High Spin Super- and Hyperdeformed Isomeric States and Long-lived Superheavy Elements

-

Amnon Marinov
The Hebrew University of Jerusalem

Warsaw University
May 12, 2010

Collaborators since 1970:

C.J. Batty	**Rutherford Laboratory**
A.I. Kilvington	**Rutherford Laboratory**
G.W.A. Newton	**Manchester University**
V.J. Robinson	**Manchester University**
J.D. Hemingway	**Research Reactor, Risley**
J.L. Weil	**Kentucky University**
A.M. Friedman	**Argonne National Laboratory**
D.S. Mather	**AWRE, Aldermaston, Berkshire**
S. Eshhar	**The Hebrew University**
D. Kolb	**Kassel University**
S. Gelberg	**The Hebrew University**
A. Pape	**Strasbourg Nuclear Institute**
R. Brandt	**Marburg University**
R.V. Gentry	**Knoxville, Tennessee**
H. W. Miller	**Boulder, Colorado**
L. Halicz	**Geological Survey of Israel**
I. Segal	**Geological Survey of Israel**
I. Rodushkin	**Anlytica, Loleà, Sweden**
Y. Kashiv	**The Hebrew University**

nuclear chart1.jpg

Special thanks I owe to Nissan Zeldes

In the nineteen sixties quite a few theoretical calculations predicted the existence of an Island of Stability around $Z = 114$ and $N = 184$, which supposed to be the next proton and neutron closed shells.

Some of these calculations predicted very long lifetimes for some isotopes in this region of up to $t_{1/2} \approx 10^9$ y

References of some pioneering theoretical works:

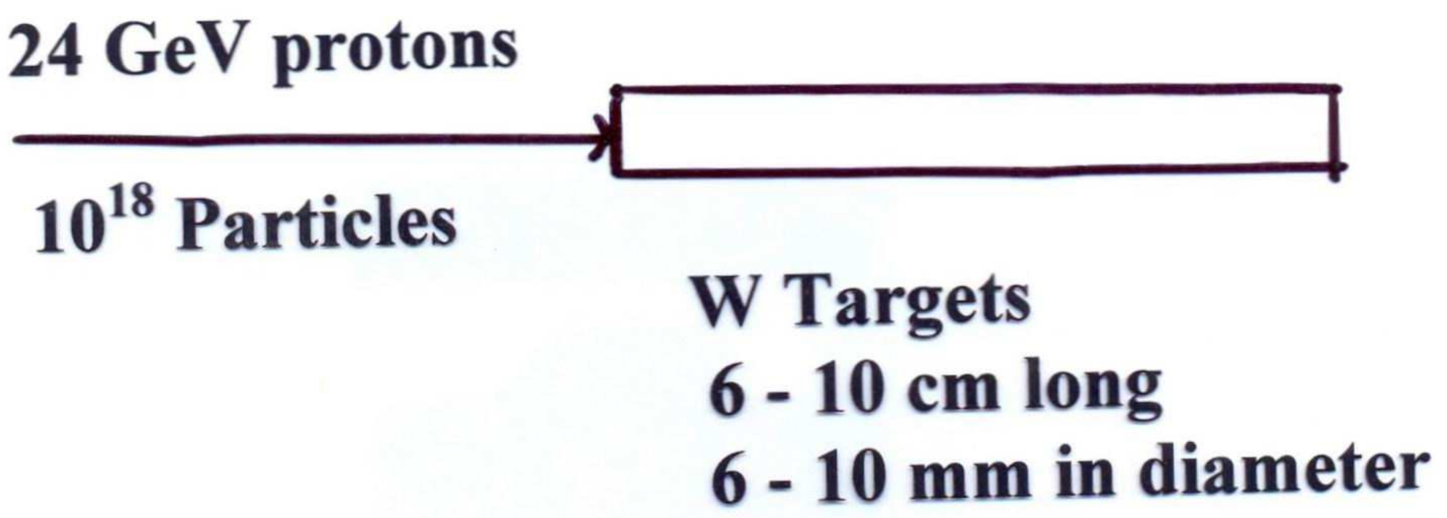
1. **V. M. Strutinskii, Yadernaya fizika 3, 614 (1964).**
2. **W. Myers and W. Swiateski, Nucl. Phys. 81, 1 (1966).**
3. **A. Sobiczewski, F. A. Gareev and B. N. Kalinkin, Phys. Lett. 22, 590 (1966).**
4. **V. M. Strutinskii, Nucl. Phys. A95, 420 (1967).**
5. **C. L. Wong, Phys. Rev. Lett. 19, 328 (1967).**
6. **Yu. A. Muzychka, V. V. Pashkevich and Strutinskii, Dubna Preprint R7-3733, 1968.**
7. **S. G. Nilsson, J. R. Nix, A. Sobiczewski, Z. Szymanski, S. Wycech, C. Gustafson and P. Möller, Nucl. Phys. A115, 545 (1968).**
8. **J. Grumann, U. Mosel, B. Fink and W. Greiner, Z. Physik 228, 371 (1969).**

These predictions excited the nuclear community and three paths of research have begun.

a) People started to build and to upgrade their heavy ion accelerators: GSI, Berkeley, Dubna and recently Riken in Japan.

b) People searched for the existence of superheavy elements (SHE) in various natural materials. The results of most of these searches were negative, but some unexplained phenomena were observed, which I will discuss later on.

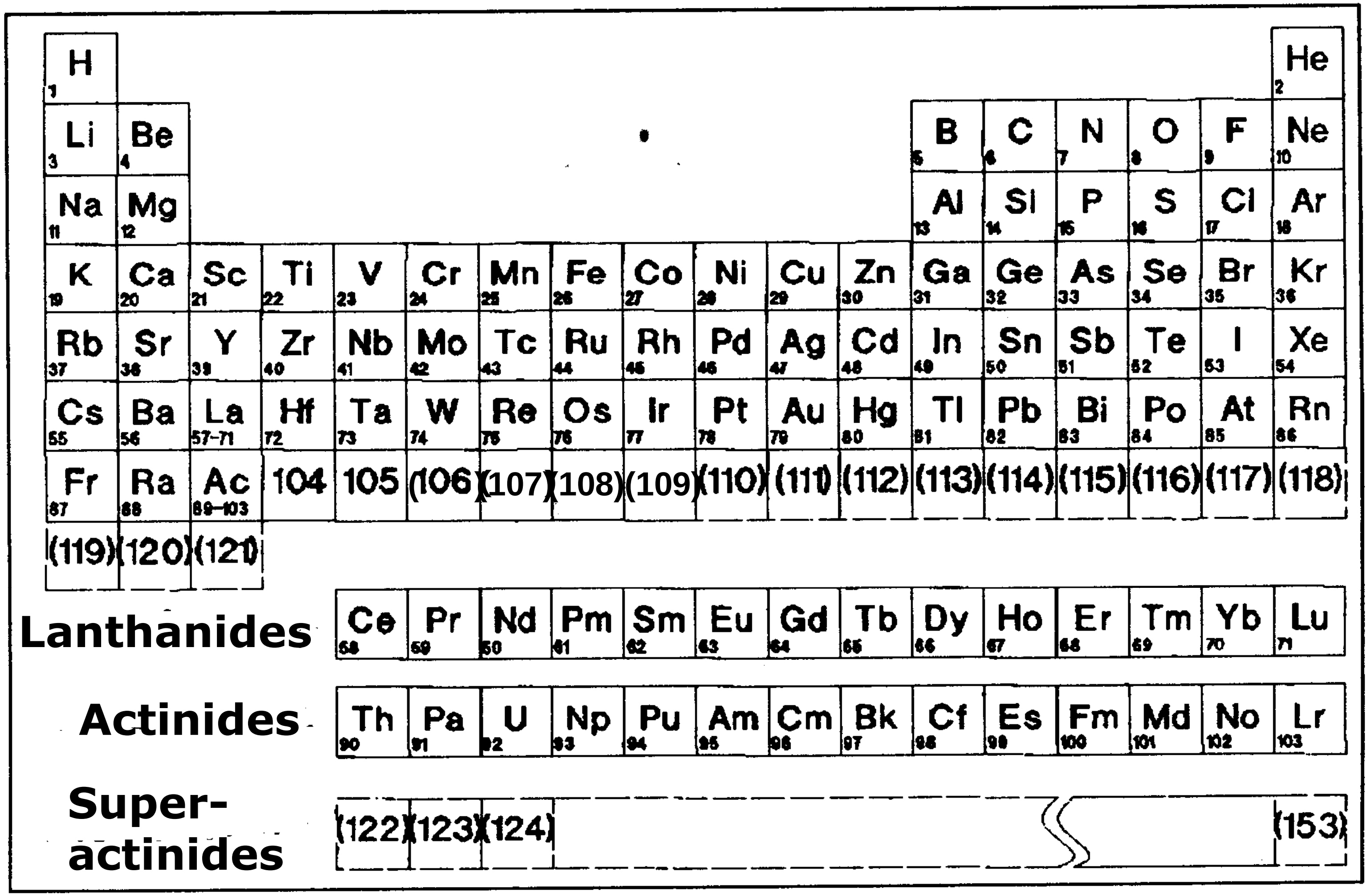
c) Our approach was to try to produce the SHE by Secondary Reaction Experiments.



Many fission and spallation fragments are produced. Some of them may perhaps have enough kinetic energy to overcome the Coulomb barrier between them and another W nucleus in the target, and via these secondary reactions produce the SHE.

In order to find the SHE we have first to separate them from the W target, and then to study their radioactive decay properties, and in particular spontaneous fission decays that are limited to very heavy nuclei.

For the separation of the SHE from the W we have to rely on the predicted chemical properties:



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	I 53	Xe 54
Cs 55	Ba 56	La 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 89-103	104	105	(106)	(107)	(108)	(109)	(110)	(111)	(112)	(113)	(114)	(115)	(116)	(117)	(118)
(119)	(120)	(121)															
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
Super-actinides				(122)	(123)	(124)											(153)

Element 114 is eka-Pb

Element 113 is eka-Tl

Element 112 is eka-Hg

Element 111 is eka-Au

Element 110 is eka-Pt

We dissolved the W target, added to it 40 μg of Pb, Tl, Hg, Au and Pt, separated them again, hoping that eka-Pb will follow the chemistry of Pb, eka-Hg (element 112) will follow the chemistry of Hg, and so on.

In two Hg sources separated from two W targets we saw fission fragments.

All together we saw about 100 fission events.

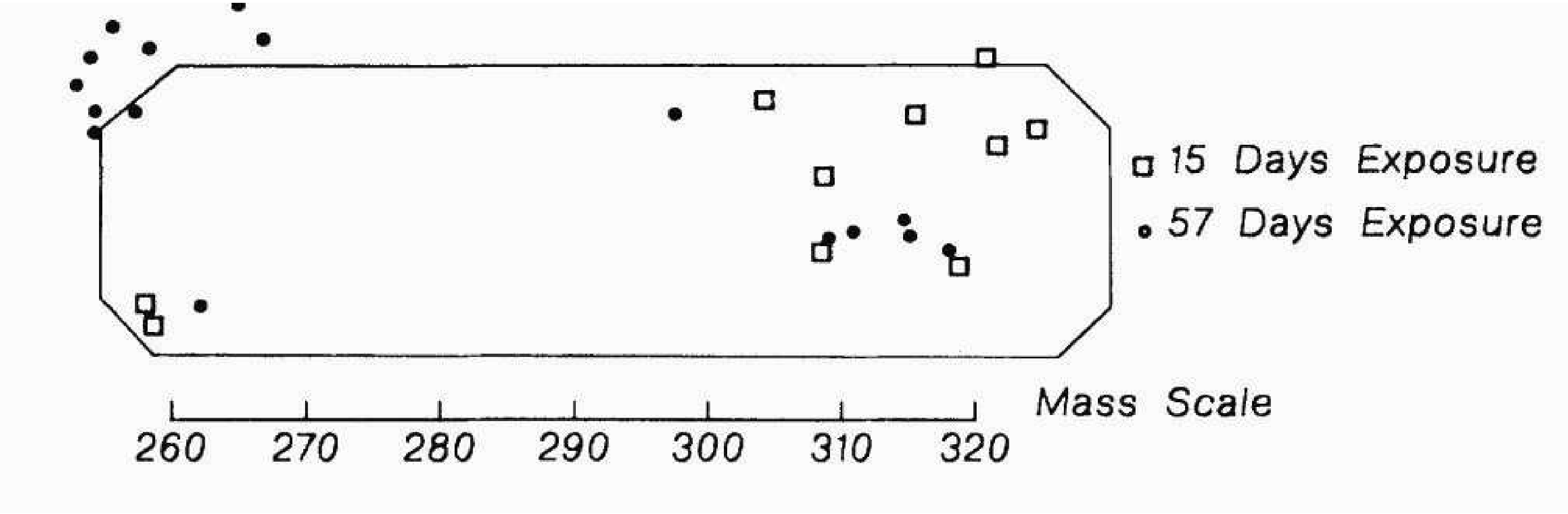
The background in these measurements was zero.

Nature 229, 464 (1971)
Nature 234, 212 (1971)

Mass Measurements

Part of the Hg source was run through a mass separator. We collected the analyzed atoms and molecules on a thin Ni foil in the focal plan of the mass separator.

We then looked for fission fragments emitted from this Ni foil using polycarbonate foils.



Species with very high masses like 308, 315, 318 that decay by fission were seen and repeated themselves several times. (Mass 317-318 repeated itself four times in four different exposures).

PRL 52, 2209 (1984)

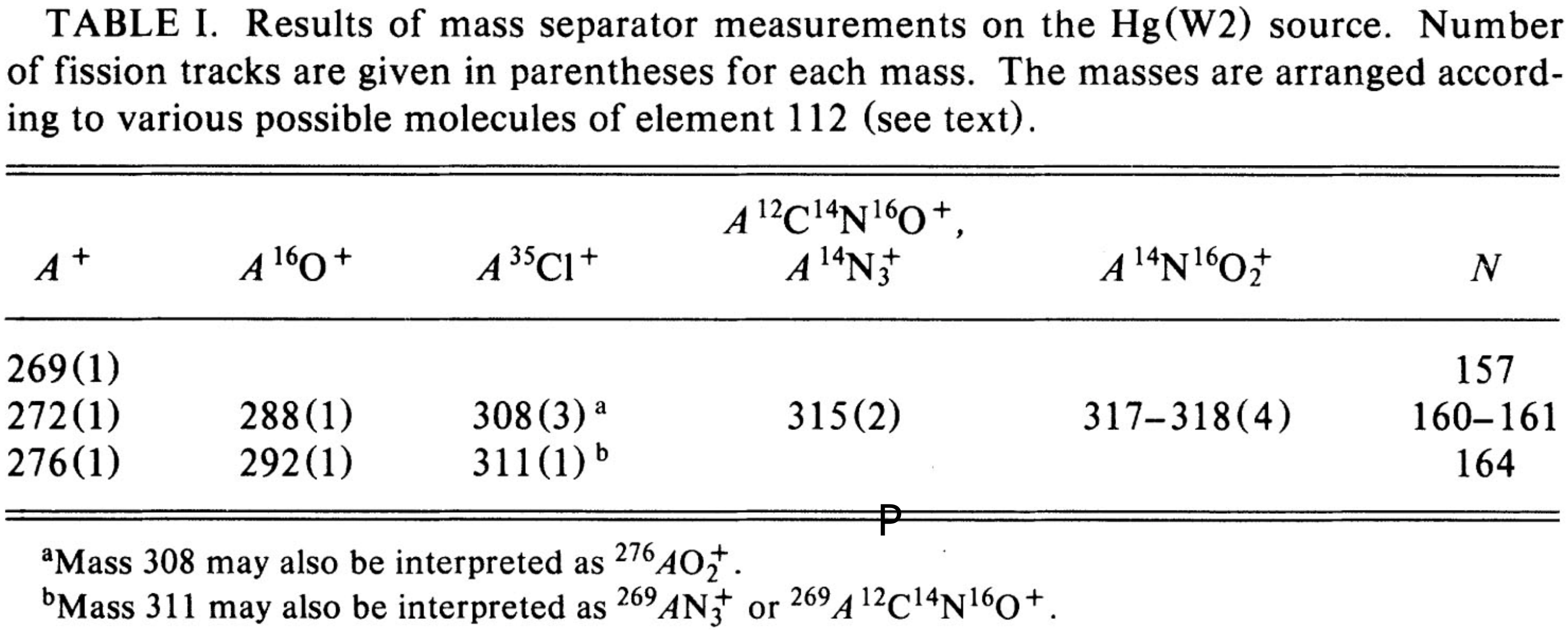


TABLE I. Results of mass separator measurements on the Hg(W2) source. Number of fission tracks are given in parentheses for each mass. The masses are arranged according to various possible molecules of element 112 (see text).

A^+	$A^{16}O^+$	$A^{35}Cl^+$	$A^{12}C^{14}N^{16}O^+$, $A^{14}N_3^+$	$A^{14}N^{16}O_2^+$	N
269(1)					157
272(1)	288(1)	308(3) [a]	315(2)	317–318(4)	160–161
276(1)	292(1)	311(1) [b]			164

P

[a] Mass 308 may also be interpreted as $^{276}AO_2^+$.

[b] Mass 311 may also be interpreted as $^{269}AN_3^+$ or $^{269}A\,^{12}C^{14}N^{16}O^+$.

PRL 52, 2209 (1984)

It is seen that 11 events can be arranged as the atom with Z = 112 and A = 272 and 4 different molecules of it.

The estimated half-life of the fission activity

$t_{1/2} \approx$ several weeks.

Deduced possible reactions:

$^{88}Sr + ^{184}W \longrightarrow ^{272}112 \quad Q_{value} = -282.4$ MeV

$^{86}Sr + ^{186}W \longrightarrow ^{272}112 \quad Q_{value} = -275.5$ MeV

Coulomb Barrier: 285 MeV

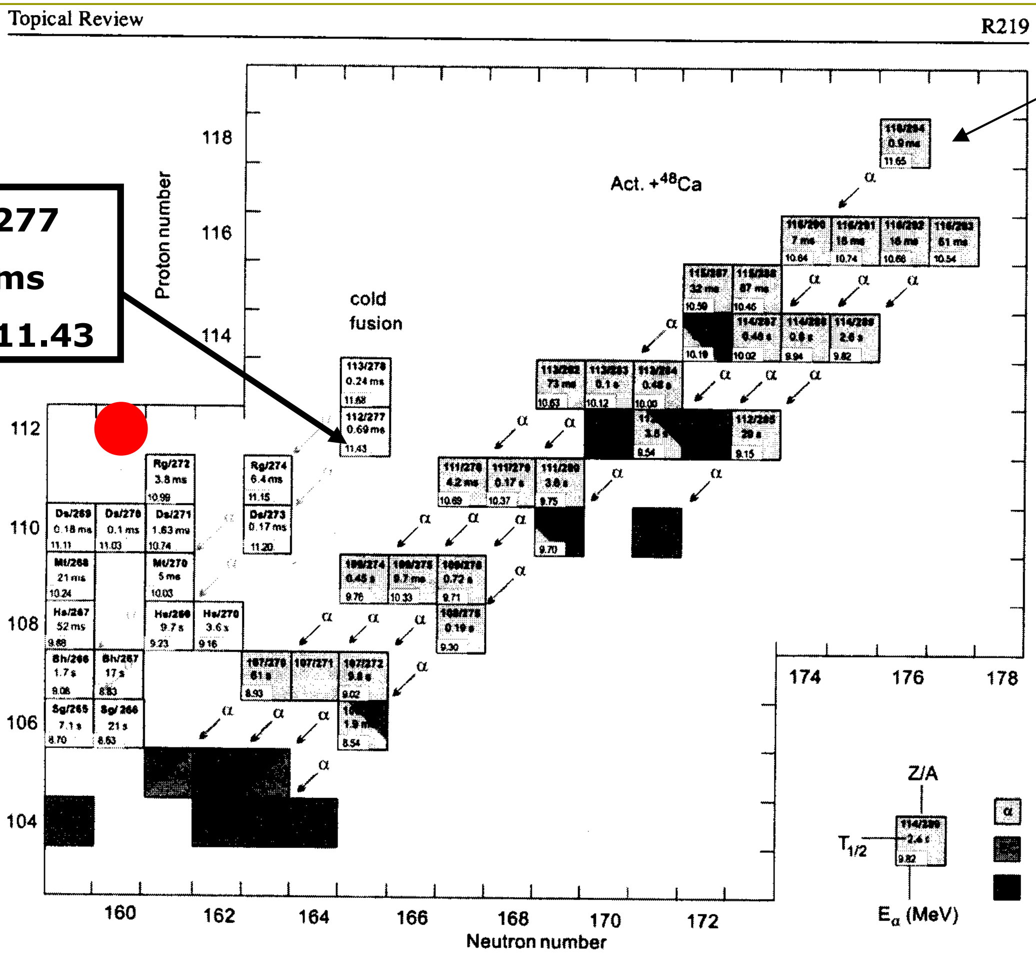
Deduced cross sections

$\sigma_{total}(^{88}Sr) \approx 1$ mb

Extrapolation from 5.5 GeV protons on U (Poskanzer, Butler and Hyde) we estimated that:

Only 5x10^{-5} of them will have enough kinetic energy to overcome the Coulomb barrier.

$\sigma_{fus}(Sr+W) \approx 4$ mb



112/277
0.69 ms
Eα = 11.43

118/294
0.8 ms
Eα = 11.65 s

Yu. Oganessian,
J. Phys. G:
Nucl. Part. Phys.
39, R165 (2007)

The typical cross sections are about 1 pb and the typical lifetimes are about ms.

Problems:

a) $t_{1/2}$ 10^9 too long

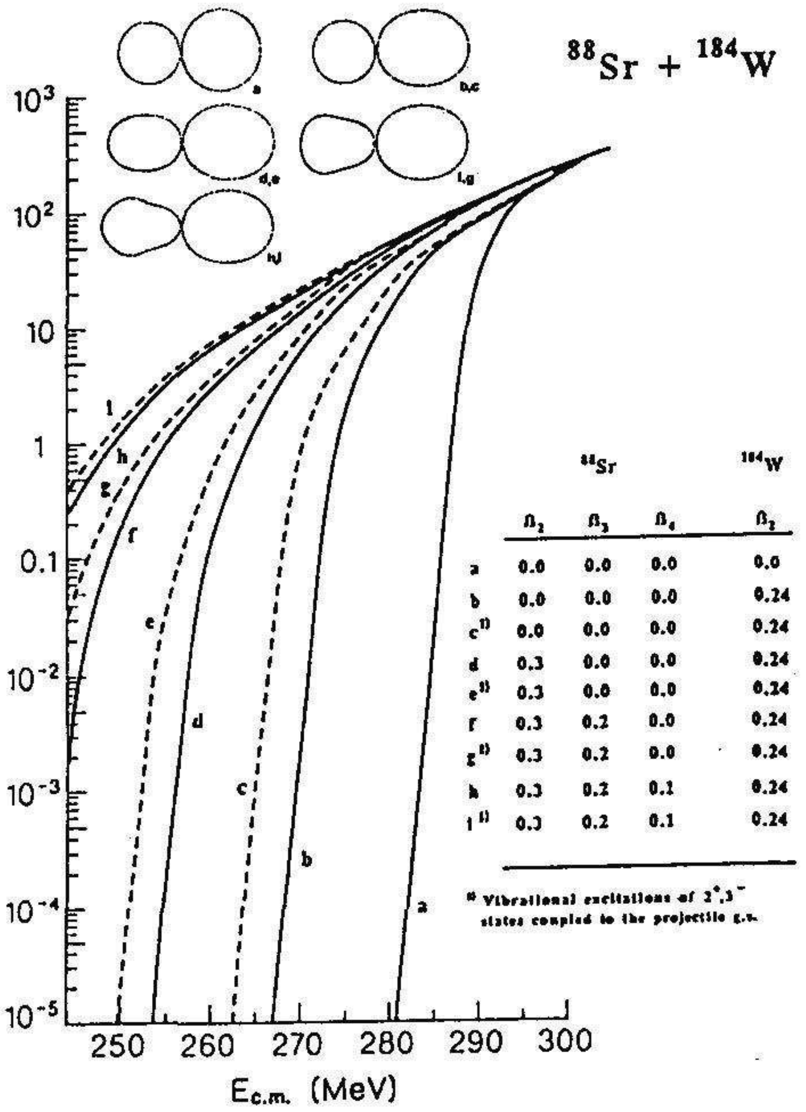
b) σ_{fus} 10^9 too large

a) Deformations:

The projectiles in the secondary reactions are not normal nuclei in their g.s. They are highly excited deformed fragments that were produced just about 5×10^{-14} s before interacting with another W nucleus in the target.

Deformations have strong effect on the fusion cross sections as was first seen by

Fusion of $^{16}O+^{148,150,152,154}Sm$ at sub-barrier energies,
R. G. Stokstad, Y. Eisen, S. Kaplanis, D. Pelte, U. Smilansky and I. Tserruya, Phys. Rev. C 21 (1980) 2427-2435.



	^{88}Sr			^{184}W
	β_2	β_3	β_4	β_2
a	0.0	0.0	0.0	0.0
b	0.0	0.0	0.0	0.24
c[1]	0.0	0.0	0.0	0.24
d	0.3	0.0	0.0	0.24
e[1]	0.3	0.0	0.0	0.24
f	0.3	0.2	0.0	0.24
g[1]	0.3	0.2	0.0	0.24
h	0.3	0.2	0.1	0.24
i[1]	0.3	0.2	0.1	0.24

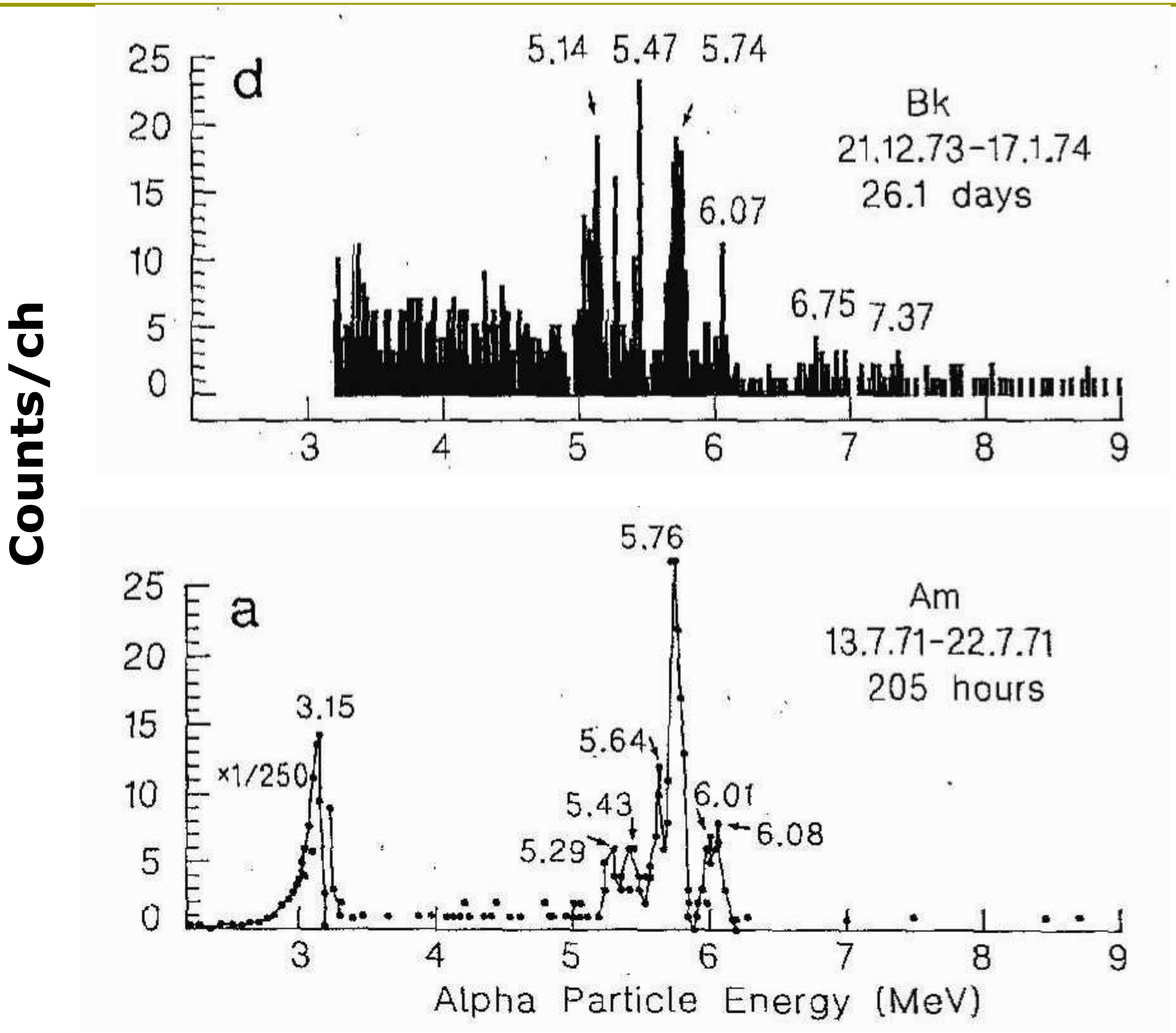
[1] Vibrational excitations of $2^+, 3^-$ states coupled to the projectile g.s.

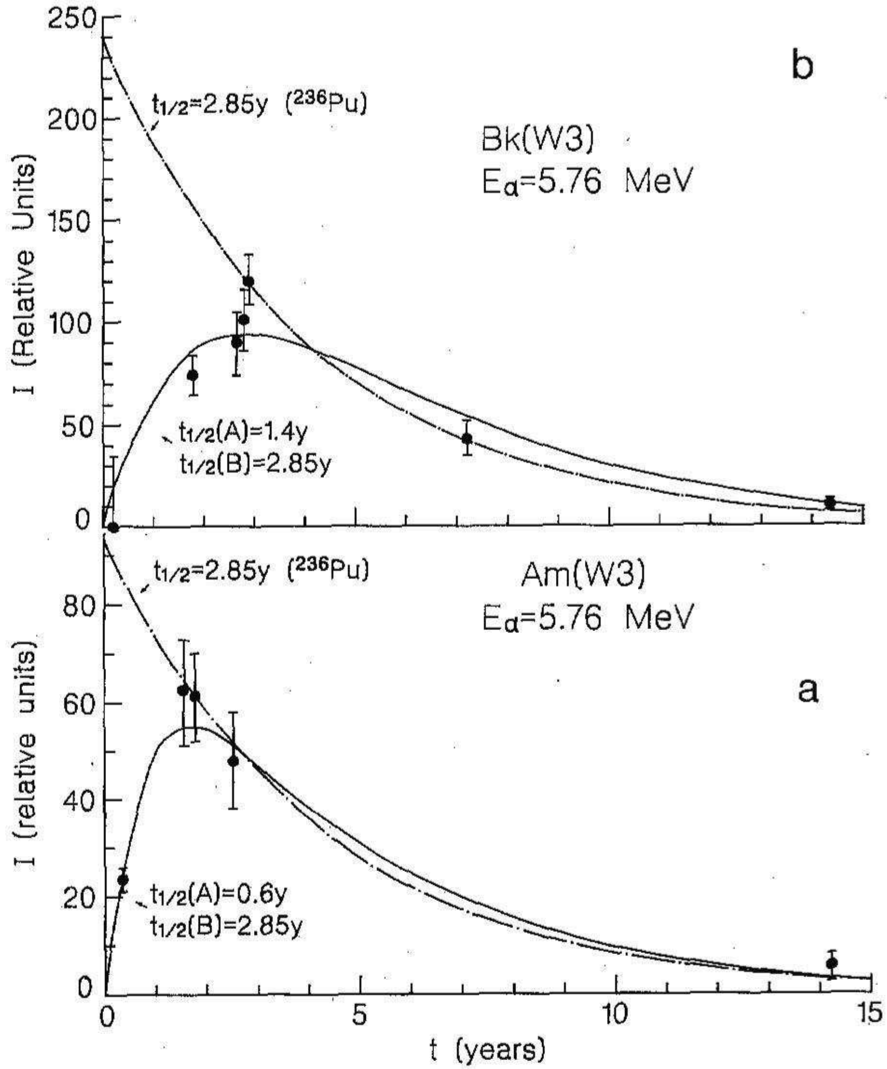
This can explain 4 to 5 orders of magnitude.

We are still missing about 4-5 orders of magnitude in the fusion cross section.

Actinides

In parallel we studied actinides that were separated from the W target.


Counts/ch
d
5.14
5.47
5.74
Bk
21.12.73–17.1.74
26.1 days
6.07
6.75
7.37
0
5
10
15
20
25
3
4
5
6
7
8
9
a
5.76
Am
13.7.71–22.7.71
205 hours
3.15
×1/250
5.64
5.43
6.01
5.29
6.08
0
5
10
15
20
25
3
4
5
6
7
8
9
Alpha Particle Energy (MeV)



The intensity of the 5.76 MeV group is first growing and then decayed with the characteristic half-life of ^{236}Pu.

Phys. Letters 191B (1987) 36-40


98
97
96
Bk
Cm
42,5 s
6.660
3,2 m
6.776
ε = 5%
10,3 m
6.456
Bk 238
144 s
Bk 240
5 m
Bk 242
7 m
Bk 243
4,5 h
Cm 238
2,4 h
Cm 239
3 h
Cm 240
27 d
Cm 241
32,8 d
Cm 242
162,94 d
Am 232
1,31 m
Am 234
2,32 m
Am 236 ?
3,7 m
3,6 m
Am 237
73,0 m
Am 238
1,63 h
Am 239
11,9 h
Am 240
50,8 h
Am 241
432,2 a
Pu 232
34,1 m
Pu 233
20,9 m
Pu 234
8,8 h
Pu 235
25,3 m
Pu 236
2,858 a
Pu 237
45,2 d
Pu 238
87,74 a
Pu 239
2,411 · 10⁴ a
Pu 240
6563 a
Np 230
4,6 m
Np 231
48,8 m
Np 232
14,7 m
Np 233
36,2 m
Np 234
4,4 d
Np 235
396,1 d
Np 236
Np 237
2,144 · 10⁶ a
Np 238
2,117 d
Np 239
2,355 d
U 229
58 m
U 230
20,8 d
U 231
4,2 d
U 232
68,9 a
U 233
1,592 · 10⁵ a
U 234
0,0055
2,455 · 10⁵ a
U 235
0,7200
U 236
U 237
6,75 d
U 238
99,2745
Pa 228
22 h
Pa 229
1,50 d
Pa 230
17,4 d
Pa 231
3,276 · 10⁴ a
Pa 232
1,31 d
Pa 233
27,0 d
Pa 234
Pa 235
24,2 m
Pa 236
9,1 m
Pa 237
8,7 m
Th 227
18,72 d
Th 228
1,913 a
Th 229
7880 a
Th 230
7,54 · 10⁴ a
Th 231
25,5 h
Th 232
100
1,405 · 10¹⁰ a
Th 233
22,3 m
Th 234
24,10 d
Th 235
7,1 m
Th 236
37,5 m
Ac 226
Ac 227
Ac 228
Ac 229
Ac 230
Ac 231
Ac 232
Ac 233
Ac 234
Pu 236
2,858 a
sf
α 5,768; 5,721…
sf; Mg 28
γ (48; 109…); e⁻
σf 160

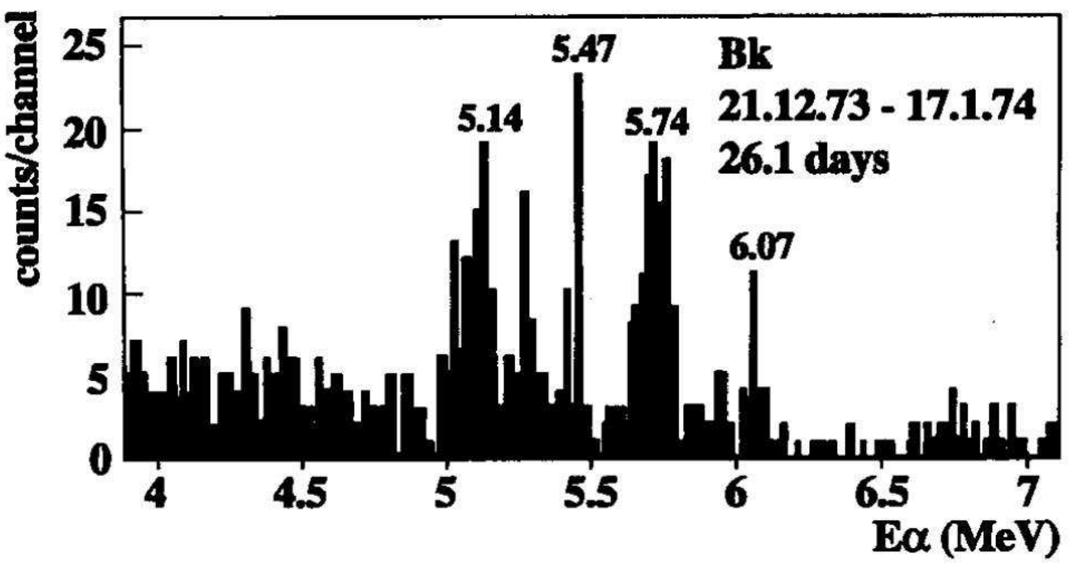
Conclusion

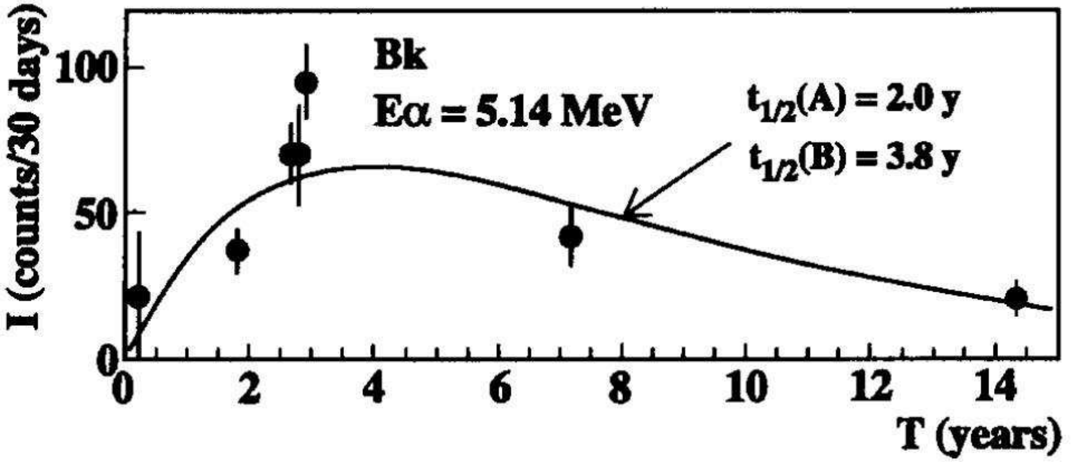
Long-lived isomeric states produced in the neutron-deficient nuclei ^{236}Am and ^{236}Bk with half-lives of about 5 orders of magnitude longer than their corresponding g.s.

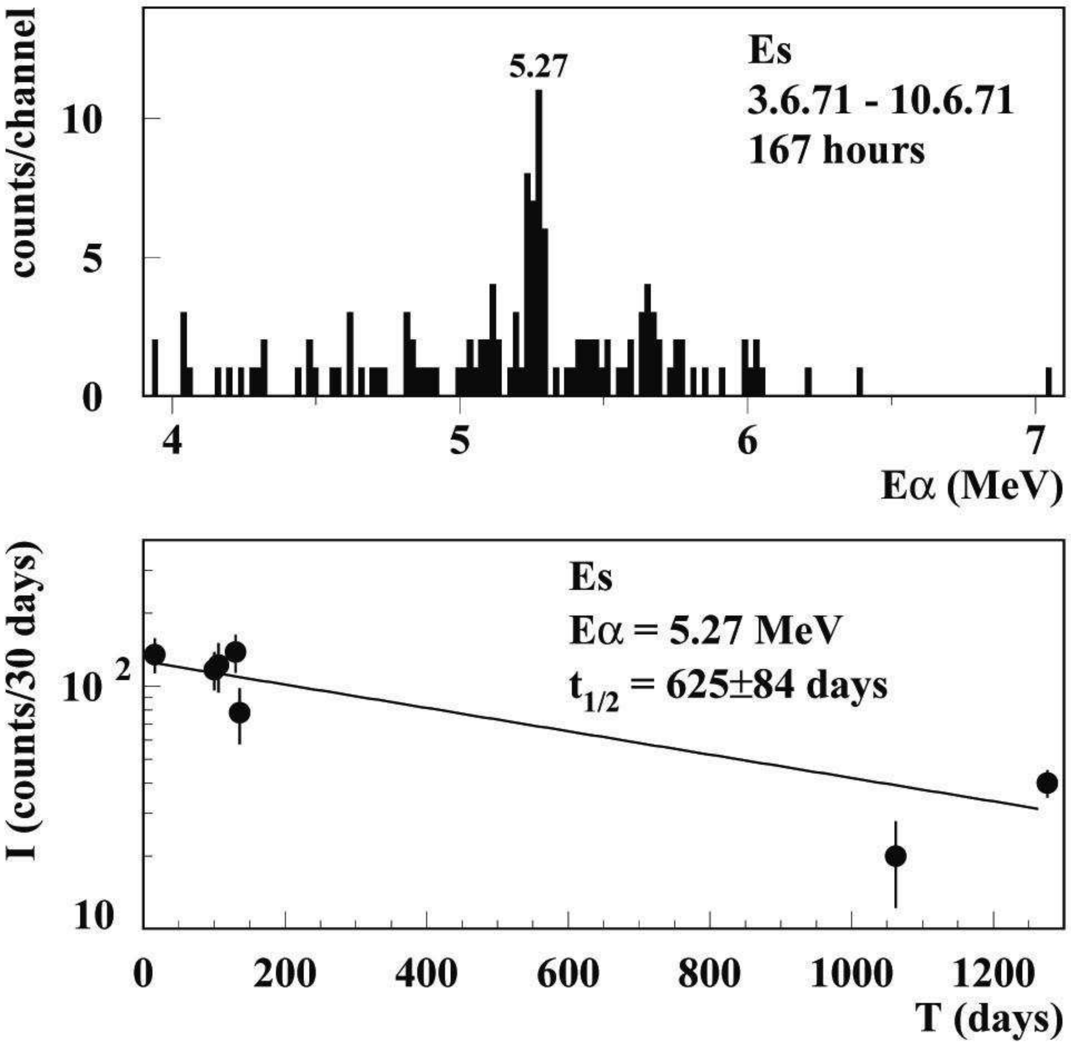
One may assume that like in the actinides a long-lived isomeric state was produced in element 112.

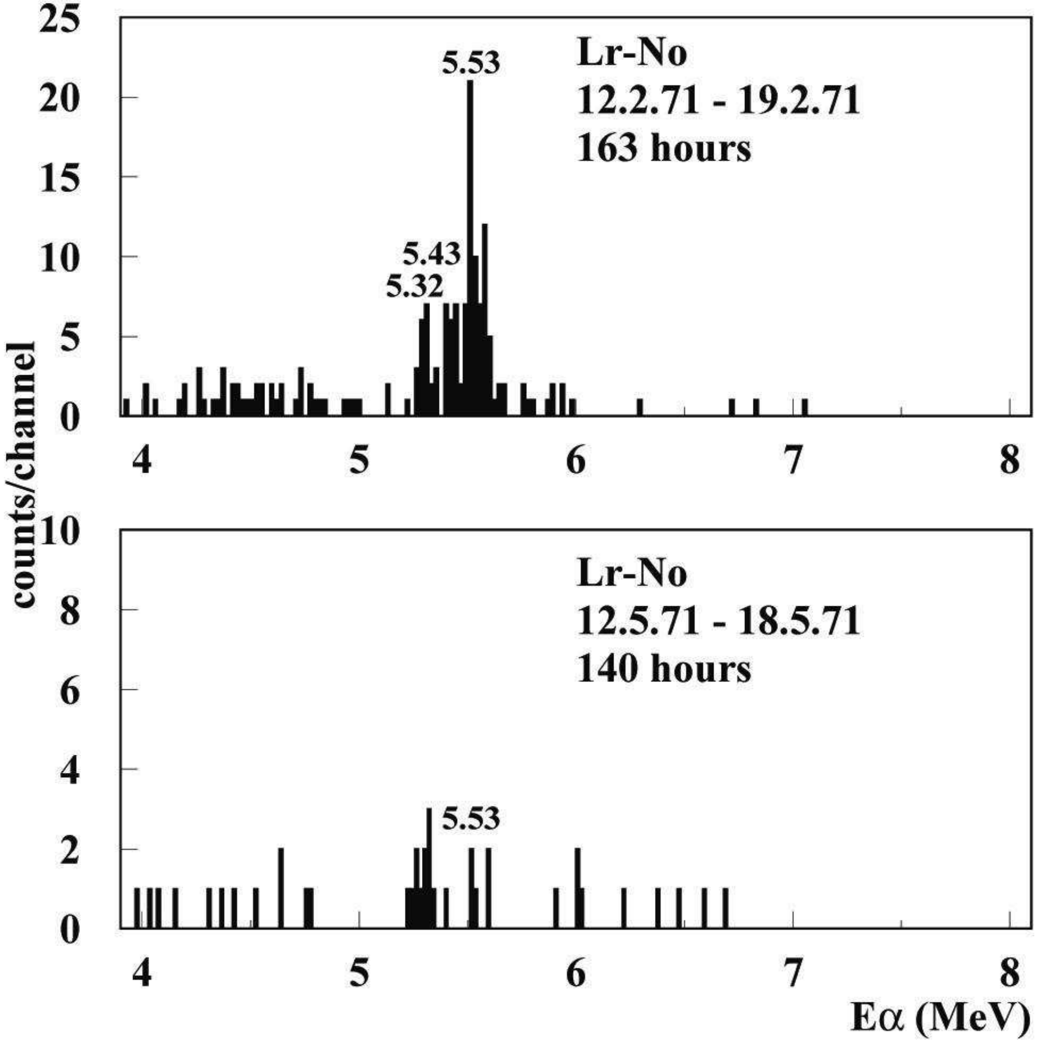
We still have the problem of the cross section.

In the actinide spectra we saw other α - particle groups.


counts/channel
25
20
15
10
5
0
5.47
5.14
5.74
6.07
Bk
21.12.73 - 17.1.74
26.1 days
4
4.5
5
5.5
6
6.5
7
Eα (MeV)


I (counts/30 days)
100
50
0
Bk
Eα = 5.14 MeV
$t_{1/2}$(A) = 2.0 y
$t_{1/2}$(B) = 3.8 y
0
2
4
6
8
10
12
14
T (years)


counts/channel
10
5
0
5.27
Es
3.6.71 - 10.6.71
167 hours
4
5
6
7
Eα (MeV)
I (counts/30 days)
Es
Eα = 5.27 MeV
t1/2 = 625±84 days
10 2
10
0
200
400
600
800
1000
1200
T (days)



counts/channel
Lr-No
12.2.71 - 19.2.71
163 hours
5.53
5.43
5.32
Lr-No
12.5.71 - 18.5.71
140 hours
5.53
25
20
15
10
5
0
10
8
6
4
2
0
4
5
6
7
8
Eα (MeV)

It is impossible to identify these α-particle groups with any known activity in the whole nuclear chart.

In addition they do not fit with the systematic of α-particles.

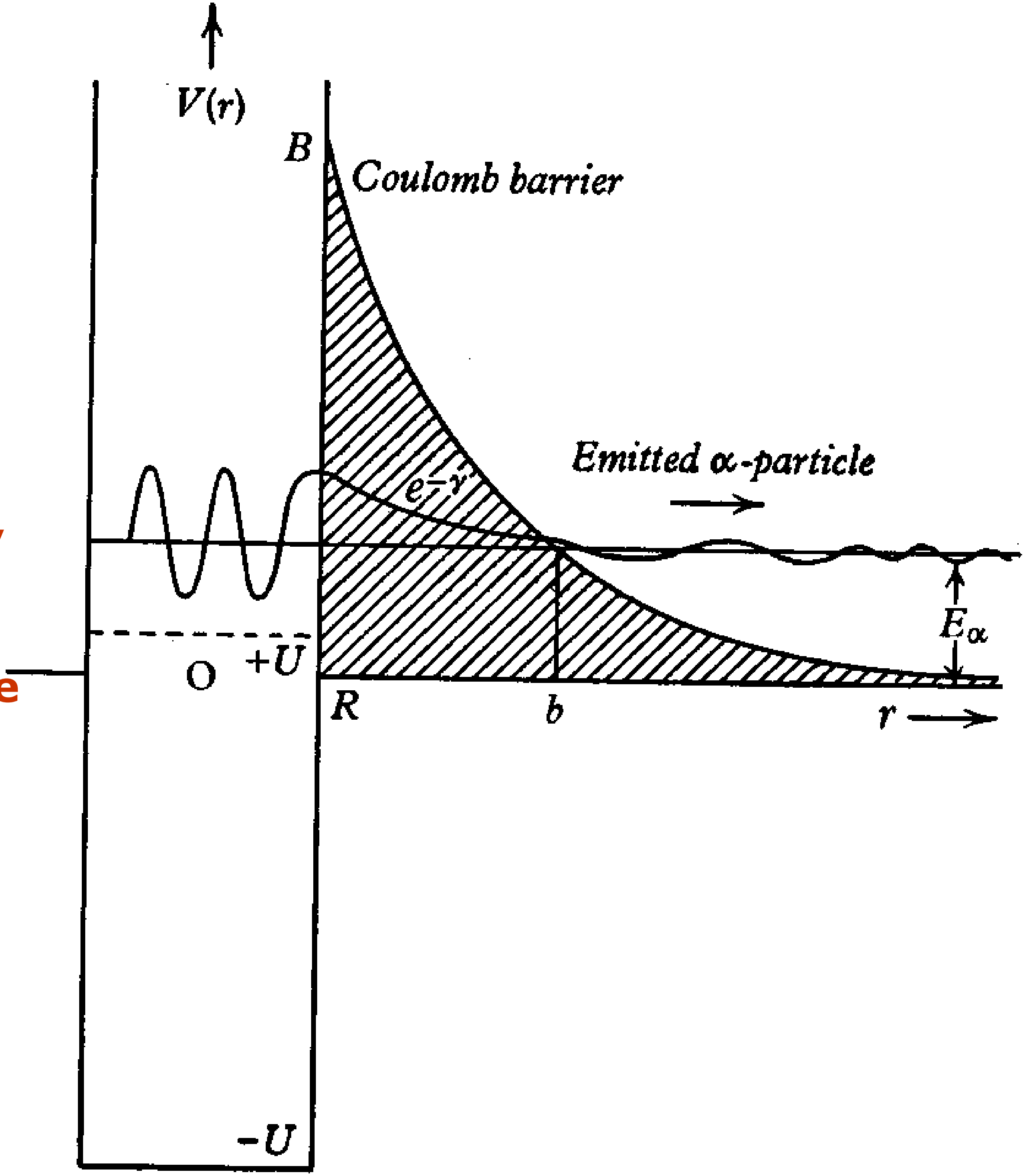
^{238}U: Eα = 4.2 MeV

$t_{1/2}$ = 4.5x10^{9} y

^{212}Po: Eα = 8.8 MeV

$t_{1/2}$ = 0.3 μs

Factor of 2 in energy corresponds to 23 orders of magnitude in lifetime



First: The energies are too low

Source	$E\alpha$ (MeV)	$t_{1/2}/s$ (Cal.)	Typical $E\alpha$ (MeV)	$t_{1/2}/s$ (Cal.)
Bk (Cm,Am)	5.14	2×10^{12}	6 - 7	2×10^{5}
Es	5.27	3×10^{13}	7 - 8	5×10^{2}
No-Lr	5.53	2×10^{13}	8 - 9	1×10^{1}

Second: These α-particles pass the Coulomb barrier too fast.

Source	$E\alpha$	$t_{1/2}/y$ (Cal.)	$t_{1/2}$ (Exp.)	Enhancement Factor
Bk (Cm,Am)	5.14	1.6×10^5	3.8 y	4.2×10^4
Es	5.27	9.5×10^5	625 d	5.5×10^5
No-Lr	5.53	6.3×10^5	26 d	8.8×10^6

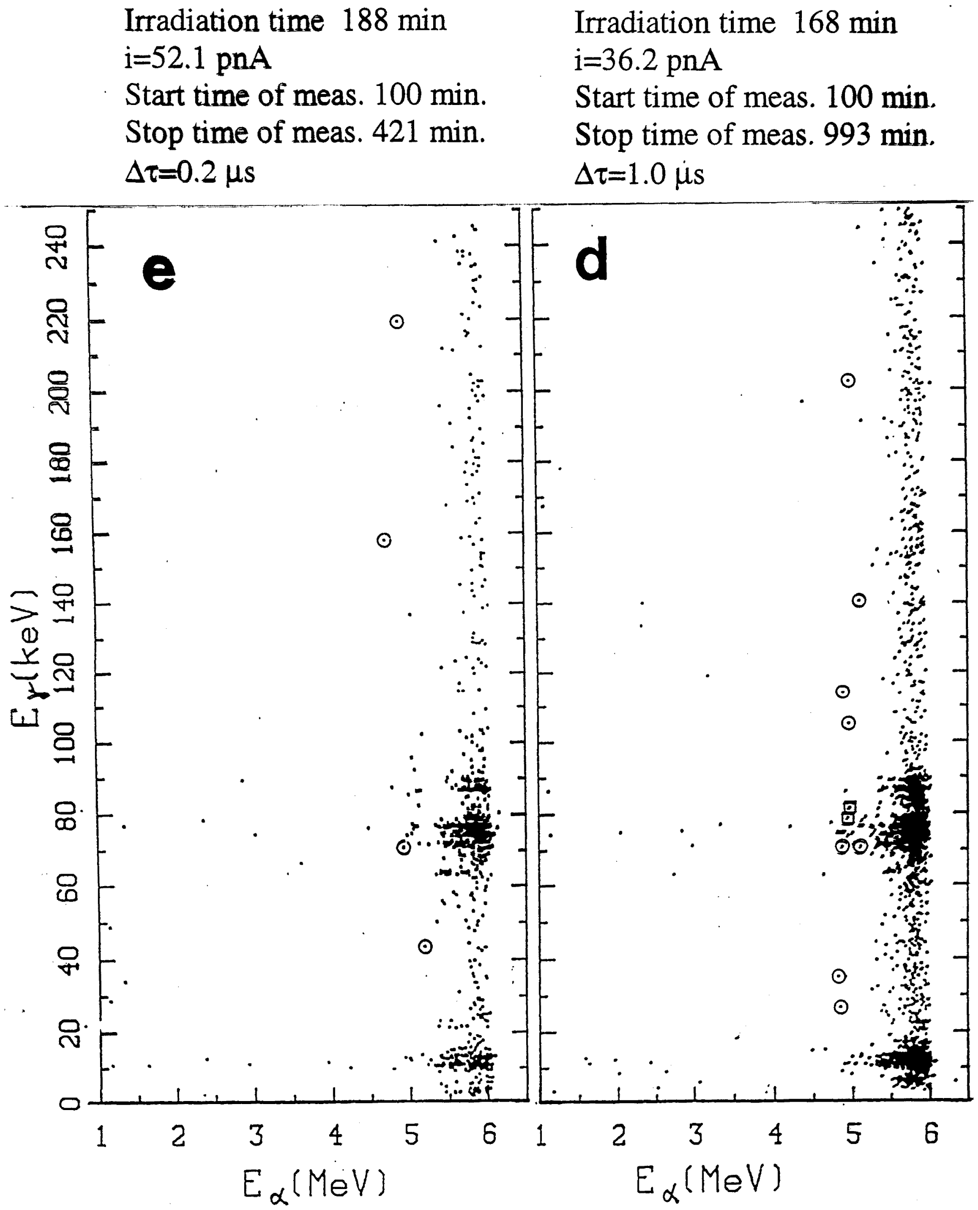
The answer to these problems was obtained from two experiments we did in Rehovot using the pelletron accelerator:

a) ^{16}O + ^{197}Au at E_{Lab} = 80 MeV; CN = ^{213}Fr

b) ^{28}Si + ^{181}Ta at E_{Lab} = 125 MeV; CN = ^{209}Fr

(This is about 10% below the Coulomb barrier)

We used catcher foil technique and measured α-γ coincidences from the catcher foil.


Irradiation time 188 min
i=52.1 pnA
Start time of meas. 100 min.
Stop time of meas. 421 min.
$\Delta\tau=0.2\ \mu s$

Irradiation time 168 min
i=36.2 pnA
Start time of meas. 100 min.
Stop time of meas. 993 min.
$\Delta\tau=1.0\ \mu s$

$^{16}O + ^{197}Au$

$E_{Lab} = 80$ MeV

We used catcher foil technique and measured off-line α-γ coincidences from the catcher foil

We found 5.2 MeV α-particle group in coincidence with various γ-rays. ($\sigma \approx 30$ nb)

It was identified as a transition from

^{210}Fr to ^{206}At

Eα for g.s. to g.s. transition is 6.54 MeV.

($t_{1/2}$ =3.18 m)

Why it decays with low energy when much higher energy is available?

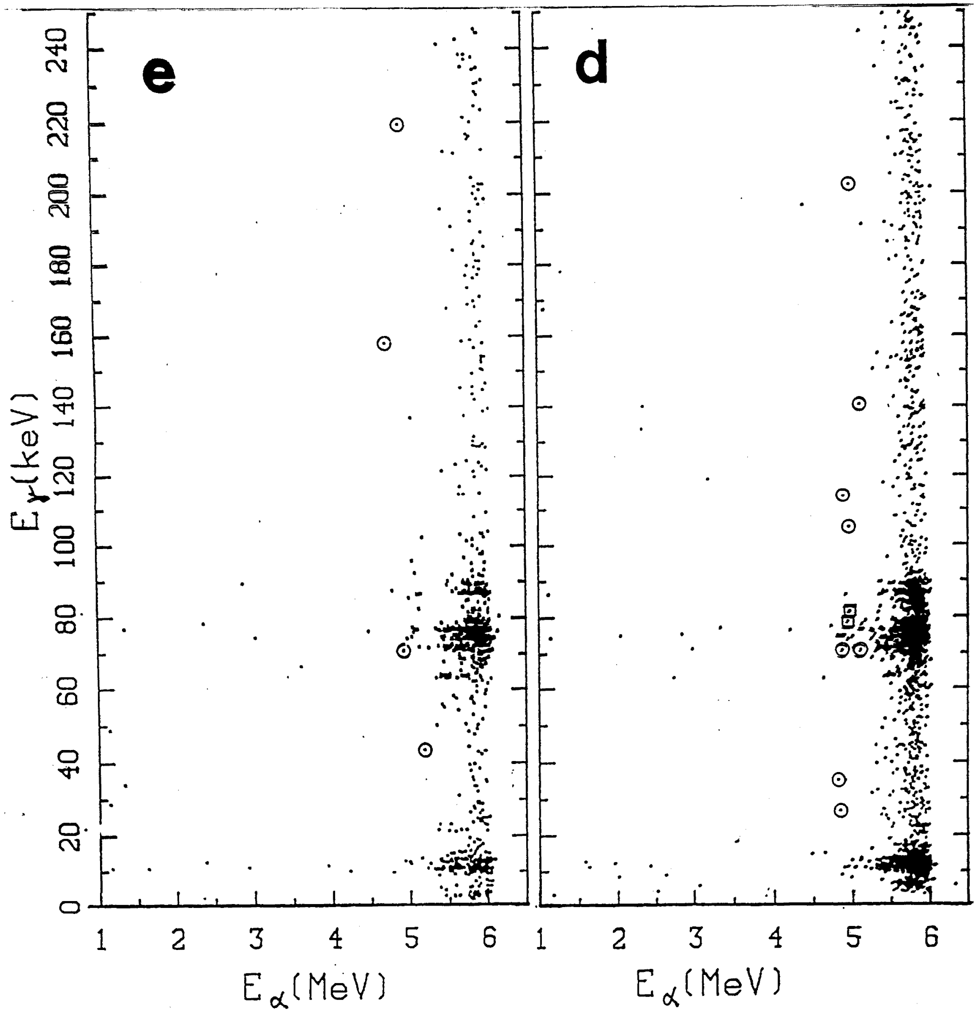
$t_{1/2}$ (Exp.) $\approx$ 90 m

$t_{1/2}$ (Cal.) =51 y

It is enhanced by a factor of 3×10^5

Irradiation time 188 min
i=52.1 pnA
Start time of meas. 100 min.
Stop time of meas. 421 min.
Δτ=0.2 μs

Irradiation time 168 min
i=36.2 pnA
Start time of meas. 100 min.
Stop time of meas. 993 min.
Δτ=1.0 μs



^{16}O + ^{197}Au E_{Lab} = 80 MeV

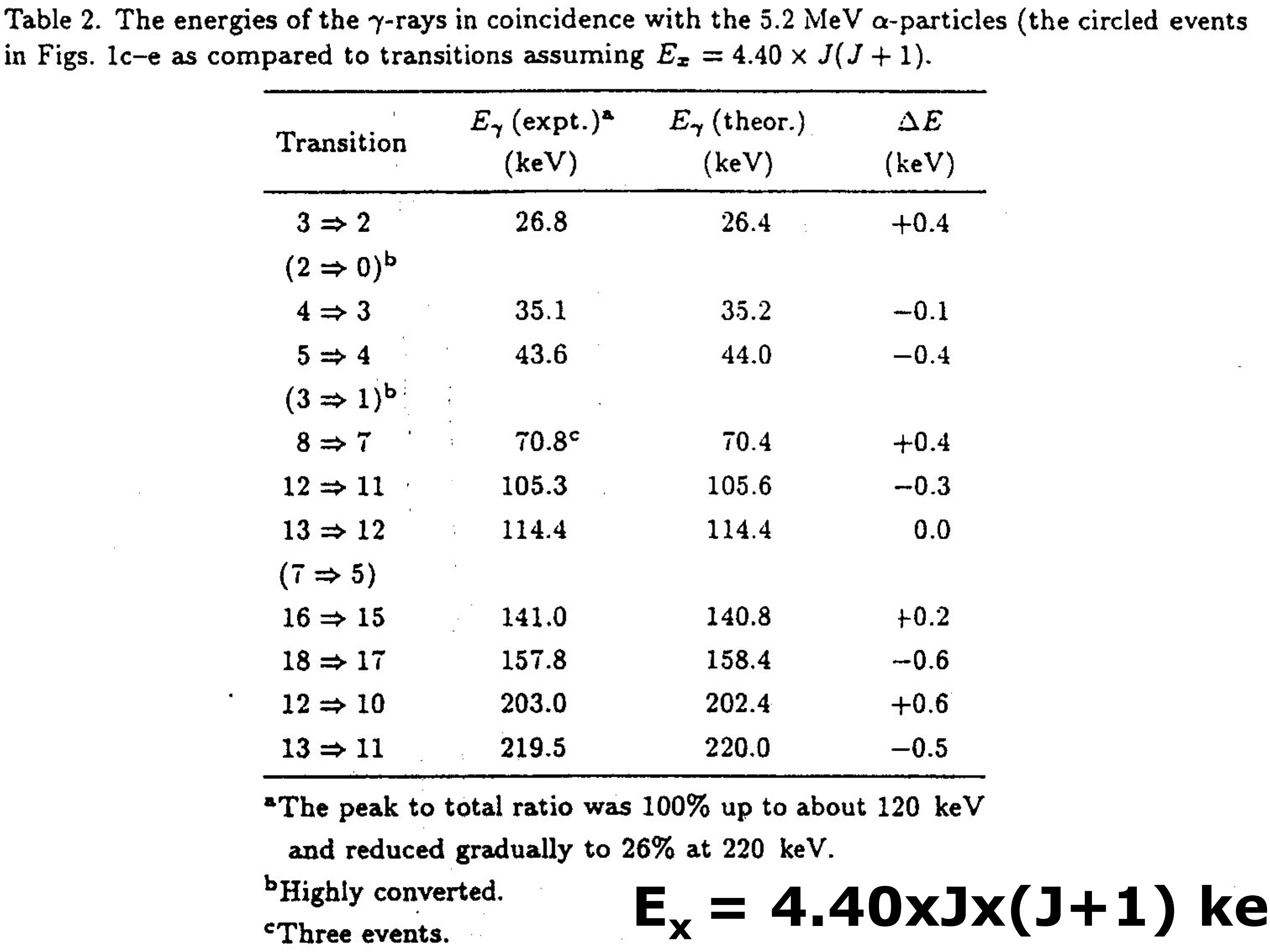


Table 2. The energies of the γ-rays in coincidence with the 5.2 MeV α-particles (the circled events in Figs. 1c–e as compared to transitions assuming $E_x = 4.40 \times J(J+1)$.

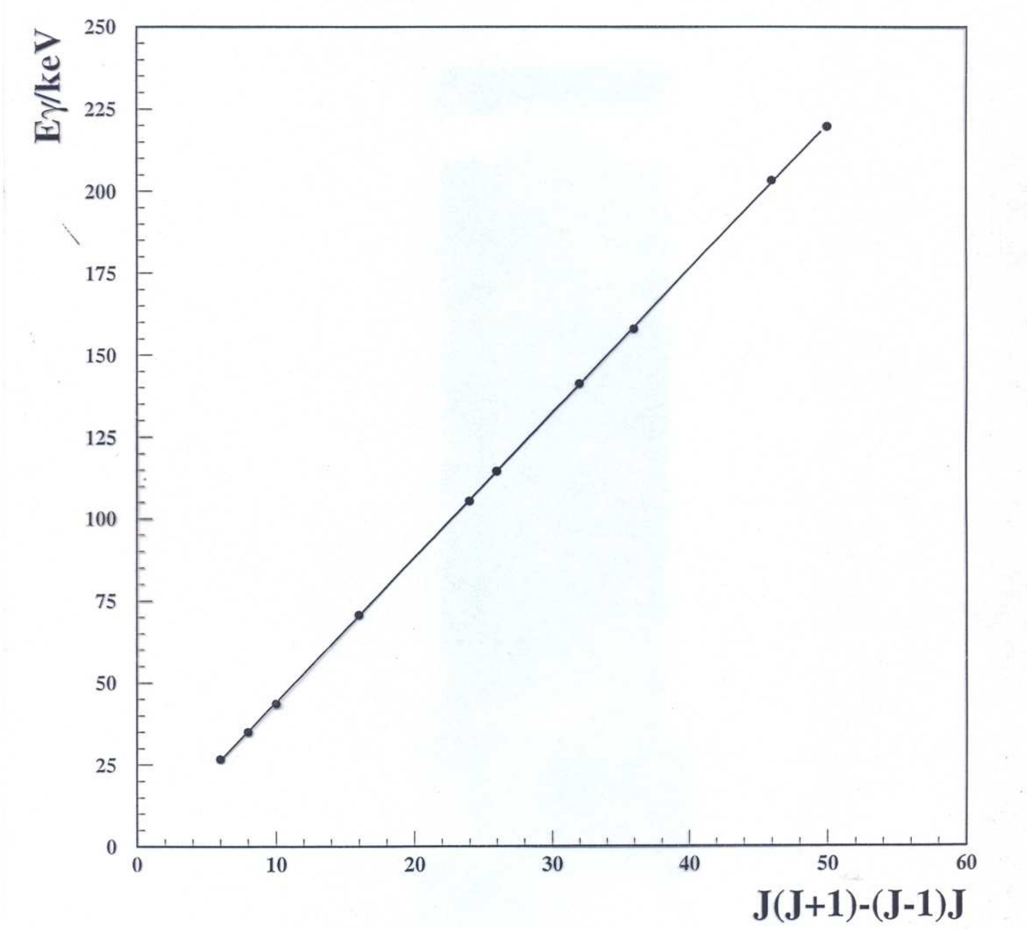
Transition	E_γ (expt.)[a] (keV)	E_γ (theor.) (keV)	ΔE (keV)
3 ⇒ 2	26.8	26.4	+0.4
(2 ⇒ 0)[b]			
4 ⇒ 3	35.1	35.2	−0.1
5 ⇒ 4	43.6	44.0	−0.4
(3 ⇒ 1)[b]			
8 ⇒ 7	70.8[c]	70.4	+0.4
12 ⇒ 11	105.3	105.6	−0.3
13 ⇒ 12	114.4	114.4	0.0
(7 ⇒ 5)			
16 ⇒ 15	141.0	140.8	+0.2
18 ⇒ 17	157.8	158.4	−0.6
12 ⇒ 10	203.0	202.4	+0.6
13 ⇒ 11	219.5	220.0	−0.5

[a] The peak to total ratio was 100% up to about 120 keV and reduced gradually to 26% at 220 keV.

[b] Highly converted.

[c] Three events.

E_x = 4.40xJx(J+1) keV



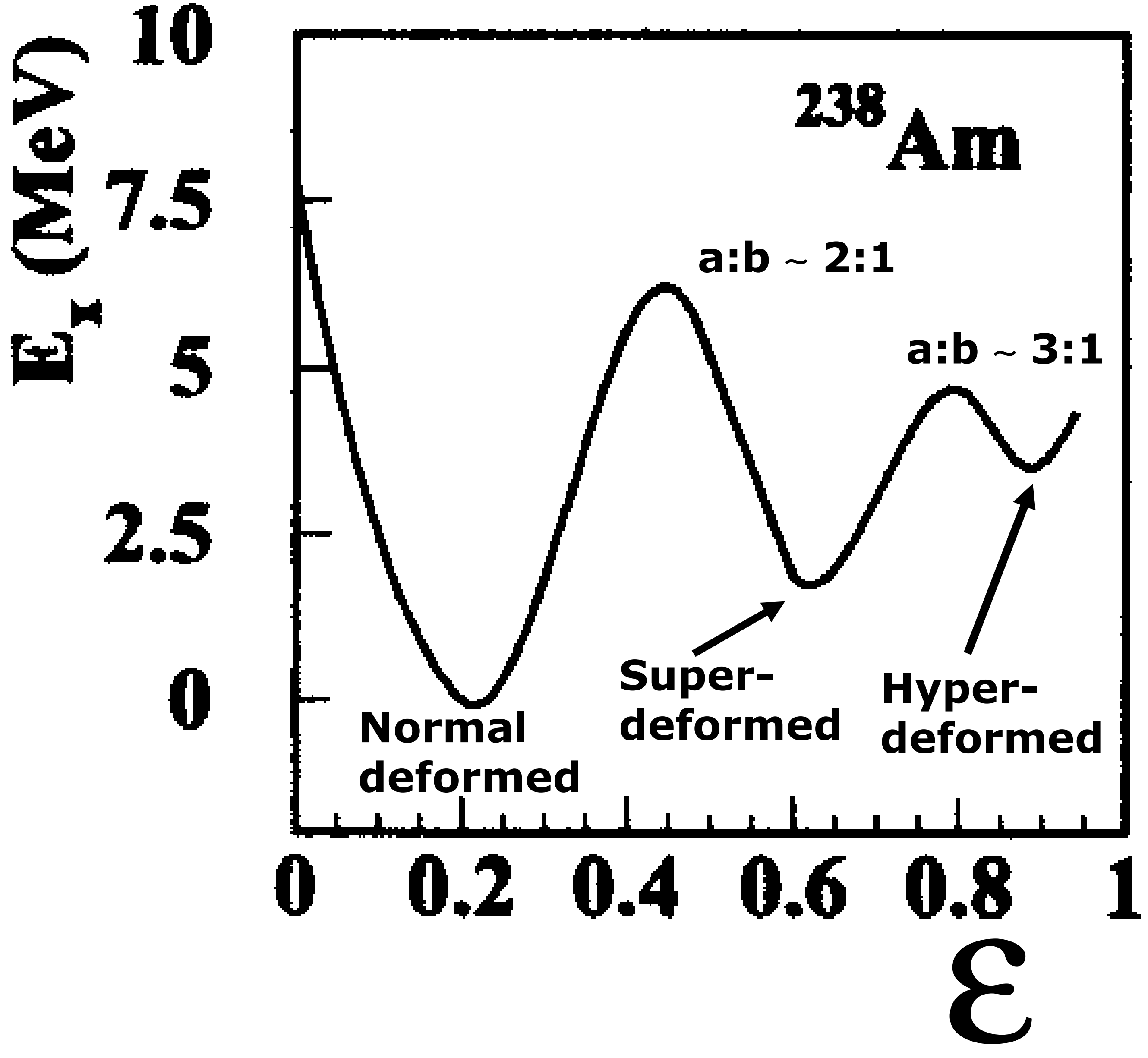
E_x = 4.40xJ(J+1)

4.40 keV is characteristic for SDB transitions

Conclusion:

The 5.2 MeV α-particles decay to a

SDB state

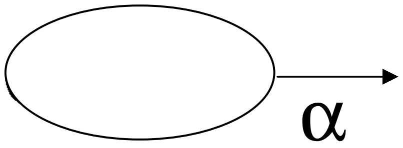


^{238}Am
E_x (MeV)
10
7.5
5
2.5
0
0
0.2
0.4
0.6
0.8
1
ε
a:b ~ 2:1
a:b ~ 3:1
Normal deformed
Super-deformed
Hyper-deformed

The potential parameters of Igo were used, but

$$R=R_0(1 + \beta_2 Y_{20}(\theta) + \beta_3 Y_{30}(\theta) + \beta_4 Y_{40}(\theta))$$

E_α	β_2	β_3	β_4	$t_{1/2}(T)(m)$	$t_{1/2}(T)/\ t_{1/2}(E)$
6.57	0.0	0.0	0.0	6.0	1.1
5.2	0.0	0.0	0.0	$2.8\text{x}10^7$	$3.1\text{x}10^5$
5.2	0.7	0.0	0.0	$1.3\text{x}10^3$	$1.4\text{x}10^1$
5.2	0.7	0.07	0.0	$3.5\text{x}10^2$	3.9
5.2	0.7	0.15	0.0	$8.2\text{x}10^1$	0.9
5.2	0.7	0.07	0.06	$1.0\text{x}10^2$	1.1



The data can consistently be interpreted in terms of a transition from a high spin state in the SD minimum of the parent nucleus to a high spin state in the SD minimum of the daughter.

Mod. Phys. Lett. A11, 861(1996)

Theoretical Predictions SDBH (^{206}At)

W. Satula et al. (1991)	**S.J. Krieger et al. (1992)**
(Macroscopic-Microscopic) (Strutinsky)	(Hartree-Fock + BCS)
7.25 MeV (ext.)	10.89 MeV(int.)

α decay to spin 18 at 1.50 (MeV); [Ex=4.40xJ(J+1)]

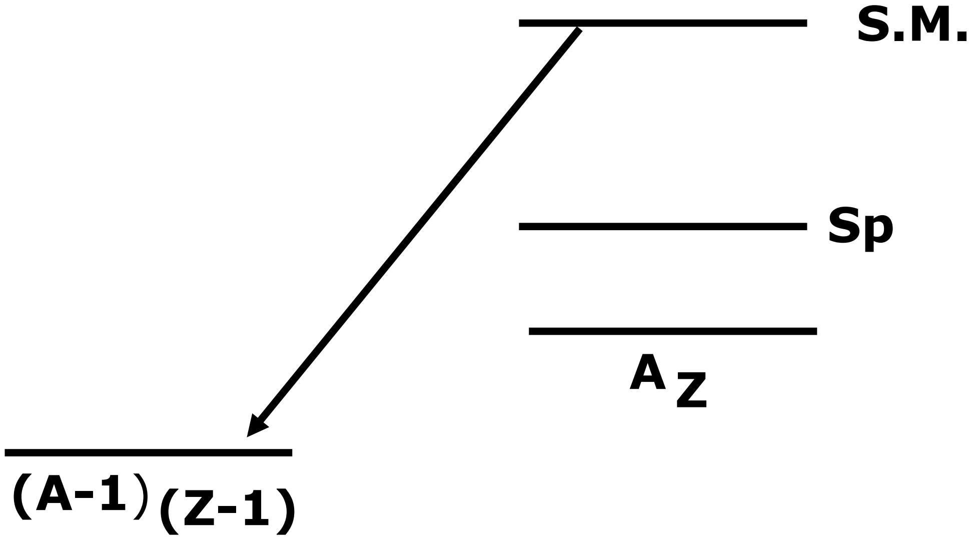
Ex in ^{206}At

≥8.75 MeV	≥12.39 MeV

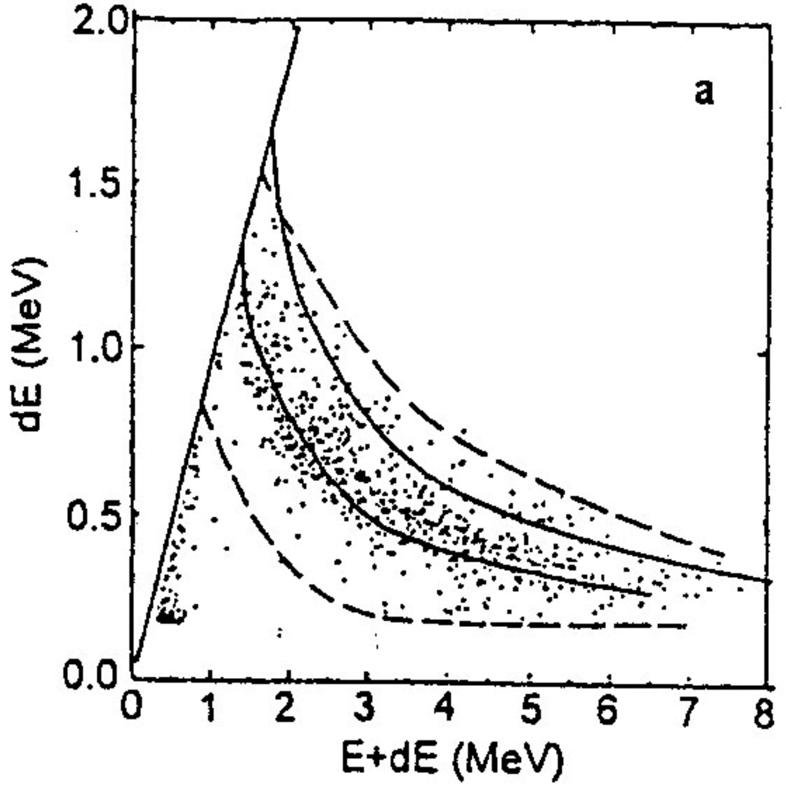
Ex in 210Fr (E_x (^{206}At)+(5.3-6.70))

≥7.35	**≥10.99**

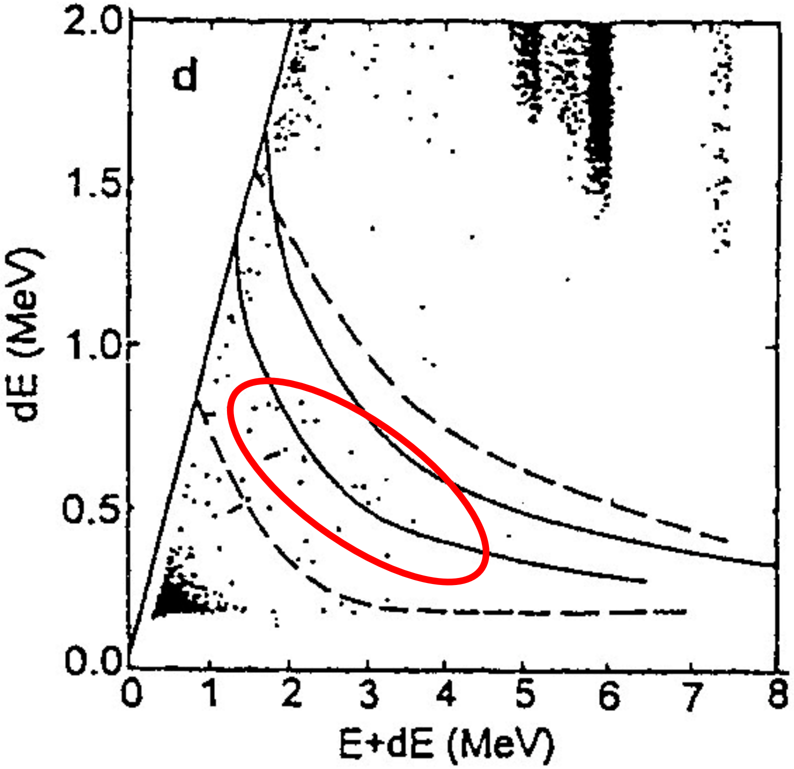
Long-lived proton radioactivity



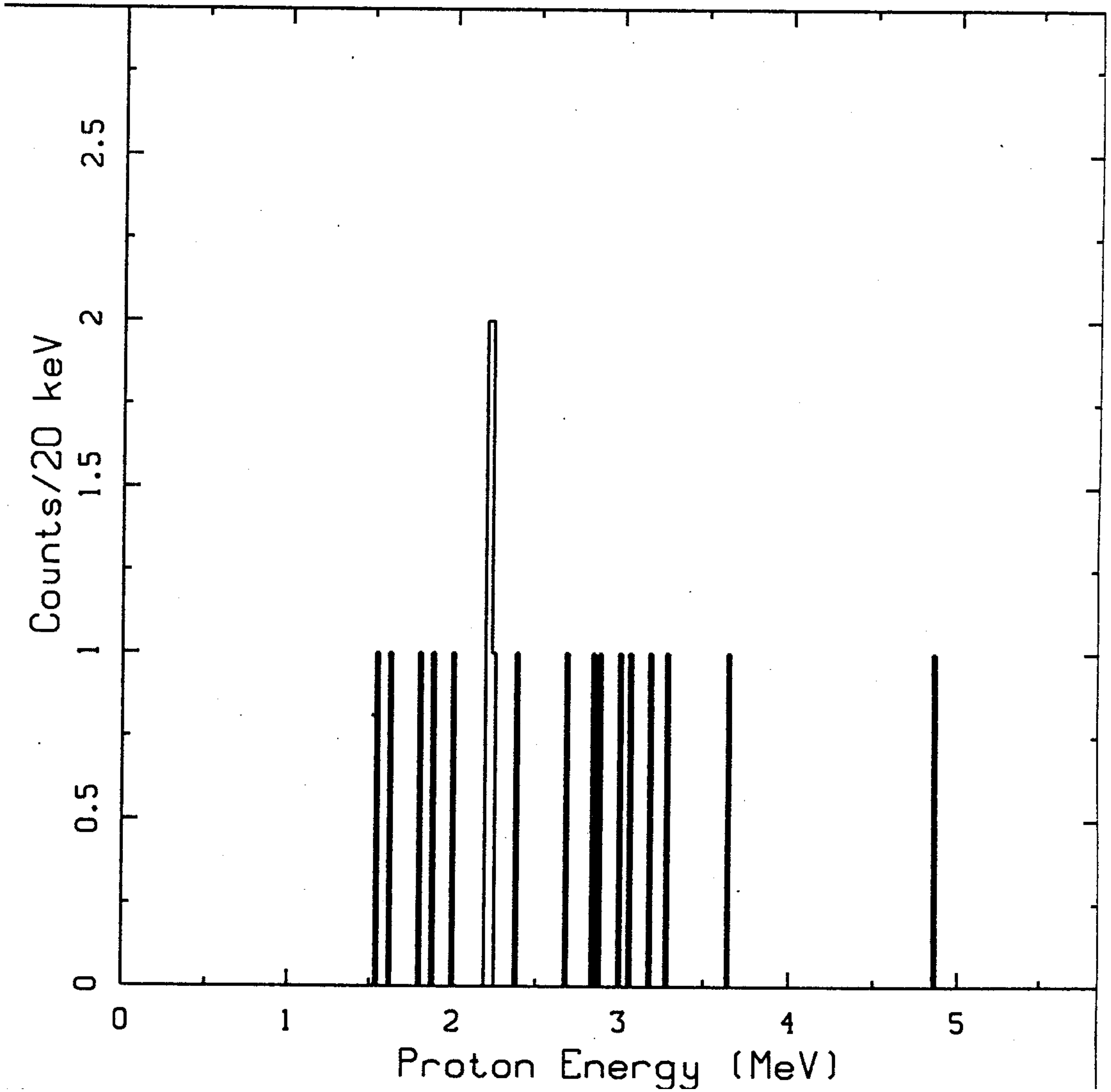
For ^{210}Fr Sp = 1.7 MeV and S.M. ≥ 7.3 MeV



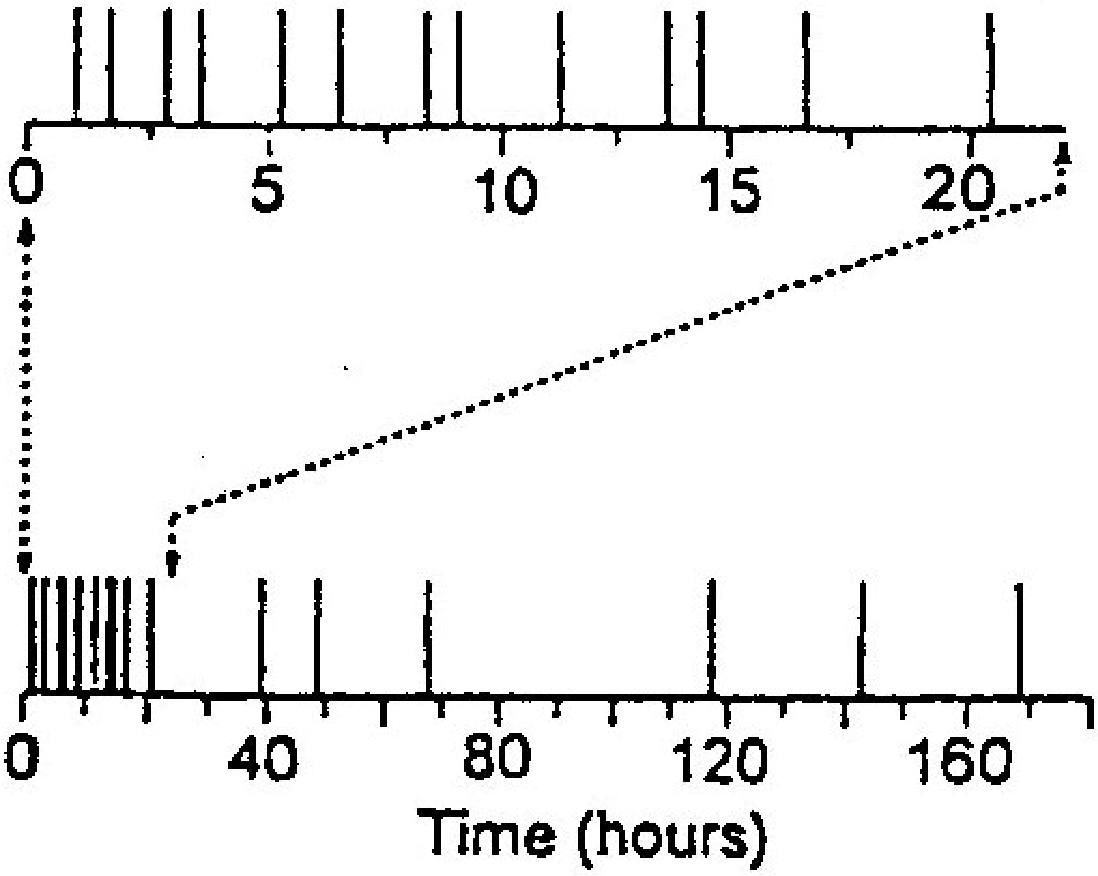
Calibration using Rd-Be source and polyethylene radiator



^{16}O+^{197}Au at 80 MeV. Sum of three measurements



E_p from 1.5 to 3.6 MeV. A small group of 5 events is seen at 2.19 MeV.



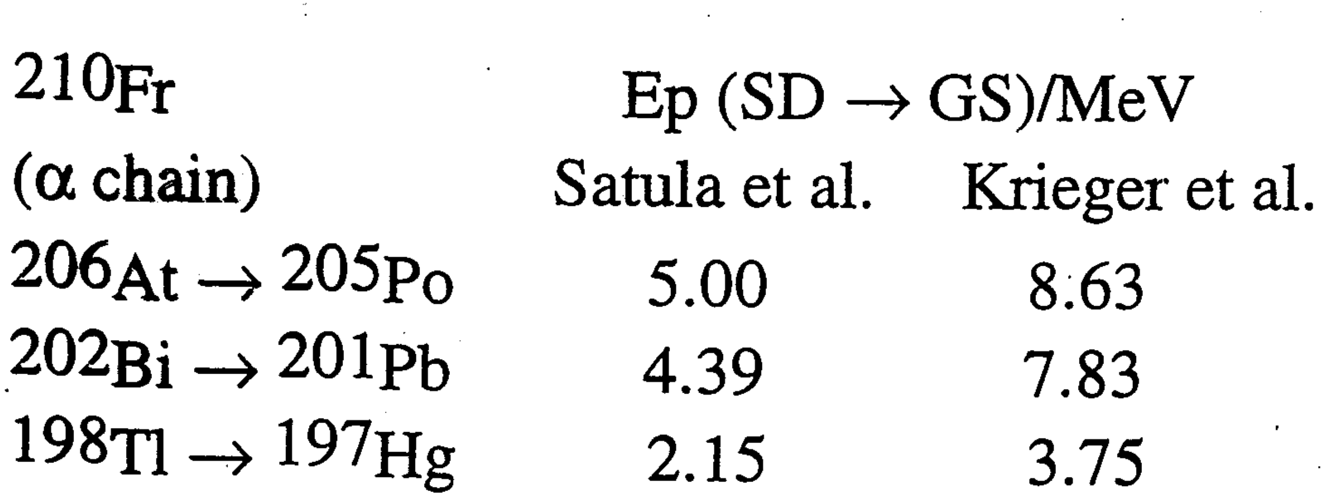
$\tau = (1/n)\Sigma t_i$

$t_{1/2}(a) \approx 6$ h

$t_{1/2}(b) \approx 70$ h

$t_{1/2}(\mathrm{Cal})$ (for $E_p = 2.19$ MeV) $\approx$ $2\mathrm{x}10^{-9}$ s

Retardation ≈ 10^{14}



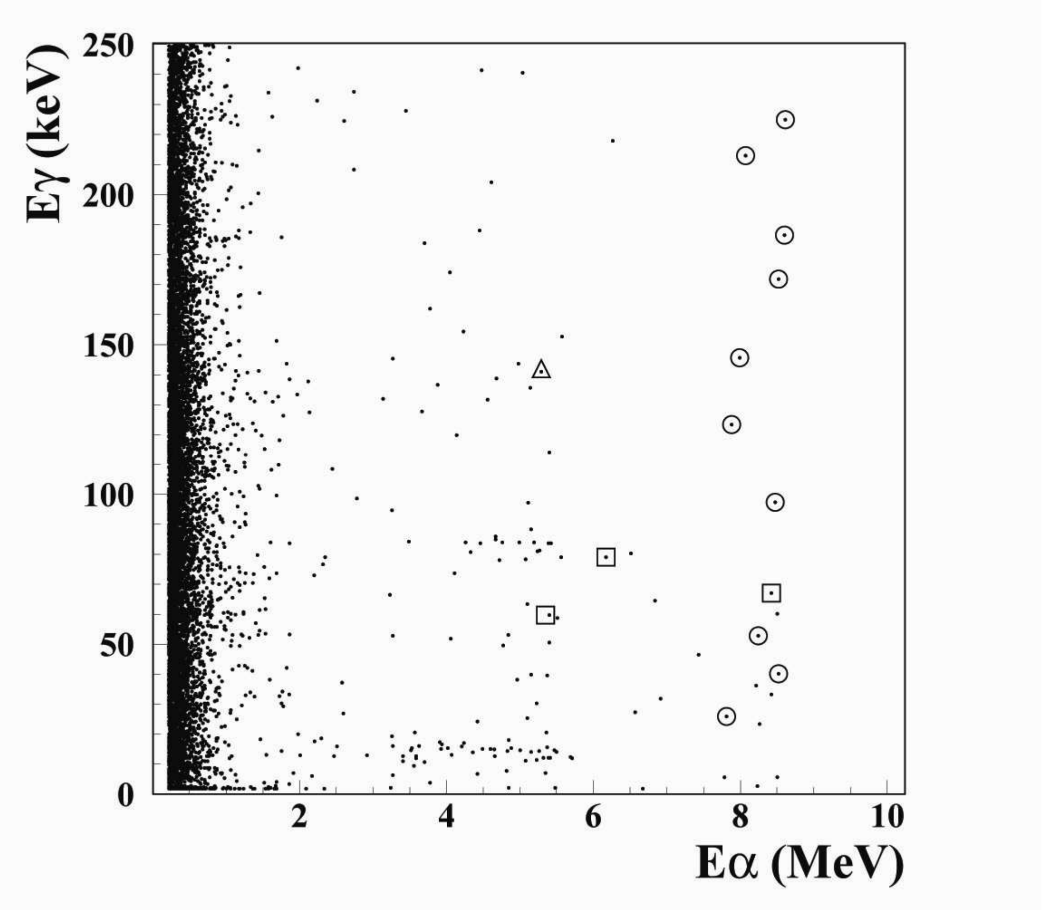
^{210}Fr	Ep (SD → GS)/MeV	
(α chain)	Satula et al.	Krieger et al.
^{206}At → ^{205}Po	5.00	8.63
^{202}Bi → ^{201}Pb	4.39	7.83
^{198}Tl → ^{197}Hg	2.15	3.75

Mod. Phys. Lett. A11, 949 (1996)

The second experiment we performed with the pelleton:

^{28}Si + ^{181}Ta at E_{Lab} = 125 MeV (This is about 10% below the Coulomb barrier)

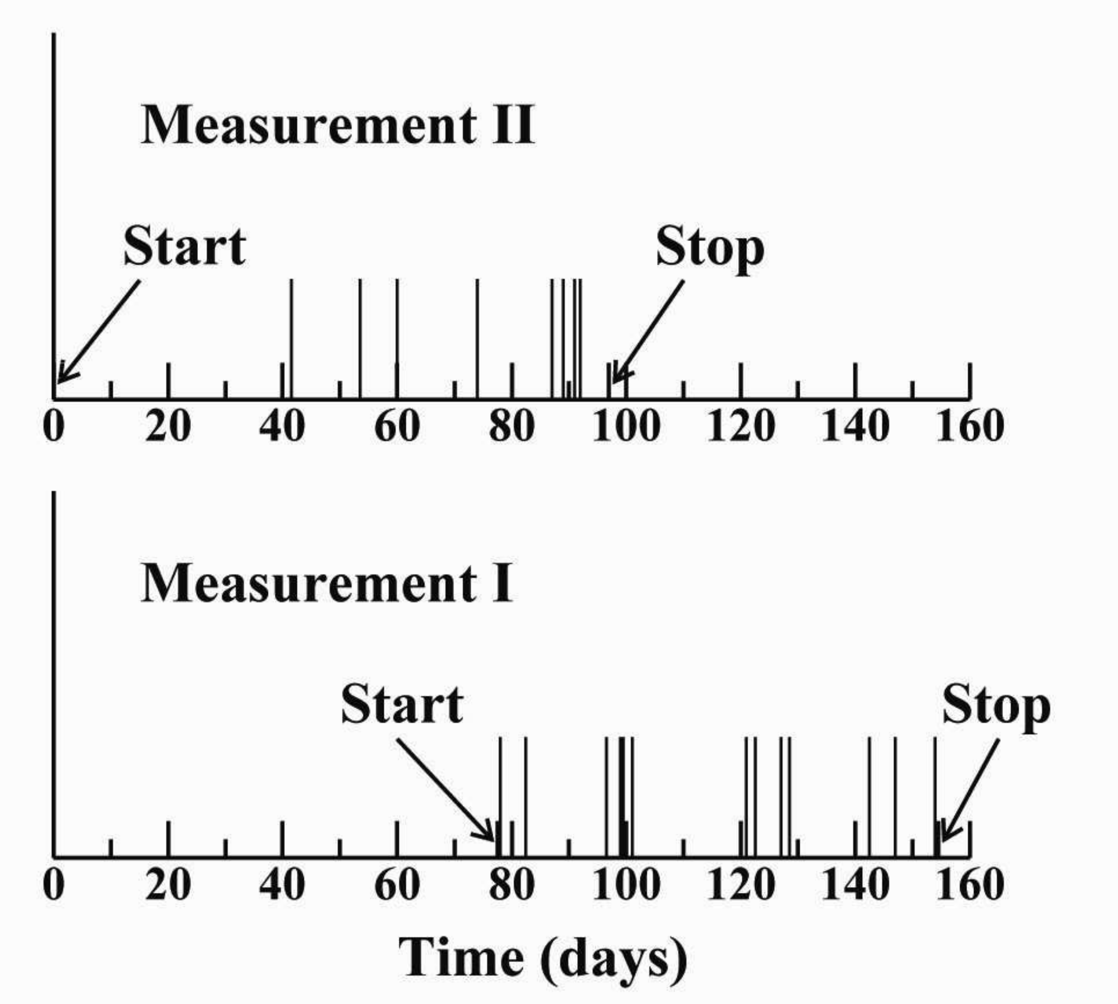
The compound nucleus is ^{209}Fr



Irradiation: T=42.5 h; i=11.5pnA; dose: 1.1x10^{16} particles.

Measurement: T_1=77.4 d; T_2=154.2 d; ΔT=76.8 d.

Catcher foil: 200 μg/cm^2 of C.

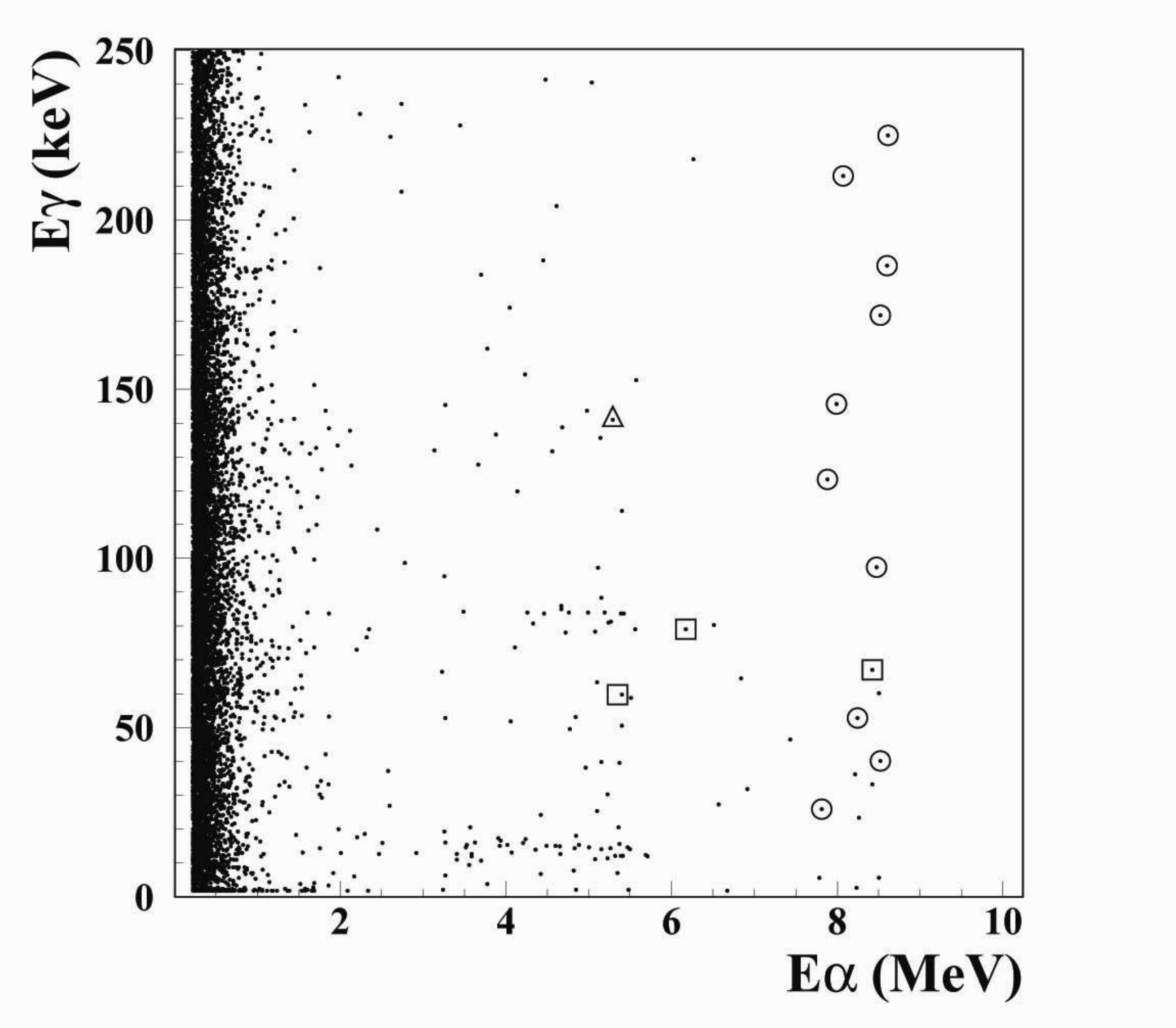


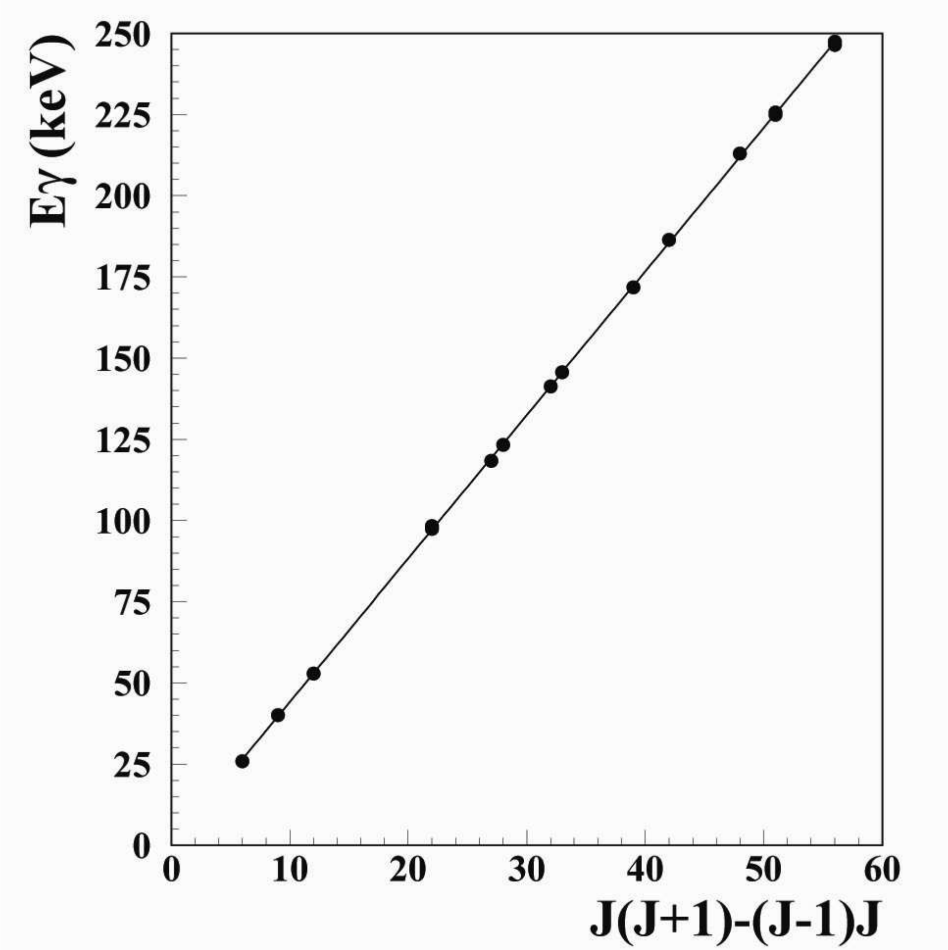
$40\ d \leq t_{1/2} \leq 2\ y$

8.6 MeV is a very high energy for α-particles.

$t_{1/2}$(Cal.) ≈ 1 μs

Retardation factor ≈ 10^{12}


Eγ (keV)
250
200
150
100
50
0
2
4
6
8
10
Eα (MeV)

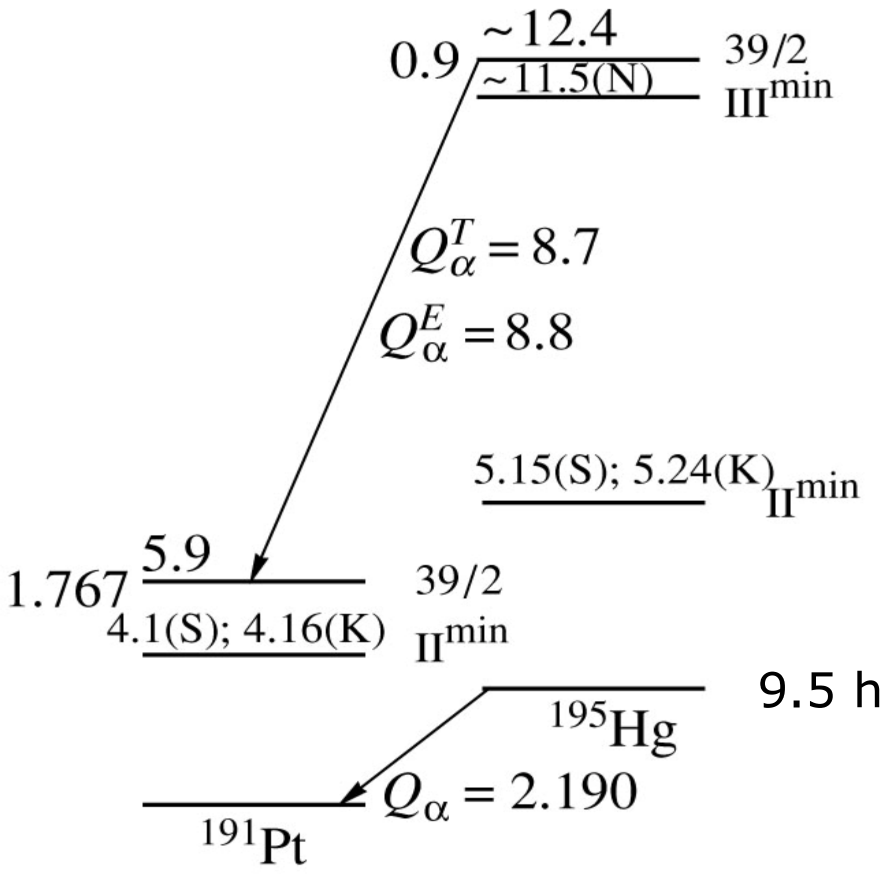


$$E_x = 4.42 \times J(J+1)$$

The 8.6 MeV α-particles decay very retardly to a high spin SD state. It cannot be a SD to a SD transition which decays very enhancely.

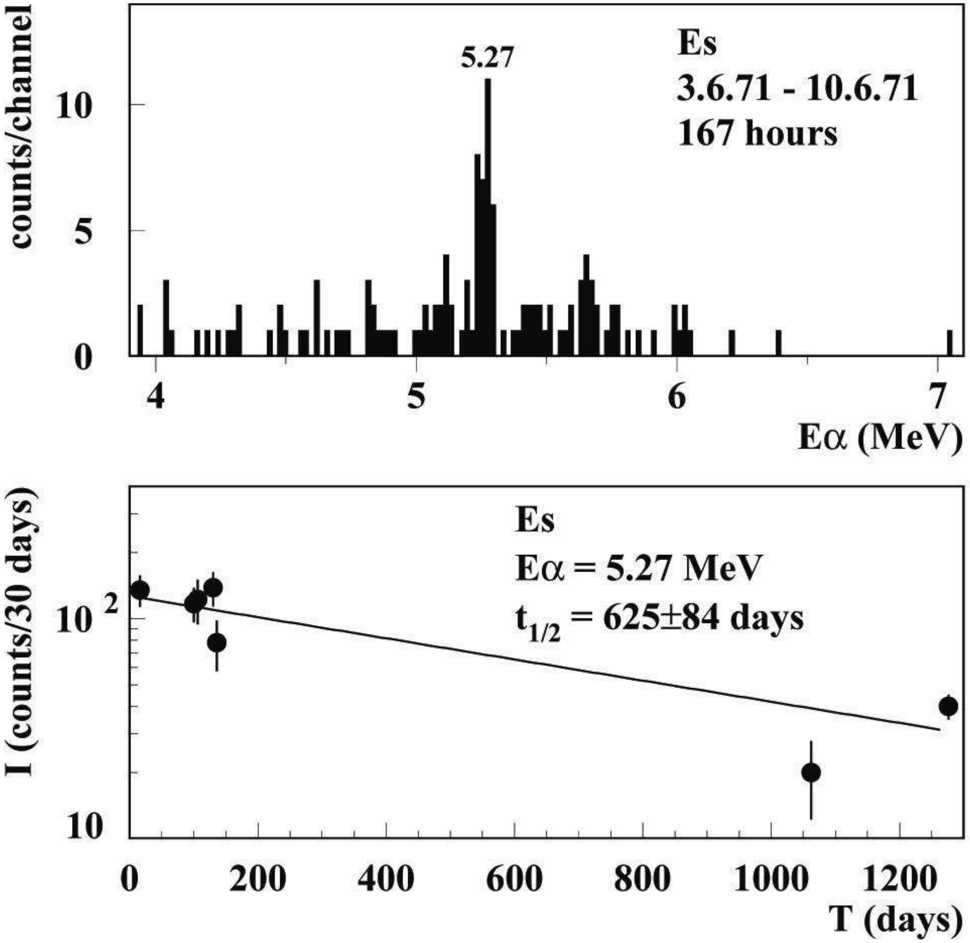
Int. J. Mod. Phys. E10 (2001) 185-208.

Consistent interpretation: A transition from a high spin Hyperdeformed (HD) state to a high spin SD state.



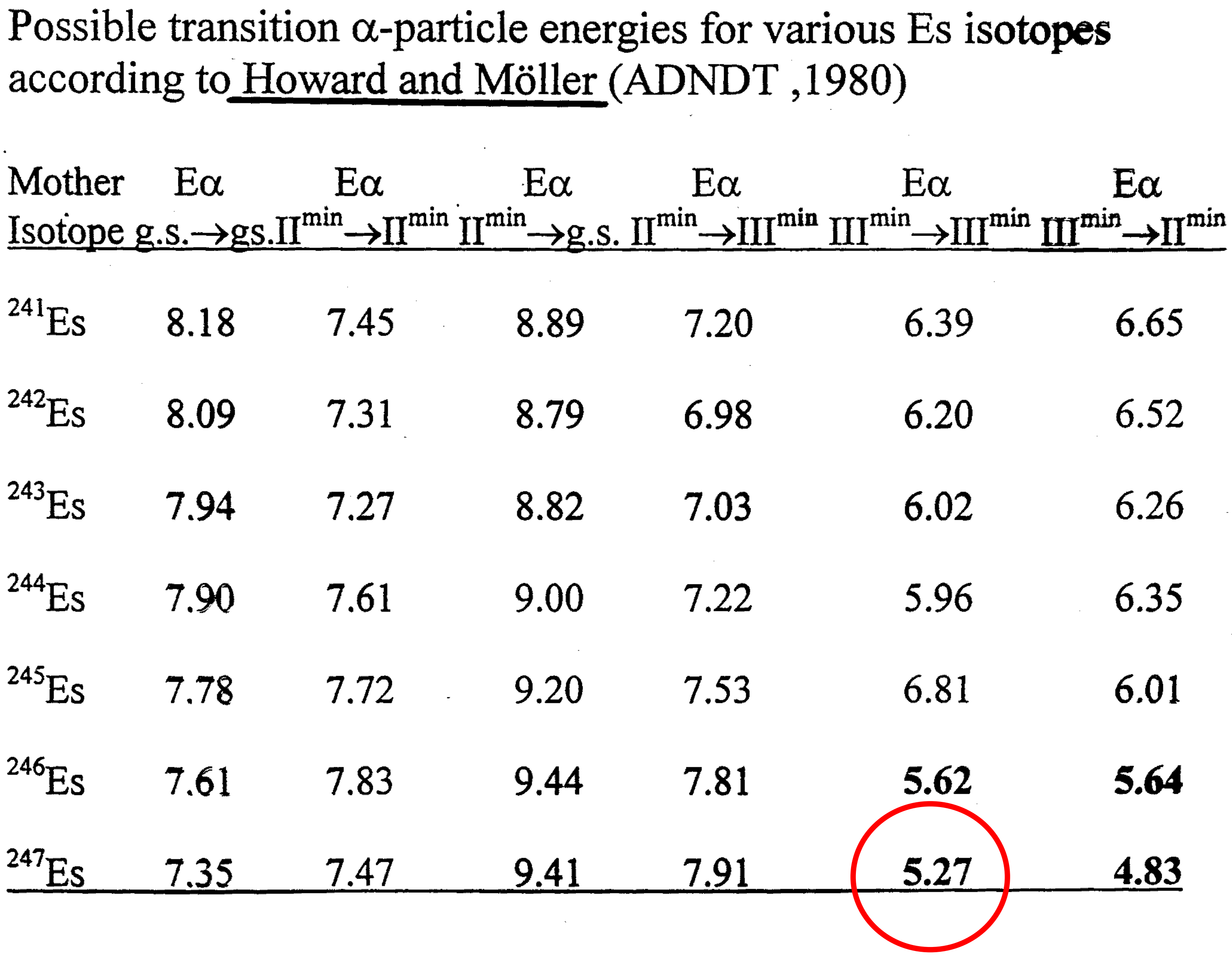
9.5 h

Coming back to the low energy α-particles in the actinides



First problem we had: Low energy of α-particles

Source	Eα (MeV)	$t_{1/2}$/s (Cal.)	Typical Eα (MeV)	$t_{1/2}$/s (Cal.)
Bk (Cm,Am)	5.14	2 x 10^{12}	6 - 7	2 x 10^{5}
Es	5.27	3 x 10^{13}	7 - 8	5 x 10^{2}
No-Lr	5.53	2 x 10^{13}	8 - 9	1 x 10^{1}



Possible transition α-particle energies for various Es isotopes according to Howard and Möller (ADNDT ,1980)

Mother Isotope	Eα g.s.→gs.II^{min}	Eα II^{min}→II^{min}	Eα II^{min}→g.s.	Eα II^{min}→III^{min}	Eα III^{min}→III^{min}	Eα III^{min}→II^{min}
^{241}Es	8.18	7.45	8.89	7.20	6.39	6.65
^{242}Es	8.09	7.31	8.79	6.98	6.20	6.52
^{243}Es	7.94	7.27	8.82	7.03	6.02	6.26
^{244}Es	7.90	7.61	9.00	7.22	5.96	6.35
^{245}Es	7.78	7.72	9.20	7.53	6.81	6.01
^{246}Es	7.61	7.83	9.44	7.81	**5.62**	**5.64**
^{247}Es	7.35	7.47	9.41	7.91	**5.27**	**4.83**

The low energy can be understood as a HD to HD transition.

Second problem we had: Enhance transition through the Coulomb barrier.

Es: $E_\alpha = 5.27$ MeV;

$t_{1/2}$ (Exp.) = 625 d = 5.4×10^{7} s
$t_{1/2}$(Cal., V.S.) = 1.5×10^{14} s

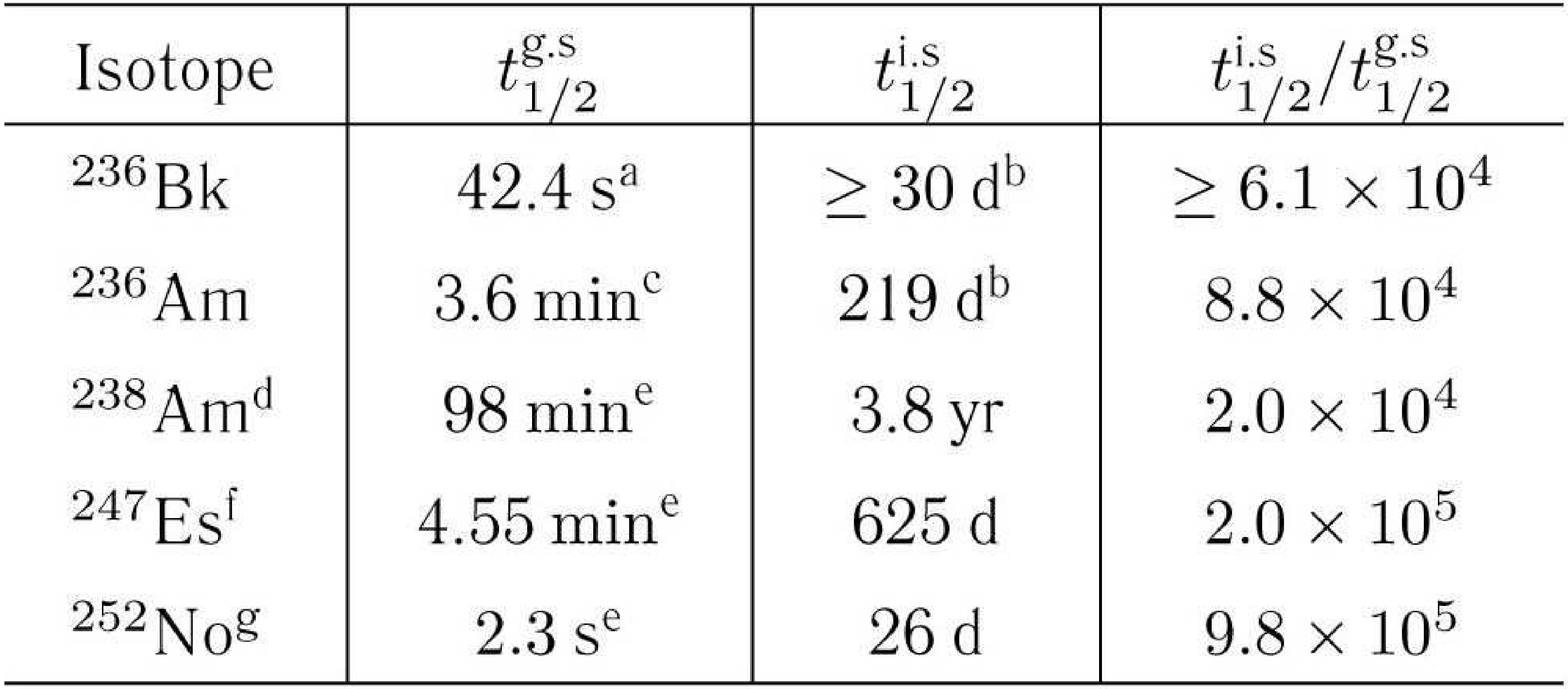
Enhancement: 2.8×10^{6}

$t_{1/2}$(Cal.) (HD → HD) = 3.3×10^{7} s.

$\beta_2 = 1.05$; $\beta_4 = 0.19$; Howard and Möller (1980)

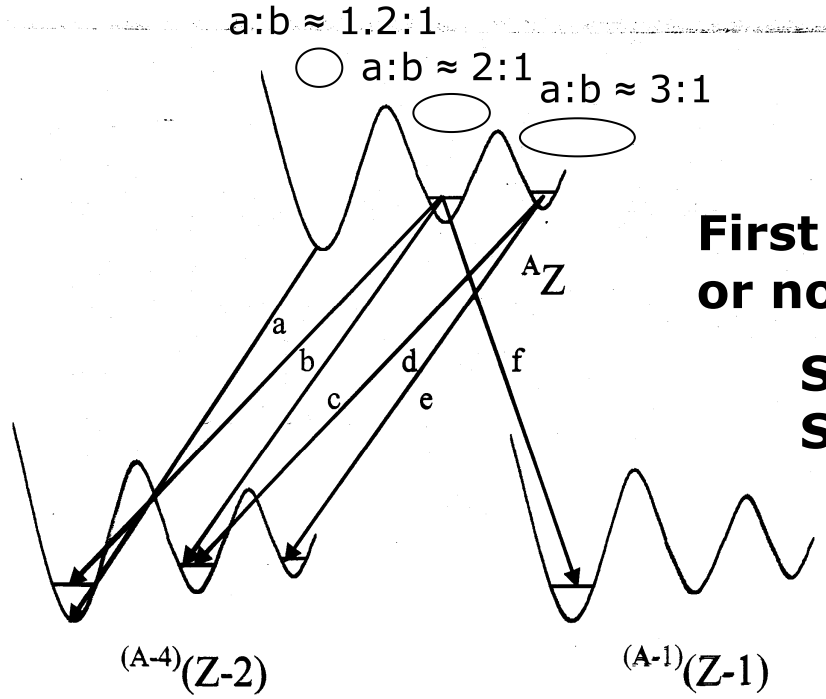
Source	Eα (MeV)	$t_{1/2}$	E_{cal} (Howard, Möller)	$t_{1/2}$(cal)/$t_{1/2}$(exp)
Bk	5.14	3.8 y 1.6 h (g.s.)	**5.13;(^{238}Am (II→II))** 5.94 (normal)	2.8
Es	5.27	625 d 4.6 m (g.s.)	**5.27; (^{247}Es (III→III))** 7.32 (normal)	~0.6
No-Lr	5.53	26 d 2.3 s (g.s.)	**~5.6; (^{252}No(III-III))** 8.42 (normal)	~3.1

The effect of the low energy is larger than the effect of the enhancement. The lifetimes of the isomeric states are larger than that of their corresponding g.s.



Isotope	$t_{1/2}^{\mathrm{g.s}}$	$t_{1/2}^{\mathrm{i.s}}$	$t_{1/2}^{\mathrm{i.s}}/t_{1/2}^{\mathrm{g.s}}$
$^{236}\mathrm{Bk}$	42.4 s^{a}	$\geq 30\ \mathrm{d}^{\mathrm{b}}$	$\geq 6.1 \times 10^4$
$^{236}\mathrm{Am}$	3.6 $\mathrm{min}^{\mathrm{c}}$	219 d^{b}	8.8×10^4
$^{238}\mathrm{Am}^{\mathrm{d}}$	98 $\mathrm{min}^{\mathrm{e}}$	3.8 yr	2.0×10^4
$^{247}\mathrm{Es}^{\mathrm{f}}$	4.55 $\mathrm{min}^{\mathrm{e}}$	625 d	2.0×10^5
$^{252}\mathrm{No}^{\mathrm{g}}$	2.3 s^{e}	26 d	9.8×10^5

Int. J. Mod. Phys. E10 (2001) 209-236



First minimum: Spherical or normal deformed

Second minimum: Suerdeformed

Third minimum: Hyperdeformed

a: $I^{min} \rightarrow I^{min}$. Normal α's.

b: $II^{min} \rightarrow I^{min}$. Retarded α's: $^{190}Ir \rightarrow ^{186}Re$.

c: $II^{min} \rightarrow II^{min}$. Enhanced α's: $^{210}Fr \rightarrow ^{106}At$; ~ $^{238}Am \rightarrow ^{234}Np$.

d: $III^{min} \rightarrow II^{min}$. Retarded α's: $^{195}Hg \rightarrow ^{191}Pt$.

e: $III^{min} \rightarrow III^{min}$. Enhanced α's: ~ $^{247}Es \rightarrow ^{243}Bk$; ~ $^{252}No \rightarrow ^{248}Fm$.

f: $II^{min} \rightarrow I^{min}$. Retarded protons: $^{198}Tl \rightarrow ^{197}Hg$ (?); $^{205}Fr \rightarrow ^{204}Rn$ (?).

Fig. 12. Summary of the new kinds of seen transitions and their properties.

The cross section for producing the compound nucleus in SD or HD shapes is much larger than its production in the normal g.s.

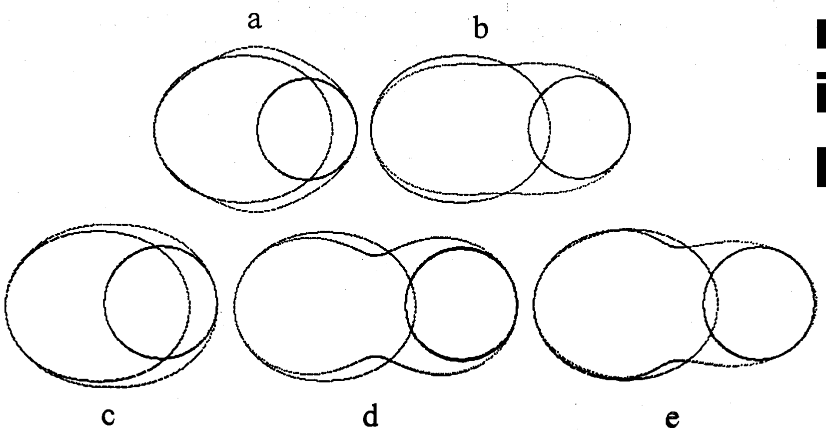


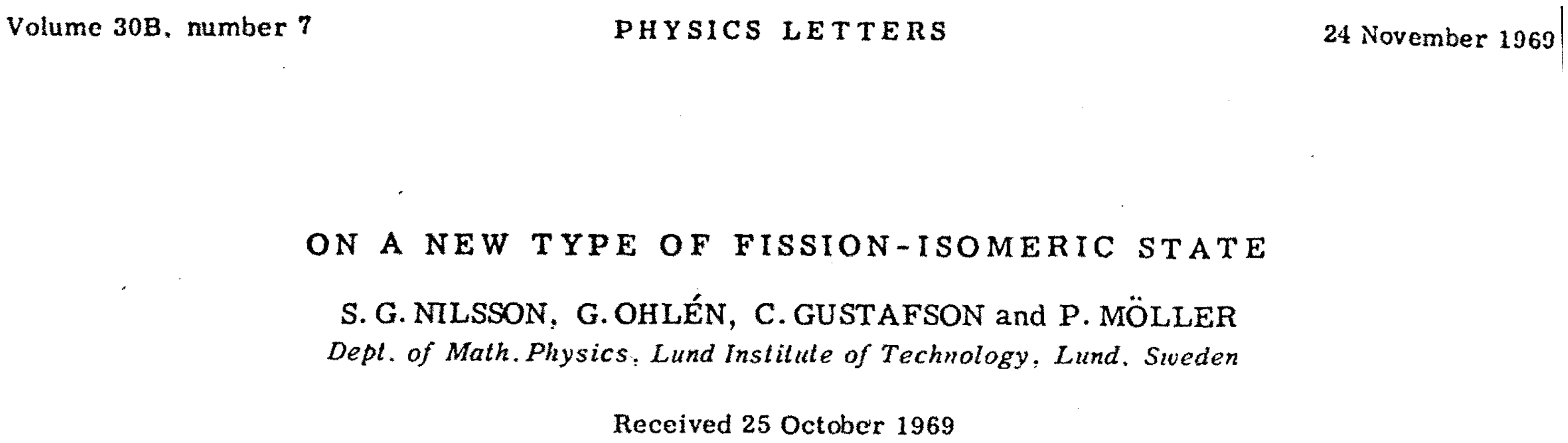
Fig. 13. Calculated shapes of two compound nuclei at various configurations together with the shapes of the corresponding projectile and target nuclei:

a) $A_{C.N.} = 239; \beta_2 = 0.2; \beta_4 = 0.08.$ b) $A_{C.N.} = 239; \beta_2 = 0.77; \beta_4 = 0.1.$
$A_{heavy} = 186; \beta_2 = 0.22.$ $A_{light.} = 53; \beta_2, \beta_3, \beta_4 = 0.0.$

c) $A_{C.N.} = 253; \beta_2 = 0.28; \beta_4 = 0.01.$ d) $A_{C.N.} = 253; \beta_2 = 1.2; \beta_4 = 0.0.$
e) $A_{C.N.} = 253; \beta_2 = 0.85; \beta_3 = 0.35; \beta_4 = 0.18.$
$A_{heavy} = 186; \beta_2 = 0.22.$ $A_{light.} = 67; \beta_2, \beta_3, \beta_4 = 0.0.$

Why the SD and HD isomers do not decay by fission?

Probably because of the high intrinsic spin.



Volume 30B, number 7 PHYSICS LETTERS 24 November 1969

ON A NEW TYPE OF FISSION-ISOMERIC STATE

S. G. NILSSON, G. OHLÉN, C. GUSTAFSON and P. MÖLLER
Dept. of Math. Physics, Lund Institute of Technology, Lund, Sweden

Received 25 October 1969

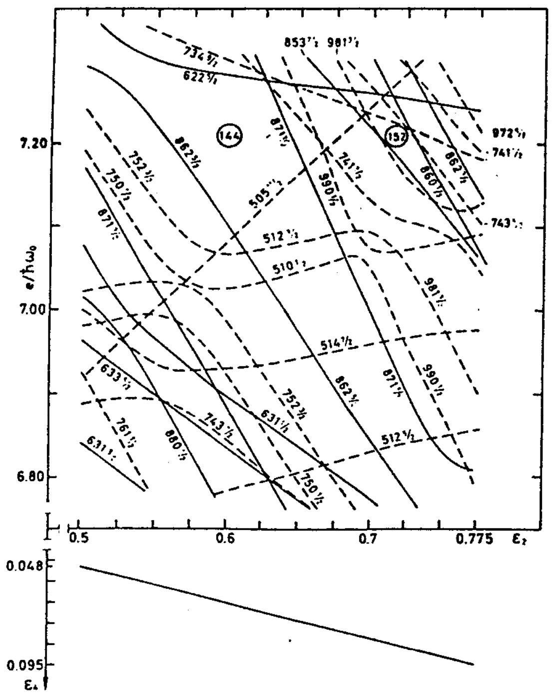


Fig. 1. Neutron orbitals for $A \approx 242$ along parts of an averaged "fission path" in the (ϵ, ϵ_4) plane [13]. (The location of the "path" is indicated at the bottom of the diagram.) Orbitals are assigned "asymptotic" quantum-numbers.

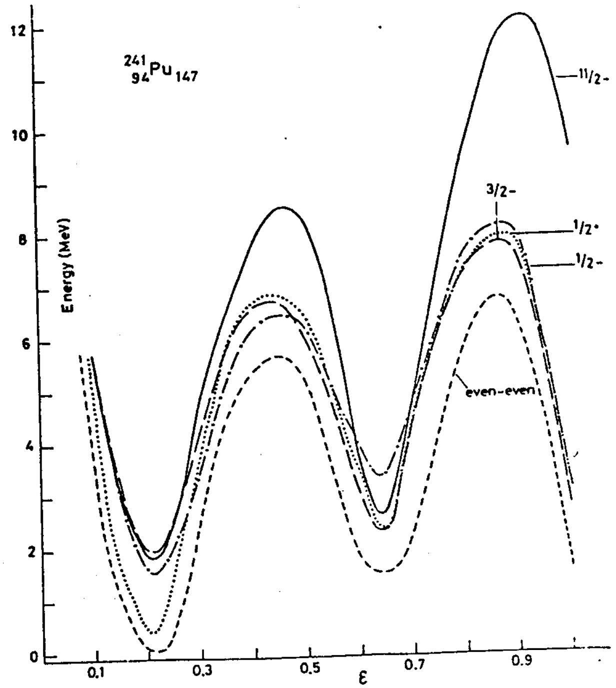
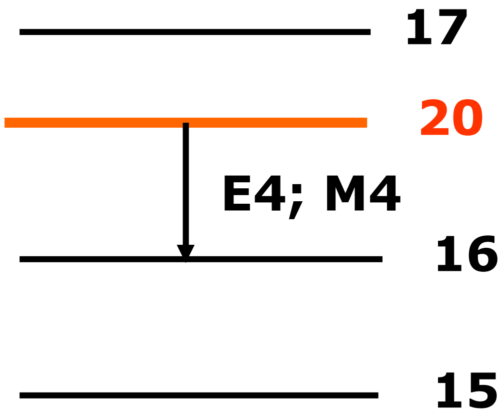


Fig. 2. Cuts through the energy surfaces corresponding to an "even" ^{241}Pu nucleus (dashed) (interpolated between ^{240}Pu and ^{242}Pu) and to minimum configurations with the odd-particle places in an optional $\frac{11}{2}^-$ (solid), $\frac{3}{2}^-$ (long-dashed), $\frac{1}{2}^+$ (dotted), and $\frac{1}{2}^-$ (dot-dashed) orbital.

4 MeV increase in the barrier which corresponds to 10^{15} longer lifetime.

Why the isomeric states do not decay by γ-rays?

Probably because they are trapped between the rotational states.

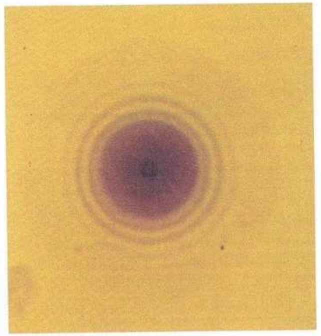


Unexplained radioactivities seen in natural materials

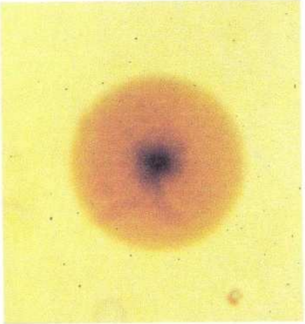
The first one is the Po halos seen in mica.

^{238}U (4.2 MeV α) ^{234}Th (β) ^{234}Pa (β) ^{234}U (4.8 MeV α) ^{230}Th (4.7 MeV α) ^{226}Ra (4.8 MeV α) ^{222}Rn (5.5 MeV α) ^{218}Po (6.0 MeV α) ^{214}Pb (β) ^{214}Bi (β) ^{214}Po (7.7 MeV α) ^{210}Pb (β) ^{210}Bi (β) ^{210}Po (5.3 MeV α).

$t_{1/2}$ (^{238}U) = 4.5x10^{9} y

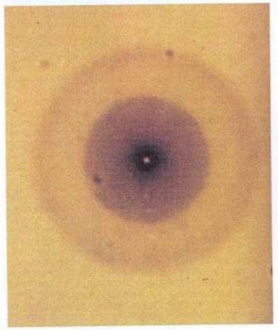


^{238}U



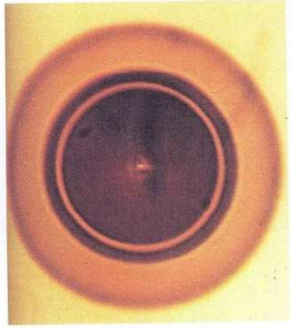
^{210}Po

$t_{1/2}$ (^{210}Po) = 138 d
$t_{1/2}$ (^{210}Pb) = 22 y
(parent)



^{214}Po

$t_{1/2}$ (^{214}Po) = 164 μs
$t_{1/2}$ (^{214}Pb) = 26.8 m
(parent)



^{218}Po

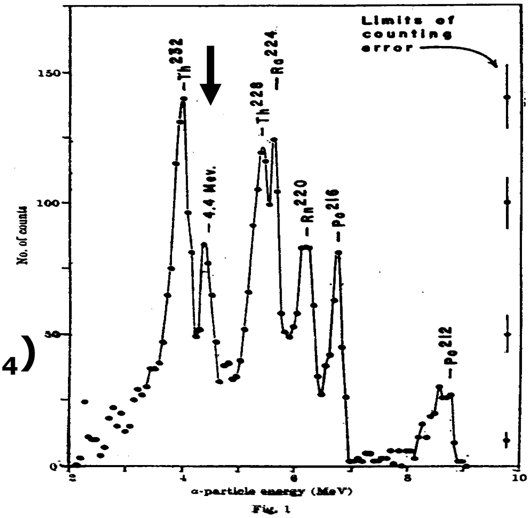
$t_{1/2}$ (^{218}Po) = 3 m

(Pictures from: R. V. Gentry, Creation's Tiny Mystery, Earth Science Associates Knoxville, Tennessee)

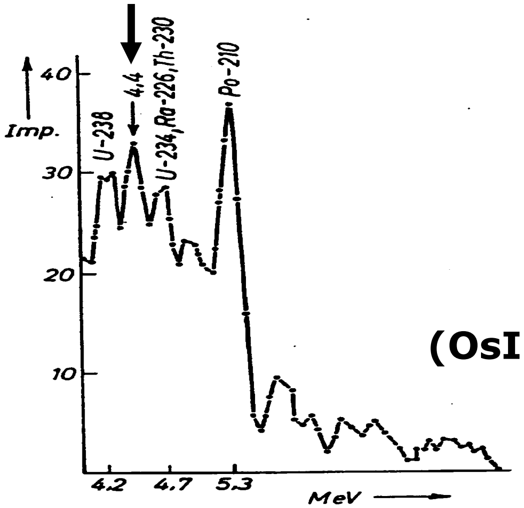
Long-lived isomeric states in the region around Po which decay by EC or beta particles can consistently interpret these puzzling halos

The second phenomenon is the observation in various materials of a low energy α-particle group of 4.5 MeV

Thorite ($ThSiO_4$) from granite

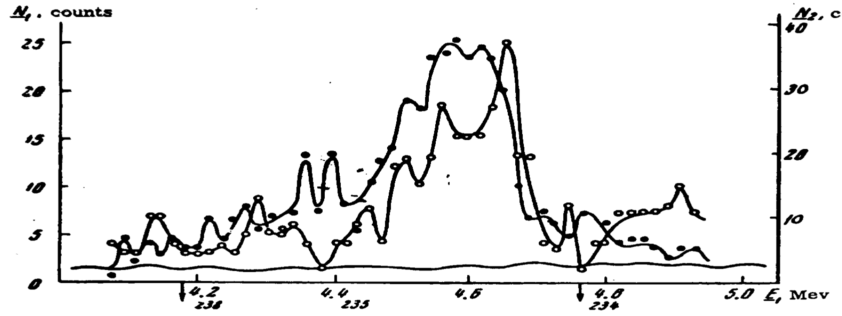


Cherry, Richardson & Adams
Nature **202**, 640 (1964)



Meier et al., Z. Naturforsch.
25 a, 79 (1970)

(OsIr)



Cherdyntsev, Zverev, Kuptsov & Kislitsina
Geokhimiya, 395 (1968).

Magnetite (Fe_3O_4) from Trans-caucacasian monazite

Distilled from strong nitric acid

Based on chemical properties it was assumed that the 4.5 MeV group is due to decay of an isotope of Hs (element 108, eka-Os)

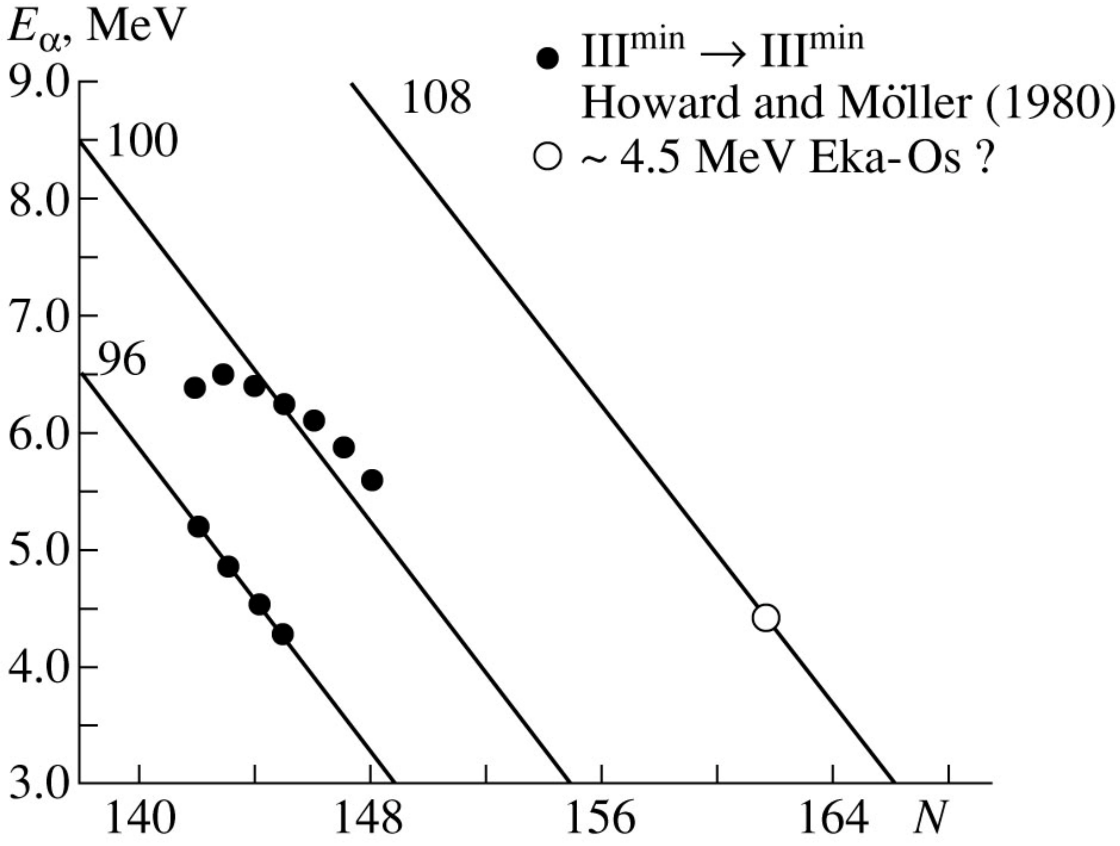
$t_{1/2} \approx 10^8$ y

However, the energy is too low (Normal energies are around 9 MeV, with $t_{1/2}$ ms to sec.) and it passes the barrier too fast

$t_{1/2}(\mathrm{Cal}) \approx 10^{16}$ y

Enhancement: 10^8

Consistent interpretation is obtained if one assumes HD to HD transition



Int. J. Mod. Phys. E12 (2003) 661.

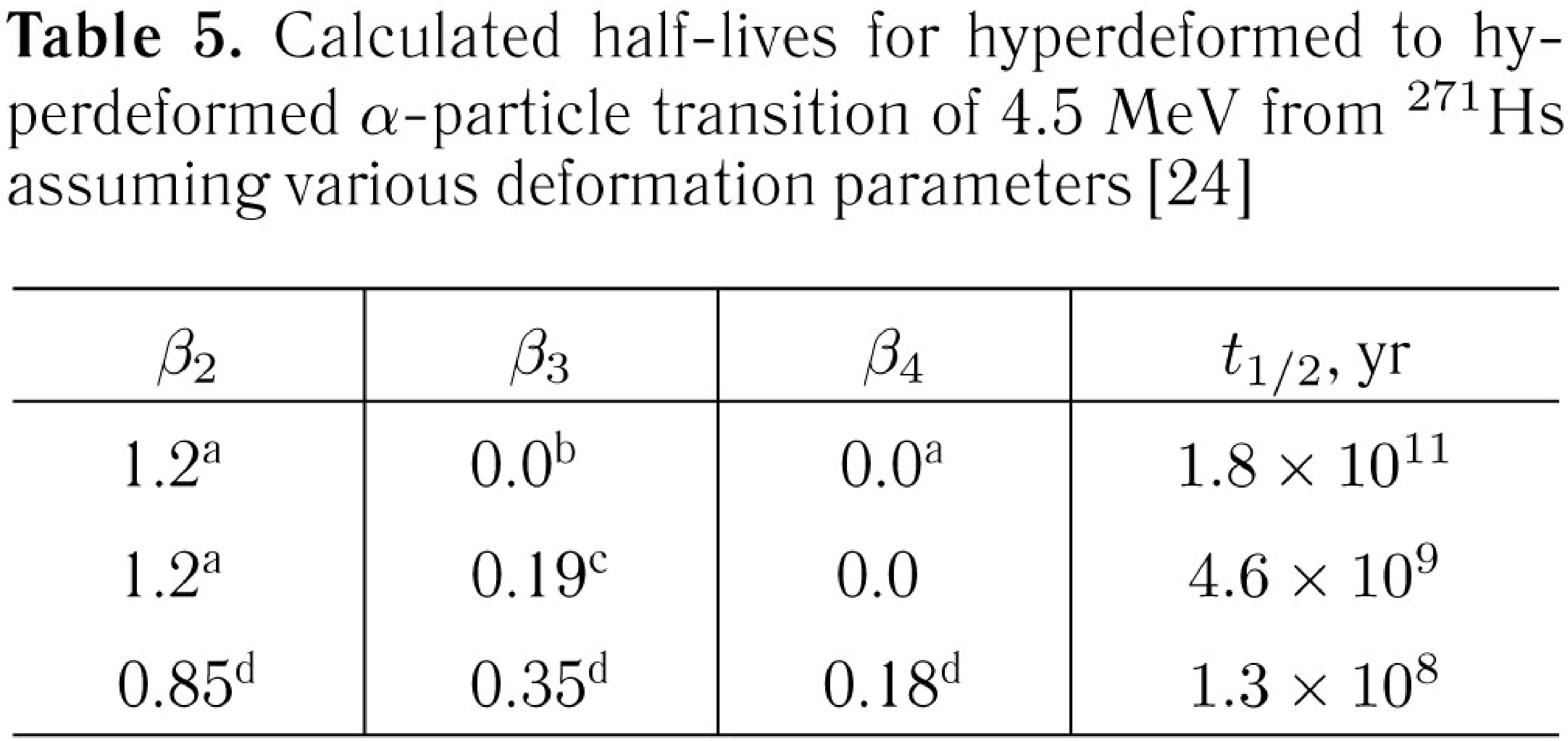


Table 5. Calculated half-lives for hyperdeformed to hyperdeformed α-particle transition of 4.5 MeV from ^{271}Hs assuming various deformation parameters [24]

β_2	β_3	β_4	$t_{1/2}$, yr
1.2[a]	0.0[b]	0.0[a]	1.8×10^{11}
1.2[a]	0.19[c]	0.0	4.6×10^{9}
0.85[d]	0.35[d]	0.18[d]	1.3×10^{8}

[d] Parameters from S. Ćwiok et al. (Phys. Lett. **B322,** 304 (1994)) for ^{232}Th.

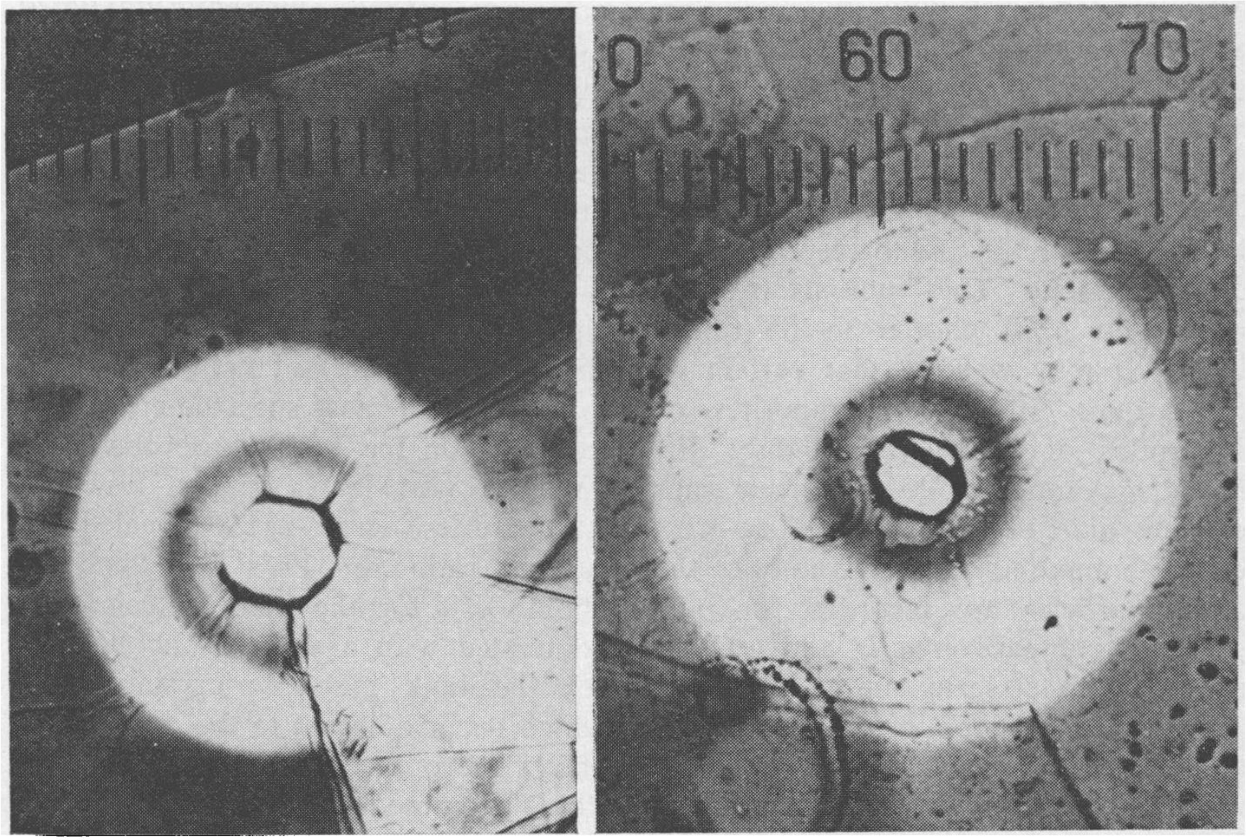


Fig. 2 (left). A giant halo approximately 57 μm in radius, presumably due to the long-range alpha particles from Po^{212} ($E = 10.55$ Mev). One scale division = 10 μm. Fig. 3 (right). A giant halo approximately 84 μm in radius, whose origin is unknown. If the halo is due to long-range alpha particles, the energy would be about 13.1 Mev. One scale division = 10 μm.

Giant Haloes

If alphas:

Left: 10.5 MeV

Right:13.1 MeV

Gentry, Sceince. 1970

10.5 MeV α-particles are expected to occur in SHE around Z = 108 to 114 with

$t_{1/2} \approx 10^{-4}$ to 1 s

13 MeV α-particles are expected to occur in SHE around Z = 122 to 126 with

$t_{1/2} \approx 10^{-4}$ s.

However, if there are long-lived isomeric states that decay by betas or low energy α-particles eventually to the g.s. of isotopes in these regions, then one can understand the existence of these giant halo.

These observations motivated us to search for long-lived isomeric states in natural materials.

Most experiments in the past searched for SHE in nature by looking for fission activities.

However: because thè radioactive decay law

$$dN/dt = - (1/\tau)xN$$

Then for $\tau \approx 10^8$ y one needs about 10^8 nuclei in order to see one dis/y

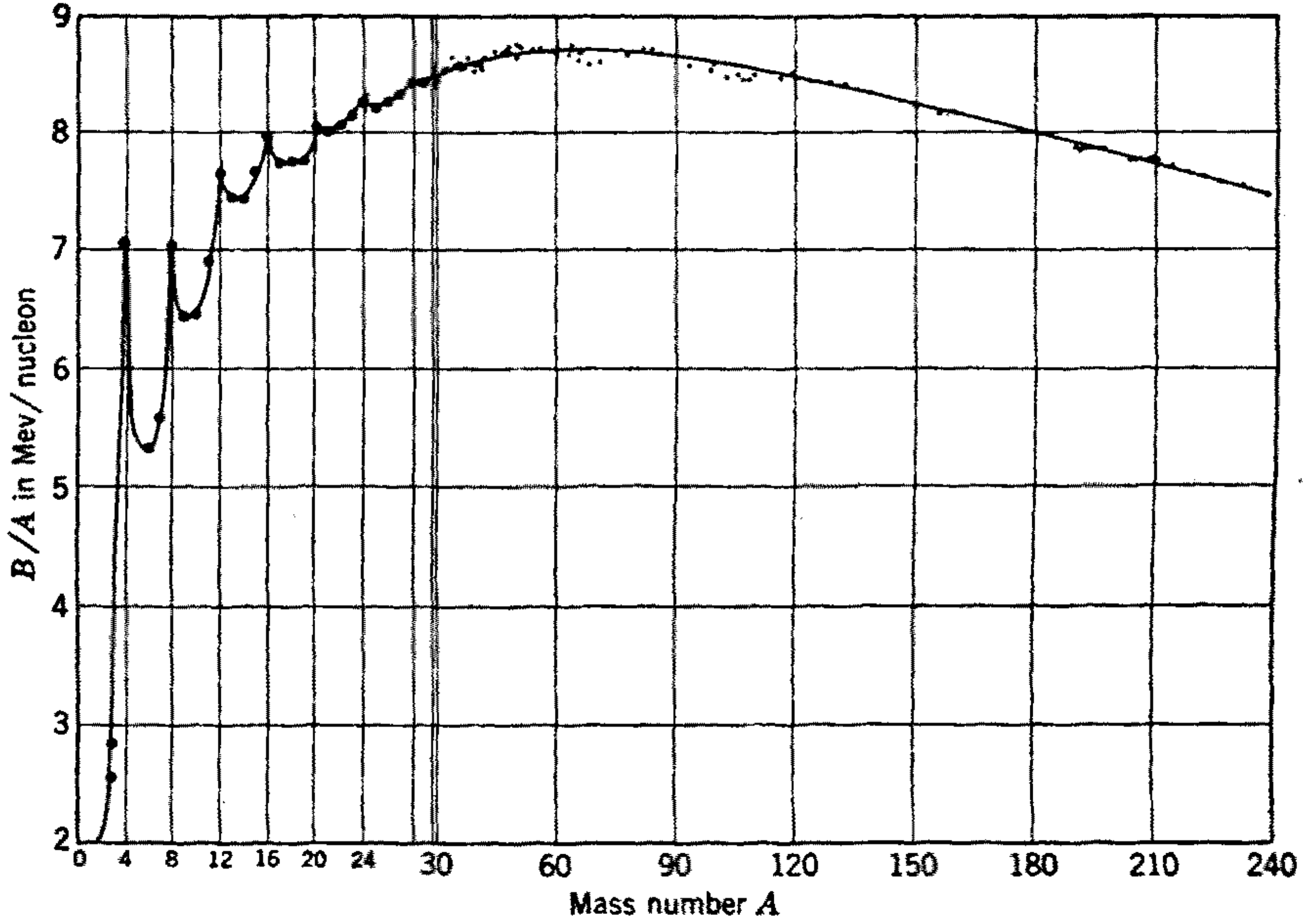
On the other hand for measuring N one needs about 10^6 atoms in an appropriate machine in order to see one event/s.

One way to measure the atomic mass number of nuclei is the AMS.

We tried it together with Michael Paul using monazite, with negative results.

Another way is to measure the accurate mass of an atom by using a high resolution mass spectrometer that is able to separate between the mass of an atom and the masses of molecules of the same mass number.

$$M_A = Z x M_H + N x M_n - BE$$



The mass of any molecule (except for multi-hydrogen molecules, or multi-Li, Be, and B molecules) is lower than the mass of an atom with the same mass number.

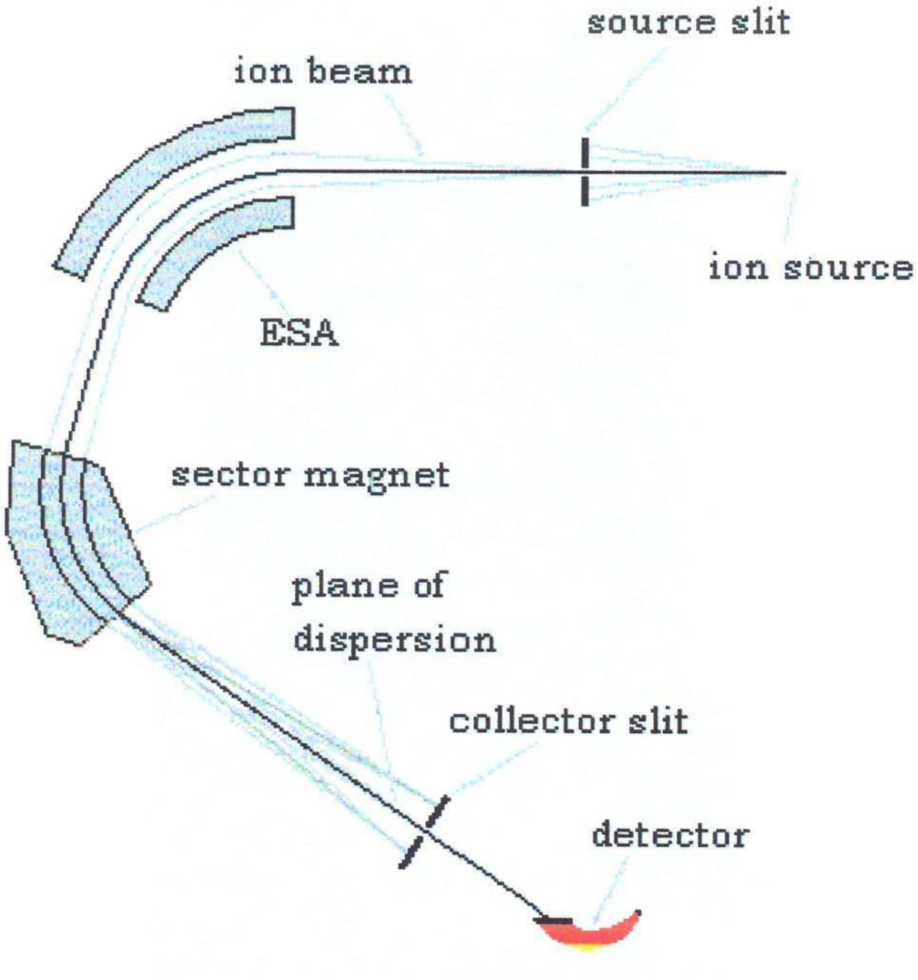
Inductively Coupled Plasma-Sector Field Mass Spectrometer (ICP-SFMS)

Plasma source at
6000-8000 K

Mostly atoms
from the source

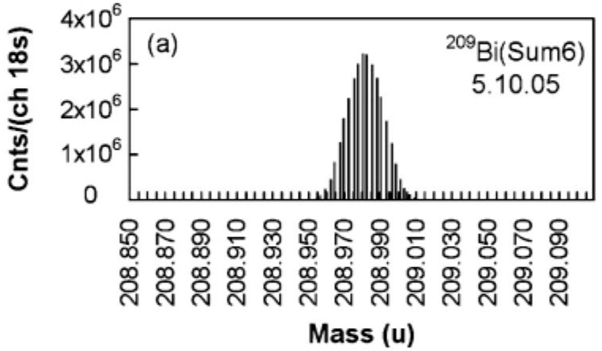
Studied material:
Solution

M/ΔM = 4000

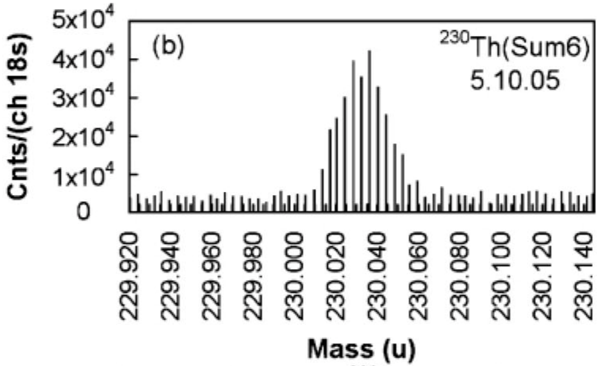


First: Measured neutron-deficient nuclei from pure Th solution

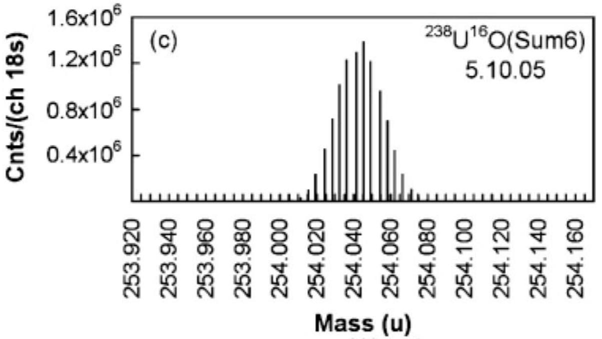
FWHM = 0.030 u



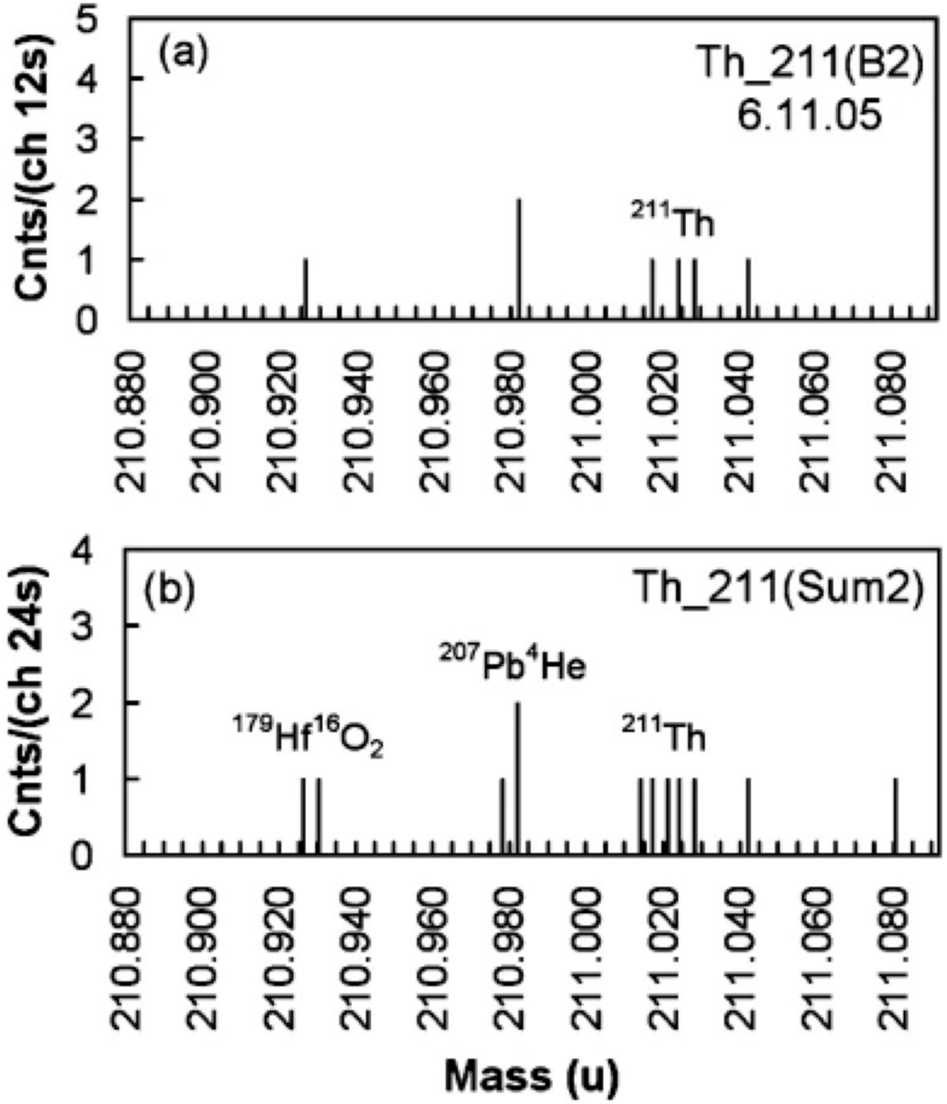
$M_{exp.}$(c.m.) = 208.981 u; M(^{209}Bi) = 208.980 u

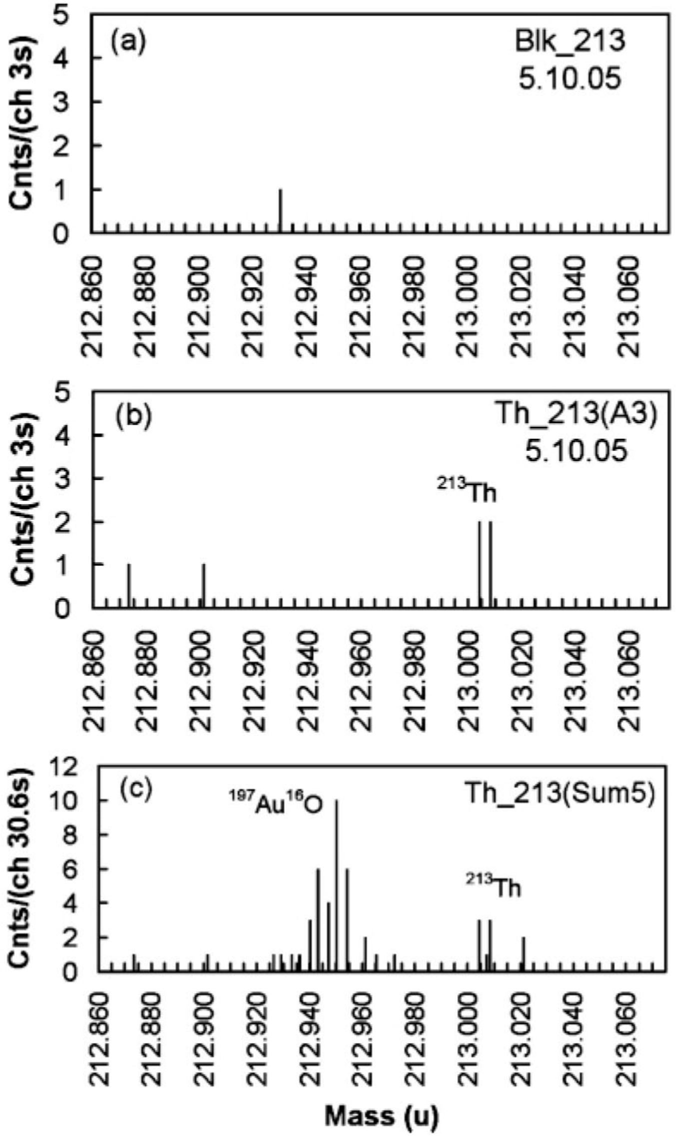


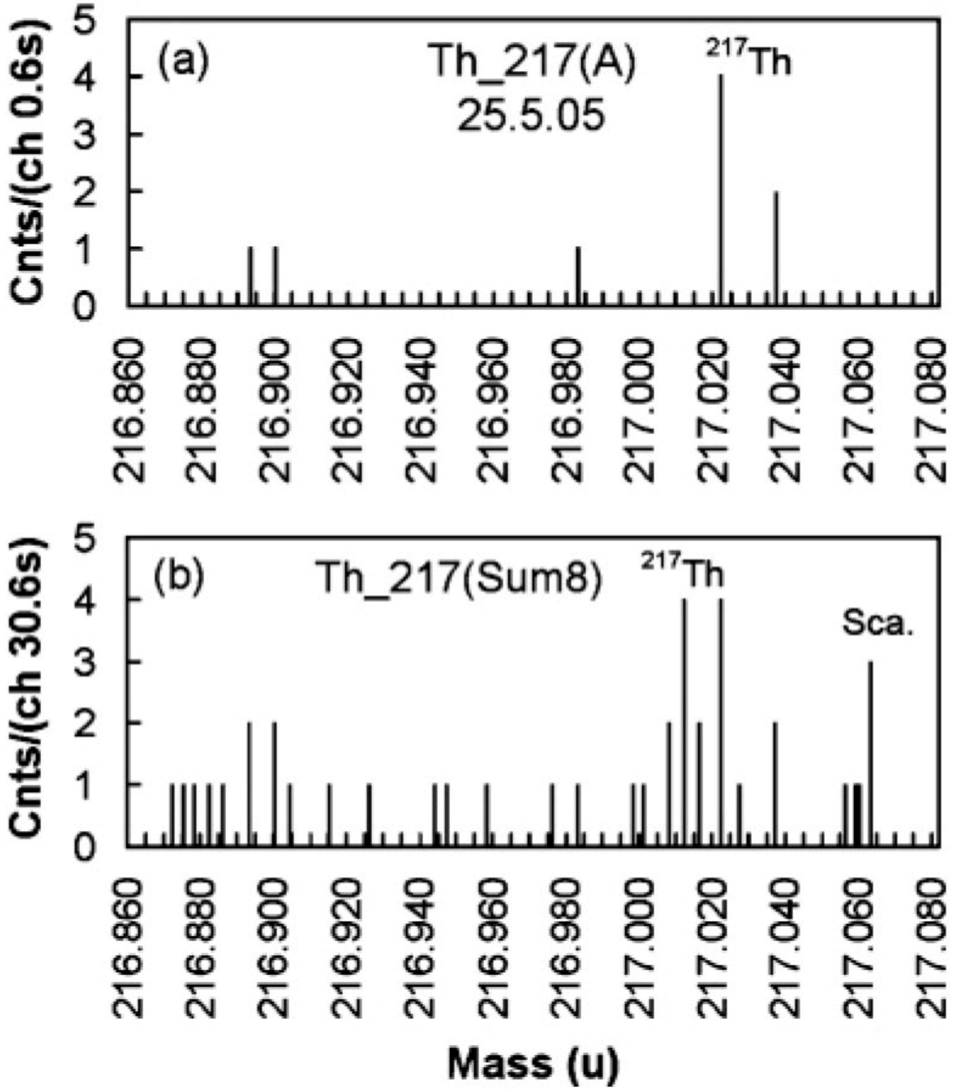
$M_{exp.}$(c.m.) = 230.035 u; M(^{230}Th) = 230.033 u



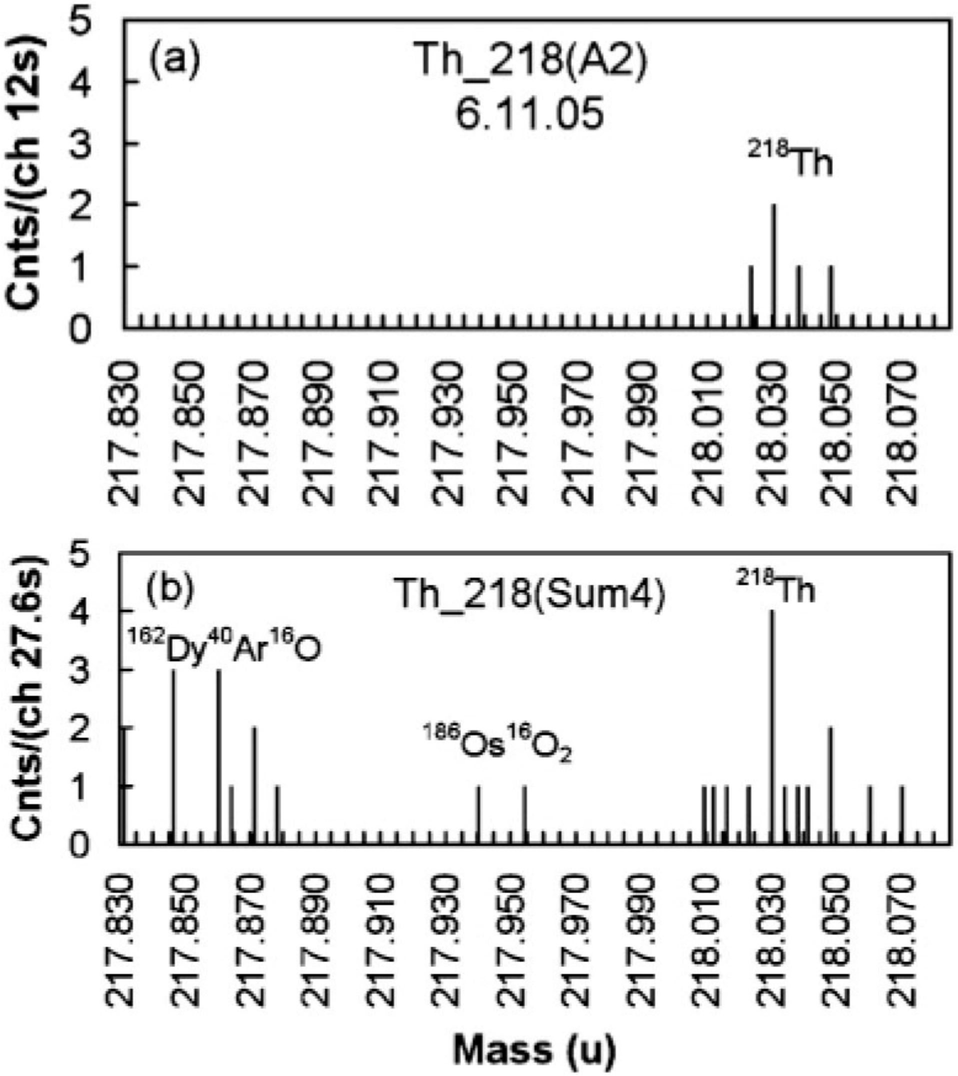
$M_{exp.}$(c.m.) = 254.043 u; M(^{238}U^{16}O) = 254.045 u


(a)
Th_211(B2)
6.11.05
^{211}Th
Cnts/(ch 12s)
0
1
2
3
4
5
210.880
210.900
210.920
210.940
210.960
210.980
211.000
211.020
211.040
211.060
211.080
(b)
Th_211(Sum2)
^{207}Pb^{4}He
^{179}Hf^{16}O$_2$
^{211}Th
Cnts/(ch 24s)
0
1
2
3
4
210.880
210.900
210.920
210.940
210.960
210.980
211.000
211.020
211.040
211.060
211.080
Mass (u)


(a)
Blk_213
5.10.05
Cnts/(ch 3s)
0
1
2
3
4
5
(b)
Th_213(A3)
5.10.05
^{213}Th
Cnts/(ch 3s)
(c)
^{197}Au^{16}O
Th_213(Sum5)
^{213}Th
Cnts/(ch 30.6s)
0
2
4
6
8
10
12
212.860
212.880
212.900
212.920
212.940
212.960
212.980
213.000
213.020
213.040
213.060
Mass (u)



(a)
Th_217(A)
25.5.05
^{217}Th
Cnts/(ch 0.6s)
0
1
2
3
4
5
216.860
216.880
216.900
216.920
216.940
216.960
216.980
217.000
217.020
217.040
217.060
217.080
(b)
Th_217(Sum8)
^{217}Th
Sca.
Cnts/(ch 30.6s)
0
1
2
3
4
5
216.860
216.880
216.900
216.920
216.940
216.960
216.980
217.000
217.020
217.040
217.060
217.080
Mass (u)


(a)
Th_218(A2)
6.11.05
^{218}Th
Cnts/(ch 12s)
0
1
2
3
4
5
217.830
217.850
217.870
217.890
217.910
217.930
217.950
217.970
217.990
218.010
218.030
218.050
218.070
(b)
Th_218(Sum4)
^{218}Th
^{162}Dy^{40}Ar^{16}O
^{186}Os^{16}O$_2$
Cnts/(ch 27.6s)
0
1
2
3
4
5
217.830
217.850
217.870
217.890
217.910
217.930
217.950
217.970
217.990
218.010
218.030
218.050
218.070
Mass (u)

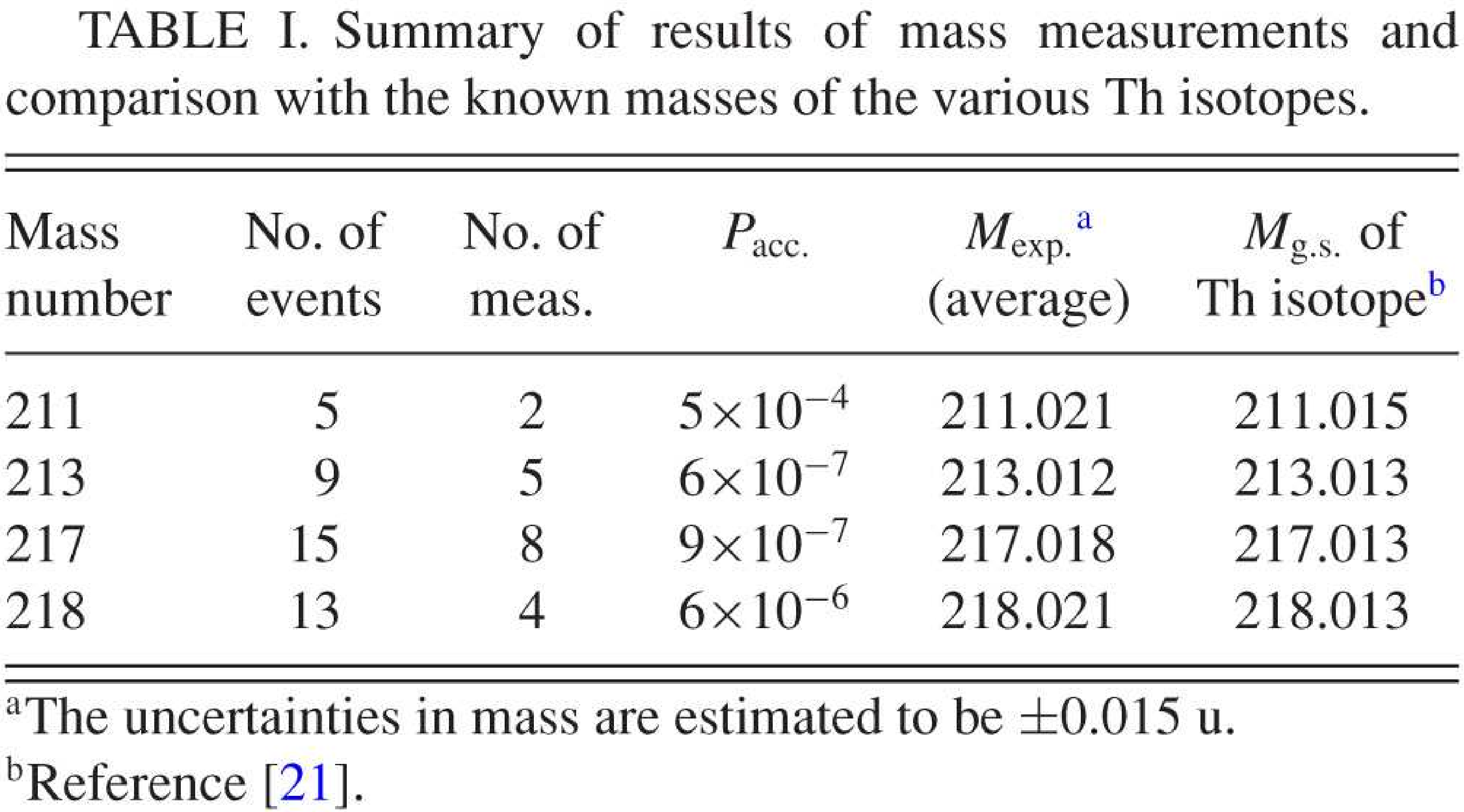


TABLE I. Summary of results of mass measurements and comparison with the known masses of the various Th isotopes.

Mass number	No. of events	No. of meas.	$P_{\mathrm{acc.}}$	$M_{\mathrm{exp.}}$[a] (average)	$M_{\mathrm{g.s.}}$ of Th isotope[b]
211	5	2	5×10^{-4}	211.021	211.015
213	9	5	6×10^{-7}	213.012	213.013
217	15	8	9×10^{-7}	217.018	217.013
218	13	4	6×10^{-6}	218.021	218.013

[a] The uncertainties in mass are estimated to be ±0.015 u.

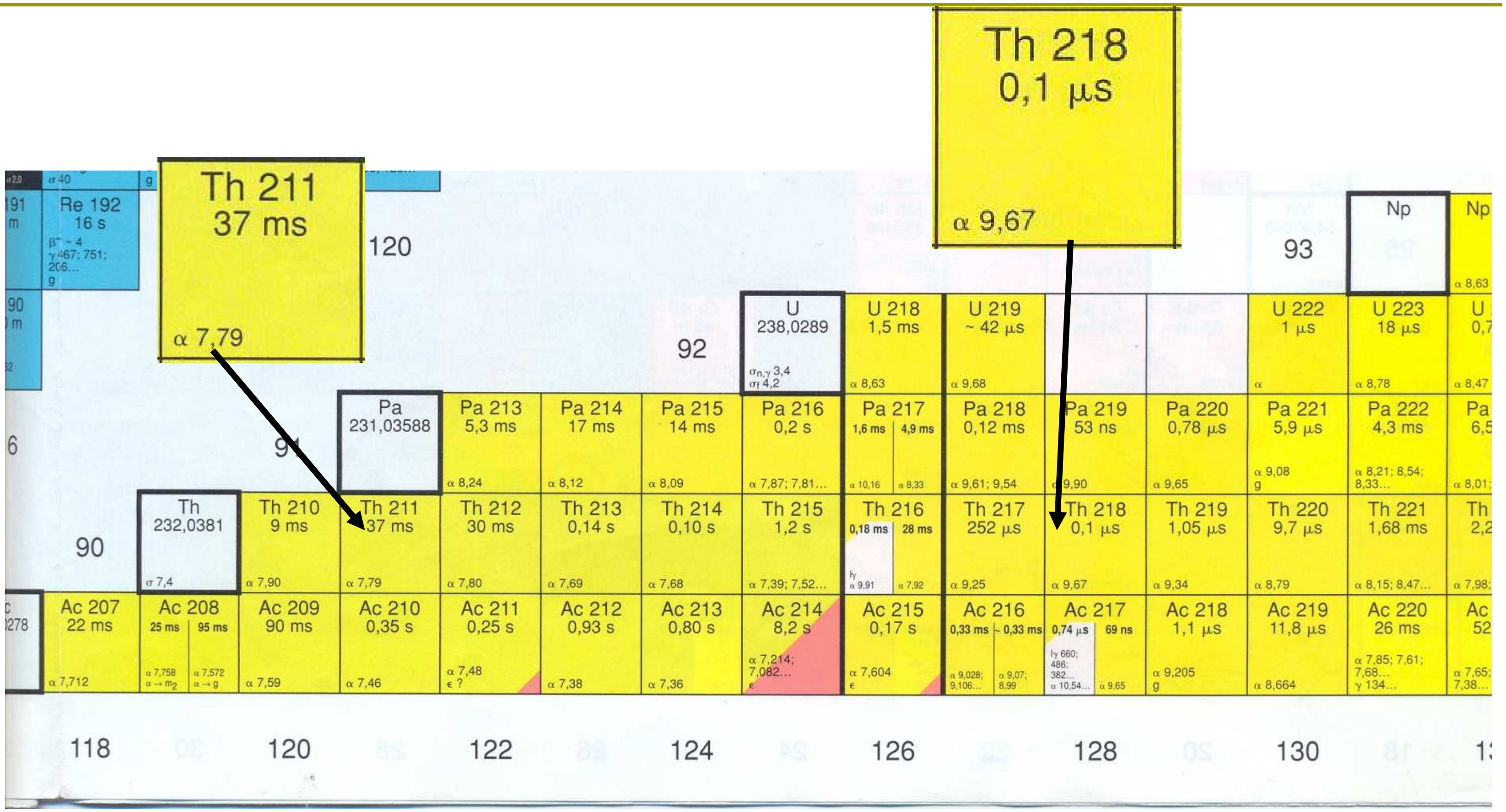
[b] Reference [21].

All together we saw 42 events in 19 independent measurements.

The relative abundance of these isotopes compared to ^{232}Th is $(1 - 10) \times 10^{-11}$

$(2-20) \times 10^{-16}$ of the solution.

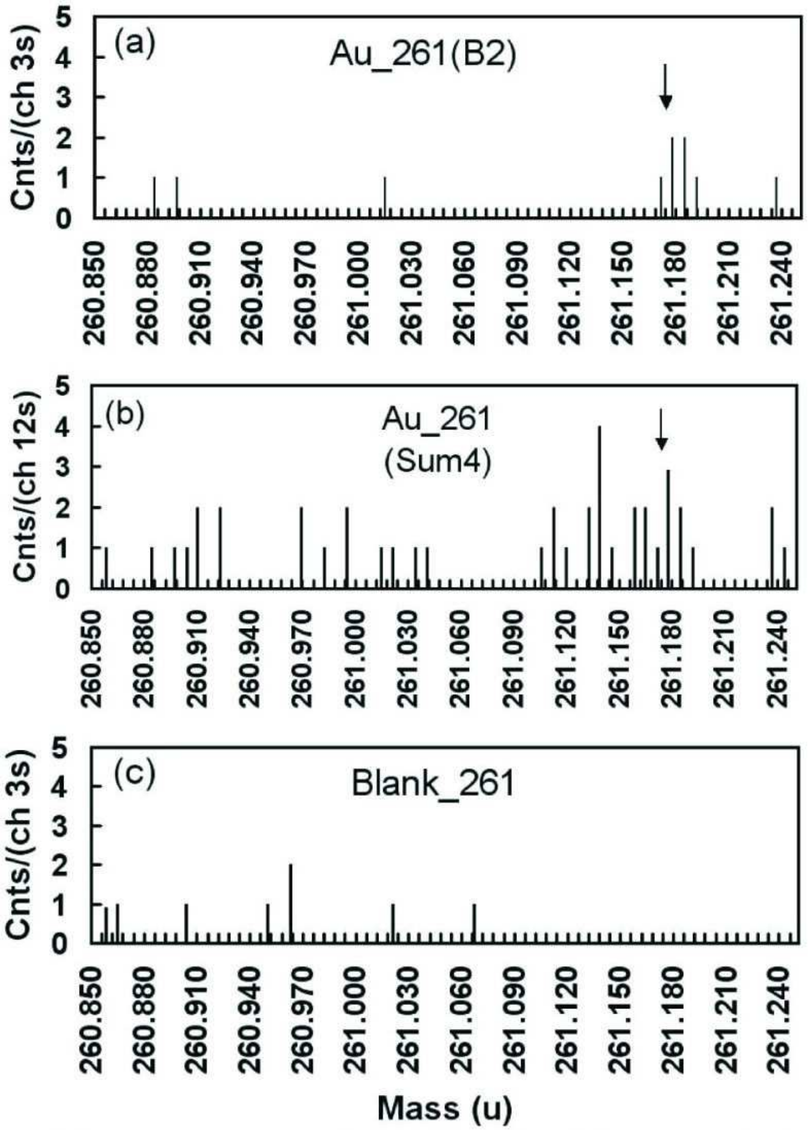
If the terrestrial concentration of these isotopes were initially the same as of ^{232}Th then $t_{1/2} \geq 10^8$ y.

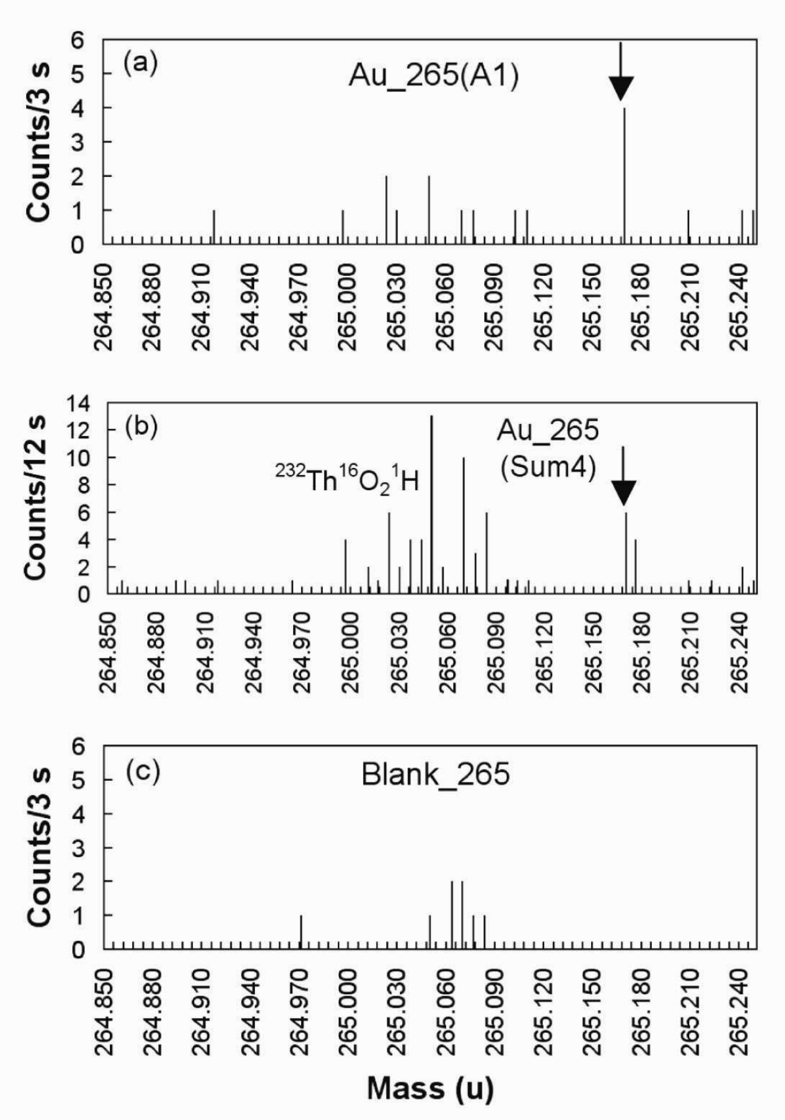

Th 211
37 ms
α 7,79
Th 218
0,1 µs
α 9,67
90
91
92
93
118
120
122
124
126
128
130

Conclusion: Long-lived isomeric states with half-lives 10^{16} to 10^{22} longer than their corresponding g.s. have been found in the neutron-deficient ^{211,213,217,218}Th nuclei.

PRC **76**, 021303(R) (2007)

Our second experiment was to search for long-lived isomeric states in pure Au solution looking for high masses, assuming that if Rg (eka-Au, element 111) exists in nature it may be found together with Au.


(a)
Au_261(B2)
Cnts/(ch 3s)
(b)
Au_261
(Sum4)
Cnts/(ch 12s)
(c)
Blank_261
Cnts/(ch 3s)
0
1
2
3
4
5
260.850
260.880
260.910
260.940
260.970
261.000
261.030
261.060
261.090
261.120
261.150
261.180
261.210
261.240
Mass (u)


(a)
Au_265(A1)
Counts/3 s
(b)
Au_265
(Sum4)
$^{232}Th^{16}O_2{}^1H$
Counts/12 s
(c)
Blank_265
Counts/3 s
264.850
264.880
264.910
264.940
264.970
265.000
265.030
265.060
265.090
265.120
265.150
265.180
265.210
265.240
Mass (u)

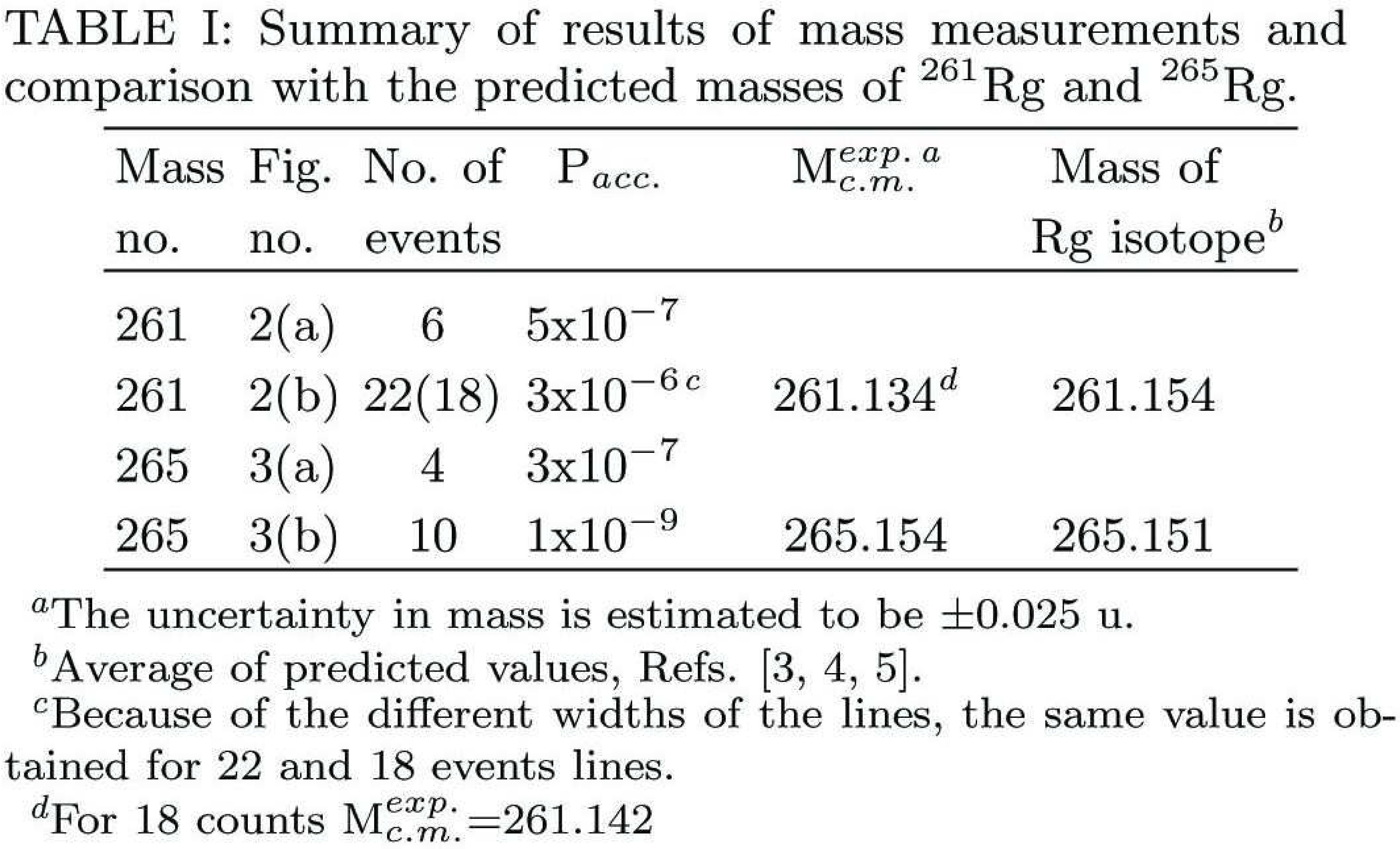


TABLE I: Summary of results of mass measurements and comparison with the predicted masses of ^{261}Rg and ^{265}Rg.

Mass no.	Fig. no.	No. of events	$P_{acc.}$	$M_{c.m.}^{exp.\,a}$	Mass of Rg isotope[b]
261	2(a)	6	$5\text{x}10^{-7}$		
261	2(b)	22(18)	$3\text{x}10^{-6\,c}$	261.134[d]	261.154
265	3(a)	4	$3\text{x}10^{-7}$		
265	3(b)	10	$1\text{x}10^{-9}$	265.154	265.151

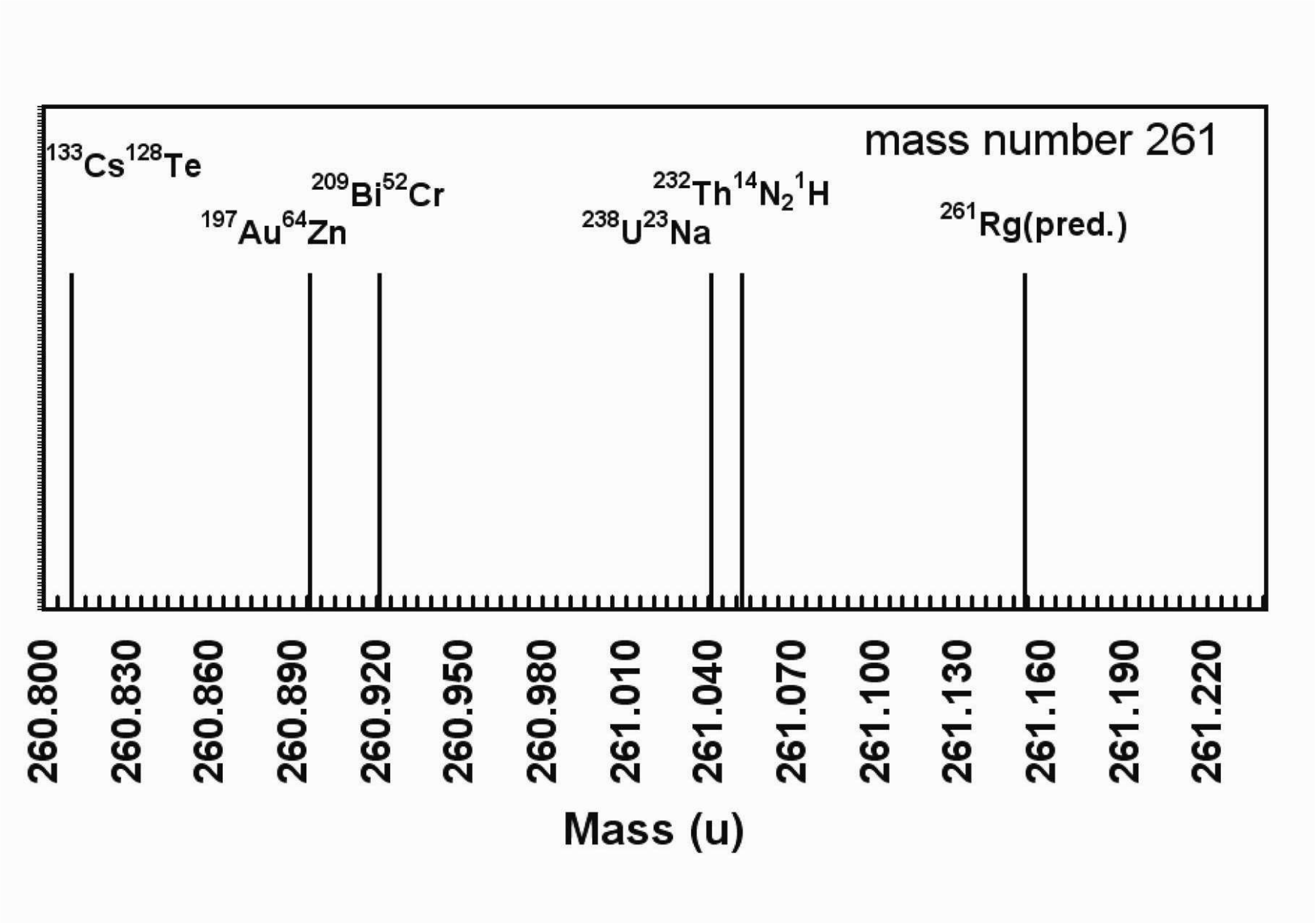
[a] The uncertainty in mass is estimated to be ±0.025 u.

[b] Average of predicted values, Refs. [3, 4, 5].

[c] Because of the different widths of the lines, the same value is obtained for 22 and 18 events lines.

[d] For 18 counts $M_{c.m.}^{exp.}$=261.142

All together we saw about 40 events in eight independent measurements.



Predictions: a) Möller *et al. (1995)* b) Koura *et al. (2005)*

c) Liran, Marinov and Zeldes (2000)

The relative abundance of these isotopes compared to ^{197}Au is about 1×10^{-10}

2×10^{-15} of the solution.

The chemical properties of Sg(106), Bh(107), Hs(108) and element 112

were found to be similar to those of their lighter homologues,

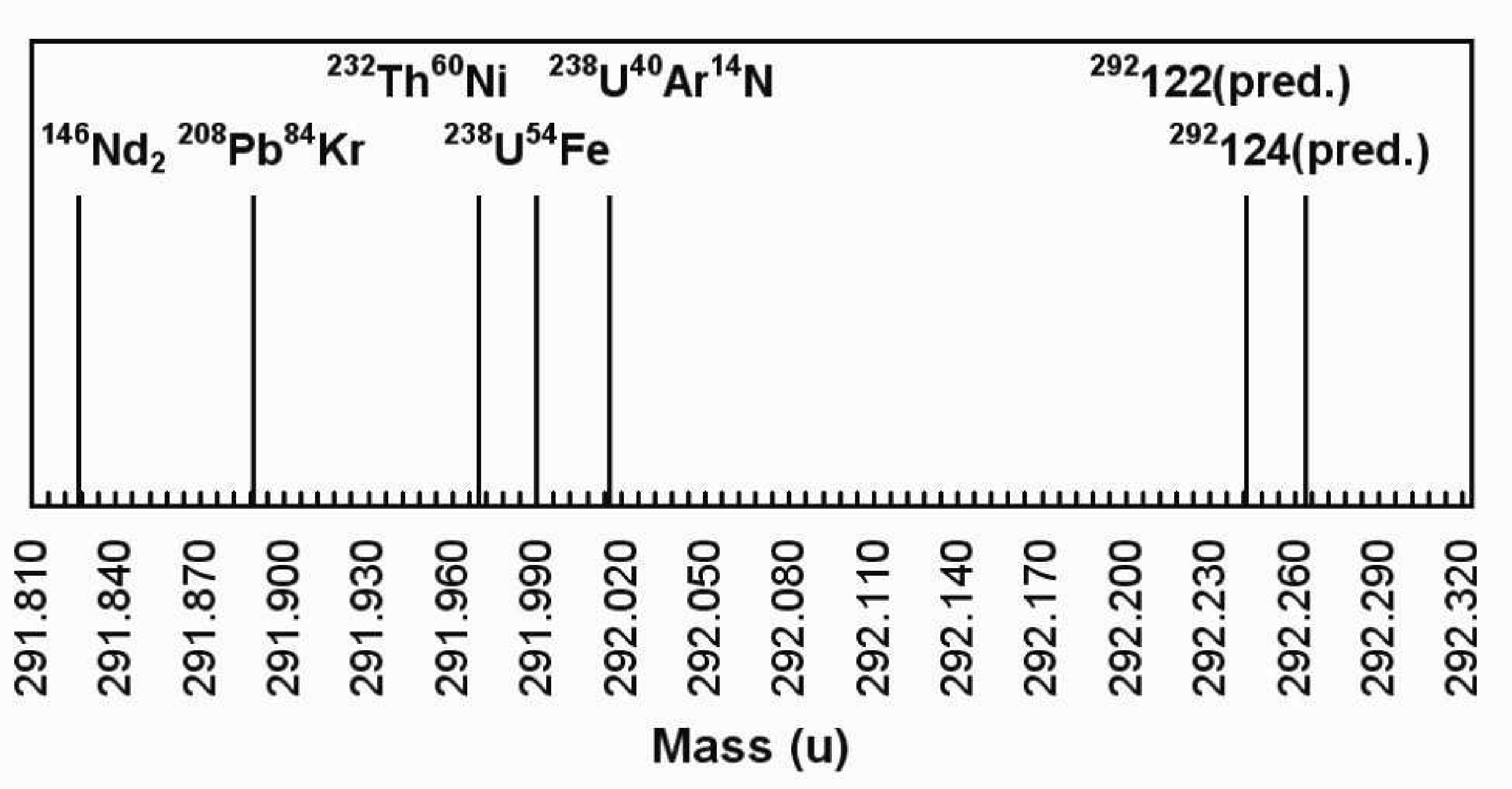
W, Re, Os, and Hg.

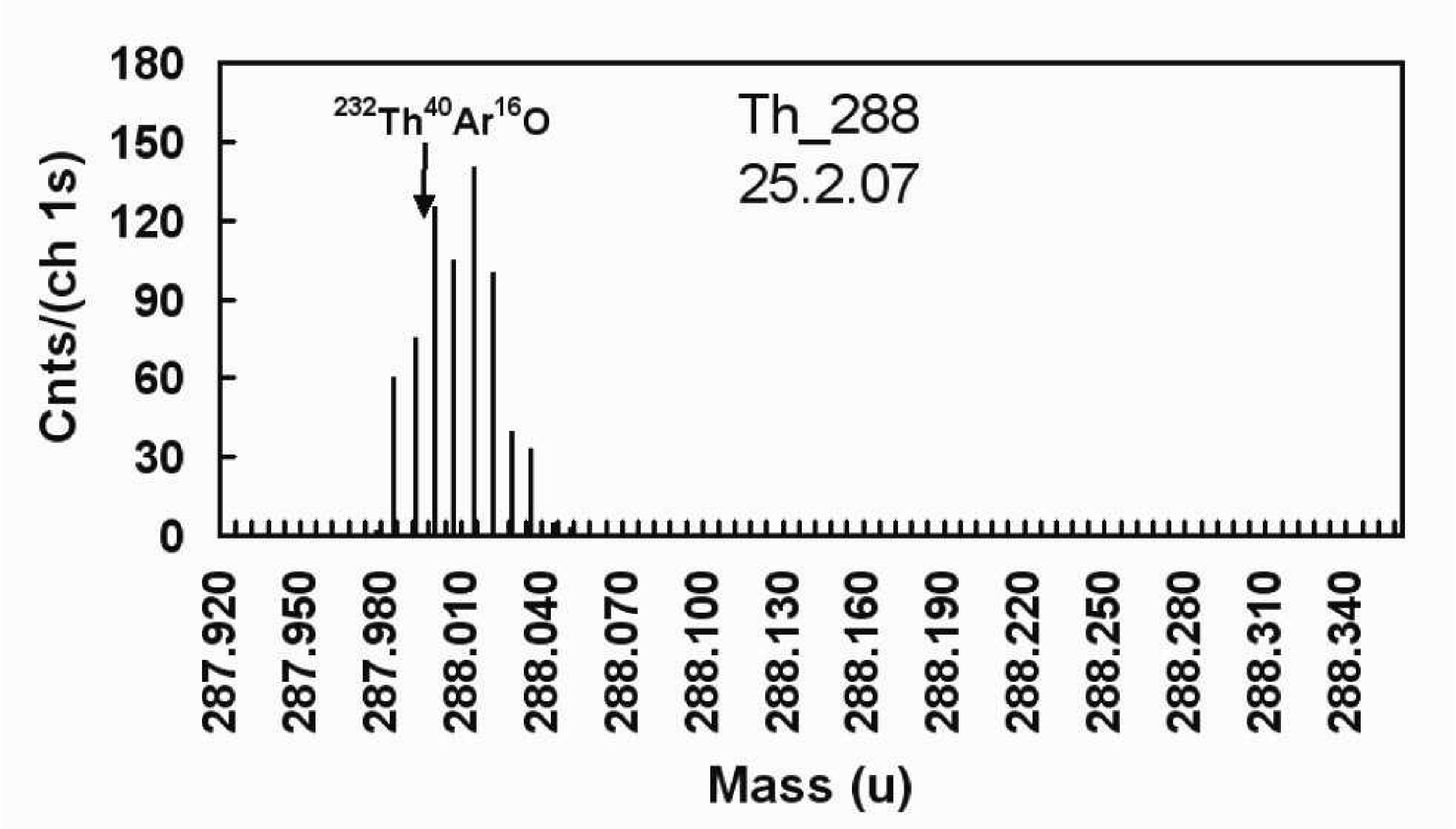
Therefore one may assume that the observed A=261 and 265 nuclei are ^{261}Rg and ^{265}Rg (element 111).

Third experiment:

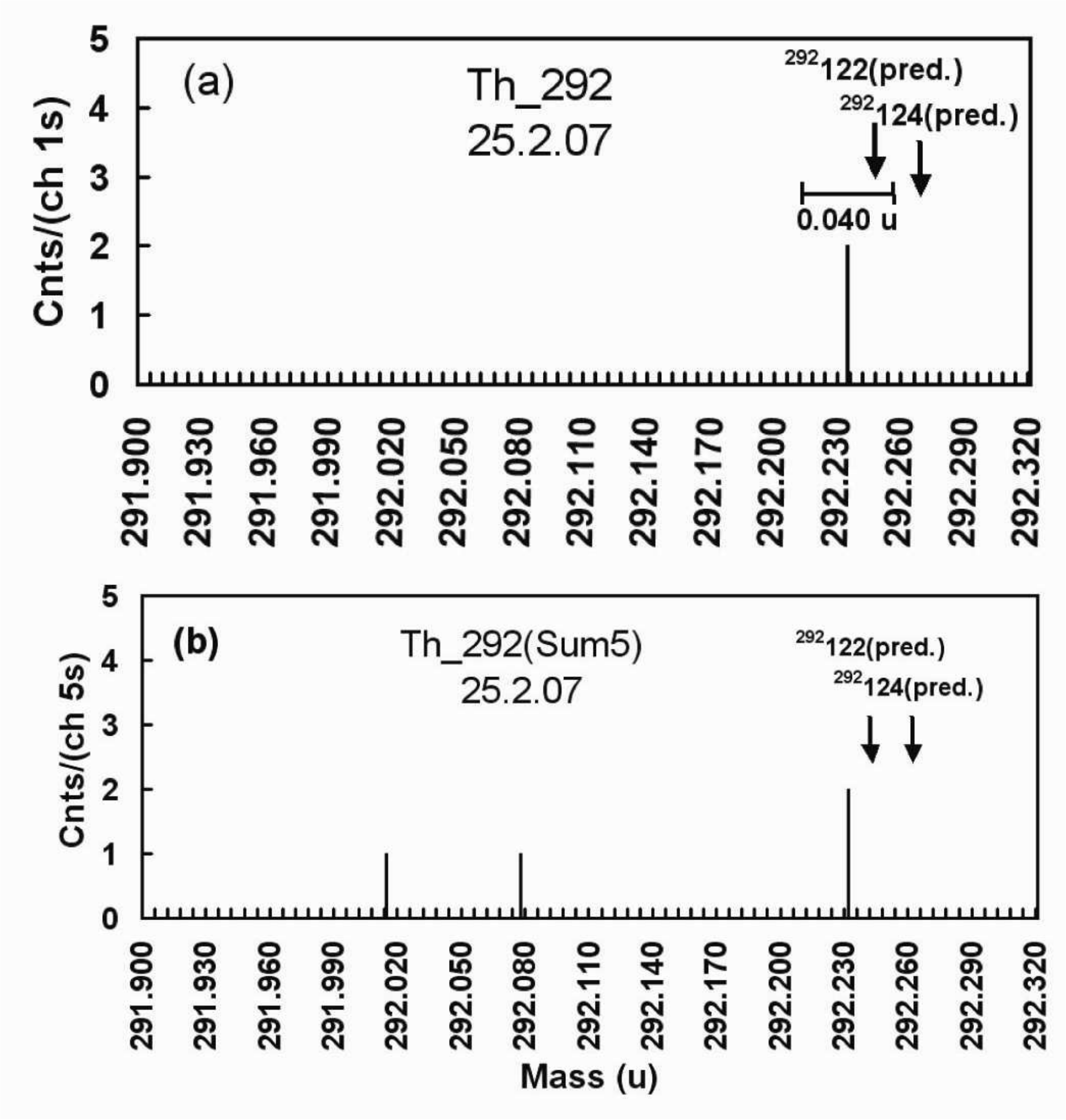
Search for SHE in Th solution but at high masses from 287 to 294, looking for superactinide nuclei. According to the extended periodic table of Seaborg elements 122 and 124 are placed as eka-Th and eka-U, respectively.

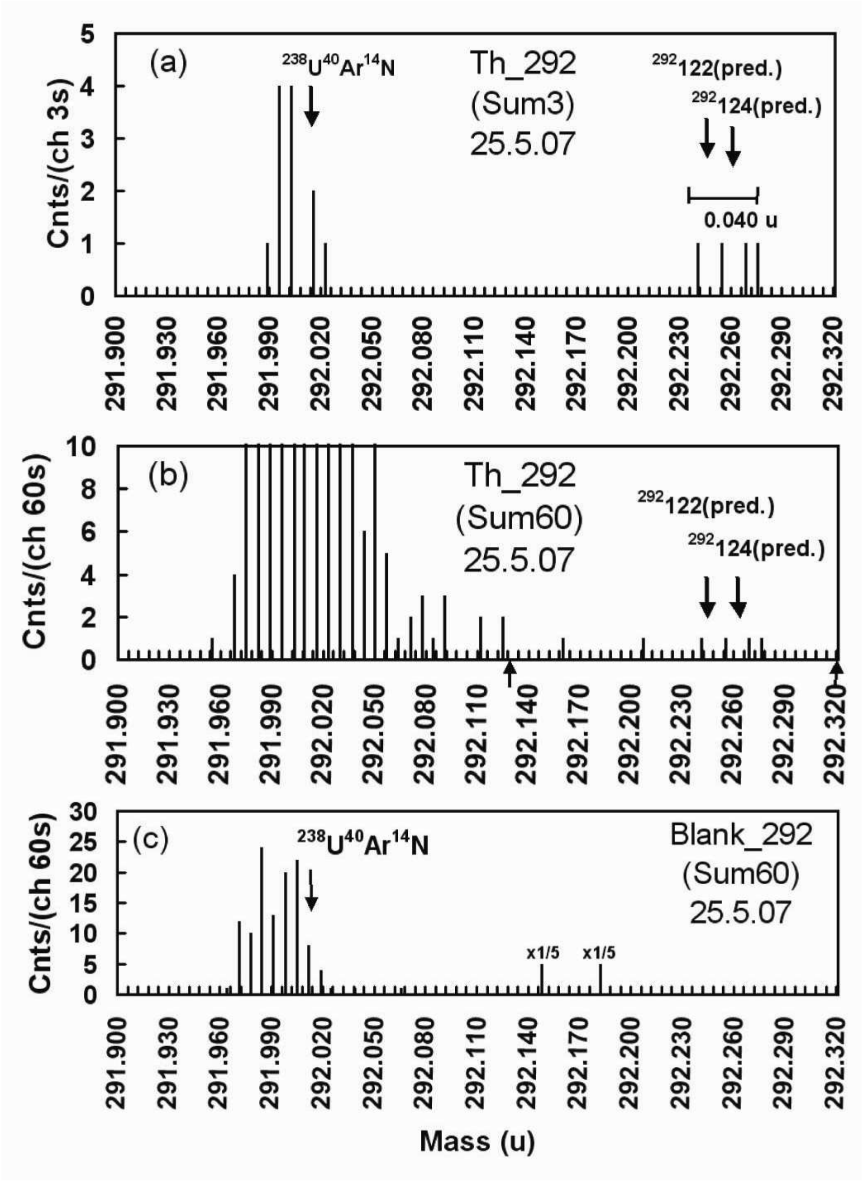
(


$^{232}Th^{60}Ni$
$^{238}U^{40}Ar^{14}N$
$^{292}122$(pred.)
$^{146}Nd_2$
$^{208}Pb^{84}Kr$
$^{238}U^{54}Fe$
$^{292}124$(pred.)
291.810
291.840
291.870
291.900
291.930
291.960
291.990
292.020
292.050
292.080
292.110
292.140
292.170
292.200
292.230
292.260
292.290
292.320
Mass (u)

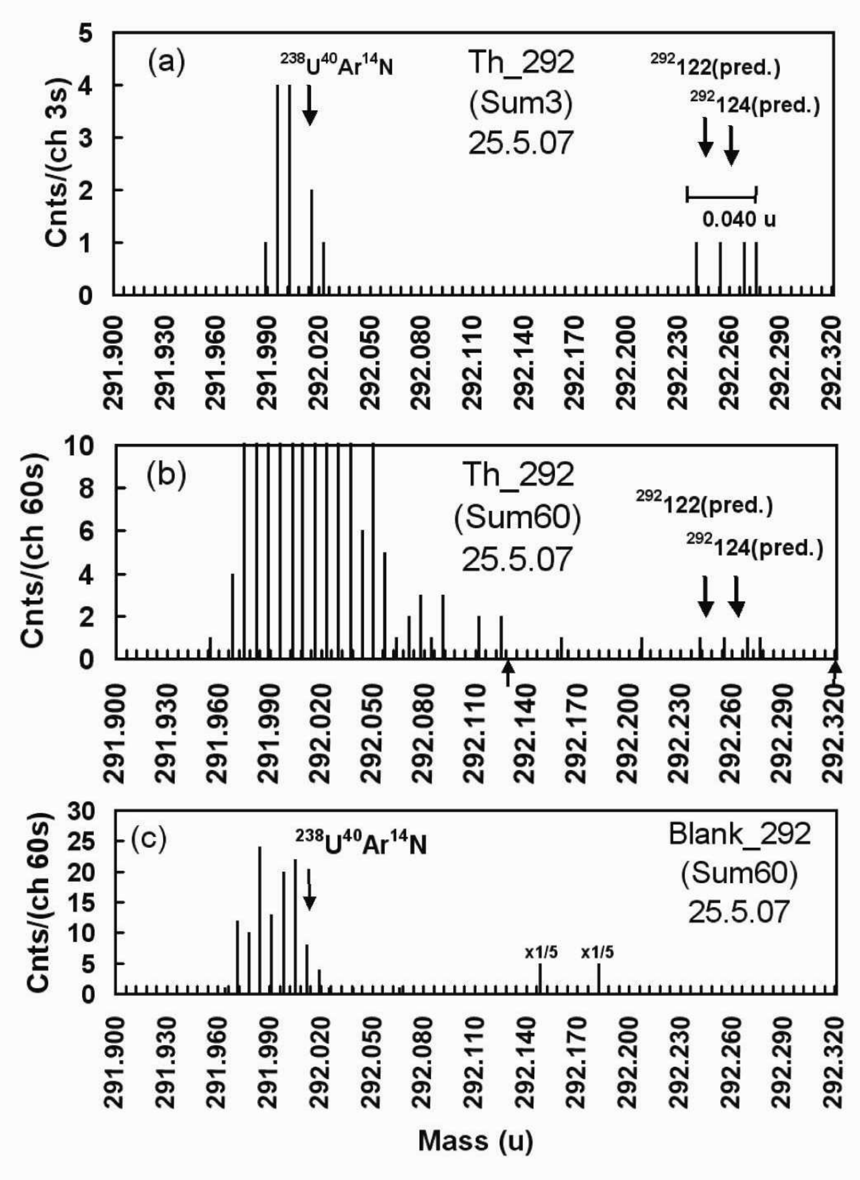


180
150
120
90
60
30
0
Cnts/(ch 1s)
$^{232}Th^{40}Ar^{16}O$
Th_288
25.2.07
287.920
287.950
287.980
288.010
288.040
288.070
288.100
288.130
288.160
288.190
288.220
288.250
288.280
288.310
288.340
Mass (u)

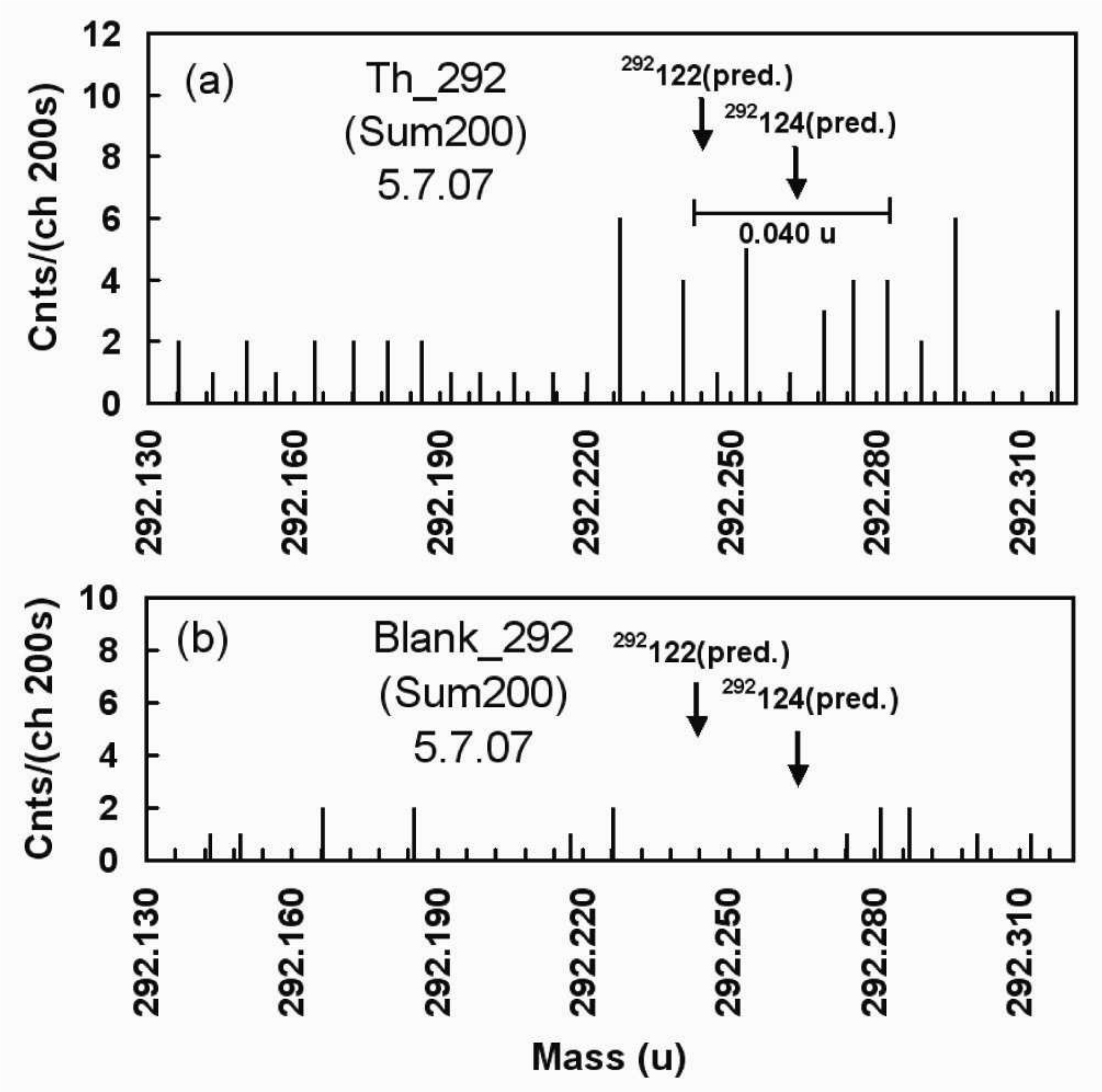

(a)
Th_292
25.2.07
^{292}122(pred.)
^{292}124(pred.)
0.040 u
Cnts/(ch 1s)
0
1
2
3
4
5
291.900
291.930
291.960
291.990
292.020
292.050
292.080
292.110
292.140
292.170
292.200
292.230
292.260
292.290
292.320
(b)
Th_292(Sum5)
25.2.07
^{292}122(pred.)
^{292}124(pred.)
Cnts/(ch 5s)
Mass (u)


(a)
$^{238}U^{40}Ar^{14}N$
Th_292
(Sum3)
25.5.07
^{292}122(pred.)
^{292}124(pred.)
0.040 u
Cnts/(ch 3s)
0
1
2
3
4
5
291.900
291.930
291.960
291.990
292.020
292.050
292.080
292.110
292.140
292.170
292.200
292.230
292.260
292.290
292.320
(b)
Th_292
(Sum60)
25.5.07
^{292}122(pred.)
^{292}124(pred.)
Cnts/(ch 60s)
0
2
4
6
8
10
291.900
291.930
291.960
291.990
292.020
292.050
292.080
292.110
292.140
292.170
292.200
292.230
292.260
292.290
292.320
(c)
$^{238}U^{40}Ar^{14}N$
Blank_292
(Sum60)
25.5.07
x1/5
x1/5
Cnts/(ch 60s)
0
5
10
15
20
25
30
291.900
291.930
291.960
291.990
292.020
292.050
292.080
292.110
292.140
292.170
292.200
292.230
292.260
292.290
292.320
Mass (u)

The reason that we don't see more events at the 60 runs is the following: One needs an abundance of about $1x10^{-10}$ in order to see 1 event/s. If the abundance is for instance $5x10^{-12}$ then one needs on the average about 20 runs of one sec. in order to see one event.



In the next experiment we focus on the limited region of 292.13u u to 292.32u


(a)
Th_292
(Sum200)
5.7.07
^{292}122(pred.)
^{292}124(pred.)
0.040 u
Cnts/(ch 200s)
0
2
4
6
8
10
12
292.130
292.160
292.190
292.220
292.250
292.280
292.310
(b)
Blank_292
(Sum200)
5.7.07
^{292}122(pred.)
^{292}124(pred.)
Cnts/(ch 200s)
0
2
4
6
8
10
292.130
292.160
292.190
292.220
292.250
292.280
292.310
Mass (u)

$M_{exp.} = 292.262 \pm 0.030$ u

Predictions (KTUY05; LMZ01)

For $^{292}121$ to $^{292}126$ are

292.236 u to 292.291 u

M(pred.) $^{292}122 = 292.243$ u

Abundance (relative to ^{232}Th):

about $1 \mathrm{x} 10^{-12}$

$t_{1/2} \geq 10^8$ y

Chemical arguments:

a) We used pure Th solution.

b) The atomic configuration of Th is $6d^2_{3/2}\,7s^2$ and its separation is based on its stable 4^+ oxidation state.

c) The accurate predicted atomic configuration of eka-Th (Z = 122) is $8s^2 7d_{3/2} 8p_{1/2}$. It is also expected to form a stable 4^+ state. (Eliav et al. (2002); Gaigalas et al. (2010))

d) Element 121 has only three electrons outside the closed shells of element 118 (eka-Rn) and it is not likely that it will form a stable 4^+ state,

e) Elements above Z=122 have more electrons. They can form 4^+ oxidation state, but also higher oxidation states.

f) If element 122 exists in nature together with Th, it is reasonable to assume that it followed Th in the chemical separation, and showed up in our measurements. However the possibility that A =292 Nucleus belongs to an element of somewhat higher Z cannot be excluded.

The predicted half-lives of nuclei around $^{292}122$ is:

$t_{1/2}$ (pred.) = 10^{-6} - 10^{-8} s.

Möller, Nix, Kratz, ADNDT (1997)

$t_{1/2}$ (exp.) $\geq 10^{8}$ y

Conclusion: What we found is an isomeric state in the nucleus

A = 292 and Z $\cong$ 122

Int. J. Mod. Phys. E19 (2010) 131

122/292

≥ 10^8 y

112/272

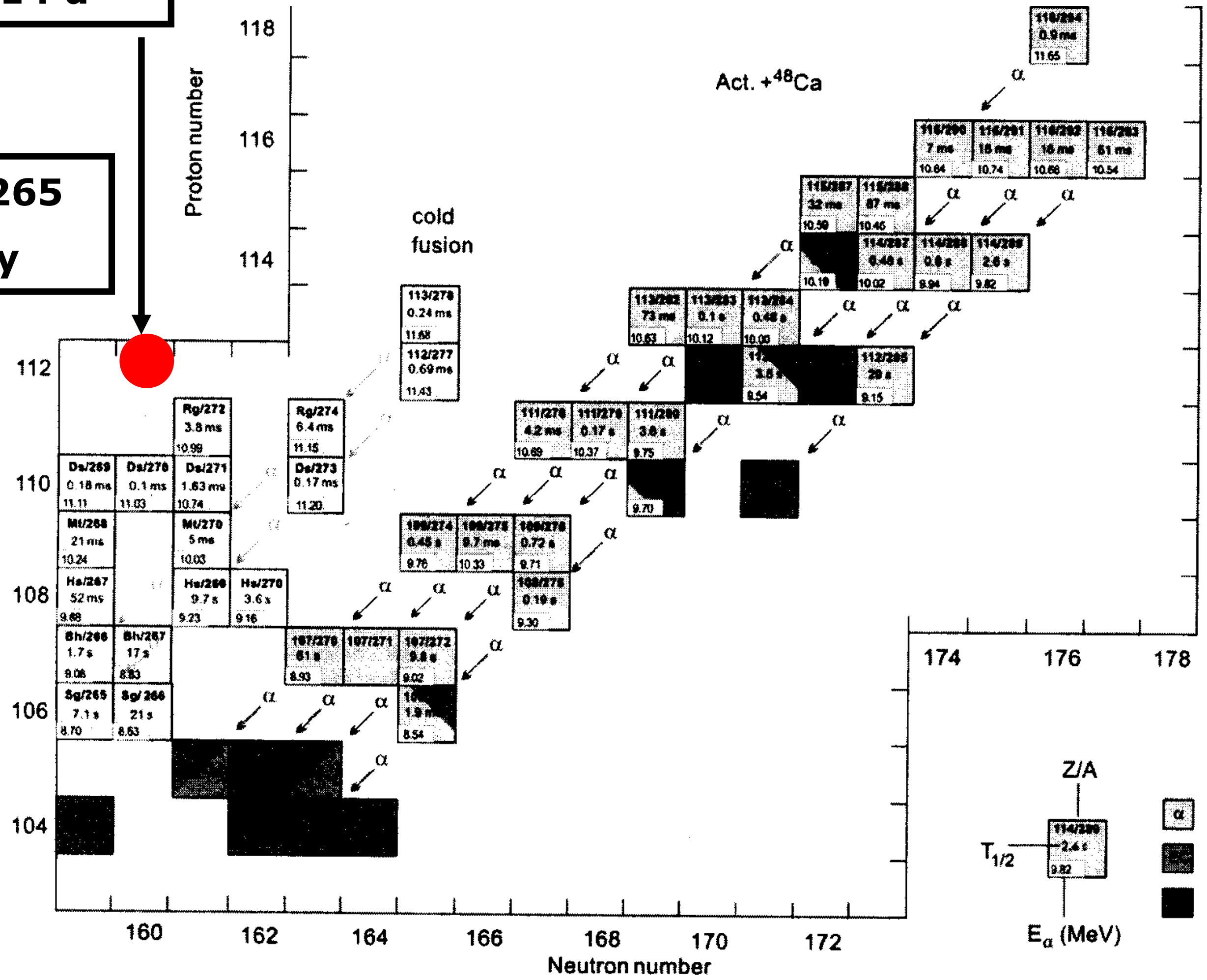
`~14 d

111/261

≥ 10^8 y

111/265

≥ 10^8 y



Yu. Oganessian, J. Phys. G: Nucl. Part. Phys. 39, R165 (2007)

Thank You for Your Attention

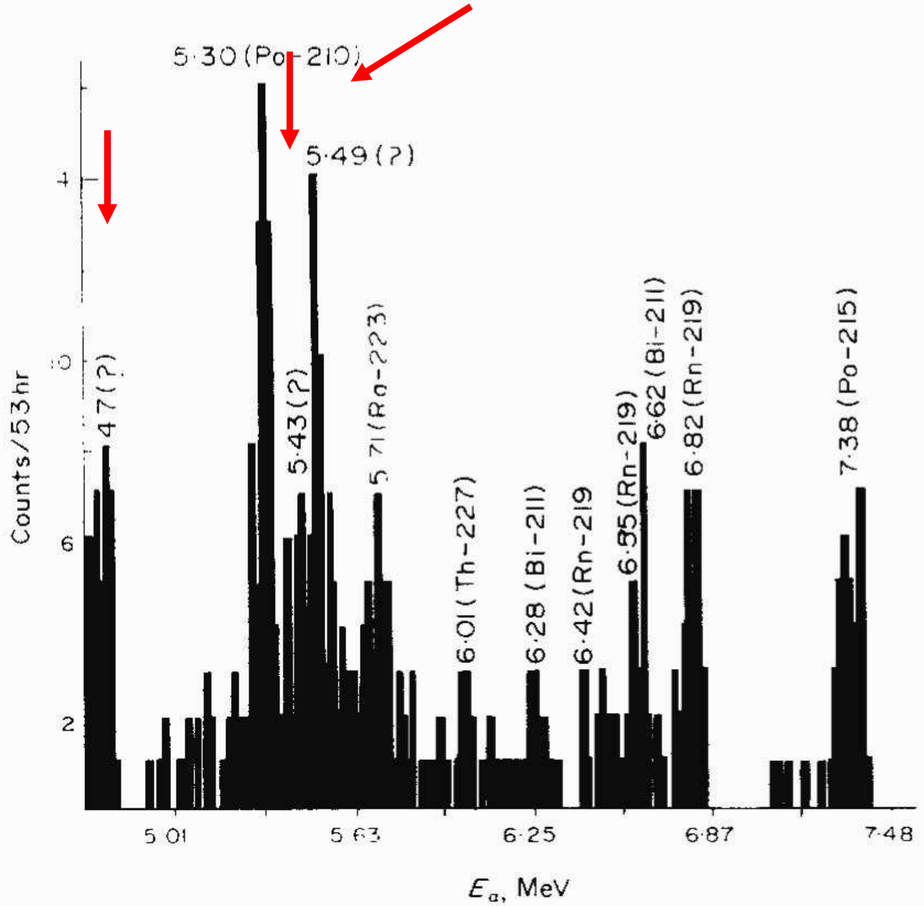


Fig. 1. Alpha-spectrum of the W II-sample. Energy calibration has been carried out with standards of Pu-239 (5·16 MeV), Am-241 (5·49 MeV) and Cm-244 (5·81 MeV).

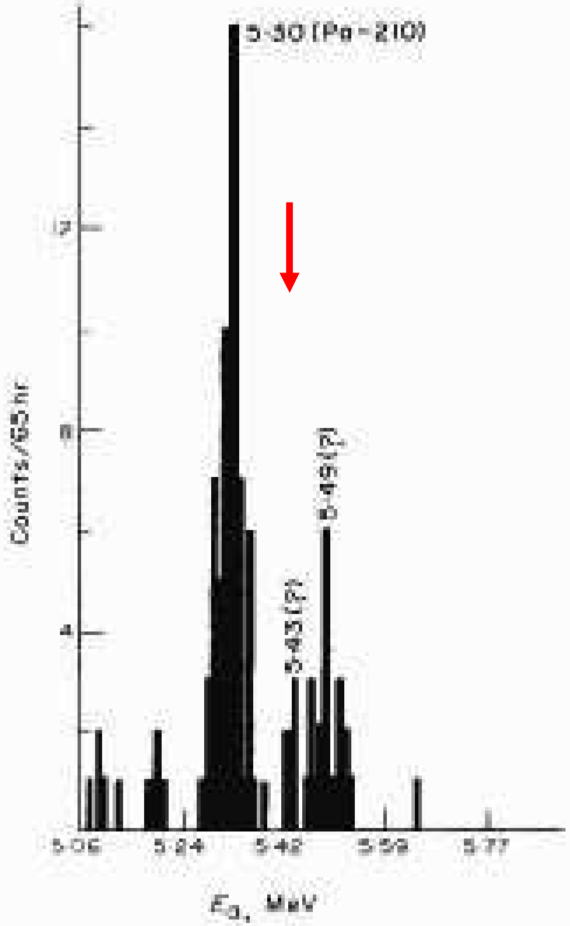


Fig. 2. Alpha-spectrum of the Au I-sample. Energy calibration as described in Fig. 1.

E. Ross et al., J. Inorg. Nucl. Chem. (1974)

- **Comparison with AMS**
- **Abundance of A=292**

ICP-SFMS	AMS (Dellinger et al.)
$\sim 1\times10^{-12}$	$<4\times10^{-15}$

- **ICP-SFMS is much simpler system than AMS.**
- **In particular, in the AMS one has to start with negative ions.**

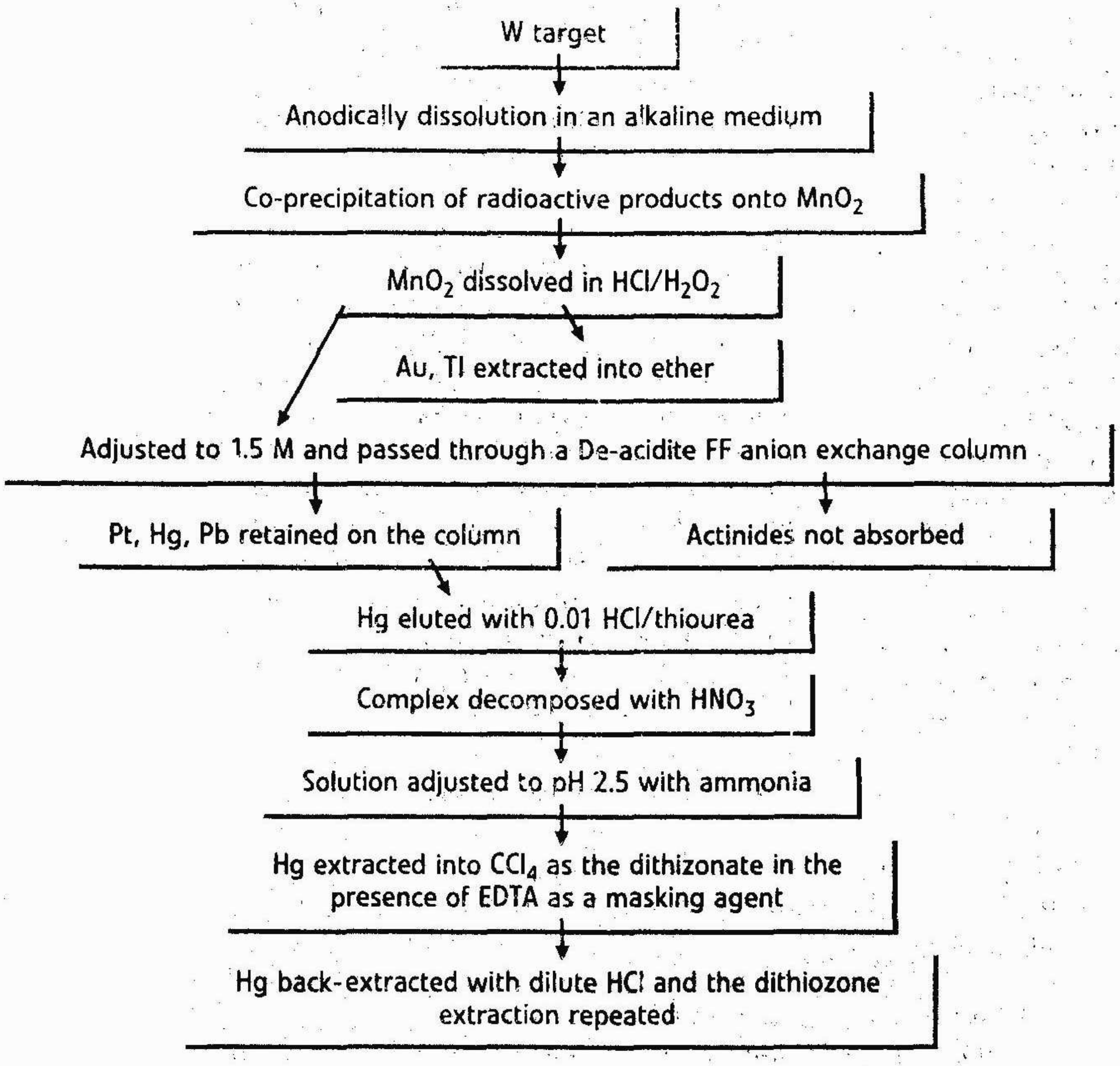
- We tried AMS and received a current of ThO^- of about 8 nA.
- Middleton (a Negetive Ion Cookbook):
- For ThO_2^- received 50 nA.

- Dellinger et al. (ThO_2^-) claim 350 nA.

- This seems unreasonable.

- Secondly: It is not certain that the current of (eka-Th)O_2^- will be the same as of ThO_2^-. For instance there is about a factor of 60 difference between:

- HfO^- (2-4 μA) and
- ThO_2^- (50 nA) (Middleton)



W target

Anodically dissolution in an alkaline medium

Co-precipitation of radioactive products onto MnO_2

MnO_2 dissolved in HCl/H_2O_2

Au, Tl extracted into ether

Adjusted to 1.5 M and passed through a De-acidite FF anion exchange column

Pt, Hg, Pb retained on the column

Actinides not absorbed

Hg eluted with 0.01 HCl/thiourea

Complex decomposed with HNO_3

Solution adjusted to pH 2.5 with ammonia

Hg extracted into CCl_4 as the dithizonate in the presence of EDTA as a masking agent

Hg back-extracted with dilute HCl and the dithiozone extraction repeated

Fig. 1. Block diagram of the chemical separation of Hg from the CERN W targets

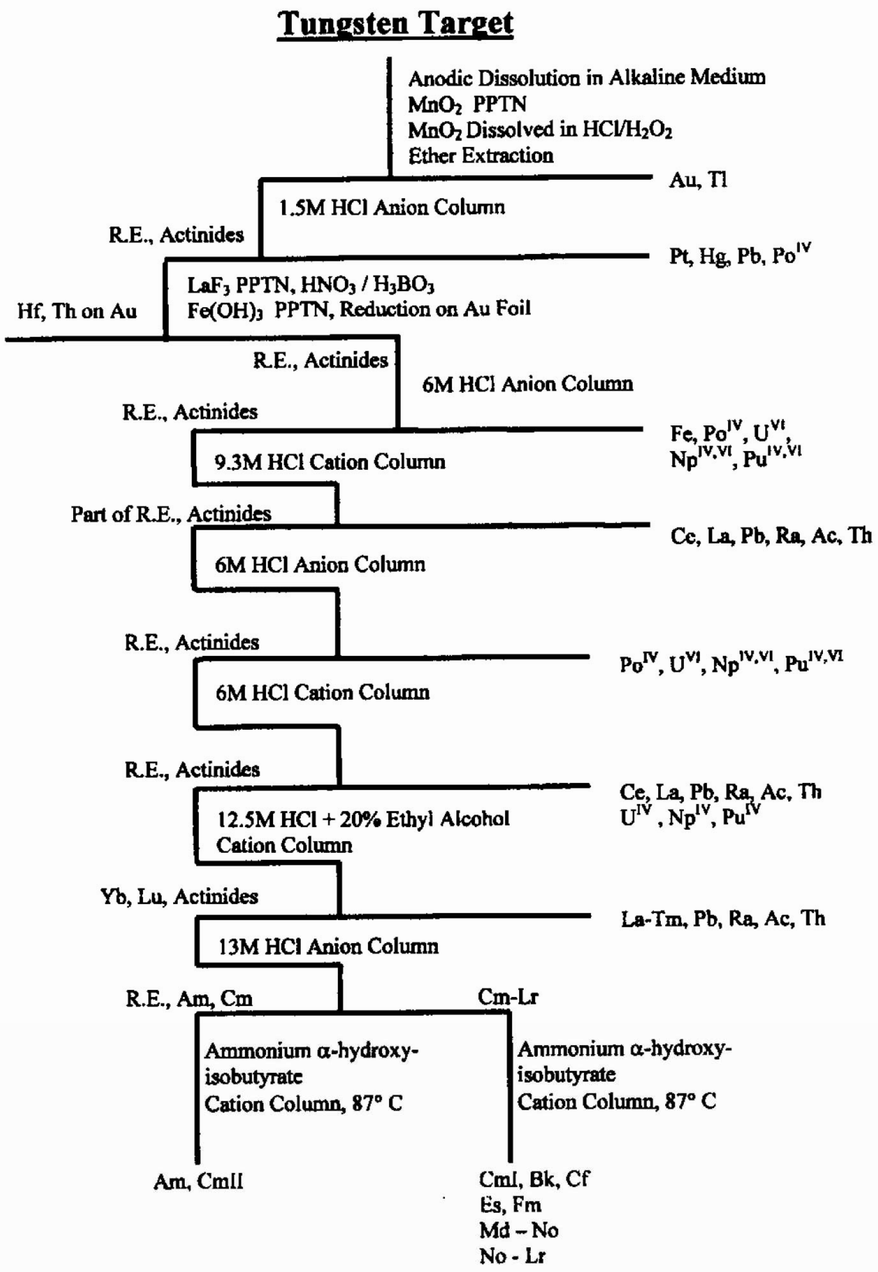
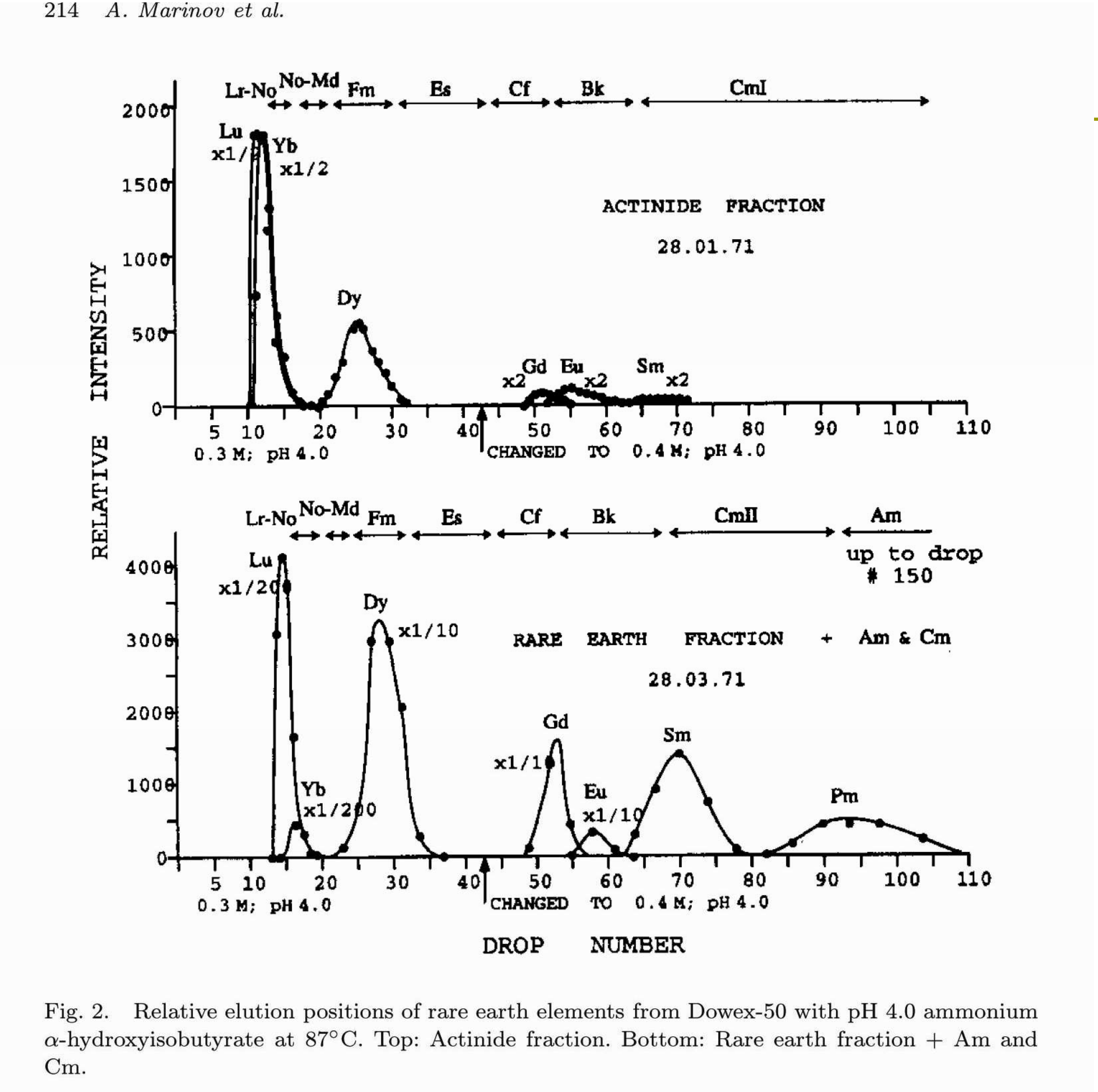


Fig. 1. Block diagram of the chemical separation of the actinide fraction from the W target.



214 *A. Marinov et al.*

Fig. 2. Relative elution positions of rare earth elements from Dowex-50 with pH 4.0 ammonium α-hydroxyisobutyrate at 87°C. Top: Actinide fraction. Bottom: Rare earth fraction + Am and Cm.