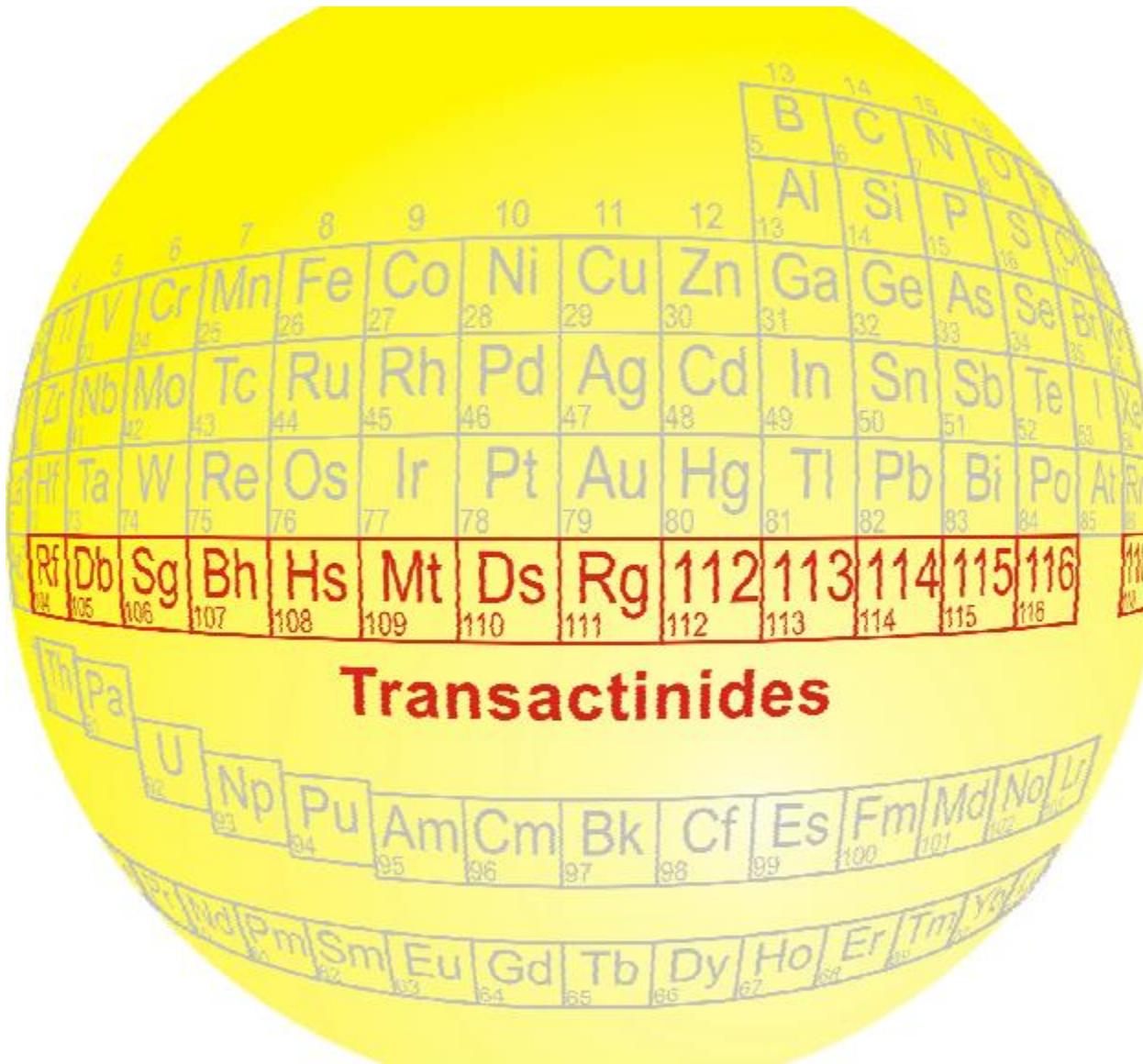


Super Heavy Elements



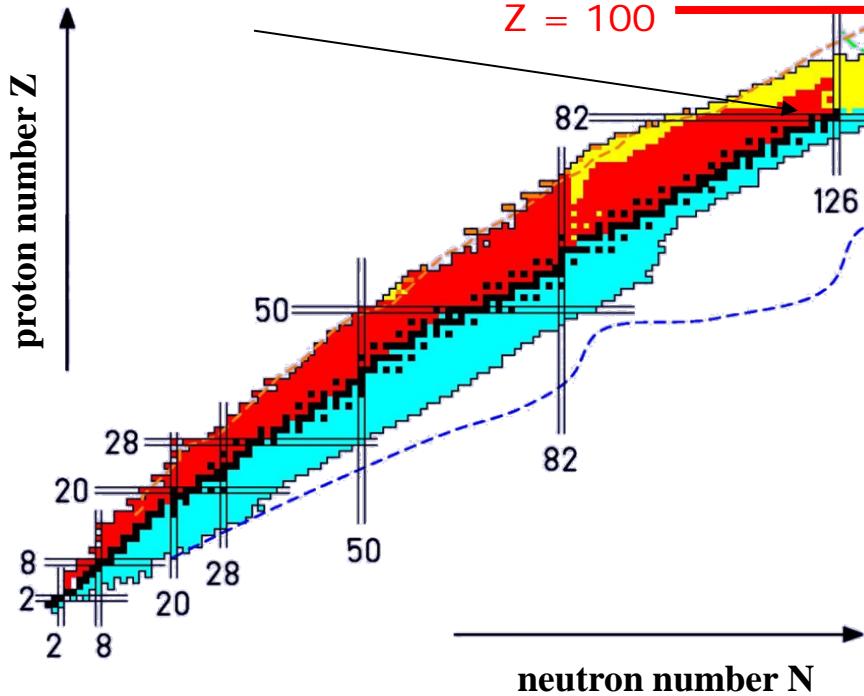
The Chart of Nuclides

- the “Playground” for Nuclear Physics

chart of nuclides:

- representation of isotopes in the Z-N plane
- isotope: atom (nucleus) of an element with different number of neutrons

Pb (lead) and Bi (bismuth)



neutron number N

?

island of stability

126
120
114

stabilisation via shell effects

SHE

U (uranium) and Th (thorium)

- black:** stable isotope
- red:** β^+ -unstable isotope
- blue:** β^- -unstable isotope
- yellow:** α -instable isotope
- green:** spontaneous fission

Periodic table of the elements

Dmitri Mendeleev (1869)

Original handwritten table by Dmitri Mendeleev, 1869.

Handwritten note: *Mon travail n'a pas encore été terminé.*

Handwritten note: *Il est nécessaire de faire des corrections et d'ajouter des éléments.*

Handwritten note: *D. Mendeleev.*

$T_i = 50$	$T_c = 90$	$? = 180$
$V = 51$	$Nb = 94$	$T_a = 182$
$C = 52$	$Ne = 96$	$W = 116$
$Ne = 55$	$Rb = 1094$	$Df = 1974$
$Fe = 56$	$Co = 1074$	$Te = 198$
$Ni = Os = 59$	$Pt = 106,6$	$Cd = 149$
$H = 1.$	$? = 8$	$I_y = 108, \quad H = 202,$
	$I_e = 94$	$Cd = 112, \quad ?$
$B = 11$	$\mathcal{Q} = 2744$	$Ue = 116, \quad Ba = 197,?$
$C = 12$	$S = 28$	$In = 118,$
$N = 14$	$P = 31$	$S = 122, \quad Bi = 210,?$
$O = 16$	$I = 32$	$Te = 128, \quad ?$
$F = 19$	$Cl = 35$	$D = 192,$
$Ar = 36$	$N = 39$	$Q = 183, \quad H = 204,$
	$Ca = 40$	$I_e = 137, \quad Pt = 217,$
	$Si = 42$	$? = 95, \quad Ca = 92,$
	$Se = 44$	$? = 56, \quad I_e = 94,$
	$Te = 60$	$? = 95,$
	$Os = 78$	$? = 188, \quad H = 118,$

Handwritten note: *Essai d'un système des éléments d'après leurs poids atomiques et fonctions chimiques par D. Mendeleff.*

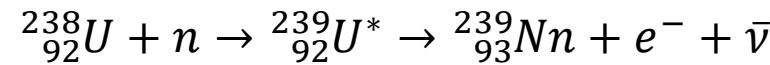
Handwritten note: *Présenté à l'Université de Berlin.*

Handwritten note: *18 $\frac{II}{I}$ 69.*

Handwritten note: *Andreas nödl, à l'Université de Berlin rapporté par -*

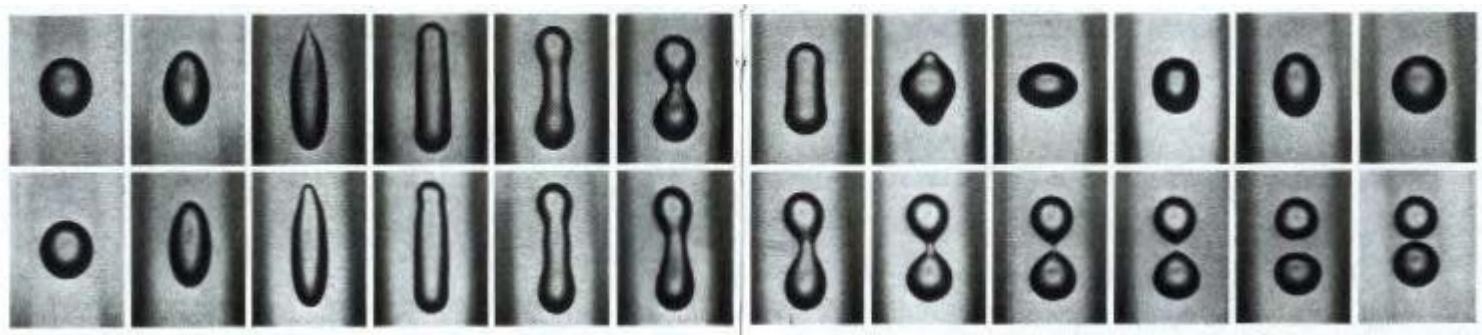
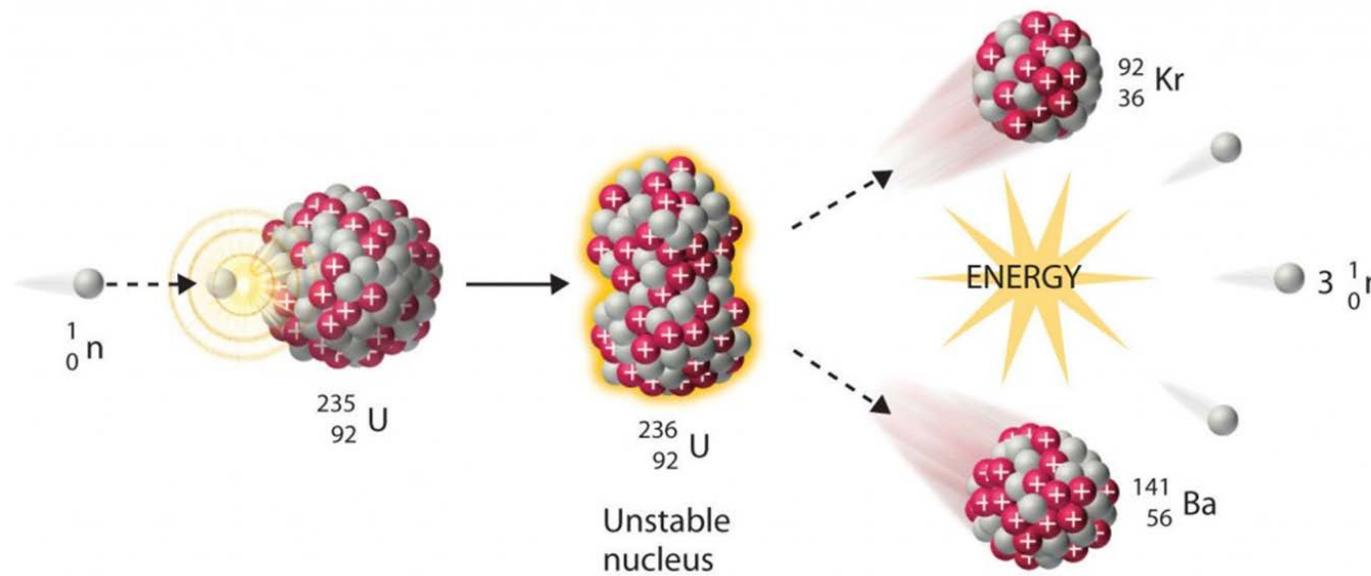


Search for transurane (1934)



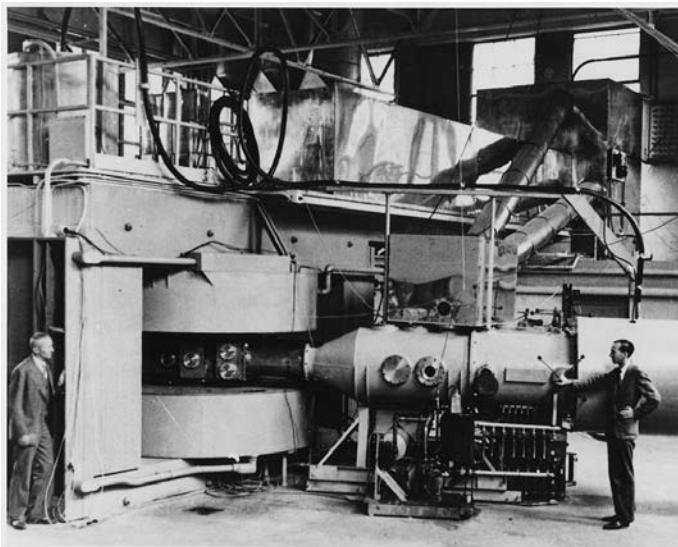
Otto Hahn and Lise Meitner

Discovery of nuclear fission (1938)



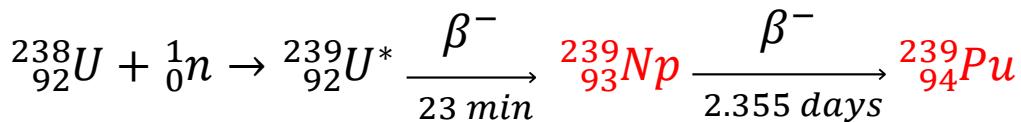
Liquid drop model: G. Gamow; Proc. Roy. Soc. London A123, 373 (1929)
N. Bohr & J.A. Wheeler; Phys. Rev. 56, 426 (1939)

Transuranium elements (1940)

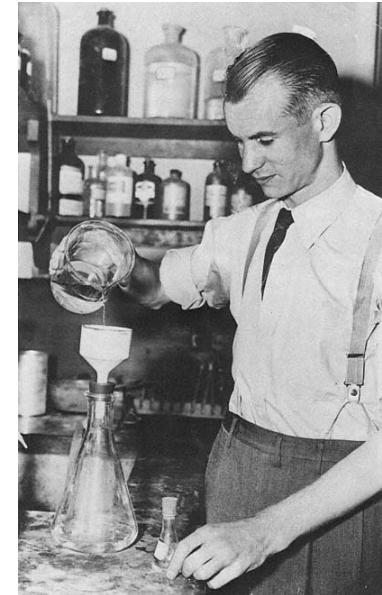


60-inch cyclotron (Berkeley, 1939)

H_2^+ , ${}^2\text{H}$ & ${}^4\text{He}$ beams ($Q/A = 1/2$)



Neptunium sphere
with Ni clad in U shells

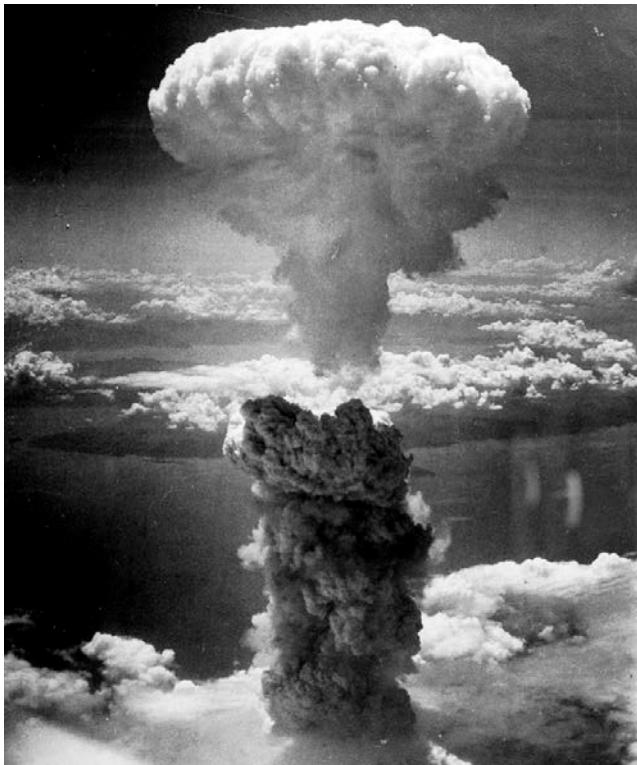
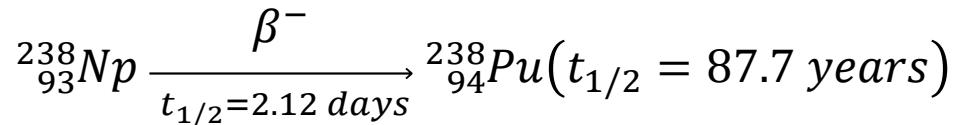
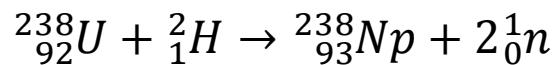


Edwin M. McMillan



Philip H. Abelson

Making new elements by simple reactions



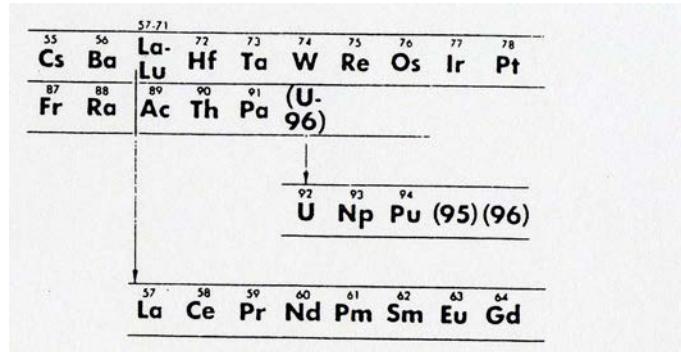
Joseph W. Kennedy 1940



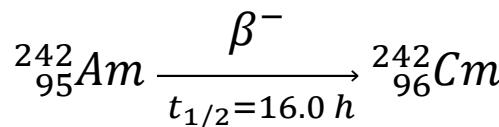
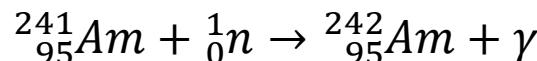
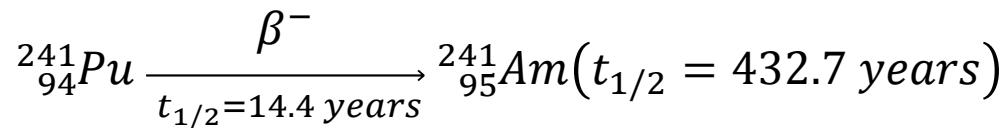
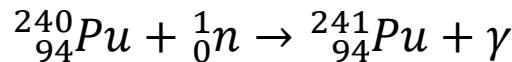
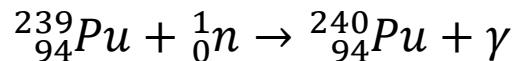
Arthur C. Wahl and
Glenn T. Seaborg (1966)

Making new elements by simple reactions – the role of chemistry

- ❖ The discovery of elements 95 (Am) and 96 (Cm)



Glenn T. Seaborg



Making new elements with nuclear weapons

- ❖ The discovery of elements 99 (Md) and 100 (Fm)

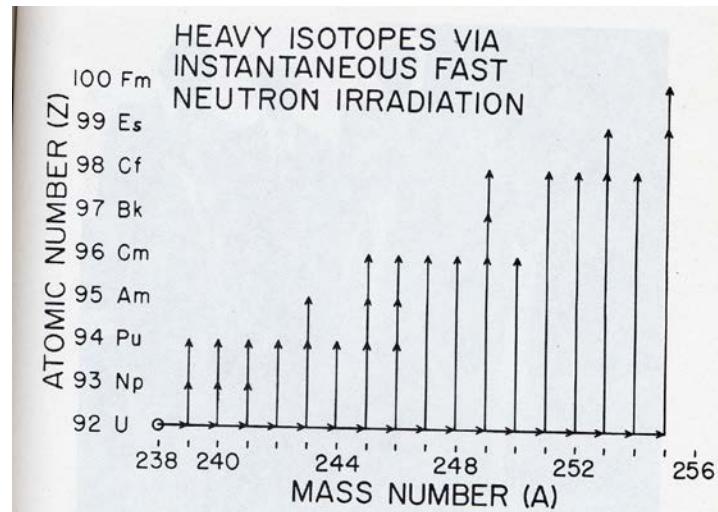


TABLE 6.1 Neutron Addition Paths to Transuranium Synthesis (Cra 74)

Neutron Addition Process	Neutron Flux ($n/cm^2 s^{-1}$)	Reaction Time	Neutron Exposure (n/cm^2)	Average Neutron Energy (keV)
High flux reactor	$\approx 5 \times 10^{15}$	0.5 years	$\approx 10^{23}$	2.5×10^{-5}
Stellar s process	$\approx 10^{16}$	$\approx 10^3$ years	$\approx 10^{26}$	≈ 10
Stellar r process	$\geq 10^{27}$	1–100 s	$> 10^{27}$	≈ 100
Nuclear explosion	$> 10^{31}$	$< 10^{-6}$ s	$\approx 10^{25}$	≈ 20

MIKE

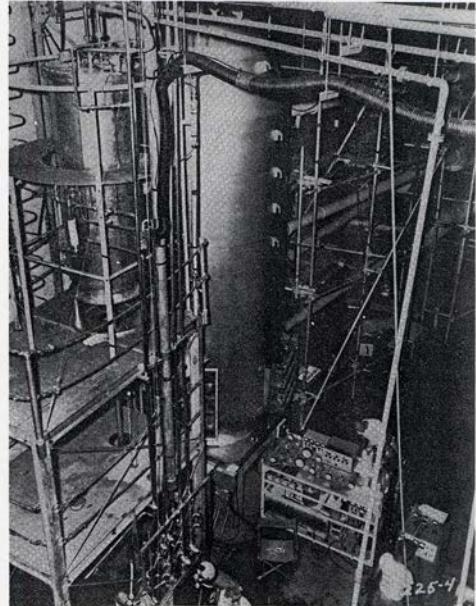
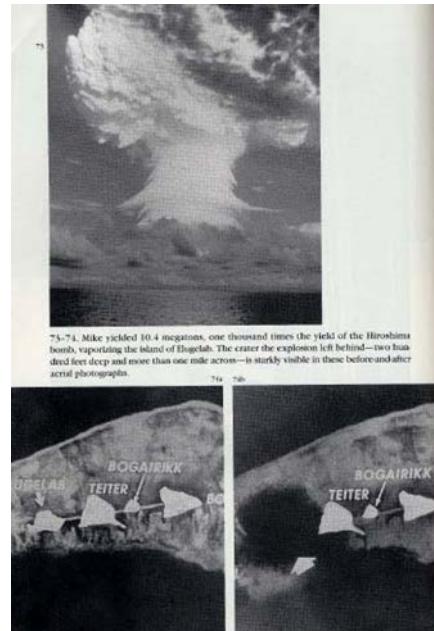


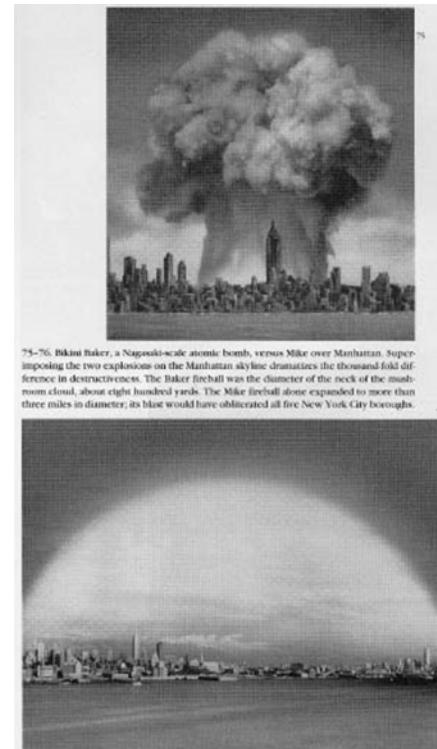
Figure 2.9 Closeup view of Mike device with its associated cryogenic equipment.

MIKE



75-74. Mike yielded 10.4 megatons, one thousand times the yield of the Hiroshima bomb, vaporizing the island of Elugelab. The crater the explosion left behind—two hundred feet deep and more than one mile across—is starkly visible in these before-and-after aerial photographs.

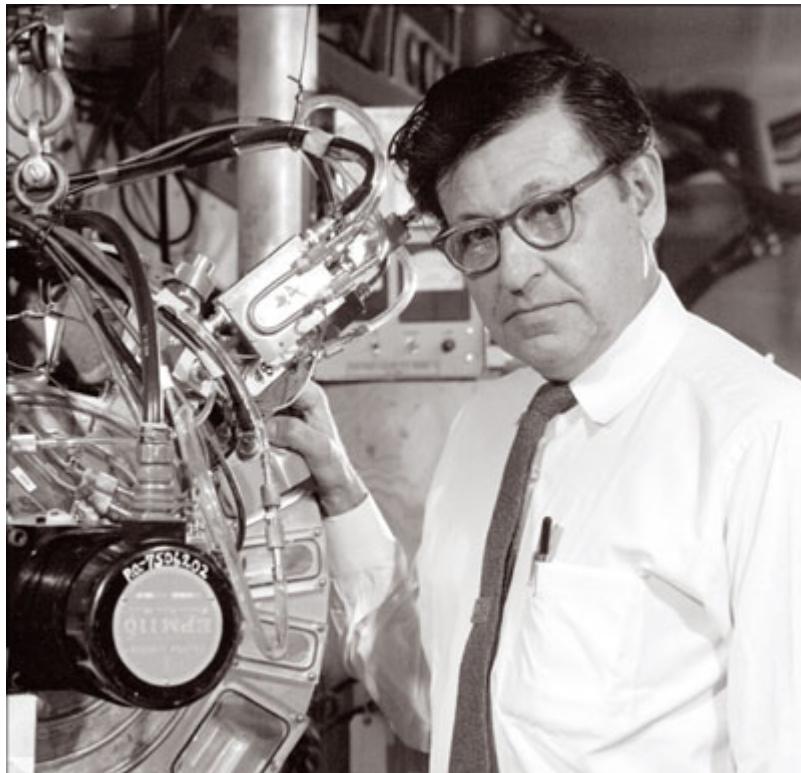
MIKE-2



75-76. Bikini Baker, a Nagasaki-scale atomic bomb, versus Mike over Manhattan. Superimposing the two explosions on the Manhattan skyline dramatizes the thousand-fold difference in destructiveness. The Baker fireball was the diameter of the neck of the mushroom cloud, about eight hundred yards. The Mike fireball alone expanded to more than three miles in diameter; its blast would have obliterated all five New York City boroughs.

Samples of the bomb debris were collected on filter papers by aircraft flying through the mushroom cloud

Using heavy ion reactions to make new elements – the Berkeley area



Albert Ghiorso



Glenn Seaborg

Synthesis of elements 101 - 106

- ❖ Making elements one atom at a time
- ❖ $^{254}\text{Es} + ^4\text{He} \rightarrow ^{256}\text{Md} + \text{n}$

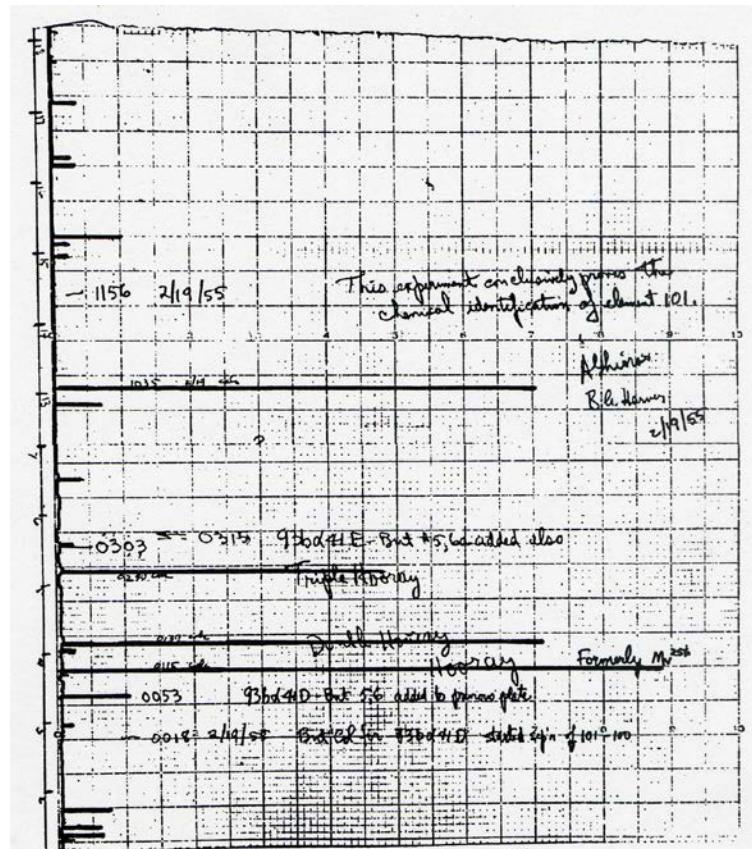
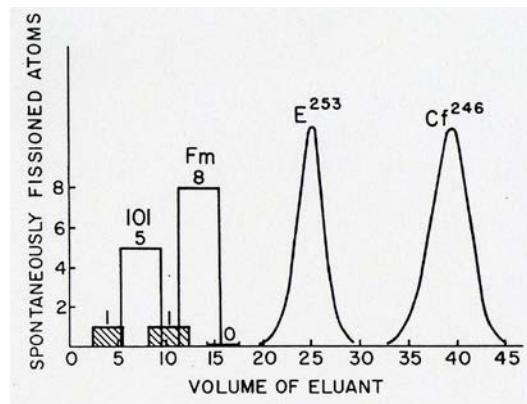
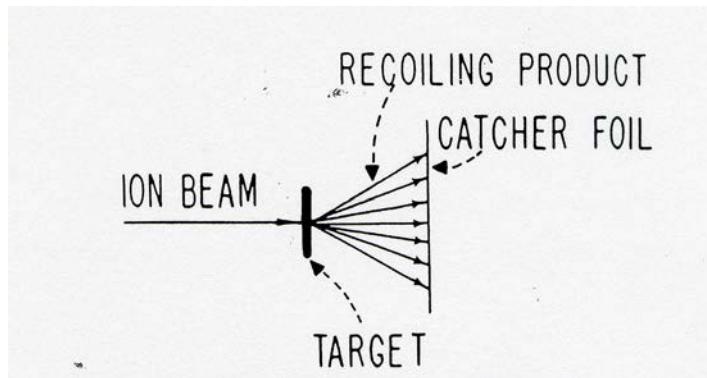


Figure 2.20 The ionization recording chart showing the first four events of the disintegration of mendelevium. The ordinate is the event time while the abscissa is the intensity of the ionization. The four pulses occurred at 1:15 a.m., 1:37 a.m., 2:40 a.m., and 10:35 a.m. on February 19, 1955. At 11:56 a.m., Ghiorso and Harvey made a note directly in the chart: "This experiment conclusively proves the chemical identification of element 101."

Hot fusion (1961-1974)

successful up to element 106 (Seaborgium)

- Coulomb barrier V_C between projectile and target nucleus has to be exceeded

$$V_C = \frac{Z_p \cdot Z_t \cdot e^2}{R_{int}} = 126.2 \text{ MeV} \quad (^{26}\text{Mg} + ^{248}\text{Cm})$$

- reaction: $a + A \rightarrow C^* \rightarrow B + b$

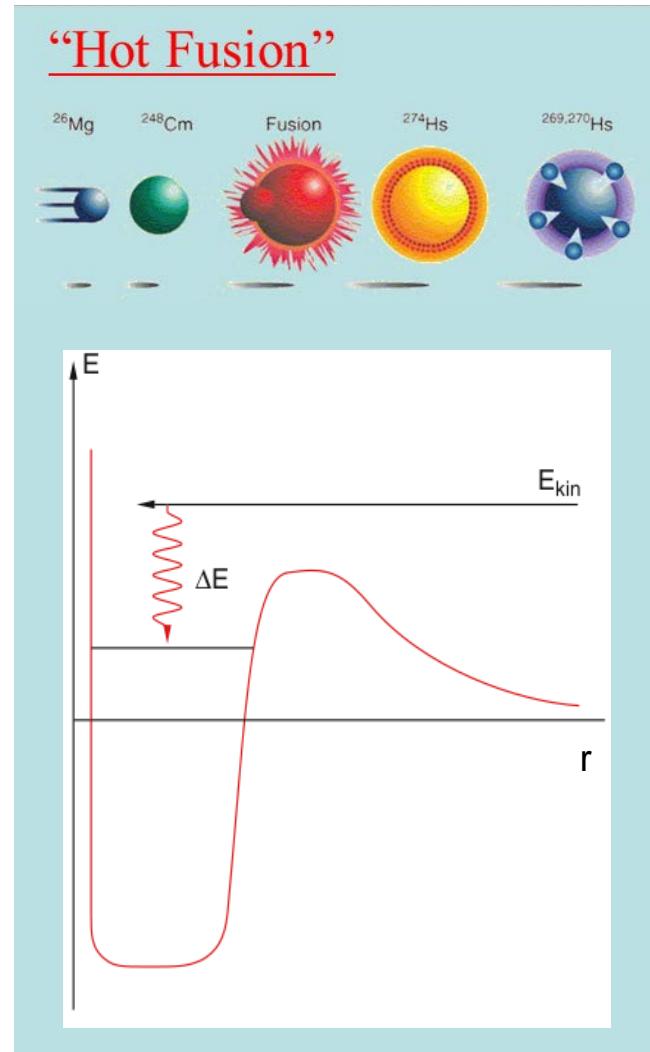
$$\Delta m = m_a + m_A - m_{CN}$$

$$\begin{aligned} \Delta m &= (25.983 + 248.072 - 274.143) * 931.478 \text{ MeV/c}^2 \\ &= -82.153 \text{ MeV/c}^2 \end{aligned}$$

- excitation energy of compound nucleus

$$E^* = E_{kin} + \Delta m \cdot c^2 = 126.2 \text{ MeV} - 82.2 \text{ MeV} = \mathbf{44.0 \text{ MeV}}$$

- approximate 4 neutrons will be evaporated to avoid fission



<http://nuclear.lu.se/database/masses/>

The problem

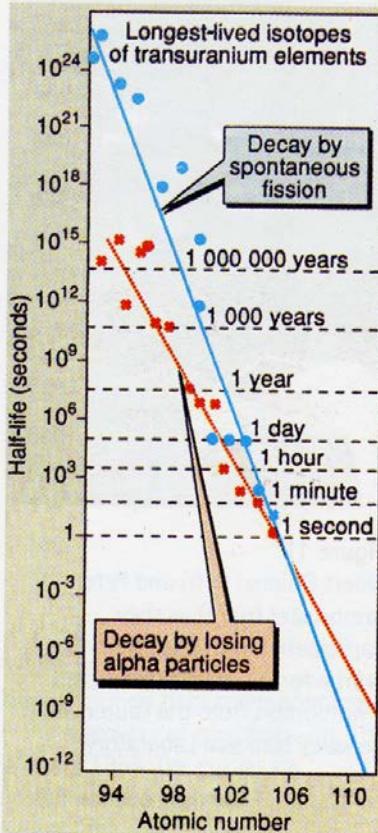


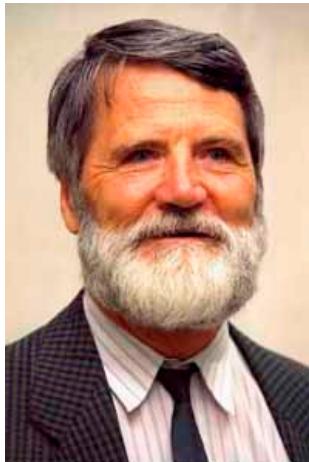
Figure 13

The half-lives of the longest-lived isotope of each element versus atomic number Z , circa 1970.

element	name	longest-lived isotope	half-life [s]
95	Americium	^{243}Am	$2.3 \cdot 10^{11}$
96	Curium	^{247}Cm	$5.0 \cdot 10^{14}$
97	Berkelium	^{247}Bk	$3.2 \cdot 10^{10}$
98	Californium	^{251}Cf	$2.8 \cdot 10^{10}$
99	Einsteinium	^{252}Es	$4.1 \cdot 10^7$
100	Fermium	^{257}Fm	$8.7 \cdot 10^6$
101	Mendelevium	^{258}Md	$4.4 \cdot 10^6$
102	Nobelium	^{259}No	$3.5 \cdot 10^3$
103	Lawrencium	^{266}Lr	$3.6 \cdot 10^4$
104	Rutherfordium	^{267}Rf	$4.7 \cdot 10^3$
105	Dubnium	^{268}Db	$1.1 \cdot 10^5$
106	Seaborgium	^{269}Sg	$1.8 \cdot 10^2$
107	Bohrium	^{270}Bh	$6.0 \cdot 10^1$
108	Hassium	^{277}Hs	$3.0 \cdot 10^1$
109	Meitnerium	^{278}Mt	$4.0 \cdot 10^0$
110	Darmstadtium	^{281}Ds	$1.4 \cdot 10^1$
111	Roentgenium	^{282}Rg	$1.2 \cdot 10^2$
112	Copernicium	^{285}Cn	$3.0 \cdot 10^1$
113	Nihonium	^{286}Nh	$8.0 \cdot 10^0$
114	Flerovium	^{289}Fl	$2.0 \cdot 10^0$
115	Moscovium	^{290}Mc	$8.0 \cdot 10^{-1}$
116	Livermorium	^{293}Lv	$6.0 \cdot 10^{-2}$
117	Tennessine	^{294}Ts	$5.0 \cdot 10^{-2}$
118	Organesson	^{294}Og	$7.0 \cdot 10^{-4}$

The solution – The Darmstadt Era

- ❖ “Cold fusion” reactions
- ❖ Bombarding Pb or Bi with heavy ions – the resulting species are borne “cold”
 - with low excitation energies – they survive better



Peter Armbruster



Gottfried Münzenberg



Sigurd Hofmann

Cold fusion (1981-1996)

- Coulomb barrier V_C between projectile and target nucleus has to be exceeded

$$V_C = \frac{Z_p \cdot Z_t \cdot e^2}{R_{int}} = 223.3 \text{ MeV} \quad (^{58}\text{Fe} + ^{208}\text{Pb})$$

- reaction: $a + A \rightarrow C^* \rightarrow B + b$

$$\Delta m = m_a + m_A - m_{CN}$$

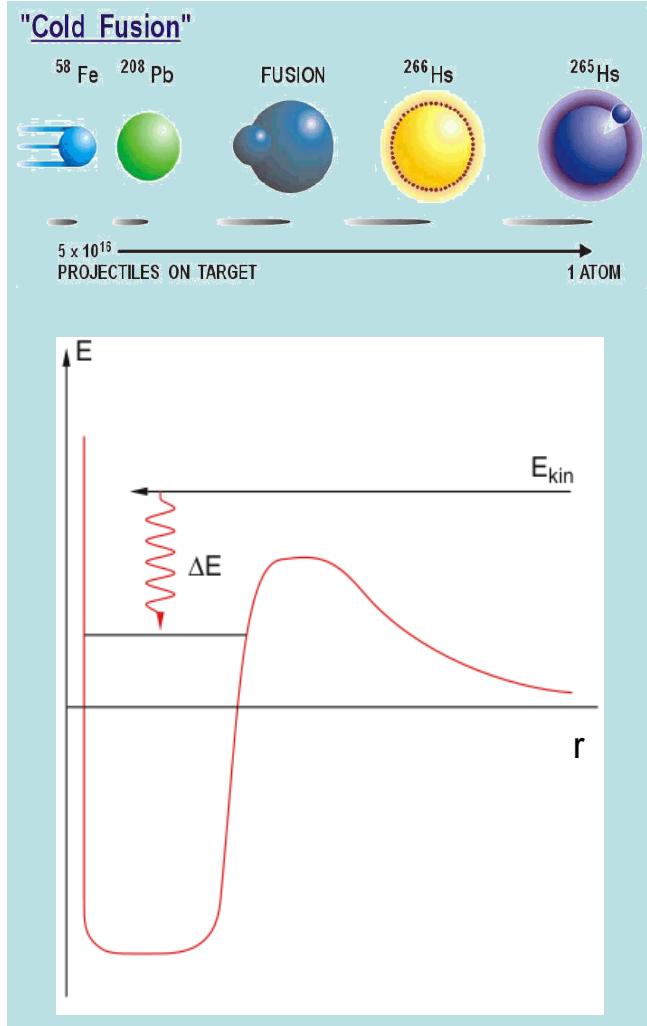
$$\begin{aligned} \Delta m &= (57.933 + 207.977 - 266.130) * 931.478 \text{ MeV/c}^2 \\ &= -205.045 \text{ MeV/c}^2 \end{aligned}$$

- excitation energy of compound nucleus

$$E^* = E_{kin} + \Delta m \cdot c^2 = 223.3 \text{ MeV} - 205.0 \text{ MeV} = \mathbf{18.2 \text{ MeV}}$$

- approximate 1-2 neutrons will be evaporated to avoid fission

reaction	Q_{gg} (MeV)	$V_C(R_{int})$ (MeV)	E^* (MeV)
$^{48}_{20}\text{Ca} + ^{208}_{82}\text{Pb} \rightarrow ^{256}_{102}\text{No}$	-153.796	175.05	21.25
$^{54}_{24}\text{Cr} + ^{209}_{83}\text{Bi} \rightarrow ^{263}_{107}\text{Bh}$	-189.911	210.00	20.09
$^{58}_{26}\text{Fe} + ^{208}_{82}\text{Pb} \rightarrow ^{266}_{108}\text{Hs}$	-205.045	223.31	18.27
$^{58}_{26}\text{Fe} + ^{209}_{83}\text{Bi} \rightarrow ^{267}_{109}\text{Mt}$	-208.526	225.86	17.33
$^{62}_{28}\text{Ni} + ^{208}_{82}\text{Pb} \rightarrow ^{270}_{110}\text{Ds}$	-223.225	238.84	15.62
$^{64}_{28}\text{Ni} + ^{209}_{83}\text{Bi} \rightarrow ^{273}_{111}\text{Rg}$	-228.52	240.78	12.26
$^{70}_{30}\text{Zn} + ^{208}_{82}\text{Pb} \rightarrow ^{278}_{112}\text{Cn}$	-244.38	252.67	8.29



<http://nuclear.lu.se/database/masses/>
G. Audi et al. Nucl. Phys. A729 (2003) 337

Excitation functions

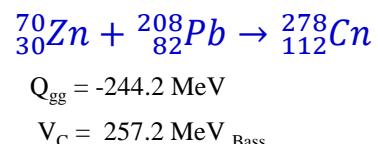
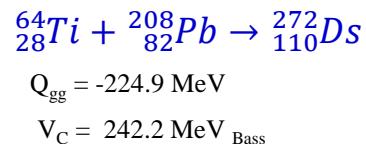
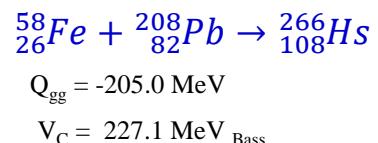
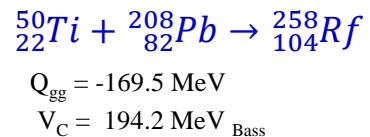
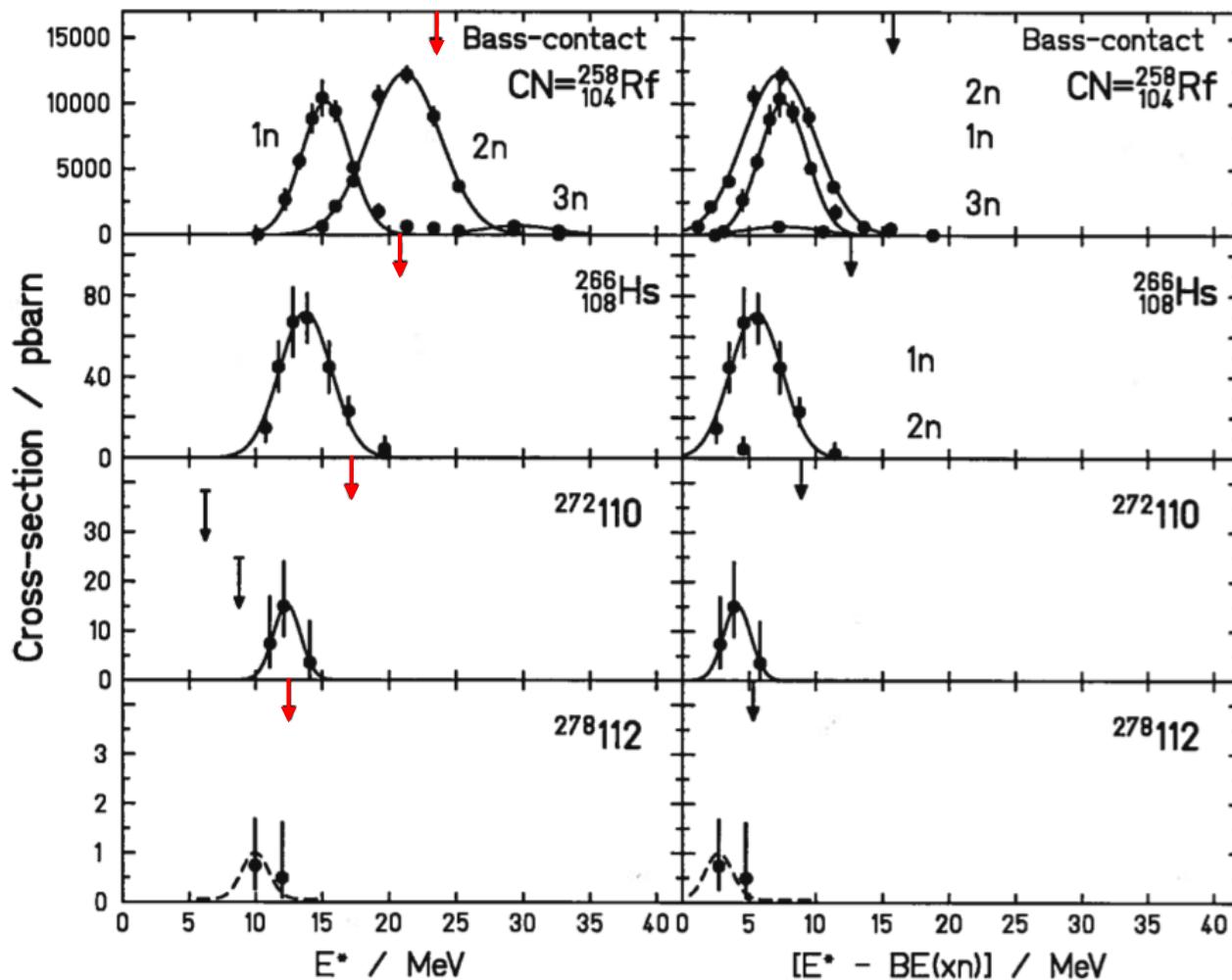
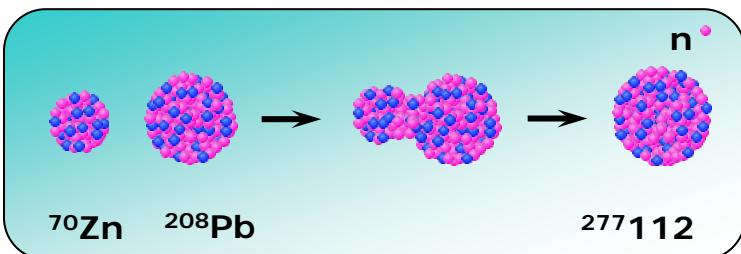


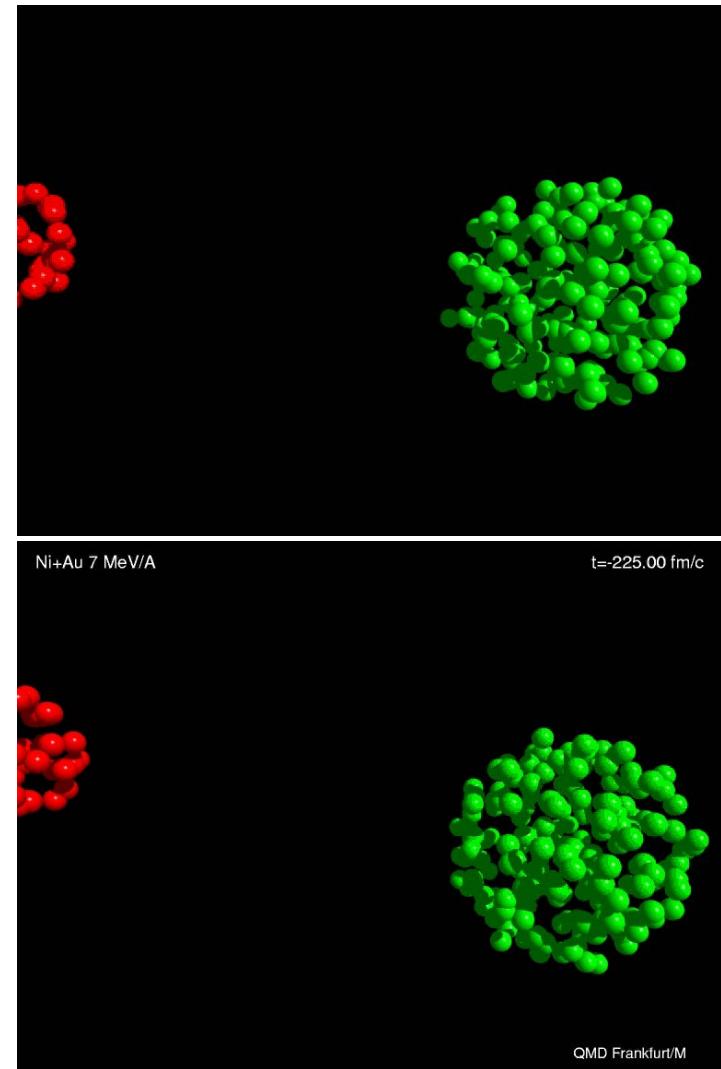
Fig. 1. Excitation functions for the production of elements 104, 108, 110, and 112 plotted versus the excitation energy of the compound system (left-hand side) and versus the excitation energy corrected by subtraction of the neutron binding energy [6] (right-hand side).

Synthesis of heavy elements

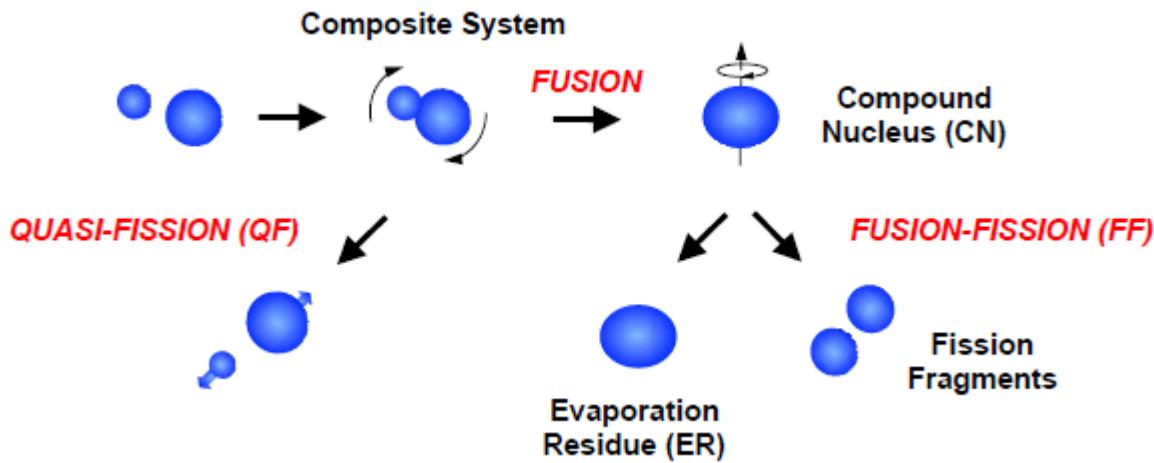


Fusion

$$-\frac{1}{10^{12}}$$



Fusion / fission competition

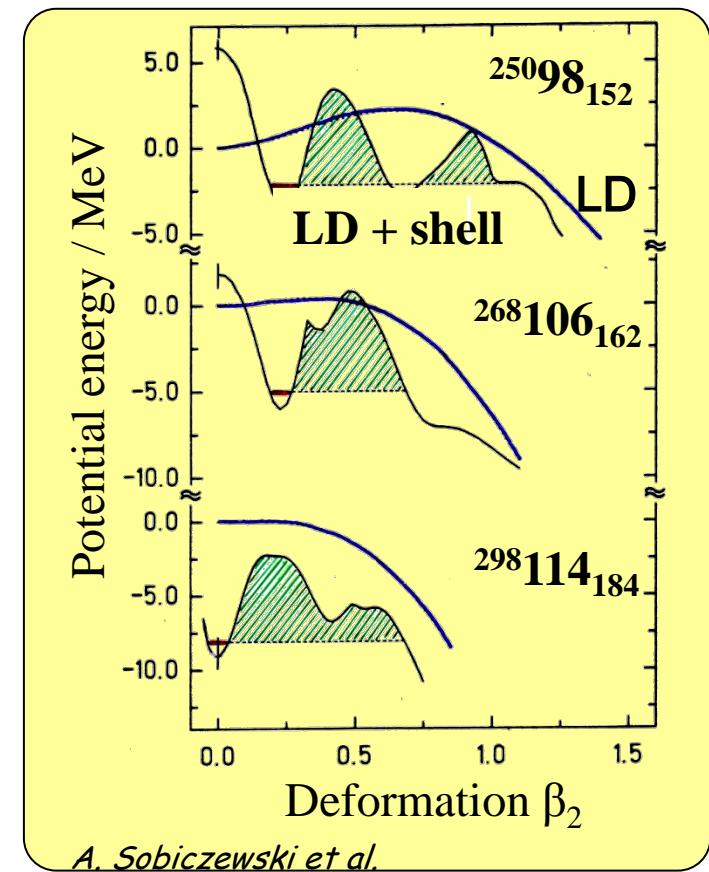


$$\sigma_{ER} = \sigma_{capture} \cdot P_{CN} \cdot P_{survival}$$

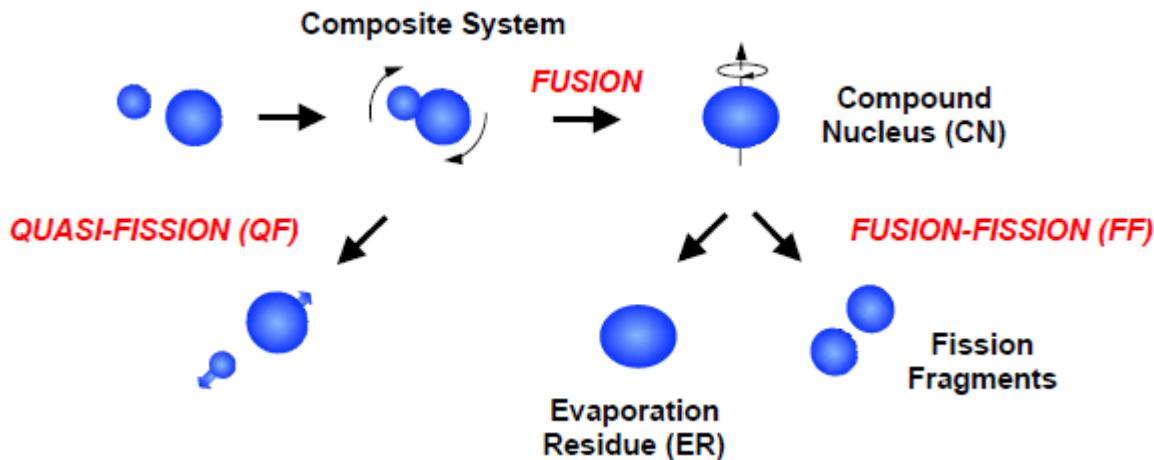
$$\sigma_{ER}^{xn}(E) = \frac{\pi}{k^2} \sum_{\ell=0}^{\infty} (2\ell + 1) \cdot \underbrace{P_{cont}(E, \ell)}_{\text{capture}} \cdot \underbrace{P_{CN}(E^*, \ell)}_{\text{formation}} \cdot \underbrace{P_{xn}(E^*, \ell)}_{\text{survival}}$$

competing: quasi-elastic quasi-fission fission

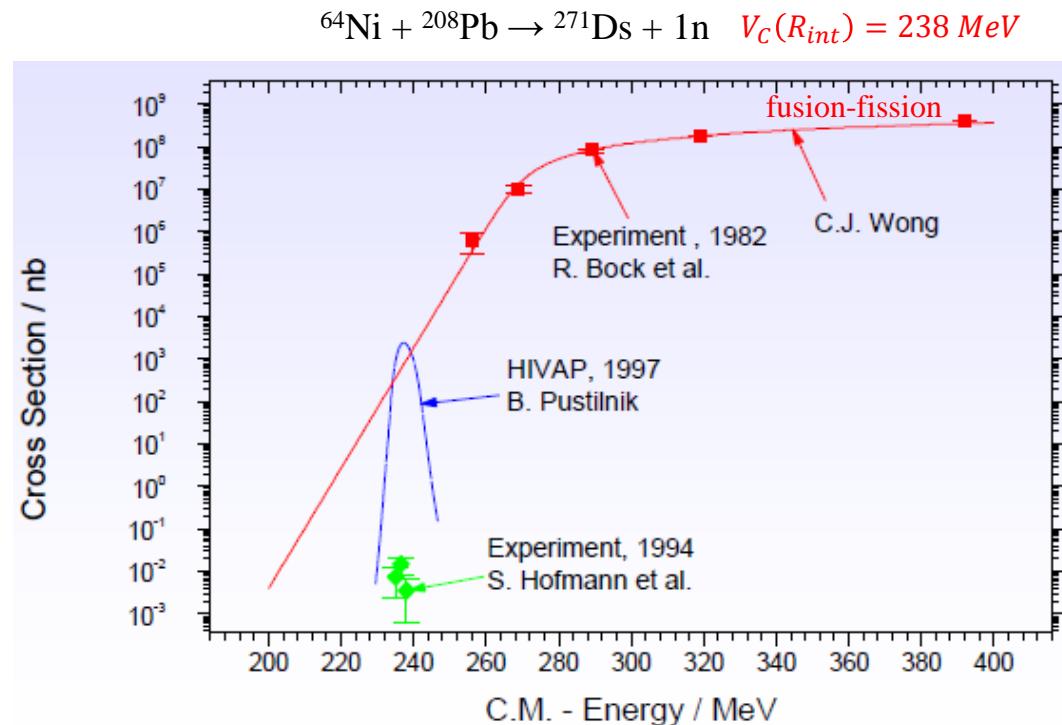
Superheavy system: $\sigma_{ER} \ll \sigma_{capture}$



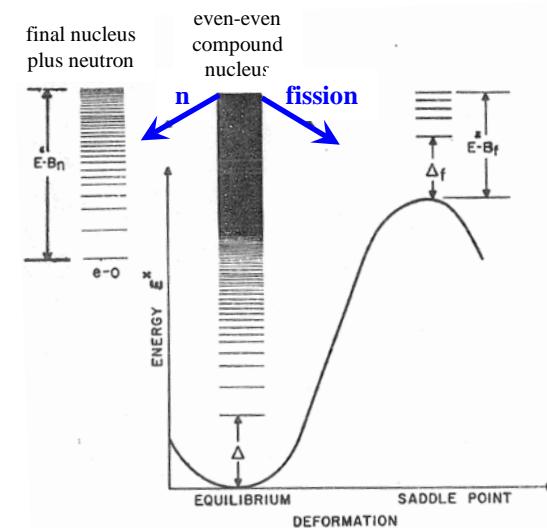
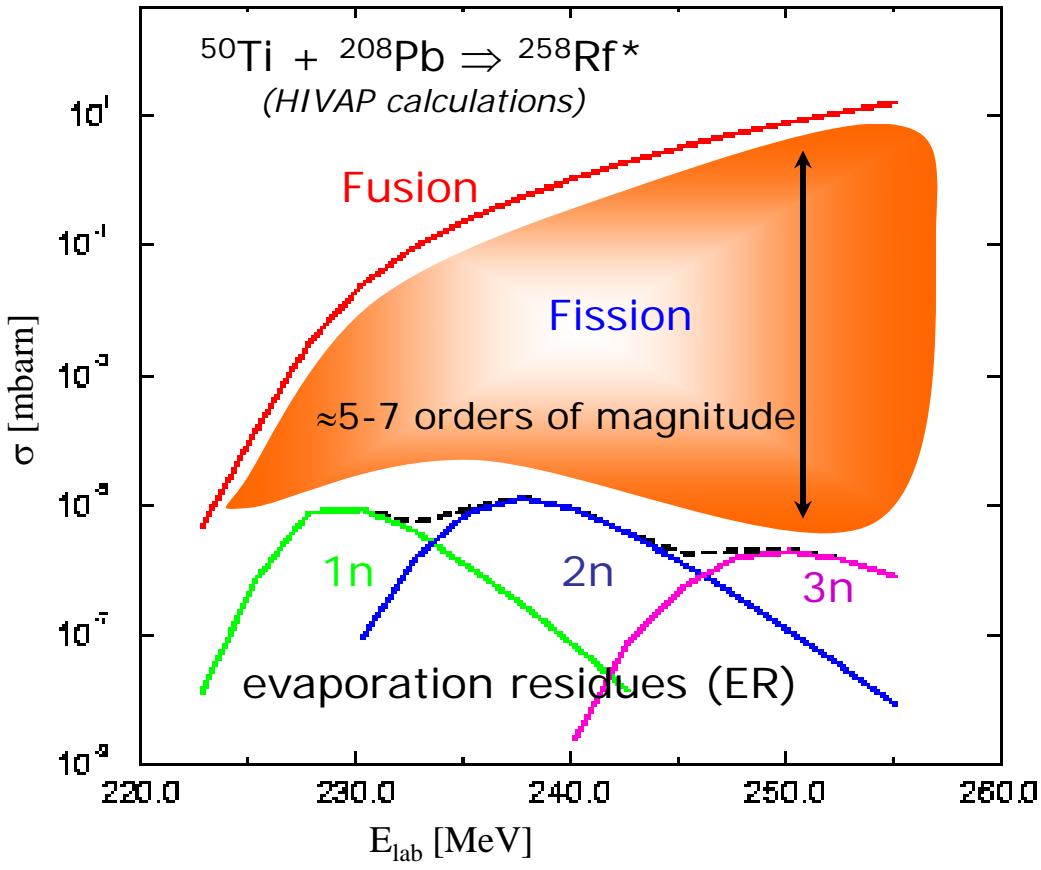
Fusion / fission competition



$$\sigma_{ER} = \sigma_{capture} \cdot P_{CN} \cdot P_{survival}$$



Fusion and evaporation



Both decay processes are determined by the level density, either from the residual nucleus or at the saddle point.

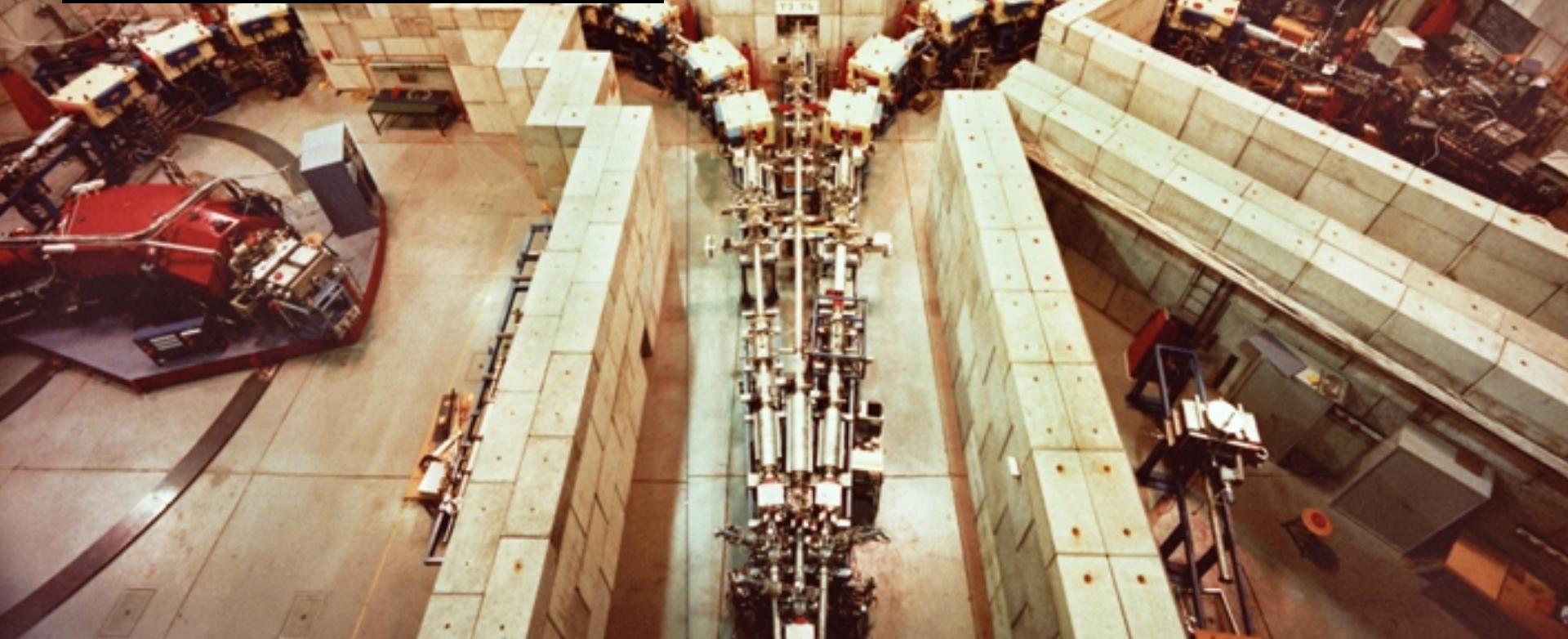
level density: $\rho(E^*) = \text{const} \cdot \exp(E^*/T)$

$$\frac{\Gamma_n}{\Gamma_f} = \frac{2 \cdot T \cdot A_{CN}^{2/3}}{K_0} \cdot \exp[(B_f - B_n)/T]$$

$$K_0 = \hbar^2 / 2 \cdot m \cdot r_0^2 \approx 11.4 \text{ MeV}$$

$$T = \sqrt{8 \cdot E^* / A_{CN}}$$

Separator for Heavy Ion Products (SHIP)



Separator for Heavy Ion Products (SHIP)

- Fusion products are slower than scattered or transfer particles

$$v_{CN} = [m_p/(m_p + m_t)] \cdot v_p$$

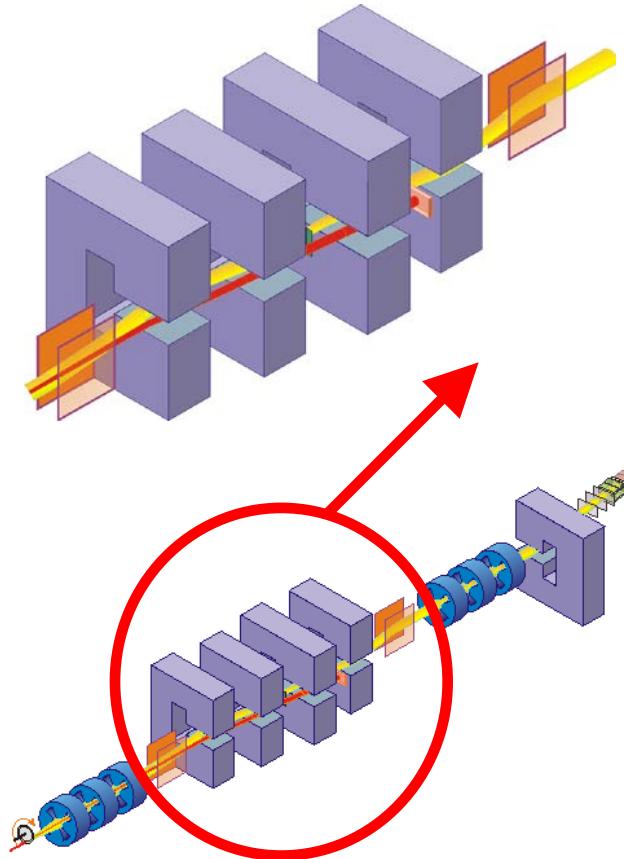
$$e \cdot q \cdot v_p \approx 10.3\% \rightarrow v_{CN} \approx 2.2\%$$

- E- and B-field are perpendicular to each other

$$B \cdot \rho = \frac{m \cdot v}{e \cdot q}$$

$$E \cdot \rho = \frac{m \cdot v^2}{e \cdot q}$$

$$F_{mag} = F_{el} \Rightarrow F_{tot} = 0$$



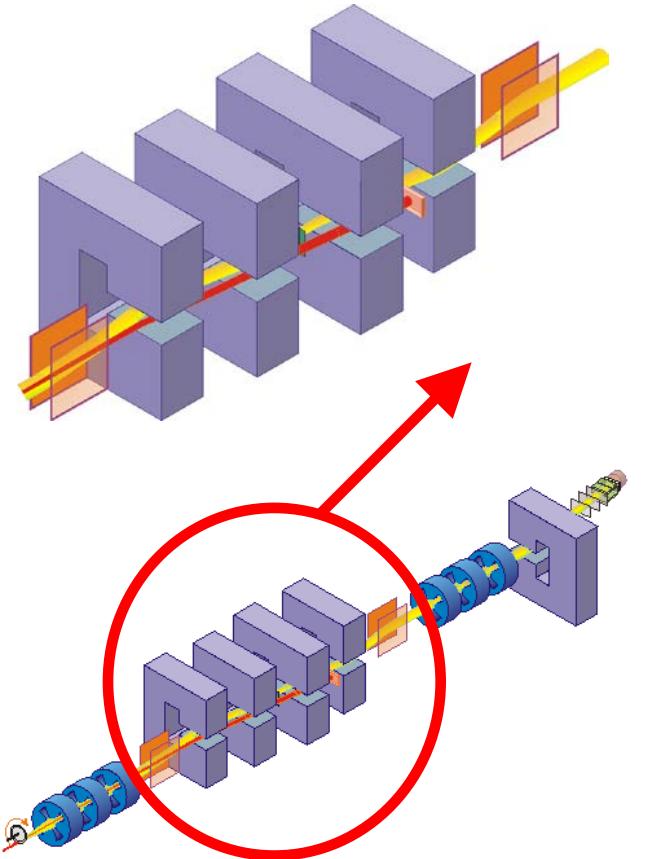
electric deflectors: ± 330 kV dipole magnets: 0.7 T max

Separator for Heavy Ion Products (SHIP)

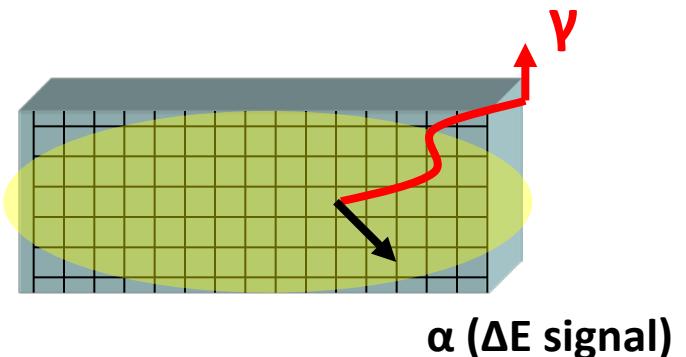
- The choice of E and B determines the transmitted velocity

$$v = \frac{E}{B}$$

- The rejected beam will be stopped on a cooled Cu plate



SHIP – stop detector



SHE will be measured in a pixel

- position sensitive Silicon detector determines the position an energy of SHE and α , β , ...

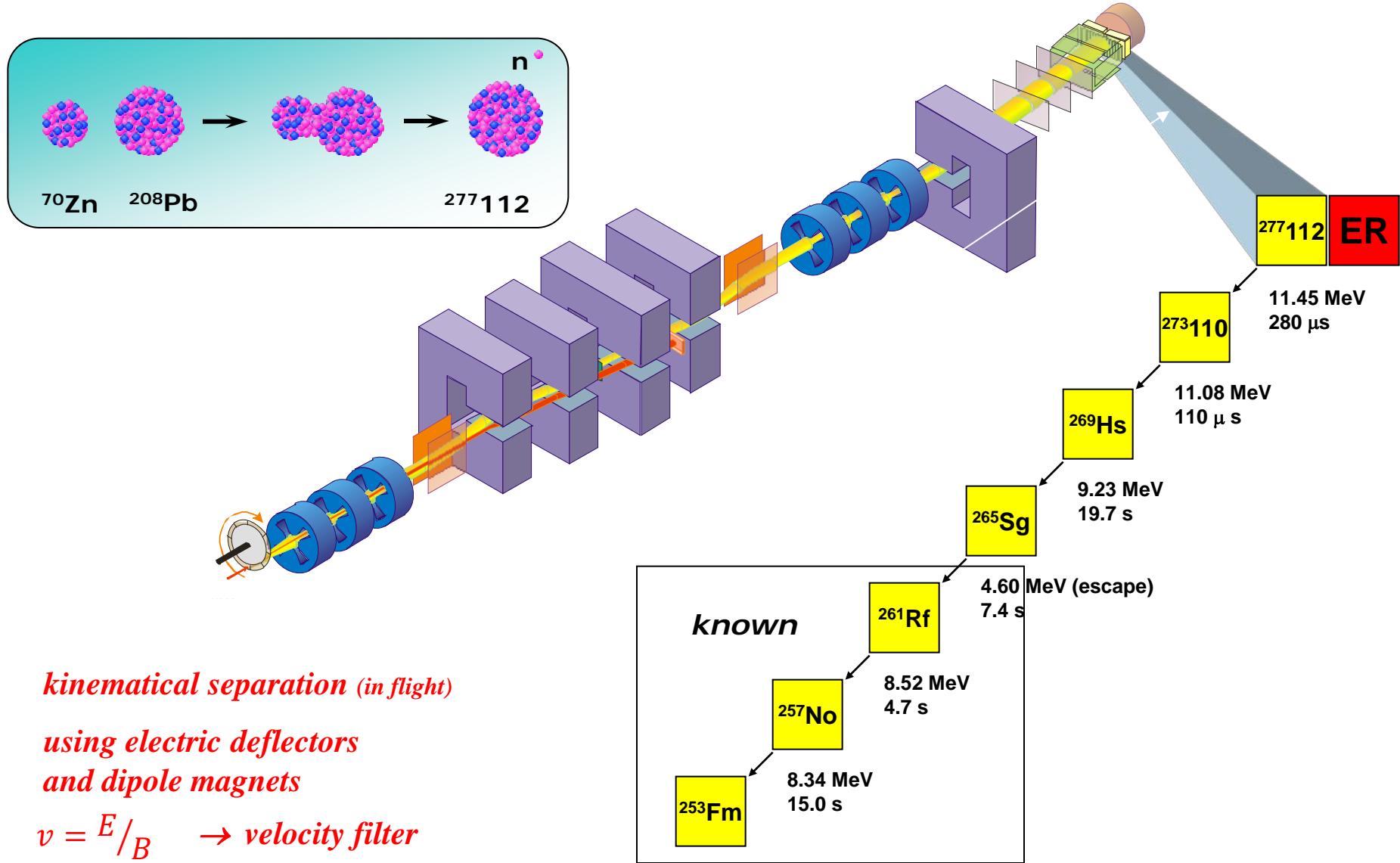
area: $27 \times 87 \text{ mm}^2$, thickness: 0.3mm, 16 strips

energy resolution $\Delta E = 18-20 \text{ keV}$ @ $E_\alpha > 6 \text{ MeV}$ (cooling 260K)

position resolution $\Delta x = 0.3 \text{ mm}$ (FWHM)

Wait for the emission of an α -particle
(or β -particle)
correlation method: implantation and decay event in the same pixel

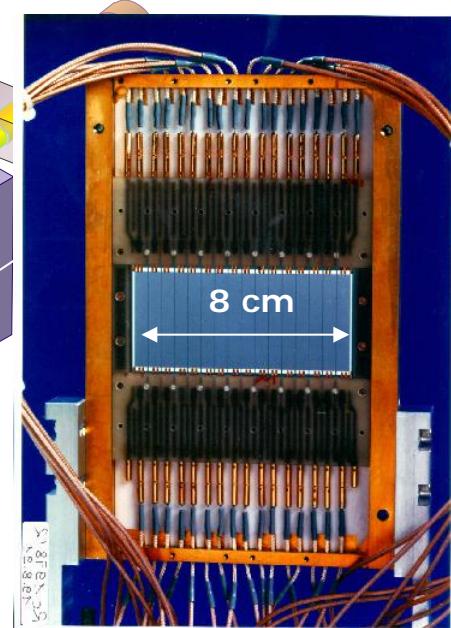
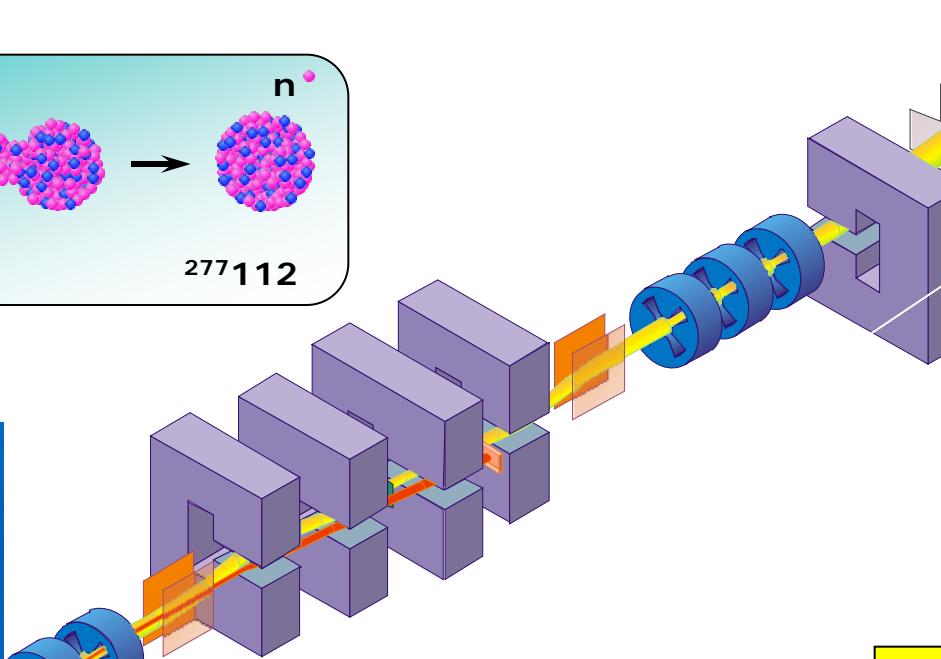
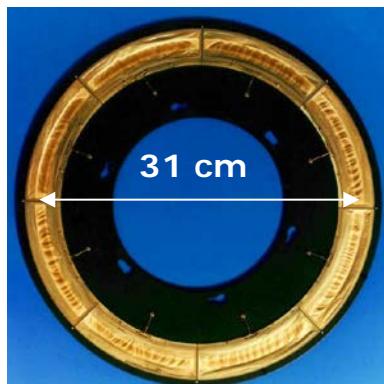
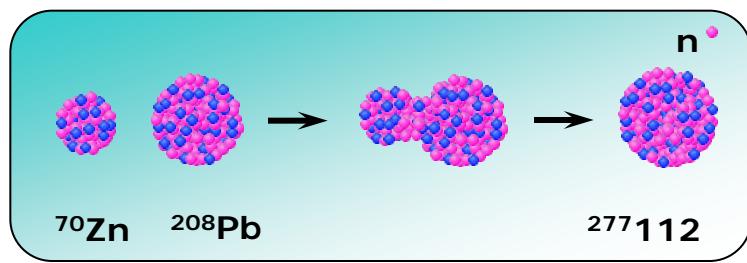
Synthesis and identification of heavy elements with SHIP



Date: 09-Feb-1996

Time: 22:37 h

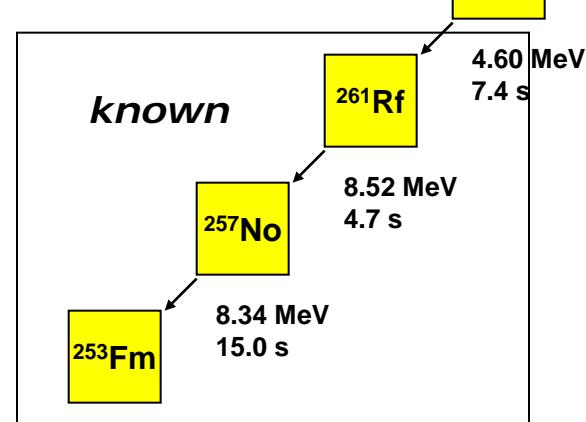
Synthesis and identification of heavy elements with SHIP



kinematical separation (in flight)

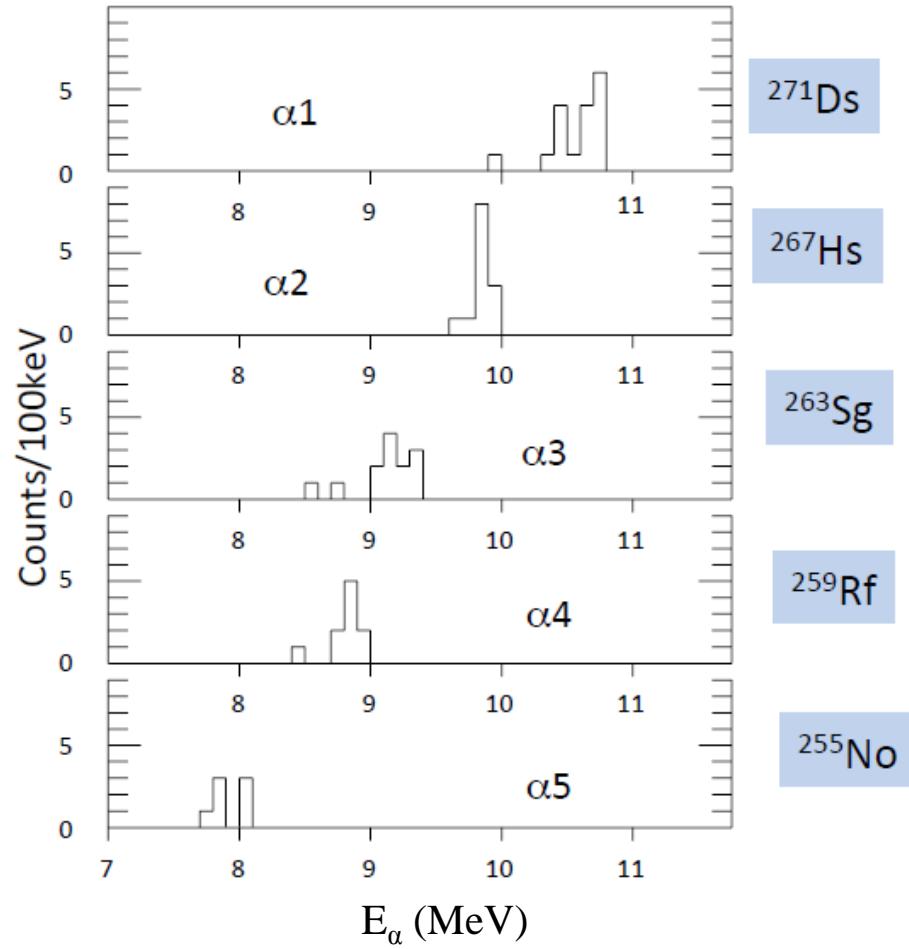
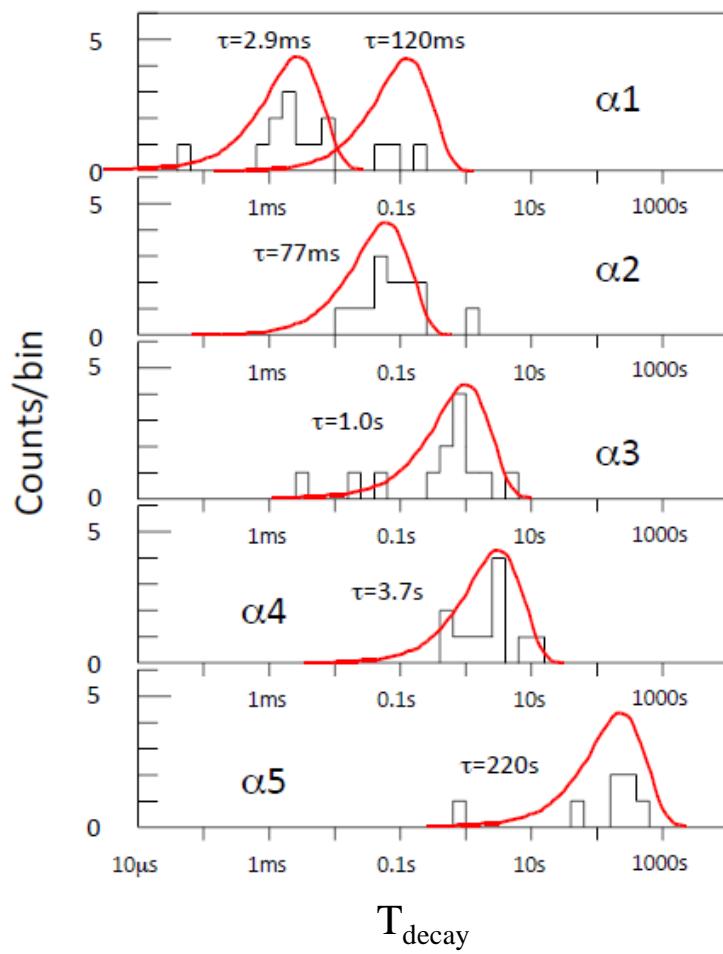
*using electric deflectors
and dipole magnets*

$$v = E/B \quad \rightarrow \text{velocity filter}$$



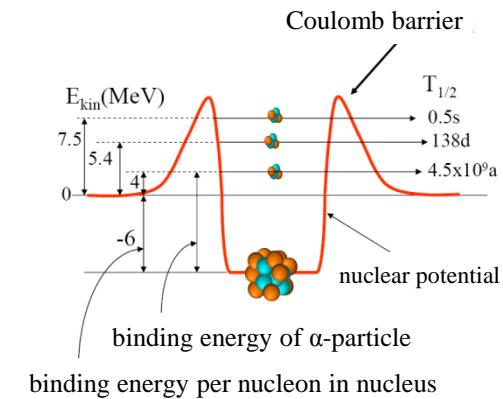
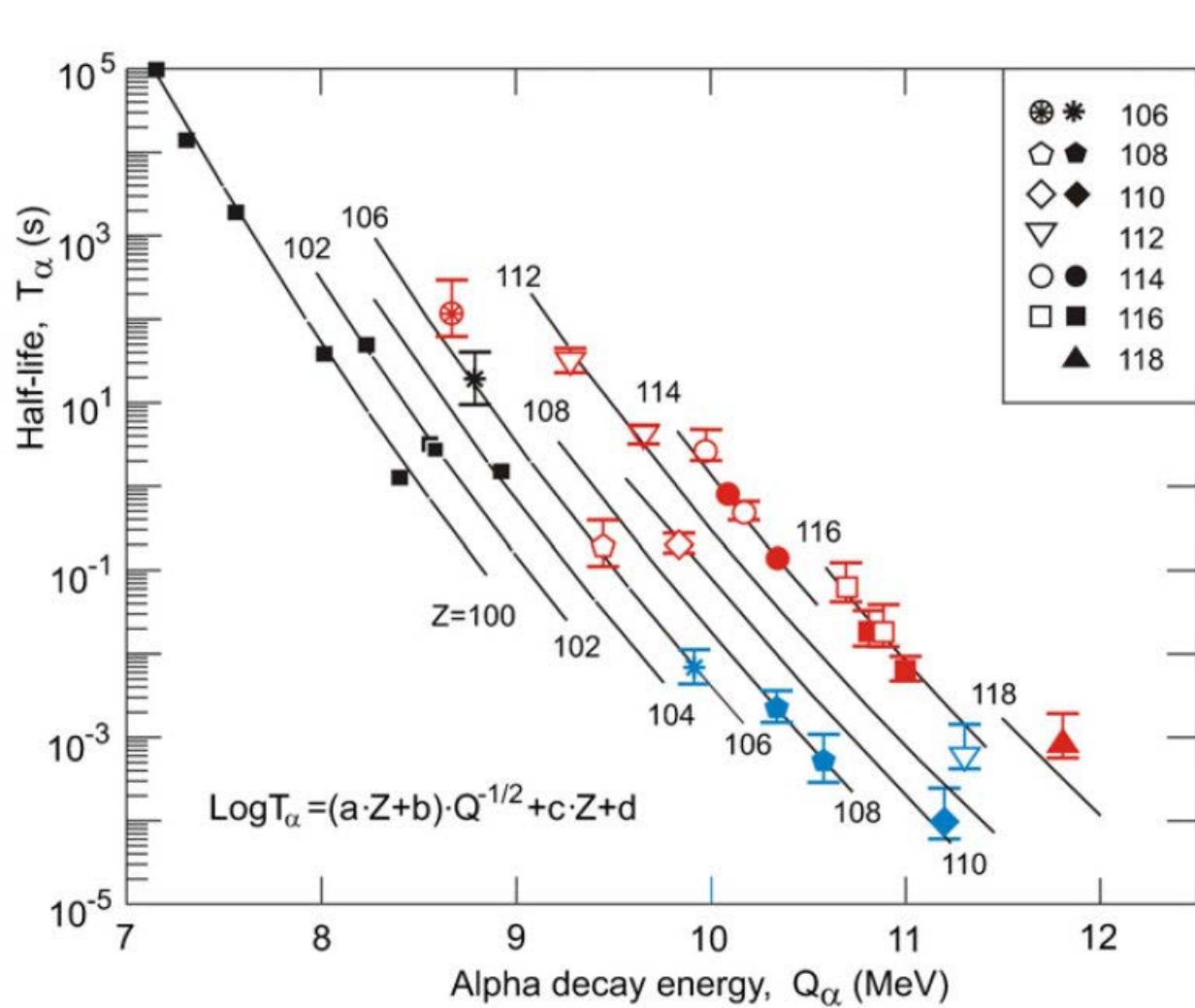
Date: 09-Feb-1996

Time: 22:37 h

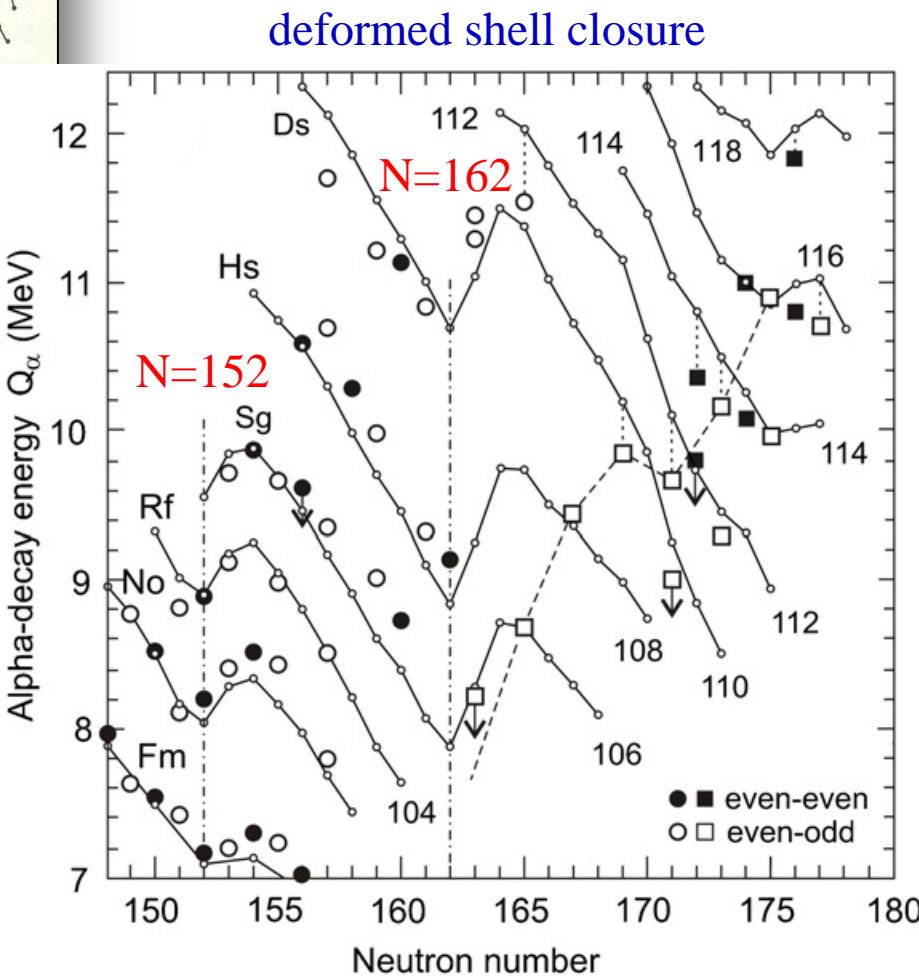
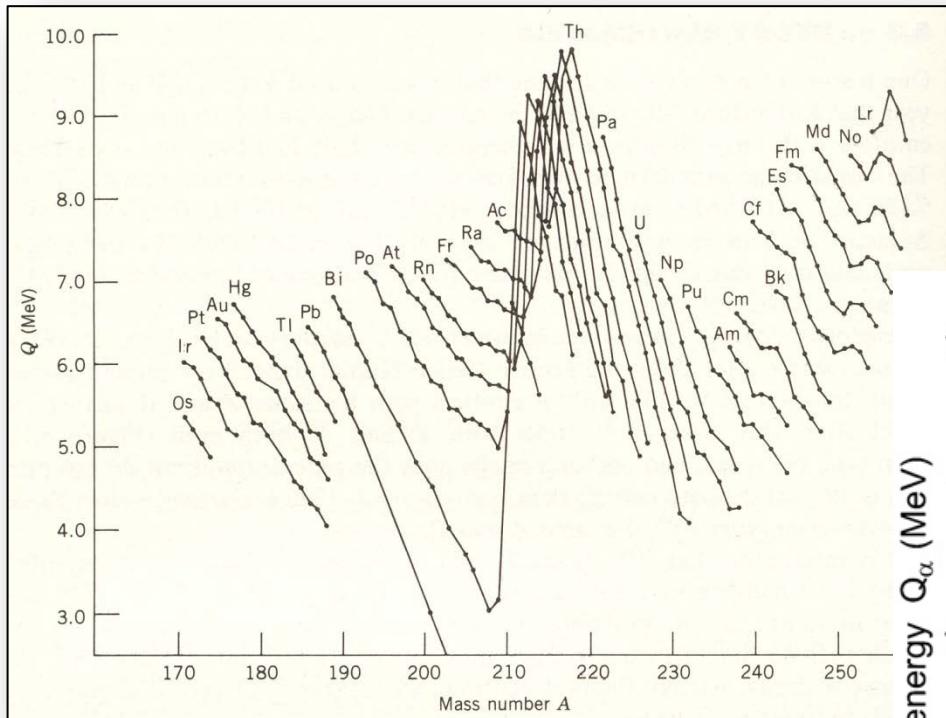


Geiger-Nuttall relationship

- The average decay properties of even mass decay chains match the Geiger-Nuttall relationship

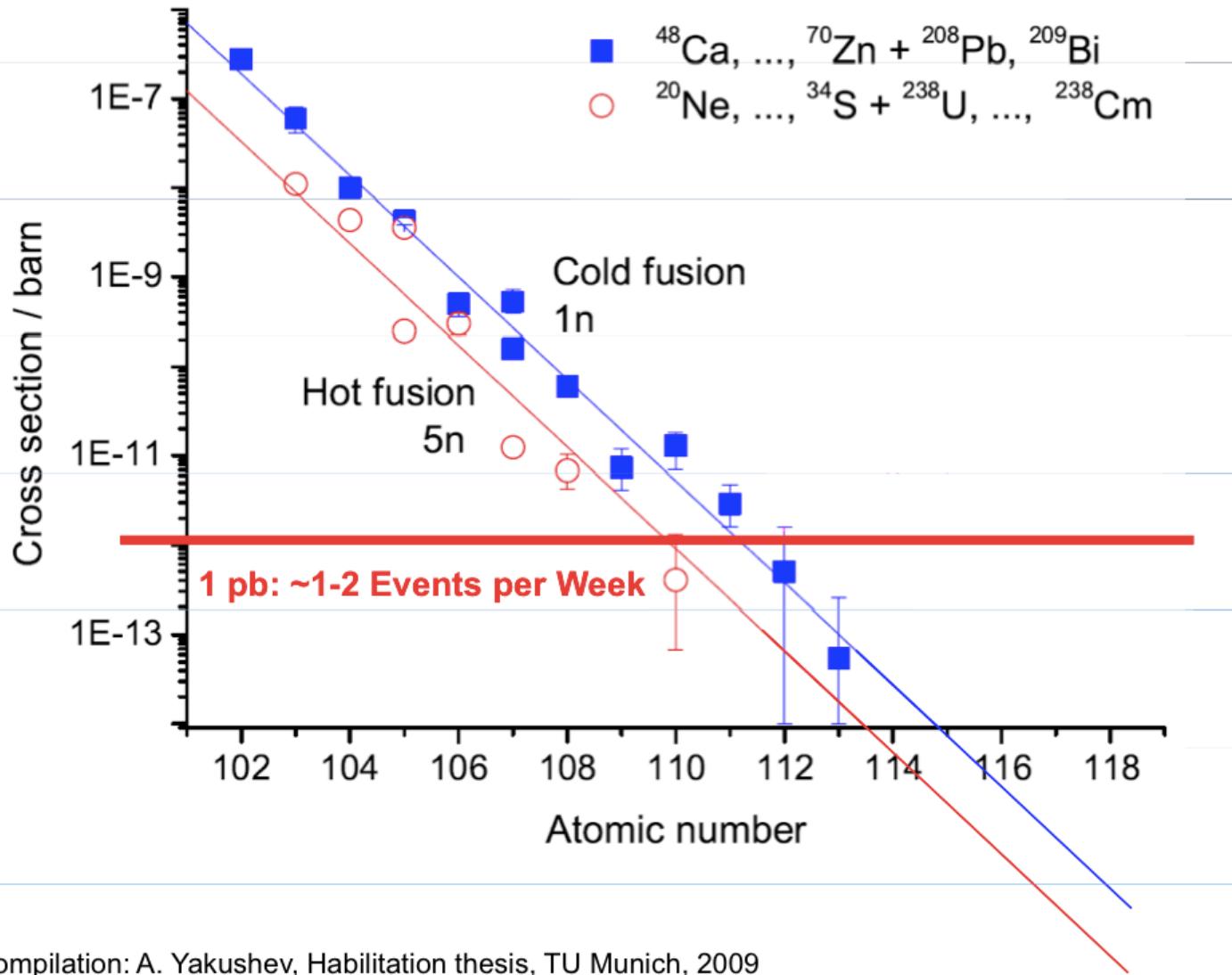


Increase of Q_α values for the isotopes with $Z = 112-118$



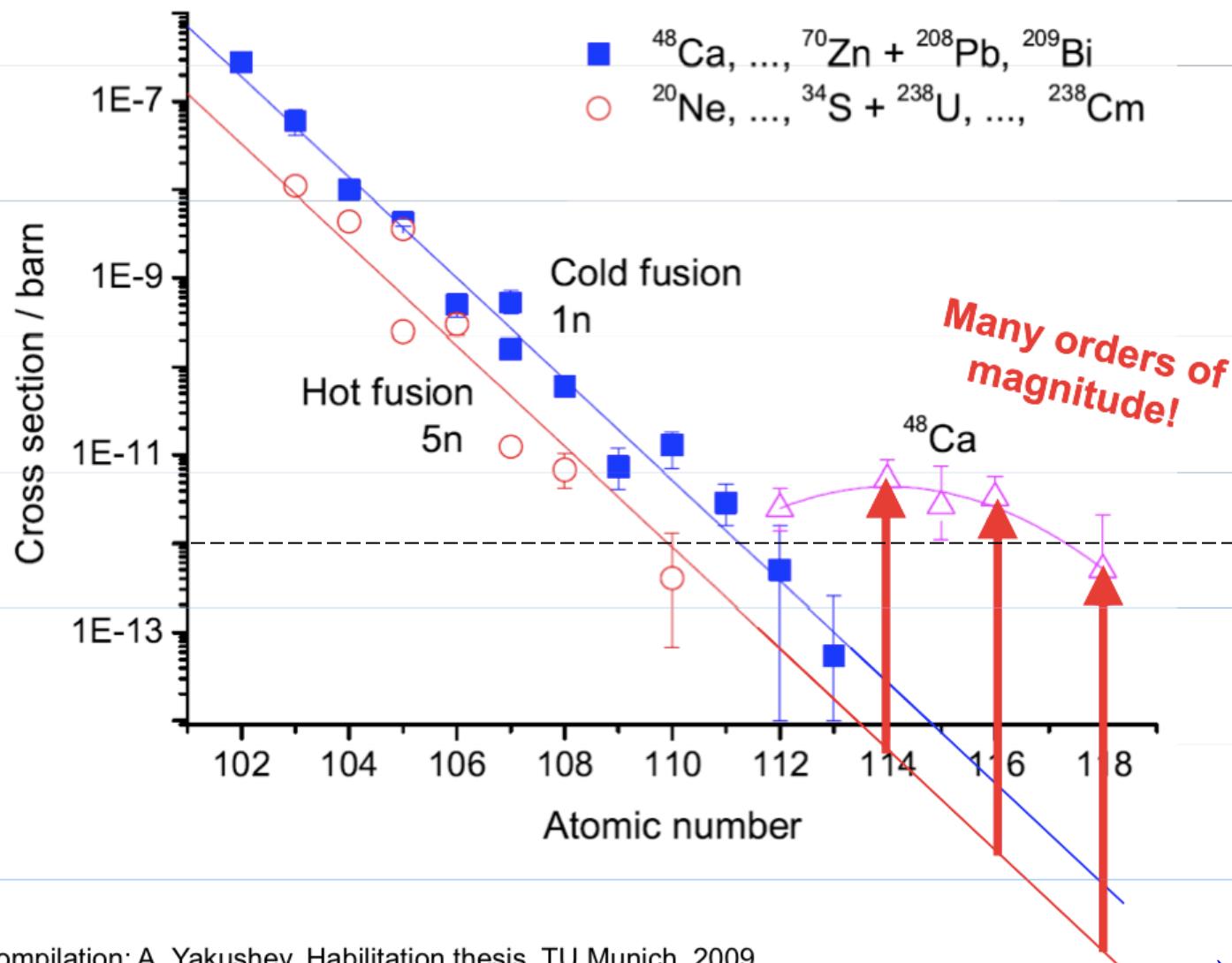
Micro-macro predictions:
nuclei with $N \geq 175$ are close to spherical ($\beta_2 \approx 0.09$)

The end of the “cold fusion” path?



Compilation: A. Yakushev, Habilitation thesis, TU Munich, 2009

“Hot fusion” – The Dubna Era



Yuri Oganessian

Compilation: A. Yakushev, Habilitation thesis, TU Munich, 2009

Using ^{48}Ca beams with actinide targets

Joint Institute of Nuclear Research (Dubna) has extended the periodic table to Z=118

- ❖ ^{48}Ca projectiles produced by U400 heavy ion accelerator

energy: 235 -250 MeV
beam intensity: 1.0 – 1.5 p μ A
consumption: 0.5 – 0.8 mg/h
beam dose: $(0.3 – 3.0) \cdot 10^{19}$



year	element	reaction	number of atoms
2000	114	$^{48}\text{Ca} \rightarrow ^{244}\text{Pu}$	50
2004	113	decay product of Z=115	8
2004	115	$^{48}\text{Ca} \rightarrow ^{243}\text{Am}$	30
2005	116	$^{48}\text{Ca} \rightarrow ^{248}\text{Cm}$	30
2006	118	$^{48}\text{Ca} \rightarrow ^{249}\text{Cf}$	3 - 4
2010	117	$^{48}\text{Ca} \rightarrow ^{249}\text{Bk}$	6

prices per 1 mg
 $^{197}\text{Au} \approx 0.03 \$$
 $^{239}\text{Pu} \approx 4 \$$
 $^{48}\text{Ca} \approx 80 \$$
 $^{249}\text{Cf} \approx 60\,000 \$$

Using ^{48}Ca beams with actinide targets

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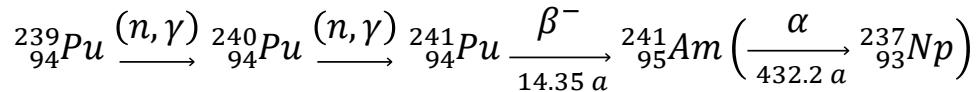
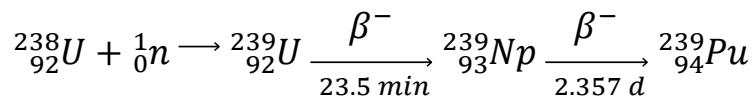
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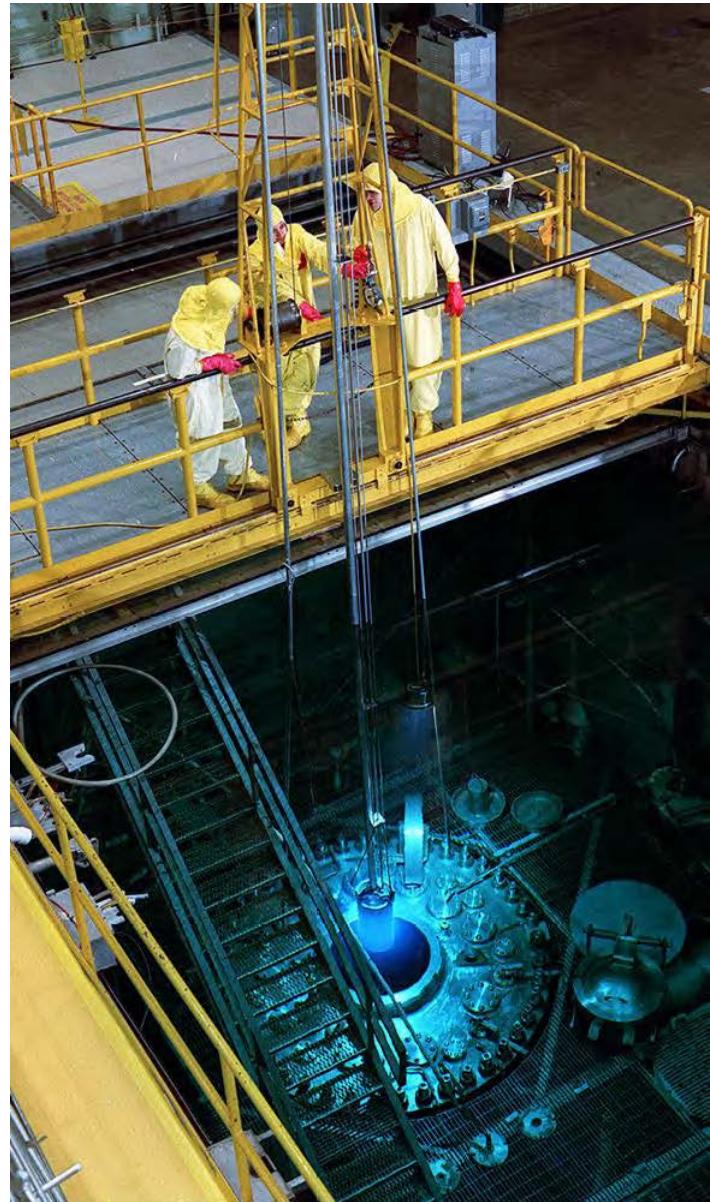
reaction	Q_{gg} (MeV)	$V_C(R_{int})$ (MeV)	E^* (MeV)
$^{48}_{20}\text{Ca} + ^{244}_{94}\text{Pu} \rightarrow ^{292}_{114}\text{Fl}$	-160.5	197.3	36.8
$^{48}_{20}\text{Ca} + ^{243}_{95}\text{Am} \rightarrow ^{291}_{115}\text{Mc}$	-170.6	199.7	29.1
$^{48}_{20}\text{Ca} + ^{248}_{96}\text{Cm} \rightarrow ^{296}_{116}\text{Lv}$	-166.6	201.1	34.5
$^{48}_{20}\text{Ca} + ^{249}_{97}\text{Bk} \rightarrow ^{297}_{117}\text{Ts}$	-170.1	203.2	33.1
$^{48}_{20}\text{Ca} + ^{249}_{98}\text{Cf} \rightarrow ^{297}_{118}\text{Og}$	-174.3	205.4	31.1

Q_{gg} and V_C (Bass) from <http://nrv.jinr.ru/nrv/webnrv/qcalc/>

Irradiation of targets at HFIR reactor (Oak Ridge)



- ❖ Irradiation in the HFIR flux trap (18 month)
 - thermal-neutron flux of
 $2.5 \cdot 10^{15}$ neutrons/(s·cm²)
 - 31 target positions
(10 – 13 targets typically irradiated)
 - produces ~35 mg ²⁵²Cf per target
(smaller quantities of Bk, Es, Fm)



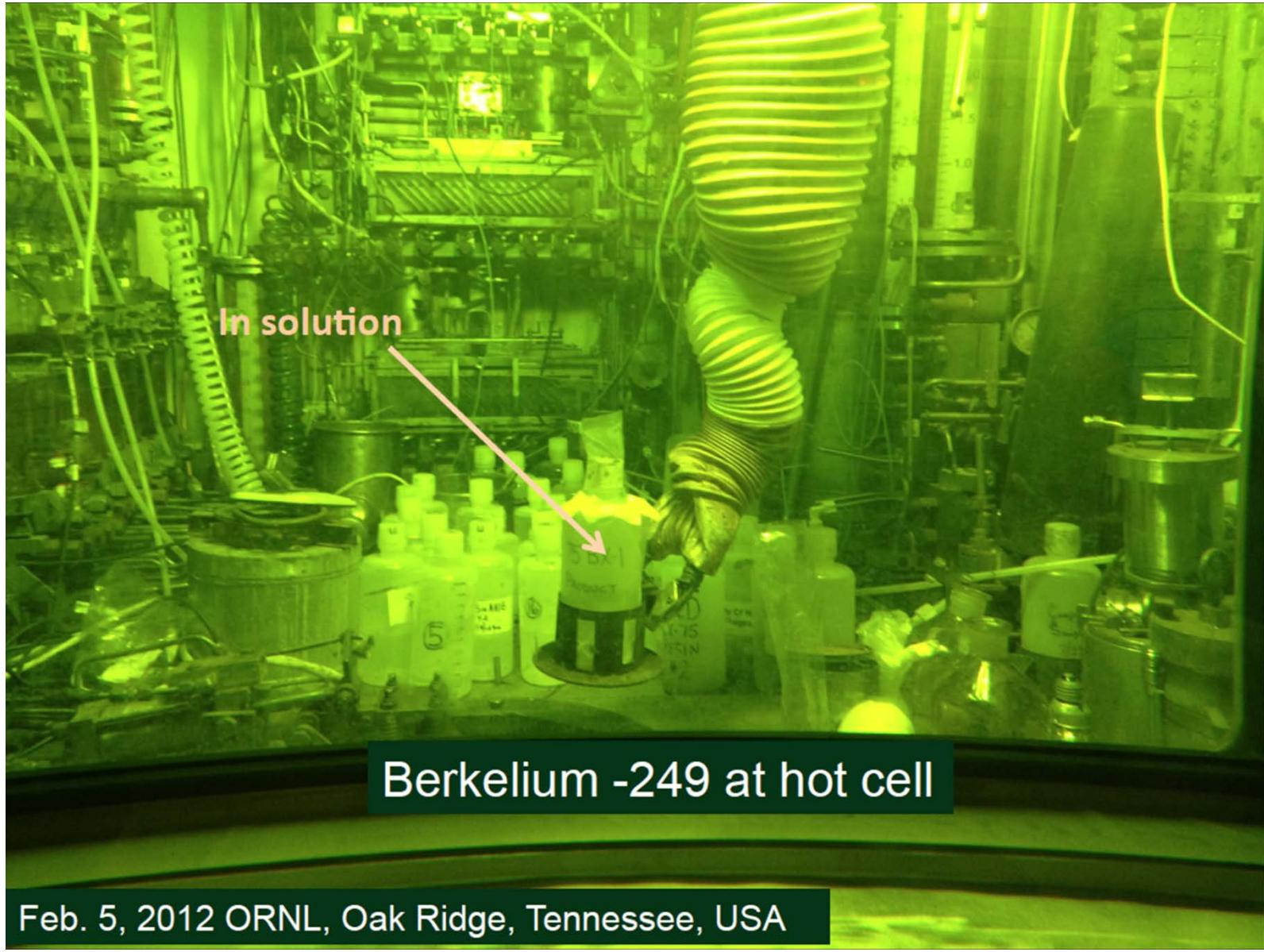
22 mg of ^{249}Bk \approx 1 M\$, 250 day irradiation in HIFR (ORNL)



$\text{Bk}(\text{NO}_3)_3$ product

- ❖ The two year experimental campaign began with a 250 day irradiation in HFIR, producing 22 milligram of ^{249}Bk , which has a 320 day half-life. The irradiation was followed by 90 days of processing at radiochemical Engineering Development Center (REDC) to separate and purify the Berkelium. The ^{249}Bk target was prepared at Dimitrovgrad and then bombarded for 150 days at the Dubna facility.

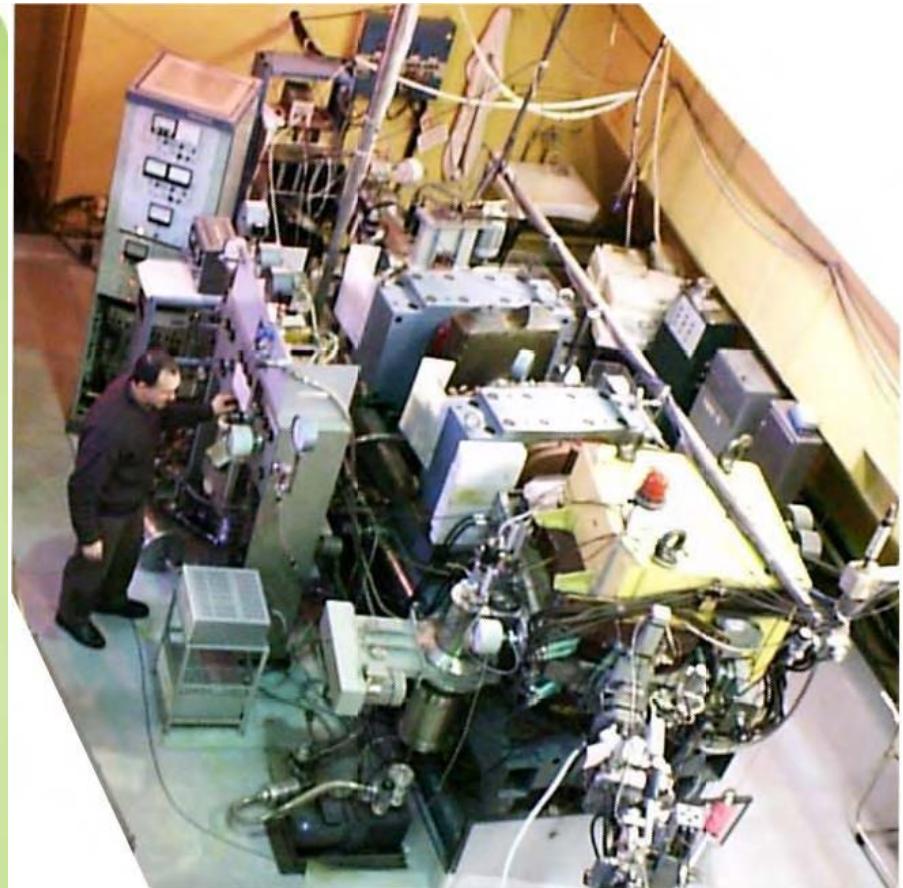
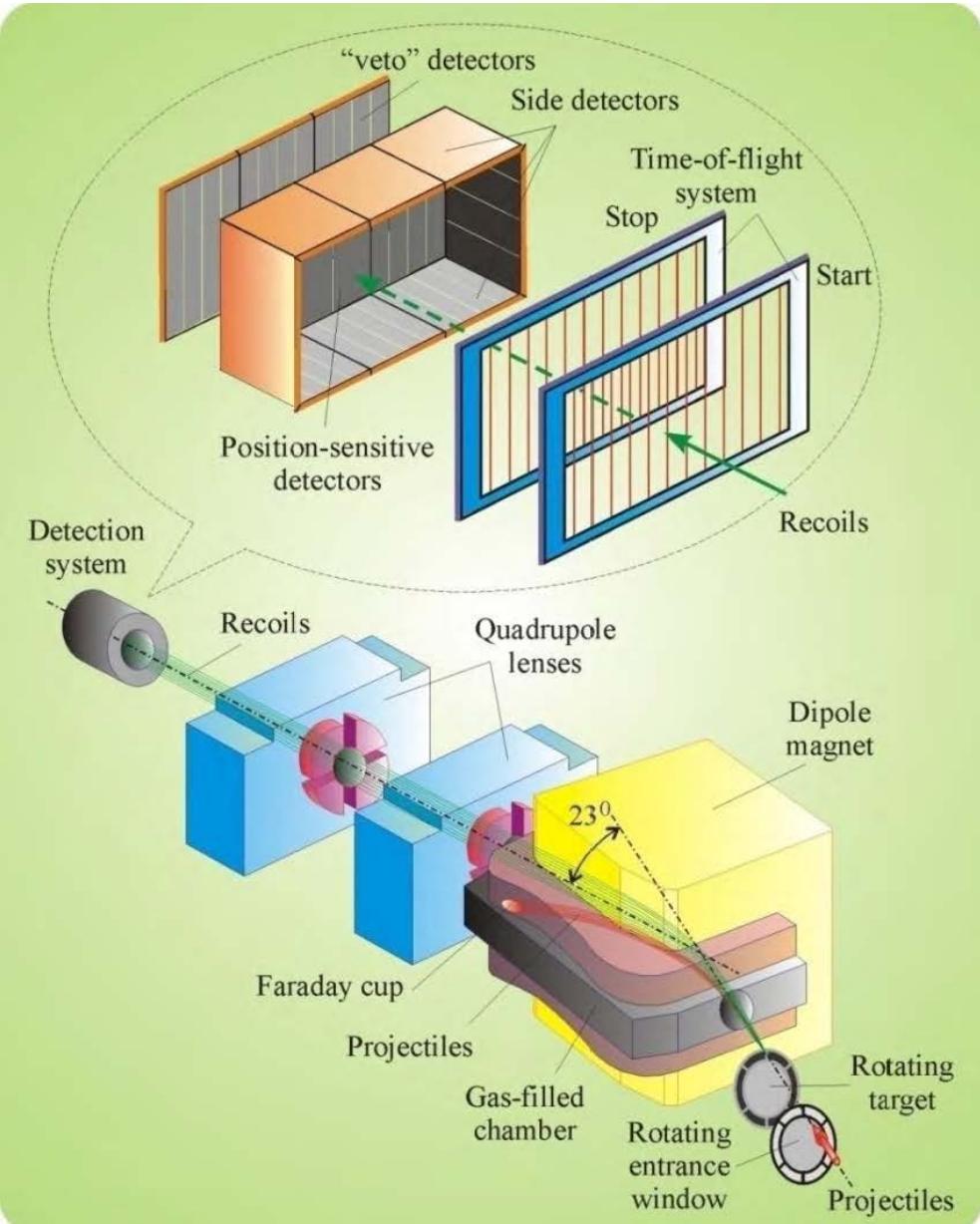
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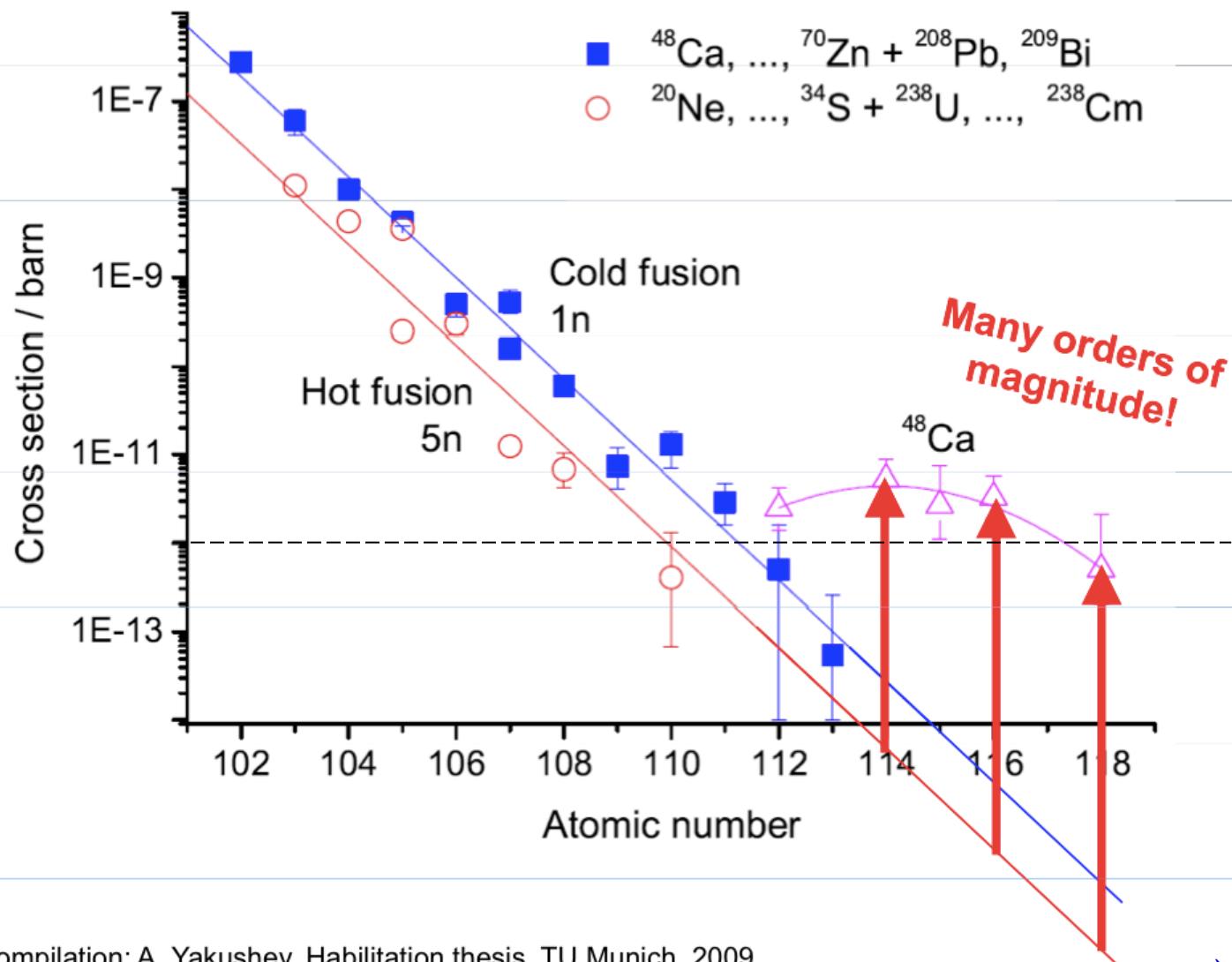
^{249}Cf target



Dubna gas filled recoil separator (DGFRS)



“Hot fusion” – The Dubna Era



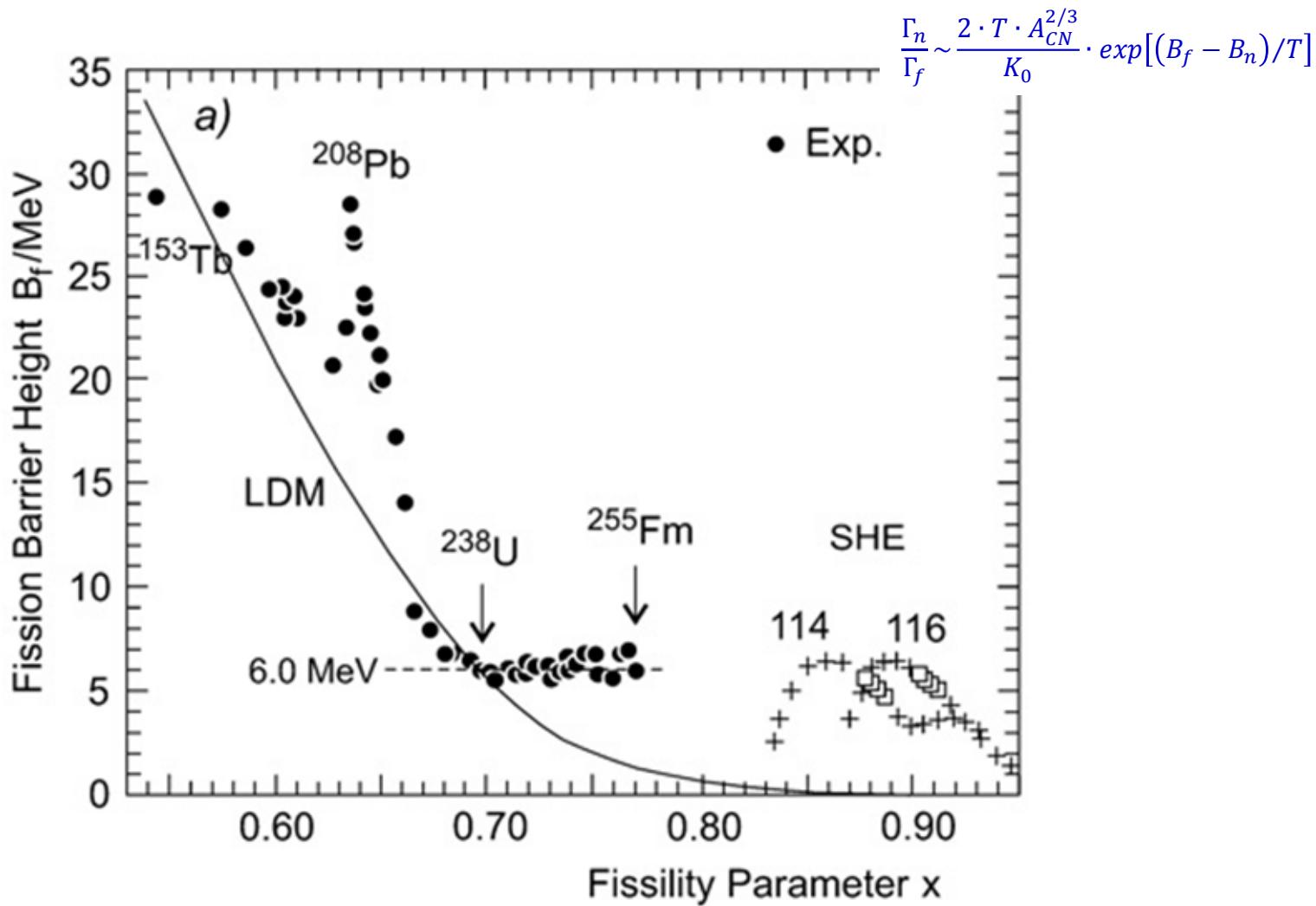
Yuri Oganessian

Compilation: A. Yakushev, Habilitation thesis, TU Munich, 2009

→ requires very efficient separation and detection techniques

Cross sections – fission barriers

$$\sigma_{ER} = \sigma_{capture} \cdot P_{CN} \cdot P_{survival}$$



Spontaneous fission half-lives of actinides

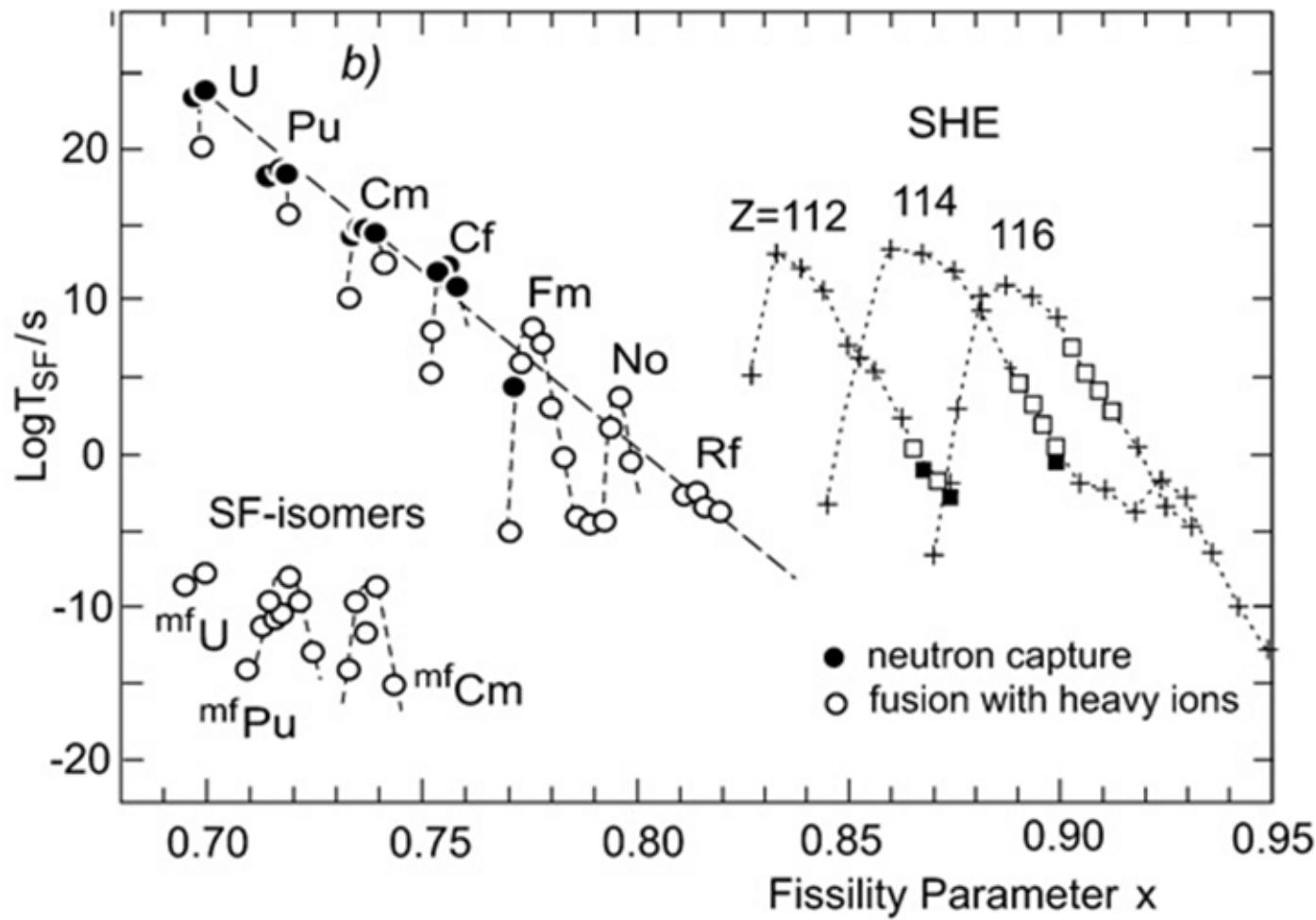


Chart of nuclides: the domain of heavy and superheavy elements

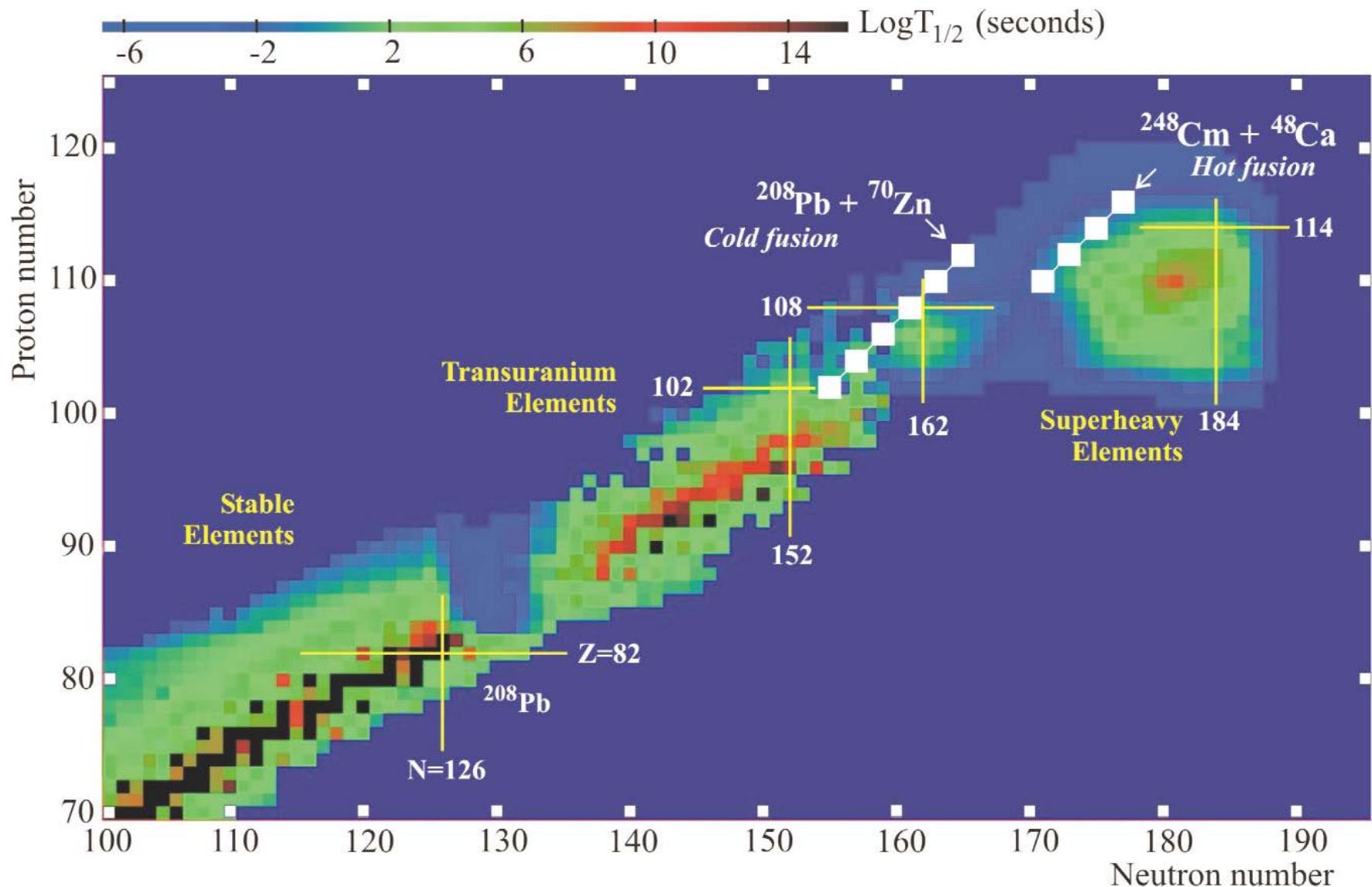
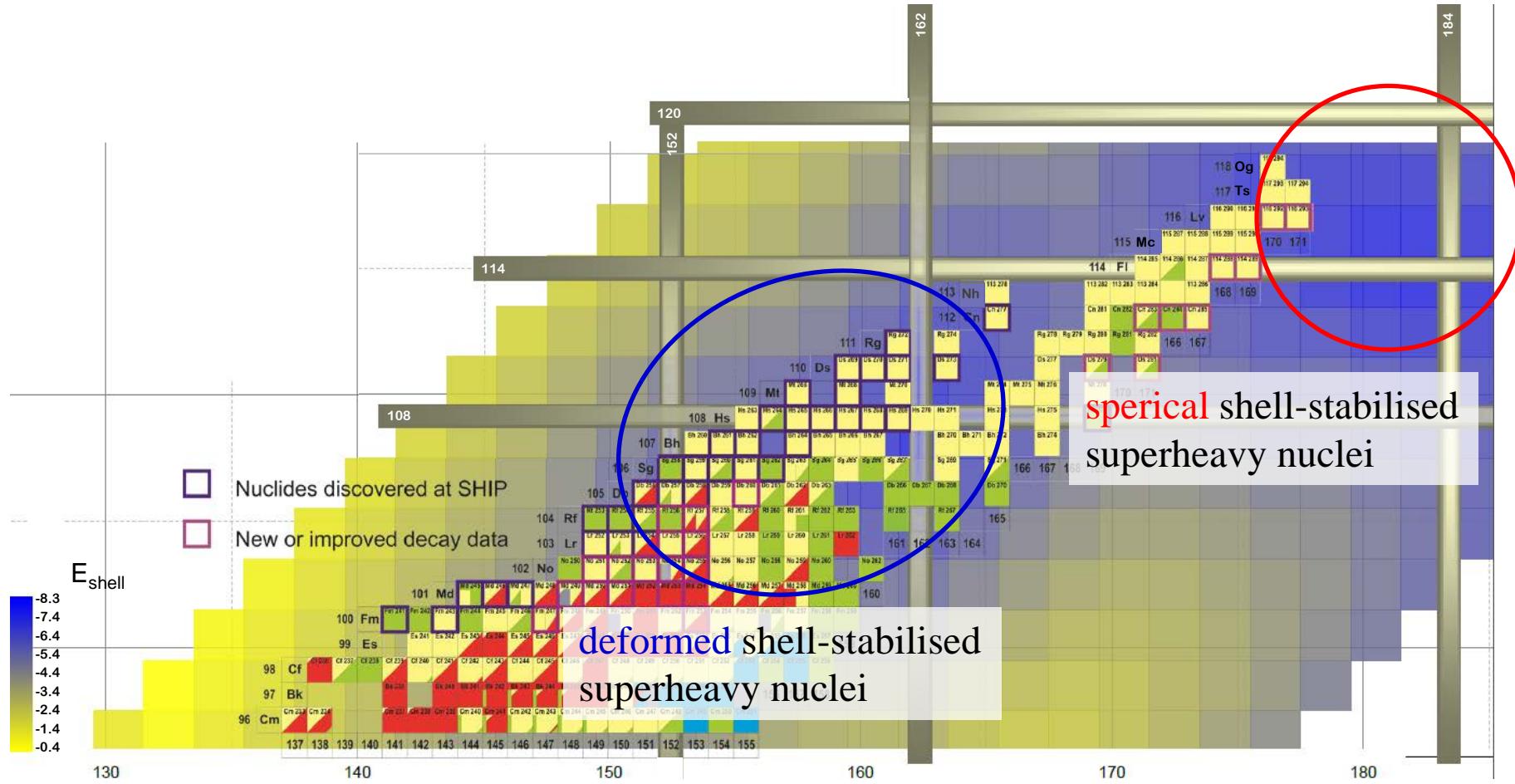


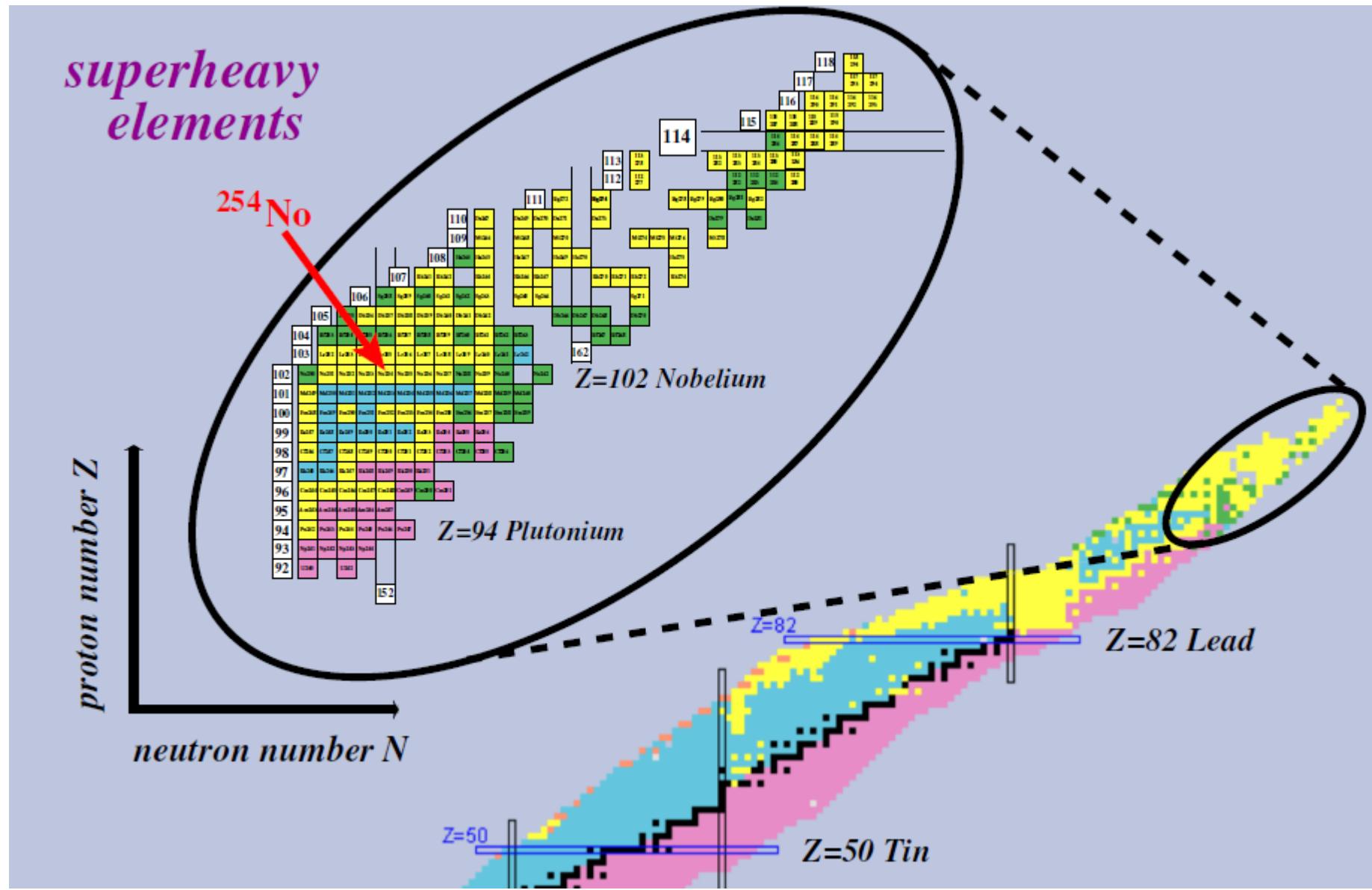
Chart of nuclides: the domain of heavy and superheavy elements



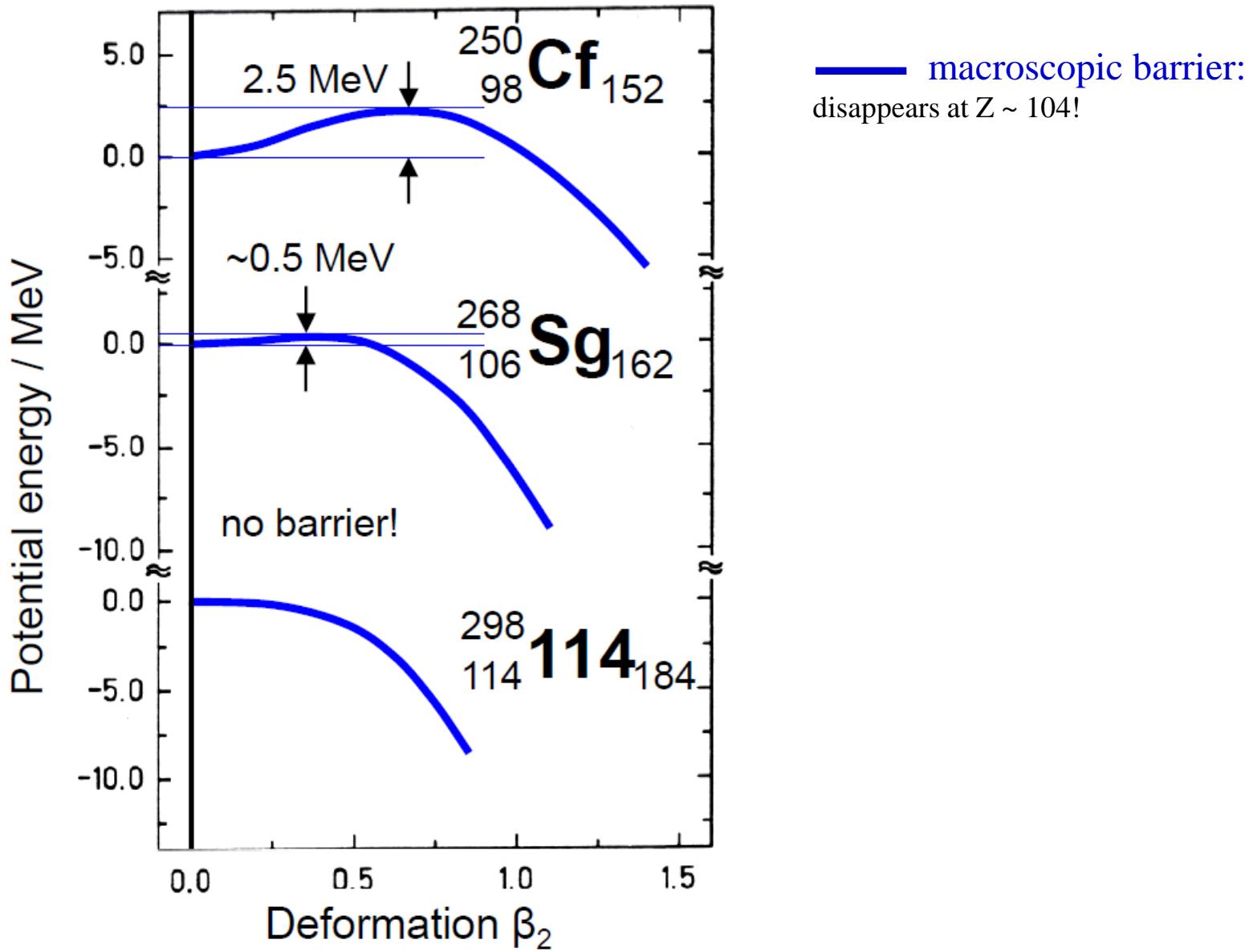
Calc.: A. Sobiczewski

Sn	Sb	Tl	In
Cd	Antimony (51)	Thallium (81)	Indium (41)
Cf	Po	Pb	Aluminum (13)
Bk	Bi	Lu	Thallium (81)
Cm	At	Yb	Antimony (51)
Cm	Rn	Tm	Indium (41)
Cm	Og	Lu	Thallium (81)

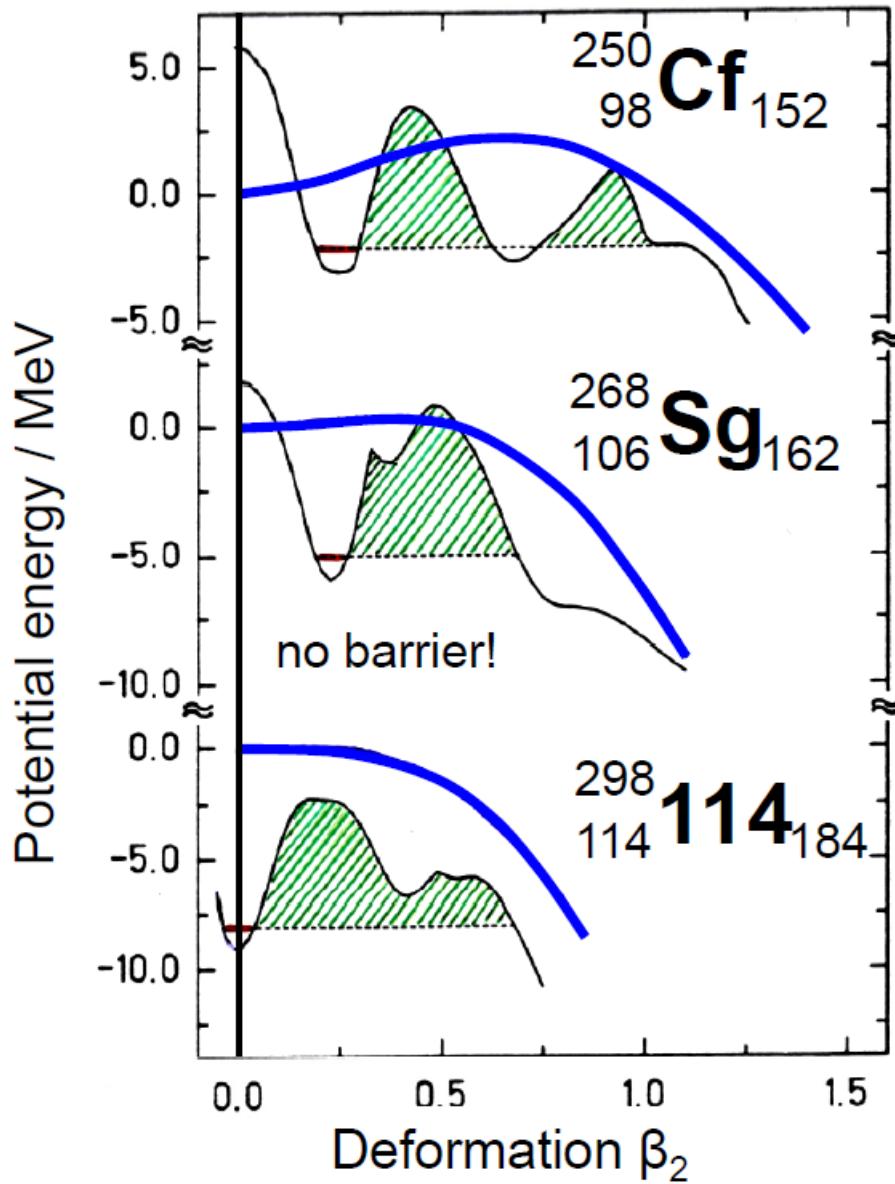
What is the structure of SHE?



Influence of shell effects on fission barrier



Influence of shell effects on fission barrier



— macroscopic barrier:

disappears at $Z \sim 104$!

— with shell structure:

spherical \leftrightarrow deformed ground state

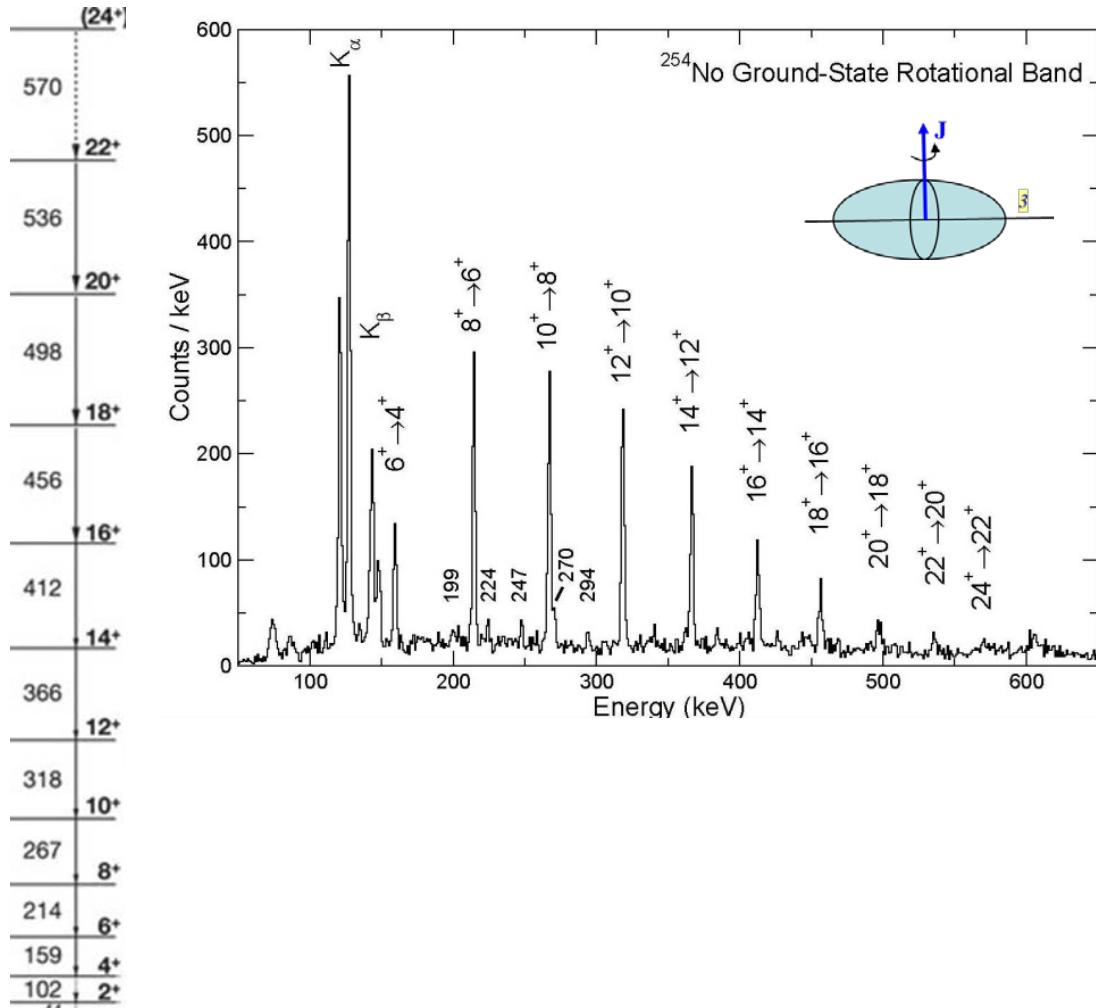
fission barrier is also > 0 for $Z \geq 104$

some fission barriers have complicated shapes,
multi-humped structure

elements exist only due to shell effects:

superheavy elements

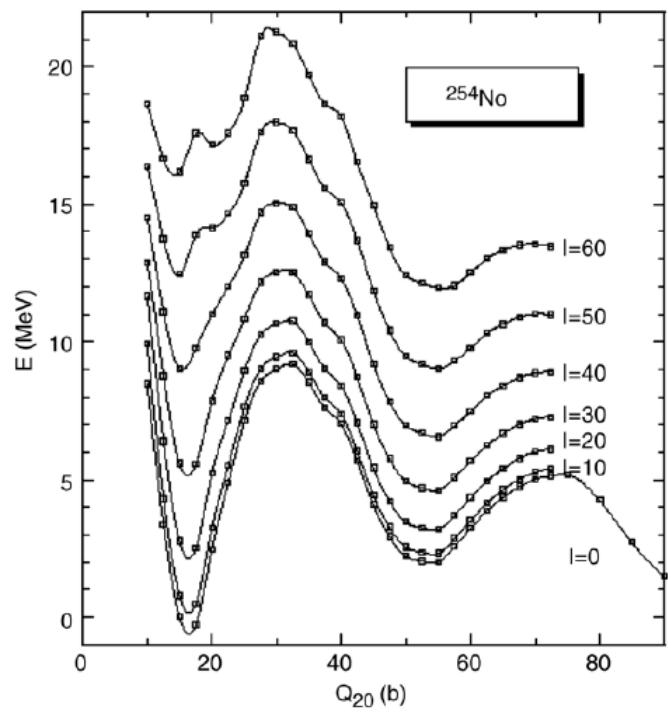
Spinning the heaviest elements



rotational energy: $E_I = \frac{\hbar^2}{2\mathfrak{J}} \cdot I(I + 1)$

$$\gamma\text{-ray energy: } E_I - E_{I-2} = \frac{\hbar^2}{2\mathfrak{J}} \cdot (4I - 2)$$

dependence of the fission barrier on spin I



Rotational Bands

(18⁺) (2372) (18⁺) (2280)

(18⁺) (2395) (18⁺) (2339)

(16⁺) (1921) (16⁺) (1845)

(16⁺) (1942) (16⁺) (1883)

(14⁺) (1508) (14⁺) (1448)

(14⁺) (1525) (14⁺) (1470)

(12⁺) (1137) (12⁺) (1091)

(12⁺) (1150) (12⁺) (1104)

(10⁺) (813) (10⁺) (779)

(10⁺) (822) (10⁺) (786)

(8⁺) (539) (8⁺) (516)

(8⁺) (545) (8⁺) (519)

(6⁺) (317) (6⁺) (304)

(6⁺) (321) (6⁺) (305)

(4⁺) (152) (4⁺) (146)

(4⁺) (154) (4⁺) (145)

(2⁺) (46) (2⁺) (44)

(2⁺) (46) (2⁺) (44)

0⁺ 0⁺ 0⁺ 0⁺

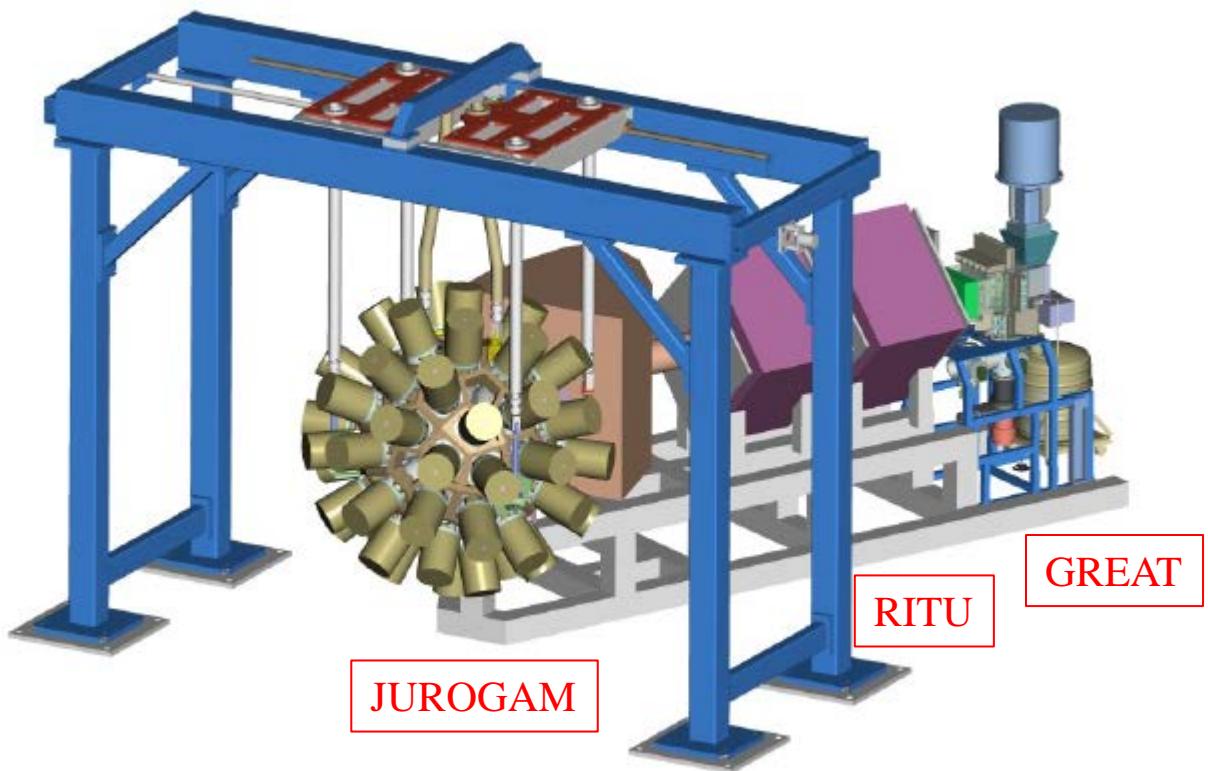
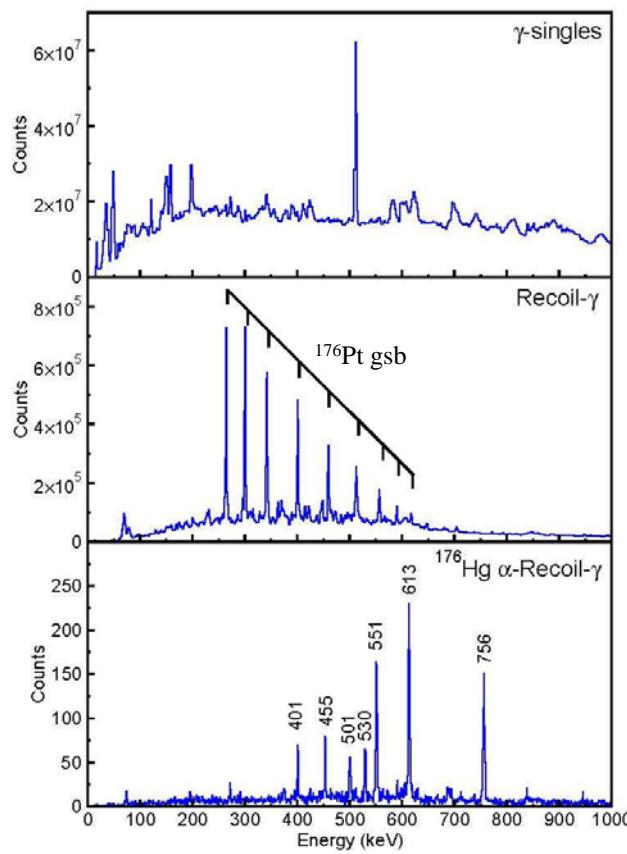
^{248}Fm

^{250}Fm

^{252}No

^{254}No

In-beam spectroscopy: recoil-decay tagging

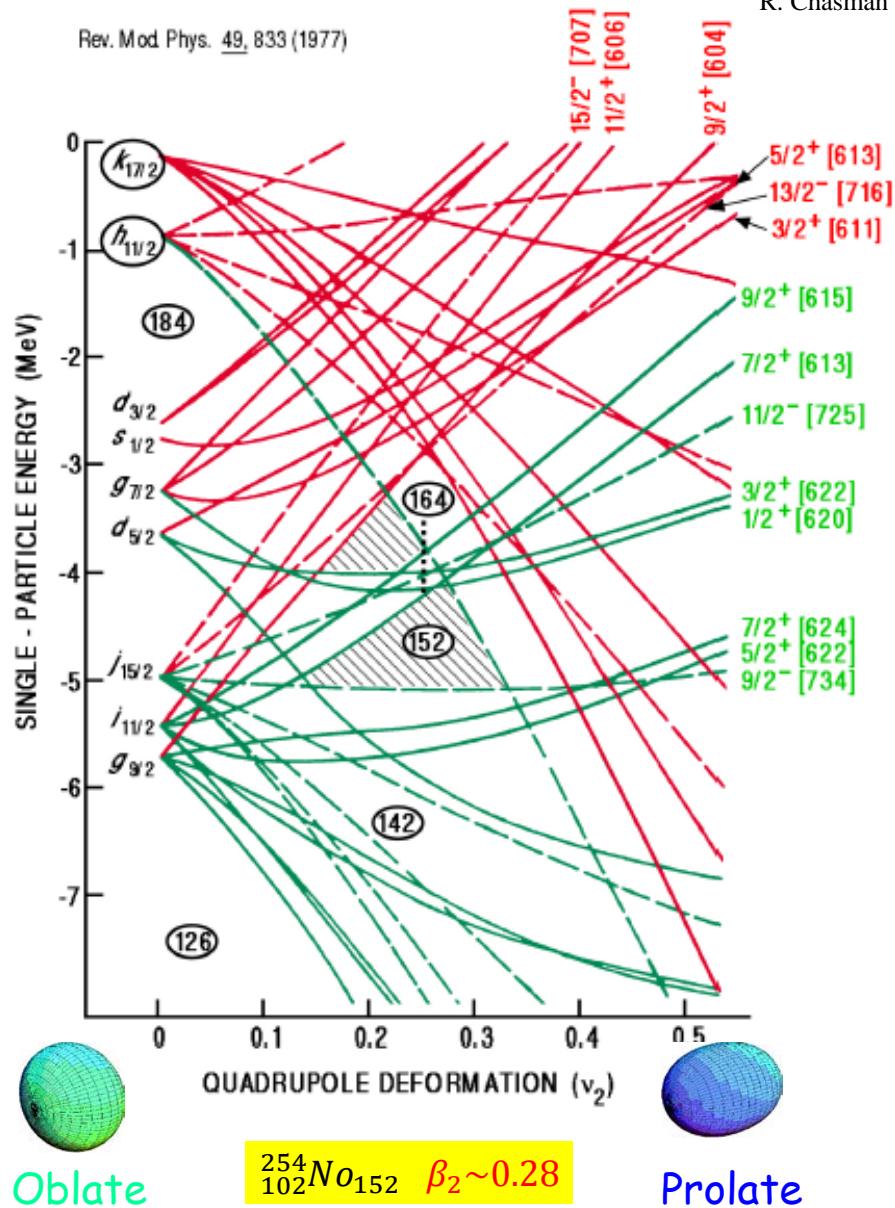


- Typical beam intensity: 6.25×10^{10} pps
- Total Gamma Ray Counting Rate: >1 MHz
- 10 nb - 3 reactions/hour
- 1 in 10^9 selectivity

Single particle orbitals

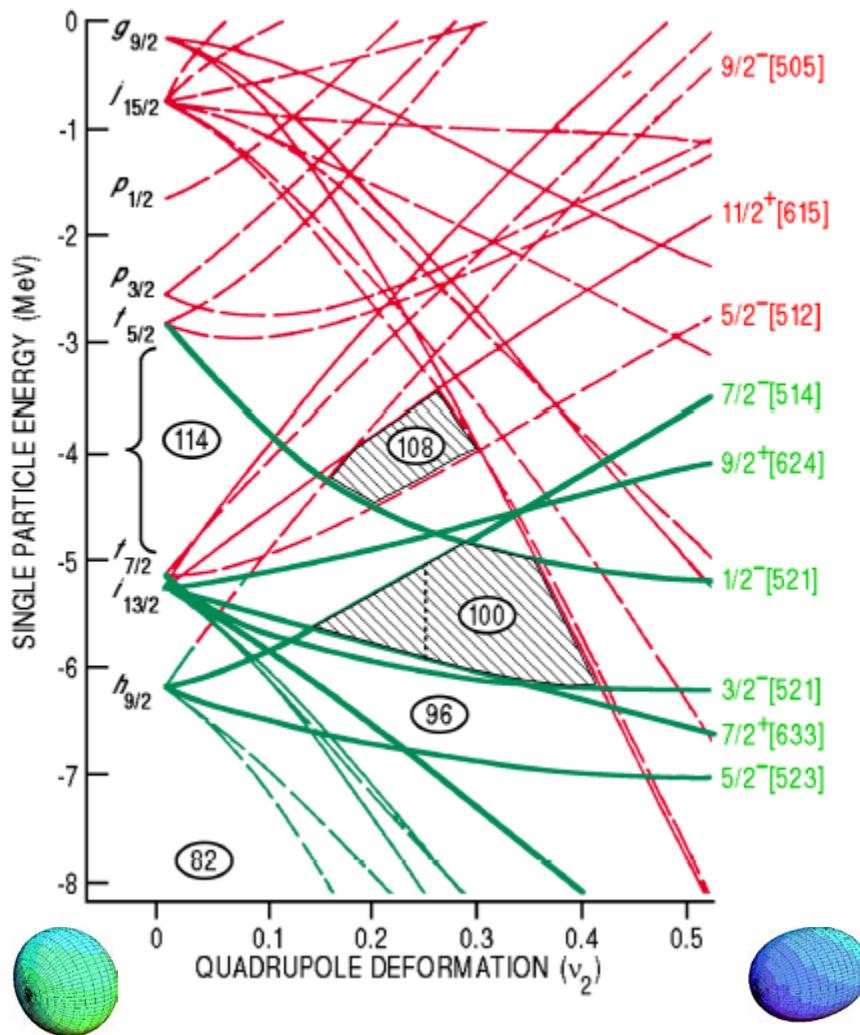
Rev. Mod. Phys. 49, 833 (1977)

R. Chasman et al. Rev. Mod. Phys. 49 (1977), 833



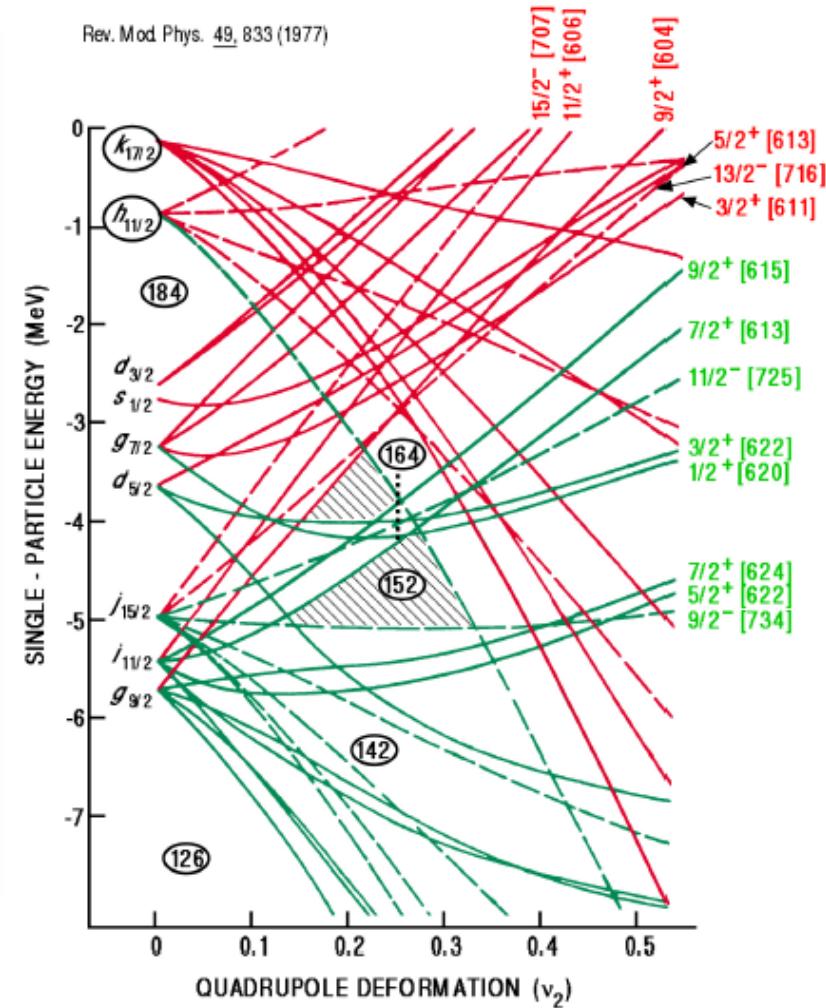
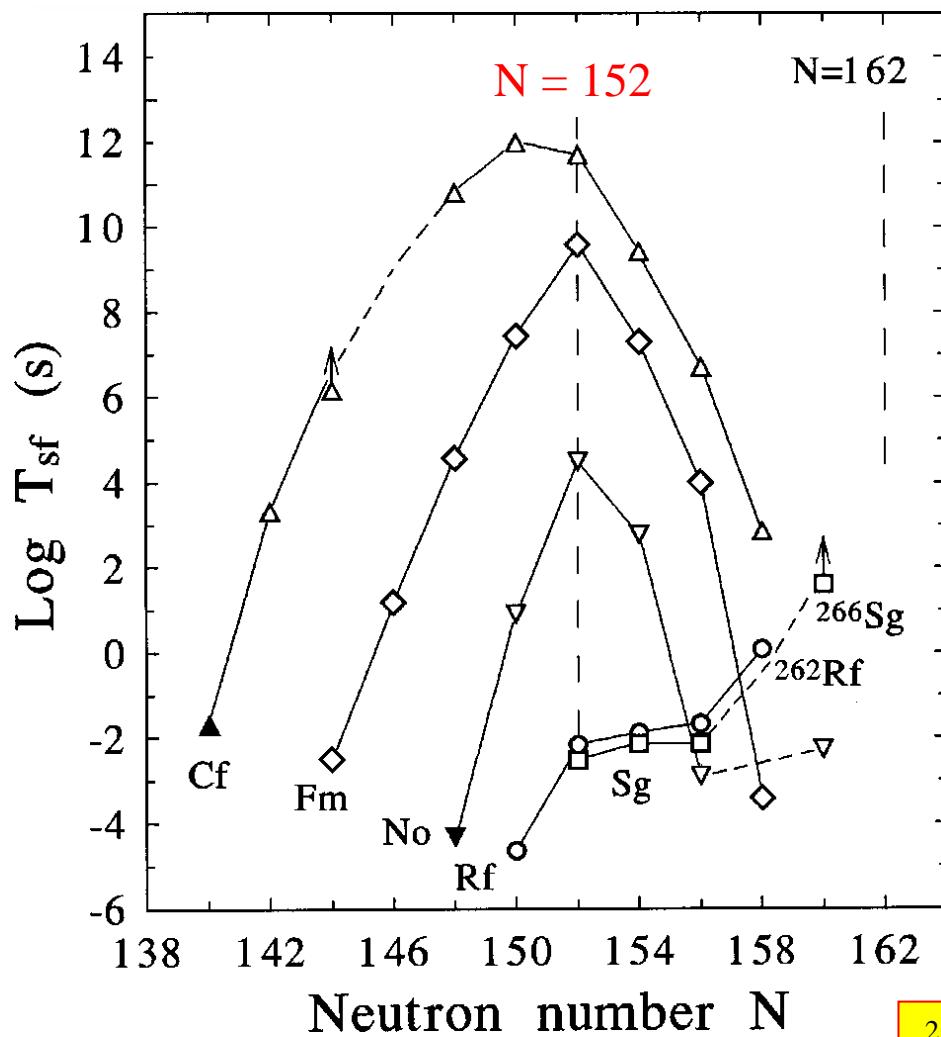
Rev. Mod. Phys. 49, 833 (1977)

ANL-P-22,033



Stability of heavy elements – Nilsson level energy gap

ANL-P-22.262

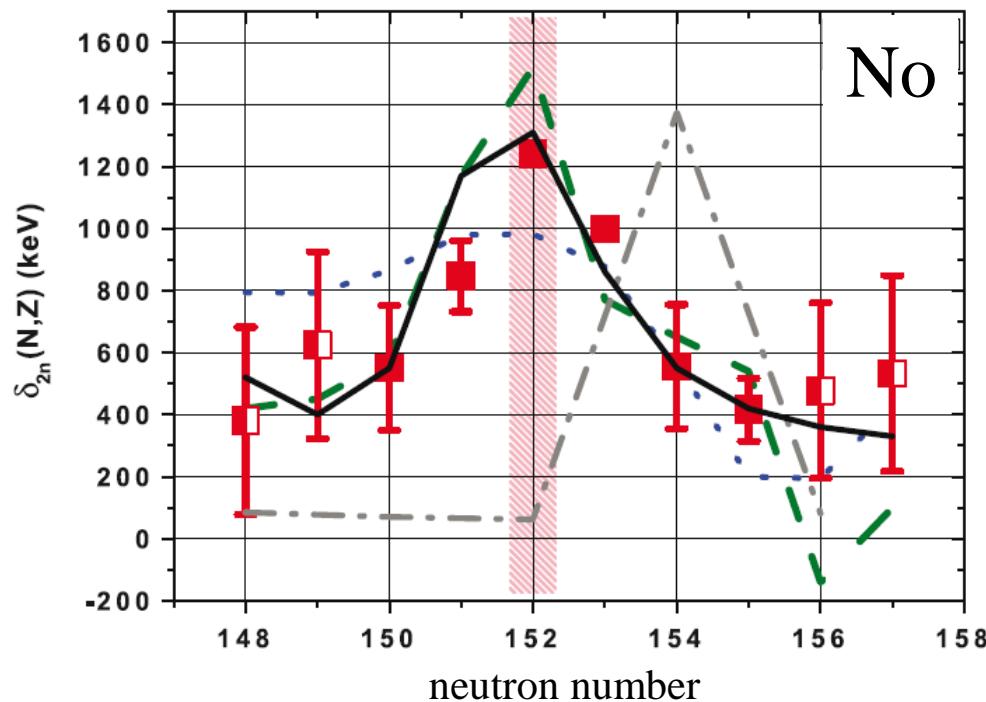


^{254}No (Z=102), ^{252}Fm (Z=100) and ^{250}Cf (Z=98)
with $N=152$
seem to be more stable than their neighbors

Shiptrap: Probing the strength of shell effects

$$\delta_{2n}(N, Z) = 2 \cdot B(N, Z) - B(N - 2, Z) - B(N + 2, Z)$$

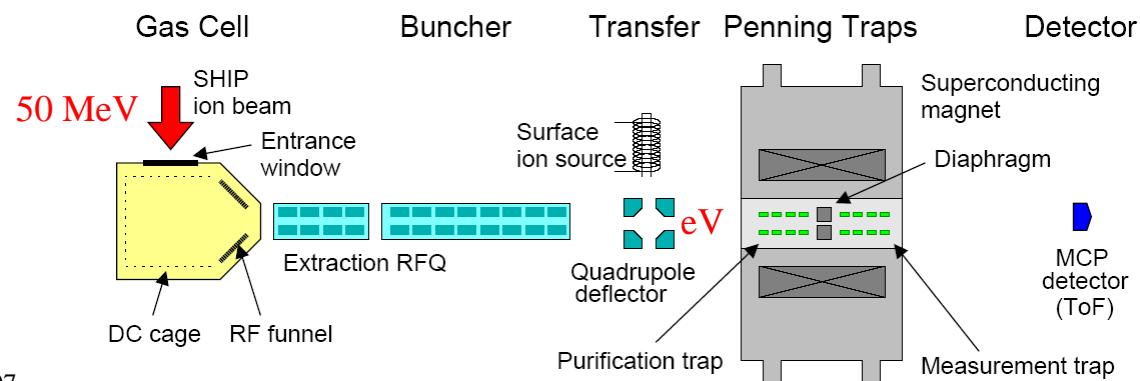
$$\delta_{2n}(N, Z) = S_{2n}(N, Z) - S_{2n}(N + 2, Z)$$



Muntian (mic-mac), Z=114 N=184
 Möller FRDM, Z=114 N=184
 TW-99, Z=120 N=172
 SkM* Z=126 N=184

$^{206}\text{Pb}(\text{Ca}, 2\text{n})^{252}\text{No}$
 $^{207}\text{Pb}(\text{Ca}, 2\text{n})^{253}\text{No}$
 $^{208}\text{Pb}(\text{Ca}, 2\text{n})^{254}\text{No}$
 $^{208}\text{Pb}(\text{Ca}, 1\text{n})^{255}\text{No}$

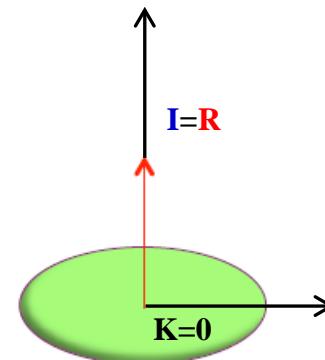
E. Minaya Ramirez et al.; Science 337 (2012), 1207



Isomeric states in even-even nuclei

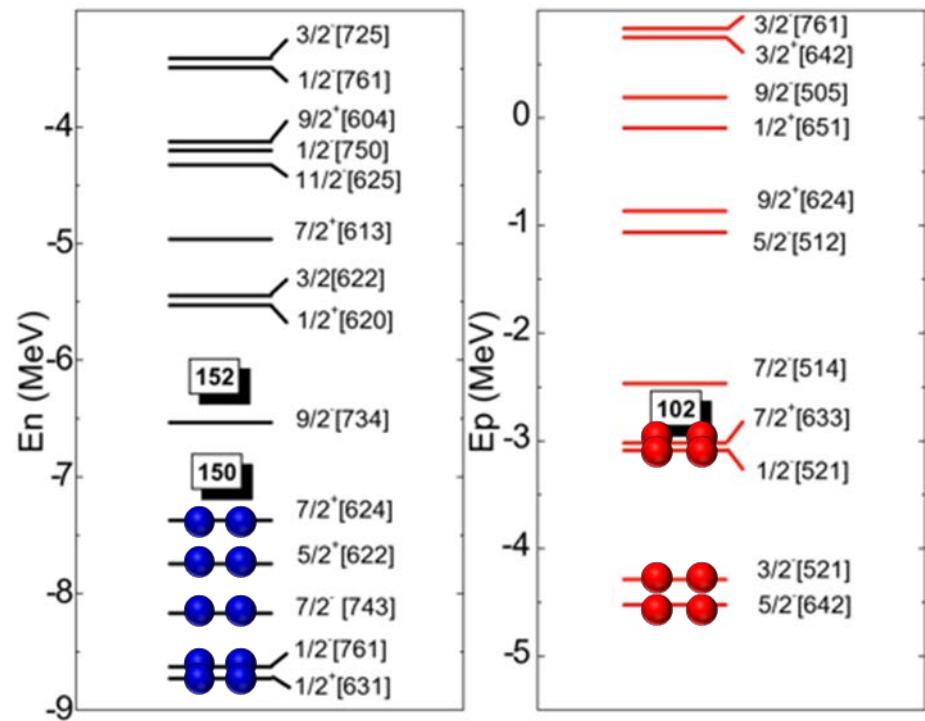
Why K-isomers occur?

- deformed nuclei
- breaking of particle pair at Fermi surface
- selection rule for electromagnetic transitions
 $\lambda \geq \Delta K$ is not fulfilled
- excitation energy of quasi-particle:
$$E = \sqrt{(\varepsilon - \lambda)^2 + \Delta^2}$$



What we can learn?

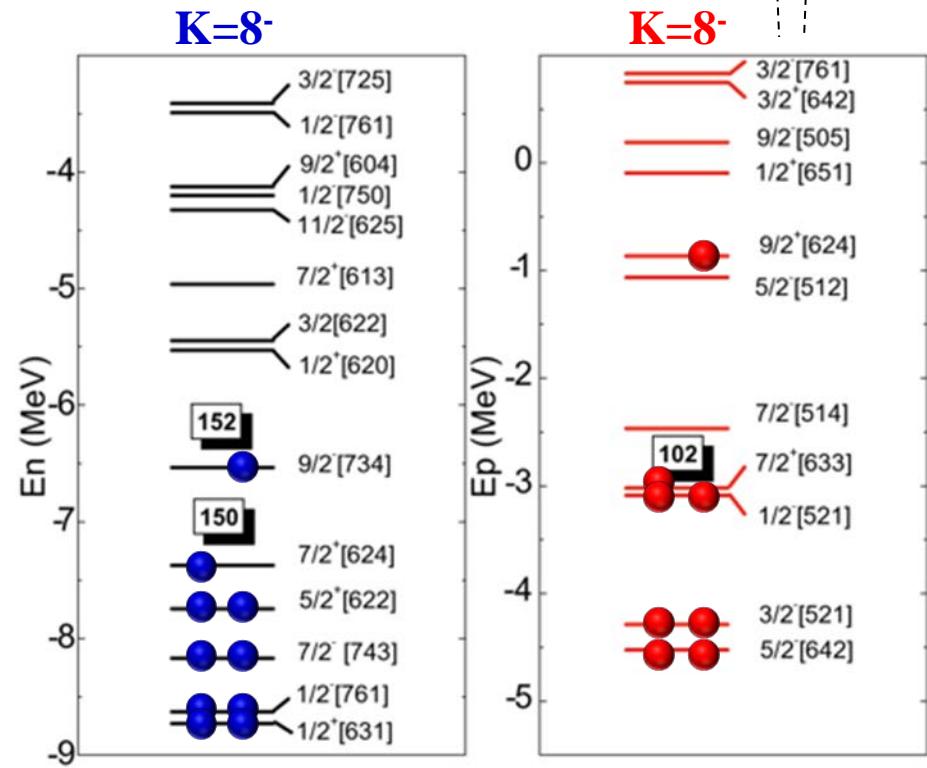
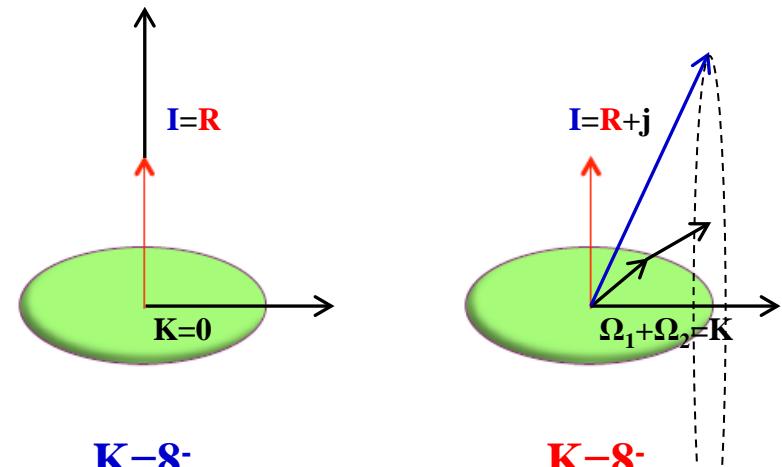
- information about Nilsson level energy gaps
- influence on stability of superheavy elements
- pairing interaction



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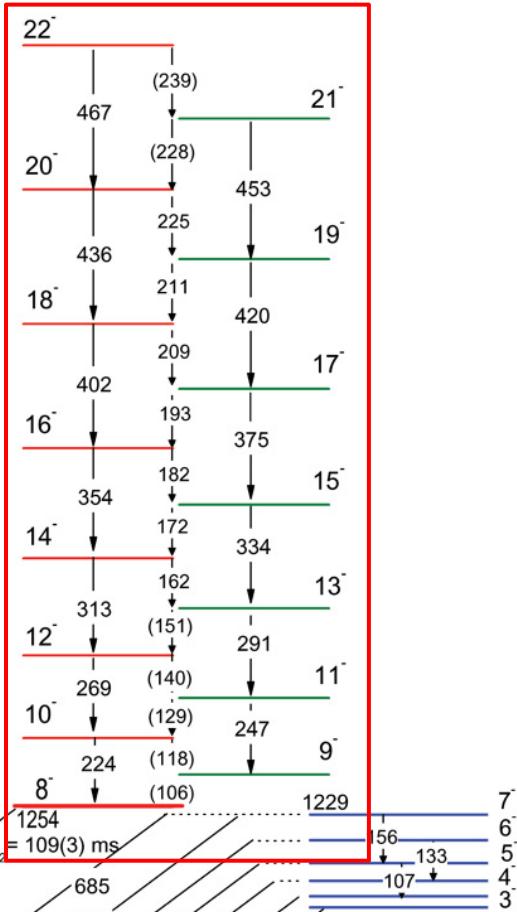
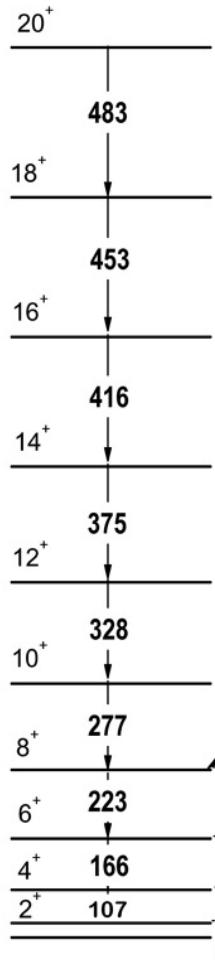
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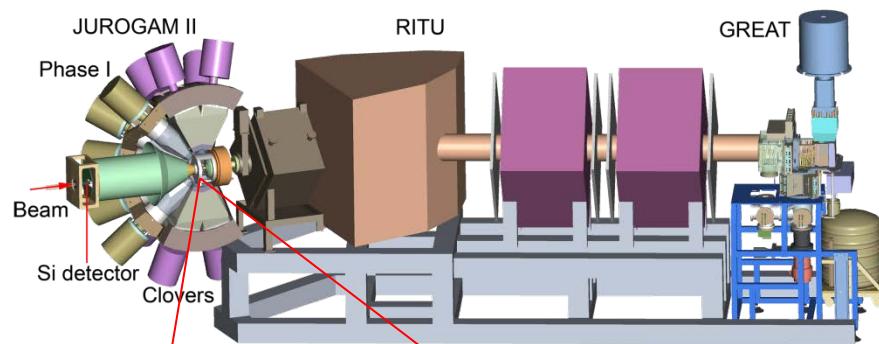
K-isomers in ^{252}No

$K=8^-$

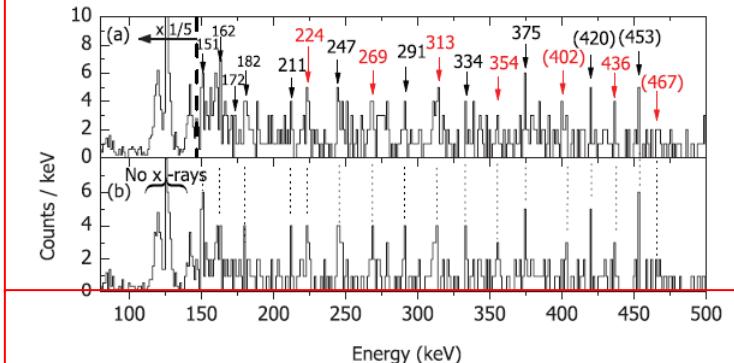
$\nu[734] 9/2^- \otimes \nu[624] 7/2^+$



$K=2^-$



$K=8^-$



B. Sulignano et al; Phys. Rev. C 86, 044318 (2012)

Neutron single particle states in ^{252}No

$7/2^+[624]$ Nilsson configuration $\text{i}_{11/2}$

$9/2^-[734]$ Nilsson configuration $\text{j}_{15/2}$

$$g(j) = \begin{cases} \frac{2\ell \cdot g_\ell + g_s}{2\ell + 1} & \text{for } j = \ell + 1/2 \\ \frac{2(\ell + 1) \cdot g_\ell - g_s}{2\ell + 1} & \text{for } j = \ell - 1/2 \end{cases}$$

proton: $g_\ell = 1$ $g_s = 5.59$

neutron: $g_\ell = 0$ $g_s = -3.83$

$$g(\text{i}_{11/2}) = +0.295 \quad g(\text{j}_{15/2}) = -0.255$$

$$g(j_1 \times j_2; J) = \frac{1}{2} \cdot (g_1 + g_2) + \frac{j_1(j_1 + 1) - j_2(j_2 + 1)}{2J(J + 1)} \cdot (g_1 - g_2)$$

$$g(\text{i}_{11/2} \times \text{j}_{15/2}; 8) = 0.09$$

$$B(M1; I \rightarrow I - 1) = \frac{3}{4\pi} [g_K - g_R]^2 \cdot K^2 |\langle IK10 | (I - 1)K \rangle|^2$$

$$B(E2; I \rightarrow I - 2) = \frac{5}{16\pi} Q_0^2 |\langle IK20 | (I - 2)K \rangle|^2$$

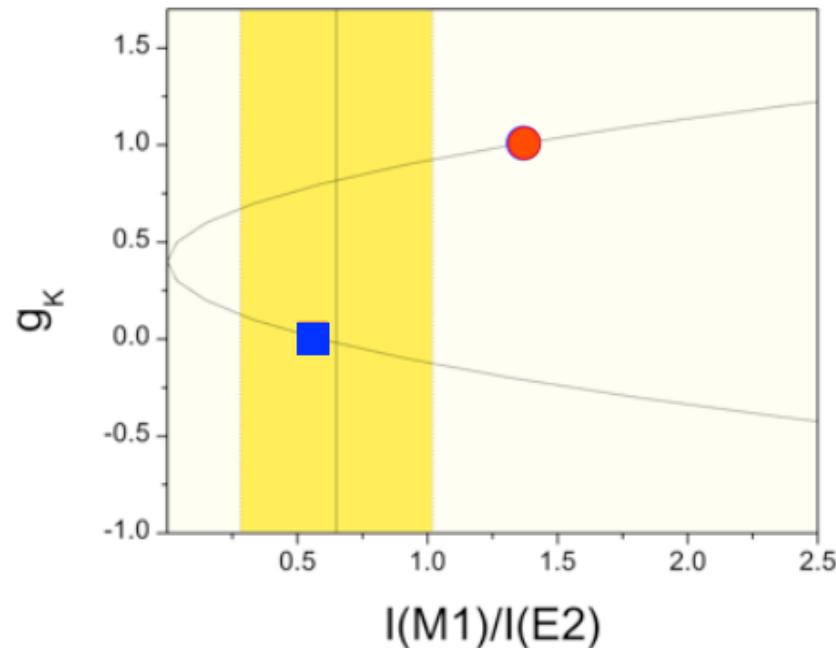
$$g_R = Z/A = 0.40$$

$$g_K \pi\pi(\text{th.}) \\ 1.001$$

Branching ratio

$$R_{\text{exp}} = I_\gamma(J \rightarrow J - 1) / I_\gamma(J \rightarrow J - 2)$$

$$R_{\text{th}} = \frac{T(M1)}{T(E2)} = \frac{1.76 [E_\gamma(M1)]^3 B(M1) s^{-1}}{1.22 [E_\gamma(E2)]^5 B(E2) s^{-1}}; \propto \frac{(g_K - g_R)^2}{Q_0^2}$$



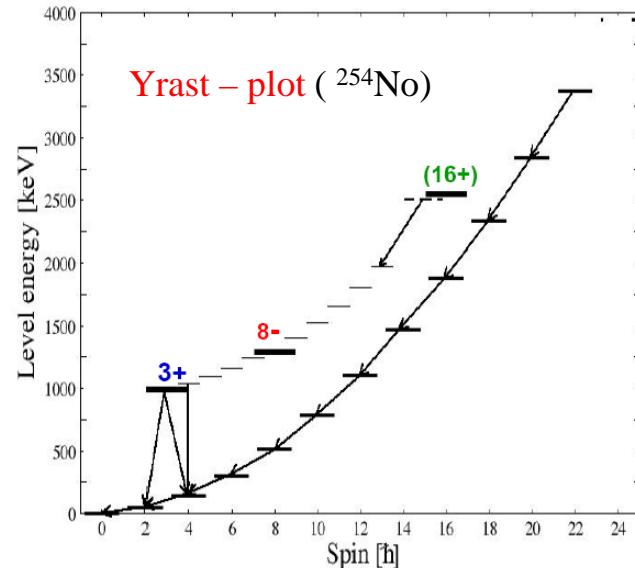
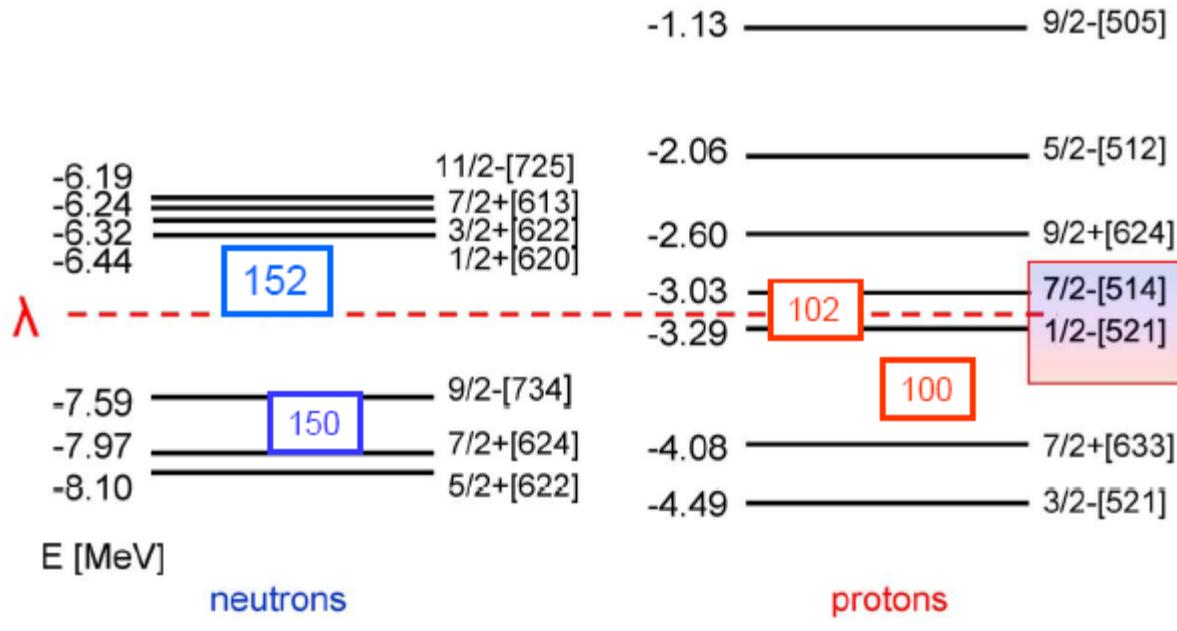
The isomer $K=8^-$ is based on 2-qn excitation

K-isomeric states

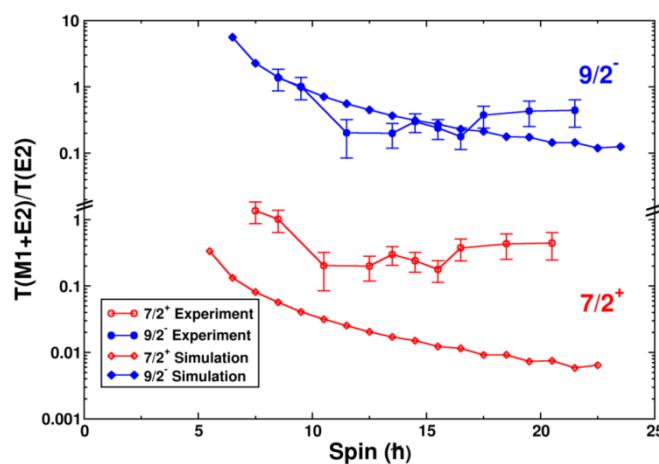
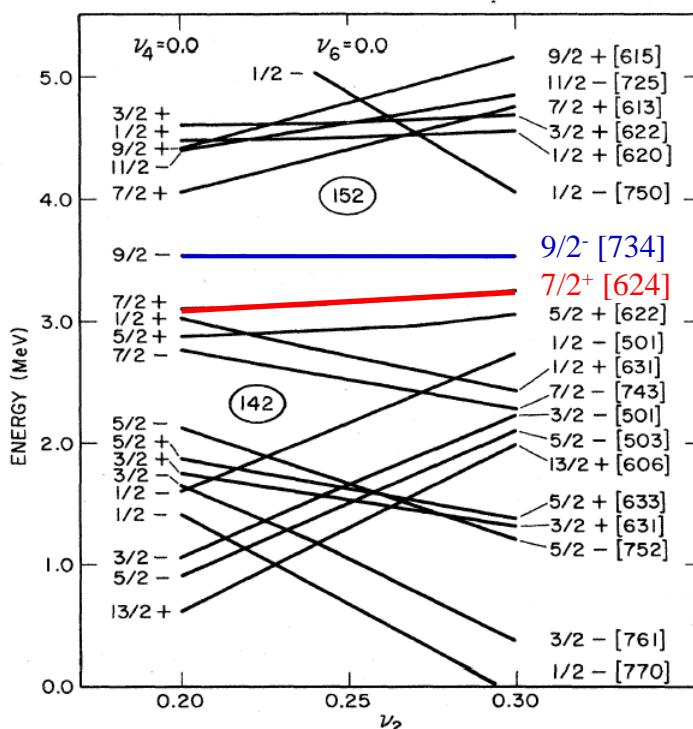
Exp. results: excitation of isomeric states

^{254}No with $Z=102$ and $N=152$ – protons will be excited easily

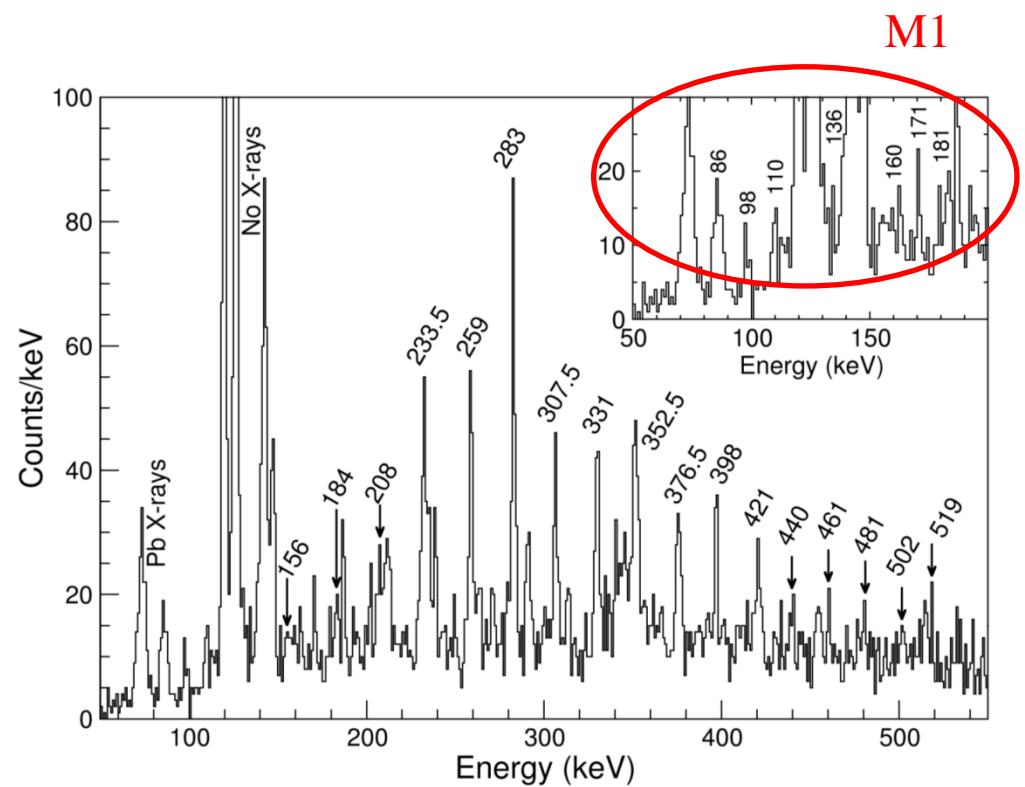
^{250}Fm with $Z=100$ and $N=150$ – neutrons will be excited easily



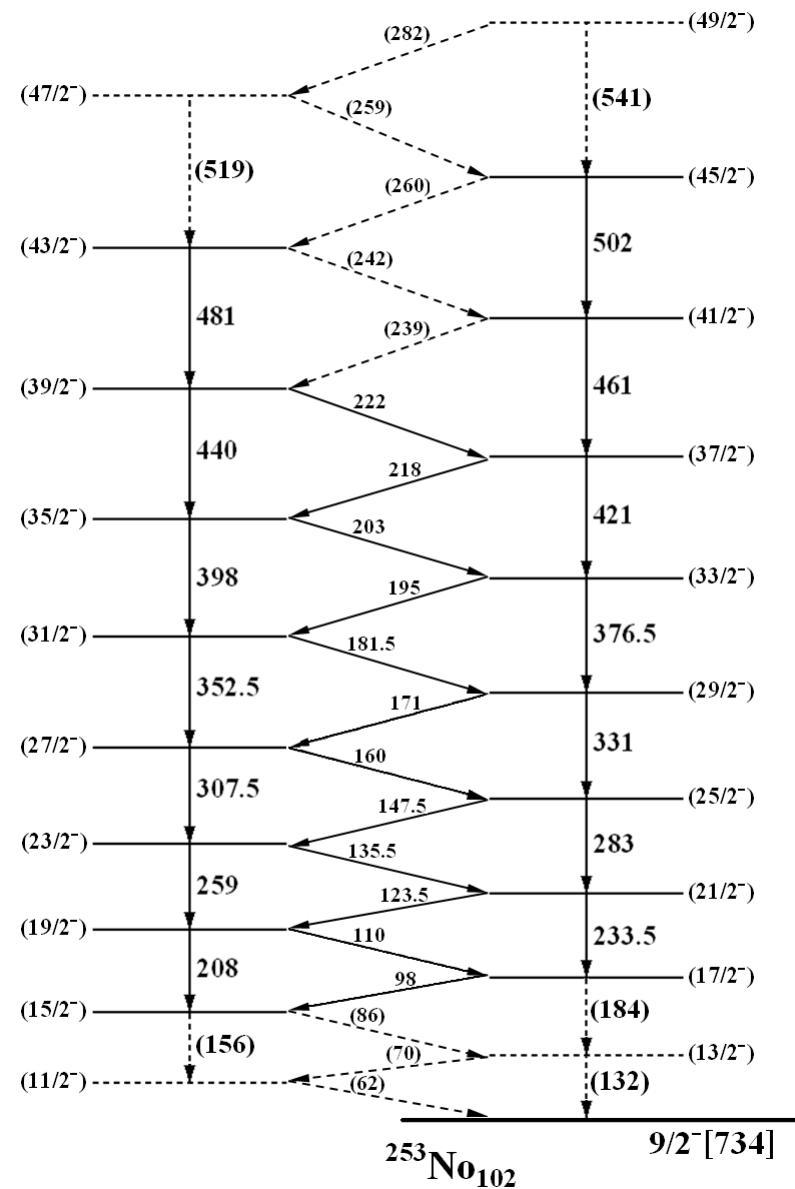
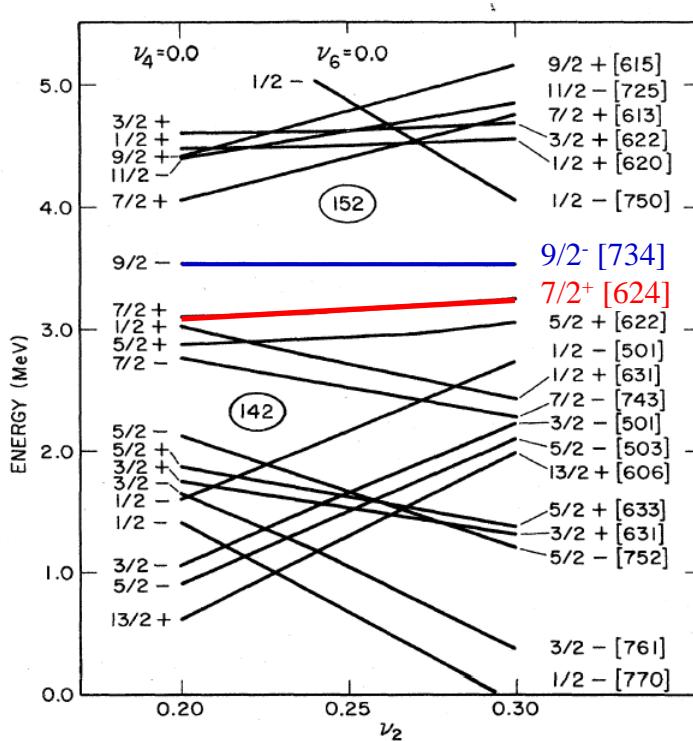
Level scheme of ^{253}No (151 neutrons)



- ❖ ground state $9/2^-$
- ❖ excited state $7/2^+$
- ❖ rotational band observed at Gammasphere & JUROGAM

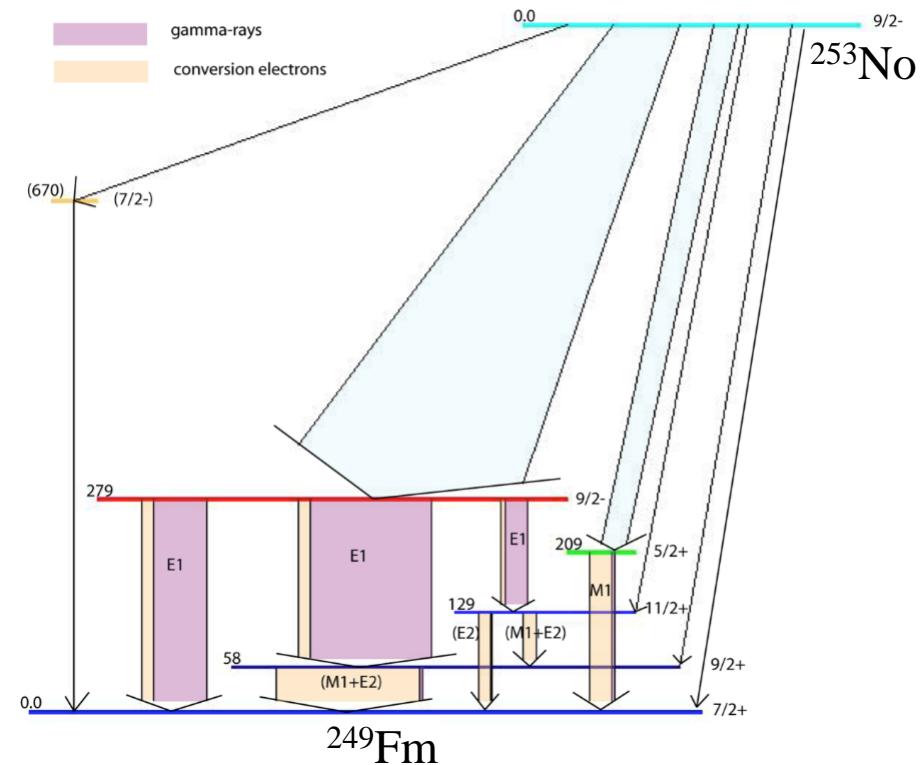


Level scheme of ^{253}No (151 neutrons)



R. Chasman et al.; Rev. Mod. Phys. 49 (1977) 833

Alpha decay of ^{253}No



In general, α -decay in even-even parents lead to the ground state of the daughter nucleus so that the emitted particle carries away as much energy as possible and as little angular momentum as possible.

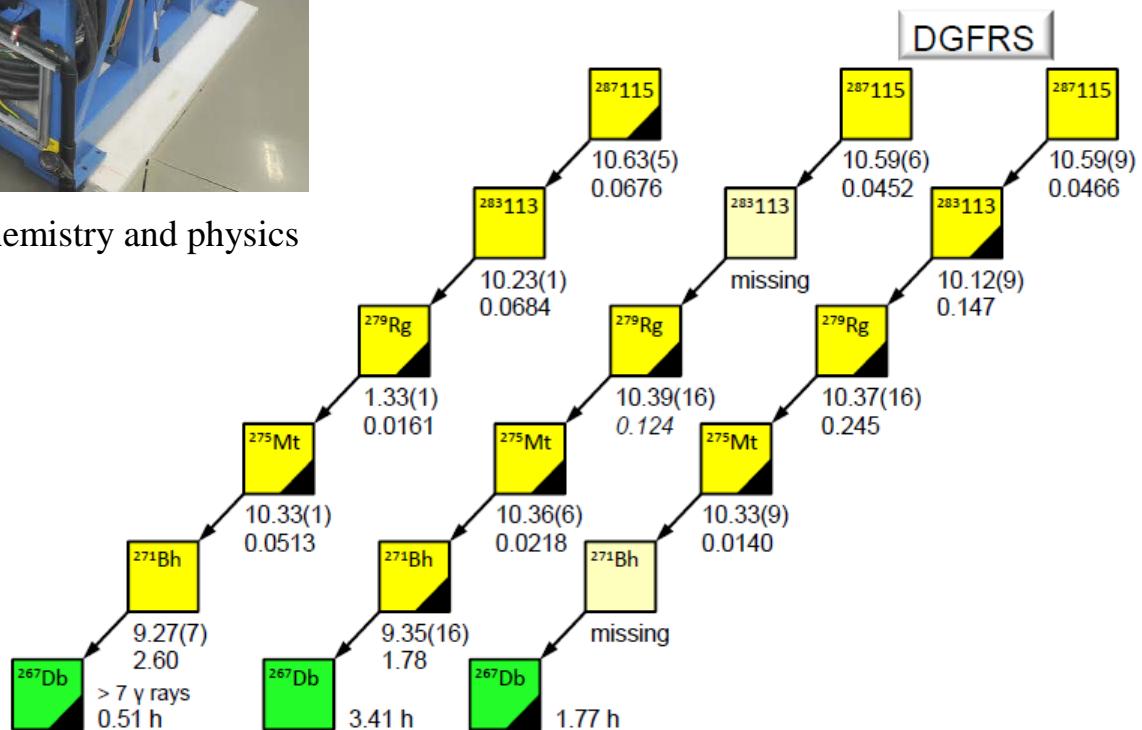
α -decays of odd-A heavy nuclei populate predominantly low-lying excited states that ***match the spin of the parent*** so that the orbital angular momentum of the α -particle can be zero.

Spectroscopy of element 115



Recoil separator for superheavy element chemistry and physics

1 chain (out of 30) is compatible with
2 chains (out of 37) associated with the
4n channel $^{287}_{115}Mc$ by Oganessian *et al.*



Dubna Gas Filled Recoil Separator

Spectroscopy of element 115



^{243}Am target wheel

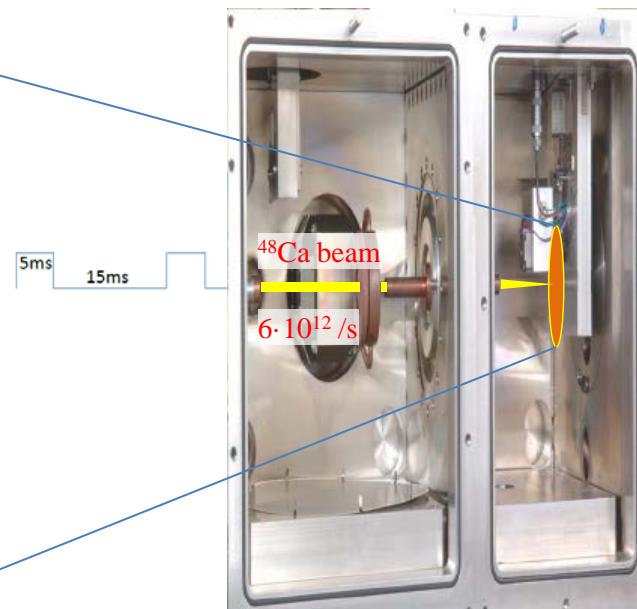
0.83 mg/cm²

20 mg, $5 \cdot 10^{19}$

> 150 MBq α, β, γ



target chamber side view



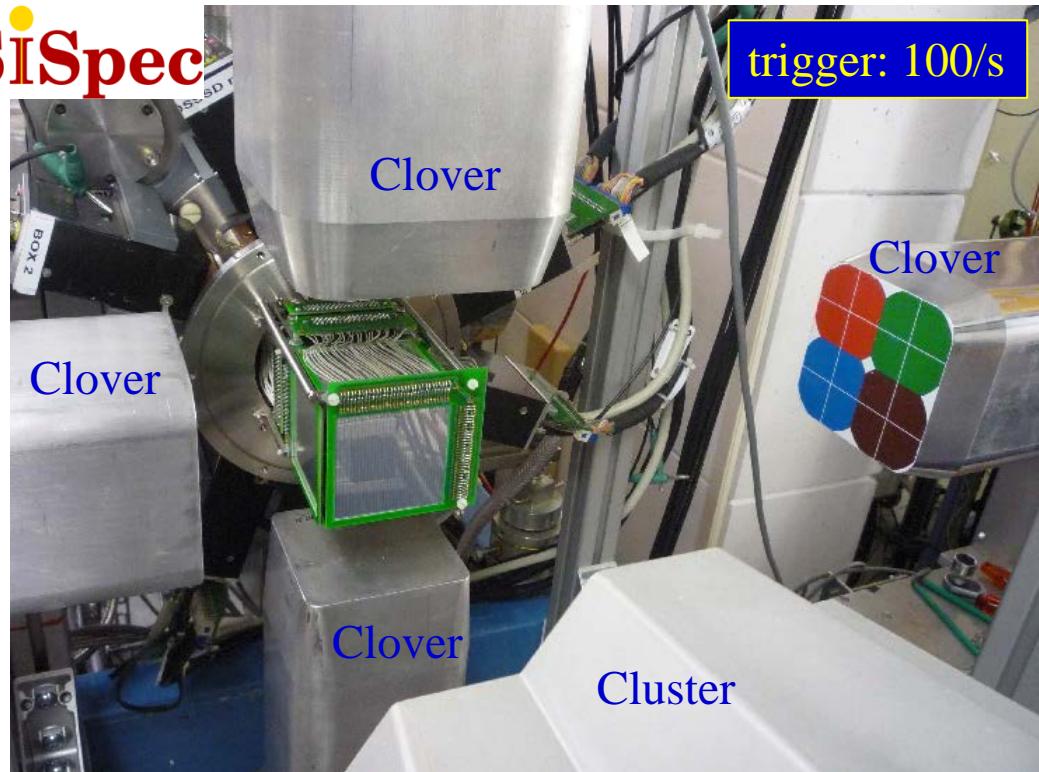
Spectroscopy of element 115

TASiSpec

Highly efficient multi-coincidence spectroscopy set-up for TASCA's very compact focal plane image

1 Implantation DSSSD (1024 pixels)
4 box-DSSSDs (1024 pixels)
 $\rightarrow \sim 80\%$ α -detection efficiency

4 Ge Clover (4·4 crystals)
1 Ge Cluster (7 crystals)
 $\rightarrow \sim 40\%$ γ -detection eff. at 150 keV



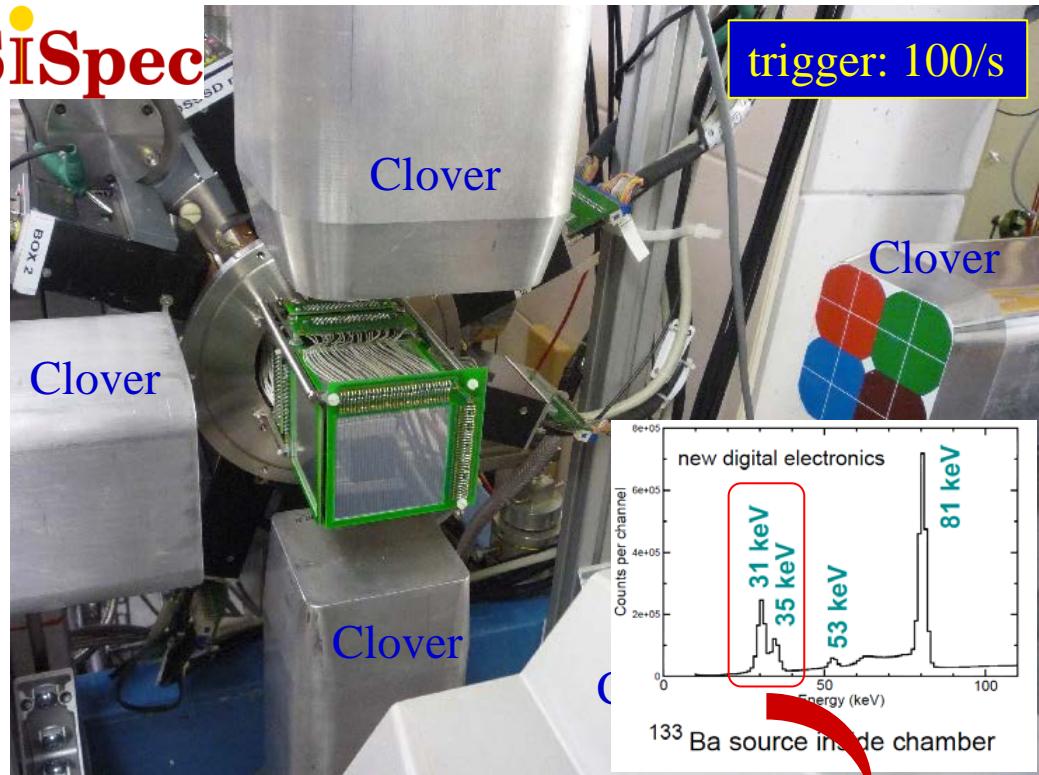
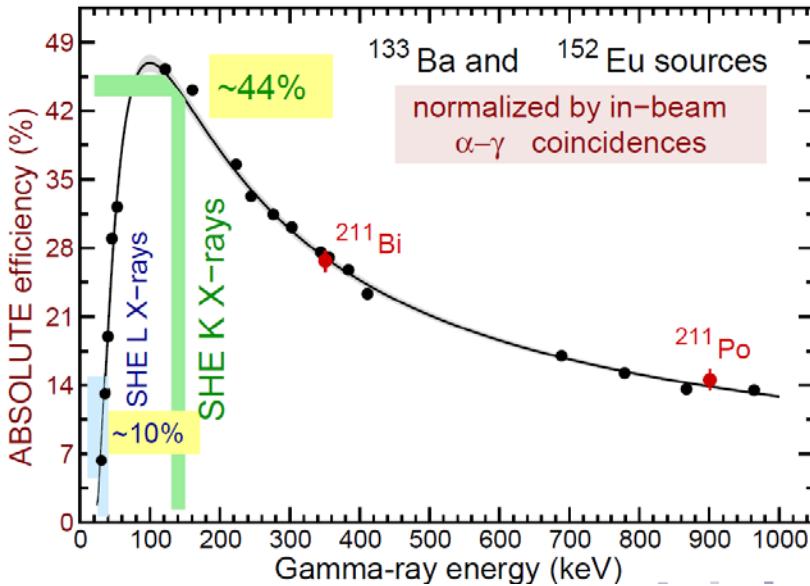
L.-L. Andersson et al.; NIM A622, 164 (2010)
L. G. Sarmiento et al.; NIM A667, 26 (2011)

TAsca Small Image mode SPECtroscopy

TASiSpec

Highly efficient multi-coincidence spectroscopy set-up for TASCA's very compact focal plane image

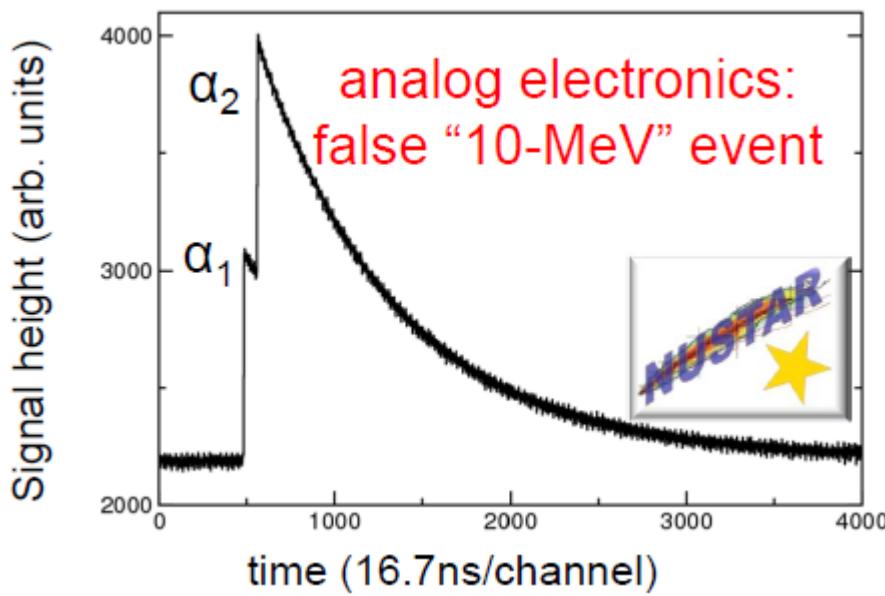
1 Implantation DSSSD (1024 pixels)
4 box-DSSSDs (1024 pixels)
 $\rightarrow \sim 80\%$ α -detection efficiency



Digital (or Sampling) electronics

96 DSSSD p-sides

60 MHz dead-time free sampling ADC
“FEBEX” cards developed at GSI-EE

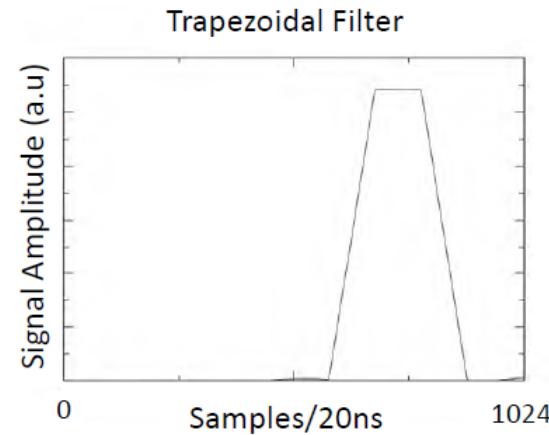


- detect summing, reduce background
- software optimization (MWD)
towards best possible resolution
- large dynamic range (linear within
0.1 – 100 MeV, time-over threshold)

25 Ge crystals

100 MHz commercial sampling ADCs,
4x SIS3302 cards, FPGA processed:

- flat-top energy
- baseline
- pile-up flagging



That allows to ...

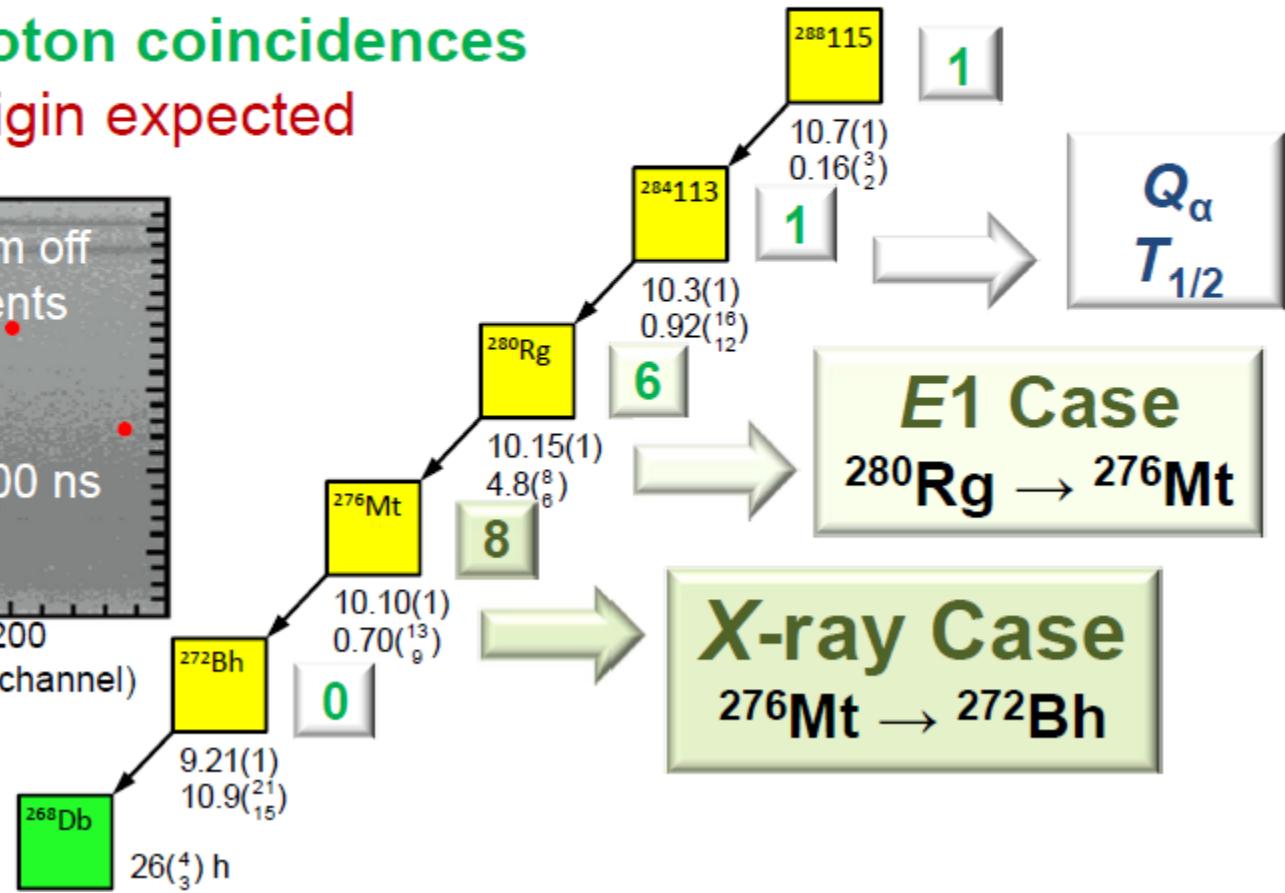
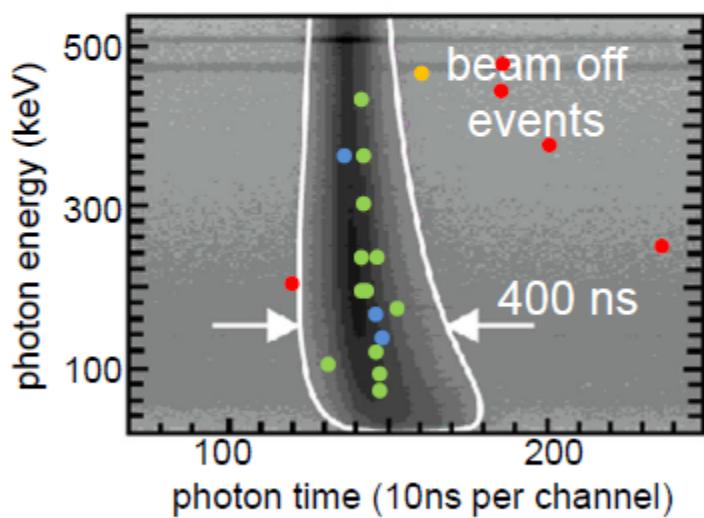
- restore baseline in software
- retain (almost) nominal Ge-detector
energy resolution

... at high counting rates.

Results – ^{288}Mc (3n – channel)

22 chains (out of 30) of ours are compatible with the
31 chains (out of 37) associated with the 3n channel $^{288}\text{Mc} \rightarrow ^{284}\text{Rg} \rightarrow ^{276}\text{Mt} \rightarrow ^{272}\text{Bh}$
by Oganessian *et al.*

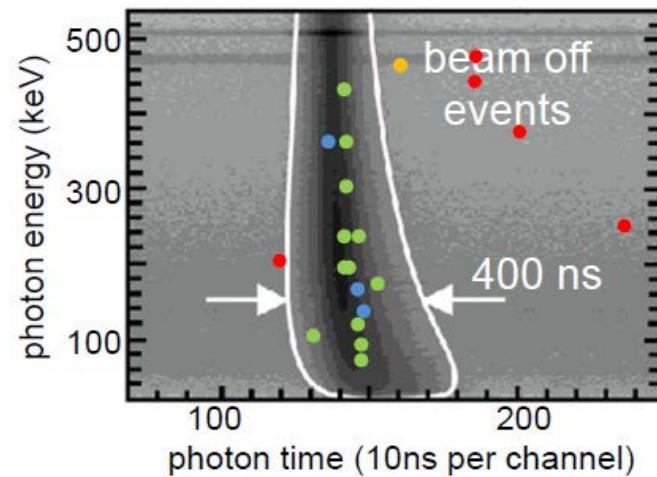
16 prompt α -photon coincidences
2-3 of random origin expected



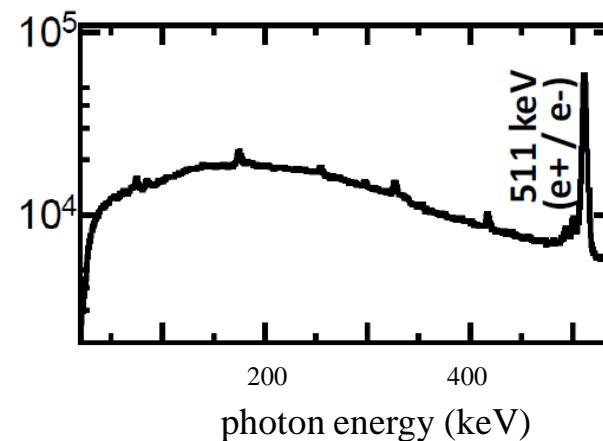
α – photon coincidences

Correlation time between implantation DSSSD detector and any Ge detector.

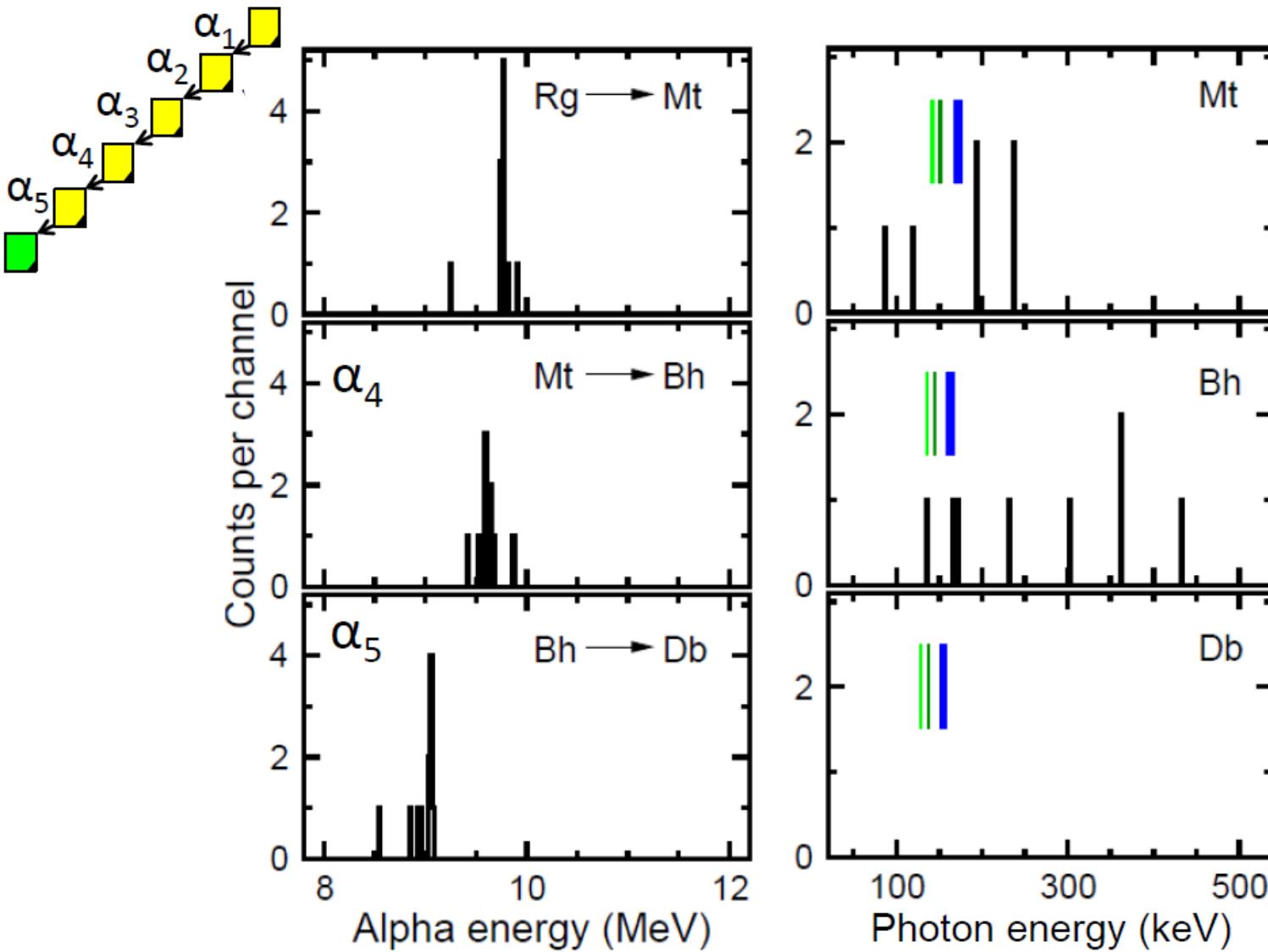
White line marks the gate for prompt coincidences.



Total projection of Ge-detector events in prompt coincidence with events in the implantation DSSSD, both accumulated during beam-off periods.



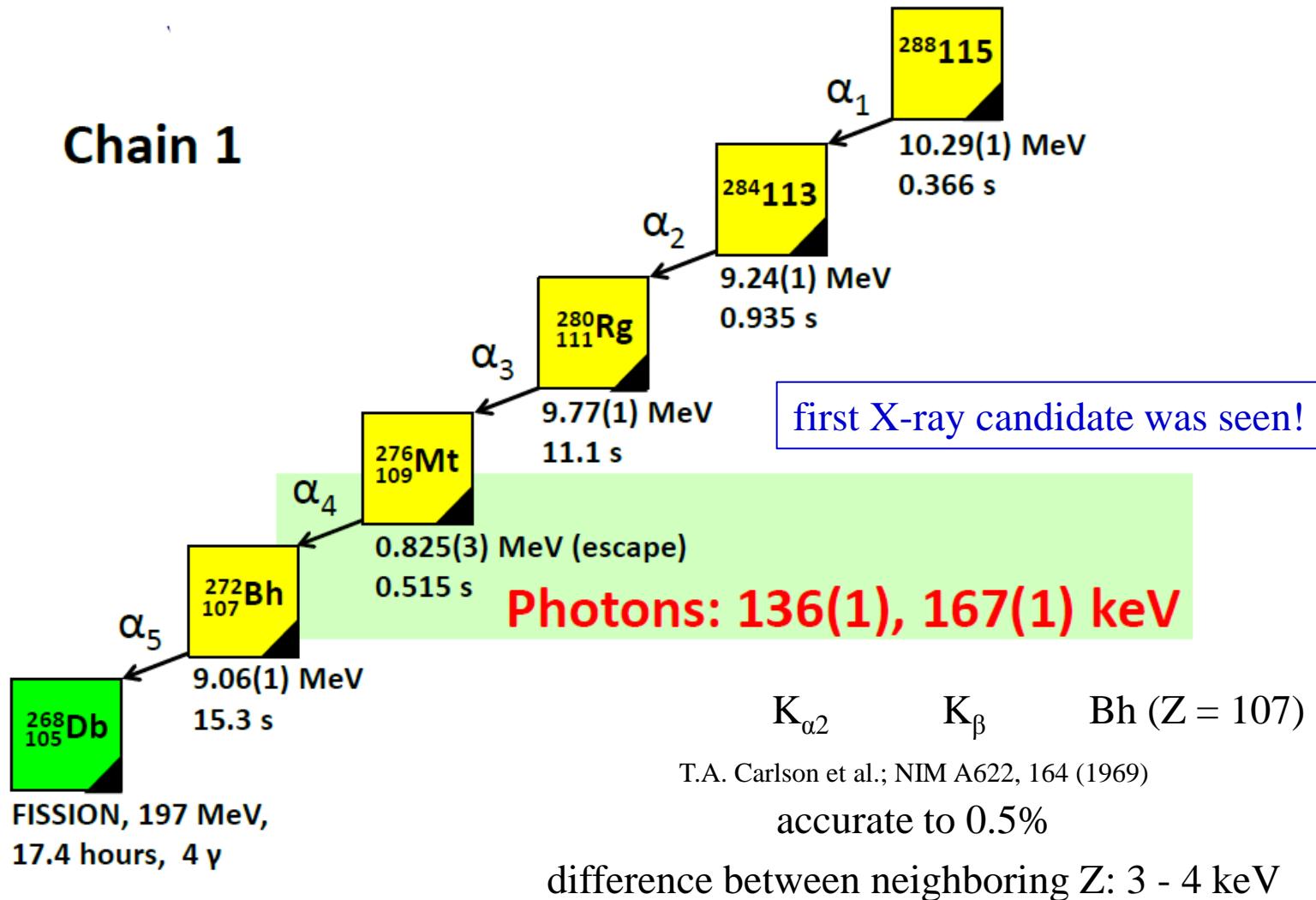
Identification of Z



X-ray
fingerprinting

Characteristic X-rays

Chain 1



Chemistry of superheavy elements

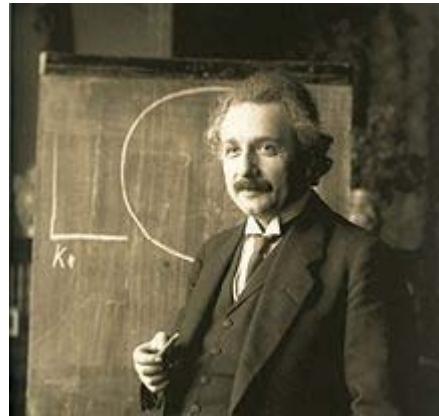
Group → 1 ↓ Period	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18		
1	H															He			
2	Li	Be										B	C	N	O	F	Ne		
3	Na	Mg										Al	Si	P	S	Cl	Ar		
4	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr	
5	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe	
6	Cs	Ba	La	*	72	73	74	75	76	77	78	79	80	81	82	83	At	Rn	
7	Fr	Ra	Ac	*	104	105	106	107	108	109	110	111	112	113	114	115	116	117	118
				*	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Og	
				*	58	59	60	61	62	63	64	65	66	67	68	69	70	71	
				*	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
				*	90	91	92	93	94	95	96	97	98	99	100	101	102	103	
					Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	

- Are the new elements in the same period?
- Does e.g. Lv show the same chemical properties as O, S, Se, Te and Po?

Chemistry of superheavy elements

relativistic effect: important for large Z

$$E = mc^2$$



- ❖ High atomic number: strong Coulomb attraction causes electrons to move faster.
- ❖ Causes relativistic mass increase $m = m_0(1 - \beta^2)^{-1/2}$, with $\beta = v/c$; and as a consequence, contraction of spherical orbitals (ns, np_{1/2})
- ❖ The s and p_{1/2} atomic orbitals contract relativistically.
- ❖ The shrinking of the inner shells results in an increased screening of the nuclear charge, and this gives rise to an expansion of the p_{3/2} and of higher angular momentum orbitals.
- ❖ Strong spin-orbit splitting

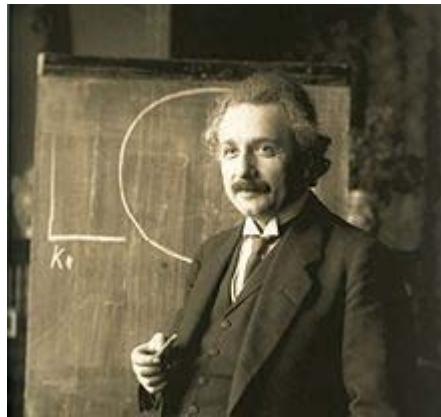
Bohr model: $E = -(2\pi^2 e^4 / n^2 h^2) \cdot m \cdot Z^2$ $r = Ze^2 / m \cdot v^2$ $v = (2\pi e^2 / n \cdot h) \cdot Z$

for hydrogen, m/m₀ = 1.0000027, for element Z=114 , m/m₀ = 1.79, for element Z=118, m/m₀ = 1.95

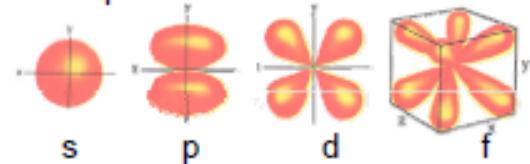
Chemistry of superheavy elements

relativistic effect: important for large Z

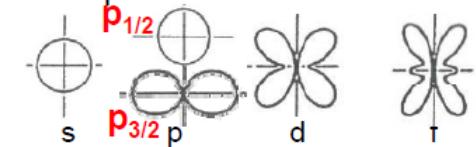
$$E = mc^2$$



Examples of **non-relativistic** orbitals



Examples of **relativistic** orbitals



Solution of the Dirac equation (relativistic quantum mechanics) for a hydrogen-like atom:

$$E_{1s} = mc^2 \sqrt{1 - (Z\alpha)^2} \sim mc^2 \cdot \left(1 - \frac{(Z\alpha)^2}{2} - \frac{(Z\alpha)^4}{8} + \dots \right)$$

relativistic effect

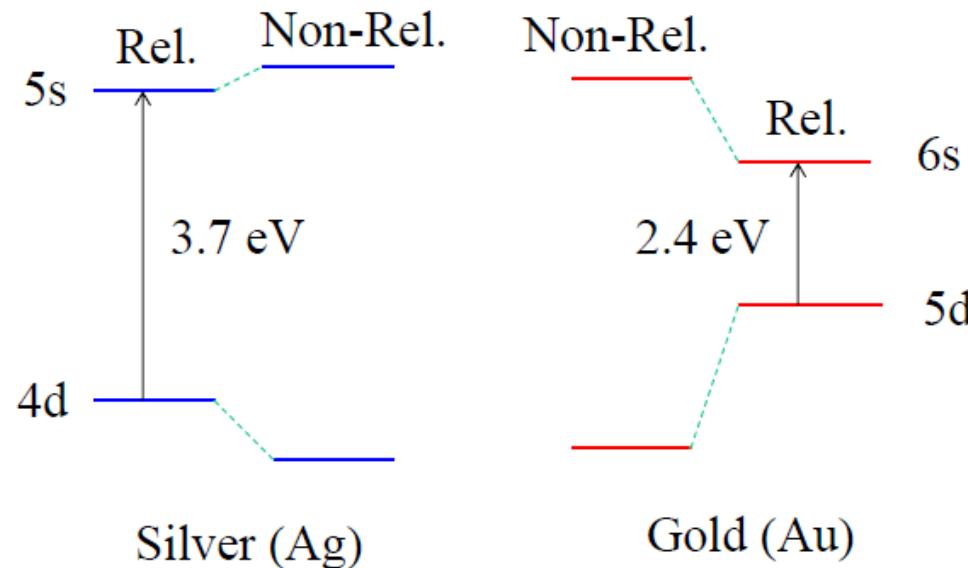
Famous example of relativistic effects: the color of gold

1	1 H																2 He		
2	3 Li	4 Be																	
3	11 Na	12 Mg																	
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr	
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe	
6	55 Cs	56 Ba	57 La	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	89 Ac	*	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og

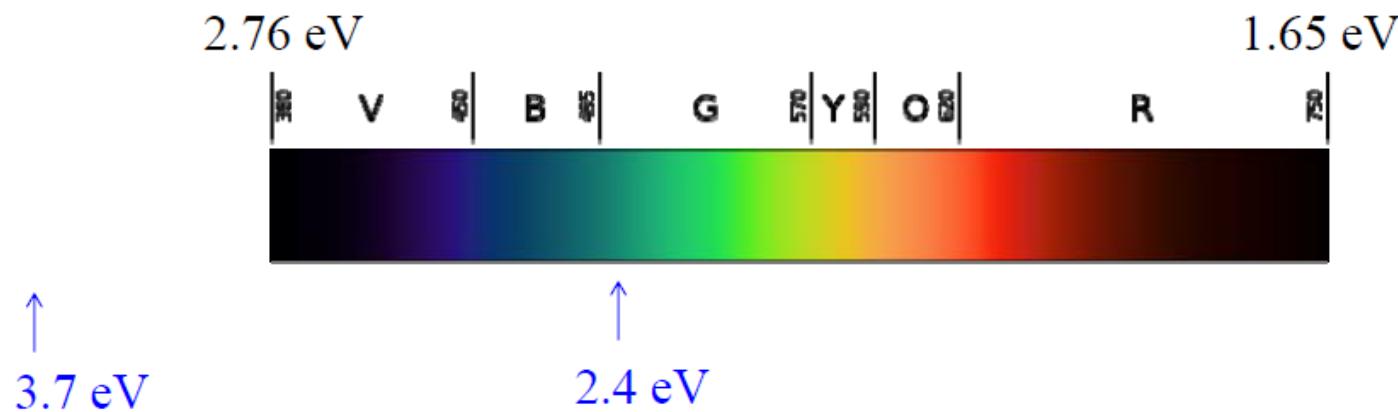


Gold looked like silver if there was no relativistic effect!

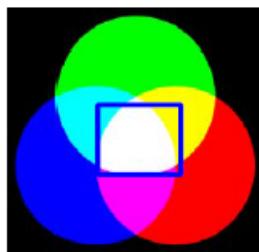
Famous example of relativistic effects: the color of gold



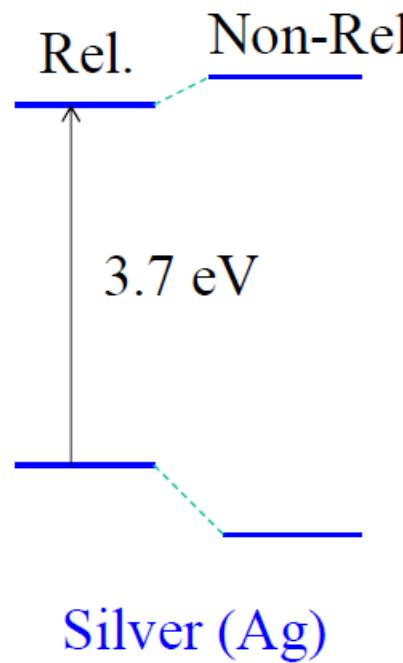
cf. visible spectrum



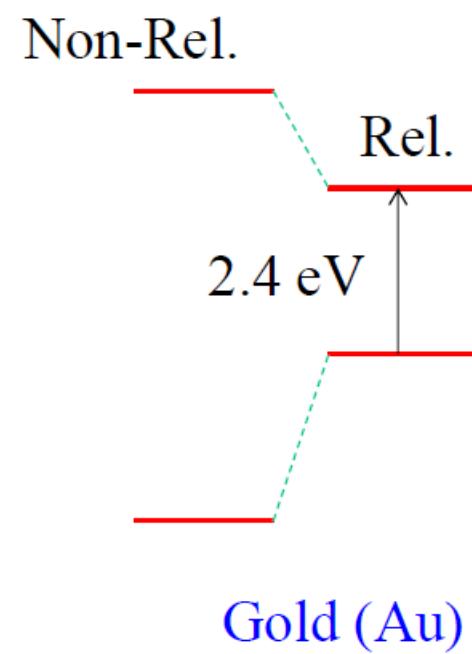
Famous example of relativistic effects: the color of gold



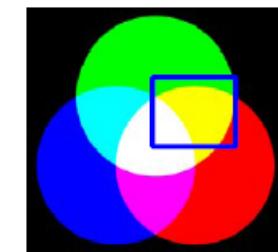
no color
absorbed



Silver (Ag)



Gold (Au)



blue: absorbed



Ag



Au

Chemistry of superheavy elements

Group→1 ↓Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18				
	1 H																2 He					
1	3 Li	4 Be															5 B	6 C	7 N	8 O	9 F	10 Ne
2	11 Na	12 Mg															13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
3	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr				
4	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe				
5	55 Cs	56 Ba	57 La	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn			
6	87 Fr	88 Ra	89 Ac	*	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og			
7				*	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu				
				*	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr				

- ❖ Gold is smaller than Silver
- ❖ Roentgenium is of the same size as Copper

Chemistry of superheavy elements

Group→1 ↓Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	
1	1 H																2 He		
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne	
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar	
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr	
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe	
6	55 Cs	56 Ba	57 La	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	89 Ac	*	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og
	*	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu				
	*	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr				

How do the relativistic effects alter the periodic table for SHE?

→ a big open question

Chemistry of superheavy elements

Group → 1 ↓ Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	
	1 H															2 He			
1	3 Li	4 Be																	
2	11 Na	12 Mg														5 B	6 C	7 N	
3	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr	
4	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe	
5	55 Cs	56 Ba	57 La	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
6	87 Fr	88 Ra	89 Ac	*	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og
7	*			*	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	
	*			*	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr	

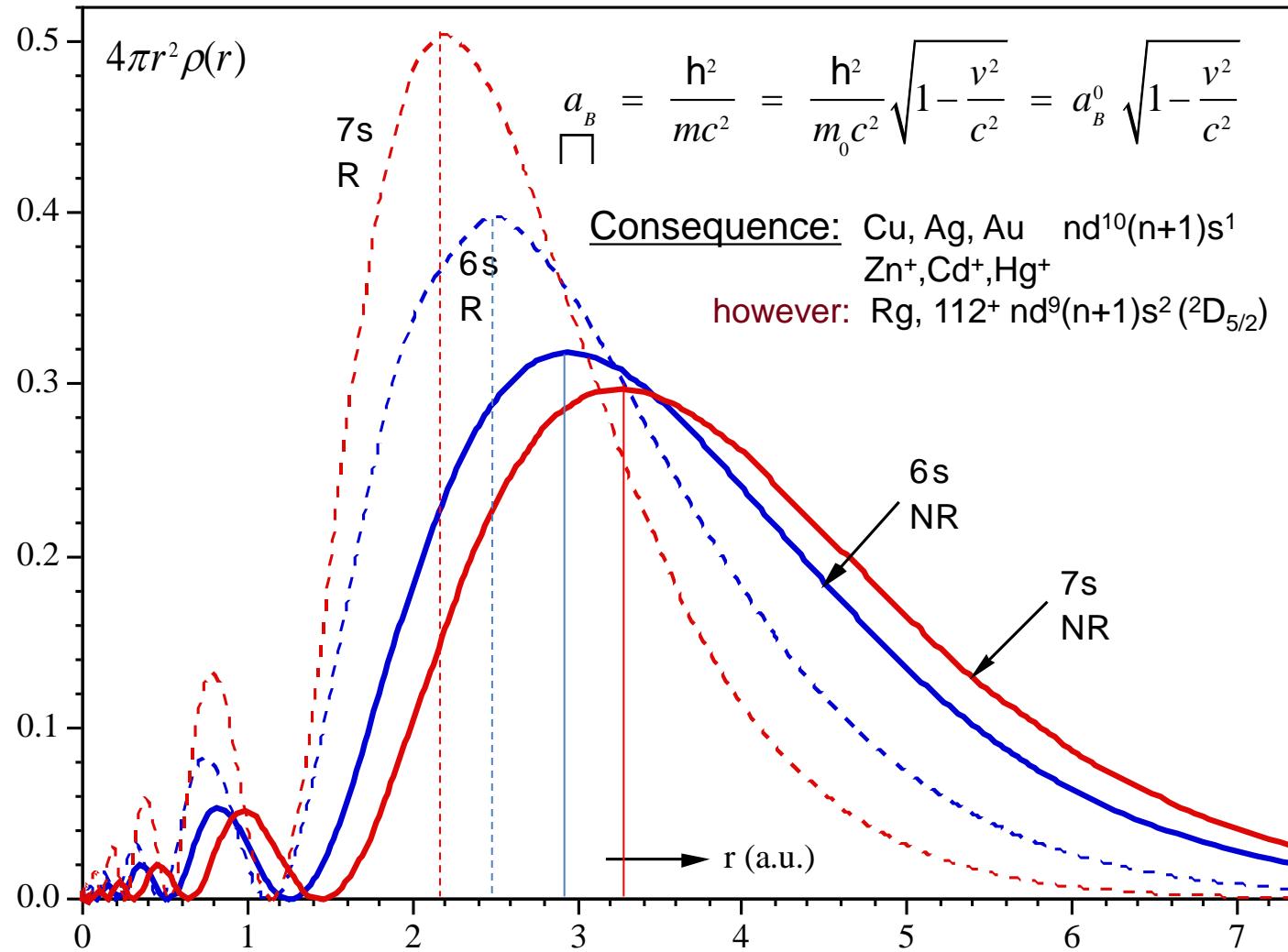
- ❖ Dubnium does not behave like Tantalum (1993)
- ❖ Sg (1997) Bh (2000) and Hs (2001) confirmed relativistic calculations predicting the expected behavior in the periodic table.
- ❖ Hassium, for instance, forms a gaseous oxide similar to Osmium.

The atomic orbitals $s_{1/2}$ and $p_{1/2}$ are contracted relativistically. The shrinking of the inner shells results in an increased *screening of the nuclear charge*, and this gives rise to an expansion of the $p_{3/2}$ and of higher angular momentum orbitals. Another relativistic effect is a *change in the spin-orbit coupling*. Both can produce drastic rearrangements of orbital levels. That is what is predicted to happen for Cn. Recent calculations indicate that the 7s orbital should be shifted below the $6d_{5/2}$ orbital due to relativistic effects. It is the large relativistic stabilization of its valence 7s orbital, combined with its closed shell electron configuration, that has led to the prediction that element 112 is chemically inert (not very reactive).

Chemistry of the superheavy elements with $Z > 118$ is believed to show relativistic effects that are so large that comparison with lighter elements or nonrelativistic results is meaningless.



Example: the relativistic 6s/7s contraction in Au and Rg



P. Schwerdtfeger

E.Eliav, U.Kaldor, P.Schwerdtfeger, B.Hess, Y.Ishikawa, *Phys. Rev. Lett.* 73, 3203 (1994).

M.Seth, P.Schwerdtfeger, M.Dolg, K.Faegri, B.A.Hess, U.Kaldor, *Chem. Phys. Lett.* 250, 461 (1996).

The limits of the periodic table

❖ Can this go forever? NO!!!

The [relativistic Dirac equation](#) gives the ground state energy as

where m_0 is the rest mass of the electron. For $Z > 137$, the wave function of the Dirac ground state is oscillatory, rather than bound.

More accurate calculations taking into account the effects of the finite size of the nucleus indicate that the binding energy first exceeds $2mc^2$ for $Z > 173$.

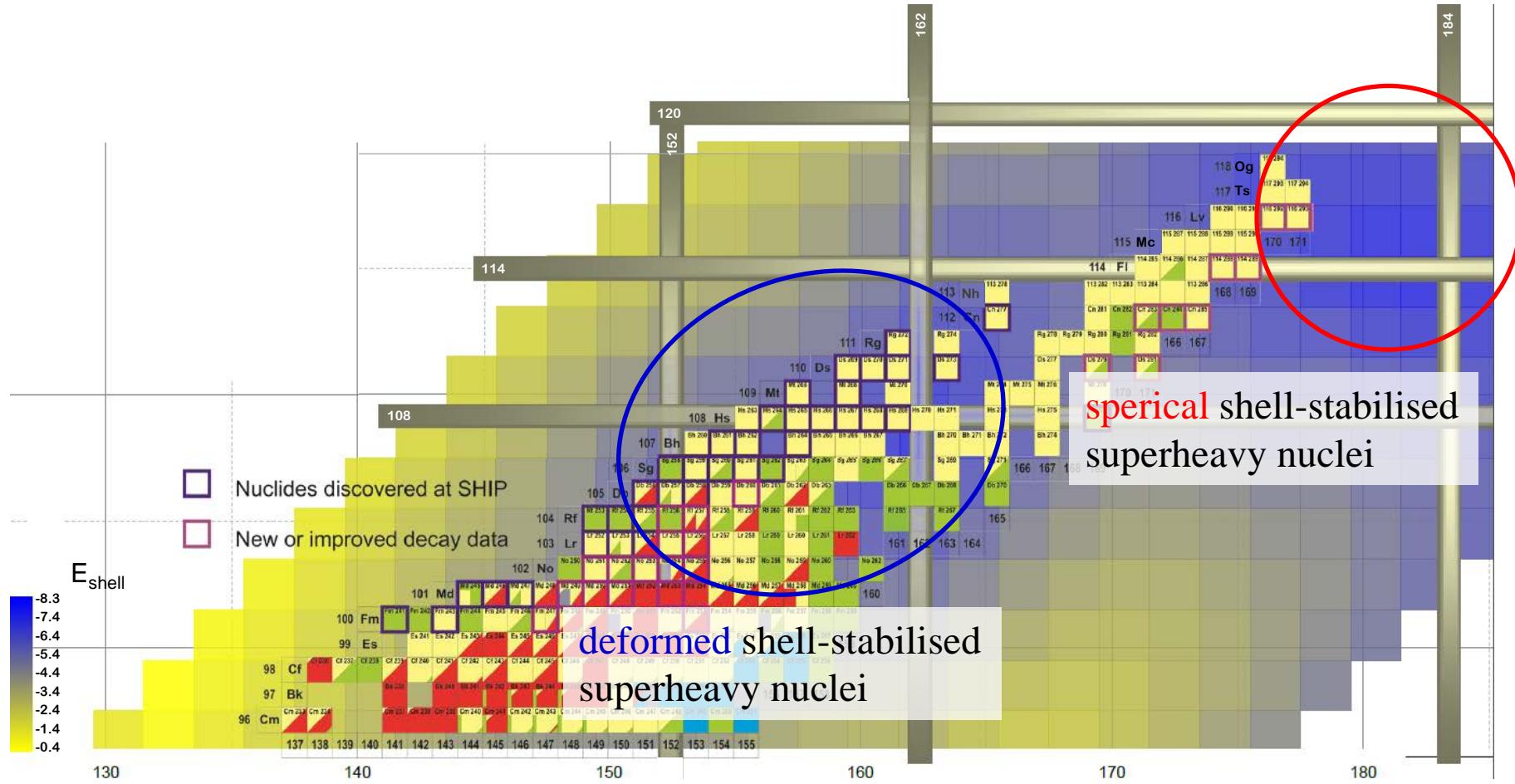
$$E = \frac{m_0 \cdot c^2}{\sqrt{1 + \frac{Z^2 \cdot \alpha^2}{n - (j + 1/2) + \sqrt{(j + 1/2)^2 - Z^2 \cdot \alpha^2}}}}$$

❖ The end of chemistry

❖ Does the periodic table has limits? Yes!!!

- At some point ($Z \sim 122$) all the electron energy levels of adjacent elements are similar so that there are no differences in their chemical behavior.

Chart of nuclides: the domain of heavy and superheavy elements



Calc.: A. Sobiczewski