Towards an Optical Nuclear Clock

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Generalized Ramsey Interrogation Schemes

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**Nuclear Clock:**
Based on an oscillator that is frequency-stabilized to a nuclear ($\gamma$-ray) transition

![Waveform and Mößbauer absorbers](image)

**Motivation:**

**Higher precision:** In most of the advanced optical clocks (trapped ion and optical lattice) field-induced shifts make a dominant contribution to the uncertainty budget (exception: Al$^+$ J=0-0). These can be reduced in a nuclear clock.

**Higher stability:** In a Mößbauer solid state nuclear clock, many absorbers may be interrogated ($>10^{10}$ instead of $\approx10^0$ (ion) or $\approx10^4$ (lattice)).

**Higher frequency:** $\rightarrow$ higher stability. EUV or even X-ray transitions may be used when suitable radiation sources become available.
Q.: Is it possible to obtain induced radioactivity by bombardment of matter with quanta of light?

A.: First of all, I have to say that, probably, there exists radioactivity of matter induced by the action of the light quanta; the difficulty of the observation of such phenomenon, if it exists, is that the effect which has to be observed is very small. The confirmation of this effect is hard but possible.

Albert Einstein

(Receiving the Academy’s honorary member diploma)

Annales Sociedad Cientifica Argentina, 107, 337 (1929)
Visible light is not matched to the nuclear scale of energy and length

Energy scales:
Visible photon: \( \hbar \omega \approx 2 \text{ eV} \)

Atomic nucleus: bound nucleon \( \Delta x \approx 5 \cdot 10^{-15} \text{ m} \)

with \( \Delta x \cdot \Delta p = \hbar \rightarrow \frac{\hbar^2}{2(\Delta x)^2 m_p} = 0.83 \text{ MeV} \)

(proton rest mass: 938 MeV)

Atomic shell: bound electron \( \Delta x \approx 10^{-10} \text{ m} \)

with \( \Delta x \cdot \Delta p = \hbar \rightarrow \frac{\hbar^2}{2(\Delta x)^2 m_e} = 3.8 \text{ eV} \)

(electron rest mass: 0.51 MeV)
Scales of electric field strengths:

\[ E = \frac{q}{4\pi\varepsilon_0 r^2} \]

shell: \[ r = 10^{-10} \text{ m} \] \[ E_H = 1,4 \cdot 10^{11} \text{ V/m} \]
nucleus: \[ r = 5 \cdot 10^{-15} \text{ m} \] \[ E_K = 5,8 \cdot 10^{19} \text{ V/m} \]

Field strength of an electromagnetic wave: related to intensity

\[ I = \frac{1}{2} \varepsilon_0 c E_L^2 \] \[ E_L = E_H \quad \rightarrow \quad I = 2,5 \cdot 10^{15} \text{ W/cm}^2 \]
\[ E_L = E_K \quad \rightarrow \quad I = 4,5 \cdot 10^{32} \text{ W/cm}^2 \]
Intensity in the focus of a short-pulse laser:

Limit intensity:

\[ I_{\text{max}} \approx N_{Ph} \cdot h \nu \cdot v \cdot \frac{V^2}{C^2} \]

Field strength in the shell: largely exceeded
Field strength in the nucleus: inaccessible

\[ N_{Ph} \approx \frac{\text{amplifier area}}{\text{transition cross section}} \approx 10^{12} \]

\[ I_{\text{max}} \approx 10^{24} \text{ W/cm}^2 \]

The nucleus is not a suitable antenna for visible light.

Lifetime for the radiation of electric multipole radiation of order $l$ from an antenna of dimension $r$:

$$\frac{1}{\tau_E(l)} = \frac{P}{\hbar \omega} \propto \omega \cdot \left(\frac{r}{\lambda}\right)^{2l} \quad \frac{r}{\lambda} \approx 10^{-8}$$

$$\tau_E(1) \approx 100 \text{ s} \quad \text{bei} \ 1 \text{ eV}$$

Long-lived excited states: Isomers

E.g. Ta-180: naturally occurring isomer, decays via E8 radiation ($l=8$) at 75.3 keV, $(r/\lambda)^{16} \approx 10^{-58}$

Half-life $> 10^{15}$ yr

0.012% abundance in natural tantalum

Using the electron shell as an antenna:

Would increase $r$ by about 5 orders of magnitude.

How to obtain a coupling between nucleus and electron?
Low-energy $\gamma$-emission is often in competition with decay via **internal conversion** (release of a free electron, most likely from the 1s shell, leads to ionization of the atom)

„Bound internal conversion“ or „Electronic Bridge“ in $\gamma$-decay

Photon is radiated from
the electron shell.
The process can be strongly enhanced in case of a resonance between nuclear and electronic transition frequencies.
Nuclear excitation via an „electronic bridge“ or NEET

• „Inverse electronic bridge“ or NEET (Nuclear Excitation by Electron Transition): Transfer of excitation from the electron shell to the nucleus

Advantages:

• Excitation energy can be supplied in a two-photon (or higher order) process

• Excitation rate may be strongly enhanced at resonance between electronic and nuclear transition frequency
Chart of Nuclides: Energy of the first excited state

Region of deformed nuclei with collective rotational and vibrational motion; partly chaotic in excited states.

Lowest (predictable) energies:
Rotation of heavy nuclei: 10-20 keV

magic nucleon numbers closed shell, deeply bound ground state

From: National Nuclear Data Center, Brookhaven National Laboratory
Nuclei with low-energy isomeric states

Pb-205    2329 eV
Hg-201    1565 eV
U-235    76.737(18) eV
Th-229    7.8(5) eV
Two close-lying band-heads: ground state and isomer
**Measurement of the energy of the Th-229 isomer**

γ-spectroscopy of two decay cascades from the 71.82-keV-level

Isomer energy:
Difference of the doublet splittings:
7.6 ± 0.5 eV
(cor.: 7.8 ± 0.5 eV, LLNL-Proc-415170)

Ground state → isomer: transition in the vacuum-ultraviolet at about 160 nm wavelength
Low-energy nuclear physics with $^{229}$Th


- highly precise and highly stable optical nuclear clock, trapped ions and doped solids. Peik and Tamm, Hudson, Kuzmich, Schumm

- test system for „new physics“: variations of fundamental constants, violation of Lorentz invariance. Flambaum et al.

  - $\gamma$-ray laser. Tkalya

  - ...

Review article:
E. Peik, M. Okhapkin, Comptes Rendus Physique 16, 536 (2015)
also available at: arXiv:1502.07322
A high-precision nuclear clock

An atomic clock shall realize the unperturbed transition frequency. The uncertainty of advanced optical clocks is often limited by field-induced shifts (e.g. from the blackbody field).

Nuclear moments are small. Field induced systematic frequency shifts can be smaller than in an (electronic) atomic clock.
How about interaction with the electrons?

Work with bare Th$^{90+}$? (Requires 110 keV ionisation energy; difficult to trap or cool)

Consider electric and magnetic hyperfine coupling.

**Magnetic:** Coupling of electronic and nuclear magnetic moments (visible in angular momentum quantum numbers and Zeeman splittings)

**Electric:** „shielding“: No linear Stark effect at the nucleus of an atom in the nonrelativistic limit. Schiff theorem (discussed in the context of parity violation, electric dipole moments)

„anti-shielding“: an external electric field gradient may be enhanced at the nucleus. Sternheimer antishielding (studied in NMR) predicted inversion and enhancement by factor -178 for Th$^{4+}$

Advantage of the nuclear over the atomic clock: (nearly) free choice of a suitable electronic state for the interrogation of the nuclear resonance. First analyzed for the Th$^{3+}$ system in:

LS coupled eigenstates of the electronic + nuclear system

Frequency shifts that only depend on $|n,L,S,J>$ are common in both levels and do not change the transition frequency.

Holds for: **scalar quadratic Stark shift**, including the effects static electric fields, collisions, blackbody AC Stark shift

**Tensor quadratic Stark** and **electric quadrupole shift**: vanish for $J<1$ or $F<1$

**Hyperfine Stark shift**: $F$-dependent, e.g. blackbody radiation shift
analog with Cs for $m_F=0-0$: $\approx 10^{-19}$ at room temperature
LS coupled eigenstates of the electronic + nuclear system

Frequency shifts that only depend on $|n, L, S, J>$ are common in both levels and do not change the transition frequency.

Holds for: **Scalar quadratic Stark shift**, including the effects of static electric fields, collisions, blackbody AC Stark shift

\[
\hbar \Delta \nu_S(\gamma, J, F, m, E) = - (2 \alpha_S(\gamma, J) + \alpha_T(\gamma, J, F) g(F, m, \beta)) \frac{|E|^2}{4}
\]

\[
g(F, m, \beta) = \frac{3m^2 - F(F + 1)}{F(2F - 1)} \left(3 \cos^2 \beta - 1 \right)
\]

$\beta$: angle between electric and magnetic field

**Tensor quadratic Stark and electric quadrupole shift**: (coupling of quadrupole moment to electric field gradient)

Have identical $m, F$ and $\beta$ dependence and vanish for $J<1$ or $F<1$
Linear Zeeman shift: use component $m_F=0 - 0$

Quadratic Zeeman shift: 

$$
\Delta f_{2\text{o}Z} = \left( g_J - \frac{m_e}{m_p} g_I \right)^2 \frac{\mu_B^2 B^2}{A}
$$

$A$: HFS splitting; Effects are comparable to other atomic clocks

Doppler shift: use ion trapping and laser cooling

→ choice of electronic state:
Half integer nuclear spin (like Th-229): $^2S_{1/2}$ or $^2P_{1/2}$
Integer nuclear spin $^1S_0$ or $^3P_0$

Options for the Th-229 trapped ion clock:
• Metastable 7s $^2S_{1/2}$ level in Th$^{3+}$ (direct laser cooling, quantum jump detection) (our proposal 2003)
• $^1S_0$ ground state in Th$^{4+}$ (m$F\neq0$, nuclear lin. Zeeman effect, but negligible quadrat. Zeeman effect (no HFS or FS splitting)) (use sympathetic cooling and quantum logic detection)

Eliminates field induced shift to a level not achievable in (electronic) atomic clocks.
Options for the Th-229 trapped ion clock:
• stretched states of $^2F_{5/2}$ ground state in Th$^{3+}$
  C. J. Campbell et al., PRL 108, 120802 (2012)
(applicable to any J quantum number. Lin. Zeeman effect corrected in averaged frequency, for example of $(m \to m')$ and $(-m \to -m')$ components)

Stretched states (of extremal $F$ and $m_F$) are eigenstates in the coupled as well as in the uncoupled basis.

$$|F = I + J, \ m_F = \pm F\rangle = |J, \ m = \pm J\rangle \otimes |I, \ m = \pm I\rangle$$

For example: Shift from room temperature blackbody radiation: analysis for stretched states $m_F=\pm(J+I)$: $\approx 10^{-22}$,
  C.J. Campbell et al., PRL 108, 120802 (2012)

Eliminates field induced shift to a level not achievable in (electronic) atomic clocks.
Detection of the Nuclear Excitation in Nuclear-Electronic Double-Resonance

Nucleus in the ground state; laser-induced fluorescence from the shell.

Laser excitation of the nucleus; change of hyperfine structure detected in intensity or polarisation of fluorescence.

Analog of Dehmelt's „electron shelving“
Observation of „quantum jumps“ in the single-ion fluorescence
Th$^{3+}$ possesses a simple level scheme. It can be laser-cooled using diode lasers and detected via resonance fluorescence at red or NIR wavelengths. Electronic and nuclear resonances are separated in energy.

Lifetime calculations:
Biémont et al., JPB 37, 4193 (2004)
Safronova et al. PRA 74, 042511 (2006)

Experiment:
Campbell et al., PRL 102, 233004 (2009)
Tests of Fundamental Physics with Atomic Clocks
Search for violations of the Equivalence Principle with different atomic clocks:
• Search for a time dependence of fundamental constants
• Test of the universality of the gravitational red shift
• Clock comparison test of Lorentz invariance
• Search for light dark matter
Th-229: the most sensitive probe in a search for variations of the fundamental coupling constants

Scaling of the $^{229}$Th transition frequency $\omega$ in terms of $\alpha$ and quark masses:


$$\frac{\delta \omega}{\omega} \approx 10^5 \left( 4 \frac{\delta \alpha}{\alpha} + \frac{\delta X_q}{X_q} - 10 \frac{\delta X_s}{X_s} \right)$$

where $X_q = m_q / \Lambda_{QCD}$ and $X_s = m_s / \Lambda_{QCD}$

$10^5$ enhancement in sensitivity results from the cancellation of $O(\text{MeV})$ contributions in the $O(\text{eV})$ transition energy.
Th-229: the most sensitive probe in a search for variations of the fundamental coupling constants

$\alpha$-sensitivity of nuclear transition frequency $\omega$ is due to the Coulomb contribution to the nuclear energy difference.

$$ \frac{\delta \omega}{\omega} = K \frac{\delta \alpha}{\alpha} $$

$$ K = \frac{E_{C,m} - E_{C,g}}{\hbar \omega} $$

Predictions vary:

$\Delta E_C \approx 0$ excitation of the unpaired neutron does not influence the proton distribution

$\Delta E_C \approx 1$ MeV required to obtain $E_{is} \approx 8$ eV from a cancellation involving Coulomb and strong interactions.

See for example:


Proposal: Use measurements of isomer shifts and atomic structure calculations

J. C. Berengut, V. A. Dzuba, V. V. Flambaum, S. G. Porsev, PRL 102, 210808 (2009)
Review articles:
E. Peik, M. Okhapkin, Comptes Rendus Physique 16, 536 (2015)
also at: arXiv:1502.07322

L. von der Wense, B. Seiferle, and P. G. Thirolf,
Measurement Techniques 60, 1178 (2018)

See also:

www.nuclock.eu
(for a comprehensive Thorium library)

L. von der Wense, Thesis LMU 2016: On the Direct Detection of $^{229m}$Th
(Springer, 2017)
Part II

Generalized Ramsey Interrogation Schemes
Rabi- and Ramsey-Excitation

2-level system with pulsed excitation. 
\( \chi \): res. Rabi frequency, \( \delta \): detuning

Excitation probability after the pulse

\[ P_2 = \int_{-\infty}^{\infty} \chi(t) e^{i \delta t/2} dt \]

(quadatic Fourier transform of the pulse)

1 Pulse (Rabi)

\[ P_2 = \frac{\chi^2}{J^2} \sin^2 \frac{\delta T}{2} \]

2 Pulses (Ramsey)

\[ P_2 = 4 \cos^2 J T \cdot \sin^2 \frac{\delta T}{2} \cdot \frac{\chi^2}{J^2} \]
Advantages of Ramsey excitation

about 0.5x narrower resonance for the same interaction time

Resonance not broadened by perturbations between the interaction zones (like B field inhomogeneity)
Light shift in Ramsey spectroscopy

Excitation with two pulses

Envelope: due to excitation in a single pulse, Strong light shift

Ramsey fringe: due to interference of two Pulses, separated by dark time, Smaller light shift

From:
High resolution excitation spectrum of the $^{171}\text{Yb}^+$ octupole transition

- 335 ms pulse duration
- 50 μW laser power
- Fourier-limited linewidth
- $Q = 2.7 \times 10^{14}$
- Light shift $\approx 4$ Hz
“Hyper-Ramsey” spectroscopy (HRS)

- Light shift $\Delta_L$ is compensated by the step frequency $\Delta_S$
- Additional $\pi$-pulse cancels the linear dependence on $\Delta_L$
  $\rightarrow$ Promises a strongly suppressed light shift
- Discriminator signal is generated by $\pm\pi/2$ phase steps

Experimental investigation of Hyper-Ramsey Spectroscopy

- HRS parameters: $T=36 \text{ ms}$, $2\tau=18 \text{ ms}$
- Largely unaffected position of central minimum
- Interleaved stabilization on $(\Delta_L-\Delta_S) = 0$ and $(\Delta_L-\Delta_S) \neq 0$
- Cubic dependence of the resonance center on $(\Delta_L-\Delta_S)$
Controlled HRS scheme

- Interleaved stabilization using Rabi spectroscopy with the same intensity and step frequency $\Delta_S$ to measure the frequency offset
- A negative feedback loop uses $v_{HRS} - v_{Rabi}$ to regulate $\Delta_S$ and ensures $|\Delta_S - \Delta_L|=0$
- Real-time measurement of the light shift
Cancellation of probe-field induced shifts

- Shifts due to the excitation / readout processes can perturb the atomic frequency.
- Strongly forbidden transitions are particularly attractive for optical clocks (narrow natural linewidth; but: Light shift or Zeeman shift associated with the excitation).
- Ramsey interrogation: spectroscopic information is accumulated during the dark period $T$.

\[ \omega_{\text{Ramsey}} = \omega_0 \rightarrow p_{\text{short}} = p_{\text{long}} \]
Stabilization in the autobalanced Ramsey scheme

- Based on frequency tuning of $\omega_{\text{Pulse}}$ and $\omega_{\text{Ramsey}}$
- Based on an additional phase step $\Delta \phi$ and $\omega_{\text{Ramsey}}$

$\pi/2 \pm 90^\circ$

- $p_{\text{short}}^- = ! p_{\text{short}}^+$
- $p_{\text{long}}^- = ! p_{\text{long}}^+$

- Two interleaved servos
- Appropriate for systems with slowly varying pulse defects

Auto-balanced Ramsey scheme – experimental results
Auto-balanced Ramsey scheme – experimental results

- Rabi-frequency reduction by phase-modulation with an EOM

equalized by EOM carrier reduction
Auto-balanced Ramsey scheme – experimental results

![Graph showing clock error vs. light shift estimate error with heating compensation and HRS annotation.]
Auto-balanced Ramsey scheme – experimental results

Pulse-defect-immunity of the auto-balanced Ramsey scheme

C. Sanner, N. Huntemann, R. Lange, Chr. Tamm, E. Peik, Phys. Rev. Lett. 120, 053602 (2018)
Auto-balanced Ramsey scheme – applications

Yb$^+$ electric octupole clock

Cs vapor cell CPT clock (coherent population trapping, driven by a modulated diode laser)

Optical lattice clock with bosonic atoms (I=0) and magnetic field-induced transition

C. Sanner, N. Huntemann, R. Lange, Chr. Tamm, E. Peik, Phys. Rev. Lett. 120, 053602 (2018)

Recent review article (theory-focussed):
Composite laser-pulses spectroscopy for high-accuracy optical clocks: a review of recent progress and perspectives

Thomas Zanon-Willette$^1$, Rémi Lefèvre$^1$, Rémi Metzdorff$^2$, Nicolas Sillitoe$^2$, Sylvain Almonacid$^3$, Marco Minissale$^4$, Emeric de Clercq$^5$, Alexey V Taichenachev$^6,7$, Valeriy I Yudin$^6,7,8$ and Ennio Arimondo$^9$
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