



Optical transitions  
in highly charged  
ions  
for atomic clocks  
with enhanced  
sensitivity to  
variation of  
fundamental  
constants

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# Motivation

Do the laws of physics change over space-time?

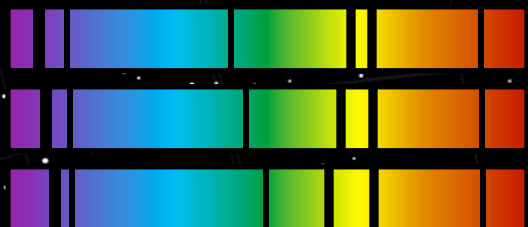
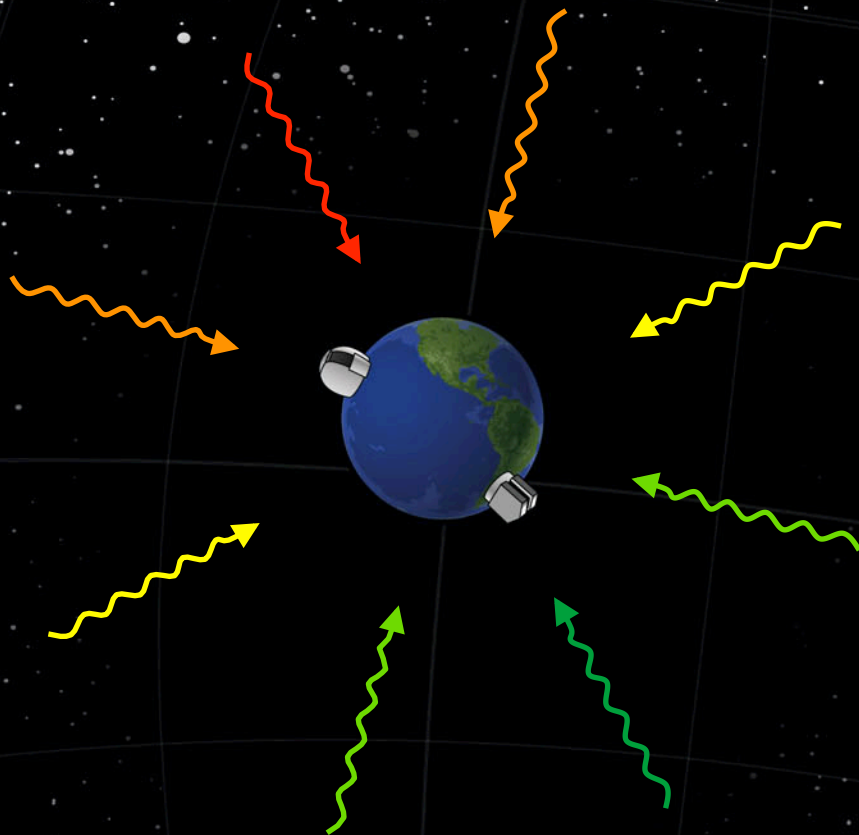
→ Do the fundamental constants change?

- ⊙ Around 25 dimensionless “fundamental constants”:  
Coupling constants, dimensionless mass ratios, mixing angles.

e.g.  $\alpha, \alpha_s, \theta_w, \frac{m_e}{\Lambda_{QCD}}, \frac{m_q}{\Lambda_{QCD}}, \dots$

- ⊙ ...plus some for General Relativity and Cosmology....
- ⊙ Cannot be predicted by theory and must be measured.
- ⊙ ∴ Should not assume constancy or homogeneity.

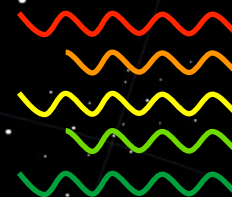
Light from  
Distant Quasars

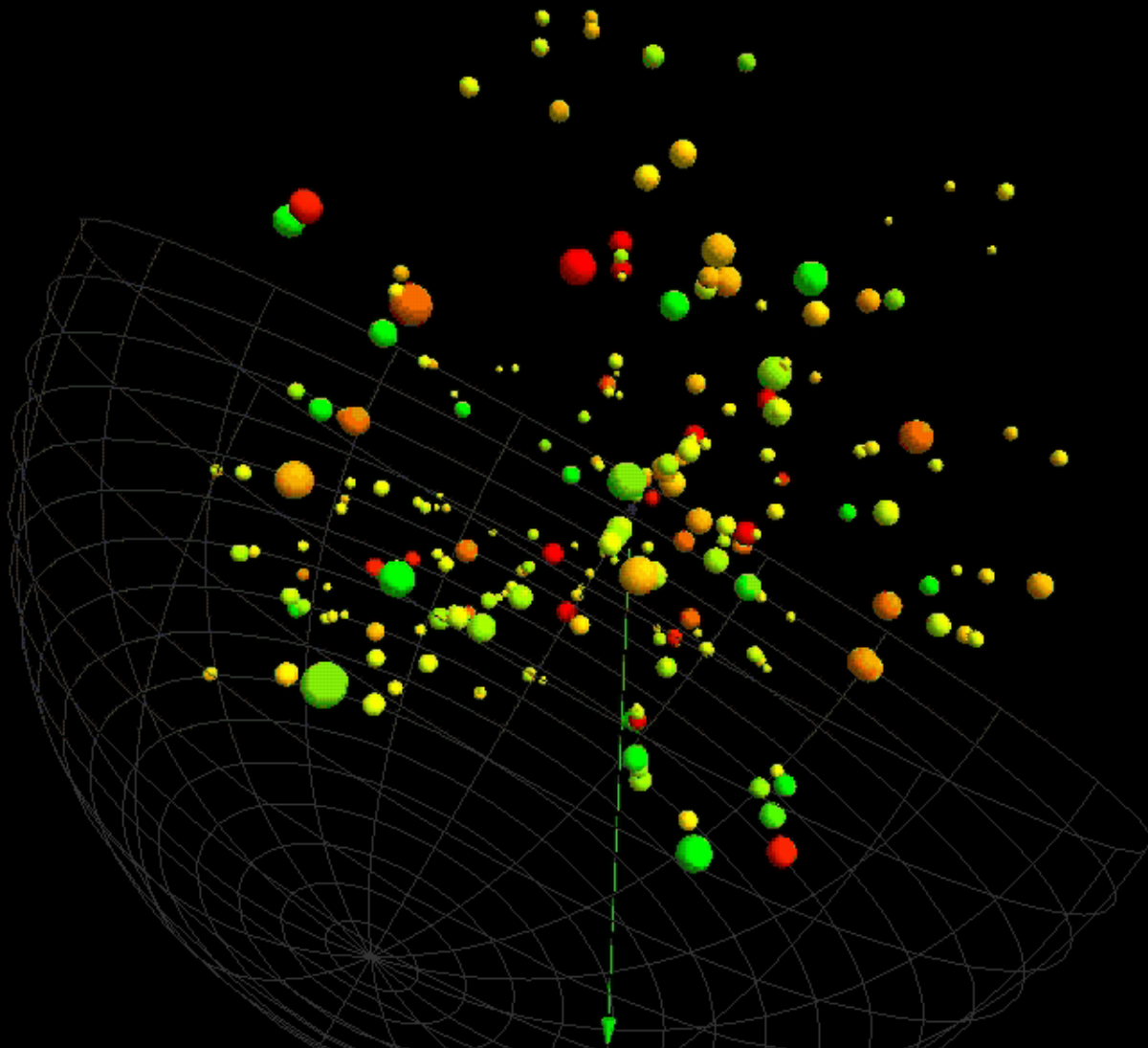


$\Delta\alpha < 0$  Negative-shifted Spectra

$\Delta\alpha = 0$  Lab-Observed Spectra

$\Delta\alpha > 0$  Positive-shifted Spectra

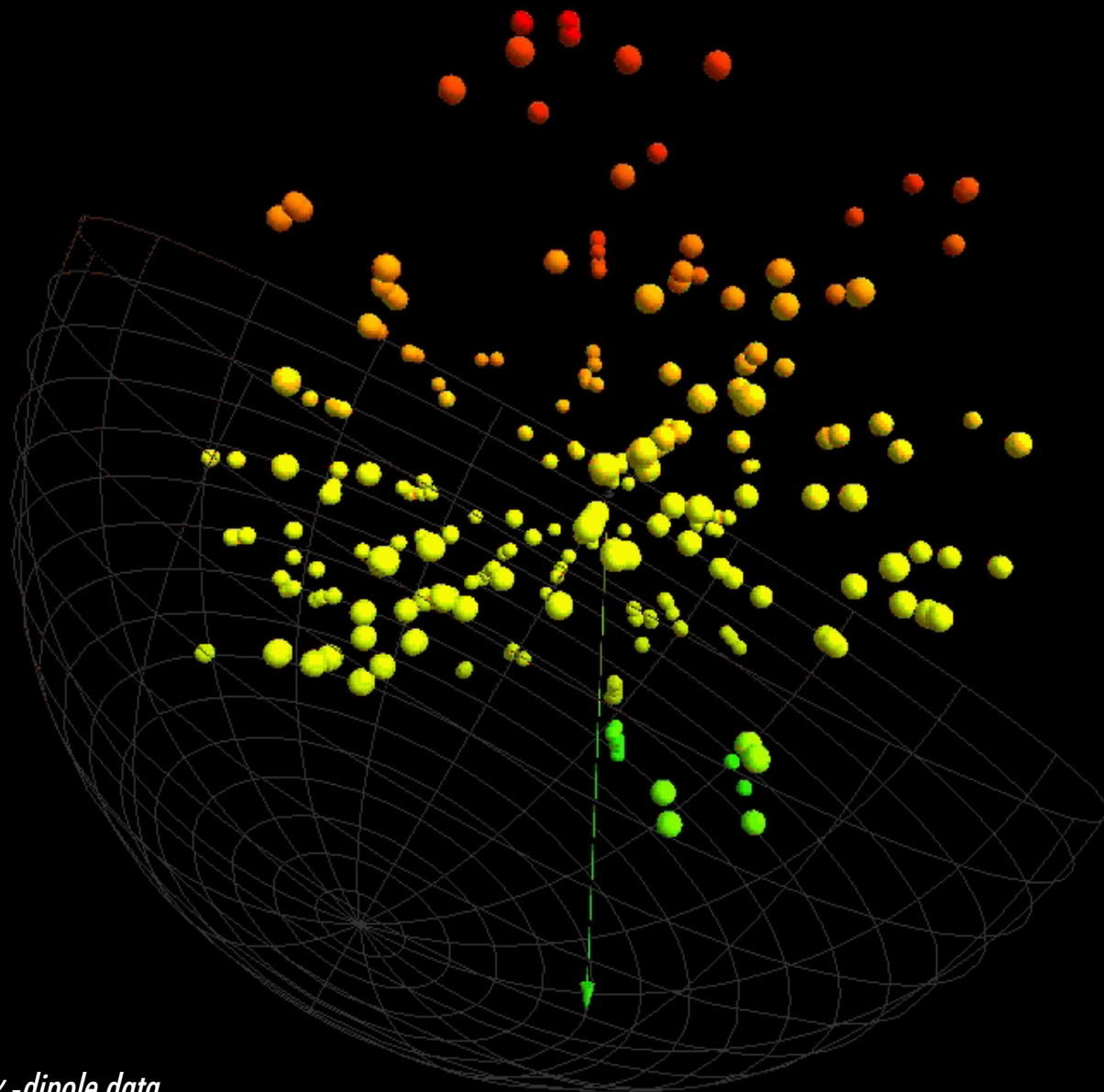




*$\alpha$ -variation from quasar absorption spectra*

*Webb, King, Murphy, Flambaum et al., PRL 107, 191101 (2011); King et al. MNRAS 422, 3370 (2012)*





*Generated  $\alpha$ -dipole data*

# Terrestrial $\alpha$ -variation

$$\frac{\dot{\alpha}}{\alpha} = 1.35 \times 10^{-18} \cos \theta \text{ yr}^{-1}$$
$$\frac{\Delta\alpha}{\alpha} = 1.4 \times 10^{-20} \cos \omega t$$



# $\alpha$ -sensitivity of single-electron transitions

Hydrogen-like ions

$$E_n = -\frac{m_e Z^2 e^4}{2\hbar^2 n^2} = -\frac{(Z\alpha)^2}{2n^2} \cdot m_e c^2$$

Relativistic corrections

$$\Delta_n = E_n \frac{(Z\alpha)^2}{v \left(j + \frac{1}{2}\right)}$$

$$\omega = \omega_0 + qx, \quad x = \left(\frac{\alpha}{\alpha_0}\right)^2 - 1$$

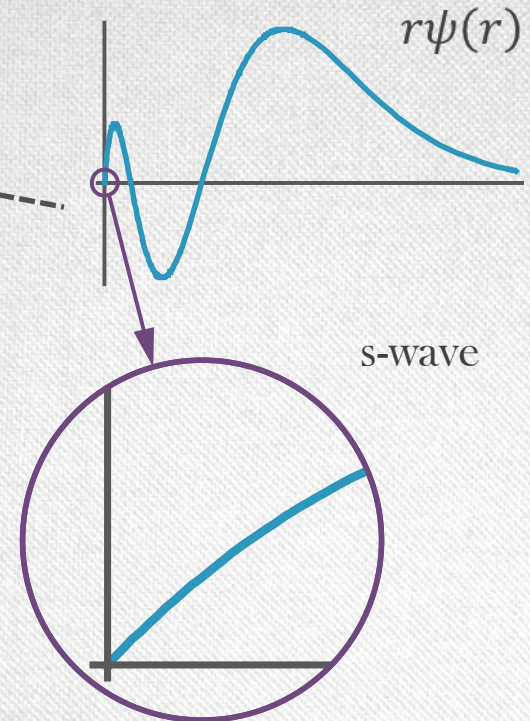
# $\alpha$ -sensitivity of single-electron transitions

$E_n = -$  Ionisation energy

$$q_n = E_n \frac{(Z\alpha)^2}{v \left( j + \frac{1}{2} \right)}$$

Effective principal quantum number

Stronger relativistic effects in heavier atoms





# Optical atomic clocks

- Required limit :  $\frac{\dot{\alpha}}{\alpha} \sim 10^{-19} \text{ yr}^{-1}$
- Best current limit ( $\text{Hg}^+$  vs  $\text{Al}^+$ ) :  $\frac{\dot{\alpha}}{\alpha} = (-1.6 \pm 2.3) \times 10^{-17} \text{ yr}^{-1}$   
*Rosenband et al., Science 319, 1808 (2008)*

$$q_n = E_n \frac{(Z\alpha)^2}{v \left( j + \frac{1}{2} \right)}$$

- $\text{Hg}^+$  clock :

$$5d^{10}6s \left( J = \frac{1}{2} \right) \rightarrow 5d^96s^2 \left( J = \frac{5}{2} \right)$$

$$\omega = 35514 \text{ cm}^{-1}$$

$$q = -52200 \text{ cm}^{-1}$$



# Optical atomic clocks

- Required limit :

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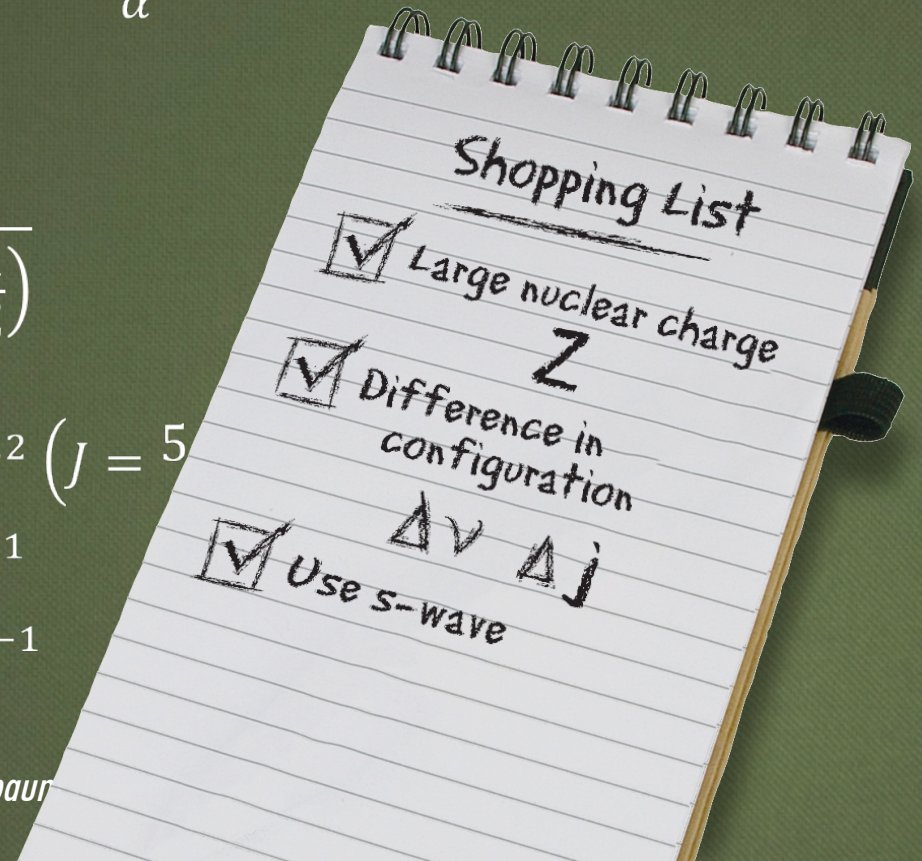
- Hg<sup>+</sup> clock :

$$5d^{10}6s \left( J = \frac{1}{2} \right) \rightarrow 5d^96s^2 \left( J = 5 \right)$$

$$\omega = 35514 \text{ cm}^{-1}$$

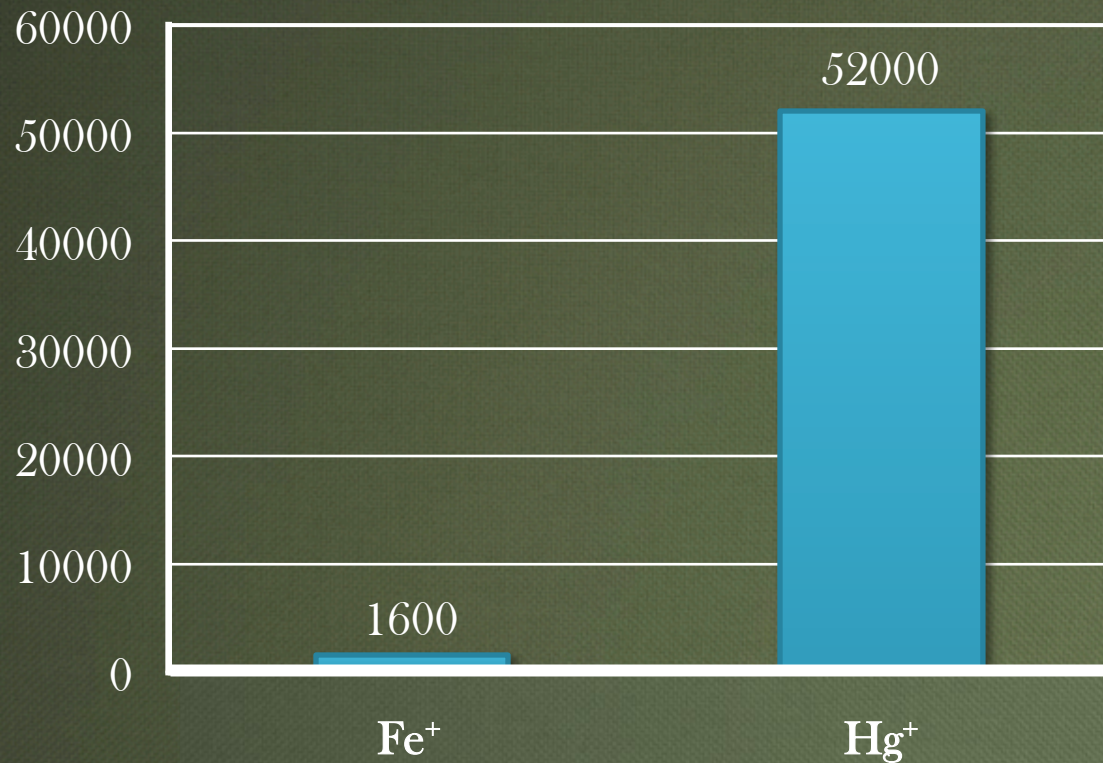
$$q = -52200 \text{ cm}^{-1}$$

*Dzuba, Flambaum, Webb, PRL 82, 888 (1999); Dzuba & Flambaum*





# Clock $q$ -values ( $\text{cm}^{-1}$ )

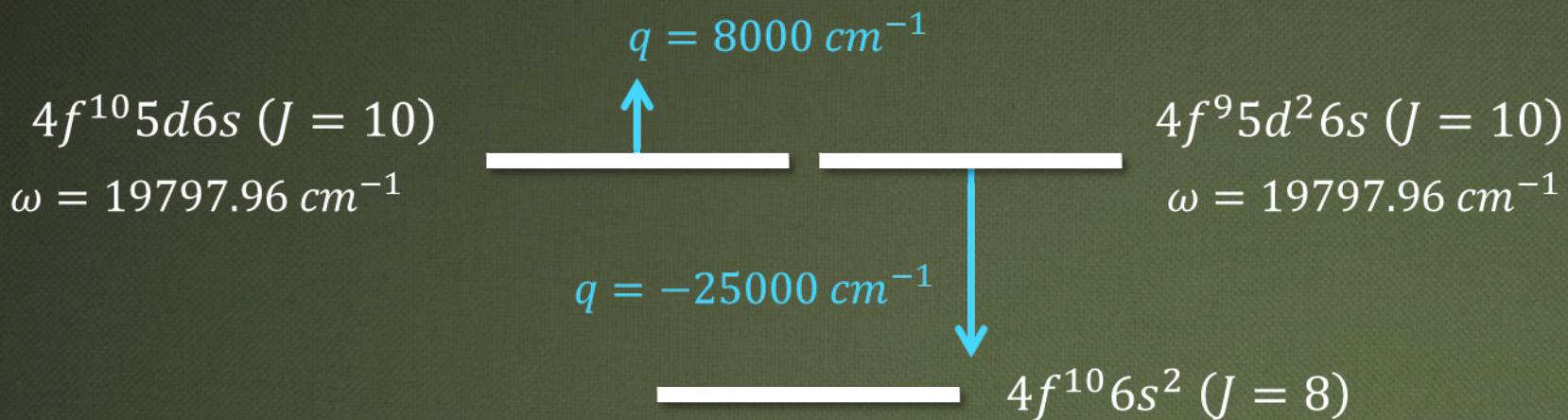




Where to find  
two orders of magnitude?



# Relative enhancement : Dy clock



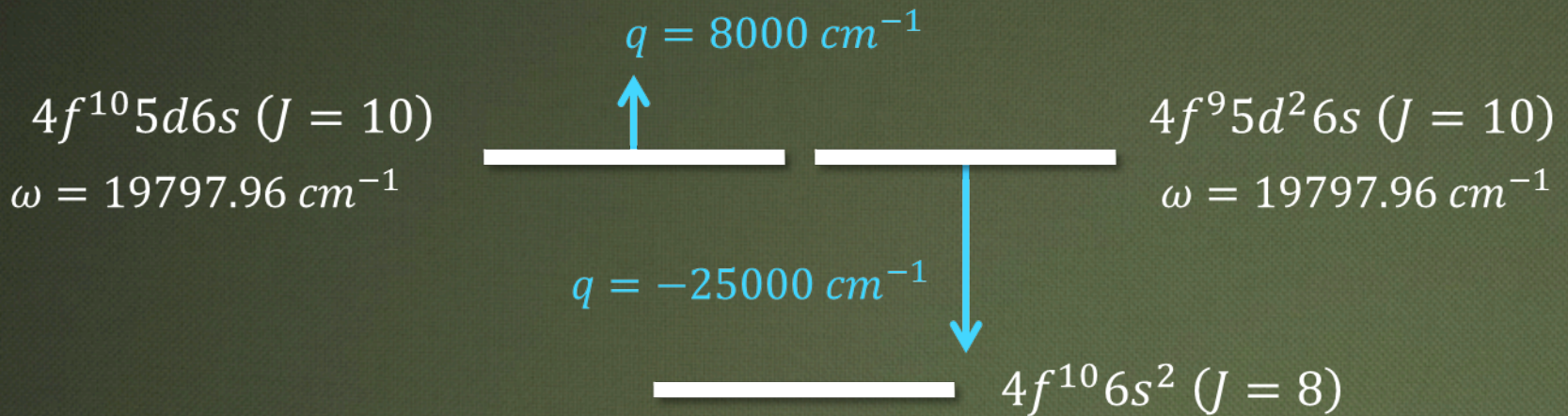
$$K = \frac{2q}{\omega}$$

$$\frac{\Delta(\omega_1/\omega_2)}{(\omega_1/\omega_2)} = (K_1 - K_2) \frac{\Delta\alpha}{\alpha}$$

- ⊙  $\text{Hg}^+$  vs  $\text{Al}^+$  :  $\Delta K = -3.2$
- ⊙ Dy :  $\Delta K \sim 10^8$



# Relative enhancement : Dy clock



$$\frac{\dot{\alpha}}{\alpha} = (-2.7 \pm 2.6) \times 10^{-15} \text{ yr}^{-1}$$

## Problem:

natural linewidths are not  
small



# Absolute Enhancement

$$q_n = E_n \frac{(Z\alpha)^2}{\nu \left(j + \frac{1}{2}\right)}$$

- ⊙  $E_n = -$  Ionisation energy
- ⊙ Use highly charged ions

$$\rightarrow q \sim Z^2 (Z_{ion} + 1)^2$$

## Problem:

transition frequencies also  
grow with  $E_n$



# Absolute Enhancement

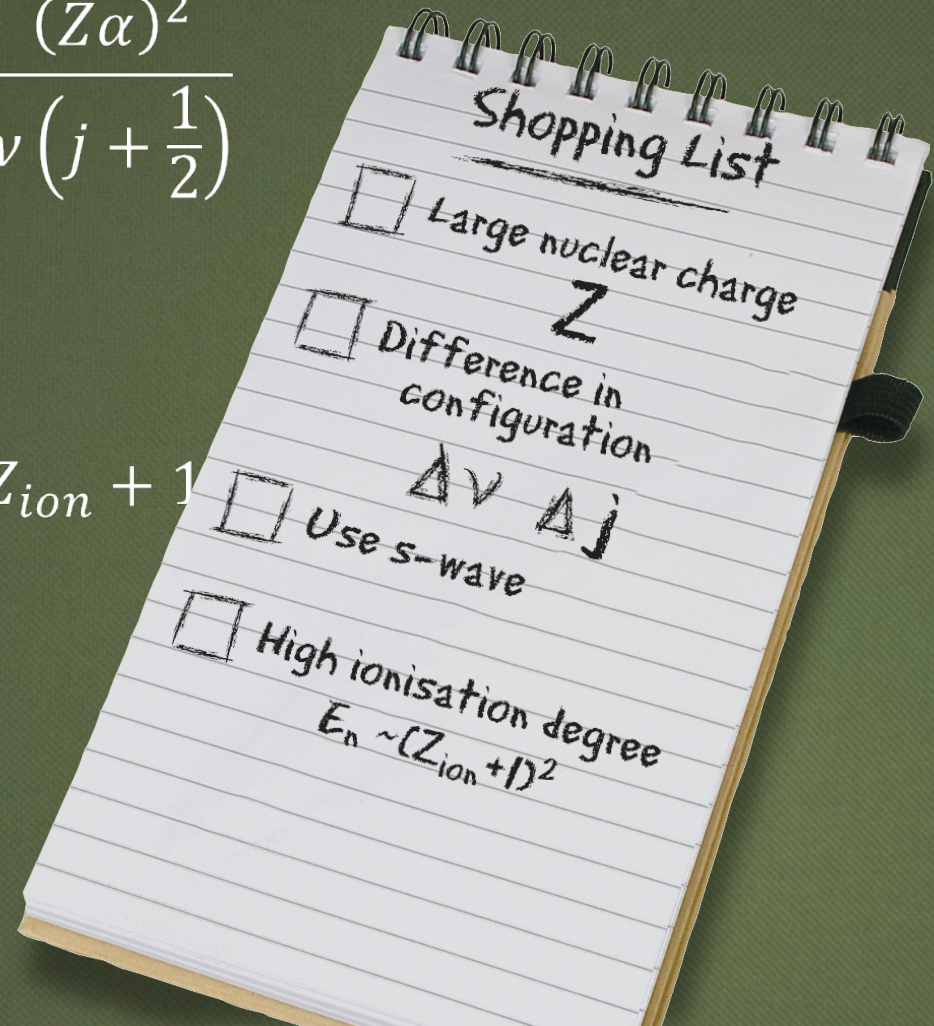
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## Problem:

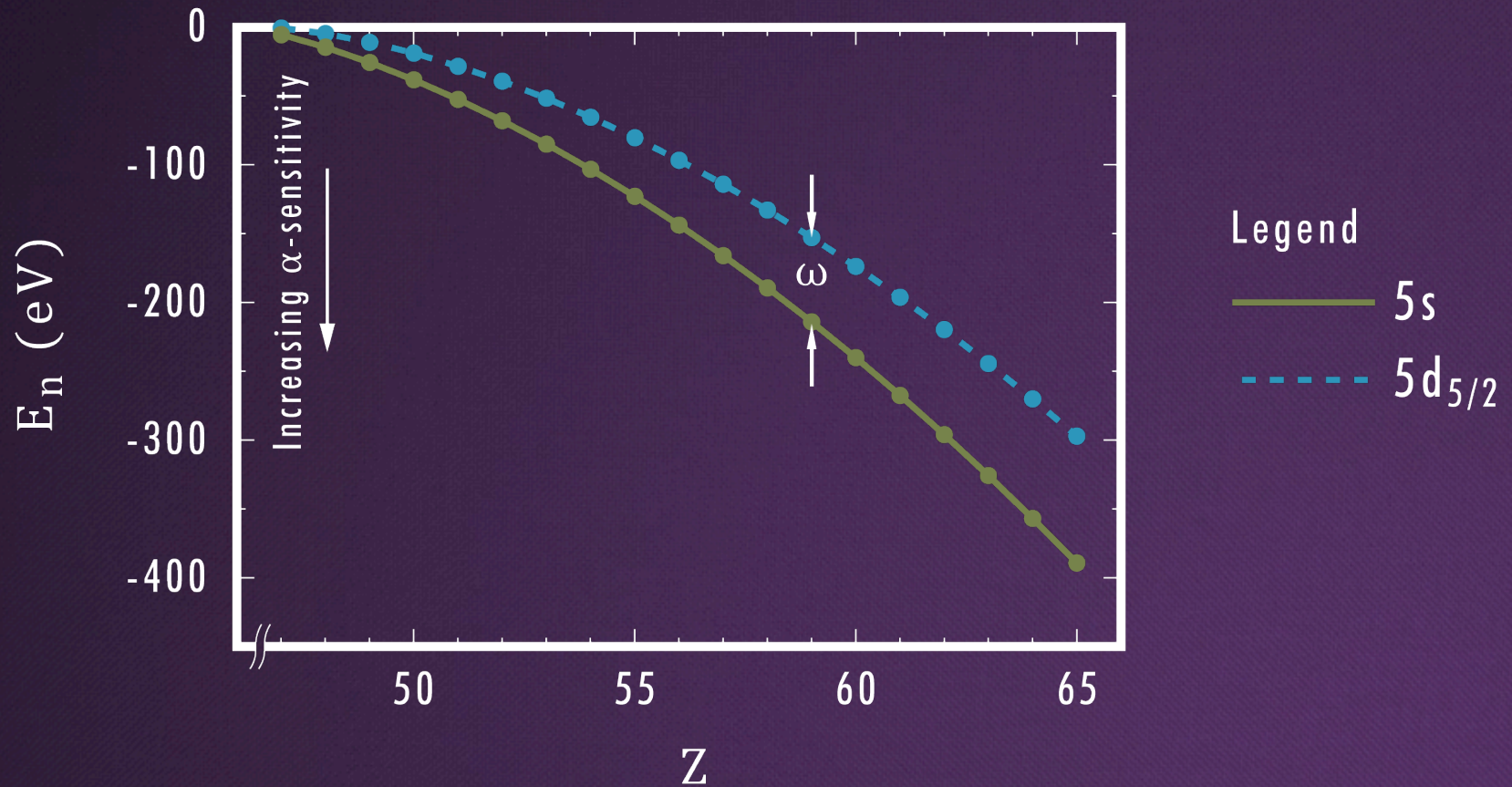
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# Ag isoelectronic sequence

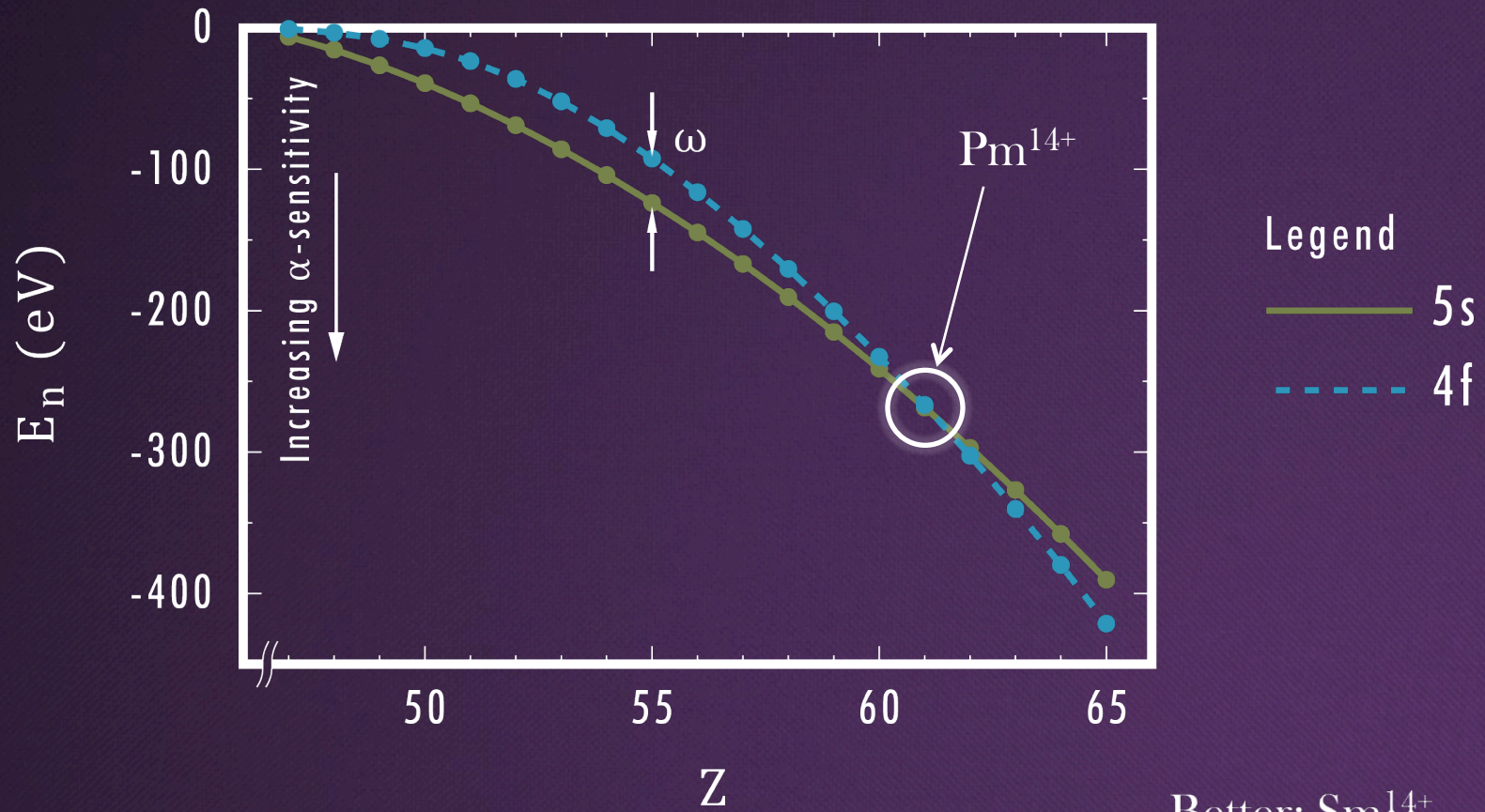
$[Kr]4d^{10} nl$  (47 electrons)





# Ag isoelectronic sequence

$[Kr]4d^{10} nl$  (47 electrons)

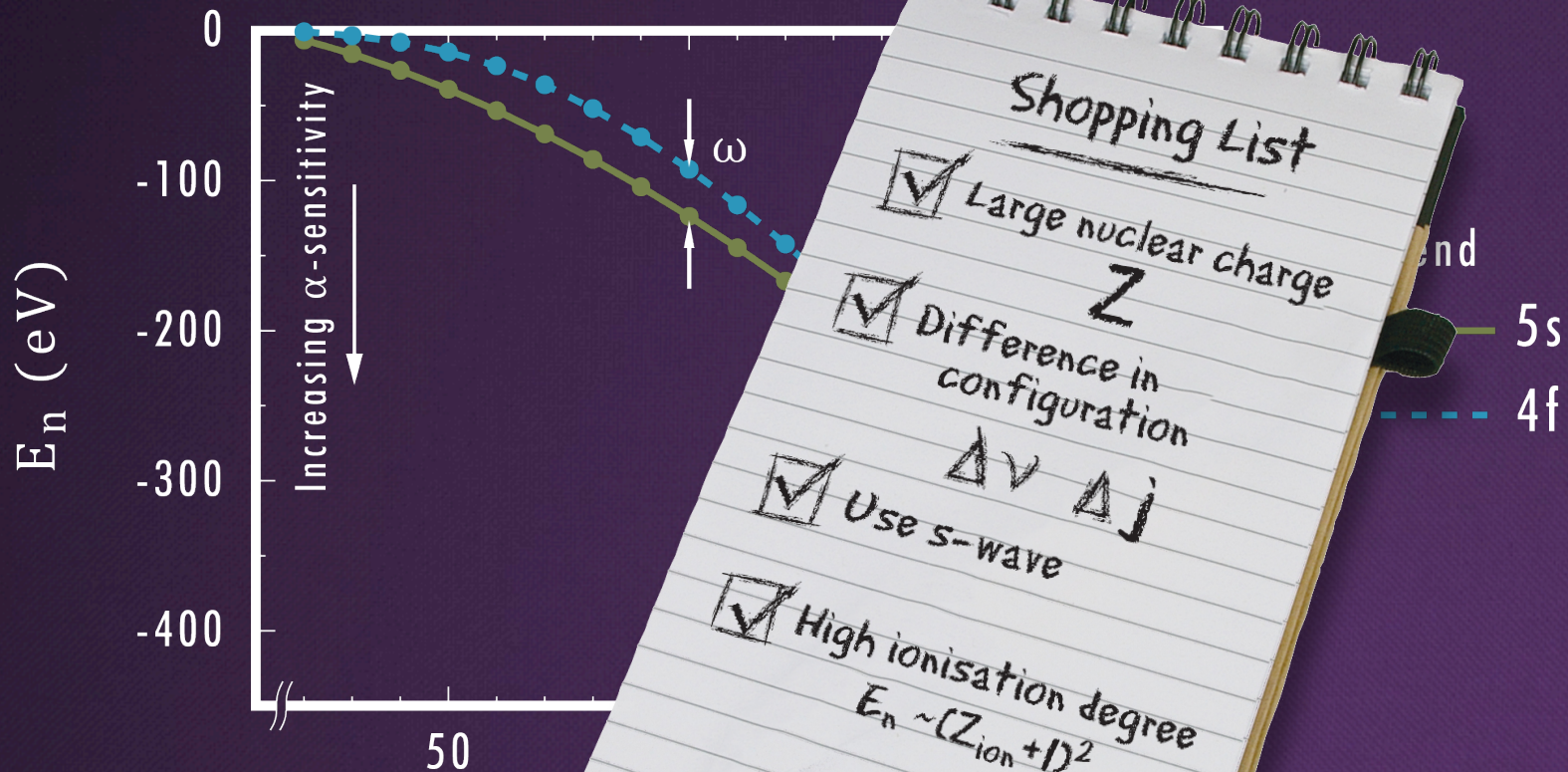


Better:  $Sm^{14+}$



# Ag isoelectronic sequence

$[Kr]4d^{10} nl$  (47 electrons)



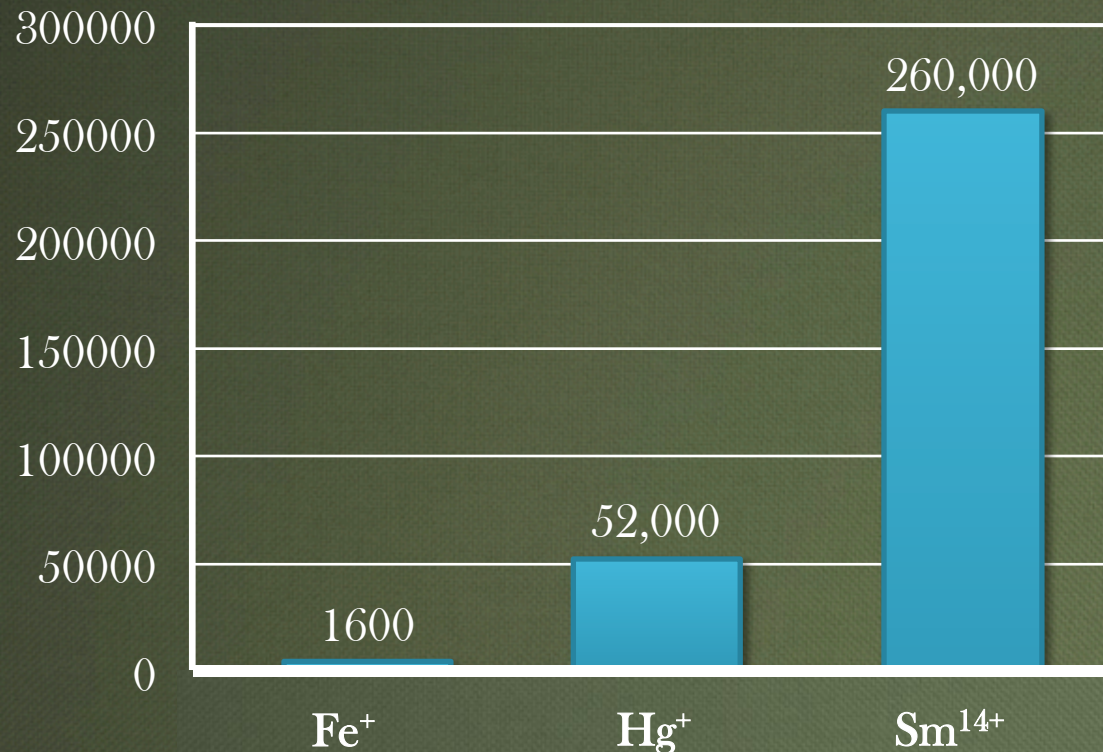
**Shopping List**

- Large nuclear charge  $Z$
- Difference in configuration  $\Delta v \Delta j$
- Use s-wave
- High ionisation degree  $E_n \sim (Z_{ion} + 1)^2$

Better:  $Sm^{14+}$



# Clock $q$ -values ( $\text{cm}^{-1}$ )



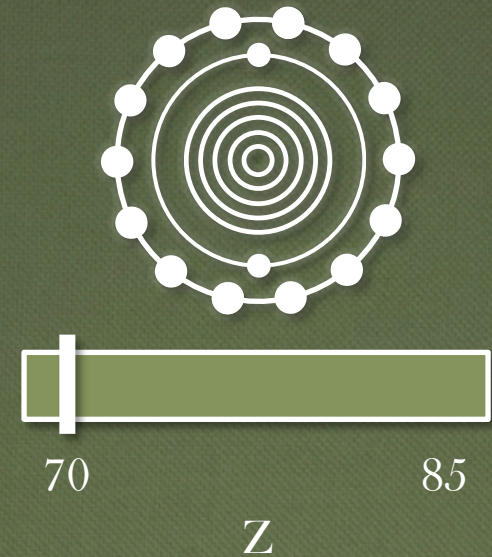


# Holes in highly charged ions

- ⦿ Use highly charged ions
- ⦿ Electrons in filled shells have maximal  $I_n$
- ⦿ Electron spends half time closer to nucleus than other electrons in the same shell

$$q \sim I_n^{3/2}$$

- ⦿ Use holes in otherwise filled shells



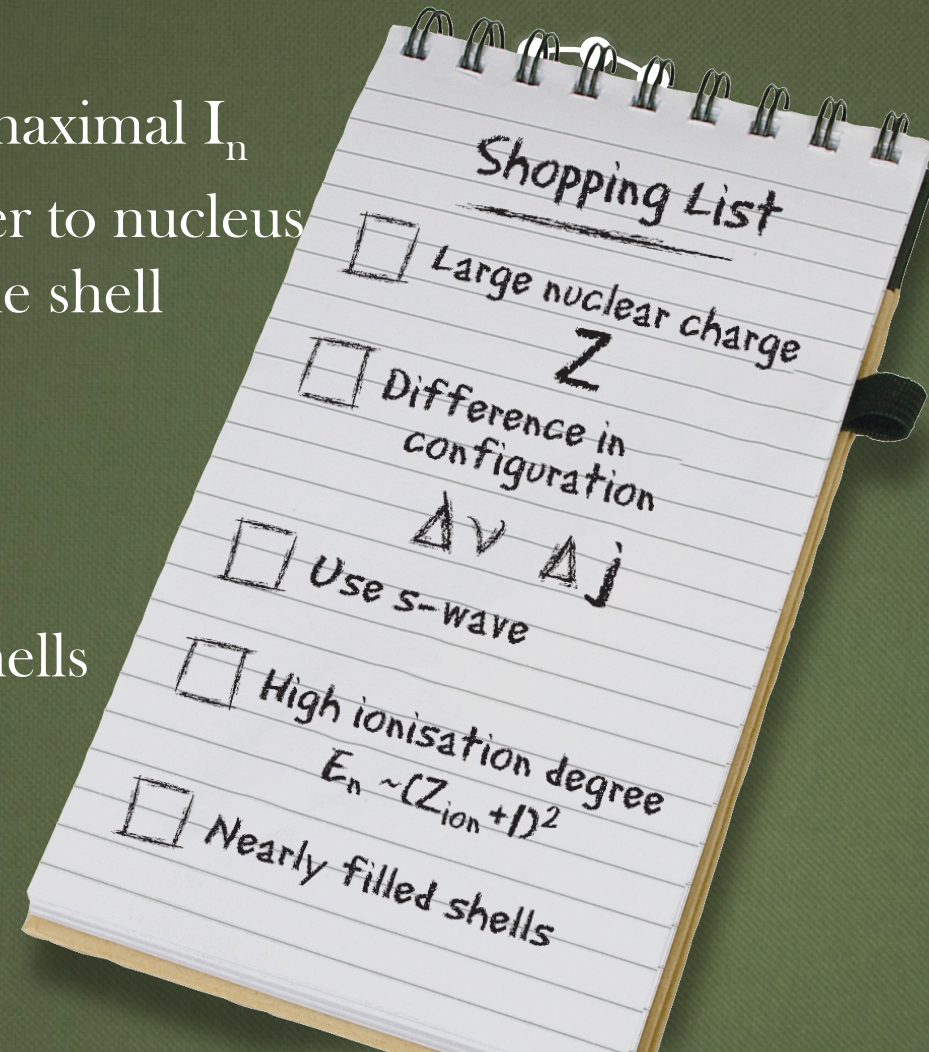


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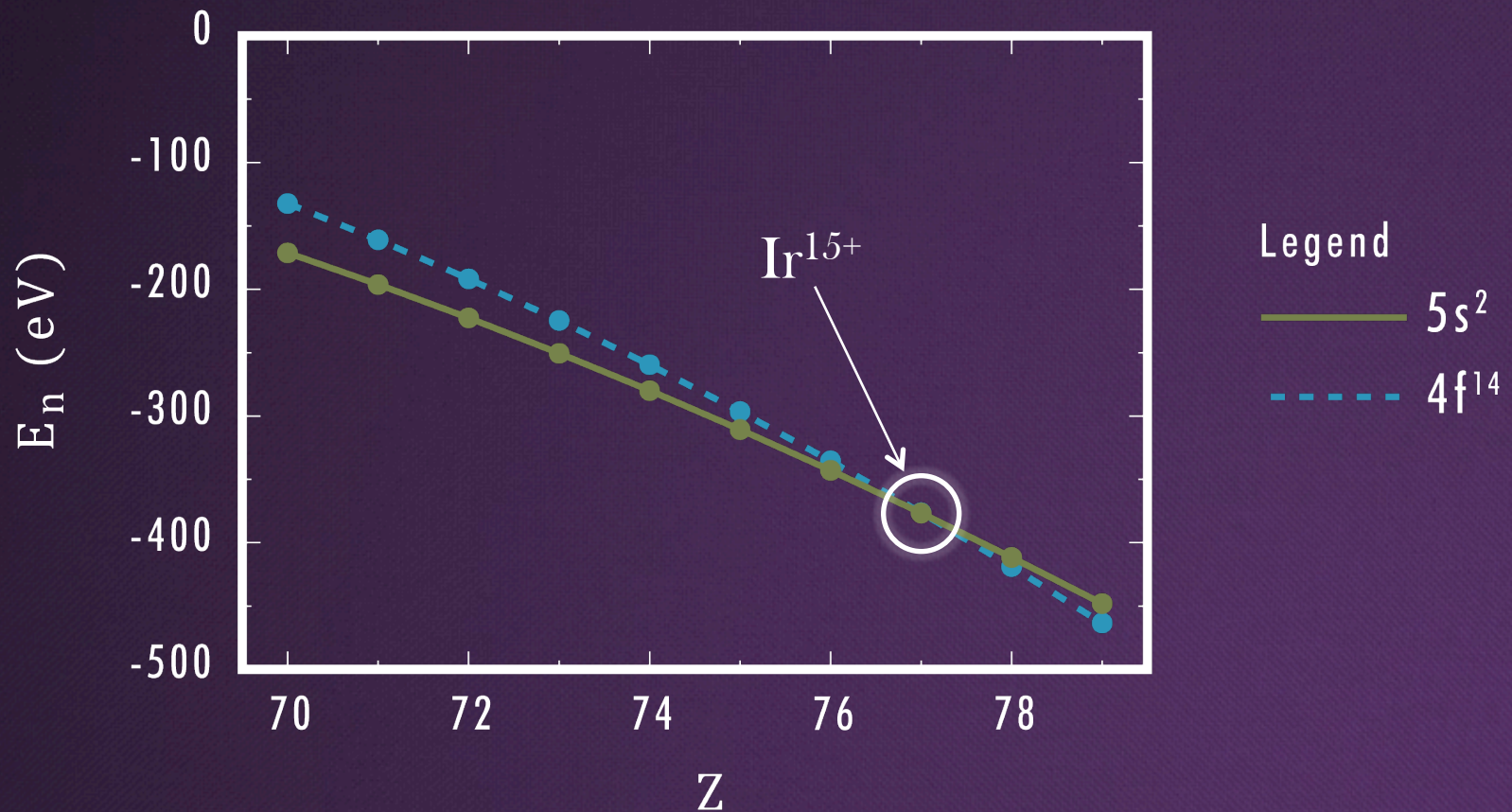
- ◉ Use holes in otherwise filled shells





# Sm isoelectronic sequence

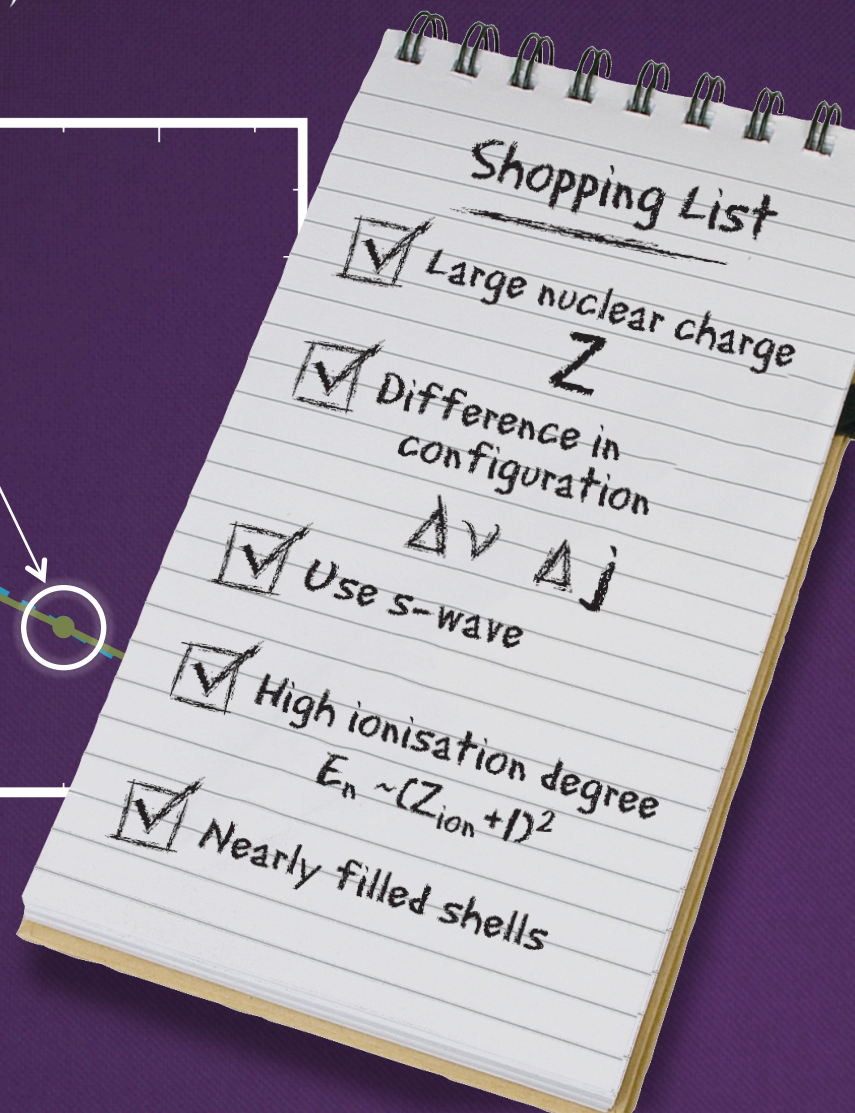
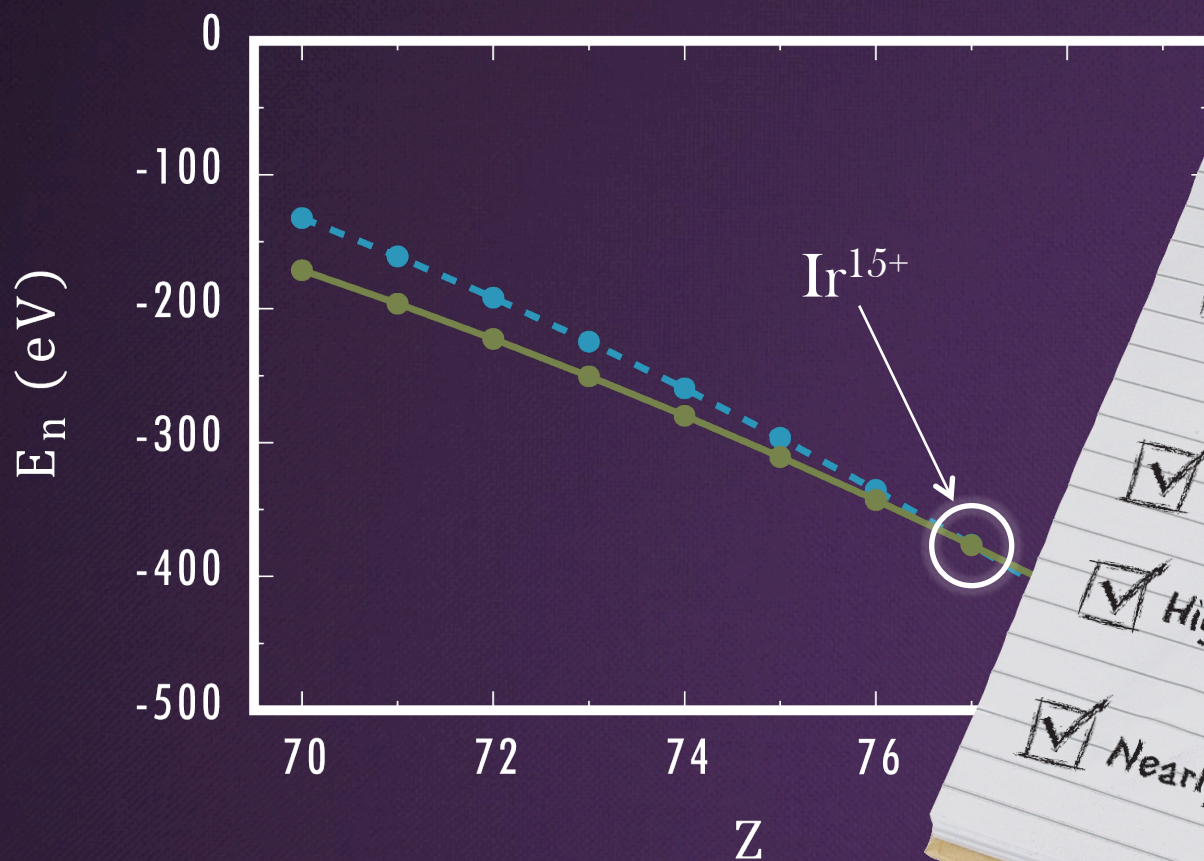
$[Kr]4d^{10} 4f^{14} 5s^2$  (62 electrons)





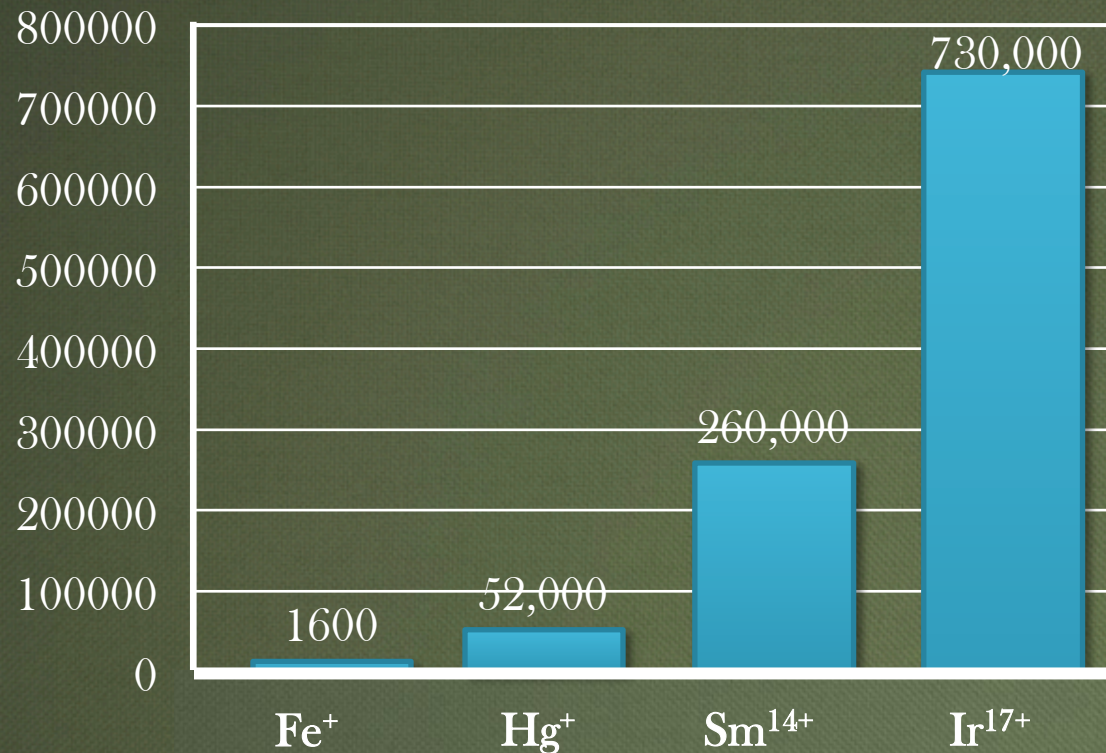
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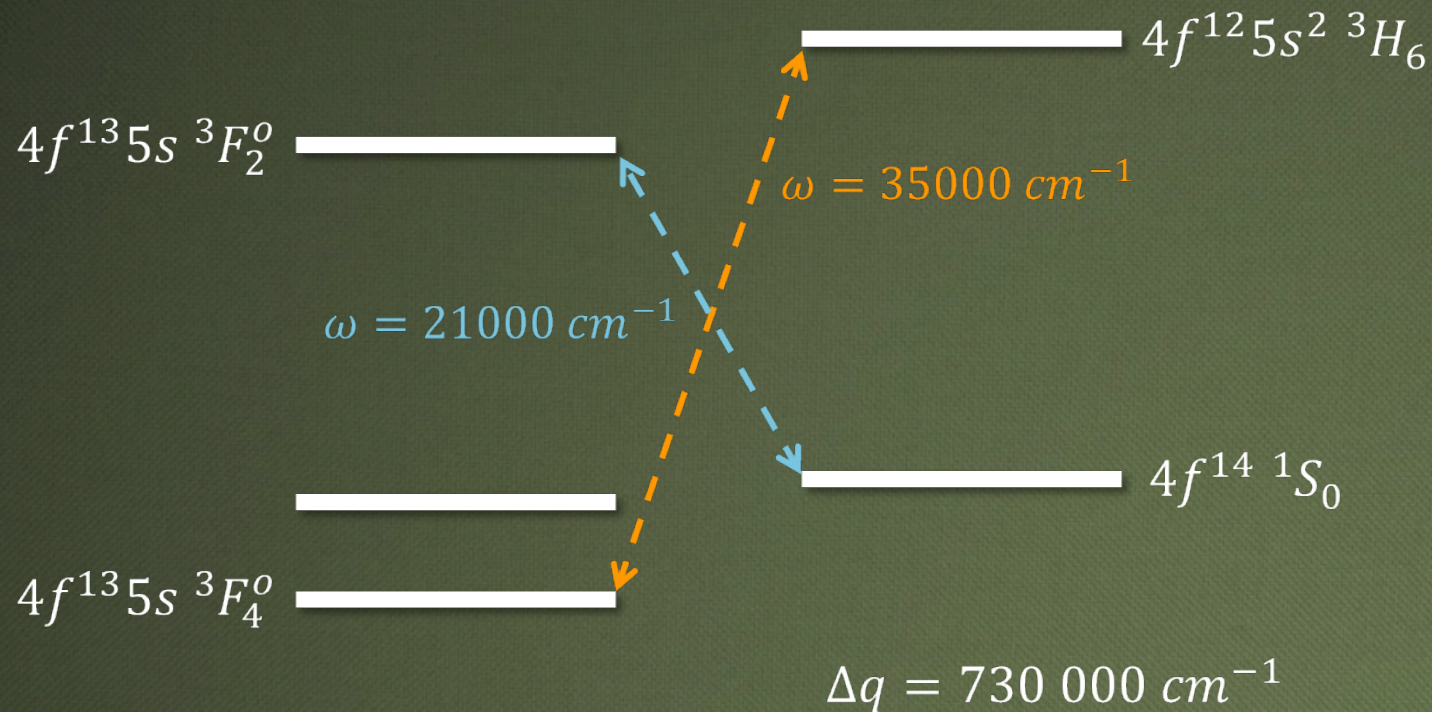


# Clock $q$ -values ( $\text{cm}^{-1}$ )





# Holes in highly charged ions : Ir<sup>17+</sup>



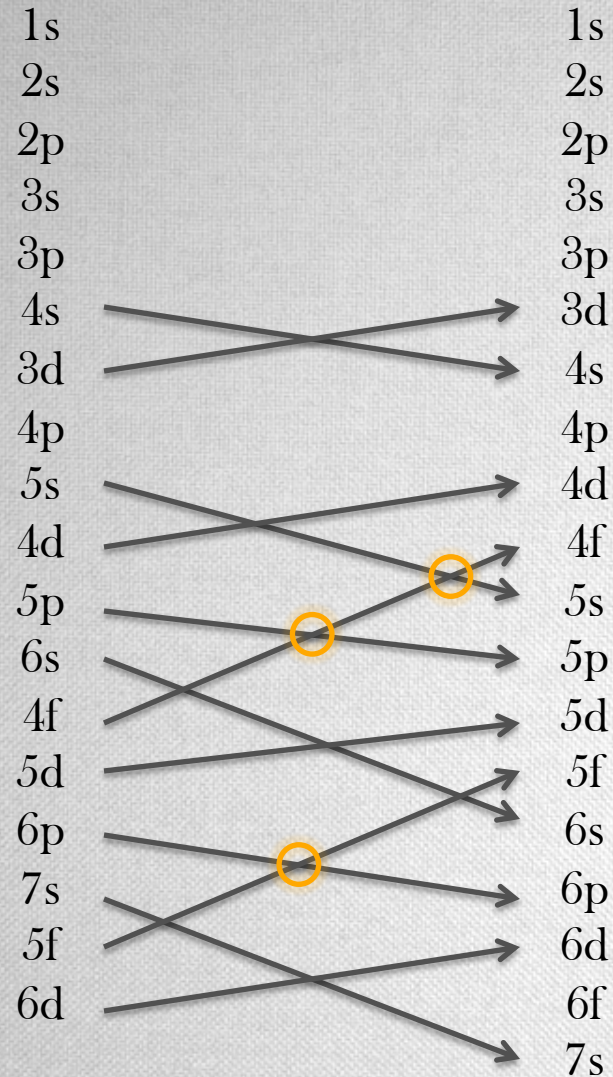
- There exists an E1 transition at 45 000 cm<sup>-1</sup> for trapping and cooling



# Finding optical transitions in highly charged ions

Periodic Table Filling

Coulomb Filling





# Finding optical transitions in highly charged ions

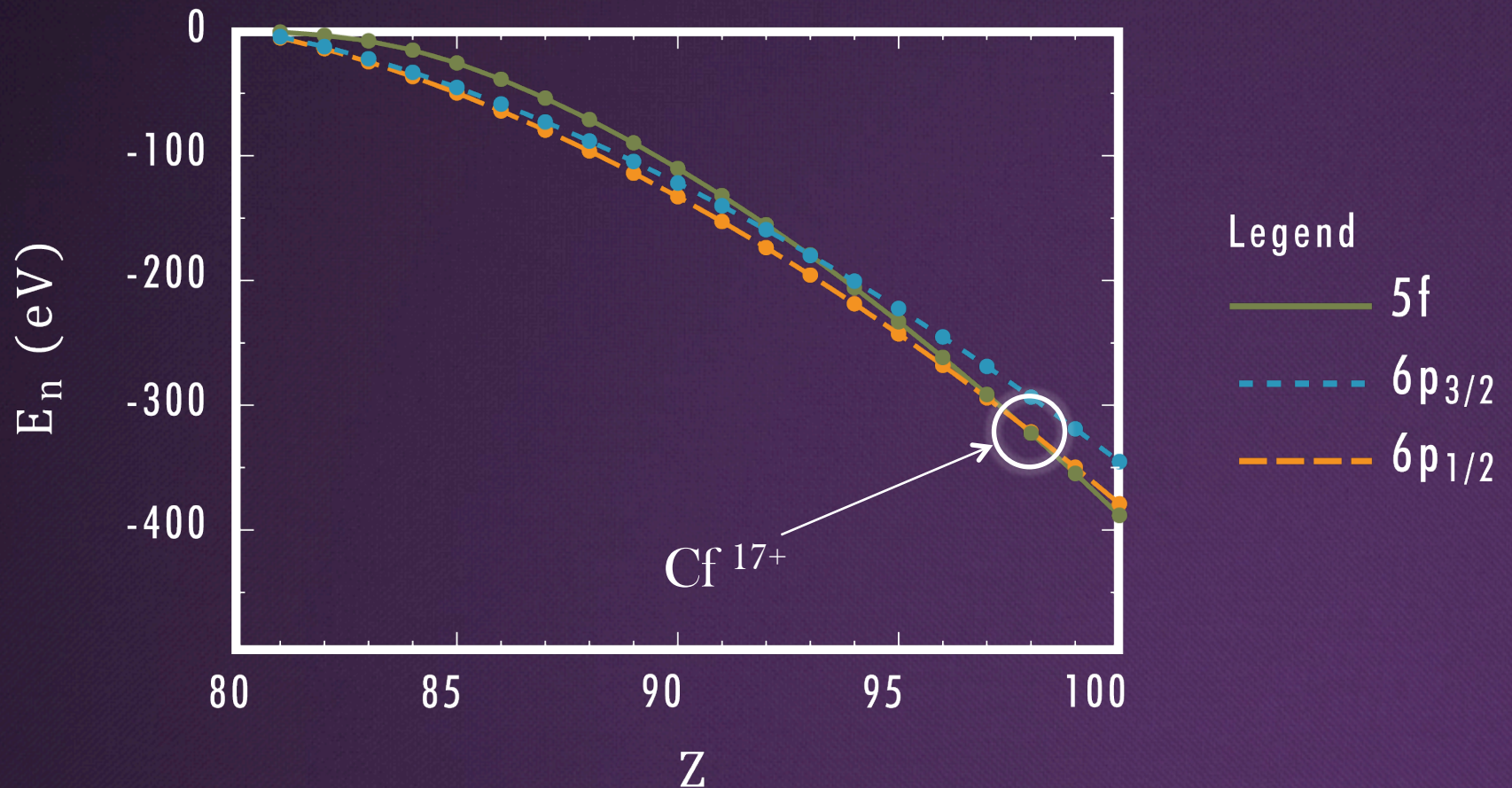
1 H 1.00794																	1 H 1.00794	2 He 4.002602
3 Li 6.941	4 Be 9.012182											5 B 10.811	6 C 12.0107	7 N 14.00674	8 O 15.9994	9 F 18.9984032	10 Ne 20.1797	
11 Na 22.989770	12 Mg 24.3050											13 Al 26.981538	14 Si 28.0855	15 P 30.973761	16 S 32.066	17 Cl 35.4527	18 Ar 39.948	
19 K 39.0983	20 Ca 40.078	21 Sc 44.955910	22 Ti 47.867	23 V 50.9415	24 Cr 51.9961	25 Mn 54.938049	26 Fe 55.845	27 Co 58.933200	28 Ni 58.6934	29 Cu 63.546	30 Zn 65.39	31 Ga 69.723	32 Ge 72.61	33 As 74.92160	34 Se 78.96	35 Br 79.904	36 Kr 83.80	
37 Rb 85.4678	38 Sr 87.62	39 Y 88.90585	40 Zr 91.224	41 Nb 92.90638	42 Mo 95.94	43 Tc (98)	44 Ru 101.07	45 Rh 102.90550	46 Pd 106.42	47 Ag 107.8682	48 Cd 112.411	49 In 114.818	50 Sn 118.710	51 Sb 121.760	52 Te 127.60	53 I 126.90447	54 Xe 131.29	
55 Cs 132.90545	56 Ba 137.327	57 La 138.9055	72 Hf 178.49	73 Ta 180.9479	74 W 183.84	75 Re 186.207	76 Os 190.23	77 Ir 192.217	78 Pt 195.078	79 Au 196.96655	80 Hg 200.59	81 Tl 204.3833	82 Pb 207.2	83 Bi 208.98038	84 Po (209)	85 At (210)	86 Rn (222)	
87 Fr (223)	88 Ra (226)	89 Ac (227)	104 Rf (261)	105 Db (262)	106 Sg (263)	107 Bh (262)	108 Hs (265)	109 Mt (266)	110 (269)	111 (272)	112 (277)		114 (289) (287)		116 (289)		118 (293)	

58 Ce 140.116	59 Pr 140.90765	60 Nd 144.24	61 Pm (145)	62 Sm 150.36	63 Eu 151.964	64 Gd 157.25	65 Tb 158.92534	66 Dy 162.50	67 Ho 164.93032	68 Er 167.26	69 Tm 168.93421	70 Yb 173.04	71 Lu 174.967
90 Th 232.0381	91 Pa 231.03588	92 U 238.0289	93 Np (237)	94 Pu (244)	95 Am (243)	96 Cm (247)	97 Bk (247)	98 Cf (251)	99 Es (252)	100 Fm (257)	101 Md (258)	102 No (259)	103 Lr (262)



# Tl isoelectronic sequence

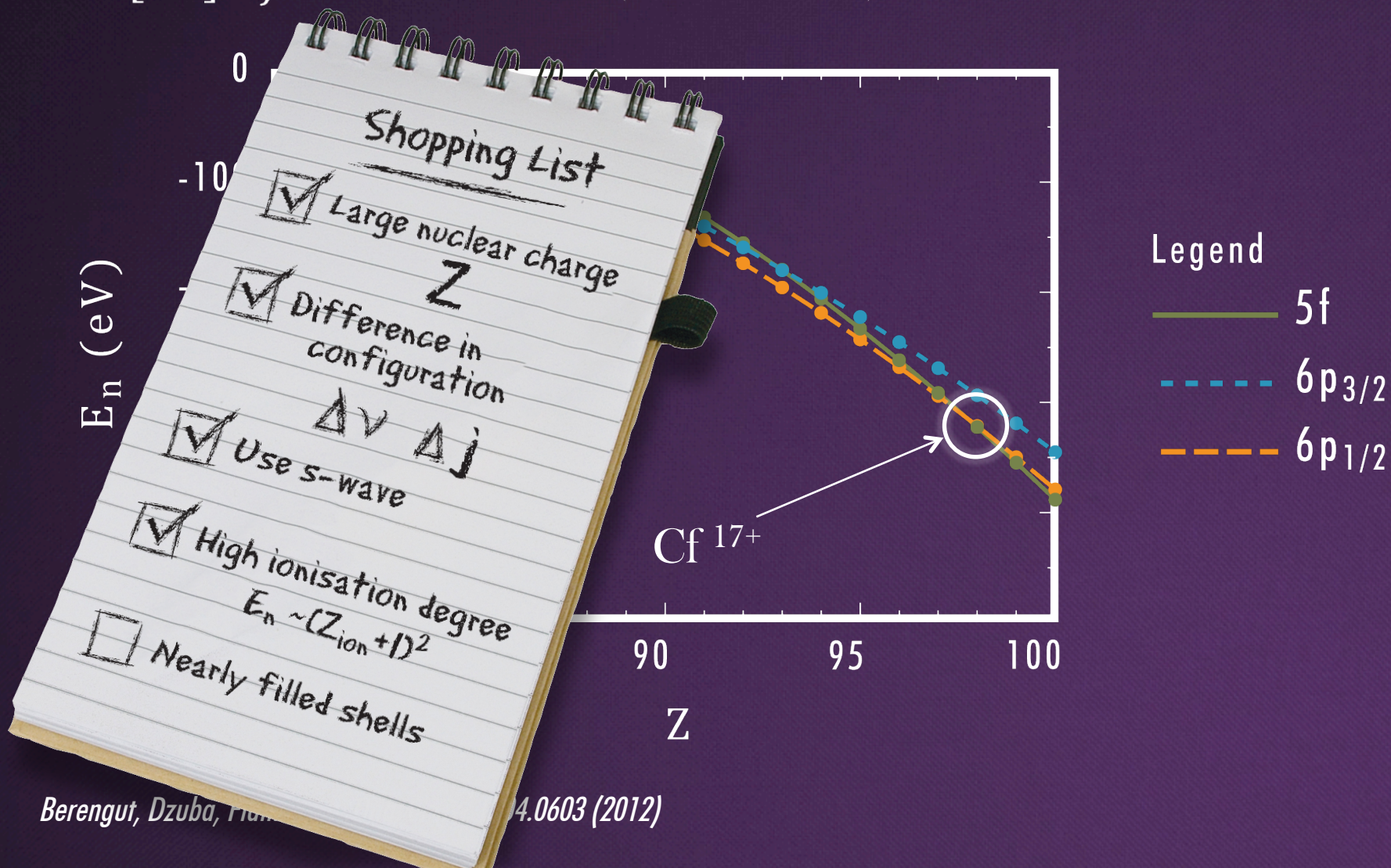
$[Xe] 4f^{14}5d^{10}6s^2 : nl$  (81 electrons)





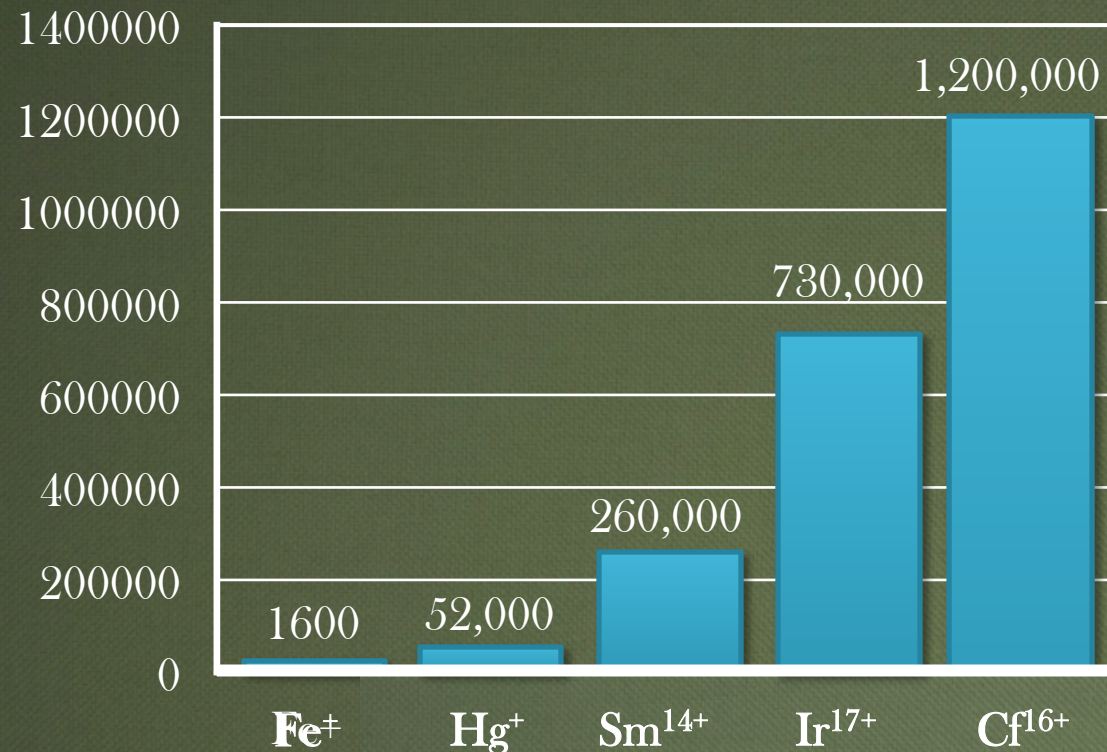
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# Clock q-values ( $\text{cm}^{-1}$ )





# HCI Clock Scaling Laws and Systematics

- ⊙ 2<sup>nd</sup> order Stark shift  $\sim 1/(Z_{ion}+1)^4$
- ⊙ Blackbody shift  $\sim 1/(Z_{ion}+1)^4$
- ⊙ 2<sup>nd</sup> order Zeeman shift  $\sim$  suppressed as  $1/\Delta E$
- ⊙ Electric quadrupole shift  $\sim 1/(Z_{ion}+1)^2$
- ⊙ Fine-structure  $\sim Z^2 (Z_{ion}+1)^2/\nu^3$
- ⊙ Hyperfine  $A$  coefficient  $\sim Z (Z_{ion}+1)^2/\nu^3$



# Thank you

Collaborators:

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Mikhail Kozlov, Sergey Porsev, Vladimir Dmitriev,  
John Webb, Julian King, Michael Murphy, Steve Curran, Elliot Koch