Candidates for cooled and trapped optical standards

– key features for the perfect atom/ion

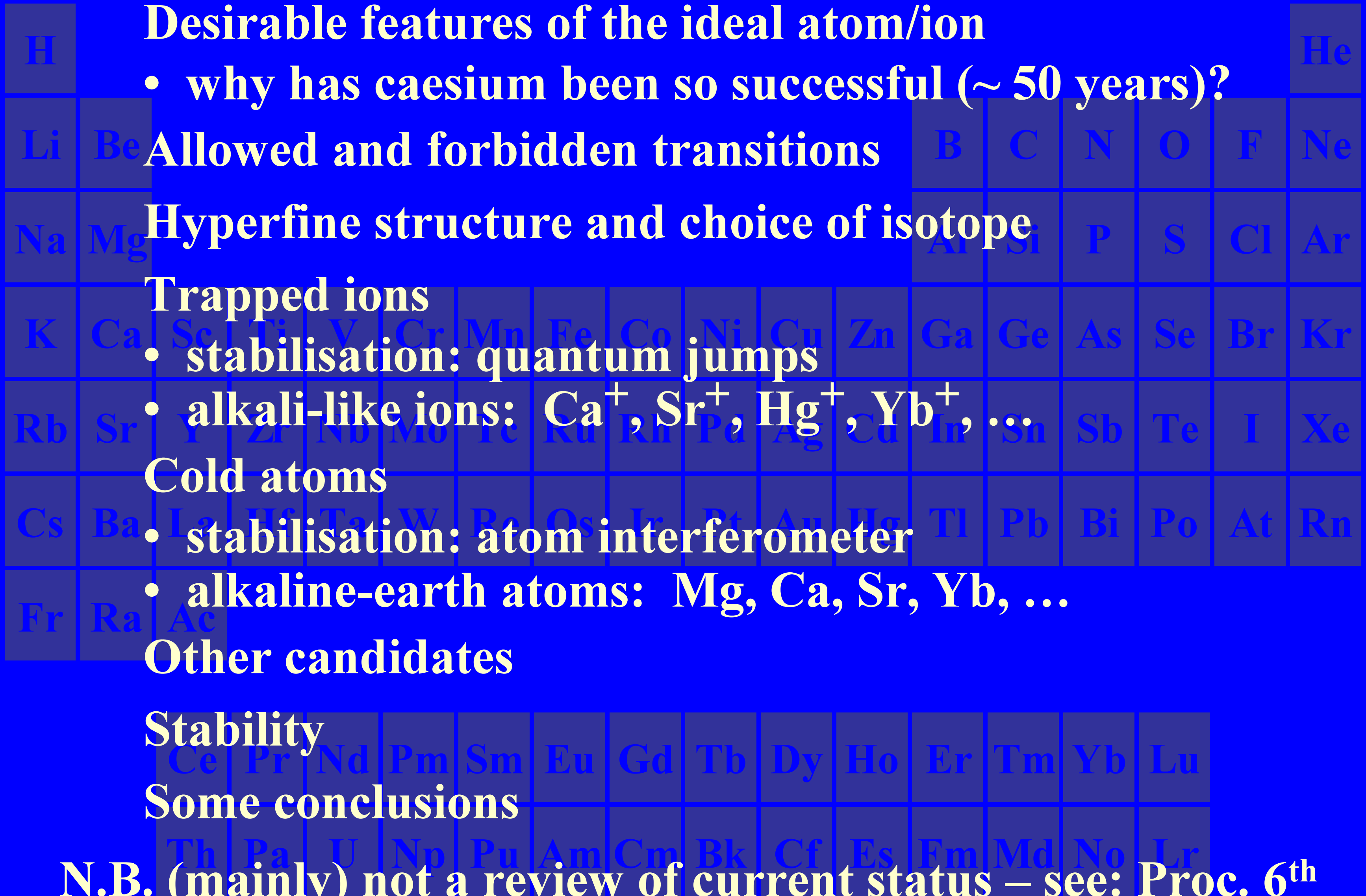
Stephen Lea

National Physical Laboratory, Teddington, Middlesex TW11 0LW U.K.

With thanks to the members of the NPL frequency standards group:

Patrick Gill, Geoff Barwood, Peter Blythe, Witold Chalupczak,
Christopher Edwards, Dale Henderson, Kazumoto Hosaka,
Guilong Huang, Hugh Klein, Valliappan Letchumanan, Helen Margolis,
Mark Oxborrow, Sarika Pugla, Dick Rowley, Alastair Sinclair,
Krzysztof Szymaniec, Stephen Webster, Peter Whibberley

Outline of talk



Desirable features of the ideal atom/ion

- why has caesium been so successful (~ 50 years)?

Allowed and forbidden transitions

Hyperfine structure and choice of isotope

Trapped ions

- stabilisation: quantum jumps
- alkali-like ions: Ca^+, Sr^+, Hg^+, Yb^+, …

Cold atoms

- stabilisation: atom interferometer
- alkaline-earth atoms: Mg, Ca, Sr, Yb, …

Other candidates

Stability

Some conclusions

N.B. (mainly) not a review of current status – see: Proc. 6th Sypm. Freq. Stds. Metrol., ed P. Gill (World Scientific 2002)

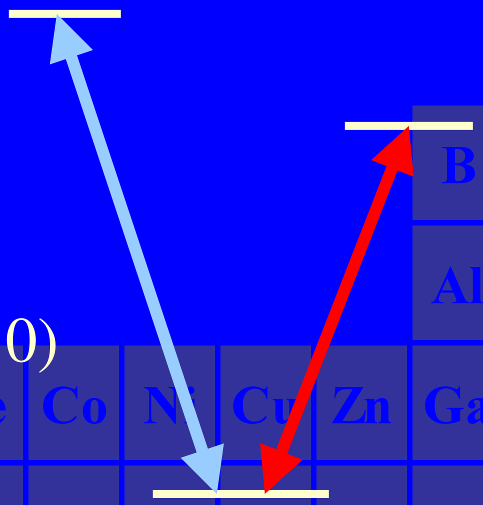
Desirable features of a primary frequency standard

It must …

- show significant gain in accuracy and stability (> × 10) over previous standard
- provide useful output signal
  - – easily compared with existing standards
- be reproducable by (at least) several standards laboratories

Would like …

- to make commercial version
- to relate directly to fundamental constants (quantum standard)



Atoms/ions

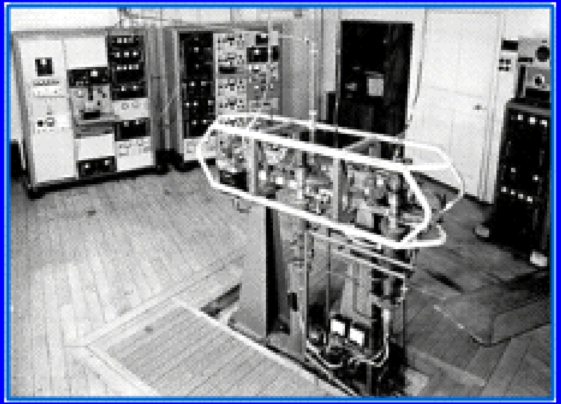
$Q > 10^{14}$

laser linewidth

$\Delta\nu < 1$ Hz

- femtosecond comb
- wavelengths accessible with existing laser technology
- probably challenging…
- theoretical understanding of atomic structure

Caesium – an ideal atom?

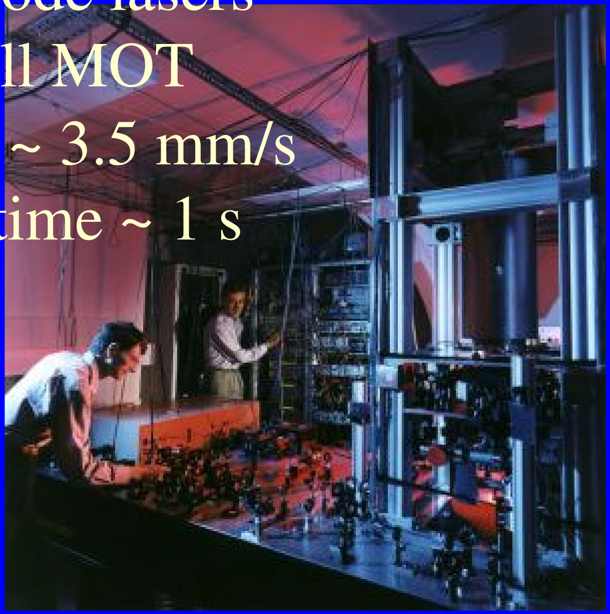


1950s: thermal beam standard

- Easy to make caesium thermal beam
- HFS transition at 9.2 GHz – X-band µW
- Simple enough to make commercial product

1990s: laser cooling – fountain standard

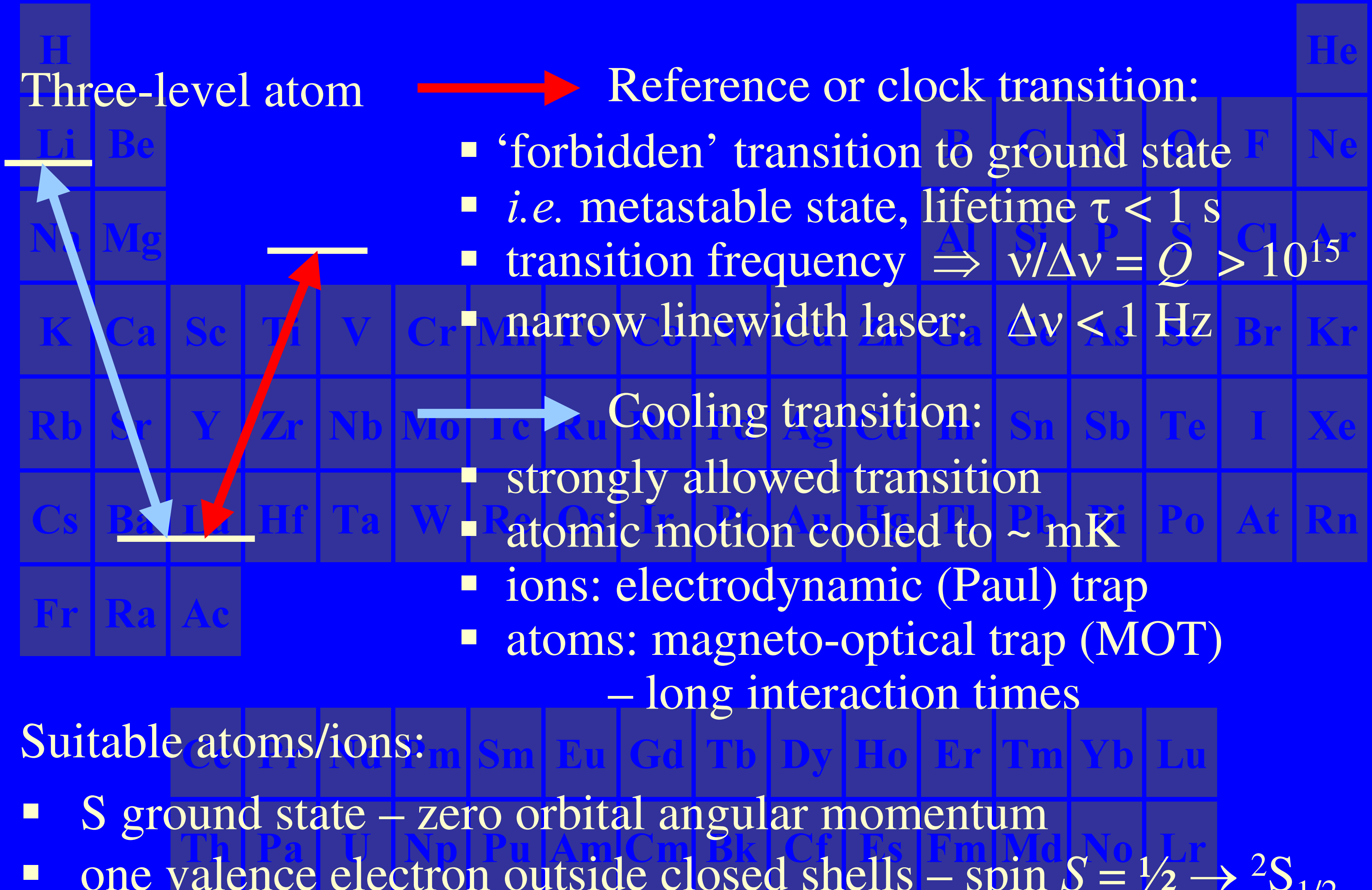
- cooling transition at 852 nm – diode lasers
- low vapour pressure – vapour cell MOT
- HFS → Sisyphus cooling to v_{rms} ~ 3.5 mm/s
- fountain geometry – interaction time ~ 1 s



2000s: present status

- accuracy $\Delta\nu/\nu = 1.0 \times 10^{-15}$
- stability $\sigma_y = 5 \times 10^{-14}\,\tau^{-1/2}$
- limits: collisions; LO stability; QPN

Laser-cooled optical frequency standard



Three-level atom

→ Reference or clock transition:

- ‘forbidden’ transition to ground state
- *i.e.* metastable state, lifetime $\tau < 1$ s
- transition frequency ⇒ $\nu/\Delta\nu = Q > 10^{15}$
- narrow linewidth laser: $\Delta\nu < 1$ Hz

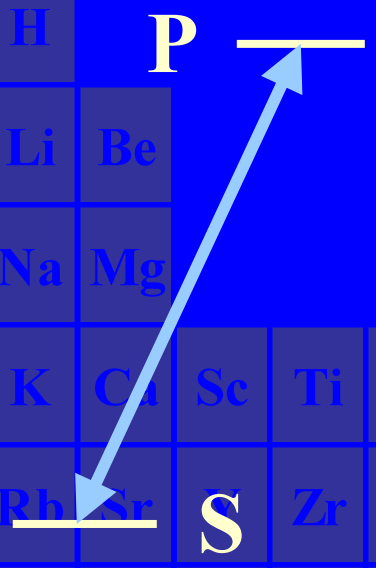
→ Cooling transition:

- strongly allowed transition
- atomic motion cooled to ~ mK
- ions: electrodynamic (Paul) trap
- atoms: magneto-optical trap (MOT)
  – long interaction times

Suitable atoms/ions:

- S ground state – zero orbital angular momentum
- one valence electron outside closed shells – spin $S = ½ \rightarrow {}^2S_{1/2}$
- two valence electrons outside closed shells – spin $S = 0 \rightarrow {}^1S_0$

Cooling transition: allowed electric dipole (*E*1)



Rate: $A = \tfrac{4}{3}\alpha\,\omega^3 \left|\langle \mathrm{S}|\boldsymbol{r}|\mathrm{P}\rangle\right|^2 / c^2$

Angular frequency – Rydberg constant:

$$\omega \sim 2\pi c R_\infty \quad \rightarrow \quad \omega \approx c R_\infty \Leftrightarrow 572\,\mathrm{nm}$$

Matrix element: $\langle \mathrm{S}|\boldsymbol{r}|\mathrm{P}\rangle \sim a_0 \;\rightarrow\; 5a_0$

$$\Rightarrow A \approx 25\alpha c R_\infty^3 a_0^2 = 2\times10^8\,\mathrm{s}^{-1}$$

$$\Rightarrow \tau \sim 5\,\mathrm{ns}, \quad \Gamma \approx 30\,\mathrm{MHz}$$

Selection rules: $\Delta L = \pm1$ (states of opposite parity)

$\Delta S = 0$ (no change of spin)

Doppler cooling:

$$\Gamma \sim 30\,\mathrm{MHz} \quad \Rightarrow \quad T_\mathrm{D} = \hbar\Gamma/2k_\mathrm{B} \sim 1\,\mathrm{mK}, \quad v_\mathrm{rms} \sim 0.3\,\mathrm{ms}^{-1}$$

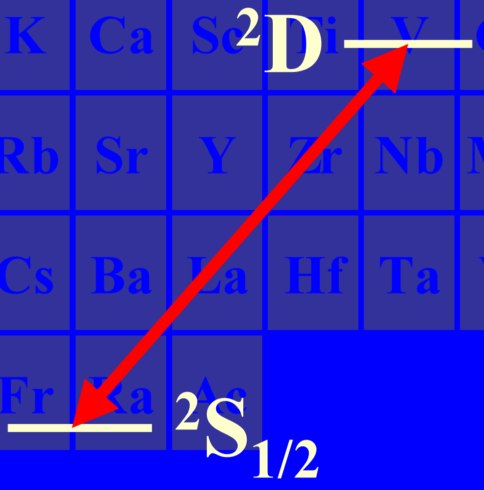
Photon recoil limit – momentum exchange with cooling light:

$$T_\mathrm{recoil} = (h/\lambda)^2/k_\mathrm{B}M_A \sim 1\,\mu\mathrm{K}, \quad v_\mathrm{rms} \sim 0.01\,\mathrm{ms}^{-1}$$

Clock transition: *E*1 forbidden

Electric quadrupole (*E*2) transition

*E*1 forbidden (parity)



Quadrupole matrix element

$$\langle \mathrm{S} | Q | \mathrm{D} \rangle \sim a_0^2$$

$$A \propto \alpha\,\omega^5 \left|\langle \mathrm{S} | Q | \mathrm{D} \rangle\right|^2 / c^4$$

$$\approx \alpha c R_\infty^5 a_0^4 = 3\,\mathrm{s}^{-1}$$

$$\Rightarrow \tau \approx 0.5\ \mathrm{s}, \quad \Gamma \approx 0.5\ \mathrm{Hz}$$

e.g. alkali-like ions: Sr$^+$, Yb$^+$, Hg$^+$…

Clock transition: *E*1 forbidden

Electric quadrupole (*E*2) transition

Two photon (2*E*1) transition

*E*1 forbidden (parity)

2-photon transition via virtual P-states

$$A \propto \alpha\,\omega^5 \sum_{\mathrm{P}} \left|\langle \mathrm{S}|\boldsymbol{r}|\mathrm{P}\rangle\langle \mathrm{P}|\boldsymbol{r}|\mathrm{D}\rangle\right|^2 \Big/ c^4$$

Hydrogen 1S – 2S $A = 8.23\ \mathrm{s}^{-1}$

$$\Rightarrow \tau = 0.12\ \mathrm{s}, \quad \Gamma = 1.3\ \mathrm{Hz}$$

Also atoms with alkali-like structure *e.g.* Ag

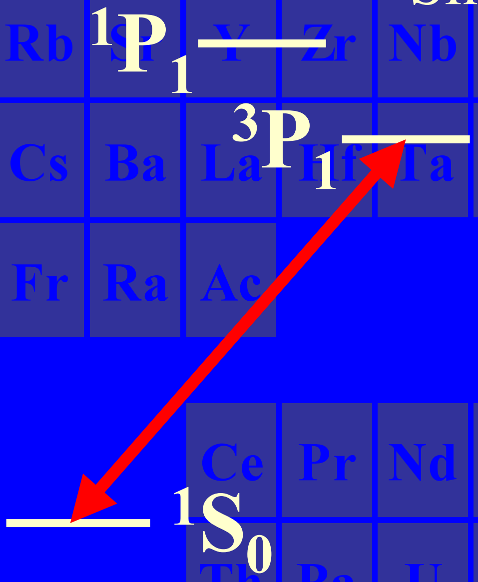
Clock transition: *E*1 forbidden

Electric quadrupole (*E*2) transition

Two photon (2*E*1) transition

Intercombination transition (2-electron systems)

Singlet ($S = 0$) – triplet ($S = 1$) transition – $\Delta S = 0$



$$|\mathrm{P}_1\rangle = |{}^3\mathrm{P}_1\rangle + \xi|{}^1\mathrm{P}_1\rangle, \quad A \propto \xi^2 A(E1)$$

$$\xi^2 \sim [\text{fine structure}/\text{gross structure}] \sim \alpha^2$$

$$A \approx 25\alpha^3 c R_\infty^3 a_0^2 = 10^4\,\mathrm{s}^{-1}$$

$$\Rightarrow \tau \approx 0.1\,\mathrm{ms}, \quad \Gamma \approx 1\,\mathrm{kHz}$$

e.g. alkaline earth atoms: Mg, Ca, … also In^+

The Zeeman effect and hyperfine structure

Linear (first order) Zeeman shift

$\Delta E = m_J g_J \mu_B B \Rightarrow$ No shift for $m_J = 0 \rightarrow m_J' = 0$

1 electron *e.g.* $^2D_{5/2}$ level $\Rightarrow m_J = \pm 1/2, \pm 3/2, \pm 5/2$

2 electrons *e.g.* $^3P_1$ level $\Rightarrow m_J = 0, \pm 1$

Hyperfine splitting: interaction of total electron angular momentum J with nuclear spin I

Mass number A even $\Rightarrow I = 0$, A odd $\Rightarrow I = 1/2$ …

$\Delta E \propto A\mathbf{I.J} \Rightarrow$ Splitting into levels described by F

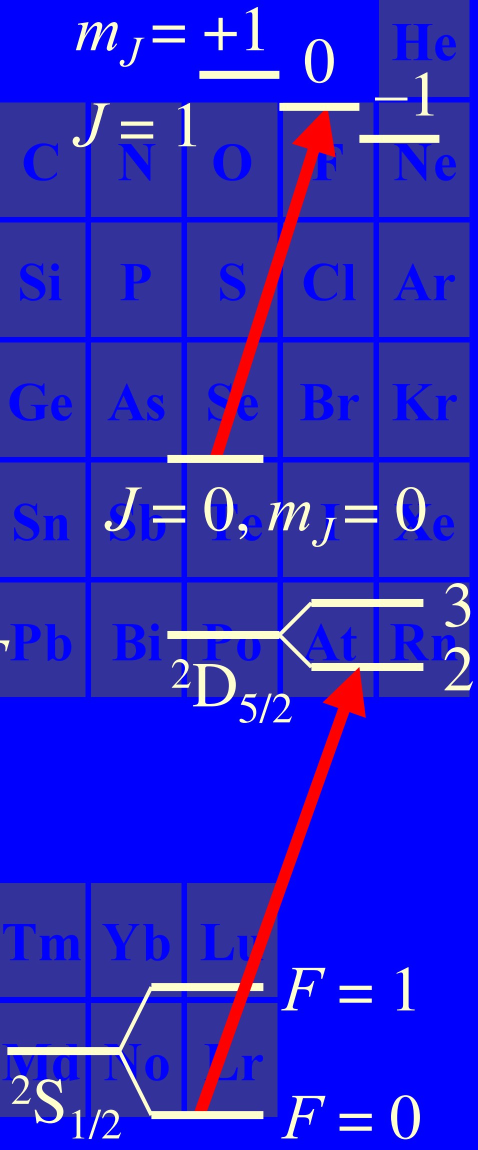
e.g. for $I = 1/2$, $F = J \pm 1/2$

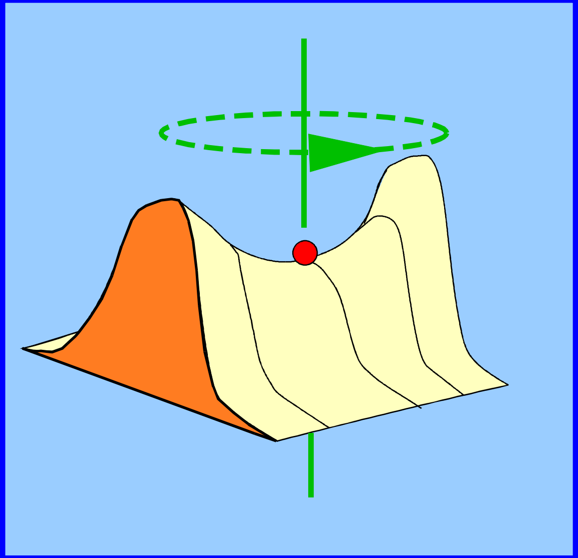
1 electron *e.g.* $^2D_{5/2}$ $F = 2$ level $\Rightarrow m_F = 0, \pm 1, \pm 2$

2 electrons *e.g.* $^3P_1$ $F = 1/2$ level $\Rightarrow m_F = \pm 1/2$

$\Rightarrow$ 1-e systems $\Rightarrow$ odd isotope

2-e systems $\Rightarrow$ even isotope





Trapped single ion

Paul (elecrodynamic) trap, drive frequency Ω_{RF}:

Ion's motion has two components:

Secular motion, $\omega_{r,z} = ½\, \beta_{r,z} \Omega_{RF}$
where β is a function of the trap geometry,
Micromotion at the drive frequency Ω_{RF}.

Typical values:

$\Omega_{RF} \sim 10$ MHz
$\beta_{r,z} \sim 0.2$
$\omega_{r,z} \sim 1$ MHz

Doppler cooling:

$$\Gamma \sim 30\ \mathrm{MHz} \quad \Rightarrow \quad T_\mathrm{D} = \hbar\Gamma / 2k_\mathrm{B} \sim 1\ \mathrm{mK}$$

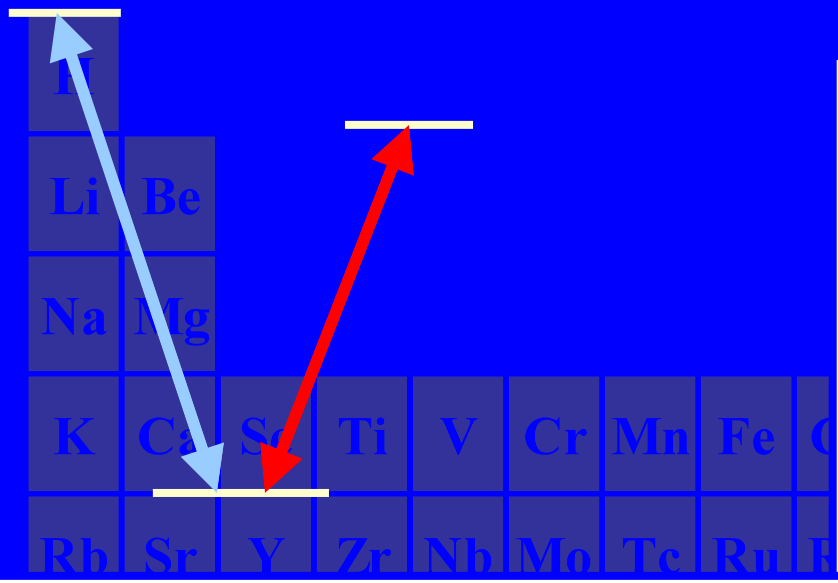
Harmonic oscillator potential:

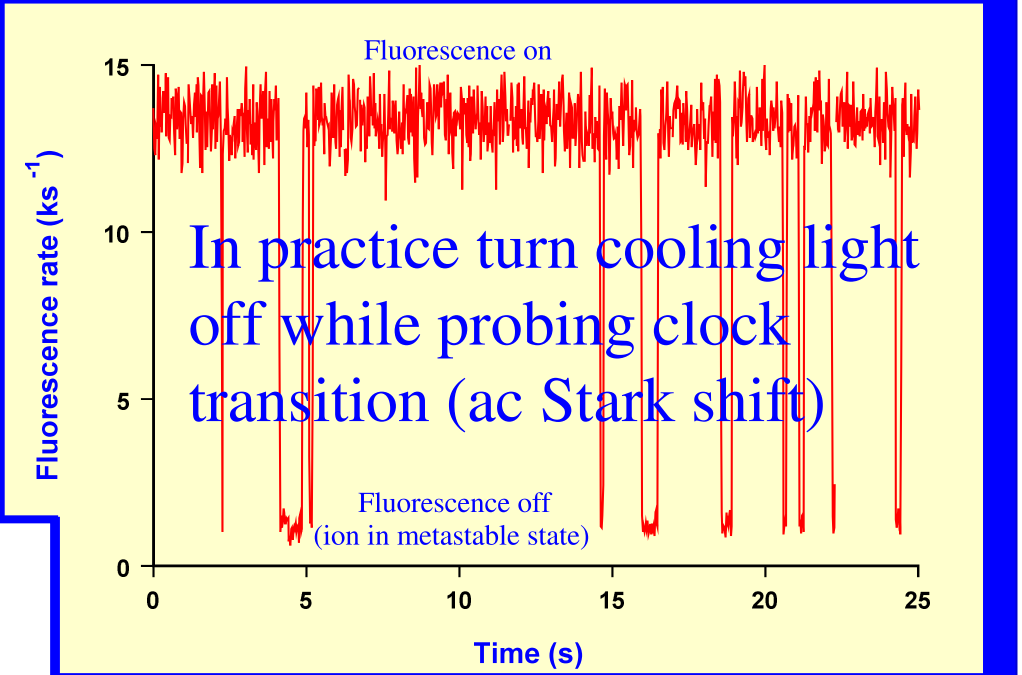
$$E_\mathrm{total} = \tfrac{1}{2}T + \tfrac{1}{2}V = \tfrac{1}{2}M_A\omega^2 x_0^2$$

$$x_0 = \frac{1}{\omega}\sqrt{\frac{\hbar\Gamma}{M_A}} \sim 50\ \mathrm{nm} \qquad \langle n \rangle = \frac{1}{2}\left(\frac{\Gamma}{\omega} - 1\right) \sim 10$$

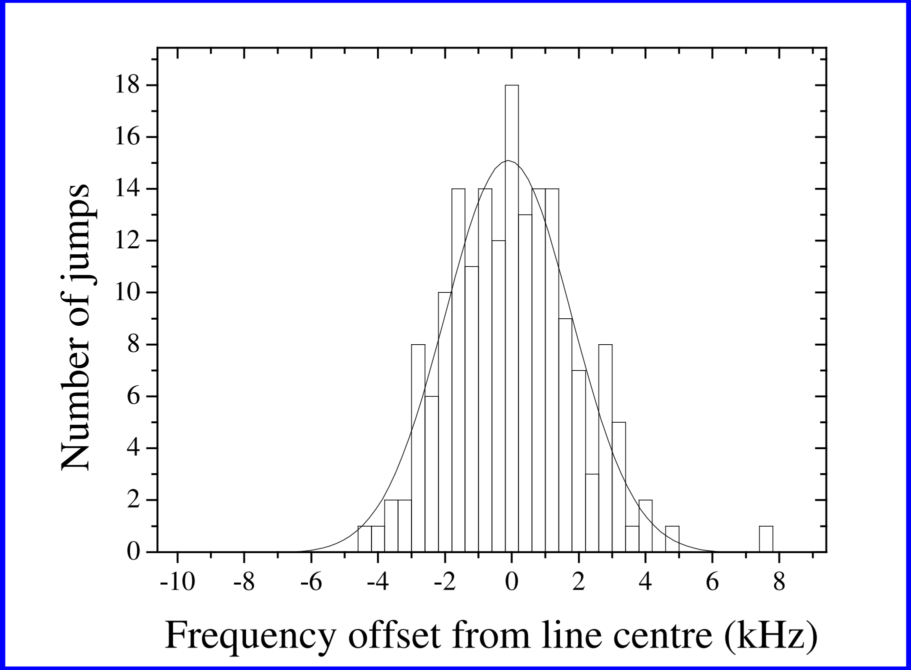
$x_0 < \lambda \Rightarrow$ Lamb-Dicke regime, Doppler shift eliminated

Stabilisation: quantum jumps





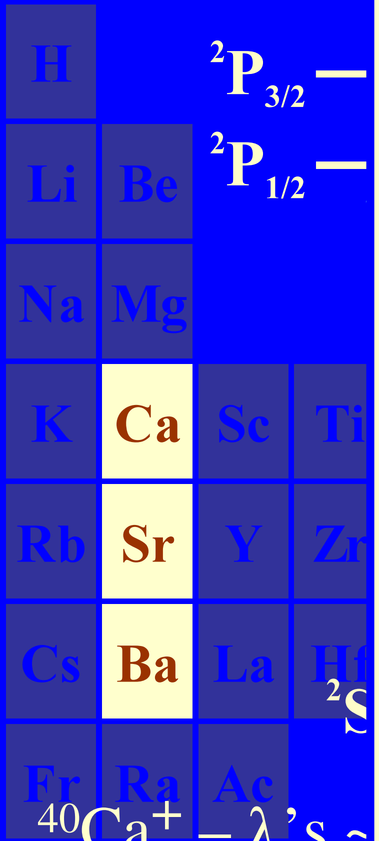
In practice turn cooling light off while probing clock transition (ac Stark shift)



Stabilisation:

Count number of quantum jumps in time $\tau > \tau_D$ at half-height on each side of line centre

Alkali-like ions – quadrupole transitions

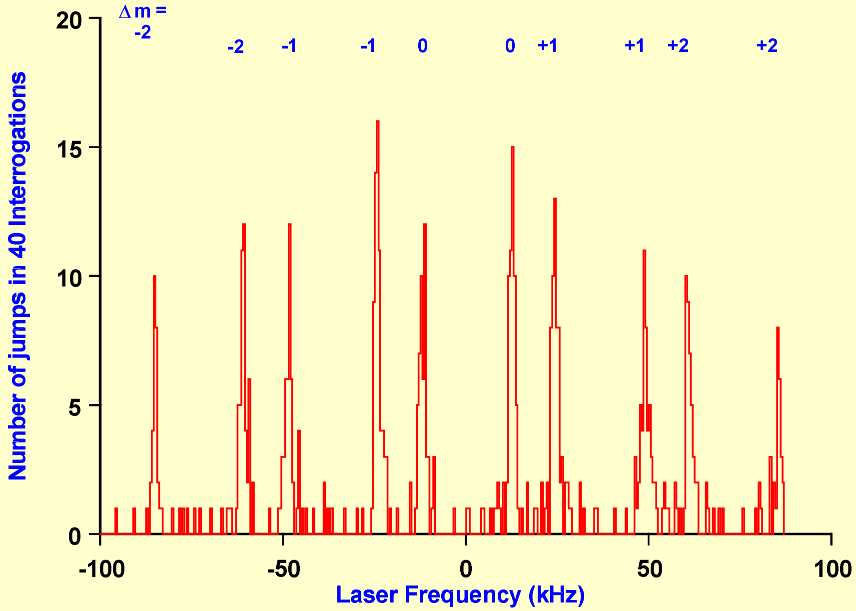


$^2P_{3/2}$

$^2P_{1/2}$

^2S

Zeeman components of the $^2S_{1/2} - {}^2D_{5/2}$ transition at 674 nm in $^{88}Sr^+$



clock nm	Hz
729	0.15
674	0.46
1761	0.003
282	1.6
411	20
435	3.1

$^{40}Ca^+$ – λ's ~

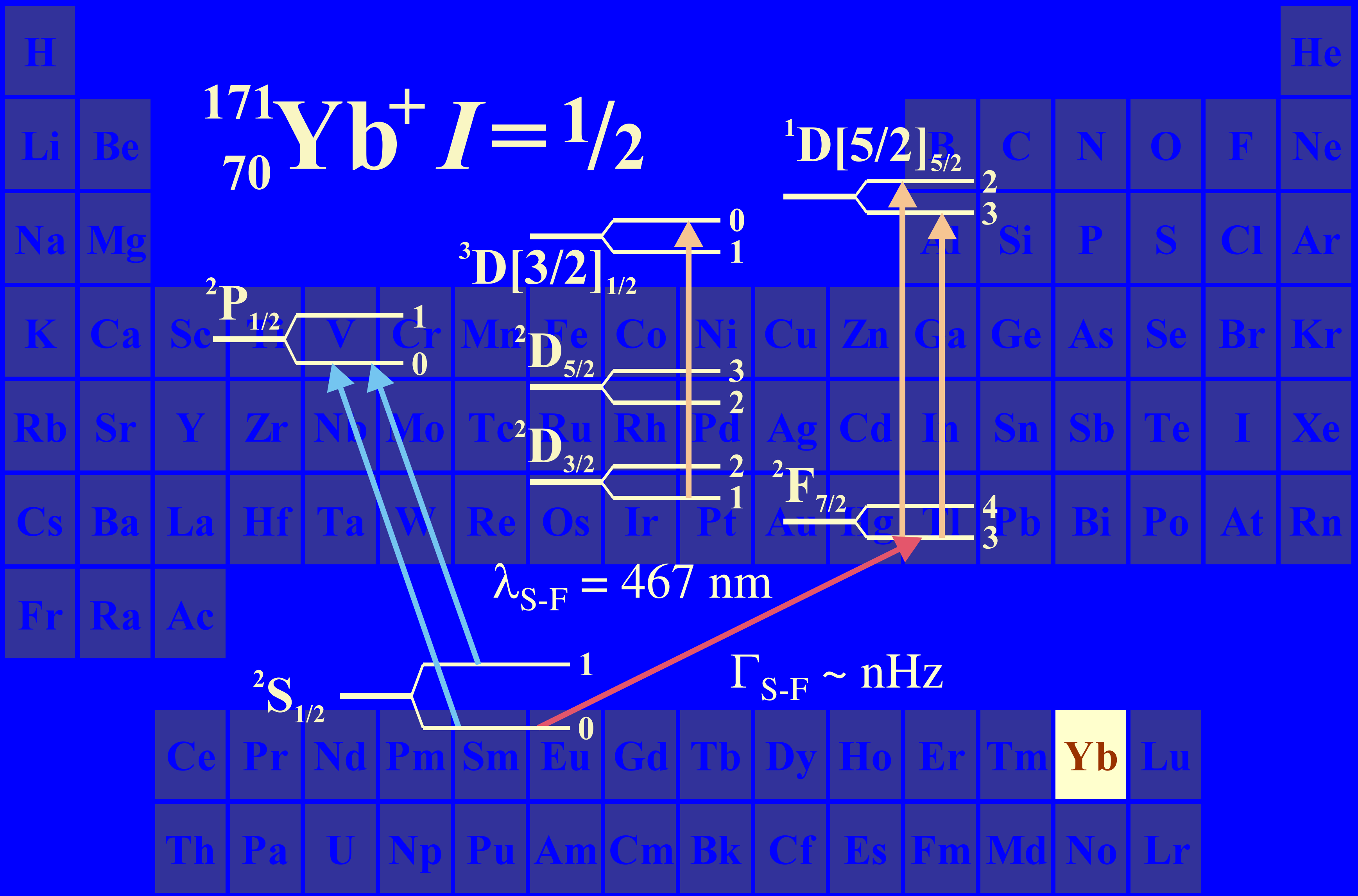
$^{88}Sr^+$ – λ's ~ OK; no HFS ⇒ $m_J \neq 0 \rightarrow 0$ NRC, NPL

$^{87}Sr^+$ – $m_F = 0 \rightarrow 0$ but $I = {}^9/_2$ … NPL

$^{137/138}Ba^+$ – clock λ in the infrared

$^{199}Hg^+$ – λ's hard, cryotrap – but NIST currently world-leading

Alkali-like ions – ytterbium



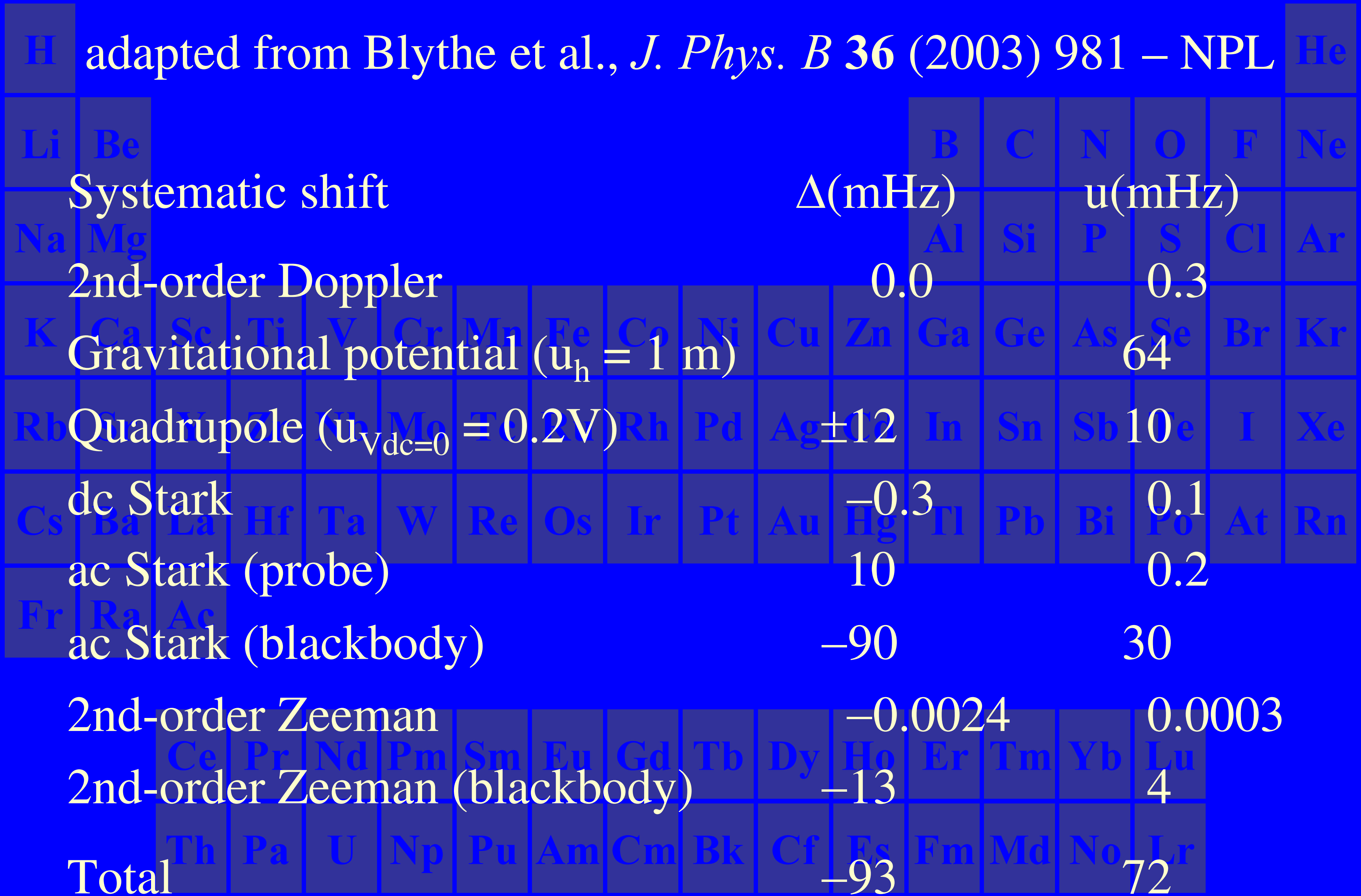
$^{171}_{70}\mathrm{Yb}^+ \; I = 1/2$

$\lambda_{S\text{-}F} = 467$ nm

$\Gamma_{S\text{-}F} \sim$ nHz

^{171}Yb^{+} $^{2}S_{1/2}$ (F=0) – $^{2}F_{7/2}$ (F=3, m_F=0): projected shifts

adapted from Blythe et al., *J. Phys. B* **36** (2003) 981 – NPL

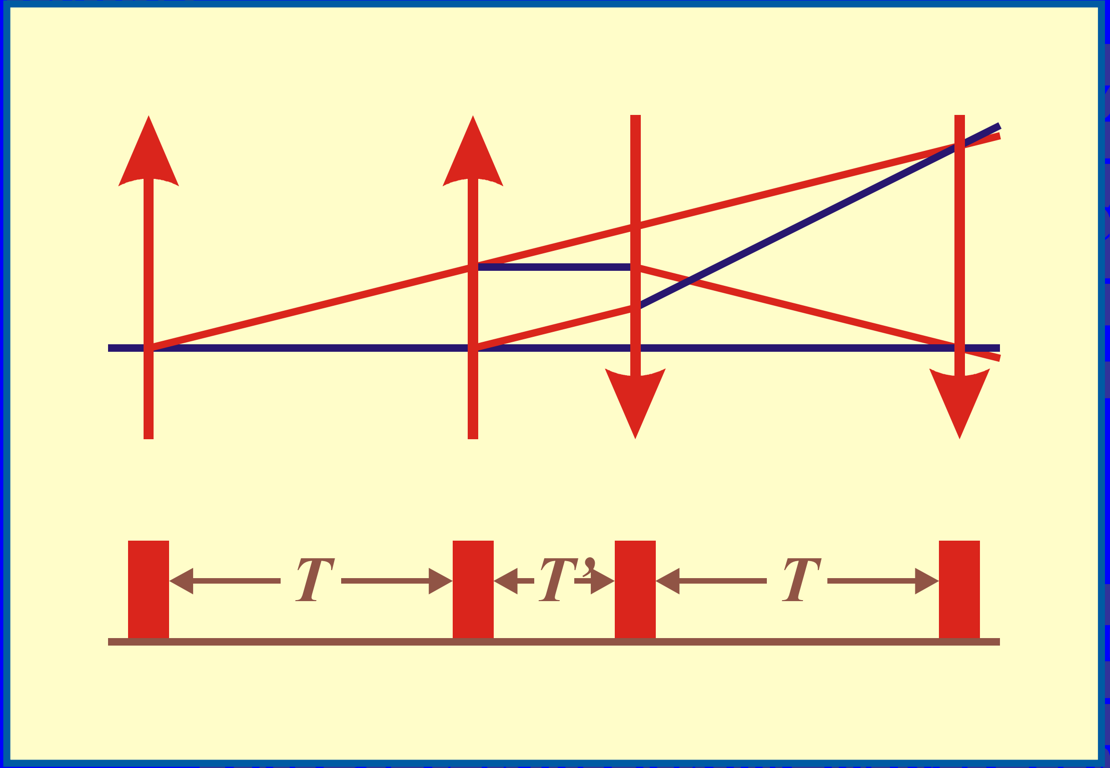


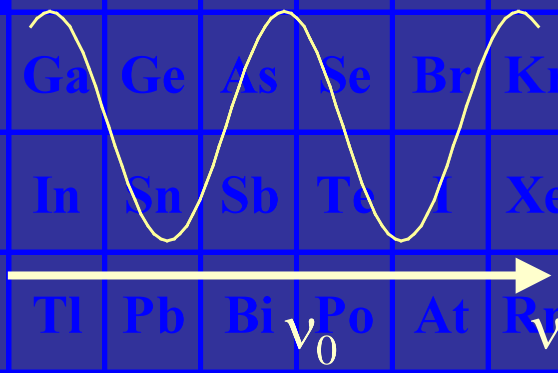
Systematic shift	Δ(mHz)	u(mHz)
2nd-order Doppler	0.0	0.3
Gravitational potential (u_h = 1 m)		64
Quadrupole ($u_{Vdc=0}$ = 0.2V)	±12	10
dc Stark	–0.3	0.1
ac Stark (probe)	10	0.2
ac Stark (blackbody)	–90	30
2nd-order Zeeman	–0.0024	0.0003
2nd-order Zeeman (blackbody)	–13	4
Total	–93	72

Cold atom interferometry

~ 10^7 atoms cooled to ~ few mK in MOT;
then free expansion of atom cloud

Ramsey-Bordé interferometer ⇒ Ramsey fringes

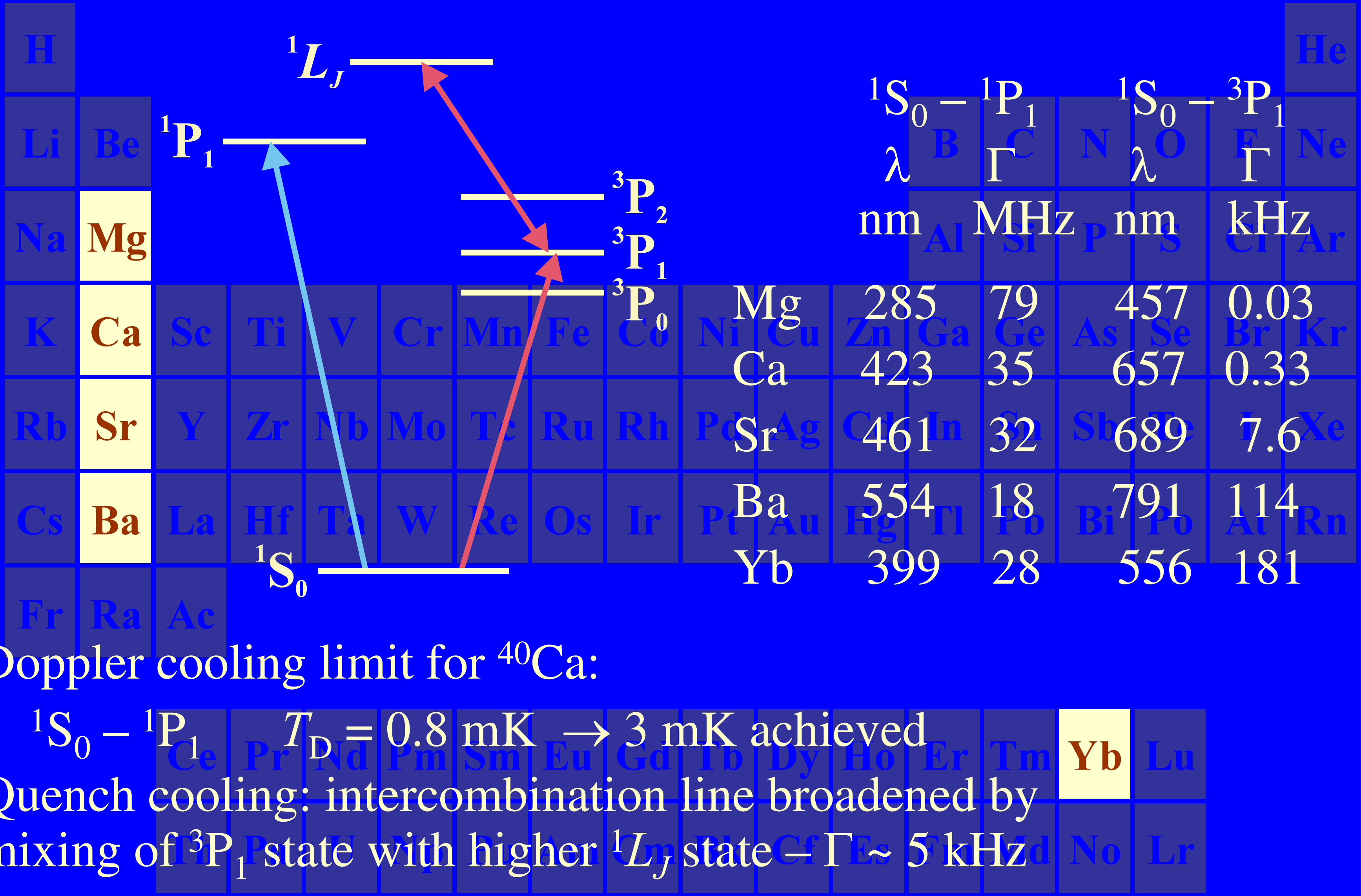




Stabilise to half-height either side of central Ramsey Fringe

Phase uniformity of laser wavefronts ⇒ Doppler shifts

Alkaline earth atoms – intercombination line



	$^1S_0 - {}^1P_1$		$^1S_0 - {}^3P_1$	
	λ nm	Γ MHz	λ nm	Γ kHz
Mg	285	79	457	0.03
Ca	423	35	657	0.33
Sr	461	32	689	7.6
Ba	554	18	791	114
Yb	399	28	556	181

Doppler cooling limit for ^{40}Ca:

$^1S_0 - {}^1P_1$ $\quad T_D = 0.8$ mK → 3 mK achieved

Quench cooling: intercombination line broadened by mixing of $^3P_1$ state with higher ^1L_J state – $\Gamma \sim 5$ kHz

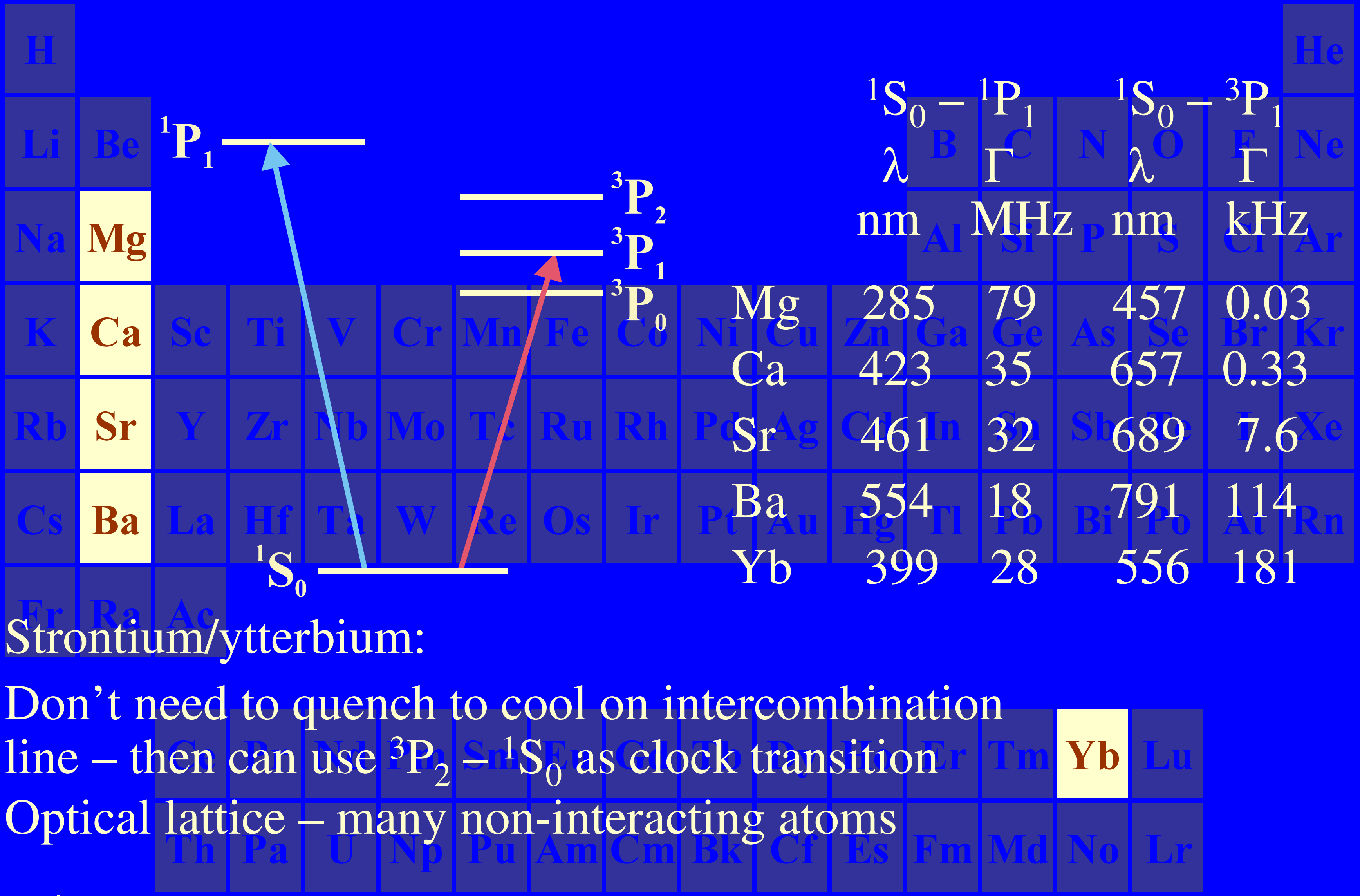
→ $T = 4\ \mu K$ (NIST), $6\ \mu K$ (PTB) ⇔ $T_{recoil} = 2 \mu K$

^{40}Ca $^{1}S_{0} - {}^{3}P_{1}$: projected shifts

from Wilpers et al., *PRL* **89** (2002) 230801
and Helmke (private communication) – PTB

Systematic shift	T = 3 mK (Hz)	6 μK (Hz)
residual 1st-order Doppler	2.6	0.15
2nd-order Doppler	0.005	0.000025
lineshape asymmetry	0.05	0.05
other phase contribution	4	0.10
magnetic field [60 Hz/(mT^2)]	0.1	0.08
quadratic Stark	0.06	0.02
blackbody radiation	4.3	0.05
servo electronics	3.2	0.10
cold atom collisions	1.8	0.26
Total	7.4	0.35

Alkaline earth atoms – intercombination line



	$^1S_0 - {^1P_1}$ λ nm	$^1S_0 - {^1P_1}$ Γ MHz	$^1S_0 - {^3P_1}$ λ nm	$^1S_0 - {^3P_1}$ Γ kHz
Mg	285	79	457	0.03
Ca	423	35	657	0.33
Sr	461	32	689	7.6
Ba	554	18	791	114
Yb	399	28	556	181

Strontium/ytterbium:

Don’t need to quench to cool on intercombination line – then can use $^3P_2 - {^1S_0}$ as clock transition

Optical lattice – many non-interacting atoms

In^+: spin-forbidden $^3P_0 - {^1S_0}$ clock transition – no quadrupole shift

Stability

Allan variance

fractional stability as function of averaging time τ

$$\sigma_y(\tau) = \left\langle \frac{\Delta f_{\mathrm{rms}}}{f_0} \right\rangle_\tau \approx \frac{\Delta f}{\pi f_0}\sqrt{\frac{T}{N\tau}}$$

Δf_{rms} – measured frequency fluctuation

N – number of atoms T – cycle time, with $\tau > T$

Quantum projection noise: statistical fluctuations in

– number of detected atoms (atom interferometer)

– number of detected quantum jumps (ions)

in cycle time T.

e.g. single ion, $N = 1$, $T \approx 1/\Gamma \sim 1$ s, $Q \sim 10^{15} \Rightarrow 10^{-16}\ \tau^{-1/2}$

(calcium) atoms, $N \sim 10^7$, $T \sim 10^{-3}$, $Q \sim 10^{12} \Rightarrow 10^{-18}\ \tau^{-1/2}$

Hydrogen – the simplest atom?

Possibility of a true quantum standard? Second ⇔ R_∞

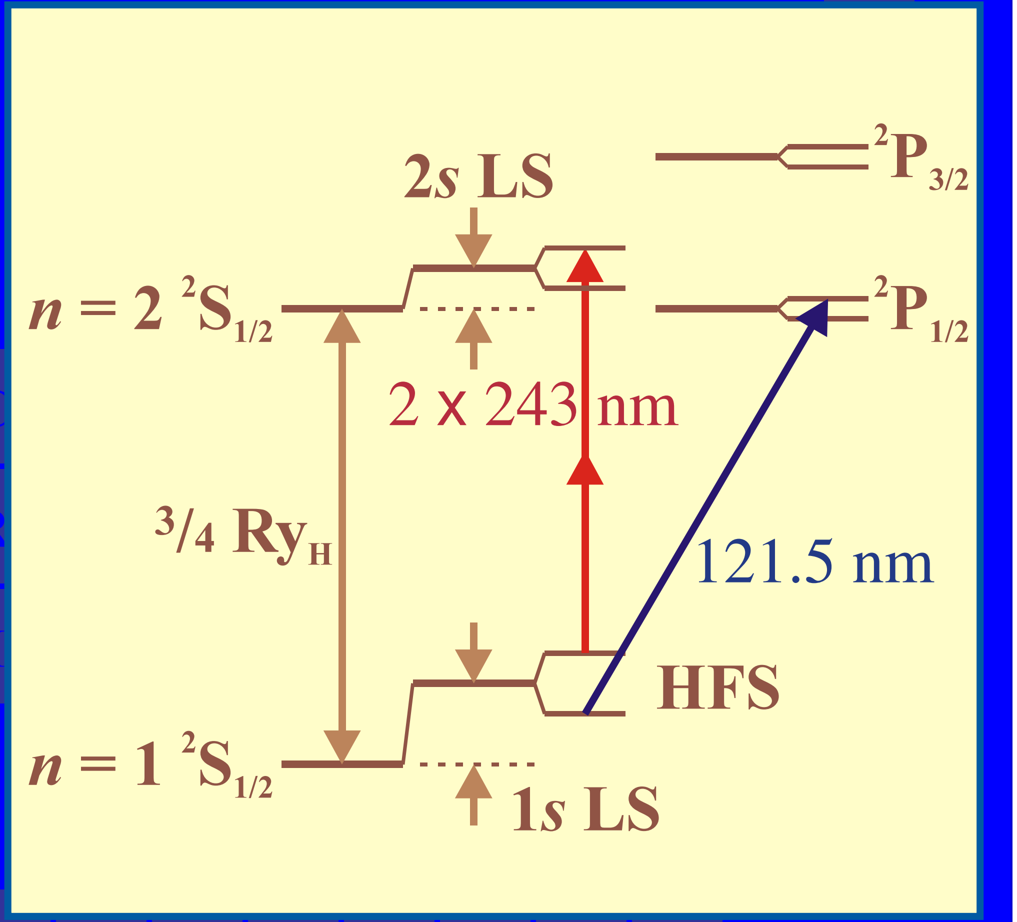
But:

Reduced mass m_e/m_p

Lamb shift – QED

HFS

Proton charge radius



Laser cooling at 121.5 nm – hard, but considerable effort underway for hydrogen/antihydrogen comparison

Other 2-photon transitions – *e.g.* Ag – atomic fountain?

Summary

- alkali-like ions:

$^{88}Sr^+$	NRC, NPL
$^{87}Sr^+$	NPL
$^{199}Hg^+$	NIST
$^{171}Yb^+$	PTB ($S_{1/2}$–$D_{3/2}$) NPL ($S_{1/2}$–$F_{7/2}$)

- alkaline earth atoms:

^{20}Mg	U. Hannover
^{40}Ca	NIST, PTB, U. Hamburg …
^{88}Sr	JILA, Tokyo, BNM-SYRTE, PTB…
^{174}Yb	various

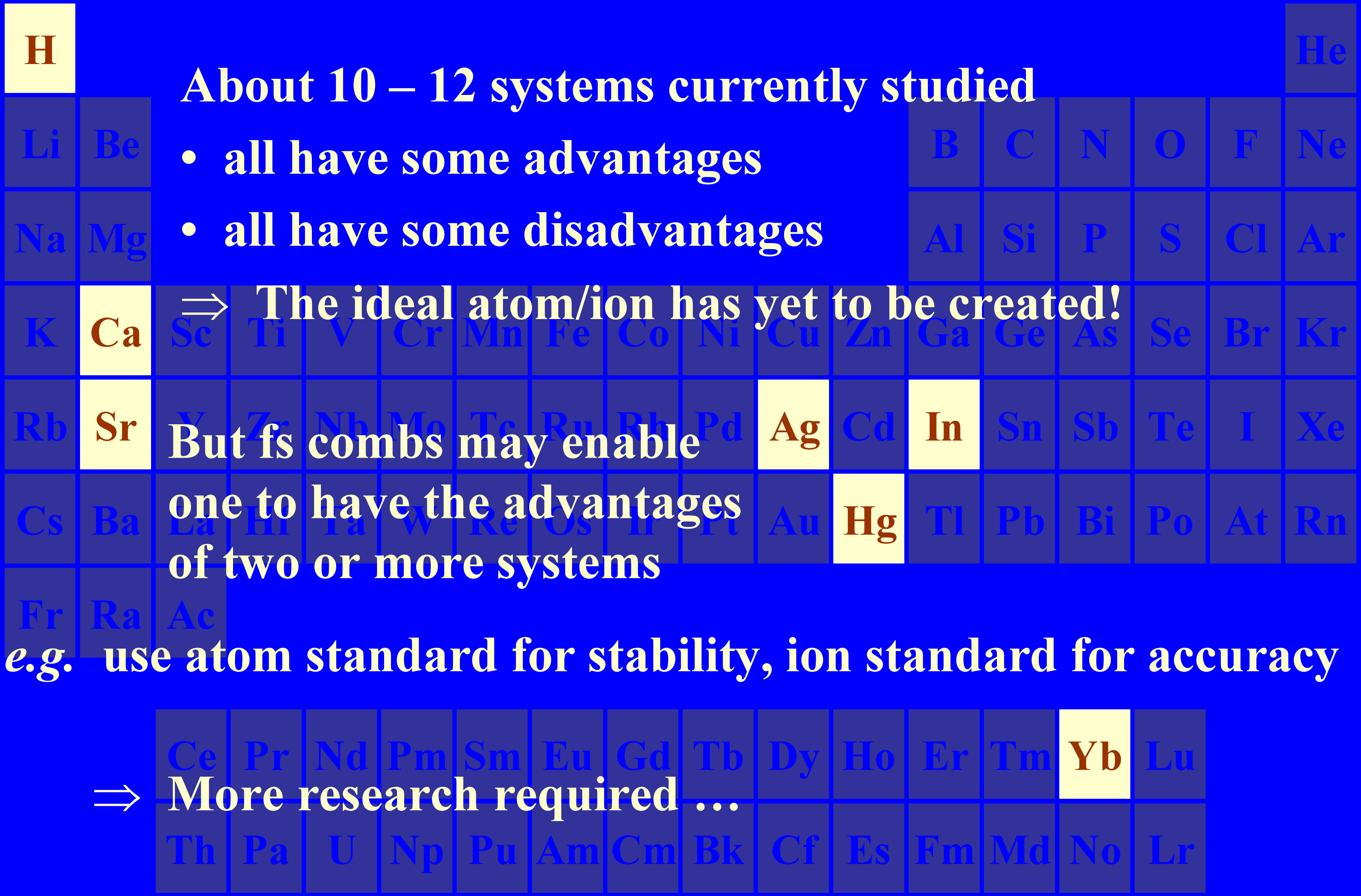
alkaline earth-like ions:

$^{115}In^+$	MPQ, Washington State

- alkali-like atoms:

^{105}Ag	MPQ, BNM-INM
^{1}H	beam: MPQ, U. Paris VI cooling: MIT

The ideal atom/ion?



About 10 – 12 systems currently studied

- all have some advantages
- all have some disadvantages

⇒ The ideal atom/ion has yet to be created!

But fs combs may enable
one to have the advantages
of two or more systems

e.g. use atom standard for stability, ion standard for accuracy

⇒ More research required …