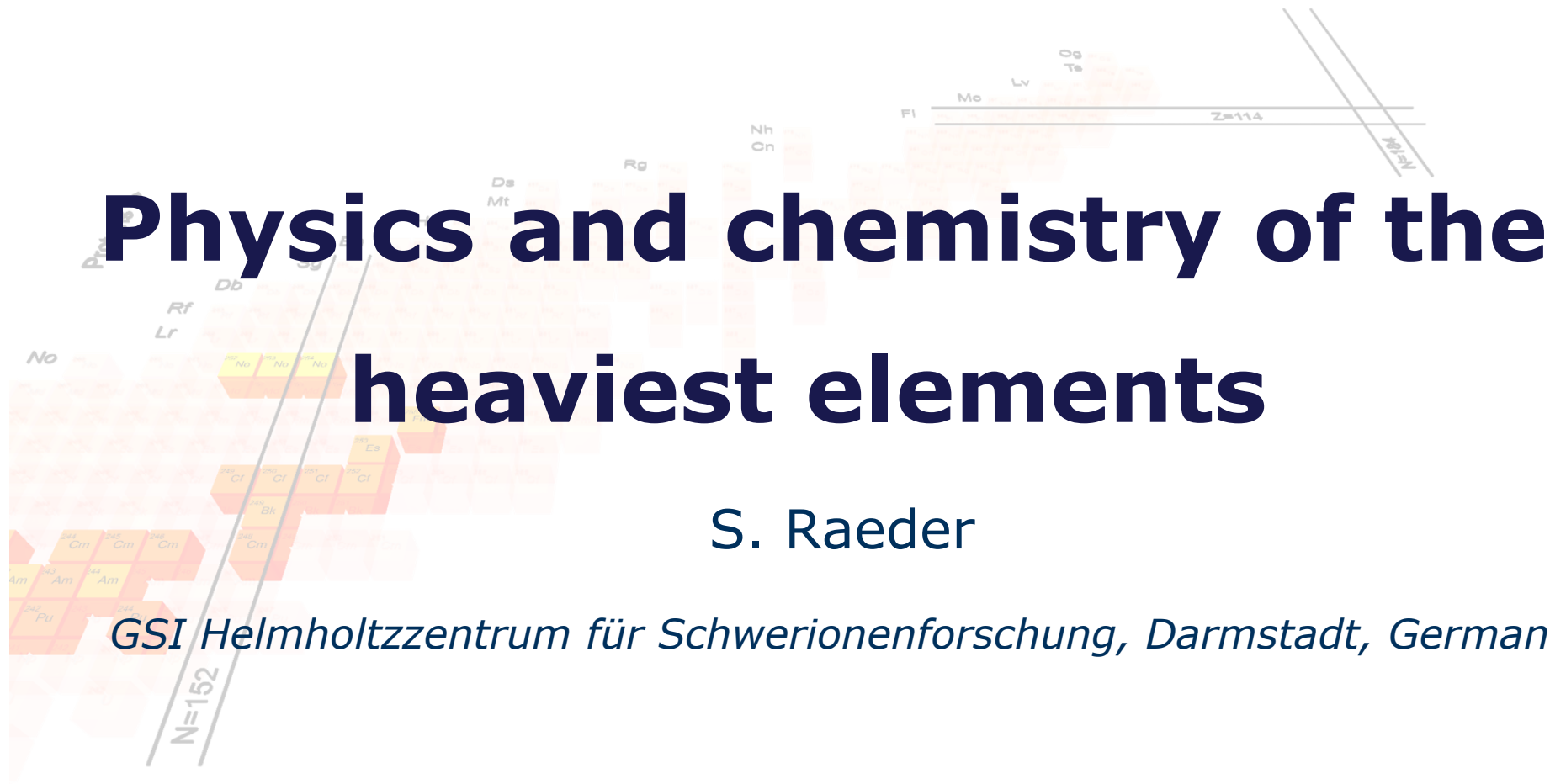


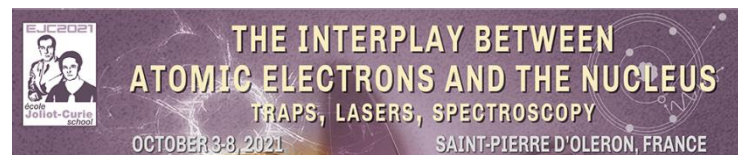
Physics and chemistry of the heaviest elements

S. Raeder

GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany



Neutrons →



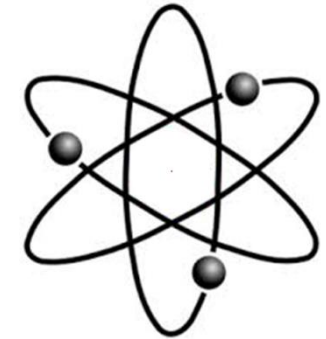
Heavy Elements - The far end of the periodic table

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 Tc	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											72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra											104 Rf 1964	105 Db 1967	106 Sg 1974	107 Bh 1981	108 Hs 1982	109 Mt 1984	110 Ds 1994	111 Rg 1994	112 Cn 1996	113 Nh 2003	114 Fl 1999	115 Mc 2003	116 Lv 2000	117 Ts 2009	118 Og 2002



Electron shell

- atomic structure
- chemical properties
- defines the element



Super Heavy Elements

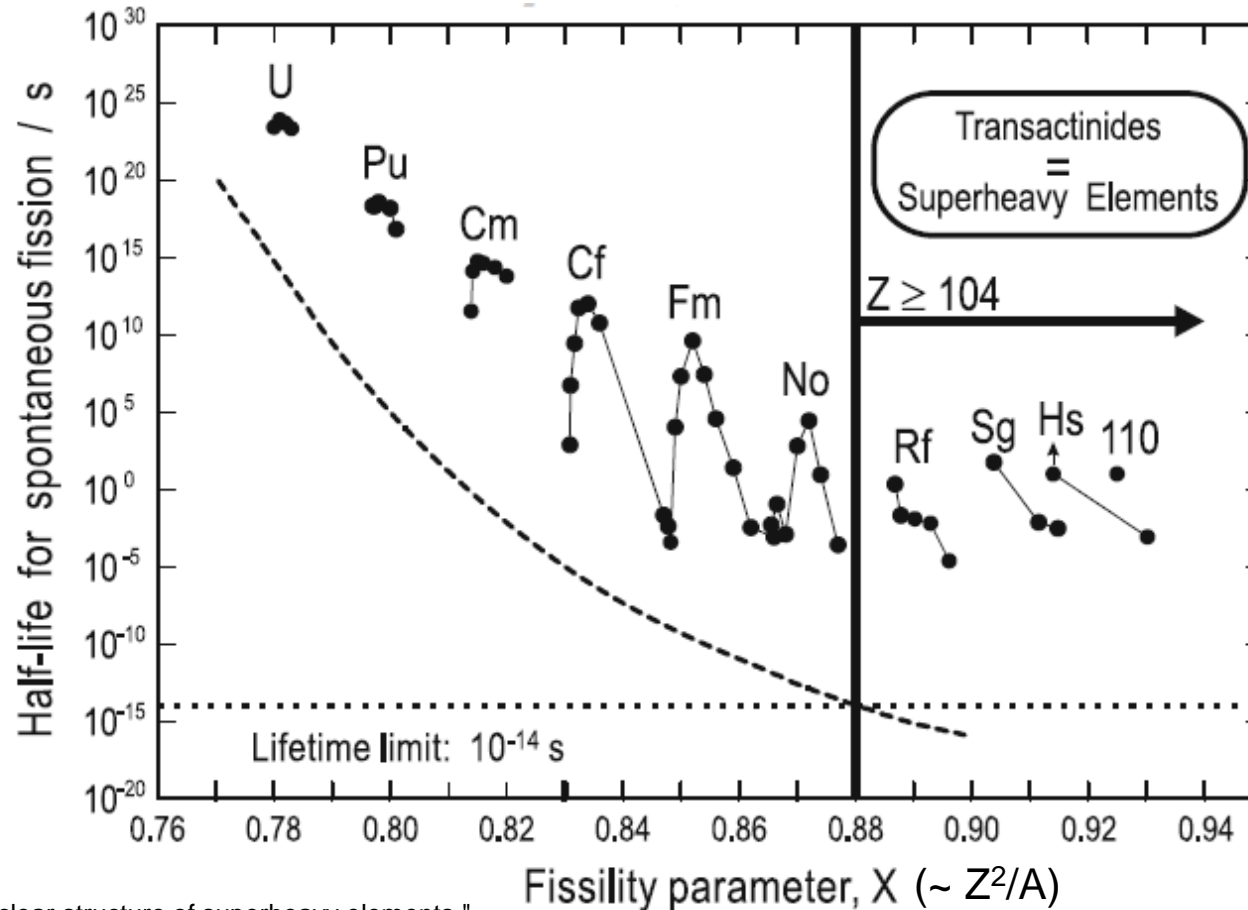
57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md 1955	102 No 1958	103 Lr 1961

Actinides

Stabilized from shell effects

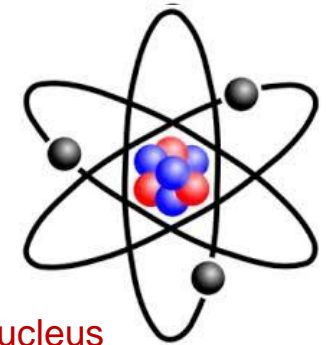
Droplet Model (DM):

$Z \geq 104$ spontaneous fission is faster than formation of the atom shell



Electron shell

- atomic structure
- chemical properties
- defines the element



Nucleus

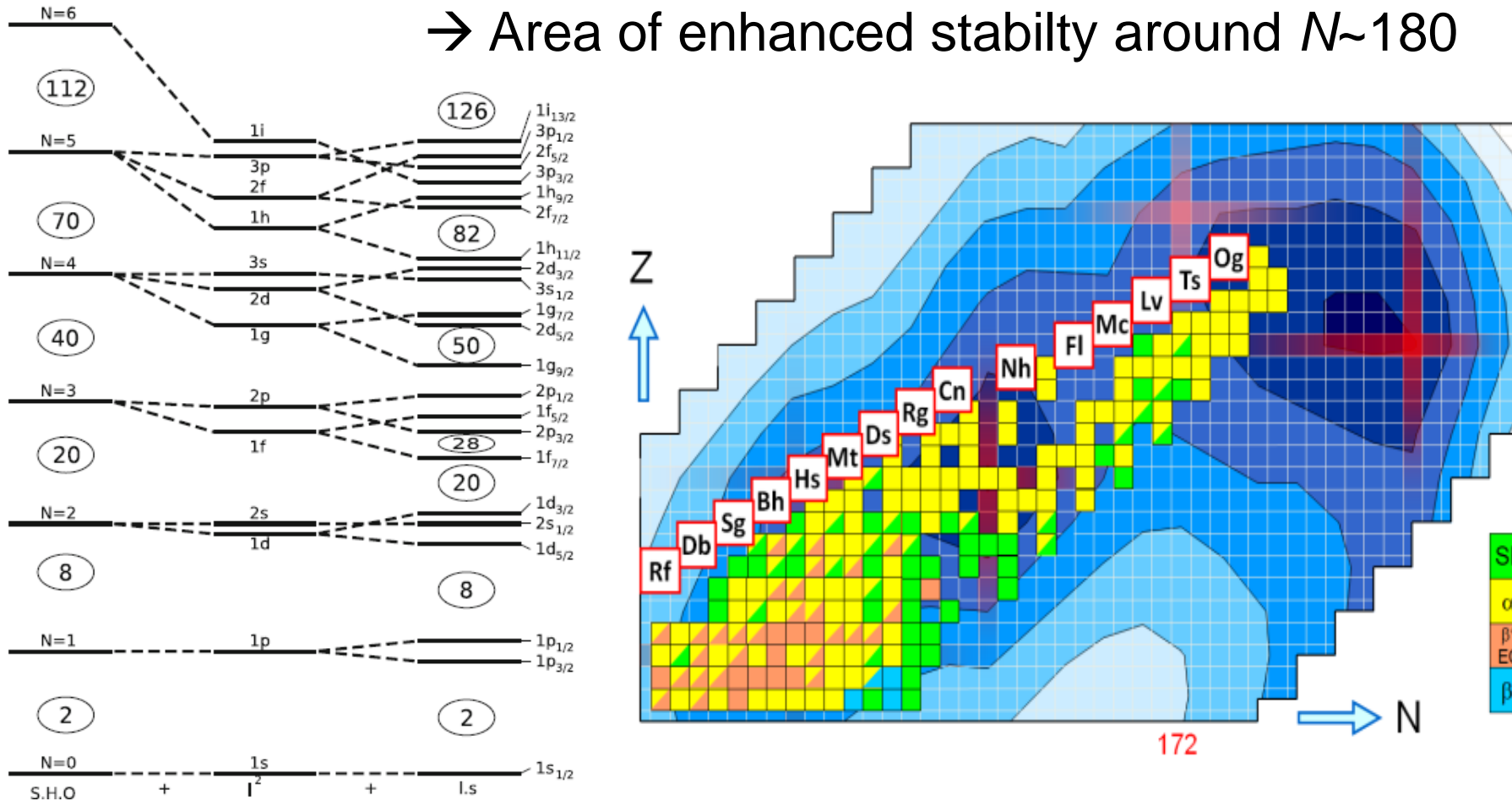
- nuclear structure
- stability of elements

Herzberg, R.-D. "Nuclear structure of superheavy elements."
The chemistry of superheavy elements. Springer, Berlin, Heidelberg, 2014. 83-133.

Stabilized from shell effects

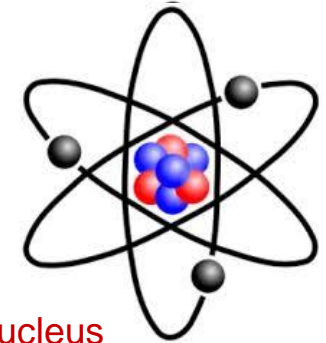
Additional stabilization from nuclear shell effects

→ Area of enhanced stability around $N \sim 180$



Electron shell

- atomic structure
- chemical properties
- defines the element



Nucleus

- nuclear structure
- stability of elements

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn	
Fr	Ra	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Ts	Og	

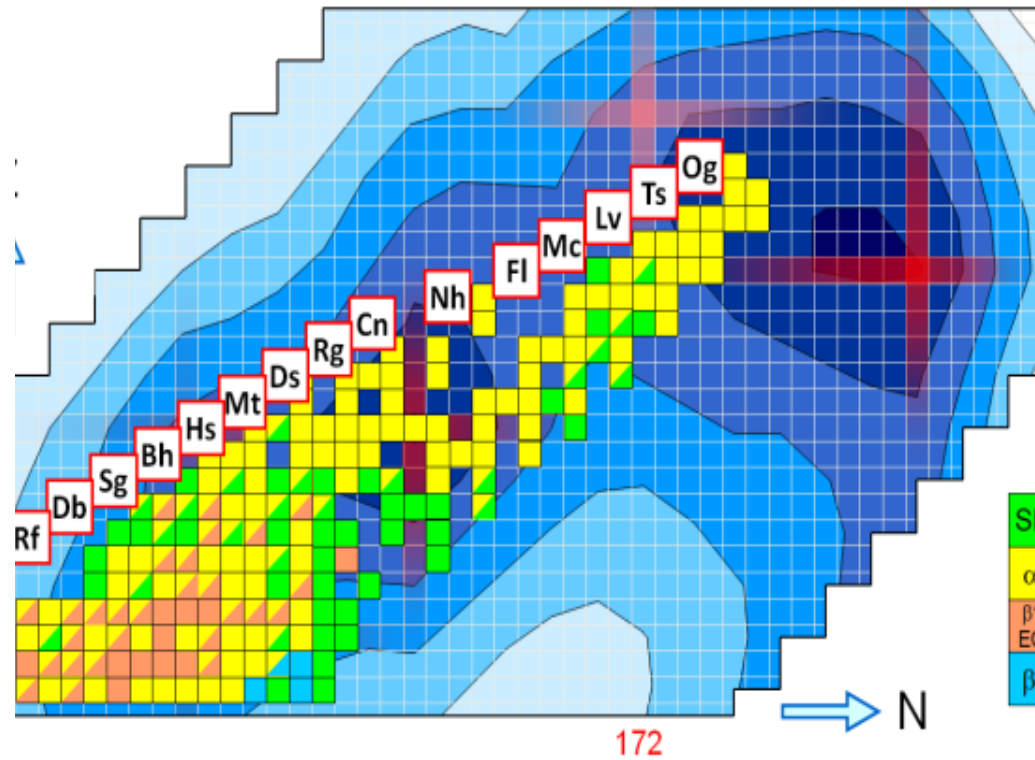
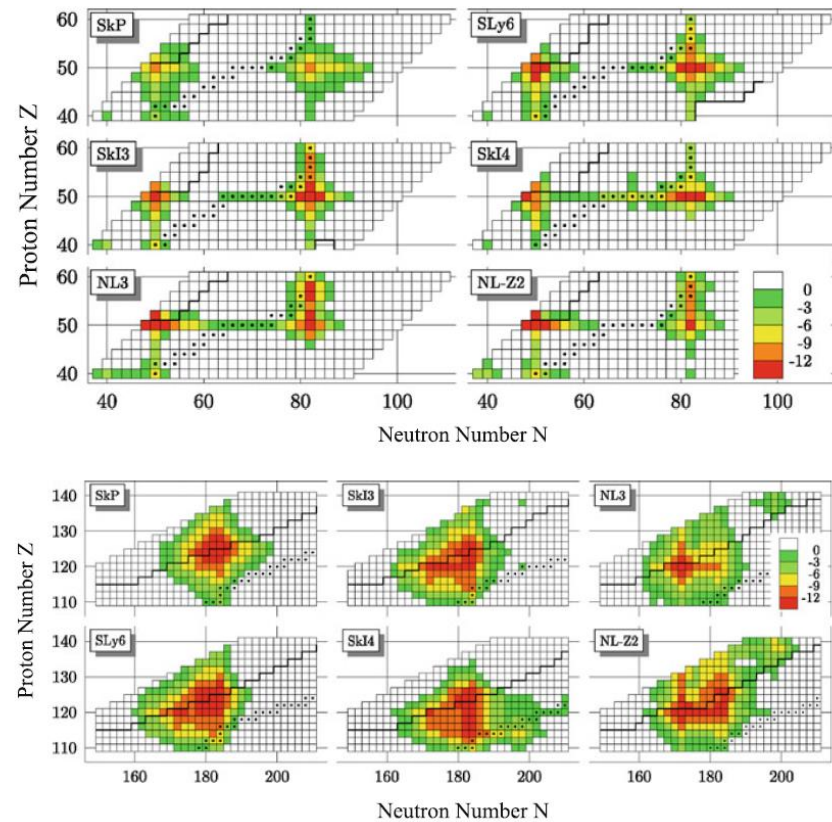
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Herzberg, Rolf-Dietmar. "Nuclear structure of superheavy elements."
The chemistry of superheavy elements. Springer, Berlin, Heidelberg, 2014. 83-133.

Stabilized from shell effects

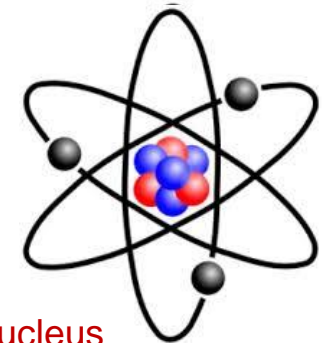
Additional stabilization from nuclear shell effects

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Electron shell

- atomic structure
- chemical properties
- defines the element



Nucleus

- nuclear structure
- stability of elements

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Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba		Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra		Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Ts	Og

La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Bender, M., Nazarewicz, W., Reinhard, P.-G.: Shell stabilization of super- and hyper-heavy nuclei without magic gaps. Phys. Lett. B 515, 42–48 (2001)

Herzberg, Rolf-Dietmar. "Nuclear structure of superheavy elements."

The chemistry of superheavy elements. Springer, Berlin, Heidelberg, 2014. 83-133.

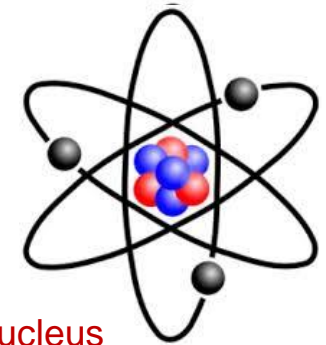
Stabilized from shell effects

Additional stabilization from nuclear shell effects

→ deformation of ground state

Electron shell

- atomic structure
- chemical properties
- defines the element

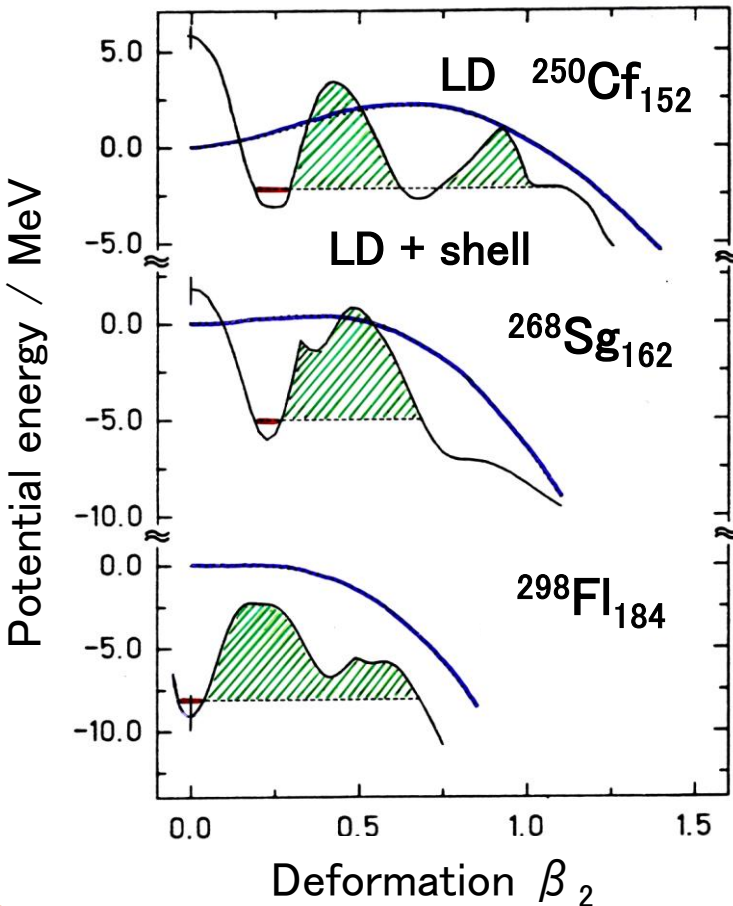
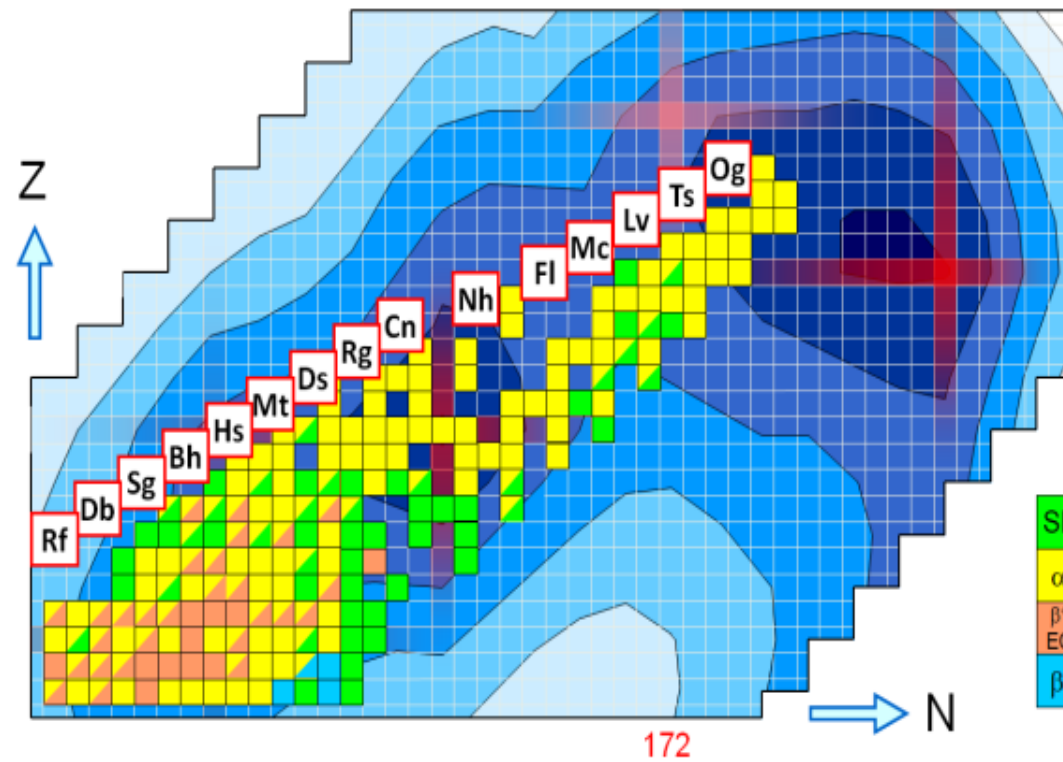


Nucleus

- nuclear structure
- stability of elements

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba		Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra		Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Ts	Og

La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr



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Stabilized from shell effects

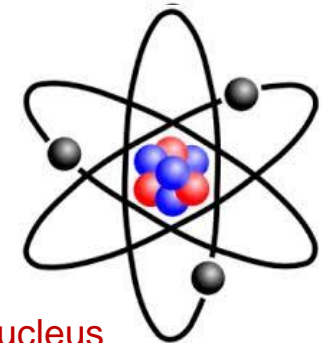
Additional stabilization from nuclear shell effects

→ deformation of ground state

Electron shell

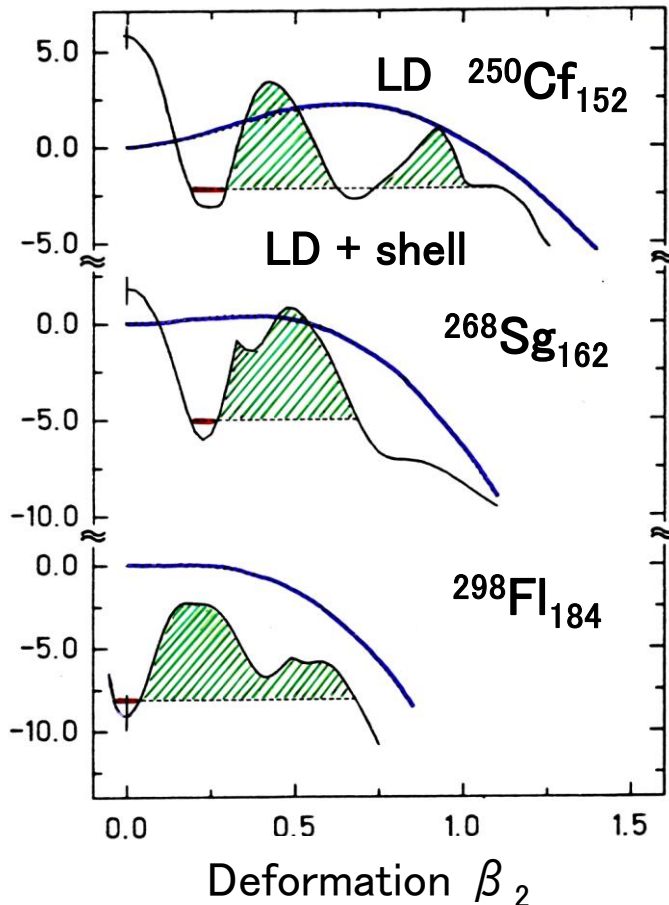
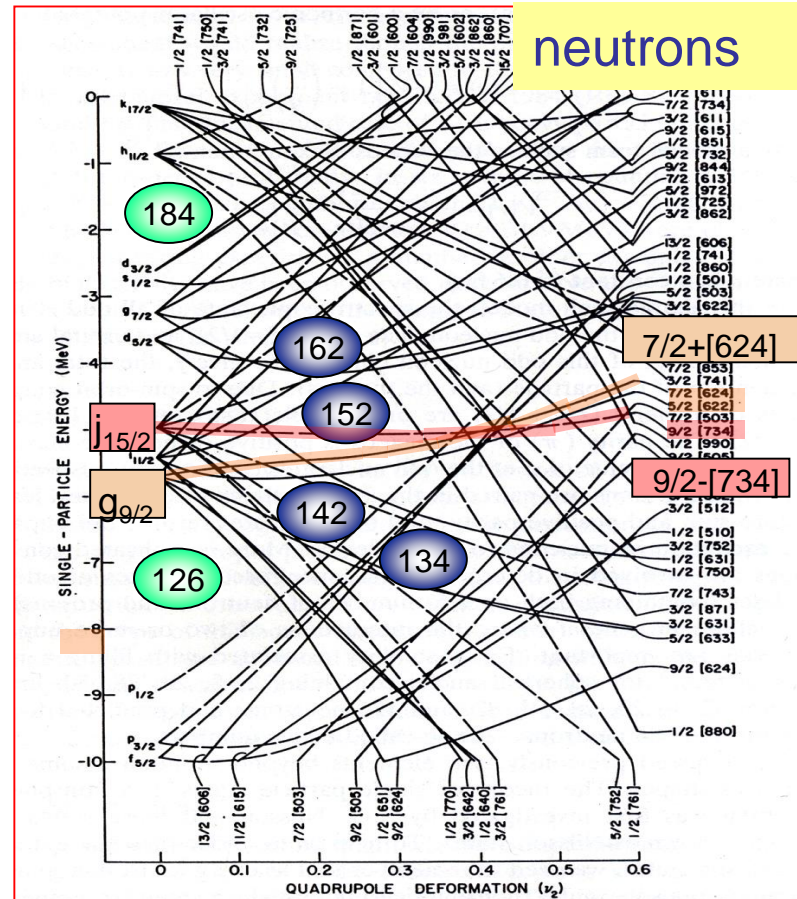
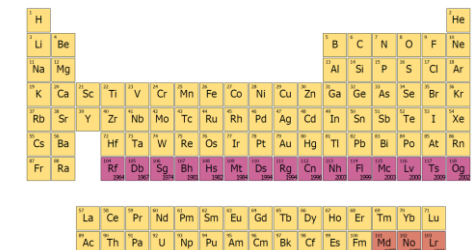
atomic structure
chemical properties

→ defines the element



Nucleus

nuclear structure
stability of elements




Herzberg, Rolf-Dietmar. "Nuclear structure of superheavy elements."
The chemistry of superheavy elements. Springer, Berlin, Heidelberg, 2014. 83-133.

Heaviest Elements

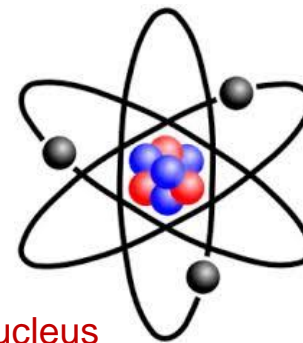
Elements at the limits of nuclear stability

- Why do SHE exist at all ? → **Shell effects**
- How are they best produced in the lab ?
- What is nuclear structure: binding energies, excitations, shape and sizes
- How do their atomic and chemical properties compare to known (lighter) elements ?

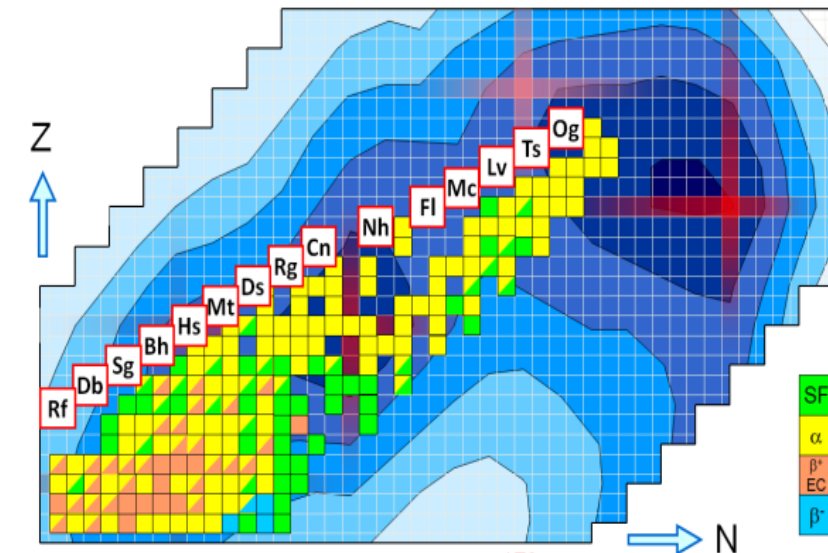


1	H																	2	He																												
3	Li	4	Be											5	B	6	C	7	N	8	O	9	F	10	Ne																						
11	Na	12	Mg											13	Al	14	Si	15	P	16	S	17	Cl	18	Ar																						
19	K	20	Ca	21	Sc	22	Ti	23	V	24	Cr	25	Mn	26	Fe	27	Co	28	Ni	29	Cu	30	Zn	31	Ga	32	Ge	33	As	34	Se	35	Br	36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

Electron shell
 atomic structure
 chemical properties
 → defines the element



Nucleus
 nuclear structure
 stability of elements

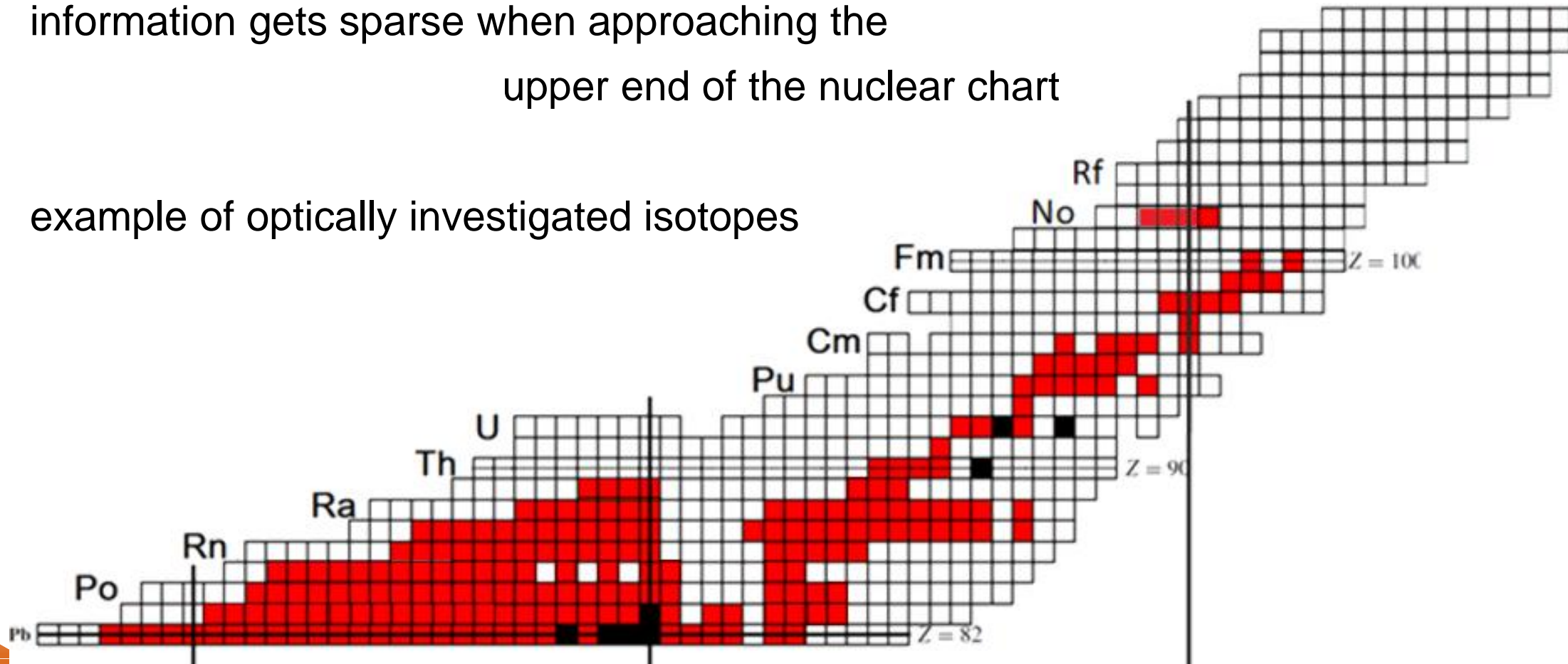


Accessing heavy elements

How to access the heaviest elements?

information gets sparse when approaching the
upper end of the nuclear chart

example of optically investigated isotopes



Accessing heavy elements

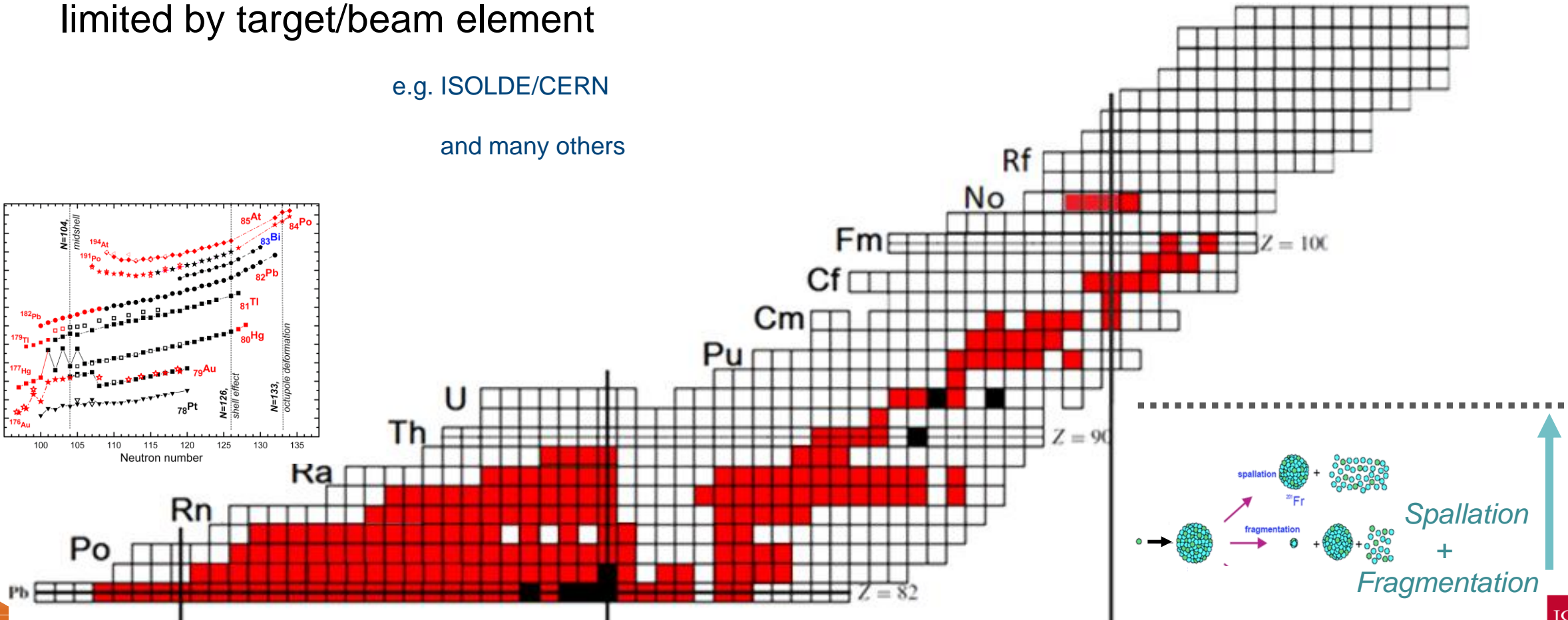
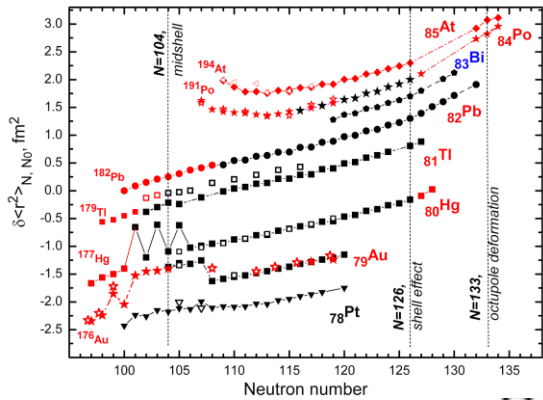
Spallation and Fragmentation

large isotope range of light elements

limited by target/beam element

e.g. ISOLDE/CERN

and many others



Accessing heavy elements

Spallation and Fragmentation

large isotope range of light elements

limited by target element

Breeding in a nuclear reactor

successive



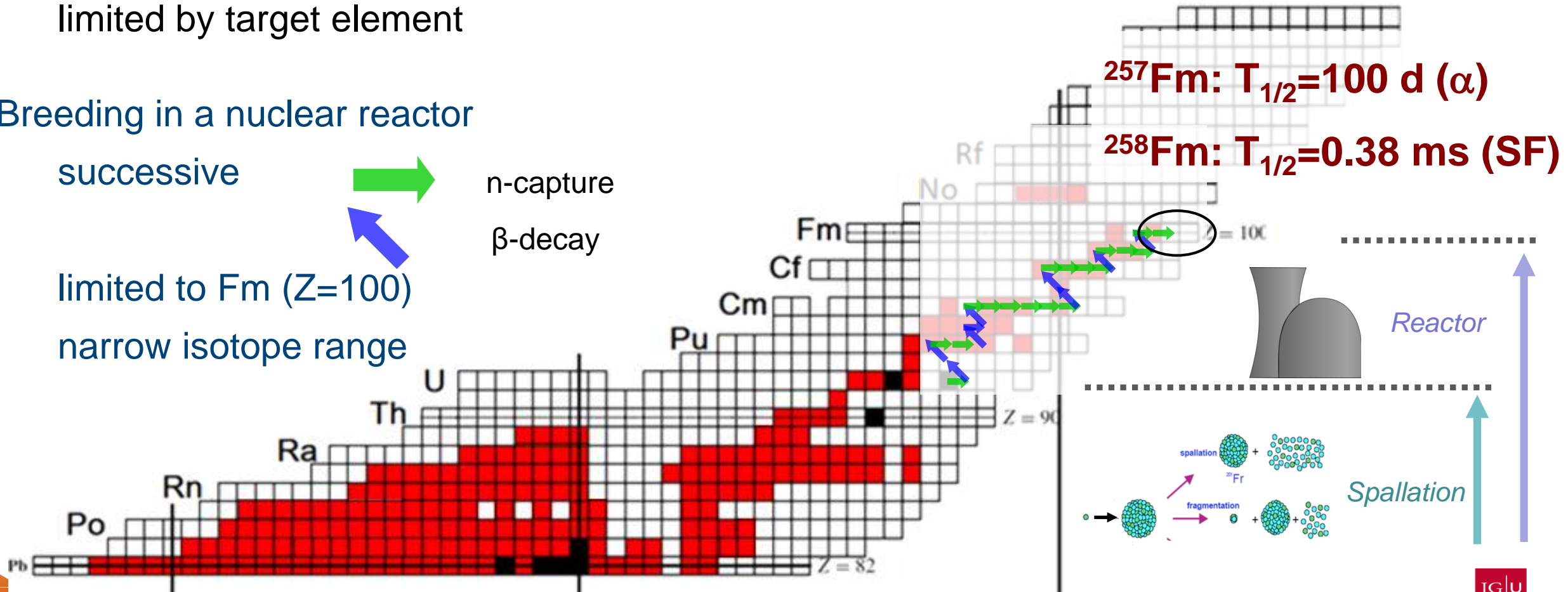
n-capture



β -decay

limited to Fm (Z=100)

narrow isotope range



Accessing heavy elements

Fusion evaporation reactions

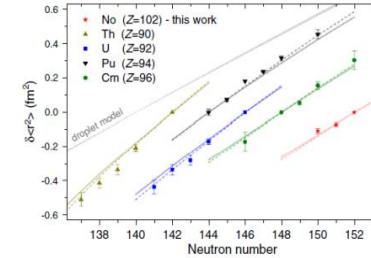
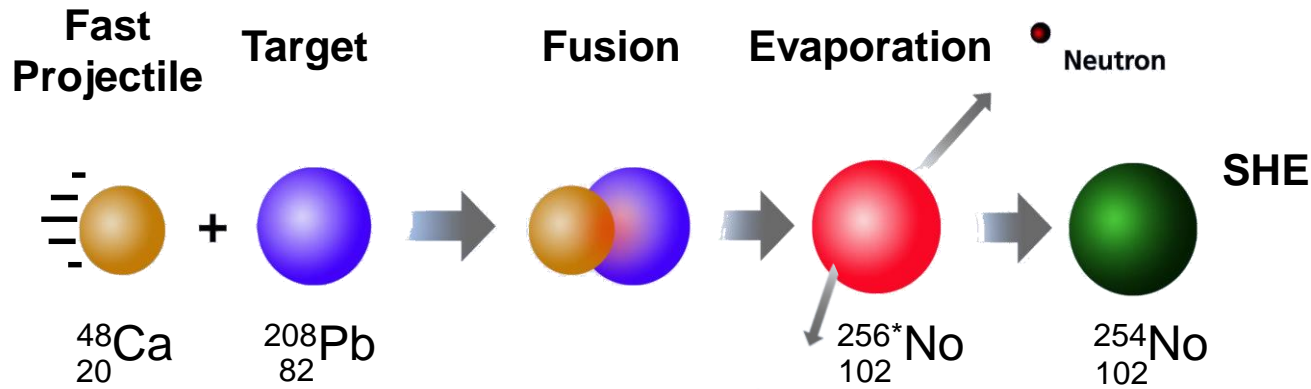
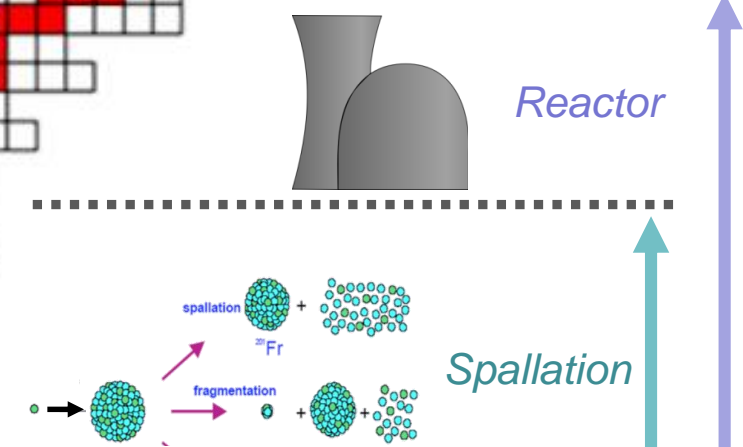
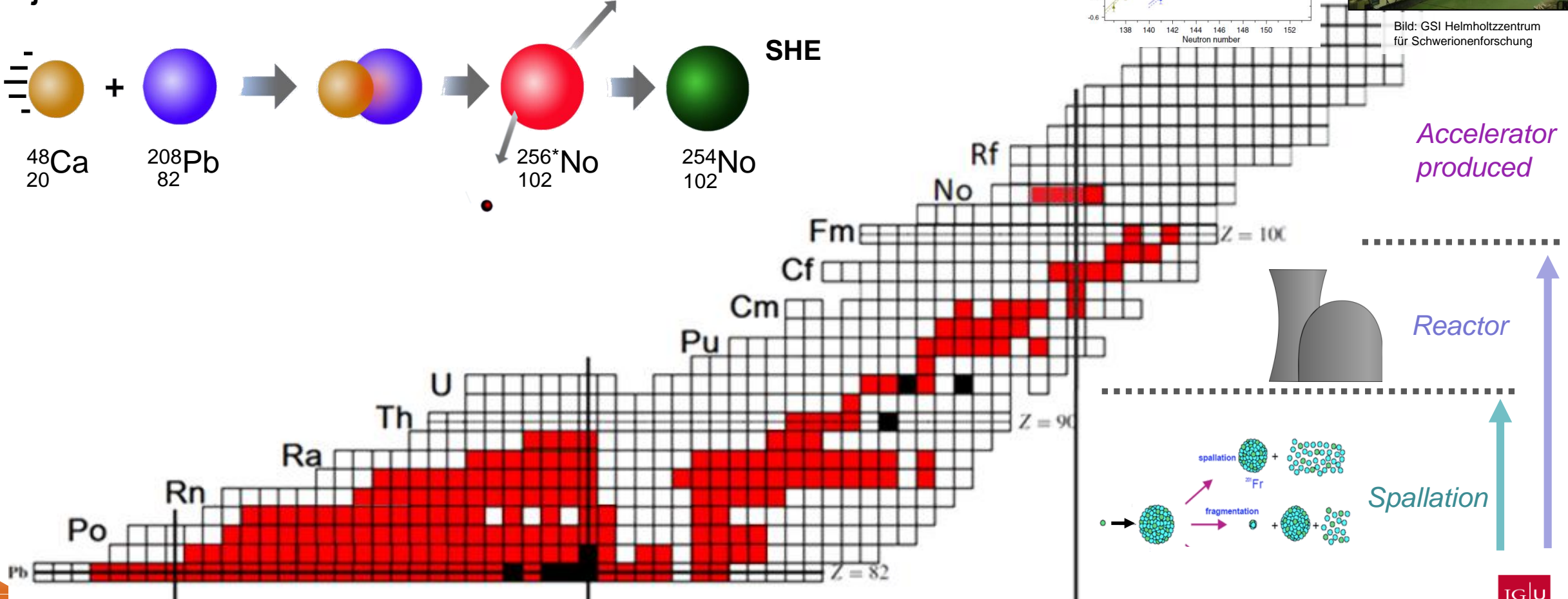


Bild: GSI Helmholtzzentrum für Schwerionenforschung



Accessing heavy elements

Fusion evaporation reactions

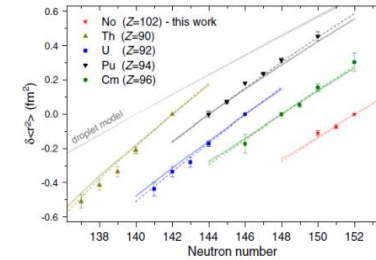
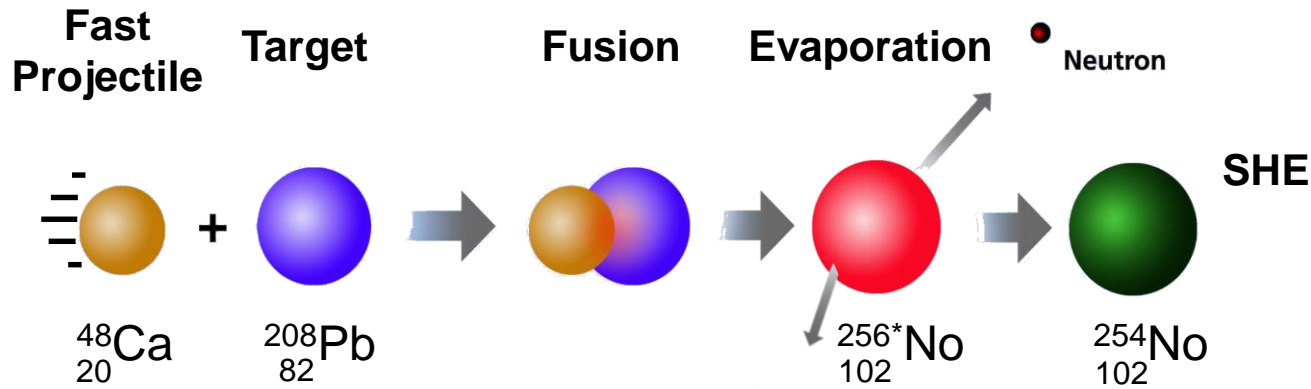
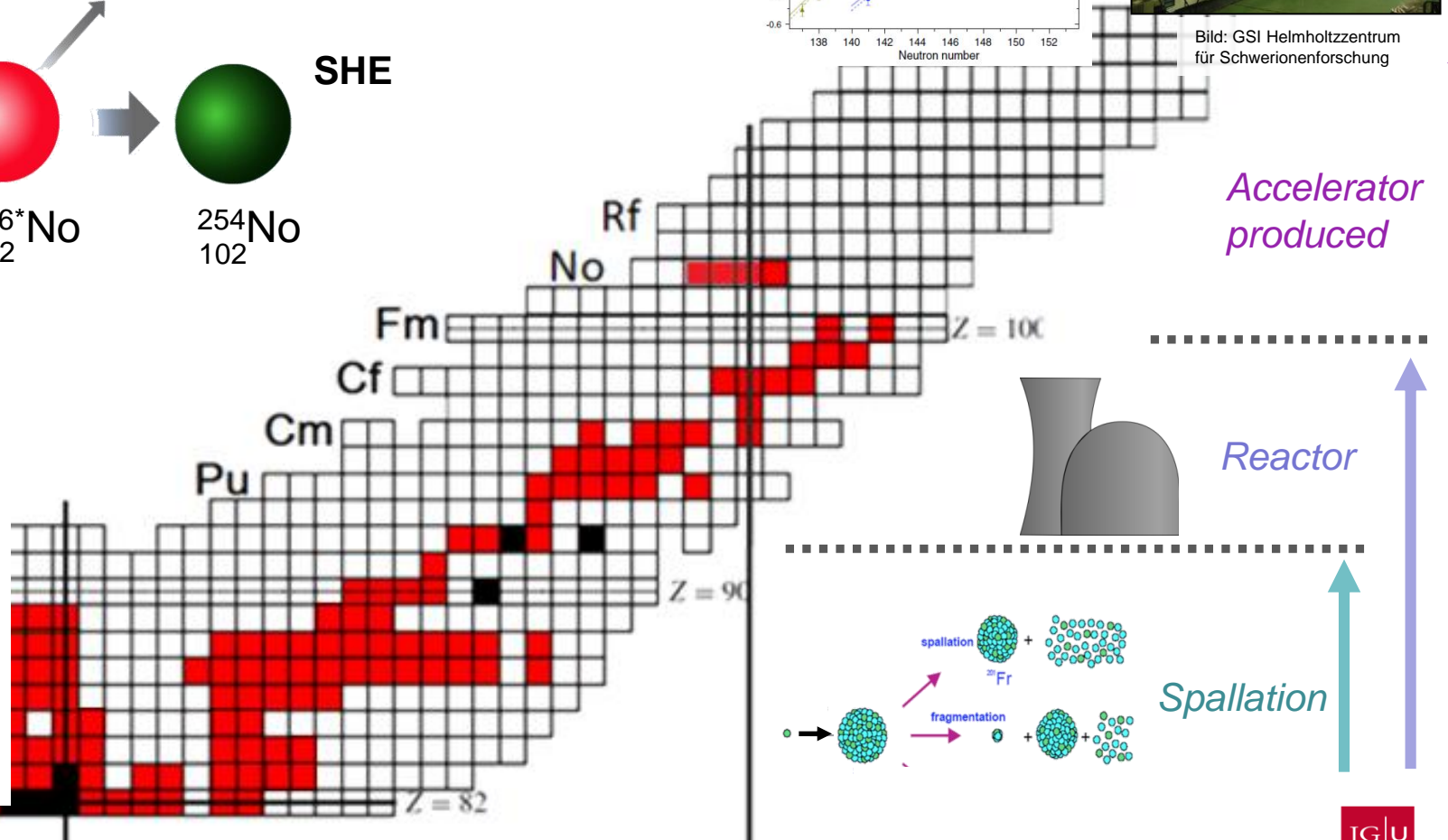
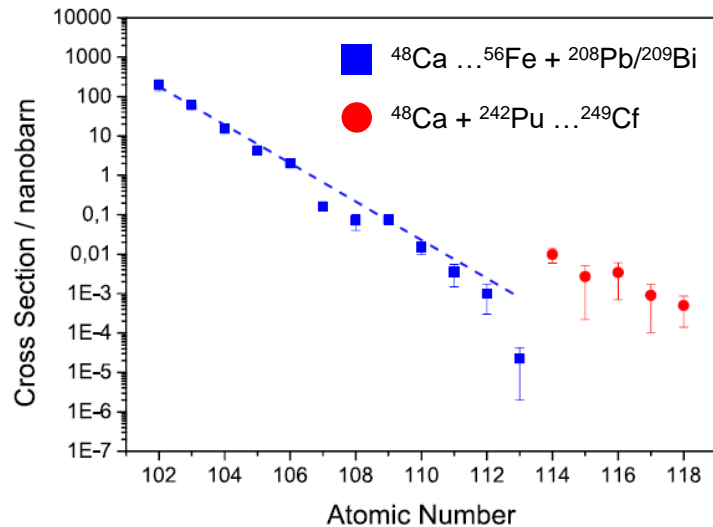
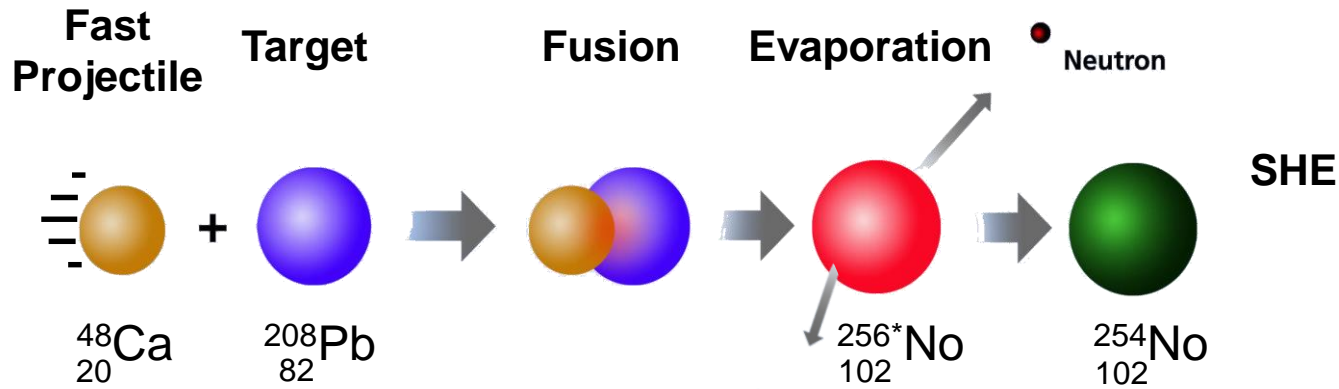


Bild: GSI Helmholtzzentrum für Schwerionenforschung



Accessing heavy elements

Fusion evaporation reactions



Also: Multinucleon transfer

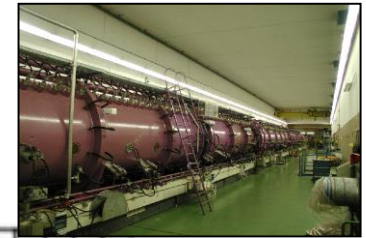
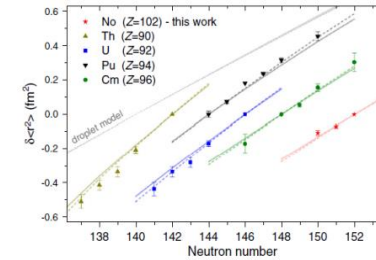
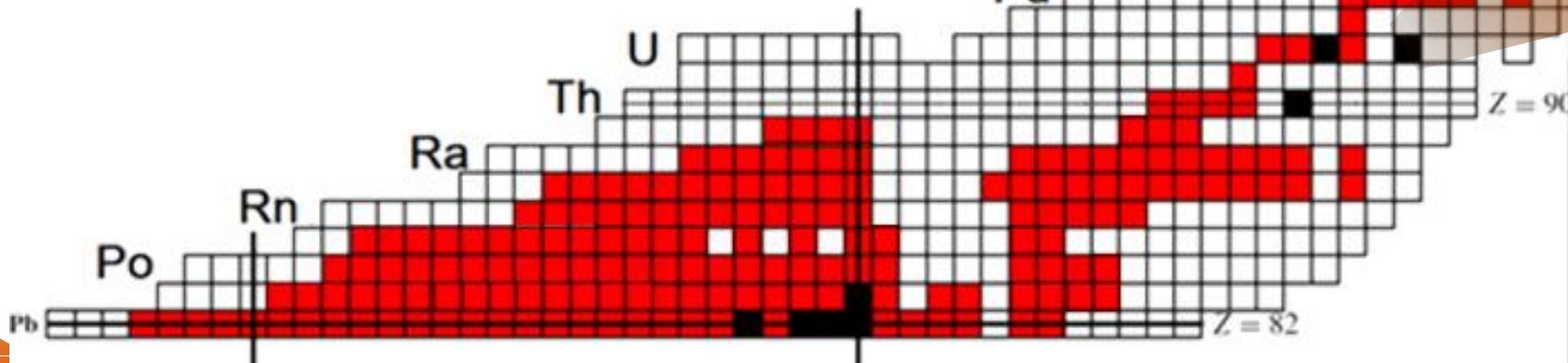
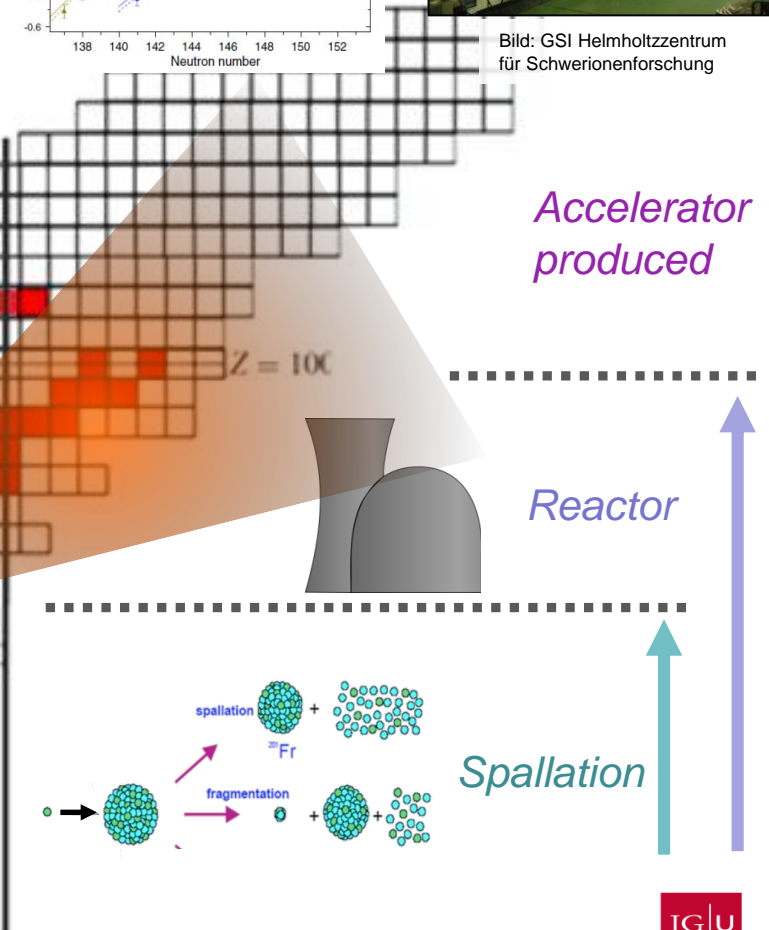


Bild: GSI Helmholtzzentrum für Schwerionenforschung



Worden E.F., et al. *J. Opt. Soc. Am.* 64.1 (1974): 77. -- P. Campbell et al., *Prog. Part Nucl. Phxs* 86 (2016) 127

Accessing heavy elements

Fusion evaporation reactions

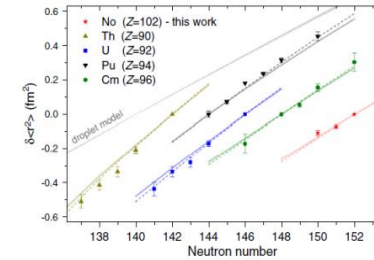
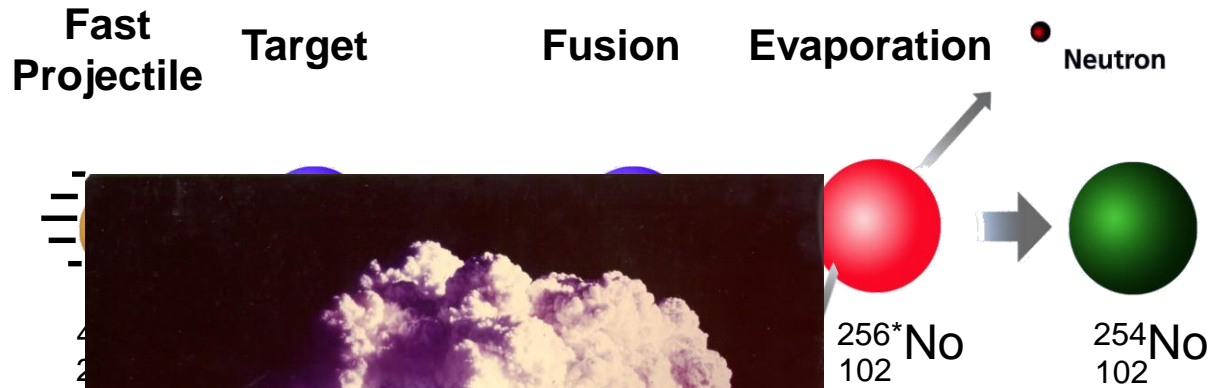
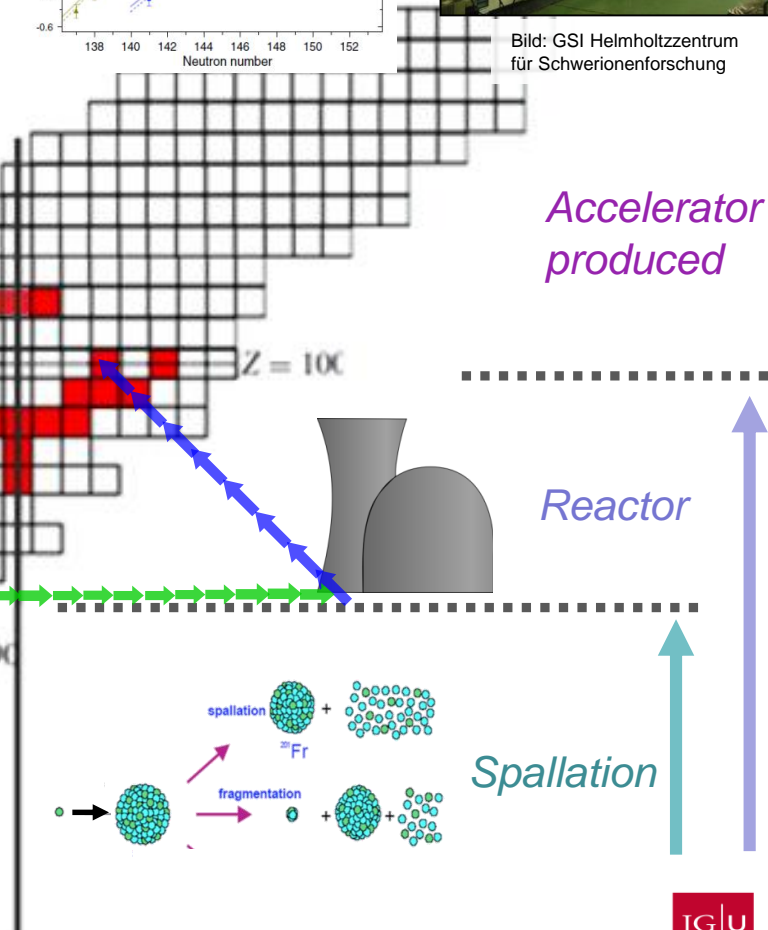
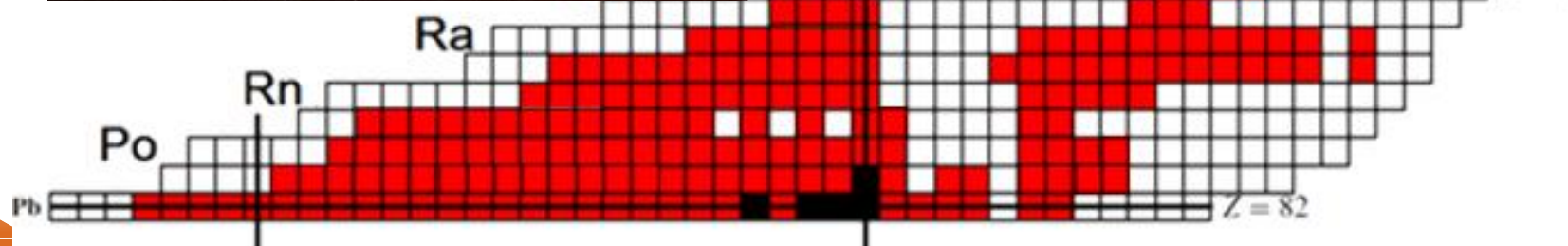


Bild: GSI Helmholtzzentrum für Schwerionenforschung



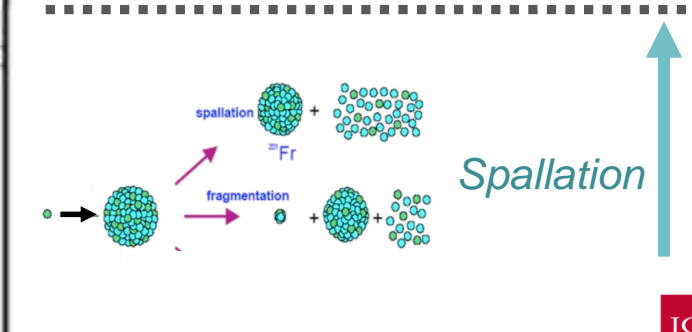
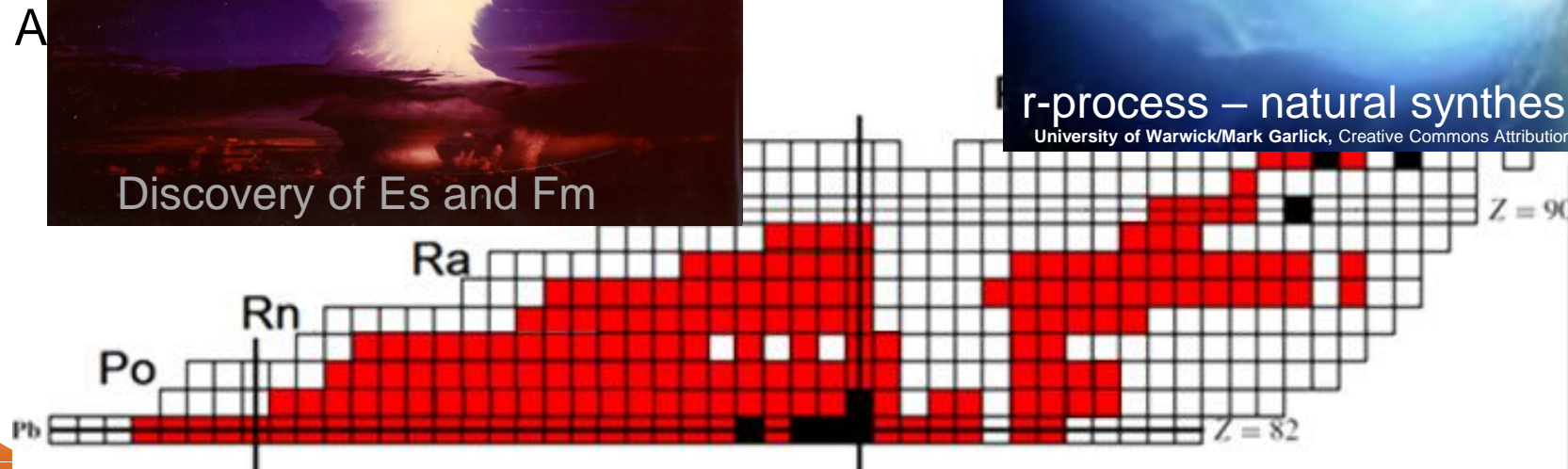
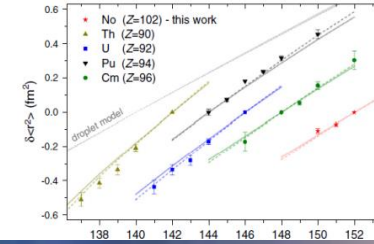
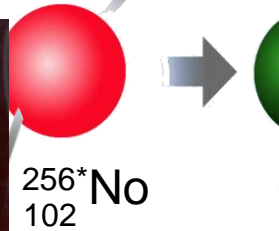
The New Elements Einsteinium and Fermium, Atomic Numbers 99 and 100, Ghiorso, A. et al, June 9, 1955.

S. Raeder – 07.10.2021 – Lecture 1 - Joliot-Curie School – Isle d'Oleron

Accessing heavy elements

Fusion evaporation reactions

Fast Projectile Target Fusion Evaporation Neutron



Accelerator produced

Reactor

The New Elements Einsteinium and Fermium, Atomic Numbers 99 and 100, Ghiorso, A. et al, June 9, 1955.

Heavy Elements - The far end of the periodic table

1 H																	2 He														
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne														
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37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe														
55 Cs	56 Ba	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md 1955	102 No 1958	103 Lr 1961	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og

Heaviest elements: Small production rates
 ⇒ On average, at any given time, at most one atom of the element under investigation exists!

Ava

89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md 1955	102 No 1958	103 Lr 1961
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tons

mg / µg / pg 50 atoms min⁻¹

~1 h

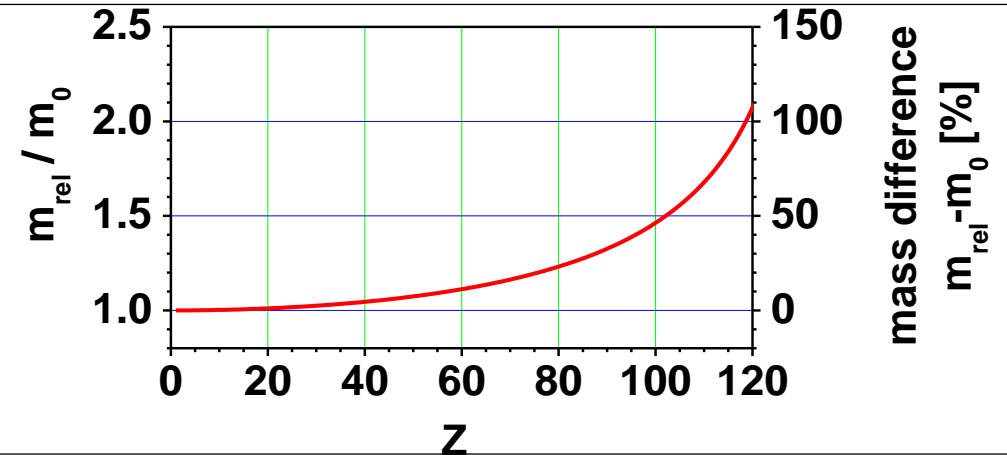
T_{1/2}

Influence of Relativity on Atomic and Chemical Properties

Relativistic speed of electrons near the atomic nucleus → mass increase

Scaling: $\sim Z^2$

⇒ **most pronounced in SHE**



Relativistic effects

1) Direct effect:

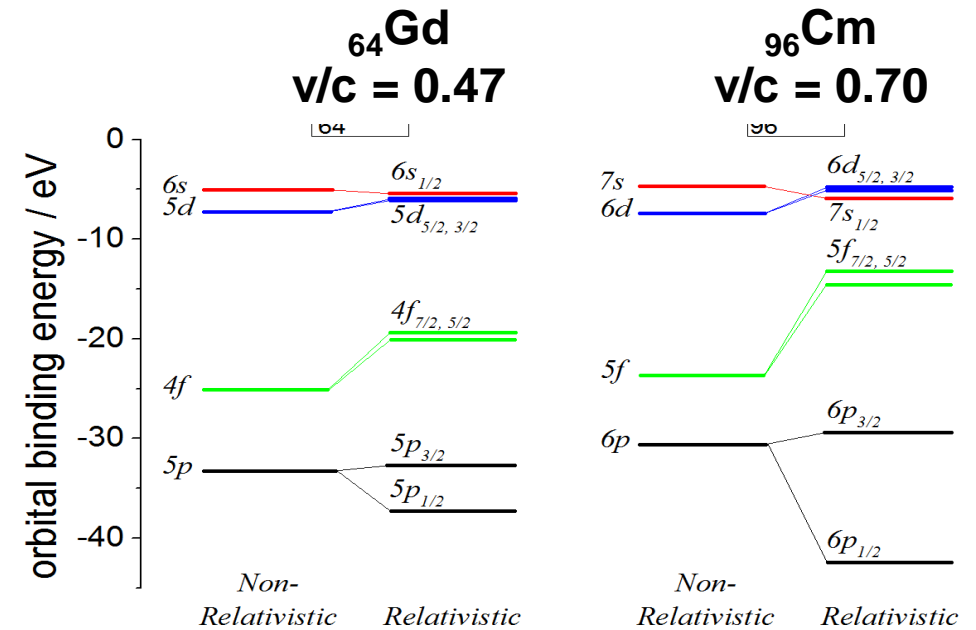
s_{1/2}; **p_{1/2}**: ⇒ stabilized / contracted

2) Indirect effect:

p_{3/2}; **d**; **f**: better shielded from core
⇒ destabilized / expanded

3) Spin-orbit splitting:

p → **p_{1/2}**/**p_{3/2}**; **d** → **d_{3/2}**; **d_{5/2}**; ..
jj-coupling lifts energy. degeneracy



J.P. Desclaux, At. Data Nucl. Data Tab. 12 (1973) 311

Chemistry with limited number of atoms

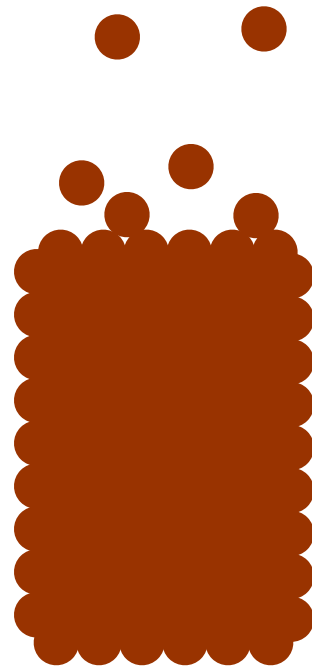
Classical chemistry



Atom-at-a-time

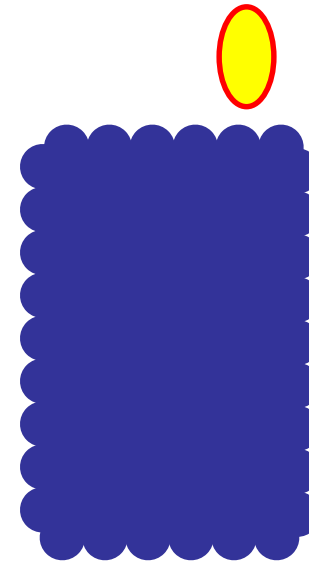
Macroamount
 10^{20} atoms

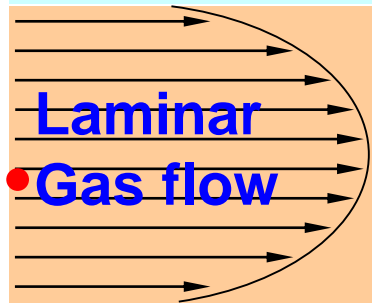
Microamount
1 atom



Gas

Solid





Chromatography column

Strength of interaction is expressed by **adsorption enthalpie** ΔH_{ads} of A on B

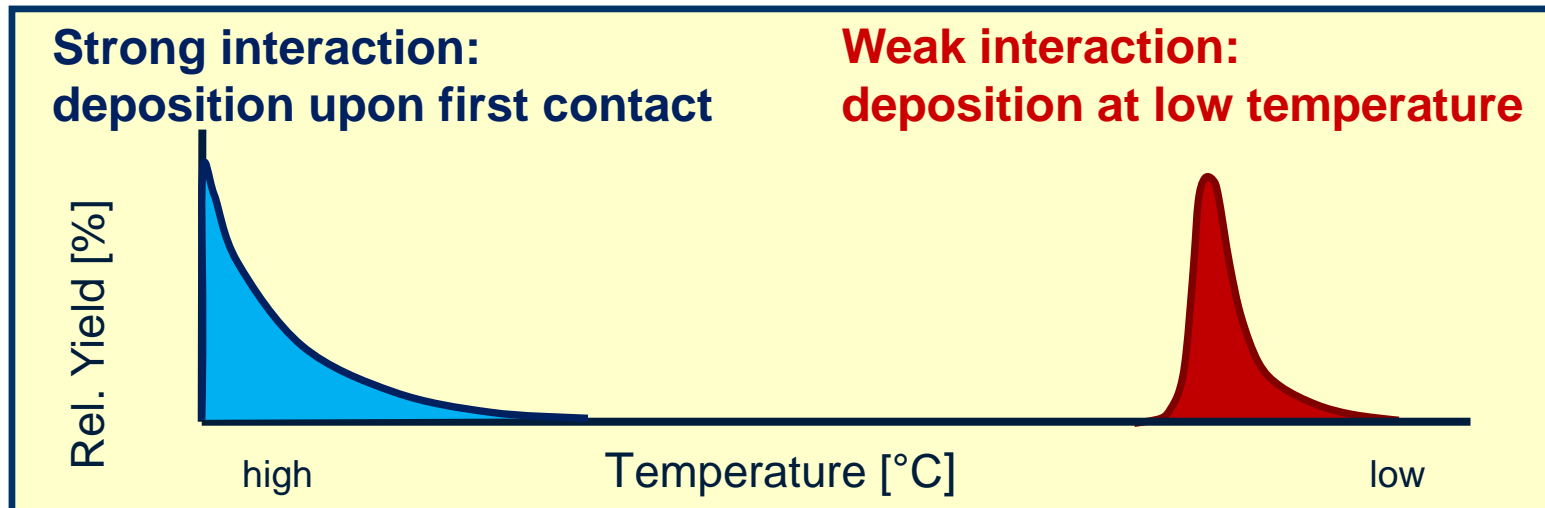
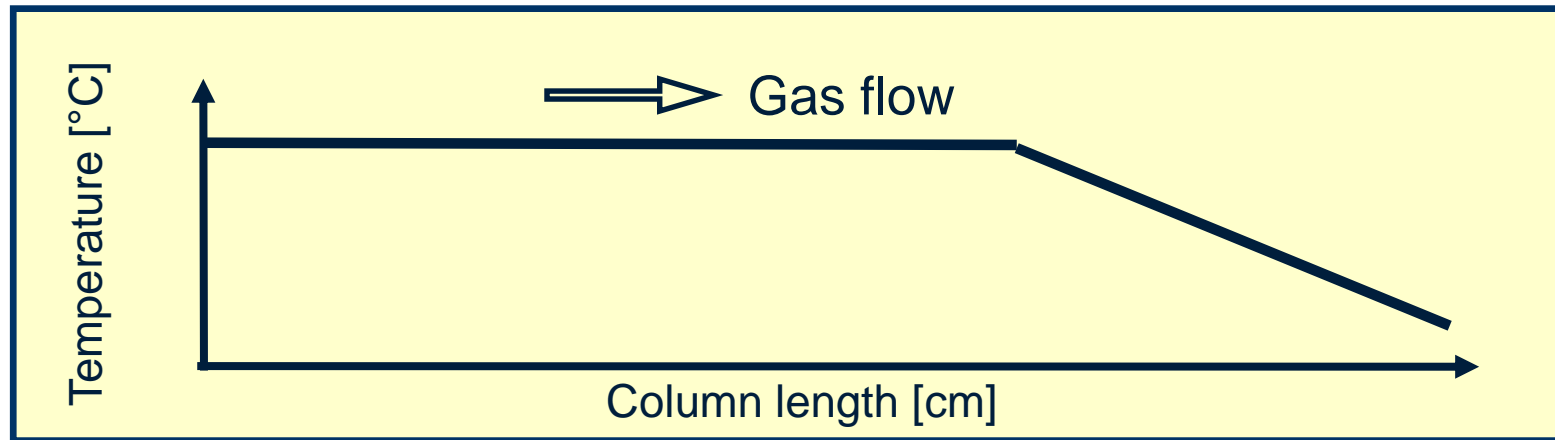
Mean time spent in adsorbed state:

Frenkel-Equation

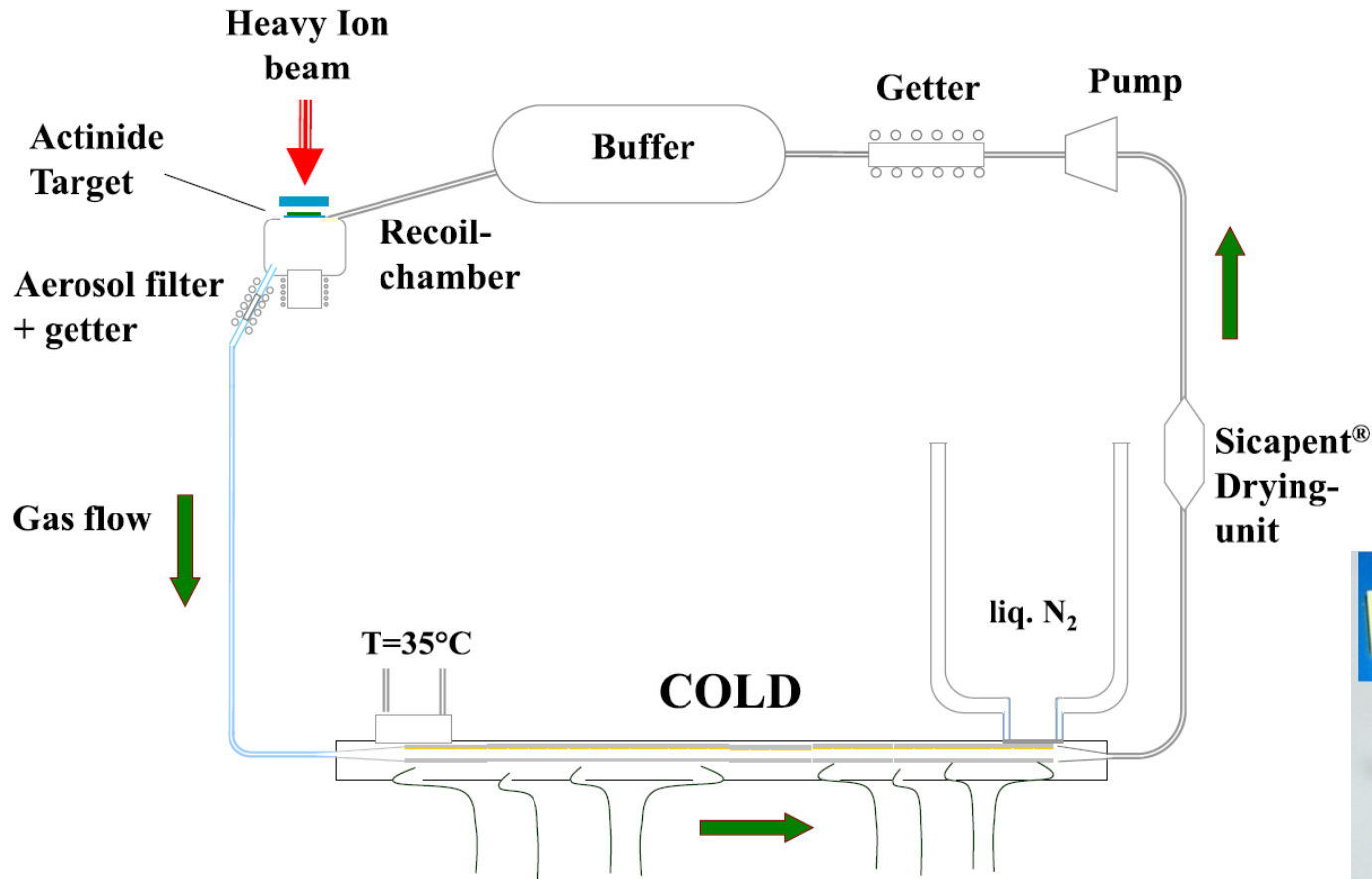
$$\tau = \tau_0 \cdot e^{-\frac{\Delta H_{ads}}{R \cdot T}}$$

R: Gas constant; T: column temperature; τ_0 =characteristic time of oscillation of the column material; e.g., SiO₂: 2.2·10⁻¹³ s

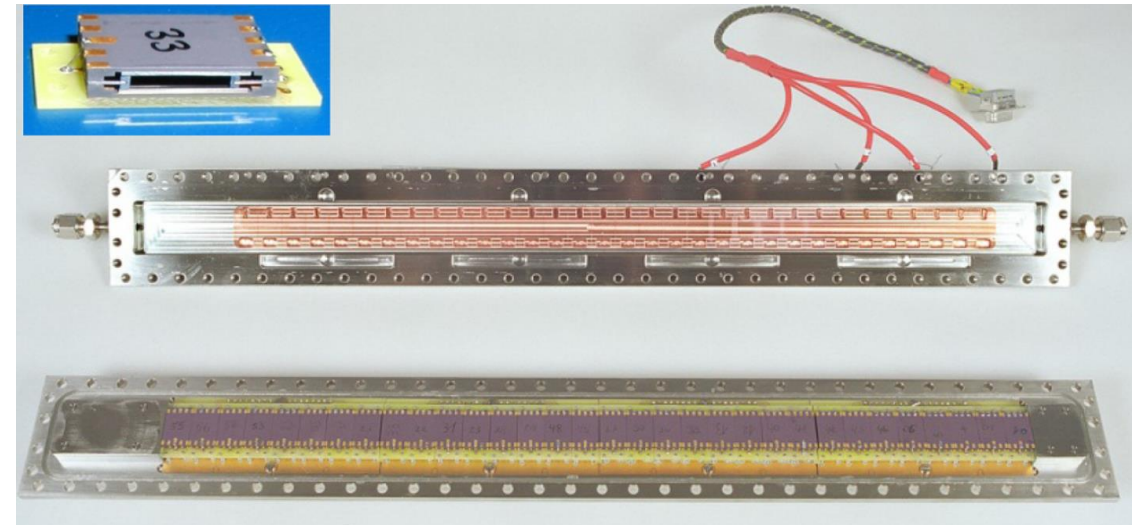
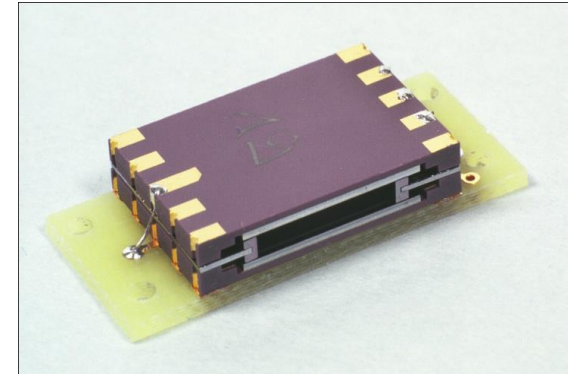
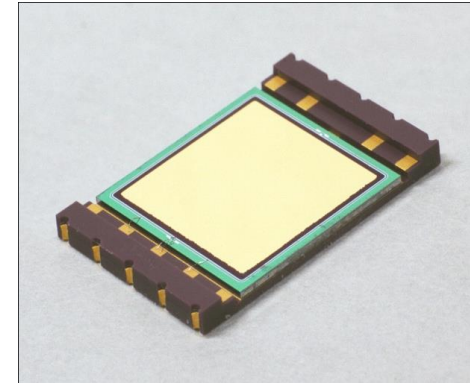
Gas phase chromatography



Experimental setup for Cn (Z=112) chemistry (PSI,FLNR)



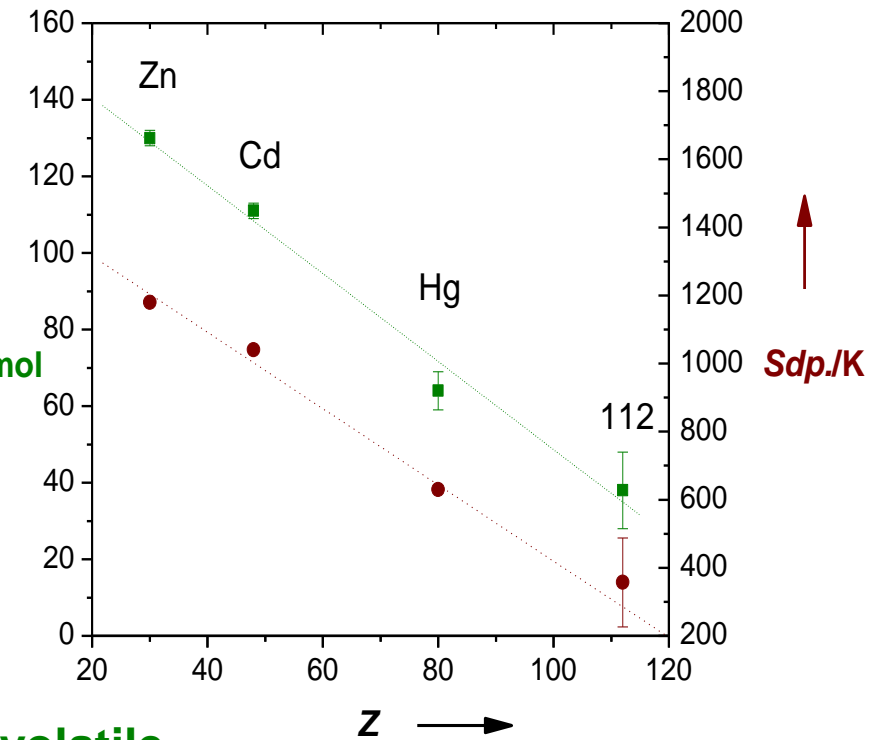
Cryo-On-Line Detector



Volatility of Cn (Z=112)

Simulations

less volatile



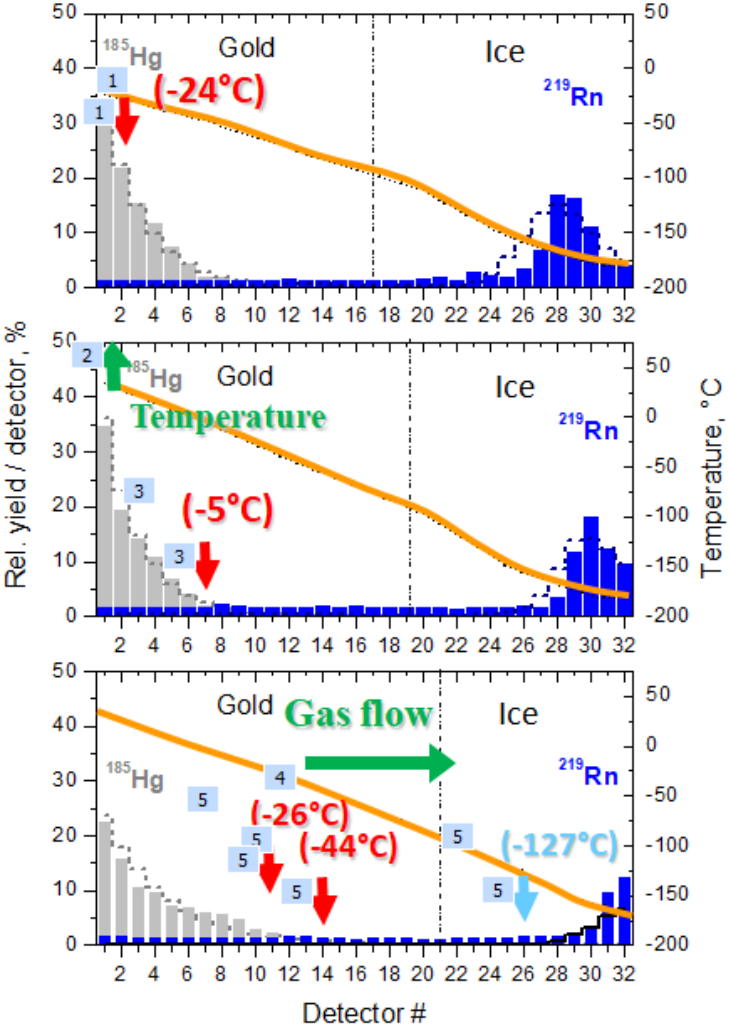
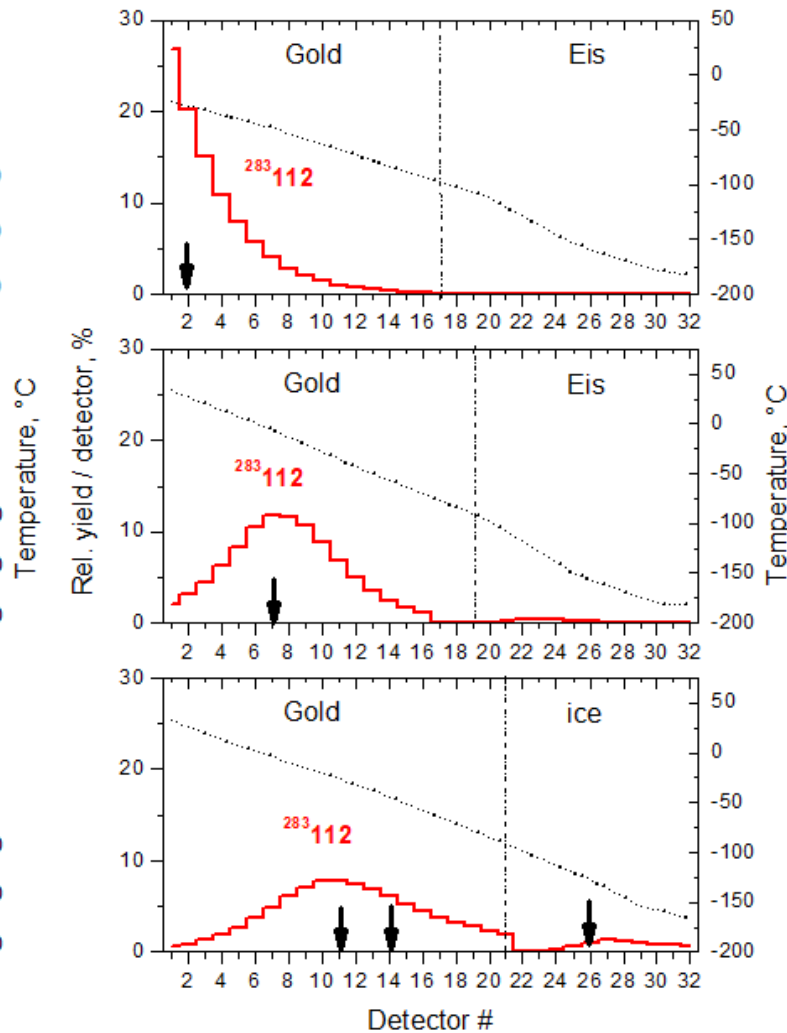
volatile

Cn keeps the trend in group 112

→ 5 Atoms required

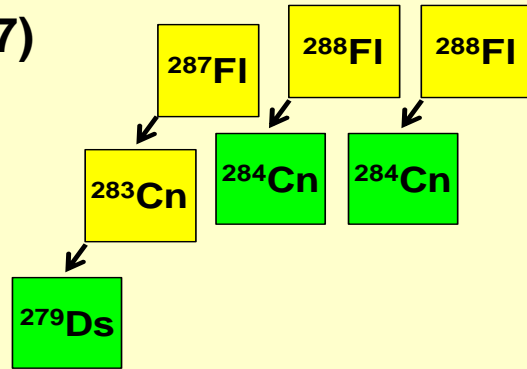
6

6



Fl (Z=114) chemistry experiments

PSI/Dubna (2007)
35 days



Conclusion: physisorption bond with Au

R. Eichler et al., Radiochim. Acta 98 (2010) 133

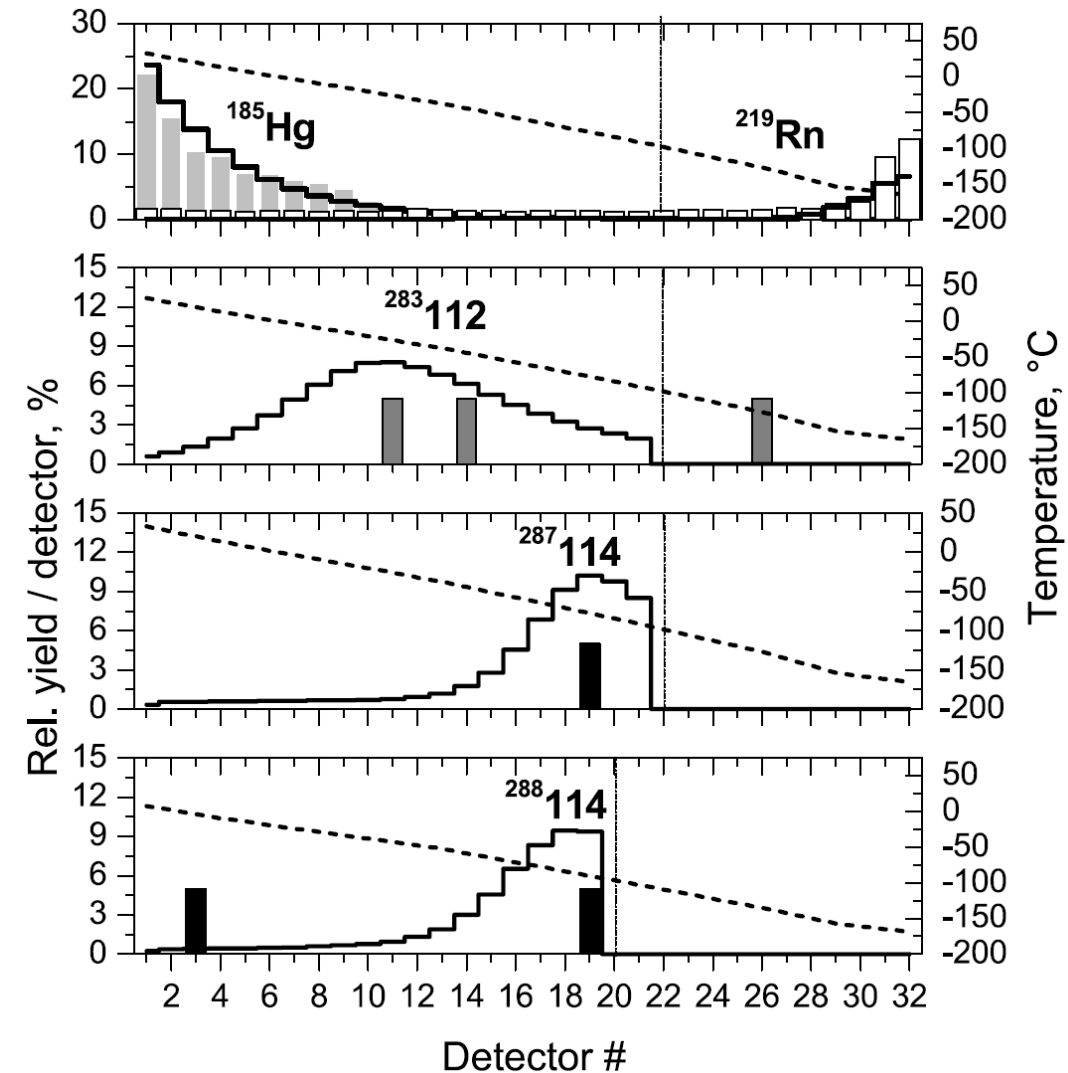
$$-\Delta H_{\text{ads}}(\text{Au})$$

Theory

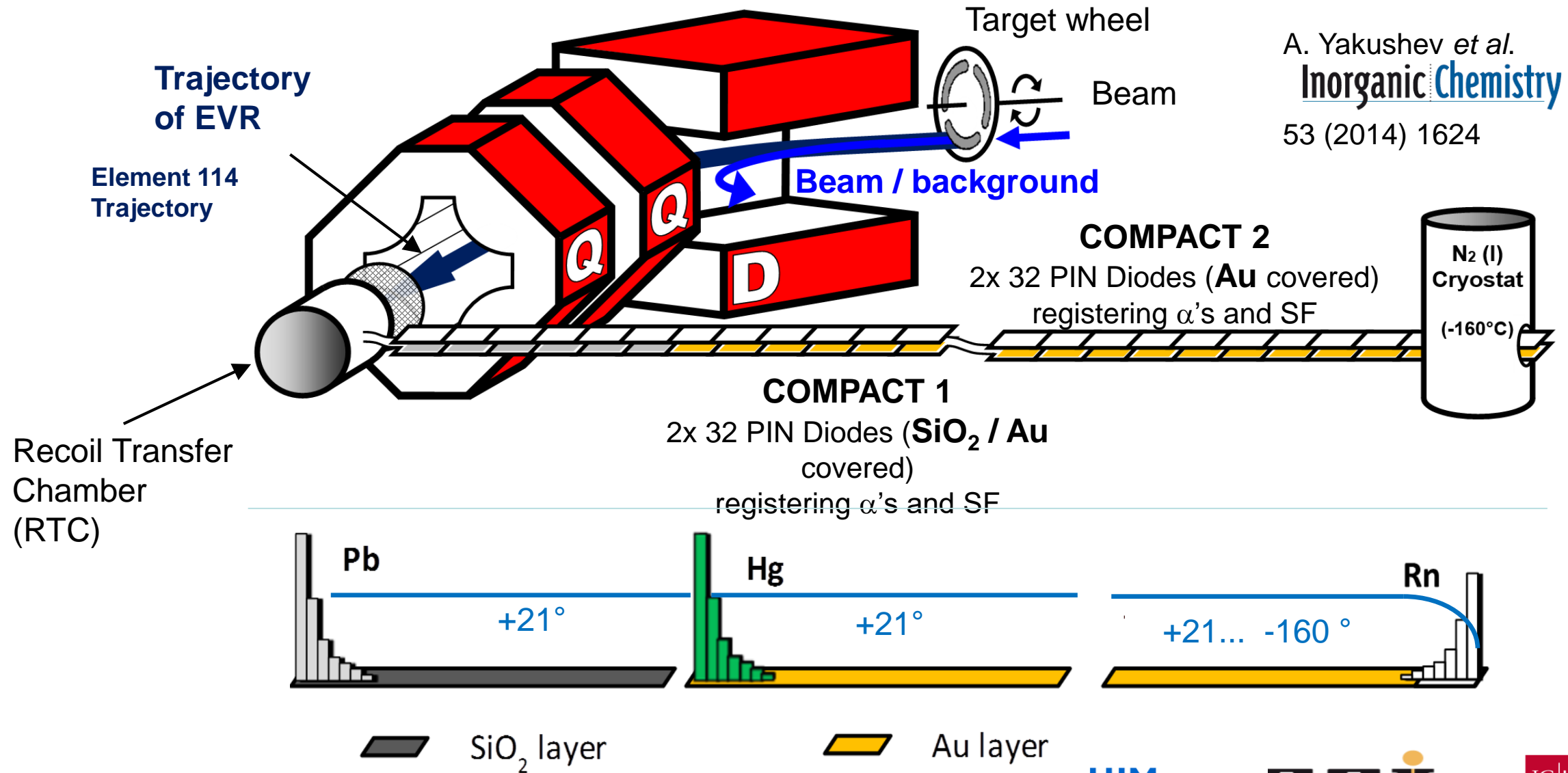
Experiment

Pb > **114** \cong Hg > **112**

Hg > **112** > **114**

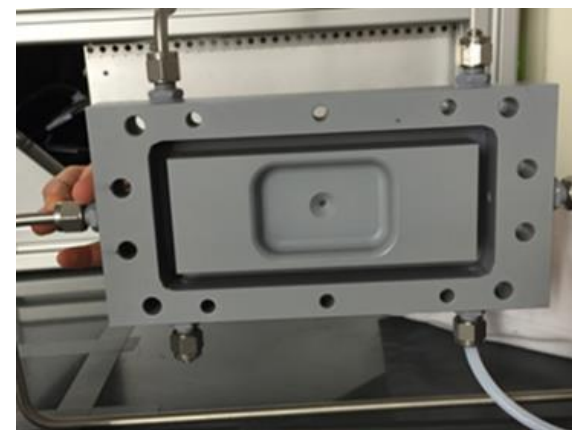
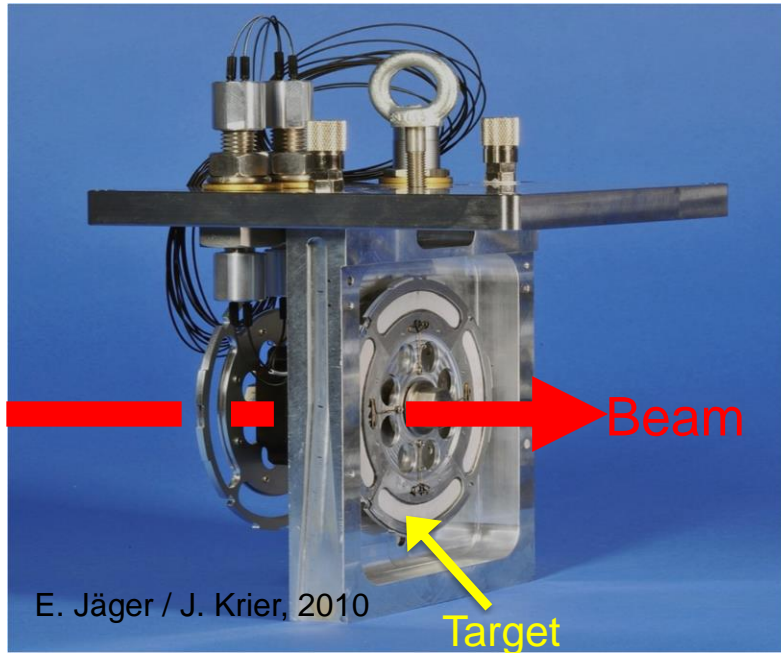


TransActinide Separator and Chemistry Apparatus – TASCA



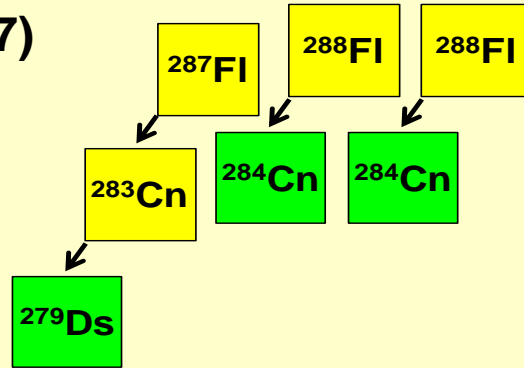
A. Yakushev *et al.*
Inorganic Chemistry
53 (2014) 1624

TransActinide Separator and Chemistry Apparatus – TASCA



Fl (Z=114) decay chains from chemistry experiments

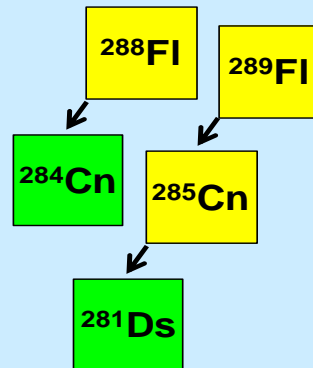
PSI/Dubna (2007)
35 days



Conclusion: physisorption bond with Au

R. Eichler *et al.*, RCA (2010)

GSI (2009)
28 days



Conclusion: metallic bond with Au

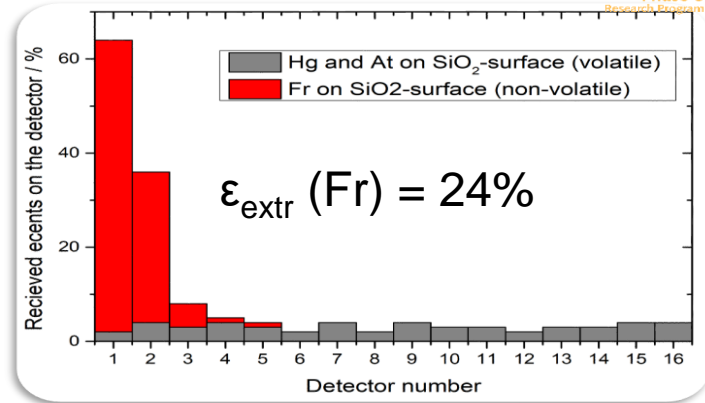
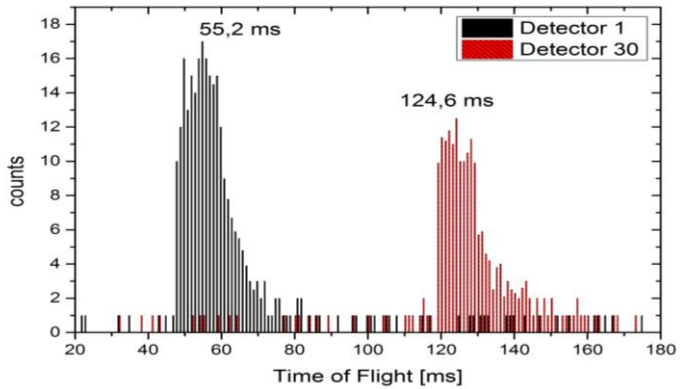
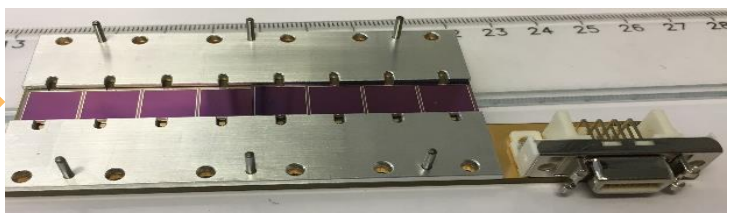
A. Yakushev *et al.*, Inorg. Chem. (2014)

Beyond Mc: Preparations for chemistry with ^{116}Lv and ^{117}Tl

First test experiments: Coupling of BGC with COMPACT and miniCOMPACT



He
50 mbar



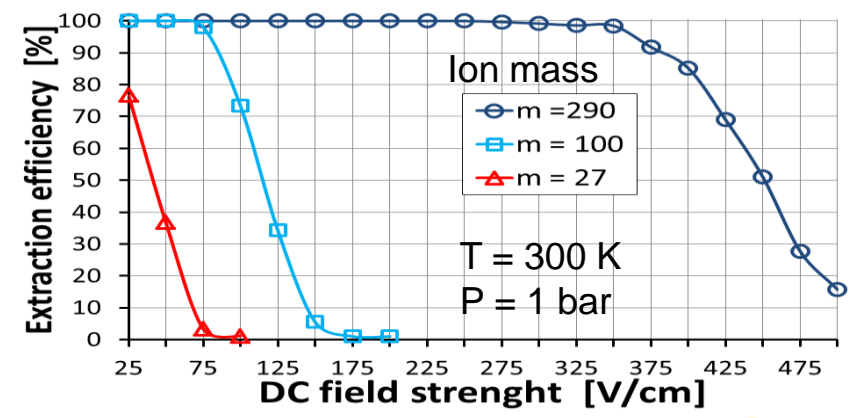
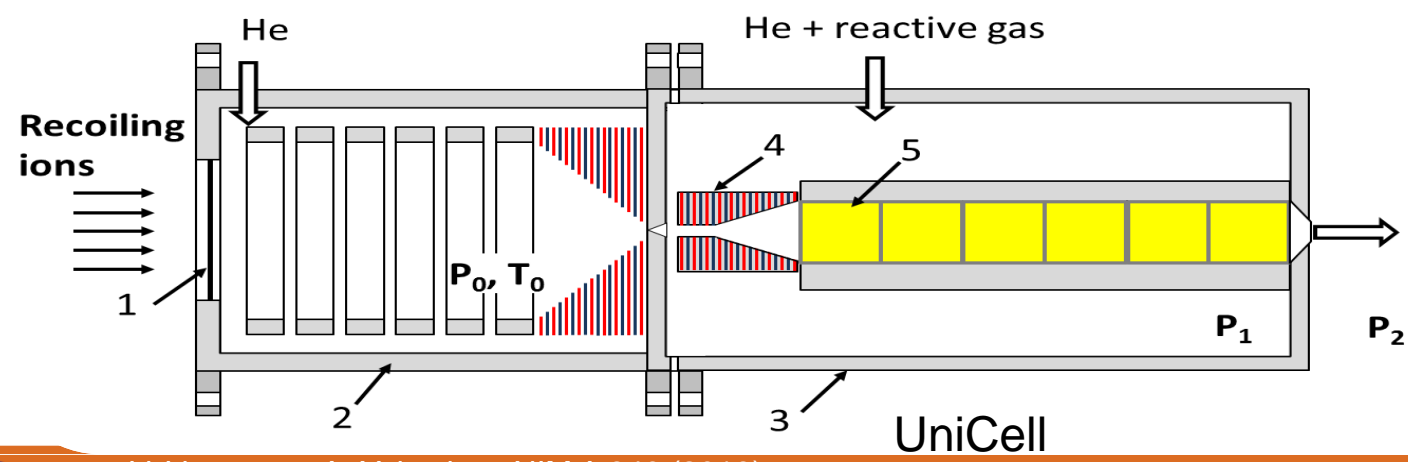
Old „SHIP“ Buffer
gas cell

miniCOMPACT

≈ 50 ms
extraction time

S. Götz *et al.*, NIM A 165090 (2021)
S. Götz *et al.*, NIM B 507 (2021) 27-35

High-pressure UniCell project for faster extraction and higher efficiency



Chemistry is possible with single atoms



Literature:

Schädel, Matthias, and Dawn Shaughnessy, eds.

The chemistry of superheavy elements. Springer Science & Business Media, 2013.