The Quest for Superheavy Elements

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The "menu"

- Why are heavy elements important?
 - » -General
 - » -Chemistry
- The evolution of the Periodic Table
- Central Issues
 - How do you make new heavy nuclei?
 - » General
 - » Detailed
 - How do you do chemistry on short-lived rare species?
- The Way Forward
 - Synthesis of new heavy nuclei
 - New/Old Ways to do Chemistry

Importance of Heavy Element Research

- A laboratory to study nuclear structure and dynamics under the influence of large Coulomb forces
- High profile research
- Results deal with fundamental principles of chemistry and physics



Spectacular Advances of the Past Fifteen Years

- Discovery of elements 110,111,112, 113,114,116
- Synthesis of elements 113, 115, 117 and 118 by "hot fusion" reactions
- First chemistry of elements 106-114



The philosophy of the Periodic Table of the Chemical Elements

- The Periodic Table is NOT a list of chemical elements in order of their atomic numbers.
- The Periodic Table is a spatial representation of the elements based upon their chemical properties.
- The Periodic Table is a "living" document whose form is a matter of experiment.

Modern Periodic Table

	1																	18
	1 H	2											13	14	15	16	17	2 He
	3	4										1	5	6	7	8	9	10
	Li	Be											В	C	N	O	F	Ne
	11	12	2	1	_	-	7	0	0	10	11	10	13	14	15	16	17	18
1	Na	Mg	3	4	5	6	1	8	9	10	11	12	Al	Si	P	S	Cl	Ar
1	19	20	21	22	23	24	25	25	27	28	29	30	31	32	33	34	35	36
	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
100.62	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54
I	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
	55	56	57-71	74	75	75	77	788	79	80	81	82	83	84	85	86	87	88
(Cs	Ba	lanthanide series	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	T1	Pb	Bi	Po	At	Rn
1	87	88	89-103	104	105	106	107	108	109	110	111	112	113	114	115	116	447	118
	Fr	Ra	actinide series	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	113	Fl	115	Lv	117	118
	7																	
			89	90	91	92	93	94	95	96	97	98	99	100	101	102	103	
			Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	
																		90
			57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	
			La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Но	Er	Tm	Yb	Lu	

The genius of Mendeleyev

REIHEN	GRUPPE 1.	GRUPPE II.	GRUPPE III.	GRUPPE IV.	GRUPPE V.	GRUPPE VI.	GRUPPE VII	GRUPPE VIII.
RE	R20	RO	R203	RO2	R205	RH ² RO ³	RH R207	RO4
1	H=1						0	nor
2	Li= 7	Be = 9,4	B=11	C=12	N=14	0=16	F=19	
3	Na = 23	Mg = 24	A1 = 27,3	Si = 28				
4	K = 39	Ca = 40	-= 44	Ti = 48	V=51			Fe = 56, Co = 59.
5	(Cu = 63)	Zn = 65	-= 68	-= 72	As = 75	50 - 70		Ni = 59, Cu = 63.
6	Rb = 85			Zr = 90	Nb = 94	Se = 78 Mo = 96	Br = 80 -=100	Ru= 104, Rh= 104,
7	(Ag = 108)	Cd = 112	In=113	Sn=118	Sb=122	Te=125		Pd = 106, Ag = 108
8	Cs = 133	Ba = 137		?C8 = 140	_	_ 10-123	J=127	
9	(-)	_						
0		-	?Er = 178	?La=180	Ta = 182	W=184	_	0s = 195, Ir = 197,
1	(Au=199)	Hg = 200	TI = 204	Pb = 207	Bi = 208			Pt = 198, Au = 199
2	_			Th = 231		U=240		

The periodic table, circa 1940

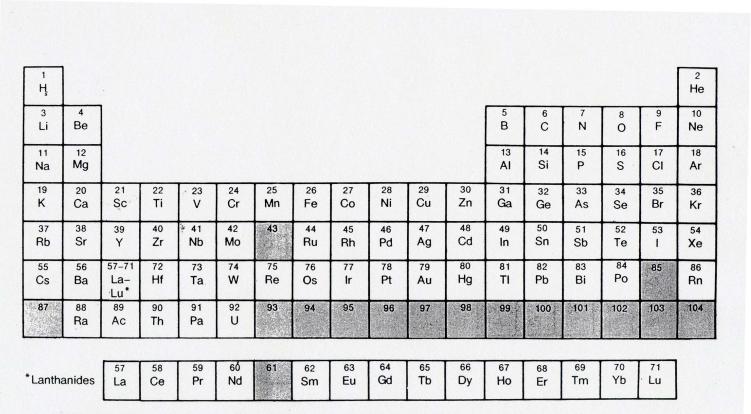
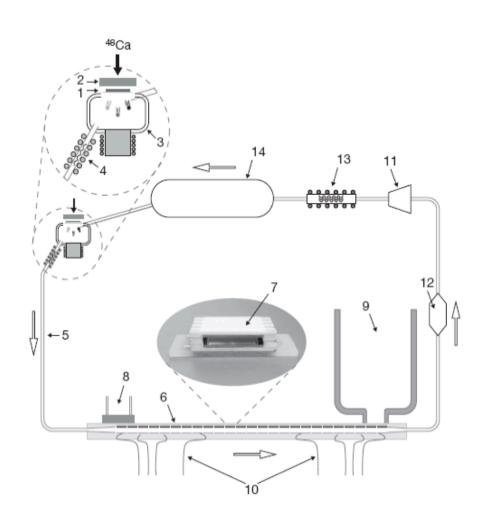
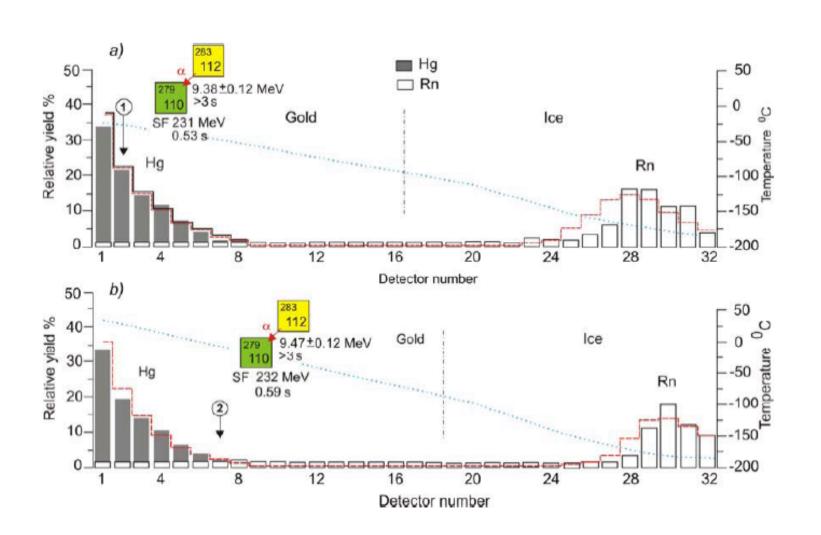


Figure 3.1 The periodic table of the 1930s; atomic numbers of then undiscovered elements are in shaded squares.

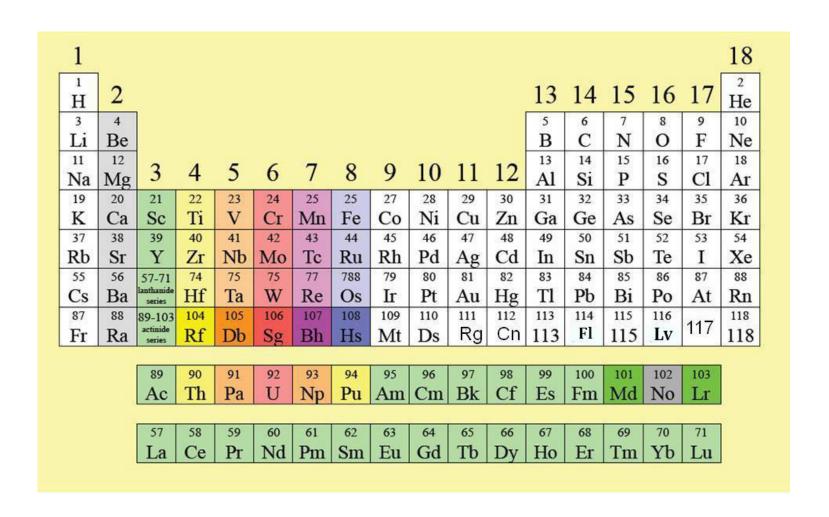
Chemistry Apparatus



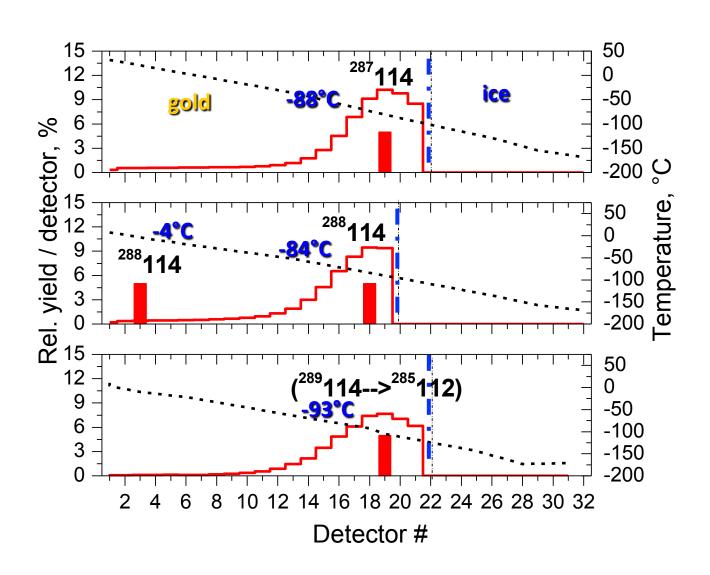
Chemistry of Element 112 (Cn)



Chemistry of the heaviest elements



114 Adsorption (2007/2008)



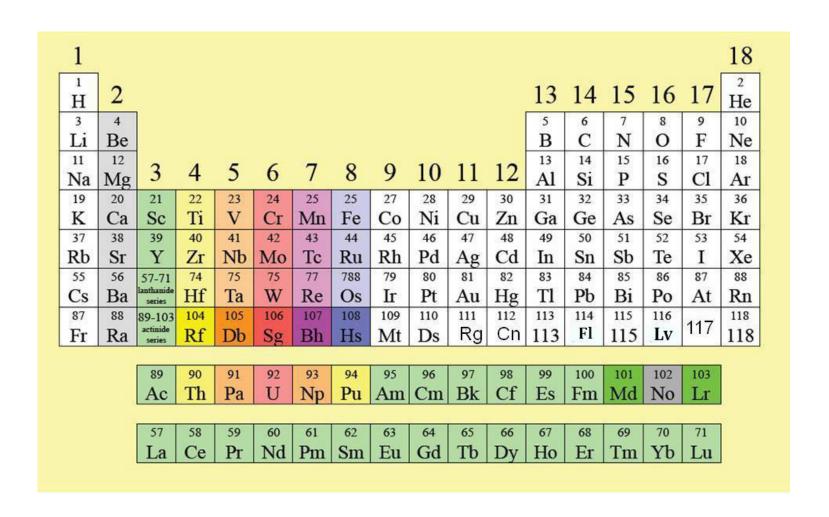
Chemistry of element 114 (FI)

• 3 events observed at Dubna by PSI group, corresponding to $^{287}114$, $^{288}114$, and $^{289}114$ from ^{48}Ca + $^{242,244}Pu$. Deposition temperatures were -72 (Au), -85(Au), and -128(ice). Conclude that element 114 seems to behave like a very volatile metal, with very weak interaction with Au - even weaker than element 112. Most likely gaseous at ambient temperatures.

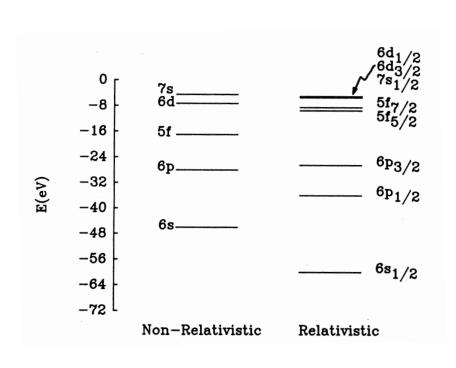
Further work on Fl

 A second thermochromatography experiment with Fl was done at GSI. The results were different from the initial study and suggested Fl was noble metal instead of being a noble gas. Further experiments are planned.

Chemistry of the heaviest elements



Relativity and the Periodic Table

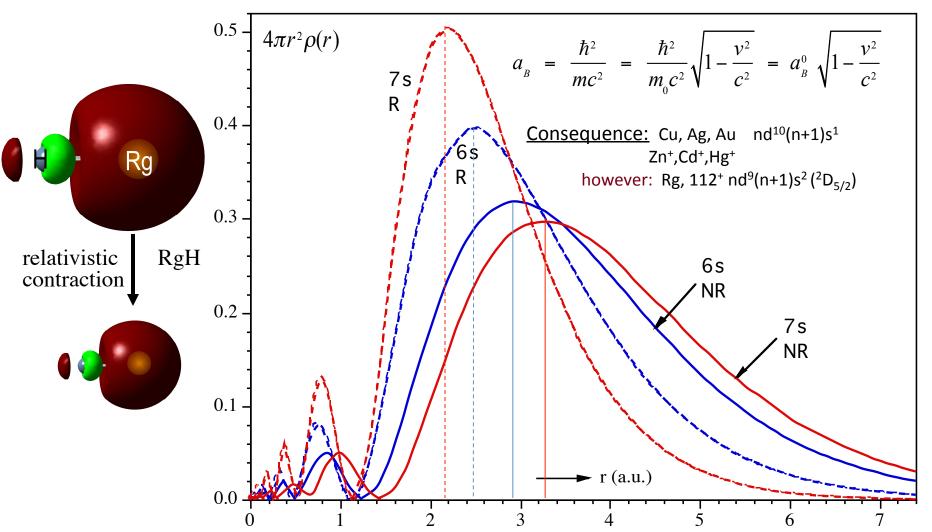


- v_{electron} ~ 0.5 c
- Relativistic effects should be important
- Primary effects
- (a) contraction of radii of $s_{1/2}$ and $p_{1/2}$ orbitals.
- (b) spin-orbit splitting
- (c) expansion of d and f orbitals



The relativistic 7s contraction in Au and Rg

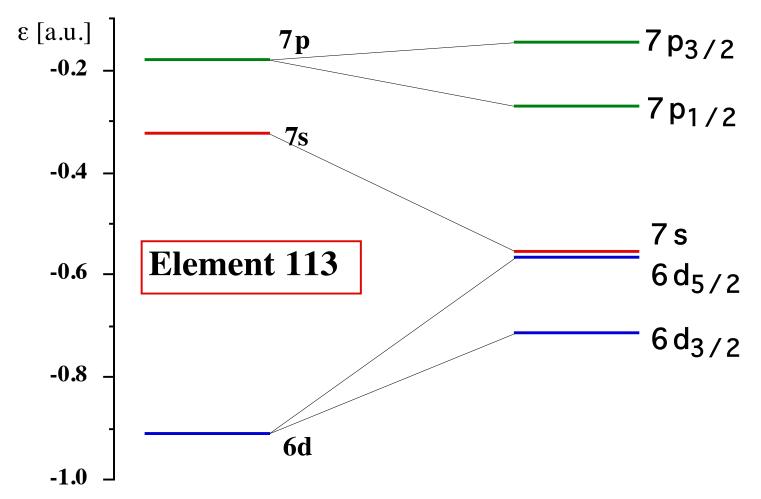
Courtesy of Peter Schwerdtfeger



E. Eliav, U. Kaldor, P. Schwerdtfeger, B. Hess, Y. Ishikawa, *Phys. Rev. Lett.* <u>73</u>, 3203 (1994). M. Seth, P. Schwerdtfeger, M. Dolg, K.Faegri, B.A. Hess, U. Kaldor, *Chem. Phys. Lett.* <u>250</u>, 461 (1996).

Relativistic shell-expansions and spin-orbit

Due to the increased relativistic shielding by the s-orbitals, the diffuse $p_{3/2}$ and higher angular momentum orbitals will expand relativistically



C. Thierfelder, P. Schwerdtfeger, A. Koers, A. Borschevsky, B. Fricke, *Phys. Rev. A* <u>80</u>, 022501-1-10 (2009).

Examples

- · Au is yellow instead of being white
 - In Au, for 6s electrons, v/c=0.58
- Radius of 6s orbital contracted by 22%
 - 5d→6s transition shifts from UV to visible
 - Enabled American gold rush



The End of Chemistry

- Does the Periodic Table have limits?
 YES!!
- At some point (Z~122) all the electron energy levels of adjacent elements are similar so that there are no differences in their chemical behaviour.

Recent Breakthroughs in SHE chemistry

- Synthesis of Sg(CO)₆
- Measurement of the first ionization potential of Lr
- Characterization of the chemistry and Periodic Table placement of Cn and Fl.

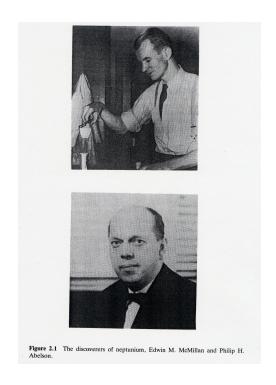
Heavy Element Nuclear Science

Making new elements by simple reactions

 The first man-made transuranium element, neptunium, Z=93

$${}^{238}_{92}U + {}^{1}_{0}n \rightarrow {}^{239}_{92}U + \gamma$$

$${}^{239}_{92}U \xrightarrow[t_{1/2}=23.5 \text{ min}]{}^{239}_{93}Np(t_{1/2} = 2.36 \text{ days})$$



Making new elements by simple reactions (cont.)

 The second man-made transuranium element, plutonium, Z=94

$${}^{238}_{92}\text{U} + {}^{2}_{1}\text{H} \rightarrow {}^{238}_{93}\text{Np} + 2{}^{1}_{0}\text{n}$$

$${}^{238}_{93}\text{Np} \xrightarrow[t_{1/2}=2.12 \text{ days}]{}^{238}\text{Pu}(t_{1/2} = 87.7 \text{ years})$$

The announcement of the discovery of Pu

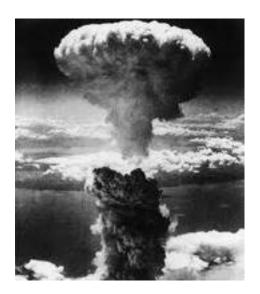


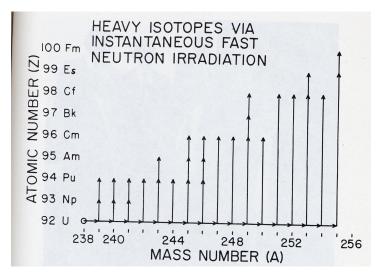




Figure 8
The co-discoverers of plutonium,
Joseph W. Kennedy (25 December
1940), Arthur C. Wahl and Glenn T.
Seaborg. Seaborg and Wahl are
shown (in February 1966) with the
sample of ²³⁹Pu in which fission was
demonstrated in 1941 (the cigar box
was that of G. N. Lewis).

Making new elements with nuclear weapons

• The synthesis of elements 99 (Md) and 100 (Fm)



Neutron Addition Process	Neutron Flux (n/cm ² s ¹)	Reaction Time	Neutron Exposure (n/cm ²)	Average Neutron Energy (keV)
High flux reactor	≈5 × 10 ¹⁵	0.5 years	≈10 ²³	2.5×10^{-5}
Stellar s process	≈10 ¹⁶	$\approx 10^3$ years	$\approx 10^{26}$	≈10
Stellar r process	≥10 ²⁷	1-100 s	>10 ²⁷	≈100
Nuclear explosion	>10 ³¹	$<10^{-6}$ s	≈10 ²⁵	≈20

MIKE

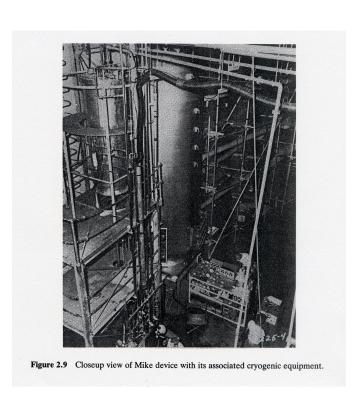




MIKE-2



Samples of the bomb debris were collected on filter papers by aircraft flying through the mushroom cloud



Using heavy ion reactions to make new elements—The Berkeley era



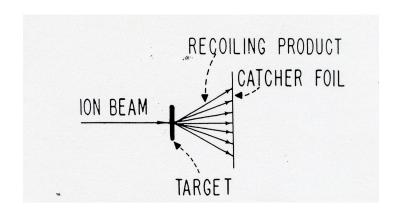


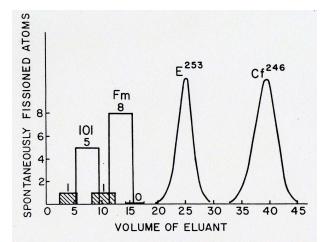
Albert Ghiorso

Glenn Seaborg

Synthesis of elements 101-106

- Making elements one atom at a time
- $^{253}Es + ^{4}He \rightarrow ^{256}Md + n$





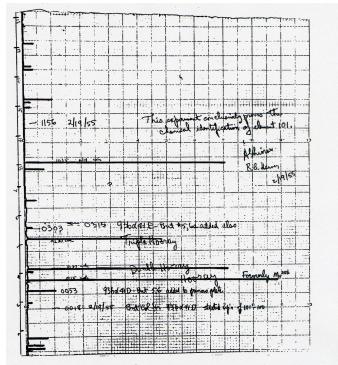
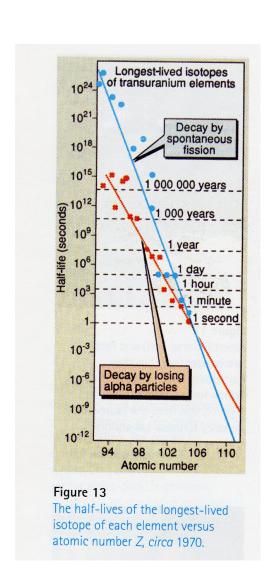


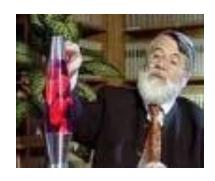
Figure 2.20 The ionization recording chart showing the first four events of the disintegration of mendelevium. The ordinate is the event time while the abscissa is the intensity of the ionization. The four pulses occurred at 1:15 a.m., 1:37 a.m., 2:40 a.m., and 10:35 a.m. on February 19, 1955. At 11:56 a.m., Ghiorso and Harvey made a note directly in the chart: "This experiment conclusively proves the chemical identification of element 101."

The problem



The Solution—The Darmstadt Era

- "Cold Fusion" Reactions
- Bombard Pb or Bi with heavy ions—the resulting species are borne "cold" -with low excitation energies—they survive better



Peter Armbruster



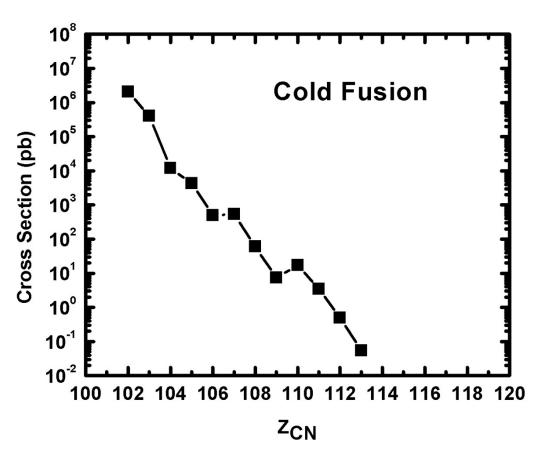
Sigurd Hofmann





Yuri Oganessian

The end of the "cold fusion" path



10 femtobarns ~ 1 atom/year: think about the human aspects of these experiments

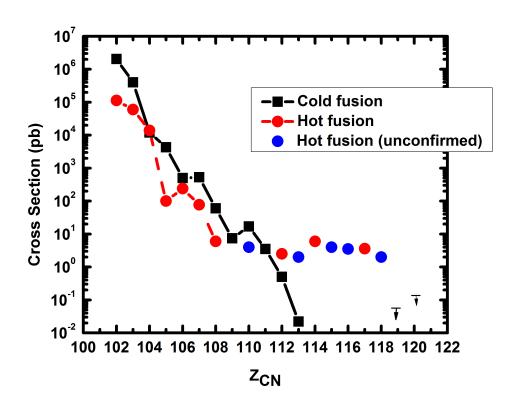
Cold and Hot Fusion

- Cold Fusion
- Pb or Bi Target
- Heavier Projectile (Ca-Kr)
- E* ~ 13 MeV (1n reaction, high survival)
- Significant fusion hindrance

- Hot (Warm) Fusion
- Actinide Target
- Lighter Projectiles (O-Ca)
- E* ~ 30 60 MeV (low survival)
- Small fusion hindrance



"Hot fusion-The Dubna Era"



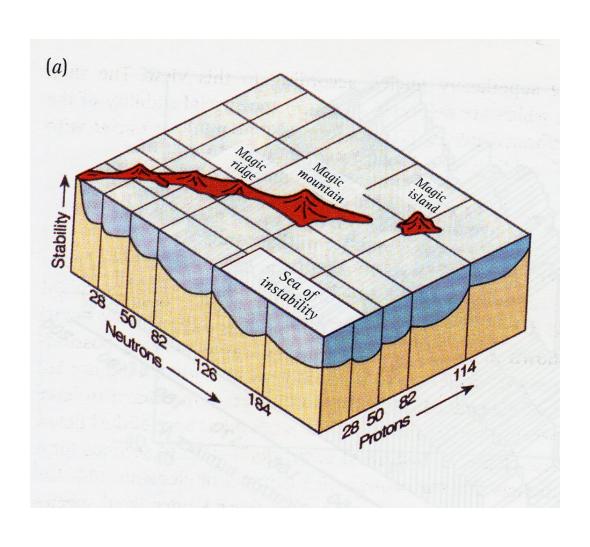


Yuri Oganessian

Are there nuclei with special stability?

- Nuclei with full shells of neutrons and protons.
- · Shell model of the nucleus
- Special stability associated with 2,8,20, 28,50,82,126 neutrons or protons. (These are called the "magic numbers")
- Where is the next proton magic number?

Allegorical View



Modern View

- The notion of an "island" may be wrong, i.e, there is a peninsula of known nuclei expected to connect to the "island"
- The position of the region of enhanced stability ("island") is a matter of controversy. N=184 is probably correct, but the proton number may be 114 or 120 or 126. Best experimental evidence favors 114, best theories suggest 120 or 126.
- Best theory suggests "magicity" may not be relevant, the region of enhanced stability may be broad.
- It is extremely difficult experimentally to get to N=184.

How do you make heavy nuclei?

- Starting material
- The latest synthesis of a chemical element was the synthesis of element 117 via the reaction ²⁴⁹Bk(⁴⁸Ca, 3n) ²⁹⁴117.
- The two-year experimental campaign began with a 250-day irradiation in HFIR, producing 22 milligrams of berkelium-249, which has a 320-day half-life. The irradiation was followed by 90 days of processing at REDC to separate and purify the berkelium. The Bk-249 target was prepared at Dimitrovgrad and then bombarded for 150 days at the Dubna facility.

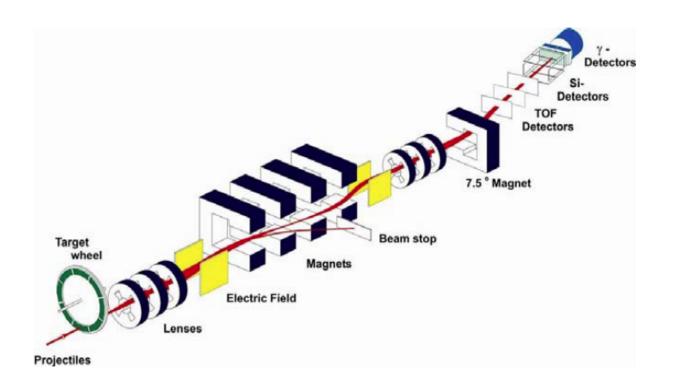
How do you make heavy nuclei?

Particle accelerators

Intense particle beams are needed.
Cross sections are ~picobarns, which
means one makes 1 atom per week. Cross
sections as low as 32 femtobarns have
been studied (1 atom/year)

How do you make heavy nuclei?

Separators



Production of Heavy Elements in Complete Fusion Reactions

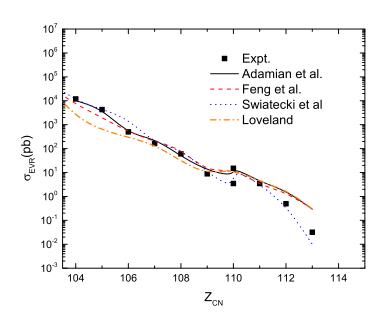
$$\sigma_{\text{EVR}}(E_{\text{c.m.}}) = \sum_{J=0}^{J_{\text{max}}} \sigma_{\text{CN}}(E_{\text{c.m.}}, J) W_{\text{sur}}(E_{\text{c.m.}}, J),$$

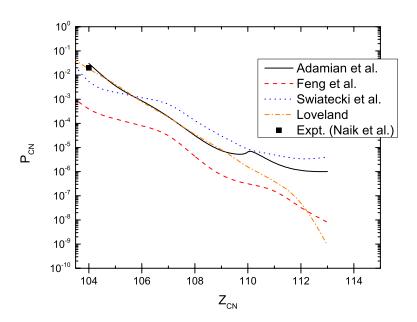
where

$$\sigma_{\text{CN}}(E_{\text{c.m.}}) = \sum_{J=0}^{J_{\text{max}}} \sigma_{\text{capture}}(E_{\text{c.m.}}, J) P_{\text{CN}}(E_{\text{c.m.}}, J),$$

 We need to know three spin-dependent quantities: (a) the capture cross section, (b) the fusion probability and (c) the survival probability, and their isospin dependence

How good are the predictions?





Capture Cross Sections

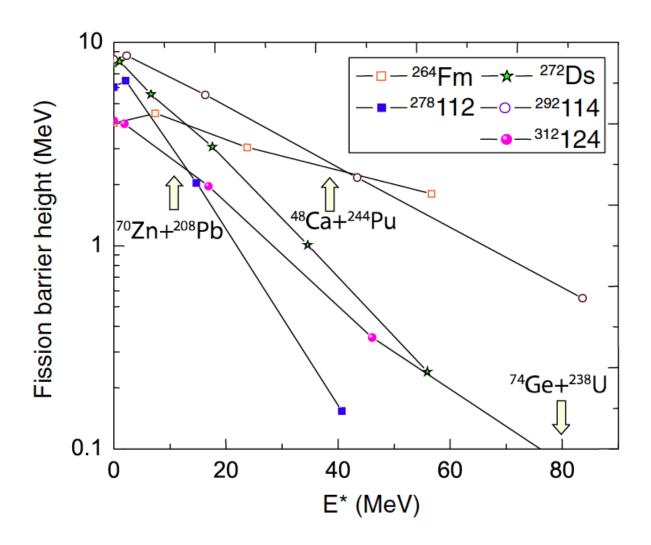
Table 1. Measured and predicted capture-fission cross sections

Proj.	Target	$^{\rm CN}$	$E_{c.m.}$	E^*	Expt.	$\sigma_{calc}[40]$	$\sigma_{calc}/\sigma_{meas}$
			(MeV)	(MeV)	(mb)	(mb)	
^{36}S	$^{208}\mathrm{Pb}$	²⁴⁴ Cf	153.9	40.	363.[43]	201.4	0.55
^{30}Si	$^{238}{ m U}$	268 Sg	133.9	40.	21[42]	18.1	0.86
58 Fe	$^{208}\mathrm{Pb}$	$^{264}\mathrm{Hs}$	245.5	40.	200.[41]	165.4	0.83
$^{26}\mathrm{Mg}$	$^{248}\mathrm{Cm}$	$^{274}\mathrm{Hs}$	122.3	40.	9[10]	17.9	2.0
^{34}S	^{238}U	$^{274}\mathrm{Hs}$	151.6	40.	20.[42]	13.3	0.67
48 Ca	^{238}U	^{286}Cn	199.2	40.	100.[41]	125.2	1.25
48 Ca	244 Pu	292 Fl	196.3	35	25.[41]	38.6	1.55
48 Ca	$^{248}\mathrm{Cm}$	$^{296}\mathrm{Lv}$	202.3	35	25[41]	56.1	2.24

Calculations done using coupled channels code (NRVP website) (Other recommended procedures are PRC 90 064622, PRC 83 054602, etc.) Capture cross sections known within a factor of 2. Is this good? NO—This is unacceptable. Capture cross sections are easy to measure and should be measured rather than calculated (a hybrid model).

Survival Probabilities (W_{sur})

- For the most part, the formalism for calculating the survival of an excited nucleus is understood.
- There are significant uncertainties in the input parameters for these calculations and the care needed to treat some situations.

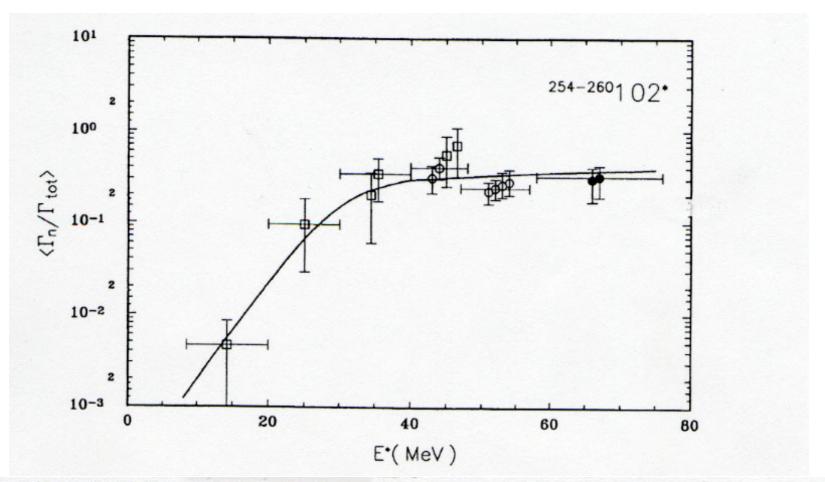


Pei et al., PRL 102, 192501

Mg + Cm
$$\rightarrow$$
 274Hs (PRL 112, 152702)

- $P_{CN} = 1$
- Subshell at N=162, Z=108
- Bf controversy
- ETFSI 2.50 MeV
- FRLDM 6.45 MeV
- Macro-Micro 4.37 MeV
 - DFT 5.1 MeV
- $(\Gamma_n/\Gamma_{tot})_1 = 0.89 + 0.13$
- Γ^{BW} is reduced by the effects of nuclear viscosity. (Kramers, 1940)

Why Hot Fusion Works



Andreyev AN et al 1994 Heavy-Ion Fusion: Exploring the Variety of Nuclear Properties (Singapore: World Scientific)
p 260

W_{sur} summary

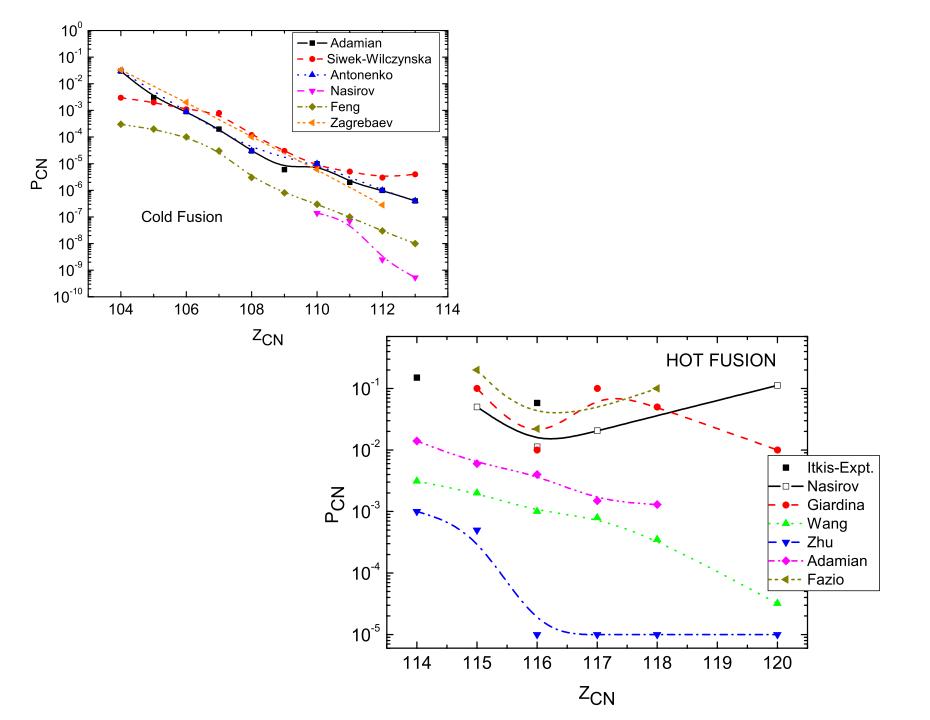
- Needed items
- Kramers correction
- Damping of shell effects
- Collective enhancement factors
- Pairing corrections
- E* (masses)
- B_f (uncertain to 0.5-1.0 MeV)

The Fusion Probability, P_{CN}

- Least well-known factor
- Hardest to measure
- A typical example

TABLE II. Predicted values of P_{CN} for the ¹²⁴Sn + ⁹⁶Zr reaction.

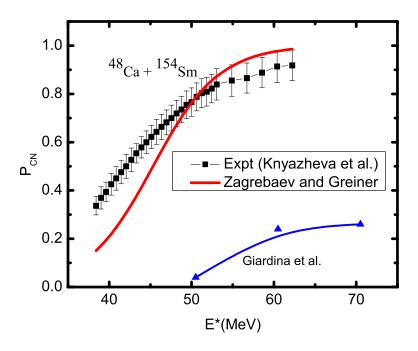
Predicted values of P_{CN}	Reference
0.56	[4]
0.13	[36]
0.008	[27]
0.0002-0.004	[37]



Excitation Energy Dependence of P_{CN}

Zagrebaev and Greiner

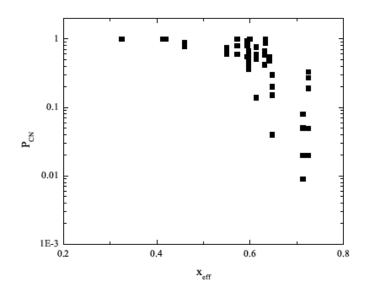
$$P_{CN}(E^*, J) = \frac{P_{CN}^0}{1 + \exp\left[\frac{E_B^* - E_{int}^*(J)}{\Delta}\right]}$$

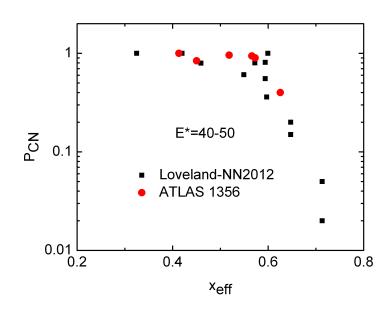


P_{CN} dependence on fissility

All data

E*=40-50 MeV





TDHF Calculations

- TDHF calculations appear to offer the best opportunity to understand (and possibly predict) P_{cN} .
- Wakhle et al. (PRL 113, 182502) use TDHF calculations to calculate P_{CN} for the reaction of ^{40}Ca with ^{238}U and its energy dependence.
- Their results agree well with the measurements of Shen et al. (PRC 36, 115)

The neutron-deficient character of our efforts

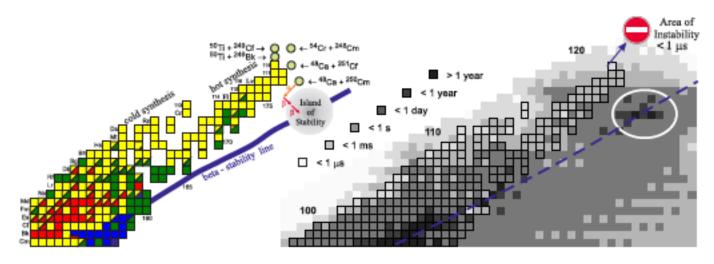
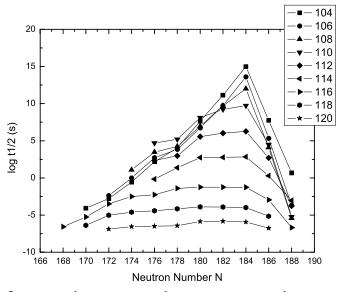


Fig. 1. Upper part of the nuclear map. Current and possible experiments on synthesis of SH elements are shown. (Right panel) Predicted half-lives of SH nuclei and the "area of instability". Known nuclei are shown by the outlined rectangles.

Zagrebaev, Karpov, and Greiner, Acta Physica Polonica B, 45,291 (2014)

Why use RNBs for producing new heavy nuclei?

 Longer half-lives of products enable more detailed atomic physics and chemical studies.

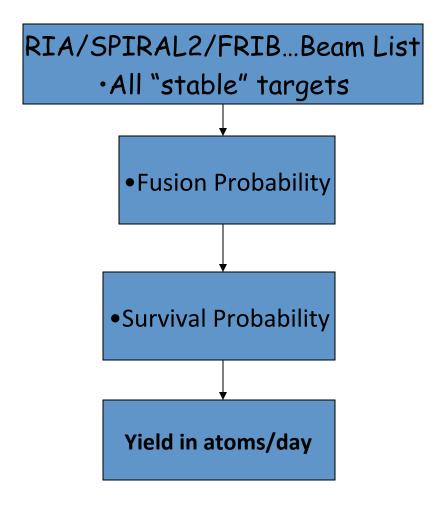


- Lowered fusion barrier due to n-rich projectiles allows lower E*
- Higher survival probabilities for n-rich products.

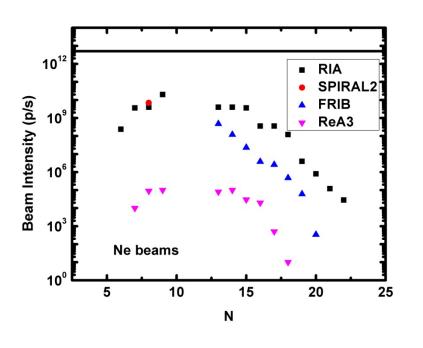


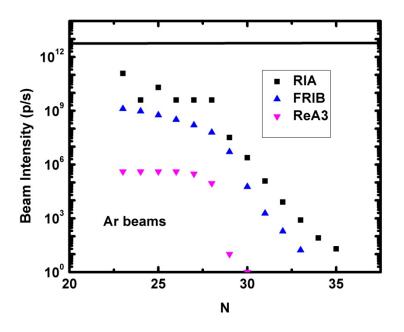
Applying what we know about the synthesis of the heaviest nuclei to the problem of making new heavy nuclei with radioactive nuclear beams

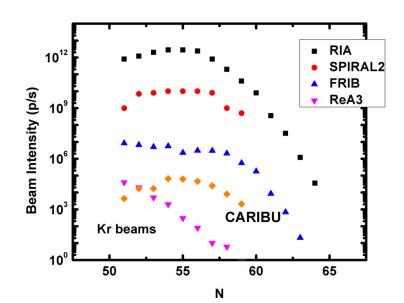
Calculational Model For RIB-Induced Reactions

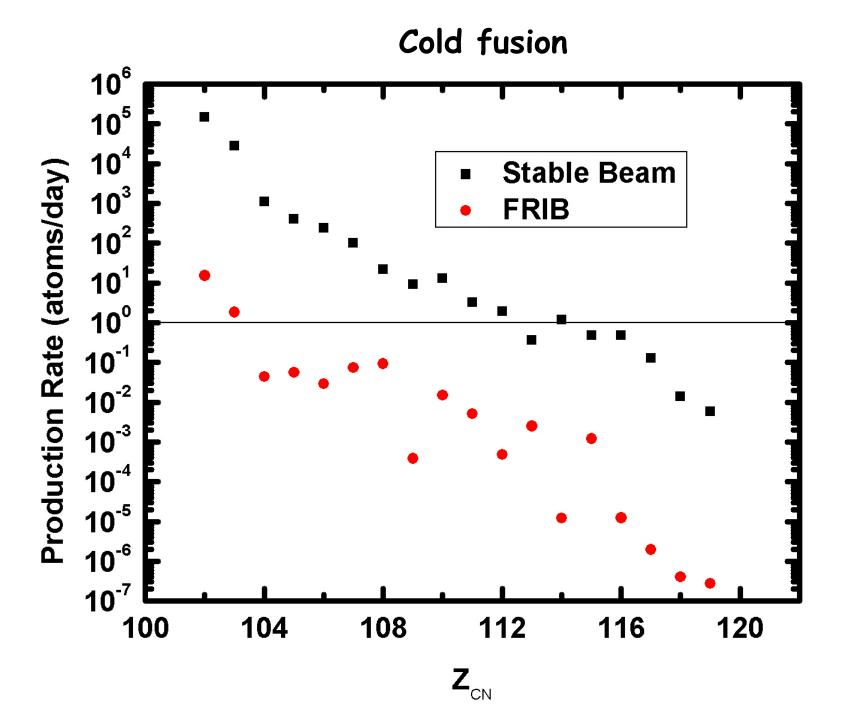


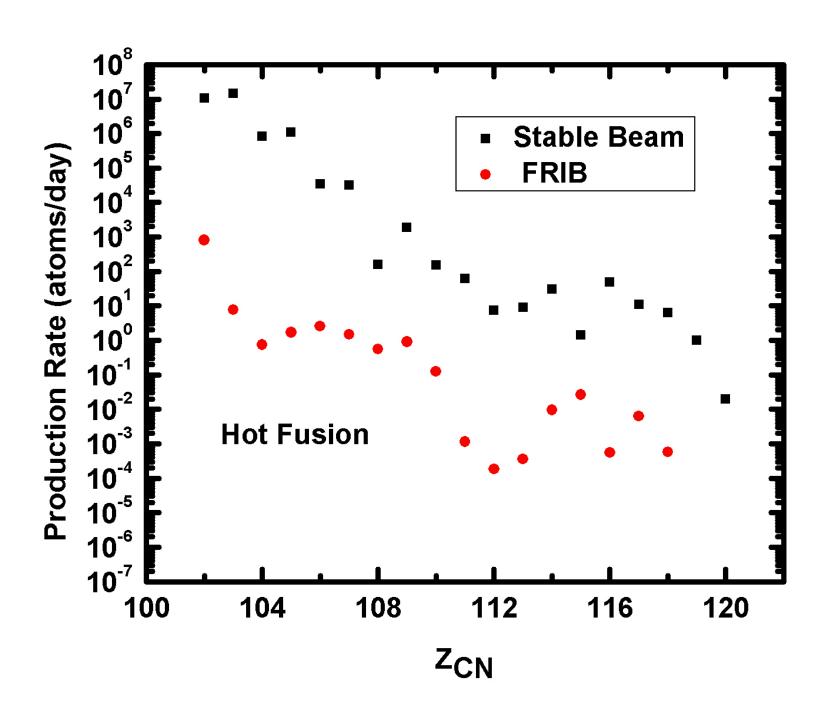












"Window to new n-rich heavy nuclei"

 There is a "window of opportunity" for making new nrich heavy nuclei using RIBs. The "window" is defined as a region where the cross sections and beam intensities lead to the production of > 10 atoms/day

Accelerator	Window
RIA	103-110
SPIRAL2	103-108
FRIB	103-107

What kind of reactions with RNBs are used to form n-rich nuclei?

Reactants	Products	FRIB Beam Intensity (p/s)	Production Rate (atoms/ day)
²³ O + ²⁵² Cf	²⁷¹ Sg + 4n	1.6 × 10 ⁶	1
³⁰ Mg + ²⁴⁴ Pu	²⁷⁰ Sg + 4n	2.1×10^7	3.2
²¹ O + ²⁵² Cf	²⁶⁹ Sg + 4n	5.0 × 10 ⁶	7.8
²⁰ O + ²⁵² Cf	²⁶⁸ Sg + 4n	4.3 × 10 ⁸	1200
²⁵ Ne + ²⁴⁶ Cm	²⁶⁷ Sg + 4n	2.3×10^7	3.2

All of the above are new isotopes of Sg

Atomic Physics and Chemistry of the Transactinides

>10 atom/day list

```
>^{265}Rf ^{252}Cf(^{16}C, 3n)

>^{266}Db ^{252}Cf(^{19}N, 5n)

>^{268}Sg ^{252}Cf(^{20}O, 4n)

>^{268}Bh ^{248}Cm(^{25}Na, 5n)

>^{264}Lr ^{248}Cm(^{19}N, 3n)
```

Reactions with Radioactive Beams

- The key factor is the production rate, not the cross sections, as RNB intensities are frequently low.
- One will not make new superheavy elements using radioactive beams.
- Most promising cases are reactions induced by light n-rich projectiles

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>^{264}Rf ^{252}Cf(^{16}C,4n)

>^{265}Db ^{249}Bk(^{20}O,4n)

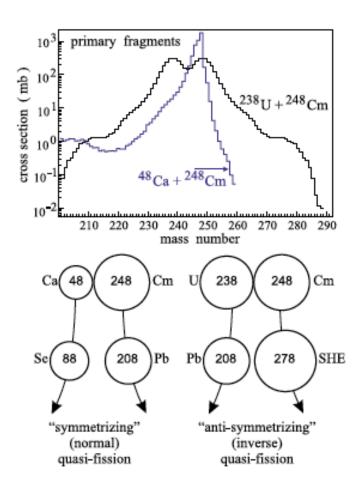
>^{268}Sg ^{252}Cf(^{20}O,4n)

>^{267}Bh ^{252}Cf(^{21}F,6n)
```

Multinucleon Transfer Reactions

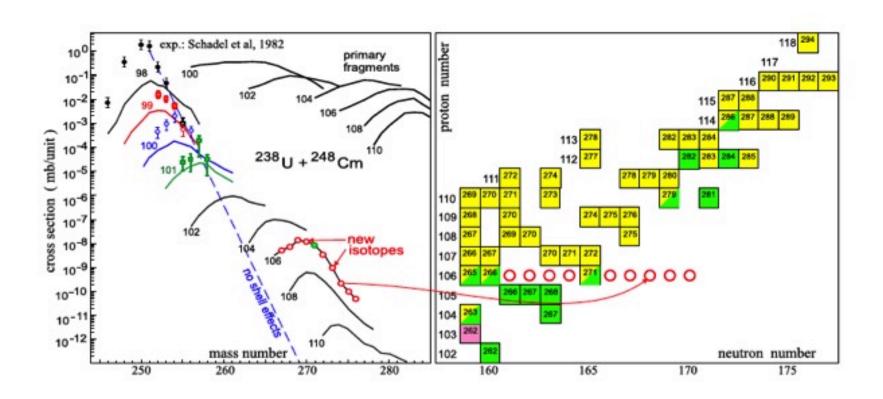
- The pioneering radiochemical studies of the 1970s and 80s at LBNL and GSI.
- The basic problem in making heavier nuclei was that the higher excitation energies that led to broader isotopic distributions caused the highly excited nuclei to fission.
- The contribution of Zagrebaev and Greiner to to emphasize the role of shell effects in these transfer reactions.

The importance of shell effects



V.I. Zagrebaev and W. Greiner, NPA (in press)

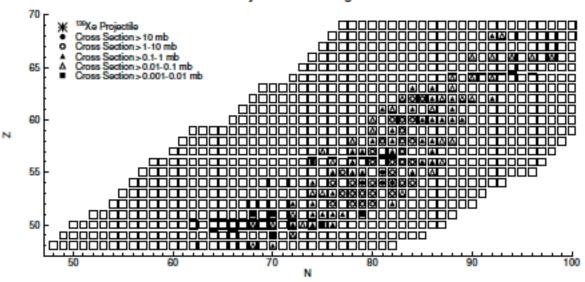
Multi-nucleon transfer reactions

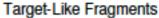


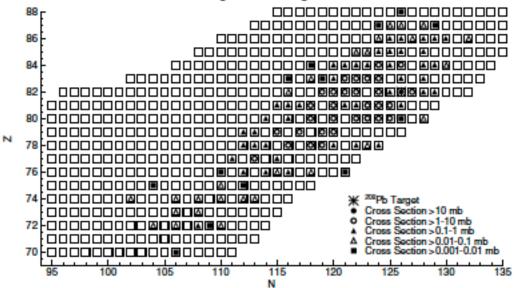
Can we test these prescriptions of Zagrebaev and Greiner?

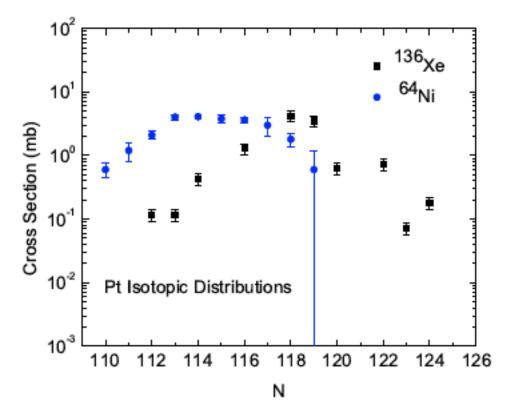
- Gammasphere study
- Over 230 nuclidic production cross sections were measured.

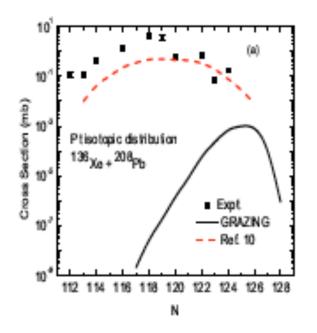
Projectile-Like Fragments

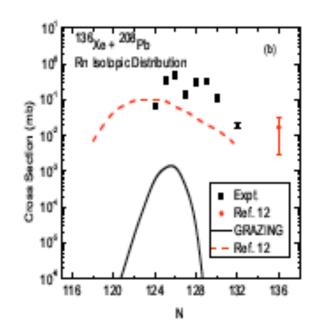












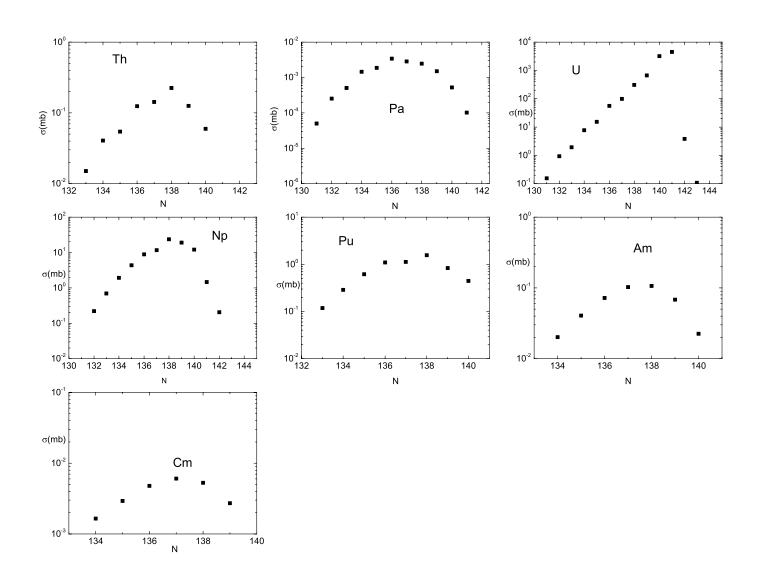
GRAZING is NOT a suitable model for large proton transfers

In all tests to date, Z-G theory agrees with or underestimates the MNT cross sections

Problem going forward

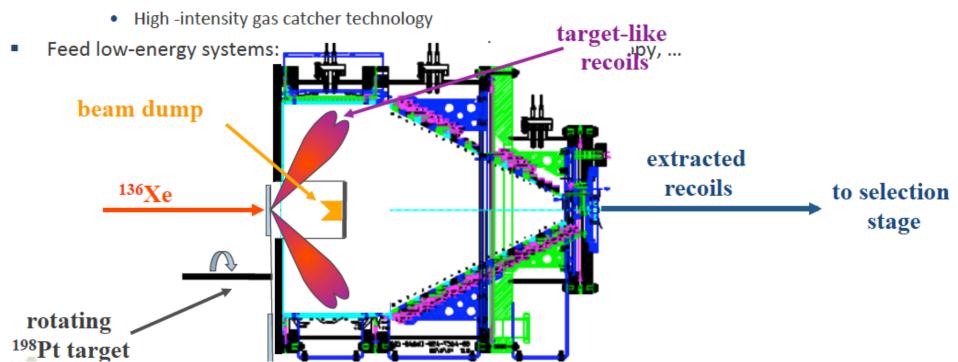
 All n-rich actinide products are long-lived beta emitters

FMA Experiment ¹³²Xe + ²³³U

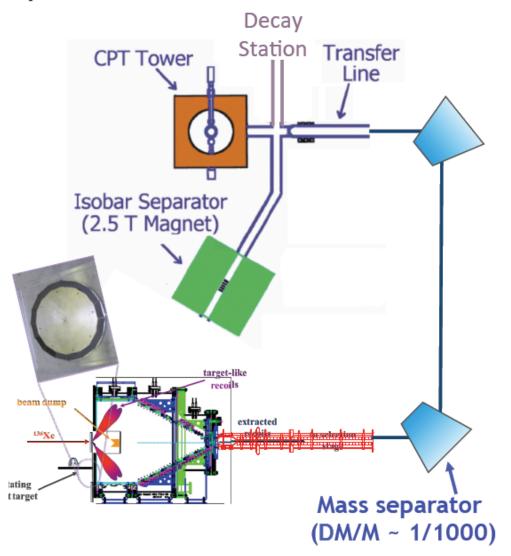


N=126 Factory--Savard

- Proposed collection system capitalizes on
 - · High primary beam intensity



The CPT-II apparatus and low-energy stations for deep-inelastic reaction products



- Designed to push back space charge limit
 - RFQ ion guide now operating in DC mode to avoid space charge build up
 - Rough mass separation by inflight mass separator before isobar separator
 - Rest of system essentially the same
- Can operate at up to 5-50 pmA while still providing required selection before precision Penning trap
- Deep inelastic reactions down to ~0.01 mb ... around ¹⁹⁸Hf on N=126 line

Conclusions about MNT

- A number of experiments confirm the validity of Z-G approach.
- Need to extend the tests of MNT to actinide nuclei
- A way forward for studying n-rich βemitters is under construction.

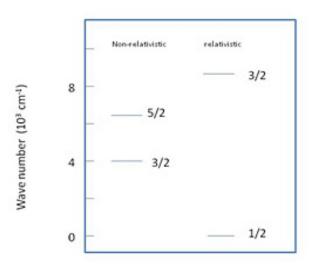
What kind of "chemistry" are we going to do?

- To confront the predictions of relativistic quantum chemistry with data, one would like to measure quantities that are directly calculable.
- Much of the wonderful atom by atom chemistry of molecular properties requires significant empirical extrapolations to be compared to data.
- These considerations drive us to focus on atomic properties that are directly measurable.

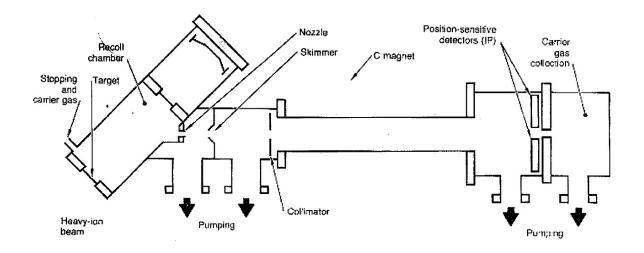
The Stern-Gerlach Experiment

- In the mid 1980's, a group at Livermore (Hulet, Cowan, Bandong, Moody, et al.)
 proposed to do a Stern -Gerlach experiment with Lr/Rf to measure the magnetic
 moment of Lr/Rf atoms. The experiment failed for technical reasons but it
 remains an intriguing possibility.
- The motivation is as follows:

Lr $5f^{14}7p7s^2$ vs. $5f^{14}6d7s^2$ Rf $6d7p7s^2$ vs $6d^27s^2$



Comparison of predicted relativistic and non-relativistic energy levels for Lr.



Production of Lr/Rf ion beam

Neutralize ion beam

Inhomogeneous magnetic field

Position sensitive detectors

Traps

 Use of traps to measure heavy nuclear masses with high accuracy

Produce nuclei with a heavy ion reaction.

Separate EVRs from beam with separator

Stop ions in gas cell

Transfer to trap

Measure masses

Implementation SHIPTRAP: Masses of ²⁵²⁻²⁵⁴No and their decay products

Conclusions

- Reviewed the history of heavy element discovery
- Summarized the chemical issues involved and a possible way forward
- Discussed the issues involved in the synthesis of new heavy nuclei and the expected future paths.

The End

Thanks!

Acknowledgements

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