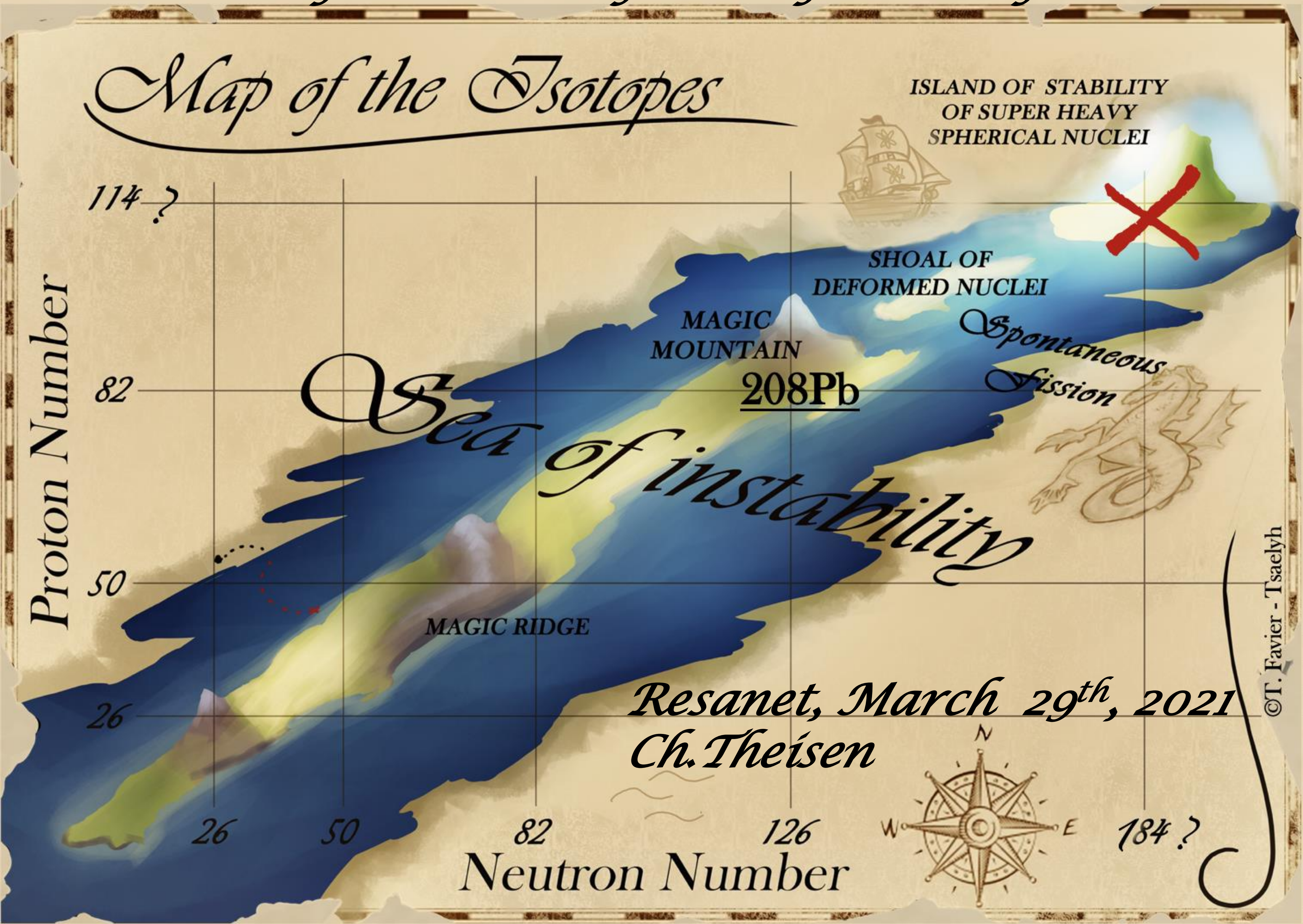
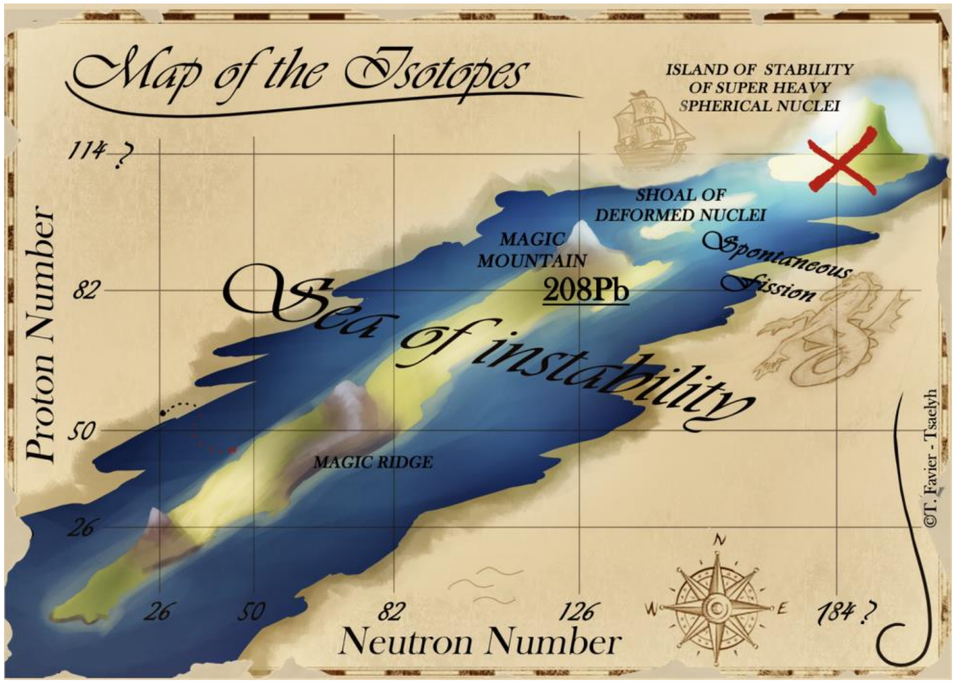
Selected topics on heavy and super-heavy nuclei



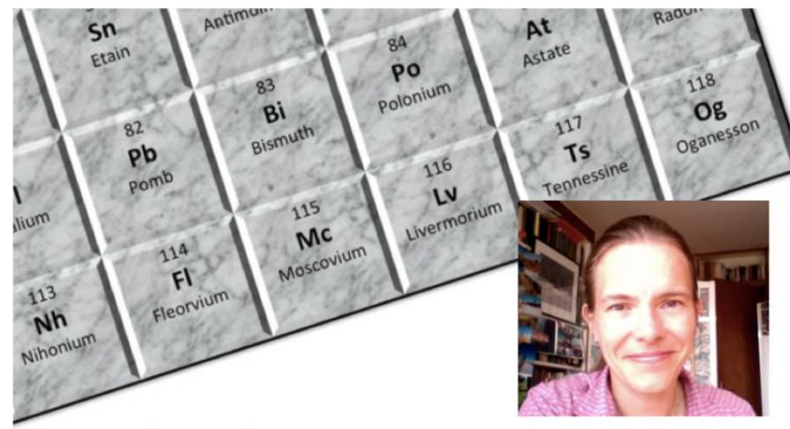
Resanet, March 29th, 2021

Ch.Theisen

A rich region for nuclear physics, atomic physics, chemistry.



Some aspects already covered by
Waely, Nov. 30th 2020.



Outlook

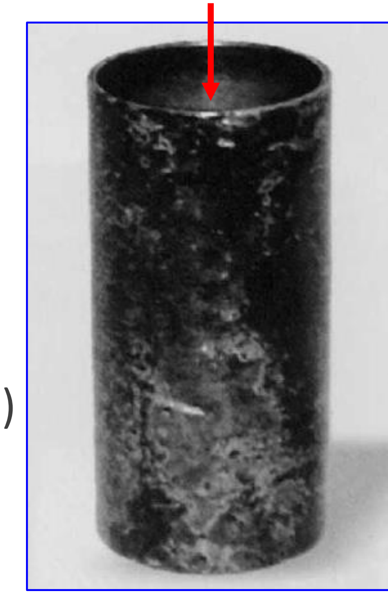
- Discovery of transuranium nuclei using neutrons
- r-process nucleosynthesis (neutrons again)
- Theoretical predictions
- Deformed shell gaps : experimental evidences and techniques
- Some trends for the future

Fermi and the neutron-induced reactions

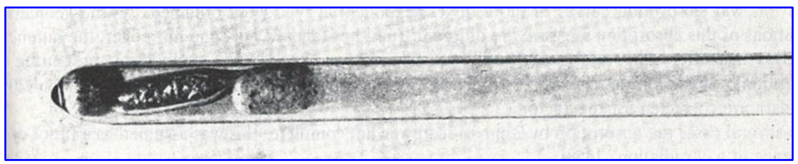
- Irradiation ${}^{A}_{Z}X + n \rightarrow {}^{A+1}_{Z}X \xrightarrow{\beta\ decay} {}^{A+1}_{Z+1}Y$
- (chemical separation)
- Detection of radioactivity (β-) Using a Geiger-Müller counter

→ lifetime and eventually β- energy using absorbers

Neutron source inside



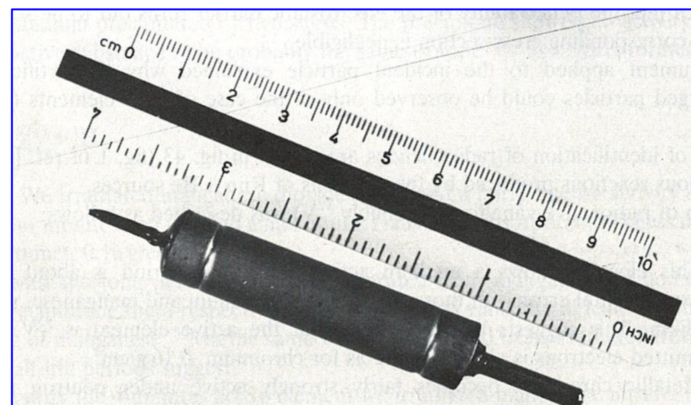
Neutron produced using Rn alpha source (800 mC) + Be → (1000 n/s/mC)



Glass tubes with Rn+Be

Systematic investigation in Roma of neutron-induced reaction along the periodic table from H to U.

About 30 new isotopes discovered !



Geiger-Müller counter

Ausonium and Hesperium

Possible Production of Elements of Atomic Number Higher than 92

By Prof. E. Fermi, Royal University of Rome

[Nature 133 (1934) 898]

In this way it appears that we have excluded the possibility that the 13 min.-activity is due to isotopes of uranium (92), palladium (91), thorium (90), actinium (89), radium (88), bismuth (83), lead (82). Its behaviour excludes also ekacæsium (87) and emanation (86).

This negative evidence about the identity of the 13 min.-activity from a large number of heavy elements suggests the possibility that the atomic number of the element may be greater than 92. If it were an element 93, it would be chemically homologous with manganese and rhenium. This hypothesis is supported to some extent also by the observed fact that the 13 min.-activity is carried down by a precipitate of rhenium sulphide insoluble in hydrochloric acid. However, as several elements are easily precipitated in this form, this evidence cannot be considered as very strong.

$$^{238}U + n \rightarrow {}^{239}U \xrightarrow{\beta^-} {}^{239}93 \xrightarrow{\beta^-} {}^{239}94$$

Elements named Ausonium and Hesperium by Franco Rasetti

Element 93 confirmed in Berlin… and much more !

1935 : neutron induced reaction repeated by chemists Hahn, Meitner and Strassmann at Kaiser Wilhelm-Institut far Chemie, Berlin (and in other places)
Compared to Fermi group, improved chemical separation, more lifetime component identified and better lifetime measurement.

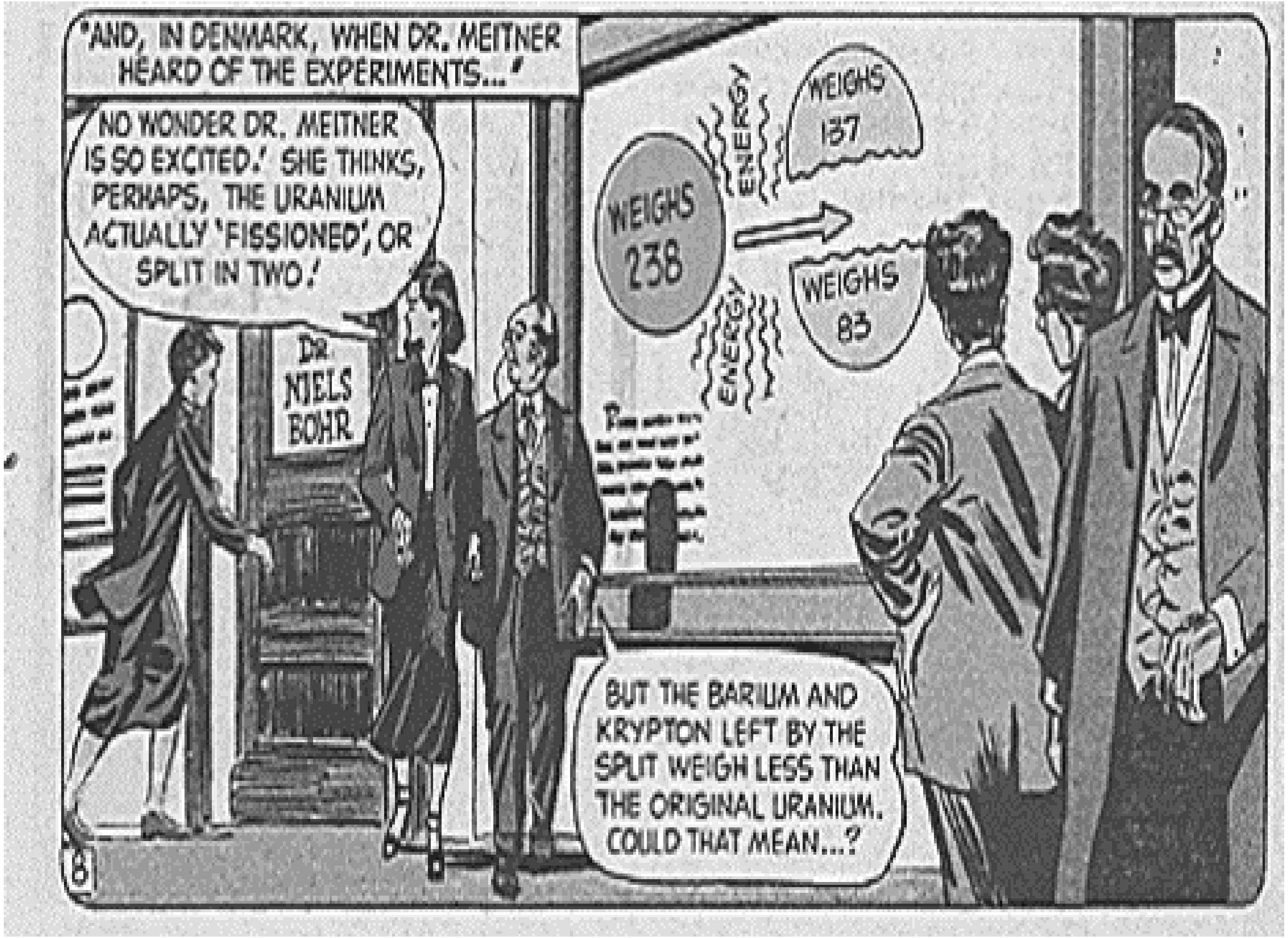
$$1.\quad U + n \longrightarrow {}^{10''}_{92}U \xrightarrow{\beta} {}^{2,2'}_{93}\text{Eka Re} \xrightarrow{\beta} {}^{59'}_{94}\text{Eka Os} \xrightarrow{\beta} {}^{66\,h}_{95}\text{Eka Ir} \xrightarrow{\beta} {}^{2,5\,h}_{96}\text{Eka Pt} \xrightarrow{\beta} {}_{97}\text{Eka Au ?}$$

$$2.\quad U + n \longrightarrow {}^{40''}_{92}U \xrightarrow{\beta} {}^{16'}_{93}\text{Eka Re} \xrightarrow{\beta} {}^{5,7\,h}_{94}\text{Eka Os} \xrightarrow{\beta} {}_{95}\text{Eka Ir ?}$$

$$3.\quad U + n \longrightarrow {}^{23'}_{92}U \xrightarrow{\beta} {}_{93}\text{Eka Re ?}$$

Meitner, Hahn, Strassmann. ZP 106 (1937) 249

P. Abelson using the Berkeley Cyclotron as a neutron source (large flux) → no conclusive results, no alpha decay found.


"AND, IN DENMARK, WHEN DR. MEITNER HEARD OF THE EXPERIMENTS..."
NO WONDER DR. MEITNER IS SO EXCITED! SHE THINKS, PERHAPS, THE URANIUM ACTUALLY 'FISSIONED', OR SPLIT IN TWO!
DR. NIELS BOHR
WEIGHS 238
ENERGY
WEIGHS 137
WEIGHS 83
ENERGY
BUT THE BARIUM AND KRYPTON LEFT BY THE SPLIT WEIGH LESS THAN THE ORIGINAL URANIUM. COULD THAT MEAN...?
8

Neptunium (93) and Plutonium (94)

- 1940 : McMillan and Alberson PR 57 (1940) 1185

Neutron from the reaction d(8MeV) + ^{8}Be.

n + ^{238}U → 2.3 days activity corresponds to ^{239}93

but not a rare-earth, not homolog to Re. Chemical properties similar to U !

Second « rare-earth » group starting from U ?

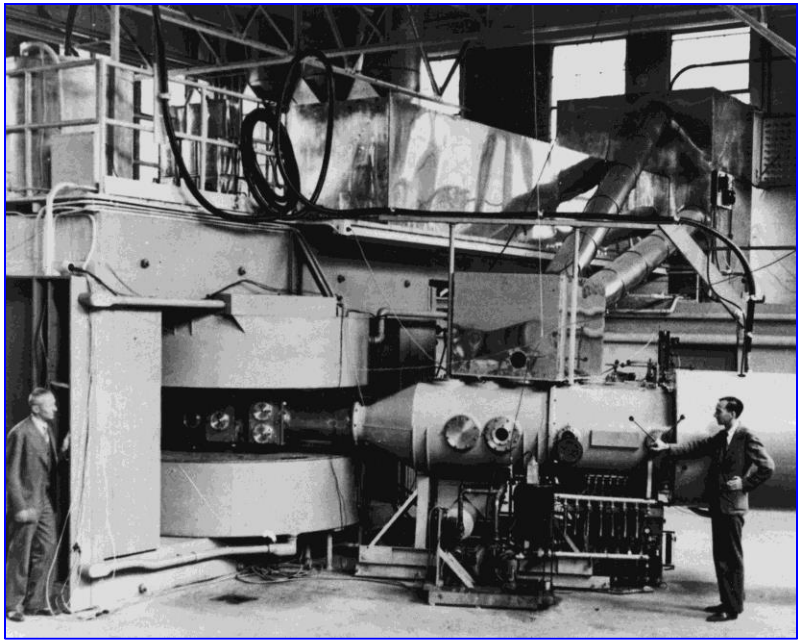
- 1940, 1941 : G. Seaborg's team

d(16 MeV)+^{238}U → ^{238}93 → ^{238}94 → ^{234}U

d(16 MeV)+^{238}U → ^{239}93 → ^{239}94

Voluntary restrictions to publish papers on fission and transuranium elements: potential application for energy production.

Berkeley, 60 inch cyclotron in 1939



Plutonium (94)

Physical Review 69 (1946) 366

Radioactive Element 94 from Deuterons on Uranium

G. T. Seaborg, E. M. McMillan, J. W. Kennedy, and A. C. Wahl
Department of Chemistry, Radiation Laboratory, Department of Physics, University of California, Berkeley, California
January 28, 1941*

WE are writing to report some results obtained in the bombardment of uranium with deuterons in the 60-inch cyclotron.

d(16 MeV)+^{238}U

* This letter was received for publication on the date indicated but was voluntarily withheld from publication until the end of the war.

Physical Review 69 (1946) 367

Radioactive Element 94 from Deuterons on Uranium

G. T. Seaborg, A. C. Wahl, and J. W. Kennedy
Department of Chemistry, Radiation Laboratory, Department of Physics University of California, Berkeley, California
March 7, 1941*

WE should like to report a few more results which we have found regarding the element 94 alpha-radioactivity formed in the 16-Mev deuteron bombardment of uranium. We sent a first report[1] of this work in a

* This letter was received for publication on the date indicated but was voluntarily withheld from publication until the end of the war.
[1] G. T. Seaborg, E. M. McMillan, J. W. Kennedy and A. C. Wahl, Phys. Rev. **69**, 366 (1946).

PHYSICAL REVIEW VOLUME 70, NUMBERS 7 AND 8 OCTOBER 1 AND 15, 1946

Properties of 94(239)

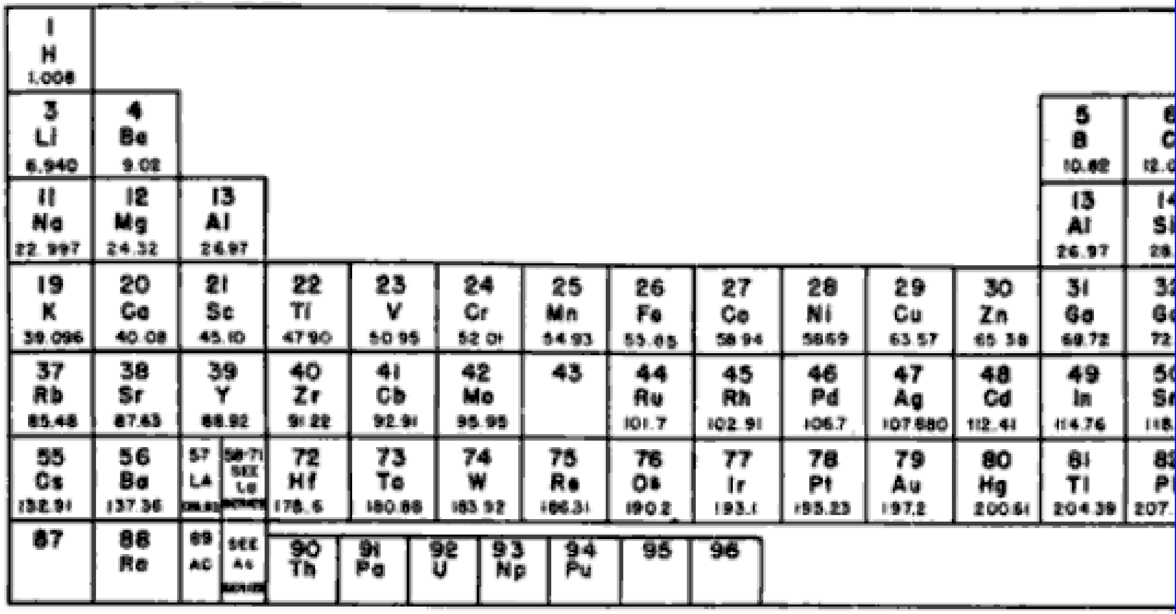
J. W. Kennedy, G. T. Seaborg, E. Segrè, and A. C. Wahl
Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California
(Received May 29, 1941)*

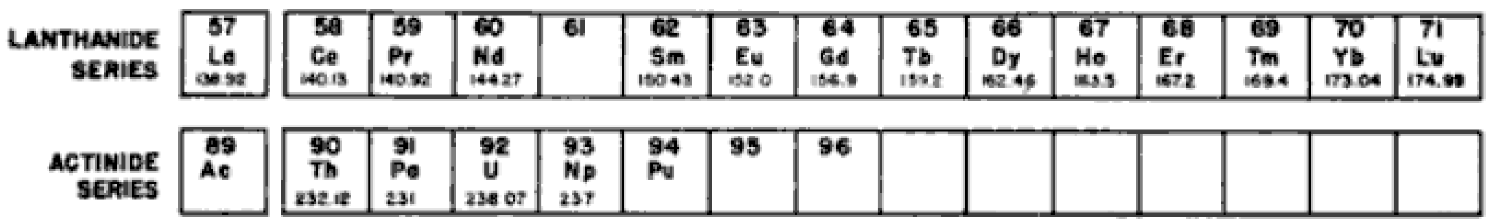
* This letter was received for publication on the date indicated but was voluntarily withheld from publication until the end of the war. The original text has been somewhat changed, by omissions, in order to conform to present declassification standards.

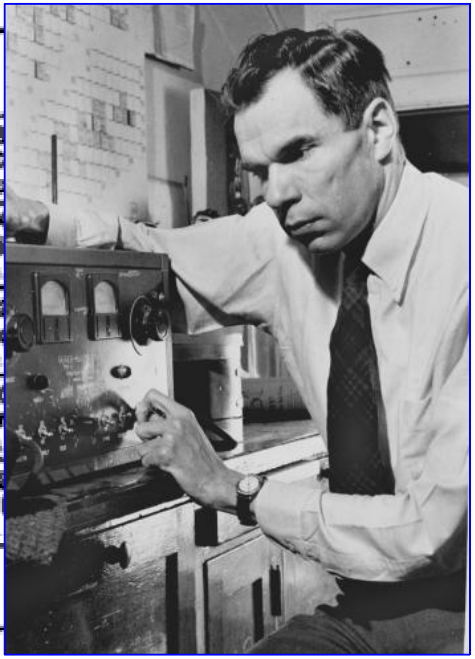
Chemical identification : what was wrong ?

I A	II A	III B	IV B	V B	VI B	VII B	VIII			I B	II B	III A	IV A	V A	VI A	VII A	O
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
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
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	(43)	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57-71 Ln	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)	86 Rn
(87)	88 Ra	89 Ac	90 Th	91 Pa	92 U	(93)	(94)	(95)	(96)	(97)	(98)	(99)					
		57 La	58 Ce	59 Pr	60 Nd	(61)	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	

Periodic table ~1930 : Z=93 same column as Mn, Tc, Re







Actinide concept : Glen Seaborg ~ 1944

Table from G. Seaborg, Science 104 (1946) 379

Z=96 Cm : Seaborg 1944 (60'' cyclotron)

$^{239}Pu(\alpha,n)^{242}Cm \xrightarrow[150d]{\alpha} {}^{238}Pu$

AECD-2182 report, [Chem. Eng. News 23 (1945) 2190]

Z=95 Am : Seaborg 1944 (60'' cyclotron)

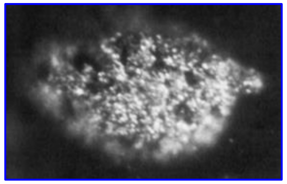
$^{238}U(\alpha,n)^{241}Pu \xrightarrow[\sim 10\,y]{\beta^-} {}^{241}Am$

AECD-2185 report, [Chem. Eng. News 23 (1945) 2190]

Z=97 Bk : Thompson 1949 (60'' cyclotron)

$^{241}Am(\alpha,2n)^{243}Bk \xrightarrow[4,8h]{EC} {}^{234}Cm$

UCRL-669 report, [PR 77 (1950) 838]

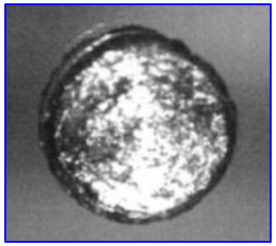


Z=98 Cf: Thompson 1950 (60'' cyclotron)

$^{242}Cm(\alpha,n)^{245}Cf \xrightarrow[44m]{\alpha} {}^{241}Cm$

UCRL-790 report [PR 78 (1950) 298], [102 (1956) 747]

(mass assignment was wrong in the 1950 paper)



First thermonuclear explosion
« Mike » November 1rst 1952,
Eniwetok Atoll
~10 Mtons

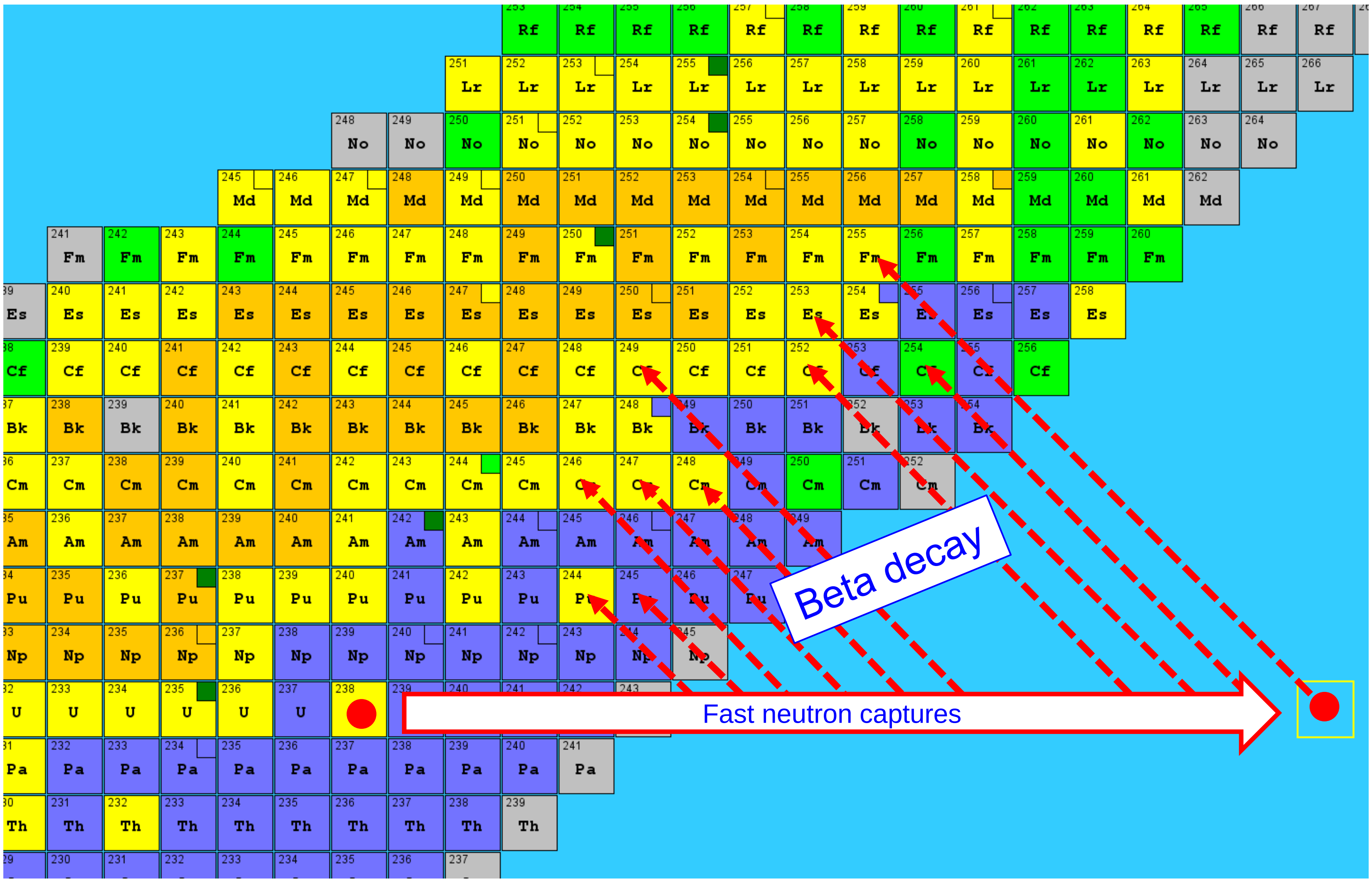
Explosion debris
collected by a plane transferred
to Los Alamos.

Results obviously classified.

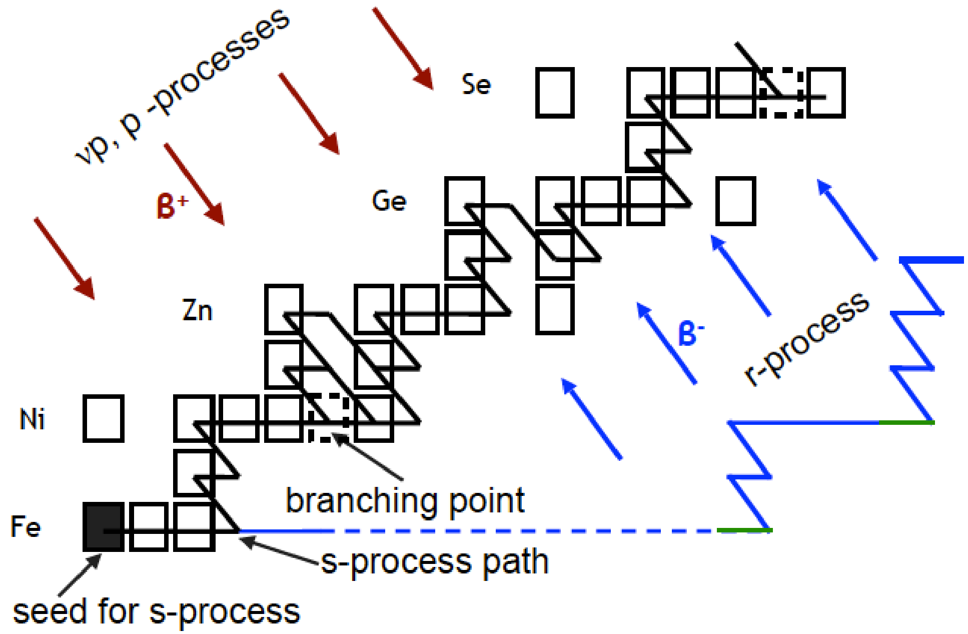
Some new alpha-rays.

Albert Ghiorso, Berkekey obtains some samples.
→ Discovery ^{253}Es and ^{255}Fm

In total 15 new isotopes discovered : ^{244,245,246}Pu, ^{246}Am, ^{246,247,248}Cm, ^{249}Bk, ^{249,252,253,254}Cf, ^{253,255}Es, ^{255}Fm


Beta decay
Fast neutron captures

Production of (super-)heavy nuclei in the universe; r-process
1957 : Burbidge, Burbidge, Fowler, and Hoyle (Reviews of Modern Physics)
Cameron (Publications of the Astronomical Society of the Pacific)



Nucleosysnthesis starting from the Fe region.
Need huge neutron flux and explosive process,
Several possible sites; core-collapse Supernovae was first considered.

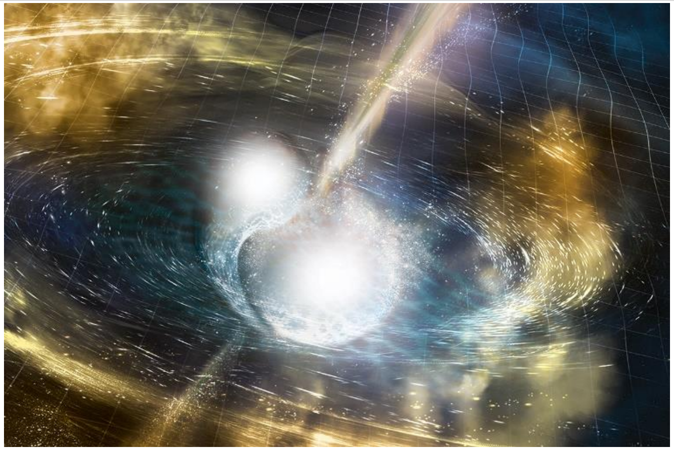
Neutron star / black hole merger

R-process site suggested in 1974 by Lattimer and Schramm "Black-Hole-Neutron-Star Collisions", Astrophys. J. 192 (1974) L145
Lattimer, Mackie, Ravenhall and Schramm Astrophys. J. 213 (1977) 225

NS-NS merger :
Symbalisty and Schramm Astrophys. J. Lett. 22 (1982) 143
Meyer Astrophys. J. 343 (1989) 254

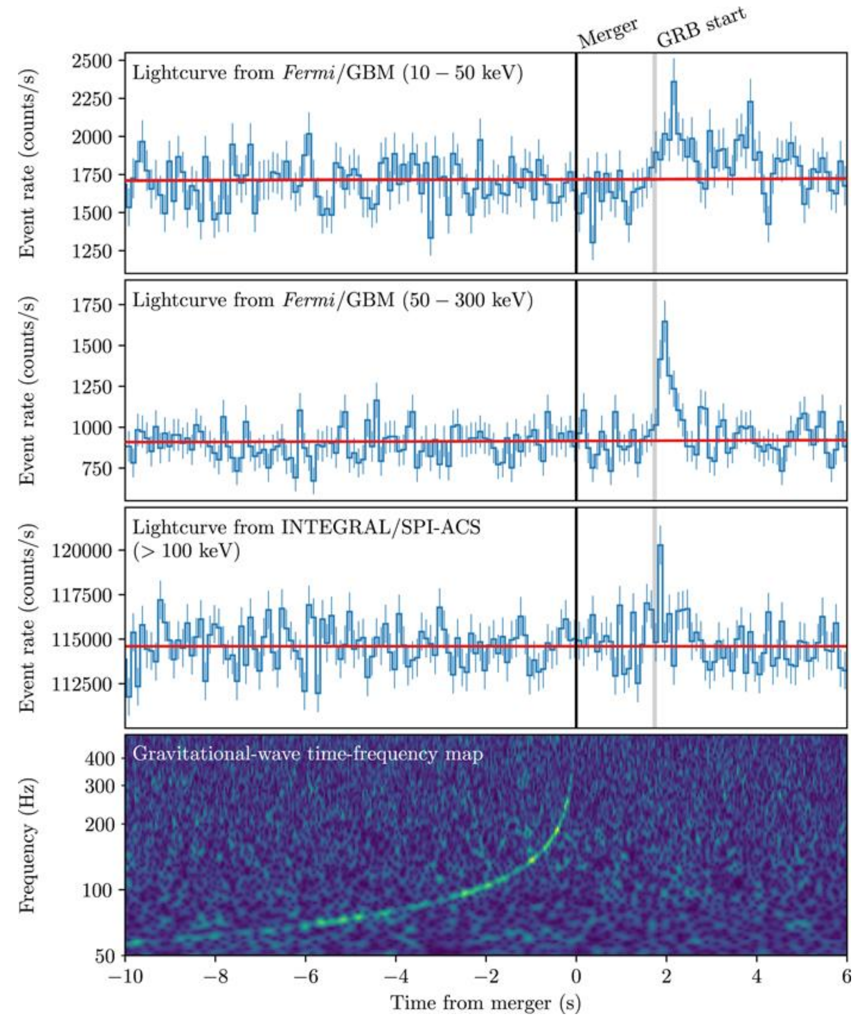
Popular candidate for r-process since ~ 2000, several predictions made.
See e.g. S.Goriely, G.Martínez-Pinedo NPA 944 (2015) 158

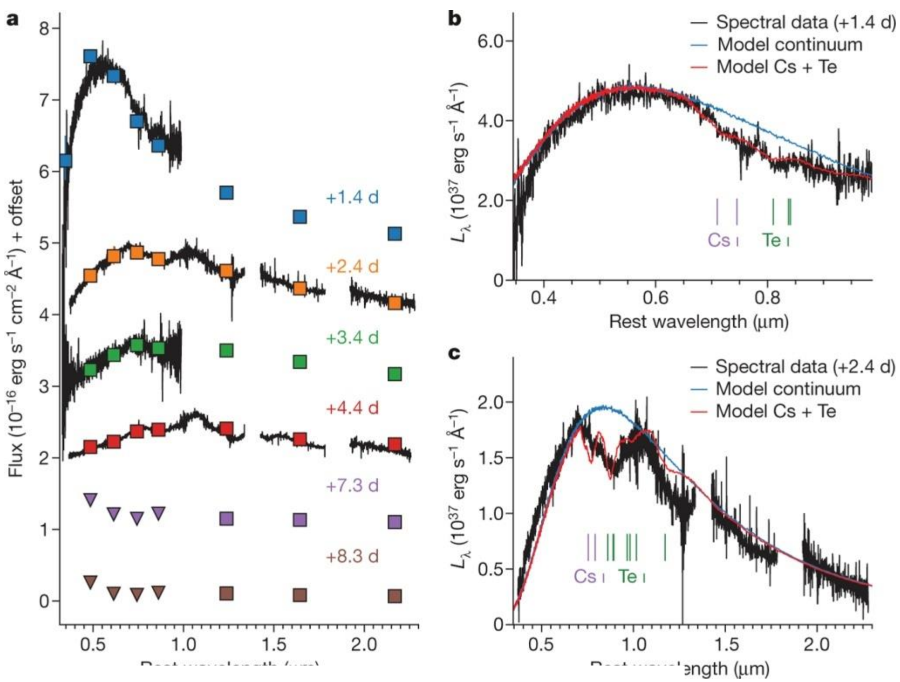
Neutron star merger 2017 08 17 GW 170817



Multi-messenger Observations

[Abbott et al. ApJ 848 (2017) L12]

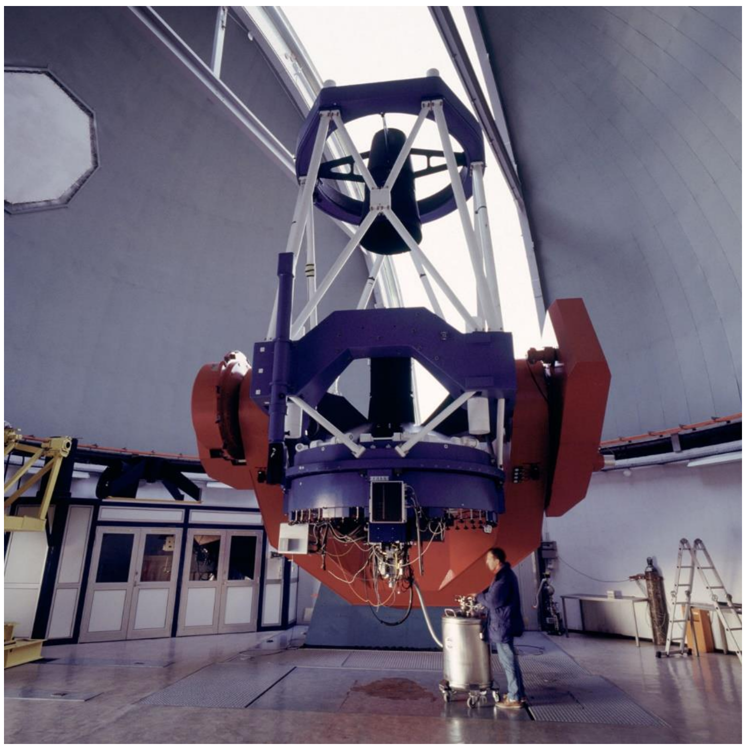




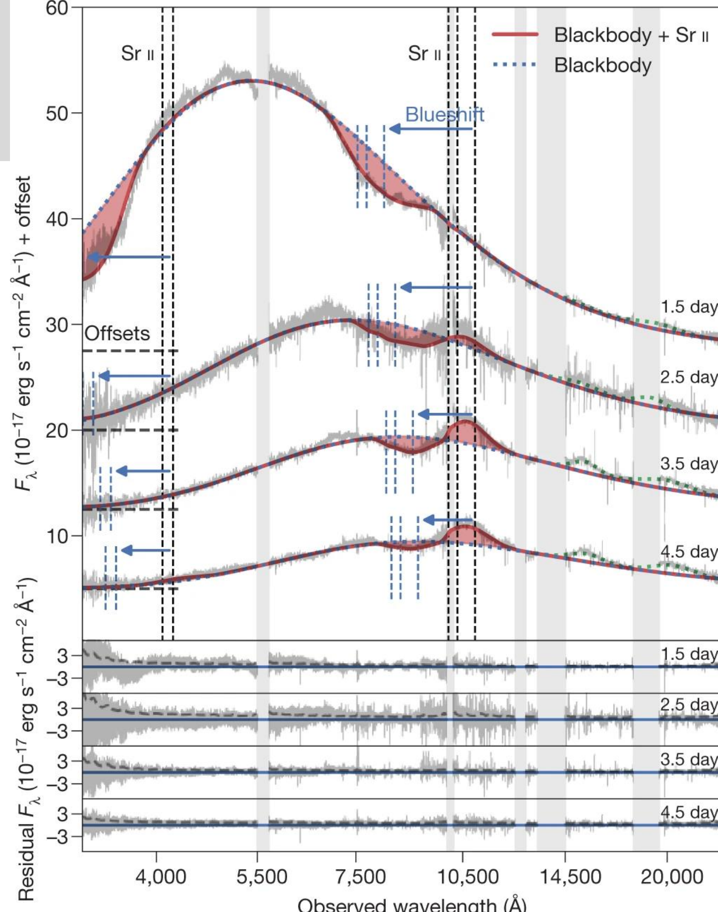
[Smartt et al. Nature 551 (2017) 75]

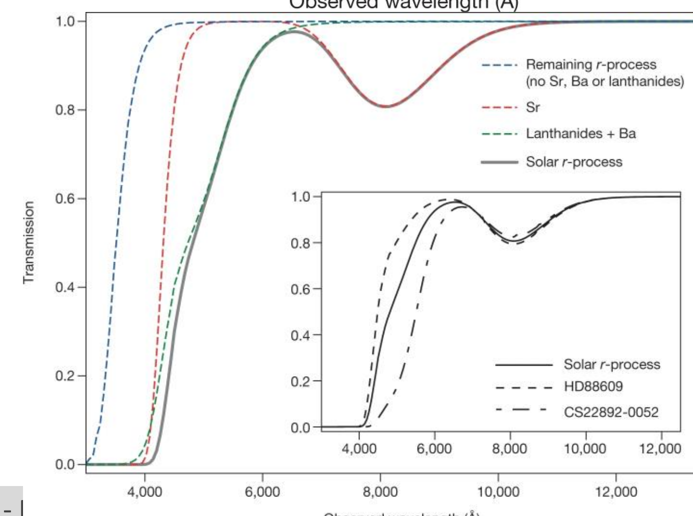
Actually not Cs + Te, but Sr. Data re-interpreted in [Watson et al., Nature 574 (2019) 497](#)

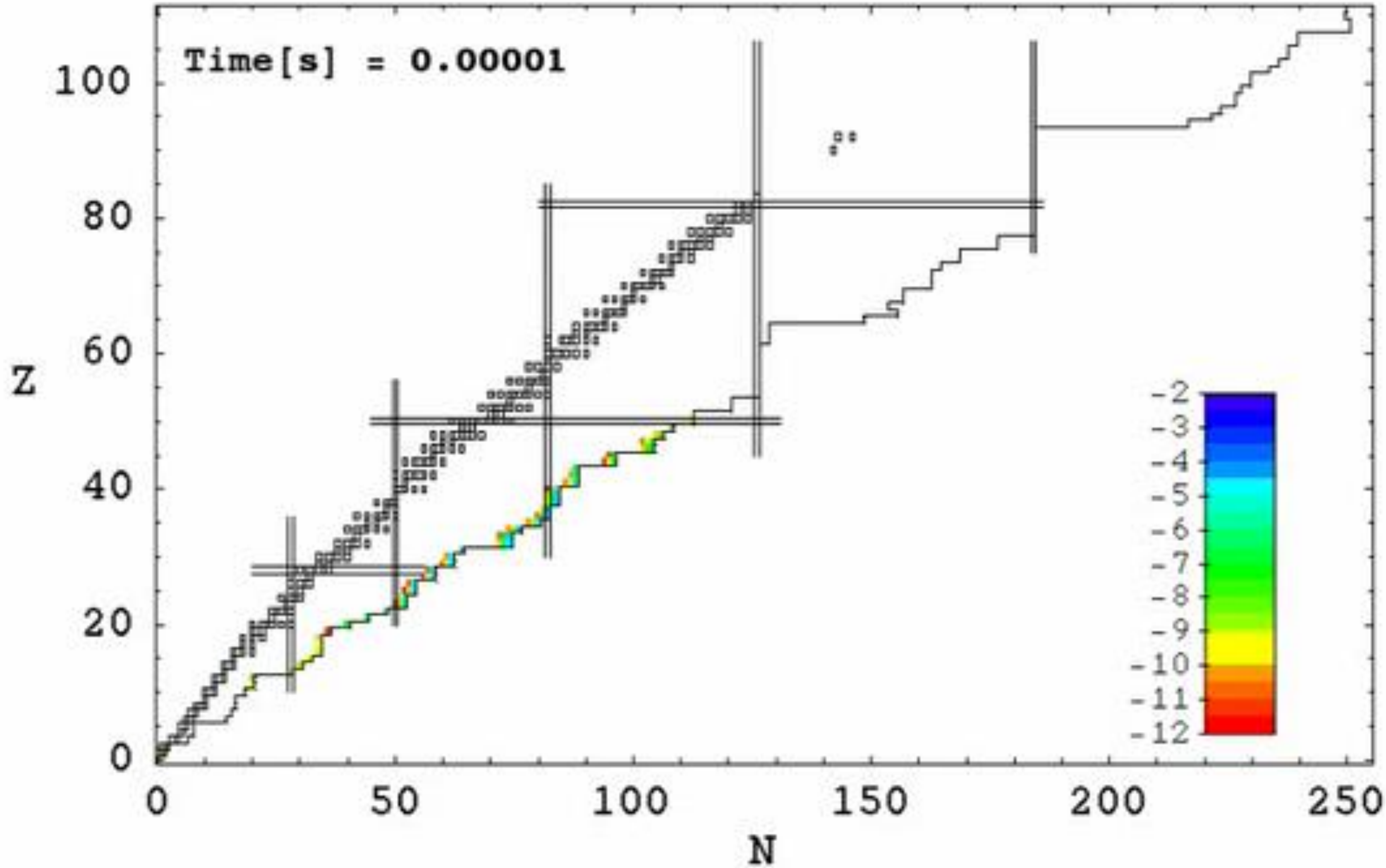
- Model includes plasma opacity
- Only x-ray line can escape because absorption
- Main component = black body



Gamma-Ray Burst Optical/Near-infrared Detector (GROND), La Silla Observatory

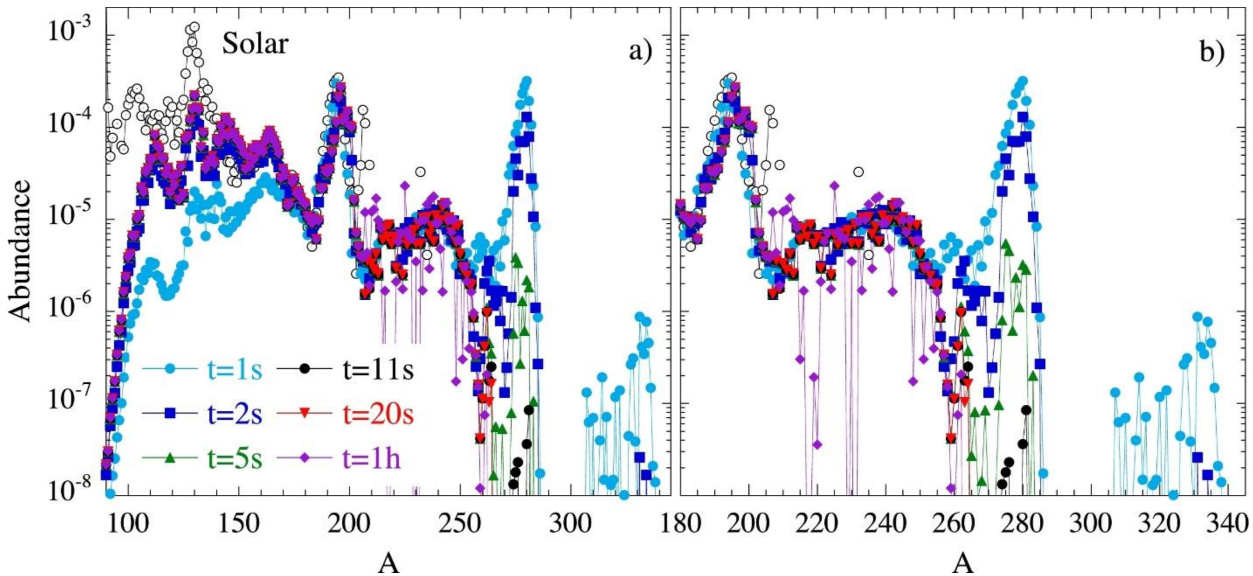




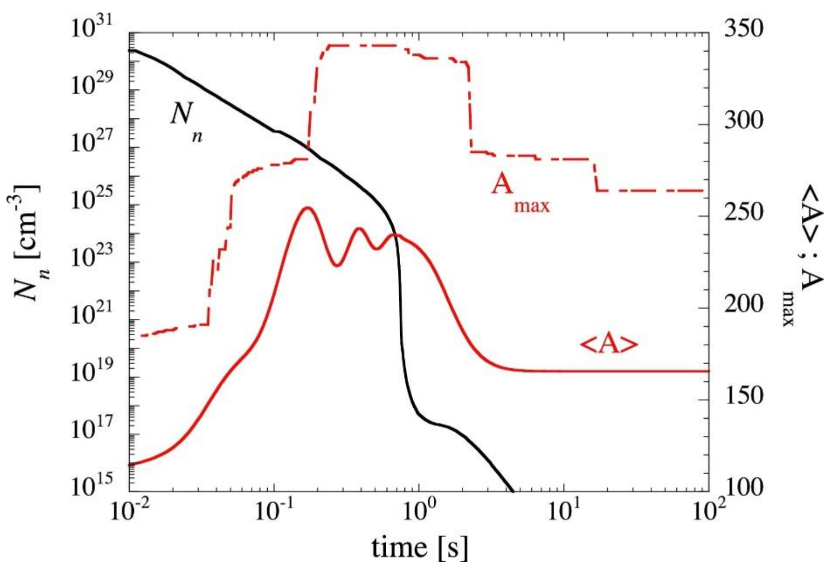


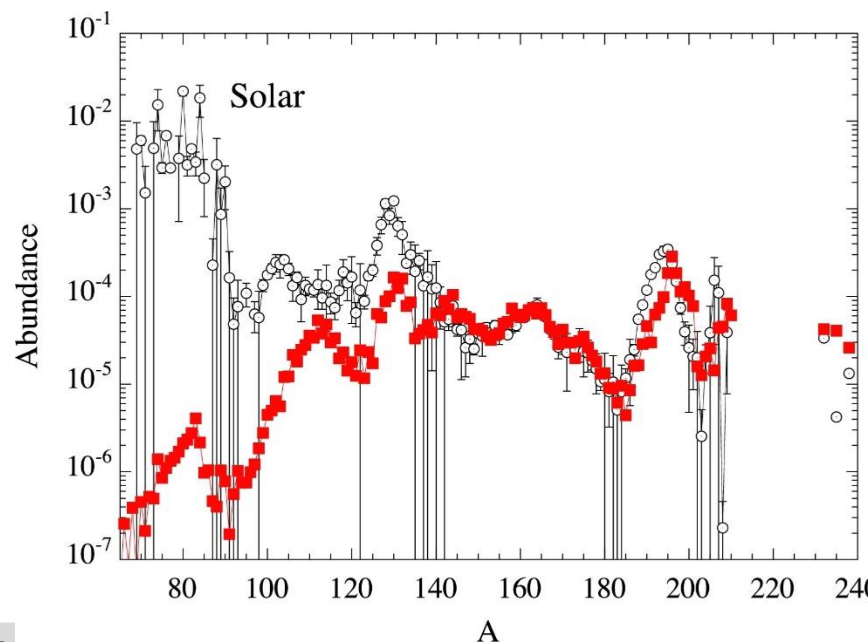
Stephane Goriely, Andreas Bauswein, Hans-Thomas Janka
https://www.youtube.com/watch?v=zouvhsFvKiM
See also S.Goriely, G.Martínez-Pinedo NPA 944 (2015) 158

Solar versus neutron-start merger abundances

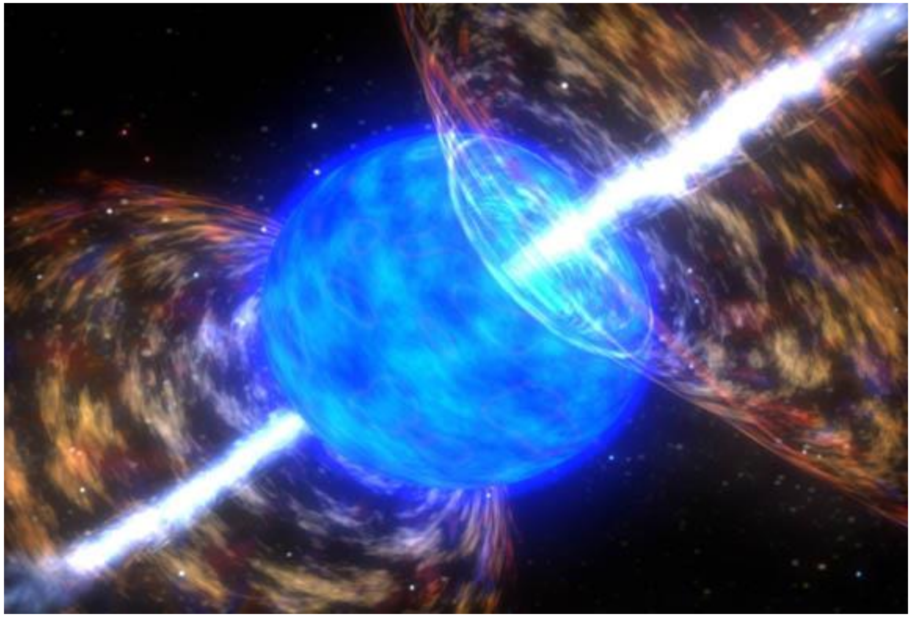


NPA 944 (2015) 158





A fresh candidate : the collapsar



Collapsar = collapse of massive rotating star (20-30 Msun) → black hole

Siegel, Barnes and Metzger [Nature 569 (2019) 241]

Less frequent than NS-merger, but r-process yield larger (x 30).
Collapsars ~ 80% heavy elements in the universe ?

Current status and ingredients of r-process nucleosynthesis

r-process site(s) not firmly identified.
r-process abundances may be due to several processes.

Some ingredients of r-process nucleosynthesis

- n-capture cross-section
- β-decay rates
- (n,γ), (γ,n) rates
- fission rates (neutron-induced, β-delayed fission) and fission fragment distribution (fission cycling)
- Neutrino physics -> (influences n,p ratio, electron fraction, etc.)
- Thermodynamics

Quantities relevant for actinide & transactinide
→ Masses, fission barriers, fission fragment distributions

Nuclear properties can not be measured in the r-process path region (fission barriers and masses difficult to measure anyway)

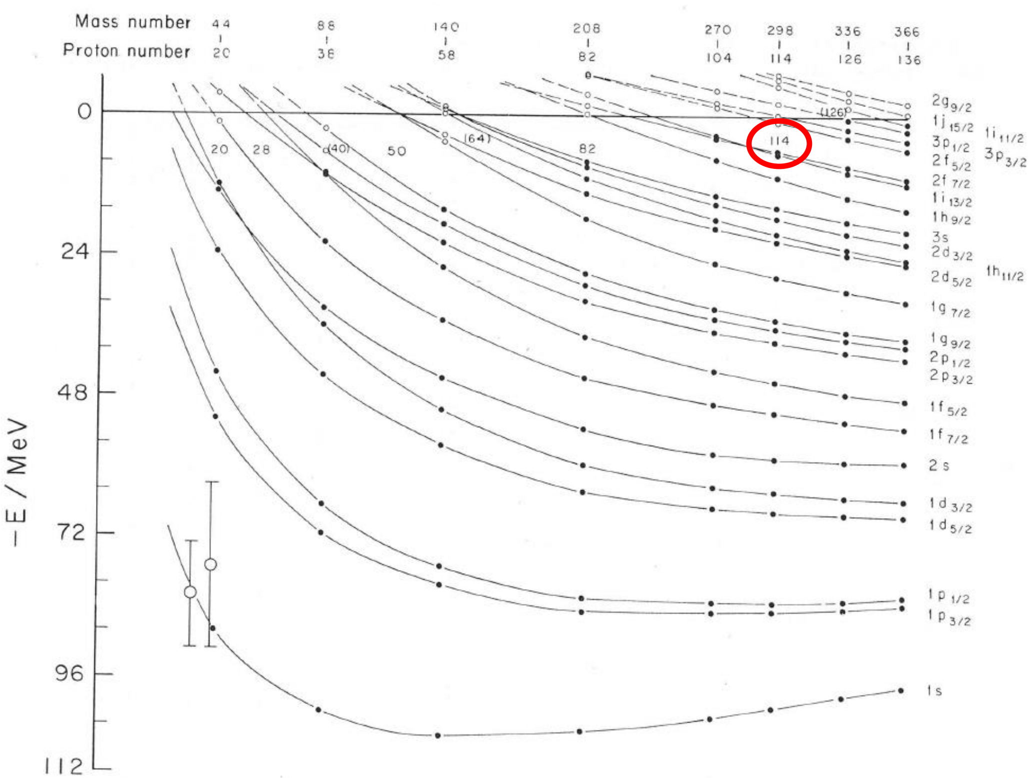
→ Theory

SHN theoretical predictions

- 1949 : The spherical shell model (Mayer, Haxel, Jensen and Suess).

… 28, 50, 82, 126, …

- 1957 : G. Scharff-Goldhaber "There may be, for instance, another region of relative stability at the doubly magic nucleus ${}_{126}X^{310}$"
- 1966 : Lysekil symposium "Why and how should we investigate nuclei far from the stability line?"



H. Meldner, Ark. Fiz. 36 (1966) 593, shell model

→ Z=114, N=184

Confirmed by C.Y. Wong PL 21 (1966) 688 (shell model)

A. Sobiczewski et al. PL 22 (1966) 500 (Woods-Saxon)

…

= calculations using phenomenological potentials

Why Z=114 and not Z = 126 ?

HFB calculations with Skyrme forces :
Vautherin and Brink PLB 1970, Vautherin, Veneroni and Brink PLB 1970, Davies and McCarthy PRC 1971, Köhler NPA 1971, Bassichis and Kerman PRC 1970 and PRC 1972, Rouben et al. PLB 1972 and PLB 1977, Saunier and Rouben PRC 1972, Beiner et al. Phys. Scr. 1974, Brack and Quentin Phys. Scr. 1974 (Z=114 = Gammelgormium) , Cusson et al. PRC 1976, Vallières and Sprung PLB 1977, Kolb ZPA 1977, Tondeur ZPA 1978 and 1980

Spherical calculation only, for few nuclei, some simplifications, mostly no pairing

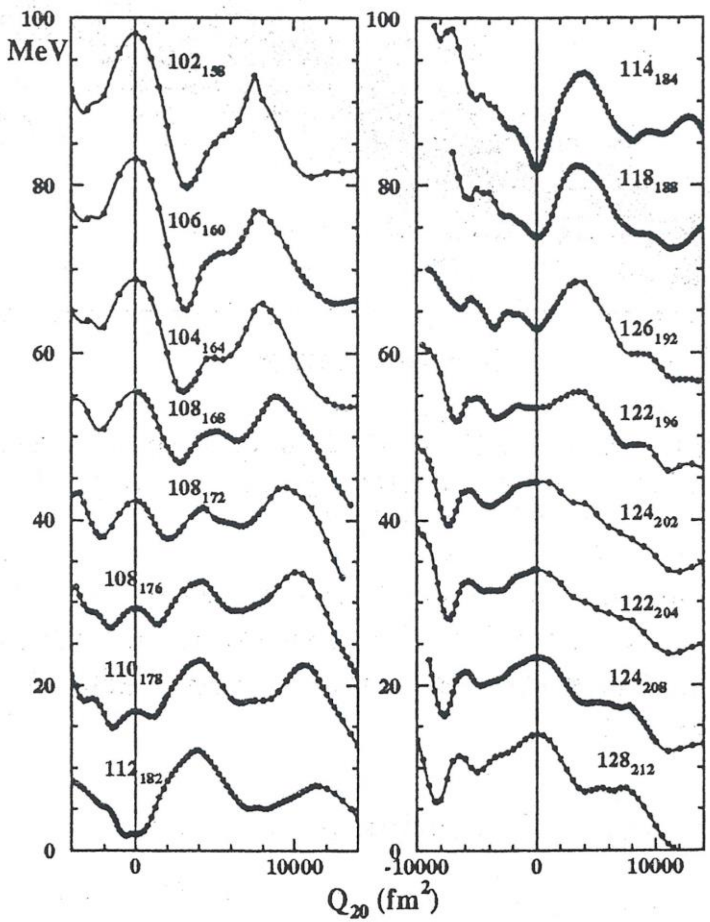
RMF calculations Gambhir et al. Ann. Phys. 1990, Boersma PRC 1993

→ Z= 114 not refuted; Z = 120, 126 or 138 also suggested

Warning = magic or higher stability ≠ stable against β decay. In many cases confusion shell gaps vs stability.

HFB calculations with Gogny force.

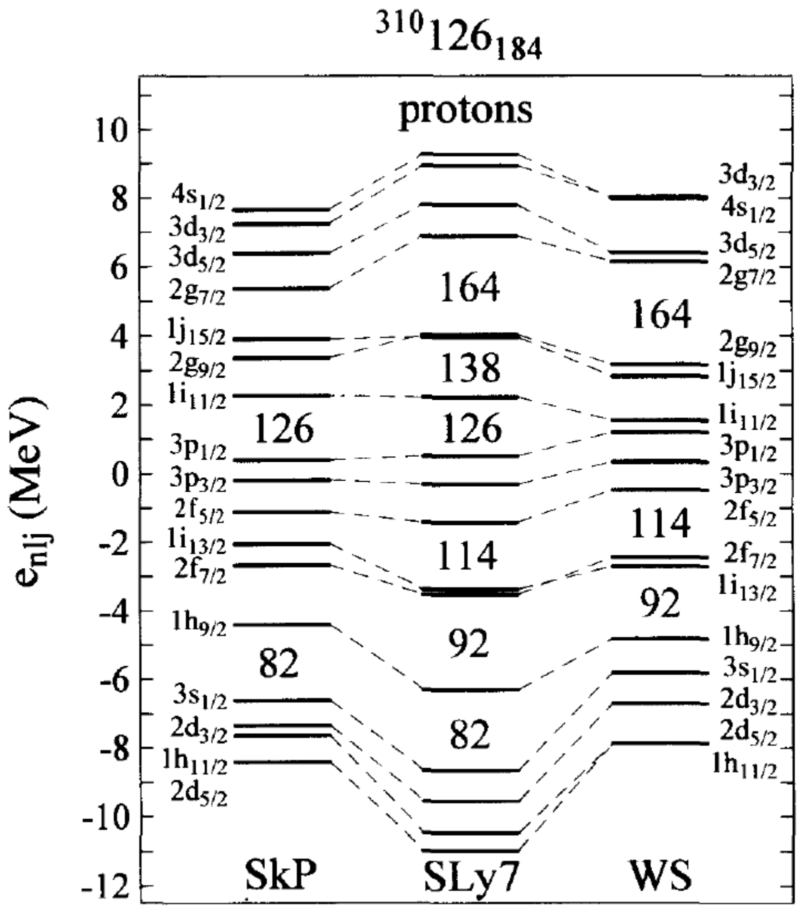
Berger, Bitaud, Dechargé, Girod, Péru, Proc. Int. Workshop XXIV on Gross Properties of Nuclei and Nuclear Excitation (1996)

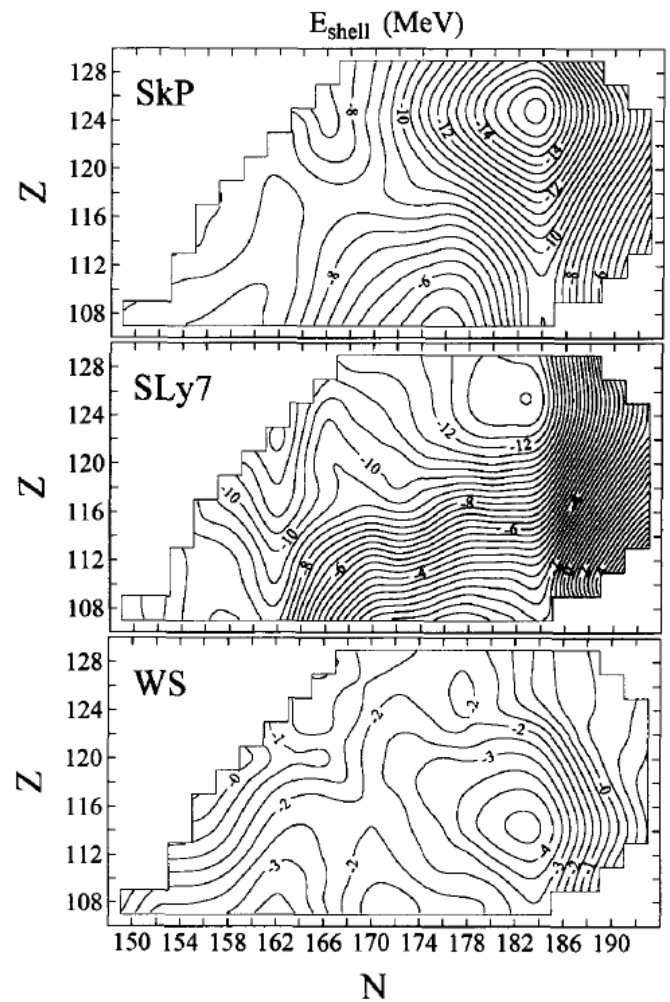


"Z=114 appears here merely as a subshell with 1.3-1.5 MeV gap, smaller than the one observed at Z=120. Actually, significant proton pairing correlations are found in Z=114 nuclei, in particular ^{298}114 – which indicates that 114 is not a magic number."

Systematic calculations using self-consistent models (spherical nuclei)

WS + Skyrme forces by Ćwiok, Dobaczewski, Heenen, Magierski and Nazarewicz. [NPA 611 (1996) 211]



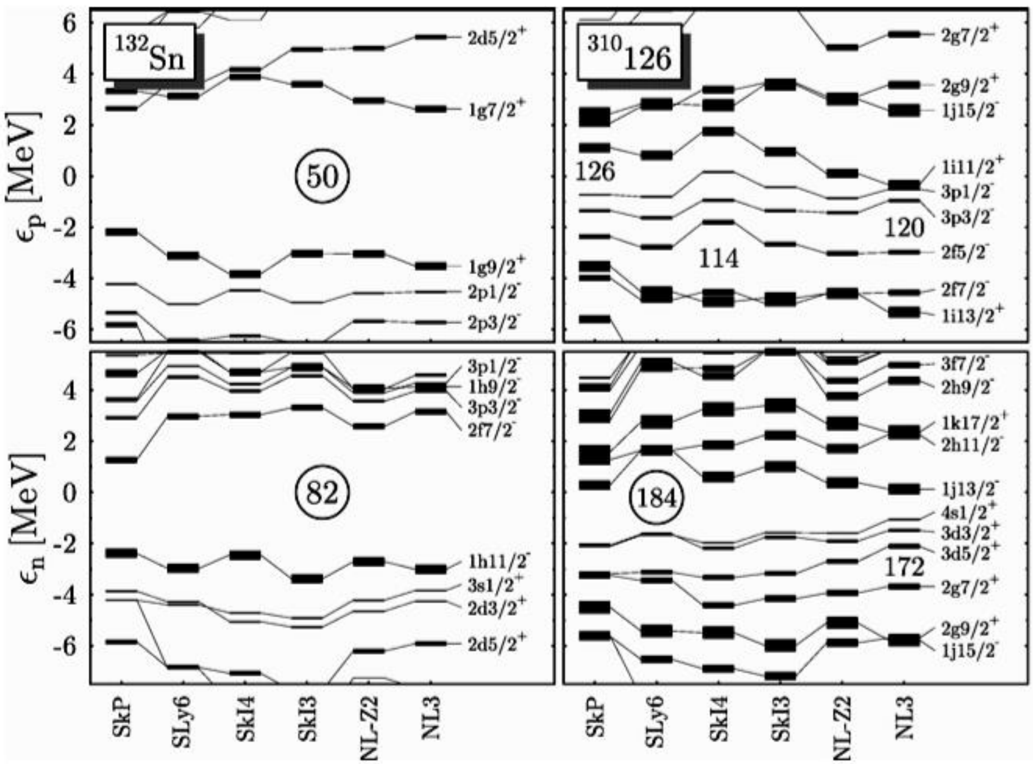


Systematic calculations using self-consistent models (spherical nuclei), Skyrme and RMF

Rutz, Bender, Bürvenich, Schilling, Reinhard, Maruhn and Greiner, PRC 56 (1997) 238,

Bender, Rutz, Reinhard, Maruhn and Greiner PRC 60 (1999) 034304

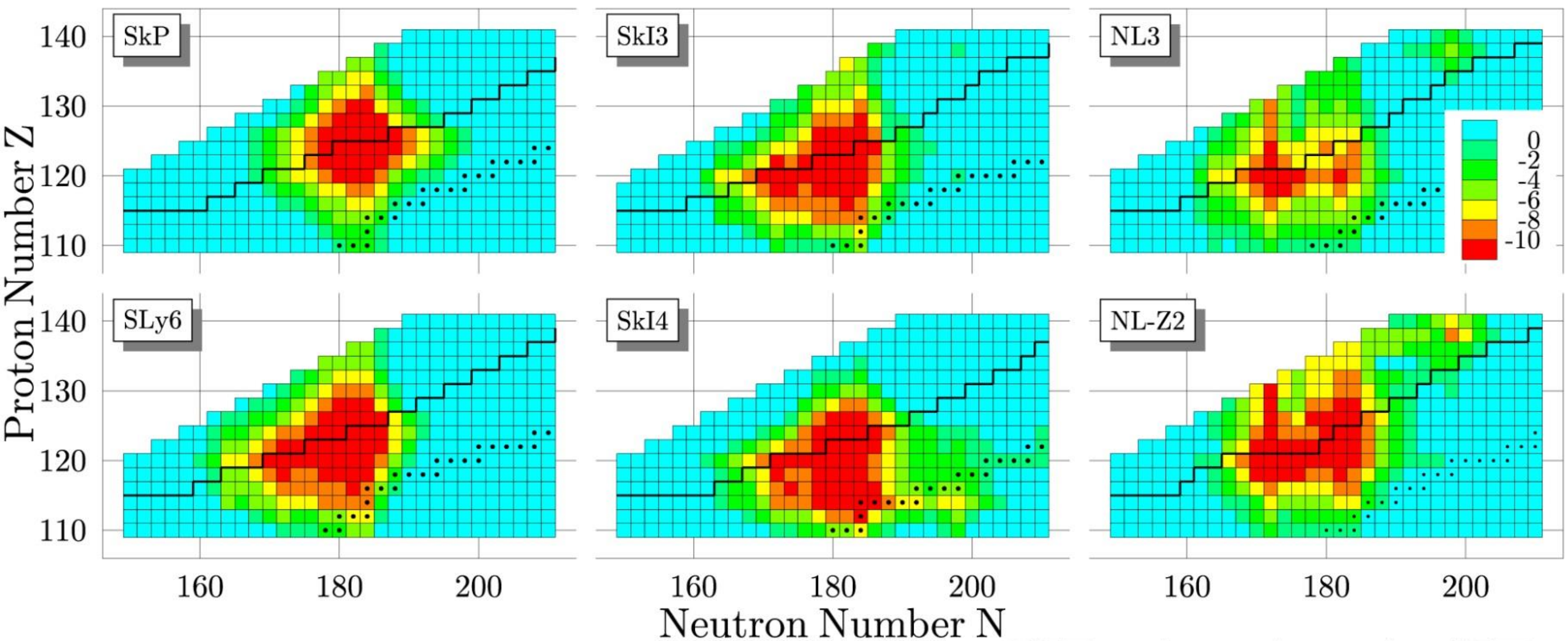
M. Bender et al., PLB 515 (2001) 42, *Shell stabilization of super- and hyperheavy nuclei without magic gaps*



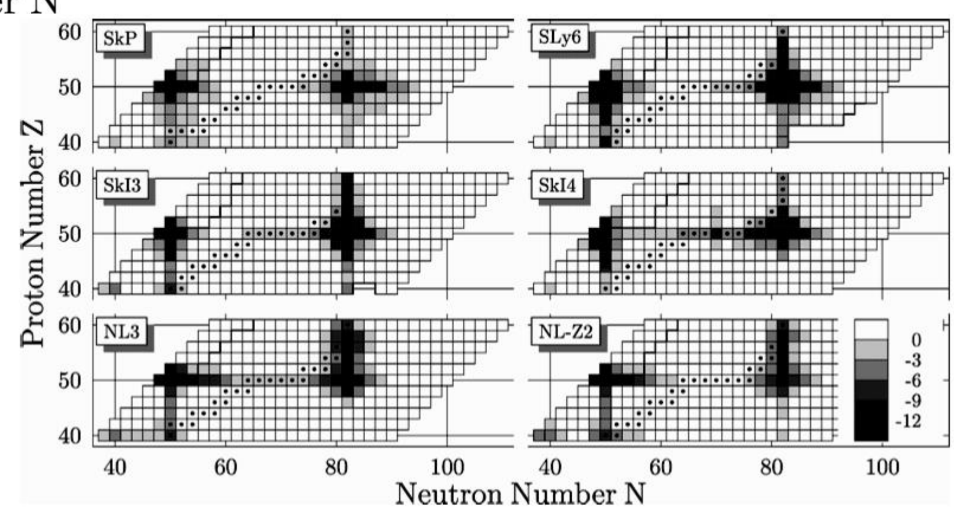
- Higher level density
- Spin orbit → orbitals flipped
- Low *j* orbitals → can modify significantly the gap but not drastically the binding energies

→ smooth island of stability

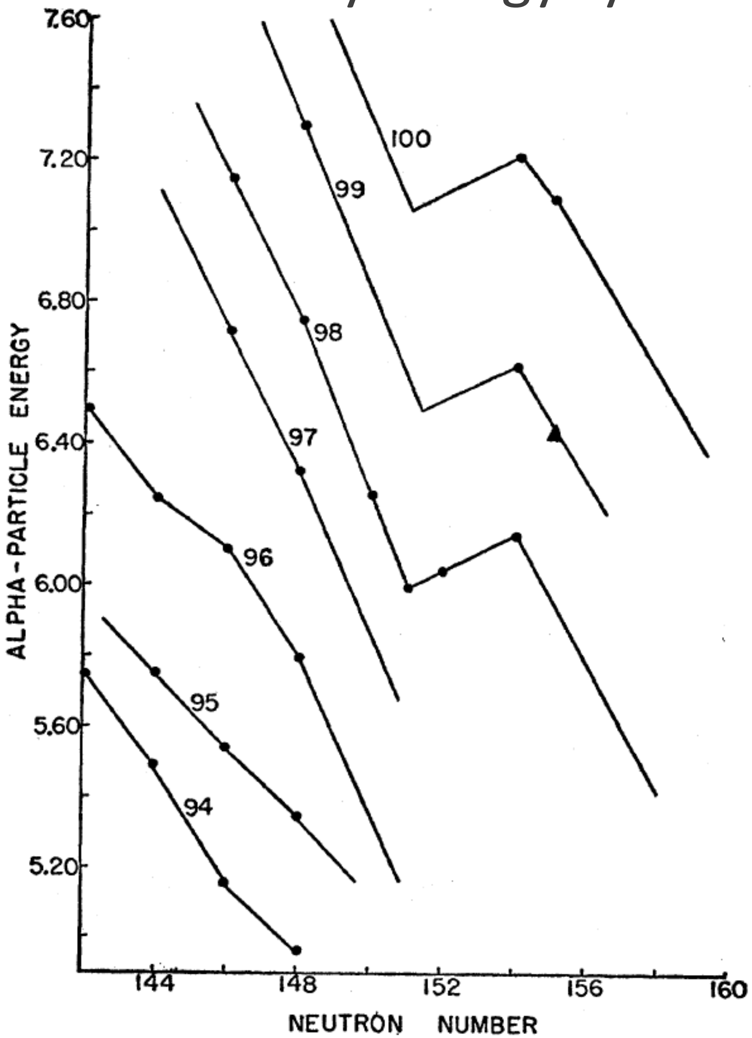
Doubly magic character of predicted SHE not as marked as lighter nuclei
Island of stability smooth and not well localized.



M. Bender et al., [PLB 515 (2001) 42]
Shell stabilization of super- and hyperheavy nuclei without magic gaps

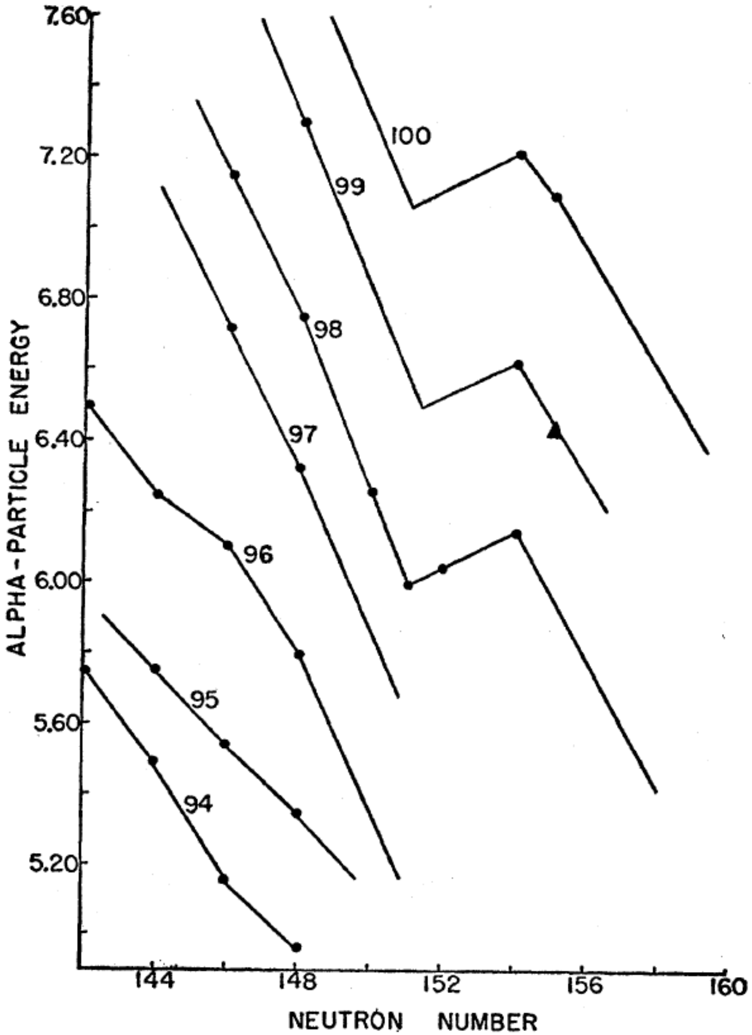


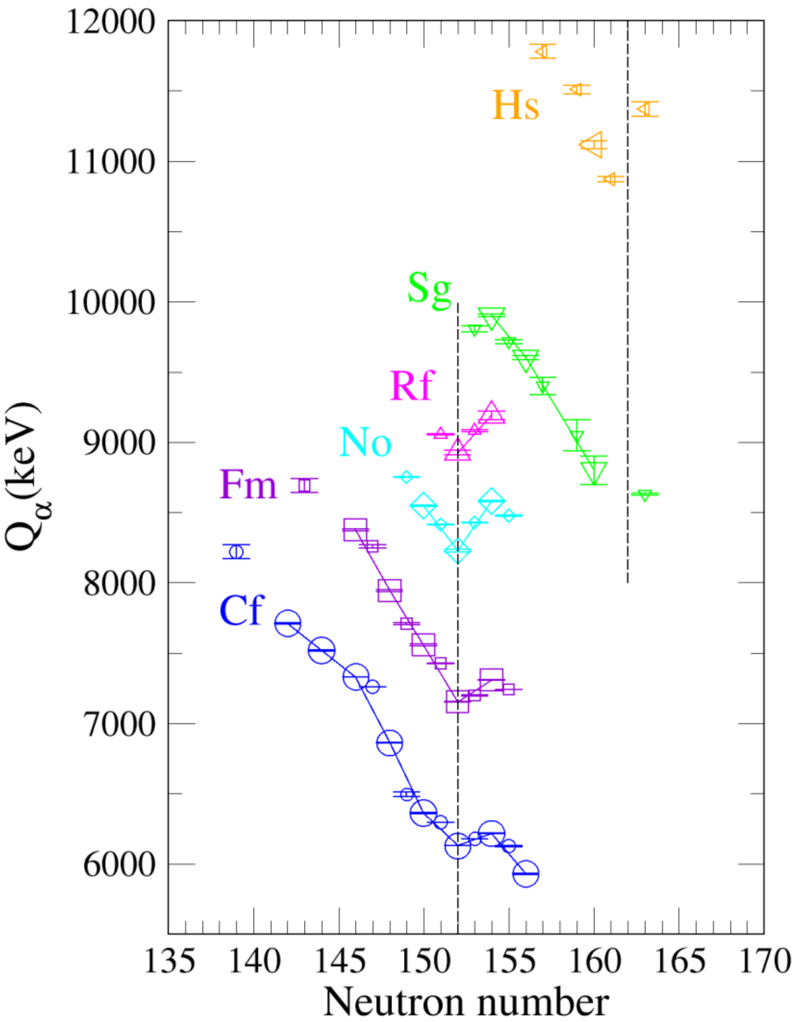
α-decay energy systematics, Ghiorso et al., PR 95 (1954) 293



pletion in the process of the filling of levels in the simple single-particle shell model may be an oversimplification because this is just the region where the strong surface coupling caused by large spheroidal distortion[10–13] or configuration interaction[14–18] of several nucleons may be important. In this connection one might expect on the basis of either the Bohr-Mottelson[12] strong surface coupling model or the de-Shalit-Goldhaber[15] configuration interaction arguments regarding trends of first excited state energies, that if the nucleon configuration at $N=152$ involves only completely filled levels, the first excited state energies should approach a maximum as is observed in the closing of other shells[19–21]; the experimental evidence so far indicates that this is not the case.[4,22] Thus it seems that the 152-neutron subshell may be of a fundamentally different nature than the major closed shells.

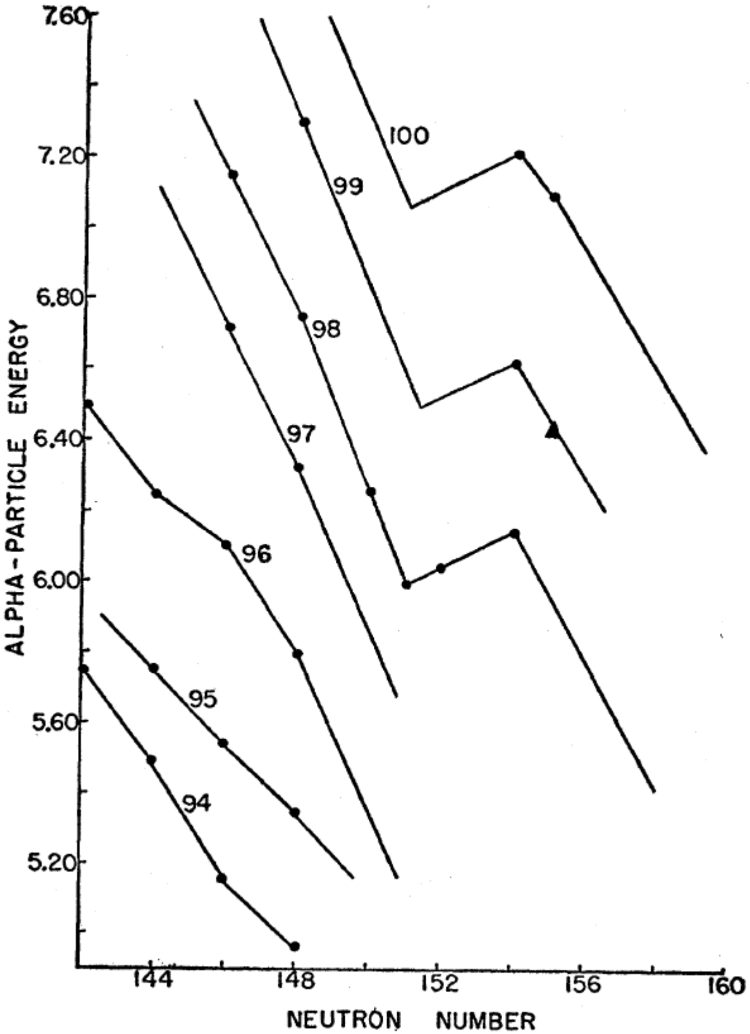
Experimental evidence



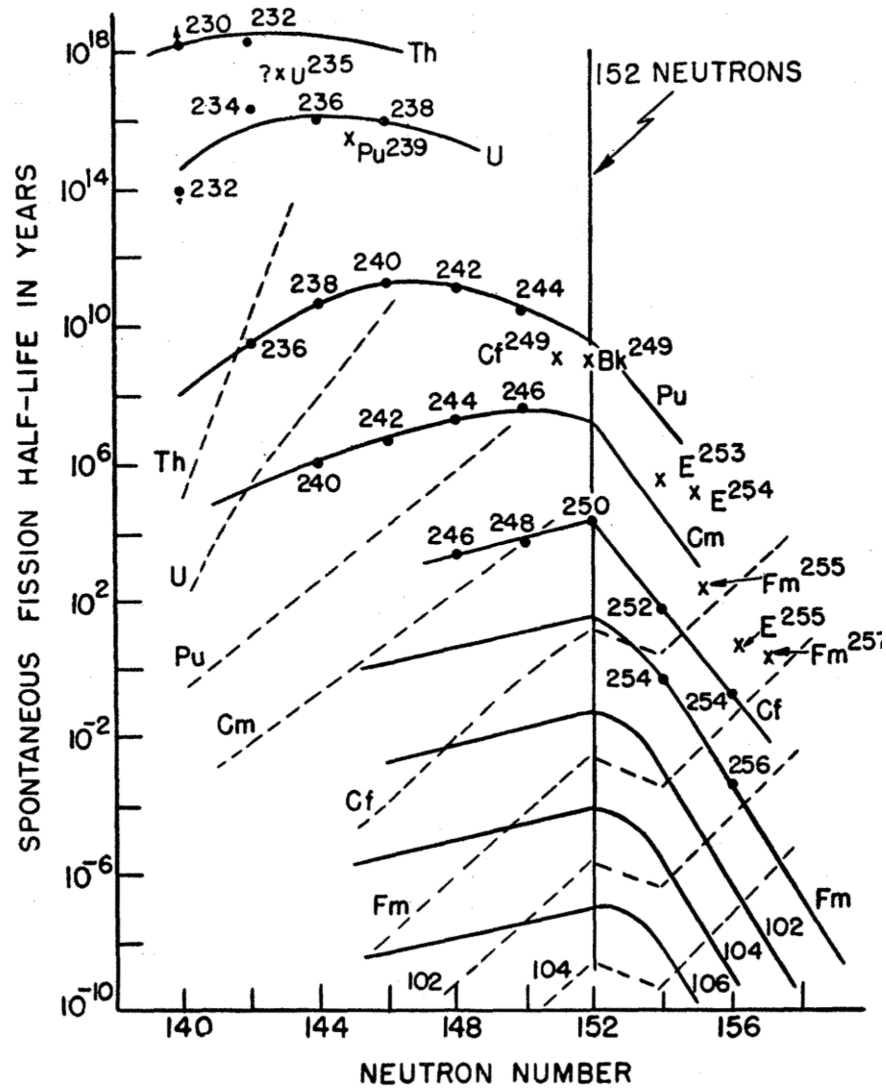


Ghiorso et al., [PR 95 (1954) 293]

Already suspected by Ghiorso in 1955

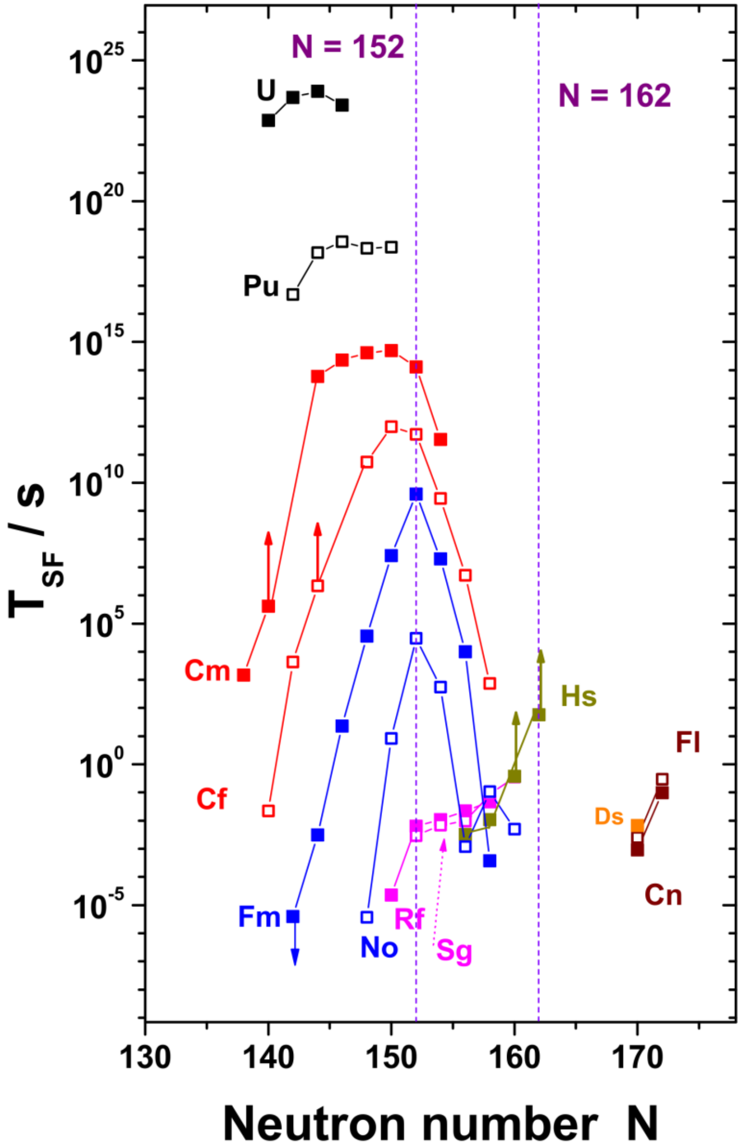


Ghiorso et al., [PR 95 (1954) 293]



Ghiorso, Proc. Int. Conf. on the Peaceful Uses of Atomic Energy, Geneva 1955, Vol. 7.

Spontaneous fission



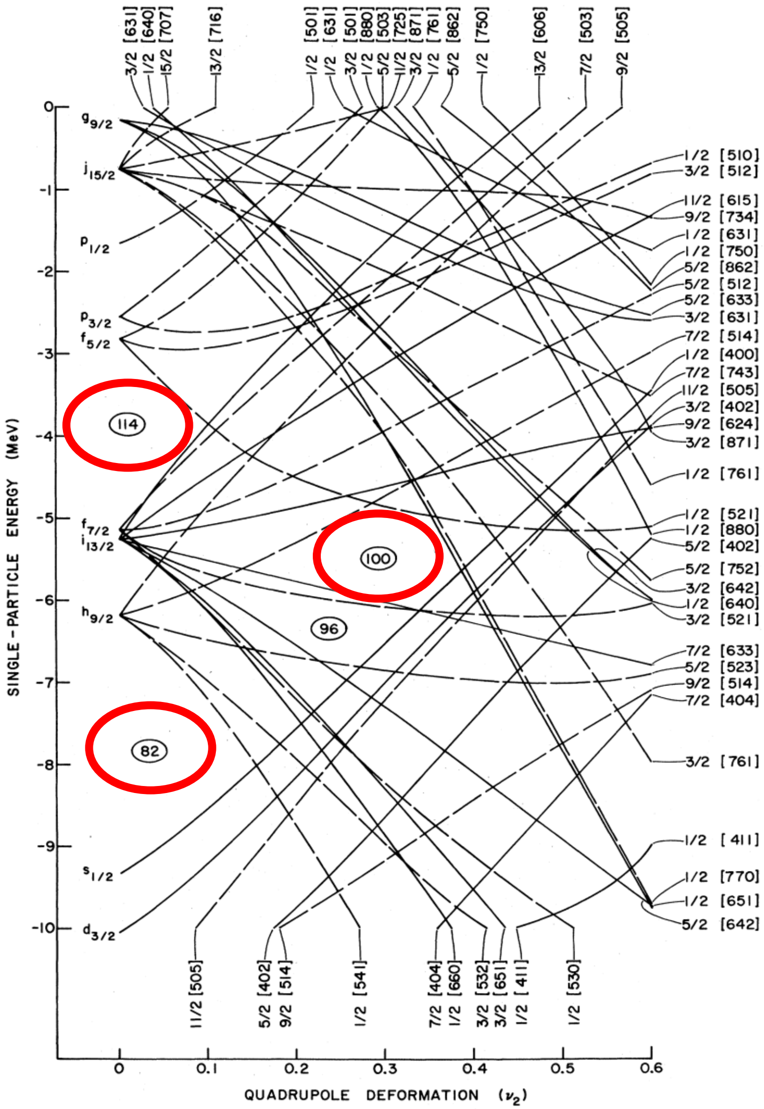
Hessberger, EPJA 53 (2017) 75

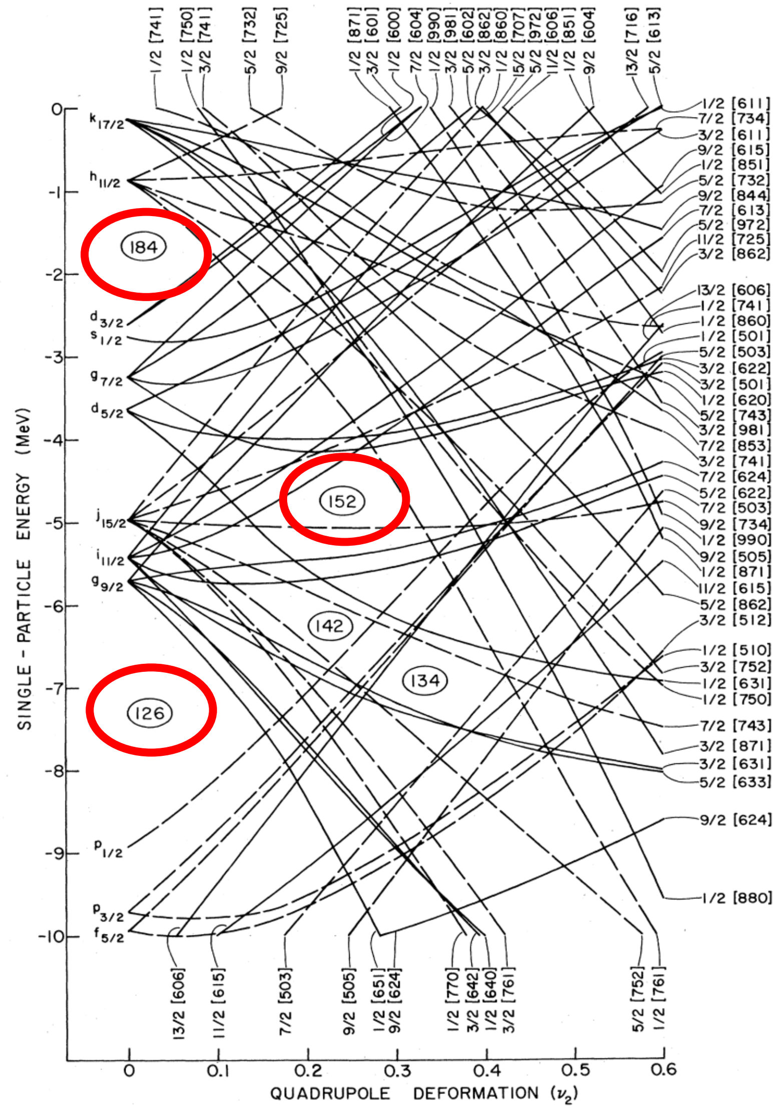
Deformed SHN nuclei ?

1949 : Townes systematics of electric quadrupole moments PR 76 (1949) 1415

1950 : Spheroidal model by J. Rainwater, unified model by Bohr and Mottelson

1955 : Nilsson deformed shell model by S.G. Nilsson



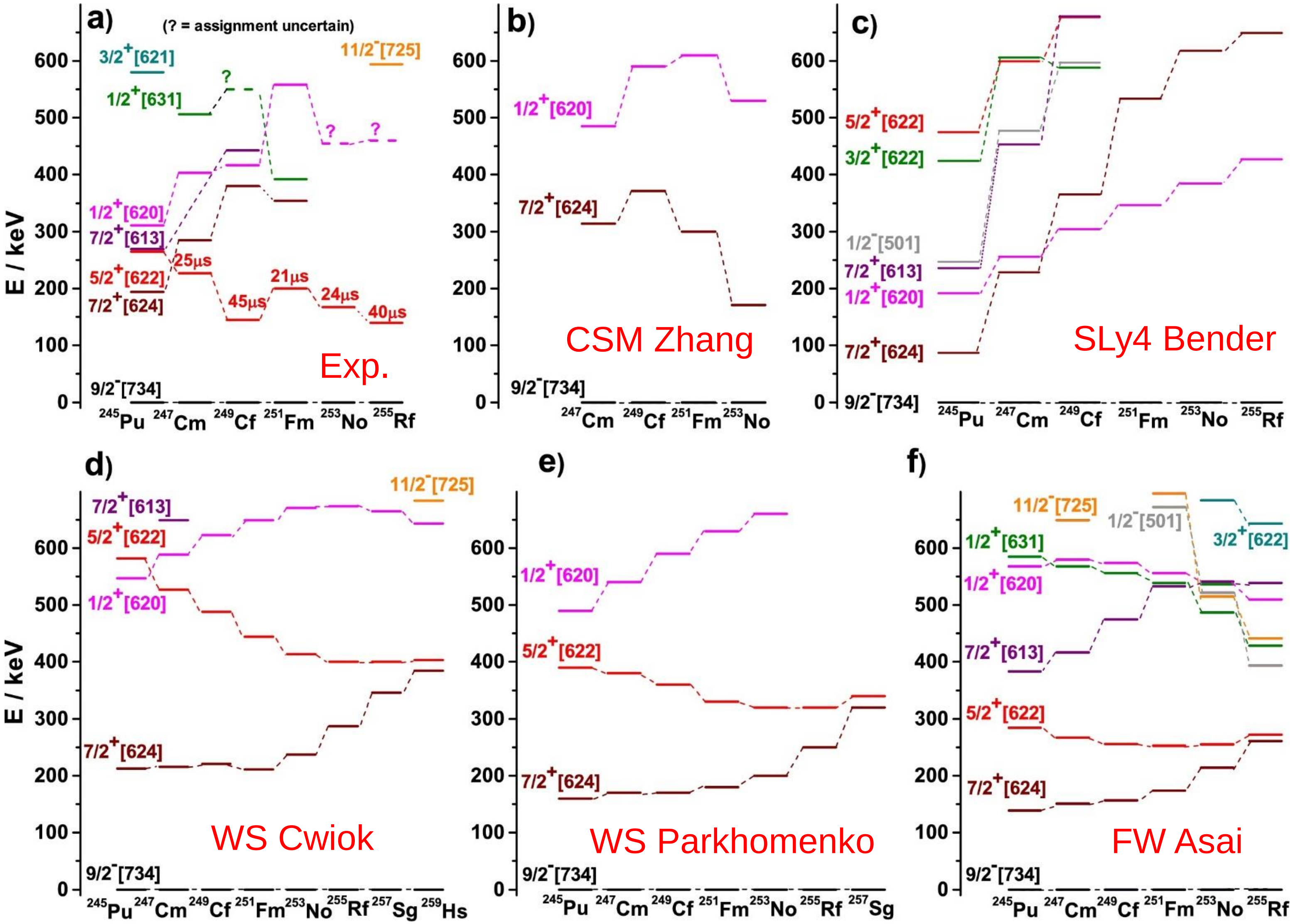


Chasman, Ahmad, Friedman, and Erskine. Rev. Mod. Phys. 49 (1977) 833

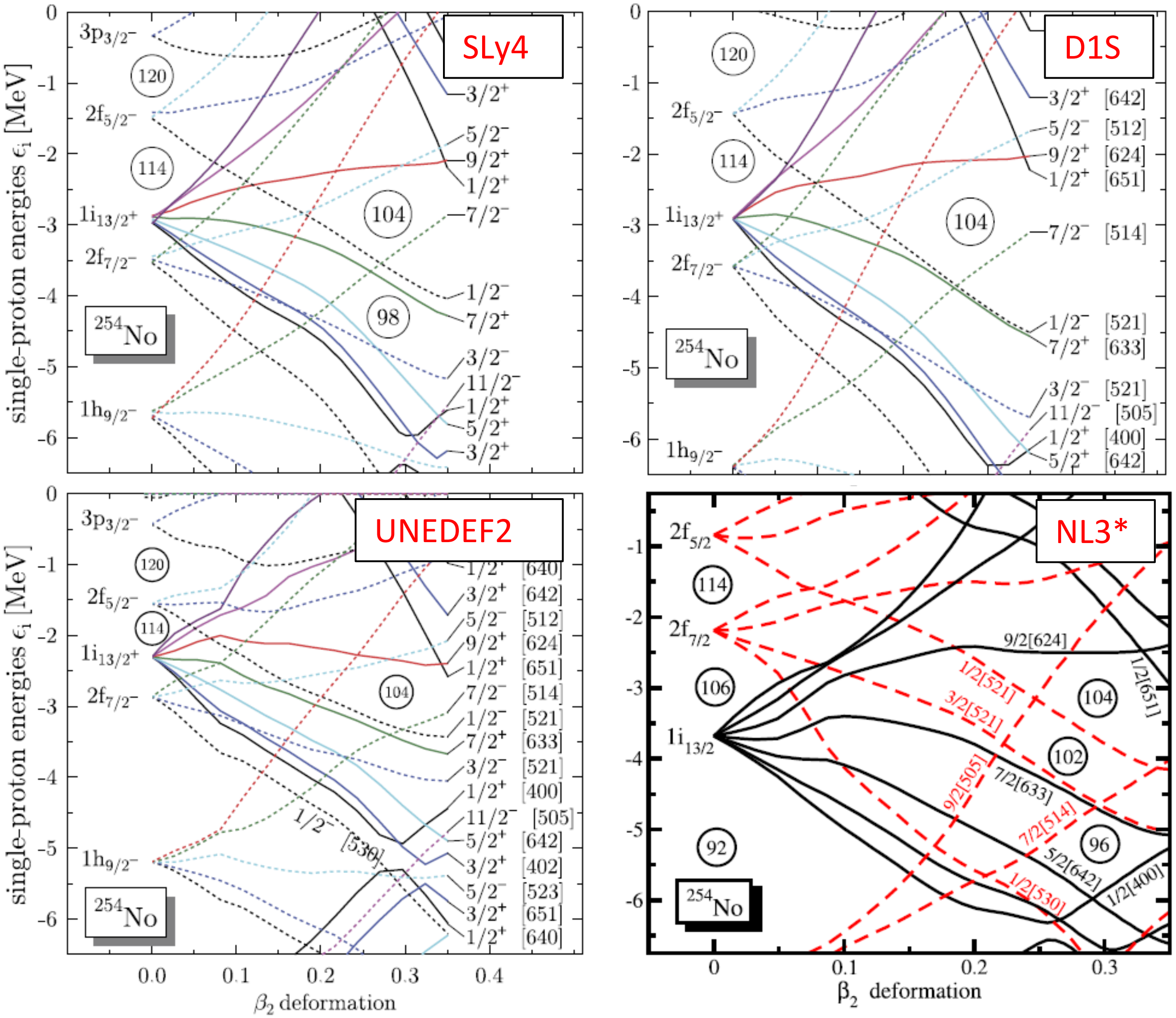
Let's do spectroscopy in the A~250 region !

→ energy, spin and parity and compare with theory

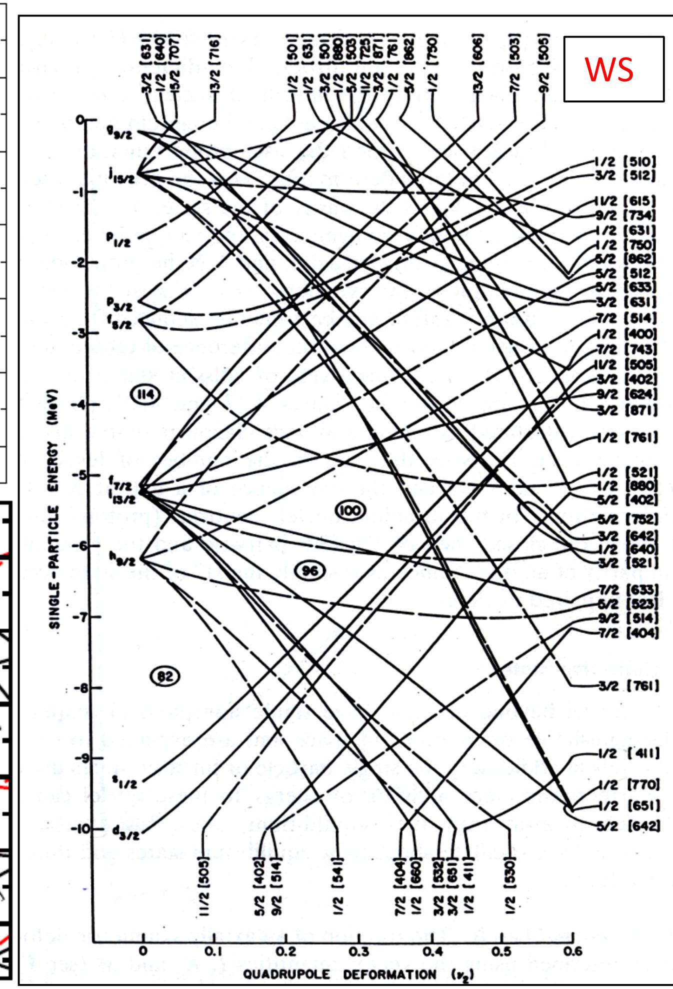
Asai, Heßberger and Lopez-Martens NPA 944 (2015) 308



Deformed shell gaps vs theory



Dobaczewski et al. [NPA 944 (2015) 388](https://doi.org/10.1016/j.nuclphysa.2015.10.001)



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

Z=100, N=152 deformed shells

Density Functional Theories fail to reproduce simultaneously Z=100 and N=152

Phenomenological potential reproduce Z=100 and N=152

- **What is the problem at Z = 100 / N = 152 ?**

Z=108, N=196 deformed shells

- **All models agree !**

SHE region

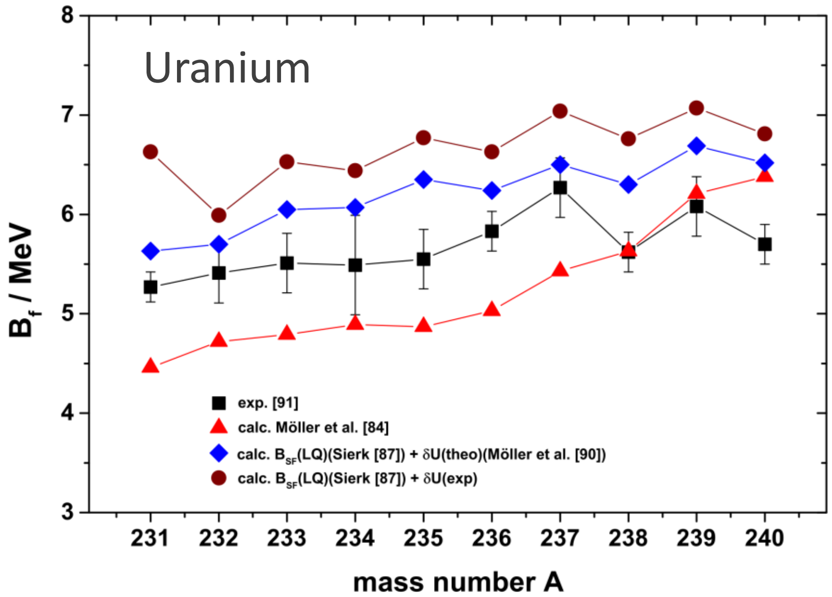
WS : Z=114 / N = 184

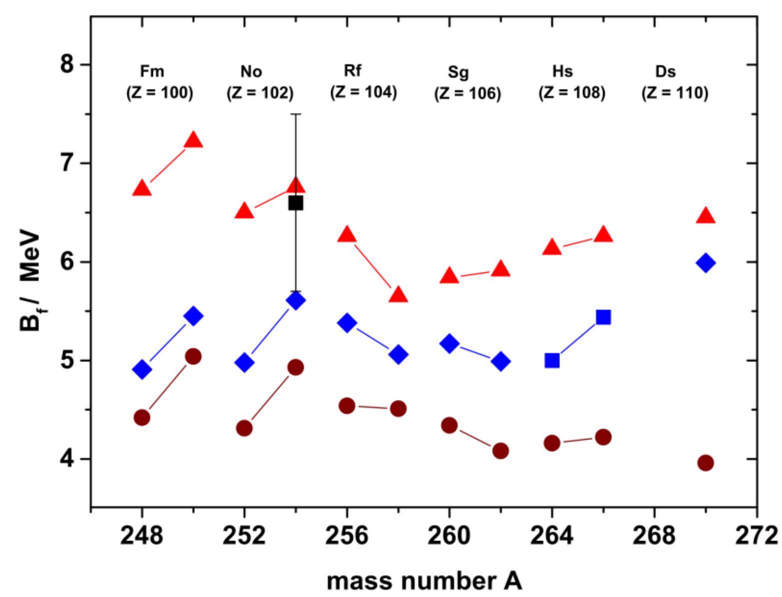
HFB : Z=126 / N =184

RMF : Z=120 / N =172

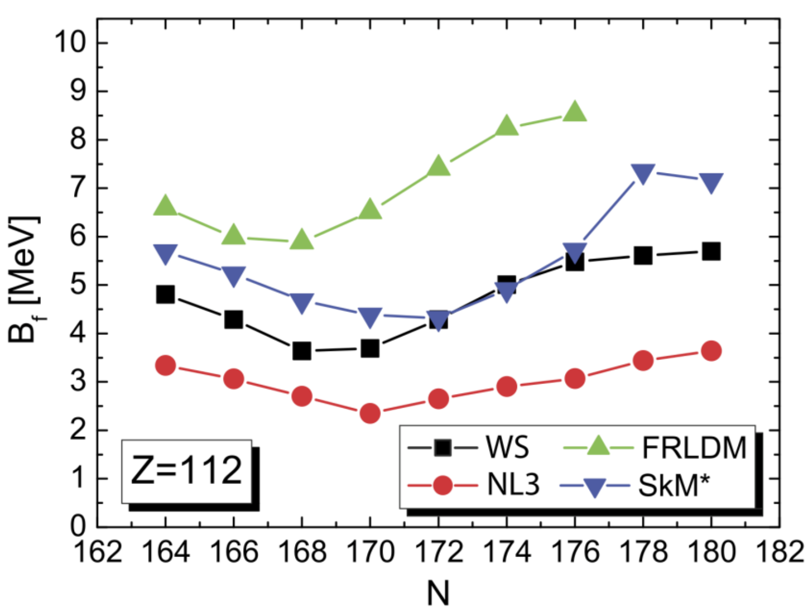
- **Who should the experimenter trust ?**

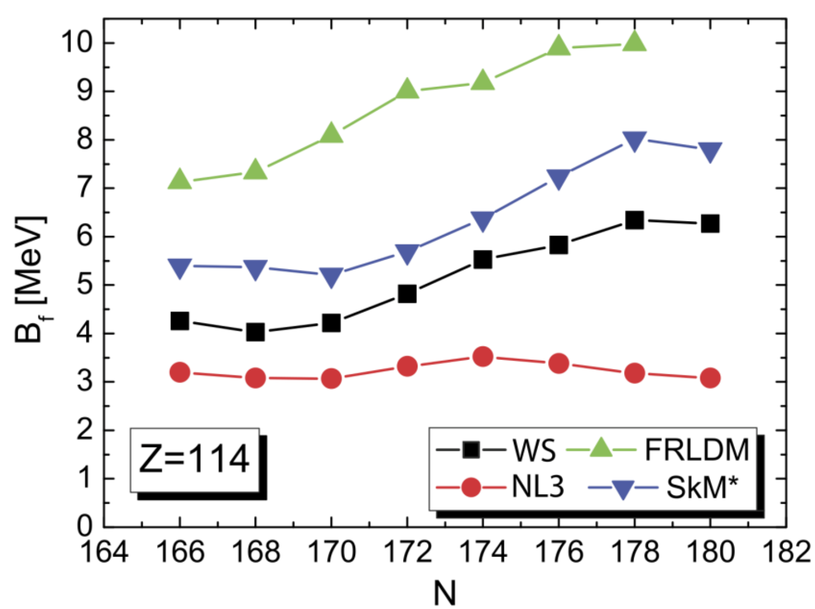
What about fission barriers ?





Hessberger, EPJA 53 (2017) 75





Baran et al., NPA 944 (2015) 442

How to measure a deformation ?

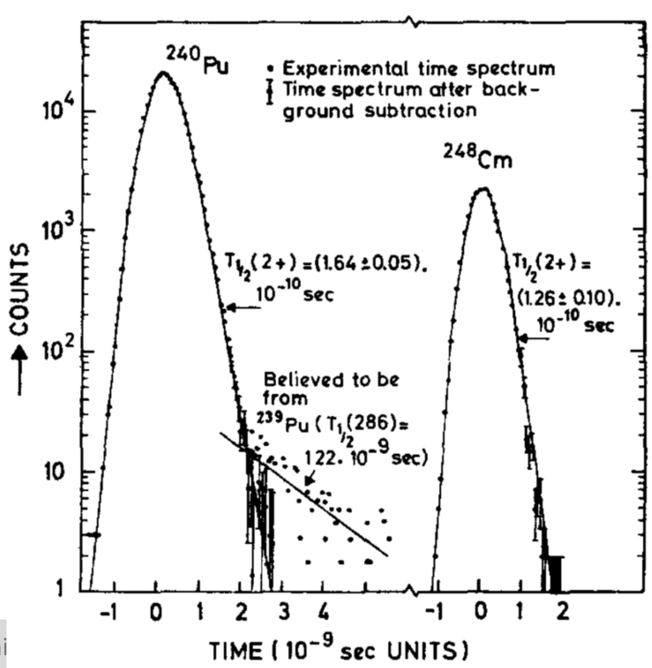
Fast timing

transition rate T = $\propto E^5$ B(E2) (1+α)

$B(E2, 0+ \rightarrow 2+) = \frac{5}{16\pi} Q_{20}^2$

Ton et al., NPA 155 (1970) 235

- 2^+ lifetime measurement ^{228}Th, ^{234,236}U, ^{238,240}Pu, ^{248}Cm
- Huge internal conversion for $2^+ \rightarrow 0^+$: fast timing measurement for low energy electrons.



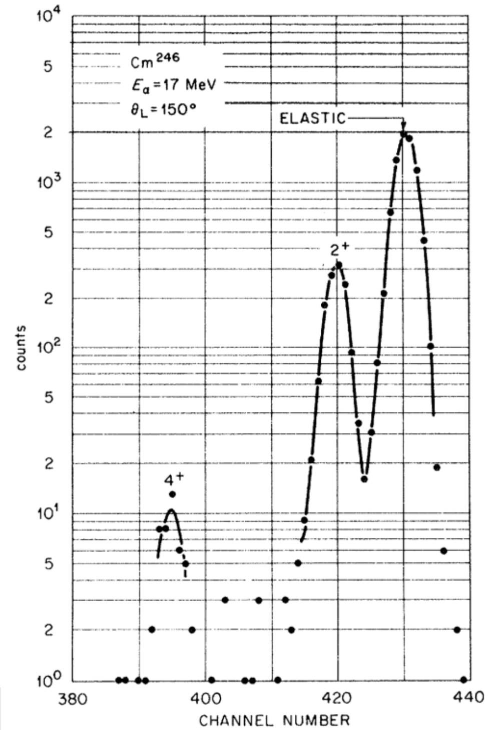
Coulex

σ(coulex) = σ(ruth) P(0+ → 2+)

P(0+ → 2+) → B(E2, 0+ → 2+)

Ford et al., PRL 27 (1971) 1232

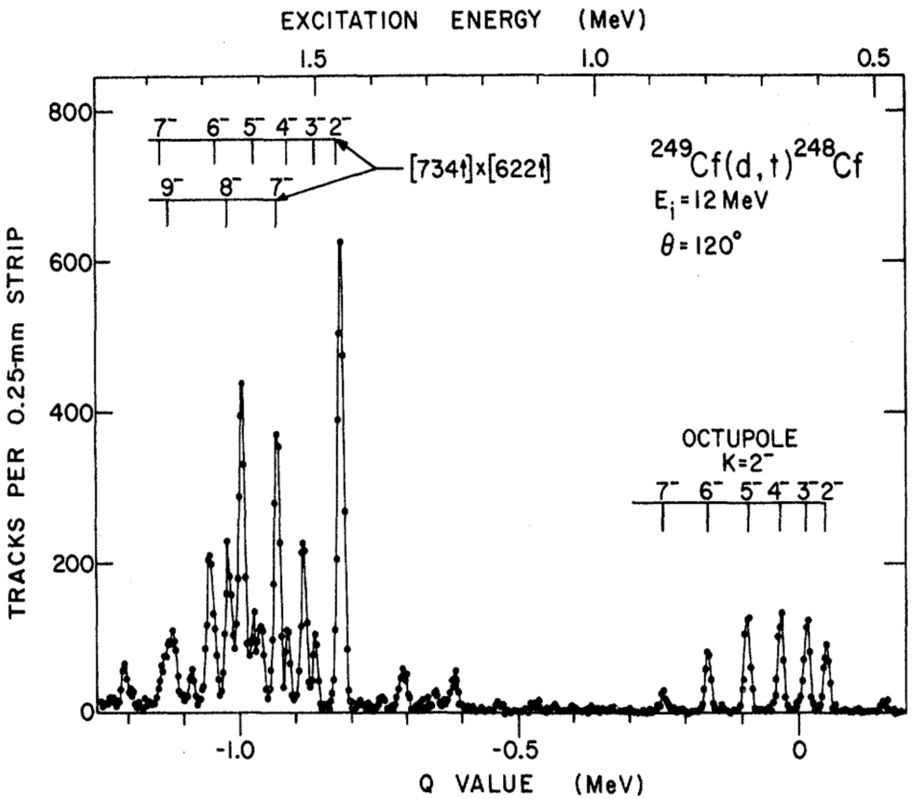
^{230,232}Th, ^{234,236,238}U, ^{238,240,242,244}Pu, ^{244,246,248}Cm, and ^{252}Cf using α beam



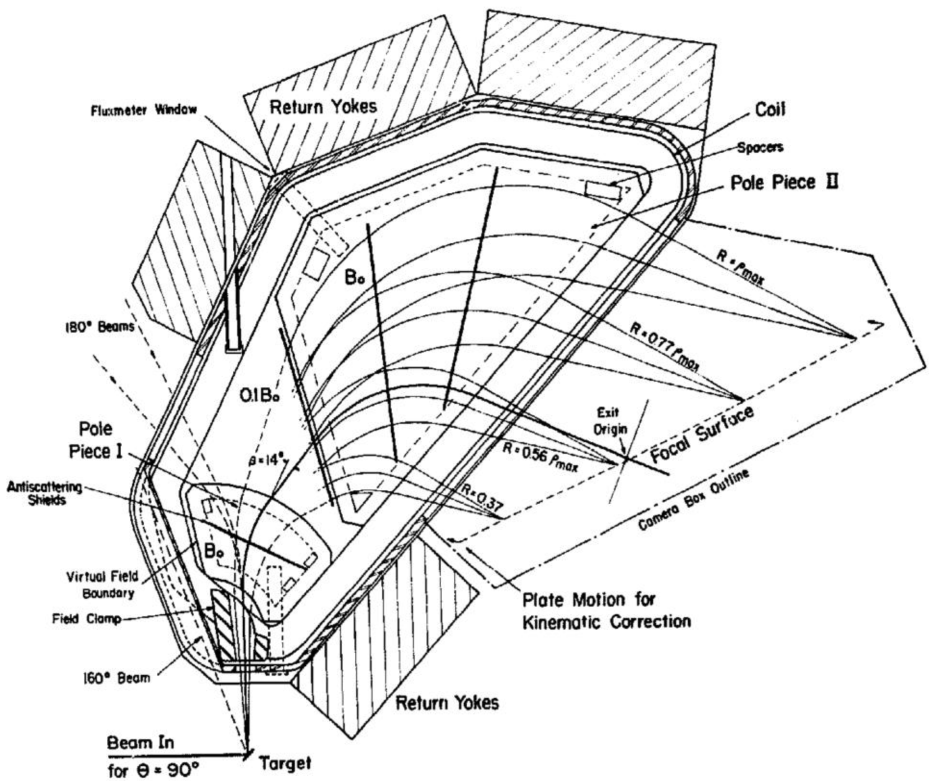
Low spin collective properties using direct reactions

Direct reactions e.g. (α,t), (d,p) or (d,t)
Ejectile detection using a spectrograph
- Enegy
- Angular distributions

→ E, J^{π}; orbital occupancy



[Yates et al. PRC 12 (1975) 442]

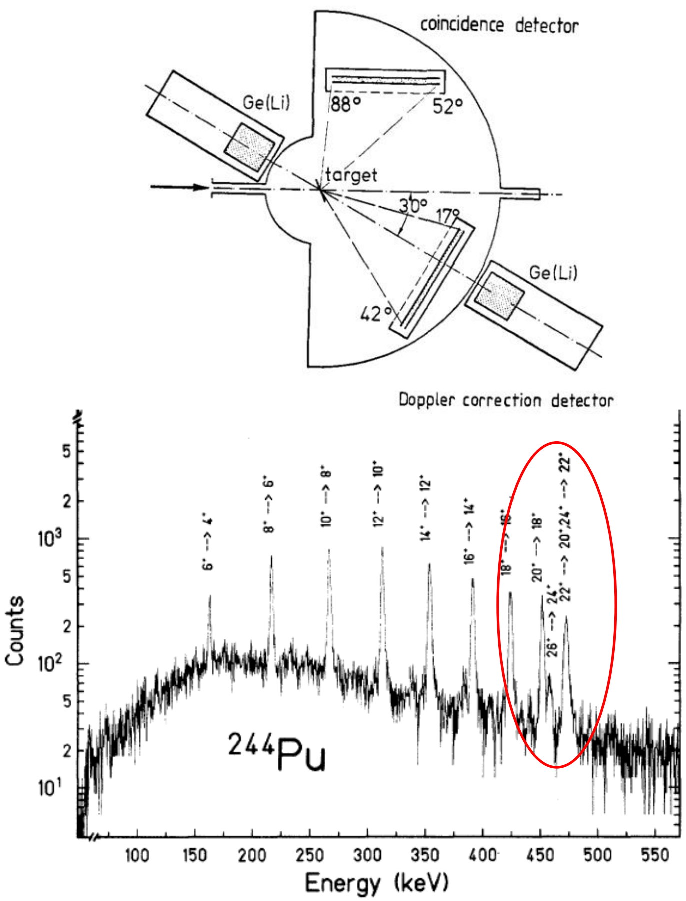


Enge NIM 162 (1979) 161

Spectroscopy using Heavy-ion Coulex

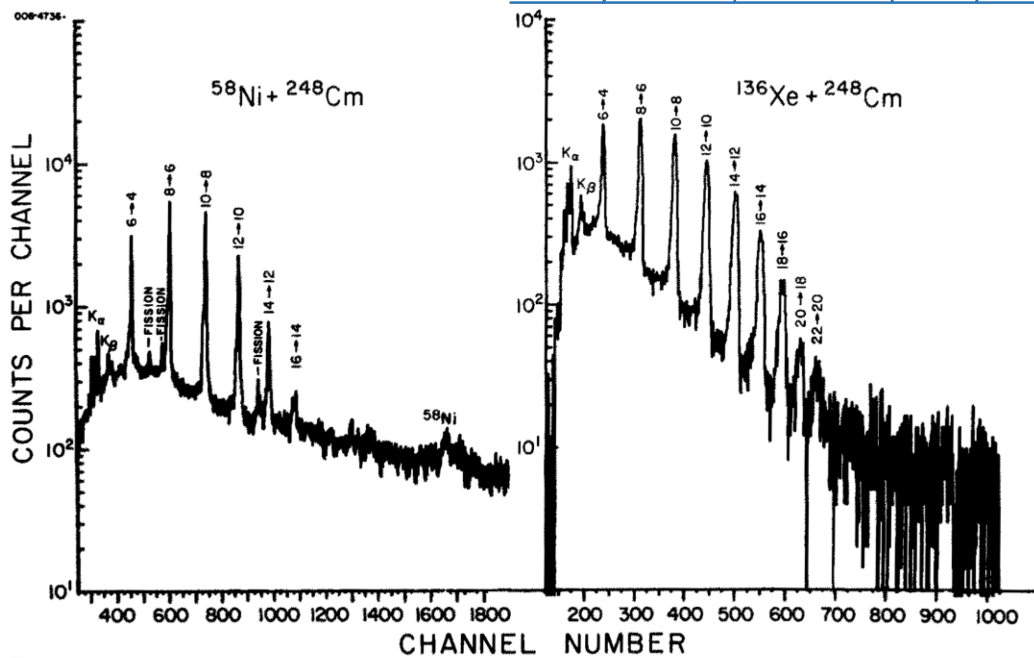
GSI 80s : ^{208}Pb beam → Coulex ^{232}Th, ^{234,236,238}U, ^{242,244}Pu, ^{248}Cm

[Speng et al., PRL 17 (1983) 1522]



LBL 80s : B(E2) ^{248}Cm using ^{58}Ni, ^{136}Xe beams

Czosnyka et al., NPA 458 (1986) 123



Still only transuranium coulex using HI beam and reference value

→ Backbending in the transuranium region

→ the alignment of a πi13/2 or νj15/2 pair ?

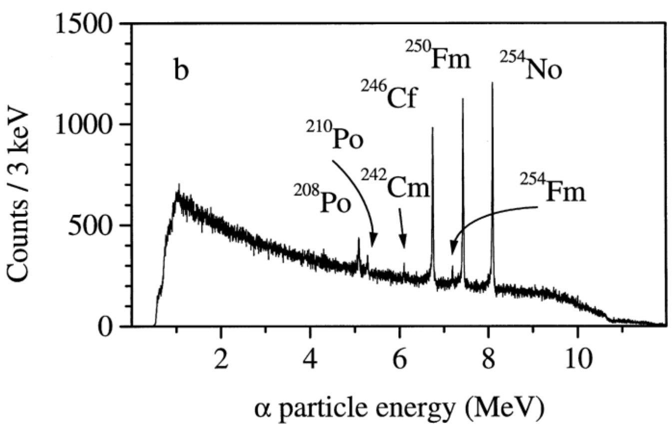
(still an open question)

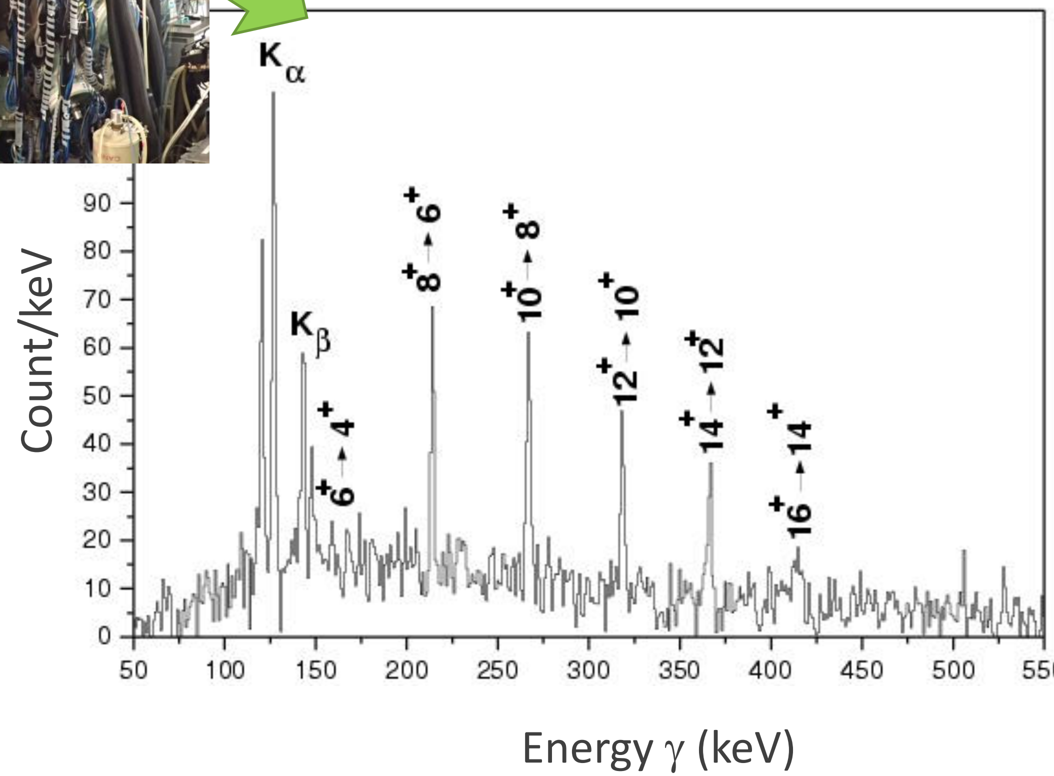
1981 : high-spin studies ^{248}Cm, coulex GSI

Noting heavier until 1997 : $^{208}Pb(^{48}Ca,2n)^{254}No$ Jyväskylä and Argonne

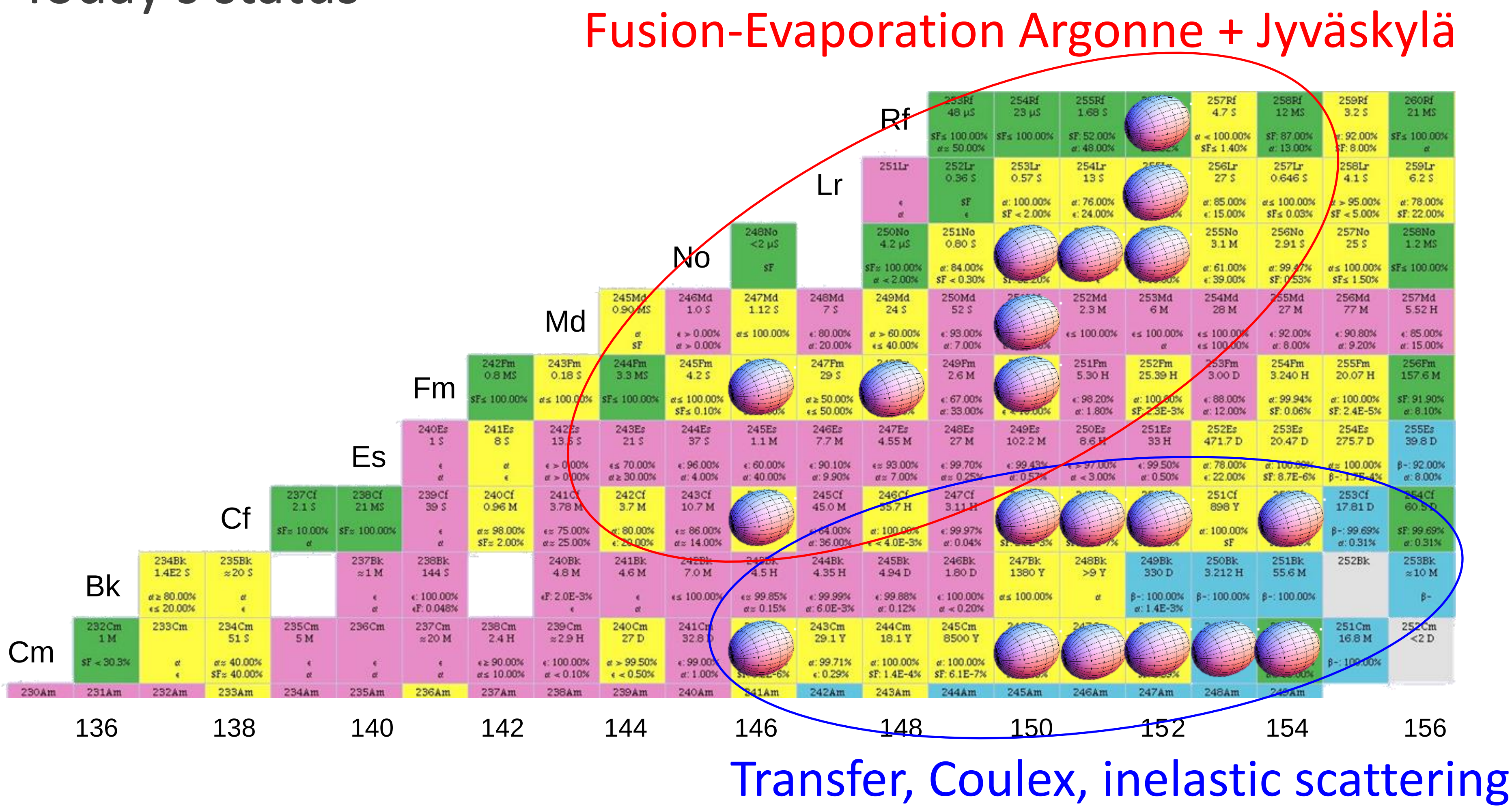
ANL : P. Reiter *et al.* PRL 82 (1999) 50

JYFL : M. Leino *et al.* EPJ A 6 (1999) 63

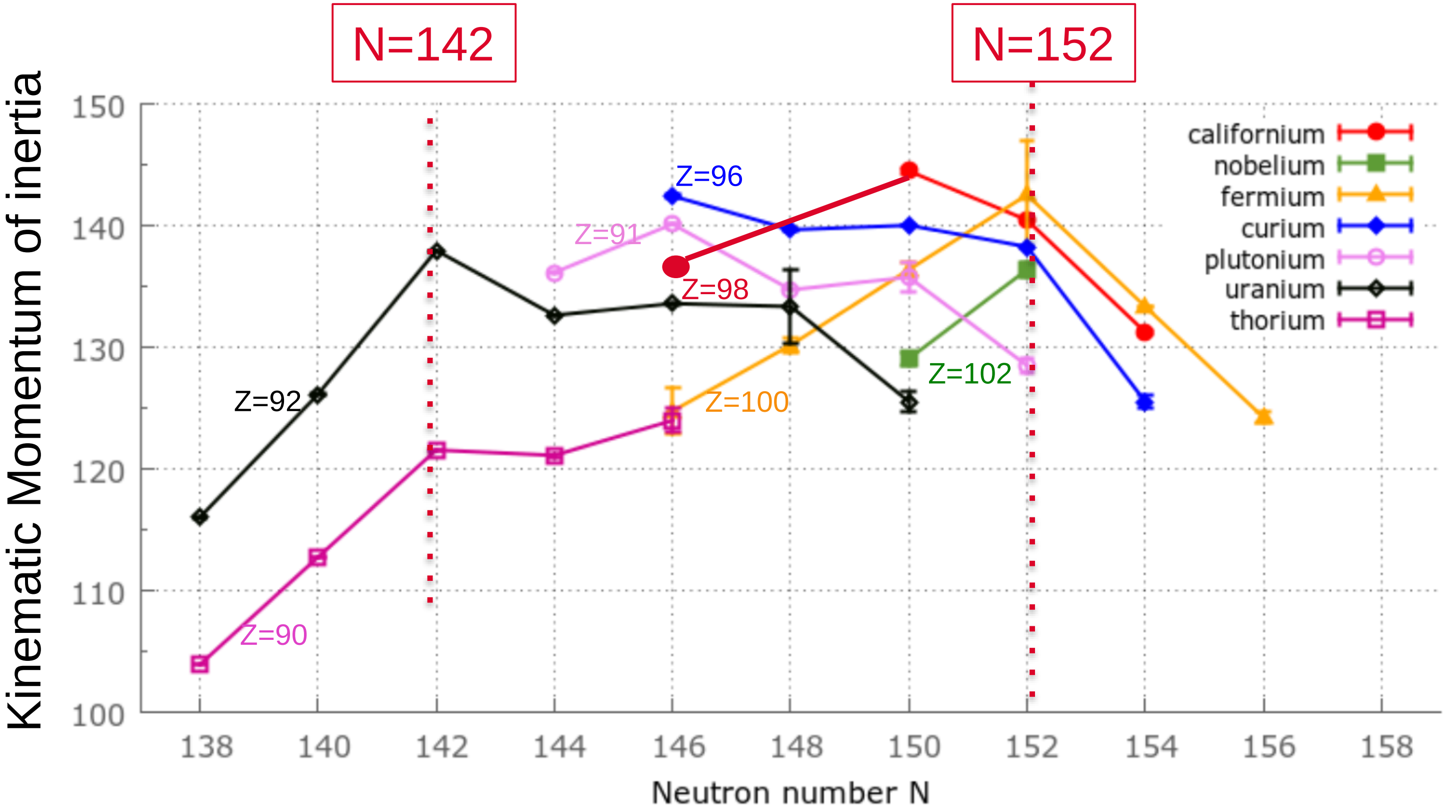




Today’s status



Moment of Inertia

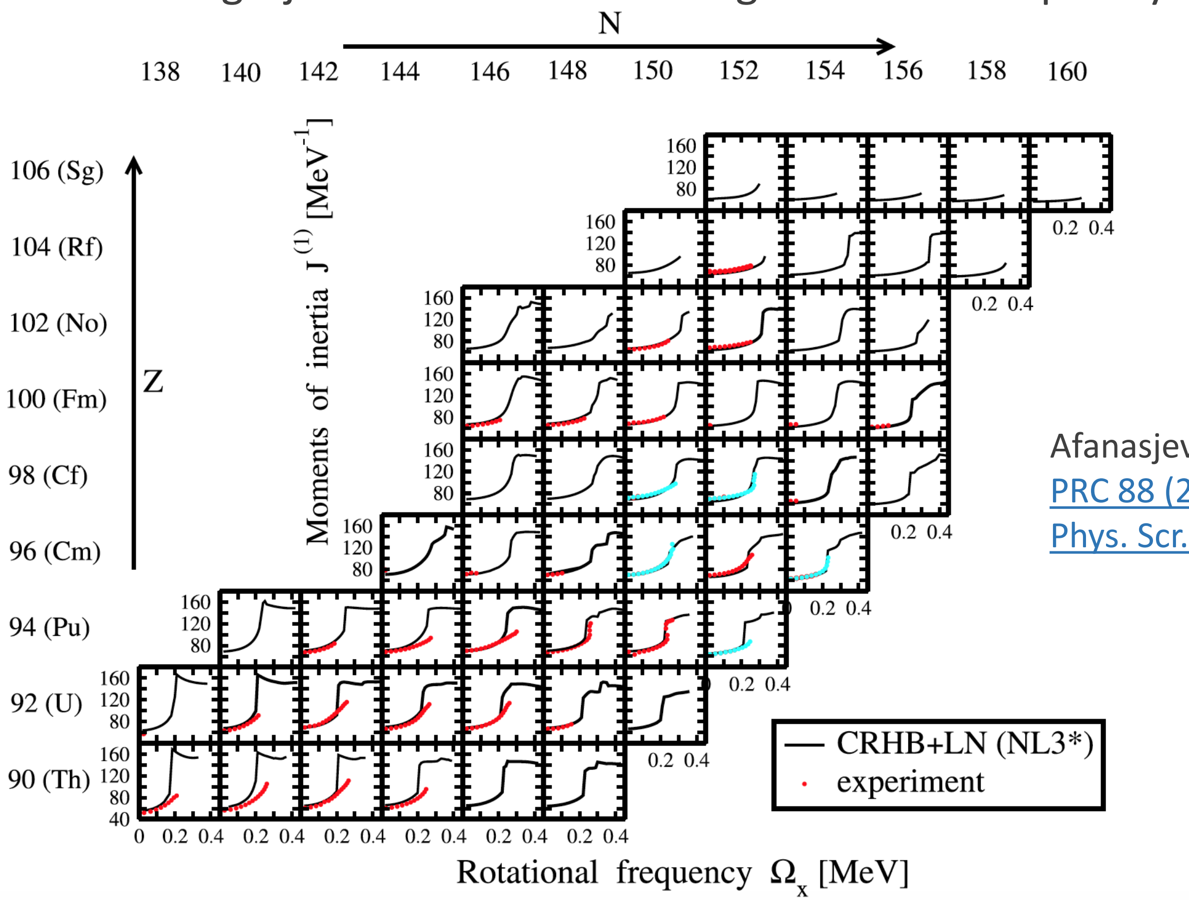


Moment of inertia vs theory

Overall good agreement

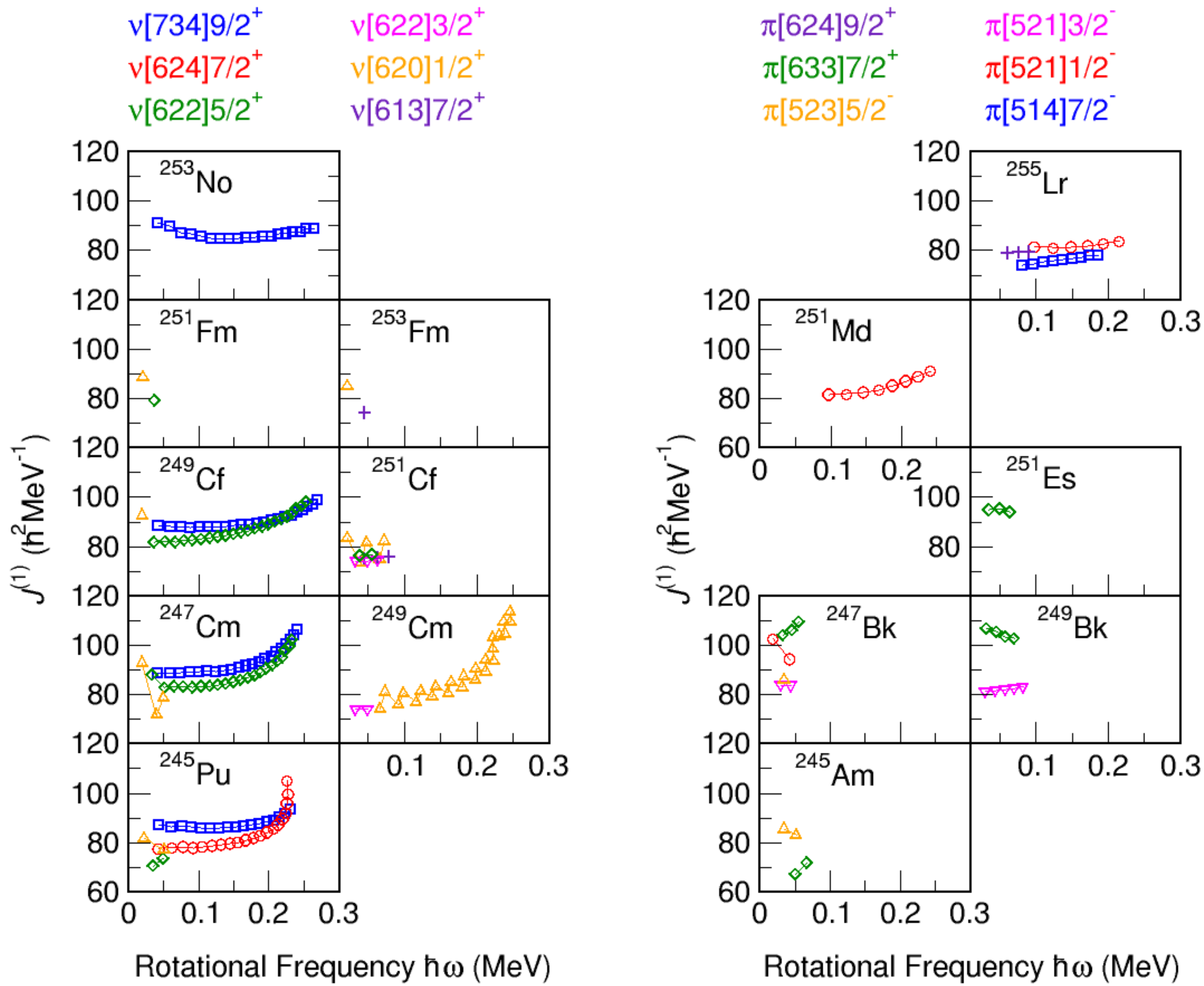
study upbending or backbending phenomena

→ Alignment of high-j orbitals with increasing rotational frequency ω

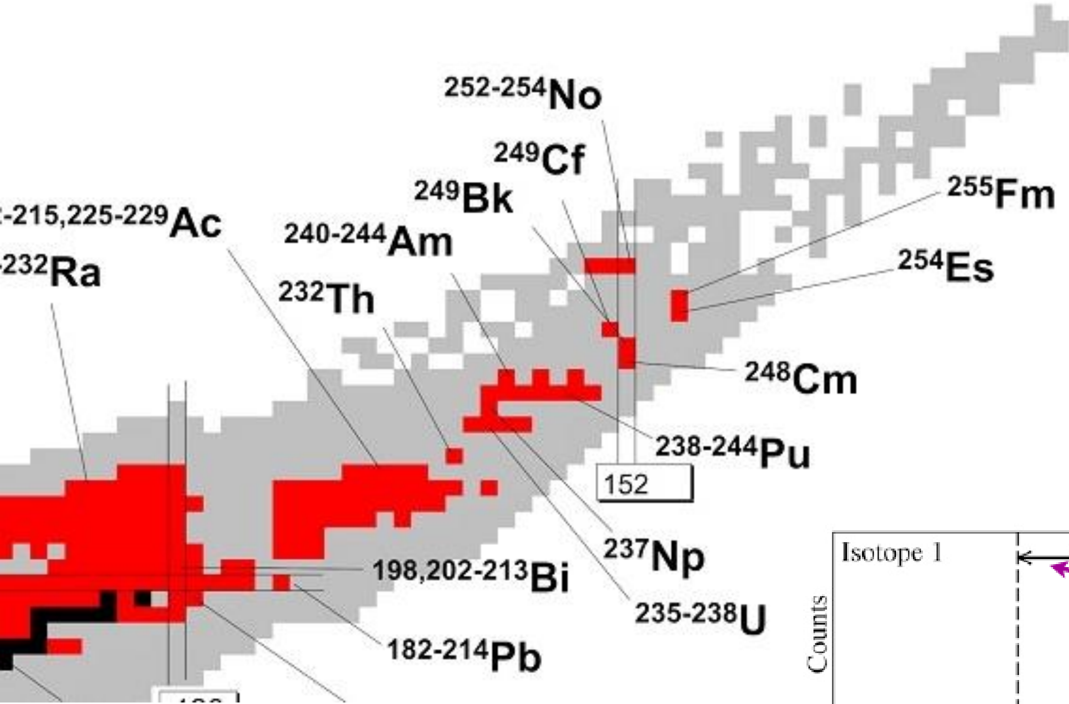


Afanasjev and Abdurazakov
[PRC 88 (2013) 014320]
[Phys. Scr. 89 (2014) 054001]

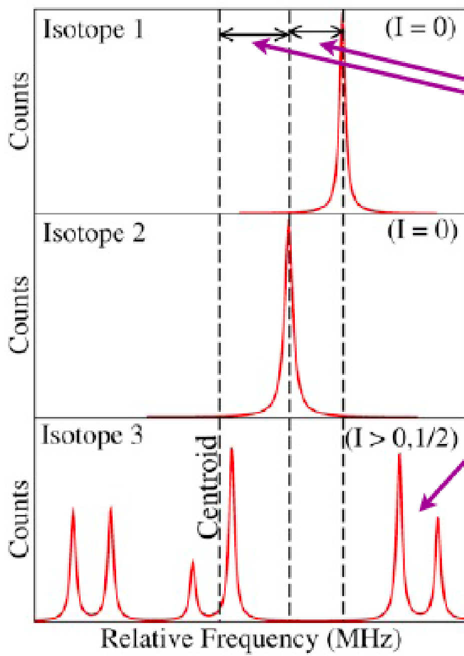
Odd-mass nuclei



Ackermann & Theisen
Phys. Scr. 92 (2017) 083002



Laser Spectroscopy Survey (GSI)
2021 01 update



Isotope Shifts
➔ $\delta\langle r^2 \rangle$

Hyperfine Structure
➔ μ
➔ Q_s ➔ $\langle\beta_2\rangle$
➔ Nuclear spin

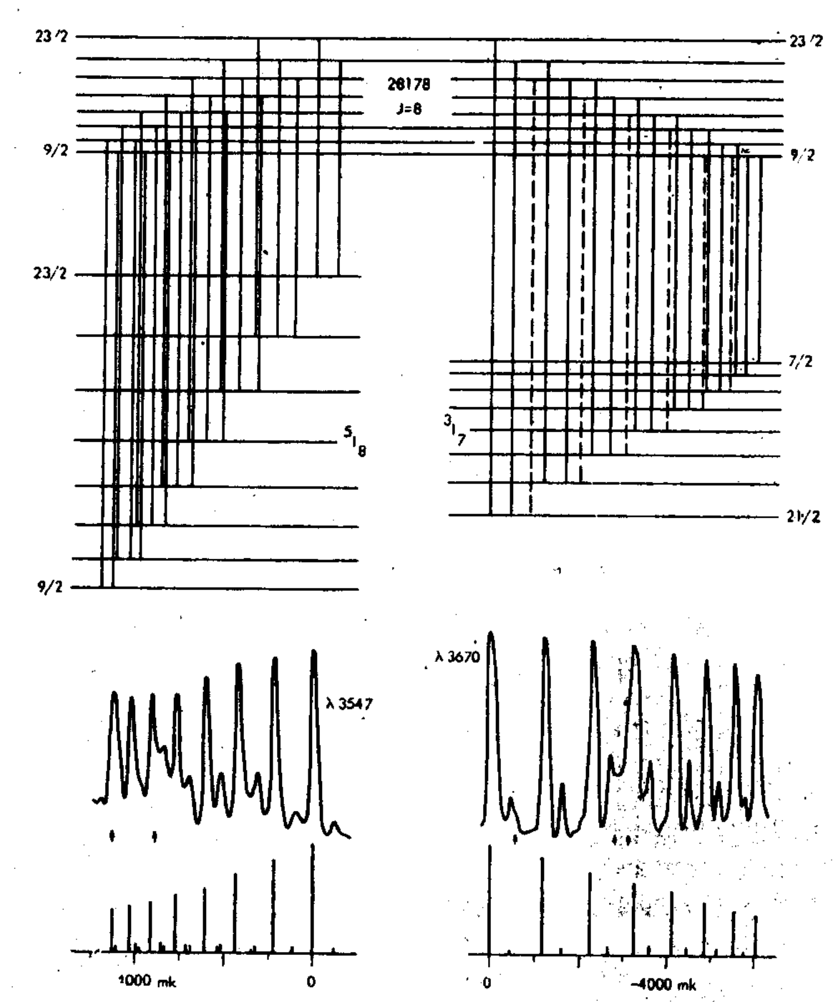
^{253}Es optics spectroscopy

E.S. Worden et al., Jour. Opt. Soc. Am. 58 (1968) 998, 60 (1970) 1297, 64 (1974) 77

Ionisation : lamp from ^{253}Es ($t_{1/2}$ = 20days) sample (0.8 µg)

53 lines observed;
23 with hyperfine structure

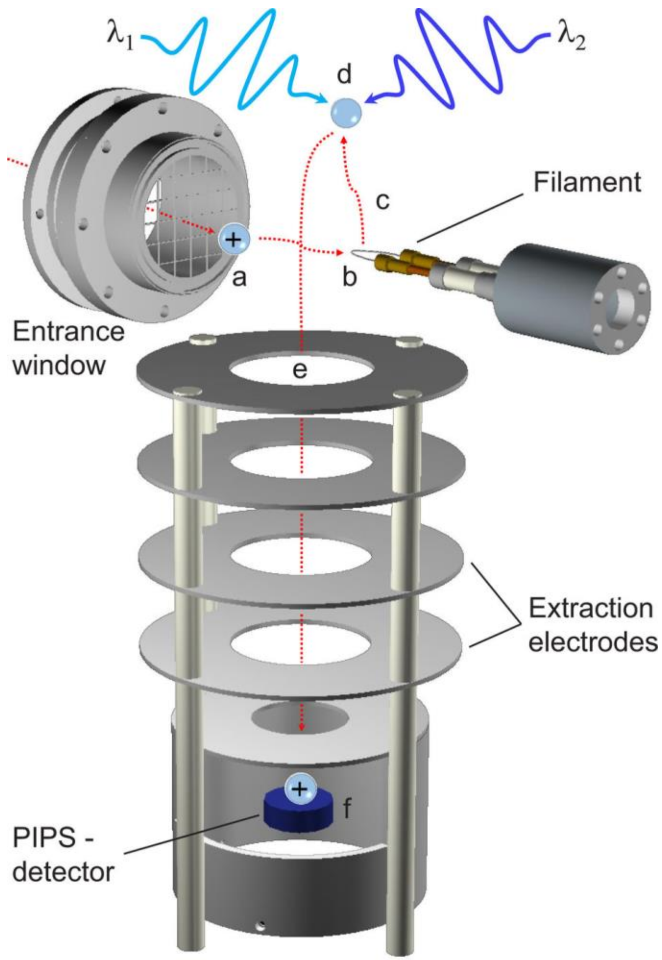
I = 7/2
$\mu = 3.6 \pm 0.4\ \mu_N$
Q = 6.0 ± 0.4 b

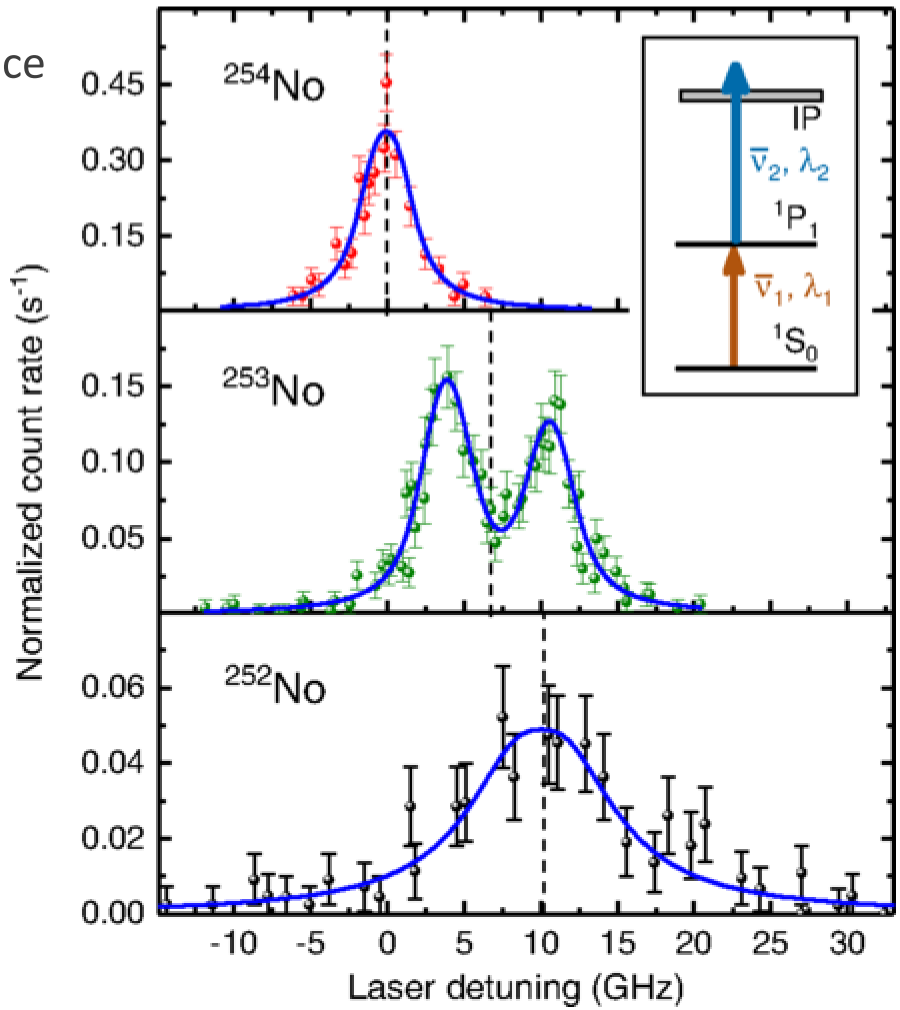


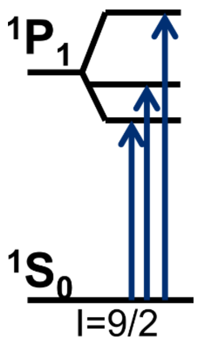
S. Raeder et al. PRL 120 (2018) 232503

RADRIS technique at SHIP

RAdioactive Decay-Detected Resonance Ionization Spectroscopy







^{253}No : I=9/2. μ =-0.527(33)(75) μ_N

Qs = +5.9(1.4)(0.9) eb

Some future trends

High beam intensities

- SHE Factory @ Dubna
- LINAG, S3, NEWGAIN @ SPIRAL2
  - → synthesis, decay spectroscopy, isomers, etc…

Precision measurements

- Mass measurement and Laser spectroscopy @ GSI
- S3 LEB (Low Energy Branch), DESIR
- Lifetime measurement ?
- Coulex ?

Reaction mechanism

- Fusion-evaporation, incomplete fusion (actinide)
- MultiNucleon Transfer reactions

Theory

- Reaction mechanism
- Structure

Selected topics on heavy and super-heavy nuclei