Towards an Optical Nuclear Clock

Generalized Ramsey Interrogation Schemes

Ekkehard Peik

Physikalisch-Technische Bundesanstalt
Braunschweig, Germany

ekkehard.peik@ptb.de
www.ptb.de/time/

Physikalisch-Technische Bundesanstalt
Braunschweig und Berlin

Distinguished Lecturer 2018/19

Advancing Technology for Humanity

Nuclear Clock:

Based on an oscillator that is frequency-stabilized to a nuclear (γ-ray) transition

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 $\approx 10^0$ (ion) or $\approx 10^4$ (lattice)).

Higher frequency: → higher stability. EUV or even X-ray transitions may be used when suitable radiation sources become available.

Special session of the National Academy of Science of Argentina on 16.04.1925

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: $$\hbar\omega \approx 2\,\mathrm{eV}$$

Atomic nucleus: bound nucleon $$\Delta x \approx 5\cdot 10^{-15}\ \mathrm{m}$$

with $$\Delta x \cdot \Delta p = \hbar \;\rightarrow\; \frac{\hbar^2}{2(\Delta \mathrm{x})^2 m_p} = 0{,}83\,\mathrm{MeV}$$

(proton rest mass: 938 MeV)

Atomic shell: bound electron $$\Delta x \approx 10^{-10}\ \mathrm{m}$$

with $$\Delta x \cdot \Delta p = \hbar \;\rightarrow\; \frac{\hbar^2}{2(\Delta \mathrm{x})^2 m_e} = 3{,}8\,\mathrm{eV}$$

(electron rest mass: 0.51 MeV)

Scales of electric field strengths:

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

shell: $r = 10^{-10}$ m $\qquad E_H = 1{,}4 \cdot 10^{11}$ V/m

nucleus: $r = 5 \cdot 10^{-15}$ m $\qquad E_K = 5{,}8 \cdot 10^{19}$ V/m

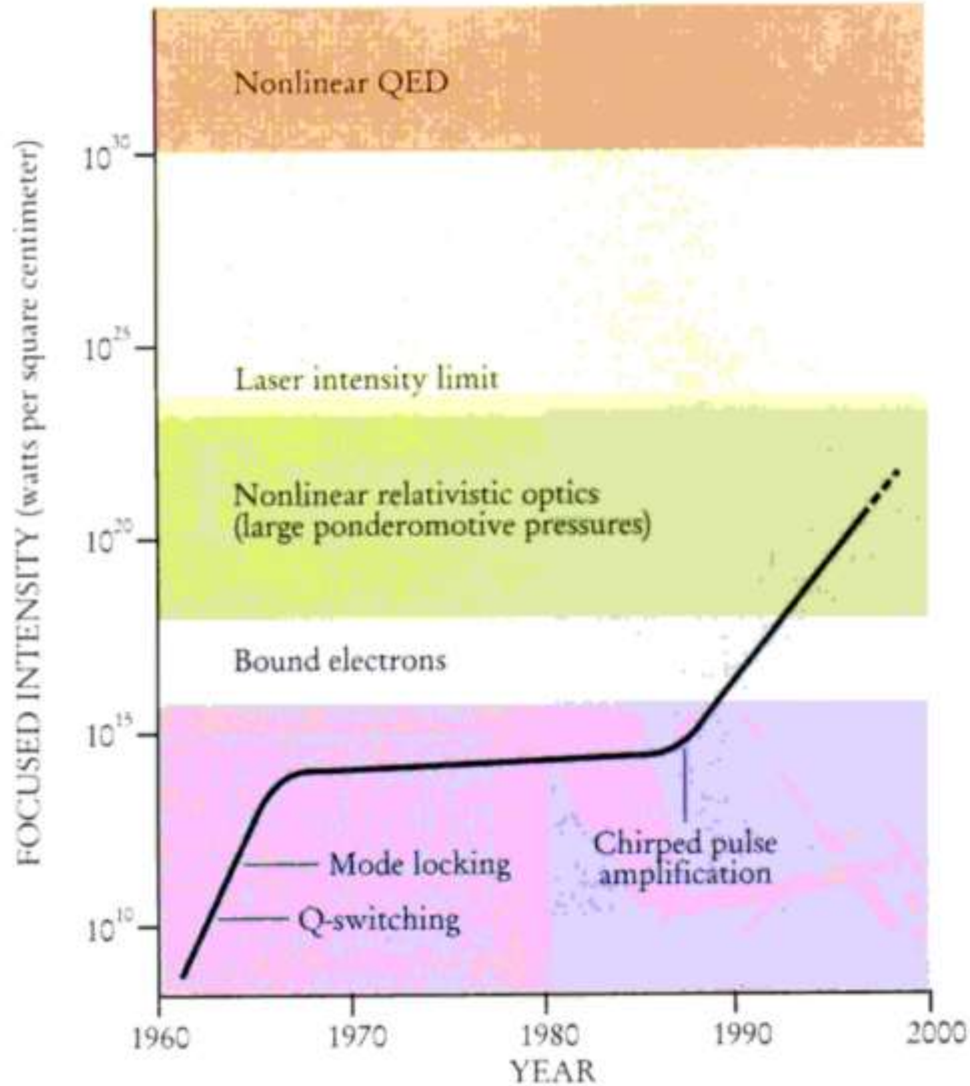
Field strength of an electromagnetic wave: related to intensity

$$I = \frac{1}{2}\varepsilon_0 c E_L^2$$

$$E_L = E_H \quad \rightarrow \quad I = 2{,}5 \cdot 10^{15} \text{ W/cm}^2$$

$$E_L = E_K \quad \rightarrow \quad I = 4{,}5 \cdot 10^{32} \text{ W/cm}^2$$

Intensity in the focus
of a short-pulse laser:



Limit intensity:

1/(min. pulse duration)

Phot. energy

1/(min. focus area)

$$I_{\max} \approx N_{Ph} \cdot h\nu \cdot \nu \cdot \frac{\nu^2}{c^2}$$

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

$$I_{\max} \approx 10^{24}\ \mathrm{W/cm^2}$$

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

G. Mourou, C. Barry, M. Perry, Phys. Today **51**(1), 22 (1998)

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 :

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

$$\tau_E(1) \approx 100\,\mathrm{s} \qquad \text{bei } 1\,\mathrm{eV}$$

Long-lived excited states: Isomers

e.g. Ta-180: naturally occuring isomer,

decays via E8 radiation (l=8) at 75.3 keV, $(r/\lambda)^{16} \approx 10^{-58}$

half-life > 10^{15} yr

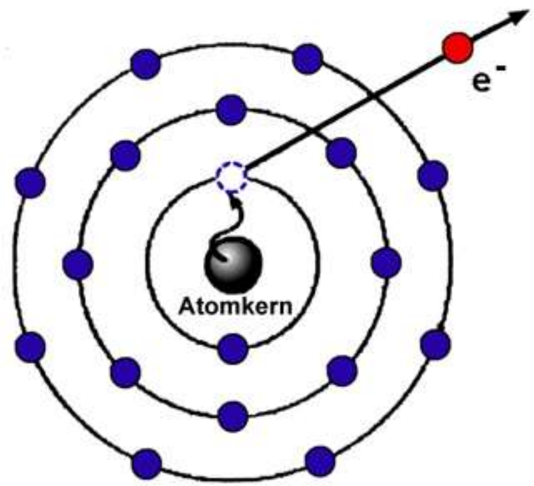
0.012% abundance in natural tantalum

Using the electron shell as an antenna:

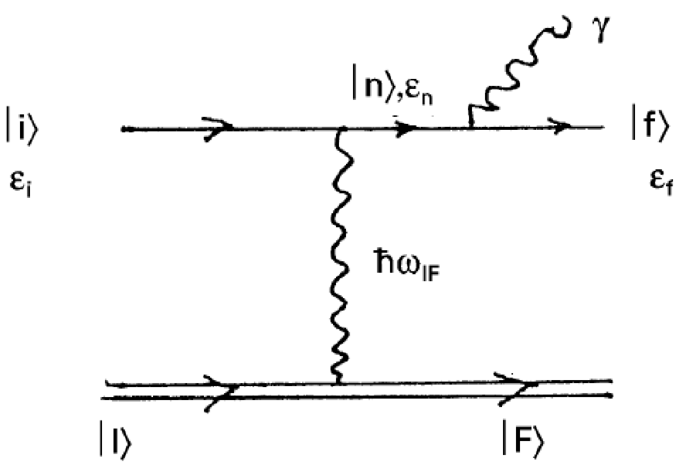
Would increase r by about 5 orders of magnitude.

How to obtain a coupling between nucleus and electron?

Low-energy γ-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 γ-decay
V. A. Krutov and V. N. Fomenko, Ann. Physik **476**, 292 (1968)

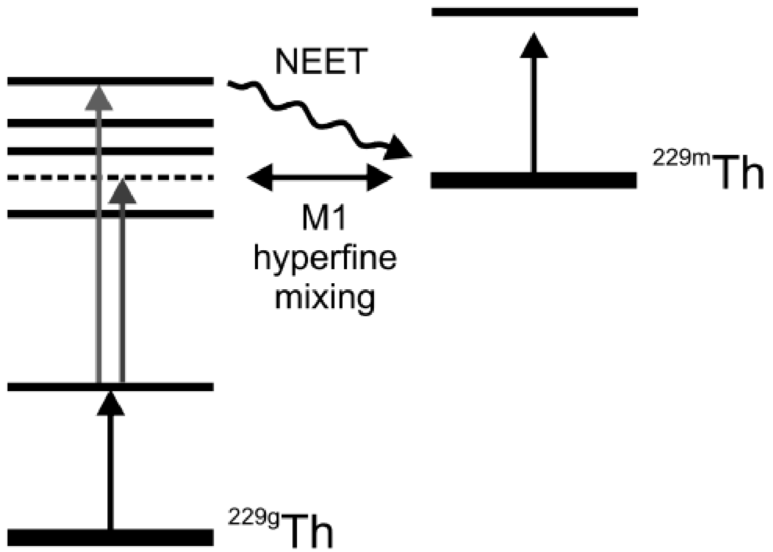


Photon is radiated from the electron shell.
The process can be strongly enhanced in case of a resonance between nuclear and electronic transition frequencies.

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

  see reviews: S. Matinyan, Phys. Rep. **298**, 199 (1998),
  E. V. Tkalya, Phys. Uspekhi **46,** 315 (2003)

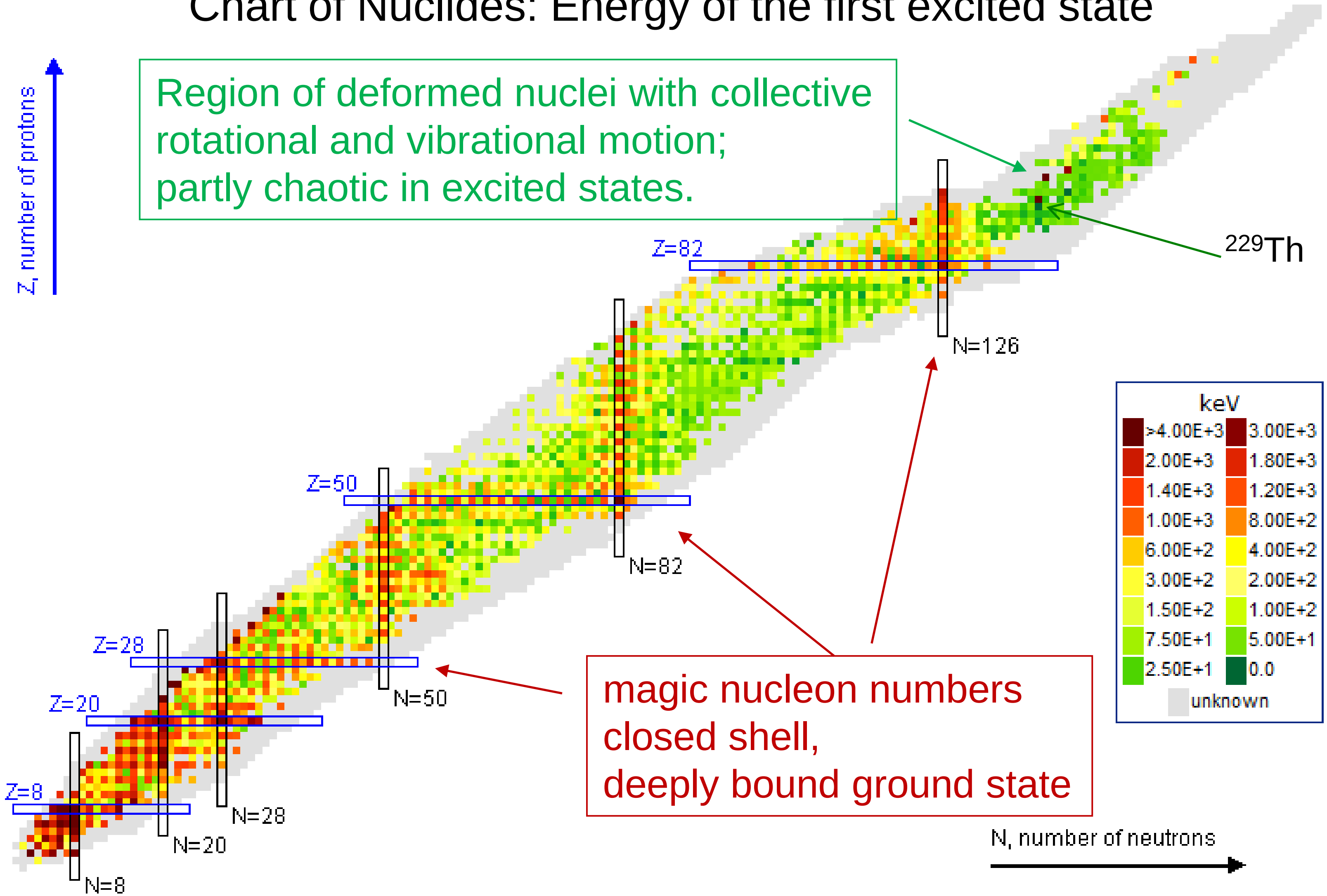


Advantages:

- 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



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

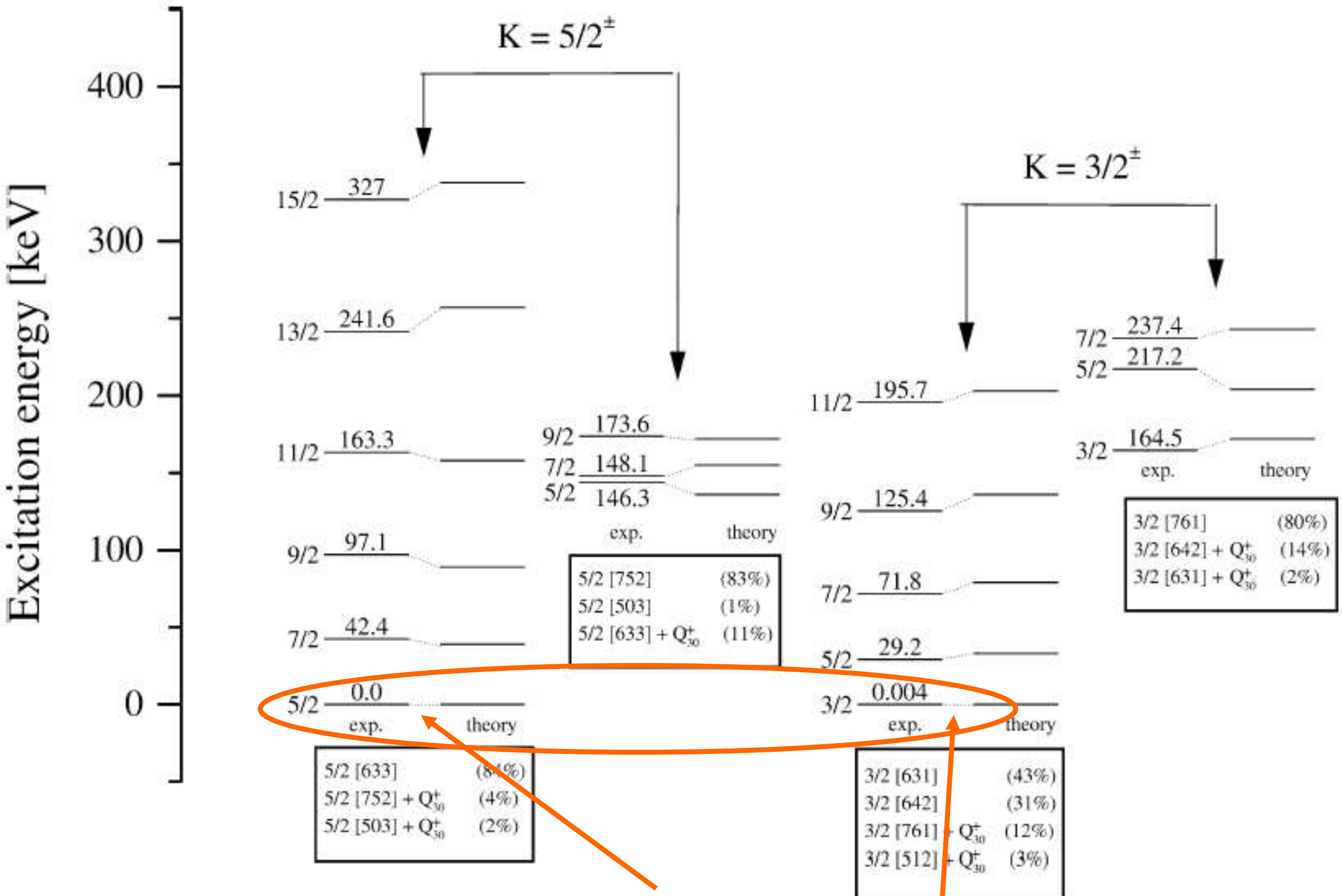
From: National Nuclear Data Center, Brookhaven National Laboratory

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

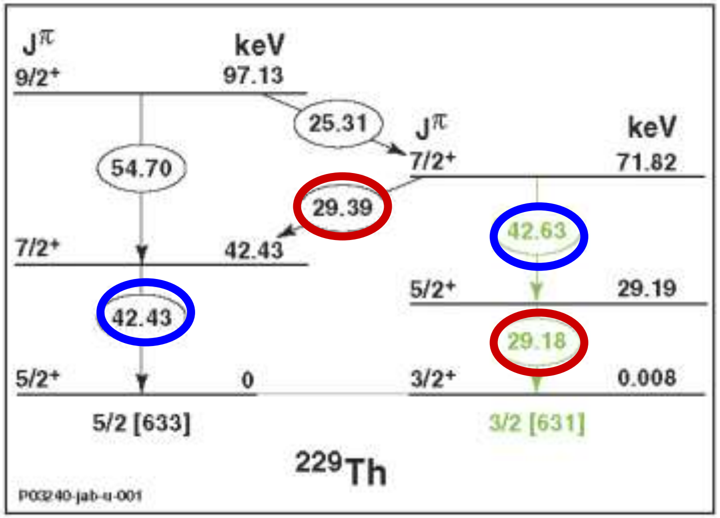
K. Gulda et al. / Nuclear Physics A 703 (2002) 45–69

The nuclear structure of ^{229}Th



Two close-lying band-heads: ground state and isomer

[Measurement of the energy of the Th-229 isomer
B. R. Beck et al. (LLNL), Phys. Rev. Lett. **98**, 142501 (2007)]

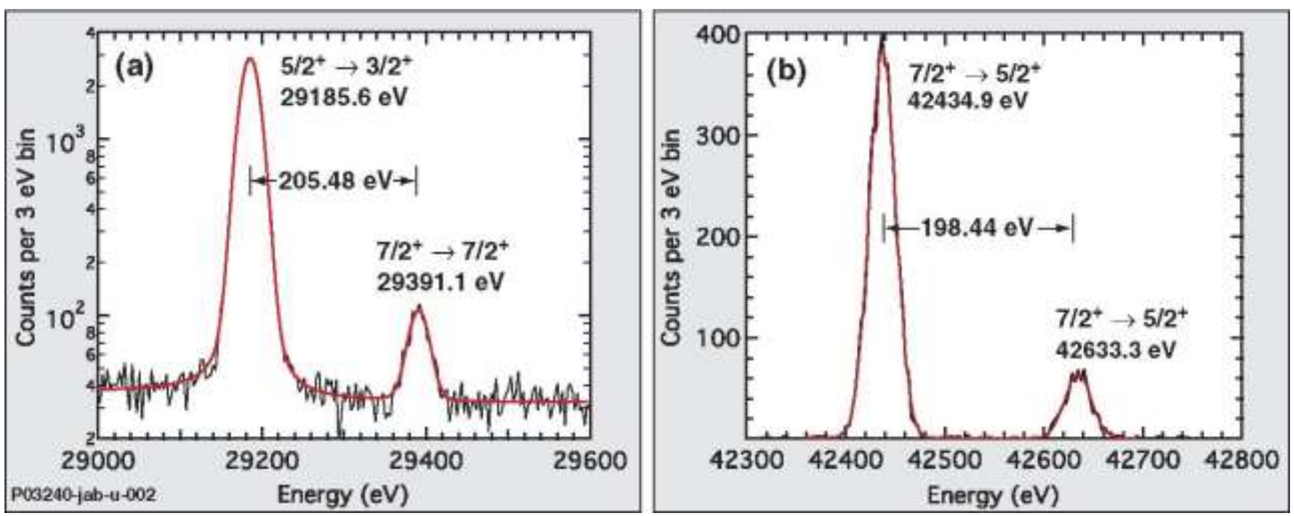


γ-spectroscopy of two decay cascades from the 71.82-keV-level

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

29 KeV lines

42 KeV lines



Ground state → isomer: transition in the vacuum-ultraviolet at about 160 nm wavelength

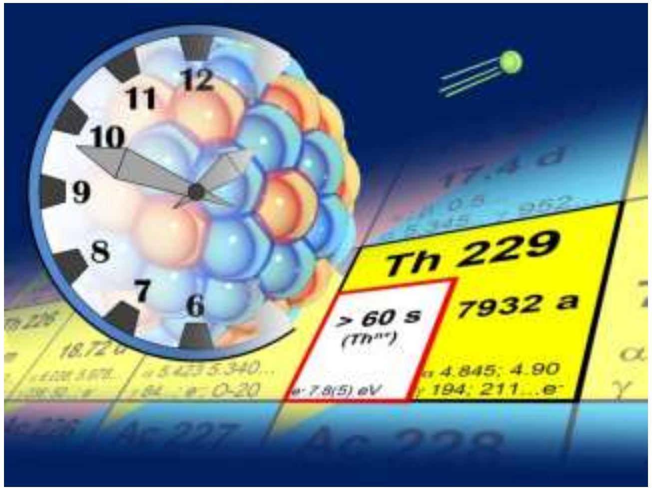
Low-energy nuclear physics with ^{229}Th

• modified decay rates in different electronic environments, electronic bridge processes. Tkalya, Karpeshin, et al.

• 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.

- γ-ray laser. Tkalya
- …



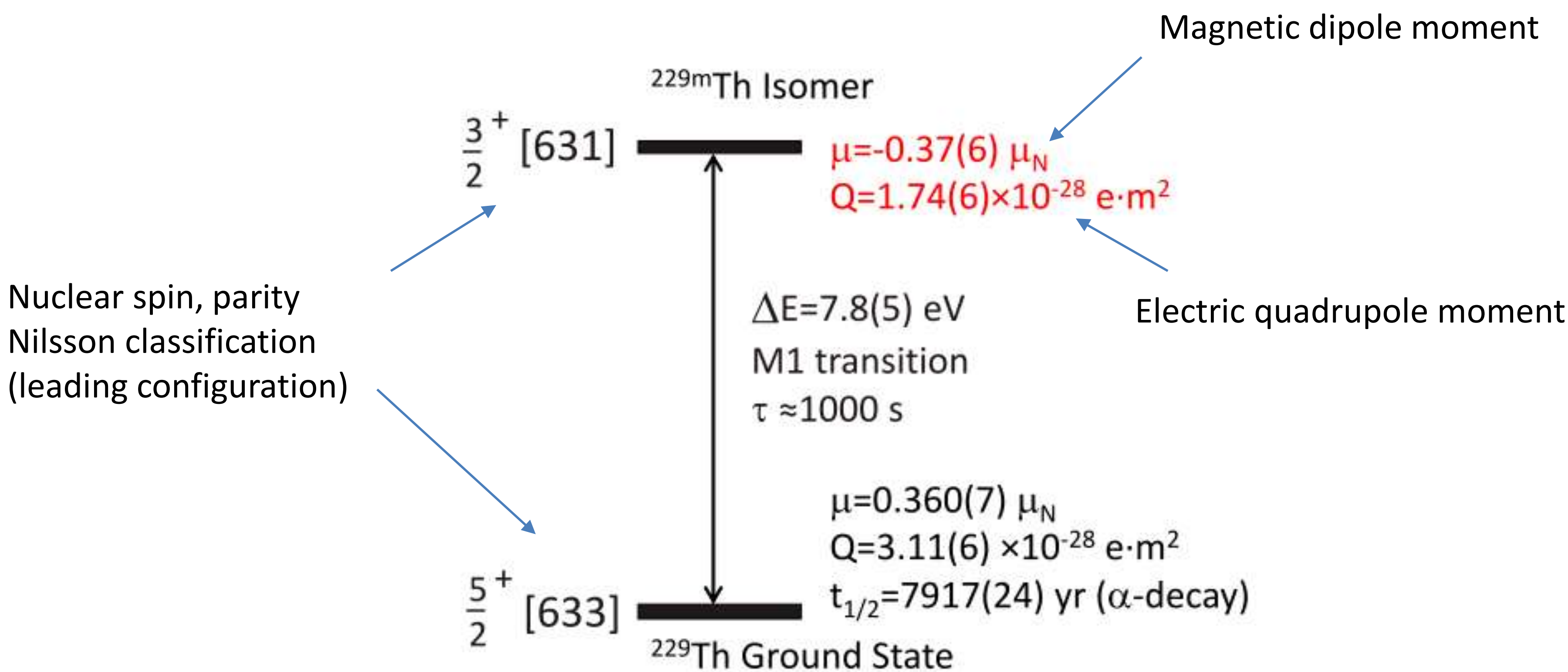
GSI

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)

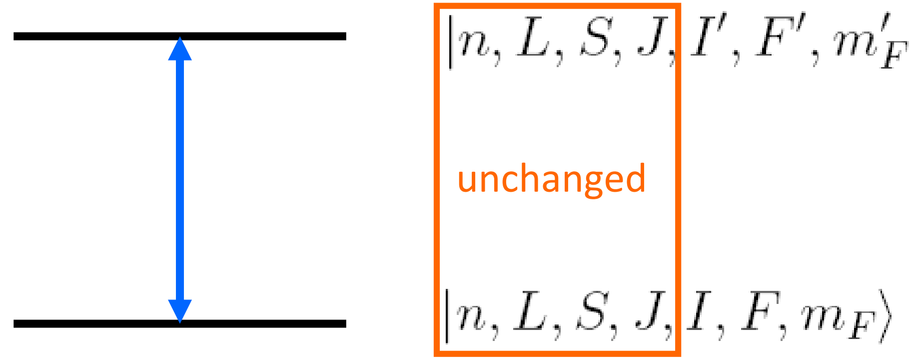
Consider electric and magnetic hyperfine coupling.

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:
E. Peik, Chr. Tamm, Europhys. Lett. **61**, 181 (2003)



LS coupled eigenstates of the
electronic + nuclear system

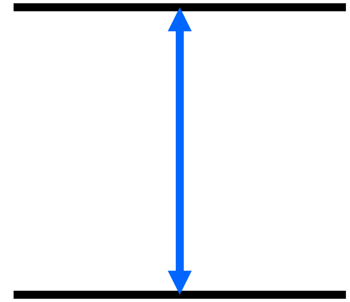
Frequency shifts that only depend on |n,L,S,J> are common in both levels and do not change the transition frequency.

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
analogy with Cs for m_F=0-0: $\approx 10^{-19}$ at room temperature

LS coupled eigenstates of the electronic + nuclear system



$$|n, L, S, J, I', F', m'_F\rangle$$

unchanged

$$|n, L, S, J, I, F, m_F\rangle$$

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

$$\begin{aligned} h\Delta\nu_S(\gamma, J, F, m, \boldsymbol{E}) &= -\left(2\alpha_S(\gamma, J) + \alpha_T(\gamma, J, F)\, g(F, m, \beta)\right) \frac{|\boldsymbol{E}|^2}{4} \\ g(F, m, \beta) &= \frac{3m^2 - F(F+1)}{F(2F-1)} \left(3\cos^2\beta - 1\right) \ , \end{aligned}$$

β: 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 β dependence and vanish for J<1 or F<1

Linear Zeeman shift: use component $m_F=0-0$

Quadratic Zeeman shift:

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

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+} (mF≠0, 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 → m‘) and (-m → -m‘) components)

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

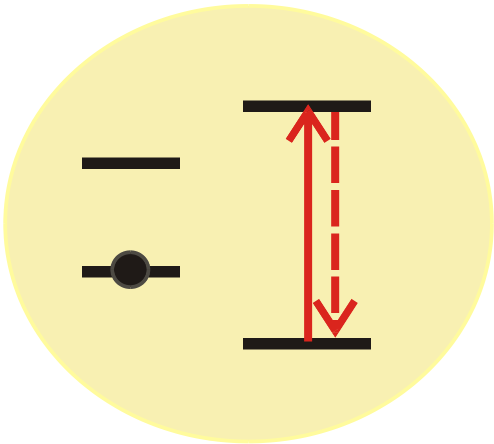
$$|F = I + J,\ m_F = \pm F\rangle = |J,\ m = \pm J\rangle \otimes |I,\ m = \pm I\rangle$$

For example: Shift from room temperature blackbody radiation:
analysis for stretched states m_F=±(J+I): ≈10^{-22},

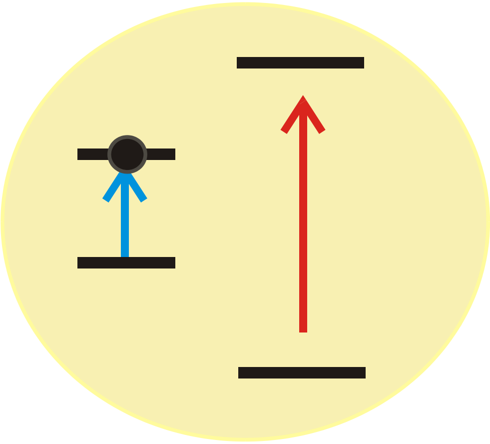
C.J. Campbell et al., PRL **108**, 120802 (2012)

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.

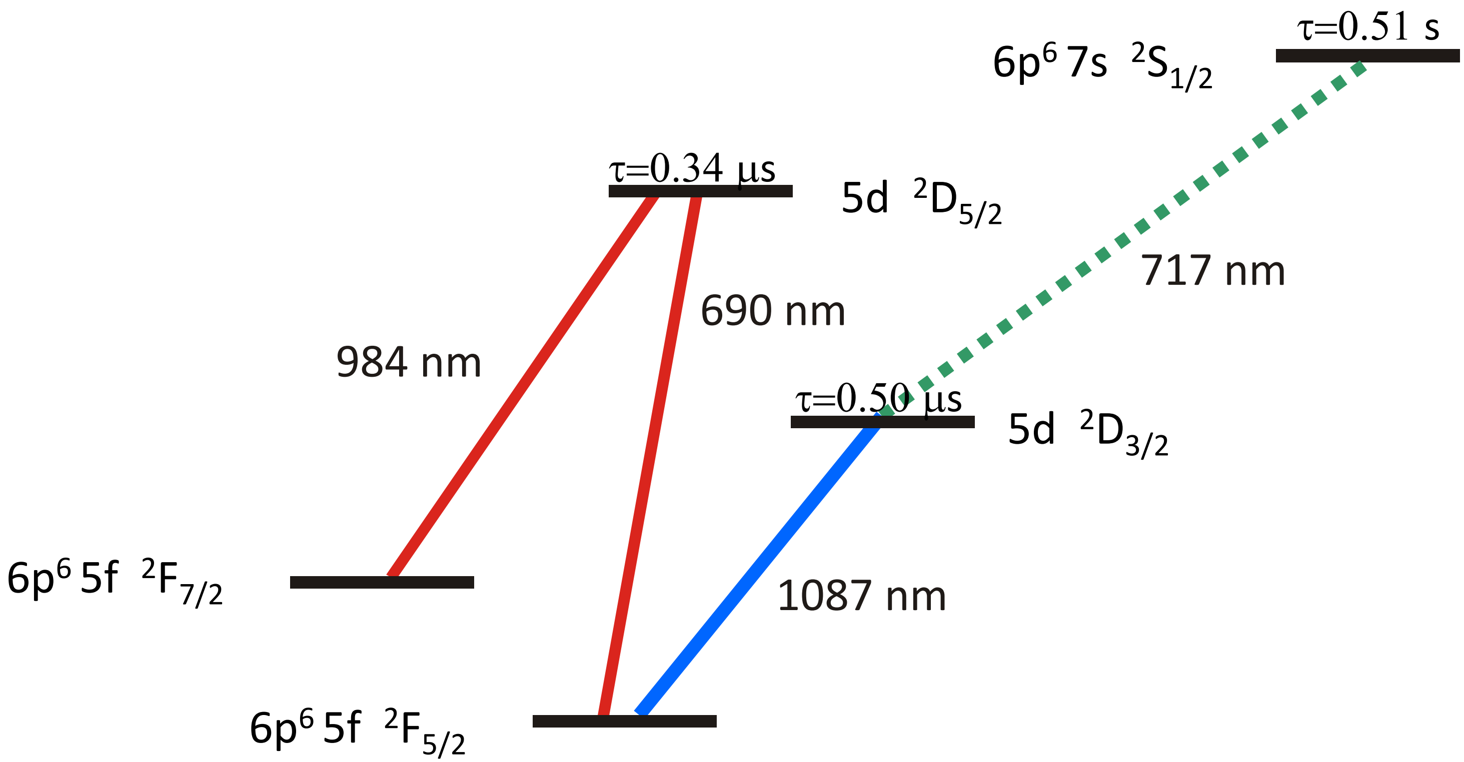


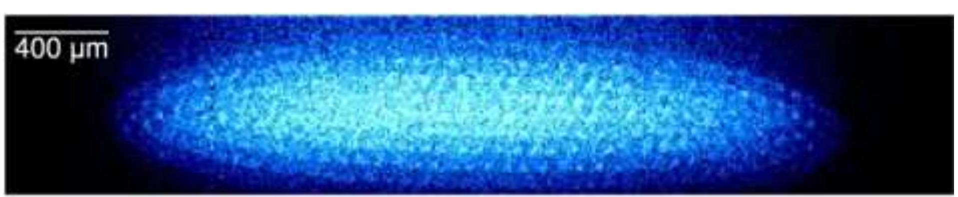
Laser excitation of the nucleus;
change of hyperfine structure detected in
intensity or polarisation of fluorescence.

Analog of Dehmelt's „electron shelving"
Observation of „quantum jumps" in the single-ion fluorescence

Electronic Level Scheme of Th^{3+}

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.



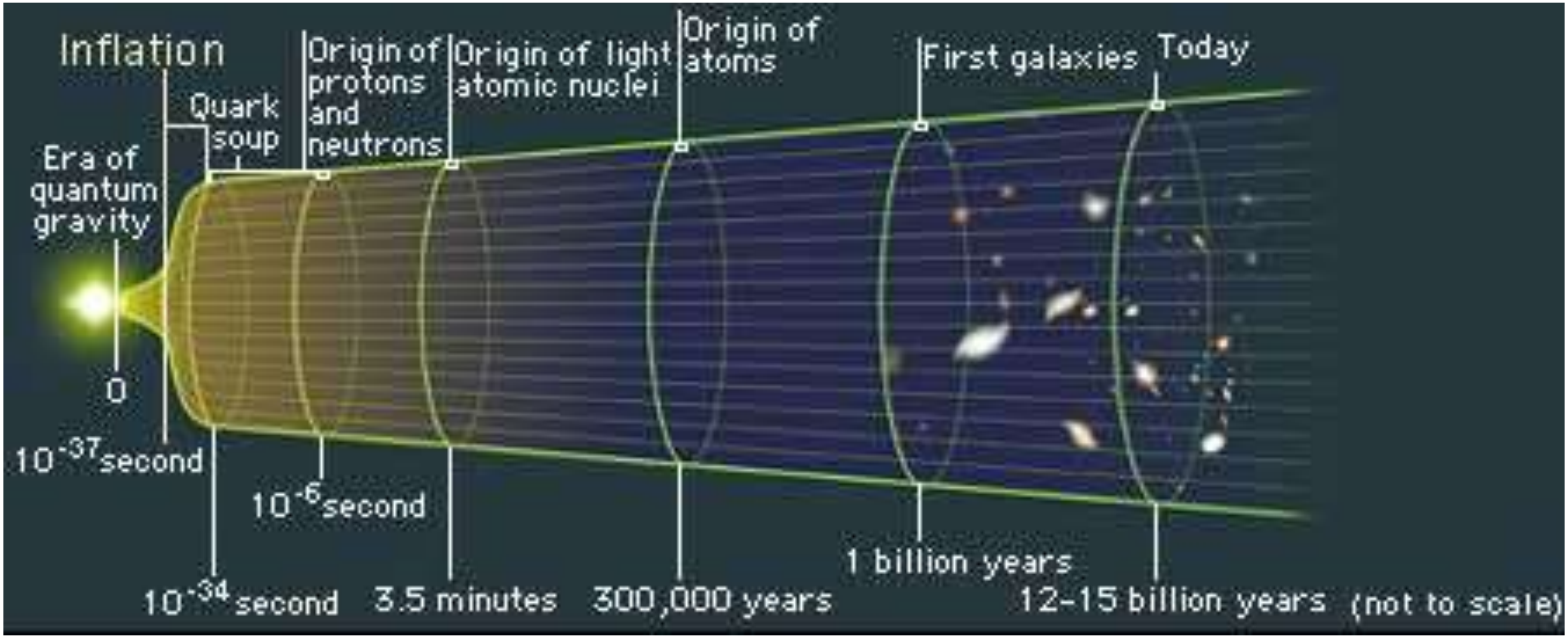


Lifetime calculations:
Biémont et al., JPB **37**, 4193 (2004)
Safronova et al. PRA **74**, 042511 (2006)
Experiment:
Campbell et al., PRL **102**, 233004 (2009)

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



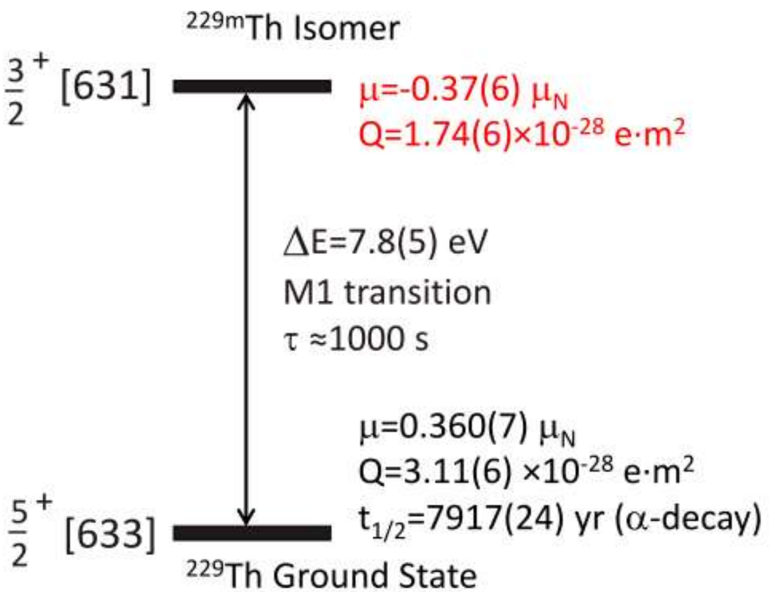
Th-229: the most sensitive probe in a search for variations of the fundamental coupling constants

Scaling of the ^{229}Th transition frequency ω in terms of α and quark masses:
V. Flambaum: Phys. Rev. Lett. **97**, 092502 (2006)

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

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

10^5 enhancement in sensitivity results from the cancellation of O(MeV) contributions in the O(eV) transition energy.



Th-229: the most sensitive probe in a search for variations of the fundamental coupling constants

α-sensitivity of nuclear transition frequency ω 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:

A. C. Hayes, J. L. Friar, P. Möller, Phys. Rev. C **78**, 024311 (2008) ($|K|<10^3$)

E. Litvinova et al., Phys. Rev. C **79**, 064303 (2009) ($|K|<4\text{x}10^4$)

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

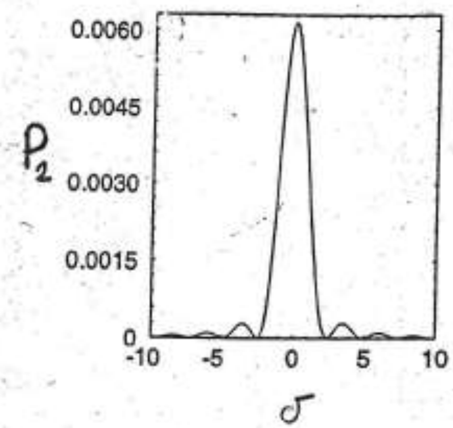
Rabi- and Ramsey-Excitation

2-level system with pulsed excitation.
χ: res. Rabi frequency, δ: detuning

Excitation probability after the pulse

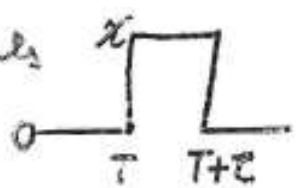
$$P_2 = \left| \int_{-\infty}^{\infty} \frac{\chi(t)}{2} e^{i\delta t} \, dt \right|^2$$

(quadatic Fourier transform of the pulse)



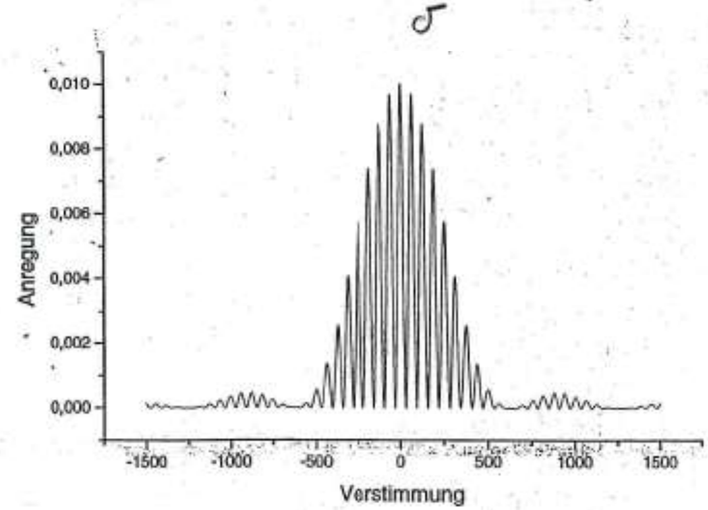
1 Pulse (Rabi)

Rechteck-Puls



$$P_2 = \frac{\chi^2}{\delta^2} \sin^2 \frac{\delta \tau}{2}$$

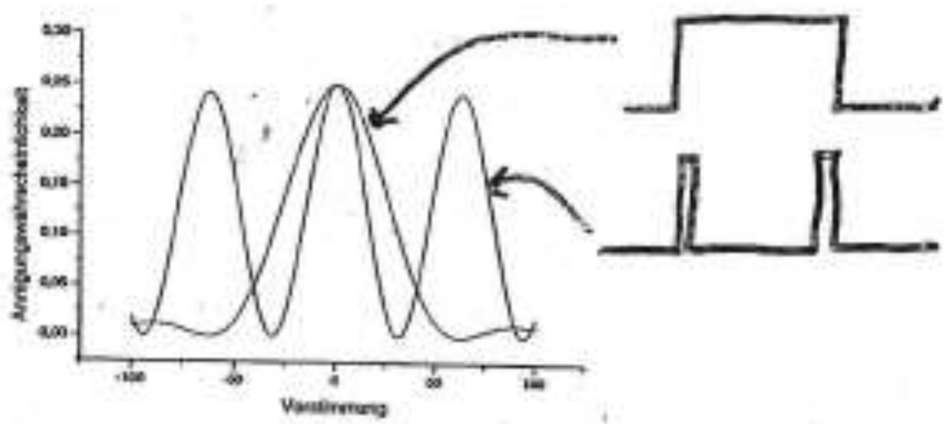
2 Pulses (Ramsey)



$$P_2 = 4 \cos^2 \delta T \cdot \sin^2 \frac{\delta \tau}{2} \cdot \frac{\chi^2}{\delta^2}$$

Advantages of Ramsey excitation

about 0.5x narrower resonance
for the same interaction time

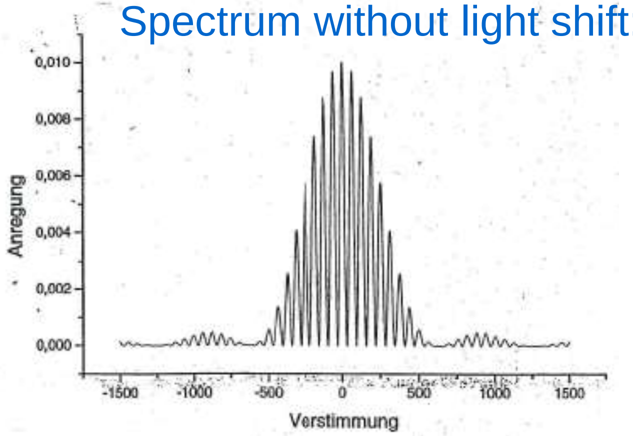


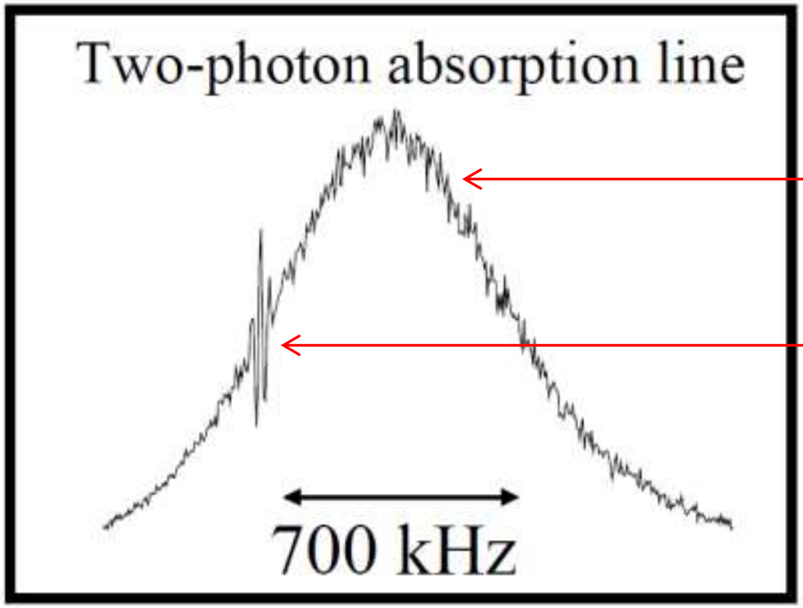
Resonance not broadened by perturbations between the interaction zones
(like B field inhomogeneity)

Light shift in Ramsey spectroscopy

Excitation with two pulses

Spectrum without light shift:



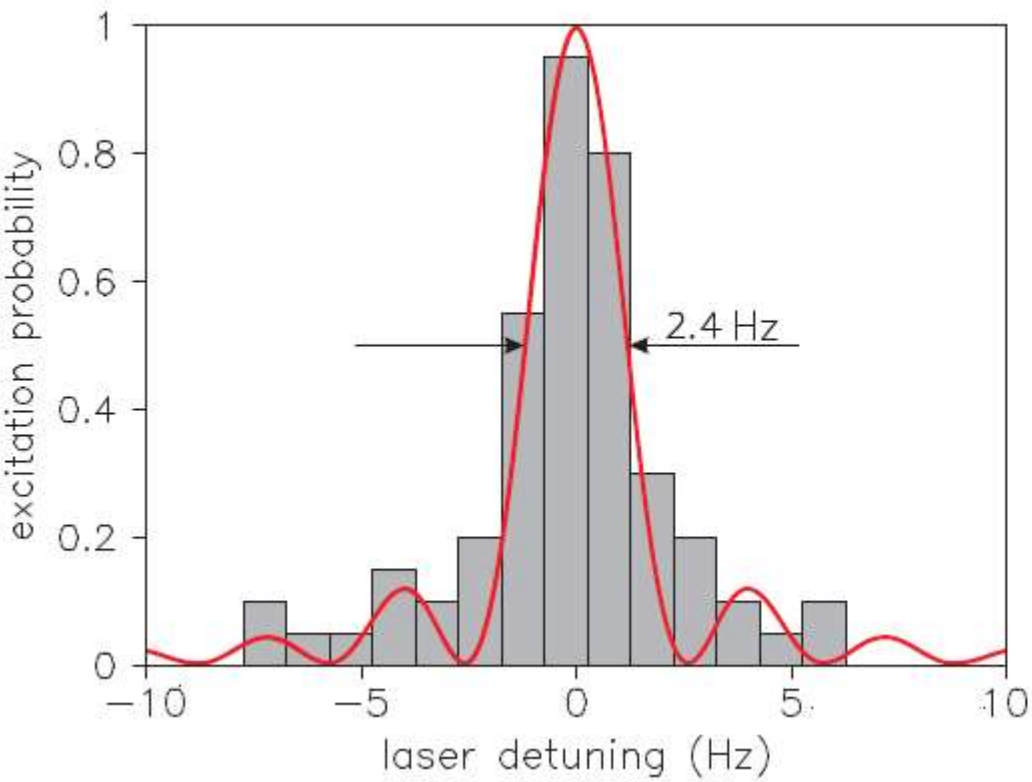


Envelope: due to excitation in a single pulse,
Strong light shift

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

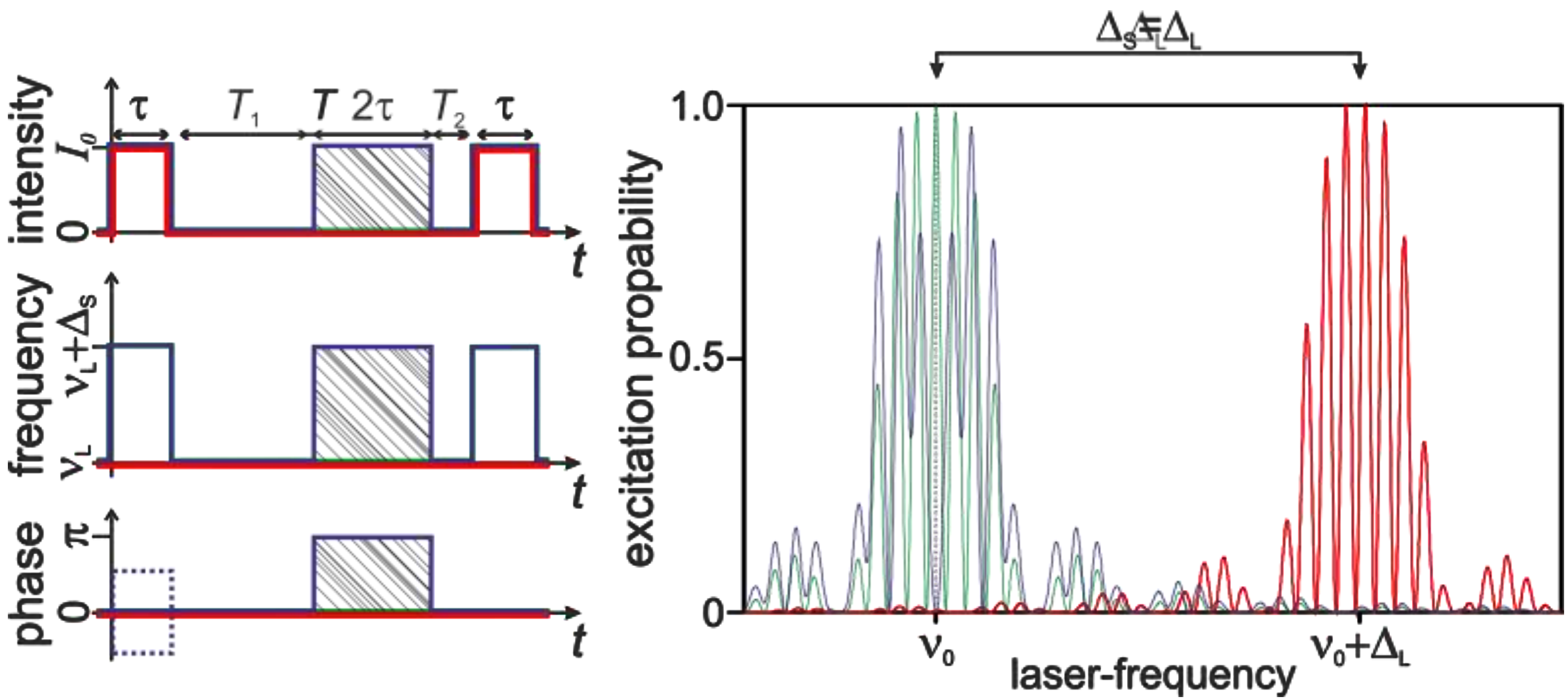
From:
M. Marocco, M. Weidinger, R. Sang,
H. Walther, Phys. Rev. Lett. 81, 5784 (1998)

High resolution excitation spectrum of the $^{171}Yb^{+}$ octupole transition



- 335 ms pulse duration
- 50 μW laser power
- Fourier-limited linewidth
- $Q = 2.7 \times 10^{14}$
- Light shift $\approx$ 4 Hz

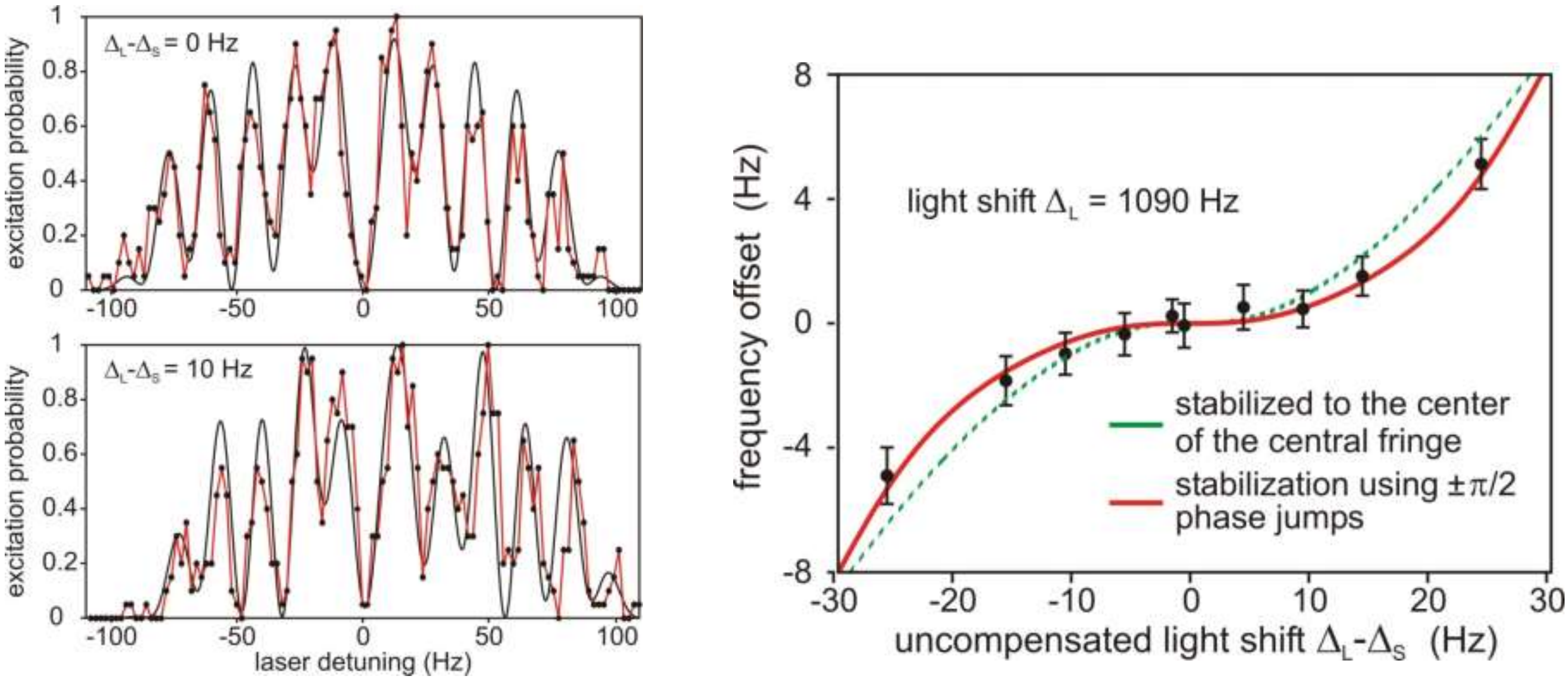
“Hyper-Ramsey” spectroscopy (HRS)



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

Yudin, Taichenachev *et al.,* Phys. Rev. A **82**, 011804(R) (2010)
Huntemann *et al.*, Phys. Rev. Lett. **109**, 213002 (2012)

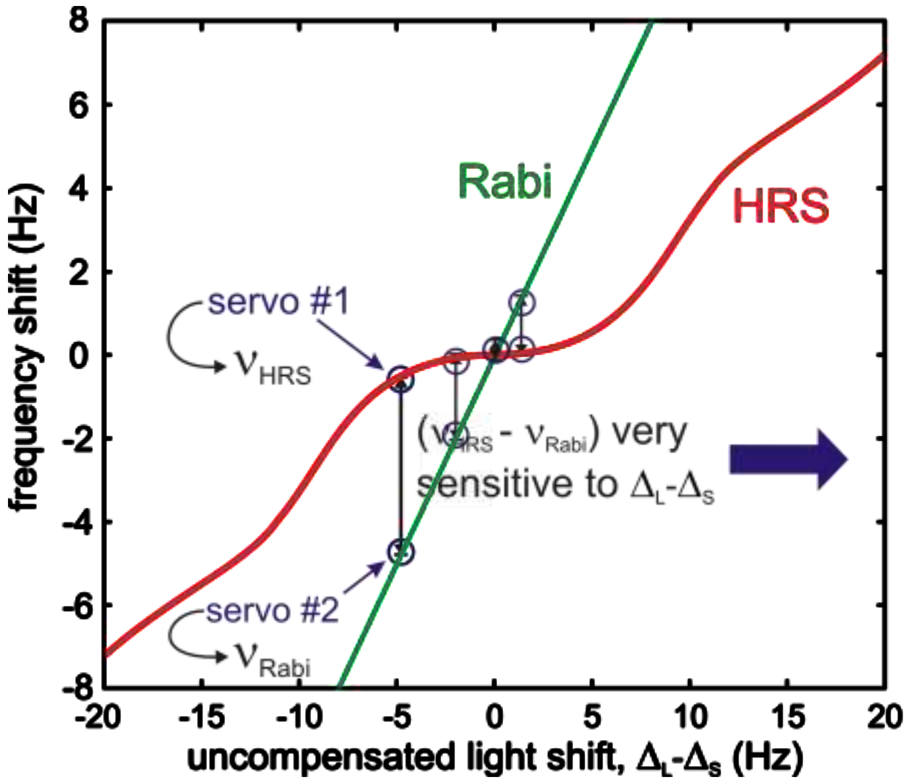
Experimental investigation of Hyper-Ramsey Spectroscopy

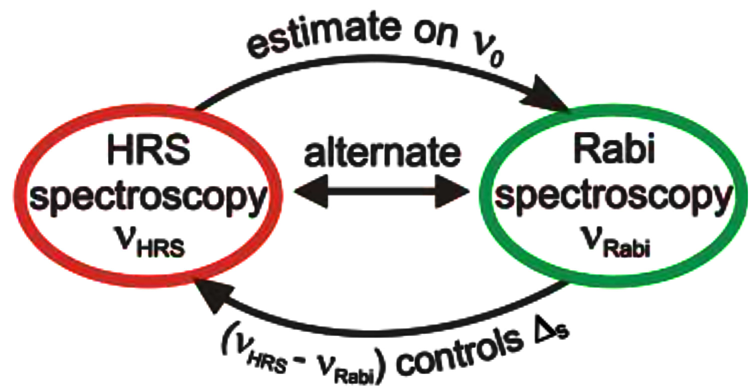


- HRS parameters: T=36 ms , 2τ=18 ms
- Largely unaffected position of central minimum
- Interleaved stabilization on $(\Delta_L\text{-}\Delta_S) = 0$ and $(\Delta_L\text{-}\Delta_S) \neq 0$
- Cubic dependence of the resonance center on $(\Delta_L\text{-}\Delta_S)$

N. Huntemann *et al.*, Phys. Rev. Lett. **109** 213002 (2012)

Controlled HRS scheme





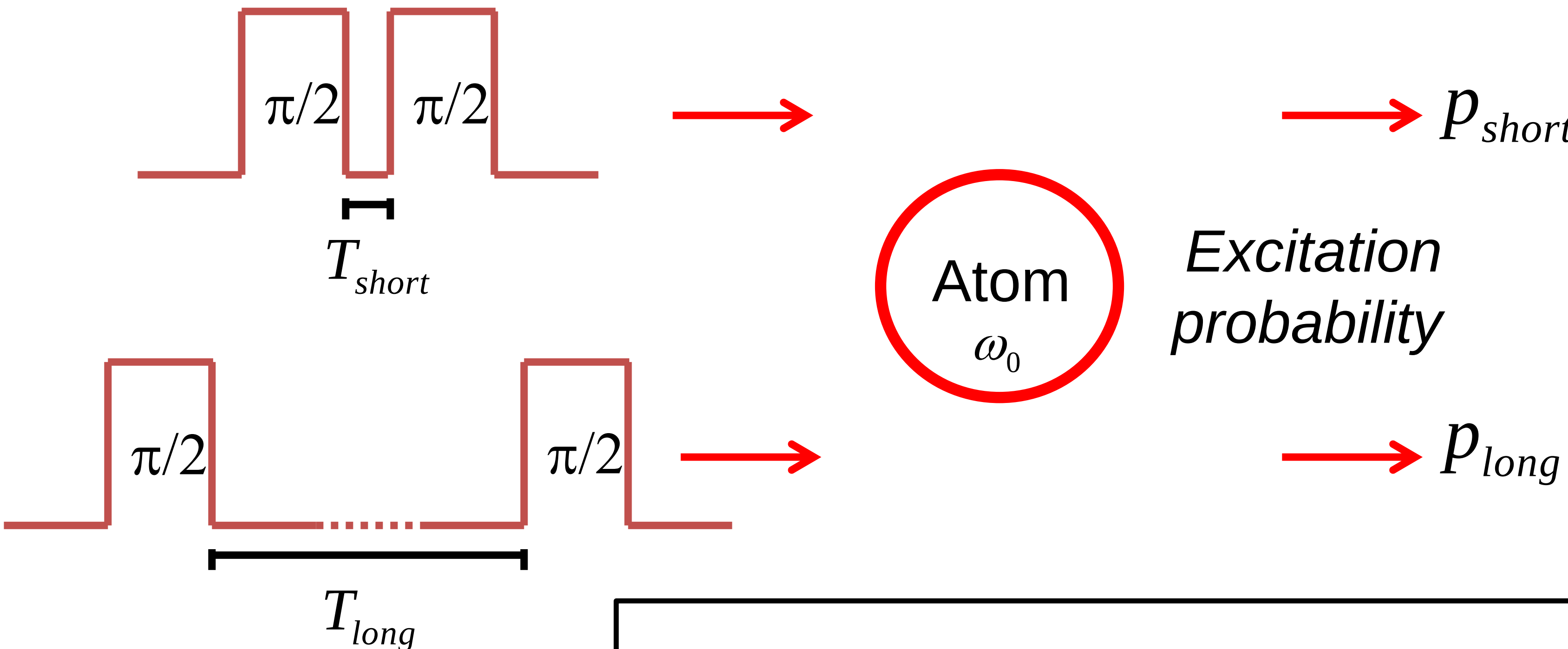
- Interleaved stabilization using Rabi spectroscopy with the same intensity and step frequency Δ_S to measure the frequency offset
- A negative feedback loop uses ν_{HRS} - ν_{Rabi} to regulate Δ_S and ensures $|\Delta_S - \Delta_L|=0$
- Real-time measurement of the light shift

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)

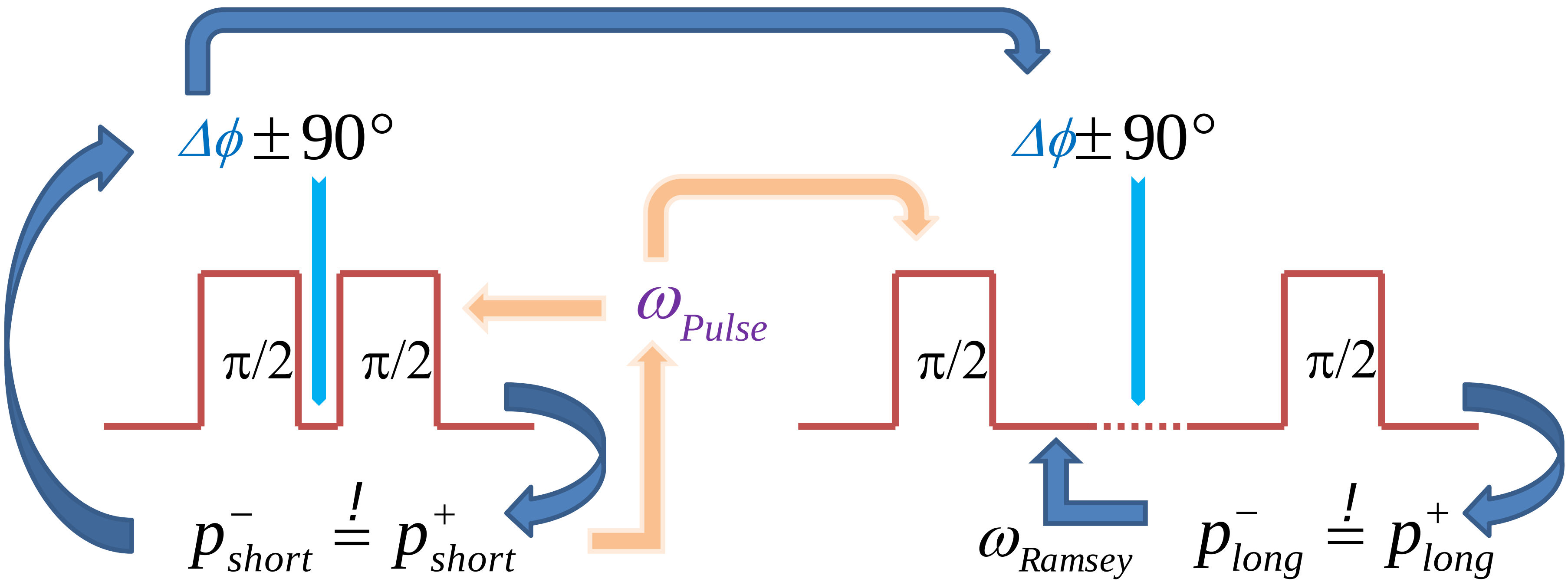
- Ramsey interrogation: spectroscopic information is accumulated during the dark period *T*.



$$\boxed{\omega_{Ramsey} = \omega_0 \rightarrow \quad p_{short} = p_{long}}$$

Stabilization in the autobalanced Ramsey scheme

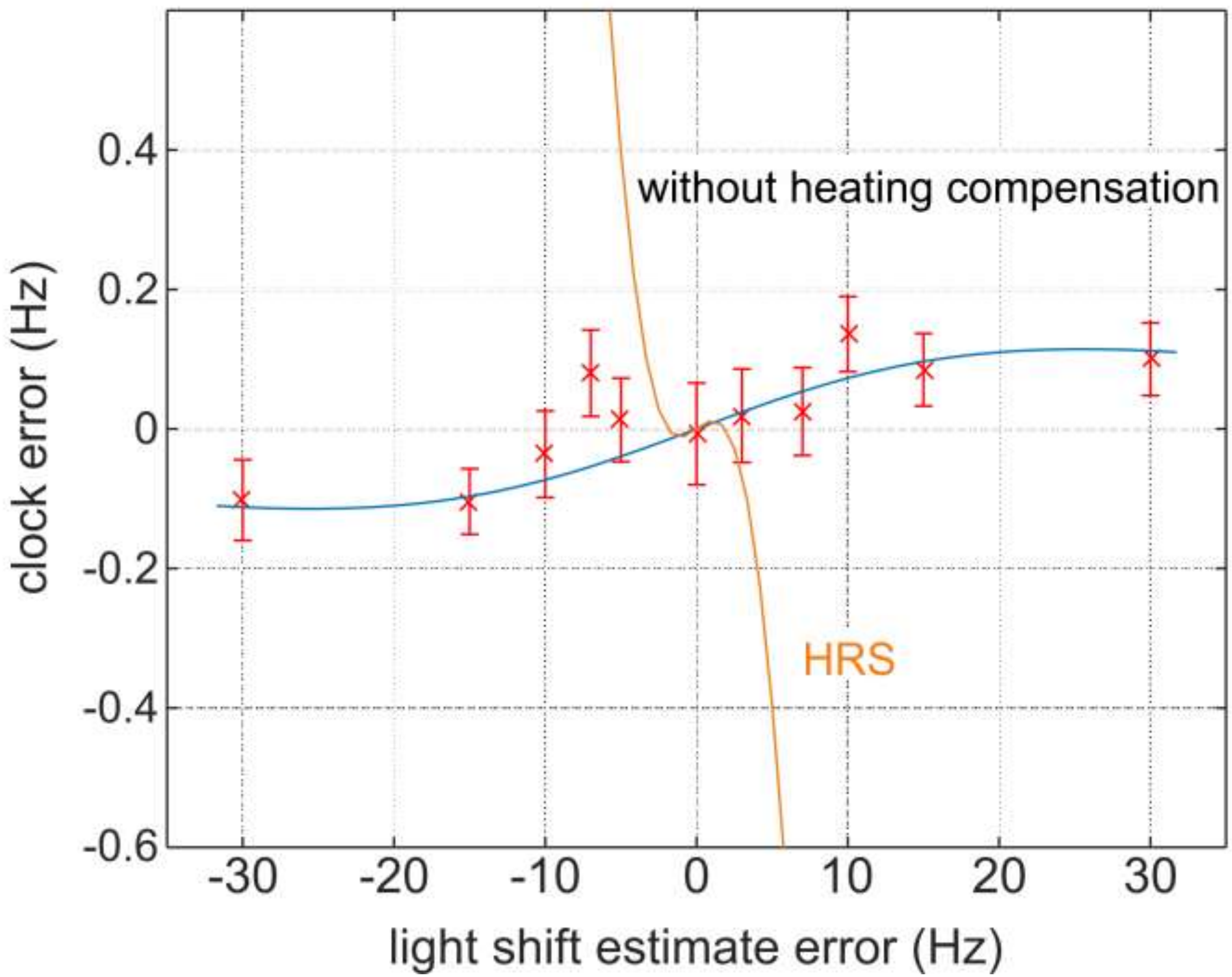
- Based on frequency tuning of ω_{Pulse} and ω_{Ramsey}
- Based on an additional phase step $\Delta\phi$ and ω_{Ramsey}



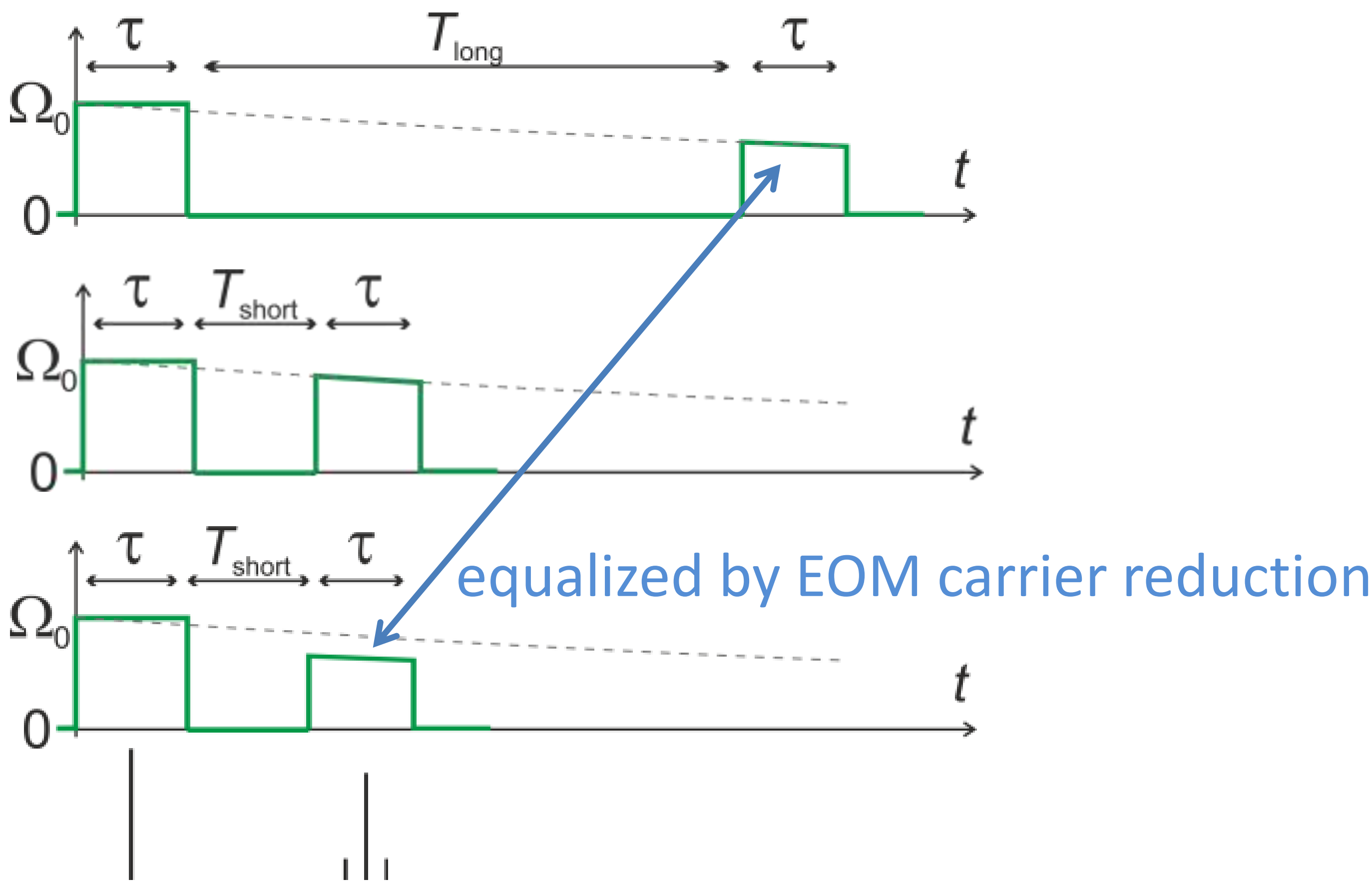
- Two interleaved servos
- Appropriate for systems with slowly varying pulse defects

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

Auto-balanced Ramsey scheme – experimental results

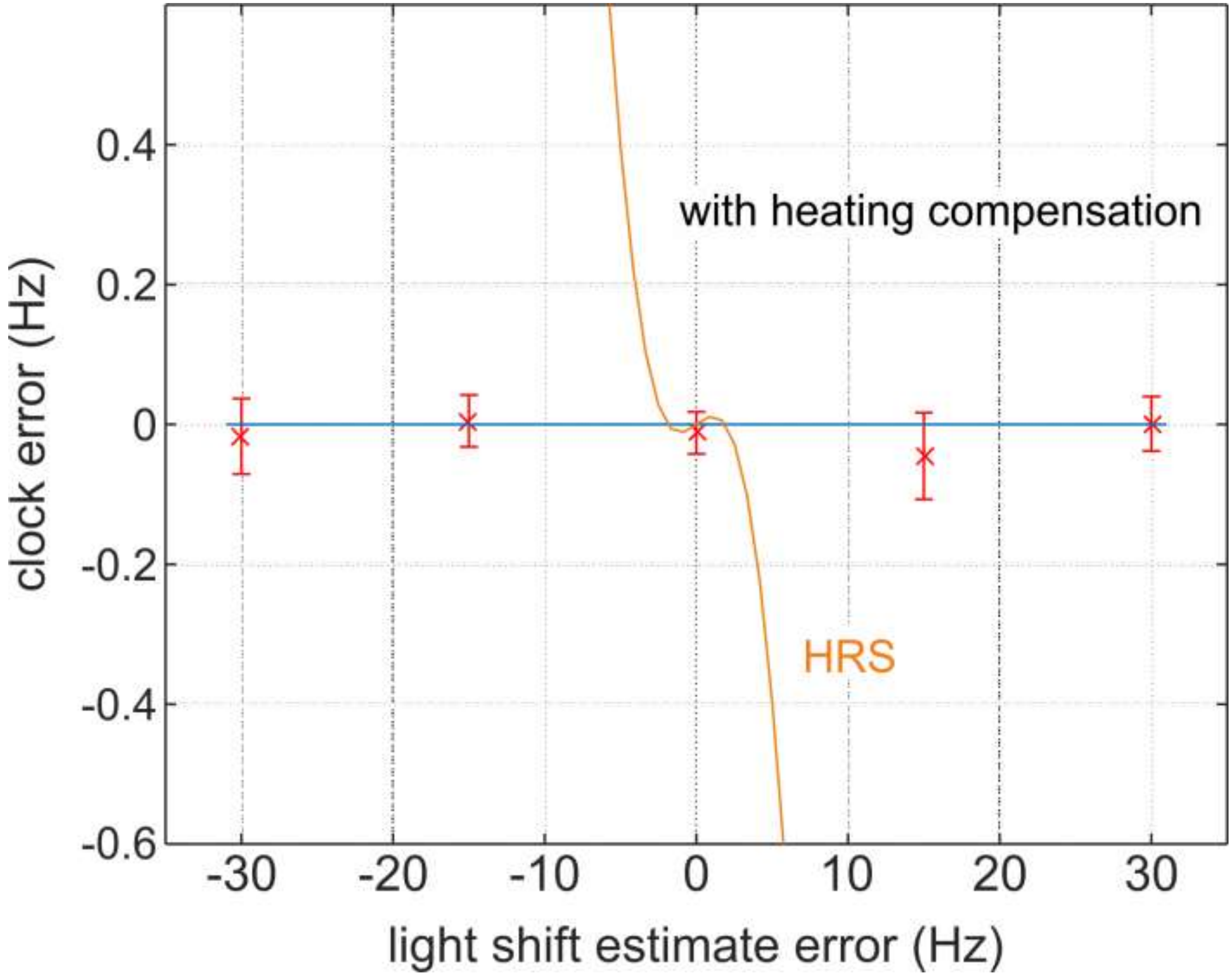


Auto-balanced Ramsey scheme – experimental results



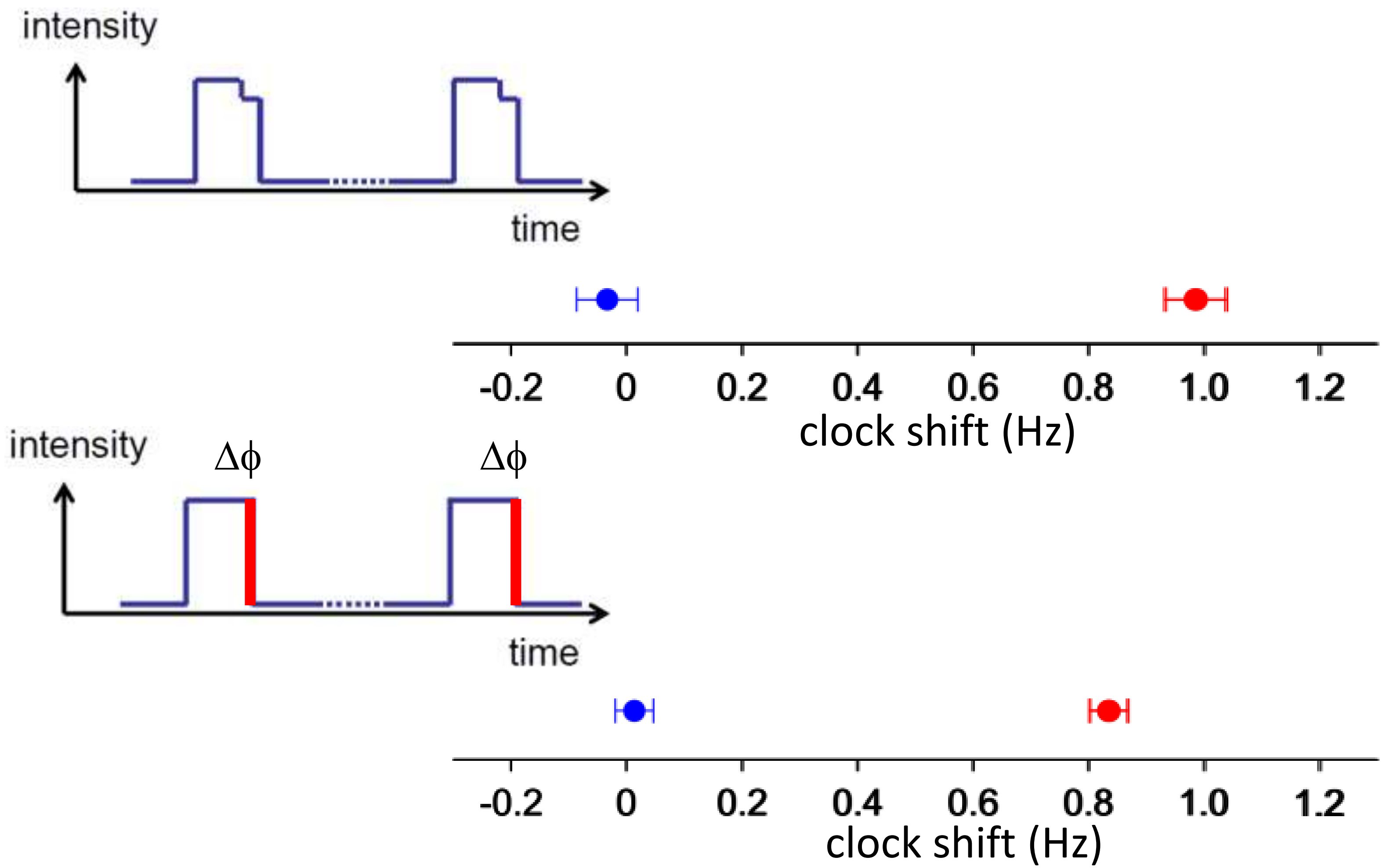
- Rabi-frequency reduction by phase-modulation with an EOM

Auto-balanced Ramsey scheme – experimental results



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 Lefevre[1], Rémi Metzdorff[2], Nicolas Sillitoe[2], Sylvain Almonacil[3], Marco Minissale[4], Emeric de Clercq[5], Alexey V Taichenachev[6,7], Valeriy I Yudin[6,7,8] and Ennio Arimondo[9]

Published 1 August 2018

Reports on Progress in Physics, Volume 81, Number 9