Coulomb energy contribution to the excitation energy in $^{229}$Th and enhanced effect of $\alpha$ variation

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Abstract – We estimated the polarization contribution of Coulomb energy to the spacing between the ground and first excited state of the $^{229}$Th nucleus as a function of the deformation parameter $\delta$. We show that despite the fact that the odd particle is a neutron, the change in Coulomb energy $\Delta U_C$ between these two states can reach several hundreds keV from this contribution. This means that the effect of the variation of the fine-structure constant $\alpha = e^2/\hbar c$ may be enhanced $\Delta U_C/E \sim 10^4$ times in the $E = 7.6$ eV “nuclear clock” transition between the ground and first excited states in the $^{229}$Th nucleus. To extract the full value of $\Delta U_C$, we propose to measure isomeric shift of atomic levels and nuclear electric quadrupole moments in $^{229}$Th.

Introduction. – Unification theories applied to cosmology suggest a possibility of variation of the fundamental constants in the expanding Universe (see, e.g., review [1]). There are hints of variation of $\alpha$ and $m_{q,e}/\Lambda_{QCD}$ in quasar absorption spectra, Big Bang nucleosynthesis and Oklo natural nuclear reactor data (see [2] and references therein). Here $\Lambda_{QCD}$ is the quantum thermodynamics (QCD) scale, and $m_q$ and $m_e$ are the quark and electron masses. However, the majority of publications report only limits on possible variations (see, e.g., [1,3–6]). A very sensitive method to study the variation in a laboratory consists of the comparison of different optical and microwave atomic clocks (see, e.g., measurements in [7–15]). An enhancement of the relative effect of $\alpha$ variation can be obtained in a transition between almost degenerate levels in the Dy atom [16]. These levels move in opposite directions if $\alpha$ varies. An experiment is currently under way to place limits on $\alpha$ variation using this transition [17], but unfortunately one of the levels has quite a large linewidth and this limits the accuracy. An enhancement of 1–5 orders exists in narrow microwave molecular transitions [18]. Some atomic transitions with enhanced sensitivity are listed in ref. [19].

In ref. [20] it was suggested that there might be five orders of magnitude enhancement of the variation effects in the low-energy transition between the ground and the first excited states in the $^{229}$Th nucleus. The existence of the enhancement was confirmed in [21]. This transition in $^{229}$Th was suggested as a possible nuclear clock in ref. [22]. Indeed, the transition is very narrow. The width of the excited state is estimated to be about $10^{-4}$ Hz [23] (the experimental limits on the width are given in [24]). The latest measurement of the transition energy [25] gives $7.6 \pm 0.5$ eV, compared to earlier values of $5.5 \pm 1$ eV [26] and $3.5 \pm 1$ eV [27]. This makes $^{229}$Th a possible reference for an optical clock of very high accuracy, and opens a new possibility for a laboratory search for the variation of the fundamental constants. Large interest in experiments exploring this possibility was expressed, for example, in several talks at a special workshop (14–18 July 2008, Perimeter Institute) devoted to the subject of variation of fundamental constants and private communications of E. Peik, P. Beiersdorfer, Zheng-Tian Lu, D. DeMille, D. Habs and J. Torgerson. As discussed in ref. [28] the part of the energy difference between the two states in $^{229}$Th that depends directly on $\alpha$, is the Coulomb energy difference. If one wants to detect the time variation of $\alpha$ one should study the variation in time of the Coulomb

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energy difference $\Delta U_C$ of the doublet of states in $^{229}$Th. A variation of the transition frequency $\nu$ is related to the variation of $\alpha$ by the equation from ref. [28]

$$\hbar \Delta \nu = \Delta U_C \frac{\delta \alpha}{\alpha},$$  \tag{1}

where $\Delta U_C$ is the contribution of the Coulomb interaction to the energy spacing between these two levels. One should mention that the energy of each state of the $^{229}$Th doublet is a result of cancellation of the total Coulomb energy and various contributions resulting from the strong interaction. We consider the Coulomb part only, because we are interested in the variation of $\alpha$.

In a recent work by Hayes et al. [29] the authors claim that there is almost no state dependence of the Coulomb energies for two states in $^{229}$Th, the upper limit being only 30 keV. The conclusion of the authors in ref. [29] is based on the notion that the deformations in the two states discussed are exactly the same, therefore the energies are the same. Note that this limit 30 keV is a result of cancellation of two very large Coulomb energies (950 MeV) of each state, therefore, ref. [29] claims that the difference is smaller than 0.03%.

In this letter we give an estimate of the effect that was not discussed in ref. [29] at all. Namely, it is a polarization contribution caused by a valence neutron in a nuclear core. Due to neutron-proton interaction this effect generates the changes in charge density. The changes depend on a particular state occupied by a valence neutron, contributing thus to the Coulomb energy difference. Even if the deformations are precisely the same, including the case when both are spherical, this effect gives non-zero result for the Coulomb energy difference.

**Shift of nuclear Coulomb energy.** — The ground state of the $^{229}$Th nucleus is $[Nn_{2}\Lambda J^P] = [633 3/2^+]$; i.e. the deformed oscillator quantum numbers are $N = 6$, $n_z = 3$, the projection of the valence neutron orbital angular momentum on the nuclear symmetry axis (internal $z$-axis) is $\Lambda = 3$, the spin projection $\Sigma = -1/2$, and the total angular momentum and the total-angular-momentum projection are $J = \Omega = \Lambda + \Sigma = 5/2$. The excited state is $[Nn_{2}\Lambda J^P] = [631 3/2^+]$; i.e. it has the same $N = 6$ and $n_z = 3$. The values $\Lambda = 1$, $\Sigma = 1/2$ and $J = \Omega = 3/2$ are different. With relation to the variation of alpha, we shall discuss below the polarization contribution to the Coulomb energy spacing between these two levels.

We start from qualitative considerations and calculate the shift of the Coulomb energy when an odd particle is added to a single-particle state $|\nu\rangle$ of the even $^{228}$Th nucleus. In an even nucleus the Coulomb energy can be presented as

$$U_C = \frac{1}{2} \int d^3 r d^3 r' \frac{\rho_c(r) \rho_c(r')}{|r - r'|},$$  \tag{2}

where $\rho_c(r)$ is the charge density. For uniformly charged sphere we obtain the well-known expression

$$U_C = \frac{3}{5} \frac{Z^2 e^2}{R},$$

which gives for $Z = 90$, and $A = 228$, the estimate, $U_C \sim 1$ GeV. The above equation can also be used to estimate the change in the Coulomb energy when one neutron is added. An addition of the neutron changes $A$, $A \rightarrow A + 1$, therefore

$$\Delta U_C = \frac{U_C}{3A},$$

that gives for $\Delta U_C$ a value of the order of $\sim 1$ MeV. This estimate gives some average macroscopic value for $\Delta U_C$ because in reality it depends on the state $|\nu\rangle$ where we put the neutron.

Adding an odd particle to the state $|\nu\rangle$, we obtain a change in the charge density

$$\langle \nu | \rho_c(r) |\nu\rangle = \rho_c(r) + \delta \rho_{\nu c}(r),$$  \tag{3}

where $\rho_c(r)$ is the charge density of a neighbour even nucleus. For the change in the Coulomb energy eq. (2) we obtain

$$\Delta U_C = \int d^3 r U_c(r) \delta \rho_{\nu c}(r),$$  \tag{4}

where $U_c(r)$ is the single-particle Coulomb potential

$$U_c(r) = \int d^3 r' \frac{\rho_c(r')}{|r - r'|}.$$  \tag{5}

Although $^{228}$Th is a deformed nucleus, for the sake of the estimate we shall neglect a small quadrupole component in the charge density $\rho_c(r)$. Effects of deformation will be discussed later. In the approximation of a uniformly charged sphere

$$U_c(r) = \left\{ \begin{array}{ll}
\frac{Z e^2}{R} \left( \frac{3}{2} - \frac{r^2}{2R^2} \right) & r \leq R, \\
\frac{Z e^2}{r} & r > R, 
\end{array} \right.$$  \tag{6}

where $R$ is a Coulomb radius of $^{228}$Th, $R = 1.2A^{1/3}$ fm.

The ground state of the nucleus $^{229}$Th is polarized due to the addition of the nucleon to the $^{228}$Th core. The added nucleon introduces an admixture of the monopole state (which is a radial excitation) [30,31] and the nuclear state is now

$$|0\nu\rangle = \sqrt{1 - \epsilon^2} |0^+; \phi_\nu\rangle + \epsilon |M; \phi_\nu\rangle,$$  \tag{7}

where $|0^+\rangle$ and $|M\rangle$ are the ground state and the isovector monopole in $^{228}$Th and $\phi_\nu$ is the wave function of the added nucleon, and

$$\epsilon = \frac{|0^+; \phi_\nu| F_N |M; \phi_\nu|}{E_0 - E_M},$$  \tag{8}

where $F_N$ is a two-body nuclear interaction.
Now we evaluate the Coulomb energy of this new polarized state \( |0^+\nu\rangle \). The correction is
\[
\Delta U_C^\nu = 2\epsilon \langle 0^+; \phi_\nu | \frac{1}{2} \sum_{i \neq j} V_C (r_i - r_j) | M; \phi_\nu \rangle, \tag{9}
\]
\( V_C (r) \) is the Coulomb potential. This correction can be written in the form
\[
\Delta U_C^\nu = 2Z/A \int \delta \rho (r) U_C (r) d^3r, \tag{10}
\]
\( \delta \rho (r) \) is the transition density between the ground state and the monopole:
\[
\delta \rho = \langle M | \psi^\dagger (r) t z \psi (r) | 0^+ \rangle.
\]
The \( Z/A \) factor takes into account the fact that only protons interact via the Coulomb interaction.

For the purpose of demonstration the strong-interaction matrix element is evaluated using a simple interaction:
\[
F_N = F_0 \delta (r_1 - r_2). \tag{11}
\]
Then
\[
(0^+; \phi_\nu | F_N | M; \phi_\nu \rangle = F_0 \int \delta \rho (r) | \phi_\nu (r) \rangle^2 d^3r. \tag{12}
\]
Finally
\[
\Delta U_C^\nu = \frac{2Z/AF_0 \int \delta \rho (r) U_C (r) d^3r \int \delta \rho (r) | \phi_\nu (r) \rangle^2 d^3r}{E_0 - E_M}. \tag{13}
\]
The result obviously depends on \( \phi_\nu \).

One form used for the monopole transitions is
\[
\delta \rho = C \left( 3 \rho + r \frac{d \rho}{dr} \right) = C \frac{1}{r^2} \frac{d (r^2 \rho)}{dr}, \tag{14}
\]
where \( C \) is a constant and \( \rho \) is the ground-state density. This form for the transition density corresponds to a change obtained by uniformly expanding the nucleus, but also can be derived from a sum rule \cite{30,31}. This transition density has a node. Because of the node in \( \delta \rho \) the result for the polarization correction will be sensitive to the spatial form of the wave function \( \phi_\nu \), and even may change sign for different single-particle states. Note that our calculation is an evaluation of the state dependence of the Coulomb interaction due to polarization caused by the addition of a neutron to the core. It is not a calculation of the Coulomb displacement energy (see ref. \cite{31}).

To obtain more quantitative estimate, let us switch to the particle-hole basis \( |ph\rangle \) instead of the single monopole resonance \( | M \rangle \). The correction to the charge density introduced in eq. (3) can be presented as follows:
\[
\delta \rho^{\nu \nu} (r) = \sum_{\lambda, \lambda'} \delta \rho_{\lambda \lambda'}^{\nu \nu} | \psi^\dagger_{\lambda'} (r) \psi_{\lambda} (r) \rangle,
\]
where \( \psi_{\lambda} (r) \) is the single-particle wave function, and \( \delta \rho_{\lambda \lambda'}^{\nu \nu} \) is the correction to the proton density matrix due to the odd nucleon added to the state \( |\nu\rangle \). Substituting this equation into eq. (4), we obtain
\[
\Delta U_C^\nu = \sum_{\lambda, \lambda'} (U_c)_{\lambda \lambda'} \delta \rho_{\lambda \lambda'}^{\nu \nu}, \tag{16}
\]
where \((U_c)_{\lambda \lambda'}\) is a matrix element of the single-particle Coulomb potential eq. (6).

The equation for \( \delta \rho_{\lambda \lambda'}^{\nu \nu} \) can be found for example in \cite{32}. It states
\[
\delta \rho_{\lambda \lambda'}^{\nu \nu} = \delta \epsilon_{\lambda} \delta \epsilon_{\lambda'} + \frac{n_{\lambda} - n_{\lambda'}}{\epsilon_{\lambda} - \epsilon_{\lambda'}} \sum_{\lambda_1, \lambda_2} \langle \lambda_1 | F_N | \lambda_2 \lambda' \rangle \delta \rho_{\lambda_1 \lambda_2}^{\nu \nu}.
\]
Here \( \langle \lambda_1 | F_N | \lambda_2 \lambda' \rangle \) is the matrix element of effective residual interaction between quasi-particles. The first term on the r.h.s. of eq. (17) exists only for an odd proton. If the odd particle is a neutron, this term is absent. The second term is the polarization correction which exists both for an odd proton and an odd neutron.

In the first order in the proton-neutron residual interaction we obtain for the correction to the proton density matrix
\[
\delta \rho_{\lambda \lambda'}^{\nu \nu} = \frac{n_{\lambda} - n_{\lambda'}}{\epsilon_{\lambda} - \epsilon_{\lambda'}} \sum_{\lambda_1, \lambda_2} \langle \lambda_1 | F_N | \lambda_2 \lambda' \rangle \delta \rho_{\lambda_1 \lambda_2}^{\nu \nu}, \tag{18}
\]
where the state \( |\nu\rangle \) refers to a neutron. The correction to Coulomb energy becomes
\[
\Delta U_C^\nu = \sum_{\lambda, \lambda'} (U_c)_{\lambda \lambda'} \frac{n_{\lambda} - n_{\lambda'}}{\epsilon_{\lambda} - \epsilon_{\lambda'}} \langle \nu | F_N^{\nu \nu} | \nu \lambda \rangle. \tag{19}
\]
Equation (19) can be presented in the following way:
\[
\Delta U_C^\nu = \int d^3r \delta U_C (r) | \psi_\nu (r) \rangle^2. \tag{20}
\]
Here we introduced the correction to the potential energy \( \delta U_C (r) \) that describes the reaction of the proton core to the presence of an additional neutron.
\[
\delta U_C (r) = \sum_{\lambda, \lambda'} (U_c)_{\lambda \lambda'} \frac{n_{\lambda} - n_{\lambda'}}{\epsilon_{\lambda} - \epsilon_{\lambda'}} | \psi^\dagger_{\lambda'} (r) \psi_{\lambda} (r) \rangle F_N^{\nu \nu} (r), \tag{21}
\]
where \( F_N^{\nu \nu} (r) \) is the density-dependent Migdal effective interaction. It follows from eq. (21) that the correction \( \delta U_C (r) \) does not depend on the neutron state \( |\nu\rangle \). However, its mean value given by eq. (20) does depend on the particular state \( |\nu\rangle \). The radial dependence of \( \delta U_C (r) \) is shown in fig. 1. The shape is not that different from the one given by eq. (14). This radial dependence shown is typical for a double charged layer. Due to small compressibility of a nucleus the change of a charge density can take place on the nuclear surface only, and in good approximation it can be presented as a double charged layer. The mean value of the correction \( \delta U_C (r) \) over the nuclear volume is negative and equal to \(-440\) keV. Due to the oscillating behaviour, the expectation value of \( \delta U_C (r) \) will obviously be state dependent. The calculations for

50005-p3
fore, we calculated the difference in Coulomb energies \( \delta U \) the change of the r.m.s. charge radius. For this reason polarization effect existing even in spherical nuclei is levels are crossed at the deformation

\[
\delta U = \frac{\delta \alpha}{\alpha} \Delta \mathcal{C}\nu \nu \text{Hz}.
\]

Table 1: Shifts of the Coulomb energy due to odd neutron for \( N = 6 \) shell orbitals.

| \( \Delta U_C \) (MeV) | \( 3d_{5/2} \) | \( 3d_{3/2} \) | \( 2g_{7/2} \) | \( 2g_{9/2} \) | \( 1i_{11/2} \) | \( 1i_{13/2} \) |
|-----------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
|                       | -0.74           | -0.93           | -0.85           | -1.38           | +0.16           | +1.30           |

\( ^{229}\text{Th} \) were performed in the following way. The main polarization effect existing even in spherical nuclei is the change of the r.m.s. charge radius. For this reason \( \delta U_C(r) \) was calculated using spherical single-particle wave functions for the core states. They were found by solving the Schrödinger equation with partially self-consistent potential [33]. The parameters \( F_{\text{in}} \) and \( F_{\text{ex}} \) of the Migdal interaction were fitted in [33] for calculations in the Lead region with full account of single-particle continuum. Deformation was accounted only for the valence neutron via Nilsson wave functions [34]. In \( ^{229}\text{Th} \) the ground state has the quantum numbers \([633 5/2^+]\) while the excited state has \([631 3/2^+]\). Both of them belong to \( N = 6 \) shell where the possible angular momenta in the Nilsson orbitals are \( L = 2, 4, 6 \). The shift of the Coulomb energy calculated due to the addition of valence neutron into these states is shown in table 1. Note, that the shift for \( 1i_{13/2} \) and \( 1i_{11/2} \) becomes positive. The radial wave function for \( L = 6 \) has a sharp peak near the nuclear surface where \( \delta U_C(r) \) is positive. The state \( 1i_{11/2} \) has smaller binding energy and its peak, compared to \( 1i_{13/2} \) state, is shifted to larger \( r \) where \( \delta U_C(r) \) becomes negative. This leads to a smaller value of the shift for \( 1i_{11/2} \) state.

The energy difference between the first excited state with quantum numbers \([631 3/2^+]\) and the ground state \([633 5/2^-] \) in \( ^{229}\text{Th} \) is extremely small (<10 eV) and cannot be reproduced in the Nilsson and other models at a reasonable deformation \( \delta \approx 0.2 \). With the standard parameters of the Nilsson model [34] these two levels are crossed at the deformation \( \delta \leq 0.1 \). Therefore, we calculated the difference in Coulomb energies \( \Delta U_C^{3/2} \) and \( \Delta U_C^{5/2} \) at different deformations, as a function of the parameter \( \eta = \delta / \kappa \), where \( \kappa = 0.05 \) is the spin-orbit constant. The result is shown in fig. 2 for \( 0 \leq \eta \leq 6 \). This interval corresponds to deformations \( 0 \leq \delta \leq 0.3 \). Note that the difference becomes negative at the deformations \( 0.15 \leq \delta \leq 0.25 \). However, the position of this interval depends on the parameters of the Nilsson model, while the fact that the difference changes sign at some deformation remains in all sets of the parameters. The value of \( \Delta U_C \) as a function of deformation changes from 1.5 MeV at zero deformation down to \(-0.5 \text{ MeV at } \delta = 0.3 \). In our approach we can hardly give a better estimate, due to the simplified treatment of deformation. However, a very small value of the Coulomb energy shift seems improbable. Note that near the Nilsson level crossing the difference of the Coulomb energies is about 1 MeV. The shift of the order from several tens keV to several hundreds keV provides an enhancement of sensitivity to changes of \( \alpha \), as it was mentioned in [20]. Indeed, the frequency shift from eq. (1) may be presented in the following form:

\[
\frac{\delta \nu}{\nu} = \frac{\Delta U_C}{100 \text{ keV}} \times \frac{\delta \alpha}{\alpha} \times 2.42 \times 10^{-19} \text{ Hz}.
\]

This shift is four-to-five orders of magnitude larger than the shift which was studied in optical atomic clocks [10,12–15]. An additional enhancement may be due to the very small width of this transition (\(10^{-1} \text{ Hz} \)). It is also instructive to present the relative enhancement:

\[
\frac{\delta \nu}{\nu} = \frac{\Delta U_C}{100 \text{ keV}} \times \frac{\delta \alpha}{\alpha} \times 1.3 \times 10^{4} \times \frac{\Delta U_C}{100 \text{ keV}} \times \frac{\delta \alpha}{\alpha}.
\]

These estimates confirm the conclusion of ref. [20] that experiments with \( ^{229}\text{Th} \) have the potential to improve the sensitivity of laboratory measurements of the variation of the fundamental constants by up to 8 orders of magnitude. Our calculation does not concentrate on the absolute value of the total Coulomb energy but on the question of variation of the Coulomb energy when one goes from one eigenstate to another. At this stage of nuclear theory it is impossible to compute differences of such energies which are a result of delicate cancellations of strong and Coulomb energies, with a precision of 7 eV. Therefore all one can do is to estimate the state-dependent variation of
the Coulomb energies. This turns out to be of the order of several tens to several hundreds of keV and leads to enhancement factors given above. Improved calculations using the completely self-consistent technique [35] and Coulomb correlations [36] may be considered in the future in order to obtain more accurate results.

We would like to bring attention to another possibility to obtain full Coulomb energy difference. We propose to resolve this problem experimentally by measuring an isomeric shift of atomic levels in $^{229}$Th. The lifetime of the excited state is long enough (1.5 hours [23]). This may give the possibility to obtain an experimental value of the change of the nuclear charge radius and hence $\Delta U_C$. To clarify the contribution of the deformation to the Coulomb energy difference one should also measure the difference of nuclear electric quadrupole moments of the ground and first excited nuclear states (using Nuclear Quadrupole Resonance or the atomic hyperfine-structure measurements; necessary atomic calculations have been started already).

In summary, we estimated the polarization contribution to the Coulomb energy difference between the ground and first excited state of the $^{229}$Th nucleus. To extract the full Coulomb energy difference including the effect of deformation, we propose to measure an isomeric shift of atomic levels and the difference in nuclear quadrupole moments since the lifetime of the excited state is long enough. This will give an experimental value of the change of the nuclear charge radius and $\Delta U_C$ and the difference in deformations.

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