JOHANNES GUTENBERG UNIVERSITÄT MAINZ

GSI

HIM HELMHOLTZ Helmholtz Institute Mainz

www.superheavies.de
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Cold fusion: From element discovery to detailed studies

An overview from GSI

Christoph E. Düllmann[1,2,3]

[1]Johannes Gutenberg University Mainz, Mainz, Germany
[2]GSI Helmholtzzentrum für Schwerionenforschung GmbH, Darmstadt, Germany
[3]Helmholtz Institute Mainz, Mainz, Germany

Outline

The key collision partners for cold fusion (and for this talk):

$^{48}Ca + ^{208}Pb$

Part I: ^{208}Pb and neighbors: element discovery, decay spectroscopy, mass spectrometry, laser spectroscopy

Part II: ^{48}Ca beam: chemical studies

Part III: Beyond ^{48}Ca and beyond ^{208}Pb

The beginning of cold fusion

1975

2.A.1:
2.N

Nuclear Physics **A239** (1975) 353—364; © *North-Holland Publishing Co., Amsterdam*

EXPERIMENTS ON THE PRODUCTION OF FERMIUM NEUTRON-DEFICIENT ISOTOPES AND NEW POSSIBILITIES OF SYNTHESIZING ELEMENTS WITH $Z > 100$

Yu. Ts. OGANESSIAN, A. S. ILJINOV, A. G. DEMIN and S. P. TRETYAKOVA
Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Dubna, USSR

Received 25 September 1974

1976

2.N

Nuclear Physics **A267** (1976) 359 – 364; © *North-Holland Publishing Co., Amsterdam*

ACCELERATION OF ^{48}Ca IONS AND NEW POSSIBILITIES OF SYNTHESIZING SUPERHEAVY ELEMENTS

G. N. FLEROV, Yu. Ts. OGANESSIAN, A. A. PLEVE, N. V. PRONIN and Yu. P. TRETYAKOV
Joint Institute for Nuclear Research, Dubna, USSR

Received 2 April 1976

E | NUCLEAR REACTIONS ^{204,206,207,208}Pb(^{48}Ca, xn), $E = 235$ MeV; measured σ for production of ^{252}102. ^{252}102 deduced $T_{1/2}$. Thick targets of separated Pb isotopes.

^{252}No cross sections from ^{48}Ca + $^{206-208}$Pb

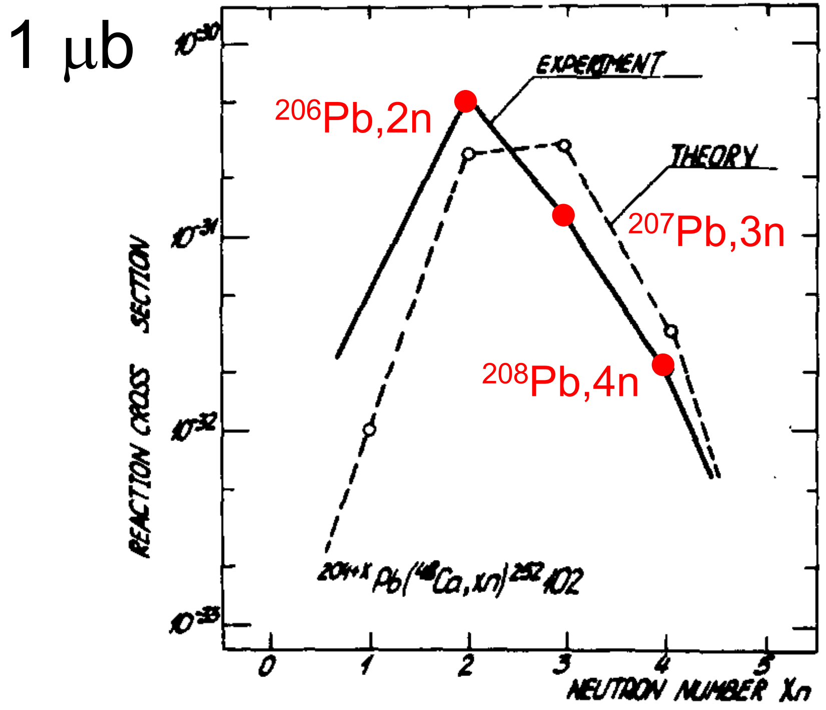


Fig. 3. Production cross sections for the isotope ^{252}102 in reactions induced by ^{48}Ca ions.

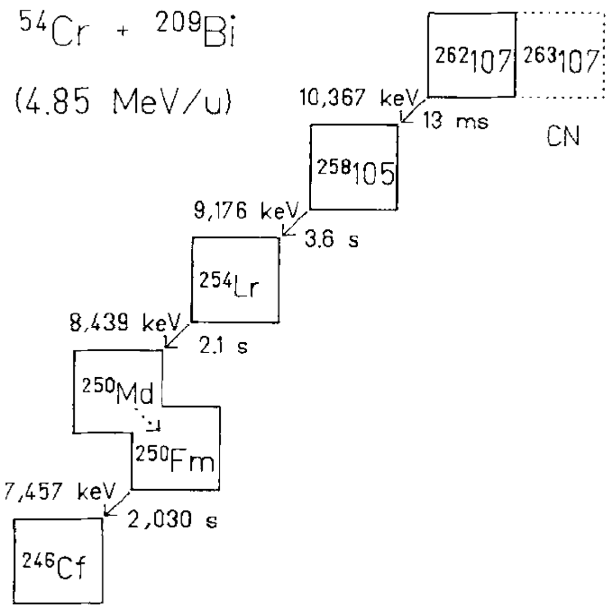
Reactions with targets around ^{208}Pb or beams of ^{48}Ca shaped the SHE field

1980s: Discovery of elements 107-109 at GSI

1981: Bohrium

^{209}**Bi**(^{54}Cr,n)^{262}Bh

Six chains

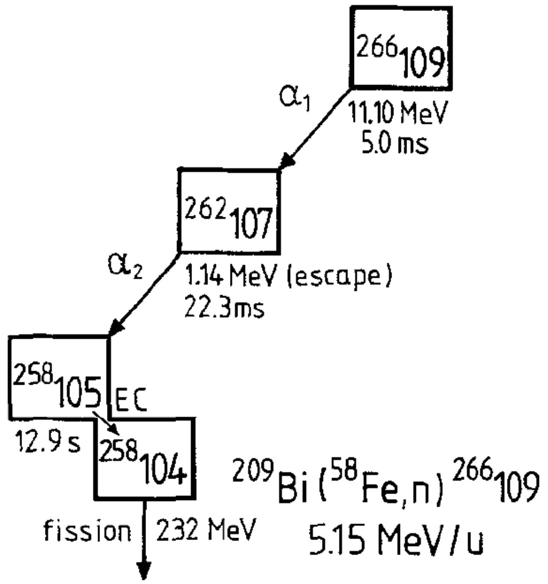


G. Münzenberg et al.,
Z. Phys. A 300 (1981) 107

1984: Meitnerium

^{209}**Bi**(^{58}Fe,n)^{266}Mt

One chain

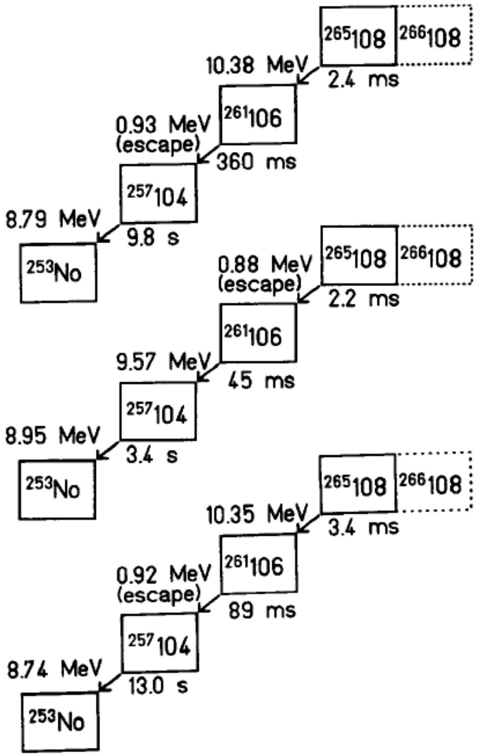


G. Münzenberg et al.,
Z. Phys. A 315 (1984) 145

1984: Hassium

^{208}**Pb**(^{58}Fe,n)^{265}Hs

Three chains



G. Münzenberg et al.,
Z. Phys. A 317 (1984) 235

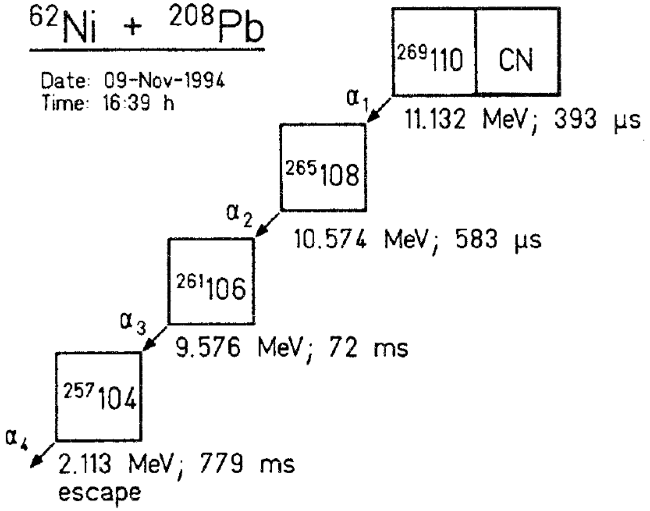
1990s: Discovery of elements 110-112 at GSI

1995: Darmstadtium

^{208}**Pb**(^{62}Ni,n)^{269}Ds

Four chains

*09.11.1994



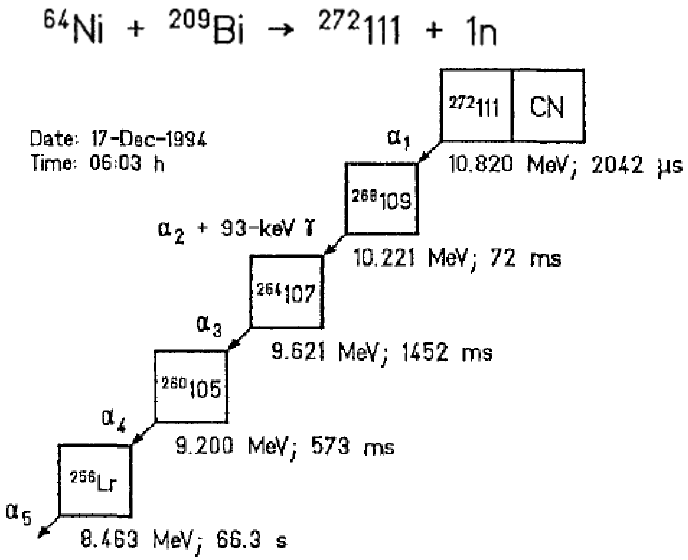
S. Hofmann et al.,
Z. Phys. A 350 (1995) 277

1995: Roentgenium

^{209}**Bi**(^{64}Ni,n)^{272}Rg

Three chains

*08.12.1994

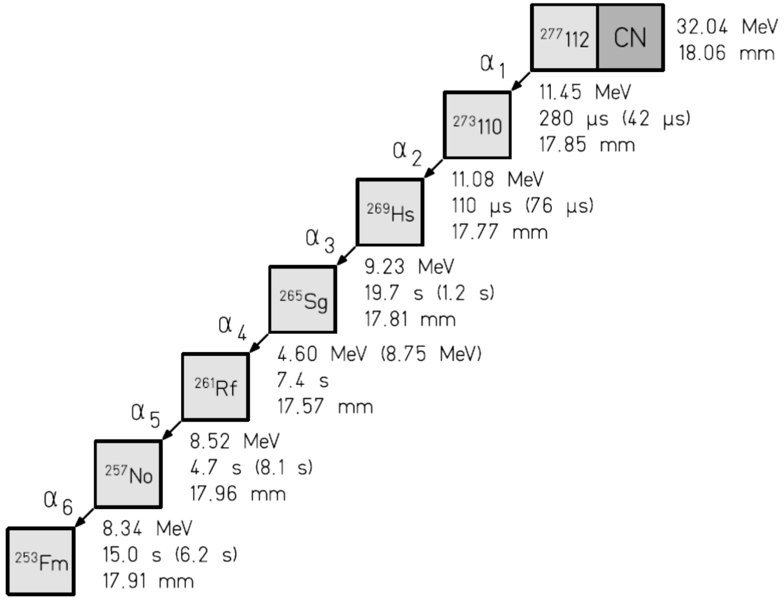


S. Hofmann et al.,
Z. Phys. A 350 (1995) 281

1996: Copernicium

^{208}**Pb**(^{70}Zn,n)^{277}Cn

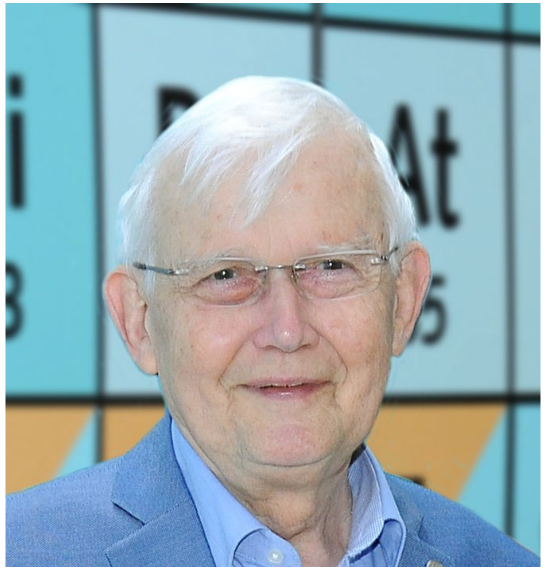
One chain



S. Hofmann et al.,
Z. Phys. A 354 (1996) 229
Eur. Phys. J. A 14 (2002) 147

We mourn our colleagues

Sigurd Hofmann
† 17.06.22

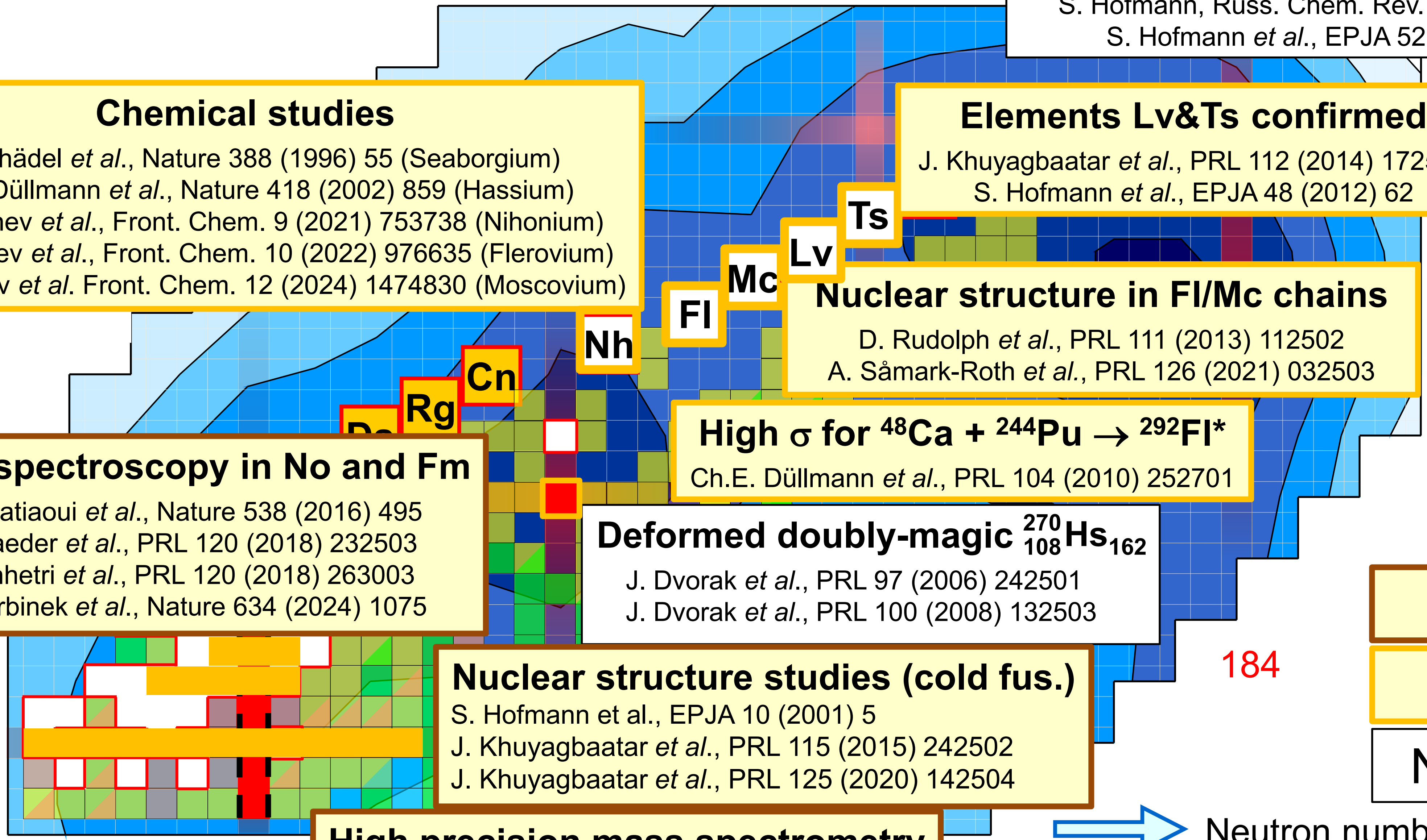


Gottfried Münzenberg
† 02.01.24

Jens V. Kratz
† 11.02.24

Peter Armbruster
† 26.06.24

SHE Highlights from GSI



Search beyond Z=118

J. Khuyagbaatar *et al.*, PRC 102 (2020) 064602 (Z=119&120)
S. Hofmann, Russ. Chem. Rev. 78 (2009) 1123 (Z=120)
S. Hofmann *et al.*, EPJA 52 (2016) 180 (Z=120)

Chemical studies

M. Schädel *et al.*, Nature 388 (1996) 55 (Seaborgium)
Ch.E. Düllmann *et al.*, Nature 418 (2002) 859 (Hassium)
A. Yakushev *et al.*, Front. Chem. 9 (2021) 753738 (Nihonium)
A. Yakushev *et al.*, Front. Chem. 10 (2022) 976635 (Flerovium)
A. Yakushev *et al.* Front. Chem. 12 (2024) 1474830 (Moscovium)

Elements Lv&Ts confirmed

J. Khuyagbaatar *et al.*, PRL 112 (2014) 172501
S. Hofmann *et al.*, EPJA 48 (2012) 62

Nuclear structure in Fl/Mc chains

D. Rudolph *et al.*, PRL 111 (2013) 112502
A. Såmark-Roth *et al.*, PRL 126 (2021) 032503

High σ for $^{48}Ca + ^{244}Pu \rightarrow ^{292}Fl^*$

Ch.E. Düllmann *et al.*, PRL 104 (2010) 252701

Laser spectroscopy in No and Fm

M. Laatiaoui *et al.*, Nature 538 (2016) 495
S. Raeder *et al.*, PRL 120 (2018) 232503
P. Chhetri *et al.*, PRL 120 (2018) 263003
J. Warbinek *et al.*, Nature 634 (2024) 1075

Deformed doubly-magic $^{270}_{108}Hs_{162}$

J. Dvorak *et al.*, PRL 97 (2006) 242501
J. Dvorak *et al.*, PRL 100 (2008) 132503

Nuclear structure studies (cold fus.)

S. Hofmann et al., EPJA 10 (2001) 5
J. Khuyagbaatar *et al.*, PRL 115 (2015) 242502
J. Khuyagbaatar *et al.*, PRL 125 (2020) 142504

High precision mass spectrometry

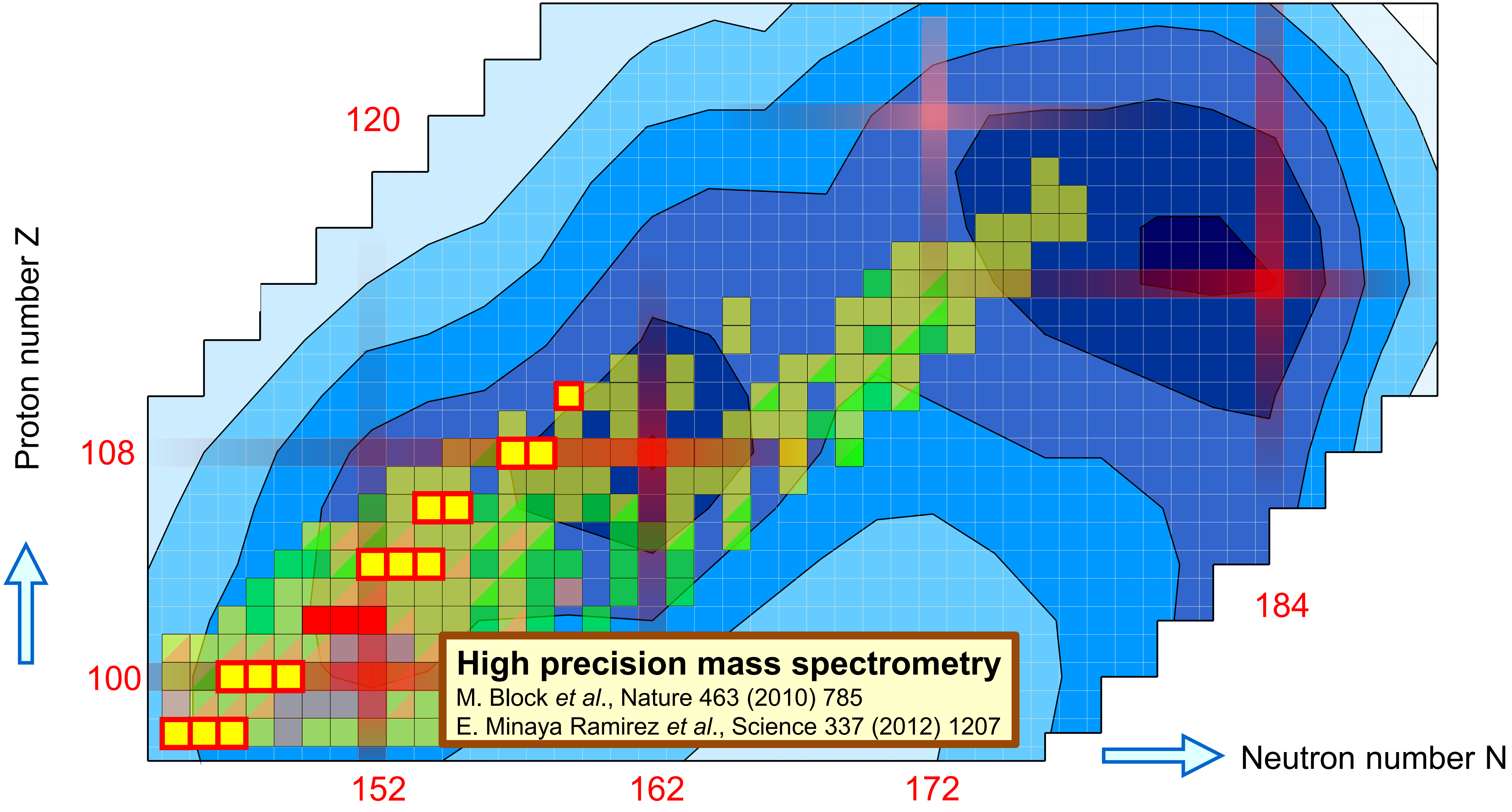
M. Block *et al.*, Nature 463 (2010) 785
E. Minaya Ramirez *et al.*, Science 337 (2012) 1207

$^{20x}Pb/^{209}Bi$

^{48}Ca beam

No ^{208}Pb, ^{48}Ca

High-precision mass spectrometry

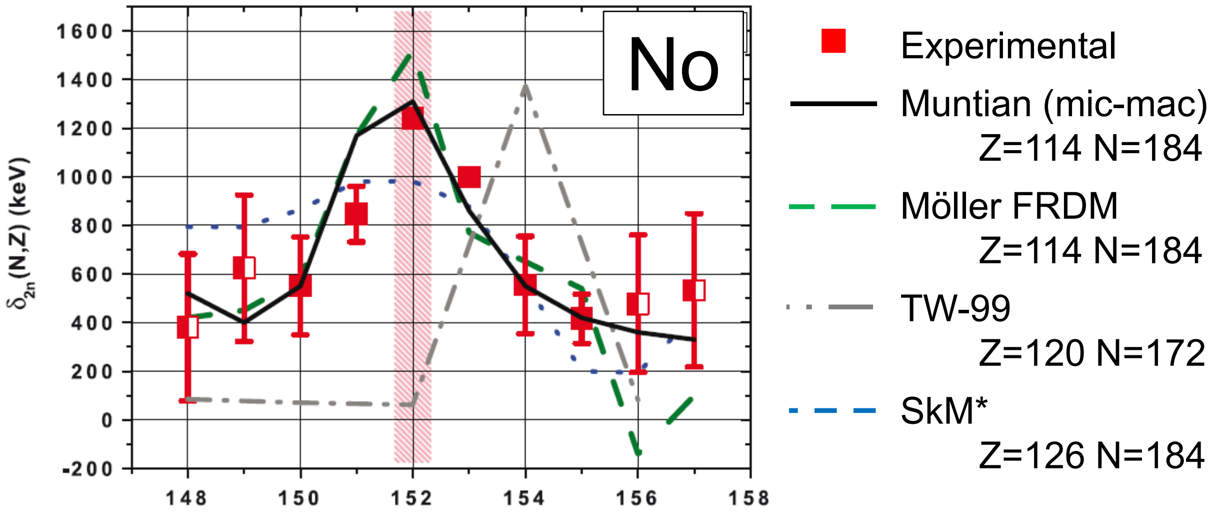


High precision mass spectrometry
M. Block *et al.*, Nature 463 (2010) 785
E. Minaya Ramirez *et al.*, Science 337 (2012) 1207

Shell mapping N=152 and 162

Direct Mapping of Nuclear Shell Effects in the Heaviest Elements

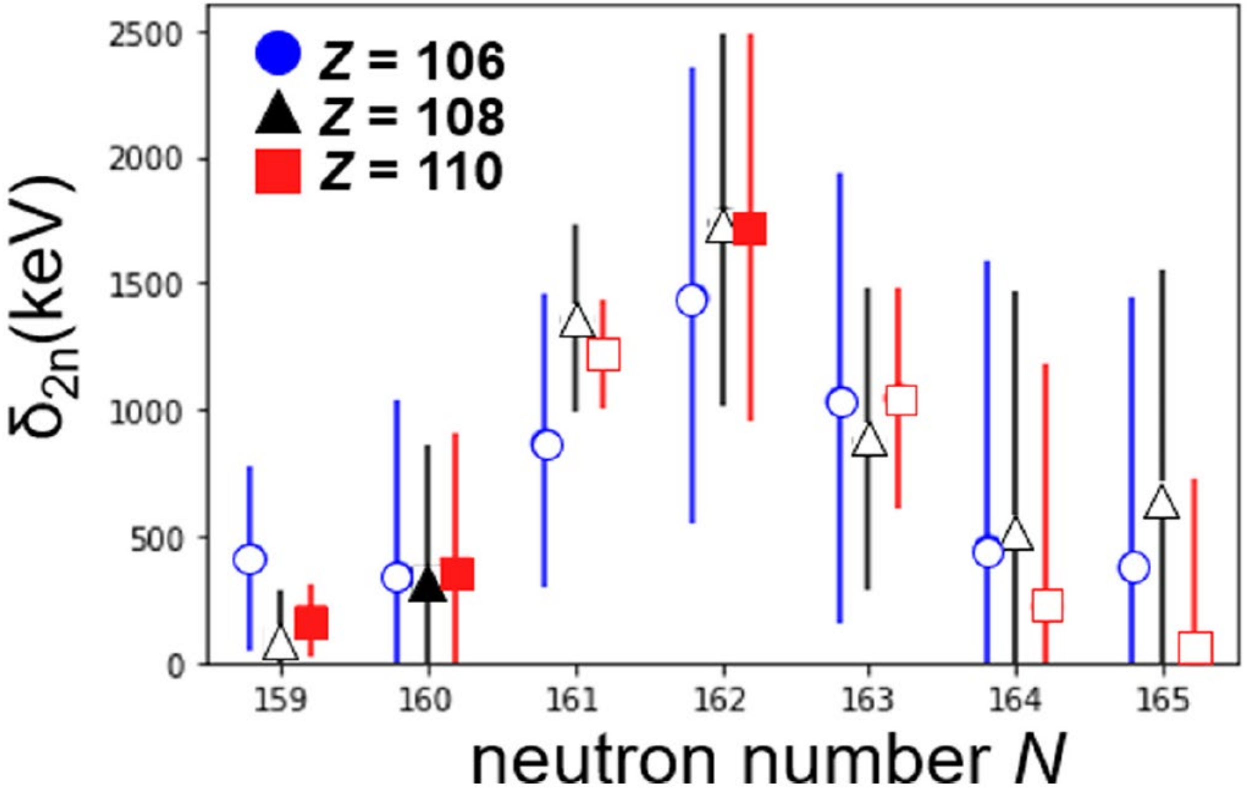
E. Minaya Ramirez,[1,2] D. Ackermann,[2] K. Blaum,[3,4] M. Block,[2*] C. Droese,[5] Ch. E. Düllmann,[6,2,1] M. Dworschak,[2] M. Eibach,[4,6] S. Eliseev,[3] E. Haettner,[2,7] F. Herfurth,[2] F. P. Heßberger,[2,1] S. Hofmann,[2] J. Ketelaer,[3] G. Marx,[5] M. Mazzocco,[8] D. Nesterenko,[9] Yu. N. Novikov,[9] W. R. Plaß,[2,7] D. Rodríguez,[10] C. Scheidenberger,[2,7] L. Schweikhard,[5] P. G. Thirolf,[11] C. Weber[11]



Science **337** (2012) 1207

The program started with $^{48}Ca + ^{208}Pb$!

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

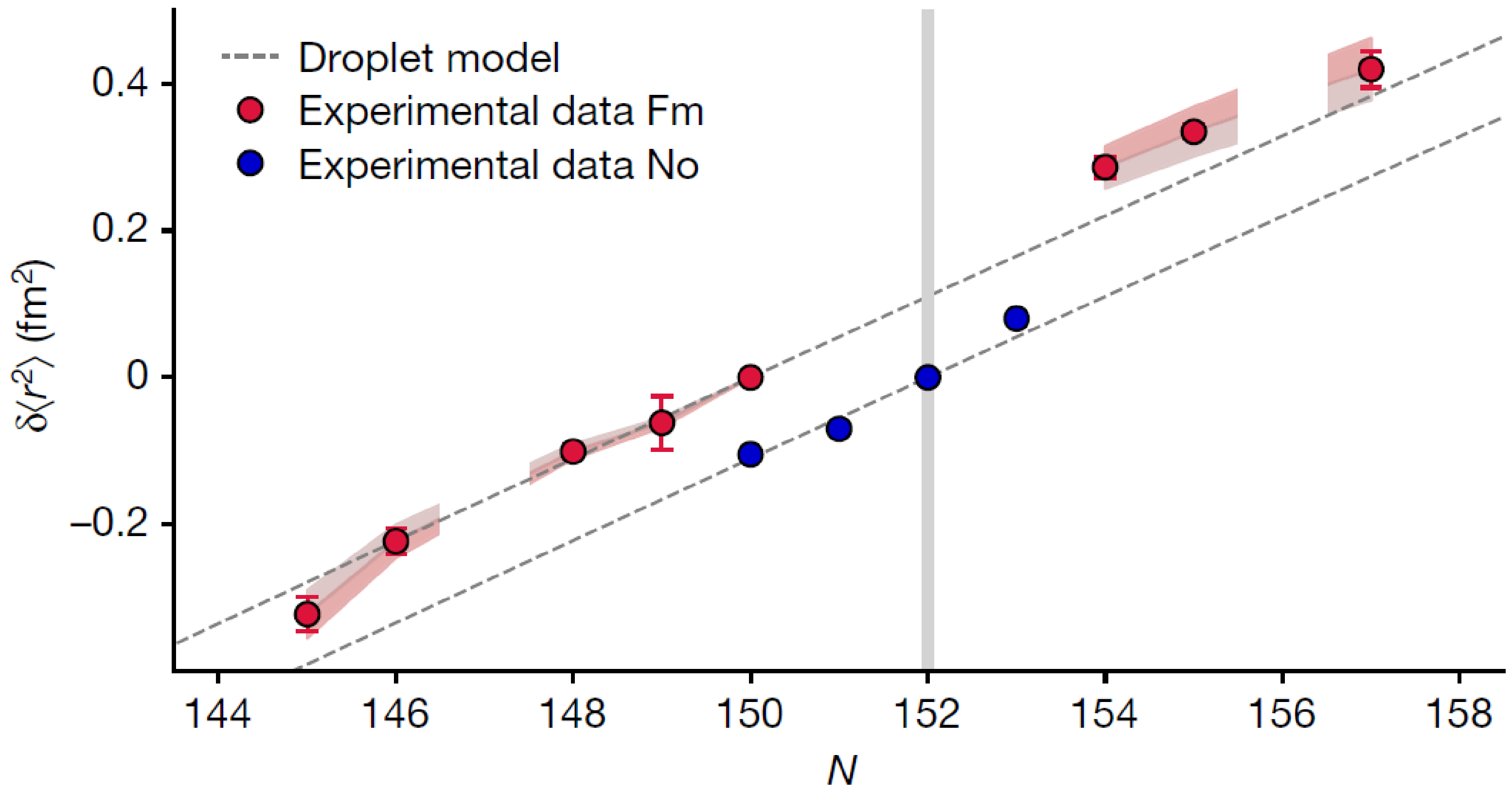


O. Kaleja *et al*, Phys. Rev. C 106 (2022) 054325

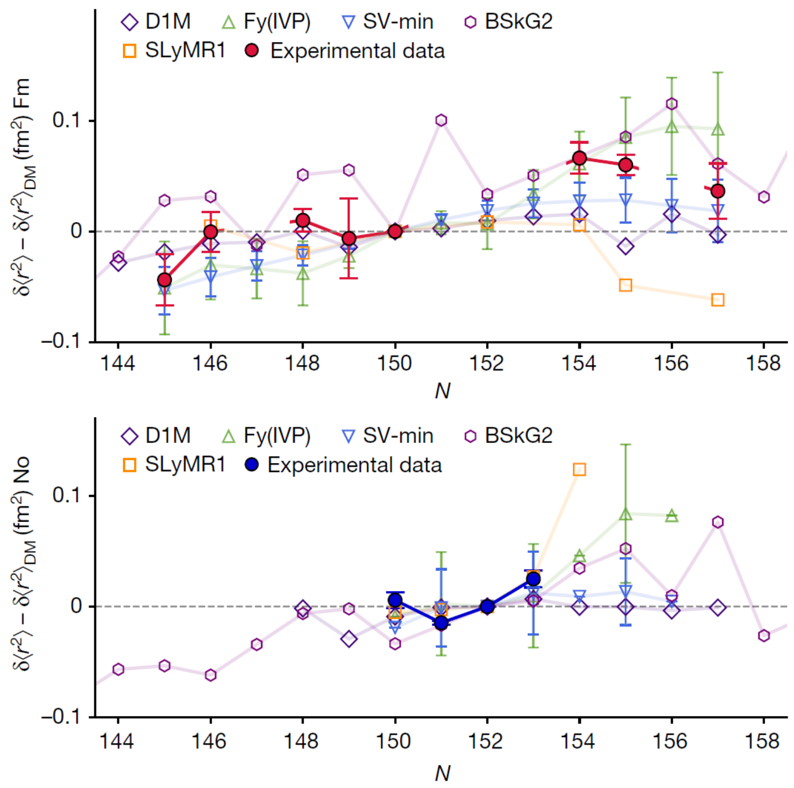
Probing shell structure with laser light

Nuclear charge radii show a smooth trend across N=152 in Fm and No nuclei

Experiment



Theory

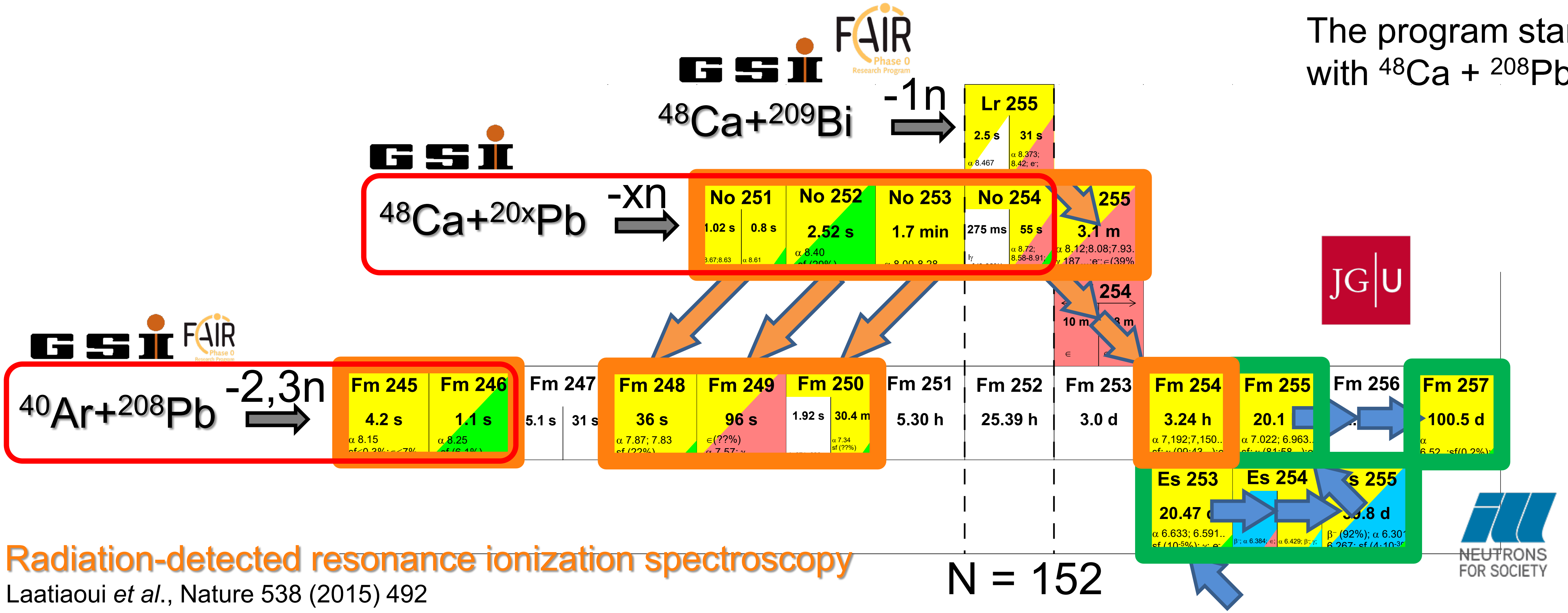


- Various theoretical model predictions reproduce the experiment.
- Shell structure influence on nuclear radii is diminished compared to lighter nuclei

J. Warbinek *et al.*,
Nature 634 (2024) 1075

Production of $_{99}$Es, $_{100}$Fm, and $_{102}$No nuclei

The program started with ^{48}Ca + ^{208}Pb!



From HFIR

Online: Radiation-detected resonance ionization spectroscopy

M. Laatiaoui *et al.*, Nature 538 (2015) 492
S. Raeder *et al.*, Phys. Rev. Lett. 120 (2018) 232503
J. Warbinek *et al.*, Nature 634 (2024) 1075

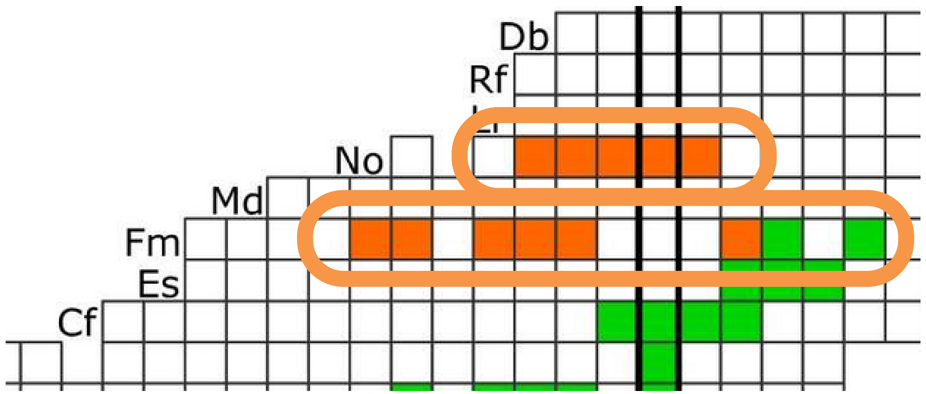
Offline: Resonance ionization spectroscopy

S. Nothhelfer *et al.*, Phys. Rev. C 105 (2022) L021302
J. Warbinek *et al.*, Nature 634 (2024) 1075

Size and shape of $_{100}$Fm and $_{102}$No nuclei around N=152

Recently investigated at GSI by laser spectroscopy

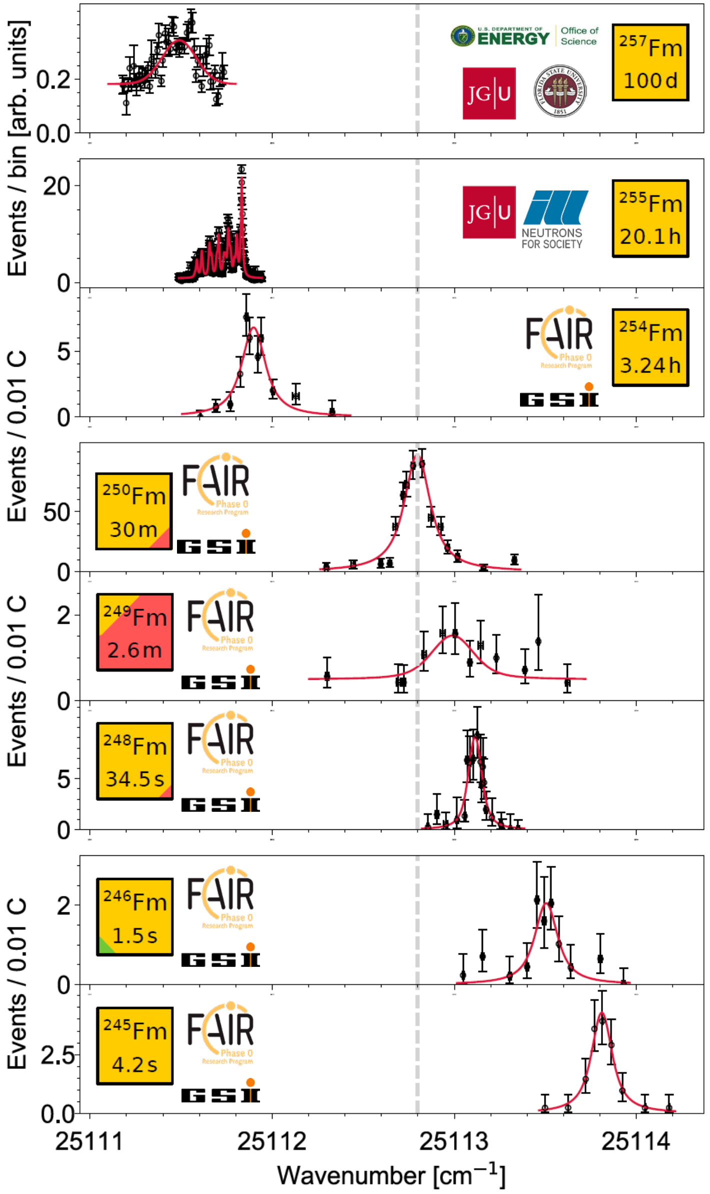
Investigated via offline laser spectroscopy

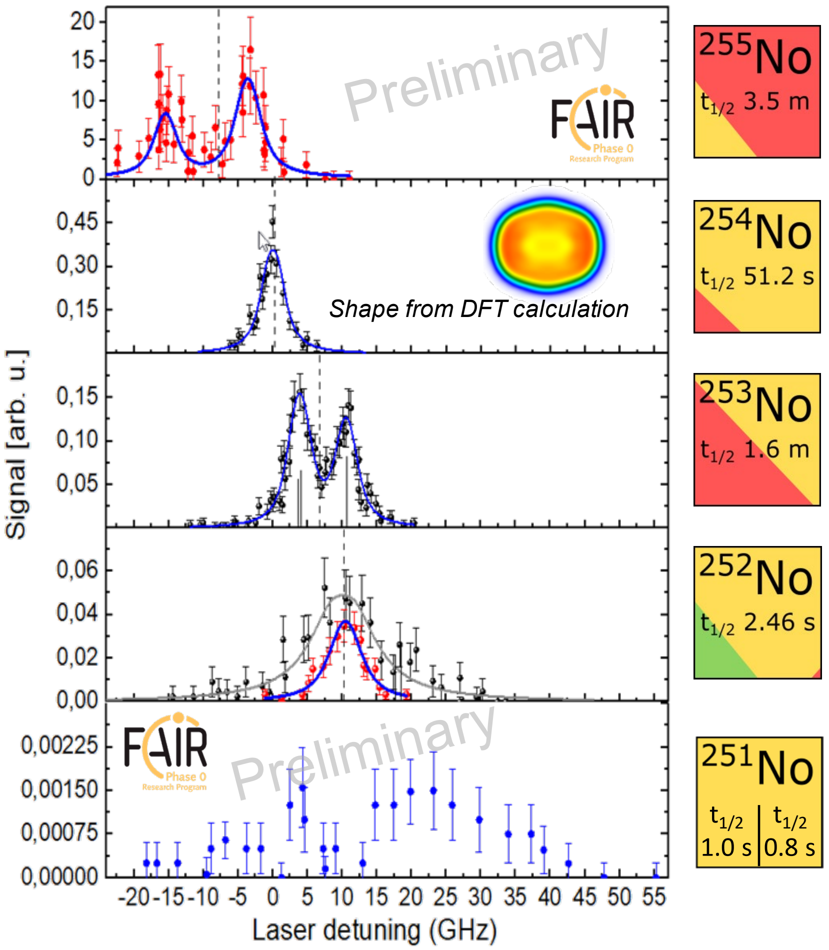


$^{252-254}$**No**: S. Raeder *et al.*, PRL 120 (2018) 232503

$^{253-255}$**Es**: S. Nothhelfer *et al.*, PRC 105 (2022) L021302

$^{249-253}$**Cf**: F. Weber *et al.*, PRC 107 (2023) 034313





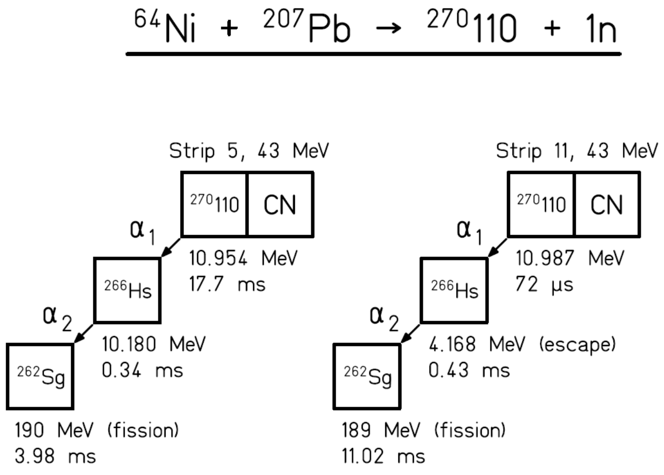
Next generation: JetRIS for improved resolution, demonstrated in ^{254}No

J. Lantis *et al.*, Phys. Rev. Res. 6 (2024) 023318

Nuclear structure examples

Ds 270 — α-decaying high-K isomer

Ds 270: 6 ms | 0.1 ms; α 10.95; | α 11.03



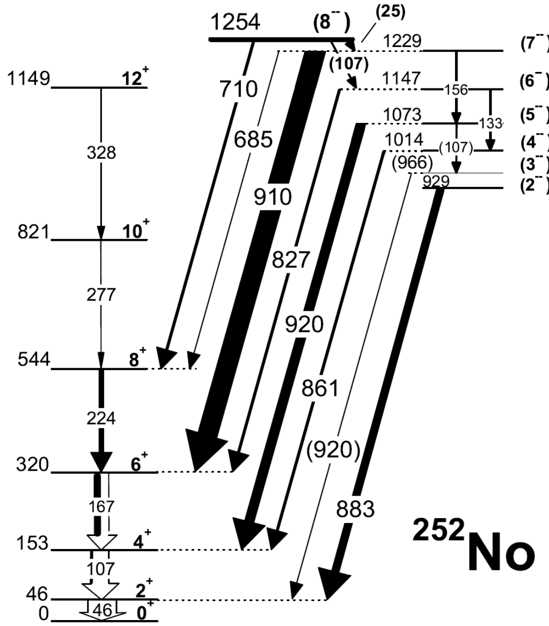
S. Hofmann *et al.*, EPJA 10 (2001) 5
D. Ackermann, NPA 944 (2015) 376

No 252 — High-*K* isomer from α-γ-studies

No 252: 109 ms | 2.52 s; Iγ 167; 910...; e⁻ | α 8.40; sf (29%)

$^{206}Pb(^{48}Ca,2n)^{252m}No$

$T_{1/2}$ = 110±10 ms



B. Sulignano *et al.*, EPJA 33 (2007) 327

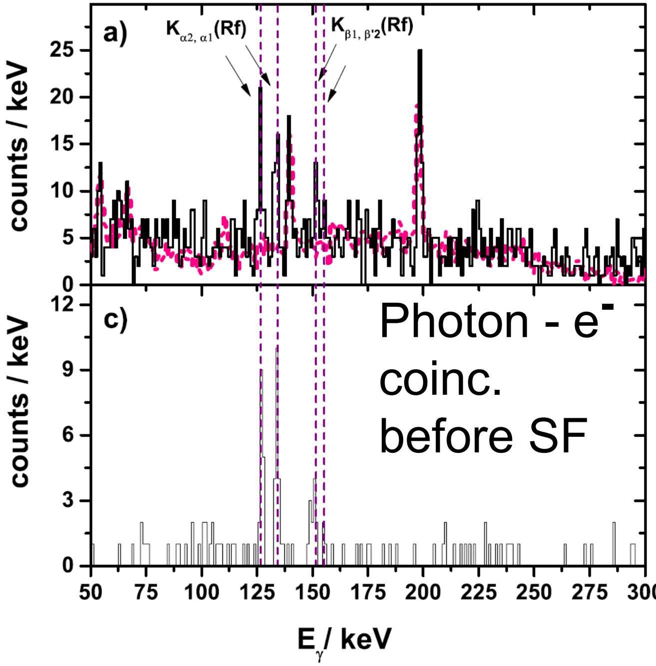
Db 258 — Delayed *X* rays after EC

Db 258: 1.9 s | 4.3 s; α 9.20; ∈? | α 9.17...; ∈; γ 221

Rf 258: 14.7 ms; sf; α 9.05 (31±11%)

$^{50}Ti + ^{209}Bi$

Photons in delayed coincidence w/ SF

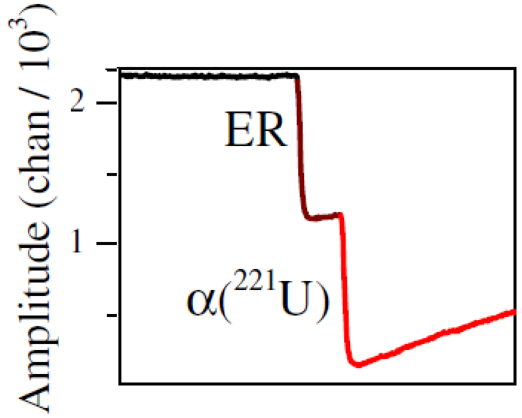


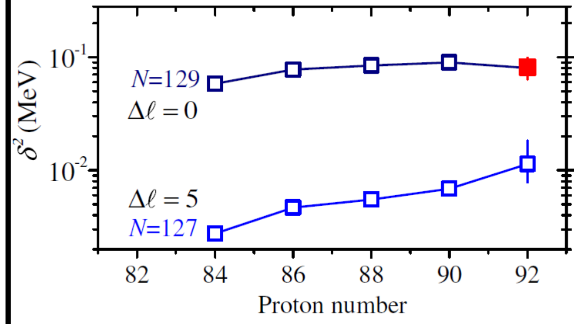
F.P. Heßberger *et al.*, EPJA 52 (2016) 328

U 221 — Sub-μs ^{221}U

U 221: 0.66 μs; α 9.71

$^{50}Ti + ^{176}Yb$





J. Khuyagbaatar *et al.*, PRL 115 (2015) 242502

Outline

The key collision partners for cold fusion (and for this talk):

$^{48}Ca + ^{208}Pb$

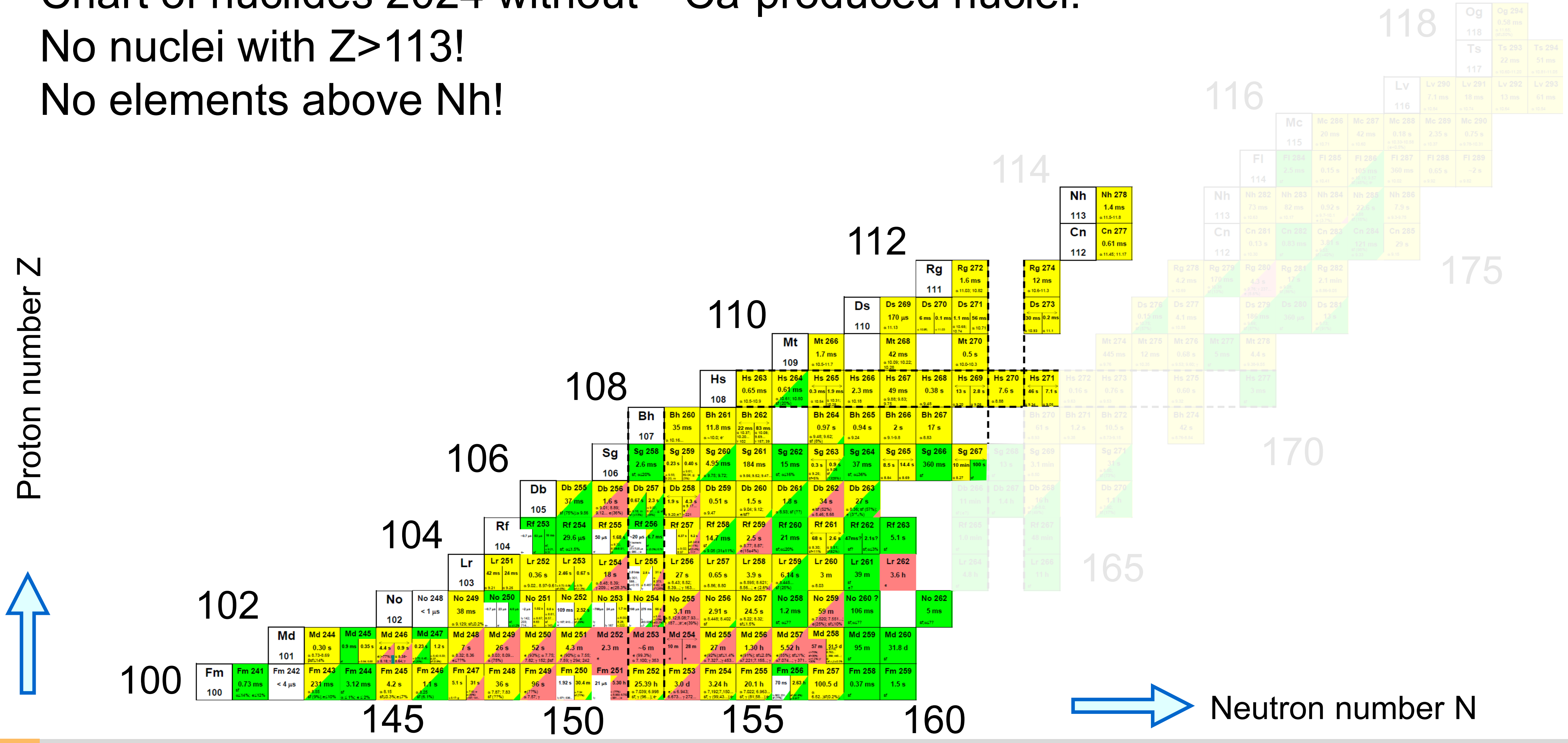
Part I: ^{208}Pb and neighbors: element discovery
decay spectroscopy
mass spectrometry
laser spectroscopy

Part II: ^{48}Ca beam: chemical studies

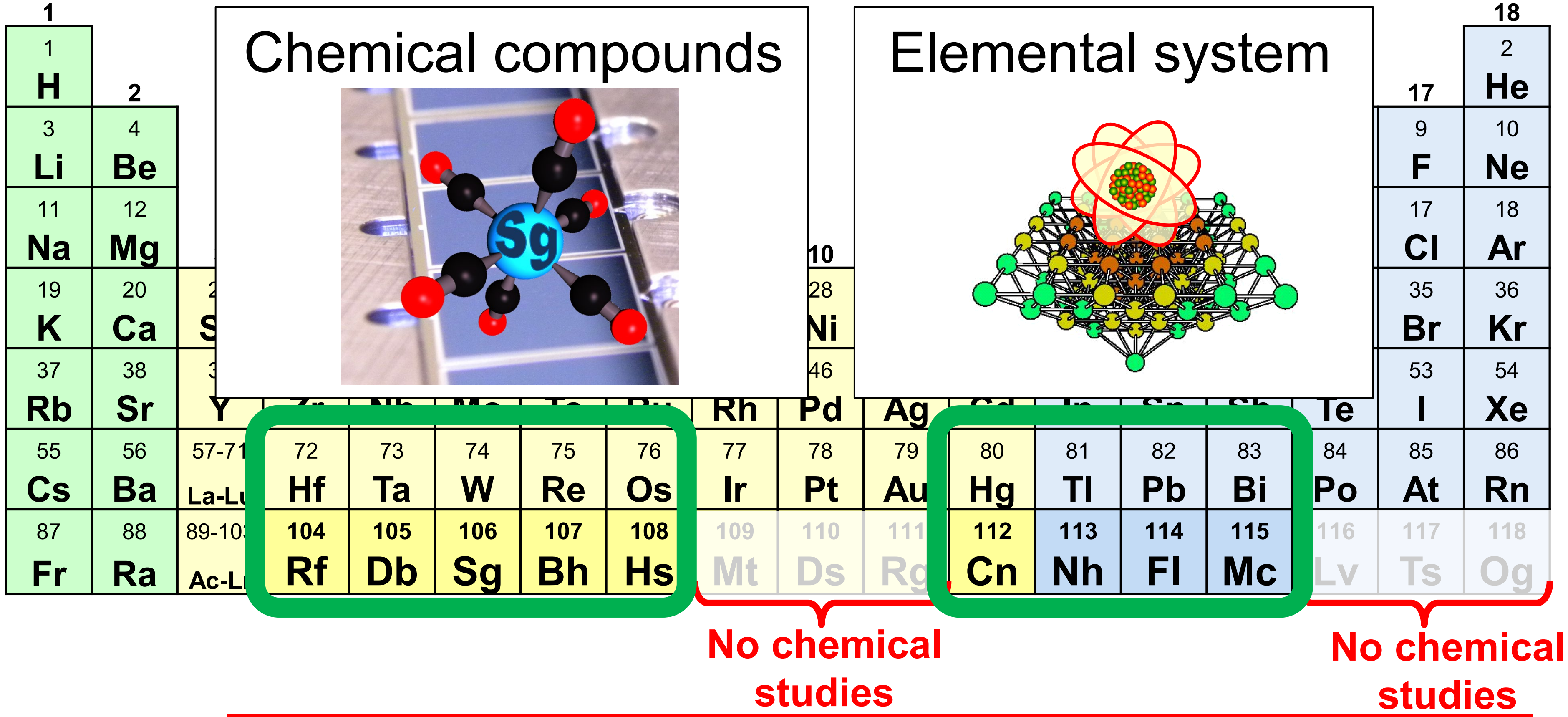
Part III: Beyond ^{48}Ca and beyond ^{208}Pb

From ^{48}Ca …^{70}Zn + ^{20x}Pb/Bi to today: the role of ^{48}Ca

Chart of nuclides 2024 without ^{48}Ca-produced nuclei:
No nuclei with Z>113!
No elements above Nh!



Current status of SHE chemistry



Are elements 112, 114, and 118 relatively inert gases?

Kenneth S. Pitzer

Department of Chemistry and Inorganic Materials Research Division of the Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

(Received 14 April 1975)

The Journal of Chemical Physics, Vol. 63, No. 2, 15 July 1975 1032

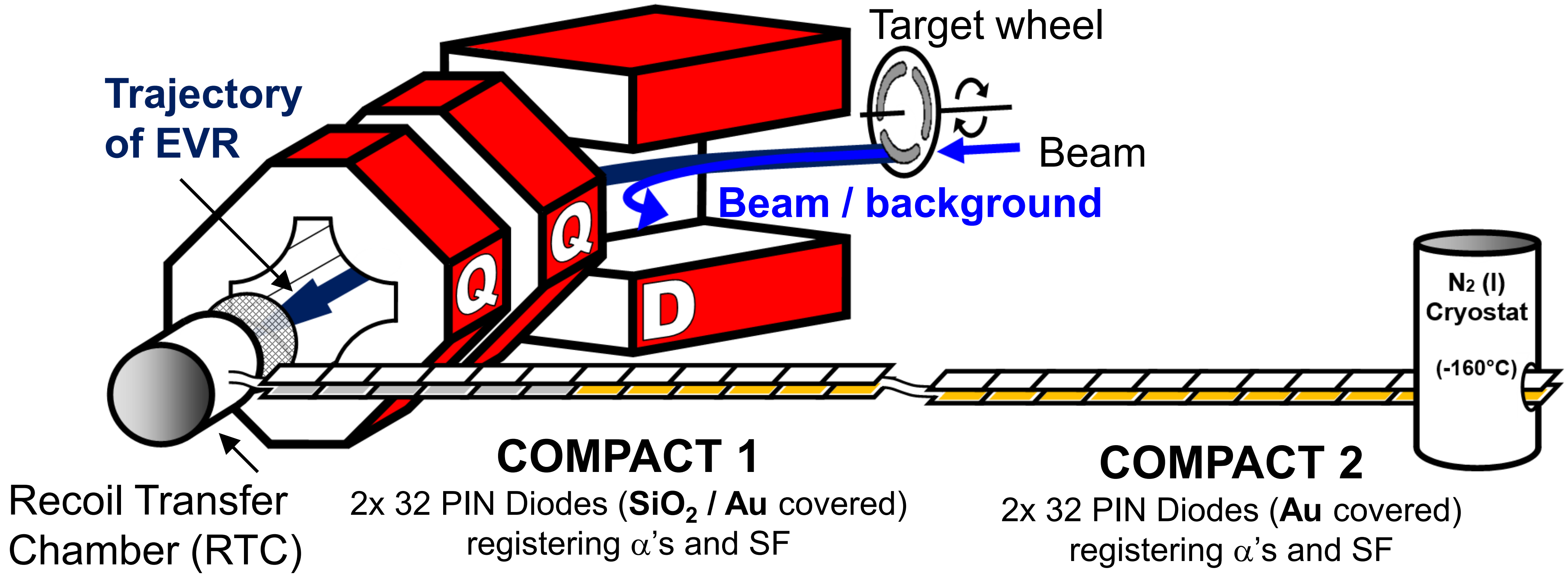
Relativistic effects of high nuclear charge on atomic orbitals affect chemical properties

Recent overview of GSI element discoveries and chemical studies

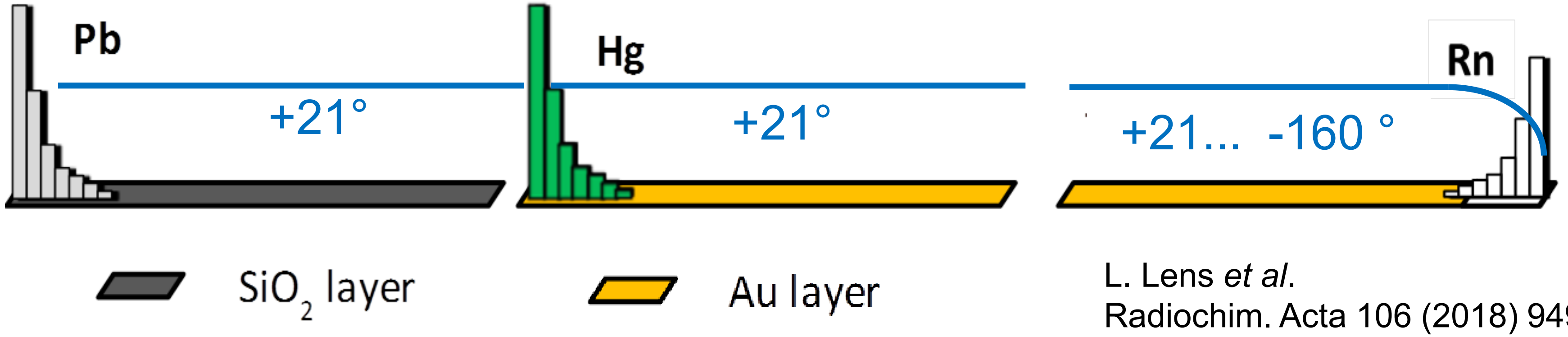
Ch.E. Düllmann *et al.*, Radiochim. Acta 110 (2022) 417

Experimental setup: chemistry behind recoil separator

TransActinide Separator and Chemistry Apparatus – TASCA



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



L. Lens *et al.*
Radiochim. Acta 106 (2018) 949

Predictions of adsorption of Cn, Nh, Fl and Mc on Au(111) and SiO_2 surface

ADF BAND calculations

- Spin-Orbit interaction
- best basis sets
- relaxation effects
- full geometry optimization

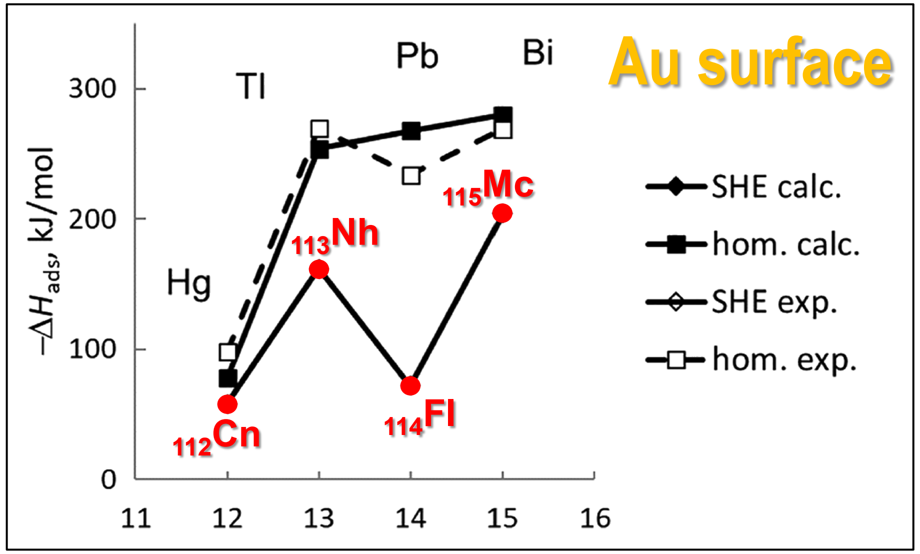
V. Pershina and M. Iliaš
Inorg. Chem. 57 (2018) 3948
Dalton Trans. 51 (2022) 7321
Inorg. Chem. 60 (2021) 9796
Inorg. Chem. 61 (2022) 15910

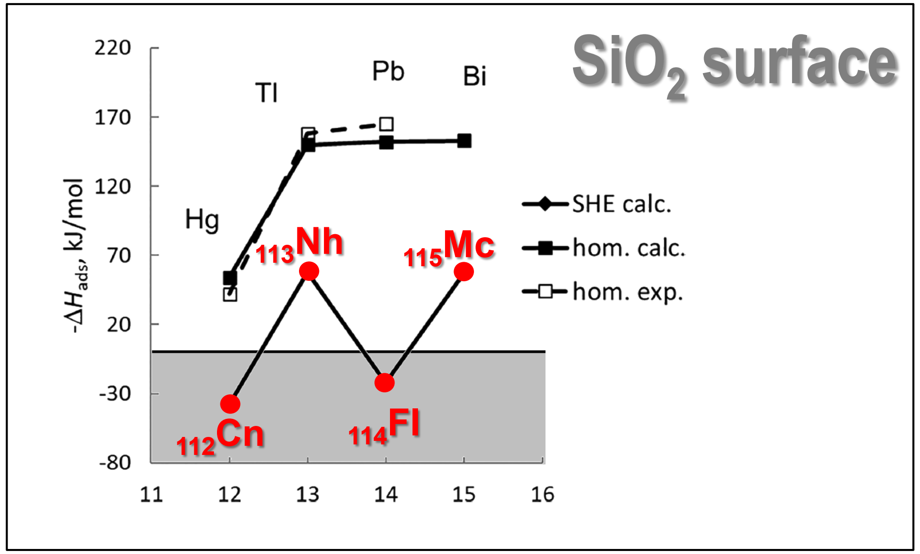
VASP calculations

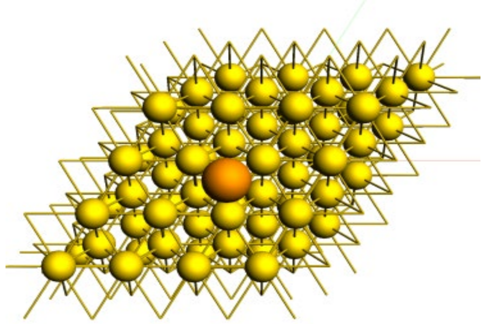
L. Trombach *et al.*,
PCCP 21 (2019) 18048

Solid state calcs.

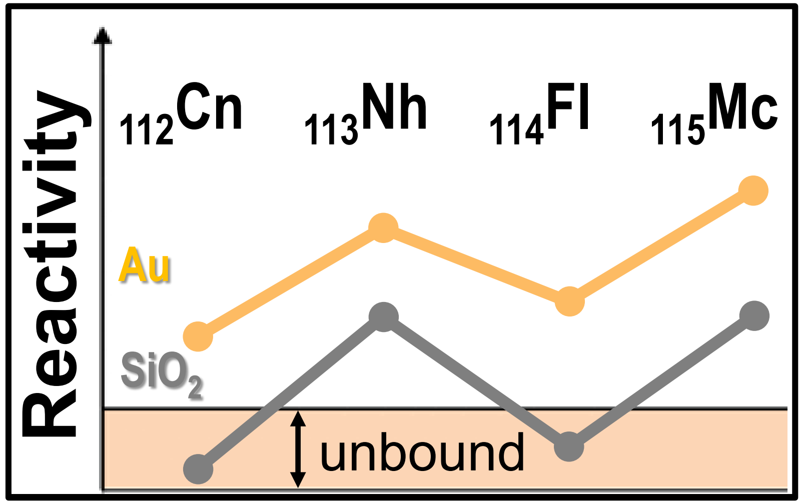
E. Florez *et al.*,
J. Chem. Phys 167, 064304 (2022)

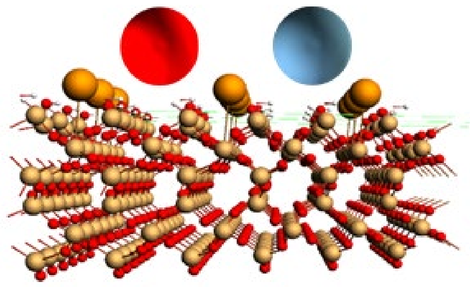






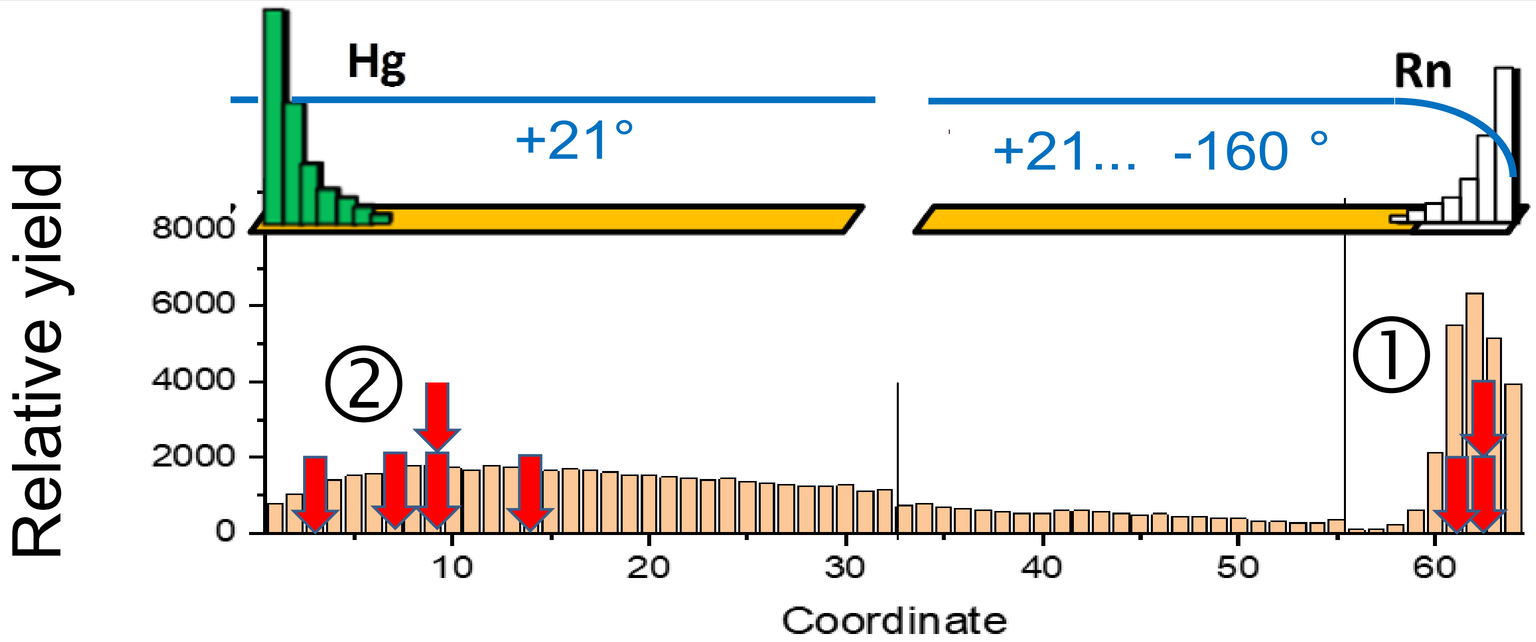
Au





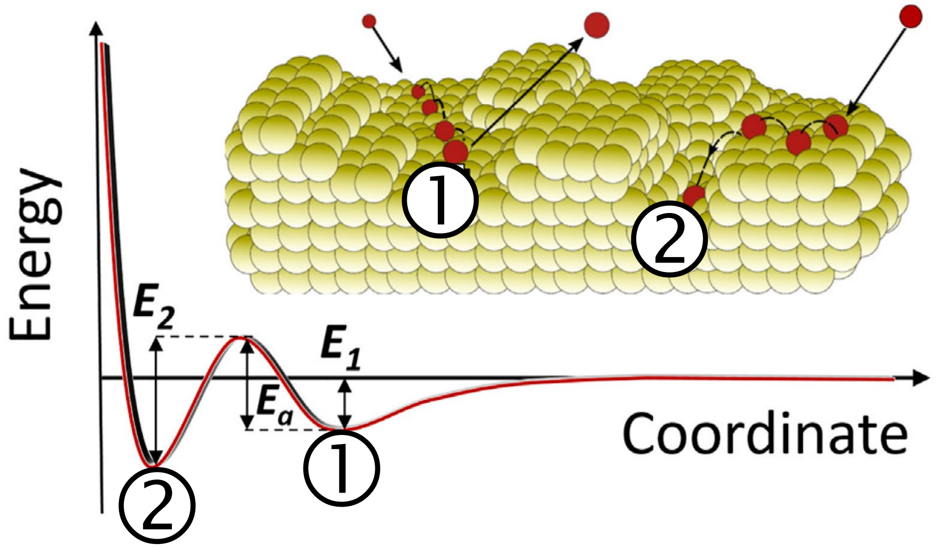
SiO_2

Interaction of Fl with an inhomogeneous Au surface



Data from 2009 – 2016 (3 runs): 8 atoms

Two peaks corresponding to two different Fl species is considered unlikely



Fl encounters the inhomogeneous Au surface, and…

-at **weakly interacting site**, desorption may occur, leading to transport unntil **adsorption by van-der Waals occurs** (peak ①)

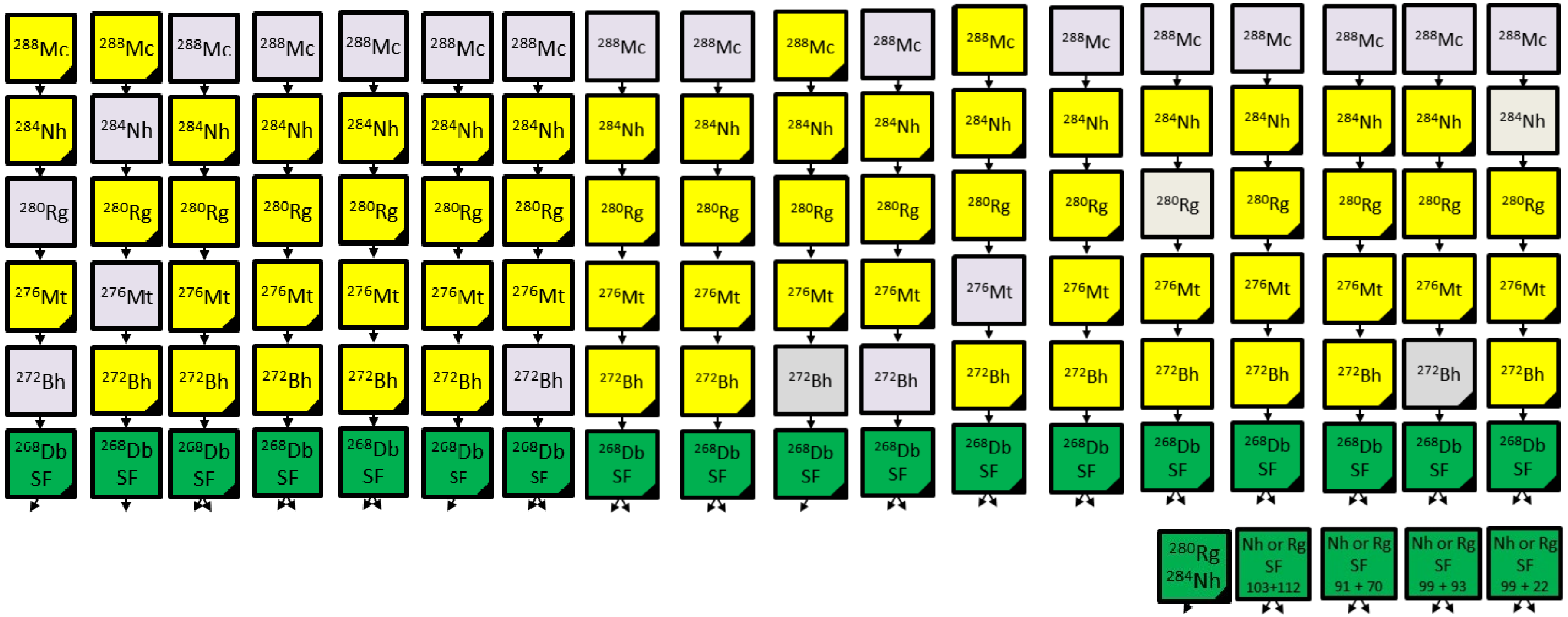
-diffusion may lead to **strongly reactive site**, where the atom is reatined due to formation of a **Fl-metal bond** (peak ②)

Flerovium is a weakly reactive metal, not a noble gas

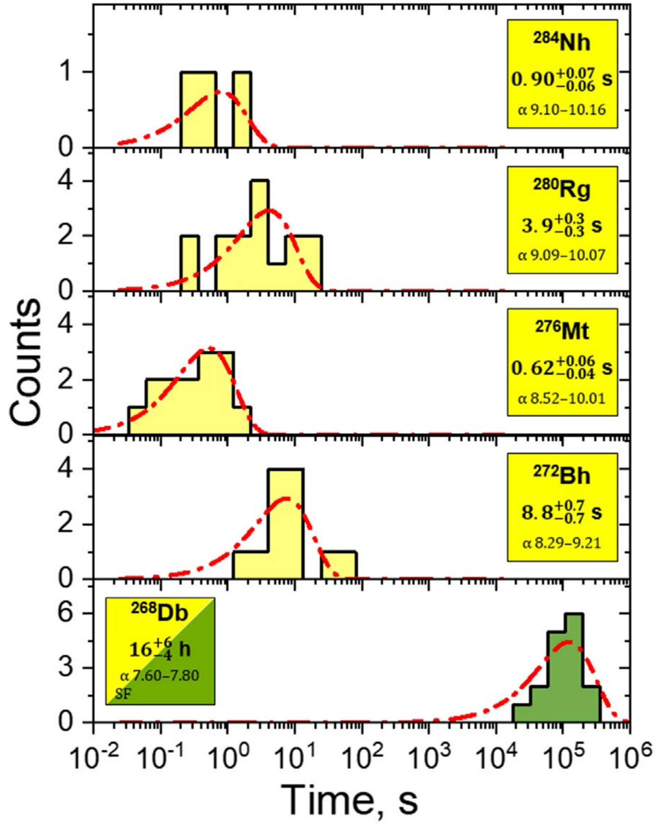
A. Yakushev *et al.*, Front. Chem. 10 (2022) 976635

Elements 113 and 115: $^{243}Am(^{48}Ca,3n)^{288}Mc \rightarrow {}^{284}Nh$

18 decay chains & 5 fissions in 2 months of beamtime



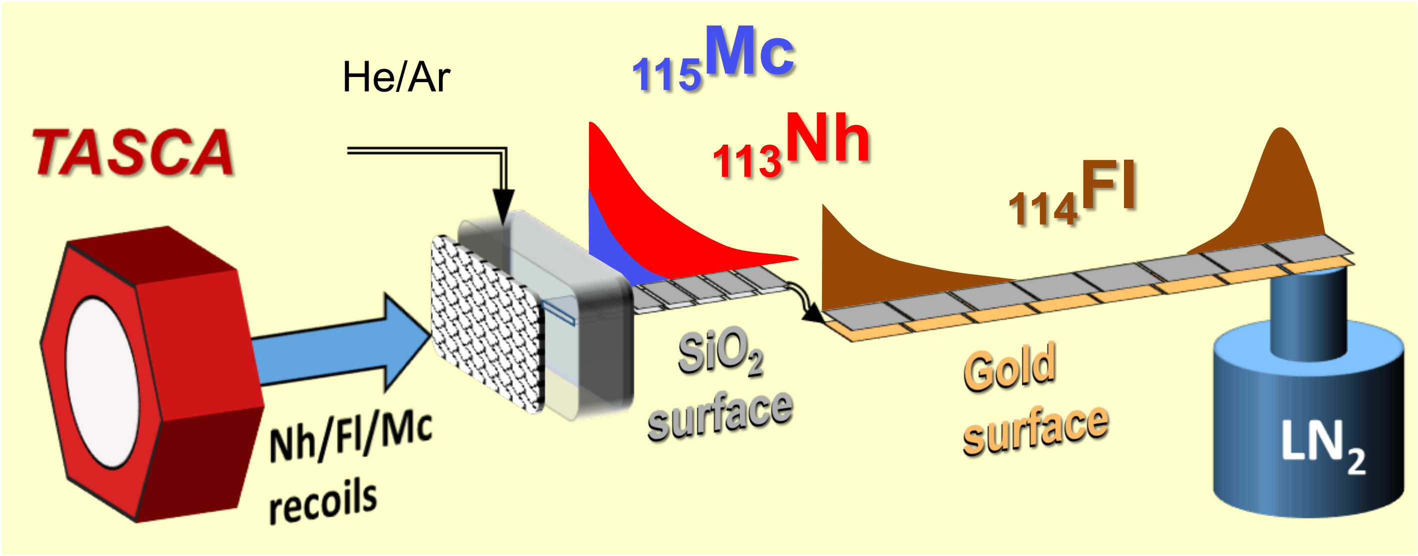
Nuclear data



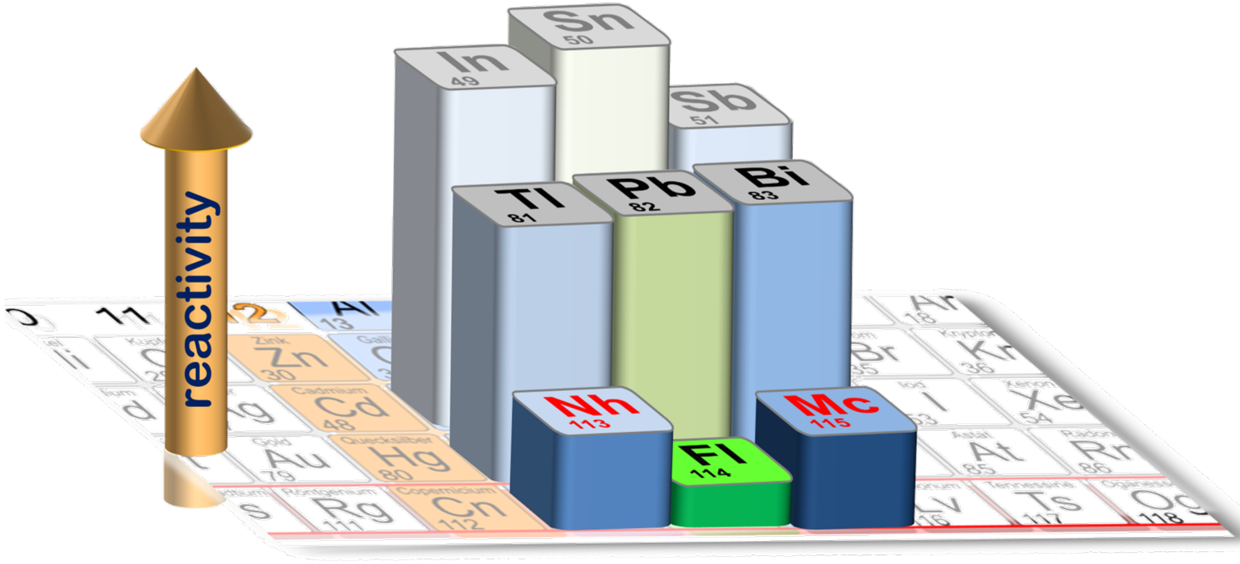
A. Yakushev *et al.*, Front. Chem. 12 (2024) 1474820

The heaviest chemically studied elements

Deposition of Nh, Fl, and Mc on SiO_2 and Au



Reactivity trends in groups 13-15



Bar height: $-\Delta H_{ads}$ on SiO_2

$7p_{1/2}$ subshell closure in $_{114}$Fl experimentally confirmed

A. Yakushev *et al*., Front. Chem. 12 (2024) 1474820

A. Yakushev *et al*., Front. Chem. 10 (2022) 976635

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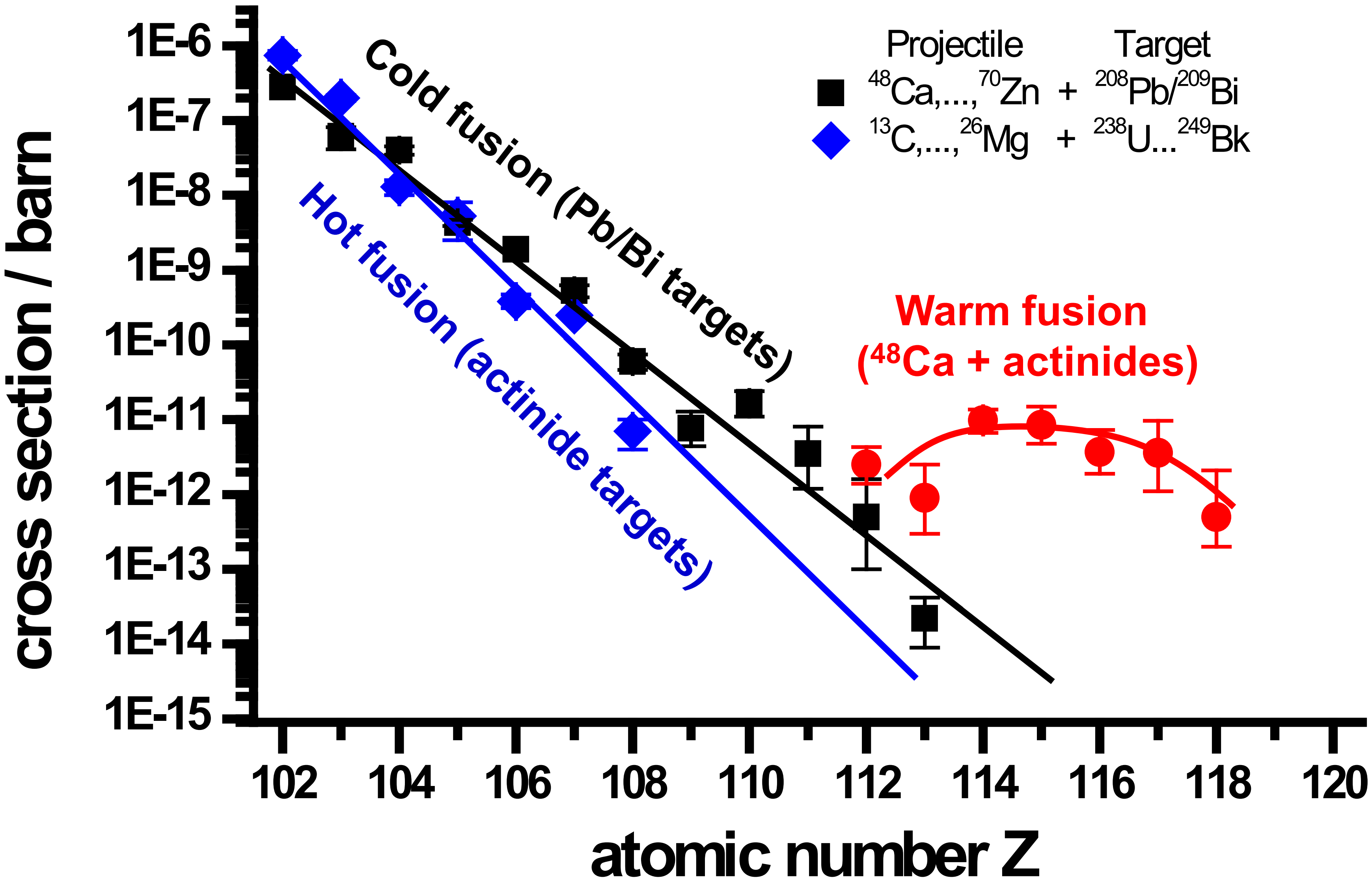
Part II: ^{48}Ca beam: chemical studies

Part III: Beyond ^{48}Ca and beyond ^{208}Pb

Cross sections decrease exponentially



Reactions leading to Z=119,120

Studied at GSI

E119

Z_{Beam}	Beam	Target	Asymmetry	E*@B_{Bass}
21	^{45}Sc	^{249}Cf		41.7
22	^{50}Ti	^{249}Bk		32.4
23	^{51}V	^{248}Cm		36.8
24	^{54}Cr	^{243}Am		31.5
25	^{55}Mn	^{244}Pu		37.7
26	^{58}Fe	^{237}Np		29.9
27	^{59}Co	^{238}U		36.7

E120

Z_{Beam}	Beam	Target	Asymmetry	E*@B_{Bass}
22	^{50}Ti	^{249}Cf		31.7
23	^{51}V	^{249}Bk		35.9
24	^{54}Cr	^{248}Cm		33.0
25	^{55}Mn	^{243}Am		34.5
26	^{58}Fe	^{244}Pu		33.9
27	^{59}Co	^{237}Np		32.9
28	^{64}Ni	^{238}U		27.3

One-event limits from GSI:

Search for 120 in ^{64}Ni+^{238}U:

E of ^{302}120: 36.4 MeV (c.o.t.)*

^{64}Ni + ^{238}U: 200 fb

S. Hofmann, Russ. Chem. Rev. 78 (2009) 1123

Search for 120 in ^{54}Cr+^{248}Cr:

^{54}Cr 304.8 MeV (c.o.t.)

^{54}Cr + ^{238}U: 580 fb

S. Hofmann et al., EPJA 52 (2016) 180
F.P. Heßberger et al., EPJA 53 (2017)1 123

Search for 120 in ^{50}Ti+^{249}Cf:

^{50}Ti @ 287.0 MeV (c.o.t.)

^{50}Ti + ^{249}Cf: 200 fb

J. Khuyagbaatar et al., PRC 102 (2020) 064602
Ch.E. Düllmann et al., unpublished

Search for 120→119 (^{249}Bk→^{249}Cf)

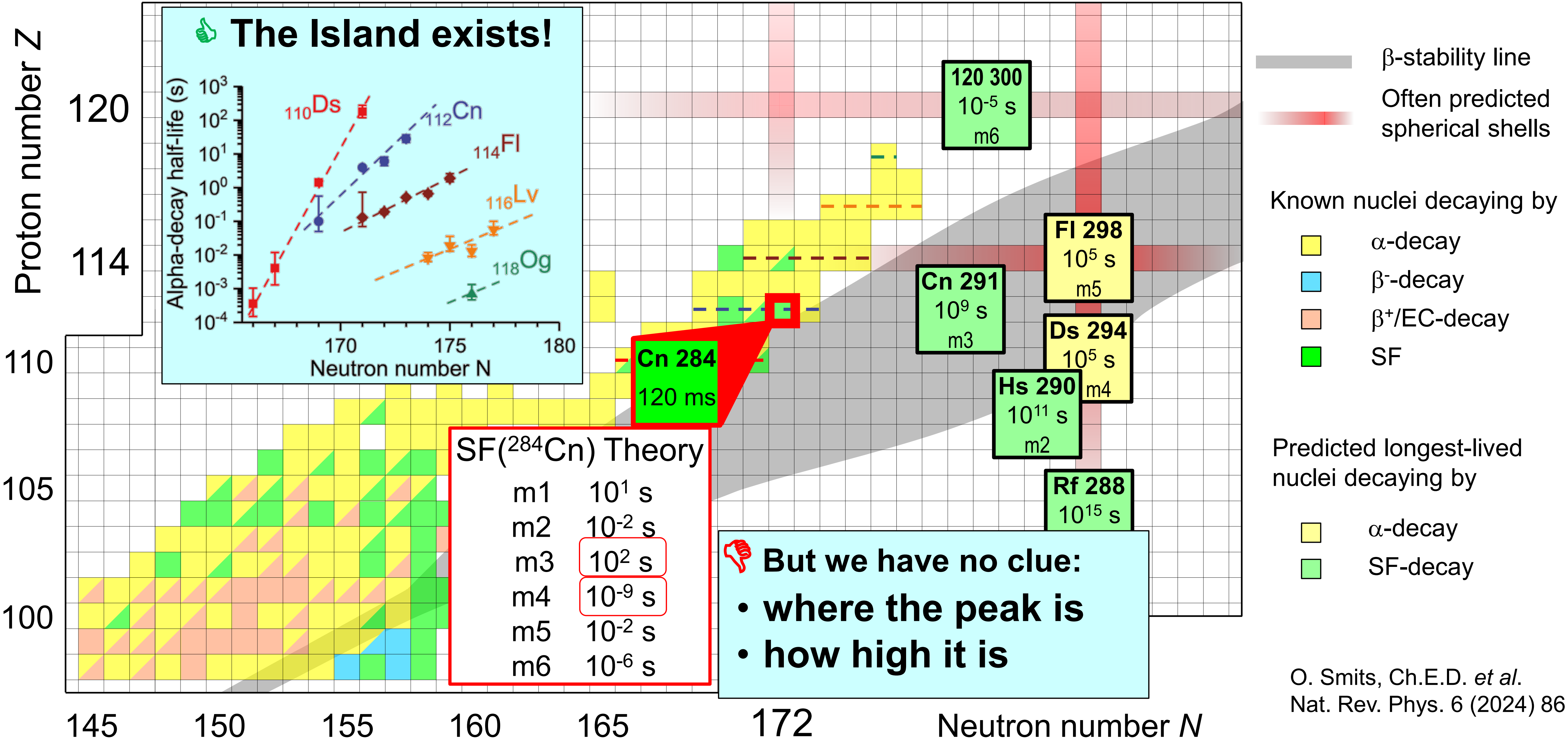
^{50}Ti @ 281.5 MeV (c.o.t.)

^{50}Ti + ^{249}Bk: 65 fb

^{50}Ti + ^{249}Cf: 200 fb

J. Khuyagbaatar et al., PRC 102 (2020) 064602

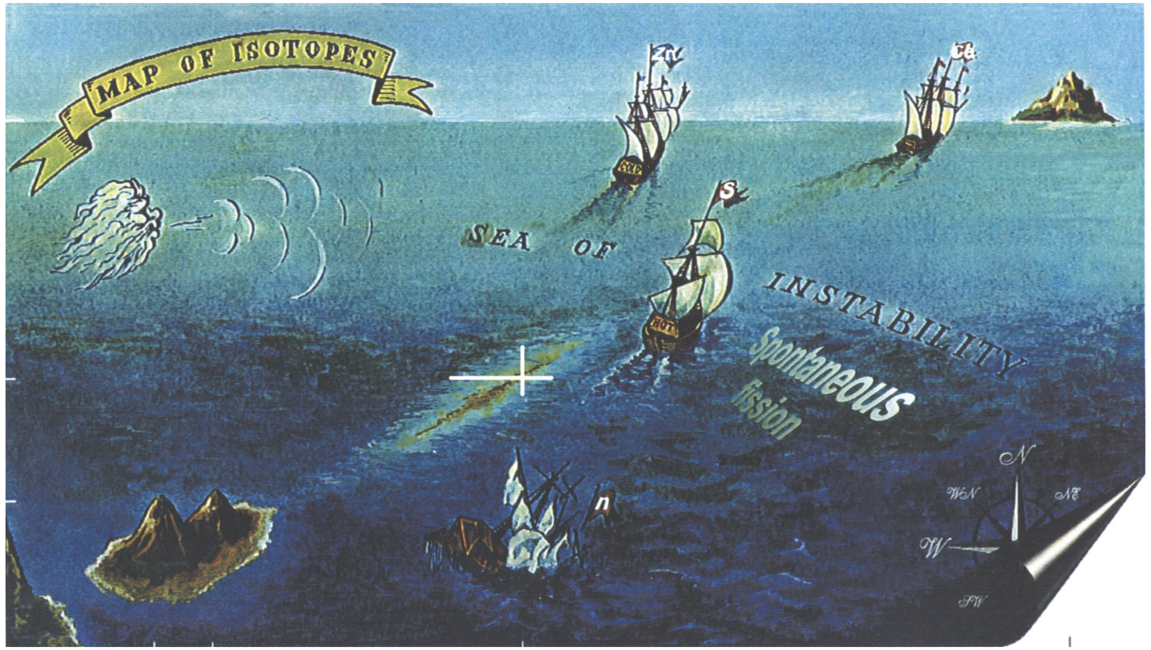
Island of Stability – Status 2024



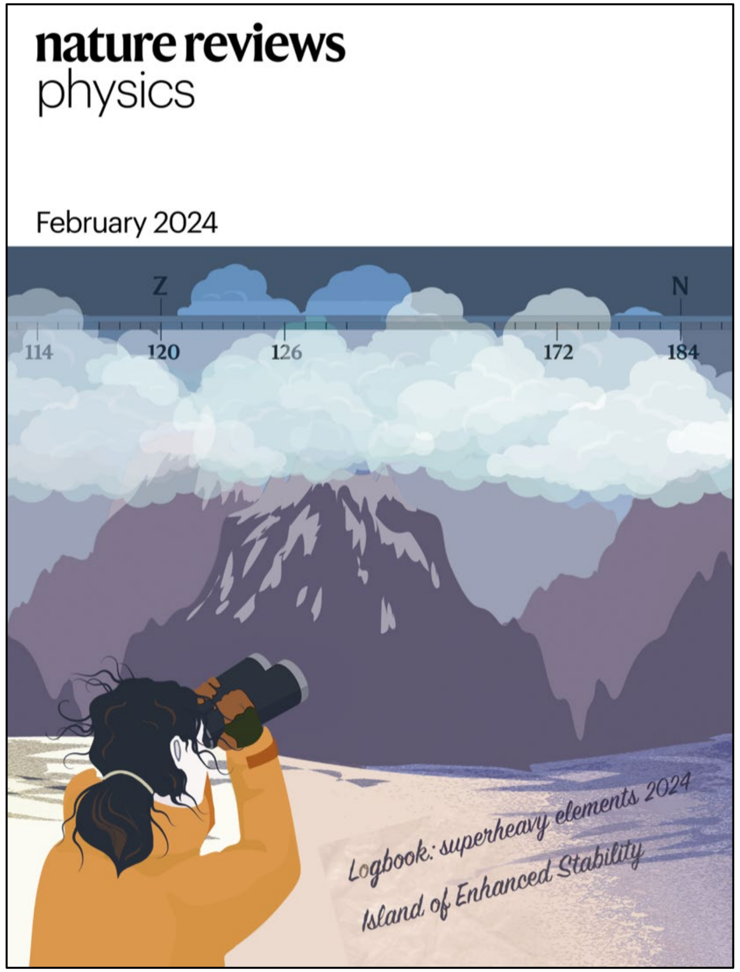
O. Smits, Ch.E.D. *et al.* Nat. Rev. Phys. 6 (2024) 86

The Island of Stability

Status 1960s



Today



Conclusions

- Cold fusion paved the way to element discovery beyond Z = 106
- High cross sections around ^{48}Ca + ^{208}Pb make this the „entry reaction“ for many new techniques SHE research
- Aspect of reaction temperature likely to remain critical for further advances to new elements
- Overarching goal remains the exploration and mapping of the island of increased stability and the end of the periodic table

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