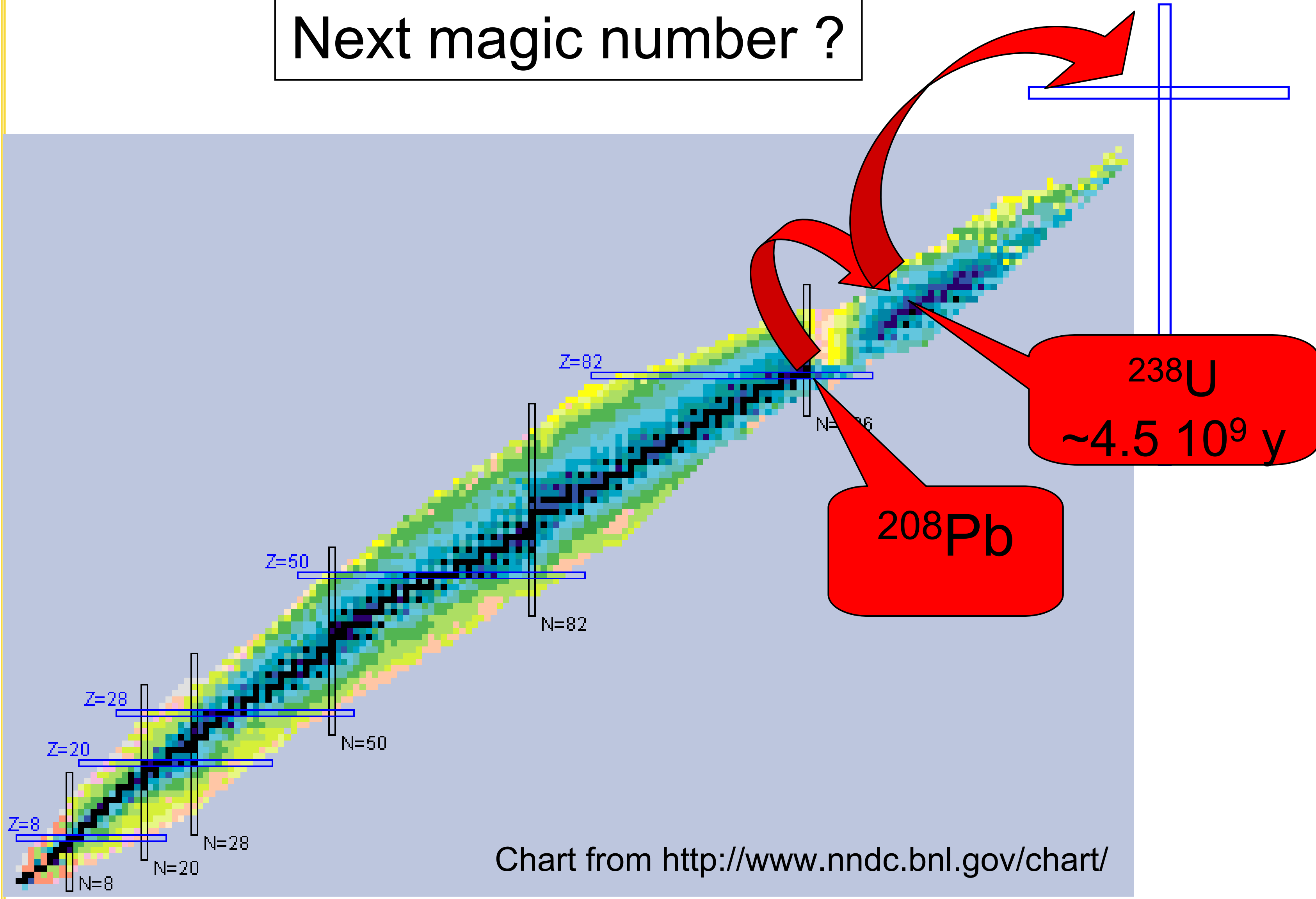
Spectroscopy of very heavy elements

Ch. THEISEN
christophe.theisen@cea.fr

Introduct

Limits of stability ?
Shell structure ?
Next magic number ?



Very heavy elements : questions

- What are the limits of the nuclear chart. Stability of nuclear matter
- Evolution of shell structure. Next magic shell number ? Is there an island of stability ? Validity and predictive power of nuclear models.
- How to synthesize heavy elements. Reaction mechanism
- Have super-heavy elements been synthesized in nature (eg supernovae) ? Do traces remain on earth, in meteorites ?
- What are the atomic properties of the heaviest elements : electron binding energies, atomic yields, …
- Chemical properties. Is the Mendeleev classification still valid ?

Outlook

- Interest and motivations
- Limits of stability : macroscopic aspects
- Decay modes
- Discovery of the heaviest elements
- Production
- Stability : microscopic aspects
- Decay spectroscopy
  - Technique, alpha decay, atomic effects, K-isomers
- Separators, spectrometers and focal plane devices
- Prompt spectroscopy
  - Recoil Decay Tagging
  - Gamma and electron spectroscopy
- Pushing the limits : facilities and devices

① ② ③ ④

Topics not covered in this lecture

(Or at least very briefly)

- Synthesis of SHE
- Reaction mechanism
- Atomic properties
- Chemistry

Very-heavy and Super-heavy elements

- Definition of an heavy element ?
  a very heavy element ?
  a super-heavy element ?

Point of view of chemist :

Actinides $90 \leq Z \leq 103$

Transactinides $104 \leq Z \leq 121$ (?)

Arbitrary point of view :

Transfermium $Z > 100$

H 1																	He 2
Li 3	Be 4																Ne 10
Na 11	Mg 12																Ar 18
K 19	Ca 20	Sc 21															Kr 36
Rb 37	Sr 38	Y 39	Zr 40	Nb 41	Mo 42	Tc 43	Ru 44	Rh 45	Pd 46	Ag 47	Cd 48	In 49	Sn 50	Sb 51	Te 52	I 53	Xe 54
Cs 55	Ba 56	La 57	Hf 72	Ta 73	W 74	Re 75	Os 76	Ir 77	Pt 78	Au 79	Hg 80	Tl 81	Pb 82	Bi 83	Po 84	At 85	Rn 86
Fr 87	Ra 88	Ac 89	Rf 104	Db 105	Sg 106	Bh 107	Hs 108	Mt 109	Ds 110	Rg 111	112	113	114	115	116	117	118
119	120																

Lanthanides	Ce 58	Pr 59	Nd 60	Pm 61	Sm 62	Eu 63	Gd 64	Tb 65	Dy 66	Ho 67	Er 68	Tm 69	Yb 70	Lu 71
Actinides	Th 90	Pa 91	U 92	Np 93	Pu 94	Am 95	Cm 96	Bk 97	Cf 98	Es 99	Fm 100	Md 101	No 102	Lr 103

Limits of stability : macroscopic point of view

Macroscopic approach : the liquid drop model

- $B(A,Z) = a_v A$ — volume – nuclear attractive force
  - $- a_s A^{2/3}$ — less binding at the surface
  - $- a_c Z^2/A^{1/3}$ — Coulomb – proton repulsion
  - $- a_a (A-2Z)^2/A$ — asymmetry
  - $+\delta A^{-1/3}$ — pairing

- Mass formula : M(A,Z) = Z.Mp + N.Mn – B(A,Z)
- « Macroscopic » limits of stability
- Proton drip line Sp = M(A-1,Z-1) + Mp – M(A,Z)
  = B(A,Z) – B (A-1,Z-1)
  Sp < 0 → not stable with respect proton emission
- Neutron drip line Sn = B(A,Z) – B (A-1,Z)
- Deformation → fission

Stability of the deformed liquid drop : fission

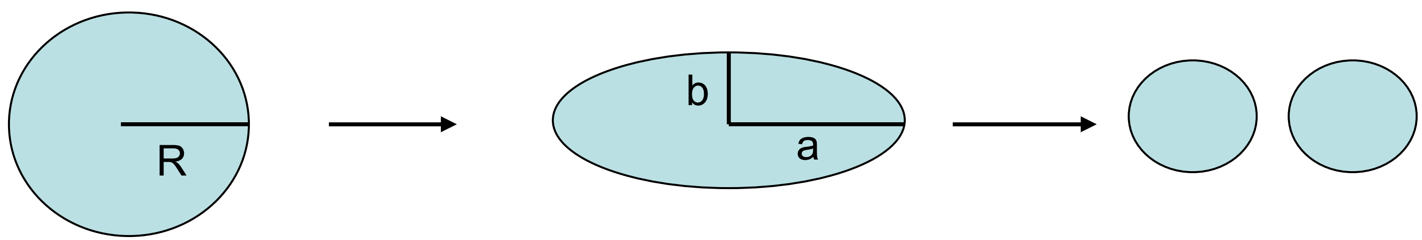
- B(A,Z,deformation) =

  $a_v A$

  $- a_s A^{2/3} (1 + 2/5\ \varepsilon^2 + \ldots)$ surface prefers spherical nuclei

  $- a_c Z^2 A^{-1/3} (1 - 1/5\ \varepsilon^2 + \ldots)$ coulomb favours deformation

  - asymmetry – pairing



$V = 4/3 \pi R^3$

$S = 4 \pi R^2$

$a = R(1+\varepsilon)$

$b = R(1+\varepsilon)^{-1/2}$

$$\varepsilon = \sqrt{1 - \frac{b^2}{a^2}}$$

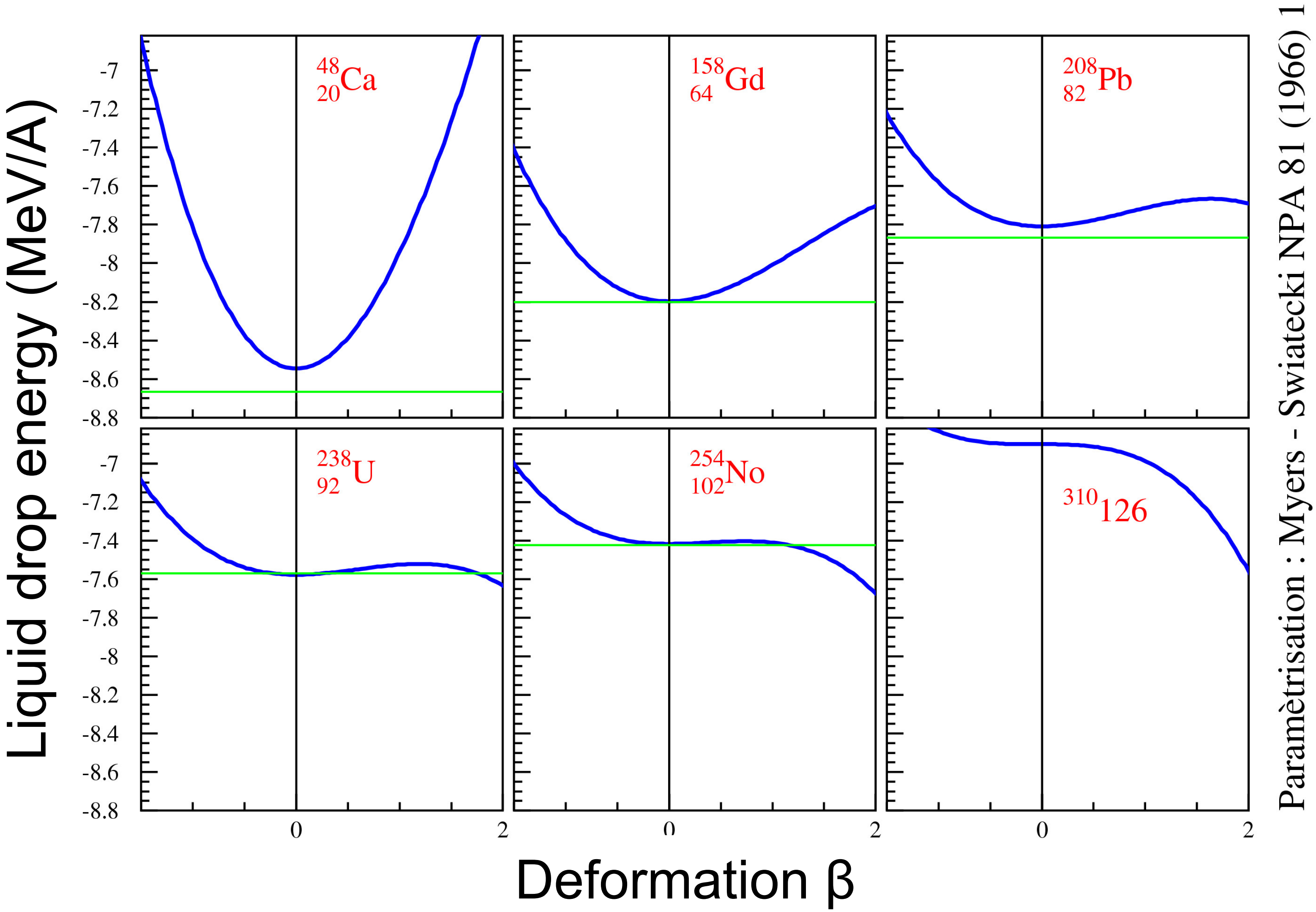
$V = 4/3 \pi a b^2$

$S = 4 \pi R^2 (1 + 2/5 \varepsilon^2 + \ldots)$

If BE(ε) -BE(ε=0)> 0: gain in energy with deformation

→ fission

Fission barrier – liquid drop



Notes

- From previous slide :

Stability = balance between surface and coulomb

Fissility parameter $x = E_{coulomb} / 2\, E_{surface}$

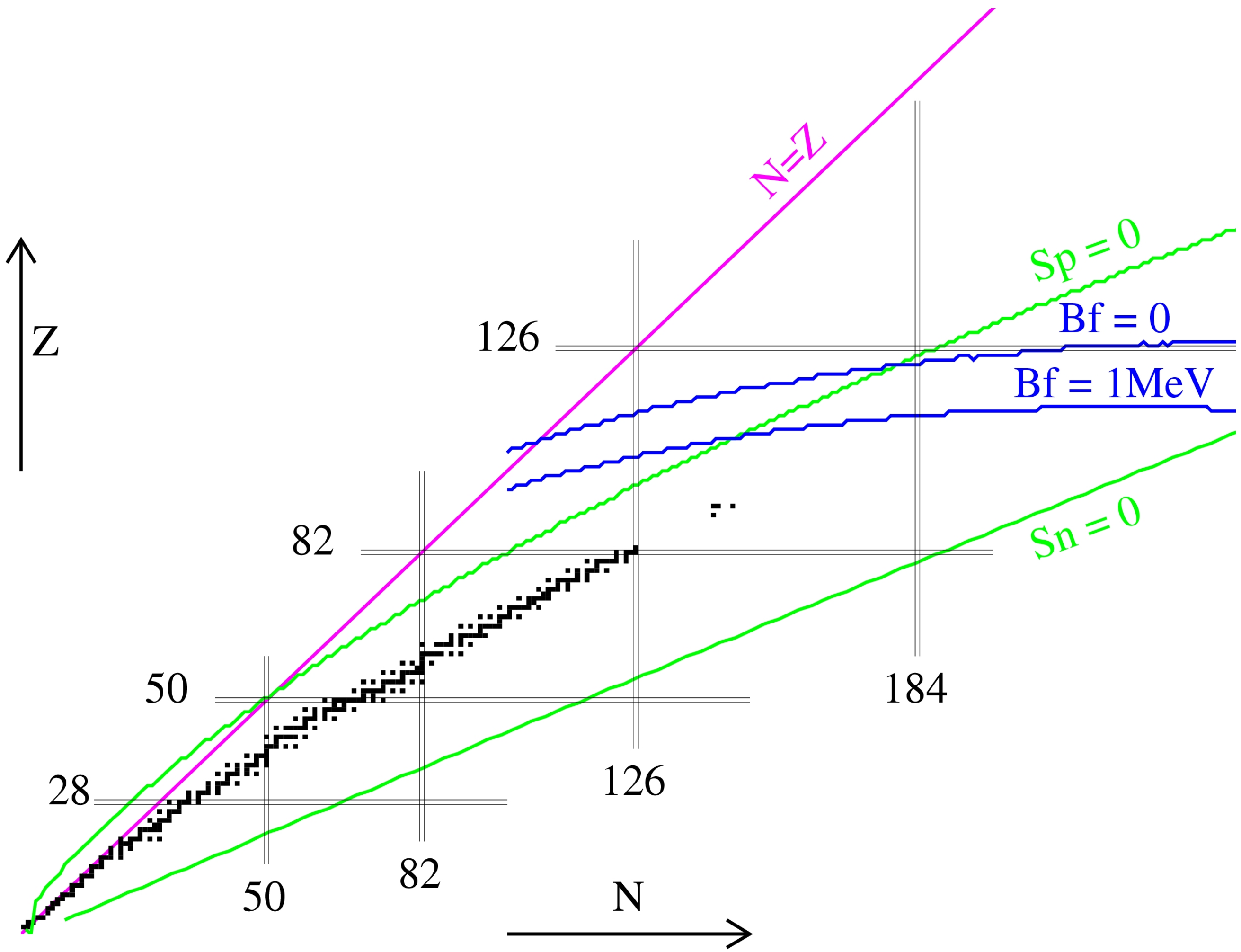
$\sim 1/50\; Z^2 / A$

scaling of the fission barrier

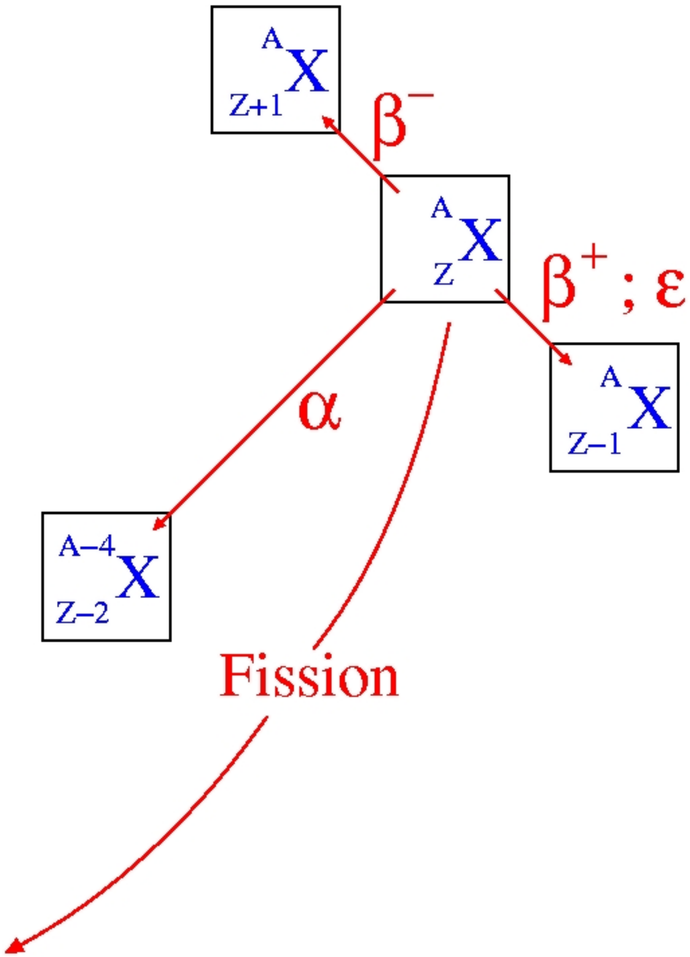
$x > 0.8$: no survival

- Possible definitions of SHE :
  - No macroscopic fission barrier
  - $B_f < 1$ MeV
  - $x > 0.8$

Limits of stability from liquid drop model



Decay modes



Alpha decay

$Q\alpha = B(A\text{-}4, Z\text{-}2) + B_{alpha} – B(A,Z)$

From liquid drop :

- $Q\alpha \uparrow$ with A and Z
- $Q\alpha \sim 0$ for A ~ 150 Mass

- Alpha decay is a favored decay mode of heavy elements

Limit of stability : positron emission

Nuclei for Z larger than 173 become unstable against positron emission.

This is because the most deeply bound electrons from the $1s_{1/2}$ shell reach an energy of -511 keV

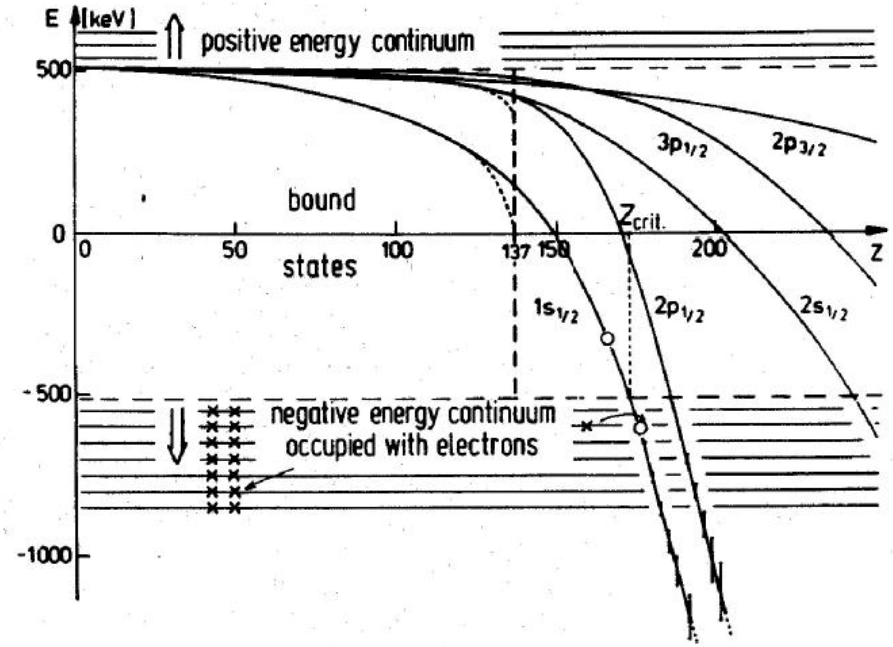
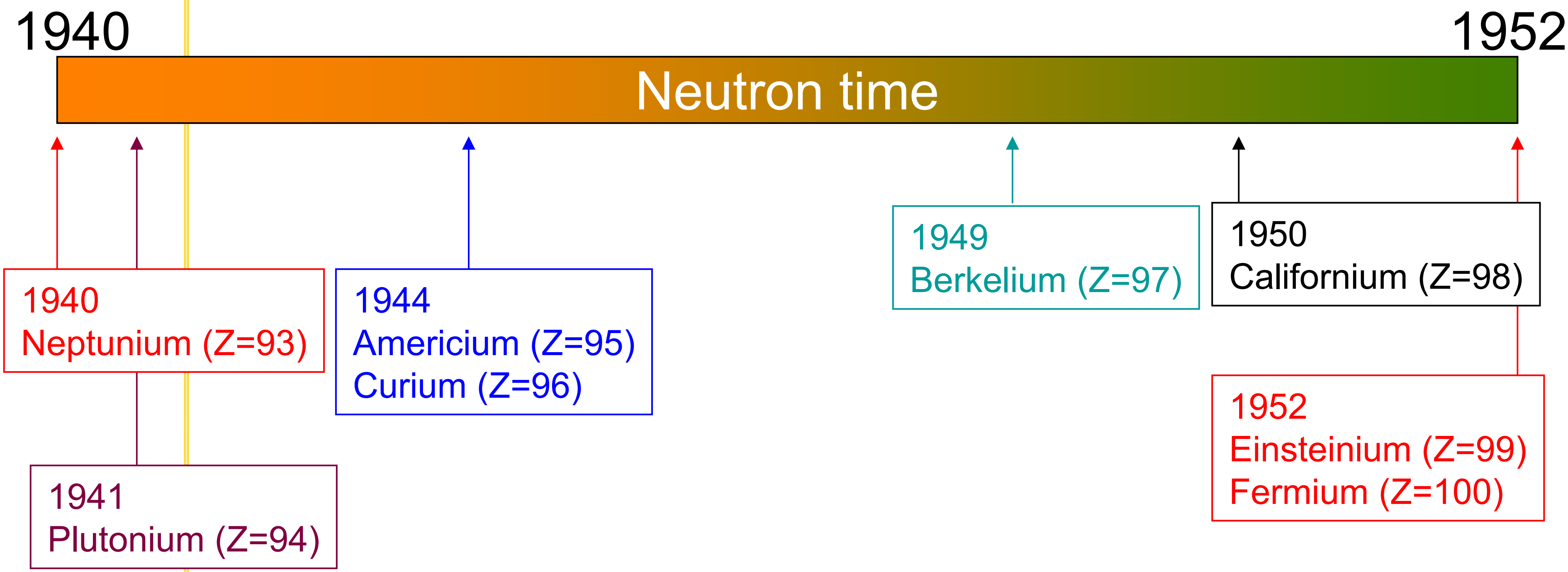


Fig. 2. Binding energies of electronic states in atoms as function of nuclear charge Z. At $Z_c = 173$ the $1s$-state dives into the negative energy continuum.

See eg W. Pieper, W. Greiner Z. Phys. A 218 (1968) 327
J. Reinhardt et al, Z. Phys. A 303 (1981) 173

History



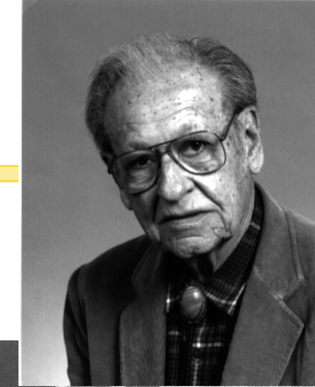
Neutron irradiation, reaction with light ions p, d, α
Mostly chemical separation

$$^{238}U_{92} + n \rightarrow {}^{239}U_{92} \rightarrow {}^{239}Np_{93} \quad (\beta \text{ decay}) \qquad (1940)$$

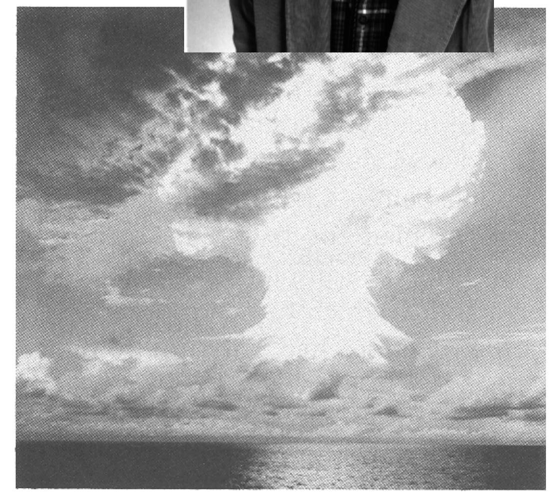
$$^{238}U_{92} + d \rightarrow {}^{238}Np_{93} + 2n \rightarrow {}^{238}Pu_{94} \quad (\beta \text{ decay}) \qquad (1941)$$

$_{99}$Es and $_{100}$Fm

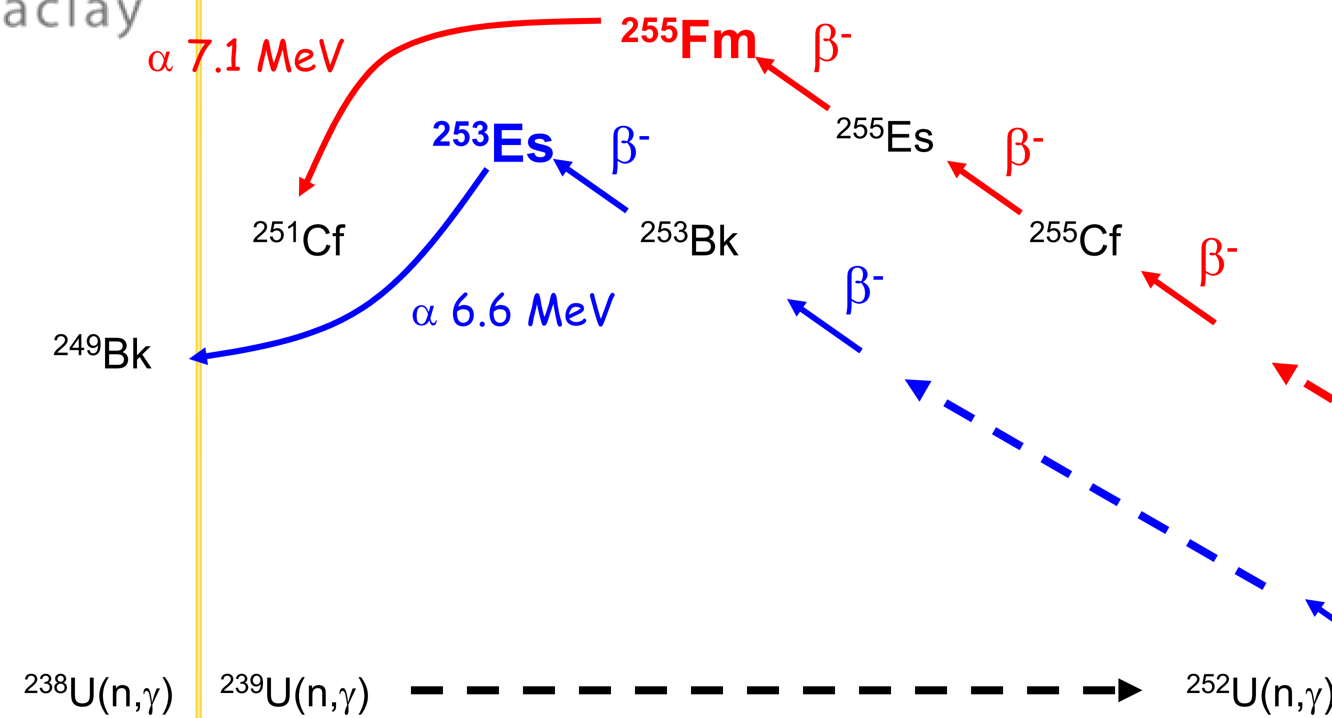
A. Ghiorso

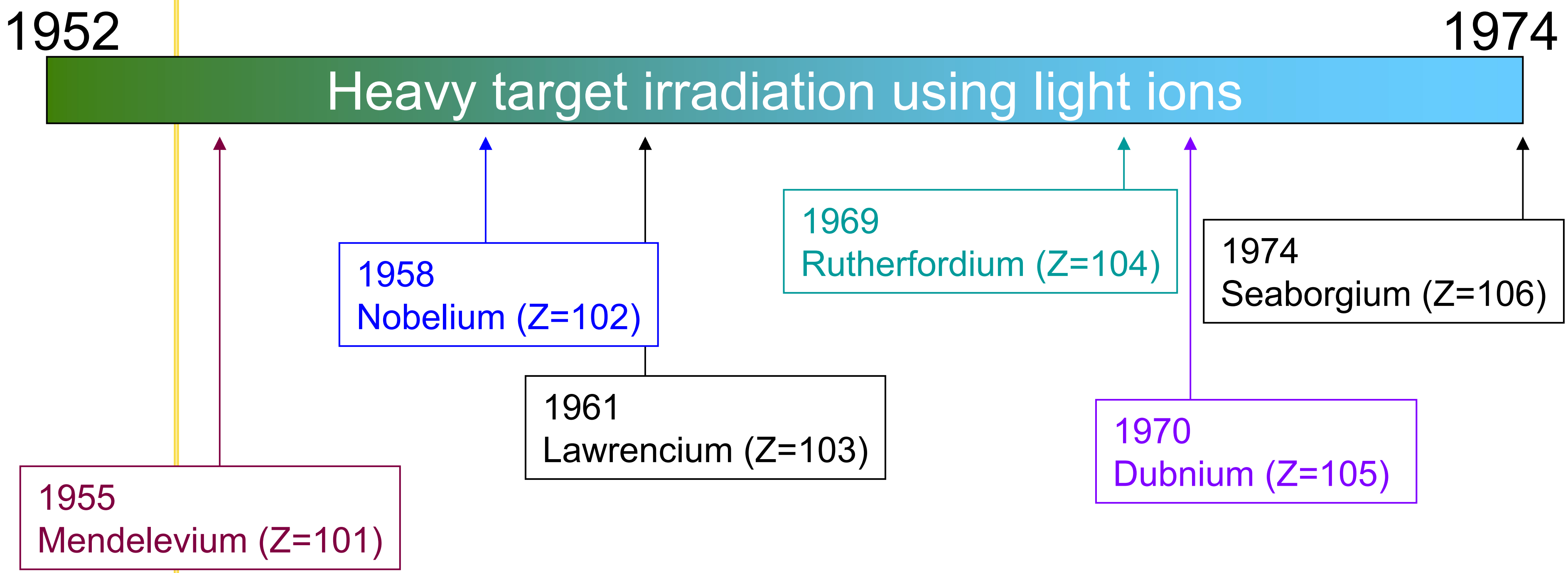


- $^{238}U(15n,7\beta^-)^{253}Es$ Chemical Separation – 1952
- $^{238}U(17n,8\beta^-)^{255}Fm$ Chemical Separation - 1953



Mike, 1/11/1952





"hot fusion" vs "cold war" USA – Soviet competition

- $^{12}C + ^{246}Cm \rightarrow ^{254}No + 4n$
- $^{15}N + ^{249}Cf \rightarrow ^{260}Db + 4n$

$_{101}$Md

A. Ghiorso, 1955

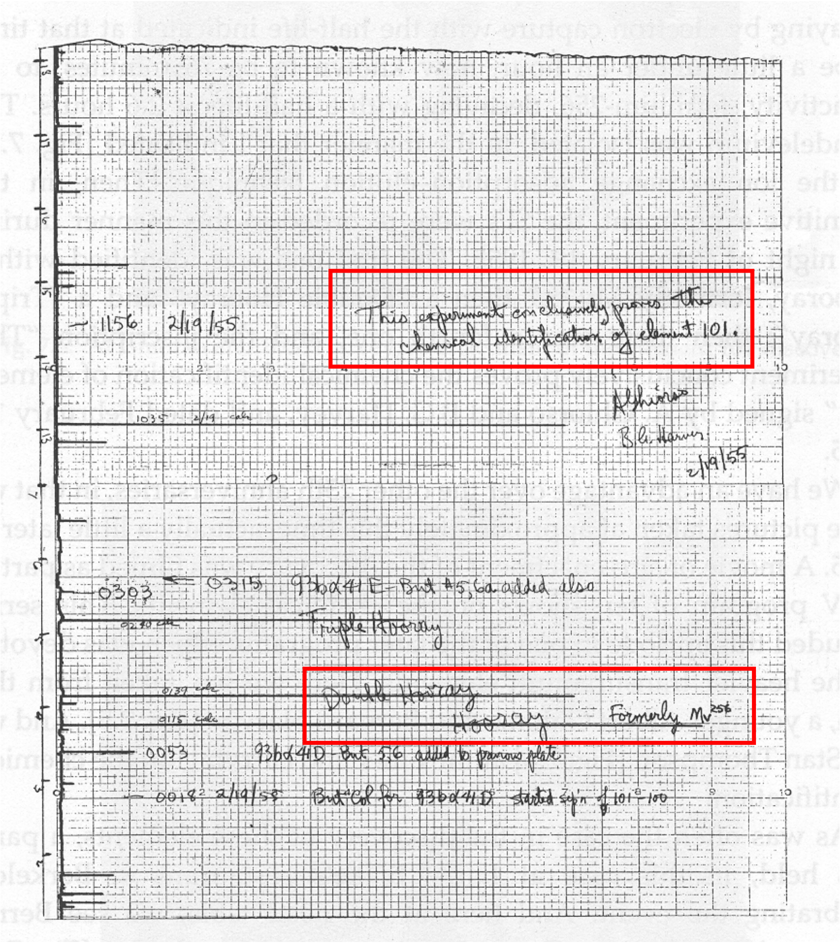
^{253}Es(α,n)^{256}Md

Target : $\simeq 10^9$ Es atoms produced using Pu irradiation

$I\alpha \simeq 10^{14}$ p.p.s.

Last element discovered using chemical separation

(17 atoms)



$_{104}$Rf

- 1964 : G.N. Flerov *et al.*, Dubna :

^{242}Pu+^{22}Ne   (→ 4n + ^{260}Rf)

Spontaneous fission $t_{1/2}$ = 14 ms.

→ Fission isomer in ^{242}Am

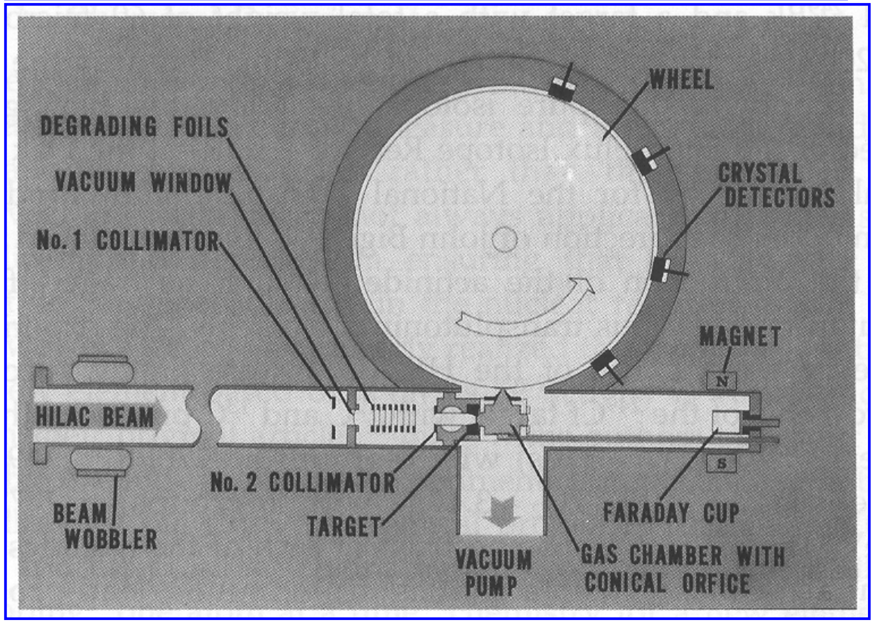
- 1969 : A. Ghiorso *et al.* :

^{249}Cf+^{12,13}C → ^{257,259}Rf →No

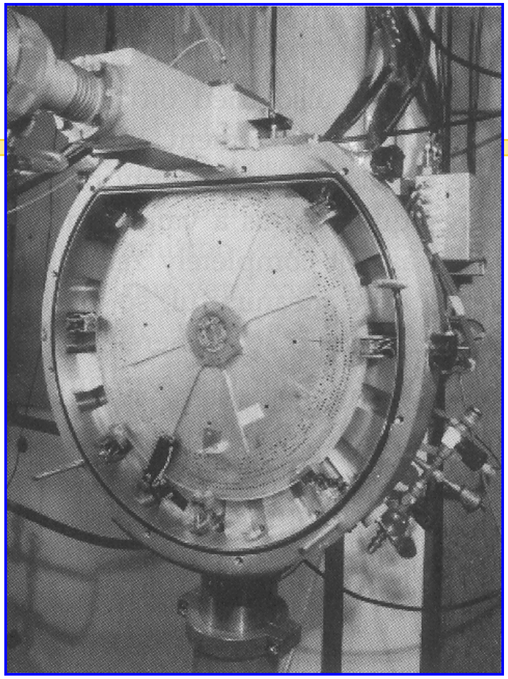
parent – daughter correlation : the genetic correlations

- Debates and controversy Berkeley/Dubna

- Joined discovery Berkeley / Dubna



$_{106}Sg$



- 1971, Berkeley $^{249}Cf + ^{18}O$: α decay candidate
- upgrade HILAC (superHILAC) α correlations analysis using a computer
- 1974 A. Ghiorso *et al.*

$^{249}Cf(^{18}O, 4n)^{263}106 \rightarrow ^{249}Rf \rightarrow ^{255}No$

- 1974 Yu.Ts. Oganessian (Dubna) :

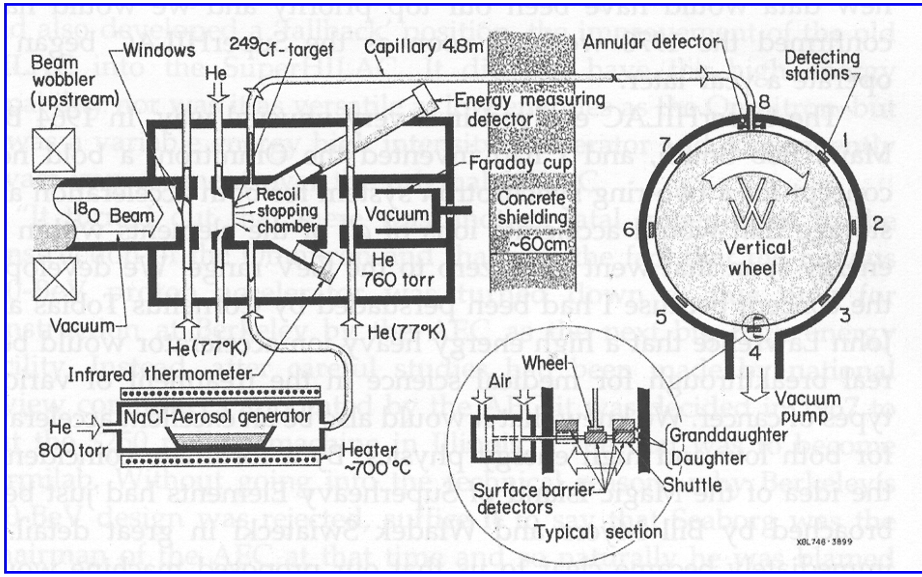
cold fusion

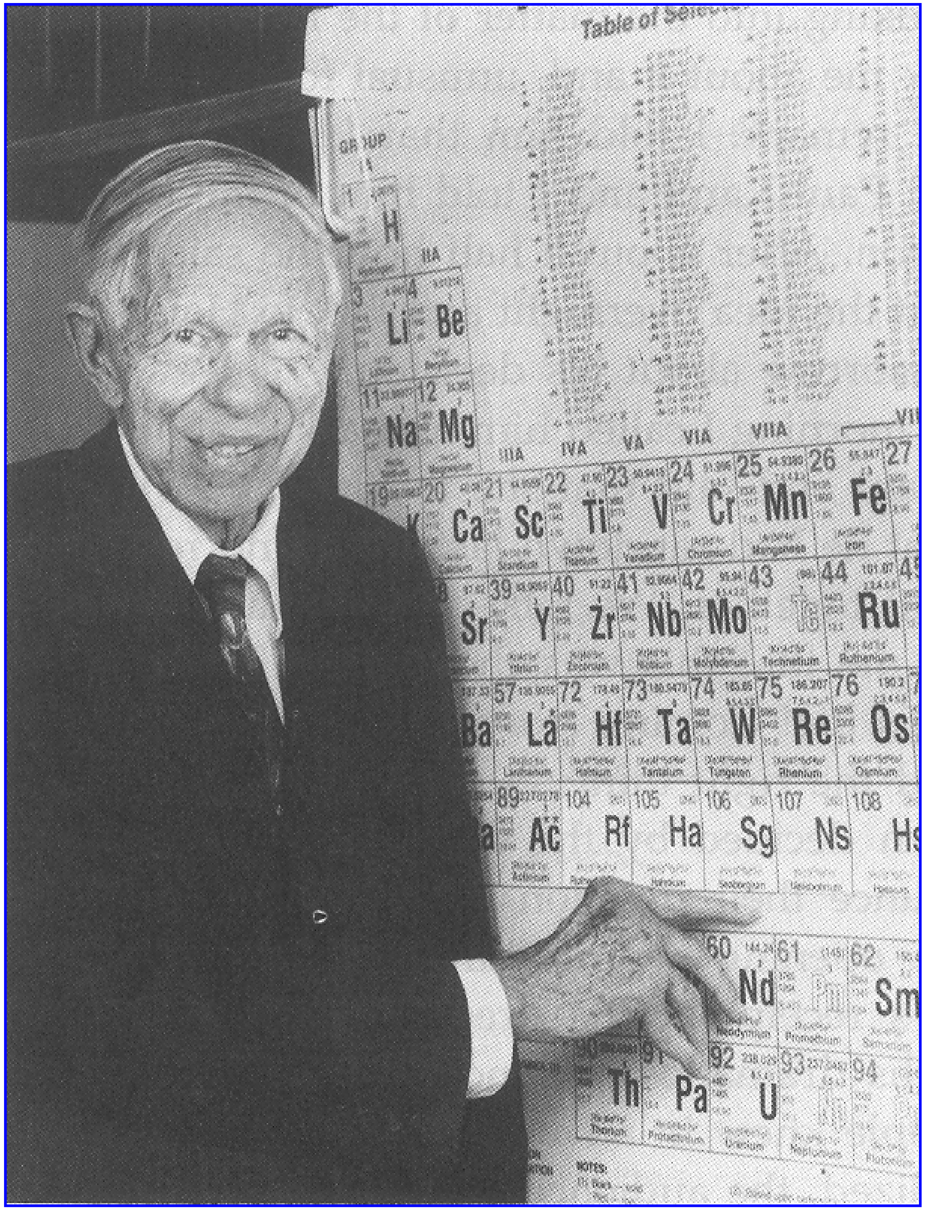
$^{208}Pb + ^{40}Ar \rightarrow ^{244,246}Fm$

$^{208}Pb(^{54}Cr,3,5n)^{257,259}106 \rightarrow S.F.$

- 1984 : S.F. entirely due to $^{256}104$
- 1997 : element named Seaborgium

- Discovery Berkeley

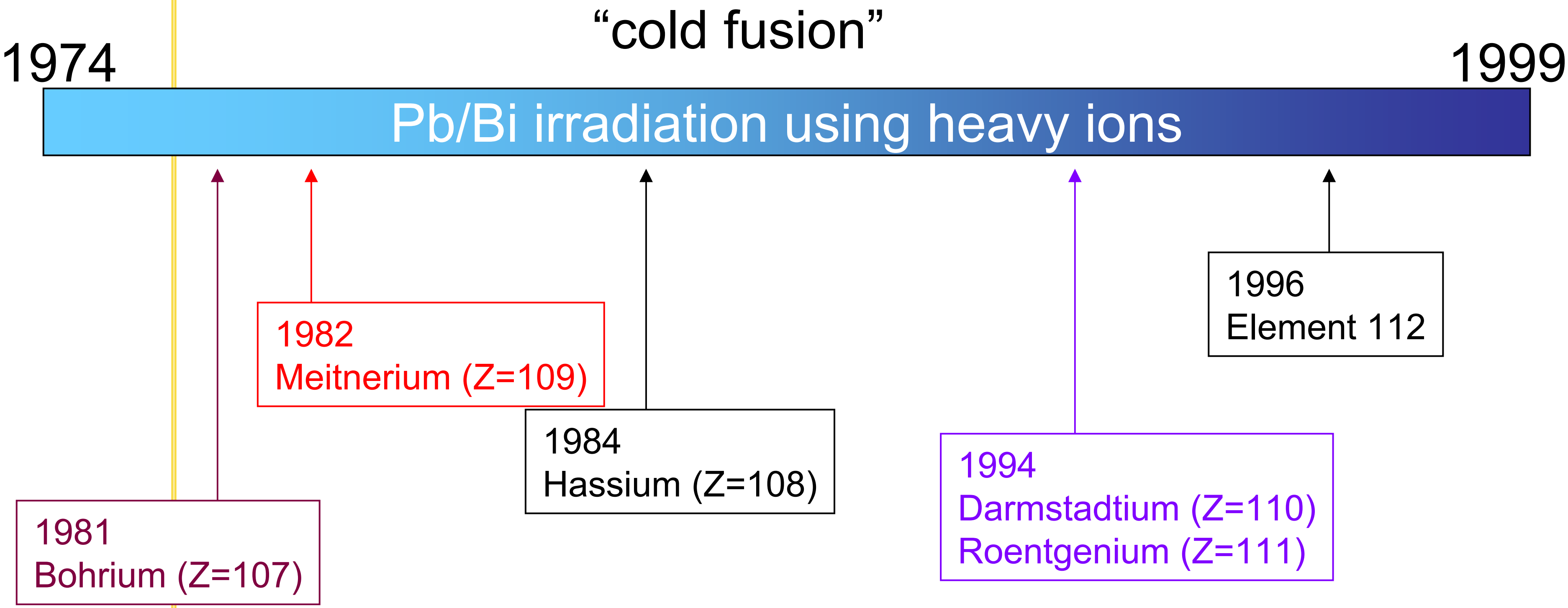




Glenn T. Seaborg
(1912-1999)

Discovered $_{94}Pu$, $_{95}Am$, $_{96}Cm$, $_{97}Bk$, $_{98}Cf$.

What's wrong on this picture ?



Examples

$^{54}Cr + ^{209}Bi \rightarrow ^{262}Bh + n$

$^{62}Ni + ^{208}Pb \rightarrow ^{269}Ds + n$

$_{107}$Bh, $_{108}$Hs, $_{109}$Mt, $_{110}$Ds, $_{111}$Rg, Z=112

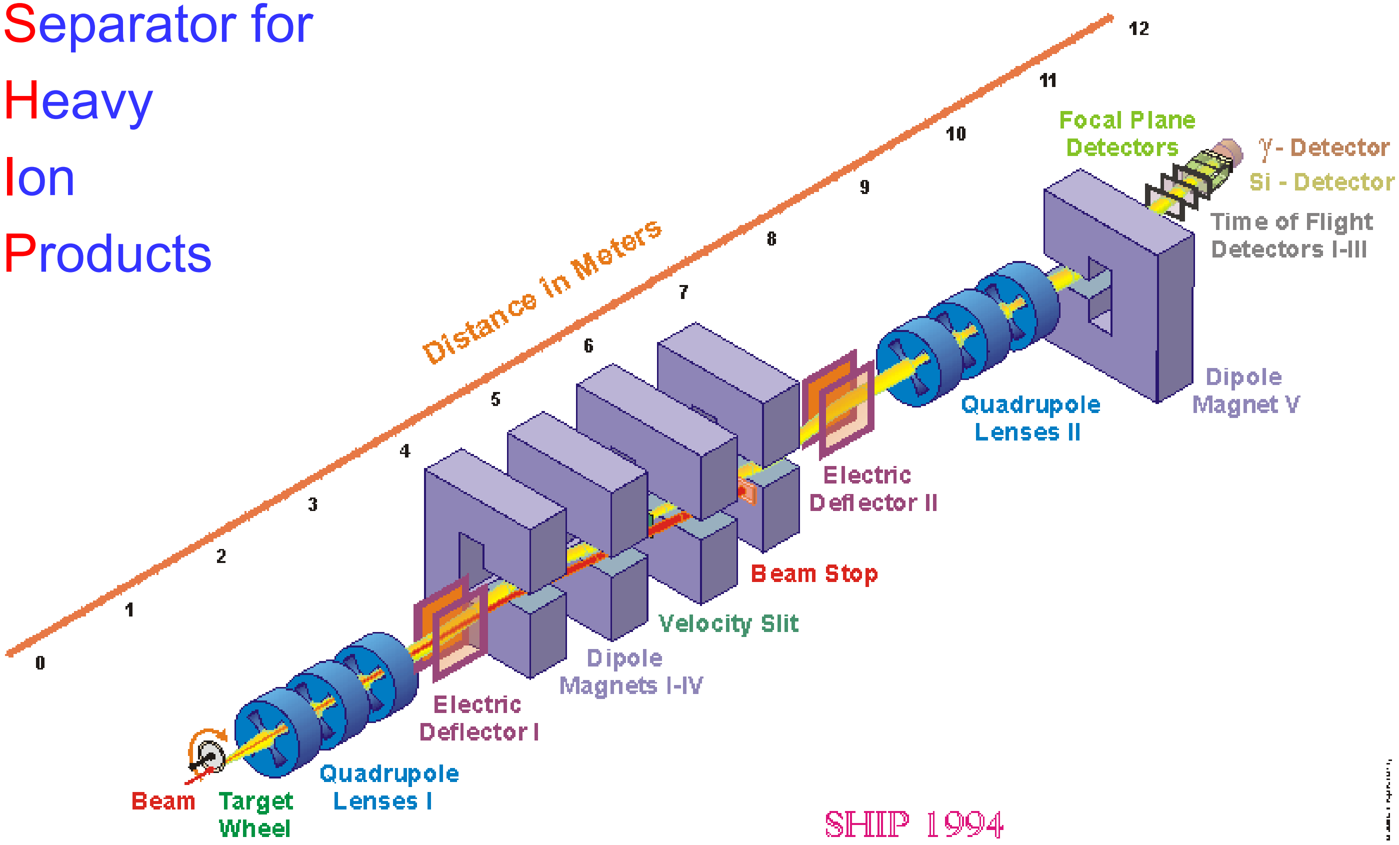
Irfu cea saclay

70th : G.S.I.; S.H.I.P. (P. Ambruster); 1975 : first UNIversal Linear ACcelerator beam

- 1981 $_{107}$Bh (G. Münzenberg *et al.)*
  $^{209}Bi(^{54}Cr,1n)^{262}Bh \rightarrow {}^{258}Db \rightarrow \ldots \rightarrow {}^{250}Fm$
- 1982 $_{109}$Mt (G. Münzenberg *et al.)*
  $^{209}Bi(^{58}Fe,1n)^{266}Mt \rightarrow {}^{262}Bh \rightarrow {}^{258}Db$
- 1984 $_{108}$Hs (G. Münzenberg *et al.)*
  $^{208}Pb(^{58}Fe,1n)^{265}Hs \rightarrow {}^{261}Sg \rightarrow {}^{257}Rf$
- 1994 $_{110}$Ds, $_{111}$Rg (S. Hofmann *et al.)*
  $^{208}Pb(^{62}Ni,n)^{269}Ds \rightarrow {}^{265}Hs \rightarrow \ldots$
  $^{209}Bi(^{64}Ni,n)^{272}Rg \rightarrow {}^{268}Mt \rightarrow \ldots$
- 1996 Z=112 *(*S. Hofmann *et al.)*
  $^{208}Pb(^{70}Zn,1n)^{277}112 \rightarrow {}^{273}Ds \rightarrow \ldots$

Irfu
cea
saclay

Separator for Heavy Ion Products



Limits of cold fusion ?

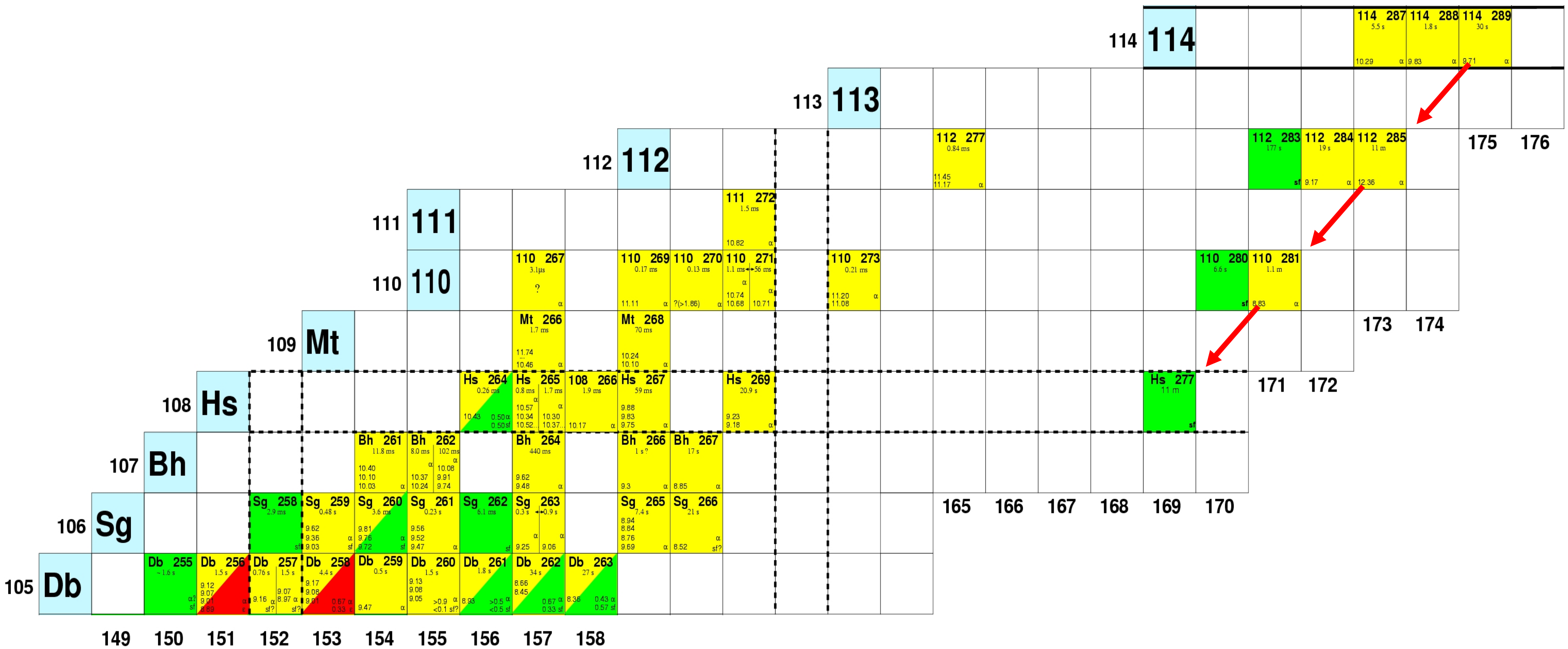
Evidences for Z = 113 at Riken (2003) $^{209}Bi(^{70}Zn,n)^{278}113$

Cross-section ~55 fb

Z=112 to 118 …

1999 Dubna, rebirth of hot fusion

Actinide targets eg $^{244}Pu(^{48}Ca,3n)^{289}114$



Z=112 to 118 …

1999 - Collaboration Dubna/Livermore

$^{244}Pu(^{48}Ca,3n)^{289}114 \rightarrow ^{285}112 \rightarrow \ldots$

2000 – element 116

$^{248}Cm(^{48}Ca,3n)^{292}116 \rightarrow ^{288}112 \rightarrow \ldots$

2003 - element 115 and 113

$^{243}Am(^{48}Ca,3n)^{288}115 \rightarrow ^{284}113 \rightarrow \ldots$

2005 - element 118

$^{249}Cf(^{48}Ca,3n)^{294}118 \rightarrow ^{290}116 \rightarrow ^{286}114 \rightarrow ^{282}112 \rightarrow \ldots$

SHE in nature

- Have SHE been produced in nucleosynthesis ?
  - The r-process
- Search for SHE in nature
  - Terrestrial sources
  - Extraterrestrial sources (meteorites, moon)
  - Cosmic sources

- More in :
  - G. Herrmann. Physica Scripta 10A (1974) 71.
  - G.N. Flerov, G.M. Ter-Akopian. Pure & Appl. Chem. 53 (1981) 909.

Production rates

Reaction rate = I beam x
Target thickness x
Cross section

Example : $^{208}Pb(^{48}Ca,2n)^{254}No$

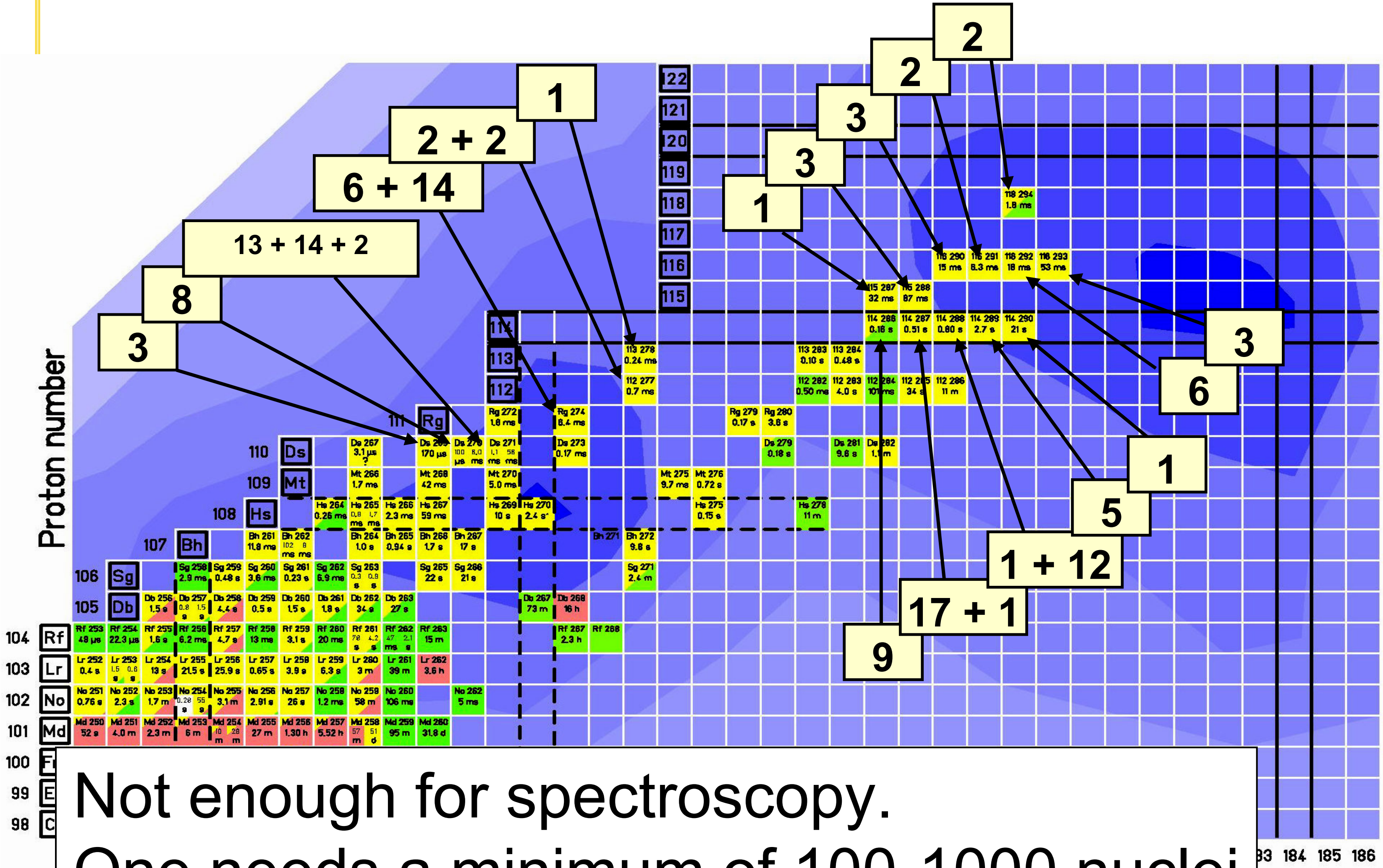
- Beam intensity
  100 pnA = $10^{-7}/1.6\ 10^{-19}\ s^{-1} = 0.625\ 10^{12}\ s^{-1}$
- Target Thickness
  $300\ \mu g/cm^2 = 300\ 10^{-6} \times N_A/208\ cm^{-2} = 8.68\ 10^{17}\ cm^{-2}$
- Cross section
  $\sigma = 2\mu b = 2\ 10^{-6} \times 10^{-24}\ cm^2 = 2\ 10^{-30}\ cm^2$

$\rightarrow$ Reaction rate ~ 1/s

$^{48}Ca + ^{238}U \rightarrow Z=112$, $\sigma \approx$ 1pb

$\rightarrow$ Reaction rate ~ 1/month

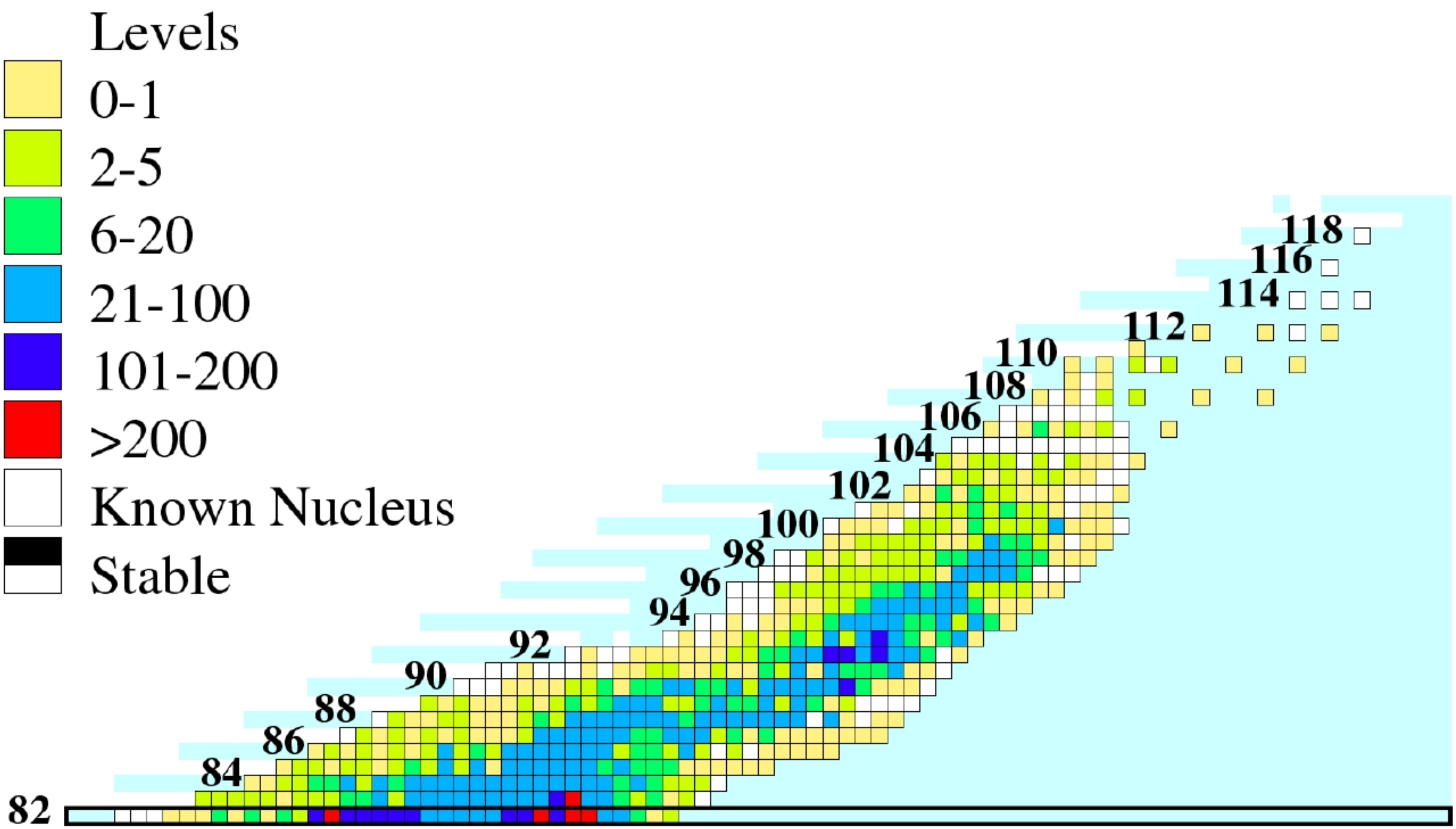
State of the art : synthesis



Not enough for spectroscopy.

One needs a minimum of 100-1000 nuclei

State of the art

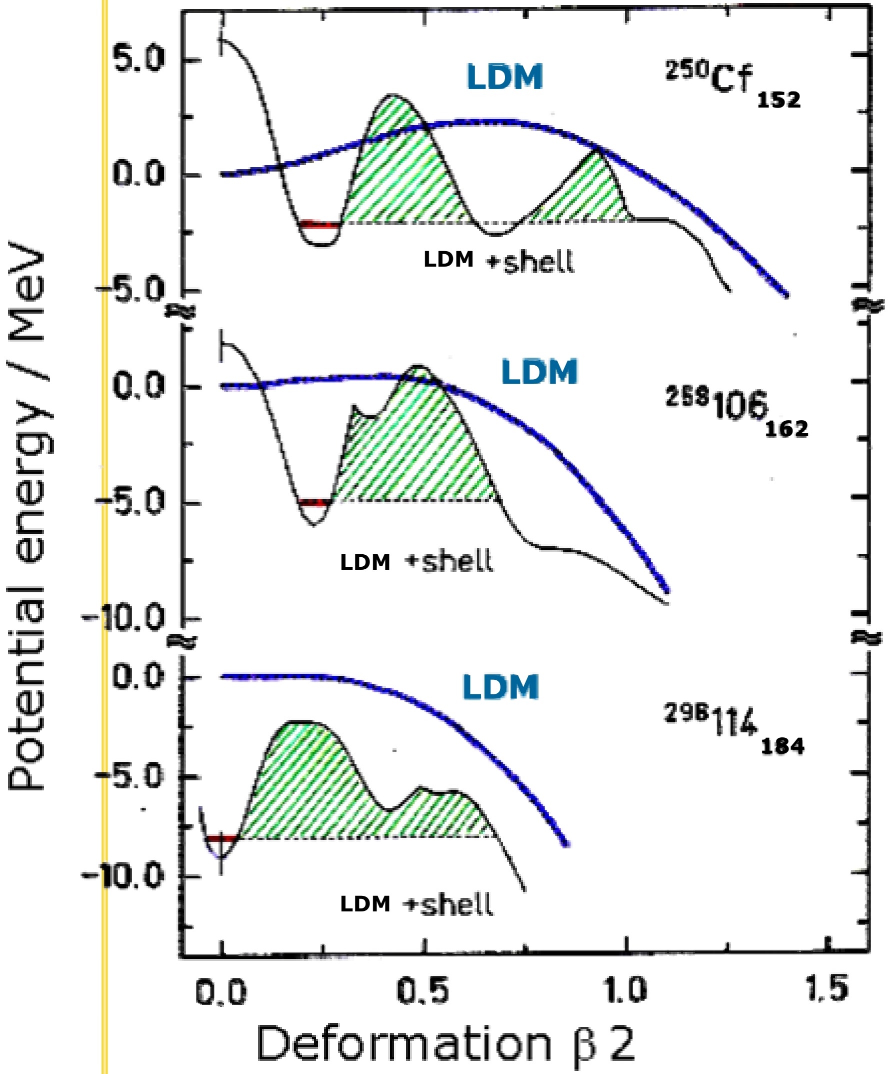


Stability of heavy elements : microscopic aspects

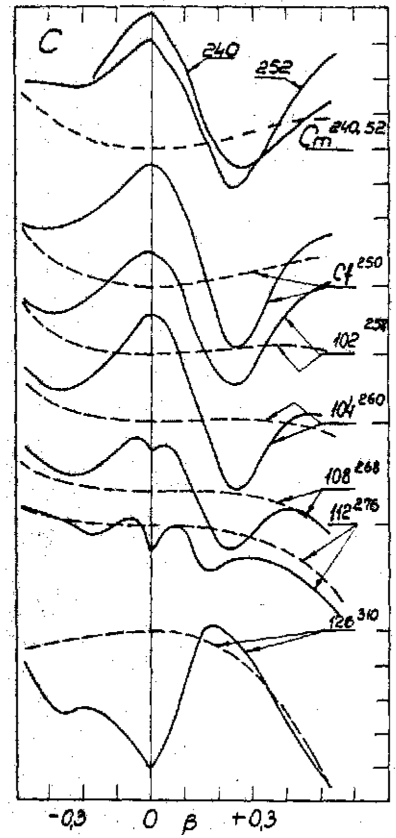
Shell effects

Oscillator :
level bunching ↔ gaps

Enhanced stability due to
Shell effects



- **Sobiczewski et al.**



V.M.Strutinsky
Nucl. Phys. A95 (1967) 420

Microscopic models

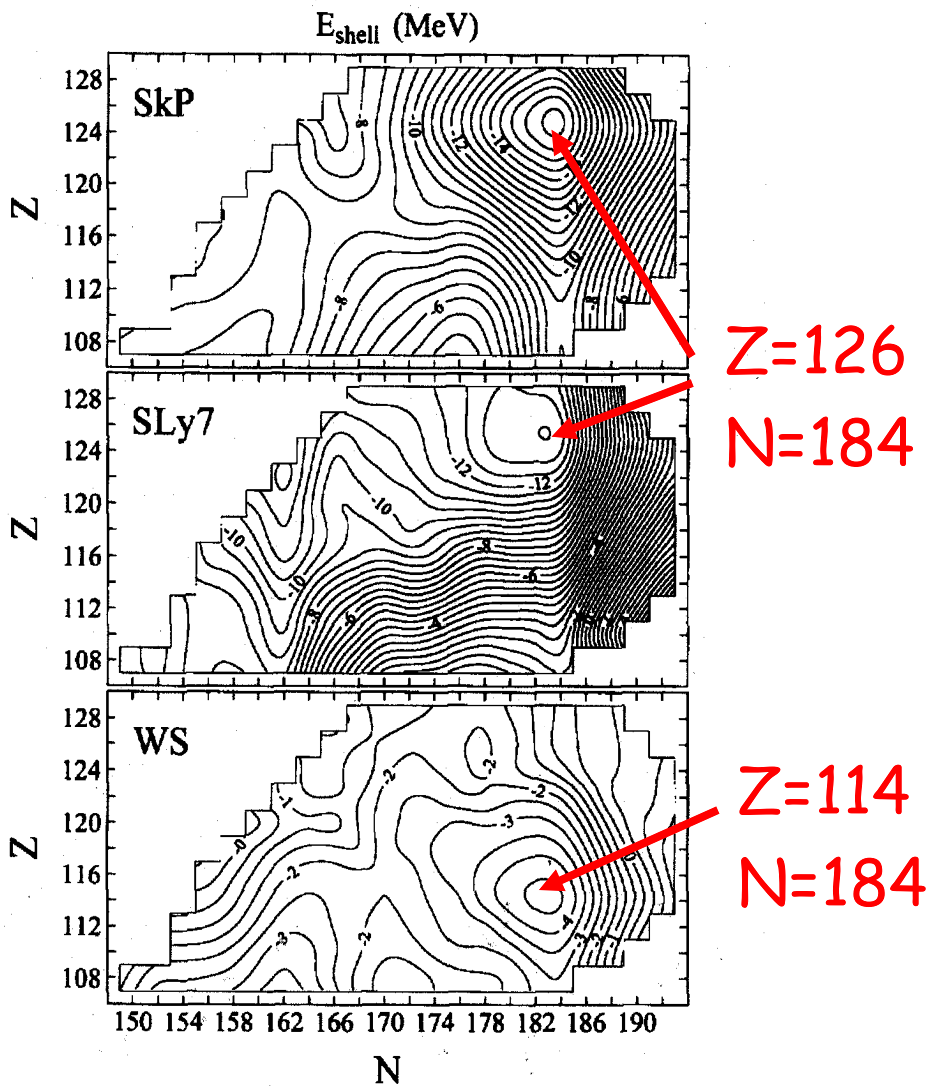
- Phenomenological
  - Nilsson, Woods Saxon

- Hartree – Fock
  - Effective nuclear interaction (Skyrme, Gogny)

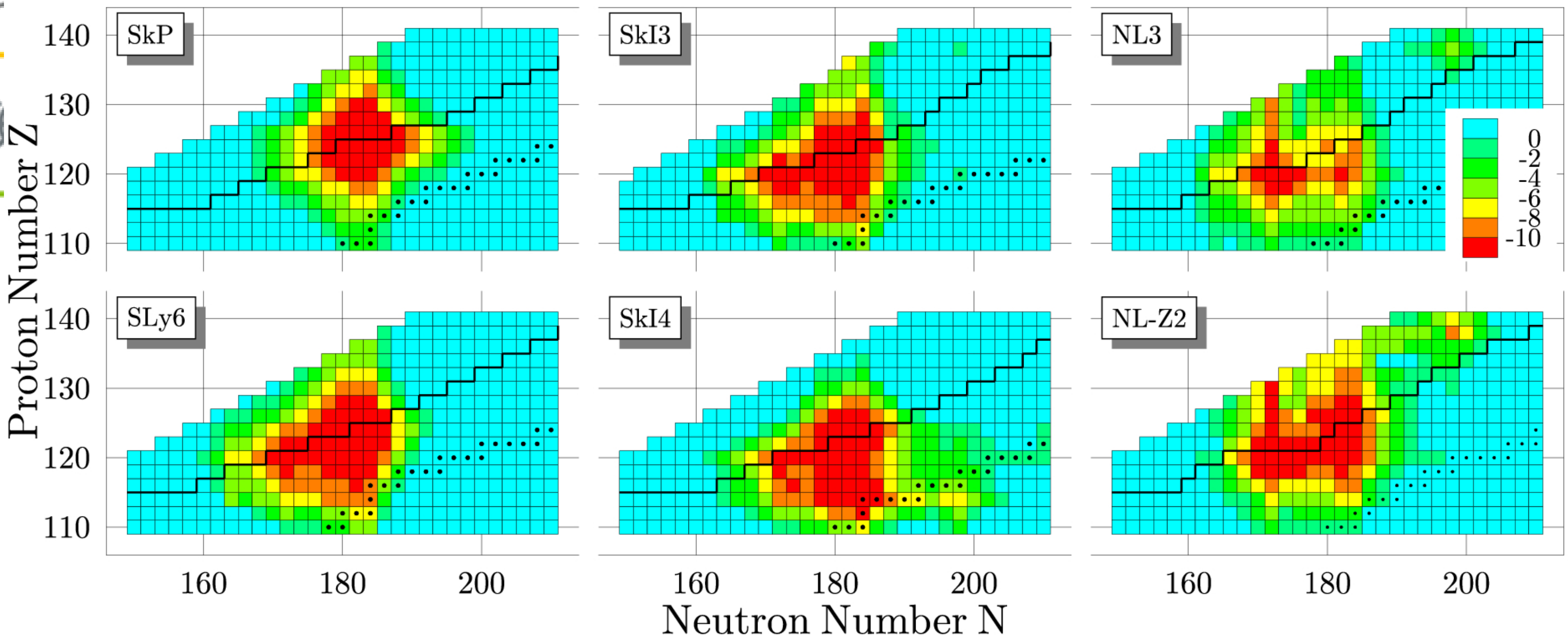
    → Mean - field

- Relativistic mean-field
  - Nucleon-nucleon interaction : meson + photon exchange

    → Mean - field



S. Cwiok *et al.* Nucl. Phys. A 611 (1996) 211



M. Bender *et al* . PL B515 (2001) 42

Magic numbers

	Z	N
W.S	114	184
HFB	126	184
RMF	120	172

Note 1 :Up to ^{208}Pb : proton and neutron magic numbers identical.

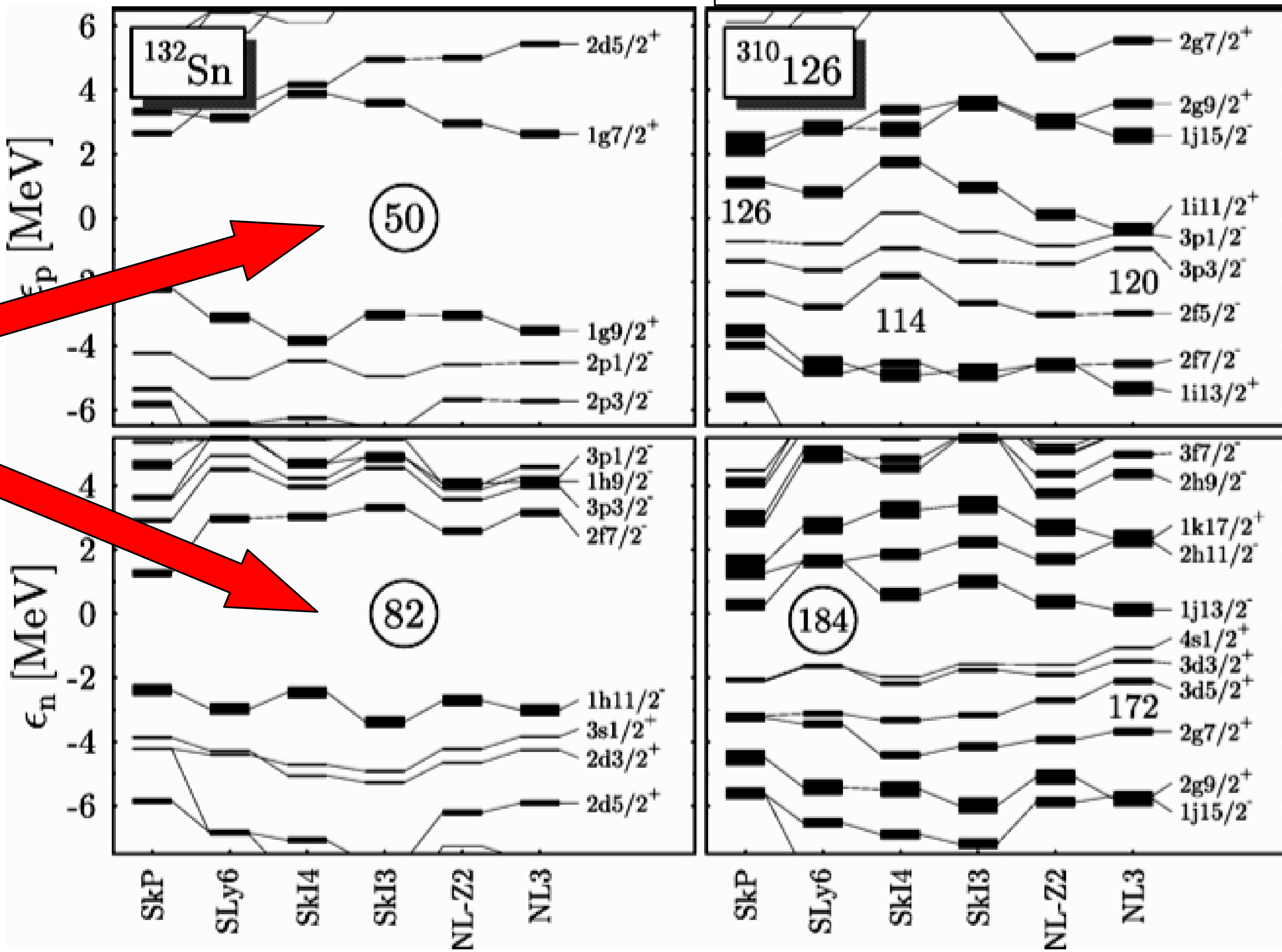
Note 2 : Models rely on extrapolations –parameters are adjusted on known cases

Theoretical challenges

Super-heavies :
Gap function of models
and not marked

Level density increases with A, Z

^{132}Sn :
Large
gap



M. Bender et al., Phys. Lett. B 515 (2001) 42

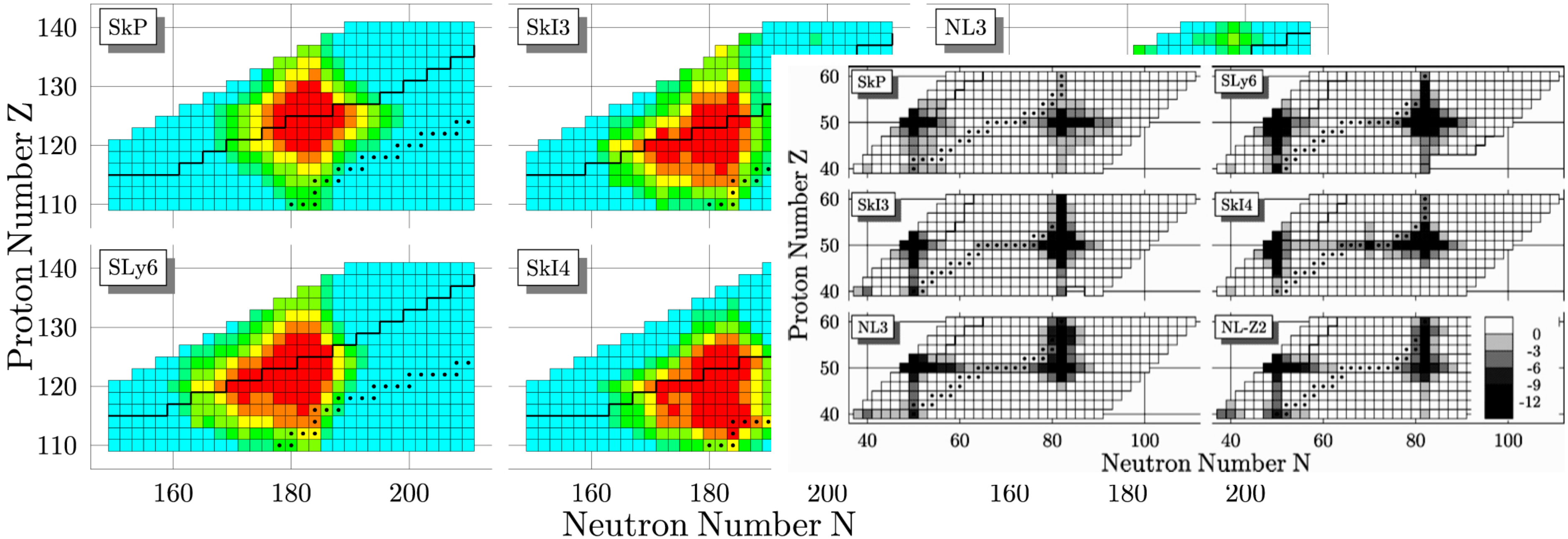
Theoretical challenges

Doubly magic character of predicted SHE not as marked as lighter Nuclei such as ^{48}Ca, ^{208}Pb, …

Do not expect a rigid spherical nucleus with a nice first 3^- excited state

Island of stability smooth and not well localized.

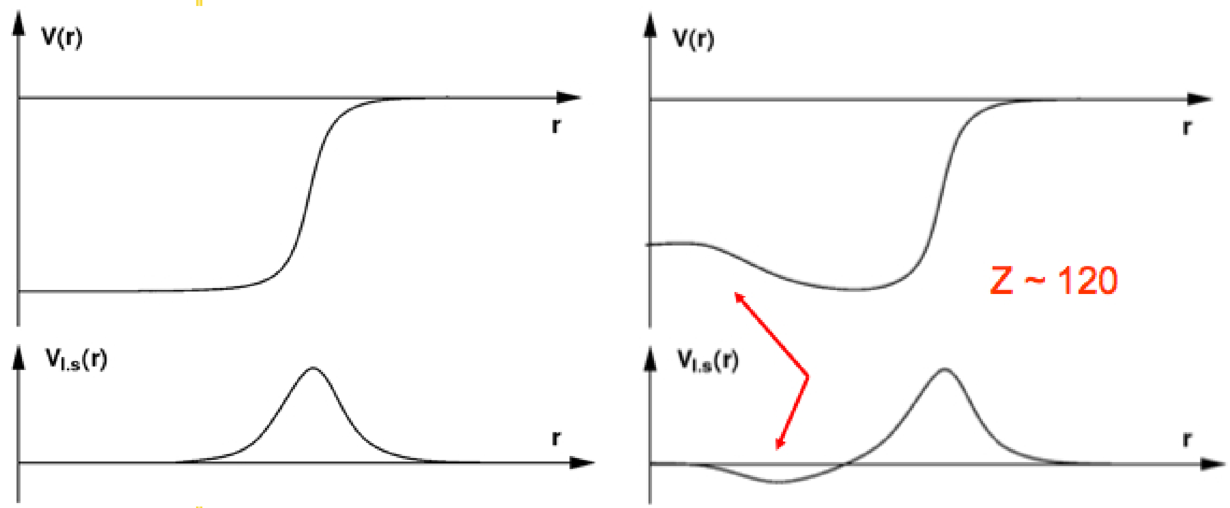
Can we still have magic numbers / nuclei ?

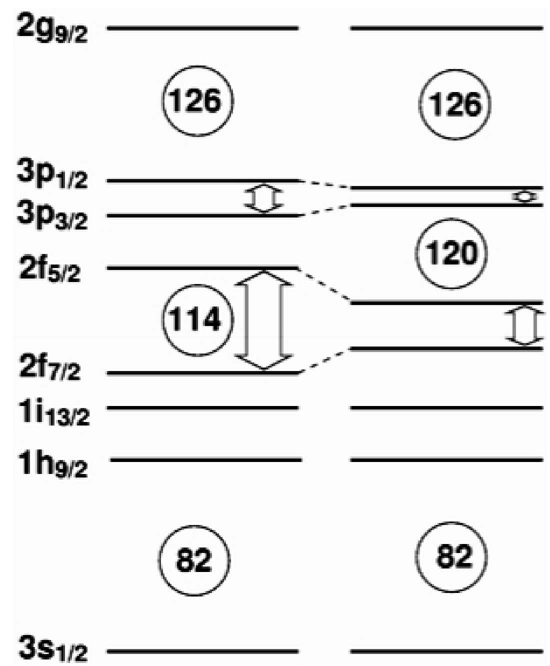


Theoretical challenges : the spin-orbit

Irfu cea saclay

$$V_{l.s}(r) = -\frac{1}{r}\frac{\partial V(r)}{\partial r}$$



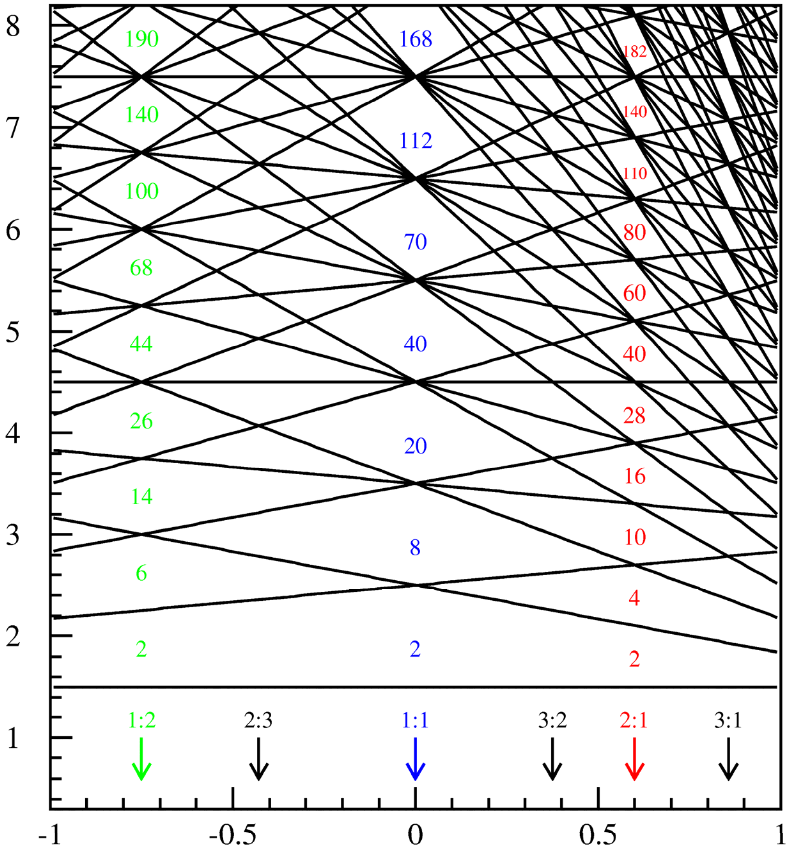


Effect of spin orbit contribution cancelled or reversed

Splitting $2f_{5/2}$ $2f_{7/2}$

Stability and deformation

Deformation → new gaps



$\delta = (\omega_\perp - \omega_z)/\omega_0$

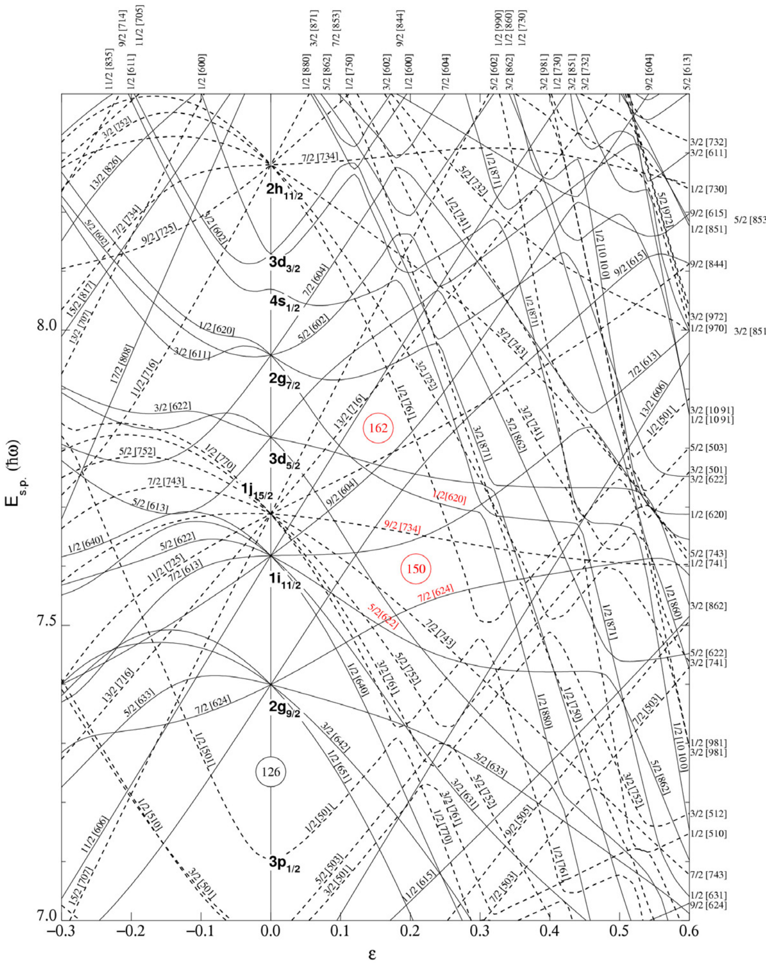
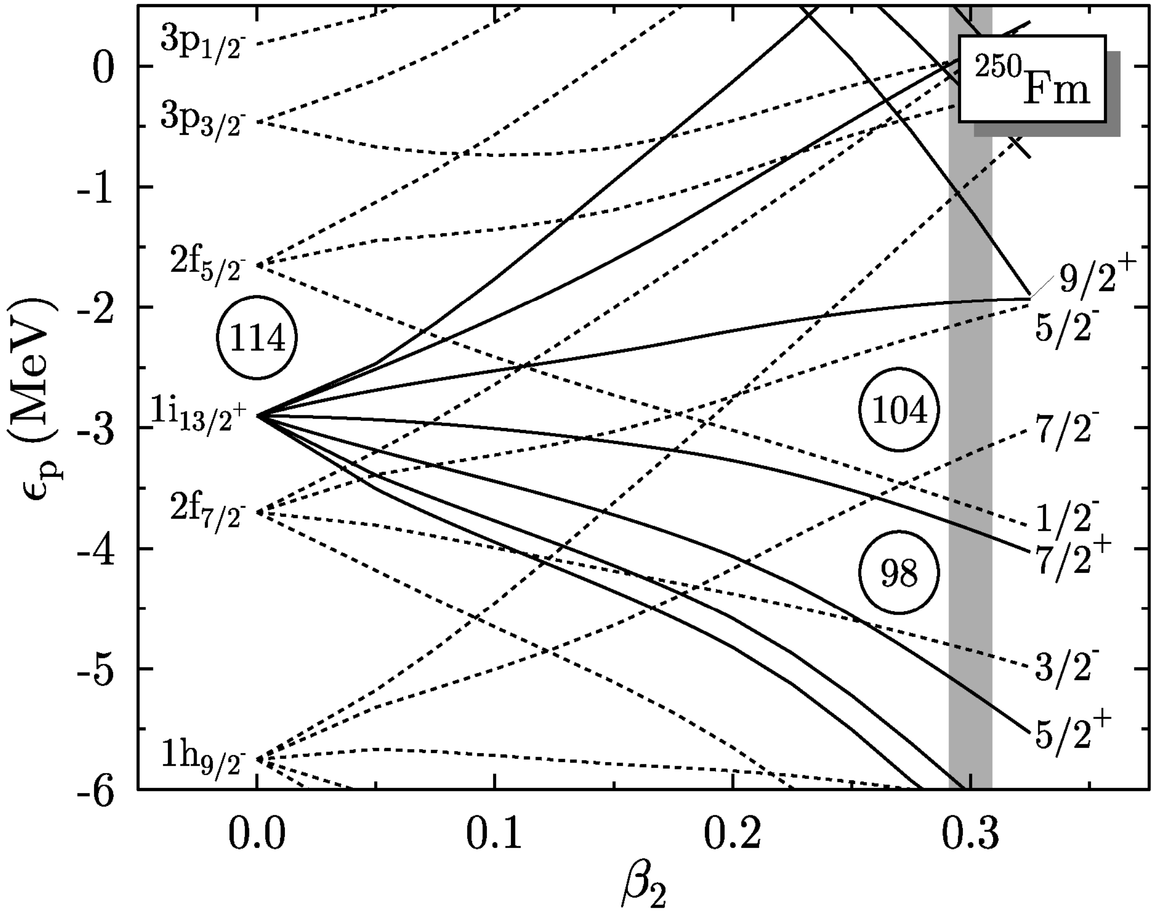


Figure 10. Nilsson diagram for neutrons, N ≥ 126 ($\varepsilon_4 = -\varepsilon_2^2/6$).

New shell gaps as a function of deformation : ^{254}No, ^{270}Hs

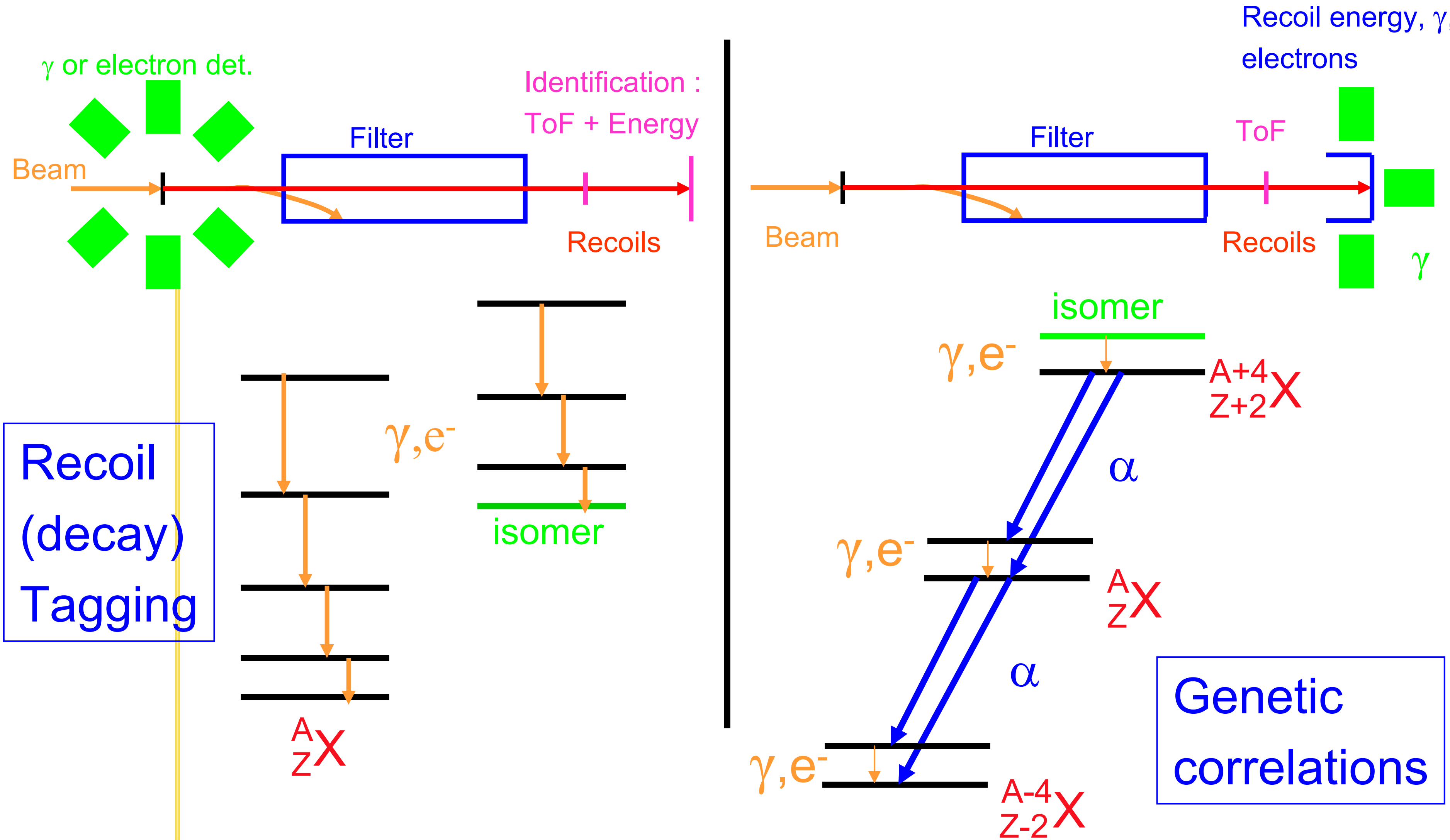


Access to high j deformed orbitals :
probe of higher lying spherical orbitals e.g. $\pi 2f_{5/2}$

Experimental techniques

Prompt spectroscopy

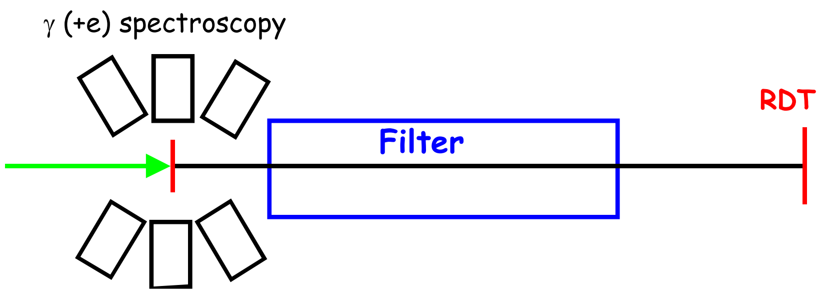
Decay spectroscopy



Recoil (decay) Tagging

Genetic correlations

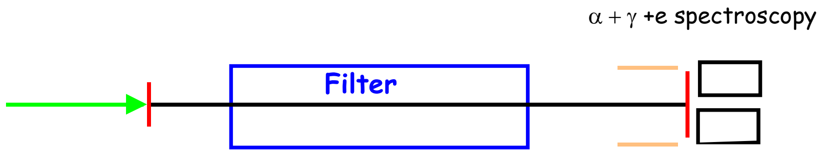
Prompt spectroscopy



- ☺ High angular momentum
- 😐 Electron spectroscopy difficult
- ☹ Counting rate limitation
- ☹ « low » statistics

Collective properties

Decay spectroscopy

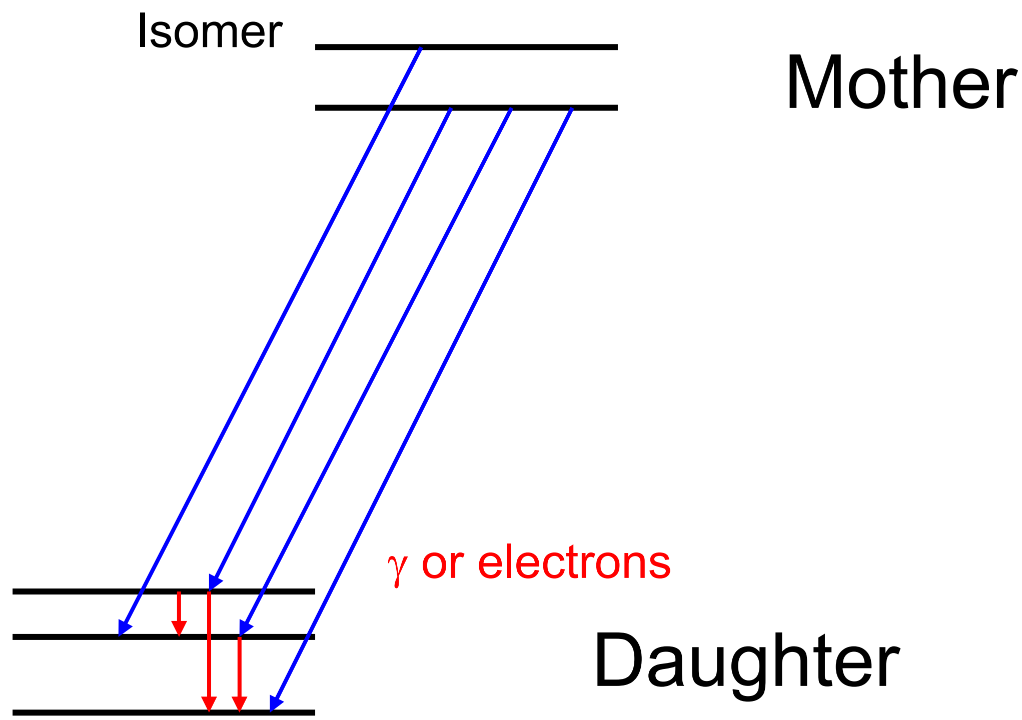


☺ High beam intensity
☺ Low counting rate after filter
☺ Spectroscopy after α decay
☹ Few states available; low angular momentum
☺ Electron spectroscopy « easy »
☺ Isomer spectroscopy

Odd nuclei, sp states

Decay spectroscopy

Alpha decay



Goal: deduce (at least)

- Qα
- level energies
- Spin and parity of levels (including g.s.)

Recoil selection

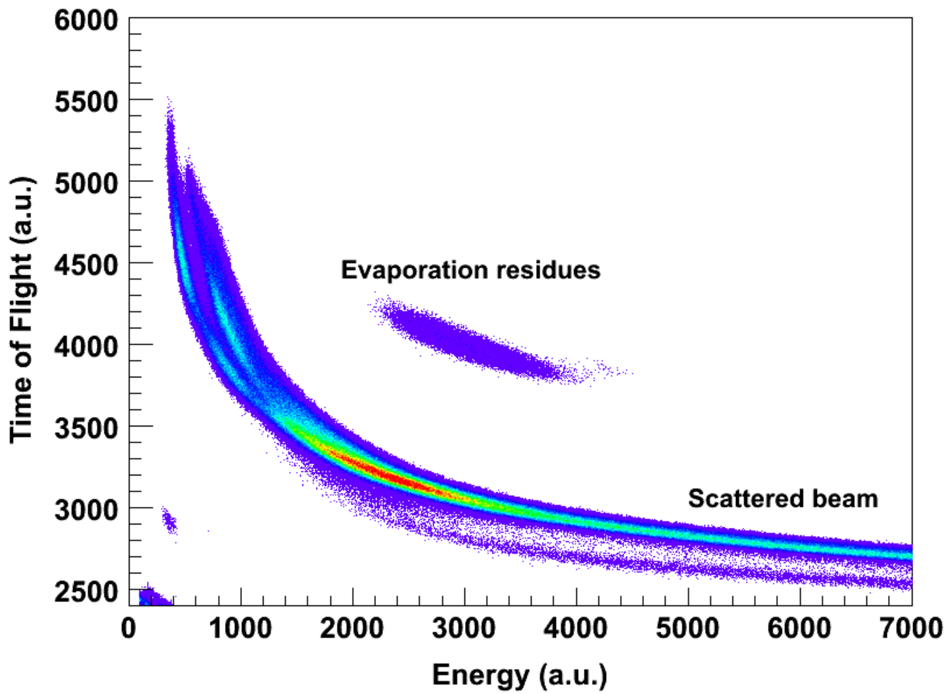
Selection using a separator or spectrometer

Then detection using

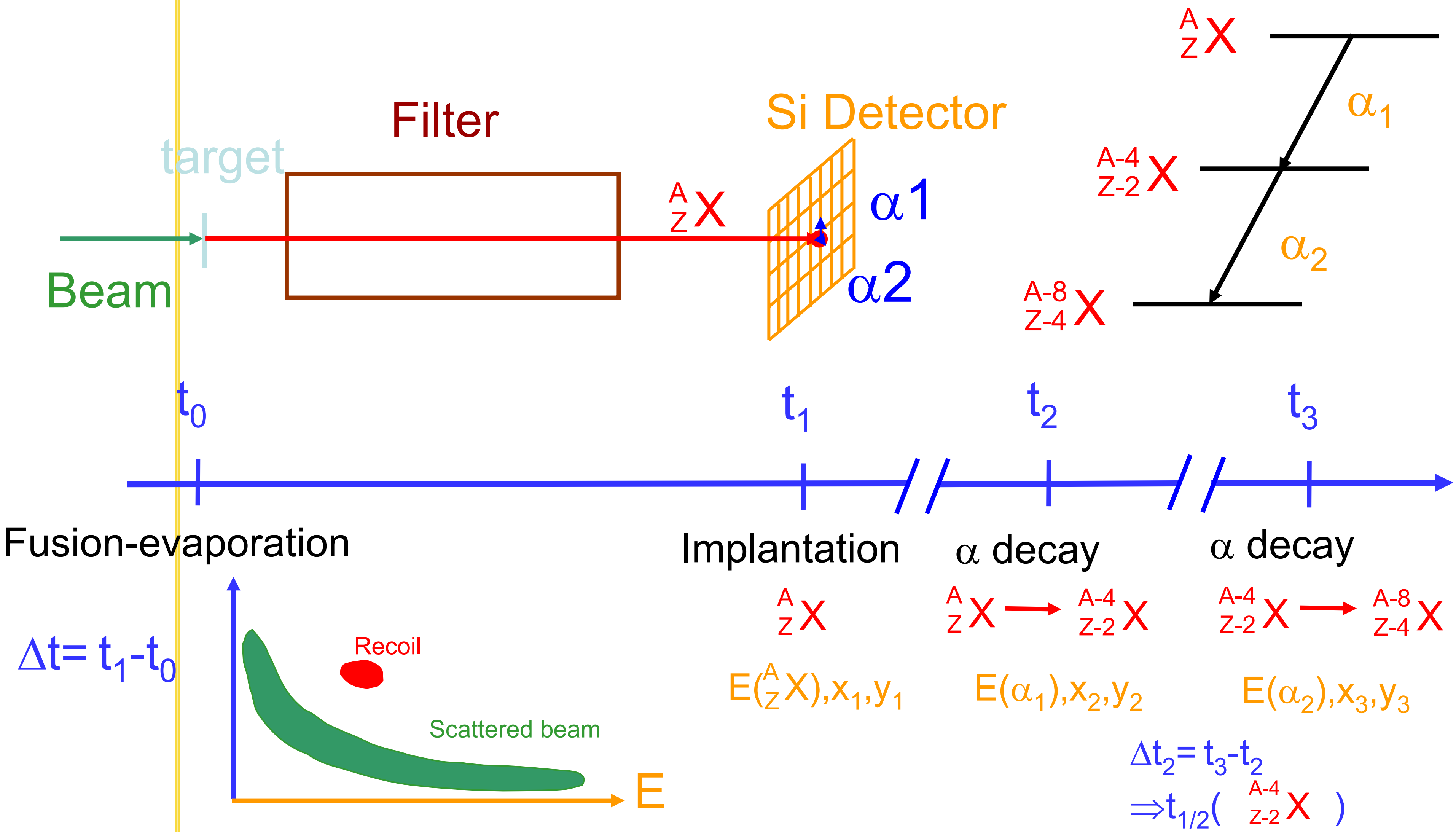
- Recoils energy + time of flight

Or

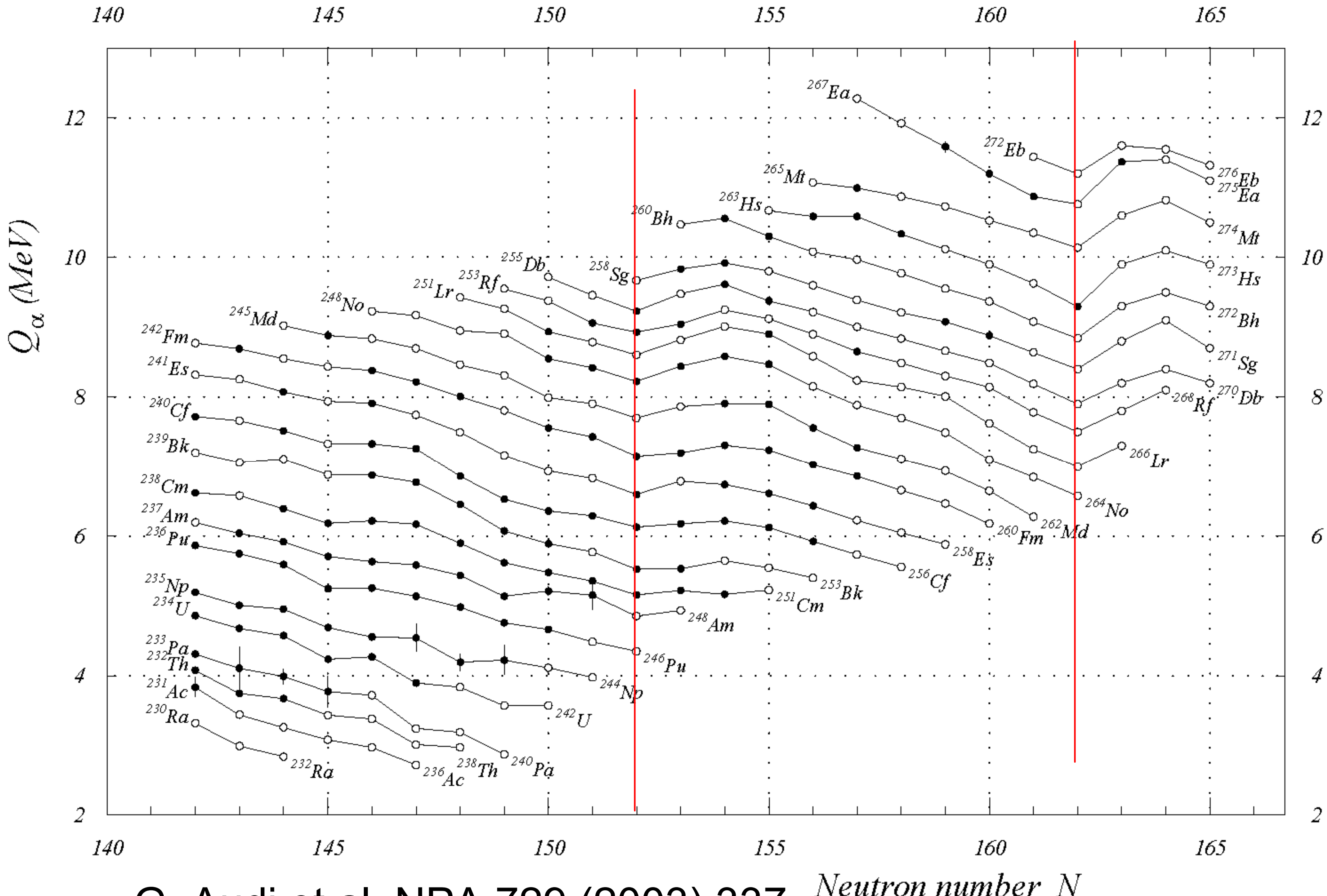
- Recoils energy + energy loss



Genetic correlations

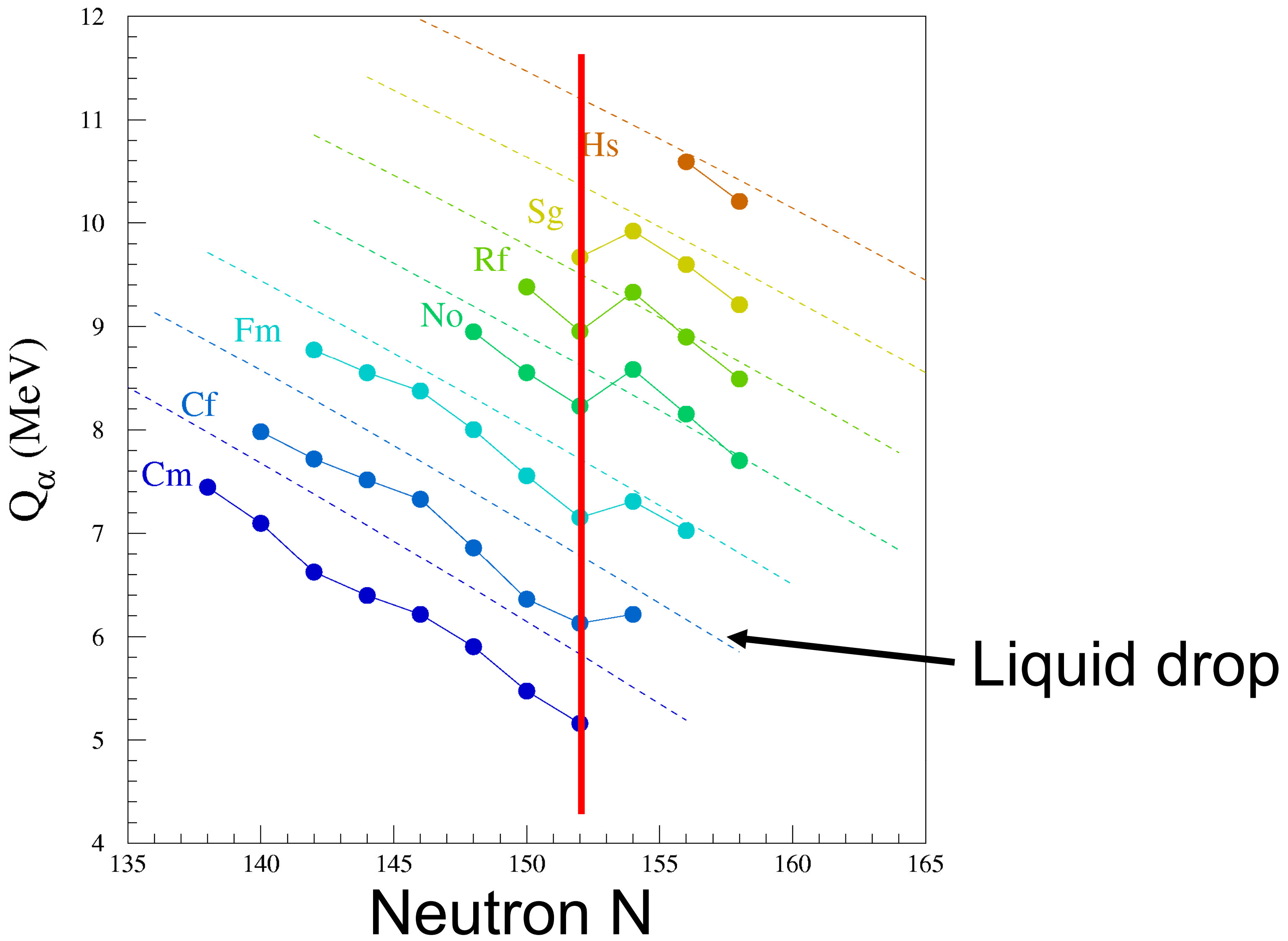


Alpha energy and shell gap



G. Audi et al, NPA 729 (2003) 337

Alpha energy and shell gap



Alpha decay : Selection rules

- $|I_i - I_f| < l_\alpha < I_i + I_f$

- $\pi_{mother} = \pi_{dauhter}\ \pi_\alpha$
- $\pi_{alpha} = (-1)^{l\alpha}$ l_α : orbital angular momentum

- Some transition from or to I=0 are forbidden
  - e.g. $0^+ \rightarrow 3^+$ forbidden but $0^+ \rightarrow 3^-$ allowed

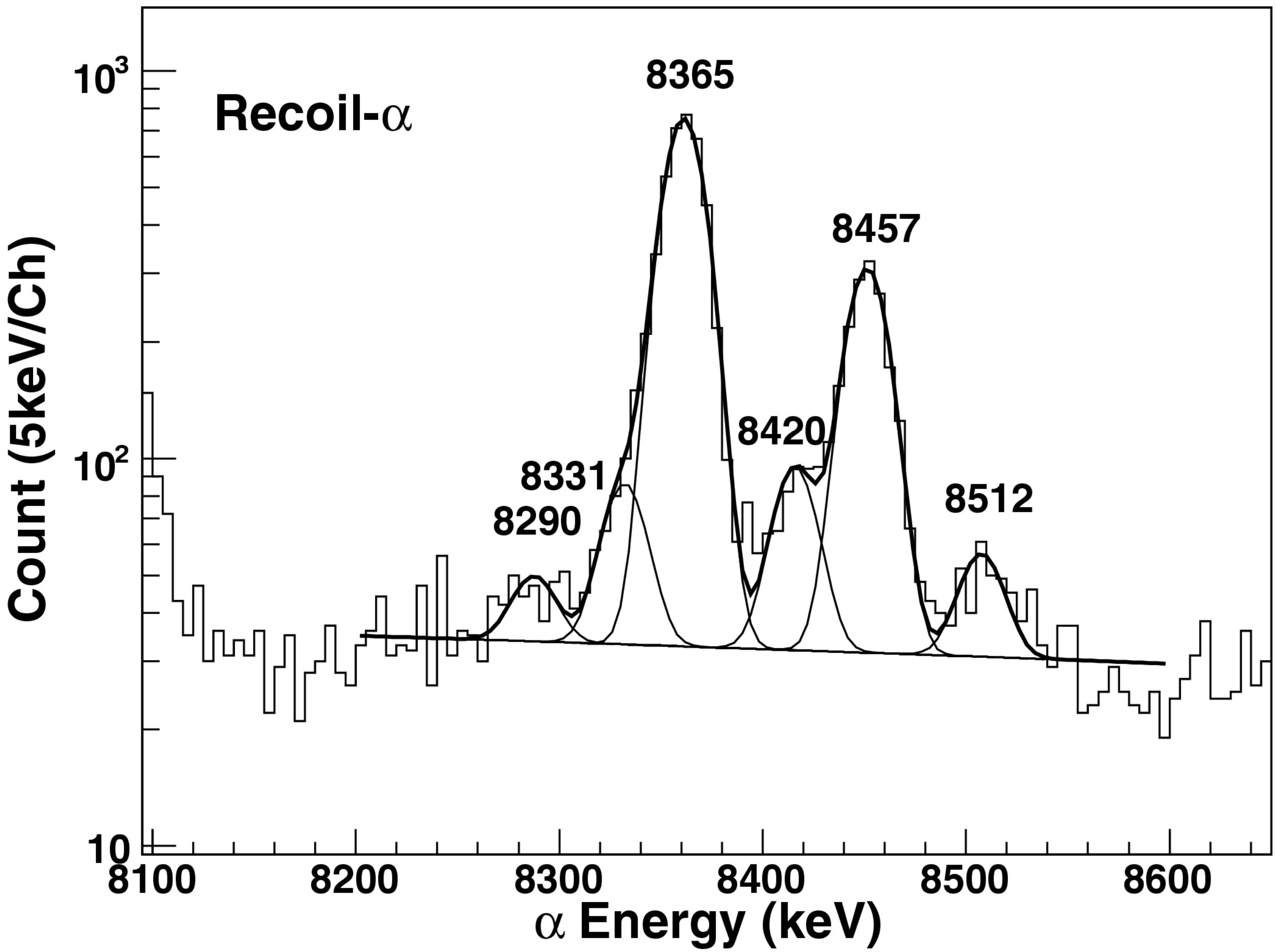
- In odd nuclei, all transitions are allowed !
  (but some l_α forbidden)

- Alpha decay is good tool for the study of single particle states, mostly odd nuclei

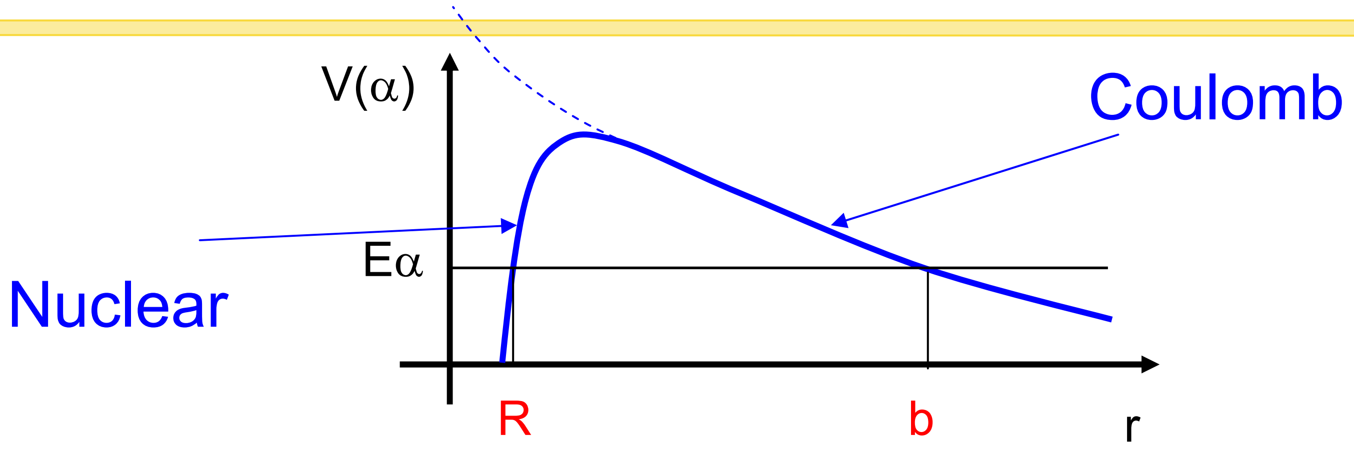
- Alpha de decay is however selective and prefers similar initial and final wave functions

alpha decay $^{255}Lr \rightarrow ^{251}Md$

What to do with this spectrum ?



Basic alpha decay formalism



$$P \approx Exp\left(-\frac{2}{\hbar}\int_R^b \sqrt{2M'_\alpha\left(V(r)+\frac{2Ze^2}{4\pi\varepsilon_0 r}+\frac{\hbar^2}{2mr^2}l(l+1)-Q_\alpha\right)}dr\right) \equiv Exp(-K)$$

$$T_{1/2} = f \ln 2 \text{ Exp K}$$

$T_{1/2}\uparrow$ with $l\uparrow$, $E\downarrow$, $Z\downarrow$

+ deformation effect : increases R then $T_{1/2}\downarrow$

Example ^{254}No

R ~ 9.2 fm
b ~ 35 fm
Vc ~ 30 MeV

f ~ 2.4 10^{21} s^{-1}

Assume excited state at 200 keV, l=0
I(exc)/I(g.s.) = 0.1

Assume same transition energy but different l
I(l=2)/I(l=0) ~ 0.7
I(l=4)/I(l=0) ~ 0.2
→ Dependence on angular momentum is small

Hindrance factor

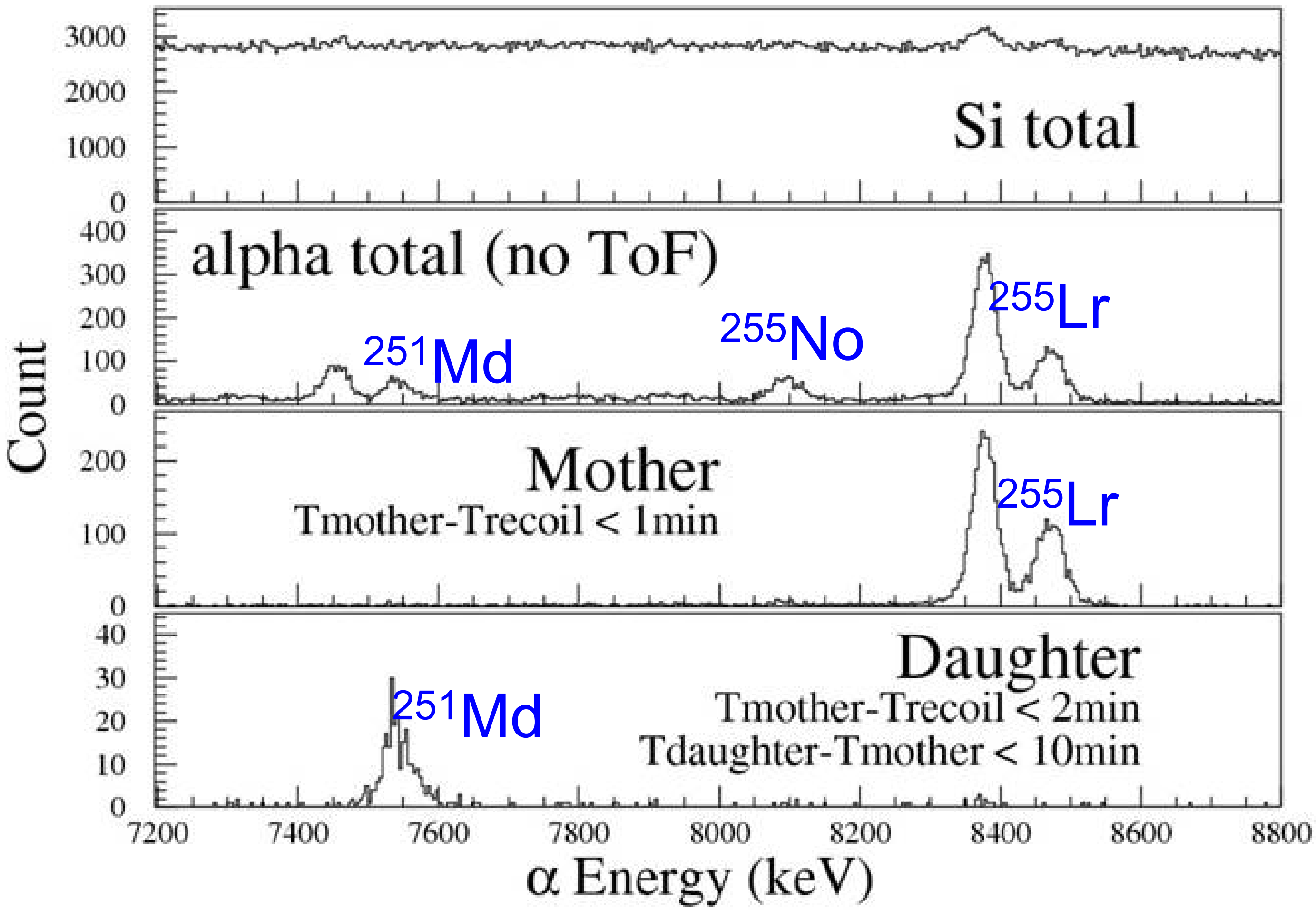
- Hindrance factor tells us whether initial and final states are similar or not
- It is somewhat similar to the *ft* value used in β decay or spectroscopic factor
- Recipe to calculate the hindrance factor :
  - Measure the alpha energy, lifetime and branching ratio
  - Deduce the experimental partial lifetime (Texp)
  - Calculate Tref : the lifetime for the same energy, but assuming a g.s. to g.s. transition in an even-even nucleus (simple "penetration" barrier problem)
    - Outer barrier b is straightforward (coulomb)
    - One can use the inner barrier R of a neighbouring even-even nucleus
  - Calculate Texp / Tref
  - You have the Hindrance Factor !!!
- What have we done :
  - We have removed the barrier penetration contribution (i.e. energy, charge, radius, potential)
  - We have still : wave function overlap and angular momentum
  - Tells us whether initial and final states are similar or not

Empirical HF values

- HF = 1-4 : same initial and final single-particle state
- HF = 4-10 : similar initial and final states
- HF = 10-100 : different single particle states, same parity, same spin projection
- HF = 100-1000 : different single particle states, parity change, same spin projection
- HF > 1000 : different single particle states, parity change, spin flip

- One needs a lot of statistics to observe a transition with a change of parity or spin flip

Example : $^{255}Lr \rightarrow {}^{251}Md \rightarrow {}^{247}Es$

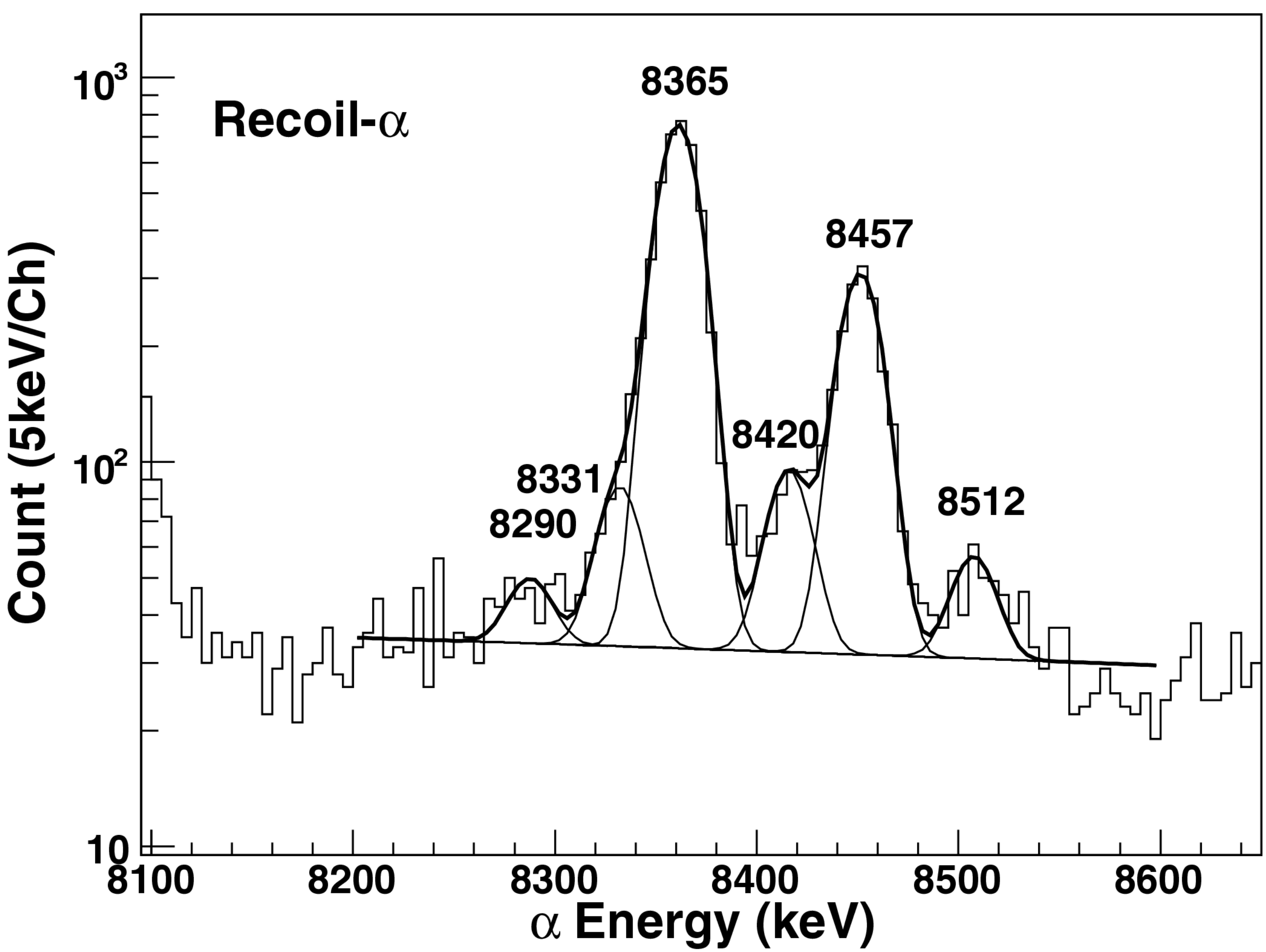


Example 1 : alpha decay $^{251}Md \rightarrow ^{247}Es$

- $E\alpha$= 7550 keV
- $T_{1/2}$ = 4.27 min
- Branching ratio : 10 %
- HF = 1.4

- Conclusion : initial and final single particle states are the same

Example 2 : alpha decay $^{255}Lr \rightarrow {}^{251}Md$



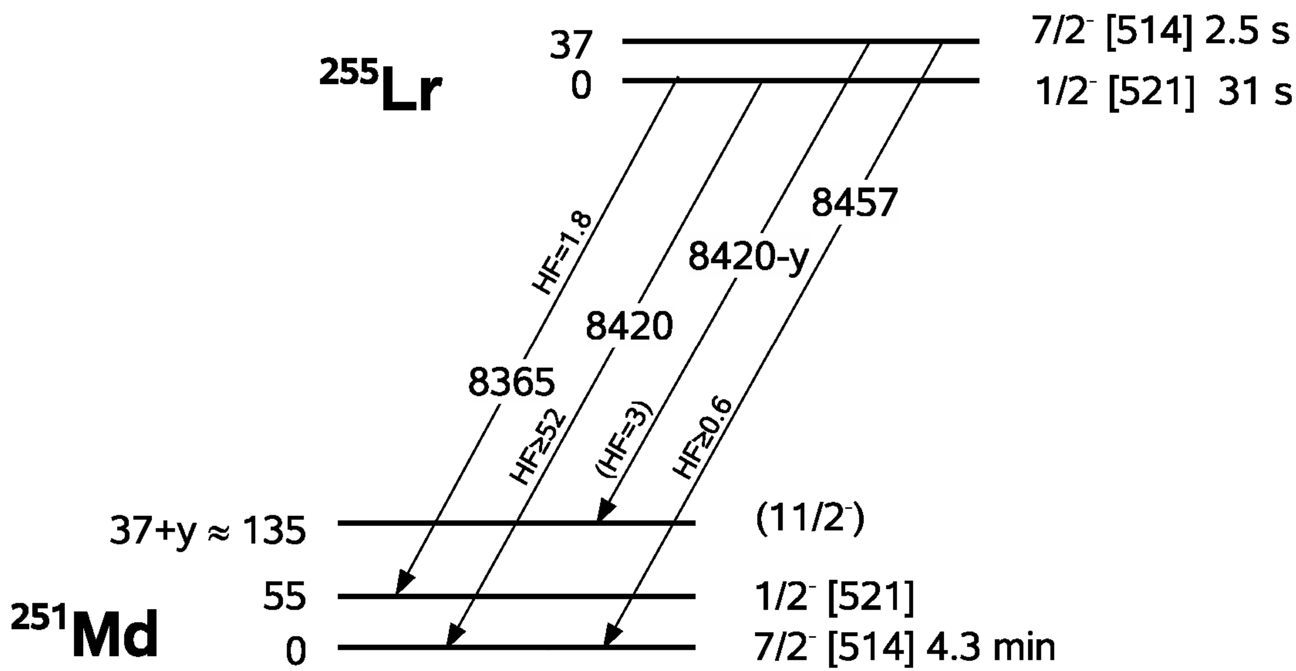
Example 2 : alpha decay $^{255}Lr \rightarrow ^{251}Md$

Isotope	Energy (keV)	Intensity (%)	Half-life	Q_α (keV)	H.F.	α branching ratio (%)
^{251}Md	7550(1)	87 ± 3	4.27 ± 0.26 min	7672(1)	1.4	10(1)
	7590(5)[a]	13 ± 3	4.3 ± 0.6 min	–	14	
	7550(1) sum	100	4.27 ± 0.26 min	7672(1)	1.3	
^{255}Lr	8290(5)[b]	1.2 ± 0.4	~ 35 s	–	–	85 (from [22])
	8365(2)	67.1 ± 1.5	31.1 ± 1.3 s	8498(2)	1.8	
	8420(10)	$\leq 3.6 \pm 0.5$	30 ± 4 s	8554(10)	≥ 52	
	8420(10)[a]	2.1 ± 0.5	2.8 ± 0.6 s	–	3[c]	
	8457(2)	26.0 ± 0.8	2.53 ± 0.13 s	8592(2)	≥ 0.6[d]	

[a] Interpreted as a line resulting from summing.
[b] Tentative assignment.
[c] Assuming an α energy of 8322 keV without summing.
[d] Possible low-energy tail due to summing.

A. Chatillon et *al*., EPJA 30 (2006) 397

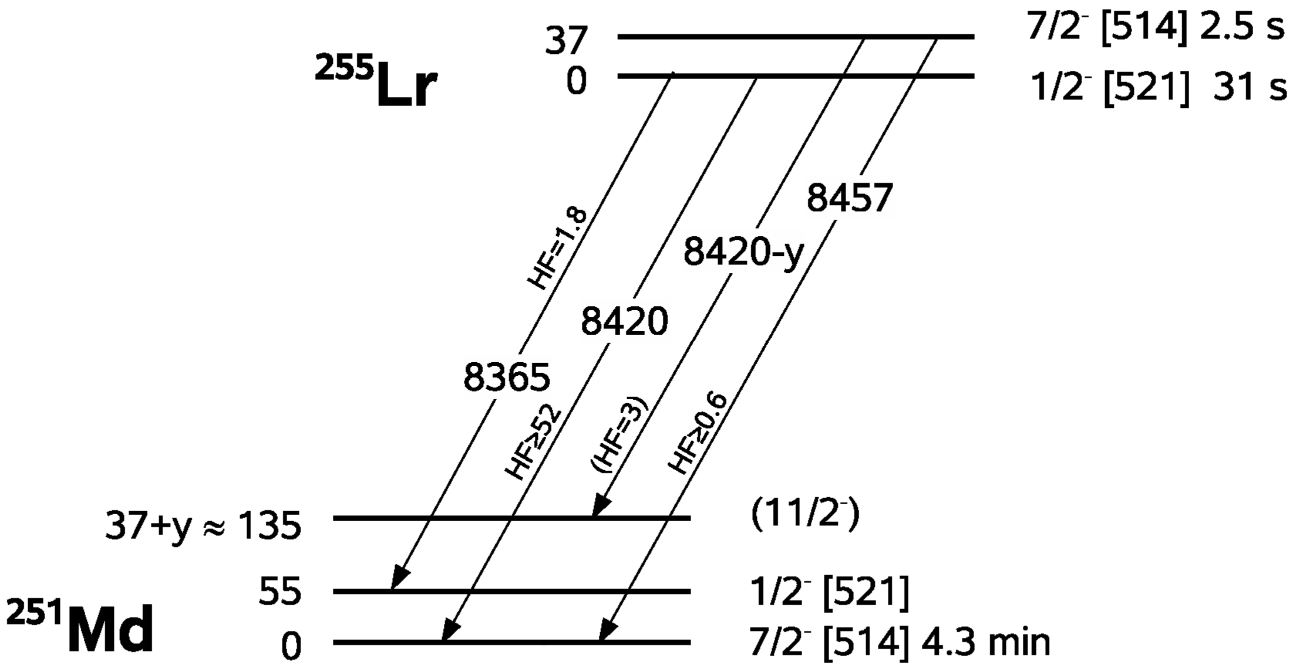
- Initial state $t_{1/2}$ ~ 30 s
  - 8365 : HF = 1.8 same initial and final single particle
  - 8420 : HF > 52 change initial and final single particle, same π, Σ
- Initial state $t_{1/2}$ ~ 2.5 s
  - 8457 : HF = 0.6 same initial and final single particle
  - 8420 : HF = 3 similar initial and final → collective excitation



Why don't we feed 7/2$^+$ [633] ?

- from 1/2$^-$ [521] : change π, change $\Sigma \rightarrow$ HF > 1000
- Assume E(7/2$^+$, ^{251}Md) = 200 keV and HF = 1000

  $\rightarrow b_\alpha \sim 0.05$ %



Why don't we feed $7/2^{+}$ [633] ?

- from $7/2^{-}$ [514] : change π, different Σ → HF > 1000
- Assume E($7/2^{+}$, ^{251}Md) = 200 keV and HF = 1000

  → b_{α} ~ 0.005 %

A. Chatillon et *al*., EPJA 30 (2006) 397