Heavy Element Synthesis Reaction Mechanisms

W. Loveland
Oregon State University

Production of Heavy Elements in Complete Fusion Reactions

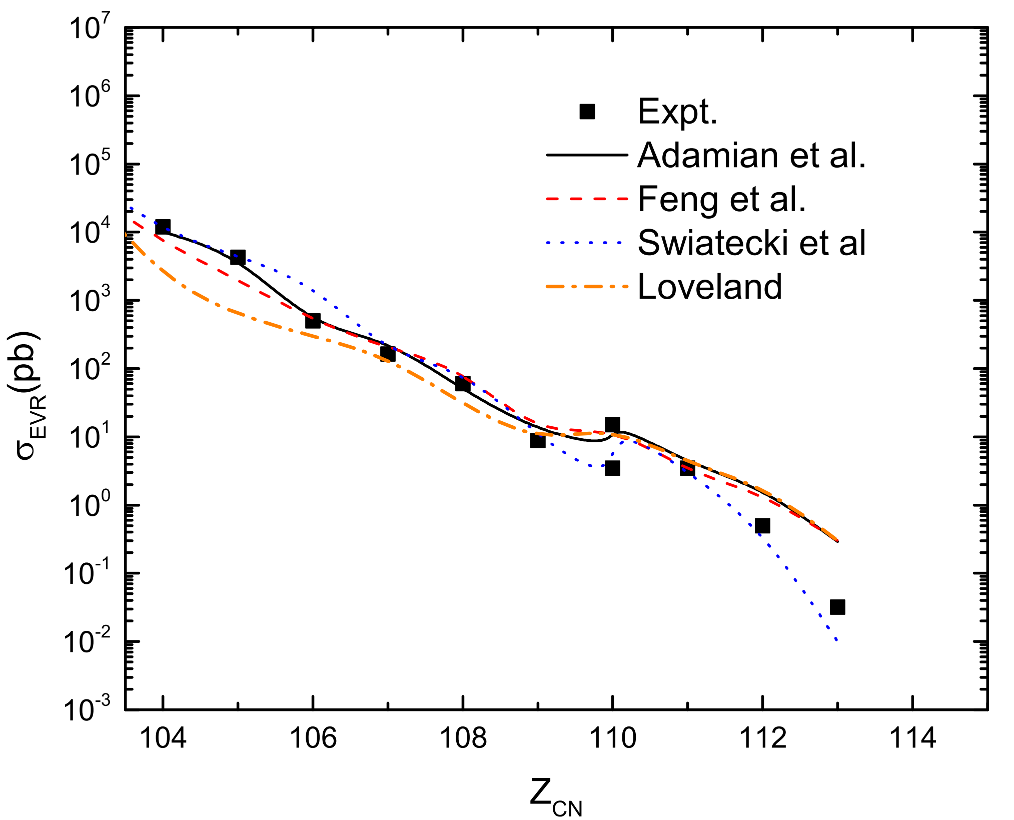
$$\sigma_{\mathrm{EVR}}(E_{\mathrm{c.m.}}) = \sum_{J=0}^{J_{\max}} \sigma_{\mathrm{CN}}(E_{\mathrm{c.m.}}, J) W_{\mathrm{sur}}(E_{\mathrm{c.m.}}, J),$$

where

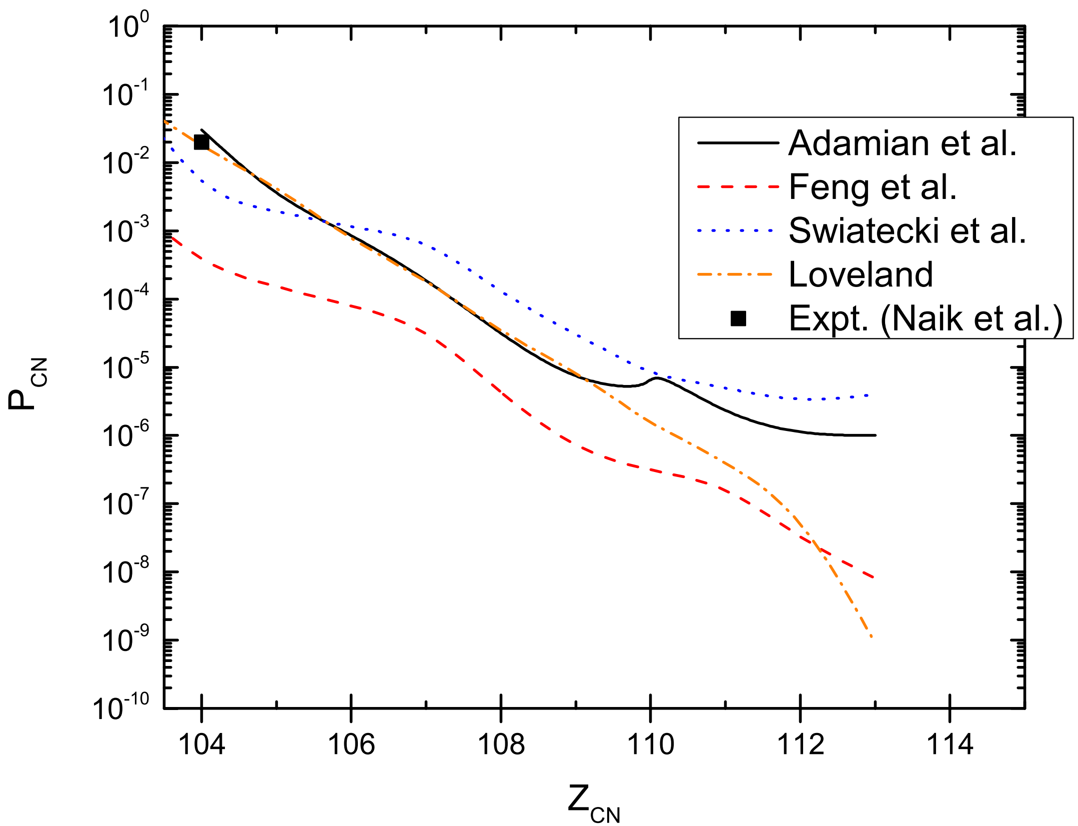
$$\sigma_{\mathrm{CN}}(E_{\mathrm{c.m.}}) = \sum_{J=0}^{J_{\max}} \sigma_{\mathrm{capture}}(E_{\mathrm{c.m.}}, J) P_{\mathrm{CN}}(E_{\mathrm{c.m.}}, J),$$

- 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

Examples of cold fusion predictions

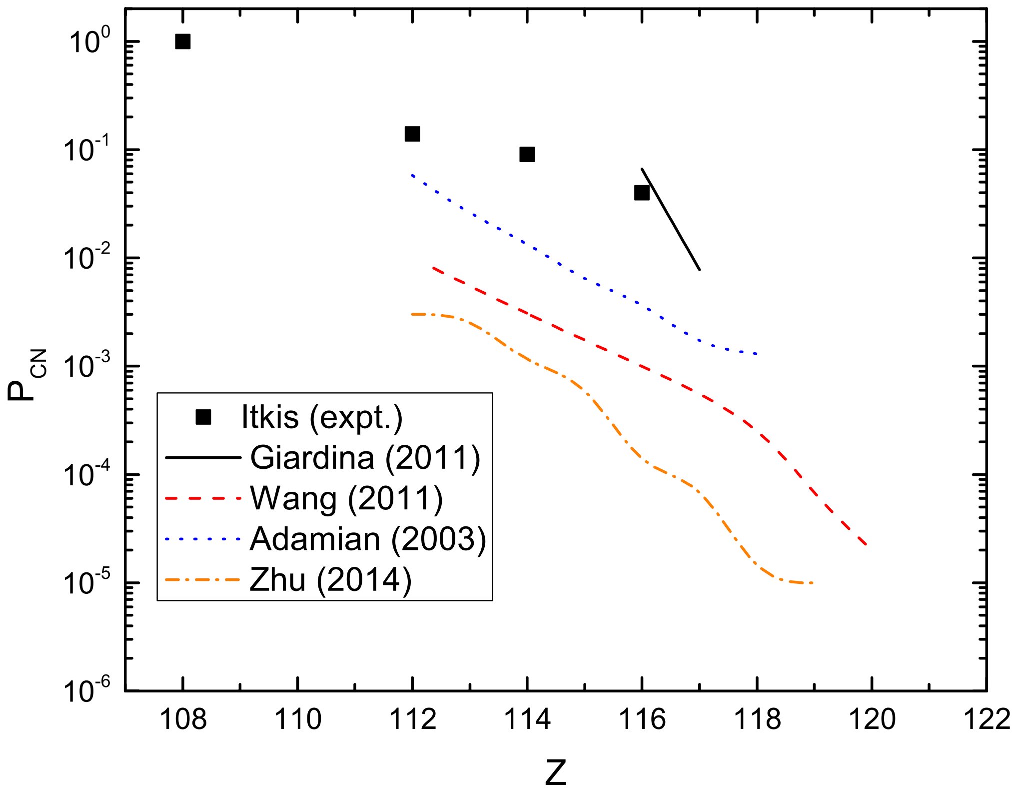


The problem



10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
P_{CN}
104
106
108
110
112
114
Z_{CN}
Adamian et al.
Feng et al.
Swiatecki et al.
Loveland
Expt. (Naik et al.)

Hot fusion examples



"How good are the model predictions of cross sections"

- Very controversial

Zagrebaev et al. (2001)

"The performed analysis makes it possible to conclude at present we are capable of calculating and predicting the values of formation cross sections of super-heavy elements with $Z > 112$ with an accuracy of two orders of magnitude at best"

Adamian, Antonenko and Scheid (2004)

"The estimated inaccuracy of our calculations of σ_{ER} is within a factor of 2-4"

Zagrebaev and Greiner (2015)

"One may conclude that our ability for the predictions of the cross sections for the production of super-heavy nuclei (both in fusion and transfer reactions) is not so good. It is different for different reactions and different combinations, but on average, accuracy of such predictions is not better than one order of magnitude"

Zagrebaev (2014)

"For unknown systems like symmetric reactions, the uncertainties will be several orders of magnitude. For reactions involving ^{50}Ti, such as the current efforts to synthesize elements 119 and 120, the uncertainties will be large. For extrapolations to the use of targets like ^{254}Es, the uncertainties will be at least one order of magnitude. But for reactions involving ^{48}Ca and "familar" targets and products, the uncertainties will be a factor of 2.

Capture Cross Sections

Table 1. Measured and predicted capture-fission cross sections

Proj.	Target	CN	$E_{c.m.}$ (MeV)	E^* (MeV)	Expt. (mb)	σ_{calc}[40] (mb)	$\sigma_{calc}/\sigma_{meas}$
^{36}S	^{208}Pb	^{244}Cf	153.9	40.	363.[43]	201.4	0.55
^{30}Si	^{238}U	^{268}Sg	133.9	40.	21[42]	18.1	0.86
^{58}Fe	^{208}Pb	^{264}Hs	245.5	40.	200.[41]	165.4	0.83
^{26}Mg	^{248}Cm	^{274}Hs	122.3	40.	9[10]	17.9	2.0
^{34}S	^{238}U	^{274}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}Cm	^{296}Lv	202.3	35	25[41]	56.1	2.24

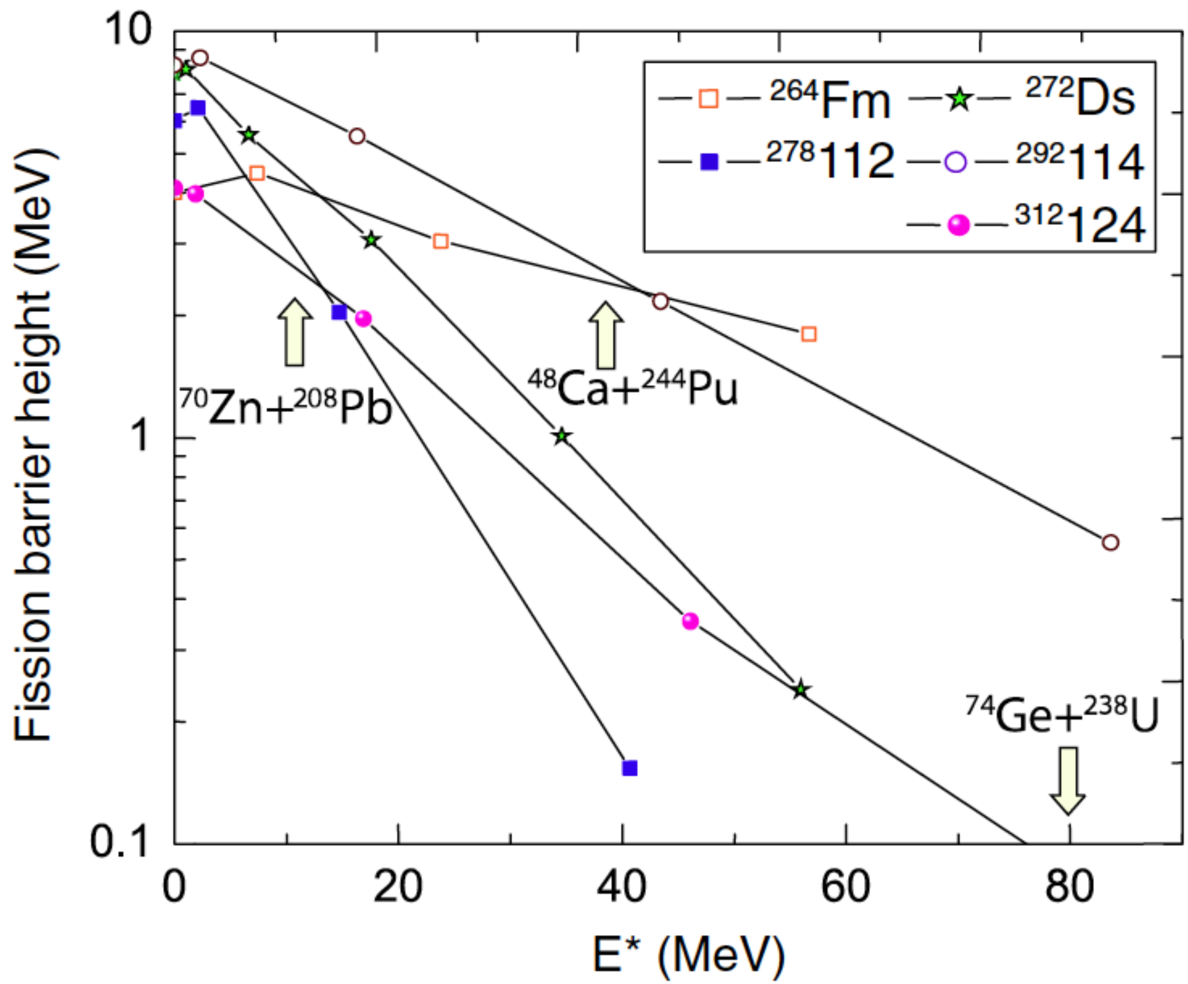
Calculations done using coupled channels code (NRVP website) (Other recommended procedures are PRC 90 064622, PRC 83 054602, etc.)

Capture cross sections known within a factor of 2. Is this good?

NO—THIS IS NOT ACCEPTABLE. Capture cross sections are easy to measure and should be measured rather than calculated.

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 $\rightarrow$ ^{274}Hs (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
- $(\Gamma_n/\Gamma_{tot})_1 = 0.89 \pm 0.13$
- Γ^{BW} is reduced by the effects of nuclear viscosity. (Kramers, 1940)

Other factors in W_{sur}

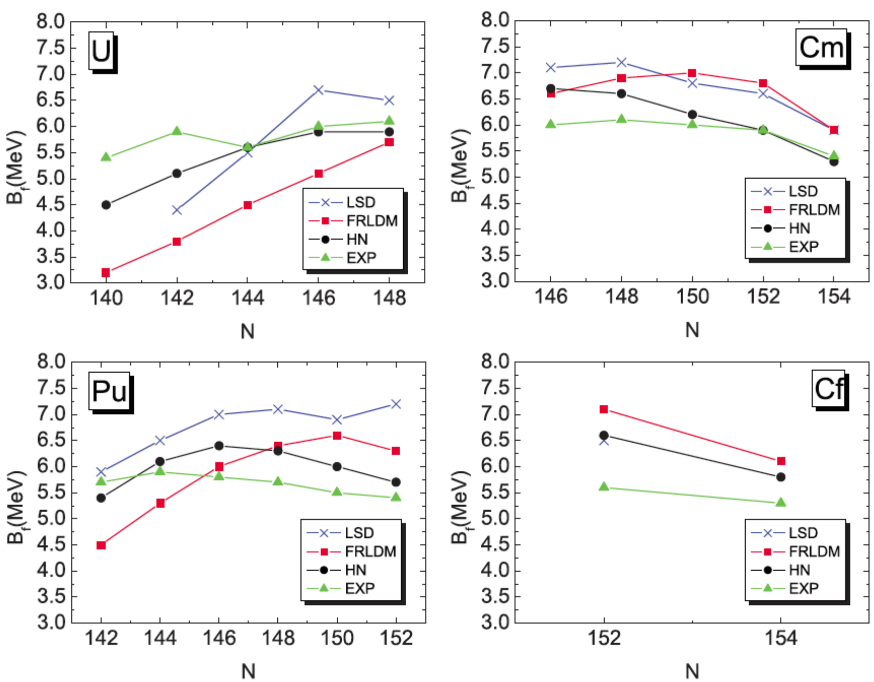
- Lu and Boilley (EPJ Web of Conf. 62, 03002) Collective effects, B_f, Kramers effects most important.
- B_f uncertainty
- Ajanasjev et al/ (IJMPE 21, 1250025) –average deviation between calculated and measured inner barrier heights was 0.7 MeV
- Bertsch et al. (J. Phys. G submitted) estimate the uncertainties in fission barrier heights to be 0.5-1.0 MeV in known nuclei.
- Sobiczewski et al. have extensively studied the uncertainties in nuclear masses and fission barriers for different models.
- For Z=92-98, the average discrepancy between measured and calculated barrier heights is 0.8 MeV

Survival Probability
(How well do we know the isospin dependence of Bf?)

PHYSICAL REVIEW C **82**, 014303 (2010)

Fission barriers for even-even superheavy nuclei

M. Kowal,[1,2,*] P. Jachimowicz,[2,3] and A. Sobiczewski[1,2]



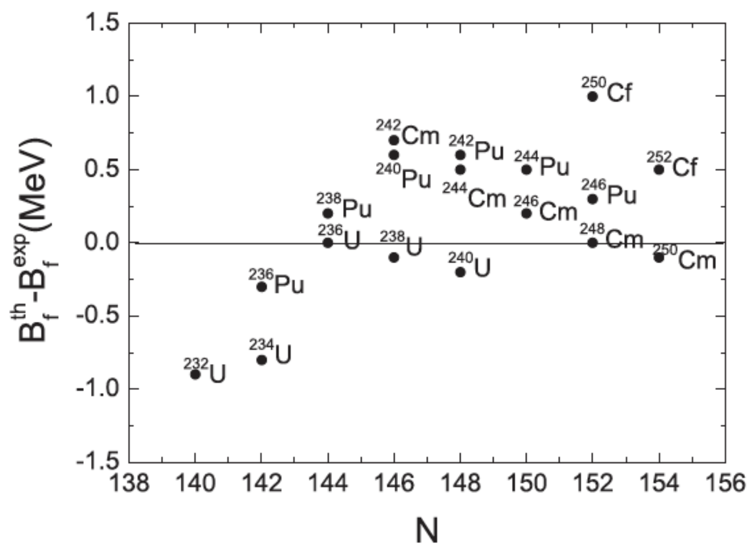
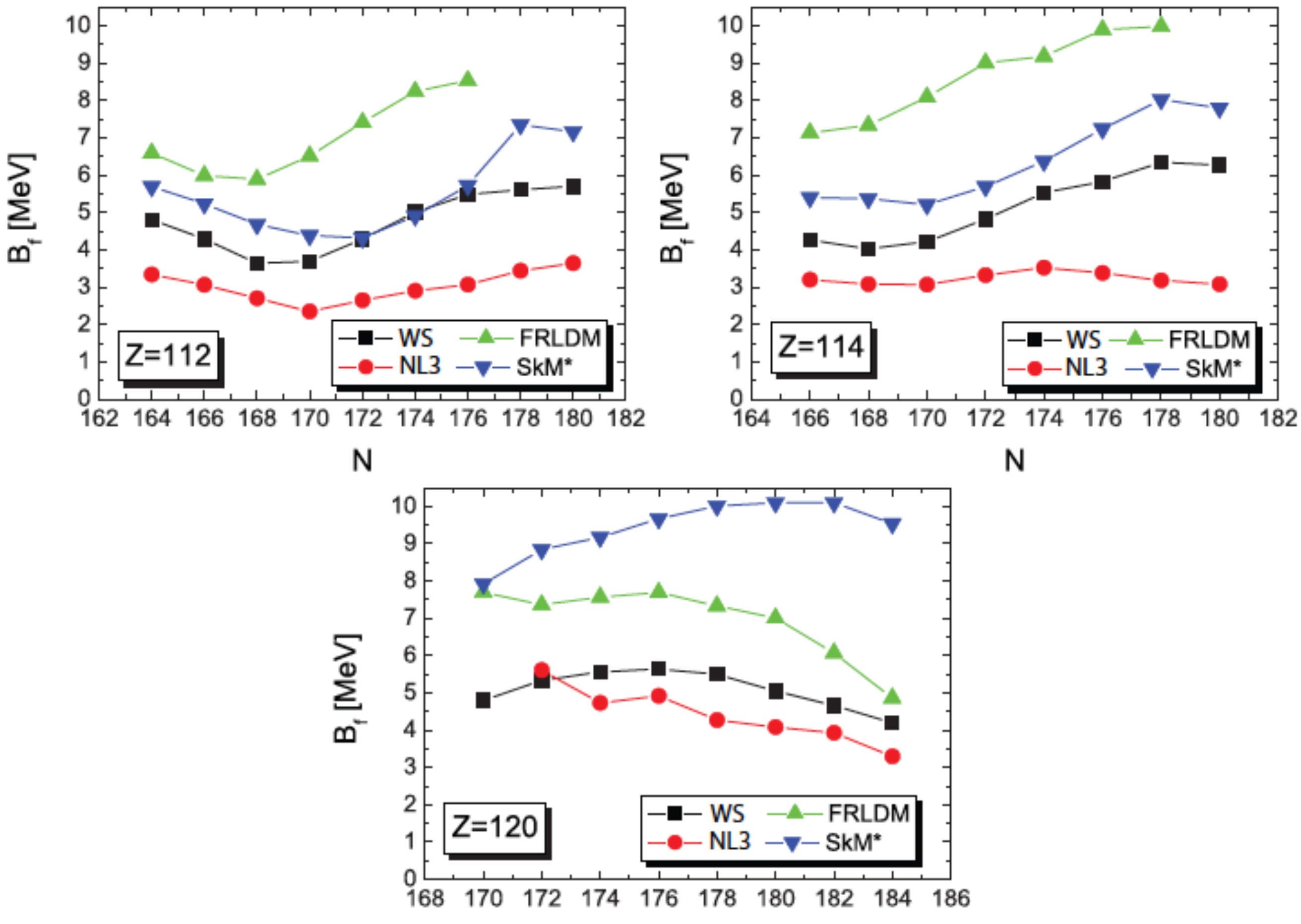


FIG. 2. Difference between theoretical and experimental [34,48] inner fission barrier heights for even-even actinide nuclei as a function of the neutron number.

The situation is more depressing if you go to the heaviest nuclei

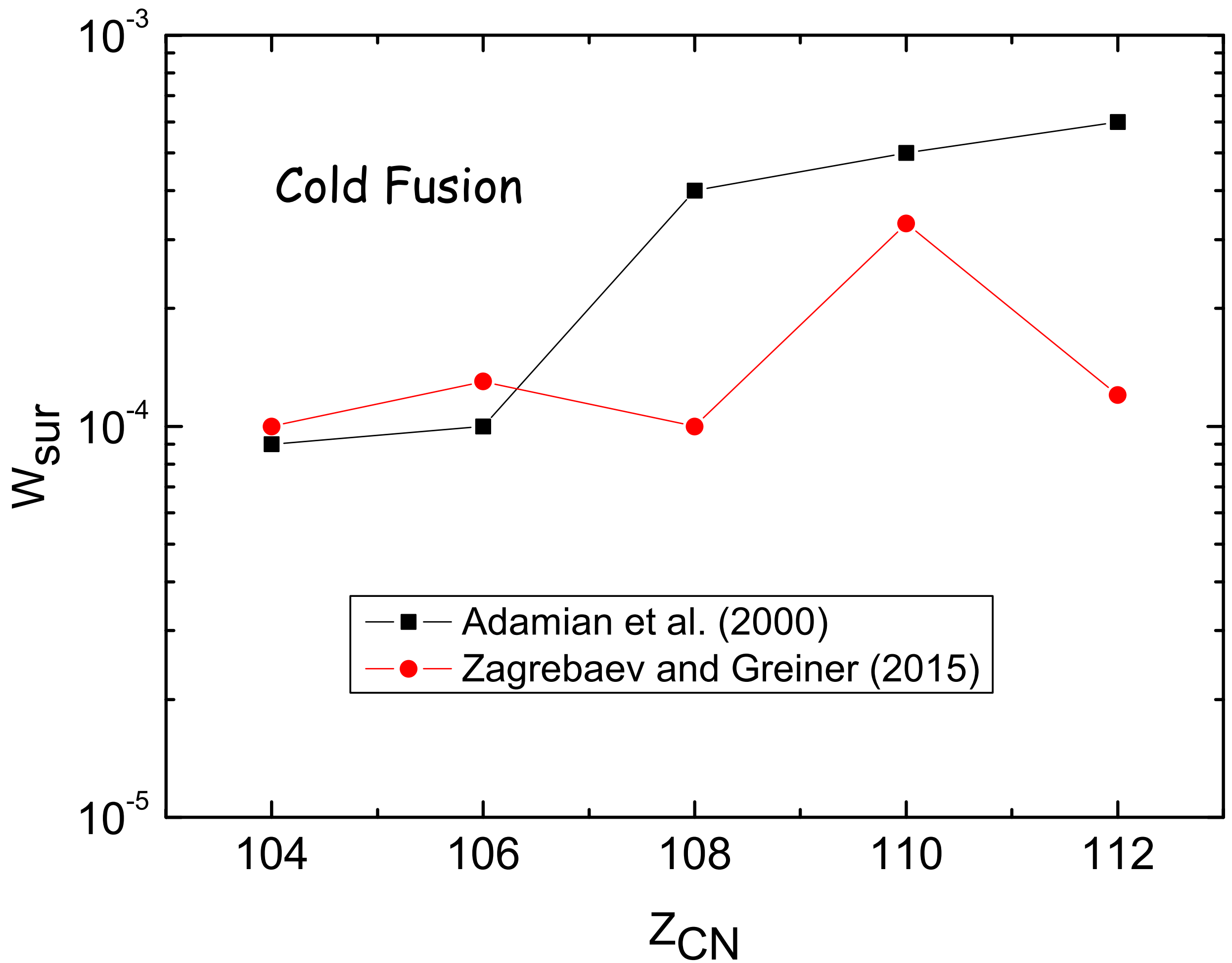


Baran et al. (2015)

W_{sur} summary

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

If B_f, masses the same, W_{sur} ~ same



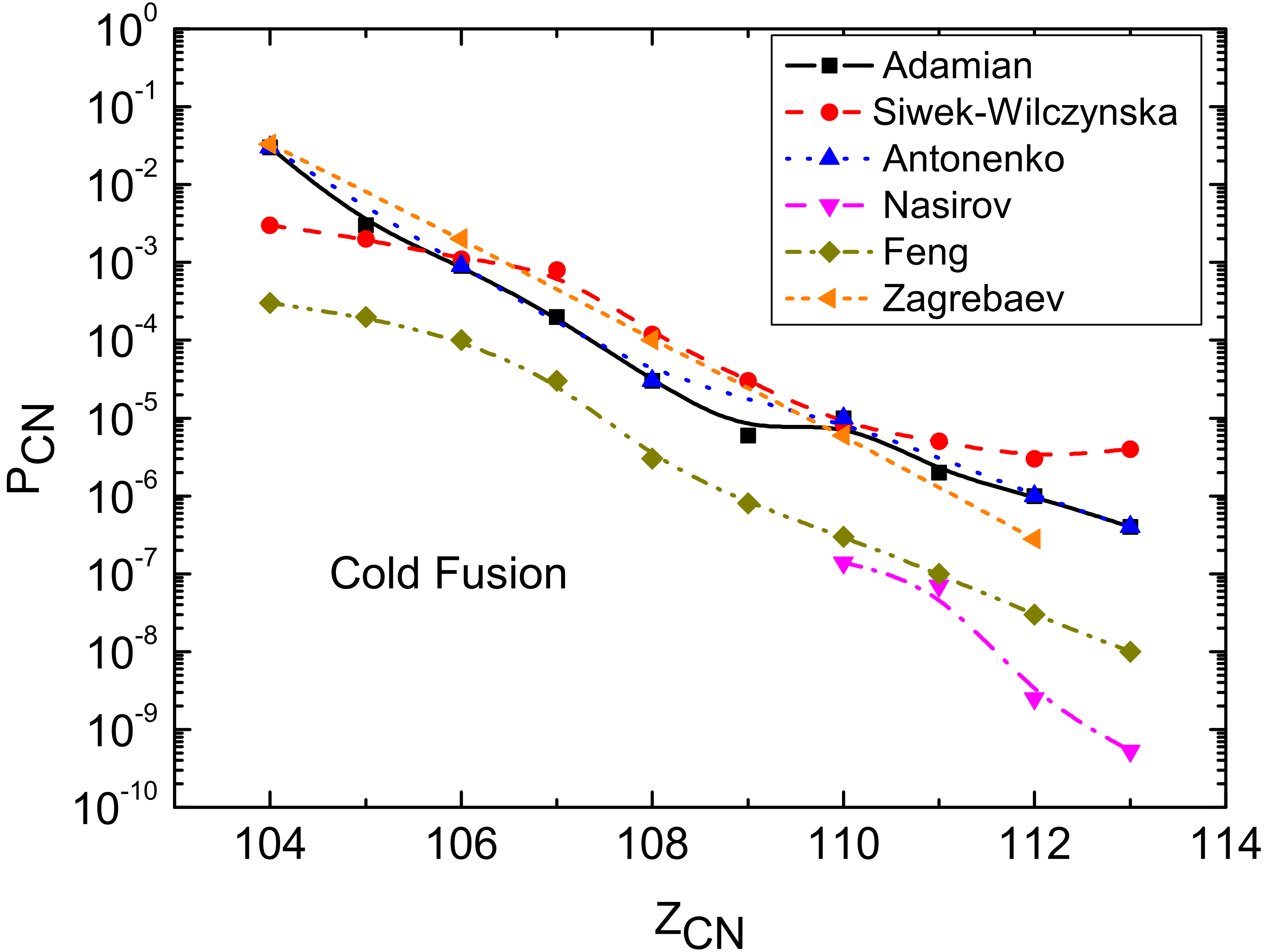
The Fusion Probability, P_{CN}

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

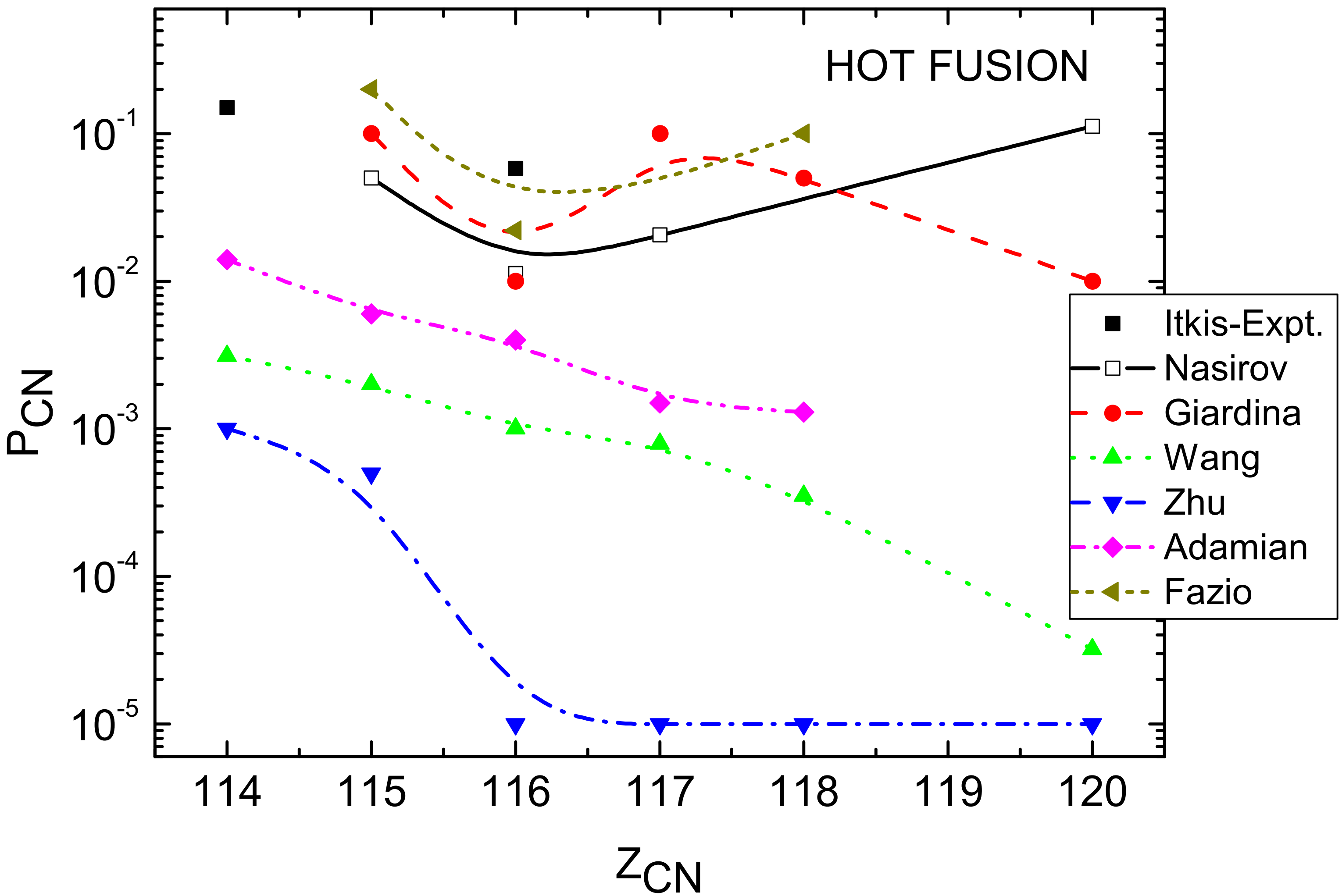
TABLE II. Predicted values of P_{CN} for the $^{124}Sn + {}^{96}Zr$ reaction.

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

P_{CN}(expt.) = 0.05



Adamian
Siwek-Wilczynska
Antonenko
Nasirov
Feng
Zagrebaev
Cold Fusion
P_{CN}
Z_{CN}
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
104
106
108
110
112
114



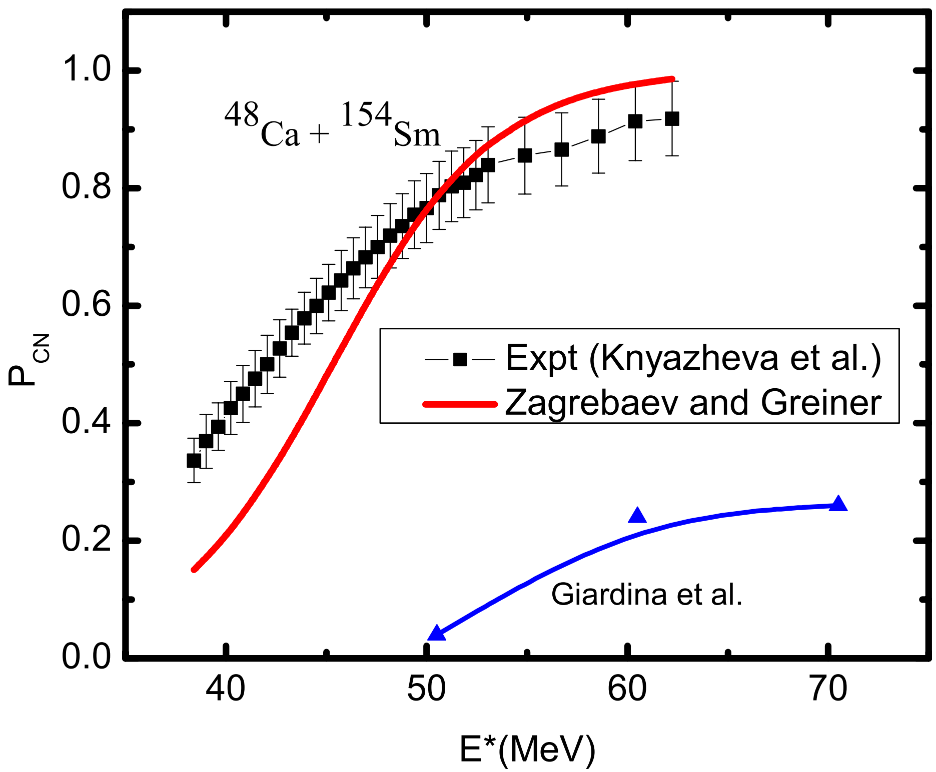
HOT FUSION
P_{CN}
Z_{CN}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
114
115
116
117
118
119
120
Itkis-Expt.
Nasirov
Giardina
Wang
Zhu
Adamian
Fazio

Excitation Energy Dependence of P_{CN}

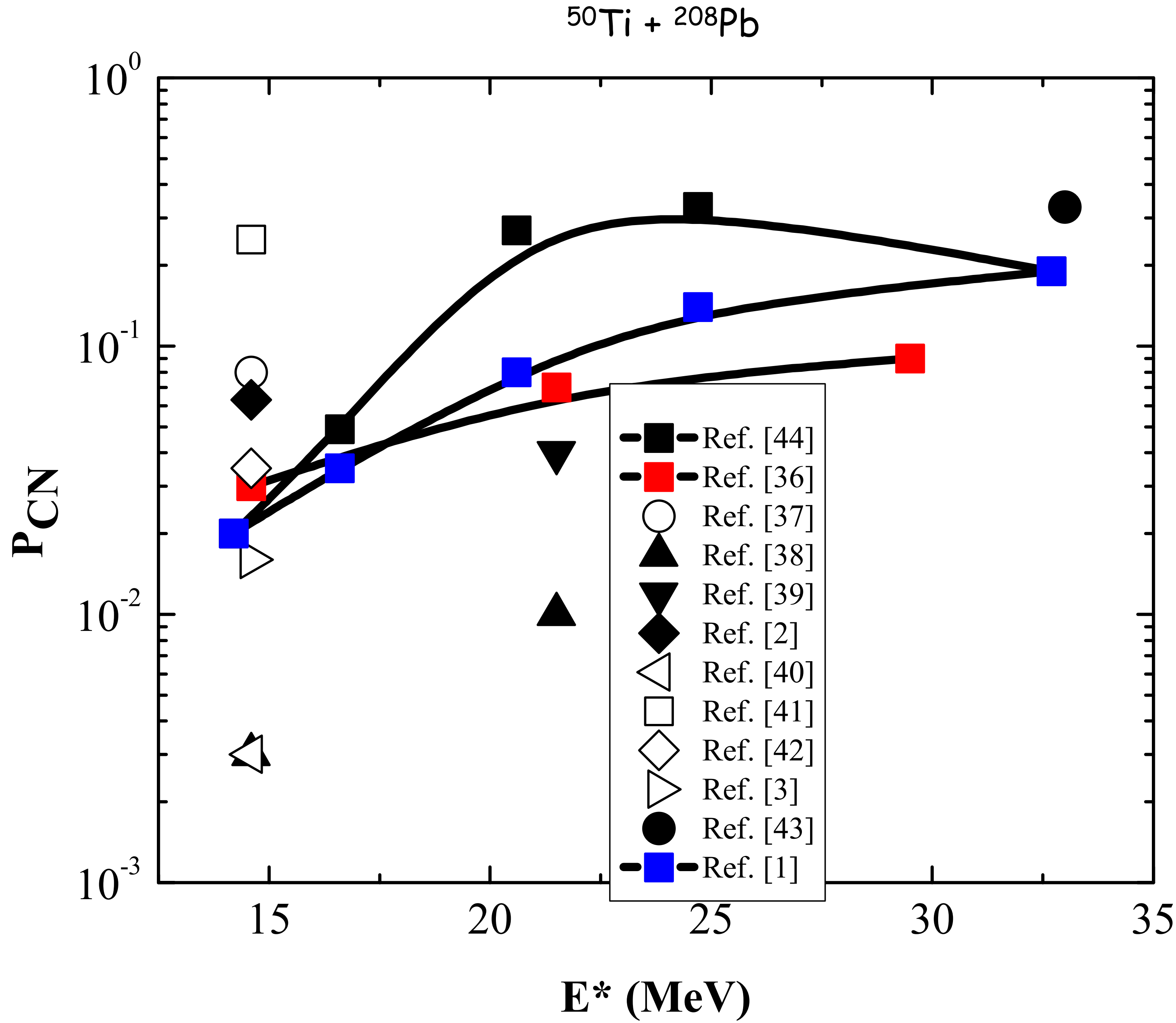
Zagrebaev and Greiner

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

P_{CN} (E*)



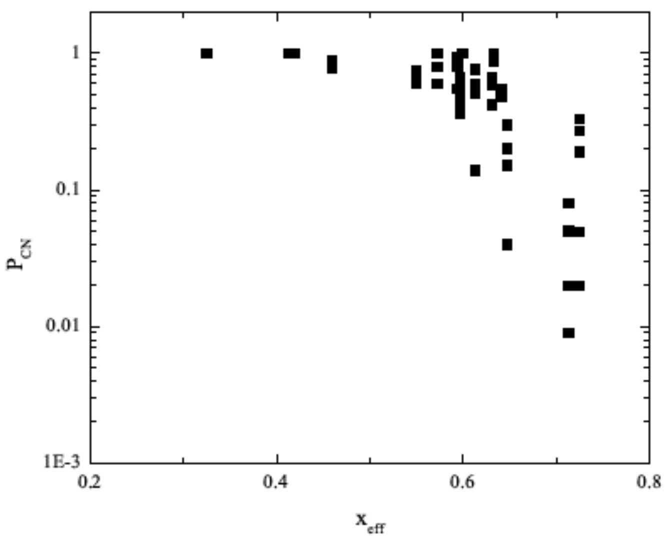
$^{48}Ca + ^{154}Sm$
Expt (Knyazheva et al.)
Zagrebaev and Greiner
Giardina et al.
P_{CN}
E*(MeV)
1.0
0.8
0.6
0.4
0.2
0.0
40
50
60
70



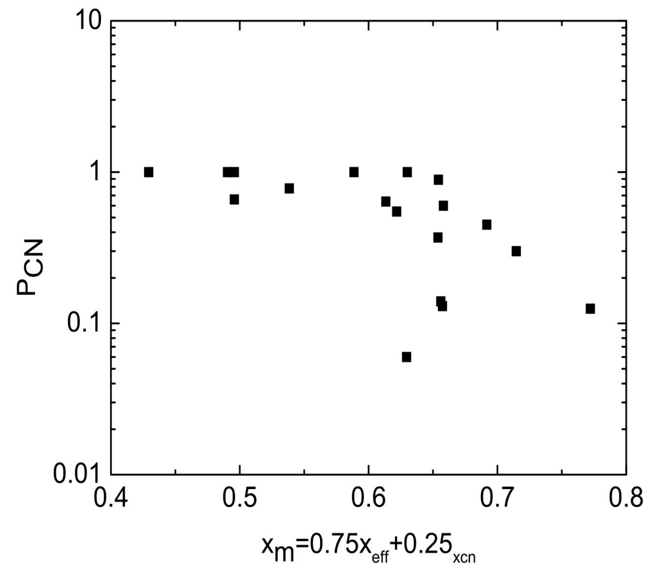
$^{50}Ti + ^{208}Pb$
10^0
10^{-1}
10^{-2}
10^{-3}
P_{CN}
15
20
25
30
35
E* (MeV)
Ref. [44]
Ref. [36]
Ref. [37]
Ref. [38]
Ref. [39]
Ref. [2]
Ref. [40]
Ref. [41]
Ref. [42]
Ref. [3]
Ref. [43]
Ref. [1]

P_{CN} dependence on fissility

All data



E*=40-50 MeV



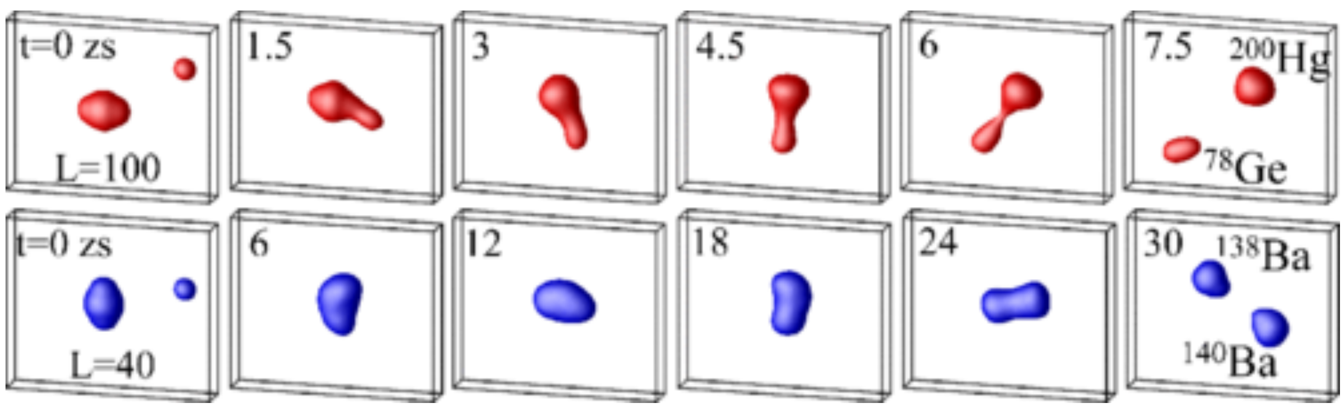
Other scaling variables include $(Z_1-Z_2)/(Z_1+Z_2)$

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)

TDHF calculations (cont.)

- Umar and co-workers (PRC 81, 064607; PRC 90, 054605) have also studied the ^{40,48}Ca + ^{238}U reactions, calculating $\sigma_{capture}$.



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
  - ^{264}Rf $^{252}Cf(^{16}C,4n)$
  - ^{265}Db $^{249}Bk(^{20}O,4n)$
  - ^{268}Sg $^{252}Cf(^{20}O,4n)$
  - ^{267}Bh $^{252}Cf(^{21}F,6n)$

Targeted Radioactive Beams

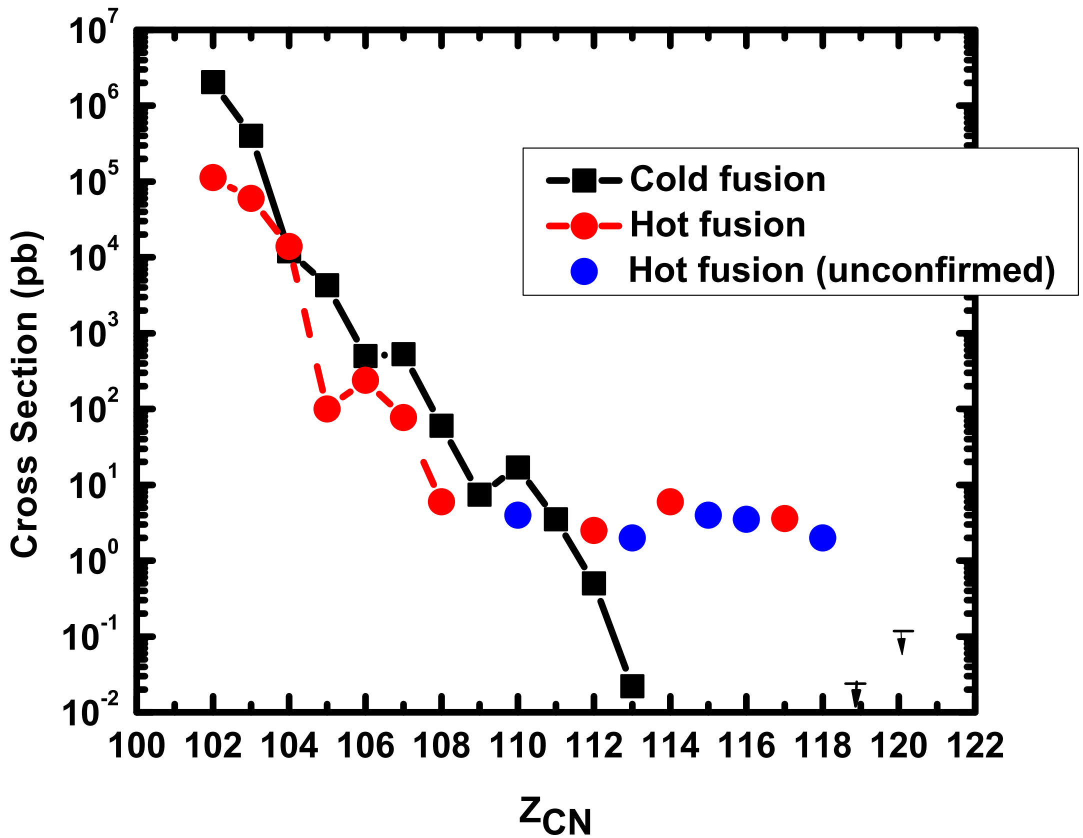
- Special opportunities may exist if RNB facilities focus on producing a beam of particular interest.
- Example: ^{46}Ar (from ^{48}Ca fragmentation)

FRIB "fast beam rate" 1.1×10^{10}

FRIB "reaccelerated beam rate" 2.3×10^{7}

Reaction	Beam Intensity (p/s)	Cross Section (pb)	Atoms/day
$^{238}U(^{48}Ca, 3n)^{283}Cn$	3×10^{12}	0.7	0.5
$^{244}Pu(^{46}Ar, 4n)^{286}Cn$	1.1×10^{10}	250	0.6
$^{244}Pu(^{46}Ar, 3n)^{287}Cn$	1.1×10^{10}	140	0.3

Multi-nucleon transfer reactions



The neutron-deficient character of our efforts

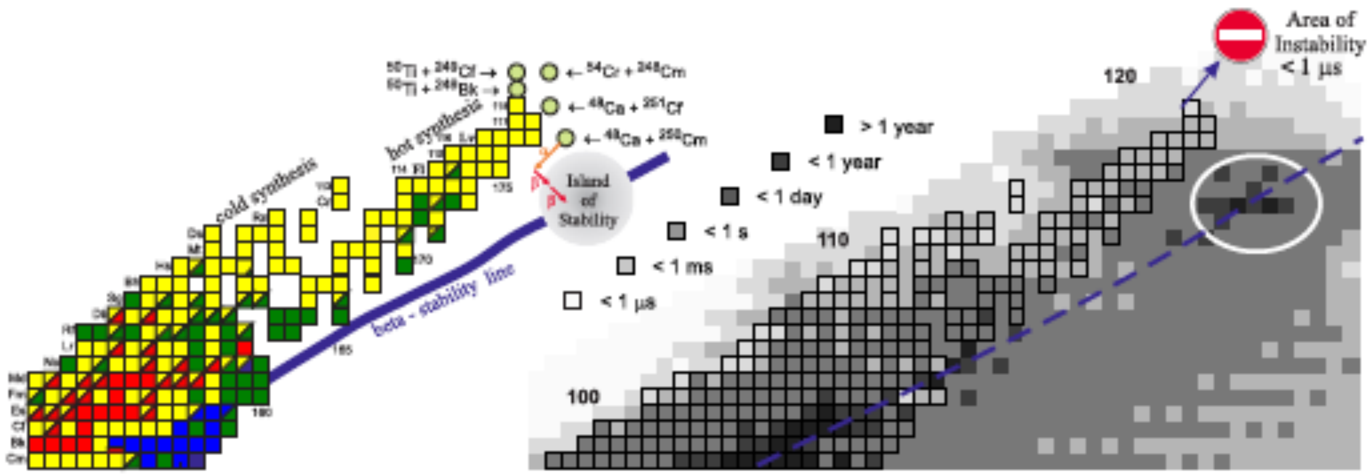
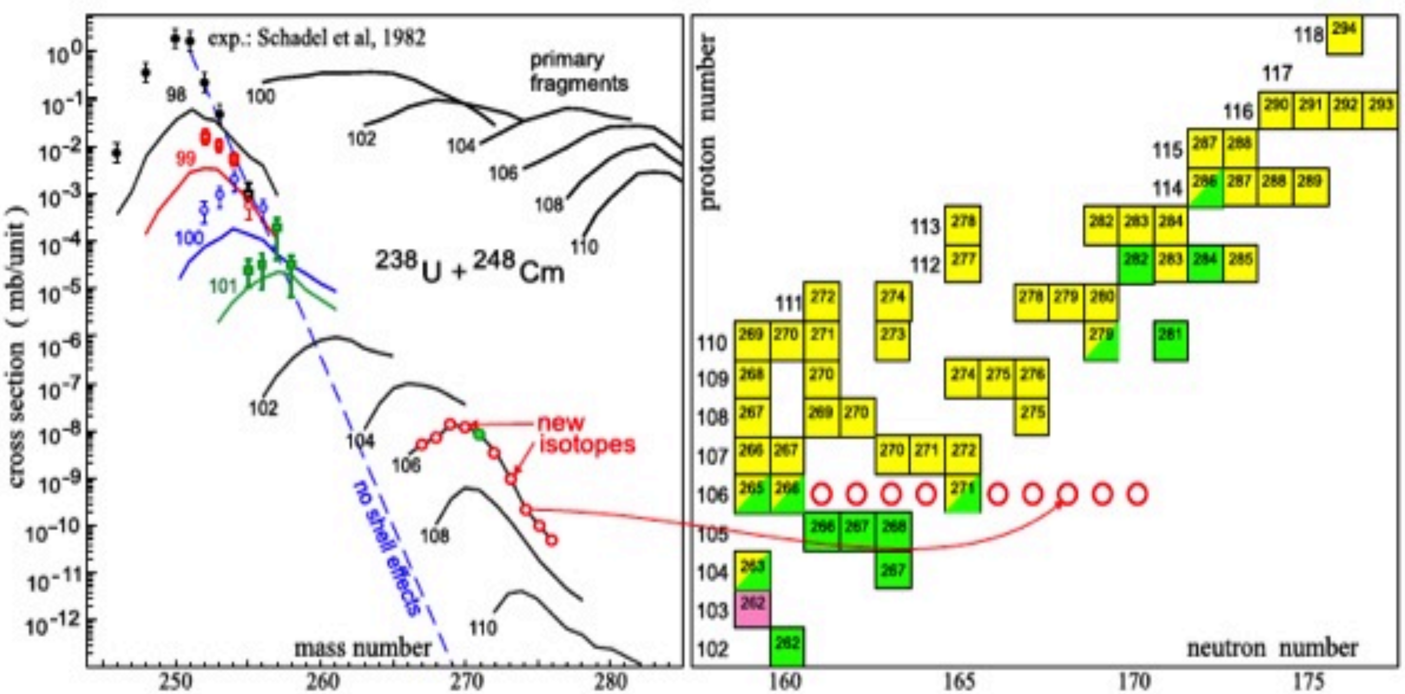


Fig. 1. Upper part of the nuclear map. Current and possible experiments on synthesis of SH elements are shown. (Right panel) Predicted half-lives of SH nuclei and the “area of instability”. Known nuclei are shown by the outlined rectangles.

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

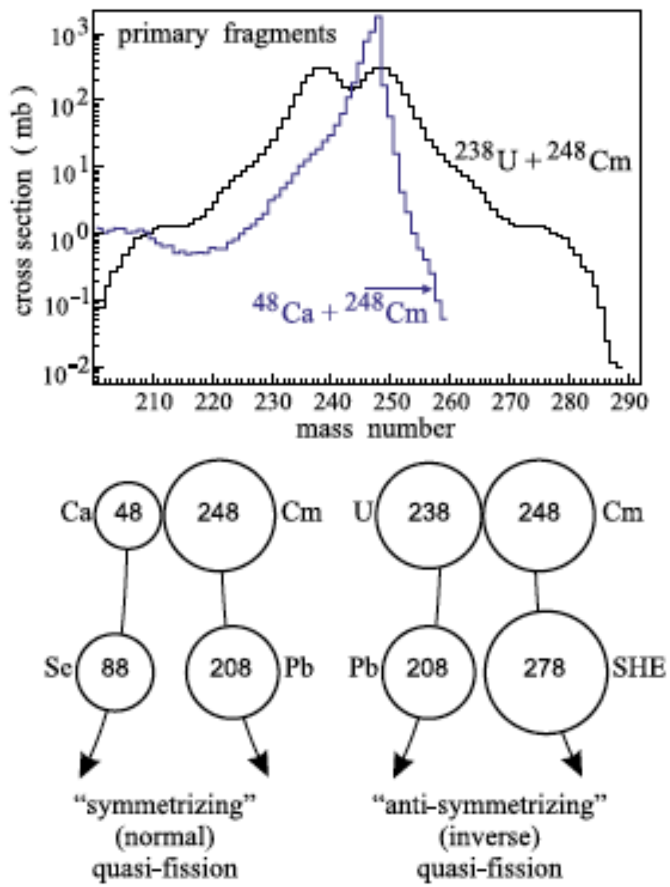
Previous work

- 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.



exp.: Schadel et al, 1982
primary fragments
$^{238}U + ^{248}Cm$
new isotopes
no shell effects
cross section (mb/unit)
mass number
proton number
neutron number

The importance of shell effects



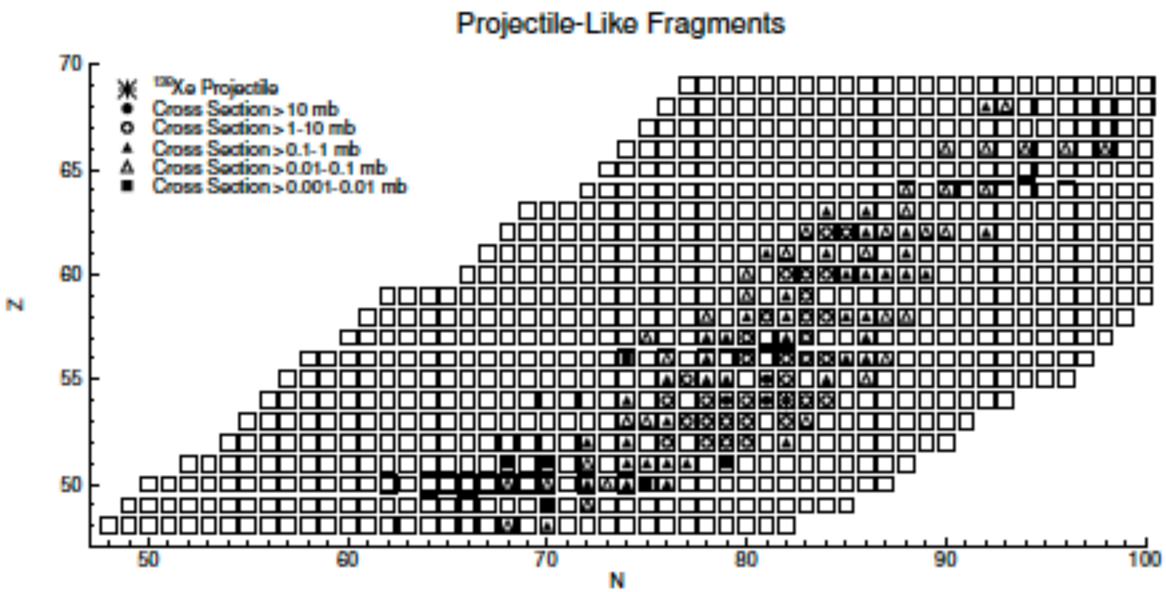
V.I. Zagrebaev and W. Greiner, NPA (in press)

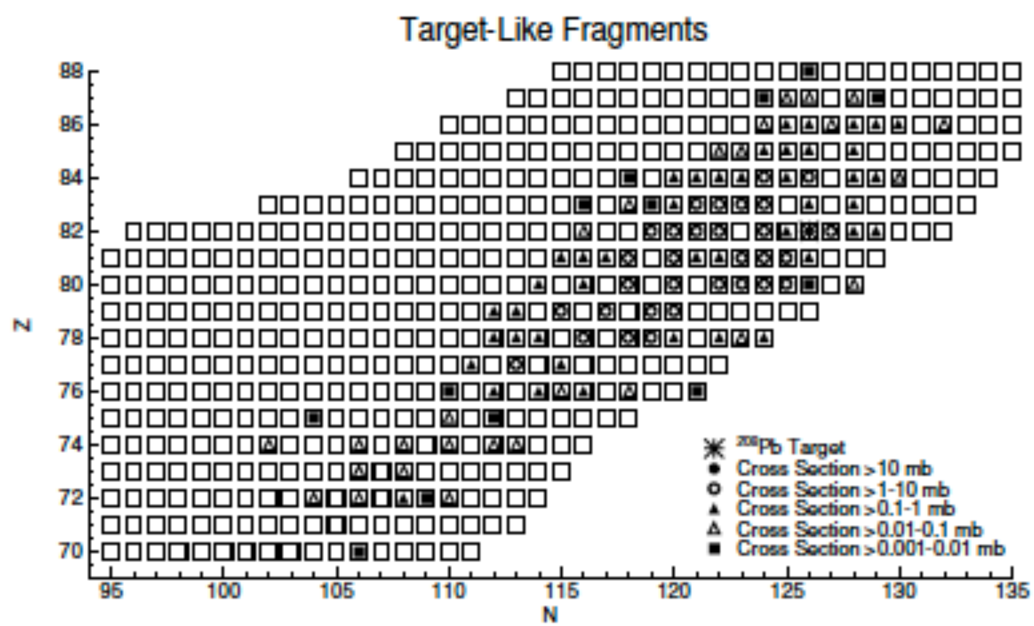
Theory tools

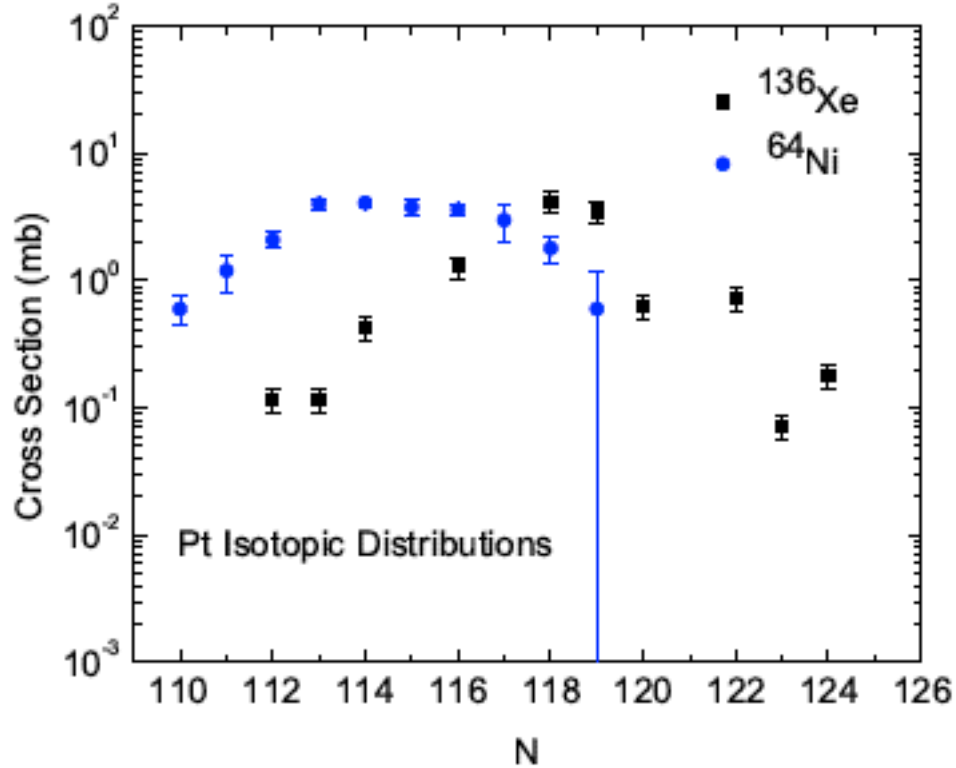
- Zagrebaev and Greiner calculations
- Semi-classical model of Pollarolo and Winther (GRAZING) and its revision (GRAZING-F) (Yanez and Loveland-2015)

$^{136}Xe + {}^{208}Pb$—Barrett et al.

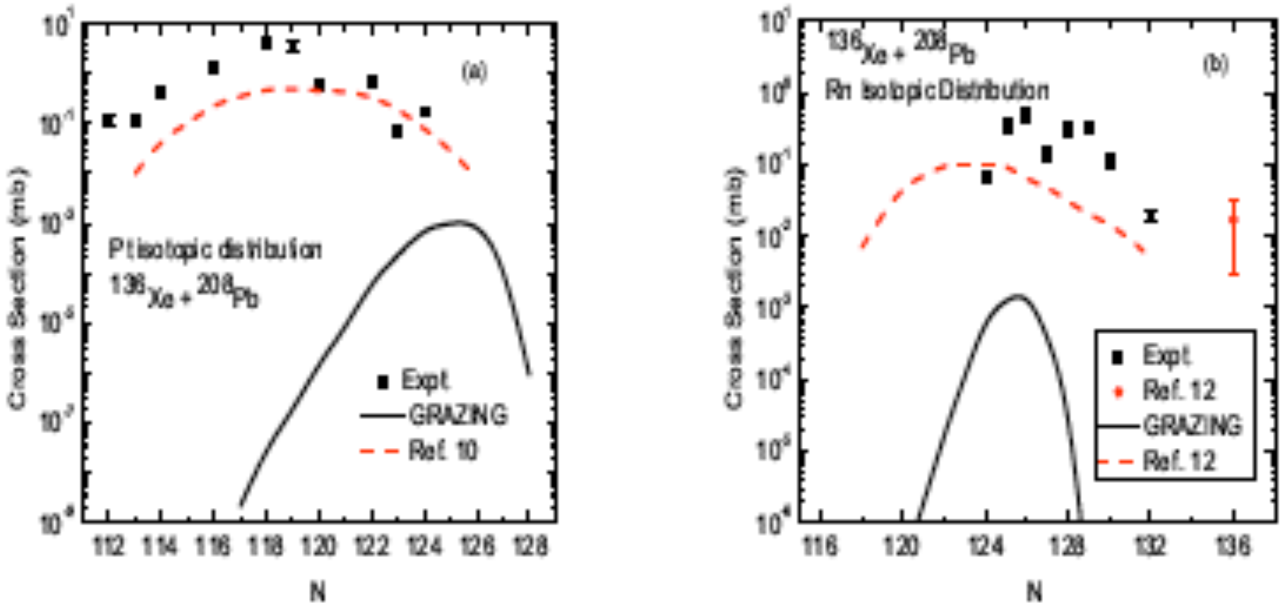
- Gammasphere study
- Over 230 nuclidic production cross sections were measured.


Projectile-Like Fragments
^{136}Xe Projectile
Cross Section > 10 mb
Cross Section > 1-10 mb
Cross Section > 0.1-1 mb
Cross Section > 0.01-0.1 mb
Cross Section > 0.001-0.01 mb
Z
N


Target-Like Fragments
^{208}Pb Target
Cross Section > 10 mb
Cross Section > 1-10 mb
Cross Section > 0.1-1 mb
Cross Section > 0.01-0.1 mb
Cross Section > 0.001-0.01 mb
Z
N



10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
Cross Section (mb)
^{136}Xe
^{64}Ni
Pt Isotopic Distributions
110
112
114
116
118
120
122
124
126
N

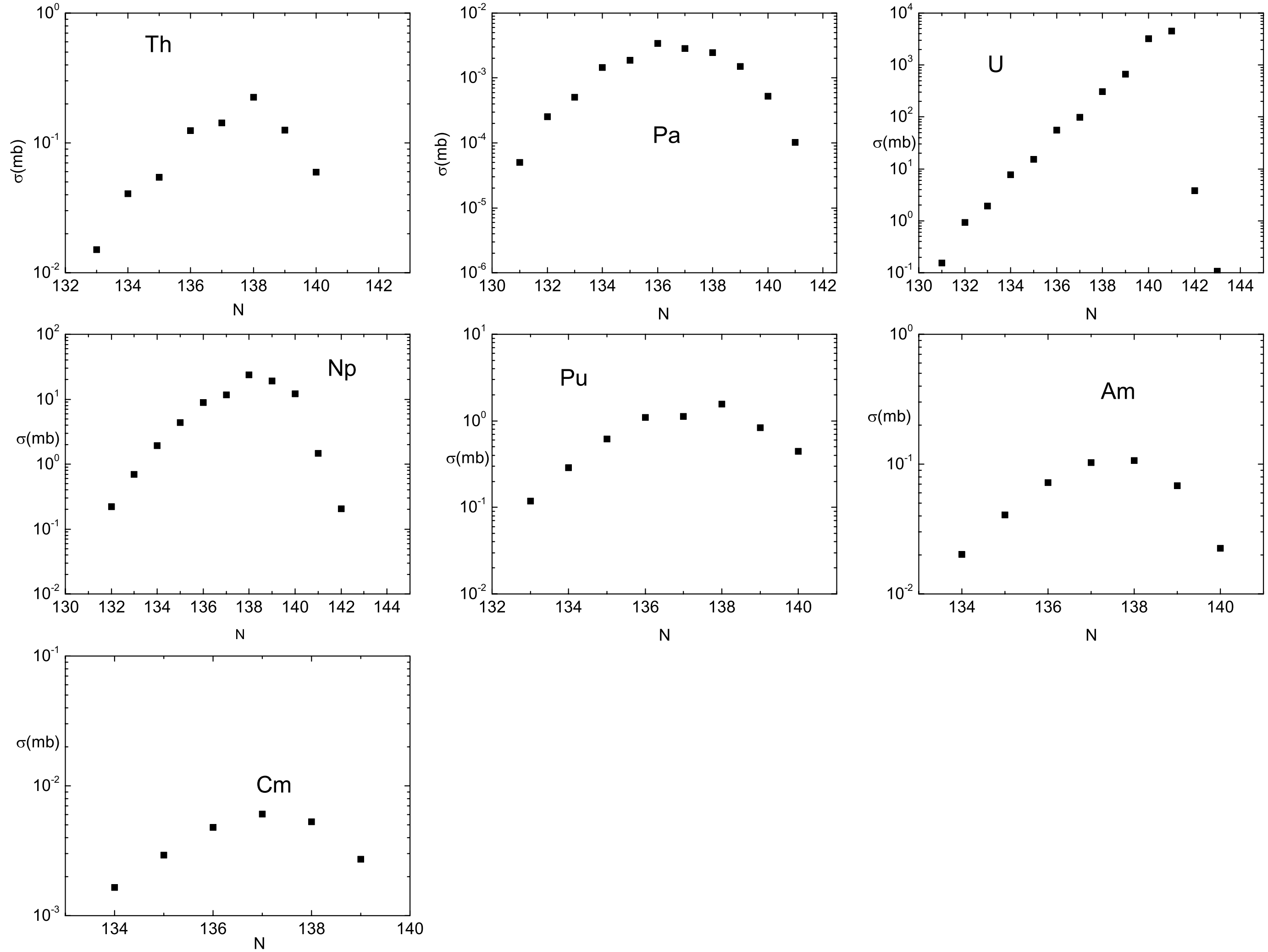


GRAZING is NOT a suitable model for large proton transfers

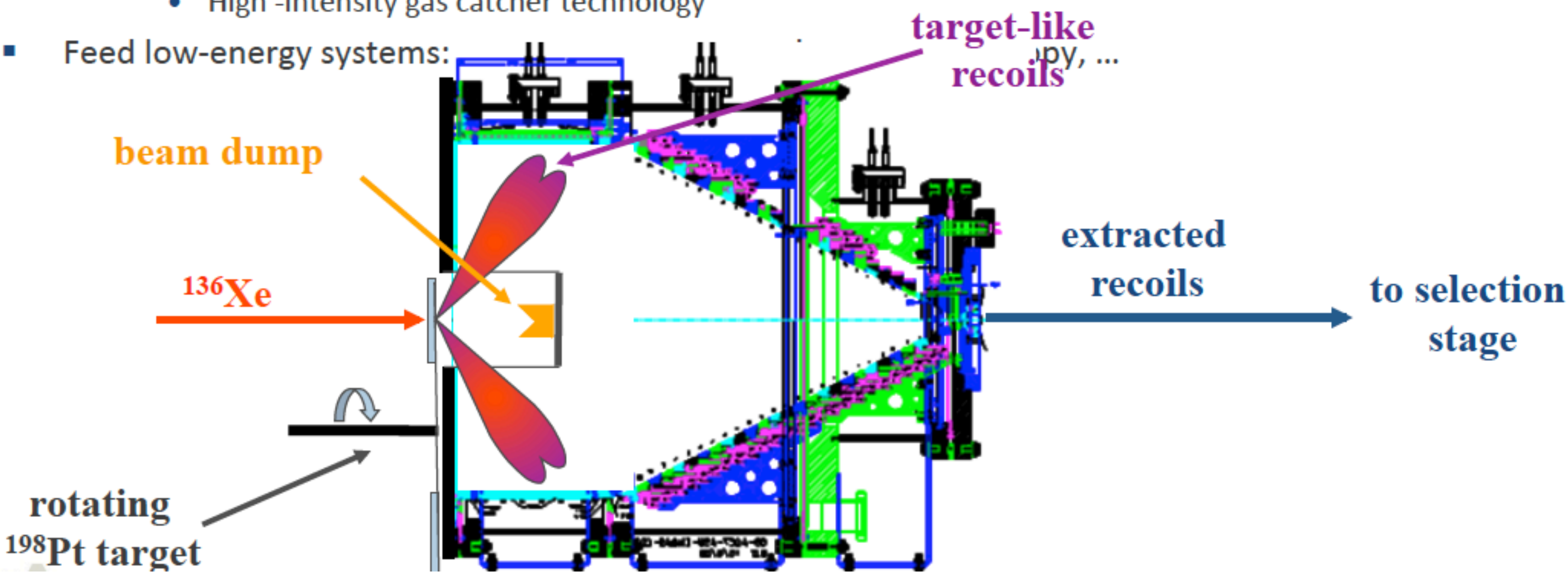
The way forward

- Vacuum separators (FMA)
- SHIP group argues that separation of products by velocity is 5 times more effective than separation by magnetic rigidity in a gas and the reaction channels can be determined directly from the velocity spectra.
- Z-G predict the most n-rich transfer products peak at 0° rather than the grazing angle
- SHIP(Heinz et al.) observes MNT products at 0-2° with cross sections down to 0.1-1 nb.
- We want to use the FMA to study MNT
- FMA (^{64}Ni + ^{208}Pb, ^{132}Xe + ^{233}U)
- N=126 factory -Guy Savard-ANL

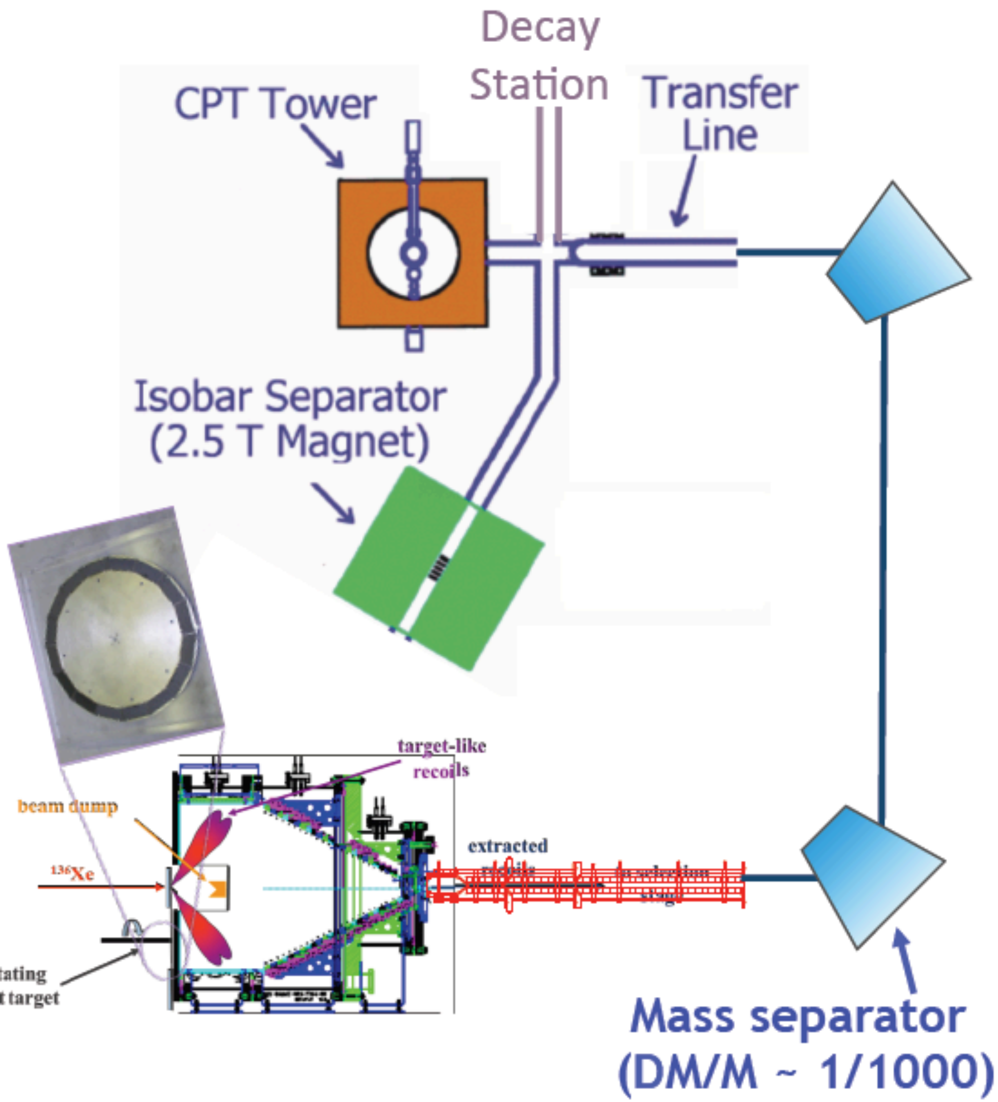
FMA Experiment $^{132}Xe + ^{233}U$



- Proposed collection system capitalizes on
  - High primary beam intensity
  - High -intensity gas catcher technology
- Feed low-energy systems: … py, …



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 in-flight 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 ^{198}Hf 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 β-emitters is under construction.

Overall Conclusions

- Measure $\sigma_{capture}$
- Better info on B_f needed
- More measurements and TDHF calculations of P_{CN}
- Uncertainty in calculated values of σ_{EVR} at least 1-2 orders of magnitude.
- RNB opportunities for making n-rich actinides exist.
- MNT appears promising and worth investigating.

Acknowledgements

- This work was supported by the U.S. Dept. of Energy, Office of Nuclear Physics.