

**Microwave and optical
spectroscopy
in r.f. traps
Application to atomic clocks**

Microwave spectroscopy for hyperfine structure measurements

Energy of a hyperfine state

$$W_F = \frac{1}{2} hAK + hB \frac{\frac{3}{2}K(K+1) - 2I(I+1)J(J+1)}{2I(2I+1)2J(2J+1)}$$

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

Hyperfine coupling constants:

A: magnetic dipole interaction

B: Electric quadrupole interaction

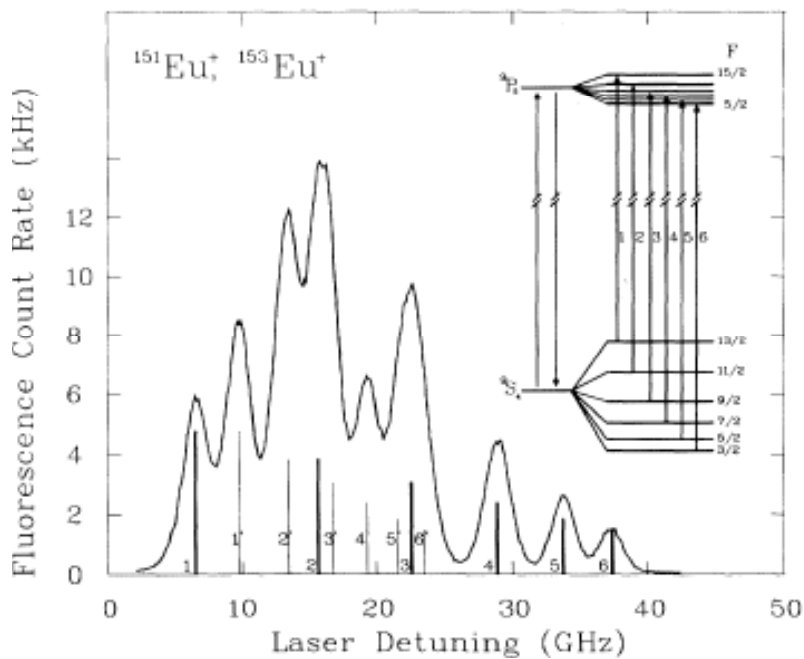
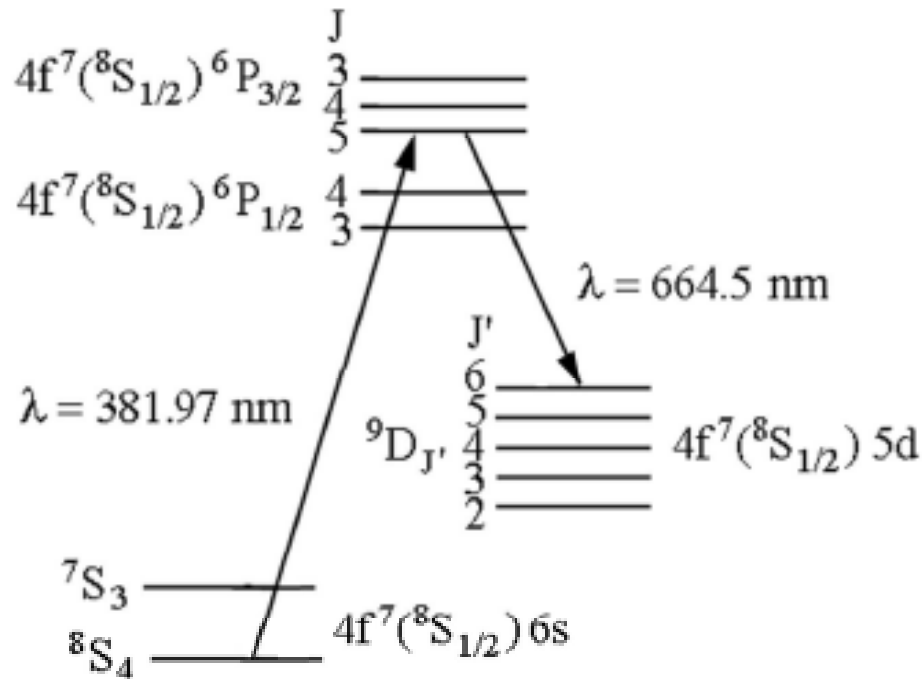
$$A_s = -\frac{1}{h} \frac{16\pi}{3} \frac{\mu_0}{4\pi} \mu_B^2 g_I |\Psi_s(0)|^2$$

$$B = \frac{1}{h} \frac{e^2}{4\pi\epsilon_0} \frac{2J+1}{2J+2} \langle r^{-3} \rangle_{nl} Q$$

Features of microwave spectroscopy:

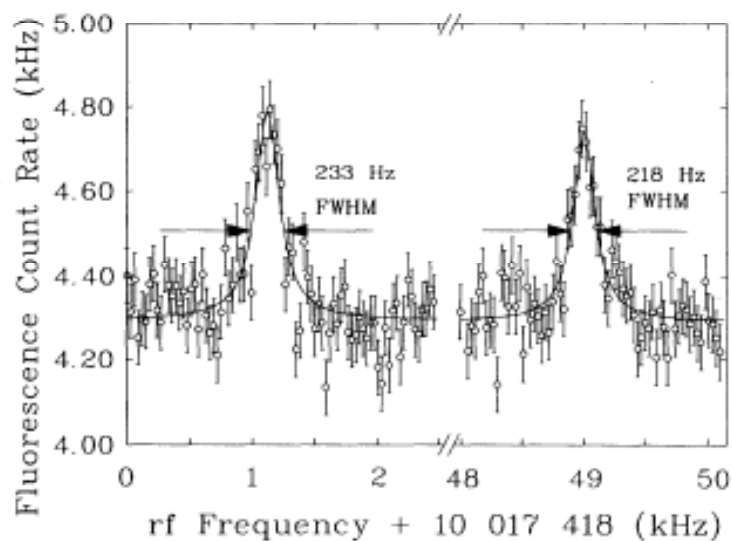
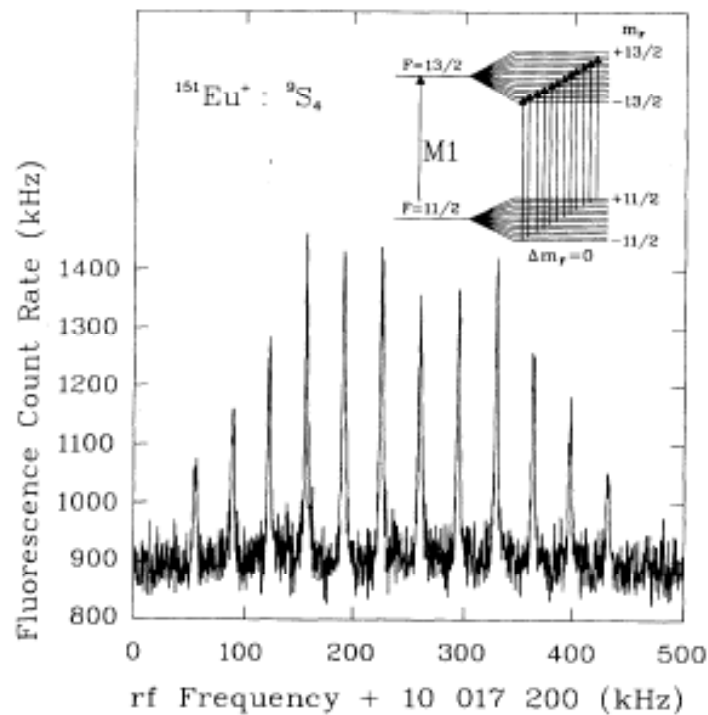
- No first order Doppler effect because of Dicke effect (ion oscillation amplitude $<$ wavelength of radiation)
- Stable and accurate radiation sources available
- State preparation by laser or spectral lamp
- Resonance detection by fluorescence light

Example I: Eu+



Optical spectrum
of stable
 Eu^+ isotopes 151, 153

Induced hyperfine transitions with Zeeman splitting



Uncertainty of resonances: 20 Hz in 10 GHz

Results for Eu isotopes

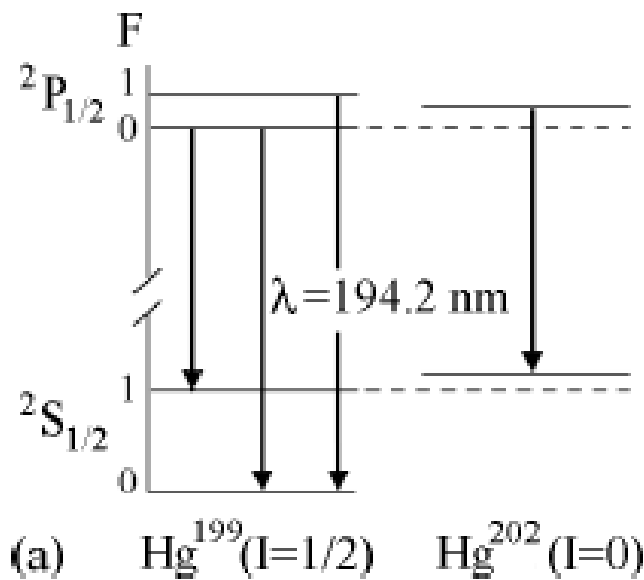
	I		A [MHz]	B [kHz]
$^{148}\text{Eu}^+$	5	6^9S_4	517.281 950 (150)	-292.63 (1.00)
$^{148}\text{Eu}^+$	5	6^7S_3	-561.647 (100)	
$^{149}\text{Eu}^+$	5/2	6^9S_4	1 585.450 570 (250)	-534.85 (1.90)
$^{150}\text{Eu}^+$	5	6^9S_4	599.010 680 (200)	-839.73 (3.00)
$^{150}\text{Eu}^+$	5	6^7S_3	-650.334 (2)	
$^{151}\text{Eu}^+$	5/2	6^9S_4	1 540.297 394 (13)	-660.862 (2.31)
$^{151}\text{Eu}^+$	5	6^7S_3	-1 672.457 109 (266)	-599.4 (3.1)
$^{153}\text{Eu}^+$	5/2	6^9S_4	684.565 993 (9)	-1 752.868 (84)
$^{153}\text{Eu}^+$	5	6^7S_3	-743.183 577 (82)	2 448.35 (8)

First step towards systematic study of **Bohr-Weisskopf effect** (distribution of magnetization over nuclear volume)

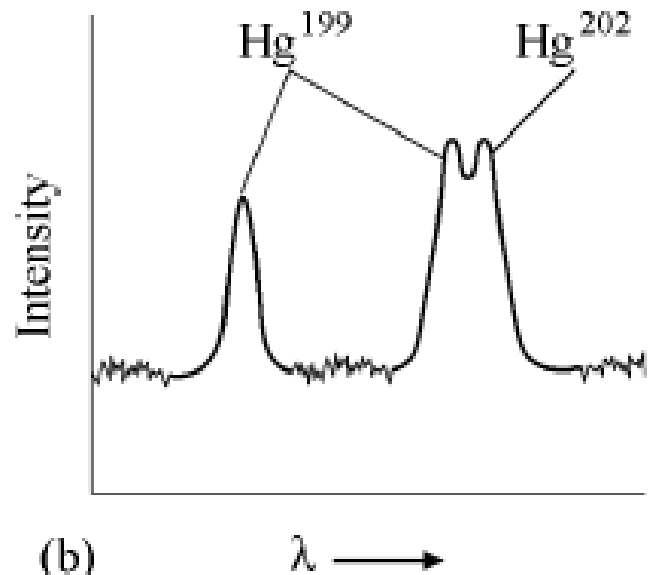
$$\Delta(1,2)=1-(A_1/A_2)(g_{I2}/g_{I1})$$

Need to measure nuclear g factors

Example II: Hg⁺



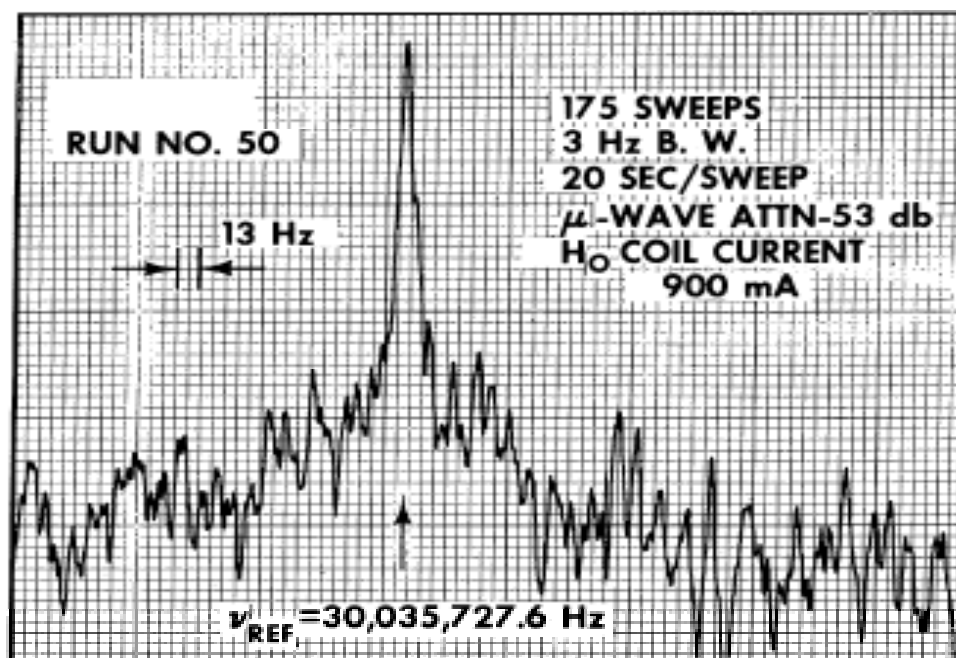
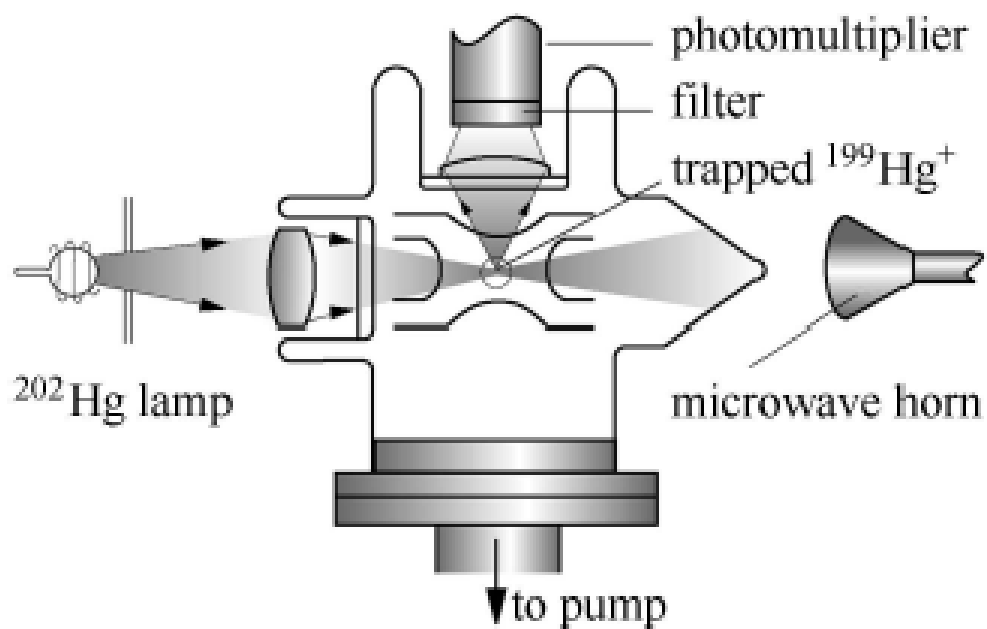
Level diagram
of Hg⁺ isotopes



Optical spectrum
of 199 Hg⁺ and 202Hg⁺

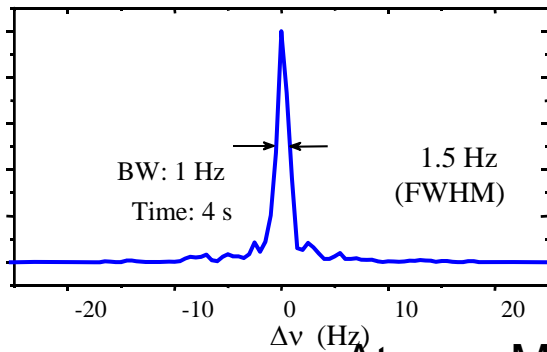
Optical pumping of one ground state hyperfine level
of 199Hg⁺ by accidental coincidence of 202Hg⁺ line
with one hyperfine component

Microwave transition between 199Hg⁺ hyperfine levels
monitored by change in fluorescence intensity

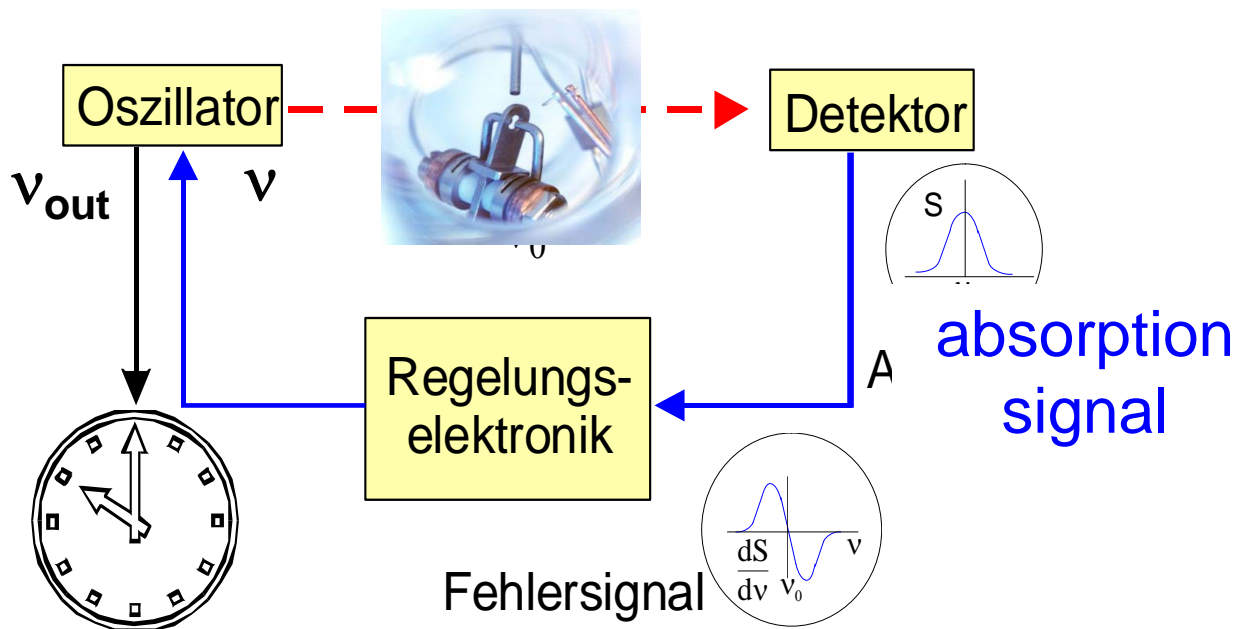


Major, Werth (1973)

Principle of Atomic Clocks



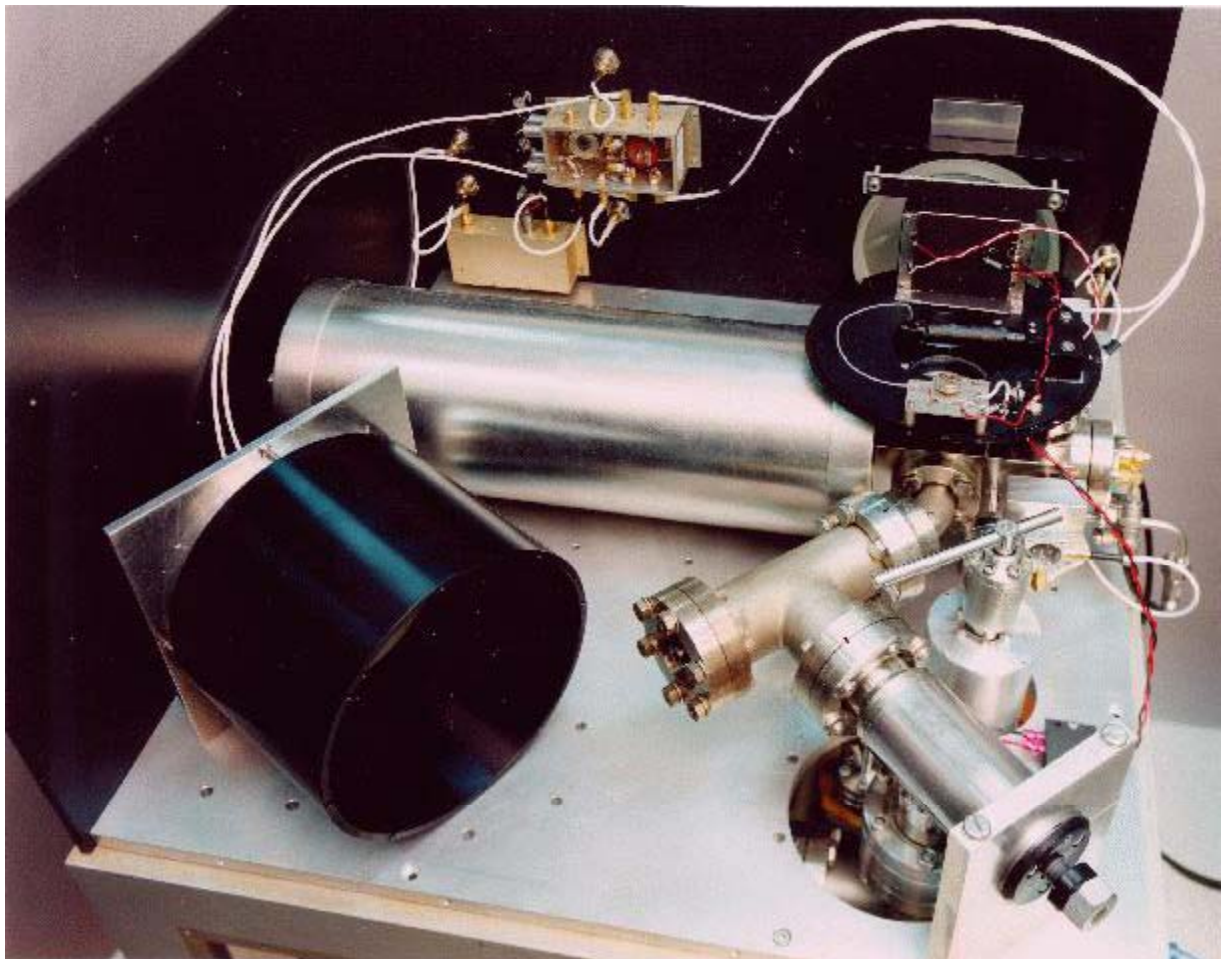
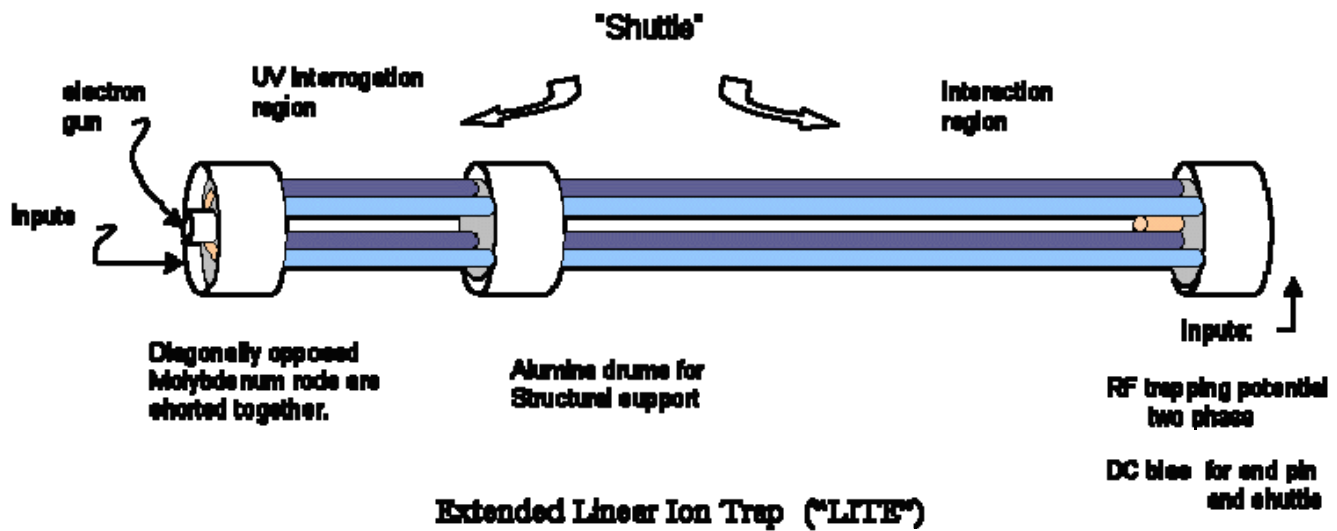
Atome, Moleküle oder Ionen



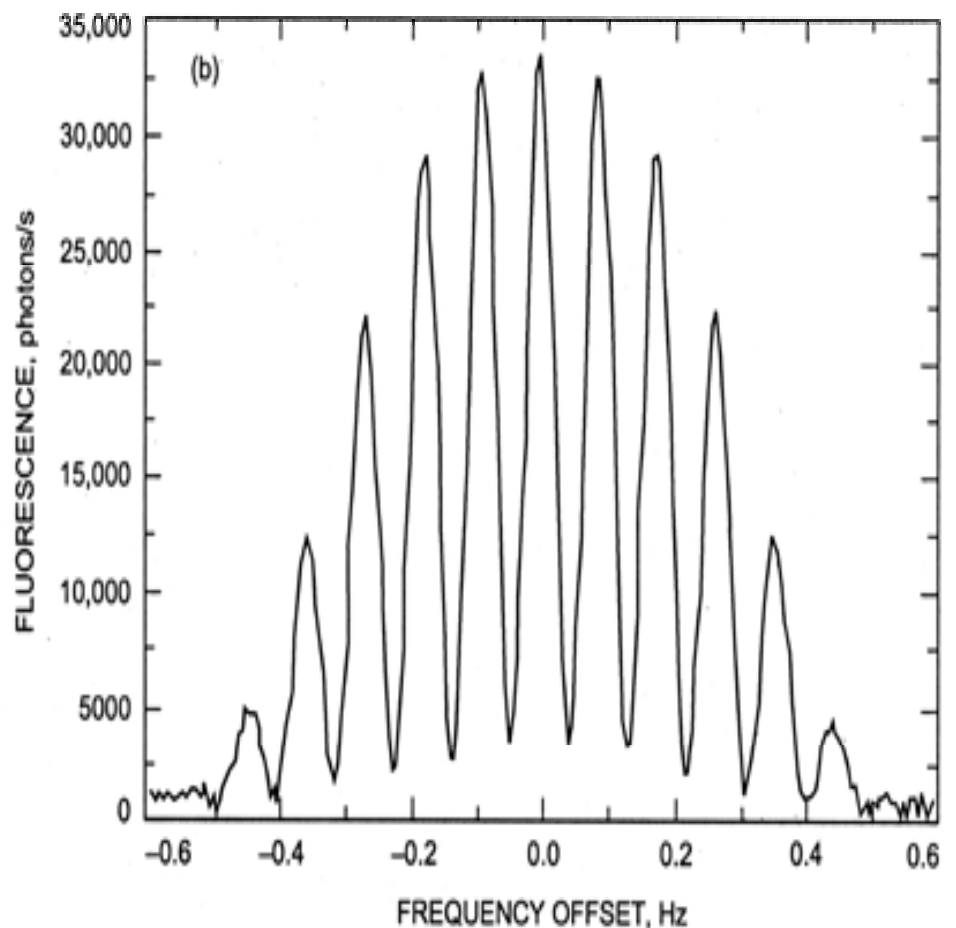
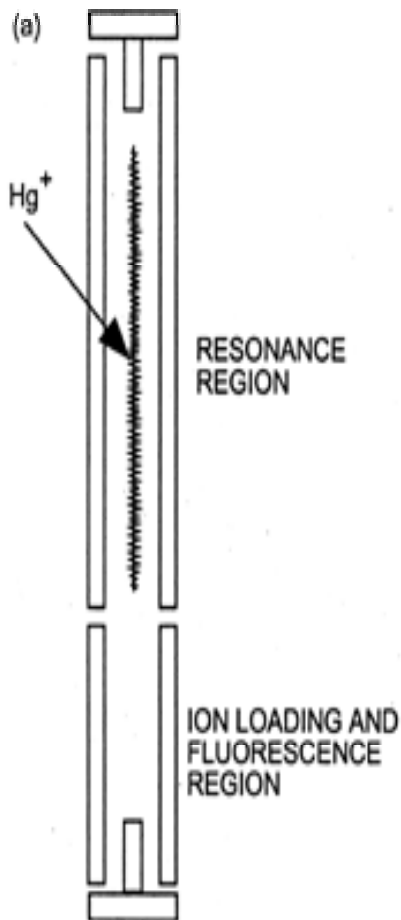
Accuracy: How accurately agrees v_{out} with v_0 ?

Stability: To what extent fluctuates v_{out} around v_0 ?

Linear ion trap at JPL for microwave frequency standard



40.9 GHz hyperfine transition in $^{199}\text{Hg}^+$
Ramsey fringes with 11,1 s interrogation time



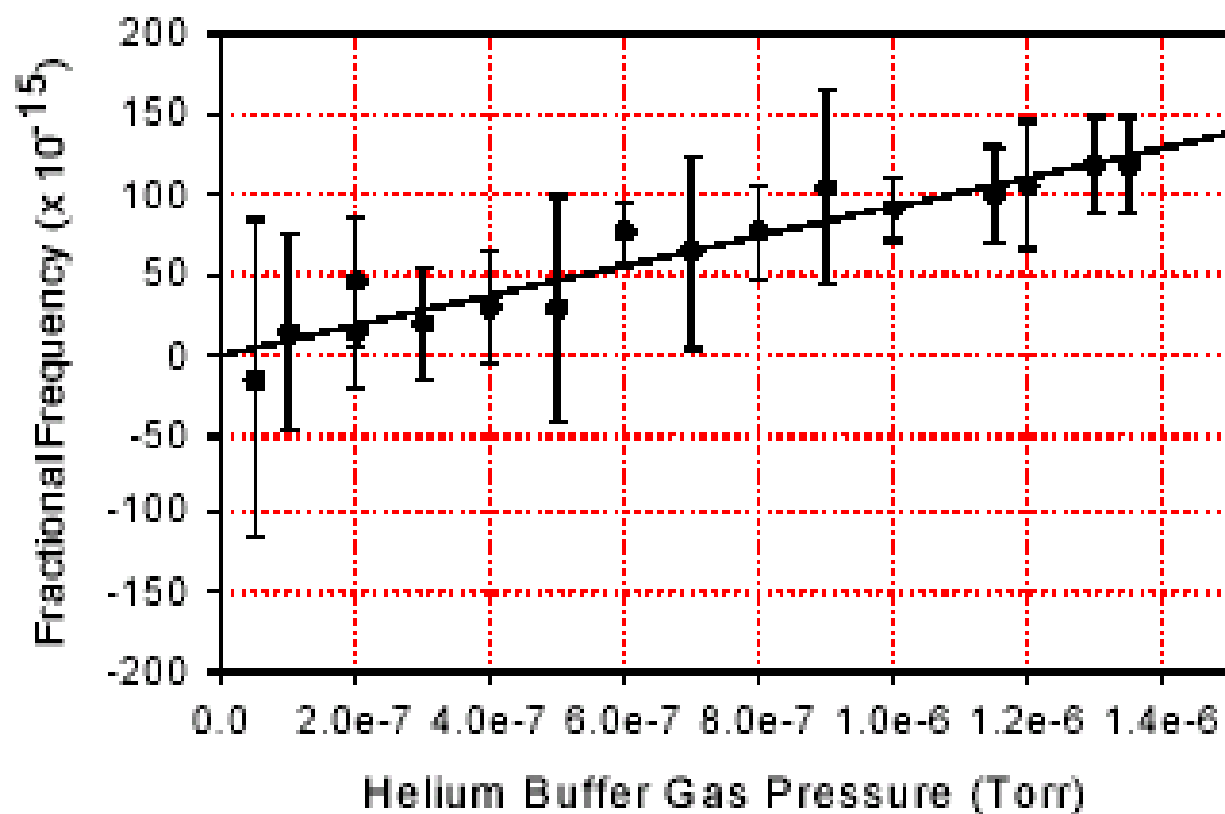
J. Tjoelker et al.,
JPL

$^{199}\text{Hg}^+$ microwave clock Uncertainty and stability

LITS Frequency Offsets & Stability			
Frequency Offsets	Magnitude	Uncertainty ($\times 10^{-15}$)	Stability ($\times 10^{-15}$)
DC Magnetic (at 0.08G)	1×10^{-11}	0.1	<0.2
Shielding (24,000)			<.002 / mG
2 nd Order Doppler			
Thermal (300K)	2×10^{-13}	<10	<0.2
Number Dependent	5×10^{-14}	<1	<0.1
Collision (pressure)			
Helium (6×10^{-6} Torr)	10^{-13}	<10	<0.2
Mercury(10^{-9} Torr)	?	?	?
Other (< 2×10^{-9} Torr)	?	?	?
Blackbody	$< 10^{-16}$	< 0.1	<0.01
Gravitational Redshift	$10^{-16}/\text{m}$	< 0.2	

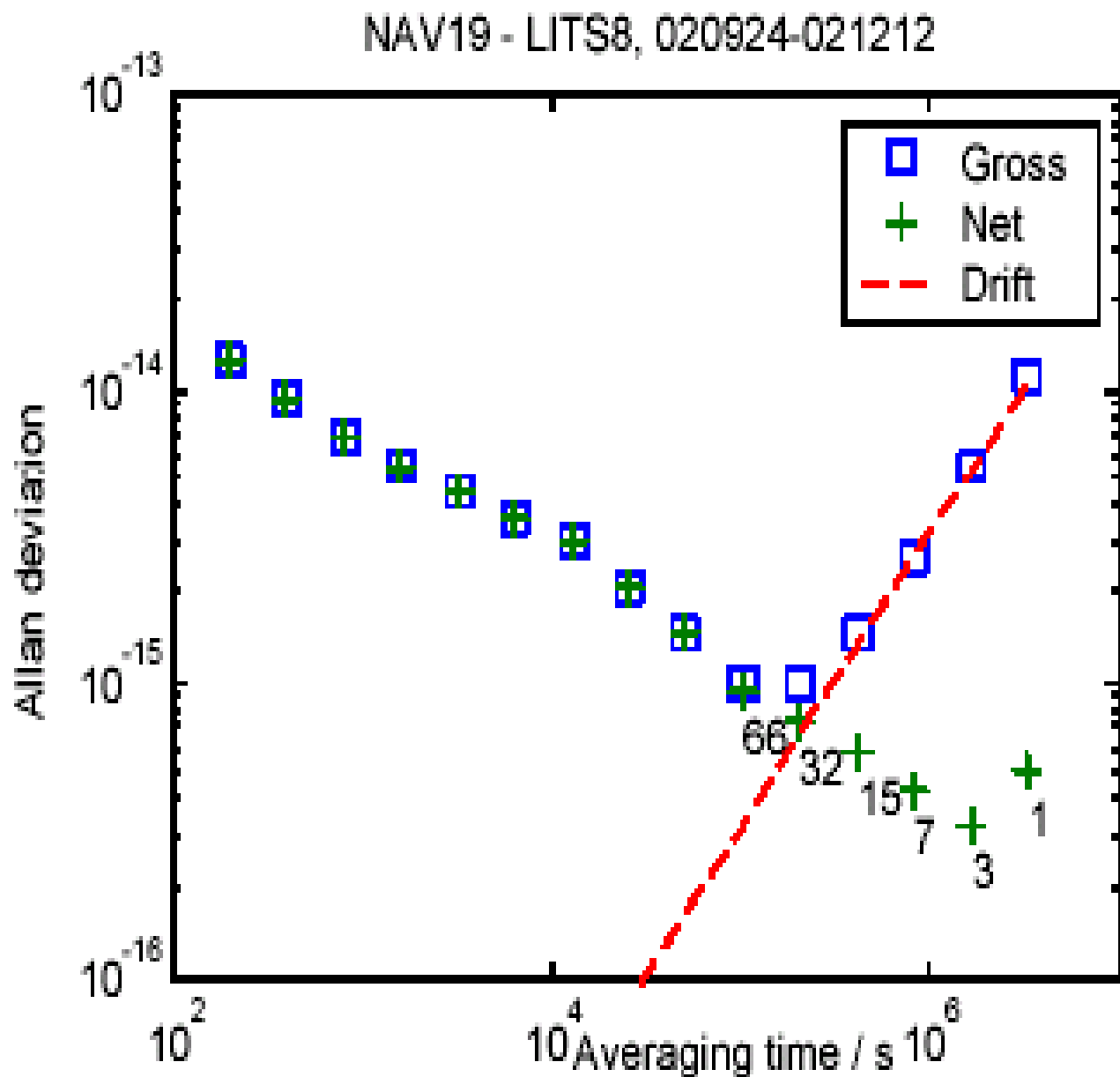
J. Tjoelker et al., Proc. 2003 Frequ. Contr. Symp

$^{199}\text{Hg}^+$ hyperfine frequency shifts
with He buffer gas pressure



Frequency stability of $^{199}\text{Hg}^+$ microwave standard

H-maser as reference



Red line: H-maser drift

J. Tjoelker et al, Prov. 2003 Frrqu.

Contr. Symp.

Optical spectroscopy

High resolution requires:

- Cooling into Dicke regime
- Long lived metastable states
- Lasers of high spectral purity

Optical clocks:

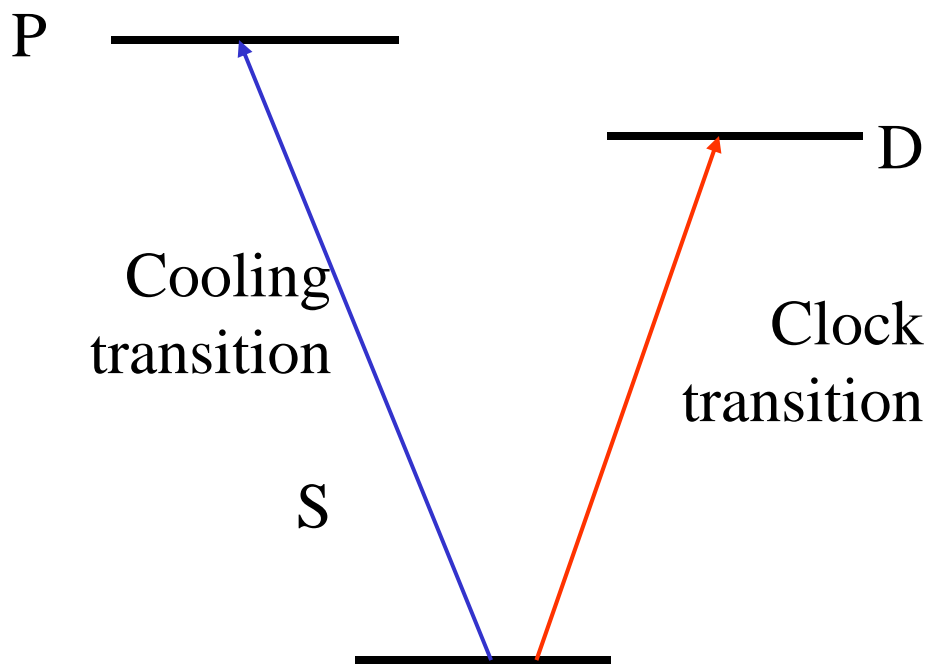
**Laser stabilization on narrow
transition to long lived metastable state,
Measurement of laser frequency**

Required level diagram

Requirement to level scheme:

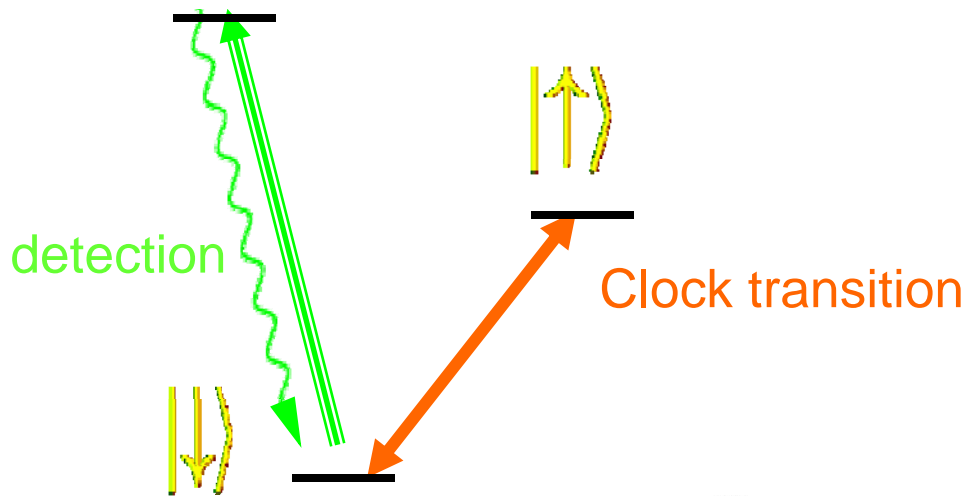
Fast E1 cooling transition

Narrow „clock“ transition

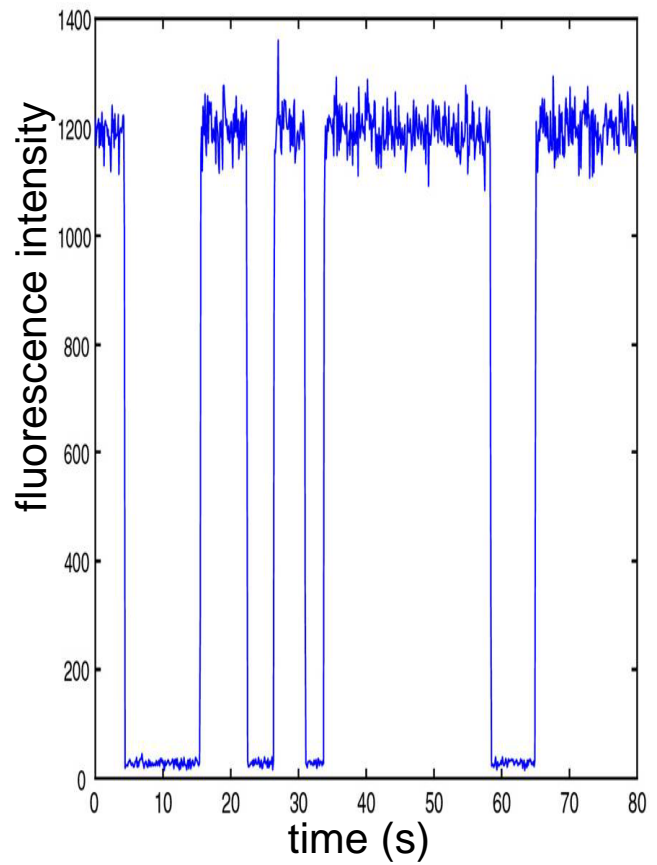


Candidate ions: Hg^+ , Yb^+ , Sr^+ , Ca^+ , In^+

State detection using electron shelving

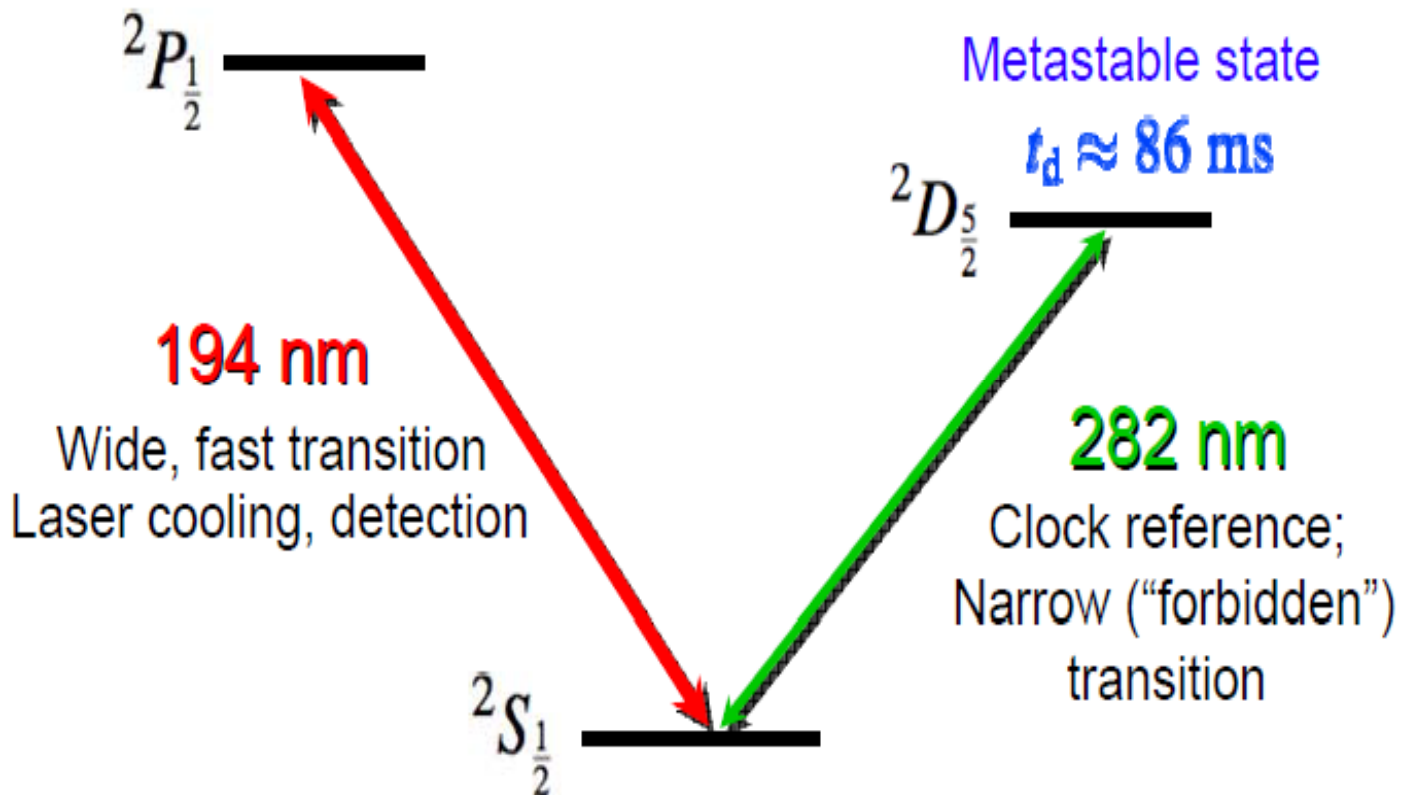


observe quantum jumps online

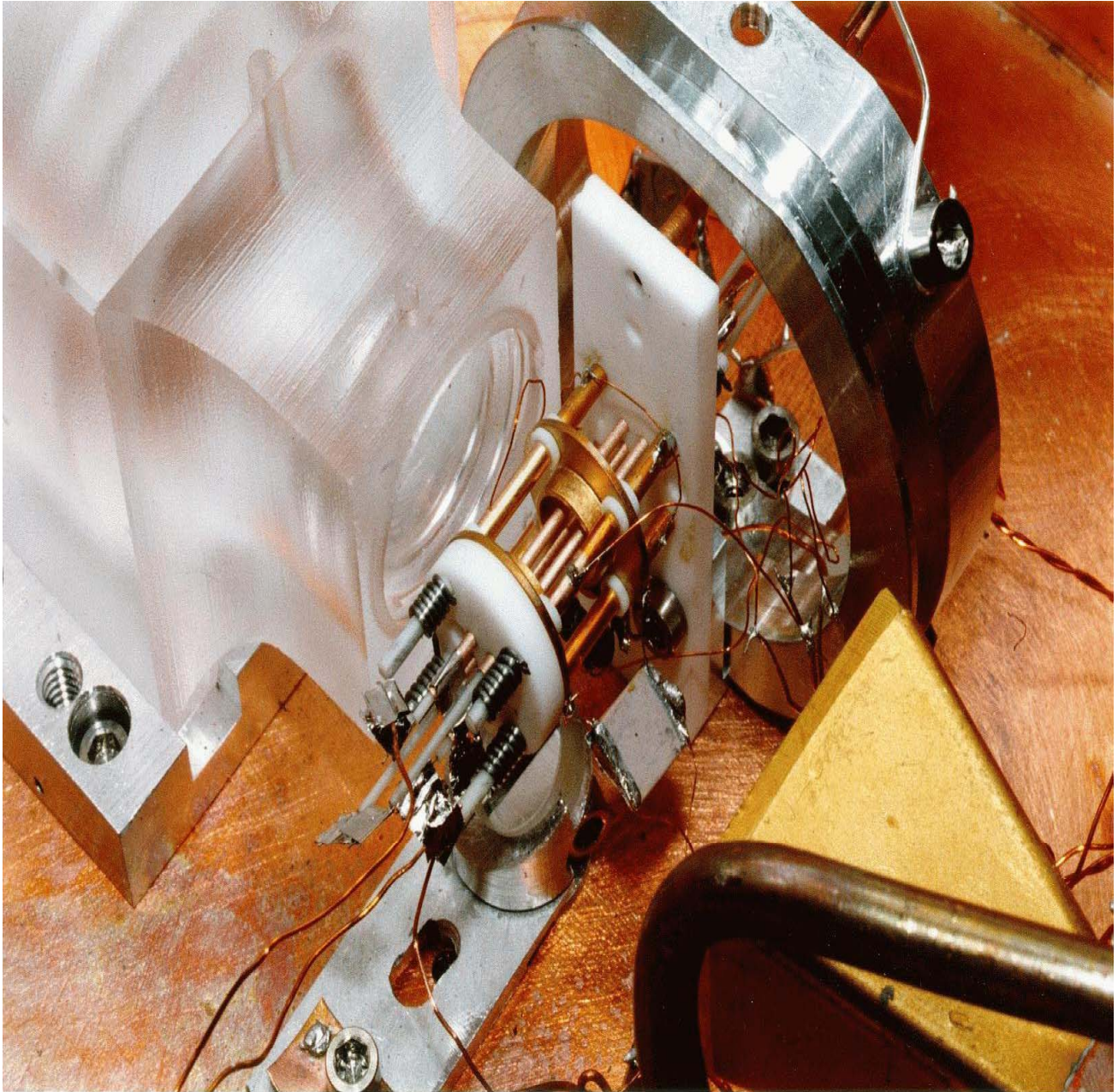


Example: $^{199}\text{Hg}^+$

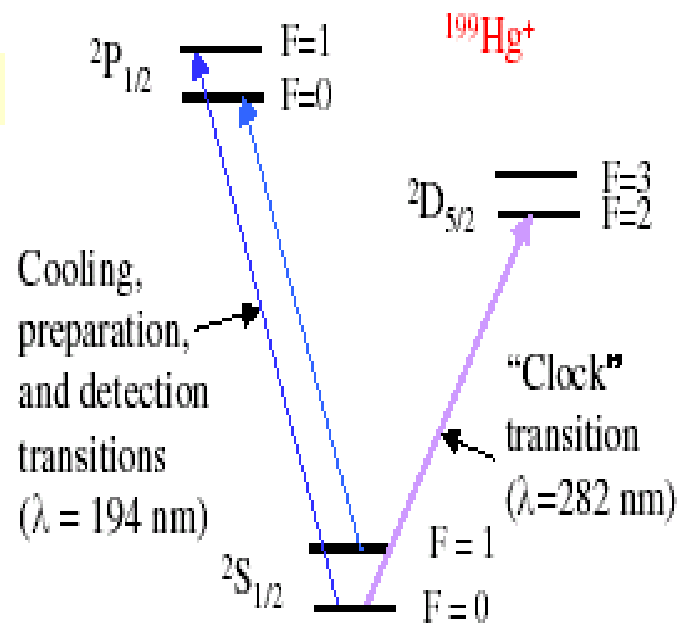
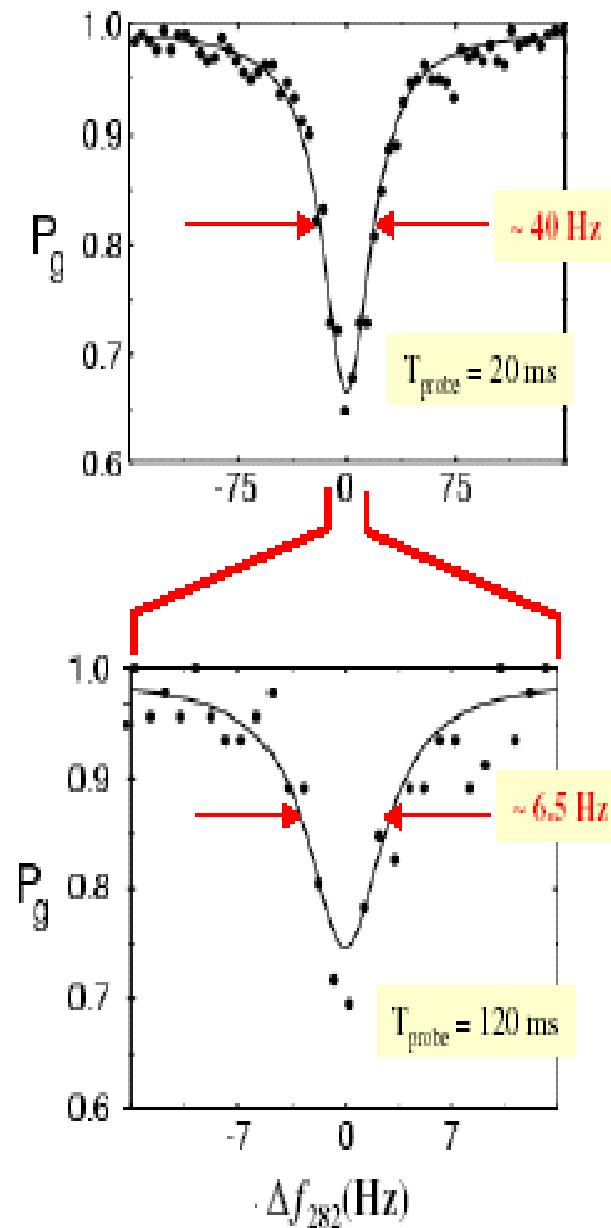
$^{199}\text{Hg}^+$ Energy Levels



Linear ion trap at NIST for Hg^+ optical clock



Quantum-jump absorption spectra of the $^2S_{1/2}(F=0) - ^2D_{5/2}(F=2)$ transition in $^{199}\text{Hg}^+$



Clock transition @
 $f_0 = 1.06 \times 10^{15} \text{ Hz}$

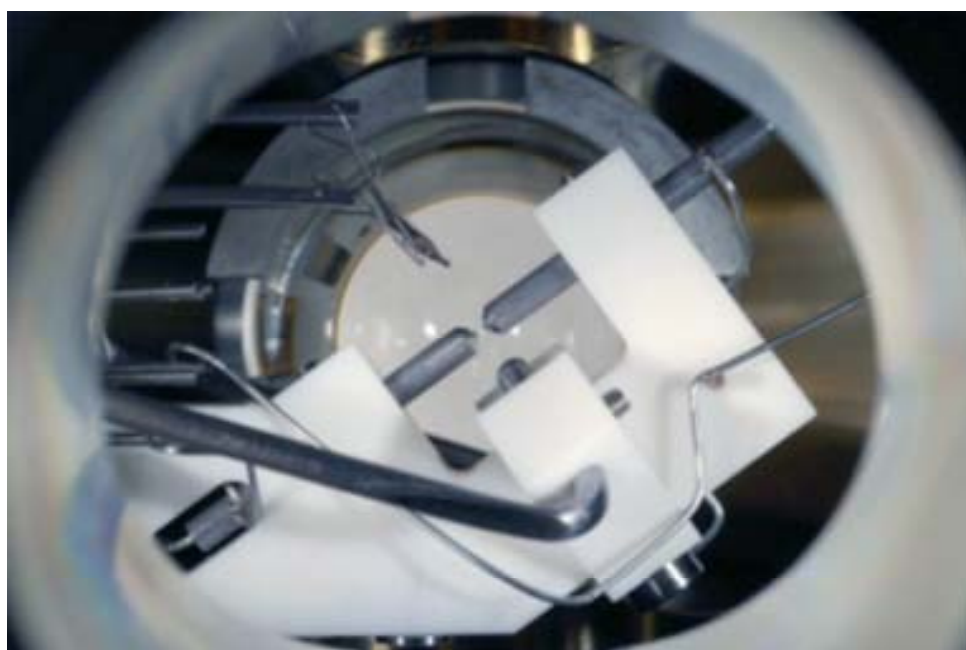
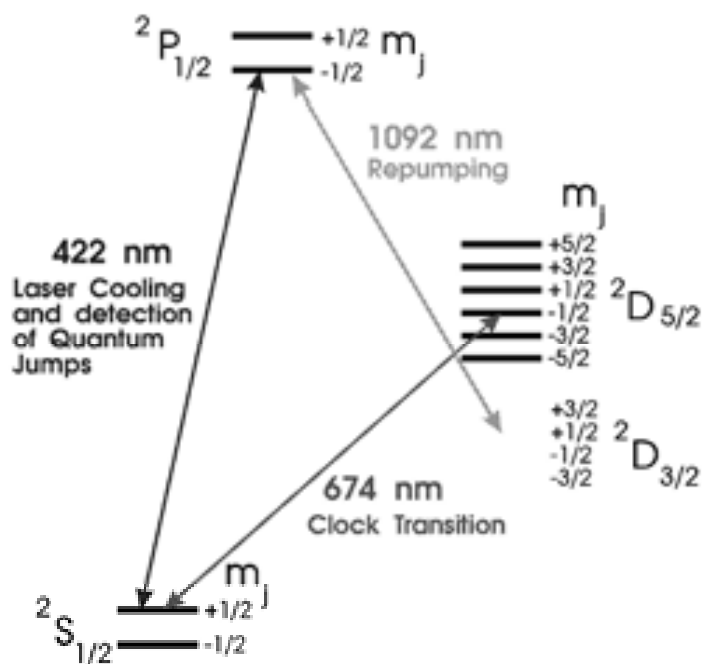
$$\frac{\Delta f_{\text{meas}}}{f_0} \approx 1.5 \times 10^{-15}$$

$(\tau = 15 - 60 \text{ s})$

J. Bergquist et al. (2002)

Work at NRC Canada: single Sr^+ optical clock

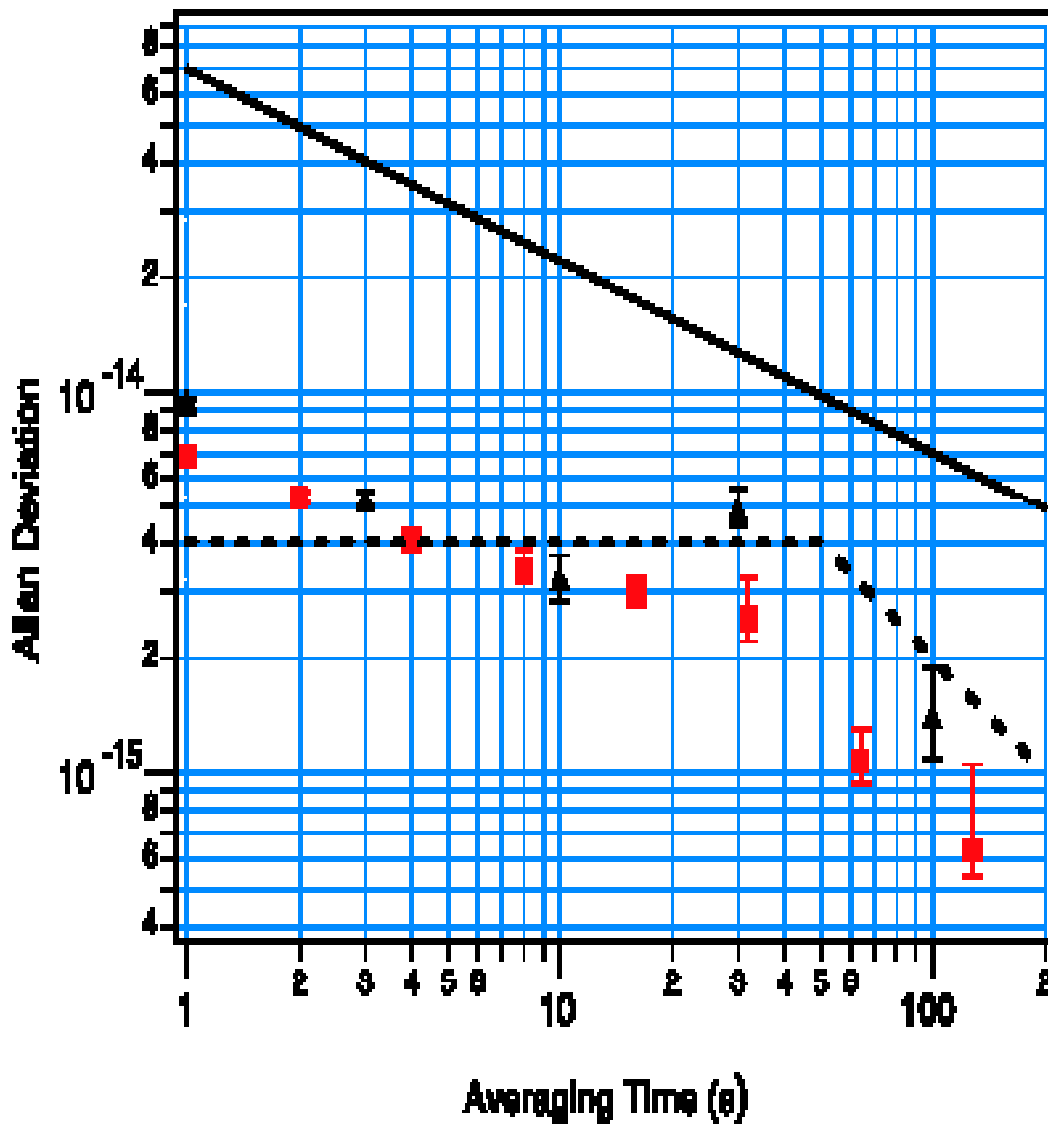
Partial Energy Level Diagram for $^{88}\text{Sr}^+$



Sources of uncertainty in single ion Sr+ clock

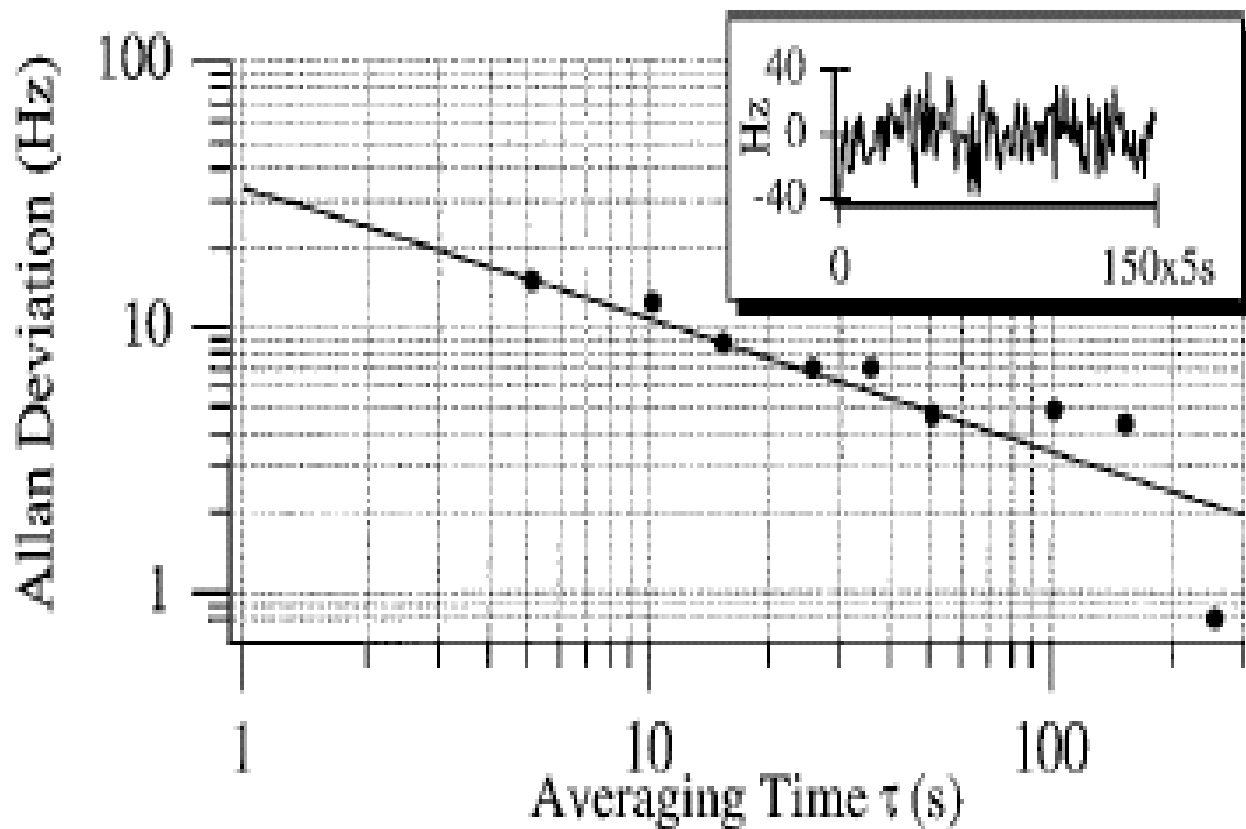
Source	Shift of line center	Magnitude
Second order Doppler effect	0.13 Hz	3×10^{-16}
Quadratic Stark shift	0.2 Hz	5×10^{-16}
Electric quadrupole shift of $4d^2D_{5/2}$ level	<0.5 Hz	$<1 \times 10^{-15}$
Blackbody ac Stark shift	0.16 Hz	4×10^{-16}
ac magnetic fields	<0.2 Hz	$<5 \times 10^{-16}$
Quadratic Zeeman shift (static field)	15 mHz	3×10^{-17}
Collisions	<10 mHz	$<2 \times 10^{-17}$

Measured stability of a single Hg^+ optical standard Cs standard (solid line) for comparison



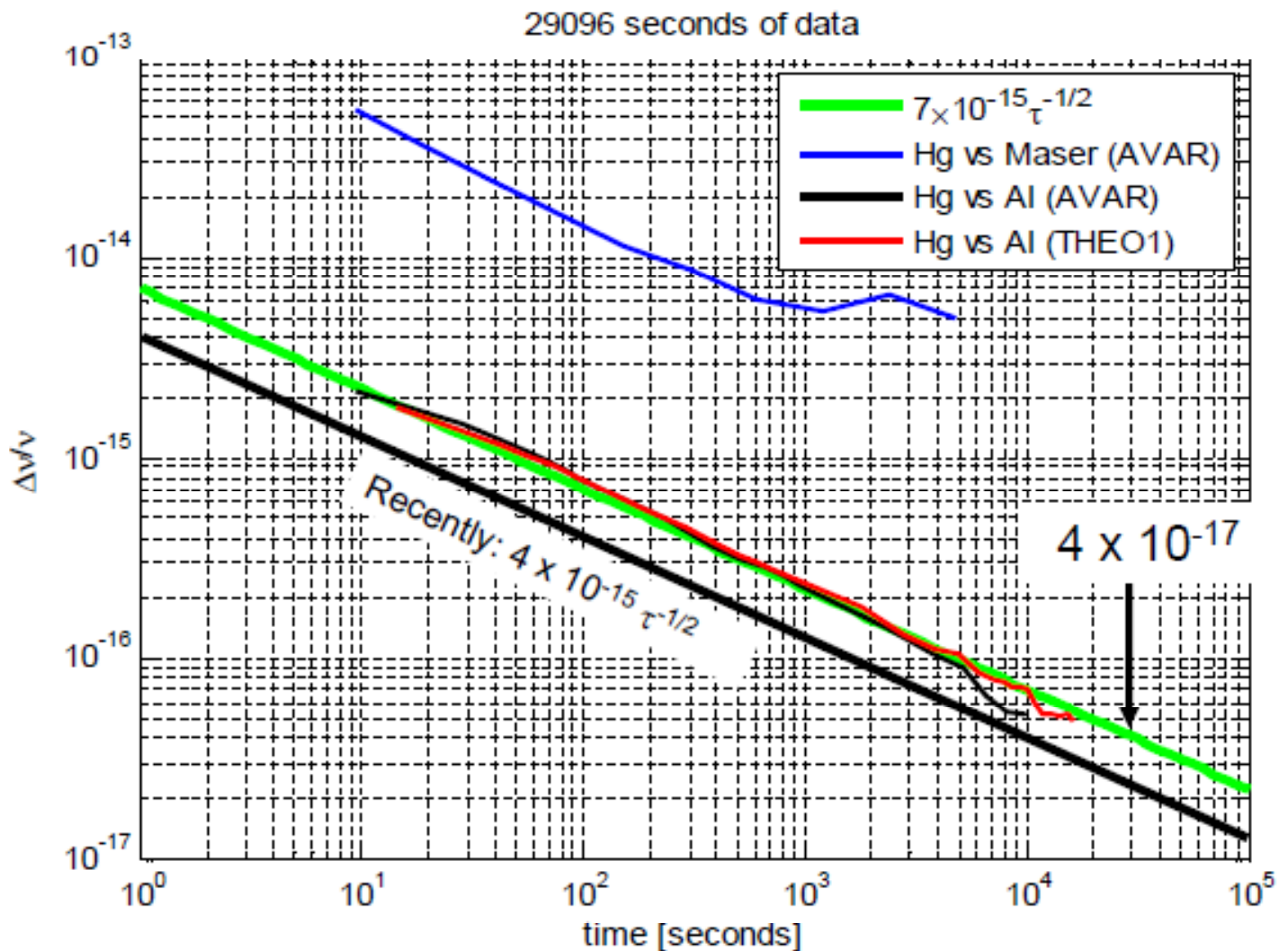
S. Diddams et al., Science 293, 825 (2001)

Frequency stability of Hg⁺ optical standard
vs. Ca standard
(K.R. Vogel et al., Opt. Lett. 26,102 (2001))



$$\sigma(\tau) = 6.4 \cdot 10^{-14} \tau^{-1/2}$$

Al⁺/Hg⁺ Stability



Systematic Frequency Shifts

- The immediate future:
Begin averaging over quadrupole shift

Error budget:

Estimated partial error budget for the near future

Effect	Correction (Hz) (at 1.06 PHz)	Fractional uncertainty $\Delta f/f_0$ (10^{-15})
Second-order Zeeman (B field uncertainty)	1.19	<0.01
$^2D_{3/2}$ quadrupole shift	0	0.01
Gravitational redshift	0.55	0.01
Micromotion shifts	0	0.01

Expected fractional systematic uncertainty: $\sim 2 \times 10^{-17}$

Measuring the frequency of optical transitions

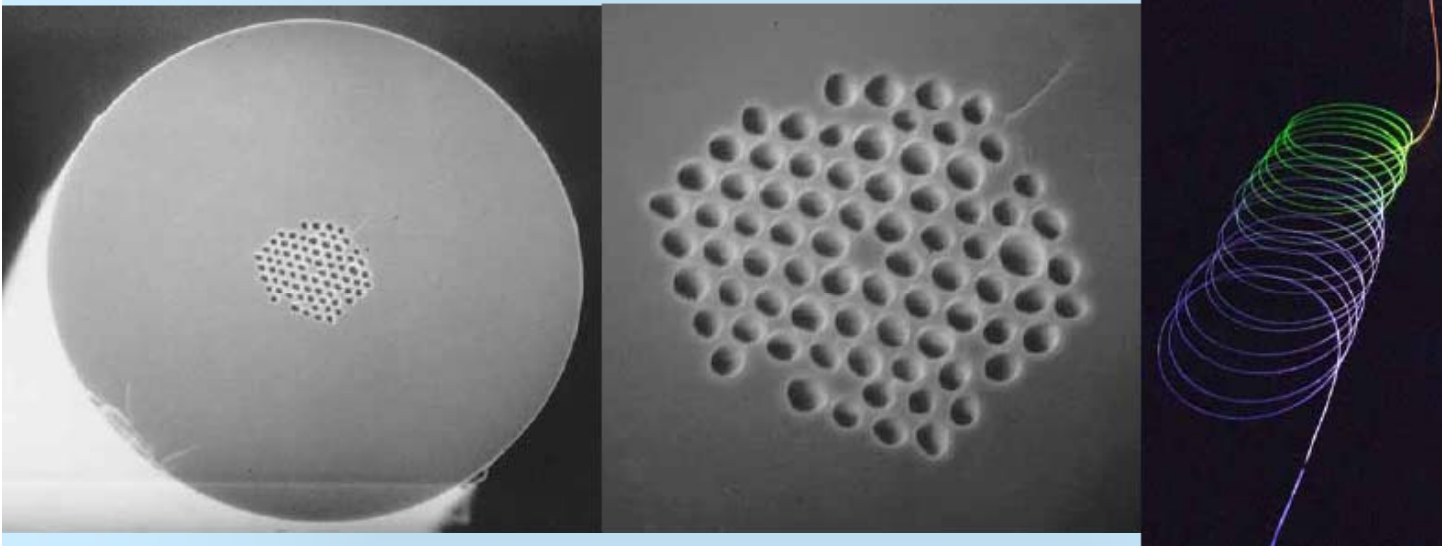
Frequency comb technique

Hänsch, Hall: Nobelprize 2005

Femtosecond Laser into photonic fiber

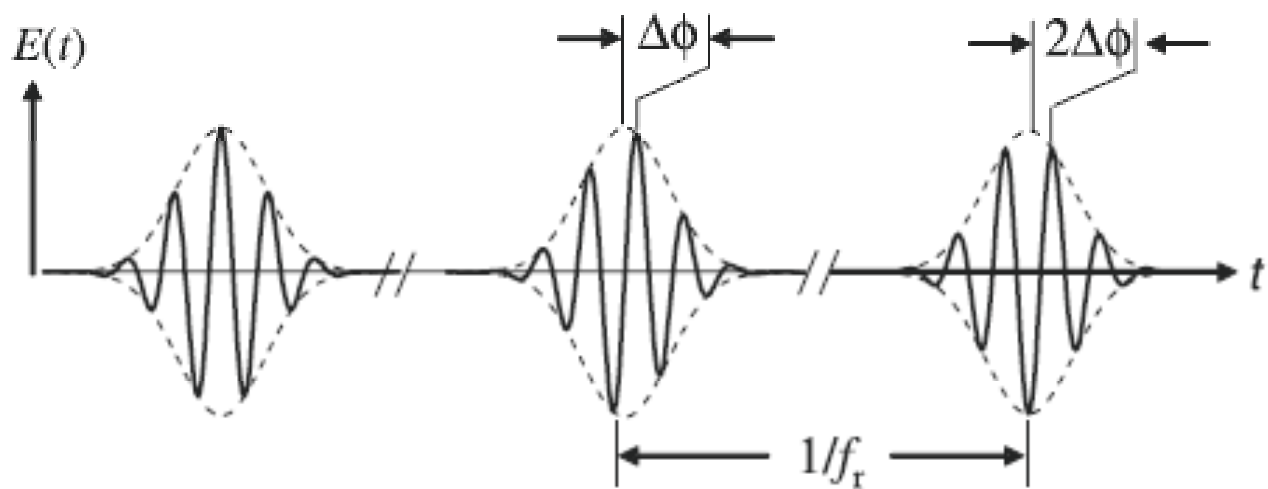
Honeycomb Microstructure Optical Fiber

CLEO, May, 1999

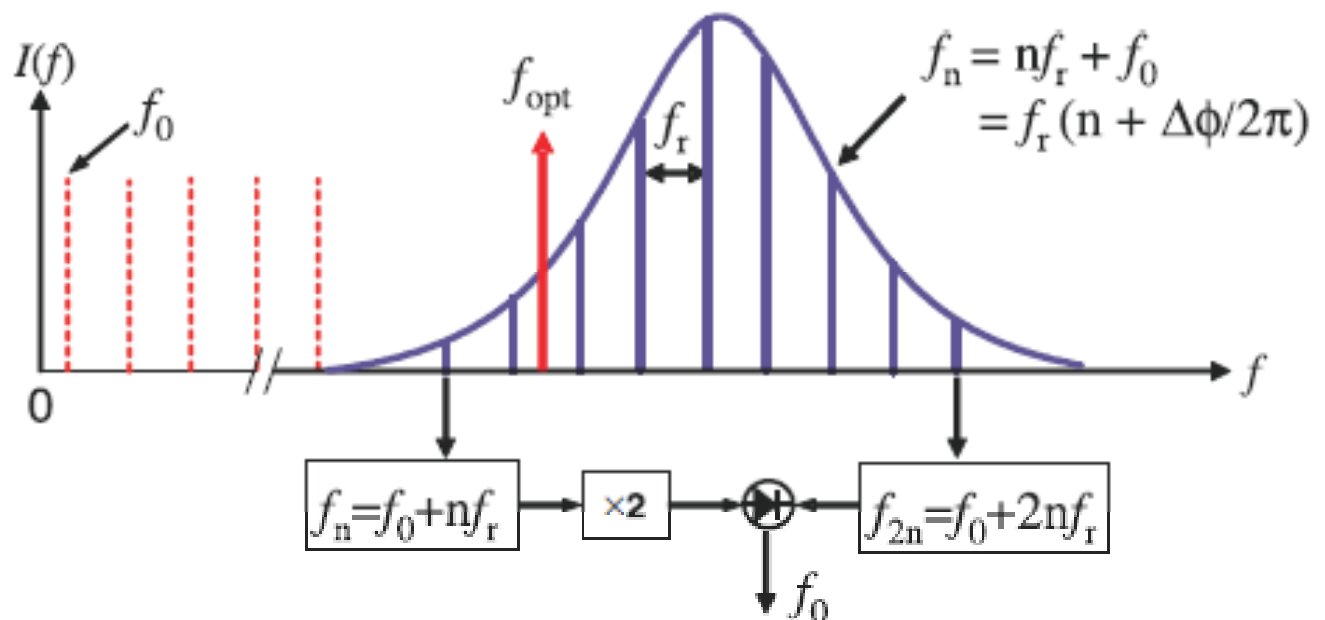


Output of optical fiber:
Discrete wavelength pulses

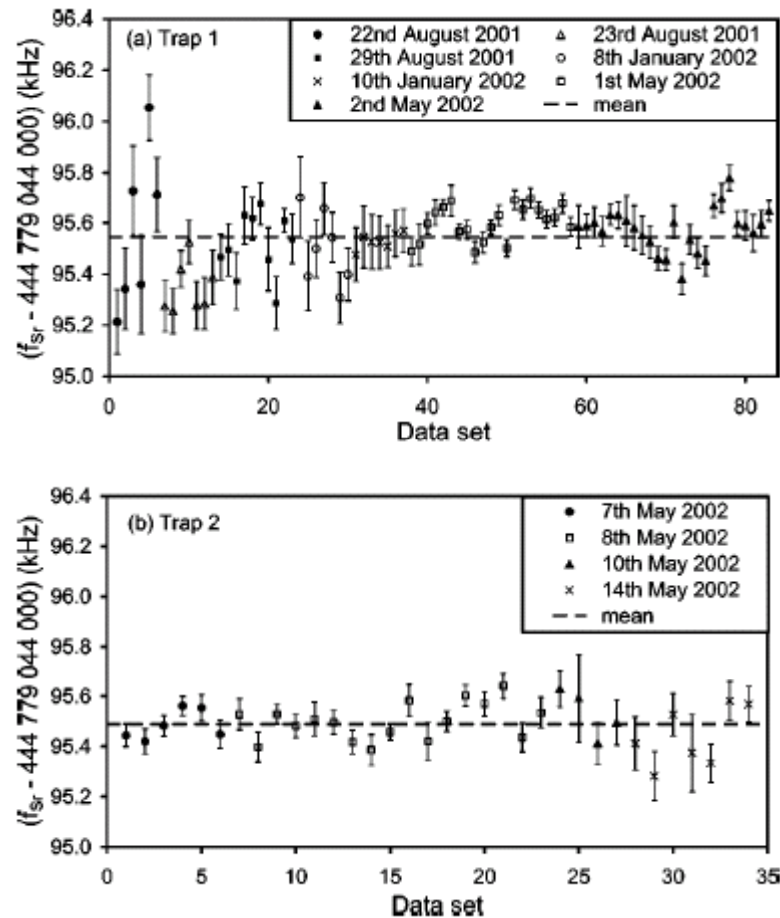
Time domain



Frequency domain



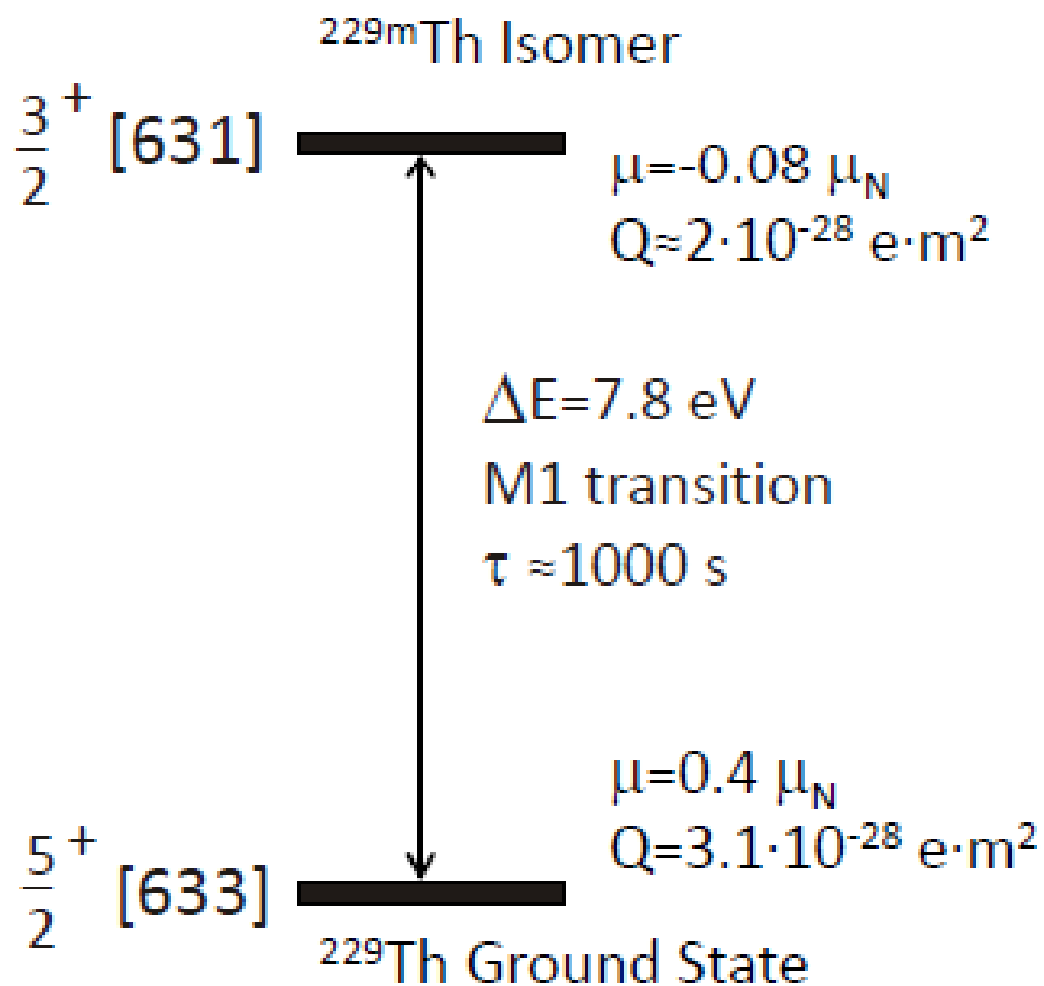
The frequency of the $^{88}\text{Sr}^+$ S-D transition measured in two different traps (NPL, 2003)



Systematic frequency shifts [Hz]

Source	Trap 1		Trap 2	
	Correction	Uncertainty	Correction	Uncertainty
Reproducibility		152		104
422 nm ac Stark shift	-48	60	0	0
1092 nm ac Stark shift	0	<3	0	<3
Servo errors	-3	3	-12	12
Other frequency shifts	0	<1	0	<1
Maser frequency	0	11	0	11
Total uncertainty for each trap		164		105

Potential future nuclear clock with ^{229}Th



The only known isomer with an excitation energy in the optical range and in the range of outer shell electronic transitions.

- Studied by C.W. Reich et al. at INL since the 1970s, established the low energy isomer, from γ -spectroscopy: 3.5 ± 1.0 eV, published in 1994
- Theoretical work by E.V. Tkalya, F.F. Karpeshin, and others isomer lifetime, coupling to electronic excitations ($\tau \sim \text{few } 1000 \text{ s}$)
- False detections of optical emission in the U-233 decay chain in 1997/98
- Proposal of nuclear laser spectroscopy and nuclear clock E. Peik and Chr. Tamm, published in 2003
- Unsuccessful search for optical nuclear excitation or decay
- More precise energy measurement from γ -spectroscopy at LLNL: 7.6 ± 0.5 eV, published in 2007
- 2011: still no direct detection of the optical transition; experimental efforts in several groups worldwide

Possible realizations of Th-229 nuclear clocks:

- Laser-cooled Th^{3+} in an ion trap
- Th ions as dopant in a transparent crystal (like CaF_2 , LiCAF etc.)

Experimental problem:

Transition energy known only to $\approx 10\%$ uncertainty, not a system for high resolution spectroscopy yet.

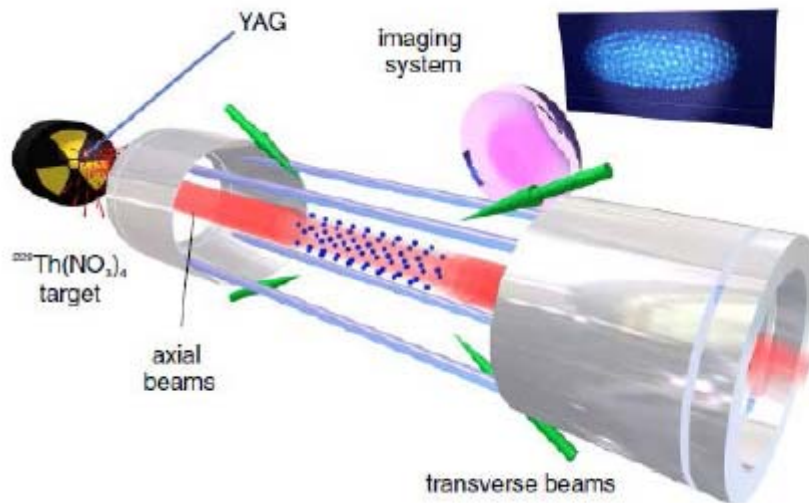
Nuclear clock with laser cooled $^{229}\text{Th}^{3+}$

- Th^{3+} possesses a much more simple level scheme (single valence e^-)
- can be laser-cooled using diode lasers & detected via resonance fluorescence in the red or NIR
- electronic and nuclear resonances are separated in energy

Estimates for long term frequency stability:

10^{-19}

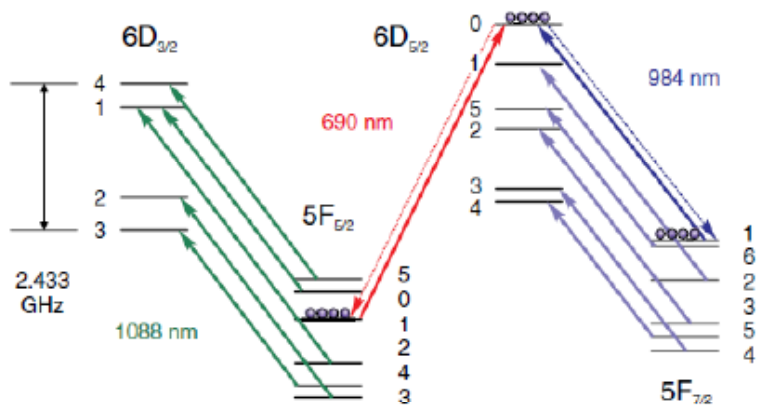
Experiment at PTB Braunschweig



- Loading via laser ablation with ns pulsed Nd:YAG (tripled)
- Trap $L = 188 \text{ mm}$ $r = 3.3 \text{ mm}$, tailored for efficient loading of ablation plume
- Trapping and cooling $10^3 - 10^4 \text{ Th}^{3+}$ ions (Th-229 & Th-231) (enhanced loading efficiency with initial buffer gas cooling)

Campbell et al., Phys. Rev.Lett **106**, 223001 (2011)

Low lying energy levels in $^{229}\text{Th}^{3+}$:

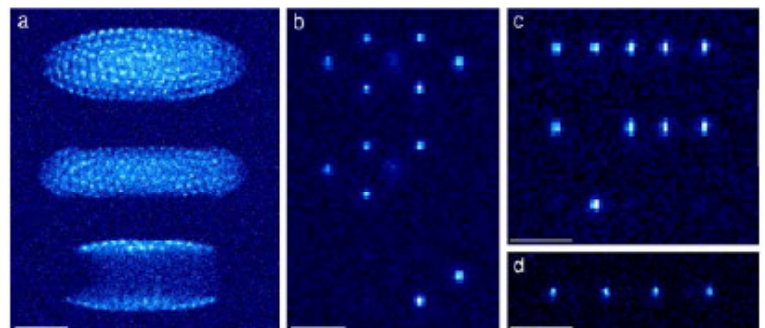


- cooling on 1088 nm line to tens of K
- cooling to tens of mK on lambda scheme
- sympathetic cooling on even isotope (no HF!) for lowest temperatures

Laser cooled ion crystals:

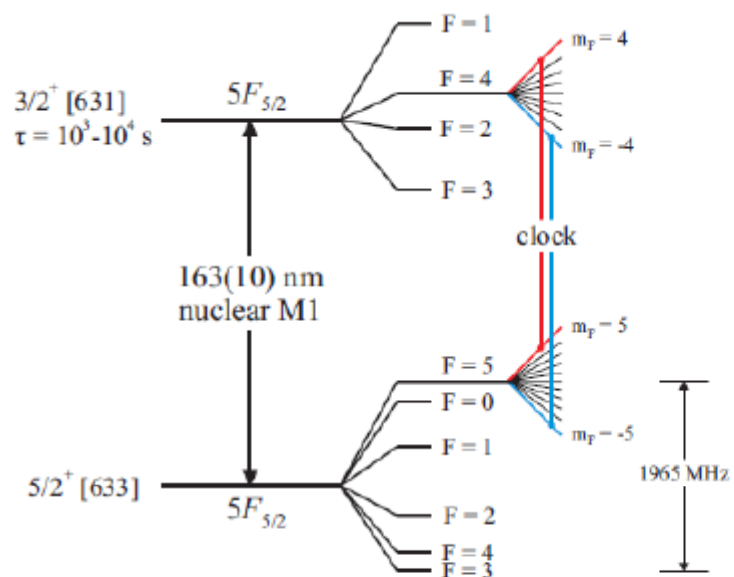
$^{229}\text{Th}^{3+}$

$^{232}\text{Th}^{3+}$



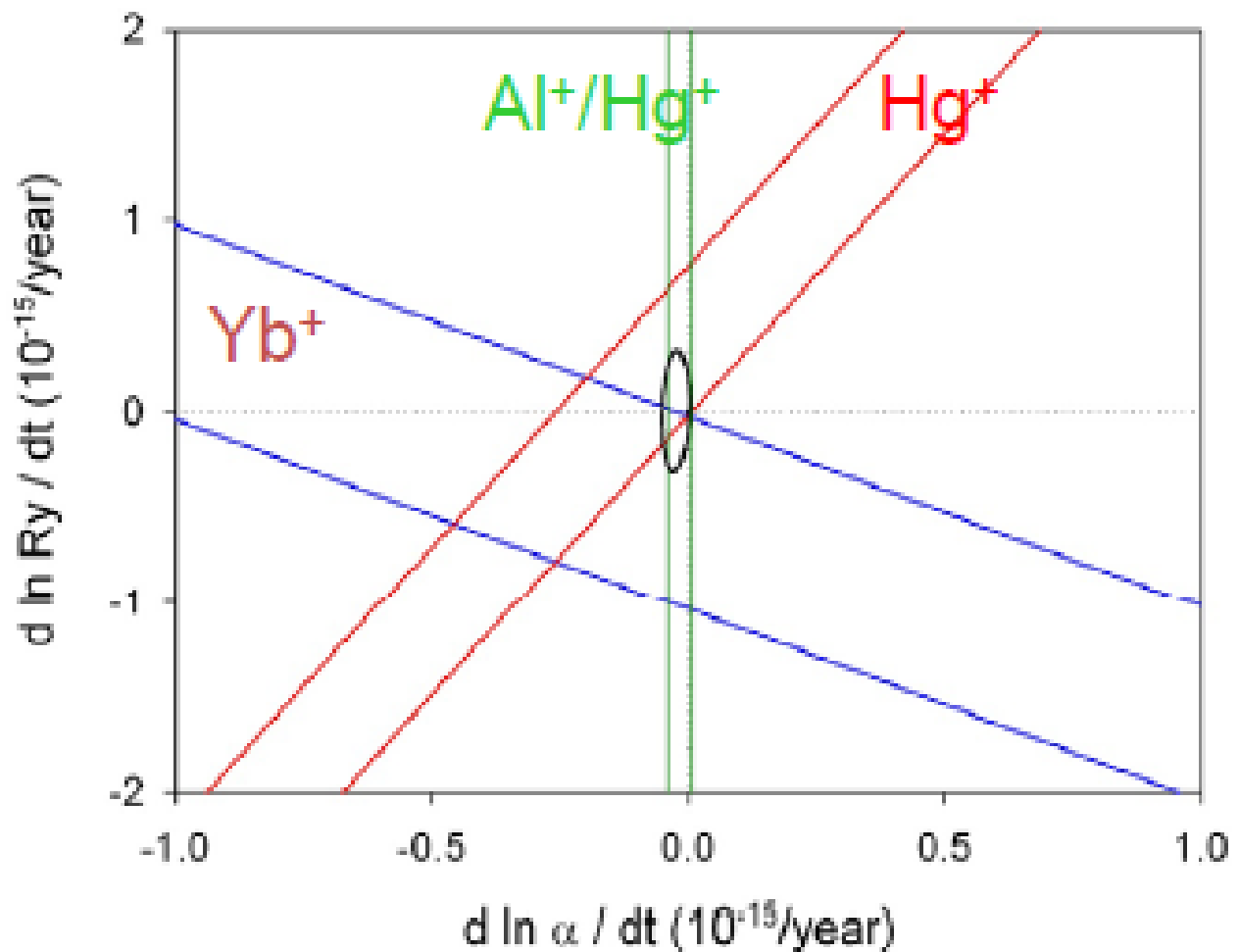
Campbell et al., Phys. Rev.Lett **106**, 223001 (2011)

Clock transition from ground state ($5F_{5/2}$):



With laser cooled and trapped i
fractional frequency inaccuracy
as low as
 10^{-19}
should be possible!

Search for variation of the finestructure constant α in time by clock comparison



Present status:

$$\frac{\partial \ln \alpha}{\partial t} = (-2.4 \pm 2.7) \cdot 10^{-17} \text{ yr}^{-1}$$

$$\frac{\partial \ln Ry}{\partial t} = (0.0 \pm 3.2) \cdot 10^{-16} \text{ yr}^{-1}$$

Summary

Ion trap frequency standards operate in the microwave domain and optical domain

Stabilities below the 10^{-15} level have been reached in both cases

Optical frequency measurements have reached high precision

Further progress is expected with potential nuclear clock