Letter

Effects of variation of the fine structure constant α and quark mass m_q in Mössbauer nuclear transitions

Pavel Fadeev [1], Julian C. Berengut [2], and Victor V. Flambaum [1,2]
[1] *Helmholtz Institute Mainz, Johannes Gutenberg University, 55099 Mainz, Germany*
[2] *School of Physics, University of New South Wales, Sydney, New South Wales 2052, Australia*

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High accuracy measurements in Mössbauer transitions open up the possibility to use them in the search for temporal and spatial variations of the fine structure constant α, quark mass m_q, and dark matter field which may lead to the variations of α and m_q. We calculate the sensitivity of nuclear transitions to variations of α and m_q. Mössbauer transitions have high sensitivity to variation of quark mass m_q and the strong interaction scale Λ_{QCD} to which atomic optical clocks are not sensitive. The enhancement factors K, defined by $\frac{\delta f}{f} = K_\alpha \frac{\delta\alpha}{\alpha}$ and $\frac{\delta f}{f} = K_q \frac{\delta m_q}{m_q}$ where f is the transition energy, may be large in some transitions. The 8-eV nuclear clock transition in ^{229}Th ($K_q \approx 10^4$) and 76-eV transition in ^{235}U ($K_\alpha \approx K_q \approx 10^3$) may be investigated using laser spectroscopy methods.

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Introduction. Mössbauer spectroscopy has been used for diverse purposes ranging from gravitational redshift of light [1] to determinations of solids, atomic, and nuclear properties. The sensitivity of Mössbauer transitions can reach the 10^{-18} level, see, e.g., Ref. [2]. Moreover, a recent paper [3] claims that for a variable perturbation the sensitivity may reach $\Delta E \approx 10^{-15}$–$10^{-17}$ eV which corresponds to a 10^{-20}–10^{-22} relative sensitivity to the frequency shift. For comparison, the best atomic clock limit on relative changes of α is $1.0(1.1) \times 10^{-18}$ per yr [4] (atomic transition frequencies depend on α due to the relativistic effects [5–7]) whereas the limit on variation of m_q is $0.71(44) \times 10^{-14}$ per yr [8] (here sensitivity to m_q comes from the nuclear magnetic moments in Cs and Rb hyperfine transitions [9]). High sensitivity motivates the study of nuclear transitions for topics of fundamental physics, such as variations of the fundamental constants [10,11], search for new particles and interactions [3,11], and search for dark matter (see below). Different possibilities to produce a nucleus in the upper state of Mössbauer transition are discussed, e.g., in Ref. [3].

Variation of the fine structure constant α. The search for temporal and spatial variations of the fine structure constant α is an ongoing interdisciplinary endeavor spanning the fields of astrophysics, molecular, atomic, nuclear, and solid-state physics [12,13]. We elucidate the usage of high-precision Mössbauer spectroscopy in the search for variation of α. The sensitivity to the change in α is encoded in the enhancement factor K_α, defined as

$$\frac{\delta f}{f} = K_\alpha \frac{\delta\alpha}{\alpha}. \tag{1}$$

A nuclear transition energy f would change by δf due to a change in α by $\delta\alpha$. Values of K_α for current atomic clocks are on the order of 0.1–10 [5–7,14–16]. In nuclei K_α may be found from the following relation [10]:

$$K_\alpha = \Delta E_{\mathrm{C}}/f, \tag{2}$$

where ΔE_{C} is the change in Coulomb energy in this transition. In the ^{229}Th 8-eV nuclear clock transition studied, e.g., in Refs. [17–36] and expected to be a highly sensitive probe for time variation in α [10,37–42], our recent analysis [11] gives K_α of 10^4.

Variation of the quark mass and strong interaction. To avoid dependence on the human units which also may vary (e.g., hyperfine transition frequency in Cs, used to define the second and hertz, which has a complicated dependence on the fundamental constants [9]), we consider variations of dimensionless parameters, such as the fine structure constant α. Another dimensionless parameter which affects nuclear transition energies is $X_q \equiv m_q/\Lambda_{\mathrm{QCD}}$, where $m_q = (m_u + m_d)/2$ is the quark mass and Λ_{QCD} is the QCD scale. We do not make any assumptions about their independent variation since in this case we must specify the units in which we measure them. Here we measure m_q in units of Λ_{QCD}, i.e., in the calculations we may keep Λ_{QCD} constant.

The energy of a nuclear transition may be presented as

$$f = \Delta E_{\mathrm{C}} + E_S, \tag{3}$$

where E_S is the difference in bulk binding energies of the excited and ground states (including kinetic and strong potential energy but excluding the Coulomb interaction energy). Thus, using experimental value of the transition energy f and calculated value of the Coulomb energy difference ΔE_{C}, we can find $E_S = f - \Delta E_{\mathrm{C}}$. The dependence of E_S on quark mass was calculated in Ref. [37],

$$\frac{\delta E_S}{E_S} = -1.45 \frac{\delta m_q}{m_q}. \tag{4}$$

TABLE I. Sensitivity of Mössbauer transitions to the variation of the fine structure constant and of the quark mass. Coulomb energy shifts ΔE_C and enhancement factors K calculated using data which we list in the Supplemental Material [48]. We present in the table the experimental errors which are determined from the errors in $\Delta\langle r^2\rangle$ values. Our estimate for the constant charge density ansatz "theoretical" error is $\approx 25\%$.

		$T_{1/2}$	$j\pi$		ΔE_C (keV)	K_α	K_q
	(keV)	excited state	gr	ex	constant density	constant density	constant density
^{57}Fe	14.4	98 ns	1/2$^-$	3/2$^-$	39 (9%)	2.7 (9%)	2.4 (14%)
^{67}Zn	93.3	9.07 μs	5/2−	1/2−	−35 (27%)	−0.37 (27%)	−1.99 (7%)
^{83}Kr	9.3	147 ns	9/2+	7/2+	−15 (25%)	−1.6 (25%)	−3.8 (15%)
^{99}Ru	90	20.5 ns	5/2+	3/2+	−59 (26%)	−0.66 (26%)	−2.40 (10%)
^{119}Sn	23.9	17.8 ns	1/2+	3/2+	−25.1 (3%)	−1.053 (3%)	−2.98 (1%)
^{121}Sb	37.2	3.5 ns	5/2+	7/2+	183 (4%)	4.91 (4%)	5.67 (5%)
^{125}Te	35.5	1.48 ns	1/2+	3/2+	−13.3 (17%)	−0.37 (17%)	−1.99 (5%)
^{127}I	57.6	1.95 ns	5/2+	7/2+	56.1 (10%)	0.97 (10%)	−0.04 (400%)
^{129}I	27.8	16.8 ns	7/2+	5/2+	−69.7 (10%)	−2.51 (10%)	−5.08 (7%)
^{149}Sm	22.5	7.6 ns	7/2−	5/2−	−5.3 (29%)	−0.24 (29%)	−1.79 (6%)
^{151}Eu	22	9.5 ns	5/2+	7/2+	−99 (29%)	−4.6 (29%)	−8.1 (24%)
^{153}Eu	83.4	0.80 ns	5/2+	7/2+	10.2 (25%)	0.12 (25%)	−1.27 (3%)
^{153}Eu	103	3.9 ns	5/2+	3/2+	321 (15%)	3.1 (15%)	3.1 (23%)
^{155}Gd	86.5	6.35 ns	3/2−	5/2+	22 (25%)	0.25 (25%)	−1.09 (8%)
^{155}Gd	105	1.18 ns	3/2−	3/2+	30 (25%)	0.28 (25%)	−1.04 (10%)
^{157}Gd	64	0.46 ms	3/2−	5/2+	−55 (25%)	−0.86 (25%)	−2.69 (12%)
^{161}Dy	25.7	29 ns	5/2+	5/2−	−29 (25%)	−1.14 (25%)	−3.10 (13%)
^{161}Dy	43.8	0.78 ns	5/2+	7/2+	6.3 (25%)	0.14 (25%)	−1.24 (4%)
^{161}Dy	75	3.2 ns	5/2+	3/2−	−31 (25%)	−0.42 (25%)	−2.06 (7%)
^{181}Ta	6	6.05 ms	7/2+	9/2−	191 (25%)	30 (25%)	43 (26%)
^{197}Au	77.3	1.91 ns	7/2+	1/2+	−42 (29%)	−0.54 (29%)	−2.24 (10%)
^{229}Th	8	10^3, s	5/2+	3/2+	−67 (13%)	−0.82 10^4(13%)	−1.19 10^4(13%)
^{235}U	76	26 m	7/2−	1/2+	$\approx$100	10^3	10^3
^{243}Am	84	2.3 ns	5/2−	5/2+	235 (25%)	2.8 (25%)	2.6 (39%)

Using Eqs. (2)–(4), we obtain

$$\frac{\delta f}{f} = K_q \frac{\delta m_q}{m_q}, \quad K_q = 1.45(K_\alpha - 1). \tag{5}$$

Possible physical origins of α and m_q variations in Mössbauer transitions. There are several possible physical origins of α and m_q variations in Mössbauer transitions, some of which we illuminate here. Many popular theories extending the Standard Model contain scalar fields ϕ which interact with quarks q as $-(\phi/\Lambda_q)m_q\bar{q}q$. Here Λ_q is the interaction constant. This interaction may be added to the mass term in the Lagrangian $-m_q\phi\bar{q}q$ and presented as a dependence of the effective quark mass $m_q(\phi) = m_q[1 + (\phi/\Lambda_q)]$ on the field ϕ (see, e.g., Refs. [43,44]). Another possibility is that an interaction $(\phi/4\Lambda_\gamma)F_{\mu\nu}F^{\mu\nu}$ between the scalar field and the electromagnetic field $F^{\mu\nu}$ may be added to the electromagnetic term in the Lagrangian $F^{\mu\nu}F_{\mu\nu}/4$. This will manifest as a dependence of the fine structure constant $\alpha(\phi) = \alpha[1 + (\phi/\Lambda_\gamma)]$ on the field ϕ (see, e.g., Refs. [43–45]). Assuming that the source and absorber of the Mössbauer radiation are separated by some distance r, the values of α and m_q can be different at these points if the field ϕ varies in space (see below).

Yukawa field ϕ. The field ϕ may vary since the interaction between the field ϕ and the Standard Model particles leads to the Yukawa field $\phi = C\exp(-mr)/r$ produced by any massive body. The coefficient C has been calculated in Ref. [44]. In this way the presence of a massive body affects the fundamental constants.

For example, in the experiment [44] variations of the field ϕ and $\alpha(\phi)$ was produced by moving a 300-kg lead mass back and forth, affecting the ratio of the transition frequencies in Dy and Cs atoms. These have different dependence on α since in Dy K_α is strongly enhanced [5–7]. In the case of Mössbauer transitions, a mass may perform oscillating motion toward emitter (or absorber) of the radiation, producing a difference in the transition frequencies between the emitter and the absorber $\delta f = f(K_\alpha\delta\alpha/\alpha + K_q\delta m_q/m_q)$ which oscillates with the frequency of the mass motion.

Alternatively, the Yukawa field ϕ may be generated on a microscopic scale. In a recent paper [3] a technique to search for new scalar and tensor interactions at the submicrometer scale is presented. They suggest to place the optically flat "attractor" (source of Yukawa field ϕ), which perturbs the Mössbauer absorber frequency, on a micropositioner. This arrangement will provide a high sensitivity to the field ϕ with mass corresponding to the submicron Compton wavelength. Importantly, the paper [3] provides estimates of the systematic effects produced by the electromagnetic interactions and concludes that they are very small: the estimated sensitivity is $\Delta E \approx 10^{-15}$–$10^{-17}$ eV which corresponds to $\delta f/f \approx 10^{-20}$–$10^{-22}$. Based on these estimates and using $\delta f = f(K_\alpha\delta\alpha/\alpha + K_q\delta m_q/m_q)$ with values of K from Table I, we obtain sensitivity to the variations $\delta\alpha/\alpha \approx \delta m_q/m_q \approx$

10^{-20}–10^{-23}. This estimate may be optimistic, but we should compare it with the current limits from atomic transitions $\delta\alpha/\alpha \approx 10^{-17}$–$10^{-18}$ and $\delta m_q/m_q \approx 10^{-14}$.

A gradient of ϕ may also be due to the Yukawa field produced by a nearby mountain or by the whole Earth if this field has a large range (a small mass). Here the situation is somewhat similar to the measurements of the gradients of the gravitational field.

Dark matter field ϕ. If we identify the scalar field with dark matter, a gradient of the field $\phi = \phi_0 \cos(\mathbf{k}\cdot\mathbf{r} - \omega t)$ appears due to the nonzero wave-vector $k \approx mv/\hbar$, where v is the speed of Earth in the reference frame of the Galaxy and $\omega \approx mc^2/\hbar$, ϕ_0 is determined by the dark matter mass density (see, e.g., Refs. [43,45]). In this case we have oscillating $m_q(\phi) = m_q[1 + (\phi/\Lambda_q)]$ and $\alpha(\phi) = \alpha[1 + (\phi/\Lambda_\gamma)]$, which depend on the position $\mathbf{r}$. Here Λ_γ is the constant of the interaction between the scalar field ϕ and photon. Therefore, the dark matter field ϕ induces oscillations in the difference of the transition energies between separated emitter and absorber of the Mössbauer radiation.

A gradient of the field ϕ may exist in the transient field of passing clumps of dark matter, Bose stars, domain walls, etc. A gradient of ϕ may also exist in the field of scalar particles captured by Earth (see, e.g., reviews [13,46] and references therein).

Comparison of transition frequencies which have different dependences on fundamental constants. Search for variations of the fundamental constants in atomic experiments has been performed using time-dependence measurement of the ratio of two transition frequencies which have different dependences on the fundamental constants. A Mössbauer transition might be compared with a transition of approximately the same frequency in a highly charged ion. It may be challenging to find such an ion transition, but they may be sought in the spectra of ions with open f shell, which are very dense.

In the case of the 8-eV nuclear clock transition in ^{229}Th, laser optical spectroscopy methods, such as frequency comb, may be used for comparison with other transitions. High-frequency sources of coherent radiation, based on the multiplication of the frequencies of the laser field, should allow one to extend this approach to 76-eV transition in ^{235}U.

Calculation of the sensitivity to α and m_q variations in nuclear transitions. To deduce K_α for a particular transition, ΔE_C must be calculated. This can be performed using measurements of the changes in the mean-square charge radius $\Delta\langle r^2\rangle$ and intrinsic quadrupole moment ΔQ_0 between the ground and the excited states [11,47],

$$\Delta E_C = \langle r^2\rangle \frac{\partial E_C}{\partial\langle r^2\rangle}\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle} + Q_0 \frac{\partial E_C}{\partial Q_0}\frac{\Delta Q_0}{Q_0}. \tag{6}$$

To extract values of intrinsic electric quadrupole moments Q_0 from experimental data for the electric quadrupole moments $Q_{\rm lab}$ we use the following relation for rotating deformed nuclei:

$$Q_{\rm lab} = Z Q_0 \frac{I(2I-1)}{(I+1)(2I+3)}. \tag{7}$$

The use of this formula in nuclei with a small or zero deformation is not justified, however, the electric quadrupole in such nuclei is small and has little effect on the final result.

To calculate derivatives $\frac{\partial E_C}{\partial\langle r^2\rangle}$ and $\frac{\partial E_C}{\partial Q_0}$ we model the nucleus as a spheroid [11,47]. In such a model,

$$E_C = E_C^0 B_C, \tag{8}$$

$$E_C^0 = \frac{3}{5}\frac{q_e^2 Z^2}{R_0}, \tag{9}$$

where q_e is the electron charge, Z is the number of protons, and for a prolate spheroid ($Q > 0$),

$$B_C = \frac{(1-e^2)^{1/3}}{2e}\ln\left(\frac{1+e}{1-e}\right), \tag{10}$$

with e being the eccentricity. For an oblate spheroid ($Q < 0$) with the eccentricity defined such that it stays positive,

$$B_C = \frac{(1+e^2)^{1/3}}{e}\arctan(e). \tag{11}$$

If one of the ΔQ_0 or $\Delta\langle r^2\rangle$ measurements is missing, one way to estimate the result is by using the ansatz of constant charge density between isomers, which is equivalent to the ansatz of constant volume [11]. In such a case, for a spheroid,

$$\frac{dQ_0}{d\langle r^2\rangle} = 1 + \frac{2\langle r^2\rangle}{Q_0}. \tag{12}$$

Note that in Refs. [11,47] we tested the accuracy of Eq. (6) and constant density ansatz Eq. (12) using results of Hartree-Fock-Bogolyubov calculations [39] of ΔE_C, ΔQ_0, and $\Delta\langle r^2\rangle$ for the ^{229}Th nuclear transition. We estimated the error in the constant density ansatz of $\approx$25%.

In Table I we compile an extensive list of ΔE_C and enhancement factors K for Mössbauer transitions. The measured values of $\Delta\langle r^2\rangle$ and Q, which we use as an input, are presented in Table I in the Supplemental Material [48]. The accuracy of the electric quadrupole moments measurements at the moment is insufficient for extraction of reliable values ΔQ_0. Therefore, we base our results on the measured values of $\Delta\langle r^2\rangle$ (which in any case gives the main contribution to ΔE_C) and constant density ansatz Eq. (12) to find ΔQ_0. Note that if we neglect ΔQ_0, the value of ΔE_C would increase. Therefore, the constant density ansatz gives us a conservative estimate of ΔE_C and K.

For some elements $\Delta\langle r^2\rangle$ is absent in the literature, to the best of our knowledge. For an estimate, we could use the constant density ansatz Eq. (12) to find $\Delta\langle r^2\rangle$ using known values of the electric quadrupole moments Q in ^{235}U 46, ^{233}U 40, ^{179}Hf 123, ^{165}Ho 95, ^{160}Ho 60, and ^{158}Ho 67.2 keV. However, the errors in ΔQ_0 and K_α are too large in these cases, so we cannot make definite predictions.

We see in Table I that the average value of $|\Delta E_C|$ in medium and heavy deformed nuclei is $\approx$ 70 keV. Therefore, we may assume $\Delta E_C \approx 70$ keV and $|K_\alpha| \approx 70$ keV$/f$ in all medium and heavy deformed nuclei where accurate data for $\Delta\langle r^2\rangle$ are not available. In light nuclei and spherical nuclei $|\Delta E_C| \approx 30$ keV.

Two exceptional transitions presented in Table I are the 8-eV nuclear clock transition in ^{229}Th and the 76-eV transition in ^{235}U. Investigation of the ^{229}Th transition using laser spectroscopy methods has long been discussed in the literature, however, new sources of coherent radiation cover the range up to 100 eV (see, e.g., Ref. [49]), so 76-eV transition in ^{235}U may be investigated using high-precision spectroscopy too. The probability of the photon emission in the bare ^{235}U nucleus is very small, but it is significantly enhanced by the electronic bridge mechanism in many-electron ions [50] (see also Ref. [51]). To avoid discharge of the 76-eV nuclear excited state by electron emission, the ionization potential of the uranium ion should exceed 76 eV. This condition is satisfied in ions with charge bigger than 6. The values of the enhancement factors for 76-eV transition in ^{235}U, $K_q \approx K_\alpha \approx 10^3$ are estimated in the Supplemental Material [48].

To summarize, we show that nuclear transitions are a sensitive tool in the search for the variation of the fine structure constant α and especially variation of the strong interaction parameter $m_q/\Lambda_{\rm QCD}$ to which atomic optical transitions are not sensitive. We calculate the sensitivity to these parameters, presented as the enhancement factors K_α and K_q, for a number of Mössbauer transitions, 8-eV transition in ^{229}Th, and 76-eV transition in ^{235}U.

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