Thorium: atom and nucleus... and clocks

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The revision of the SI will come into force on the World Metrology Day, May 20th, 2019.

All seven base units of the SI will be defined by fixing numerical values of seven "defining constants" such as, for example, the Planck constant, the elementary electric charge, or the Boltzmann constant.

As atomic physicists we are interested very much in second.
Atomic clocks based on hyperfine transitions

Very accurate clocks can be constructed by locking an electronic oscillator to the frequency of an atomic transition.

A second is defined as the duration of 9,192,631,770 cycles of microwave light absorbed or emitted by the hyper-fine transition of cesium-133 atoms undisturbed by external fields:

1 second = 9,192,631,770 cycles transition

The relative standard uncertainty of the Cs clocks is about $10^{-16}$.

Can the accuracy of atomic clocks be further improved? And why do we need this?
Are fundamental constants constant?

From the viewpoint of Standard Model the answer is clear:

**Constants are not constant!**

In early hot Universe masses of leptons and coupling constants are the same.

Physically correct question would be: how strong is **present variation** of constants?
Variation of fine-structure constant

Based on the data on quasars obtained by the Very Large Telescope a dipole-like structure in the variation of the fine-structure constant across the observable universe was reported in 2010.

The approaches, made in this work, are under discussion.

We need other methods to search for the variation of constants!

We know that even hydrogen atom is more complicated as described in simple Bohr formula:

$$\nu_{ik} = R_\infty \, c \, \left[ \frac{1}{n_i^2} - \frac{1}{n_k^2} \right]$$

We can consider transitions between:

- Fine-structure levels:
  $$\nu_{ik} = R_\infty \, c \, F_{fs}(\alpha, r_p, Z \ldots)$$

- Hyperfine-structure levels:
  $$\nu_{ik} = R_\infty \, c \, \alpha^2 \frac{m_e}{m_p} \, g \, F_{hfs}(\alpha, r_p, Z \ldots)$$
Variation of constants: Atomic spectroscopy

Different atomic levels can be shifted in different ways under the variation of the fine structure constant (as well as other constants).

Recent experiment with Cs and Yt$^+$ atomic clocks have improved the limit on the time variation of $\alpha$:

$$\frac{1}{\alpha} \frac{d\alpha}{dt} = -0.20(20) \times 10^{-16}/\text{yr}$$

Constraints on temporal variations of constants

Constraints on temporal variations of $\alpha$ and $\mu = m_e/m_p$
from comparisons of atomic transition frequencies.

N. Huntemann et al.,
Atomic clocks based on hyperfine transitions

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\[ 1 \text{ second} = 9,192,631,770 \text{ cycles transition} \]

The relative standard uncertainty of the Cs clocks is about $10^{-16}$.

Can the accuracy of atomic clocks be further improved? And why do we need this?
Atomic clocks: Improving accuracy

A second is defined by the transition in an atom undisturbed by external fields. But can we really “decouple” our atom from environment?

External fields lead to the shift of the energy levels (and, hence, of frequencies). For electric field, for example:

$$\Delta E \approx -\frac{1}{2} \alpha_0 \mathcal{E}^2 + \ldots$$

$\alpha_0$ - is the polarizability which describes the response of an atom to external field.

Alternative systems are searched which are less “coupled” to external fields. This will allow to make more accurate measurements.
Since 1990’s the $^{229}\text{Th}$ isotope attracts considerable attention as a promising candidate for the development of nuclear clocks.

What is so special with $^{229}\text{Th}$?
What is (un)known about $^{229}\text{mTh}$?

- **Energy of the isomeric state:**
  - Experimental results
  \[
  \mu_{iso} = -0.37(6) \mu_N, \\
  Q_{iso} = 1.74(6) |e| b
  \]

- **Angular momentum:** $J = 3/2^+$

- **Nuclear momenta:**
$^{229}$Th is in the focus of research

Intense studies are currently done on Th!

Many new results have been reported two weeks ago at the Spring Meeting of DPG in Rostock.

In our present work we are interested in nuclear momenta of $^{229}$Th isotope. We can extract them from hyperfine structure studies.
Nuclear properties and atomic spectroscopy

During the recent years many advances have been done in the high-precision atomic spectroscopy.

Measurements of atomic structure and interactions opens up new possibilities for studies of nuclear properties.

Atomic (ionic) levels can be modified owing to interactions with nuclei.

- **Mass shift** due to finite nuclear mass
  \[ M_N \]
- **Field shift** due to non-zero charge radii
  \[ \langle r^2 \rangle_A > 0 \]
- **Hyperfine effects** due to nuclear momenta
Hyperfine splitting in hydrogen atom

For the case of ground $1s_{1/2}$ state ($j=1/2$) of hydrogen, HF interaction results in splitting of energy level into two levels.

One can observe transition between two HF levels: famous 21 cm line in astrophysics!

21 cm radiation is used, for example, to measure radial velocities of spiral arms of Milky Way.

Analysis of the properties of galaxies.
Hyperfine structure: Reminder

The leading contributions arise due to magnetic dipole (M1) and electric quadrupole (E2) coupling resulting in energy shifts:

\[ \Delta E_{M1} = \frac{1}{2} AC \]

\[ \Delta E_{E2} = B \frac{C(C + 1) - J(J + 1)I(I + 1)}{2IJ(2J - 1)(2I - 1)} \]

\[ C = F(F + 1) - J(J + 1) - I(I + 1) \]

The most important “building blocks” here are the **hyperfine constants A and B** that account for the nuclear as well as electronic structure of an atom.

Example: hyperfine structure of hydrogen atom
Hyperfine constants A and B

For an electronic state characterized by its total angular momentum \( J \), parity \( p \), and further quantum numbers \( \gamma \), the hyperfine constants are obtained as:

\[
A = \frac{\mu_I}{I} \frac{1}{\sqrt{J(J+1)(2J+1)}} \langle \gamma J^p \| T^1 \| \gamma J^p \rangle \\
B = 2Q \left[ \frac{J(2J-1)}{(J+1)(2J+1)(2J+3)} \right]^{1/2} \langle \gamma J^p \| T^2 \| \gamma J^p \rangle
\]

The A and B constants are proportional to the nuclear magnetic dipole \( \mu_I \) and electric quadrupole \( Q \) moments.

From atomic physics side the reduced matrix elements \( \langle \gamma J \| T^k \| \gamma J \rangle \) of the hyperfine interaction operators enter into A and B.
Hyperfine constants A and B

In atomic theory we can calculate only "atomic part" of hyperfine constants:

\[
A = \frac{\mu_I}{I} \frac{1}{\sqrt{J(J+1)(2J+1)}} \langle \gamma J^p || T^1 || \gamma J^p \rangle
\]

\[
B = 2Q \left[ \frac{J(2J-1)}{(J+1)(2J+1)(2J+3)} \right]^{1/2} \langle \gamma J^p || T^2 || \gamma J^p \rangle
\]

We can plug in experimental or (nuclear) theory values of nuclear dipole and quadrupole momenta to find A and B. Or...

High-precision atomic calculations are required to find the reduced matrix elements \( \langle \gamma J^p || T^2 || \gamma J^p \rangle \)!
Many-electron ions: Theory

In quantum theory, states of an atom are described by their energy values and by wave-functions:

\[ E_n, \quad \Psi(r_1, r_2, r_3, ..., r_N) \]

The wave function is a function of 3N coordinates, where \( N \) is the number of electrons! How to deal with this huge dimension?

For heavy ions like Pb the starting point is the Dirac equation:

\[
\hat{H} = \sum_i \left( -i\hbar \mathbf{c} \alpha_i \cdot \hat{\mathbf{V}}_i + V(r_i) + m_e c^2 \gamma^0 \right) + \sum_{i<j} V(|r_i - r_j|)
\]

Hamiltonians of individual electrons

e-e interactions
Theory of many-electron systems

The Hamiltonian of many-electron atoms reads as:

$$\hat{H} = \sum_i \left( -i\hbar c \alpha_i \cdot \hat{\nabla}_i + V(r_i) + m_e c^2 \gamma^0 \right) + \sum_{i<j} V(|r_i - r_j|)$$

We can easily construct the many-electron wavefunctions and energies if we neglect the electron-electron interaction term:

$$\Psi_{Jp}(r_1, r_2, r_3, ..., r_N) = \sum_S d_S \begin{vmatrix} \varphi_1(r_1) & \ldots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \ldots & \varphi_N(r_N) \end{vmatrix}$$

Where $\varphi_i(r_i)$ are solutions of individual Hamiltonians:

$$\left( -i\hbar c \alpha_i \cdot \hat{\nabla}_i + V(r_i) + m_e c^2 \gamma^0 \right) \varphi_i(r_i) = \varepsilon_i \varphi_i(r_i)$$

Which potential to use here?
Theory of many-electron systems

We can construct the many-electron wavefunctions as:

$$\Psi_{f\ell}(r_1, r_2, r_3, ..., r_N) = \sum_s d_s \left| \begin{array}{ccc} \phi_1(r_1) & \cdots & \phi_N(r_1) \\ \vdots & \ddots & \vdots \\ \phi_1(r_N) & \cdots & \phi_N(r_N) \end{array} \right|$$

Where $\phi_i(r_i)$ are solutions of individual Hamiltonians:

$$(-i\hbar c \alpha_i \cdot \hat{V} + V_{eff}(r_i) + m_e c^2 \gamma^0) \phi_i(r_i) = \epsilon_i \phi_i(r_i)$$

Just Coulomb electron-nucleus interaction?

$$V_{eff}(r_i) = -\frac{Ze^2}{r}$$

Too rough! We need to account for e-e interactions

It is usually convenient to include a part of the e-e interaction already in the one-electron orbitals by introducing a screening potential

$$V_{DF}(r_1) \psi(r_1) = \sum_c \int dr_2 \psi^*_c(r_2) \frac{\alpha}{r_{12}} \times [\psi_c(r_2) \psi(r_1) - \psi_c(r_1) \psi(r_2)]$$

But... We need to account for the rest of e-e interactions!
Configuration interaction method

We can use CSF as basis functions to construct more accurate wavefunction

\[ \Psi_{jp}(r_1, r_2, r_3, \ldots, r_N) = \sum_r c_r \sum_s d_s \left| \begin{array}{cc} \varphi_1(r_1) & \varphi_N(r_1) \\ \vdots & \vdots \\ \varphi_1(r_N) & \varphi_N(r_N) \end{array} \right| \]

Summation over configurations  
State of particular symmetry \( |\gamma, j^p M\rangle \)

Ground state = \[ \begin{array}{c} + \\ + \\ + \end{array} \]

Here we virtually excite only valence electron while core electrons are „frozen“!
To make more accurate calculations we need also to „open the core“. 
Configuration interaction method

We can use CSF as basis functions to construct more accurate wavefunction

$$\Psi_{JP}(r_1, r_2, r_3, \ldots, r_N) = \sum_r c_r \sum_s d_s \begin{bmatrix} \varphi_1(r_1) & \ldots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \ldots & \varphi_N(r_N) \end{bmatrix}$$

Summation over configurations  
State of particular symmetry $|\gamma_r J^P M\rangle$

In the CI method, the energy levels of the system and the mixing coefficients $c_r$ are obtained by solving the secular equation

$$\text{det}\{(\gamma_r J^P M | \sum_i \hat{h}_i + \sum_{i<j} V(|r_i - r_j|) |\gamma_s J^P M\rangle - E_r \delta_{rs}\} = 0$$

In present calculations the $V(|r_i - r_j|)$ usually accounts for (frequency-dependent) Breit interaction corrections.
Hyperfine constants A and B

In atomic theory we can calculate only “atomic part” of hyperfine constants:

\[
A = \frac{\mu_I}{I} \frac{1}{\sqrt{J(J+1)(2J+1)}} \langle \gamma J^p \| T^1 \| \gamma J^p \rangle
\]

\[
B = 2Q \left[ \frac{J(2J-1)}{(J+1)(2J+1)(2J+3)} \right]^{1/2} \langle \gamma J^p \| T^2 \| \gamma J^p \rangle
\]

We can plug in experimental or (nuclear) theory values of nuclear dipole and quadrupole momenta to find A and B. Or...

The way around, we can compare with experimental values of A and B in order to extract information about nuclear momenta:

\[
\frac{A_{\text{exp}}}{A_{\text{theo}}/\mu_I} = \mu_I \quad \frac{B_{\text{exp}}}{B_{\text{theo}}/Q} = Q
\]
Experimental and theoretical studies have been performed for Th$^{2+}$ ion whose ground-state configuration is 5f 6d ("Radon core + 2 electrons").
Hyperfine spectra of $^{229}\text{Th}^2+$ and $^{229m}\text{Th}^2+$

Not all levels have been analyzed in the experiment.
Hyperfine structure of $^{229}$Th$^{2+}$

First, theory and experiment have been compared for the hyperfine structure of $^{229}$Th$^{2+}$ ion (i.e. when nucleus is in its ground state!)

<table>
<thead>
<tr>
<th>configuration</th>
<th>$J^\pi$</th>
<th>energy [cm$^{-1}$]</th>
<th>$A$[MHz]</th>
<th>$B$[MHz]</th>
</tr>
</thead>
<tbody>
<tr>
<td>[Rn] + 5$f$6$d$</td>
<td>4$^-$</td>
<td>0</td>
<td>64(17)</td>
<td>81(4)</td>
</tr>
<tr>
<td>[Rn] + 6$d^2$</td>
<td>2$^+$</td>
<td>63</td>
<td>143(47)</td>
<td>162(8)</td>
</tr>
<tr>
<td>[Rn] + 5$f^2$</td>
<td>4$^+$</td>
<td>15148</td>
<td>38(3)</td>
<td>72(3)</td>
</tr>
<tr>
<td>[Rn] + 5$f$6$d$</td>
<td>1$^-$</td>
<td>20711</td>
<td>109(36)</td>
<td>90(4)</td>
</tr>
<tr>
<td>[Rn] + 5$f^2$</td>
<td>4$^+$</td>
<td>21784</td>
<td>8(36)</td>
<td>26(2)</td>
</tr>
</tbody>
</table>
Hyperfine structure of $^{229}\text{mTh}^{2+}$

Extraction of the nuclear magnetic dipole moment

<table>
<thead>
<tr>
<th>configuration</th>
<th>$J^\Pi$</th>
<th>energy [cm$^{-1}$]</th>
<th>$A/\mu_{iso}$ [MHz/$\mu_N$]</th>
<th>$A$ [MHz] exp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>[Rn] + 6$d^2$</td>
<td>2$^+$</td>
<td>63</td>
<td>660 CI+MBPT 750 MCDF</td>
<td>-263(29)</td>
</tr>
<tr>
<td>[Rn] + 5$f6d$</td>
<td>1$^-$</td>
<td>20711</td>
<td>506 CI+MBPT 419 MCDF</td>
<td>-151(22)</td>
</tr>
</tbody>
</table>

\[
\frac{A_{exp}}{A_{theo}/\mu_I} = \mu_I
\]

Extraction of the nuclear electric quadrupole moment

<table>
<thead>
<tr>
<th>configuration</th>
<th>$J^\Pi$</th>
<th>energy [cm$^{-1}$]</th>
<th>$B/Q_{iso}$ [MHz/eb]</th>
<th>$B$ [MHz] exp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>[Rn] + 6$d^2$</td>
<td>2$^+$</td>
<td>63</td>
<td>22 CI+MBPT 23 MCDF</td>
<td>53(65)</td>
</tr>
<tr>
<td>[Rn] + 5$f6d$</td>
<td>1$^-$</td>
<td>20711</td>
<td>270 CI+MBPT 229 MCDF</td>
<td>498(15)</td>
</tr>
</tbody>
</table>

\[
\frac{B_{exp}}{B_{theo}/Q} = Q
\]

Experimental results

\[
\mu_{iso} = -0.37(6) \mu_N,
Q_{iso} = 1.74(6) |e| b
\]

R. Müller et al, PRA 98 (2018) 020503(R)
We have performed first theoretical calculations for the hyperfine constants of $^{229}\text{Th}^{2+}$ and $^{229m}\text{Th}^{2+}$.

When combined with experimental results our theoretical calculations allows yet another way to extract dipole and quadrupole moments of isomeric nuclear states.