating operator into the calculation can be reduced to the simple rescaling of the kinetic energy term in the Dirac equation (we use atomic units),

$$\left(\frac{\partial f}{\partial r} + \frac{\kappa}{r}f\right)(1+s) - [2 + \alpha^2(\epsilon - \hat{V})]g = 0,$$
$$\left(\frac{\partial g}{\partial r} - \frac{\kappa}{r}g\right)(1+s) + (\epsilon - \hat{V})f = 0. \tag{16}$$

Here f and g are upper and lower radial components of the single-electron wave function ψ, which in the spherically symmetric case can be written in the form

$$\psi_{\kappa mn}(\mathbf{r}) = \frac{1}{r}\begin{pmatrix} f_n(r)\Omega_{\kappa m}(\mathbf{n}) \\ i\alpha g_n(r)\Omega_{-\kappa m}(\mathbf{n}) \end{pmatrix}, \tag{17}$$

where ($\mathbf{n} = \mathbf{r}/r$), s in ((16)) is the rescaling parameter, and $s = 0$ corresponds to the Dirac equation with no extra operator. The value of s should be chosen to ensure the linear dependence of the energy shift on s. In practice, it can be taken between 10^{-4} and 10^{-3}. The potential $\hat{V}$ in (16) includes the nuclear and electronic parts. The electronic part is usually the self-consistent Hartree-Fock-Dirac potential. Note that the EEP violating perturbation operator δH (the parameter s) is included into the self-consistent procedure and produces a correction to the Hartree-Fock potential (in the linear approximation in δH, this is equivalent to the random-phase approximation (RPA) with exchange).

The rest of the calculations are the same as for the energies. One can use the many-body perturbation theory (MBPT), configuration interaction (CI), or any other technique suitable for a many-electron atom and perform calculations for several values of the rescaling parameter s, including $s = 0$. Then the linear energy shift with respect to s is extracted. The advantage of this approach comes from the fact that the accuracy for the energy shift is expected to be similar to the accuracy for the energy which may be controlled by the comparison of the calculated and experimental energies. This is important since a strong sensitivity of the EEP violating energy shift to the many-body effects makes it difficult to estimate the accuracy of the calculations.

The actual choice of the computational technique is dictated by the configuration of external electrons. Table I lists popular optical clock transitions. At least three different types of configurations can be found here. For atoms with one external electron and no core change in the clock transition (e.g., the $5s - 4d$ transition in Sr^+ and $6s - 5d$ transition in Yb^+), we use the correlation potential method with second-order correlation potential [12]. For atoms with two valence electrons (Al^+, Sr, Yb, Hg), we use the combination of the configuration interaction (CI) with the many-body perturbation theory [13,14]. Finally, for the clock transition involving excitation from the atomic core (the $4f^{14}6s^2\mathrm{S}_{1/2} - 4f^{13}6s^{22}\mathrm{F}_{7/2}$ transition in Yb^+, the $5d^{10}6s$ ${}^1\mathrm{S}_0 - 5d^96s^{22}\mathrm{D}_{5/2}$ transition in Hg^+), we use a version of the CI method for the many-valence-electron atoms [15,16].

The results of the calculations are presented in Table I. The largest sensitivity can be achieved when monitoring the ratio of the frequencies of E2 and E3 transitions in Yb^+ ($\Delta R = 3.38$, see Table I). This ratio has been recently measured to about 10^{-16} accuracy [17]. However, separate sets of data dated between January and July (or between July and December) are not available. For other clock transitions, the typical value of the measured frequency ratios is also $\sim 10^{-16}$ (see, e.g. [18,19]), and a typical value of ΔR is $\Delta R \sim 0.2$–0.8 (see Table I).

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

Atom/Ion	Ground state			Clock state		$\hbar\omega$ [cm^{-1}]	R
Al^+	$3s^2$	${}^1\mathrm{S}_0$	-	$3s3p$	${}^3\mathrm{P}_0^o$	37393	1.00
Sr^+	$5s$	${}^2\mathrm{S}_{1/2}$	-	$4d$	${}^2\mathrm{D}_{5/2}$	14556	1.20
Sr	$5s^2$	${}^1\mathrm{S}_0$	-	$5s5p$	${}^3\mathrm{P}_0^o$	14317	1.03
Yb	$6s^2$	${}^1\mathrm{S}_0$	-	$6s6p$	${}^3\mathrm{P}_0^o$	17288	1.20
Yb^+	$6s$	${}^2\mathrm{S}_{1/2}$	-	$5d$	${}^2\mathrm{D}_{3/2}$	22961	1.48
Yb^+	$6s$	${}^2\mathrm{S}_{1/2}$	-	$4f$	${}^2\mathrm{F}_{7/2}$	21419	−1.9
Hg	$6s^2$	${}^1\mathrm{S}_0$	-	$6s6p$	${}^3\mathrm{P}_0^o$	37645	1.40
Hg^+	$5d^{10}6s$	${}^2\mathrm{S}_{1/2}$	-	$5d^96s^2$	${}^2\mathrm{D}_{5/2}$	35515	0.2

Several extra steps are needed to calculate the relativistic factors for microwave transitions, e.g. the hyperfine transition in caesium which serves as the primary frequency standard. First, the wave functions are found using Eq. (16) with the rescaled kinetic energy operator. Second, some standard technique is used to calculate the hyperfine structure including the many-body corrections. We use the correlation potential method (see, e.g. [12,20,21]) to calculate the relativistic factors for the hyperfine structure of the ground state of all alkali atoms from Li to Cs. This *ab initio* method provides accuracy ~1% for the hyperfine structure of the alkali atoms. The method includes solving the Hartree-Fock-Dirac equations in an external hyperfine filed [equivalent to the random phase approximation (RPA)] to account for the core polarization effect. The RPA equations are similar to Eq. (16) but with the right-hand side containing the hyperfine interaction operator. The kinetic energy terms in the RPA equations have also been rescaled. As for the energies, the calculations are done for several values of the rescaling parameter s.

In the nonrelativistic limit, the effect of the kinetic energy rescaling results in $R_k = 2.5$ for the hyperfine splitting. Indeed, the hyperfine splitting is due to the "relativistic" magnetic interaction and depends on the kinetic energy differently from the energies in the electrostatic atomic potential. The direct analytical calculation based on Eq. (16) shows that the kinetic energy scales as $1/(1+s)^2$ while the hyperfine structure scales as $1/(1+s)^5$; i.e., the ratio of the parameters R is equal to 5/2. This result is confirmed by the numerical calculations.

There is another effect which we should take into account in the case of the hyperfine transitions. We should take the EEP violating interaction in the gauge invariant form replacing momentum $\mathbf{p}$ by $\mathbf{p} - e/c\mathbf{A}$, where A is the electromagnetic vector potential:

$$\delta H = c_{00}\frac{2}{3}\frac{U}{c^2}\frac{(\mathbf{p} - e/c\mathbf{A})^2}{2m}. \tag{18}$$

TABLE II. Relativistic factors for the hyperfine clock transitions in atoms.

	H	Li	Na	K	Rb	Cs
Z	1	3	11	19	37	55
R	1.50	1.50	1.51	1.53	1.66	1.89

[The relativistic form of the EEP violating operator, which we actually use in the hyperfine structure calculations, is proportional to $c\gamma_0\gamma^j(p_j - e/cA_j)$]. This means that the EEP violating correction gives the additional factor $(1 + c_{00}\frac{2}{3}\frac{U}{c^2})$ to the magnetic interaction. This correction effectively replaces the relativistic factor R_k for the hyperfine interaction, which was obtained by the rescaling of the kinetic energy, by $R = R_k - 1$. For example, the nonrelativistic limit for R will be 1.5 (instead of 2.5 for R_k). To clarify this statement, it is useful to note that R_k scales in the nonrelativistic limit as $1/(1+s)^5$, and the value of the rescaling parameter s at the physical point is $s = \frac{1}{2}c_{00}\frac{2}{3}\frac{U}{c^2}$.

The calculated relativistic factors for the hyperfine transitions are presented in Table II.

The EEP violating correction also affects the magnetic interaction between atomic electrons and gives the EEP violating correction to the Breit interaction Hamiltonian. However, the contribution of the Breit interaction to energies of optical transitions is negligible.

(a) Al$^+$ vs Hg$^+$. Figure 1 shows the results of the measurements of the ratio of frequencies of Al$^+$ and Hg$^+$ clock transitions performed between December 2006 and November 2007 in Ref. [22]. Note that we approximately reproduce Fig. 3(A) of this paper. The measurements are fitted by the cosine function $A\cos(2\pi t/1\ \mathrm{yr}) + B$. The least mean square fitting leads to $A = (0.26 \pm 0.50) \times 10^{-16}$, $B = (4.3 \pm 0.4) \times 10^{-16}$. Substituting A into Eq. (9) and using the relativistic factors $R_1 = 1$ for Al$^+$ and $R_2 = 0.2$ for Hg$^+$ (see Table I), we obtain

$$c_{00} = (-3.0 \pm 5.7) \times 10^{-7}. \tag{19}$$

We can also use the data to extract the limit on the gravity-related variation of the fine structure constant κ_α. Using (15) and the values $K_{\alpha 1} \approx 0$ for Al$^+$ and $K_{\alpha 2} \approx -3$ for Hg$^+$ [23], we get

$$\kappa_\alpha = (5.3 \pm 10) \times 10^{-8}. \tag{20}$$

This represents an improvement of 5 times the previous result with dysprosium [24] (see below).

(b) Dysprosium. The dysprosium atom has a unique pair of degenerate states of the opposite parity, state A $4f^{10}6s5d$, $J = 10$ and state B $4f^9 5d^2 6s$, $J = 10$, both having energy $E_{A,B} = 19797.96\ \mathrm{cm}^{-1}$ above the ground state. Due to the extremely small energy interval between these states, many effects relevant to new physics are strongly enhanced [25–27]. The transition between these states was used to study parity nonconservation [28], time variation of the fine structure constant [2,9,24], LLI and EEP violation [2,8], and the search for dark matter [29]. The latest study of the coupling of the variation of the fine structure constant to gravity reveals [24]

$$\kappa_\alpha = (-5.5 \pm 5.2) \times 10^{-7}. \tag{21}$$

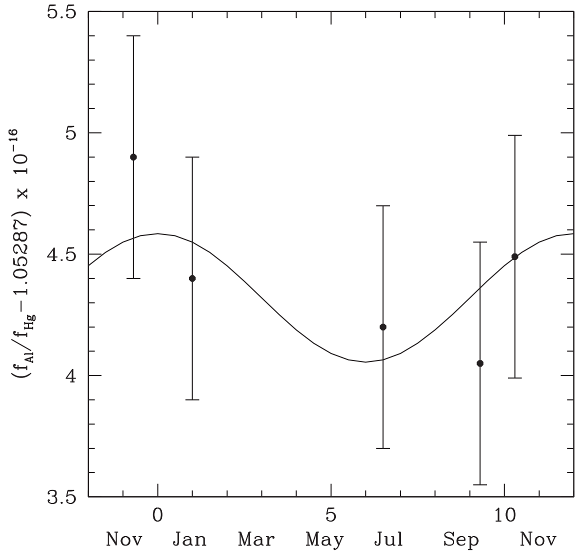

FIG. 1. Fit by $A\cos(\omega t) + B$ the results of the measurements of the frequency ratio for the clock transitions in Al$^+$ and Hg$^+$ [22] between November 2006 and November 2007.

We can use this result together with Eq. (14) to obtain the value of c_{00}. To do so, we also need to know Δq and δK_{ab}. Both values come from the atomic calculations. The sensitivity coefficients q for states A and B of Dy ($\Delta q = q_A - q_B$) were calculated in Refs. [16,30]. The values are $q_A = 7952\ \mathrm{cm}^{-1}$, $q_B = -25216\ \mathrm{cm}^{-1}$. Both values are stable and reliable. The value of δK_{ab} was calculated in Ref. [2]. However, we believe that this number is inaccurate. Therefore, we performed new, more accurate calculations and performed several tests to be sure that there are no mistakes. Our new calculations use the configuration interaction (CI) method in the Hartree-Fock-Dirac basis. For the ten valence electrons, we obtain $\langle p^2/2m\rangle_A = 57.632$ a.u., and $\langle p^2/2m\rangle_B = 57.803$ a.u., leading to $\delta K_{ab} = 0.174$ a.u. These values agree well with what is expected from the virial theorem linking kinetic and total energies. The CI energy of ten external electrons of Dy is $E^{\mathrm{CI}}_{A,B} = -61.89$ a.u. for both states A and B. The small difference between $|E^{\mathrm{CI}}|$ and $\langle p^2/2m\rangle$ can be attributed to the relativistic effects. In contrast, the values calculated and used in Ref. [2] are too large and strongly disagree with the virial theorem. In the end, our present value for δK_{ab} is 43 times smaller (with the minus sign) than in Ref. [2]. To exclude any mistake in the new calculation we performed it using two different methods. One is via the energy shift due to the rescaling procedure [see Eq. (16)], and the other is as a value of the matrix element of the EEP violating operator. The results are in perfect agreement.

Substituting the numbers into Eq. (14), we obtain

$$c_{00} = (6.4 \pm 6.0) \times 10^{-7}. \tag{22}$$

This result agrees with the result of Ref. [2] if the latter is corrected by a factor of 43.

(c) Hg^+ and Sr optical clocks, Rb and hydrogen. Frequency of the electric quadrupole clock transition in Hg^+ at $\hbar\omega = 35515\ \mathrm{cm}^{-1}$ (last line of Table I) was measured against the caesium clock to a very high precision in [31]. The limit on the modulated change of the frequency during a year was found to be

$$\frac{\Delta(\omega_{\mathrm{Hg}^+}/\omega_{\mathrm{Cs}})}{(\omega_{\mathrm{Hg}^+}/\omega_{\mathrm{Cs}})} = (0.7 \pm 1.2) \times 10^{-15}. \tag{23}$$

Substituting this number into Eq. (7) and using the values $R = 0.2$ for Hg^+ from Table I, the value $R = 1.89$ for Cs from Table II and the value for the change of the Sun's gravitational potential $\Delta U/c^2 \approx 3.3 \times 10^{-10}$ [7,8], we get

$$c_{00} = (1.8 \pm 3.2) \times 10^{-6}. \tag{24}$$

In Ref. [32], the values of parameters β [see Eq. (6)] have been measured using a comparison of the frequencies of the ${}^1\mathrm{S}_0$-${}^3\mathrm{P}^o_0$ clock transition in Sr against the caesium primary frequency standard. The result reads $\beta(\mathrm{Sr}) - \beta(\mathrm{Cs}) = (6.6 \pm 9.1) \times 10^{-7}$. Using (6) and (7) and the values of R from Tables I and II, we obtain

$$c_{00} = (1.4 \pm 2.0) \times 10^{-6}. \tag{25}$$

TABLE III. Limits on the EEP violating parameter c_{00} from different experiments.

Clocks	Ref.	c_{00}
$\mathrm{Al}^+/\mathrm{Hg}^+$	[22]	$(-3.0 \pm 5.7) \times 10^{-7}$
Dy/Cs	[2]	$(6.4 \pm 6.0) \times 10^{-7}$
$\mathrm{Hg}^+/\mathrm{Cs}$	[31]	$(1.8 \pm 3.2) \times 10^{-6}$
Sr/Cs	[32]	$(1.4 \pm 2.0) \times 10^{-6}$
Rb/Cs	[33]	$(4.8 \pm 4.2) \times 10^{-7}$
H/Cs	[34]	$(0.4 \pm 5.4) \times 10^{-6}$

In Refs. [33,34], the values of β have been measured using comparisons of the frequencies of the hyperfine transitions Rb/Cs and H/Cs: $\beta(\mathrm{Rb}) - \beta(\mathrm{Cs}) = (7.4 \pm 6.5) \times 10^{-7}$ and $\beta(H) - \beta(\mathrm{Cs}) = (0.1 \pm 1.4) \times 10^{-6}$. Again, using (6) and (7) and the data from Table II, we obtain

$$c_{00} = (4.8 \pm 4.2) \times 10^{-7}, \qquad c_{00} = (0.4 \pm 5.4) \times 10^{-6} \tag{26}$$

correspondingly. The results are summarized in Table III.

We see that the best limits provided by the current method on the value of c_{00} for the electron come from the frequency measurements in Al^+ vs Hg^+ (20) and Dy vs Cs (22), which stand at the 10^{-7} level. The current fractional accuracy for the Al^+ vs Hg^+ measurements is 10^{-16} [22]. Since optical clocks approach a fractional accuracy of 10^{-18} (see, e.g. [35–38]), further progress in limiting the gravity-dependent EEP violating interaction is possible. Many existing measurements can probably be used for this purpose if the date of the measurements is known.

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