The Impact of Superheavy Elements on the Chemical and Physical Sciences

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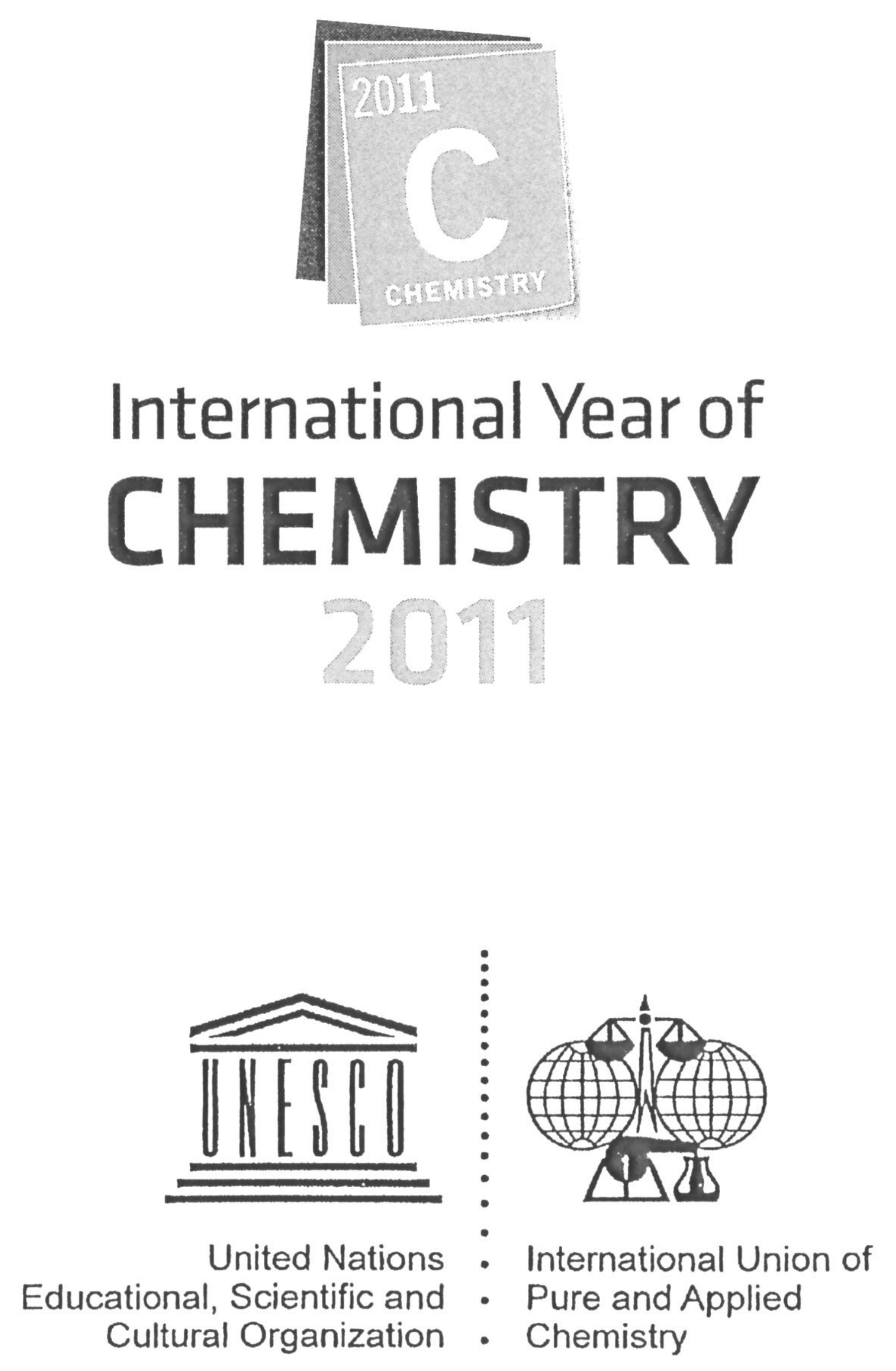
Invited talk presented at the 4th International Conference on the Chemistry and Physics of the Transactinide Elements, 5 – 11 September, 2011, Sochi, Russia

The menue:

- **The international year of chemistry 2011**
- **The Periodic Table of the elements – 1871 till today**
- **Atomic structure: Relativistic effects in the electron shells of heavy atoms**
  Relativity in the test tube
  One-atom-at-a-time chemistry
  Aqueous chemistry of rutherfordium
  Gas-phase chemistry of element 114
- **Nuclear structure: Nuclear stability and nuclear shell effects**
  Fission isomers
  The role of isomers for $Z \geq 100$
  In-beam spectroscopy and decay spectroscopy: ^{254}No
  Neutron shell closures in transuranium nuclei
- **Search for the next spherical proton shell**
  α-decay energies at $Z=82$ and at $Z=114$
  Periodicity of nuclear structure properties within the IBA and consequences
- **Summary**



International Year of CHEMISTRY 2011

United Nations
Educational, Scientific and
Cultural Organization

International Union of
Pure and Applied
Chemistry

International journal for chemical aspects of nuclear science and technology

RADIOCHIMICA ACTA

International Year of Chemistry 2011
Special Issue
Heavy Elements

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Volume 99 7-8/2011

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Radiochimica Acta – Special issue „Heavy Elements“

Y. Nagame and M. Hirata
Production and properties of transuranium elements

A. Sobiczewski
Theoretical descriptionof superheavy nuclei

S. Hofmann
Synthesis of superheavy elements by cold fusion

Yu. Oganessian
Synthesis of the heaviest elements in 48Ca-induced reactions

R.-D. Herzberg and D.M. Cox
Spectroscopy of actinide and transactinide nuclei

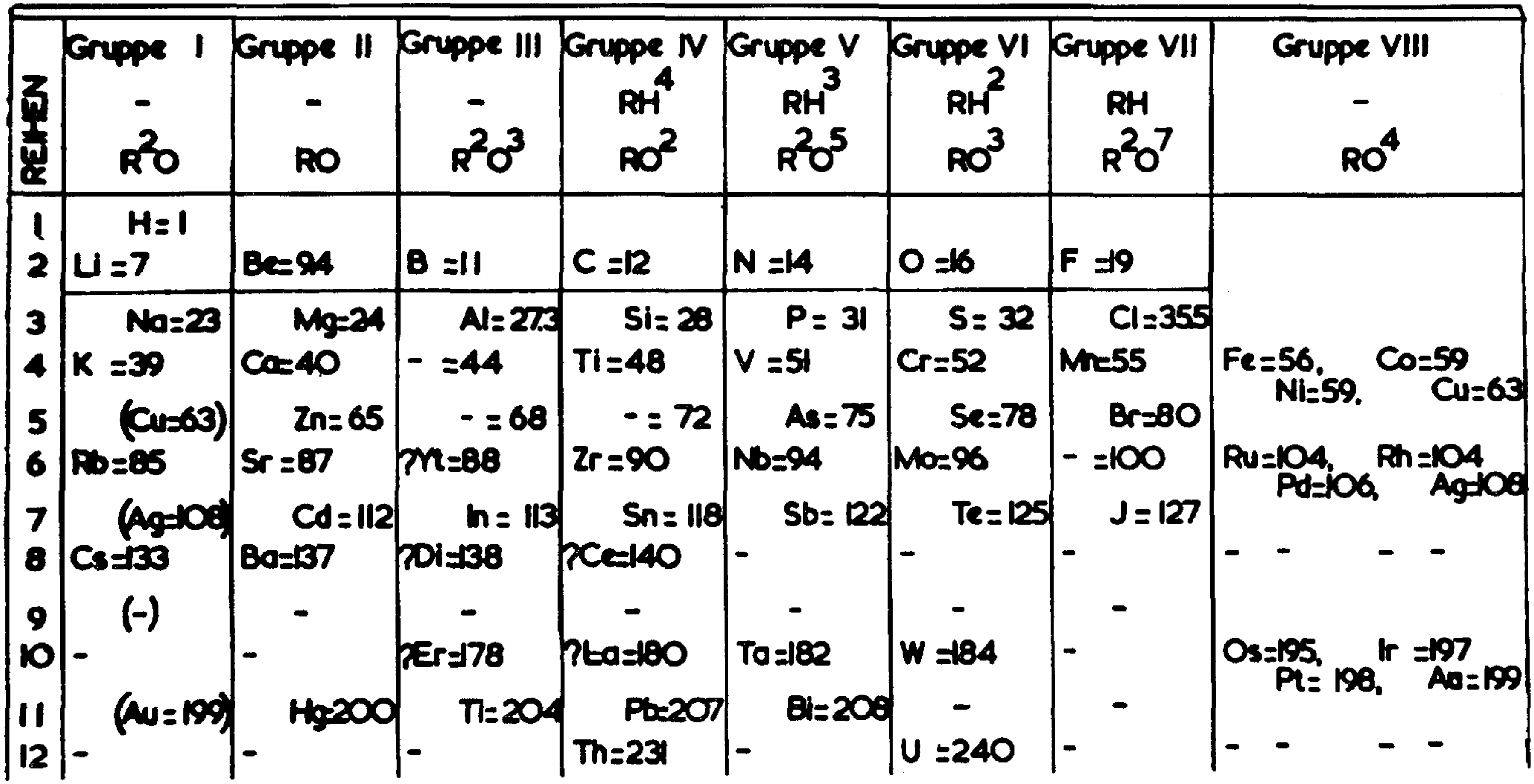
V. Pershina
Relativistic electronic structure studies on the heaviest elements

J.V. Kratz
Aqueous-phase chemistry of the transactinides

H.W. Gäggeler
Gas chemical properties of heaviest elements

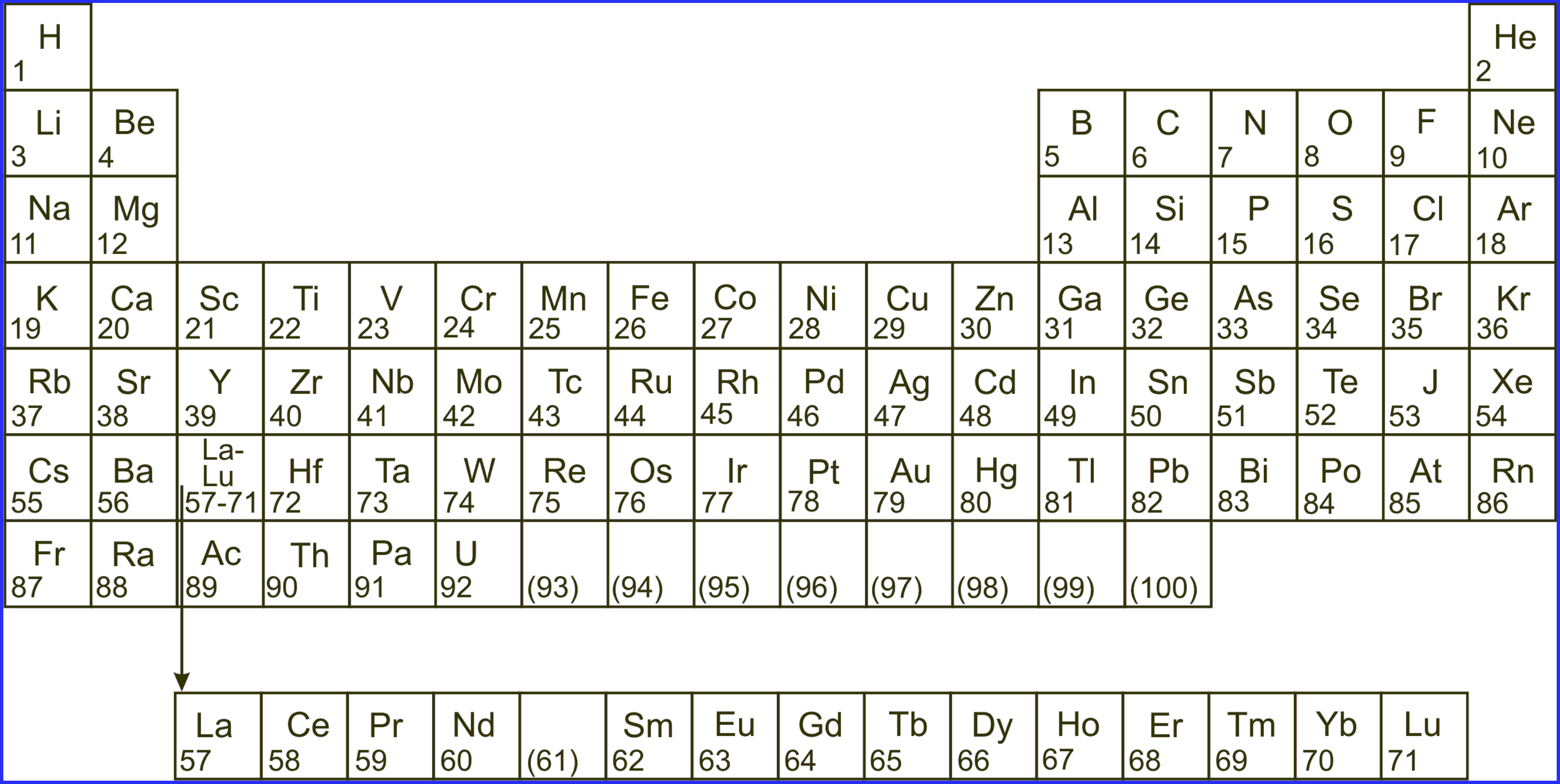
Ch.E. Düllmann
Superheavy element studies with preseparated isotopes

Mendeleev Periodic Table of 1871



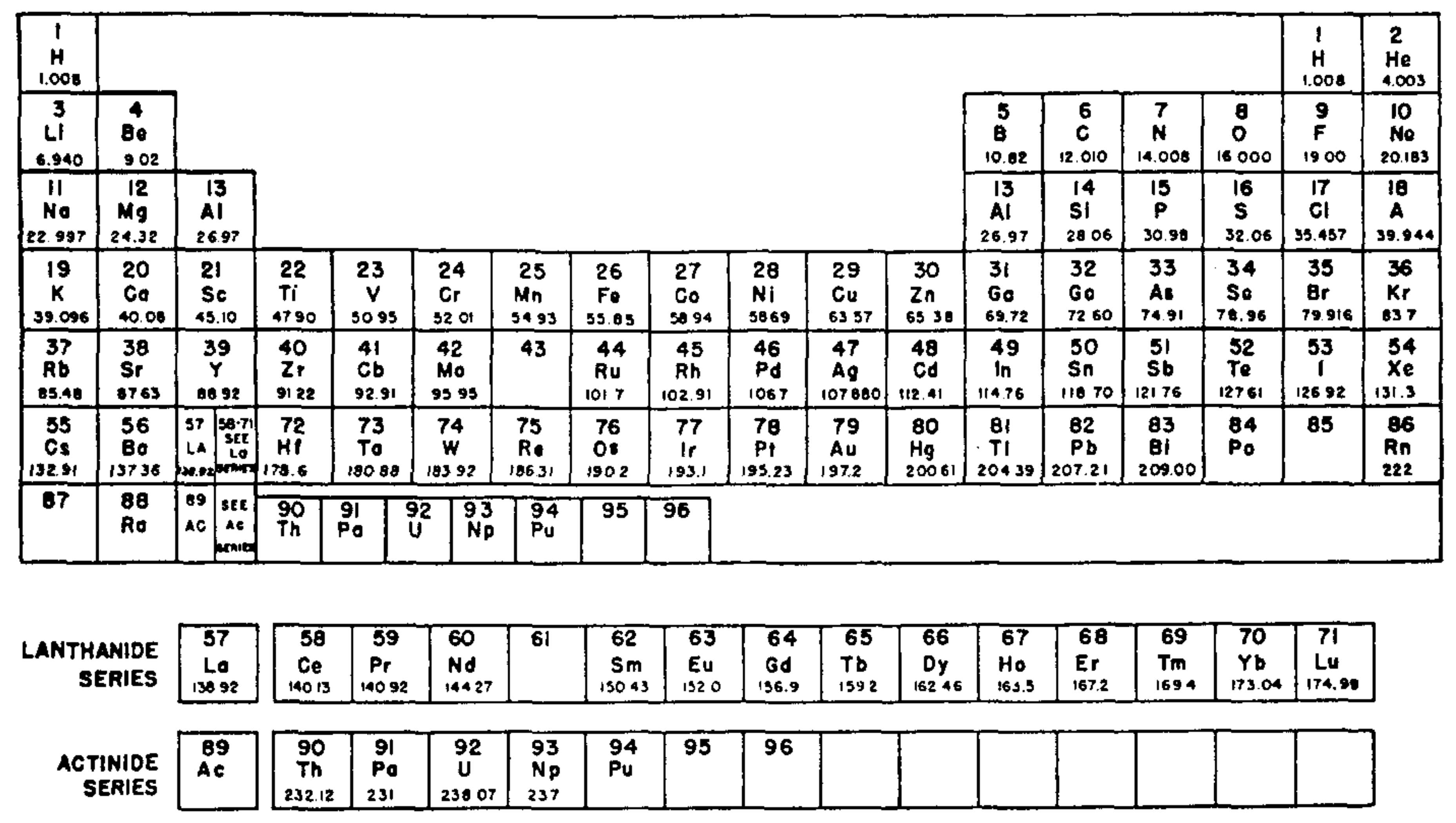
REIHEN	Gruppe I	Gruppe II	Gruppe III	Gruppe IV	Gruppe V	Gruppe VI	Gruppe VII	Gruppe VIII
	–	–	–	RH^4	RH^3	RH^2	RH	–
	R^2O	RO	R^2O^3	RO^2	R^2O^5	RO^3	R^2O^7	RO^4
1	H=1							
2	Li=7	Be=9.4	B=11	C=12	N=14	O=16	F=19	
3	Na=23	Mg=24	Al=27.3	Si=28	P=31	S=32	Cl=35.5	
4	K=39	Ca=40	–=44	Ti=48	V=51	Cr=52	Mn=55	Fe=56, Co=59, Ni=59, Cu=63
5	(Cu=63)	Zn=65	–=68	–=72	As=75	Se=78	Br=80	
6	Rb=85	Sr=87	?Yt=88	Zr=90	Nb=94	Mo=96	–=100	Ru=104, Rh=104, Pd=106, Ag=108
7	(Ag=108)	Cd=112	In=113	Sn=118	Sb=122	Te=125	J=127	
8	Cs=133	Ba=137	?Di=138	?Ce=140	–	–	–	– – – –
9	(–)	–	–	–	–	–	–	
10	–	–	?Er=178	?La=180	Ta=182	W=184	–	Os=195, Ir=197, Pt=198, Au=199
11	(Au=199)	Hg=200	Tl=204	Pb=207	Bi=208	–	–	
12	–	–	–	Th=231	–	U=240	–	– – – –

Pre-World War II Periodic Table



H 1																	He 2
Li 3	Be 4											B 5	C 6	N 7	O 8	F 9	Ne 10
Na 11	Mg 12											Al 13	Si 14	P 15	S 16	Cl 17	Ar 18
K 19	Ca 20	Sc 21	Ti 22	V 23	Cr 24	Mn 25	Fe 26	Co 27	Ni 28	Cu 29	Zn 30	Ga 31	Ge 32	As 33	Se 34	Br 35	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	J 53	Xe 54
Cs 55	Ba 56	La- Lu 57-71	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	Th 90	Pa 91	U 92	(93)	(94)	(95)	(96)	(97)	(98)	(99)	(100)				
		La 57	Ce 58	Pr 59	Nd 60	(61)	Sm 62	Eu 63	Gd 64	Tb 65	Dy 66	Ho 67	Er 68	Tm 69	Yb 70	Lu 71	

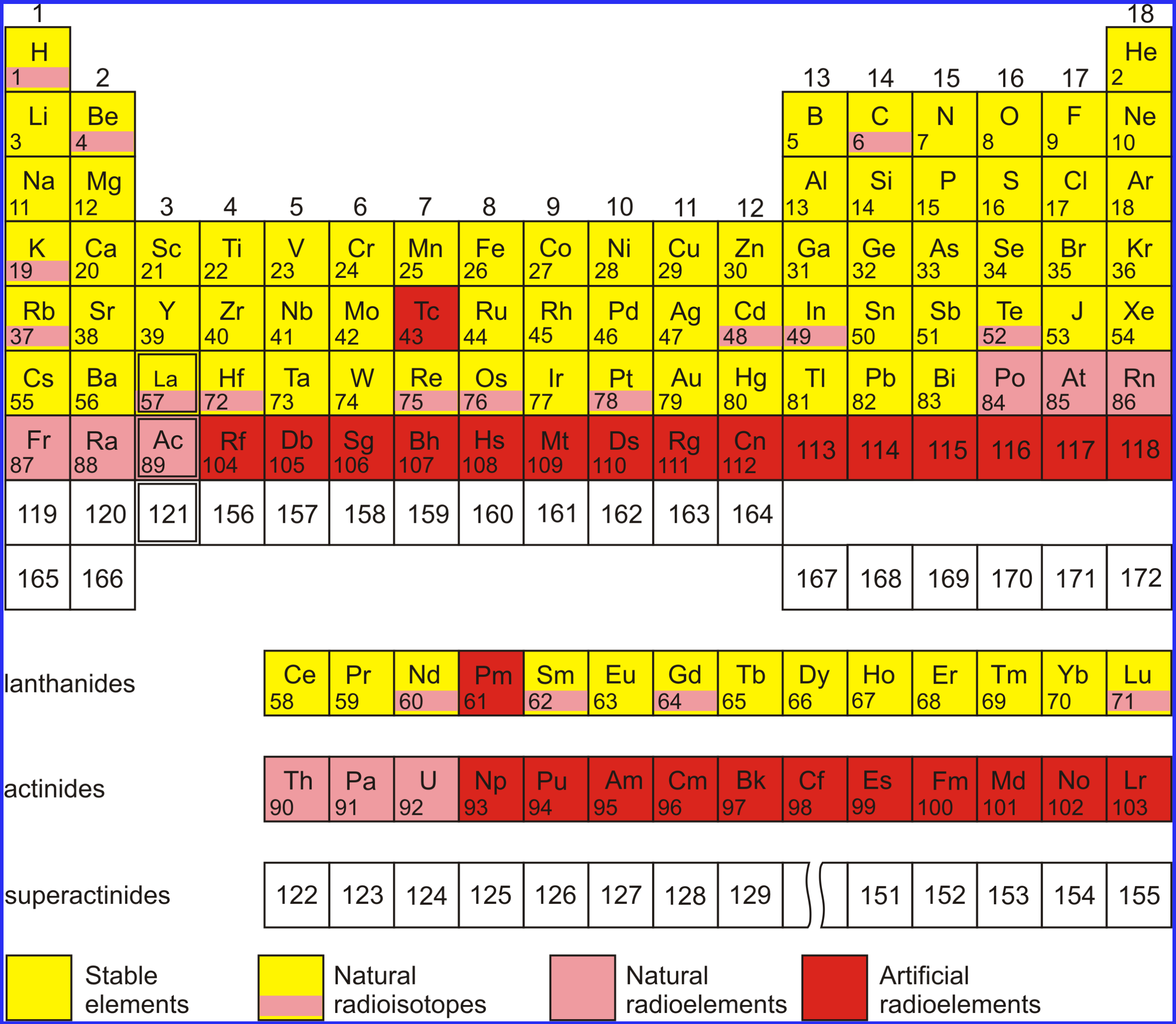
G. T. Seaborg, Chemical and Engineering News 23, 2190 (1945)



1 H 1.008																1 H 1.008	2 He 4.003
3 Li 6.940	4 Be 9 02											5 B 10.82	6 C 12.010	7 N 14.008	8 O 16 000	9 F 19 00	10 Ne 20.183
11 Na 22. 997	12 Mg 24.32	13 Al 26.97										13 Al 26.97	14 Si 28 06	15 P 30.98	16 S 32.06	17 Cl 35.457	18 A 39.944
19 K 39.096	20 Ca 40.08	21 Sc 45.10	22 Ti 47 90	23 V 50 95	24 Cr 52 01	25 Mn 54 93	26 Fe 55.85	27 Co 58 94	28 Ni 5869	29 Cu 63 57	30 Zn 65 38	31 Ga 69.72	32 Ge 72 60	33 As 74.91	34 Se 78.96	35 Br 79.916	36 Kr 83 7
37 Rb 85.48	38 Sr 87 63	39 Y 88 92	40 Zr 91 22	41 Cb 92.91	42 Mo 95 95	43	44 Ru 101 7	45 Rh 102.91	46 Pd 1067	47 Ag 107 880	48 Cd 112.41	49 In 114.76	50 Sn 118 70	51 Sb 121 76	52 Te 127 61	53 I 126 92	54 Xe 131.3
55 Cs 132.91	56 Ba 137 36	57 LA 138.92 58-71 SEE La SERIES	72 Hf 178.6	73 Ta 180 88	74 W 183 92	75 Re 186.31	76 Os 190 2	77 Ir 193.1	78 Pt 195.23	79 Au 197.2	80 Hg 200 61	81 Tl 204 39	82 Pb 207.21	83 Bi 209.00	84 Po	85	86 Rn 222
87	88 Ra	89 AC SEE AC SERIES	90 Th	91 Pa	92 U	93 Np	94 Pu	95	96								

LANTHANIDE SERIES	57 La 138 92	58 Ce 140 13	59 Pr 140 92	60 Nd 144 27	61	62 Sm 150 43	63 Eu 152 0	64 Gd 156.9	65 Tb 159 2	66 Dy 162 46	67 Ho 163.5	68 Er 167.2	69 Tm 169 4	70 Yb 173.04	71 Lu 174.99
ACTINIDE SERIES	89 Ac	90 Th 232.12	91 Pa 231	92 U 238 07	93 Np 237	94 Pu	95	96							

Present Periodic Table 2011



1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
H 1																	He 2
Li 3	Be 4											B 5	C 6	N 7	O 8	F 9	Ne 10
Na 11	Mg 12											Al 13	Si 14	P 15	S 16	Cl 17	Ar 18
K 19	Ca 20	Sc 21	Ti 22	V 23	Cr 24	Mn 25	Fe 26	Co 27	Ni 28	Cu 29	Zn 30	Ga 31	Ge 32	As 33	Se 34	Br 35	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	J 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	Cn 112	113	114	115	116	117	118
119	120	121	156	157	158	159	160	161	162	163	164						
165	166											167	168	169	170	171	172

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
superactinides	122	123	124	125	126	127	128	129		151	152	153	154	155

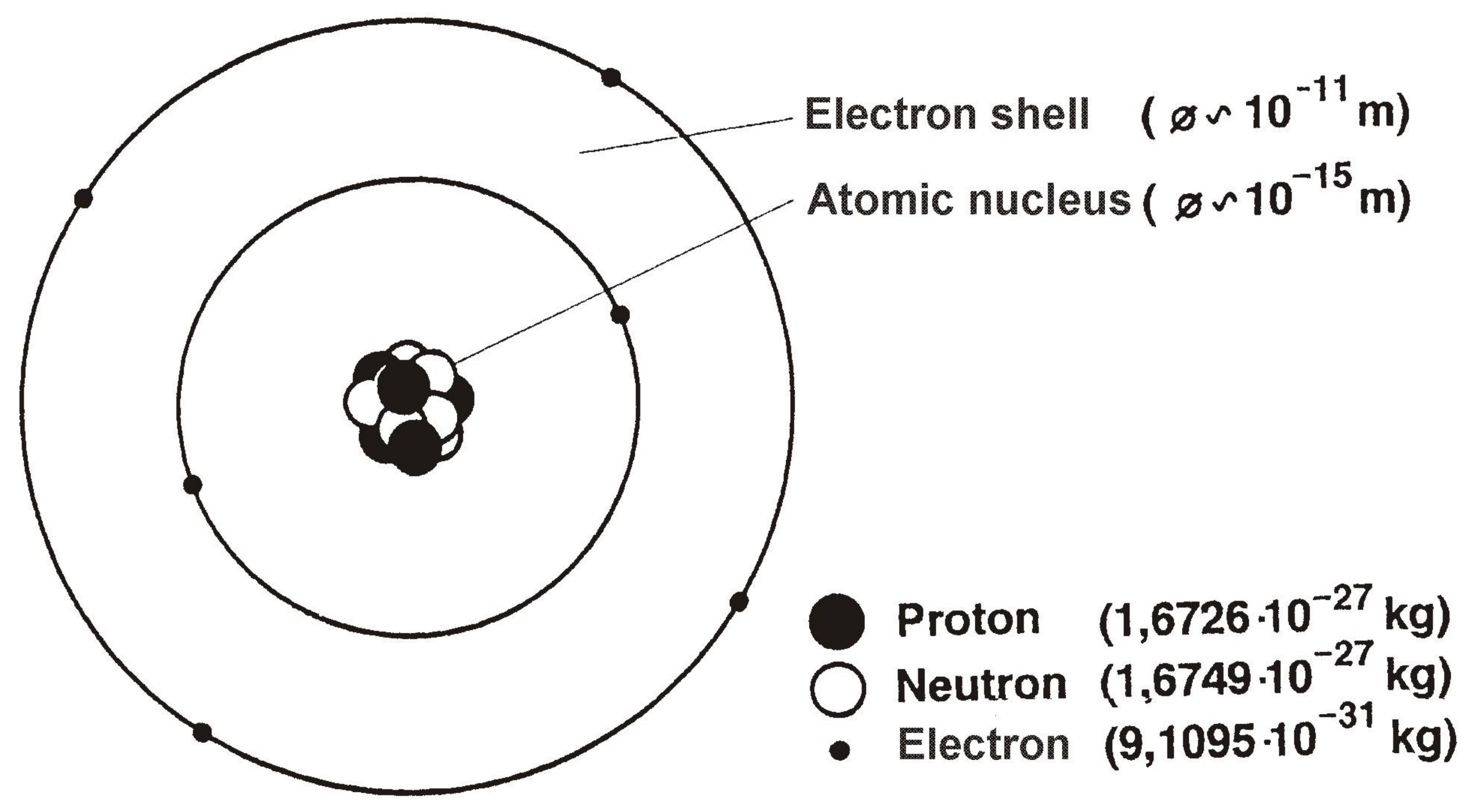
Stable elements

Natural radioisotopes

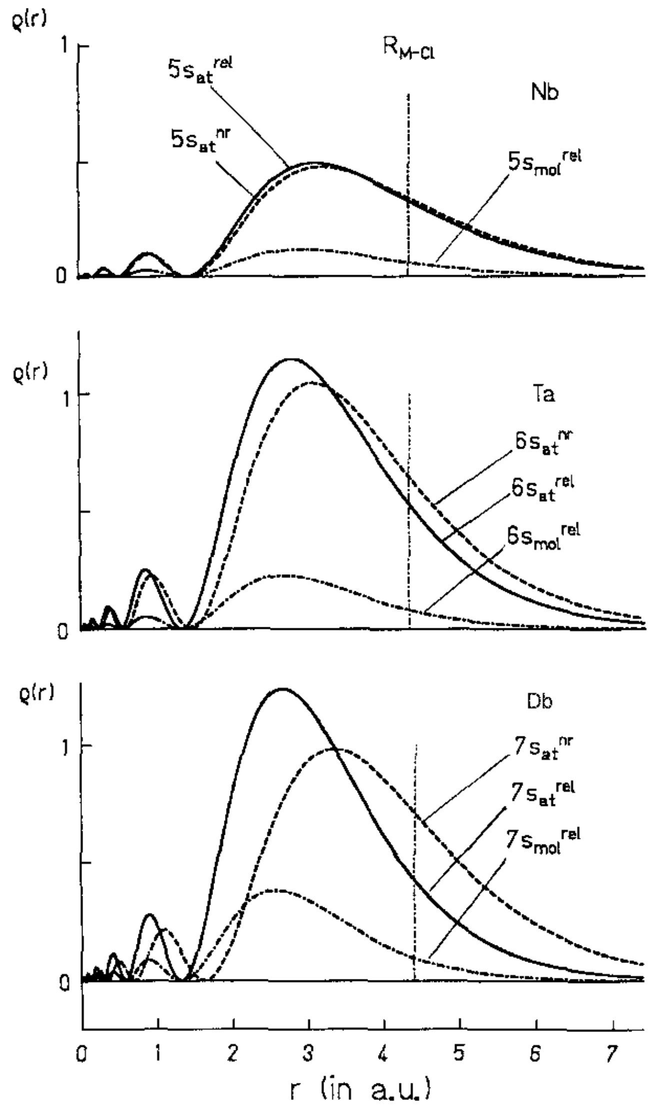
Natural radioelements

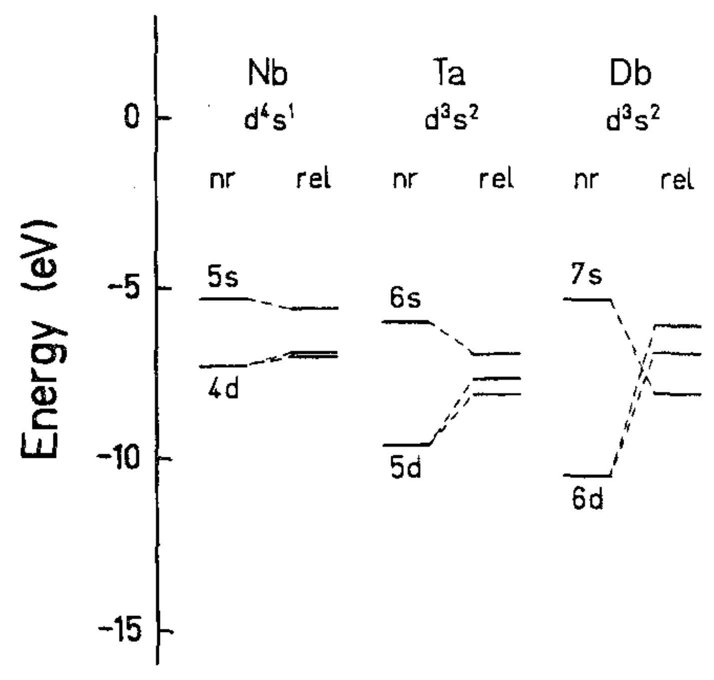
Artificial radioelements

Atomic Structure

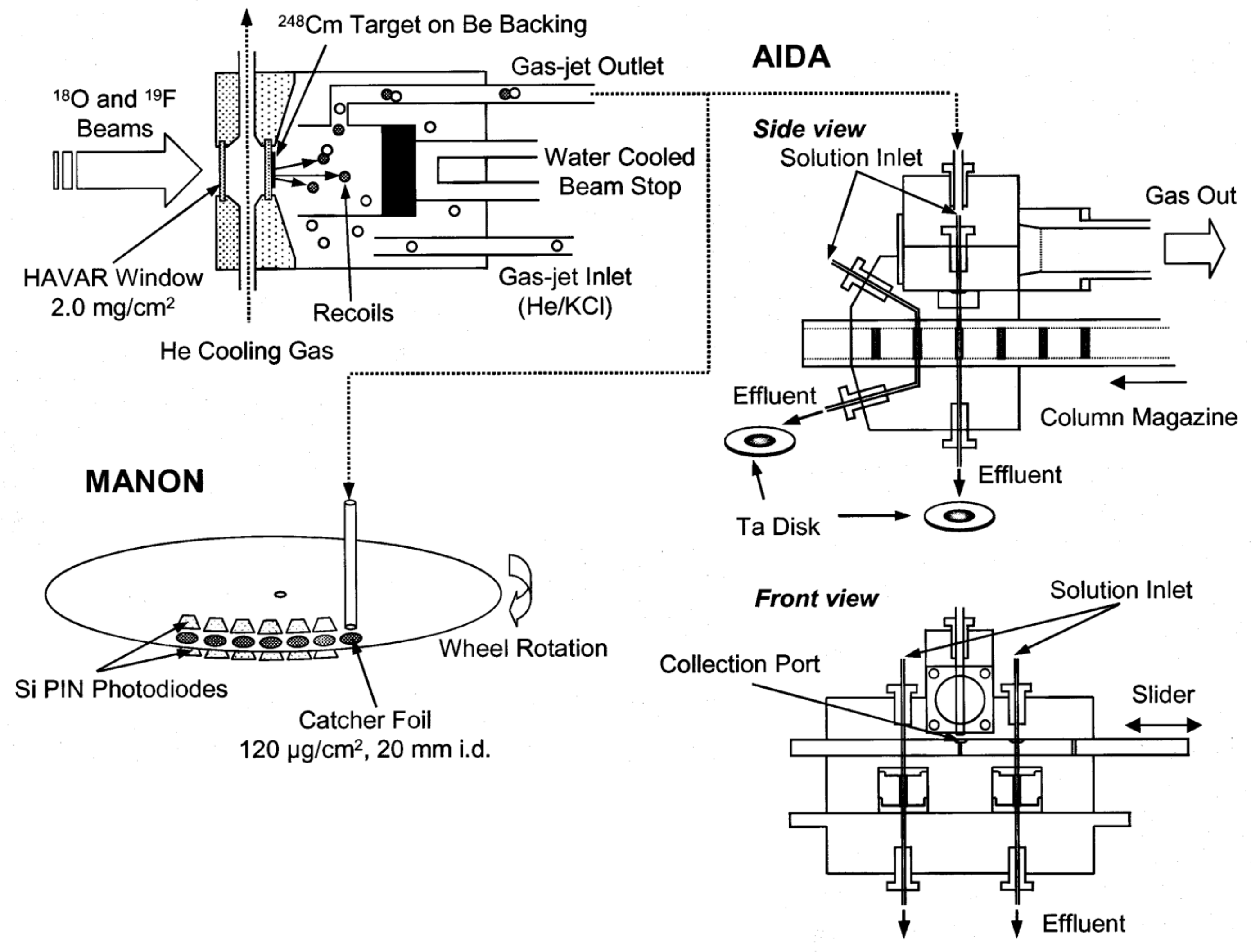


Relativistic Effects

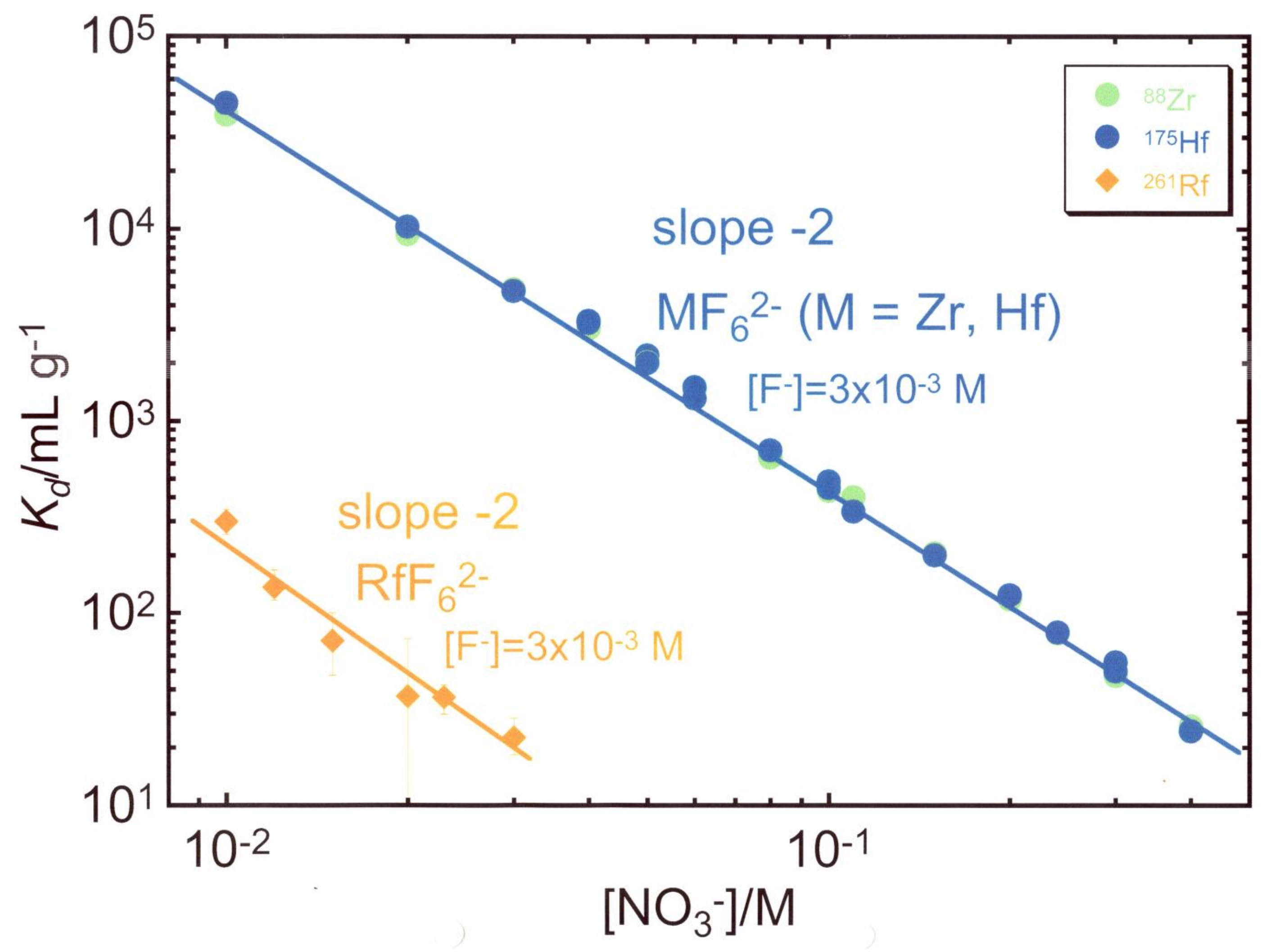




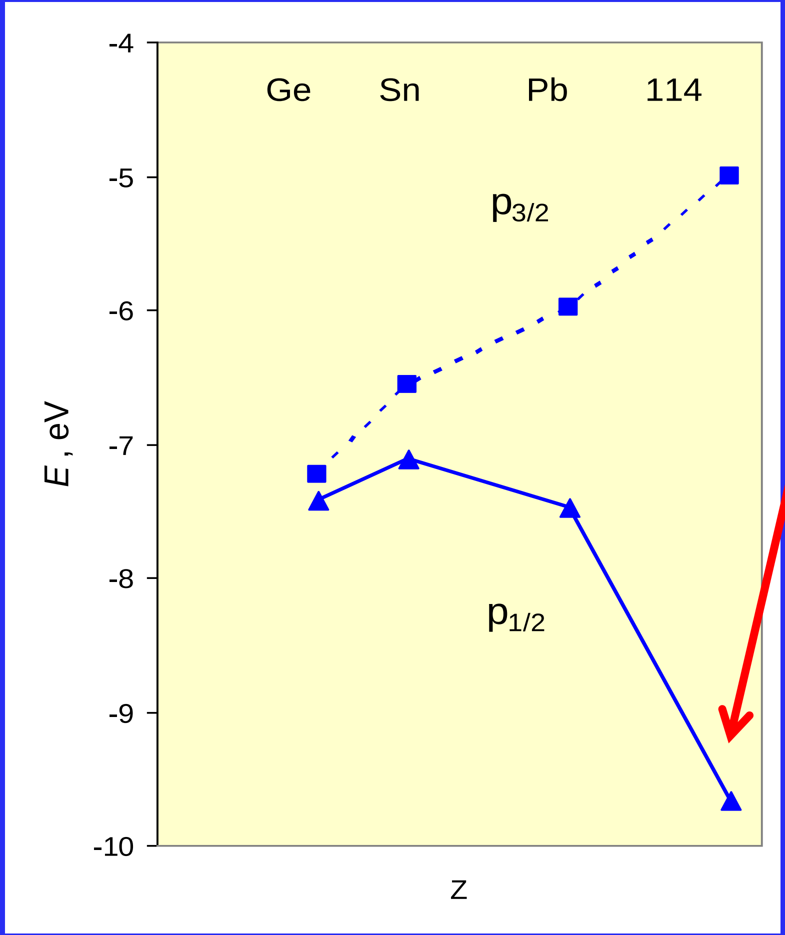
One - atom - at - a - time chemistry
Aqueous Chemistry of Rf



Aqueous Chemistry of Rf



Direct and indirect relativistic effects



$$m = m_0 / \sqrt{(1-(v/c)^2}$$

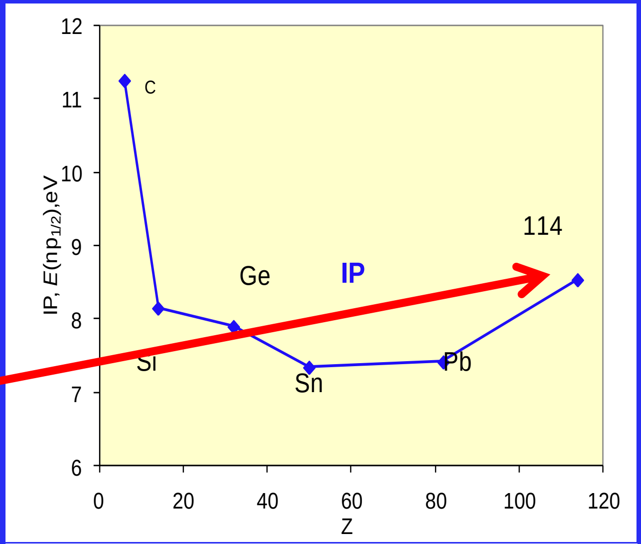
$$a_0 = 4\pi\varepsilon_0\hbar^2 / me^2$$

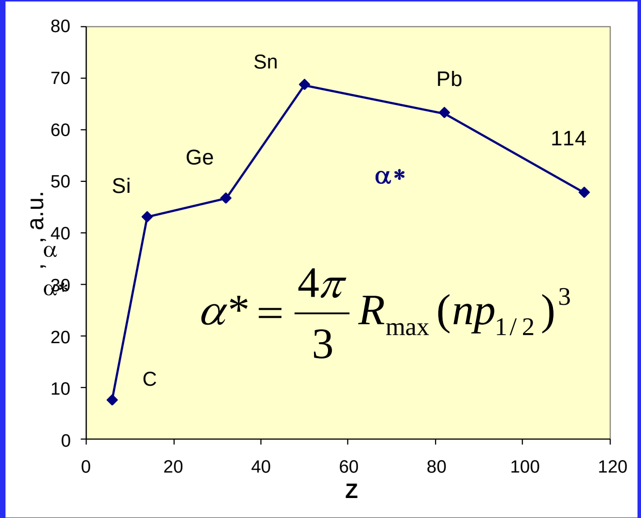
Direct:

contraction and **stabilization** of $s_{1/2}$ and $p_{1/2}$ orbitals

Indirect:

expansion and destabilization of $p_{3/2}$ and d orbitals





$$\alpha^* = \frac{4\pi}{3} R_{\max}(np_{1/2})^3$$

Relativistic calculations of E114 atomic properties
(*V. Pershina et al. J. Chem. Phys.* ***128***, *024707 (2008)*)

Summary of Predicted Properties of Elements 112 and 114

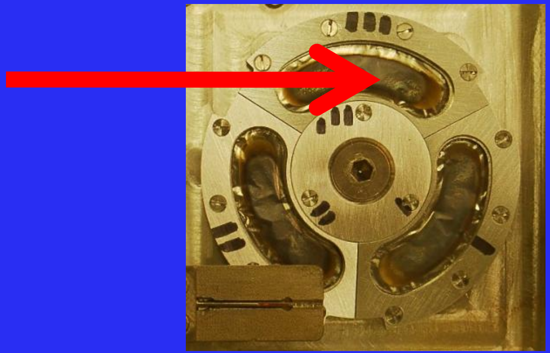
Property	112	114
Electronic configuration	$d^{10}s^2$	$s^2p_{1/2}^2$
IP, eV	11.97	8.54
α, a.u.	27.4	29.5
AR, a.u.	3.21	3.30
R_{vdW}, a.u.	3.75	3.94
ΔH_{ads}(quartz), kJ/mol	-27	-21
ΔH_{ads}(gold), kJ/mol	-65	-92

Element 114 should be more reactive than 112 !!!

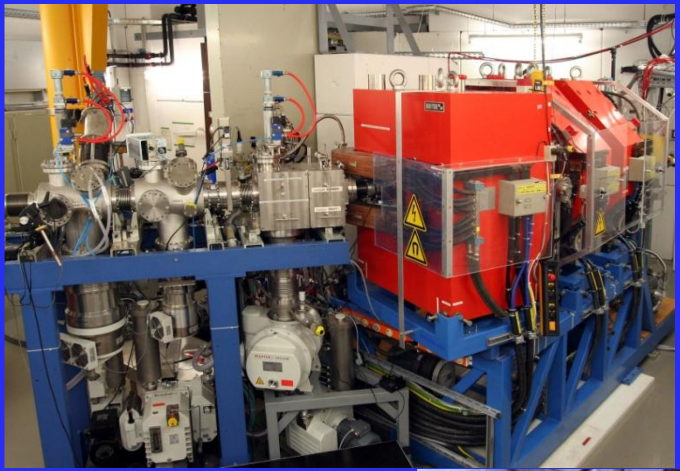
4*c*-DFT and *ab initio* DC calculations [V. Pershina *et al*., J. Chem. Phys. (2008)]

114 Chemistry

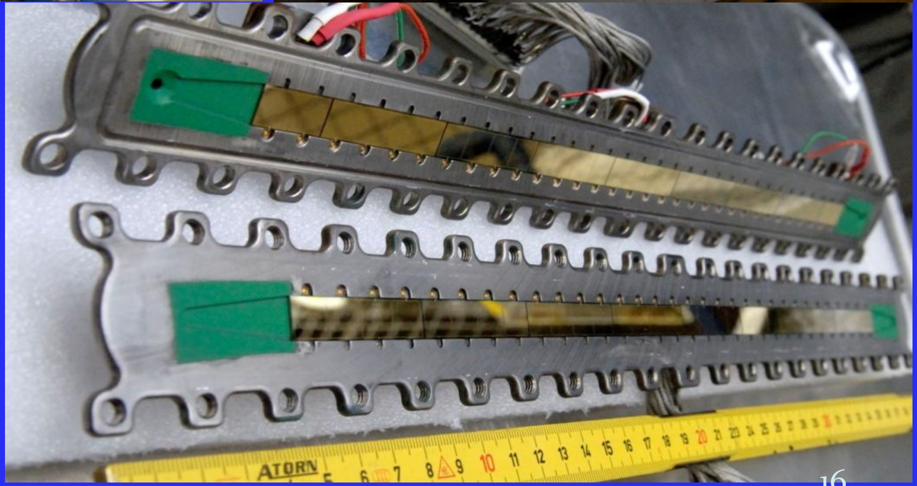
Beam: $^{48}Ca^{+10}$
(5.475 MeV/u)



TASCA / SIM

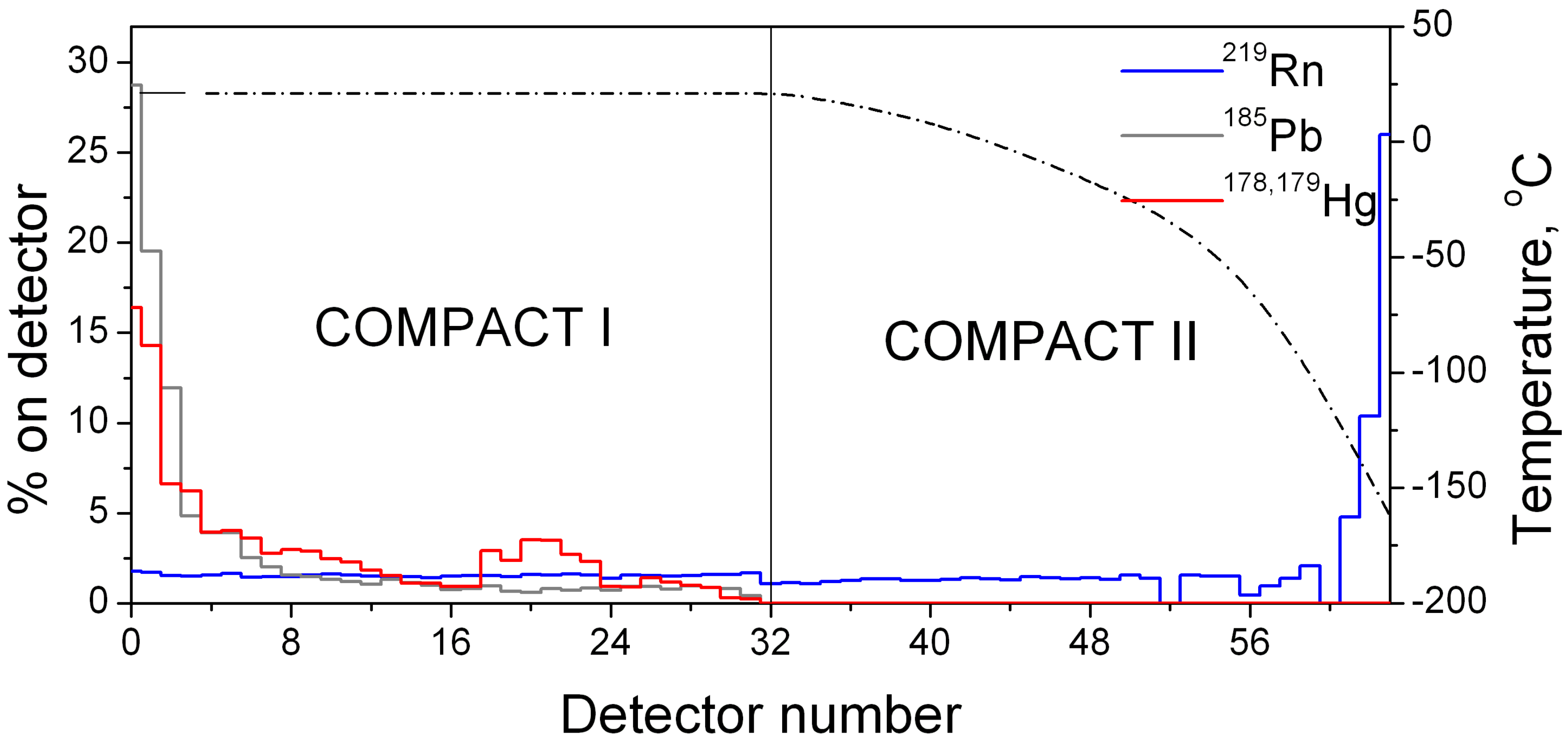


RTC / COMPACT



Target: $^{244}PuO_2$
Backing 2.5 μm Ti
Segment 1: 440 $\mu g/cm^2$
Segment 2: 771 $\mu g/cm^2$
Segment 3: 530 $\mu g/cm^2$

Adsorption of Pb, Hg and Rn in COMPACT



Monte Carlo simulation and thermodynamical calculation

Using the model of the mobile adsorption

$$\frac{\Delta S_{ads}}{R} = \ln\left(\frac{A}{V\cdot\upsilon_b}\cdot\sqrt{\frac{RT}{2\pi\cdot M}}\right)+\frac{1}{2};\quad \Delta S_{ads} = -236\,Jmol^{-1}K^{-1}$$

$$\exp\left(\frac{-\Delta H_{ads}}{RT}\right) = \frac{t_R^{IC}\cdot Q_0\cdot T_c}{s\cdot T_0\cdot L}\exp\left(\frac{-\Delta S_{ads}}{R}\right)$$

$t_R = T_{1/2} / \ln2$
$s = 2.1\ cm^2$
$Q = 22\ cm^3/s$
$L = 32\ cm$
$\nu_b = 4.2\cdot10^{12}$

a lower limit of the adsorption enthalpy value can be calculated with retention time t_R and column length L:

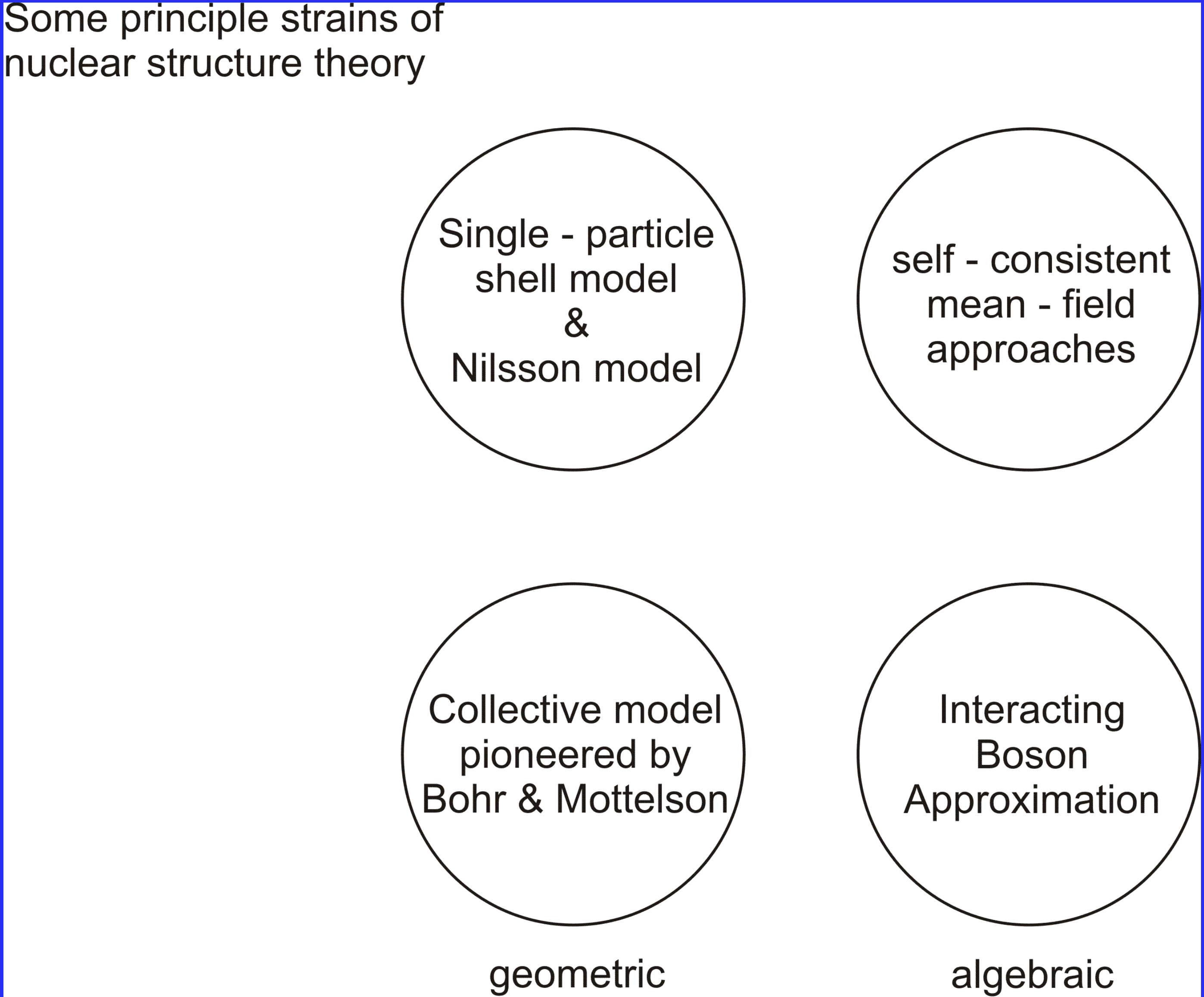
$-\Delta H_{ads}(Au) > 48$ kJ/mol

$-\Delta H_{ads}$(Au): Pb > Hg > **114** > **112**

Relativistic effects have created a new category of elements in the Periodic Table:

112 an 114 are gaseous metals

Nuclear Structure



Some principle strains of nuclear structure theory

Single - particle shell model & Nilsson model

self - consistent mean - field approaches

Collective model pioneered by Bohr & Mottelson

Interacting Boson Approximation

geometric

algebraic

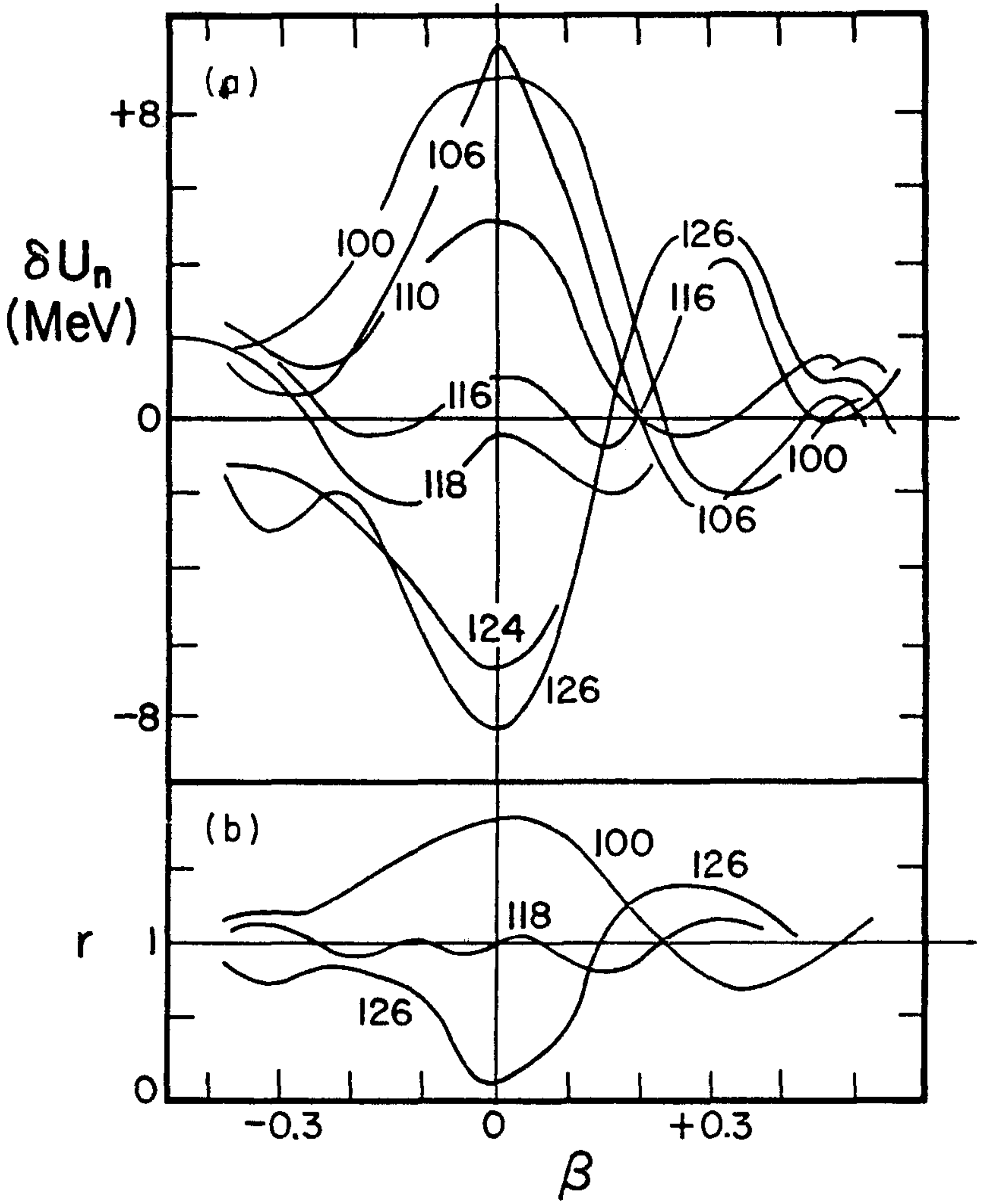
V.M. Strutinsky 1967 $$E = E_{LDM} + \sum_{n,p} (\delta U + \delta P)$$

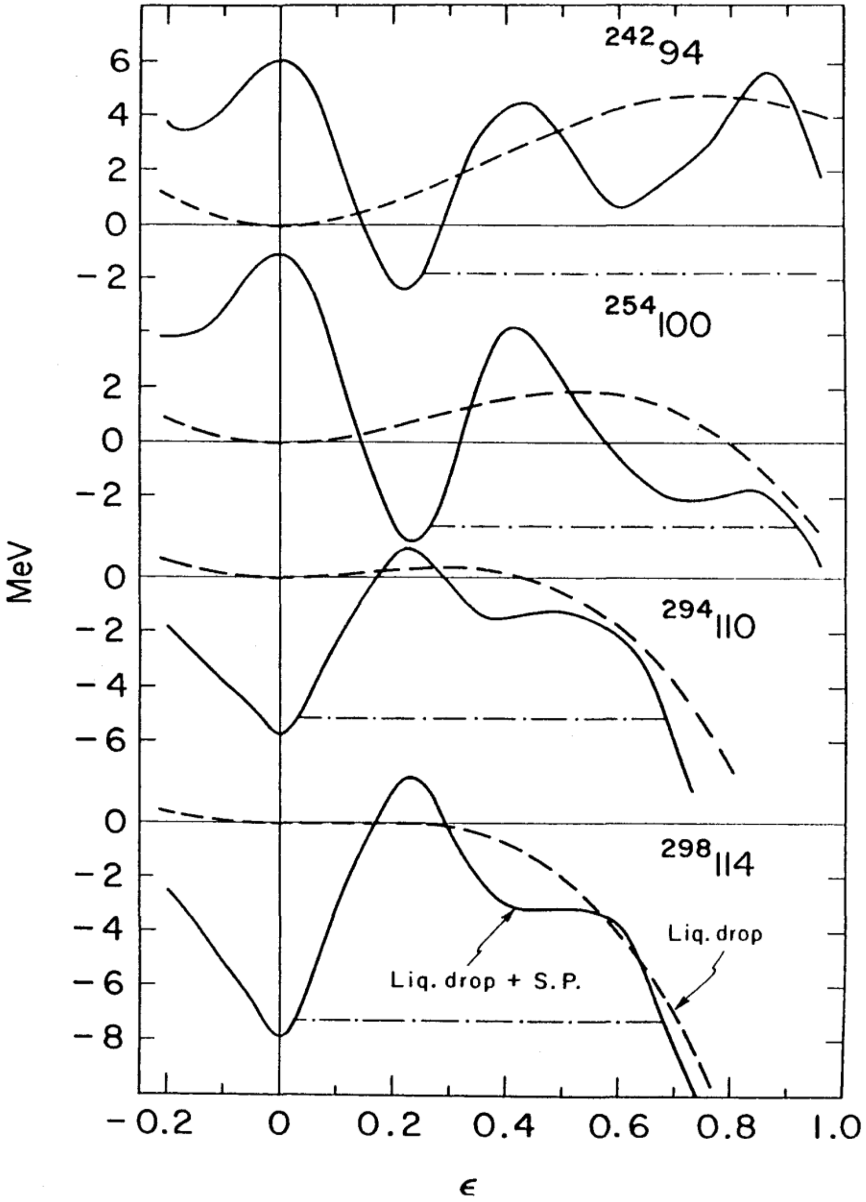
Shell correction $\delta U = U - \tilde{U}$ with $U = \sum_{\nu} 2\varepsilon_{\nu} n_{\nu}$

and $\tilde{U} = 2\int_{-\infty}^{\lambda} \varepsilon\, \tilde{g}(\varepsilon)\, d\varepsilon$

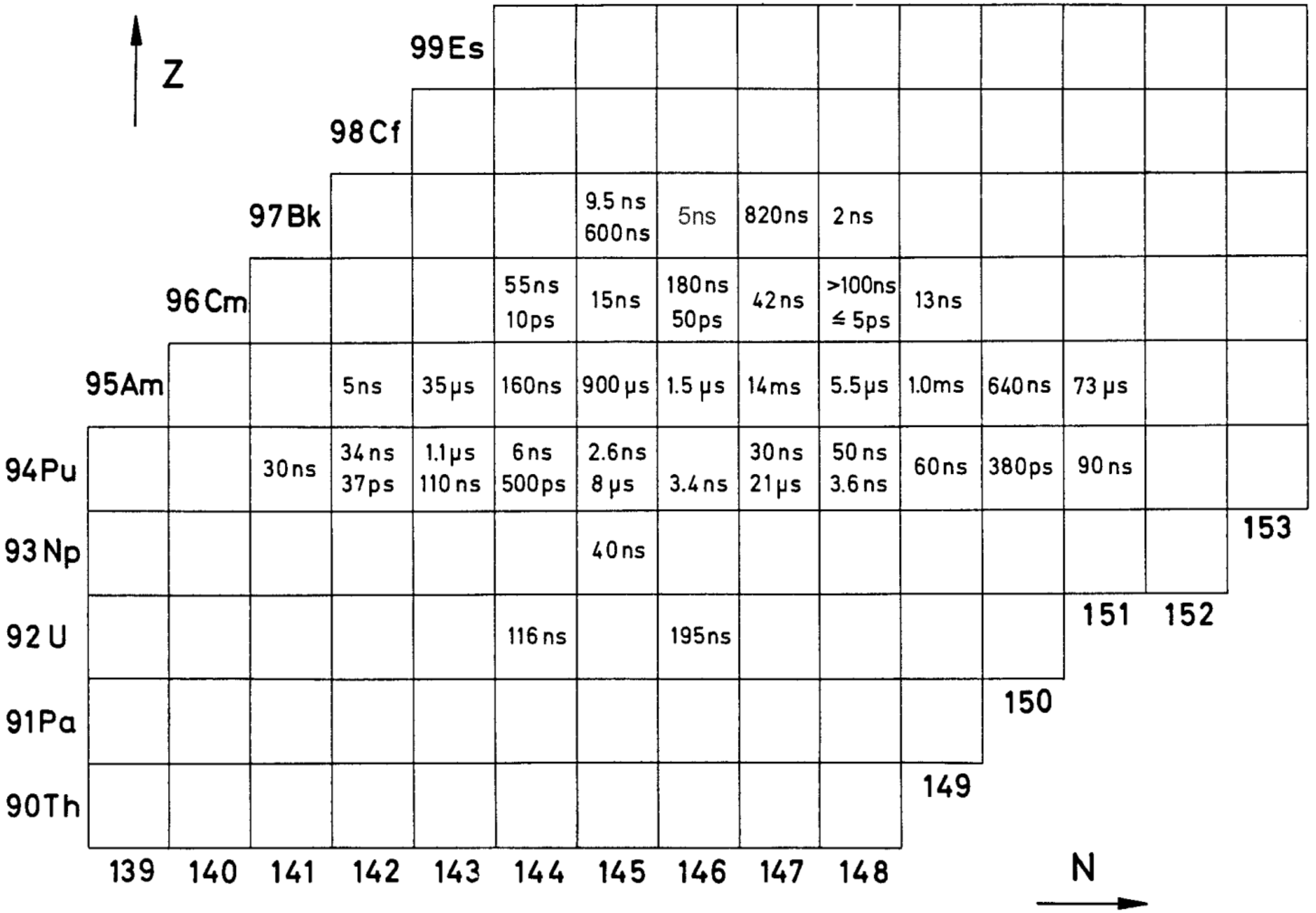
where $\tilde{g}(\varepsilon)$ is a uniform distribution of single-particle states
λ is the chemical potential defined by
$N = 2\int_{-\infty}^{\lambda} \tilde{g}(\varepsilon)\, d\varepsilon$, and $N =$ total number of particles

Philosophy: systematic errors arising from the calculation of the total energy from a single-particle model will cancel, and only effects associated with the special degeneracies and splitting of levels in the shell-model potential will remain as a shell correction.

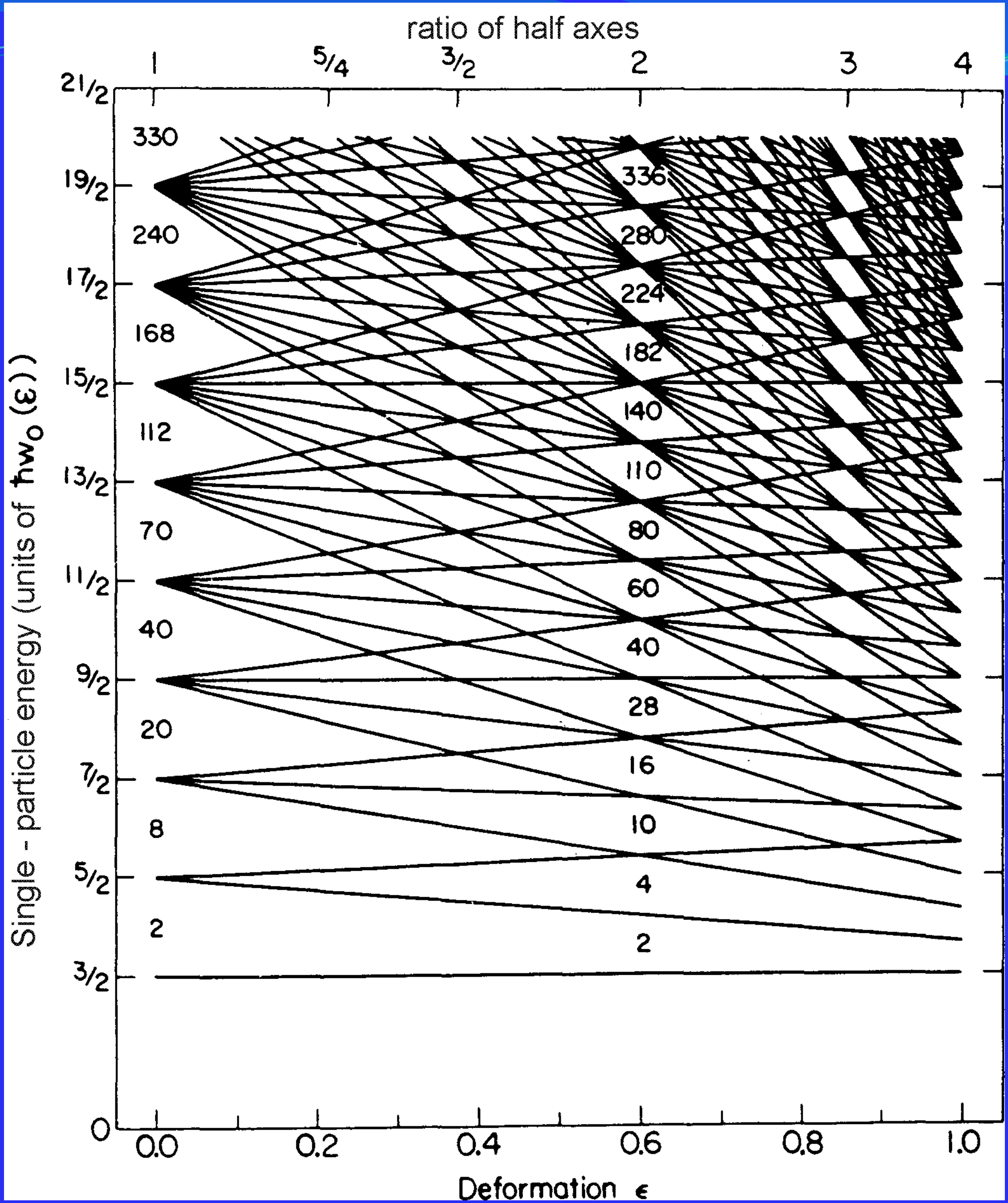

(a)
+8
0
−8
δU_n (MeV)
106
100
110
126
116
116
118
124
126
100
106
(b)
100
126
118
126
r
1
0
−0.3
0
+0.3
β


$^{242}94$
$^{254}100$
$^{294}110$
$^{298}114$
Liq. drop
Liq. drop + S.P.
MeV
6
4
2
0
−2
2
0
−2
0
−2
−4
−6
0
−2
−4
−6
−8
−0.2
0
0.2
0.4
0.6
0.8
1.0
ϵ

Fission Isomers

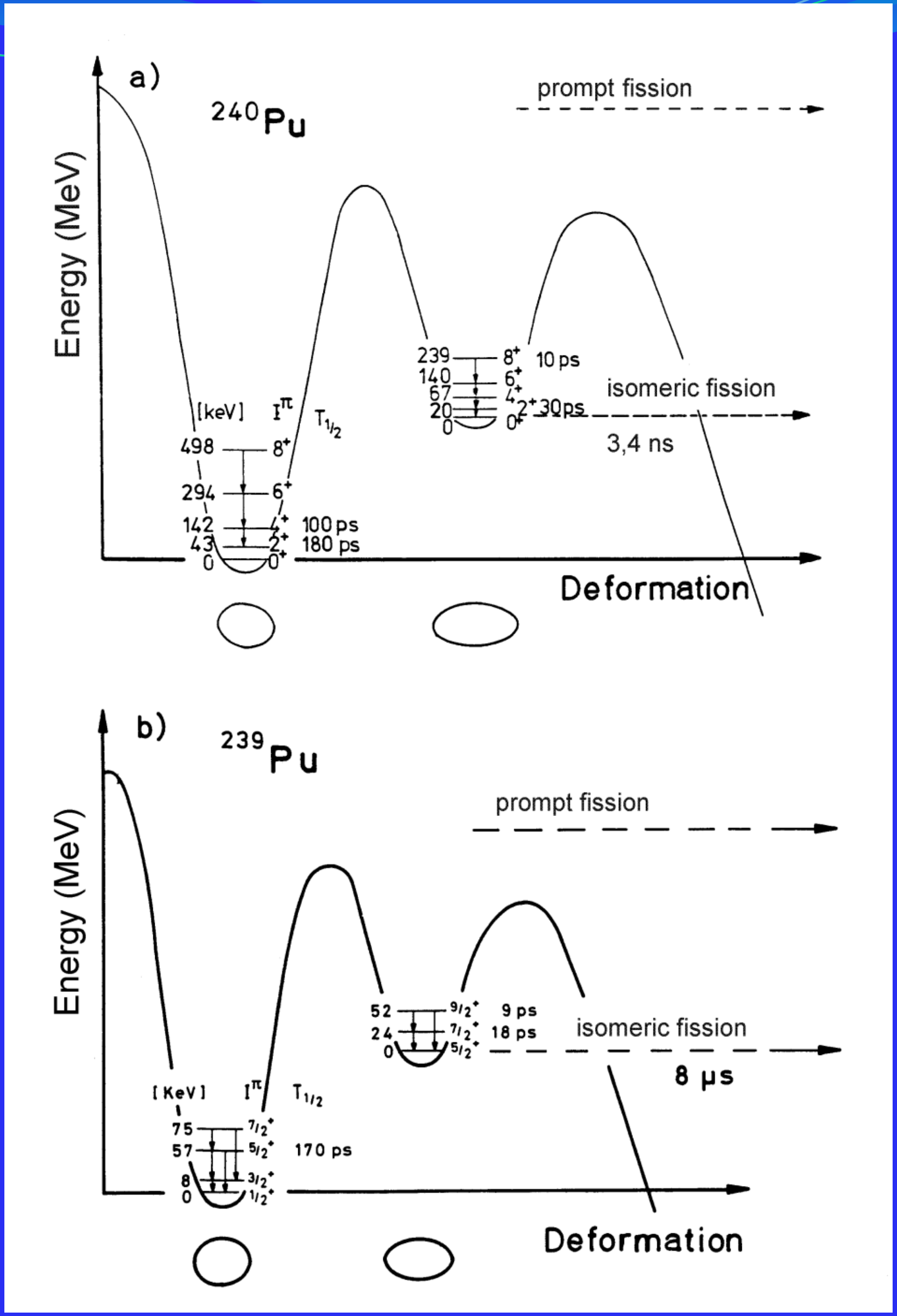


	139	140	141	142	143	144	145	146	147	148	149	150	151	152	153
99Es															
98Cf															
97Bk							9.5 ns 600 ns	5ns	820ns	2 ns					
96Cm						55ns 10ps	15ns	180ns 50ps	42ns	>100ns ≤ 5ps	13ns				
95Am				5ns	35μs	160ns	900μs	1.5μs	14ms	5.5μs	1.0ms	640ns	73μs		
94Pu			30ns	34ns 37ps	1.1μs 110ns	6ns 500ps	2.6ns 8μs	3.4ns	30ns 21μs	50ns 3.6ns	60ns	380ps	90ns		
93Np							40ns								
92U						116ns		195ns							
91Pa															
90Th															


ratio of half axes
1
5/4
3/2
2
3
4
21/2
19/2
17/2
15/2
13/2
11/2
9/2
7/2
5/2
3/2
0
330
240
168
112
70
40
20
8
2
336
280
224
182
140
110
80
60
40
28
16
10
4
2
Single - particle energy (units of ħω₀(ε))
0.0
0.2
0.4
0.6
0.8
1.0
Deformation ε

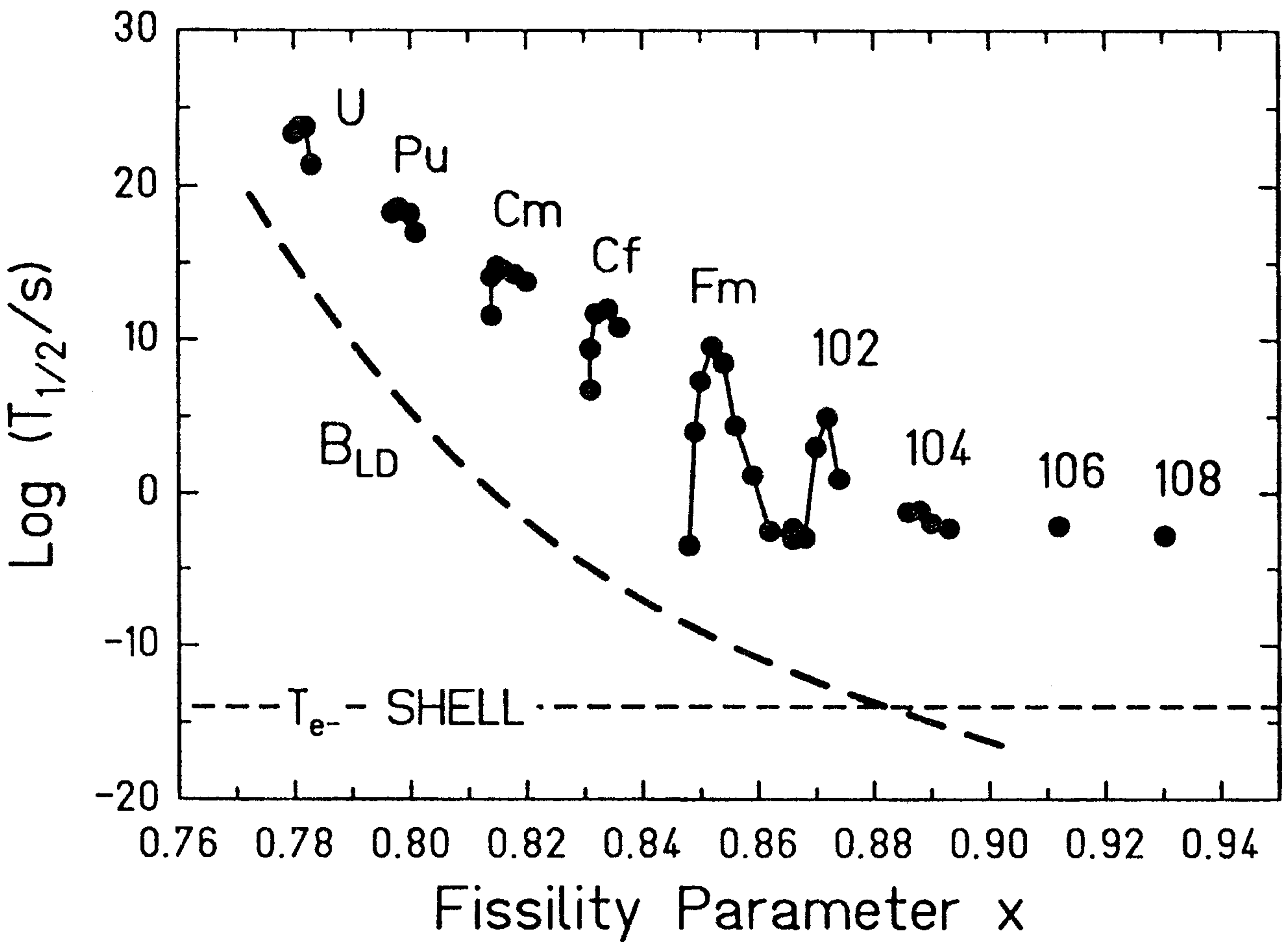
D. Habs, V. Metag (1978)

Charge – plunger technique

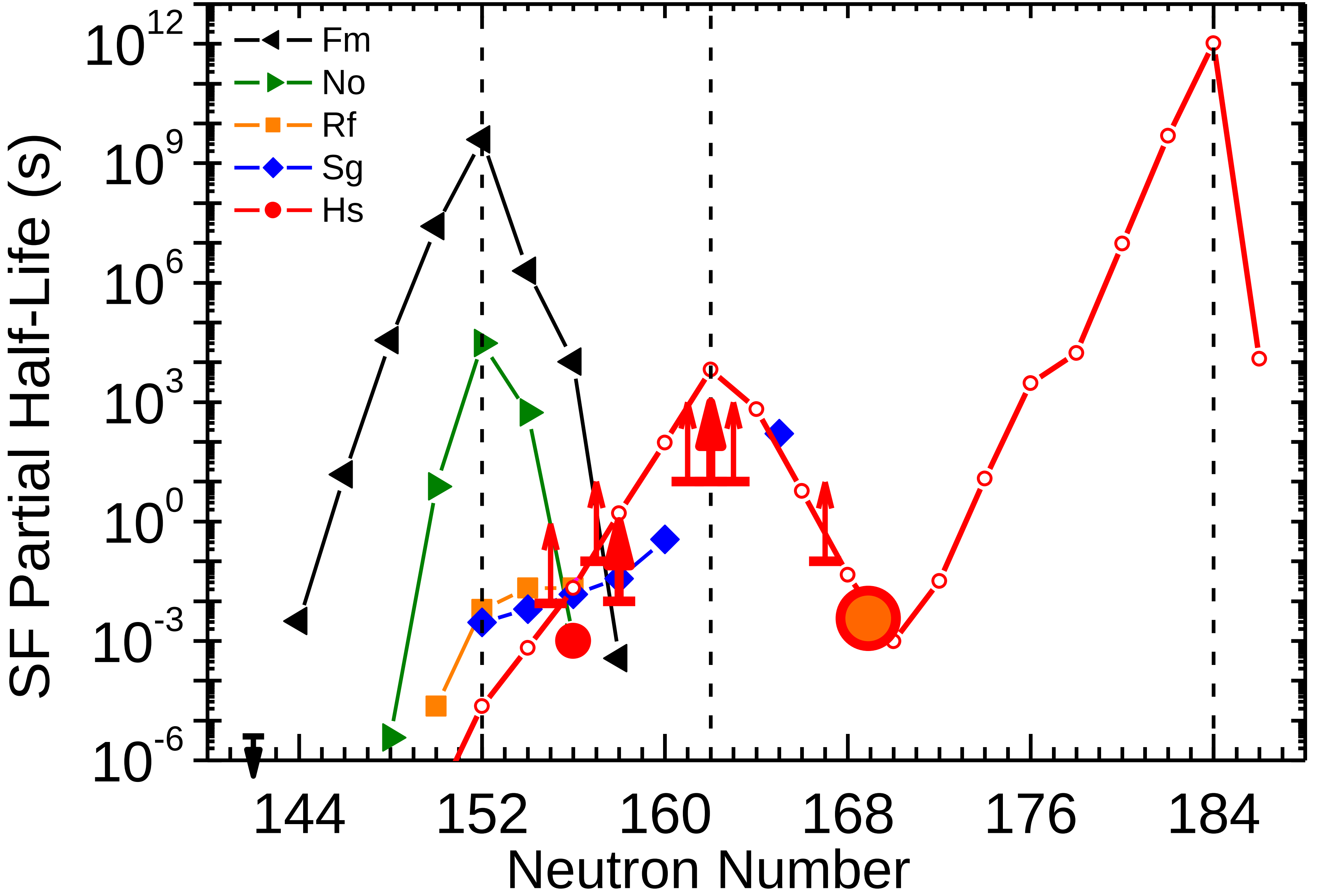


Comparison of experimental and theoretical quadrupole moments and deformations in the first and second minimum of ^{239}Pu.

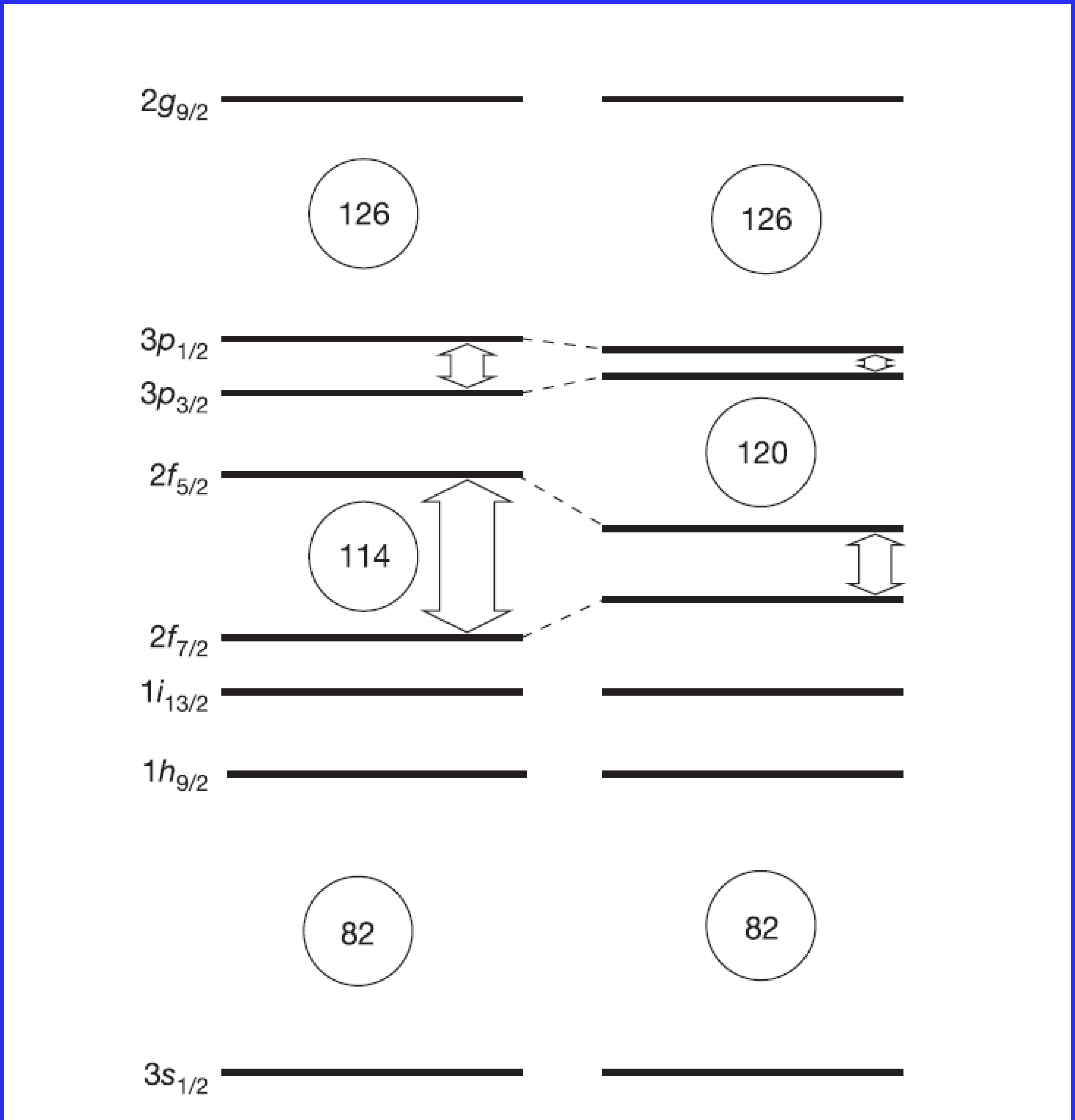
	1. Minimum	2. Minimum
Q_{exp}	$(11{,}3 \pm 0{,}5)$ b	(36 ± 4) b
Q_{theor} (^{240}Pu)		38 b [Lit.] 35 b [Lit.]
$(c/a)_{exp}$	$(1{,}30 \pm 0{,}05)$	$(2{,}0 \pm 0{,}1)$


30
20
10
0
−10
−20
Log (T_{1/2}/s)
0.76
0.78
0.80
0.82
0.84
0.86
0.88
0.90
0.92
0.94
Fissility Parameter x
U
Pu
Cm
Cf
Fm
102
104
106
108
B_{LD}
T_{e-} SHELL

Integral data: Systematics of SF Half-Lives



Search for the next proton shell

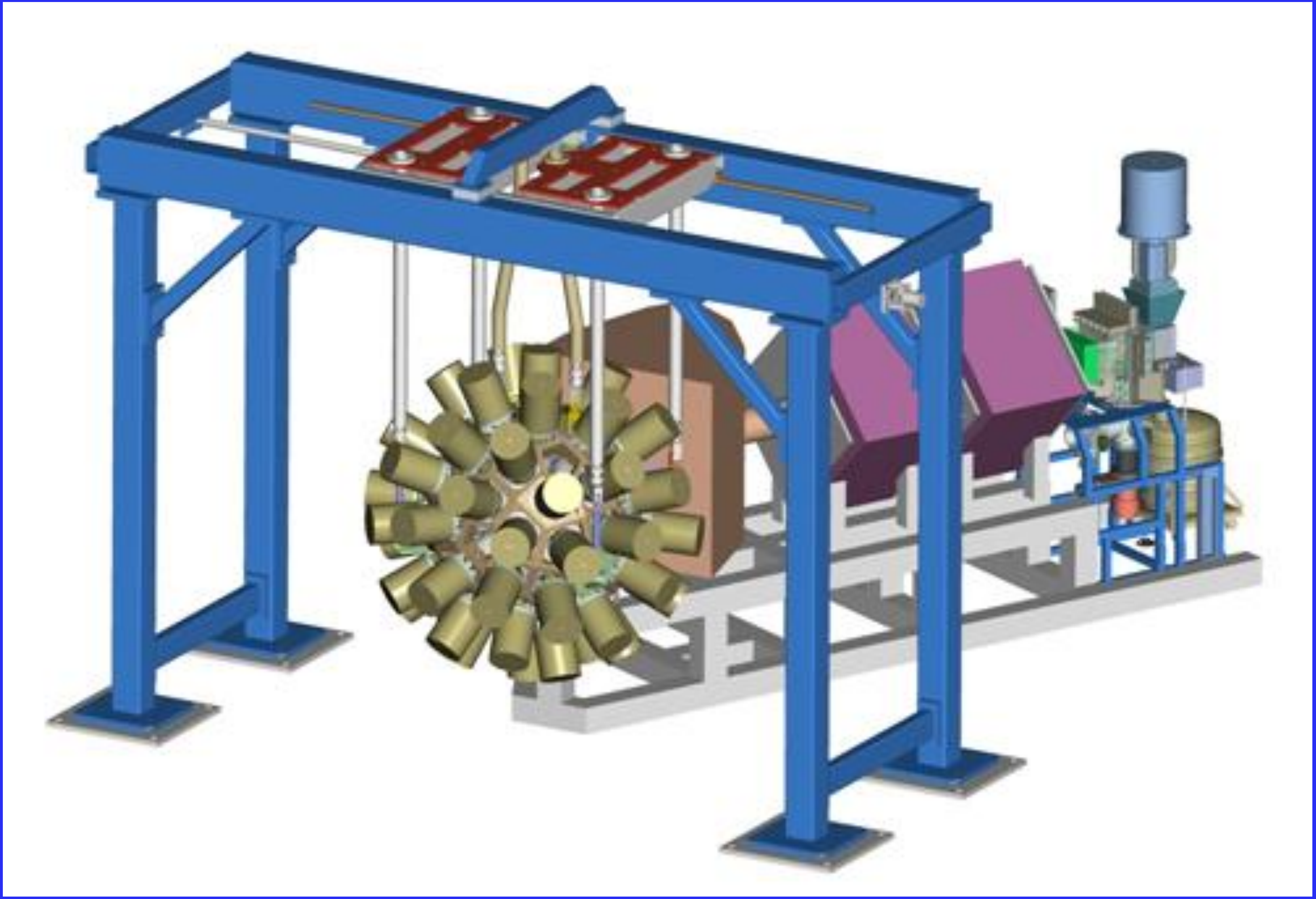


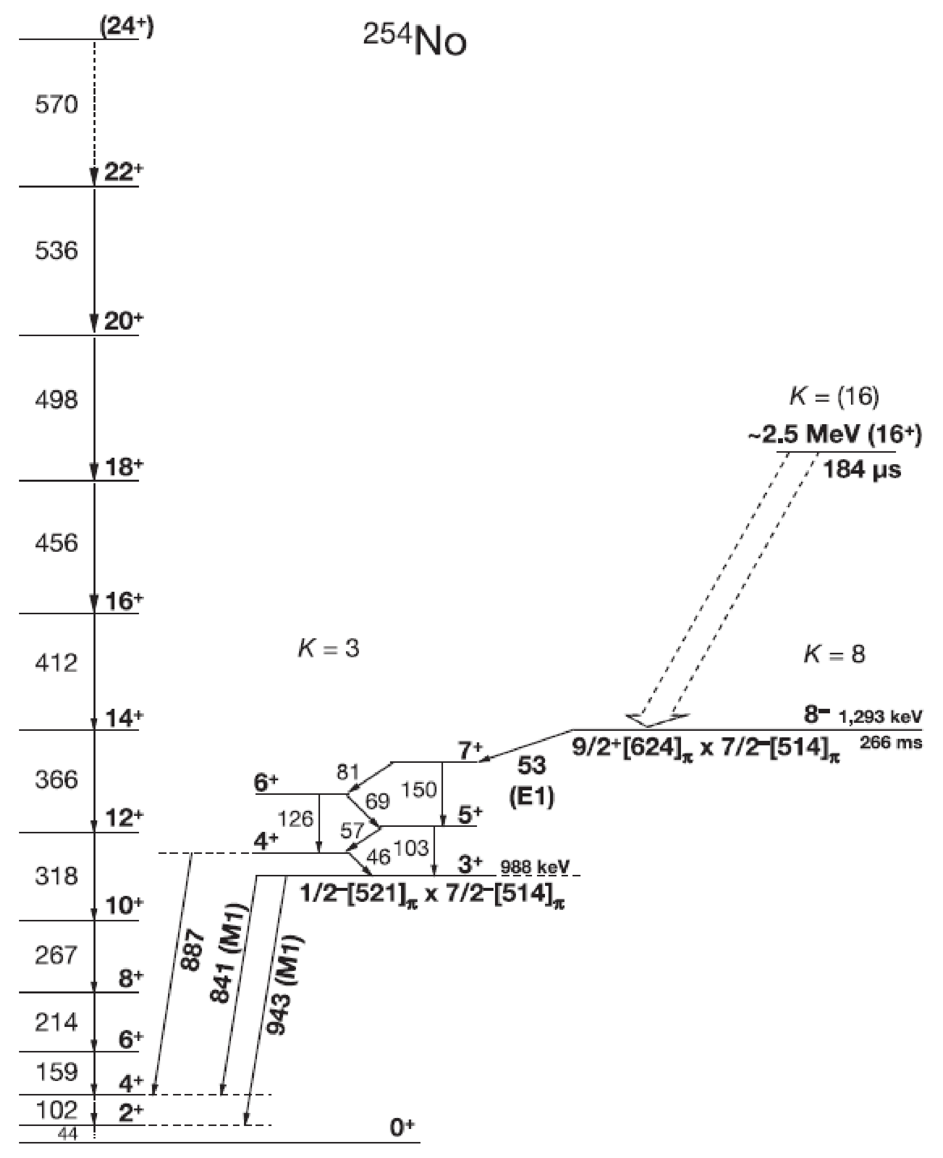
Look at experimental data for superheavy elements:

- **integral data for a few atoms such as half lives, decay modes, and Q_α- values**

- **spectroscopic studies in nuclei approaching the "island of stability" such as ^{254}No (Z=102, N=152).**
  These are deformed nuclei and the degenerate spherical single-particle orbitals split in a well defined manner into Nilsson components according to the projection of the angular momentum onto the symmetry axis of the nucleus, the K quantum number.
  Orbitals above the spherical proton shell, e.g., $2f_{5/2}$, whose low-spin components come close to the Fermi level in a prolate nucleus, play a key role in the formation of excited states in nuclei near ^{254}No.
  In particular, K isomers give a very clear and unique experimental signature through their decay times and paths.

RITU: Recoil – Decay – Tagging (RDT)

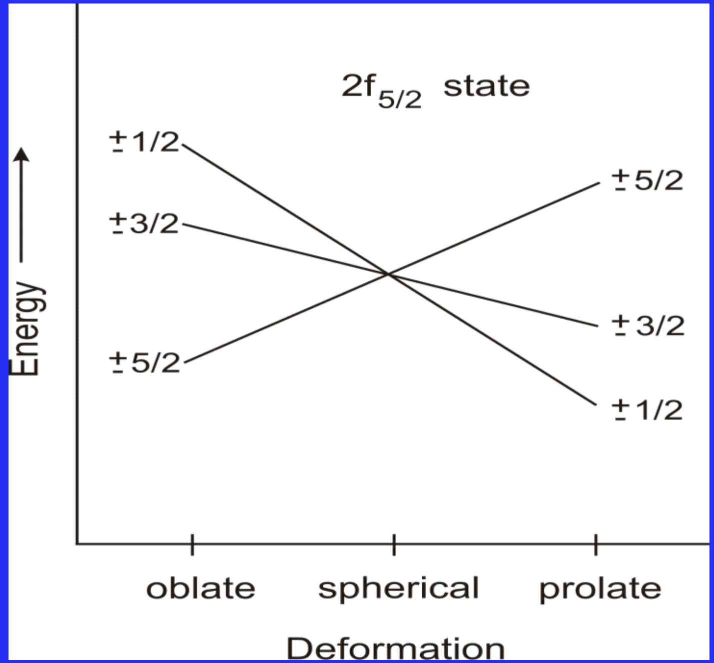




^{254}No
(24+)
570
22+
536
20+
498
18+
456
16+
412
14+
366
12+
318
10+
267
8+
214
6+
159
4+
102
2+
44
0+
K = (16)
~2.5 MeV (16+)
184 µs
K = 3
K = 8
8− 1,293 keV
9/2+[624]π x 7/2−[514]π
266 ms
7+
53
(E1)
81
6+
69
150
5+
126
4+
57
46
103
3+
988 keV
1/2−[521]π x 7/2−[514]π
887
841 (M1)
943 (M1)

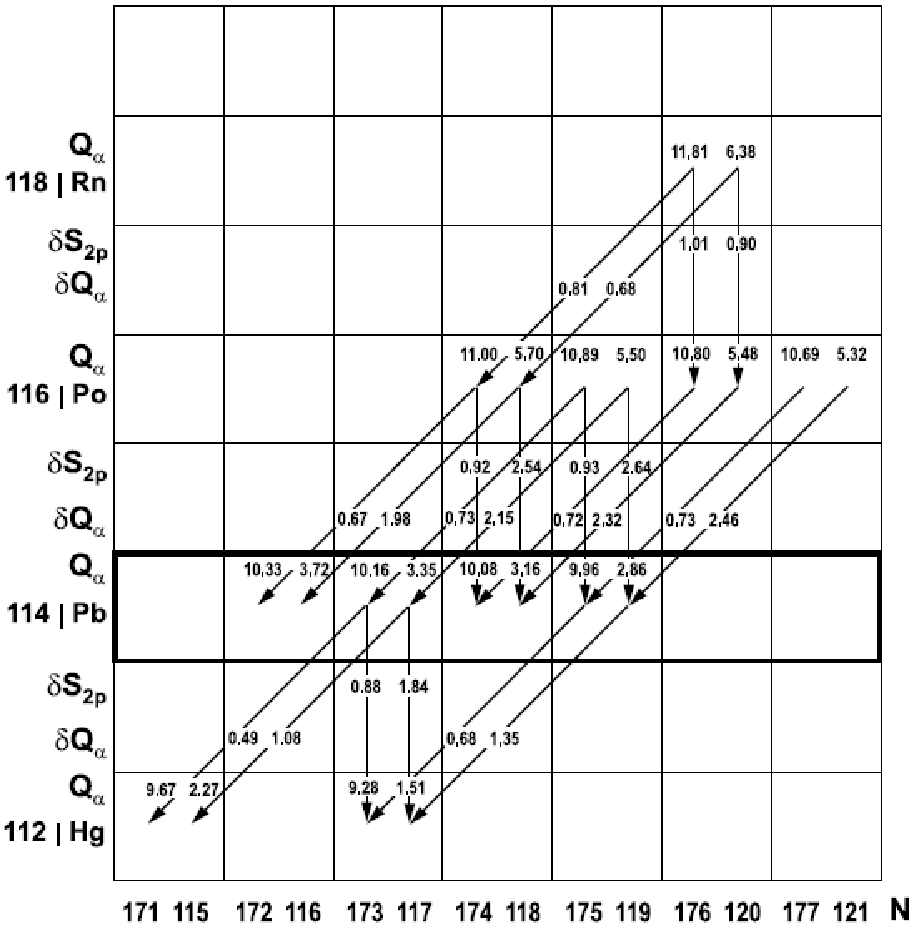
The message:

The 3+ band is built on a two-quasiparticle configuration ($1/2^-[521]\mathrm{x}7/2^-[514])3^+$ of which the $1/2^-[521]$ component stems from the spherical $2f_{5/2}$ proton orbital, the other one from $1h_{9/2}$. Thus, any calculation that gets the 3^+ energy right, also has the $f_{5/2}$ and $h_{9/2}$ spherical orbitals in the right place.



This is especially challenging for the self-consistent models where the high-l orbitals are systematically shifted to too high energies, i.e. the proton $i_{13/2}$ ends up between the $f_{7/2}$ and the $f_{5/2}$ removing 114 as a gap.

P. Armbruster



$\delta\overline{S}_{2n}$ (114) = 0,25 MeV

δS_{2p}^{shell} (114) = (-0.02 ± 0.1) MeV

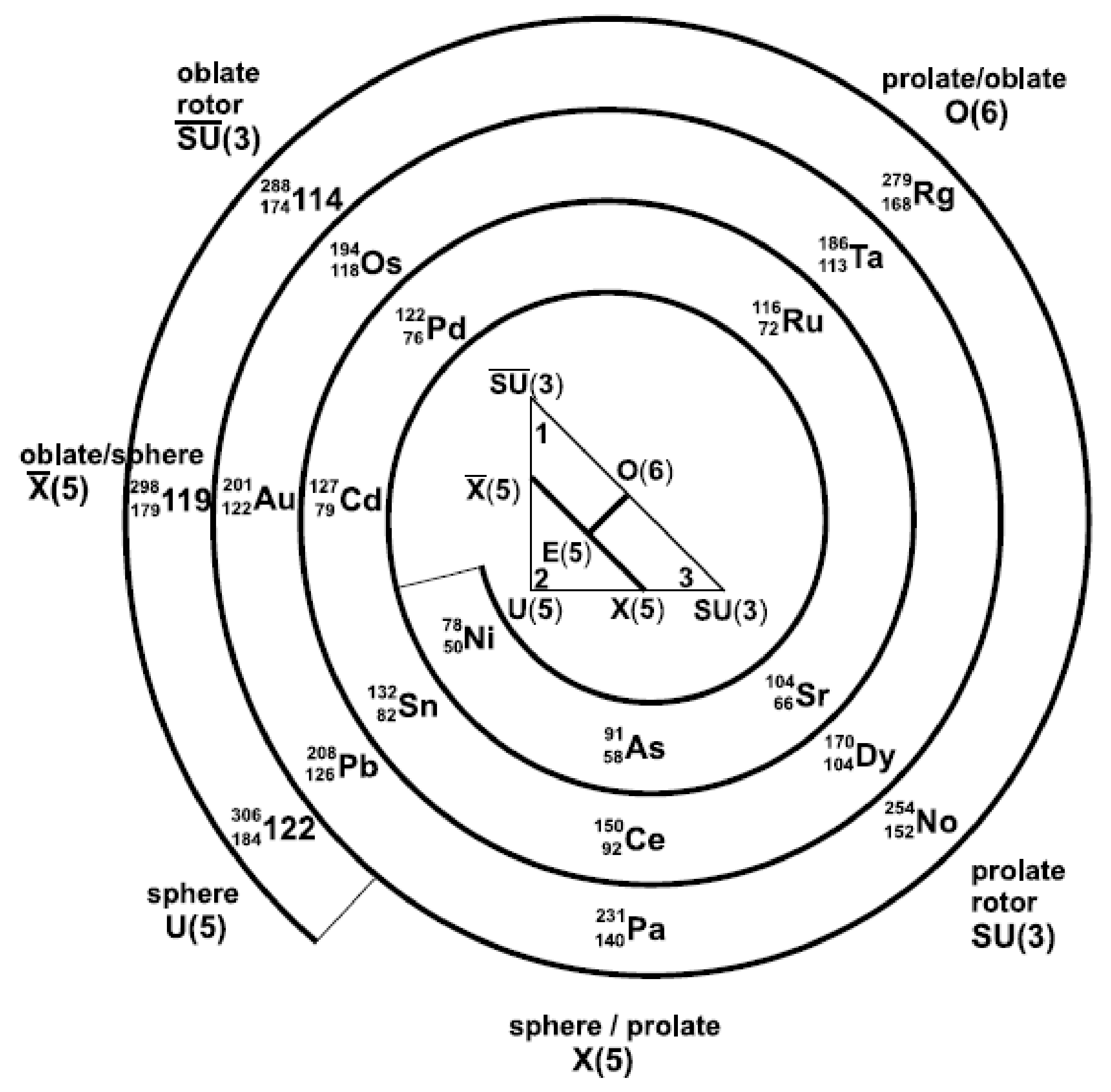
$\delta Q_{\alpha}^{shell}$ (114) = (0,05 ± 0,06) MeV

$\delta\overline{S}_{2n}$ (Pb) = 0,44 MeV

δS_{2p}^{shell} (Pb) = (1.22 ± 0.02) MeV

$\delta Q_{\alpha}^{shell}$ (Pb) = (1.21 ± 0.02) MeV

Interacting Boson Approximation (IBA)

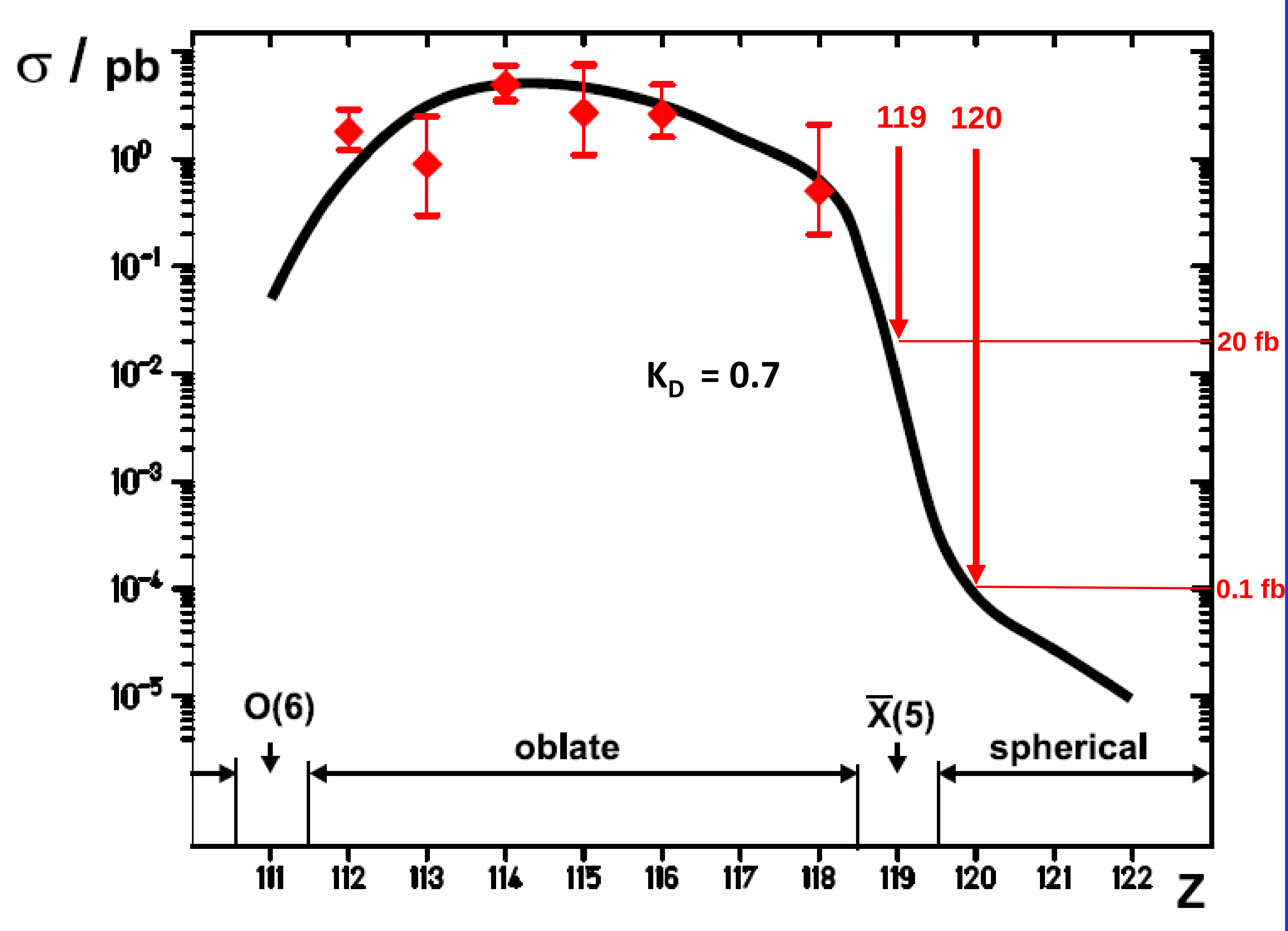


P. Armbruster: Shifting the closed proton shell to Z = 122 – A possible scenario to understand the production of superheavy elements Z =112 – 118, Eur. Phys. J. A37, 159 (2008)

IBA periodicity:

- Next closed proton shell is Z = 122.
- Z = 115 ±3 are oblate.
- Oblate nuclei (Z = 112 – 118) are stabilized against fission by a common gain factor of 10 (p^{shape}).
- For spherical nuclei, collective enhancement of level densities at the saddle point causes a loss factor of 10^{-2} (p^{shape}).
- Damping of shell effects = fission barrier heights with $K_D = \exp(-\gamma/E^*)$ (Ignatyuk)
- $p^{hindrance}(Z) = C \exp[-(0.5/\log e)(Z-Z_0)]$

$$\sigma(Z) = \sigma_{capture} \cdot p^{hindrance} \cdot p^{shape} \cdot W^{survival}(Z)$$


σ / pb
10^0
10^-1
10^-2
10^-3
10^-4
10^-5
119
120
20 fb
0.1 fb
K_D = 0.7
O(6)
oblate
X̄(5)
spherical
111
112
113
114
115
116
117
118
119
120
121
122
Z

Summary:

- **Periodic Table of the Elements**
  - one quarter of all elements are synthetic transuranium elements
  - they have altered the architecture of the Table: actinide and superactinide series

- **Chemical Science**
  - relativistic effects change chemical properties in a given group in a non-linear fashion
  - there are primary, secondary relativistic effects and spin-orbit splitting
  - sub-shell closures give rise to a new category of elements in the Periodic Table: gaseous metals

- **Physical Science**

- shell effects dominate the nuclear structure of transuranium elements
- these give rise to superdeformed shape isomers (fission isomers) in the actinides (U – Bk)
- superheavy elements (Z≥104) are unique elements that owe their existence exclusively to nuclear shell effects @ N = 152, N = 162, and N = 184
- at this time, a building lot is the question for the next spherical proton shell. This urgently needs further theoretical and experimental efforts
- The cross sections for the syntheses of Z=119 and Z=120 will give us important information about the “end of the Periodic Table of the Elements”.

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