Thorium: atom and nucleus… and clocks

Andrey Surzhykov and Robert Müller

Technische Universität Braunschweig /
Physikalisch-Technische Bundesanstalt (PTB)

New System of Units

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*.

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Atomic clocks based on hyperfine transitions

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

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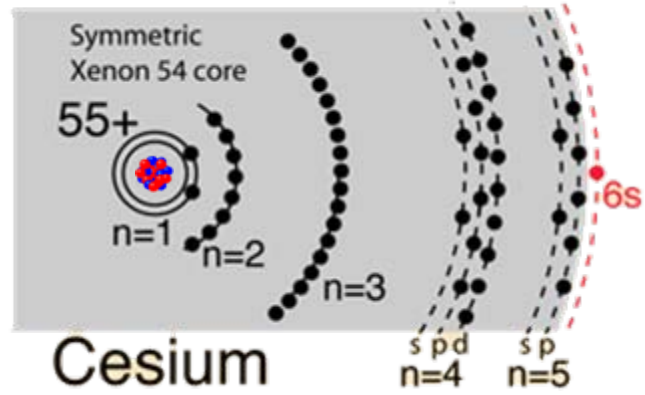
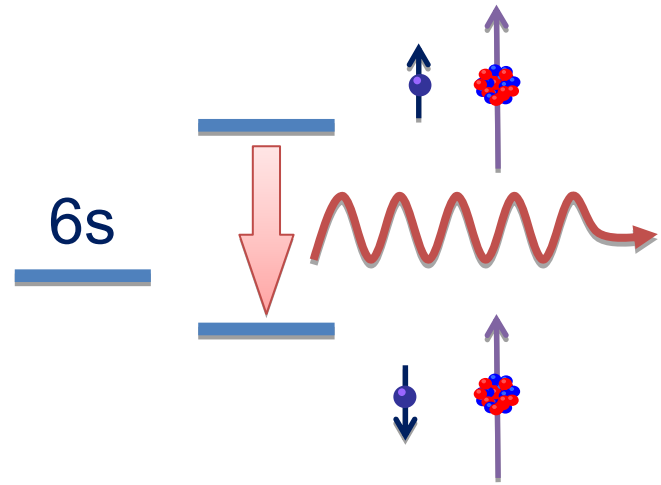


Quelle des Bildes: hyperphysics.phy-astr.gsu.edu



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

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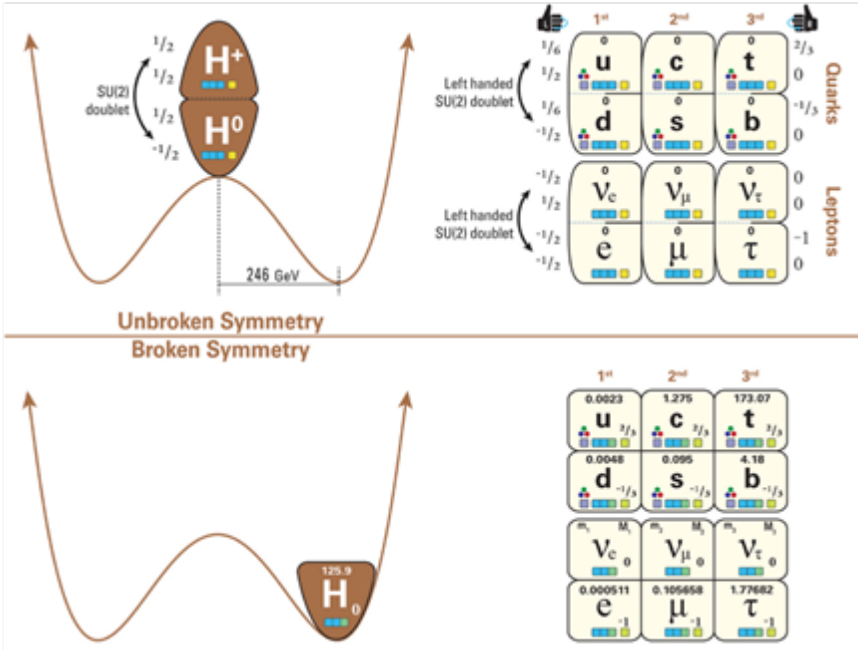
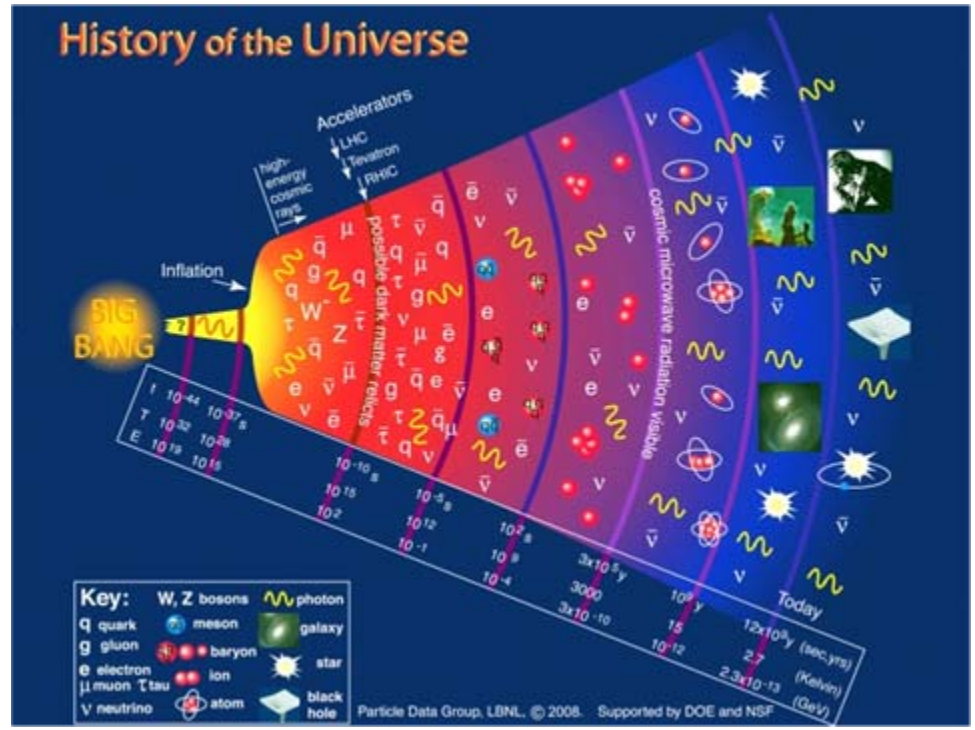
Are fundamental constants constant?

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?

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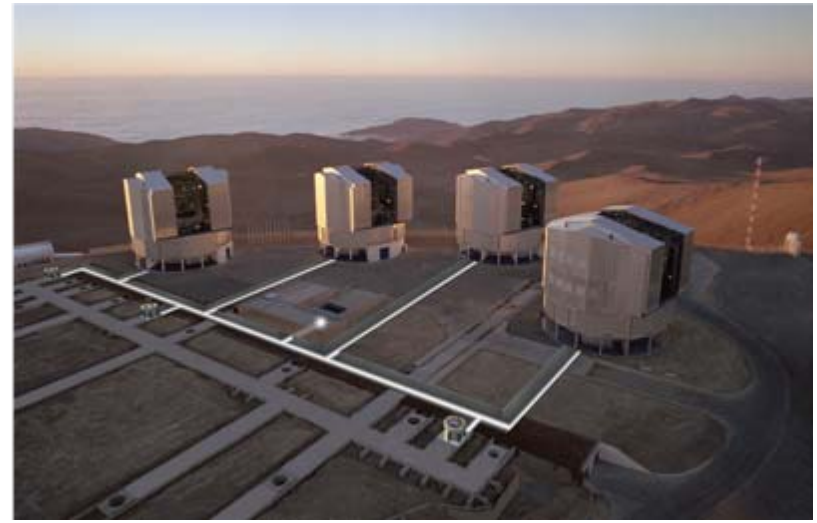
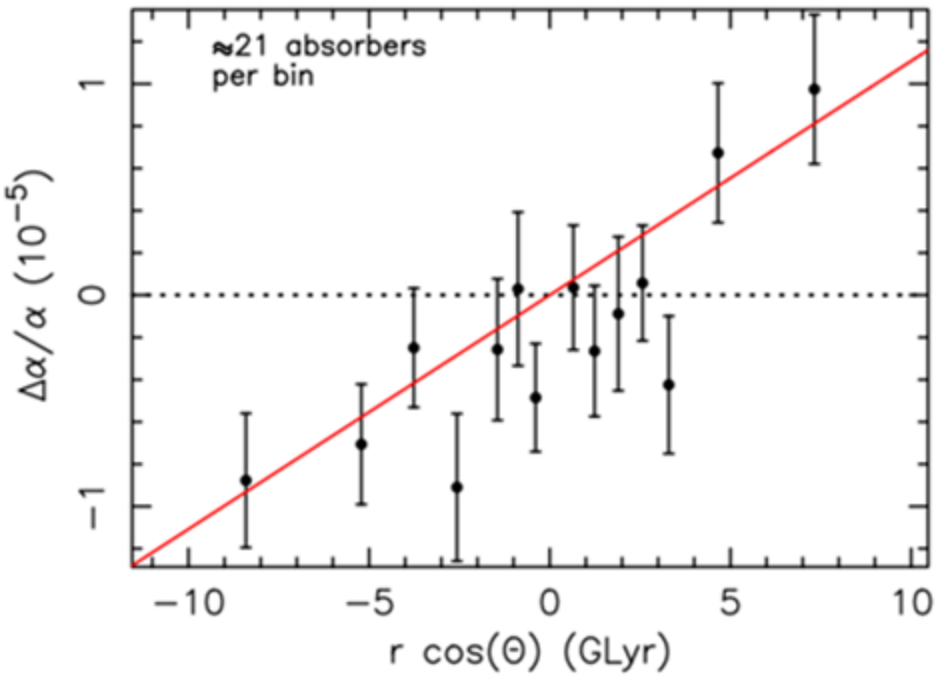
Variation of fine-structure constant

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.



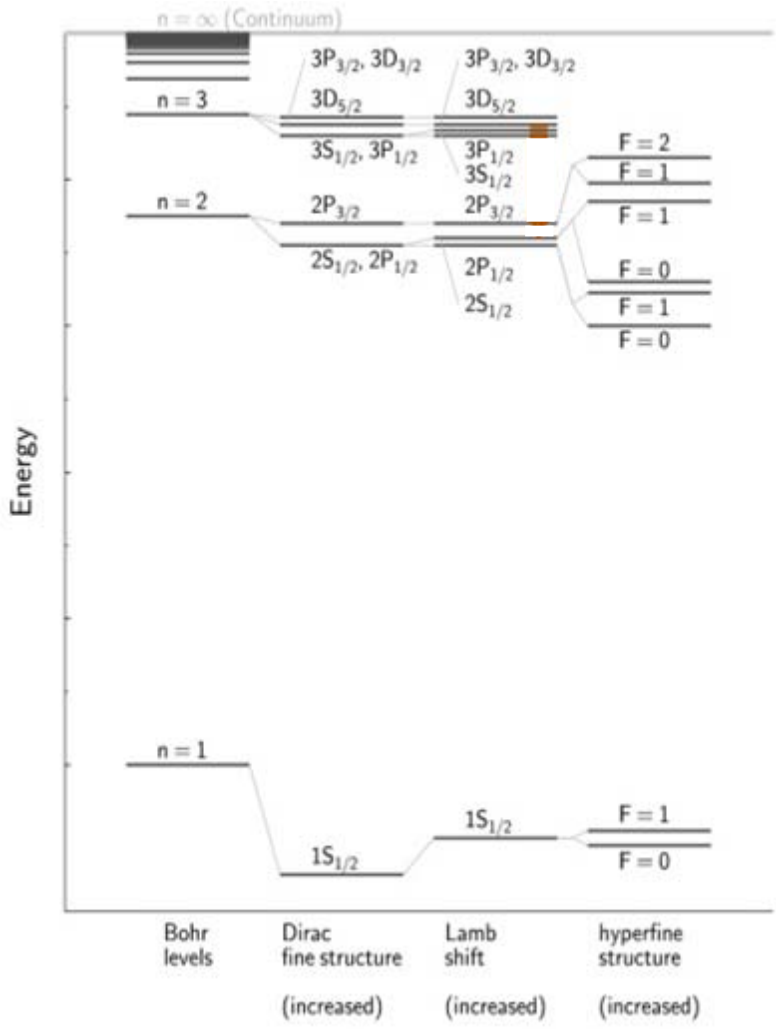


The approaches, made in this work, are under discussion.

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

J. K. Webb *et al.*, Phys. Rev. Lett. **107**, 191101 (2011)

Variation of constants: Atomic spectroscopy



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:

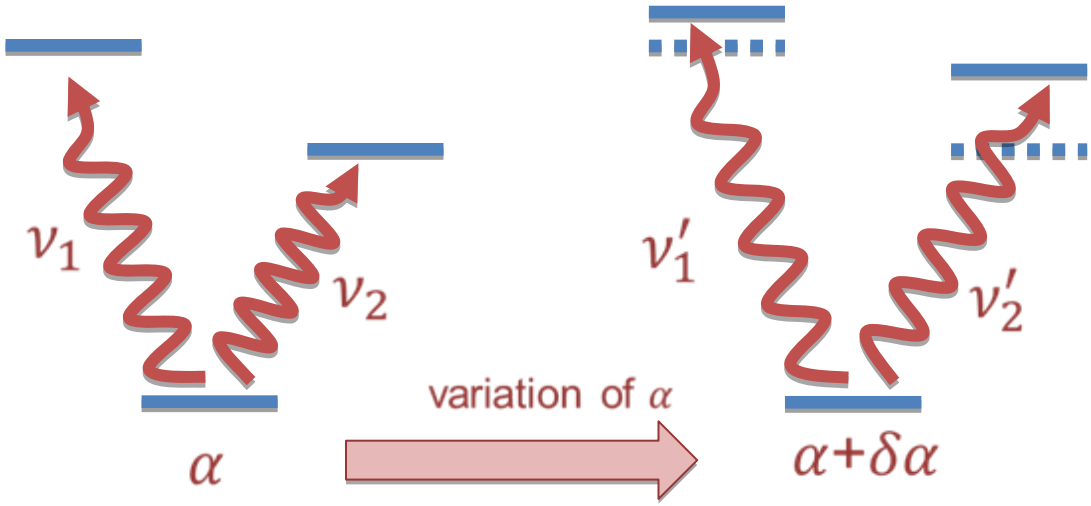
- Fine-structure levels:

$$\nu_{ik} = R_\infty \, c \, F_{fs}(\alpha, r_p, Z \ldots)$$

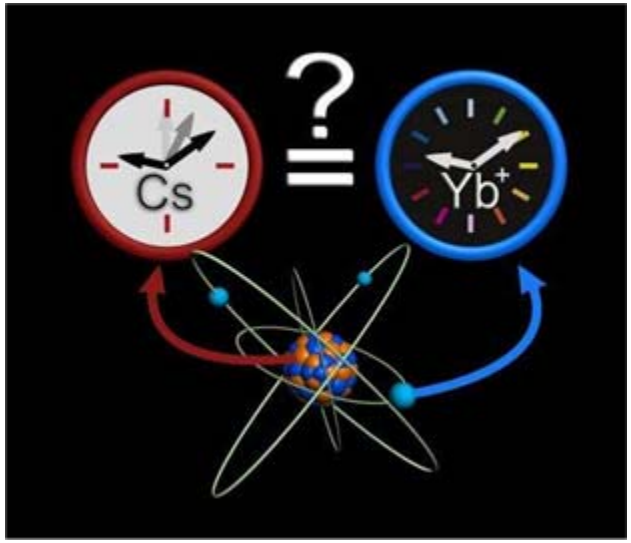
- Hyperfine-structure levels:

$$\nu_{ik} = R_\infty \, c \, \alpha^2 \frac{m_e}{m_p} g \, F_{hfs}(\alpha, r_p, Z \ldots)$$

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 α:

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

N. Huntemann *et al.*, Phys. Rev. Lett. 113, 210802 (2014)
R. M. Godun *et al.*, Phys. Rev. Lett. 113, 210801 (2014)

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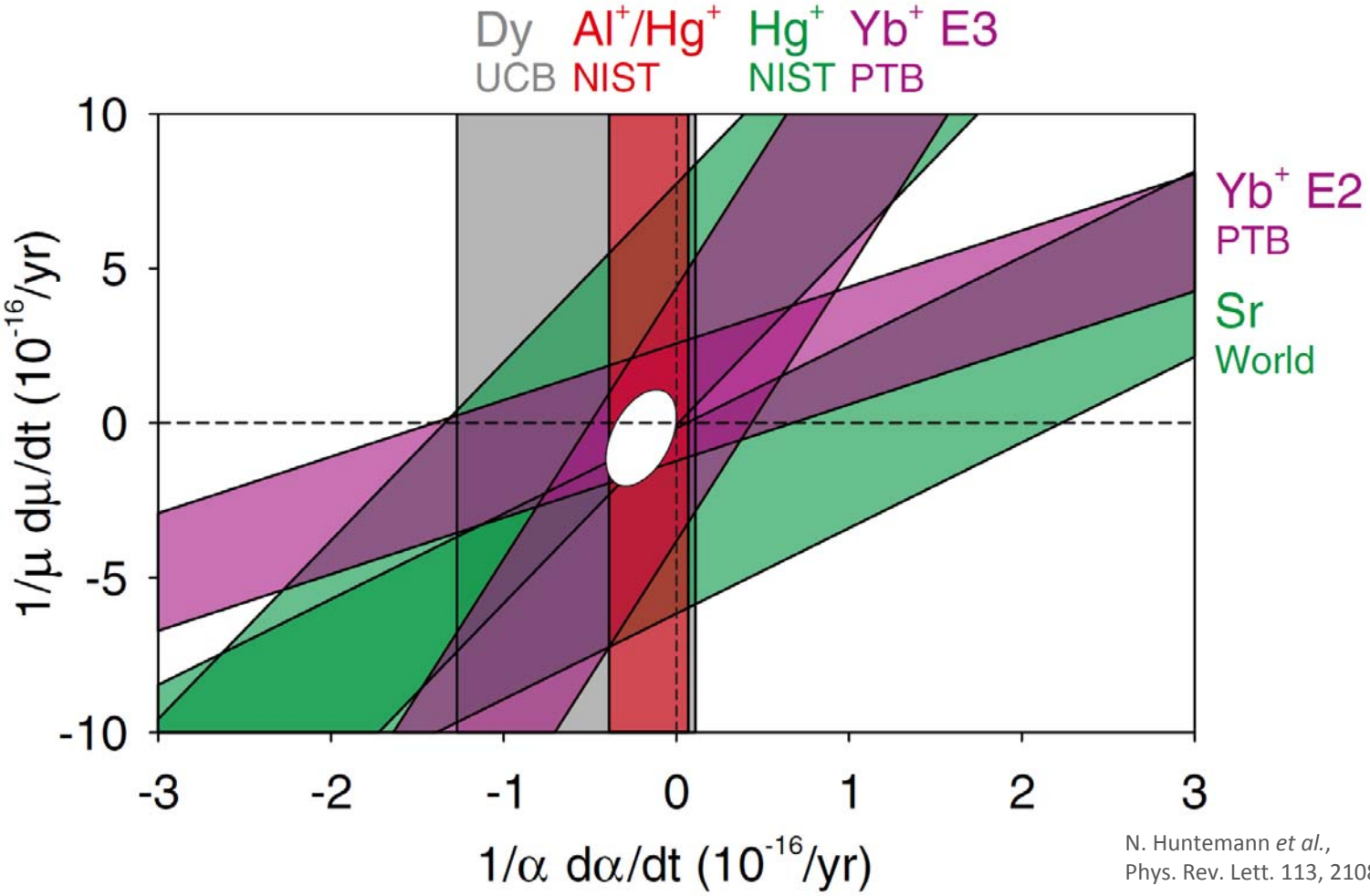
Constraints on temporal variations of constants



N. Huntemann *et al.*,
Phys. Rev. Lett. 113, 210802 (2014)

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

Atomic clocks based on hyperfine transitions

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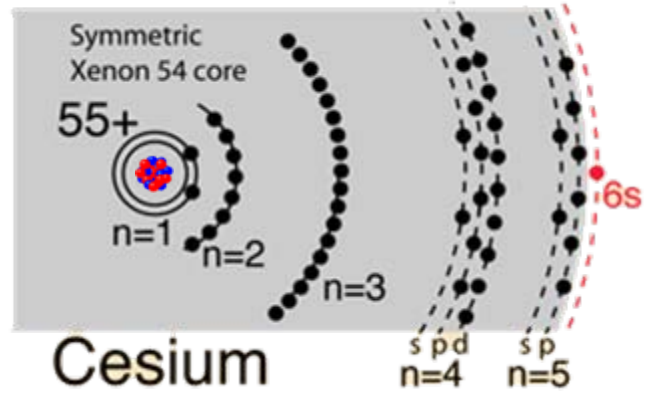

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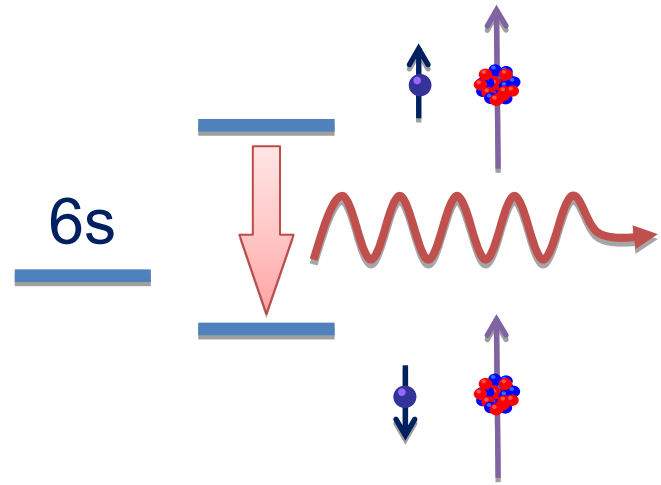

Can the accuracy of atomic clocks be further improved? And why do we need this?

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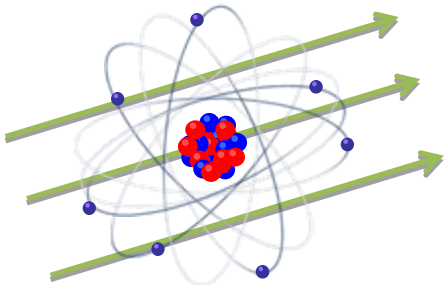
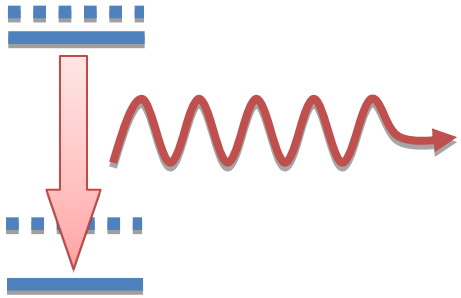
Atomic clocks: Improving accuracy





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

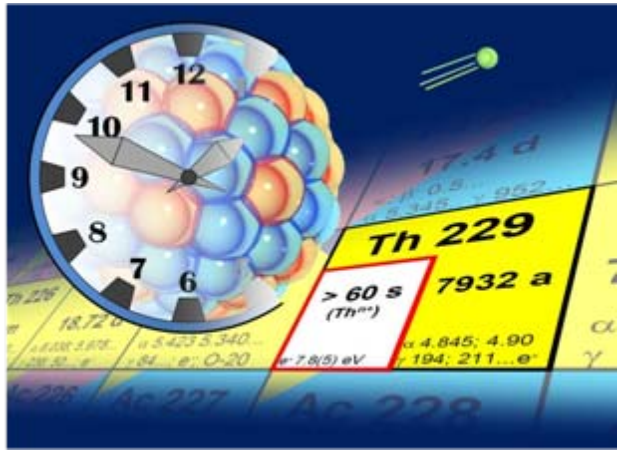
External fields lead to the shift of the energy levels (and, hence, of frequencies). For electric field, for example:

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

α_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.

Nuclear clocks: Even higher accuracy



Physica Scripta. Vol. 53, 296–299, 1996

Processes of the Nuclear Isomer ^{229m}Th(3/2$^+$, 3.5 ± 1.0 eV) Resonant Excitation by Optical Photons

E. V. Tkalya[1] and V. O. Varlamov

Laboratory of Theory of Radiation Processes, Nuclear Safety Institute of Russian Academy of Sciences, Bolshaya Tulskaya – 52, Moscow, 113191, Russia

and

V. V. Lomonosov and S. A. Nikulin

Institute of General Nuclear Physics, Russian Research Center the "Kurchatov Institute", Kurchatov Square 1, Moscow, 123182, Russia

Received June 29, 1995; accep

properties of solids by means of measuring the half-life time of isomeric levels and energies of emitted photons; development of a high stability nuclear source of light for metrology; creation of γ-laser in the optical range, and so on.

Since 1990's the ^{229}Th isotope attracts considerable attention as a promising candidate for the development of nuclear clocks.

What is so special with ^{229}Th?

EUROPHYSICS LETTERS 15 January 2003

Europhys. Lett., **61** (2), pp. 181–186 (2003)

Nuclear laser spectroscopy of the 3.5 eV transition in Th-229

E. Peik(*) and Chr. Tamm

Physikalisch-Technische Bundesanstalt - Bundesallee 100
38116 Braunschweig, Germany

(received 17 June 2002; accepted in final form 11 November 2002)

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What is (un)known about ^{229m}Th?

- Energy of the isomeric state:

NATURE | ARTICLE

日本語要約

Direct detection of the ^{229}Th nuclear clock transition

Lars von der Wense, Benedict Seiferle, Mustapha Laatiaoui, Jürgen B. Neumayr, Hans-Jörg Maier, Hans-Friedrich Wirth, Christoph Mokry, Jörg Runke, Klaus Eberhardt, Christoph E. Düllmann, Norbert G. Trautmann & Peter G. Thirolf

Affiliations | Contributions | Corresponding author

Nature **533**, 47–51 (05 May 2016) | doi:10.1038/nature17669
Received 16 December 2015 | Accepted 16 March 2016 | Published online 04 May 2016

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Abstract

Abstract • References • Author information • Extended data figures and tables

Today's most precise time and frequency measurements are performed with optical atomic clocks. However, it has been proposed that they could potentially be outperformed by a nuclear clock, which employs a nuclear transition instead of an atomic shell transition. There is only one known nuclear state that could serve as a nuclear clock using currently available technology, namely, the isomeric first excited state of ^{229}Th (denoted ^{229m}Th). Here we report the direct detection of this nuclear state, which is further confirmation of the existence of the isomer and lays the foundation for precise studies of its decay parameters. On the basis of this direct detection, the isomeric energy is constrained to between 6.3 and 18.3 electronvolts, and the

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

Experimental results

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

Article

Laser spectroscopic characterization of the nuclear-clock isomer ^{229m}Th

Johannes Thielking, Maxim V. Okhapkin, Przemysław Głowacki, David M. Meier, Lars von der Wense, Benedict Seiferle, Christoph E. Düllmann, Peter G. Thirolf & Ekkehard Peik

Nature **556**, 321–325 (2018)
doi:10.1038/s41586-018-0011-8
Download Citation

Atomic and molecular interactions with photons

Nuclear physics

Received: 15 September 2017
Accepted: 13 February 2018
Published: 18 April 2018

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^{229}Th is in the focus of research

Rostock 2019 – wissenschaftliches Programm

Bereiche | Tage | Auswahl | Suche | Aktualisierungen | Downloads | Hilfe

A: Fachverband Atomphysik

A 18: Precision Spectroscopy of atoms and ions V (Th 229)

Mittwoch, 13. März 2019, 14:00–16:00, S HS 2 Physik

Auswahlstatus für diese Sitzung: nicht ausgewählt

⊕	14:00	A 18.1	Excitation of $^{229}Th^{2+}$ with a two-photon electron transition — •Robert A. Müller, Andrey V. Volotka, Stephan Fritzsche, and Andrey Surzhykov
⊕	14:15	A 18.2	Excitation of ^{229}Th in VUV-transparent crystals — •Brenden Nickerson and Adriana Pálffy
⊕	14:30	A 18.3	Electronic level structure investigations in Th^+ for the excitation of the nuclear isomer — •David-Marcel Meier, Johannes Thielking, Gregor Zitzer, Maksim Okhapkin, Robert Müller, Andrey Surzhykov, and Ekkehard Peik
⊕	14:45	A 18.4	Towards a ^{229m}Th energy determination with 40 µeV accuracy — •L. von der Wense, B. Seiferle, Ch. Schneider, J. Jeet, I. Amersdorffer, N. Arlt, F. Zacherl, R. Haas, D. Renisch, Pa. Mosel, Ph. Mosel, M. Kovacev, U. Morgner, Ch.E. Düllmann, E.R. Hudson, and P.G. Thirolf
⊕	15:00	A 18.5	Gamma spectroscopy to measure the ^{229}Th isomer energy using a 2-dimensional array of metallic magnetic microcalorimeters — •Jeschua Geist, Daniel Hengstler, Christian Schötz, Sebastian Kempf, Loredana Gastaldo, Andreas Fleischmann, Christian Enss, Georgy A. Kazakov, Simon Stellmer, and Thorsten Schumm
⊕	15:15	A 18.6	Measurement of Fundamental Constants by Spectroscopy of the Molecular Hydrogen Ion — Soroosh Alighanbari, Gouri Giri, Michael Hansen, Florin Constantin, Vladimir Korobov, and •Stephan Schiller
⊕	15:30	A 18.7	Hauptvortrag: Towards a precise energy determination of the ^{229}Th nuclear isomer — •Benedict Seiferle, Lars v.d. Wense, Ines Amersorffer, and Peter G. Thirolf

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.

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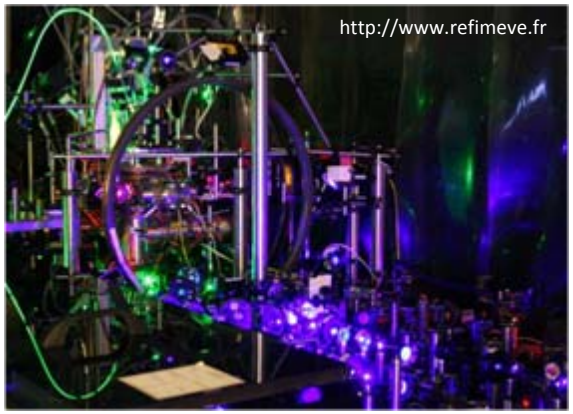
Nuclear properties and atomic spectroscopy



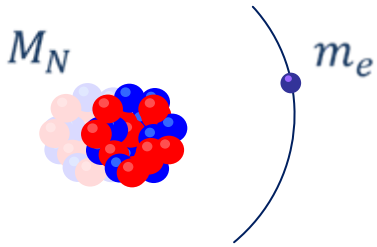
During the recent years many advances have been done in the high-precision atomic spectroscopy.

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.

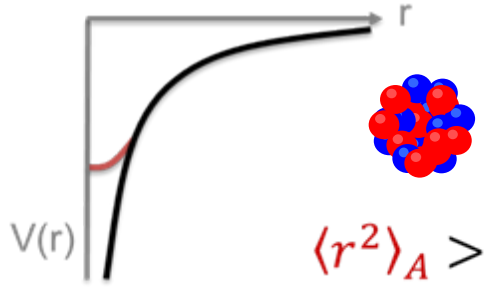
Mass shift

due to finite nuclear mass



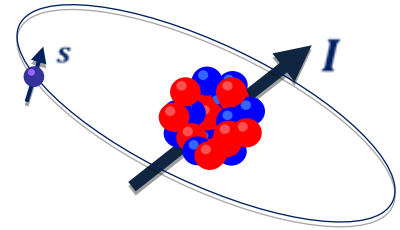
Field shift

due to non-zero charge radii



Hyperfine effects

due to nuclear momenta



Isoptope shift

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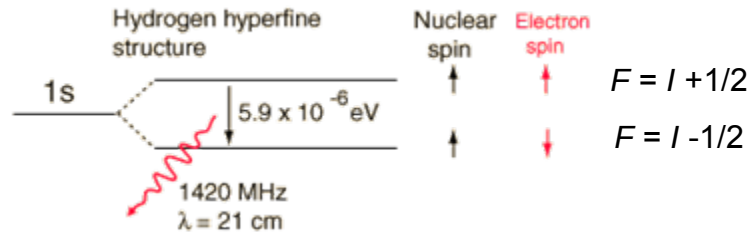
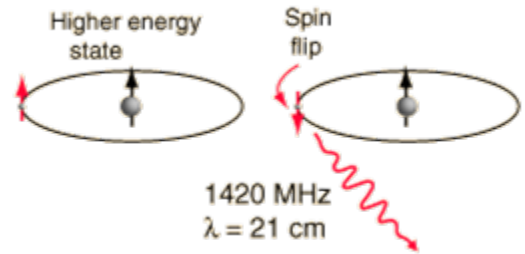
Hyperfine splitting in hydrogen atom

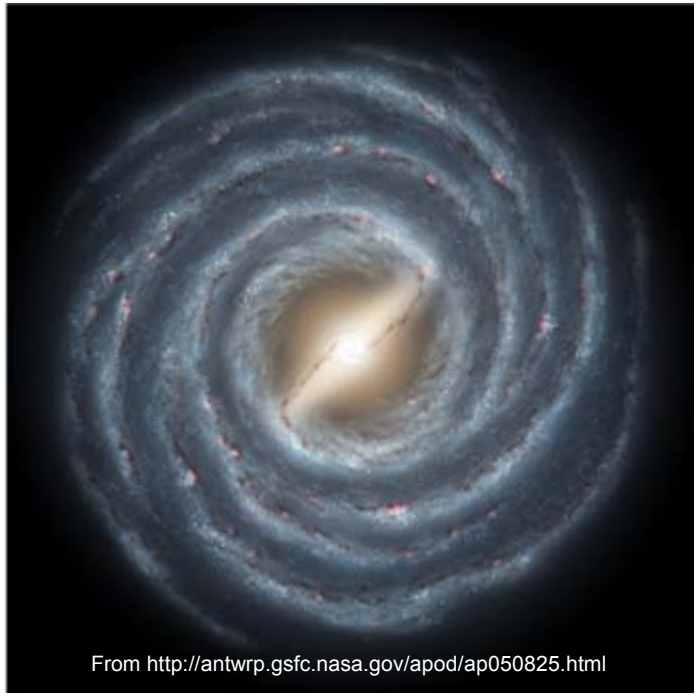
For the case of ground $1s_{1/2}$ state (j=1/2) of hydrogen, HF interaction results in splitting of energy level into two levels.

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





From: http://hyperphysics.phy-astr.gsu.edu



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

Analysis of the properties of galaxies.

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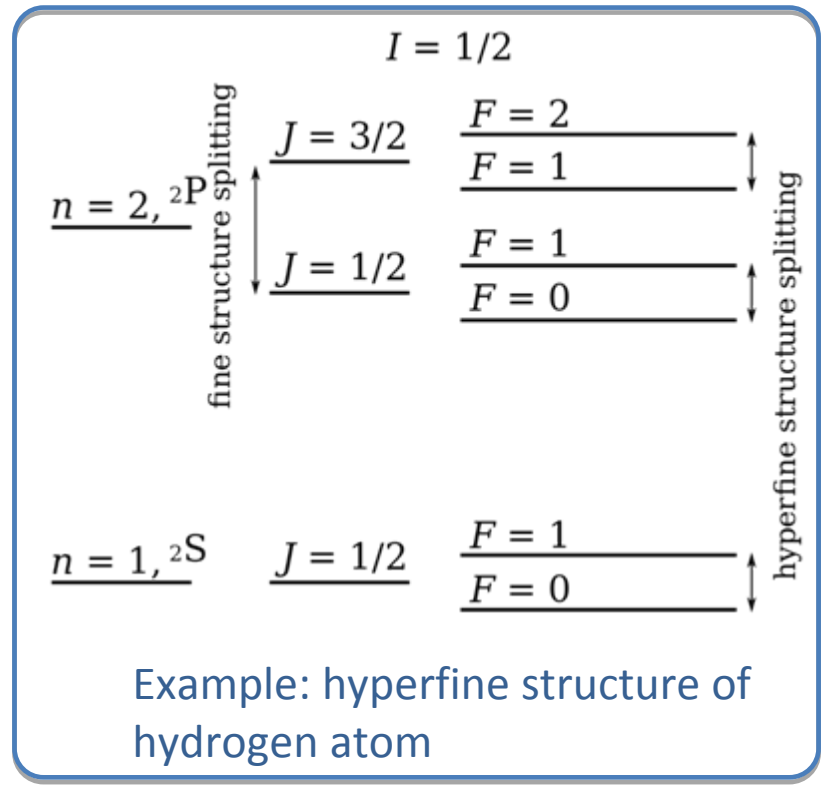
Hyperfine structure: Reminder

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

$$\Delta E_{M1} = \frac{1}{2} A\, C$$

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

$$C = F(F+1) - J(J+1) - I(I+1)$$



Example: hyperfine structure of hydrogen atom

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.

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Hyperfine constants A and B

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

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

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

The A and B constants are proportional to the nuclear magnetic dipole μ_I and electric quadrupole Q moments.

From atomic physics side the reduced matrix elements $\langle \gamma J \| \boldsymbol{T}^k \| \gamma J \rangle$ of the hyperfine interaction operators enter into A and B.

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Hyperfine constants A and B

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

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

„atomic parts“

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

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 $\langle \gamma J^p \| \boldsymbol{T}^2 \| \gamma J^p \rangle$!

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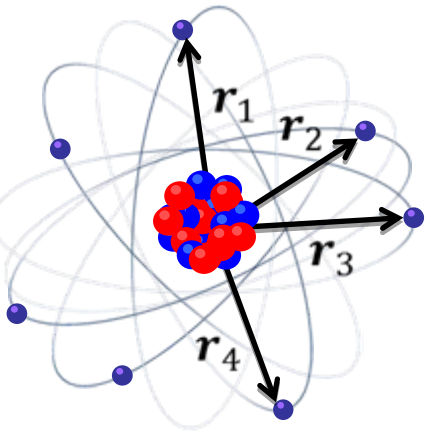
Many-electron ions: Theory



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

$$E_n\ ,\ \Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \ldots, \boldsymbol{r}_N)$$

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

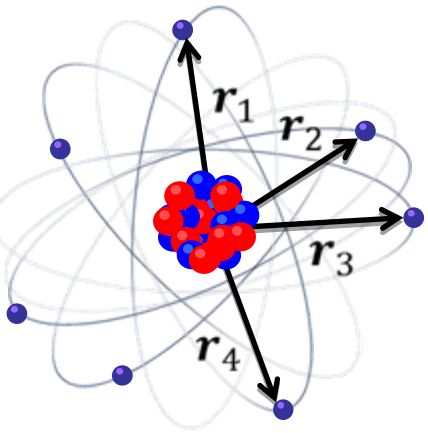
For heavy ions like Pb the starting point is the Dirac equation:

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

Hamiltonians of individual electrons

e-e interactions

Theory of many-electron systems



The Hamiltonian of many-electron atoms reads as:

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

Hamiltonians of individual electrons

e-e interactions

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

$$\Psi_{J^p}(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \ldots, \boldsymbol{r}_N) = \sum_s d_s \begin{vmatrix} \varphi_1(r_1) & \cdots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \cdots & \varphi_N(r_N) \end{vmatrix}$$

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

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

Which potential to use here?

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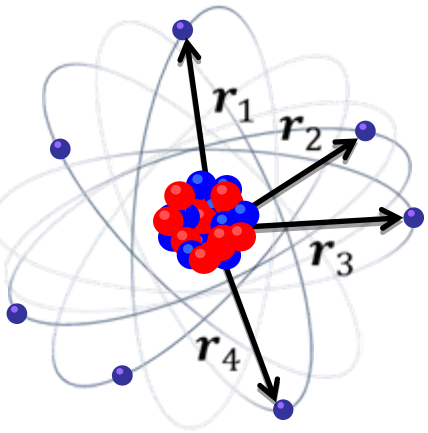
Theory of many-electron systems



We can construct the many-electron wavefunctions as:

$$\Psi_{J^p}(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \ldots, \boldsymbol{r}_N) = \sum_s d_s \begin{vmatrix} \varphi_1(r_1) & \ldots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \ldots & \varphi_N(r_N) \end{vmatrix}$$

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

$$\left(-i\hbar c\boldsymbol{\alpha}_i \cdot \hat{\nabla}_i + V_{eff}(\boldsymbol{r}_i) + m_e c^2 \gamma^0\right) \varphi_i(r_i) = \varepsilon_i \varphi_i(r_i)$$

Just Coulomb electron-nucleus interaction?

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

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

$$V_{\mathrm{DF}}(\boldsymbol{r}_1)\,\psi(\boldsymbol{r}_1) = \sum_c \int d\boldsymbol{r}_2\, \psi_c^+(\boldsymbol{r}_2)\, \frac{\alpha}{r_{12}} \times [\psi_c(\boldsymbol{r}_2)\,\psi(\boldsymbol{r}_1) - \psi_c(\boldsymbol{r}_1)\,\psi(\boldsymbol{r}_2)]$$

But… We need to account for the rest of e-e interactions!

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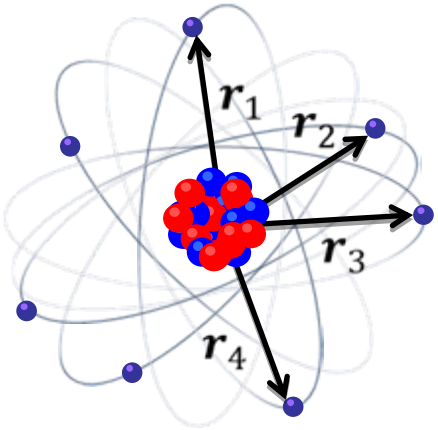
Configuration interaction method

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

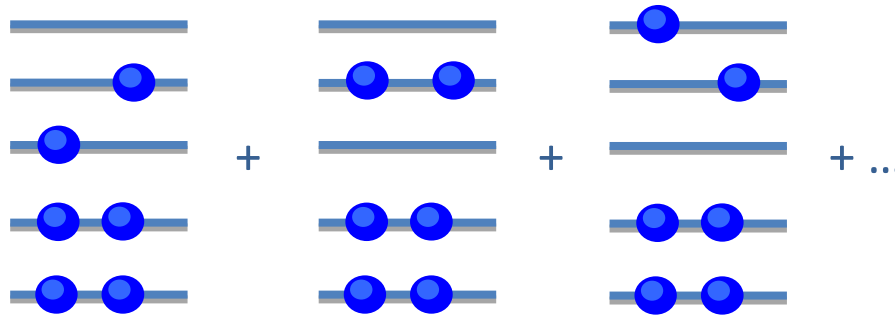


$$\Psi_{J^P}(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \ldots, \boldsymbol{r}_N) = \sum_r c_r \sum_s d_s \begin{vmatrix} \varphi_1(r_1) & \ldots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \ldots & \varphi_N(r_N) \end{vmatrix}$$

Summation over configurations

State of particular symmetry $|\gamma_r\, J^P M\rangle$

Ground state =



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

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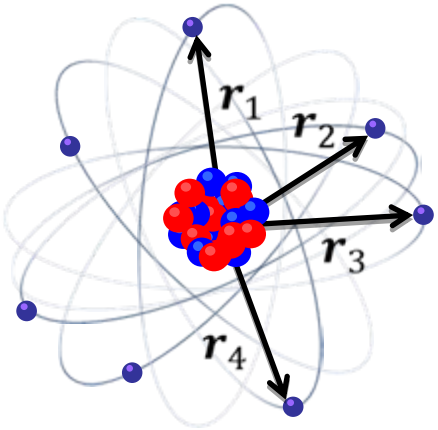
Configuration interaction method

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



$$\Psi_{J^P}(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_3, \dots, \boldsymbol{r}_N) = \sum_r c_r \sum_s d_s \begin{vmatrix} \varphi_1(r_1) & \dots & \varphi_N(r_1) \\ \vdots & \ddots & \vdots \\ \varphi_1(r_N) & \dots & \varphi_N(r_N) \end{vmatrix}$$

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

$$det\left\{\langle \gamma_r J^P M | \sum_i \hat{h}_i + \sum_{i<j} V(|\boldsymbol{r}_i - \boldsymbol{r}_j|) | \gamma_s J^P M \rangle - E_r \delta_{rs}\right\} = 0$$

In present calculations the $V(|\boldsymbol{r}_i - \boldsymbol{r}_j|)$ usually accounts for (frequency-dependent) Breit interaction corrections.

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Hyperfine constants A and B

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

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

„atomic parts“

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

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:

$$\frac{A_{exp}}{A_{theo}/\mu_I} = \mu_I \qquad \frac{B_{exp}}{B_{theo}/Q} = Q$$

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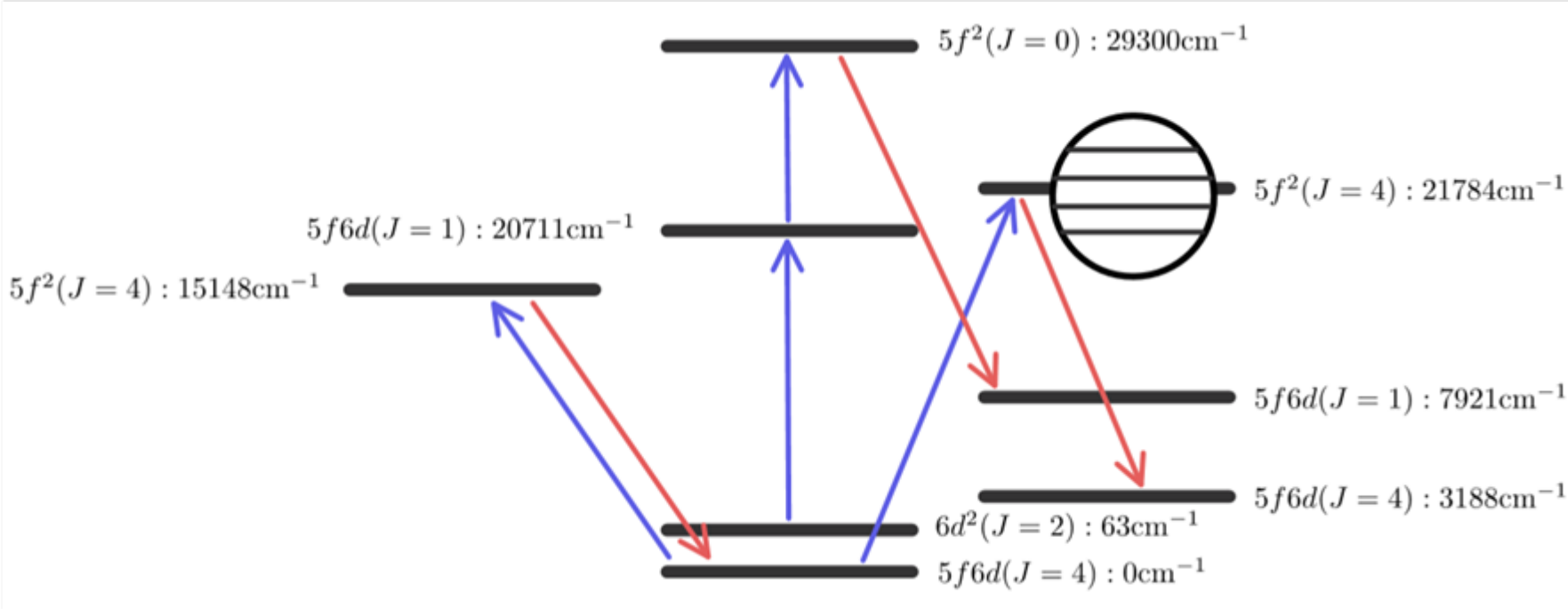
Hyperfine spectra of $^{229}Th^{2+}$ and $^{229m}Th^{2+}$



Experimental and theoretical studies have been performed for Th^{2+} ion whose ground-state configuration is 5f 6d („Radon core + 2 electrons“).

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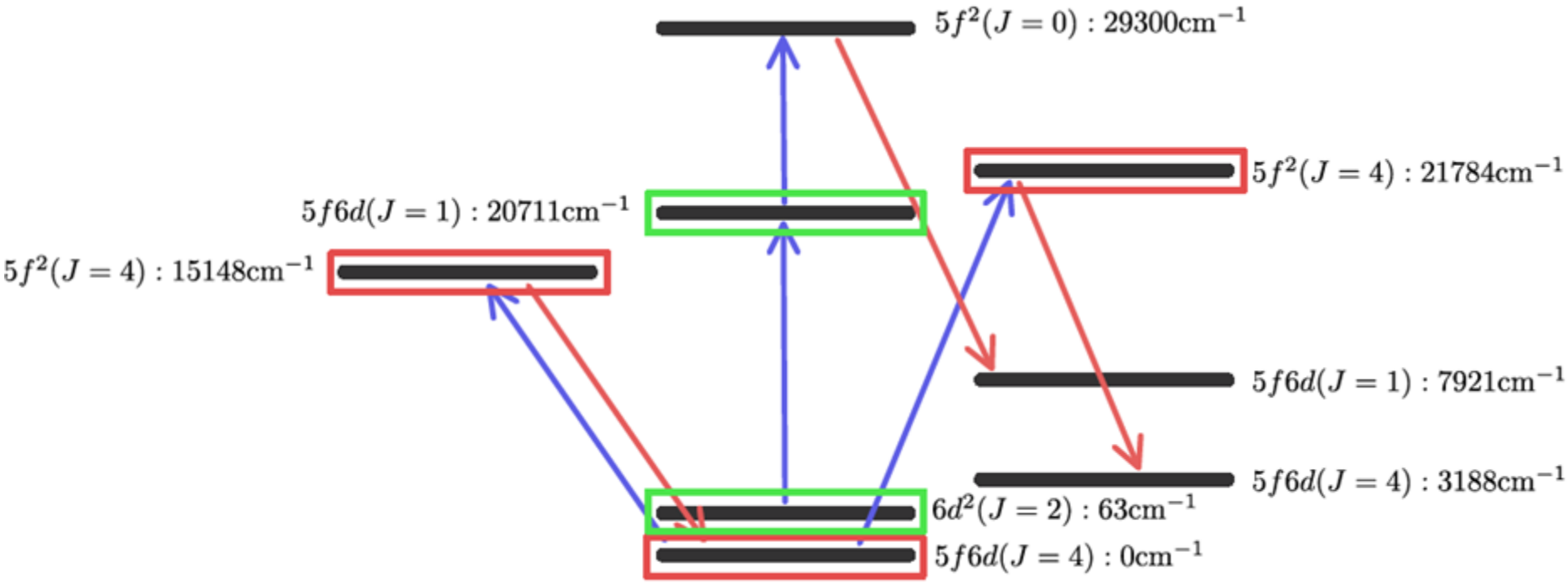
Hyperfine spectra of $^{229}Th^{2+}$ and $^{229m}Th^{2+}$



Not all levels have been analyzed in the experiment.

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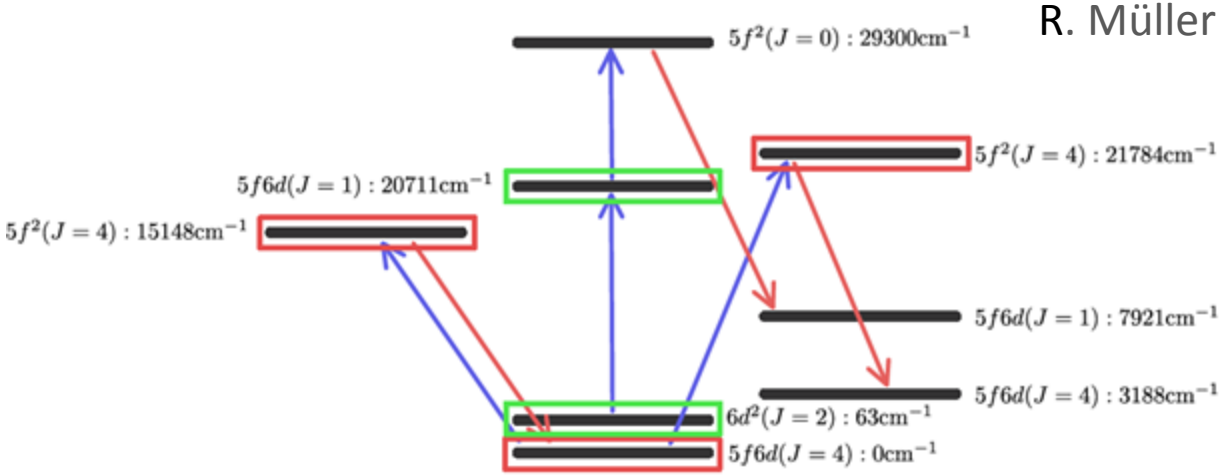
Hyperfine structure of $^{229}Th^{2+}$

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

energy level			A[MHz]			B[MHz]		
configuration	J^{Π}	energy $[cm]^{-1}$	CI+MBPT	MCDF	exp.	CI+MBPT	MCDF	exp.
$[Rn]+5f6d$	4^-	0	64(17)	81(4)	–	3287(630)	3008(260)	–
$[Rn]+6d^2$	2^+	63	143(47)	162(8)	151(8)	68(23)	71(7)	73(27)
$[Rn]+5f^2$	4^+	15148	38(3)	72(3)	–	1221(390)	1910(200)	–
$[Rn]+5f6d$	1^-	20711	109(36)	90(4)	88(5)	839(220)	689(110)	901(18)
$[Rn]+5f^2$	4^+	21784	8(36)	26(2)	–	65(21)	39(45)	–

R. Müller *et al*, PRA 98 (2018) 020503(R)



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Hyperfine structure of $^{229m}Th^{2+}$

Extraction of the nuclear magnetic dipole moment

energy level			A/μ_{iso} [MHz/μ_N]		A [MHz]
configuration	J^Π	energy [cm^{-1}]	CI+MBPT	MCDF	exp.
$[\mathrm{Rn}]+6d^2$	2^+	63	660	750	−263(29)
$[\mathrm{Rn}]+5f6d$	1^-	20711	506	419	−151(22)

$$\frac{A_{exp}}{A_{theo}/\mu_I} = \mu_I$$

Extraction of the nuclear electric quadrupole moment

energy level			B/Q_{iso} [MHz/eb]		B [MHz]
configuration	J^Π	energy [cm^{-1}]	CI+MBPT	MCDF	exp.
$[\mathrm{Rn}]+6d^2$	2^+	63	22	23	53(65)
$[\mathrm{Rn}]+5f6d$	1^-	20711	270	229	498(15)

$$\frac{B_{exp}}{B_{theo}/Q} = Q$$

R. Müller *et al*, PRA 98 (2018) 020503(R)

Experimental results

$\mu_{iso} = -0.37(6)\mu_N$,

$Q_{iso} = 1.74(6)$ |e| b

✓

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Summary

- We have performed first theoretical calculations for the hyperfine constants of $^{229}Th^{2+}$ and $^{229m}Th^{2+}$.

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