Fusion hindrance in the synthesis of superheavy elements

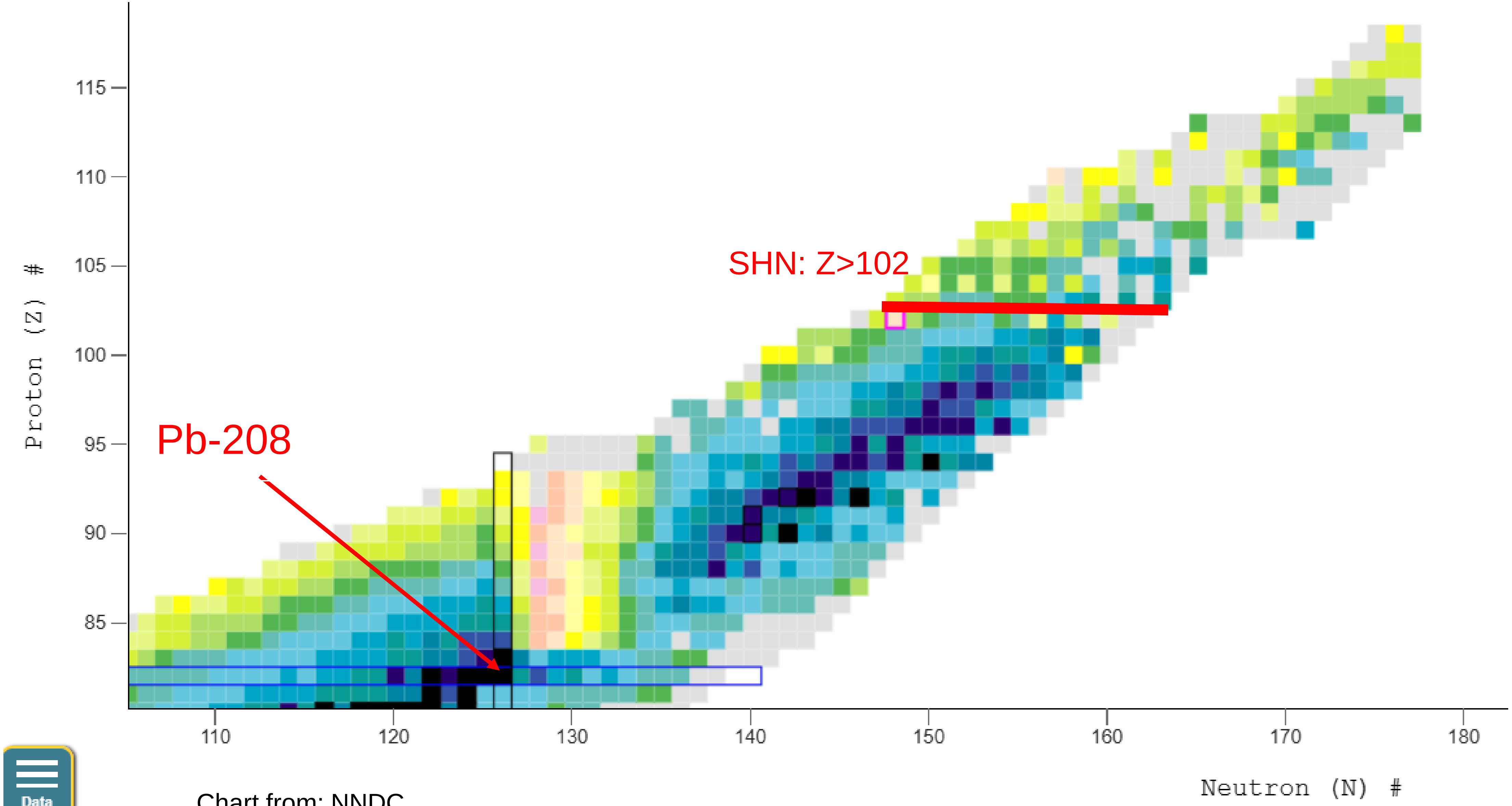
By Filip Agert

LUND UNIVERSITY

LTH FACULTY OF ENGINEERING

GANIL

Where are the superheavy nuclei? (SHN)

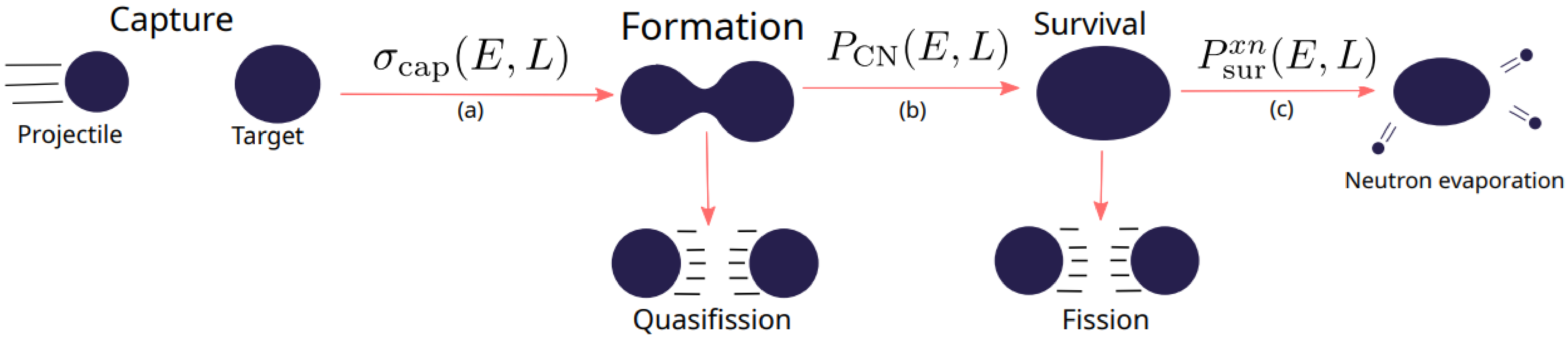


Data

Chart from: NNDC

How are superheavy elements synthesised?

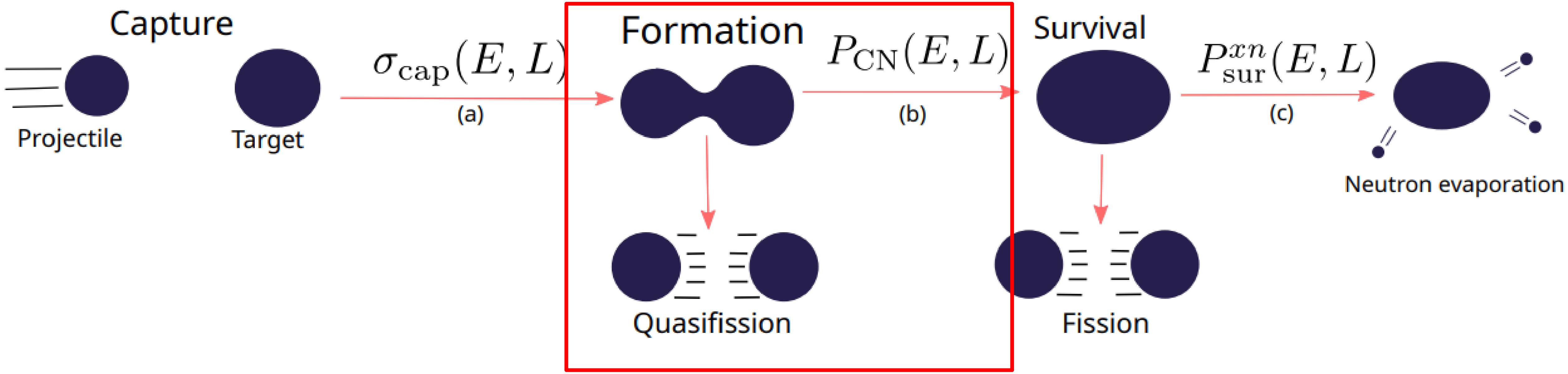
3-stages



$$\sigma^{xn}_{er}(E) = \sum_{L=0}^{\infty}(2L+1)\sigma_{\text{cap}}(E,L)P_{\text{CN}}(E,L)P^{xn}_{\text{sur}}(E,L)$$

How are superheavy elements synthesised?

3-stages



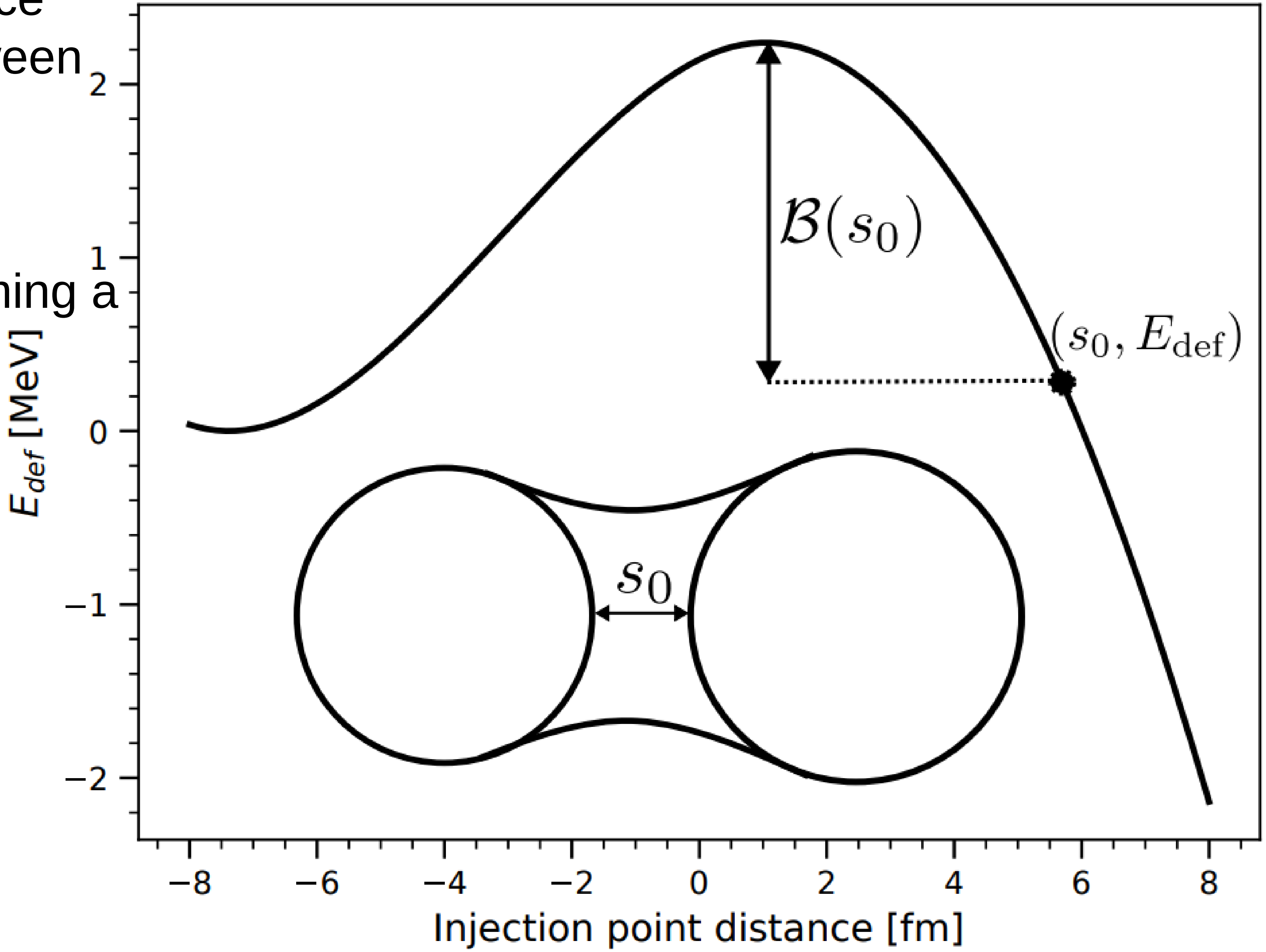
$$\sigma_{er}^{xn}(E) = \sum_{L=0}^{\infty}(2L+1)\sigma_{\text{cap}}(E,L)\boxed{P_{\text{CN}}(E,L)}P_{\text{sur}}^{xn}(E,L)$$

Supervisor:
David Boilley

Thanks to:
Dieter Ackermann

1D potential energy surface
Function of distance between projectile-target.

What is probability of forming a compound nucleus?



Langevin Equation of motion

Langevin EOM is an EOM for a deterministic+random force

$$\ddot{s} + \beta \dot{s} - \omega^2 (s - s_{\mathrm{sad}}) = r(t).$$

s - Separation of target-projectile.
β - Friction.
ω - Angular frequency of parabolic barrier.
r(t) - Random force (Gaussian noise).

Simplifies when $\beta \gg \omega$ in the overdamped limit.

Langevin Equation of motion

Solve for s(t) by applying the Laplace transform.

$$
\begin{aligned}
s(t) &= \mathcal{L}^{-1}\{S(\sigma)\}, \\
&= (s_0 - s_{\mathrm{sad}})e^{-\frac{\omega^2}{\beta}t} + s_{\mathrm{sad}} + \frac{1}{\beta}\int_0^t r(\tau)e^{-\frac{\omega^2}{\beta}(t-\tau)}d\tau.
\end{aligned}
$$

Formation probability given by integrating a Gaussian up to the saddle point.

$$
P_{\mathrm{CN}}(t, s_0) = \int_{-\infty}^{s_{\mathrm{sad}}} \frac{1}{\sqrt{2\pi}\sigma_s} \exp\left(-\frac{(s - \langle s(t)\rangle)^2}{2\sigma_s^2}\right) ds
$$

Langevin Equation of motion

Take the limit as t → infinity

$$P_{\mathrm{CN}} = \frac{1}{2}\mathrm{erfc}\left(\sqrt{\frac{\mathcal{B}(s_0)}{\mathcal{T}}}\right)$$

With T temperature, dependent on energy,
B barrier height, dependent on potential energy surface.

Langevin Equation of motion

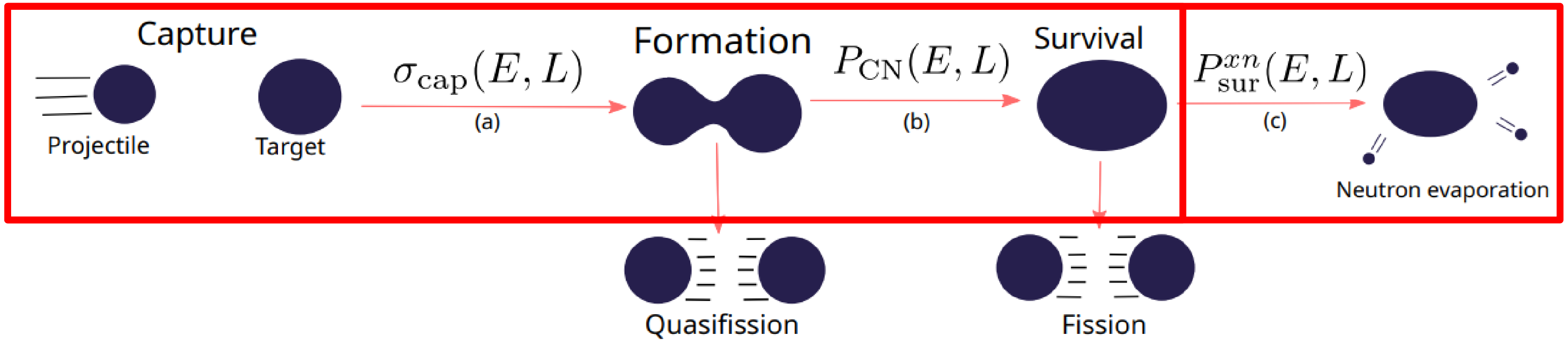
Similar derivation even when not overdamped.

$$P_{\mathrm{CN}}(E_{cm},L)=\frac{1}{2}\mathrm{erfc}\left(\sqrt{\frac{\mathcal{B}(s_0)}{\mathcal{T}}}-\frac{1}{\beta/2\omega+\sqrt{1+\beta^2/4\omega^2}}\sqrt{\frac{K(E_{cm},L)}{\mathcal{T}}}\right)$$

K - kinetic energy
Ecm - Center of mass energy
L - Angular momentum for partial wave

Two free parameters of the model to be fit to data:
β – Friction parameter
s_0 – Injection point distance

Fit parameters to data



$$\sigma^{xn}_{er}(E) = \sum_{L=0}^{\infty}(2L+1)\sigma_{\text{cap}}(E,L)P_{\text{CN}}(E,L)P^{xn}_{\text{sur}}(E,L)$$

Cannot fit to formation cross section data due to ambiguous measurement.

Evaporation residue cross section data not ideal
- Bad statistics
- Little available data

σ_{CN}~ mb to μb
σ_{er} ~ nb to fb

Survival part introduces other sources of uncertainty:
- Fission barrier height
- Neutron separation energy

Fit parameters to data

$$\sigma_{er}^{xn}(E) = \sum_{L=0}^{\infty}(2L+1)\sigma_{\text{cap}}(E,L)P_{\text{CN}}(E,L)P_{\text{sur}}^{xn}(E,L)$$

$$P_{\text{surv}}^{x\text{n}}(E_{A_0}^*) = \prod_{i=1}^{x} \frac{\Gamma_{\text{n},i-1}(E_{A_{i-1}}^*)}{\Gamma_{\text{n},i-1}(E_{A_{i-1}}^*) + \Gamma_{\text{f},i-1}(E_{A_{i-1}}^*)}$$

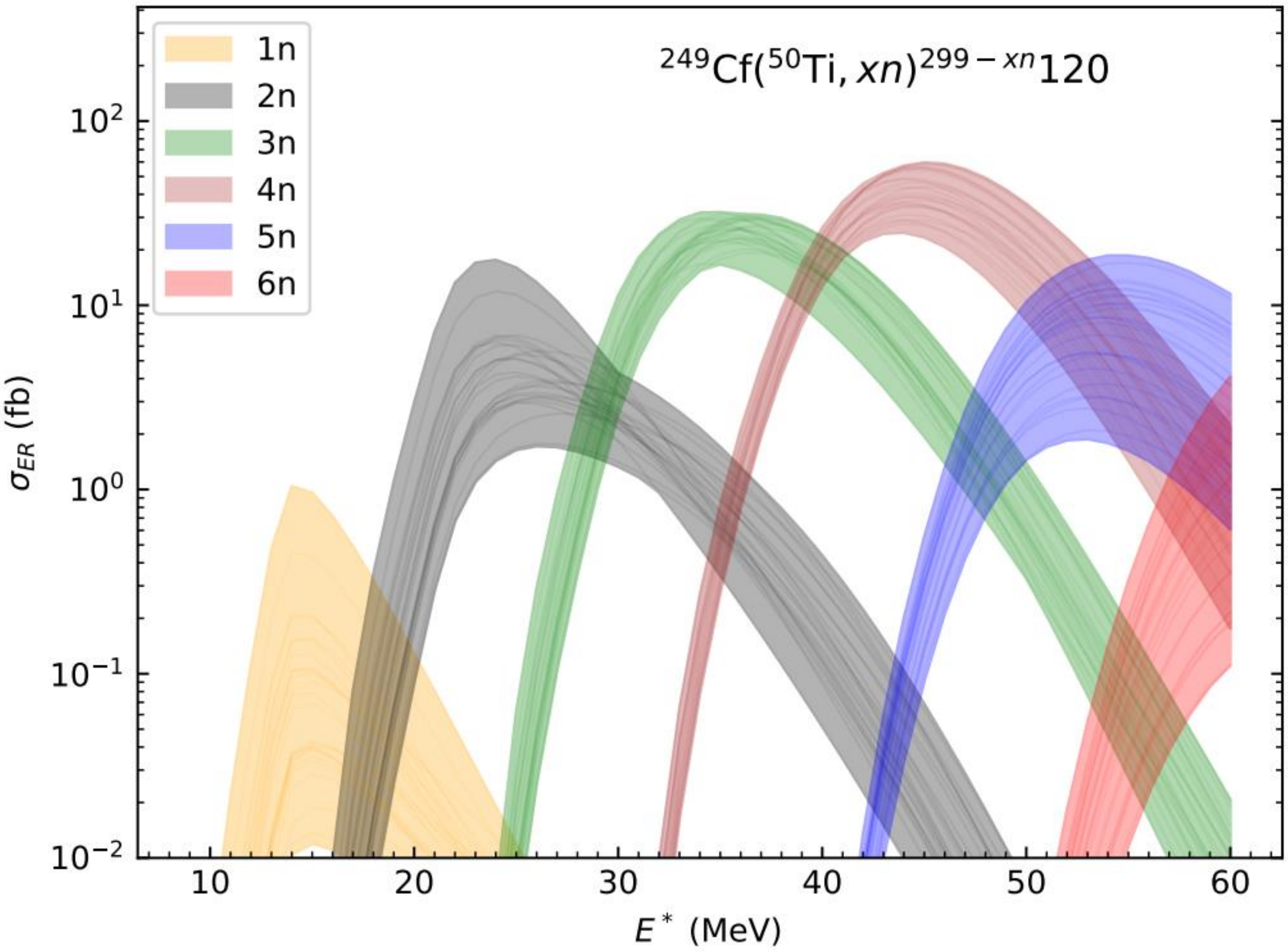
Γ – Decay width for neutron and fission channels, depends on:

- Level density parameter, a
- Fission friction parameter β
- Fission barrier height
- Neutron separation energy
- Etc…

Effect of survival stage parameters on prediction

1. Refit the formation model parameters to each combination of the survival probability settings. 144 combinations.
2. Pick the 25 best combinations scoring the lowest loss function.
3. Predict reaction.

Shaded area is spread in prediction
Factor 3 spread in the 4n channel.



Effect of survival stage parameters on prediction

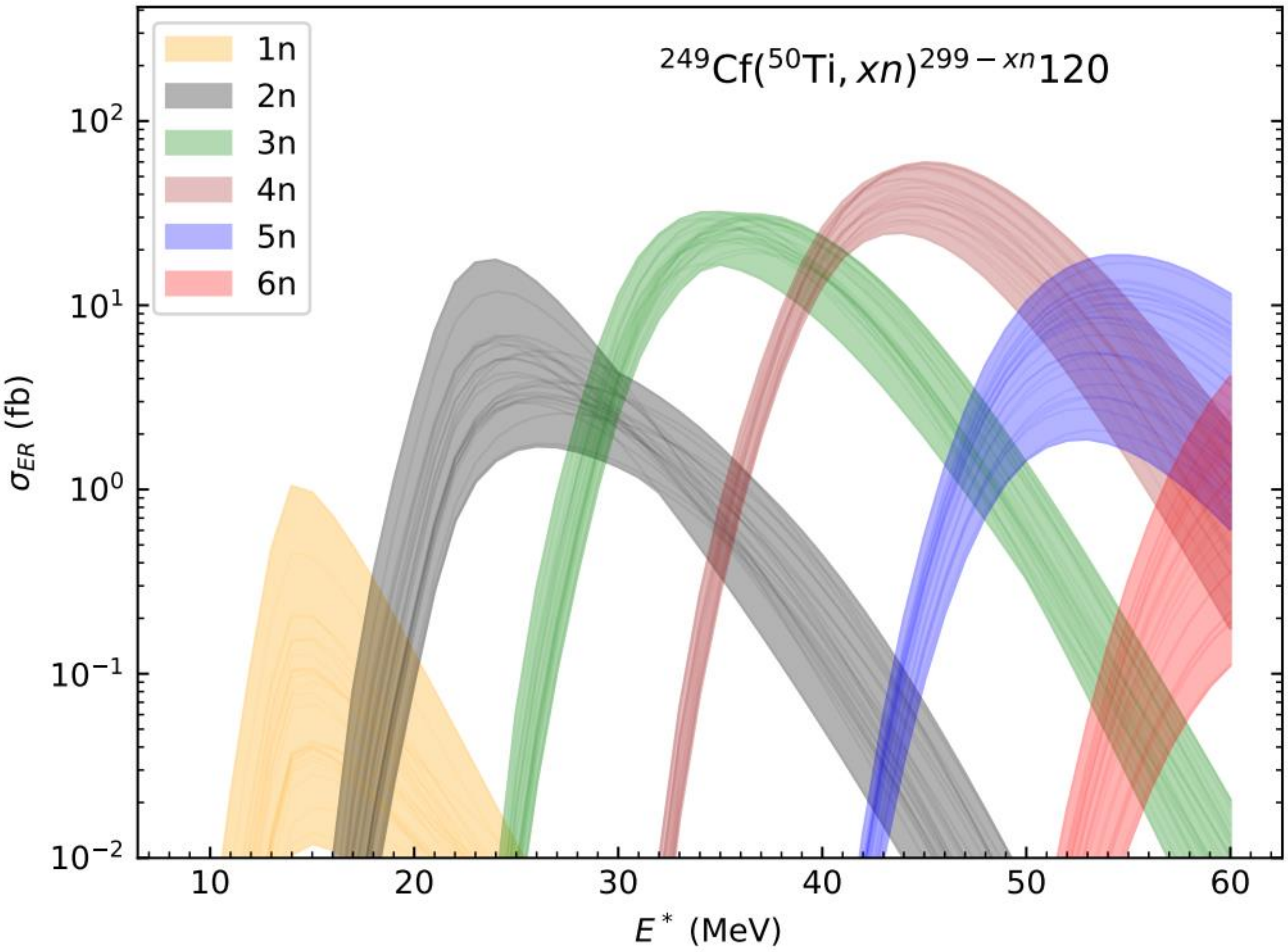
1. Refit the formation model parameters to each combination of the survival probability settings. 144 combinations.
2. Pick the 25 best combinations scoring the lowest loss function.
3. Predict reaction.

Shaded area is spread in prediction
Factor 3 spread in the 4n channel.

$\beta \sim 0.7 * 10^{21}\ s^{-1}$ in best fit

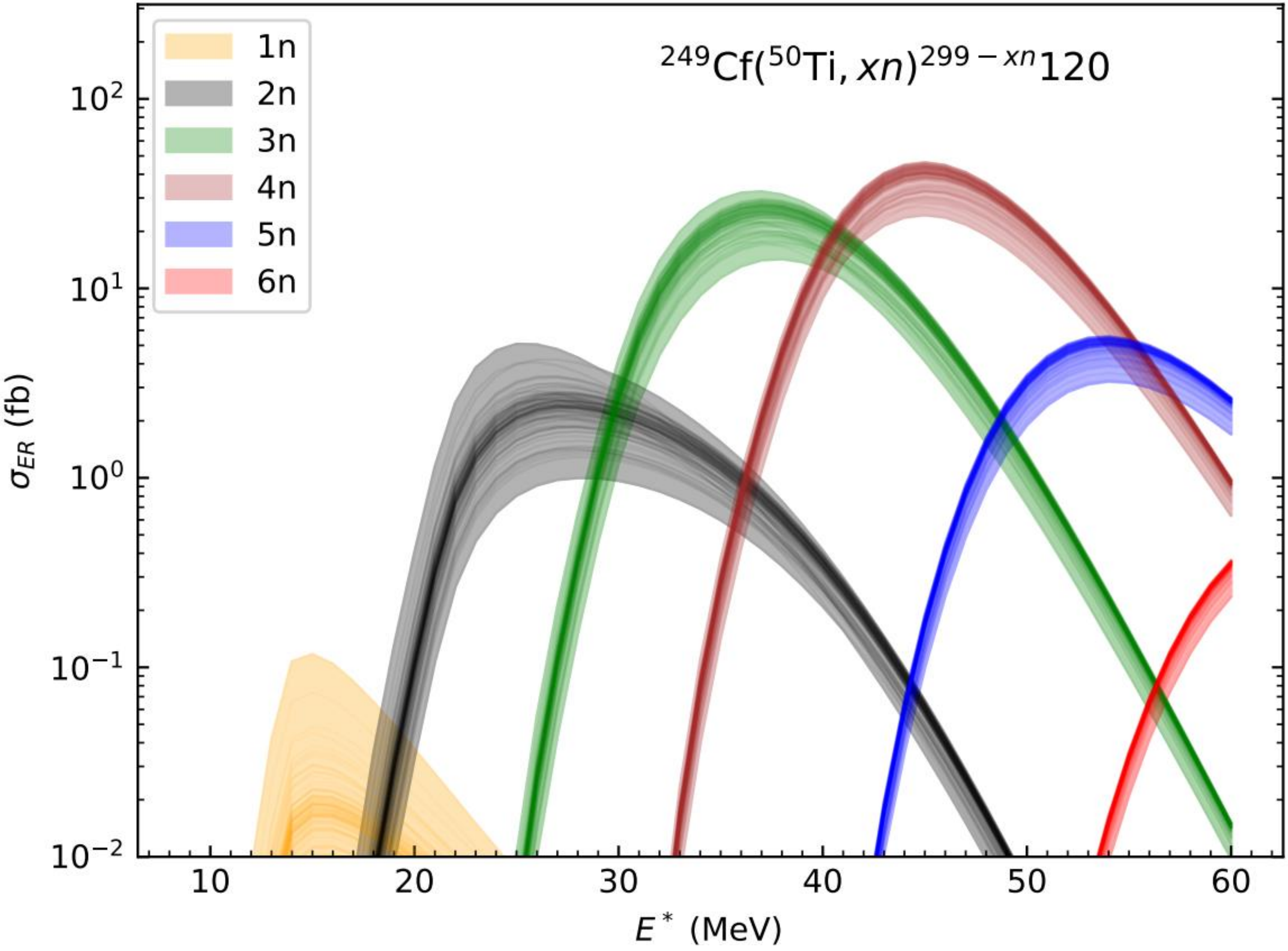
But varies from 10^{17} to 10^{21} in the top 25 combinations.

β model dependent.



Effect of data bootstrapping

1. Randomly select 85% of points in full dataset.
2. Fit the model to these points.
3. Repeat 100 times.
4. Figure shows the different predictions this produces.



Outlook for 1D model

- Remove model dependence of survival part by using Bohr's hypothesis
  - Survival probability independent of entrance channel.
  - Compare two reactions leading to same CN.
- Quantify uncertainty in survival part
  - Need covariance matrices from Micmac models

    Expected 1 order of magnitude uncertainty from uncertainty in in fission barriers alone.

LUND UNIVERSITY

LTH

FACULTY OF ENGINEERING

Supervisors:
G. Carlsson, M. Albertsson, A. Idini

Five dimensional solution

Five shape coordinates define an potential energy surface.
FRLDM potential is used.

α – Mass asymmetry
c_{neck} – Neck radius
Q_2 – Elongation
ε_l – Left fragment deformation
ε_r – Right fragment deformation

Collectively denoted χ

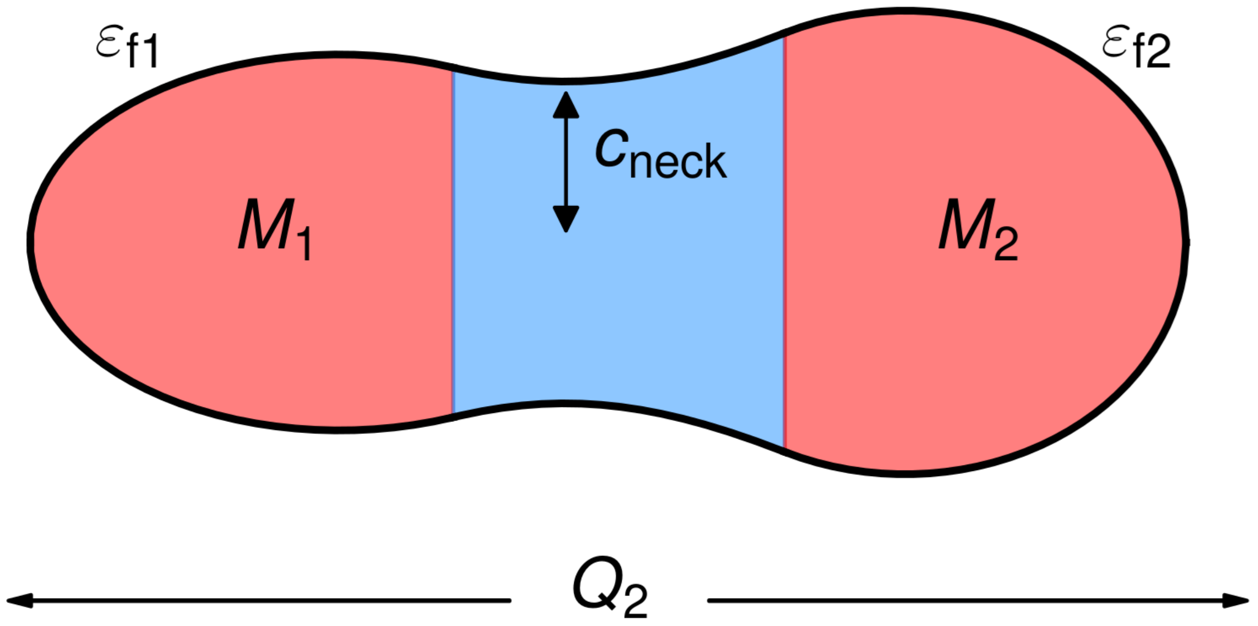


Fig from Martin Albertsson, private communication

P. Möller et.al. At. Data Nucl. Data Tables **109–110**, 1-204 (2016).

Five dimensional solution

How to solve Langevin EOM?
In overdamped limit, motion is approximated by Brownian motion.

Solve by random walk on the potential energy surface. $U(\chi)$

With probability of moving to a neighbor related to level densities

$$P_{i\to j} = \min\left[1, \rho_j(E^*(\chi_j))/\rho_i(E^*(\chi_i))\right]$$

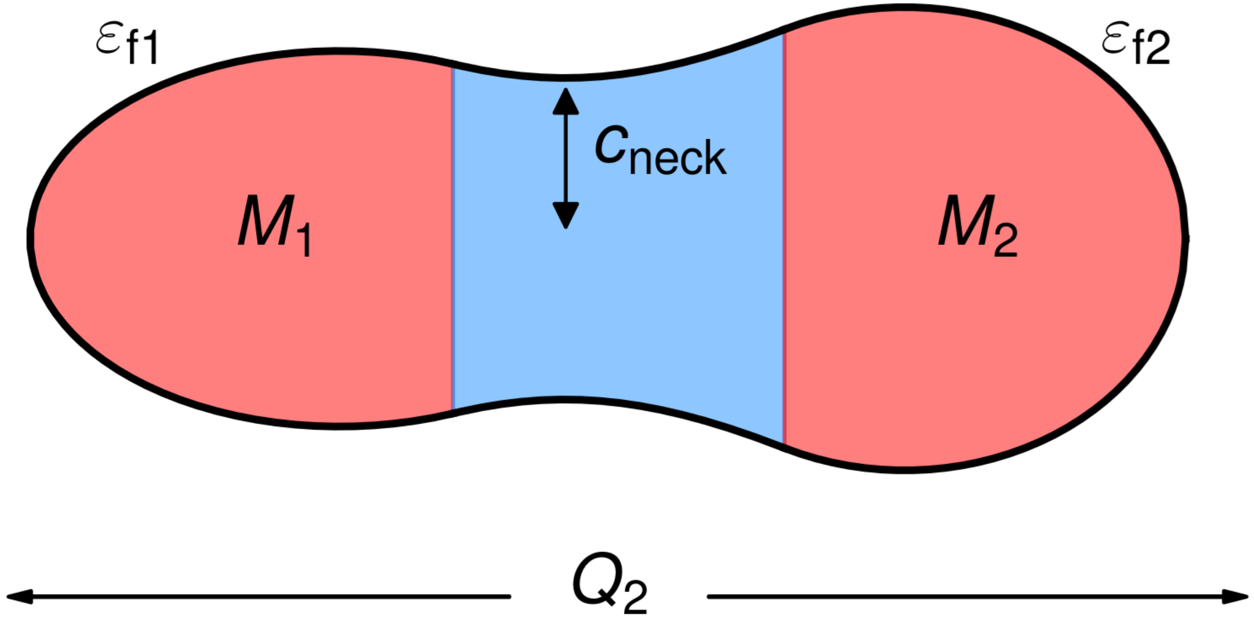


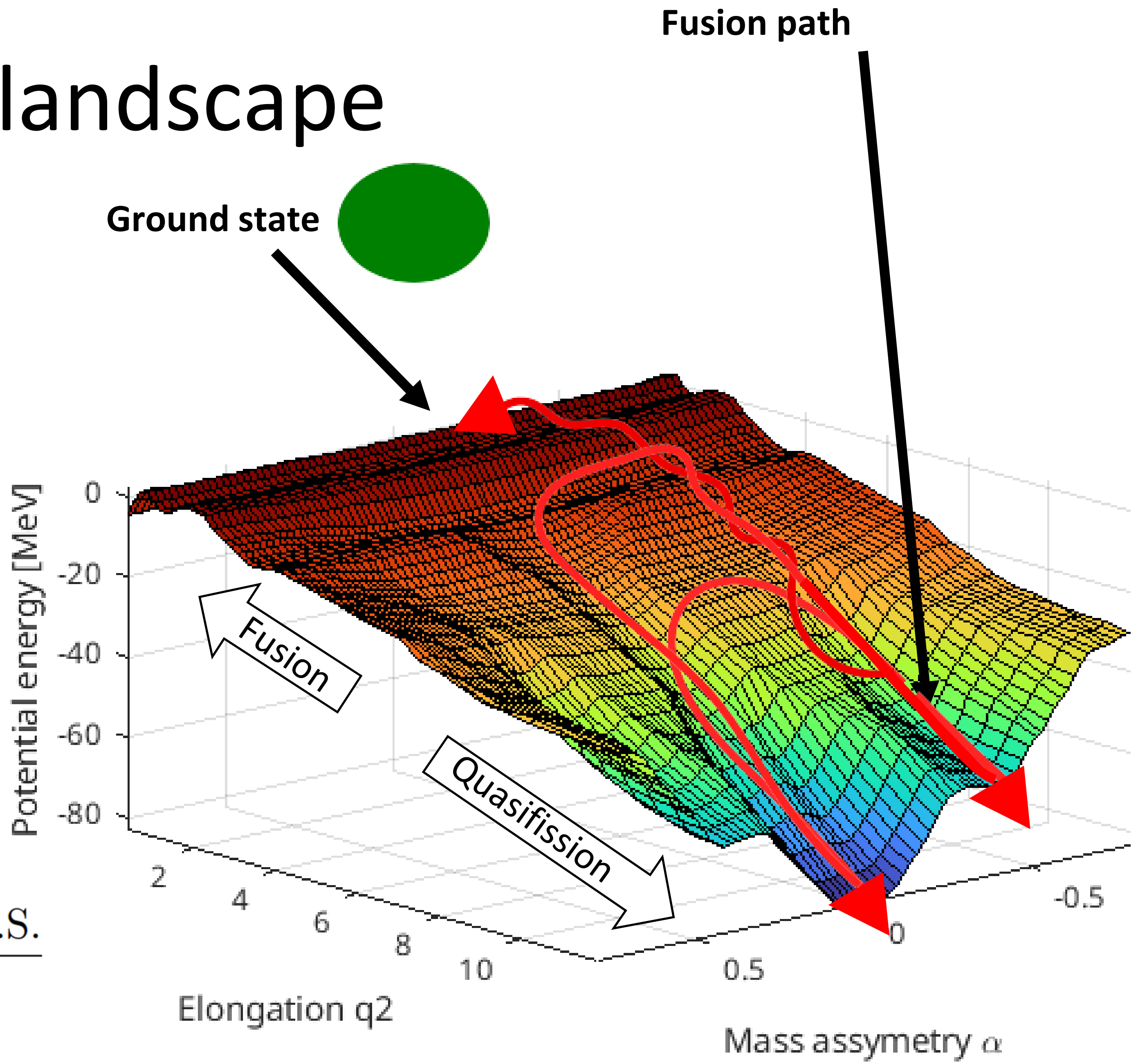
Fig from Martin Albertsson, private communication

Potential energy landscape

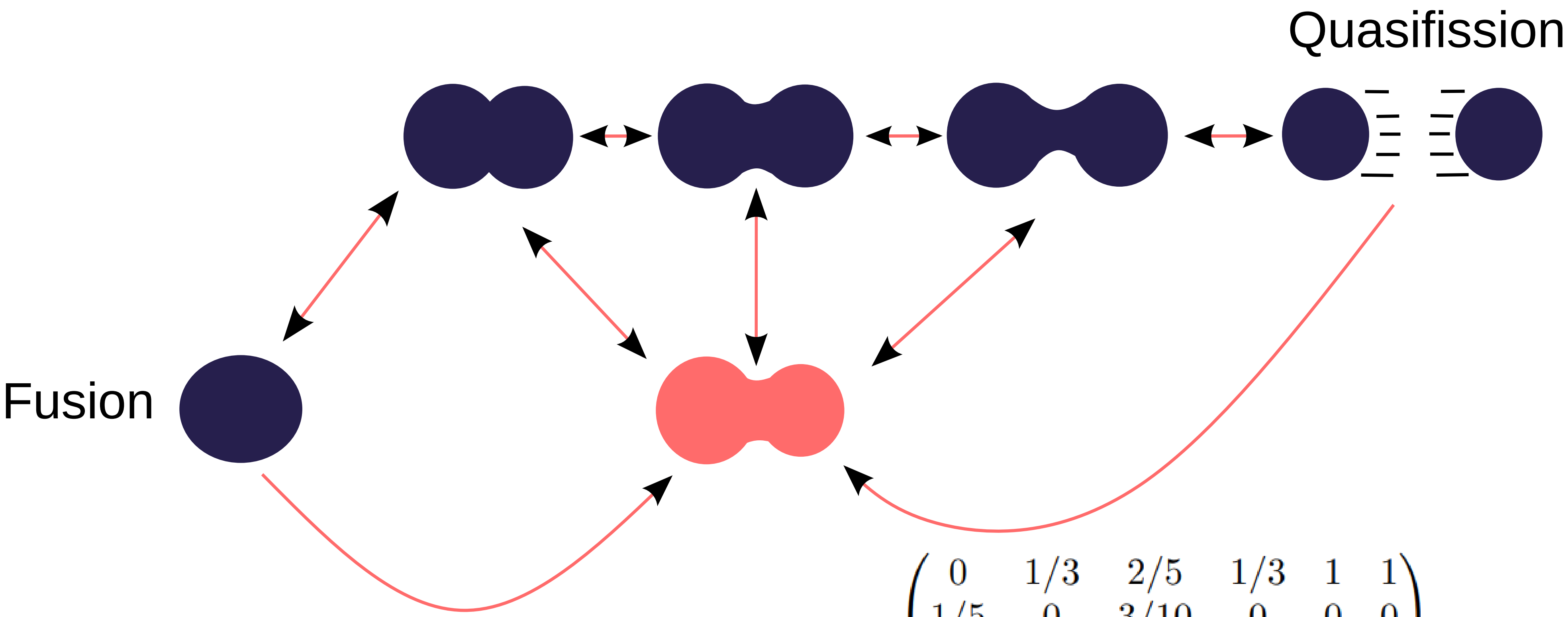
Use FRLDM to compute energy of millions of shapes.

Shape changes with a diffusive process on potential surface.

$$P_{\mathrm{CN}} = \frac{\#\ \text{Random-walks moving into G.S.}}{\#\ \text{Total random-walks}}$$



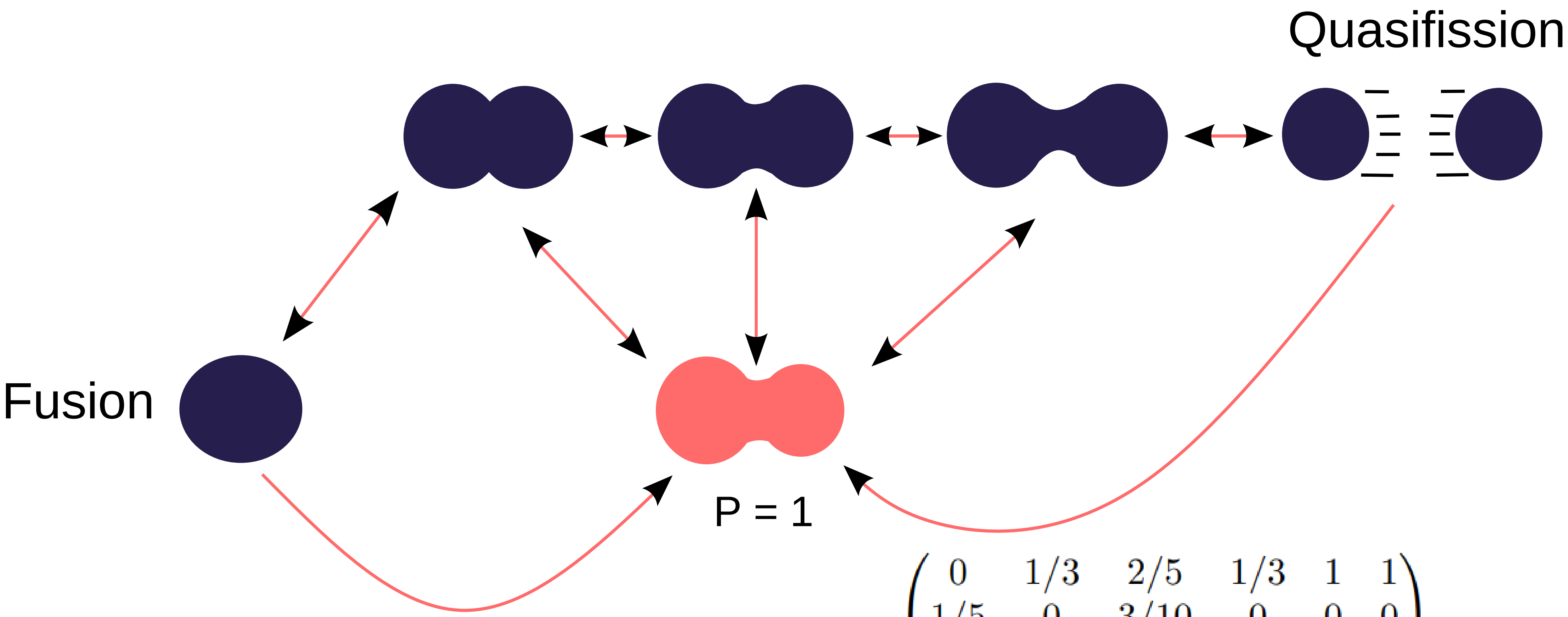
Markov chain



A is a ~ $10^7 \times 10^7$ sparse matrix
filling of order ~ 10^{-5}

$$A = \begin{pmatrix} 0 & 1/3 & 2/5 & 1/3 & 1 & 1 \\ 1/5 & 0 & 3/10 & 0 & 0 & 0 \\ 2/5 & 1/3 & 0 & 1/3 & 0 & 0 \\ 2/5 & 0 & 2/5 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1/3 & 0 & 0 \end{pmatrix}$$

Markov chain

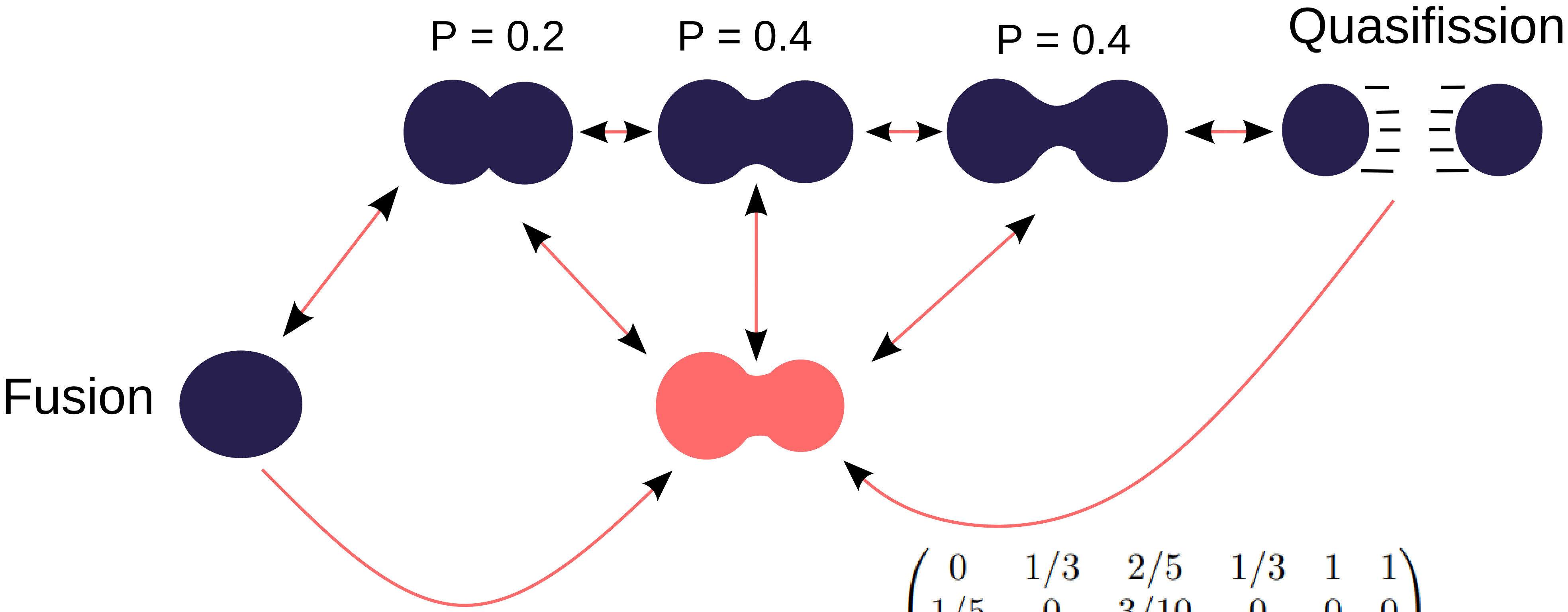


$$A^n p^0 = p^n$$

$$A = \begin{pmatrix} 0 & 1/3 & 2/5 & 1/3 & 1 & 1 \\ 1/5 & 0 & 3/10 & 0 & 0 & 0 \\ 2/5 & 1/3 & 0 & 1/3 & 0 & 0 \\ 2/5 & 0 & 2/5 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1/3 & 0 & 0 \end{pmatrix}$$

$$p^0 = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}$$

Markov chain

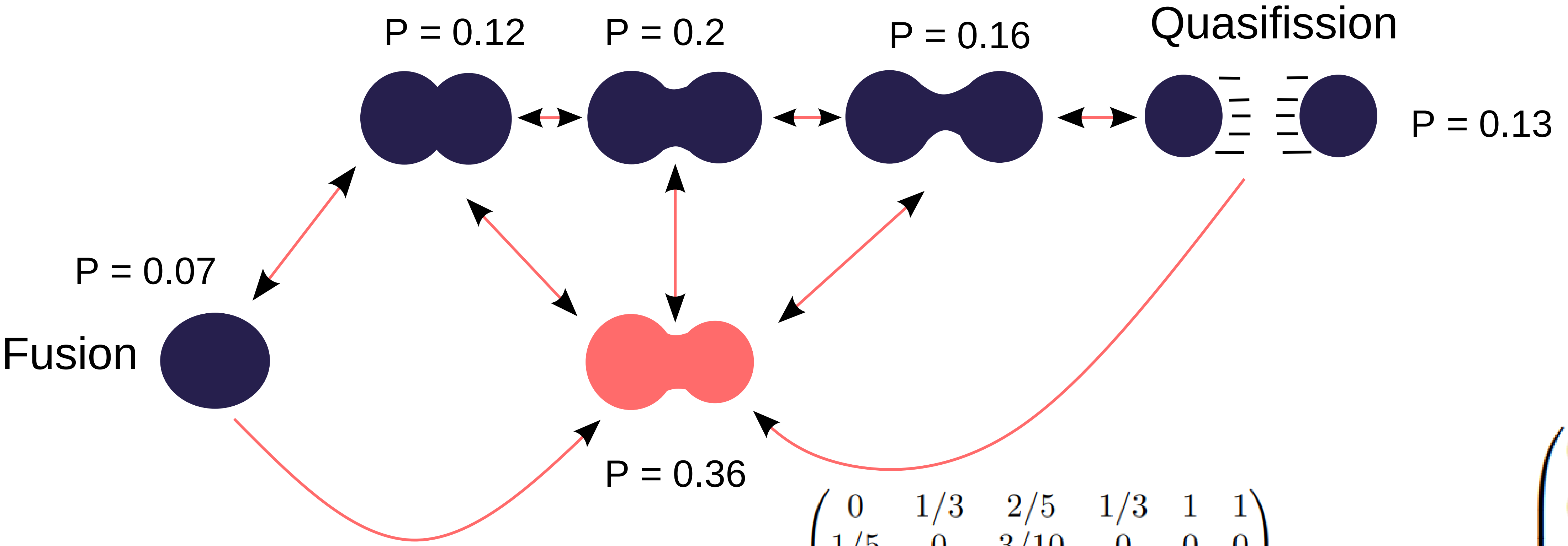


$$A^n p^0 = p^n$$

$$A = \begin{pmatrix} 0 & 1/3 & 2/5 & 1/3 & 1 & 1 \\ 1/5 & 0 & 3/10 & 0 & 0 & 0 \\ 2/5 & 1/3 & 0 & 1/3 & 0 & 0 \\ 2/5 & 0 & 2/5 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1/3 & 0 & 0 \end{pmatrix}$$

$$p^1 = \begin{pmatrix} 0 \\ 0.2 \\ 0.4 \\ 0.4 \\ 0 \\ 0 \end{pmatrix}$$

Markov chain

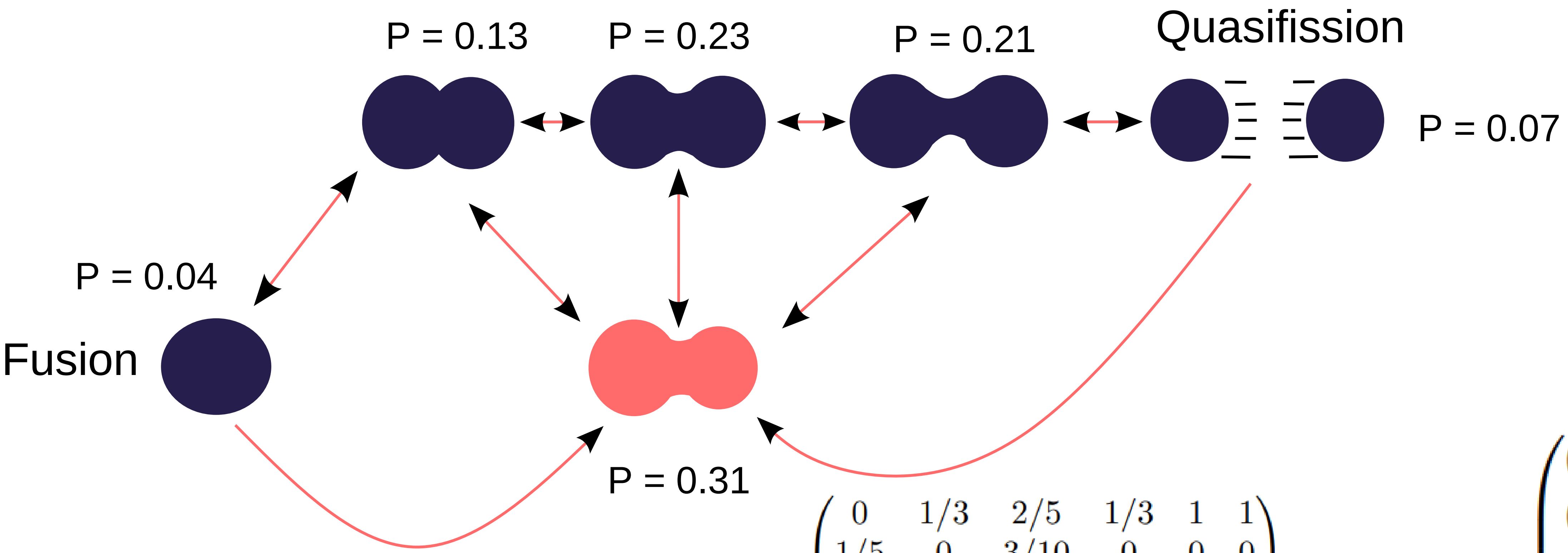


$$P_{\mathrm{CN}} = \frac{0.07}{0.07 + 0.13} \approx 0.43$$

$$A = \begin{pmatrix} 0 & 1/3 & 2/5 & 1/3 & 1 & 1 \\ 1/5 & 0 & 3/10 & 0 & 0 & 0 \\ 2/5 & 1/3 & 0 & 1/3 & 0 & 0 \\ 2/5 & 0 & 2/5 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1/3 & 0 & 0 \end{pmatrix}$$

$$p^2 = \begin{pmatrix} 0.36 \\ 0.12 \\ 0.2 \\ 0.16 \\ 0.07 \\ 0.13 \end{pmatrix}$$

Markov chain



$$P_{\mathrm{CN}} = \frac{0.04}{0.04 + 0.07} \approx 0.38$$

$$A = \begin{pmatrix} 0 & 1/3 & 2/5 & 1/3 & 1 & 1 \\ 1/5 & 0 & 3/10 & 0 & 0 & 0 \\ 2/5 & 1/3 & 0 & 1/3 & 0 & 0 \\ 2/5 & 0 & 2/5 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1/3 & 0 & 0 \end{pmatrix}$$

$$p^{\infty} = \begin{pmatrix} 0.31 \\ 0.13 \\ 0.23 \\ 0.21 \\ 0.04 \\ 0.07 \end{pmatrix}$$

Markov chain

Convergence of the vector p means that $Ap^{\infty} = p^{\infty}$

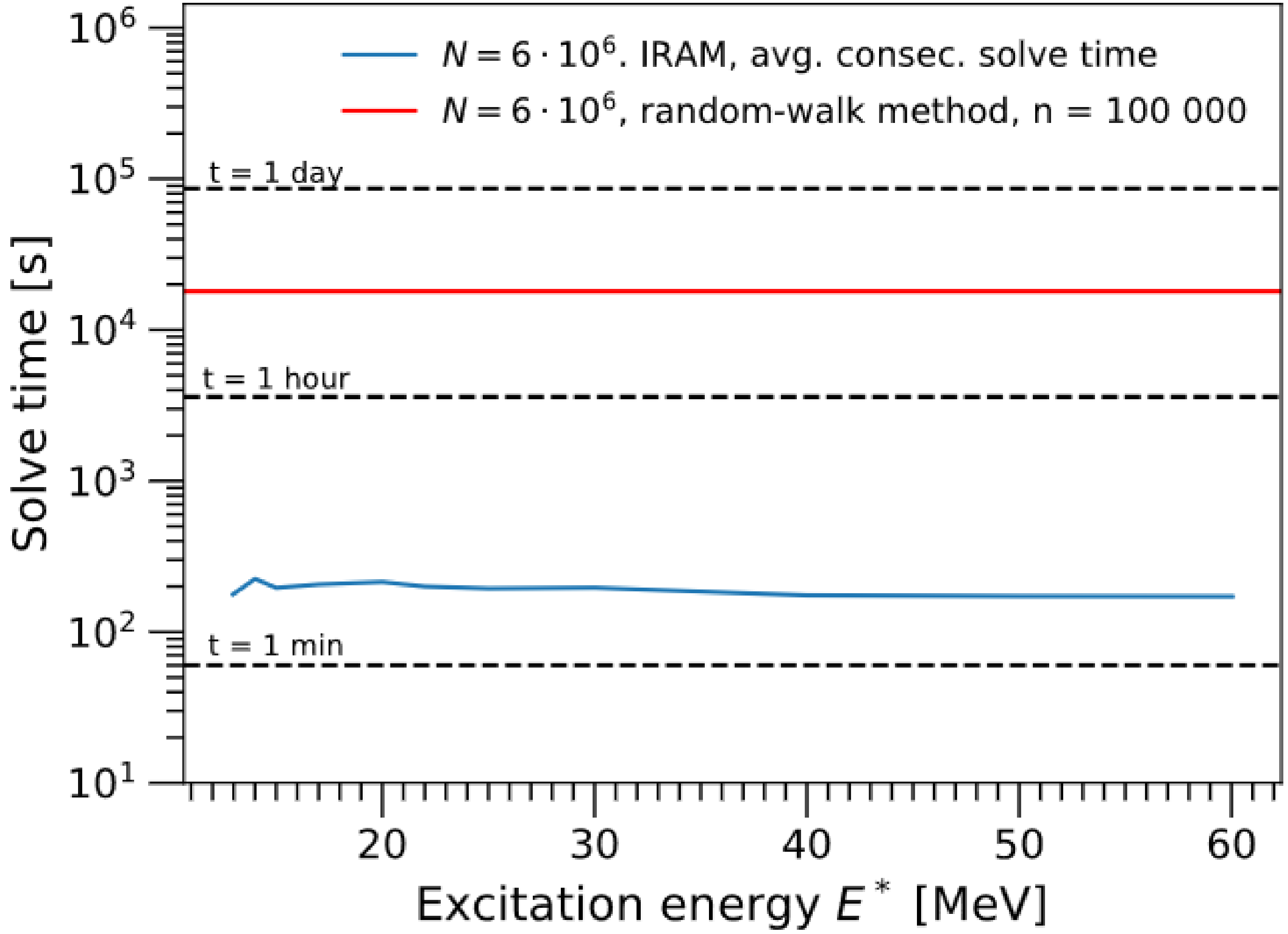
Eigenvalue problem with eigenvalue 1.

$|\lambda| \leq 1$ for all of A’s eigenvalues

Use implicitly restarted Arnoldi method (IRAM) to find eigenvalue and eigenvector of interest.

Implementation: ARPACK-NG (github.com/opencollab/arpack-ng)

Solve time for one partial wave $P_{CN}(E, L)$



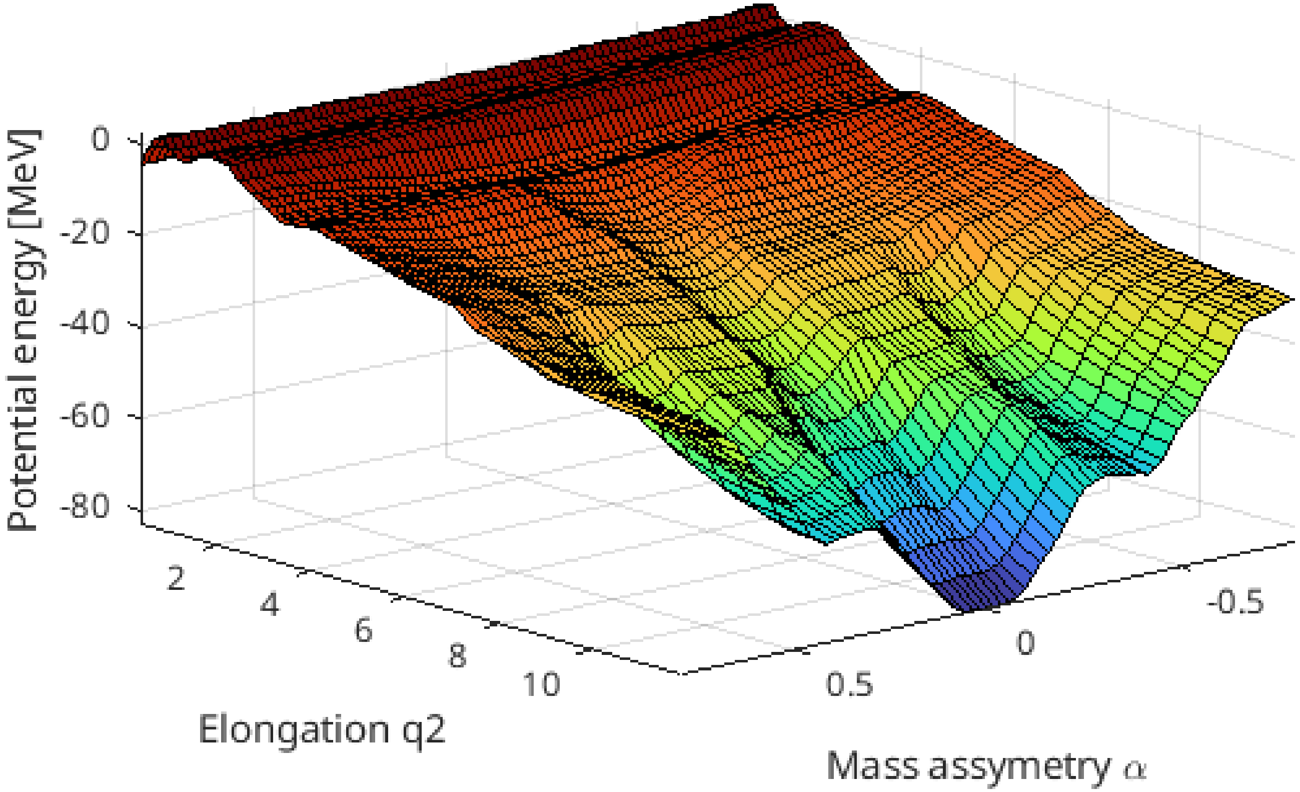
Starting location on PES?

Once nuclei collide, they hav
Friction between nucleons cc
excitation energy.

$$\mu\ddot{R} = -\nabla_R U_I(R) - \boxed{\beta(R)\dot{R}}$$

$$\beta(R) = \frac{1}{2} m_n \rho \bar{v} \pi c^2_{\mathrm{n}}$$

Use Langevin Eq. of motion u



Fusion path

$$\mu \ddot{R} = -\nabla_R U_I(R) - \beta(R)\dot{R} - \sigma(R)r(t)$$

Friction along 1D path in 5D landscape.
Starting condition:

- Distance q_2: Large
- Neck c_{neck}: Small.
- Mass asymmetry α: Projectile & Target
- Deformations ε: G.S deformation.

Path defined by following the gradient of PES in the parameters ε_l, ε_r, c_{neck} to lower elongations.

α frozen to initial value.

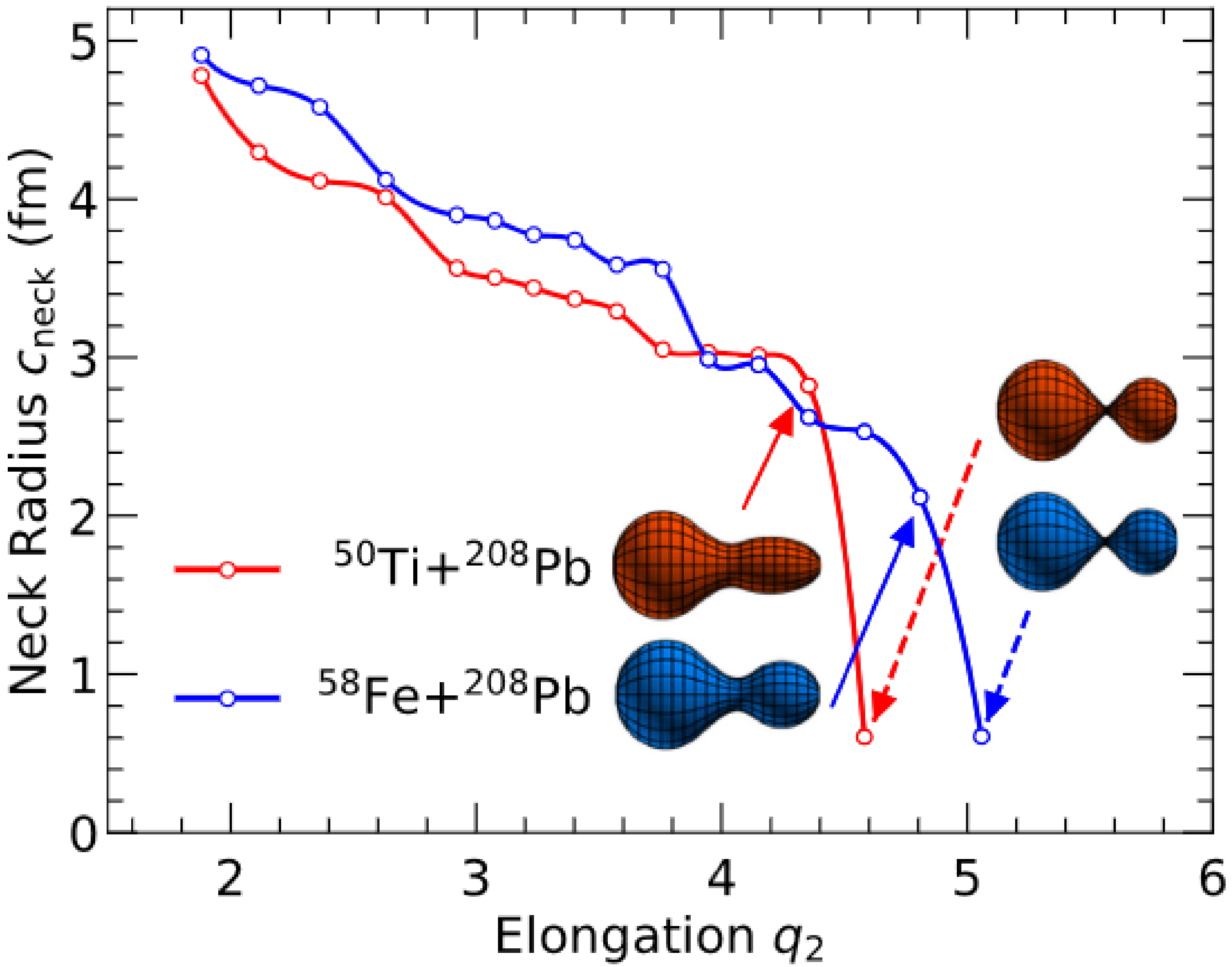


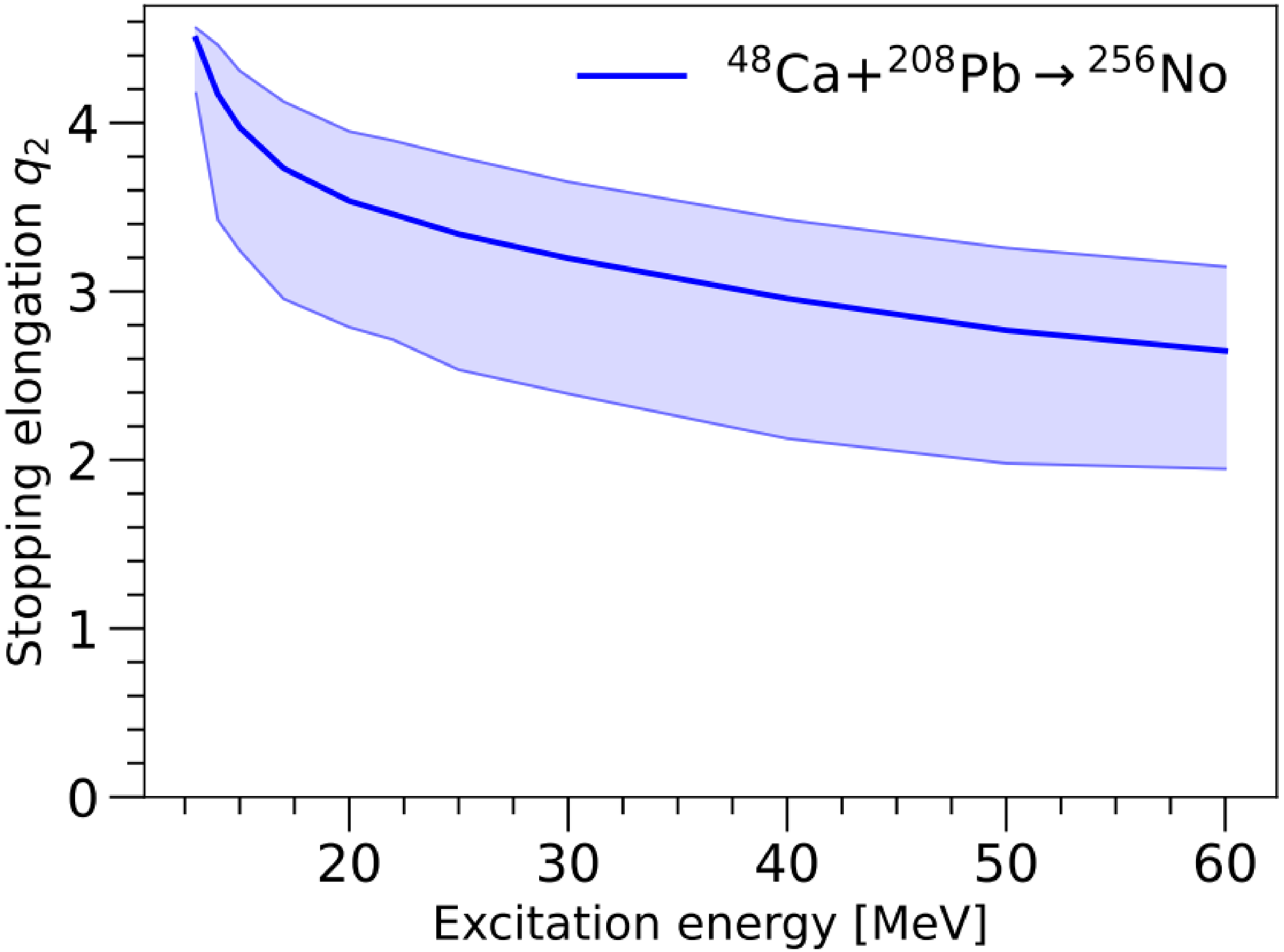
Fig. from M. Albertsson, et.al, Phys. Rev. C. 110, (2024)

Fusion path

$$\mu \ddot{R} = -\nabla_R U_I(R) - \beta(R)\dot{R} - \sigma(R) r(t)$$

Friction along 1D path in 5D landscape.
Starting condition:

- Distance q_2: Large
- Neck c_{neck}: Small.
- Mass asymmetry α: Projectile & Target
- Deformations ε: G.S deformation.



Path defined by following the gradient of PES in the parameters ε_l, ε_r, c_{neck} to lower elongations.

α frozen to initial value.

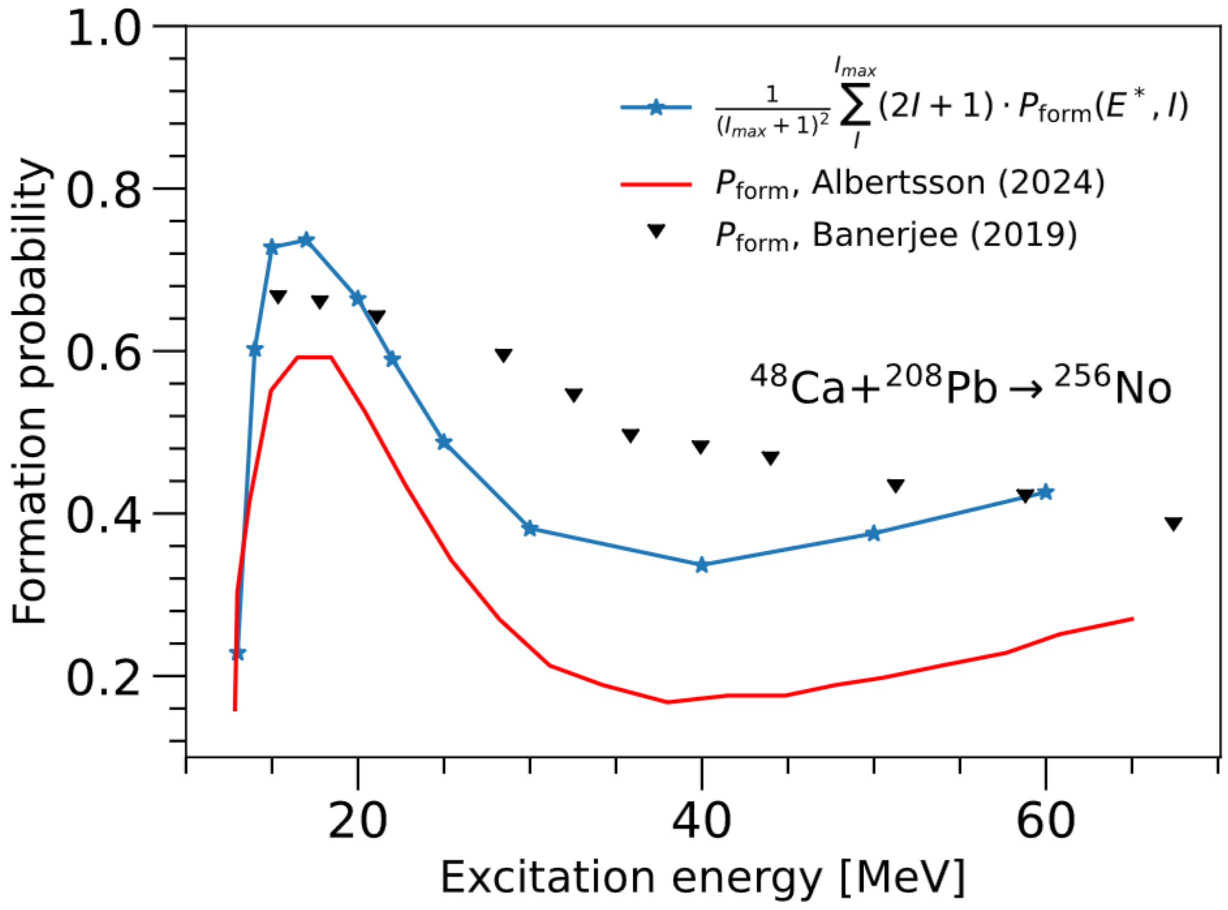
Formation probability

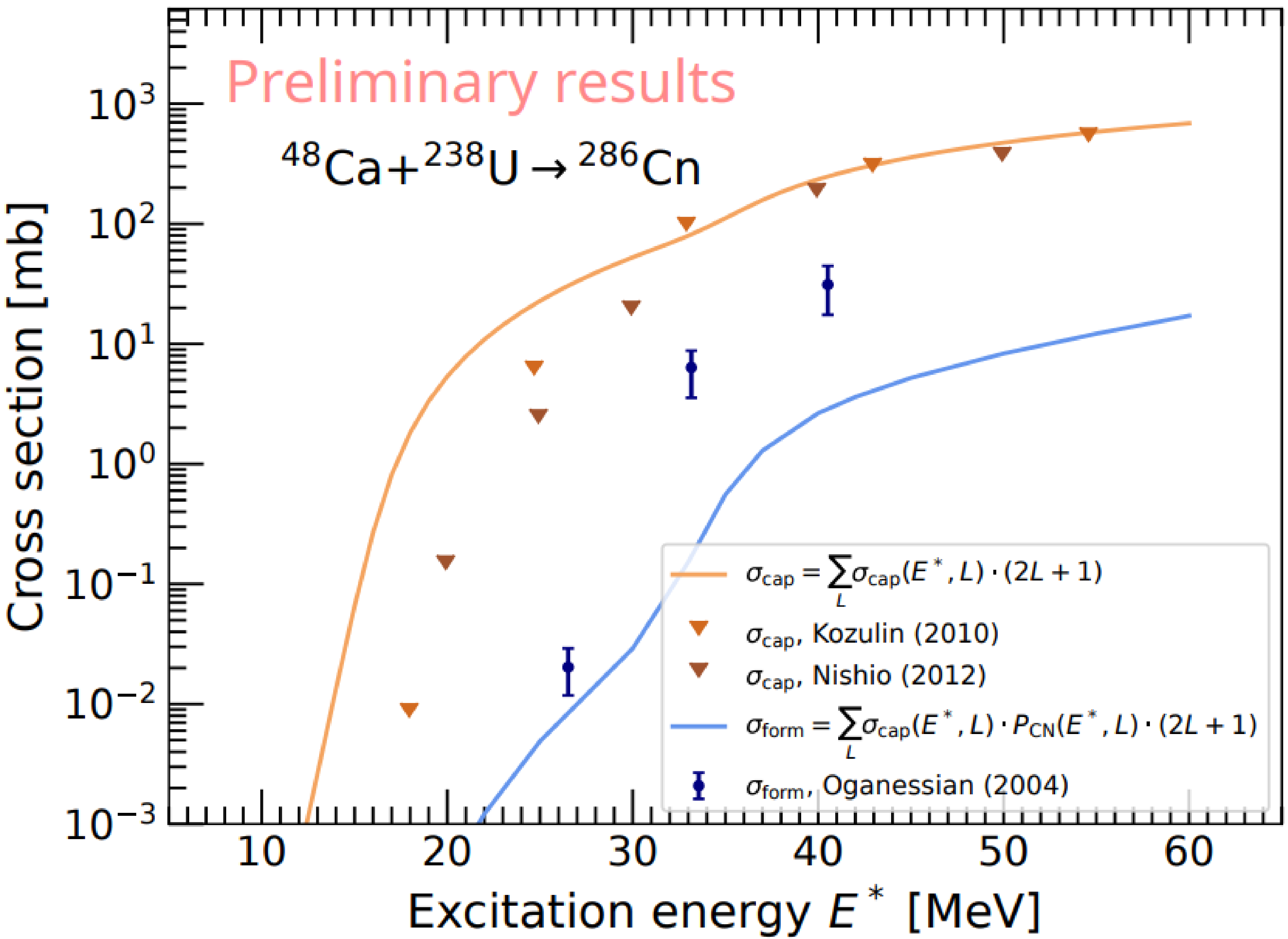
Difference between blue and red calculation is the choice of 1D fusion path to run friction on.

Blue – Follow gradient in parameters ε_l, ε_r, c_{neck}

Red – Follow gradient in parameters ε_l, ε_r, c_{neck}, α

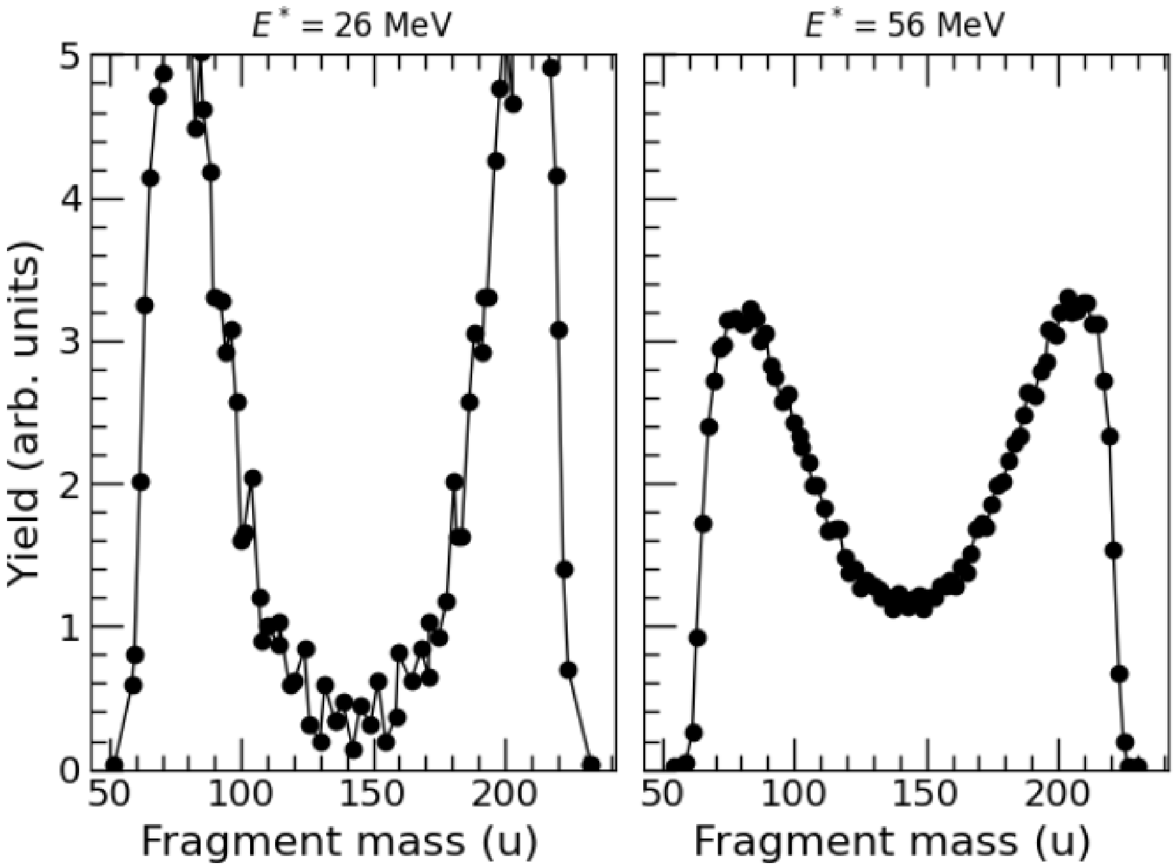
Sensitive to choice of path.

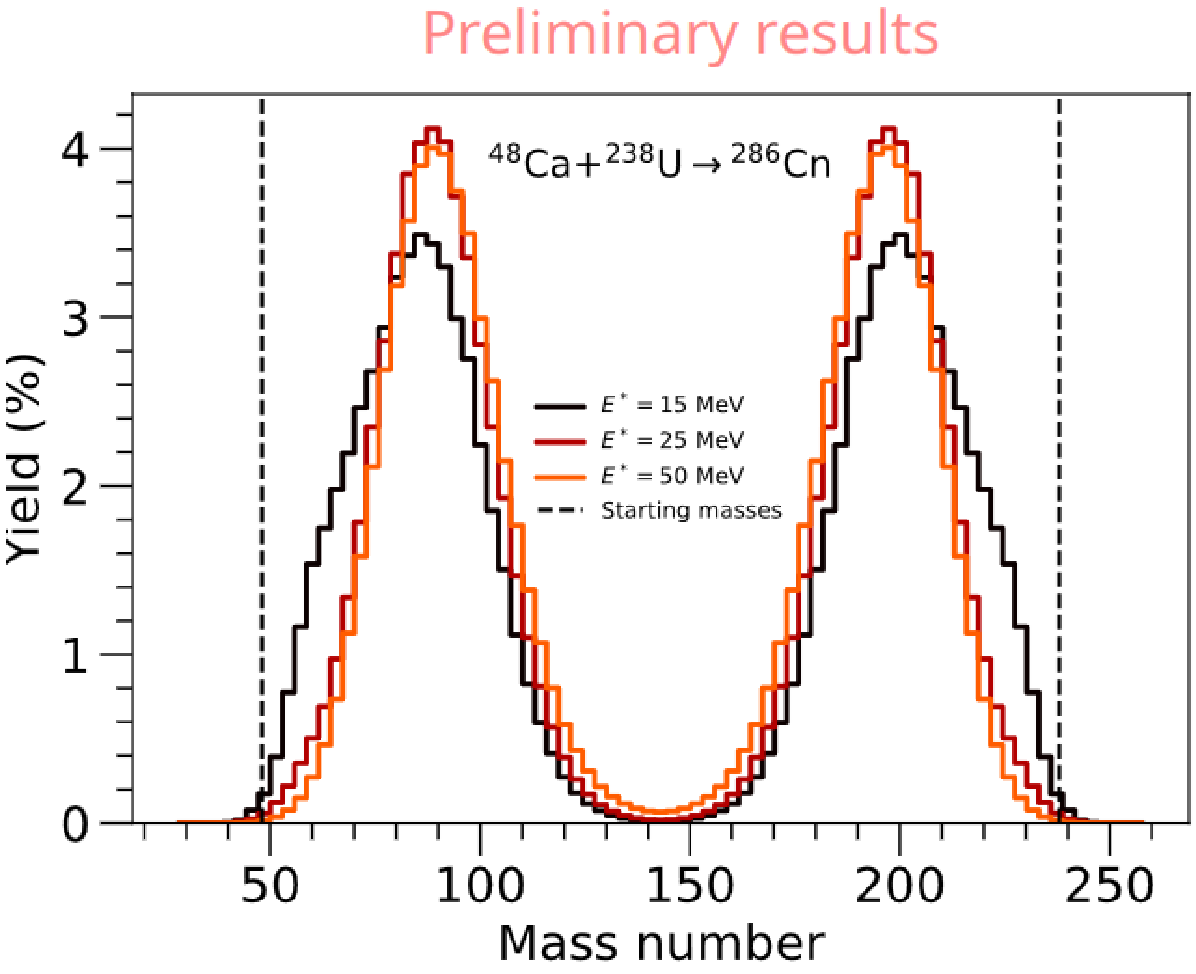




Preliminary results
$^{48}Ca+^{238}U \rightarrow ^{286}Cn$
Cross section [mb]
Excitation energy E^* [MeV]
$\sigma_{cap} = \sum_L \sigma_{cap}(E^*, L) \cdot (2L+1)$
σ_{cap}, Kozulin (2010)
σ_{cap}, Nishio (2012)
$\sigma_{form} = \sum_L \sigma_{cap}(E^*, L) \cdot P_{CN}(E^*, L) \cdot (2L+1)$
σ_{form}, Oganessian (2004)

Quasifission fragment mass distribution





M. G. Itkis, et.al, Eur. Phys. J. A **58**, 178 (2022).

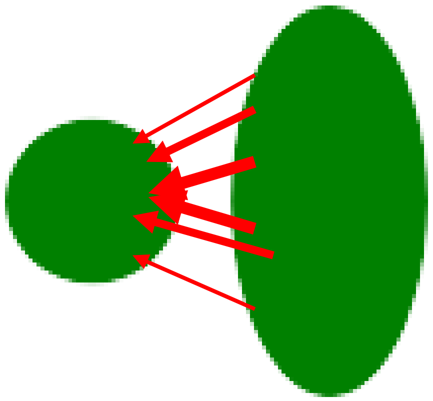
Outlook for 5D model

- Compute survival probability
- Extend to odd nuclei
- Add degrees of freedom in potential
    - Z/N degree of freedom in target/projectile
- Formation probability sensitive to start location:
    - Friction in a higher dimensional path.

Thank you for your attention!

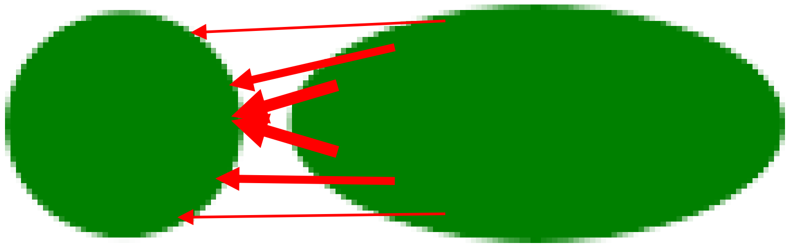
Capture

Side collision



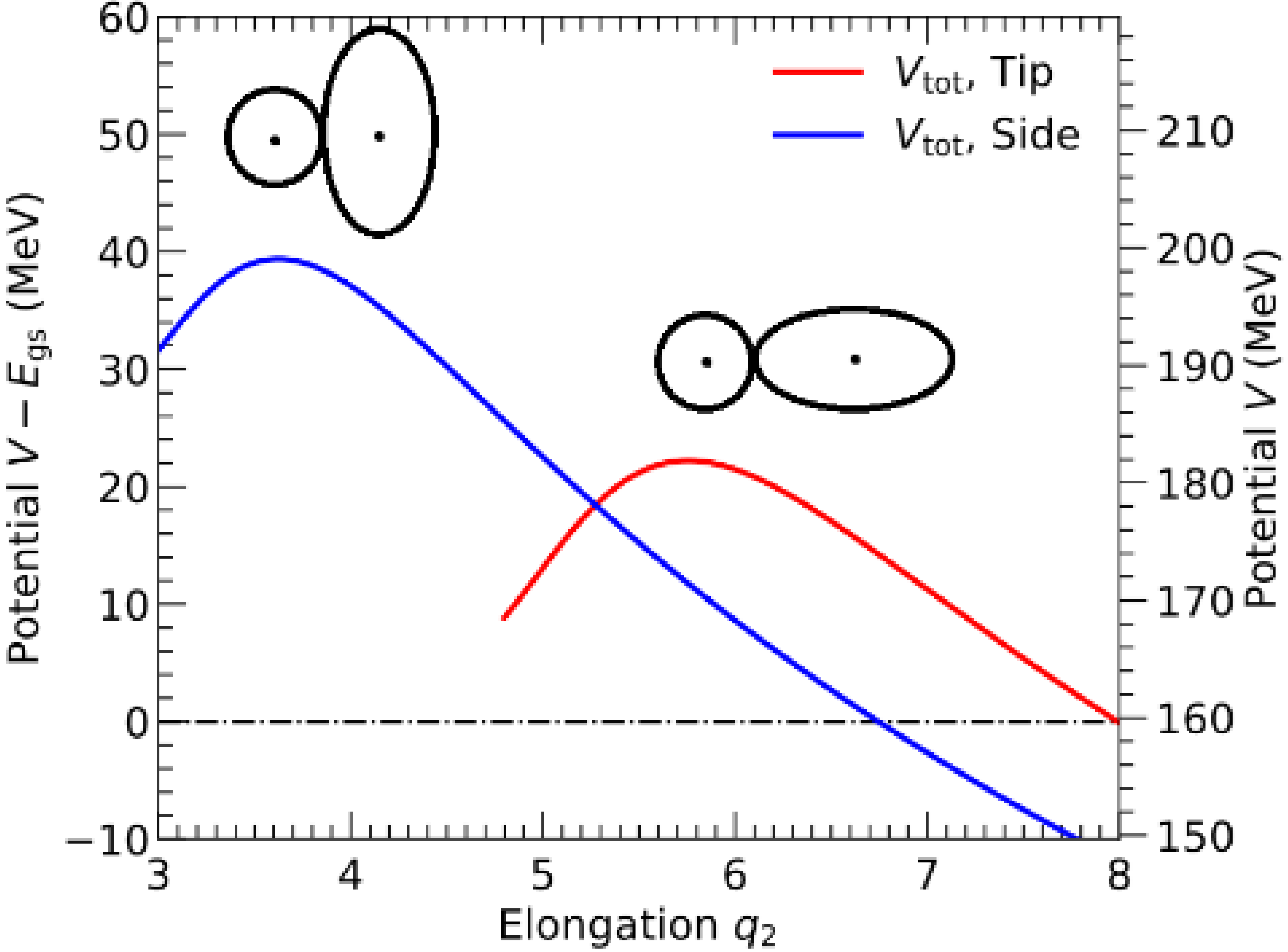
More repulsion
Larger Coulomb barrier

Tip collision



Less repulsion
Smaller Coulomb barrier

Capture



V_{tot}, Tip
V_{tot}, Side
Potential $V - E_{gs}$ (MeV)
Potential V (MeV)
Elongation q_2
60
50
40
30
20
10
0
−10
210
200
190
180
170
160
150
3
4
5
6
7
8

Capture

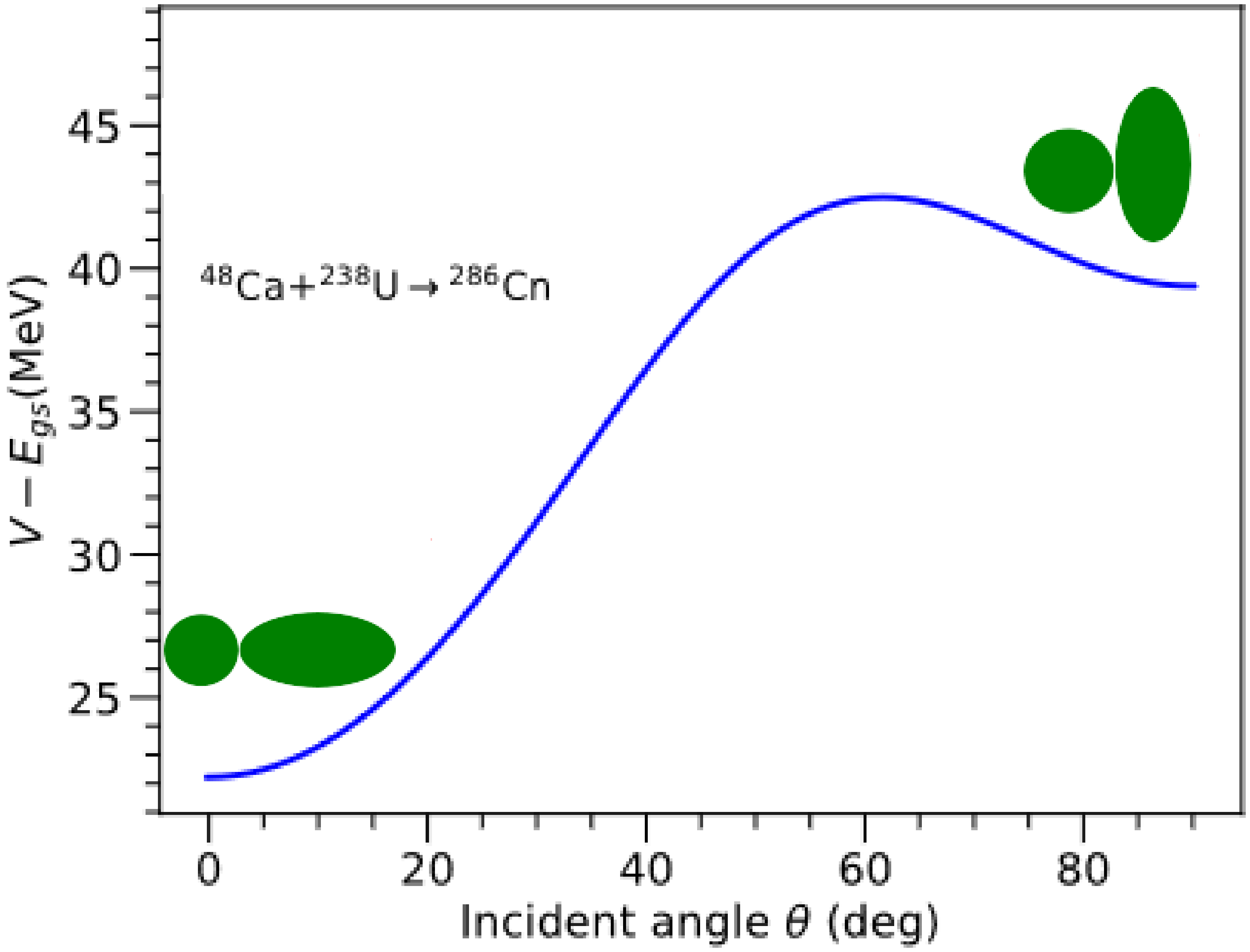
Incident angle distribution:

$\sin\theta$

0 degrees = tip

90 degrees = side

Side collision more likely

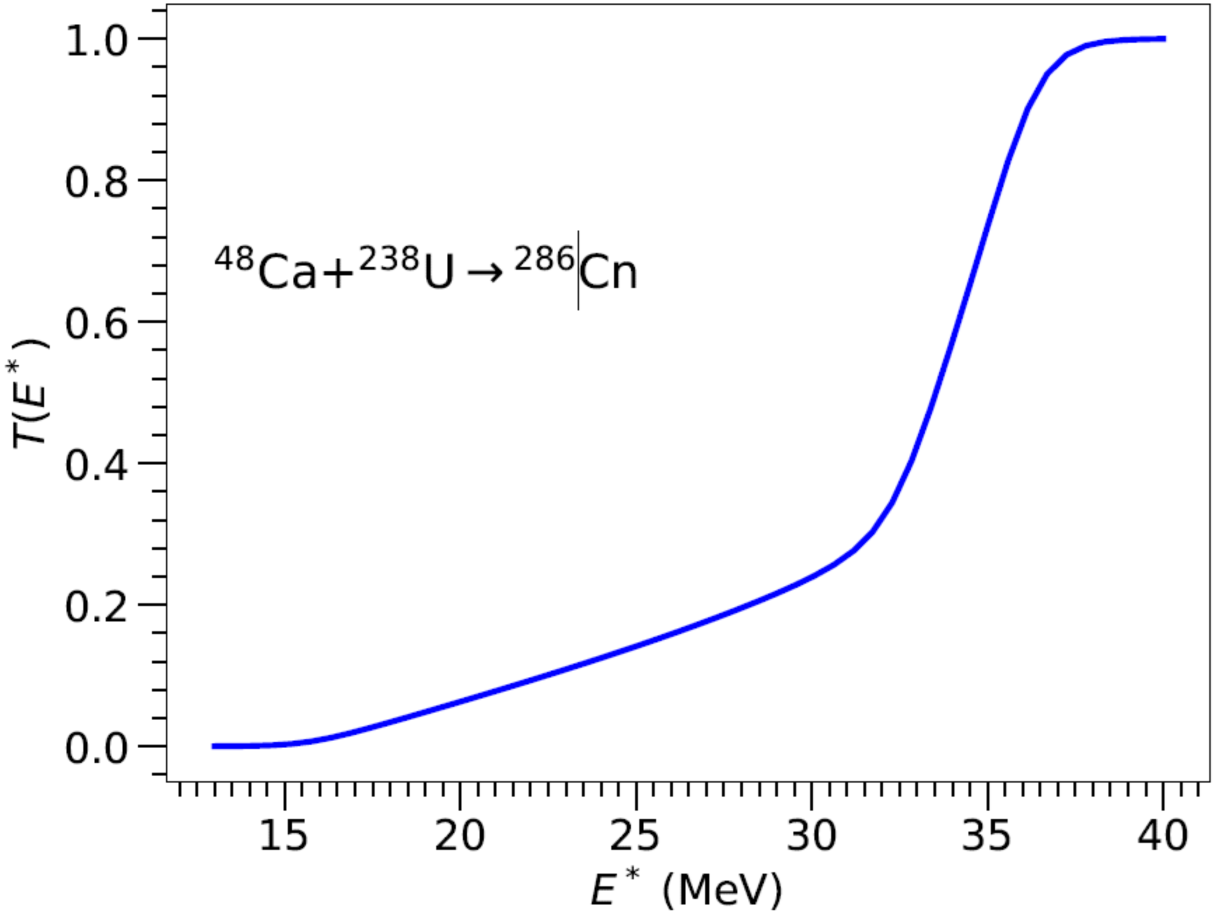


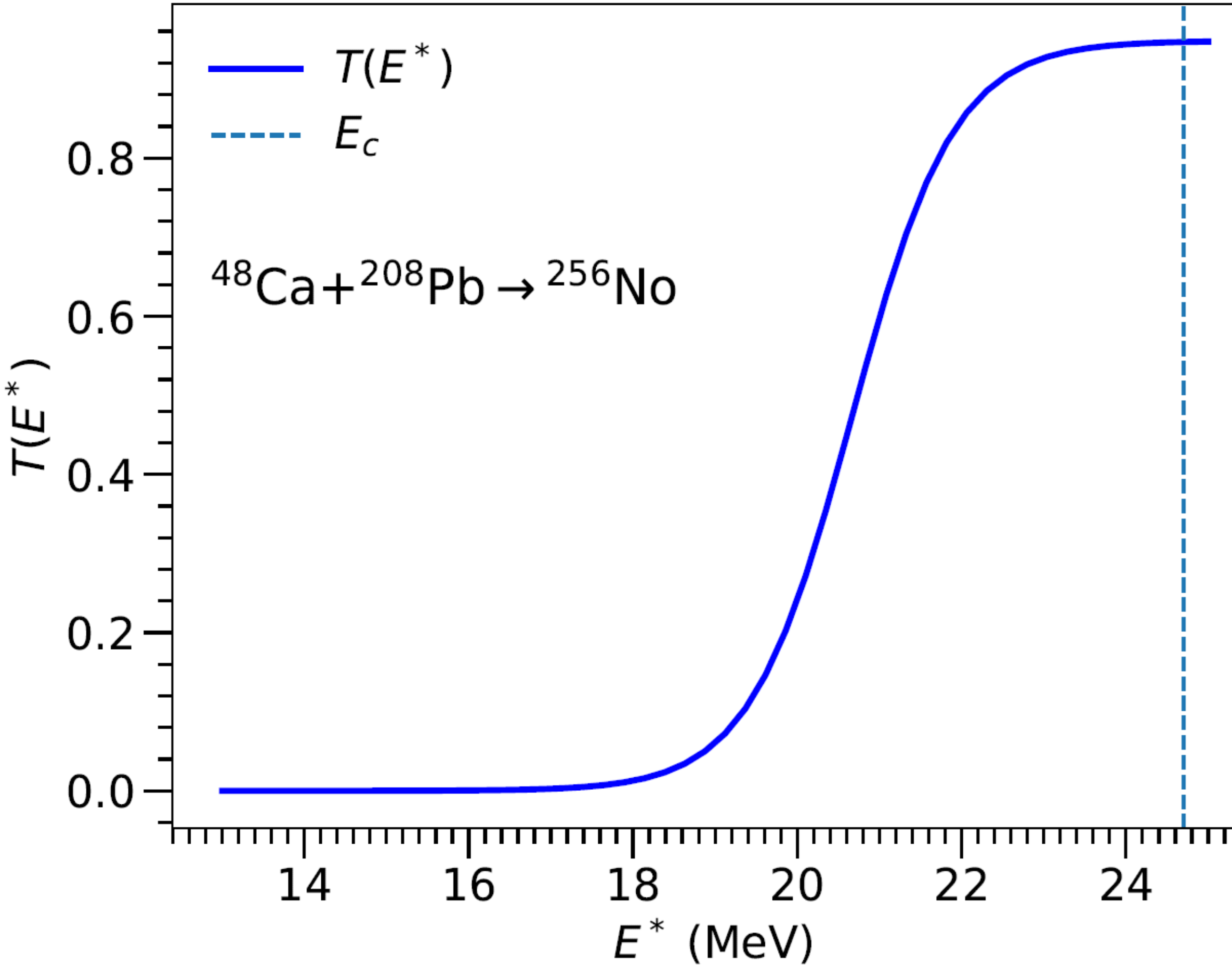
$$T(E^*, \theta, I) = \frac{1}{1 + \exp\left(\frac{2\pi}{\hbar\Omega(\theta,I)}\left[V_B(\theta, I) - E^*\right]\right)},$$

Capture

Integrate transmission prob over all angles

$$T(E^*) = \int_0^{\pi/2} T(E^*, \theta) \sin\theta d\theta$$

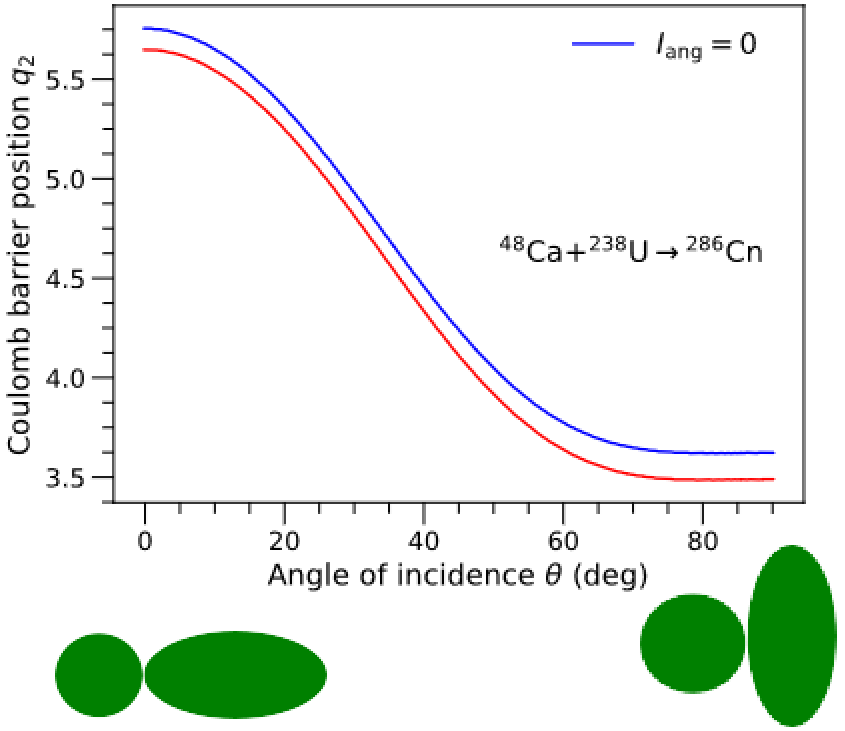


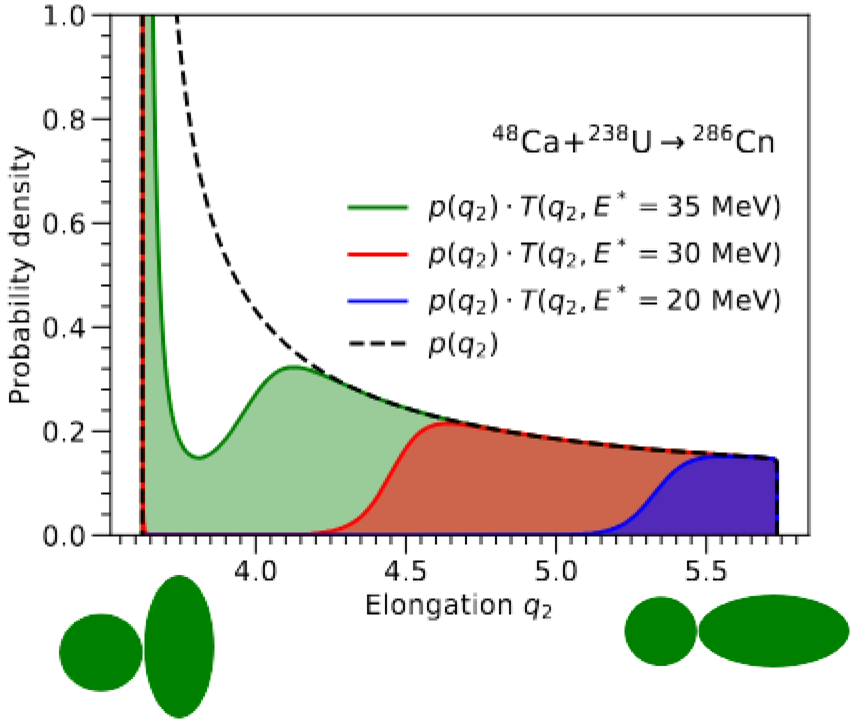


Capture

Variable transform angle into elongation to get Coulomb barrier position distribution. Coulomb barrier position determines injection point distance in the formation stage.

$$p(q_2) = \frac{1}{\frac{dq_2}{d\theta}} \sin\theta(q_2)$$



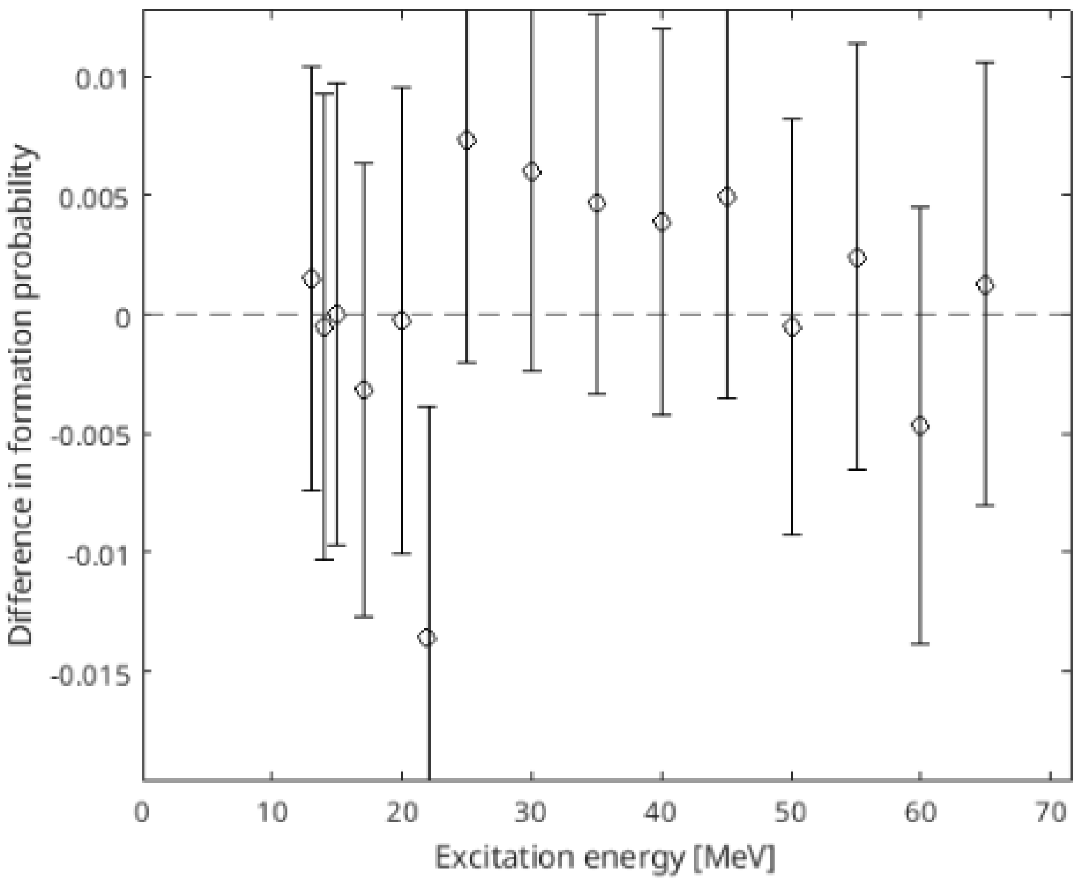


Linear systems method

- Did not spend too much time on improving the linear systems method as the performance was already two orders of magnitude better
- There are methods that can use starting guesses. You could try using them instead. This should improve the performance even further. One idea:
  - Inverse iteration aka inverse power method.

Method validation

Test eigenvector method against random walk algorithm.



15 calculations. 95% error bars

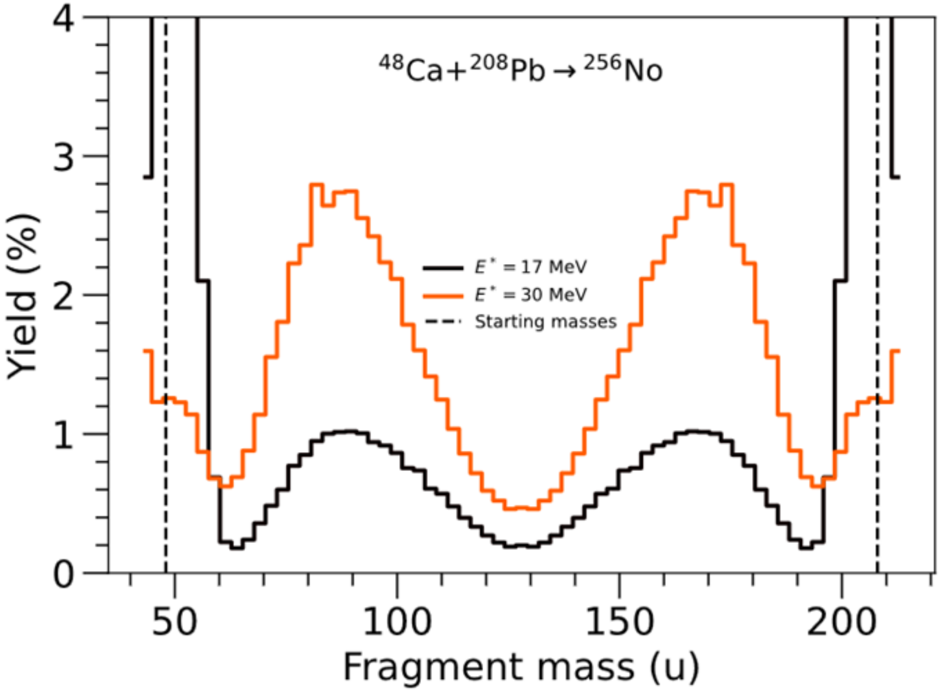
(0.95)^15 = 46% =>

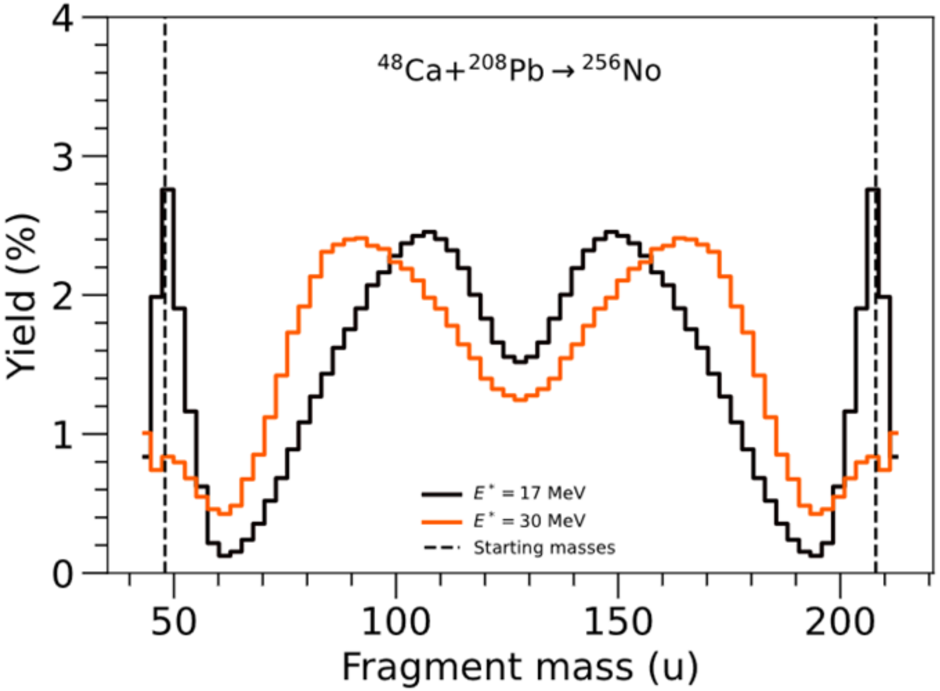
Probability of at least one calculation being outside error bars = 54%

$$\sigma_{\hat{p}} = \sqrt{\frac{\hat{p}(1-\hat{p})}{n}}.$$

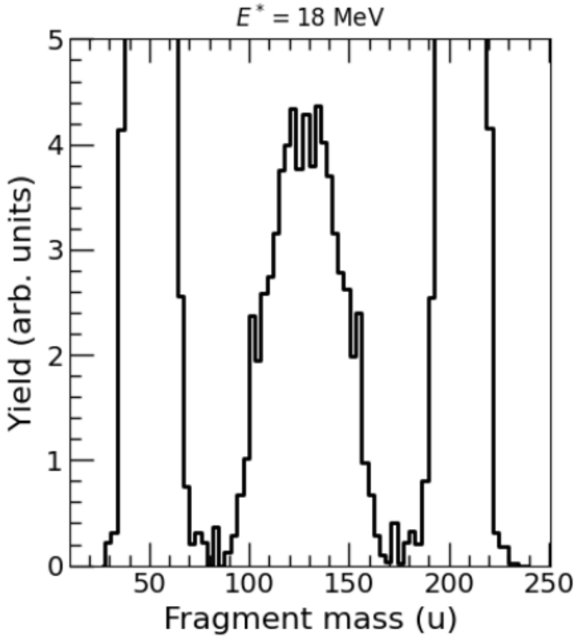
$$n \geq \frac{4 \times 1.96^2(1-\hat{p})}{\hat{p} \times 10^{-2d}}.$$

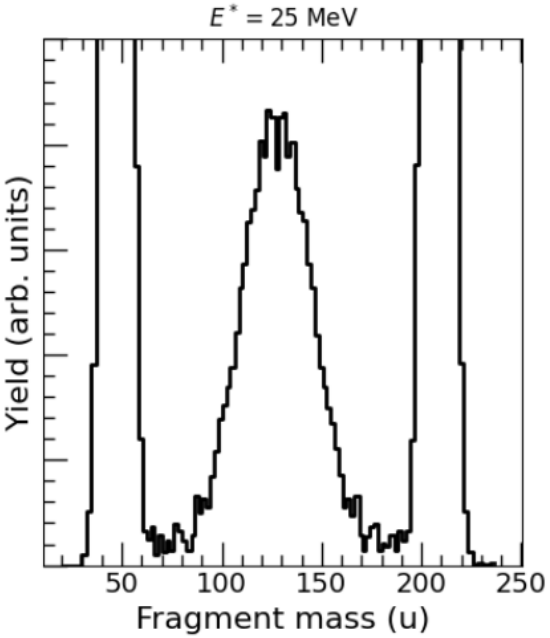
Prediction



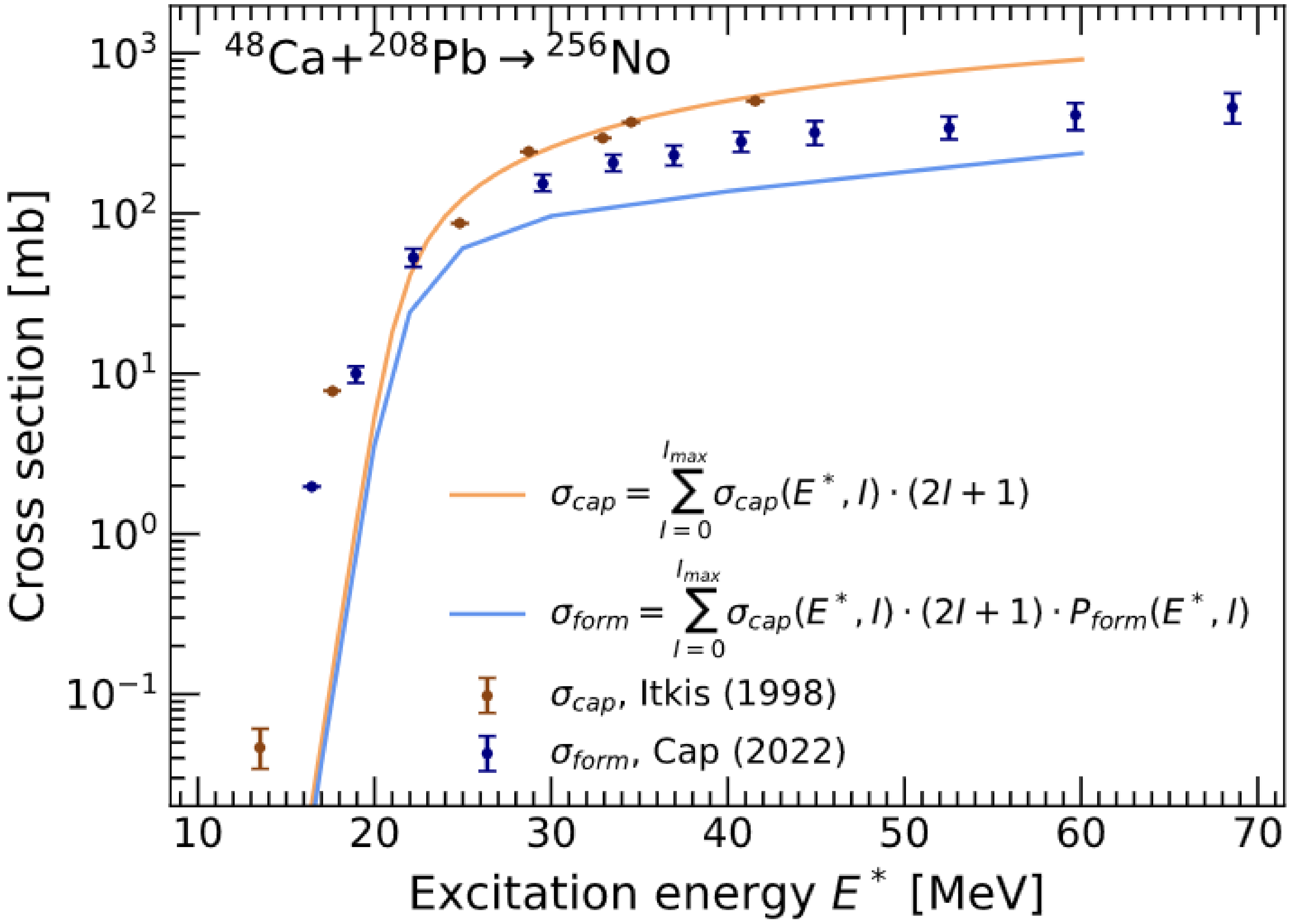


Exp data:





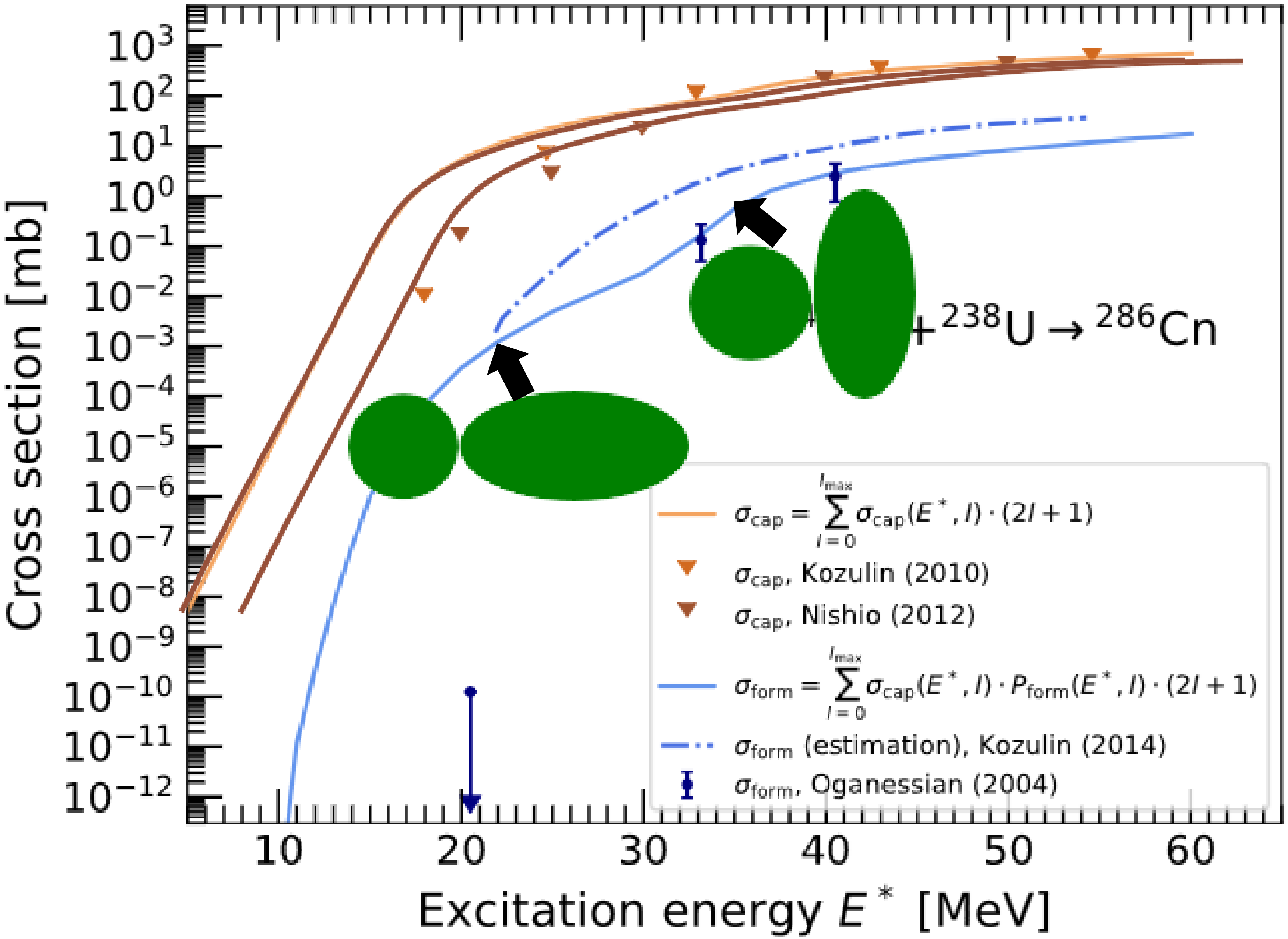
Results



$^{48}Ca+^{208}Pb \rightarrow ^{256}No$
Cross section [mb]
10^3
10^2
10^1
10^0
10^{-1}
10
20
30
40
50
60
70
Excitation energy E^* [MeV]
$\sigma_{cap} = \sum_{l=0}^{l_{max}} \sigma_{cap}(E^*, l) \cdot (2l+1)$
$\sigma_{form} = \sum_{l=0}^{l_{max}} \sigma_{cap}(E^*, l) \cdot (2l+1) \cdot P_{form}(E^*, l)$
σ_{cap}, Itkis (1998)
σ_{form}, Cap (2022)

Results

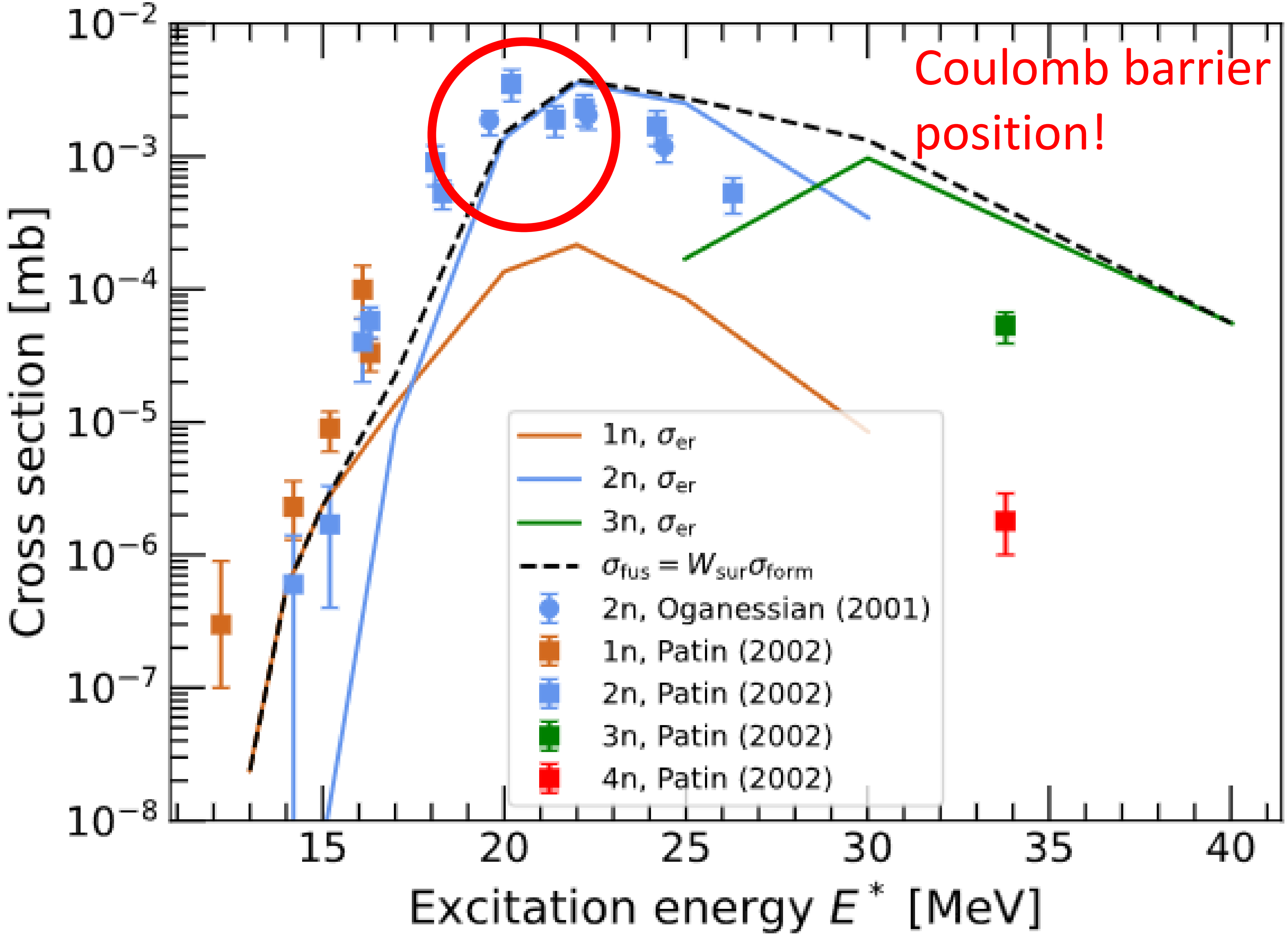
Cn officially named 2010



3. Survival stage

Results

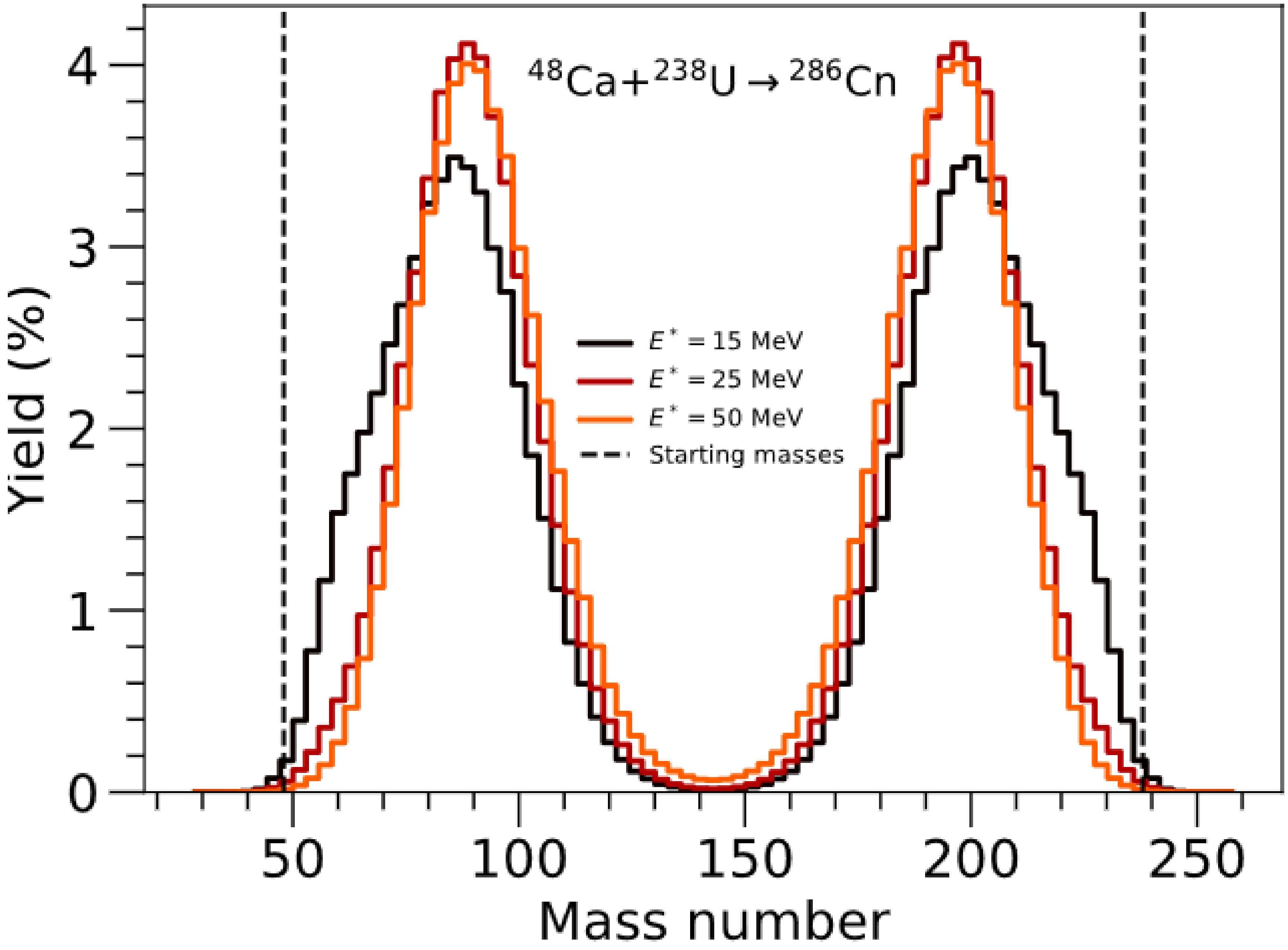
No discovered
1950s-1960s
(disputed)



Thank you for listening!

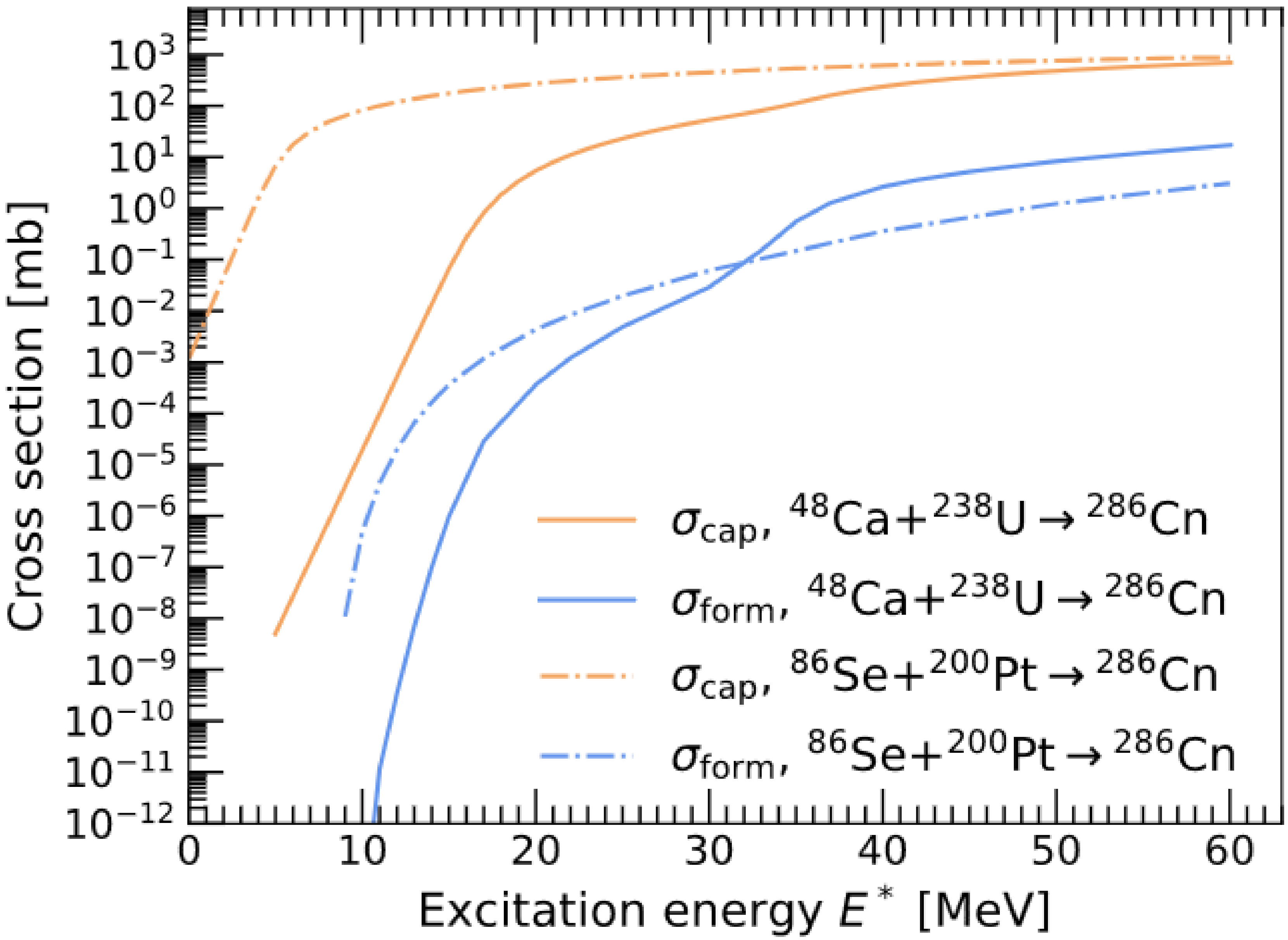
Results

Cn officially named 2010



Results

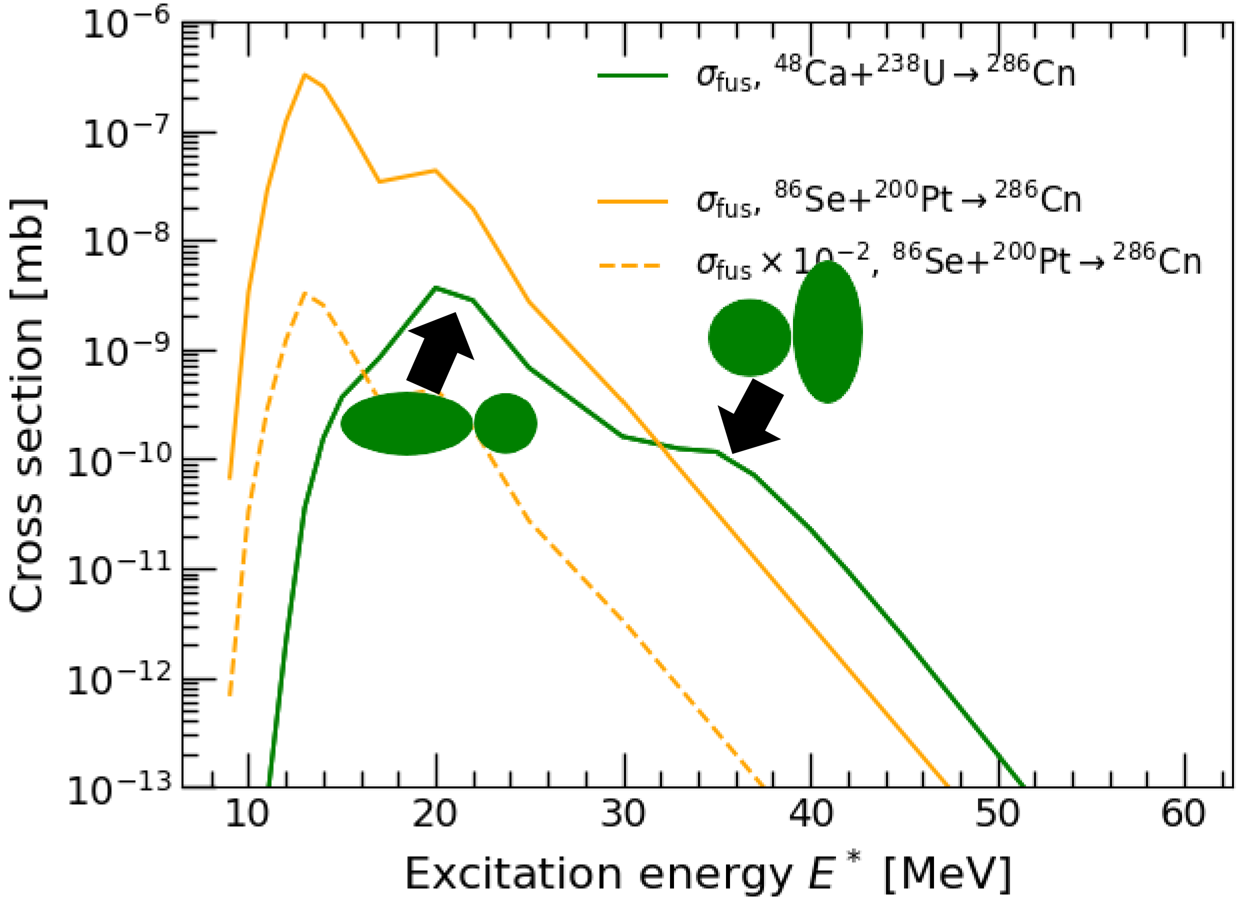
Cn officially named 2010



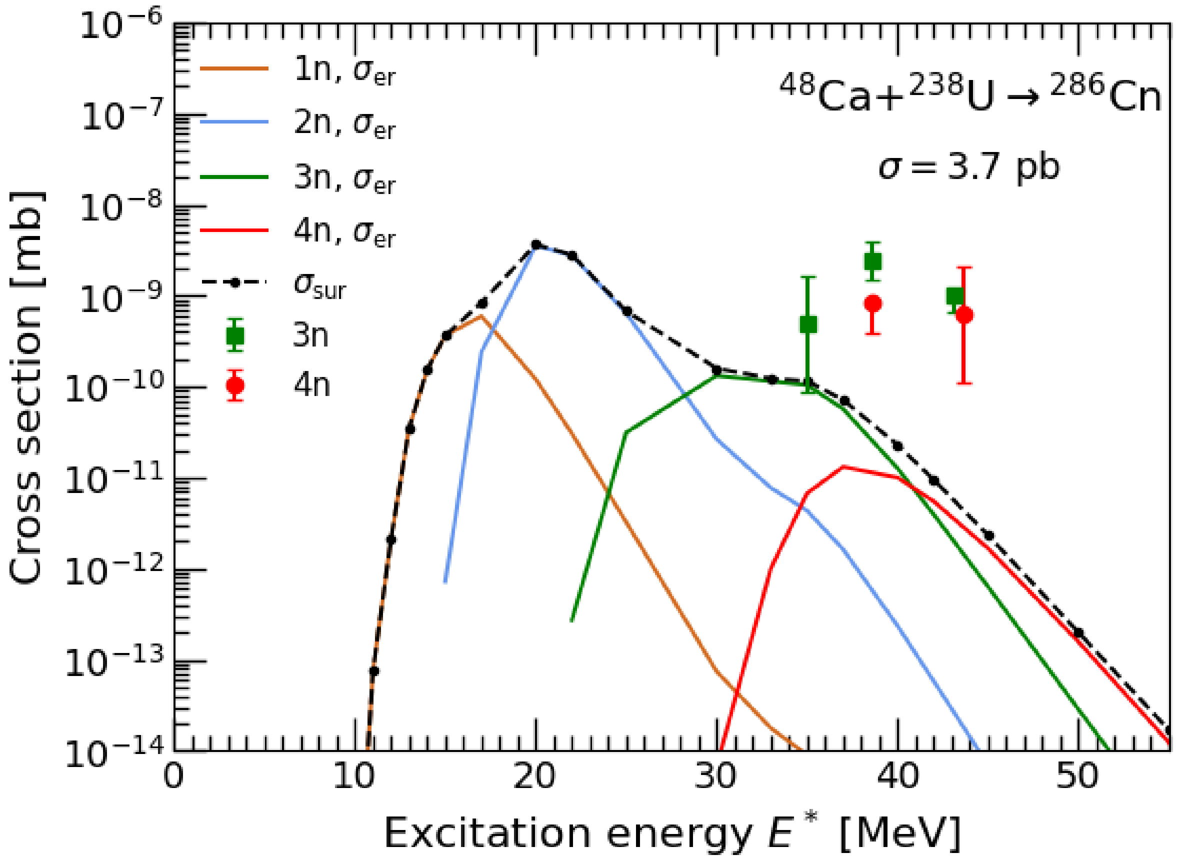
Results

Survival probability from Kewpie2

Cn officially named 2010



Results



^{48}Ca+^{238}U → ^{286}Cn
$\sigma = 3.7$ pb
1n, σ_{er}
2n, σ_{er}
3n, σ_{er}
4n, σ_{er}
σ_{sur}
3n
4n
Cross section [mb]
Excitation energy E^* [MeV]

Results

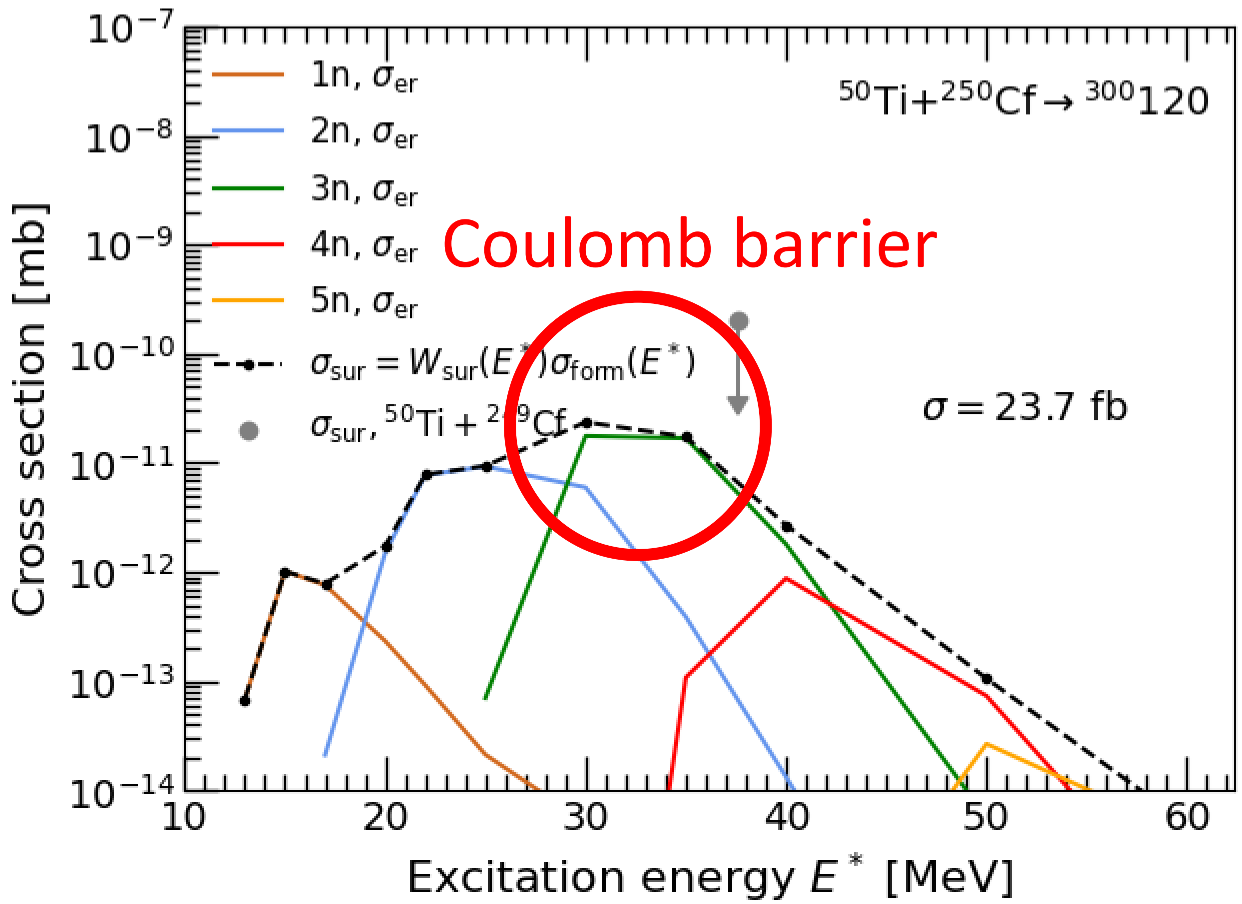
Survival prob from

Kewpie2

Exp. Data for

In figure for

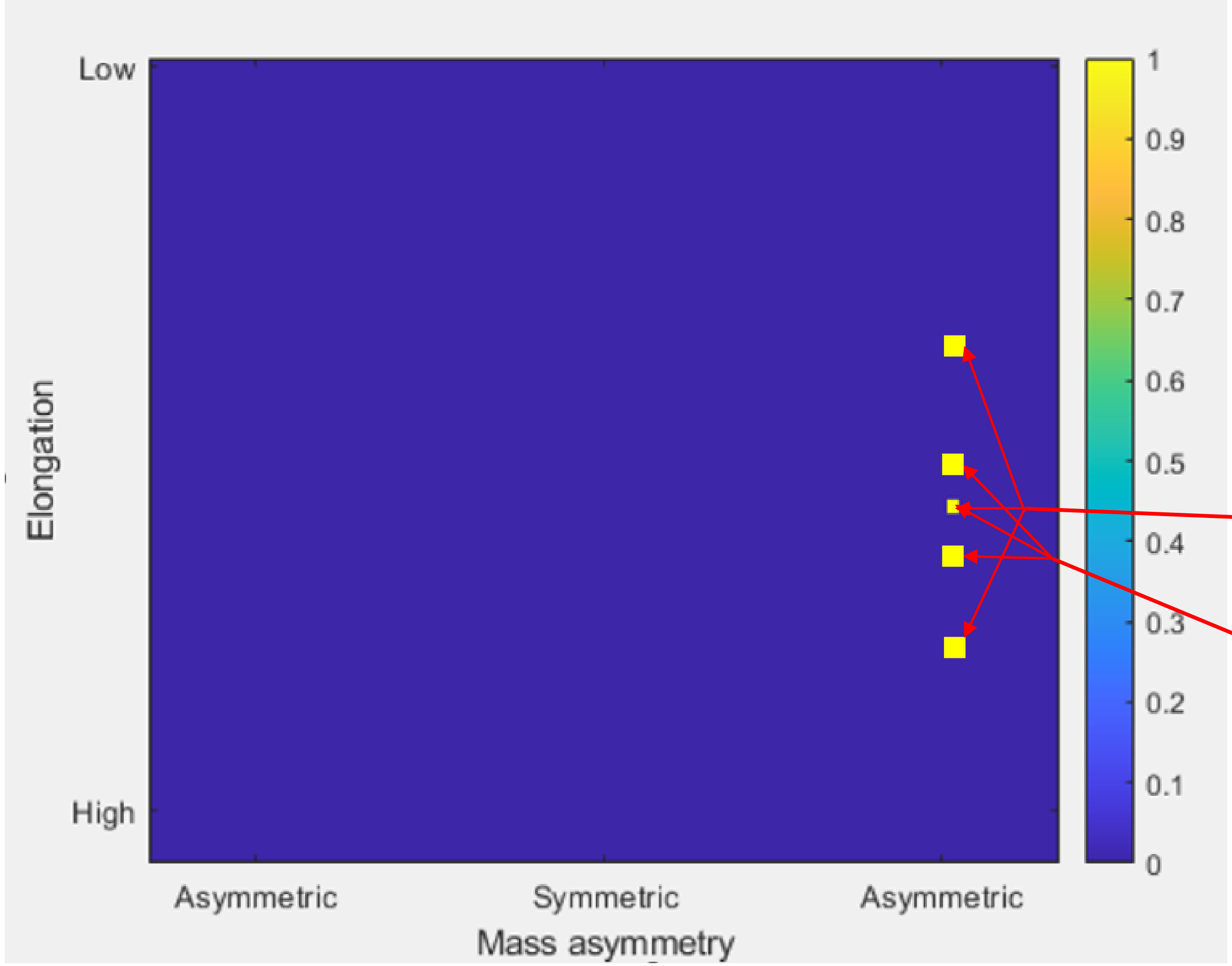
$^{299}120$



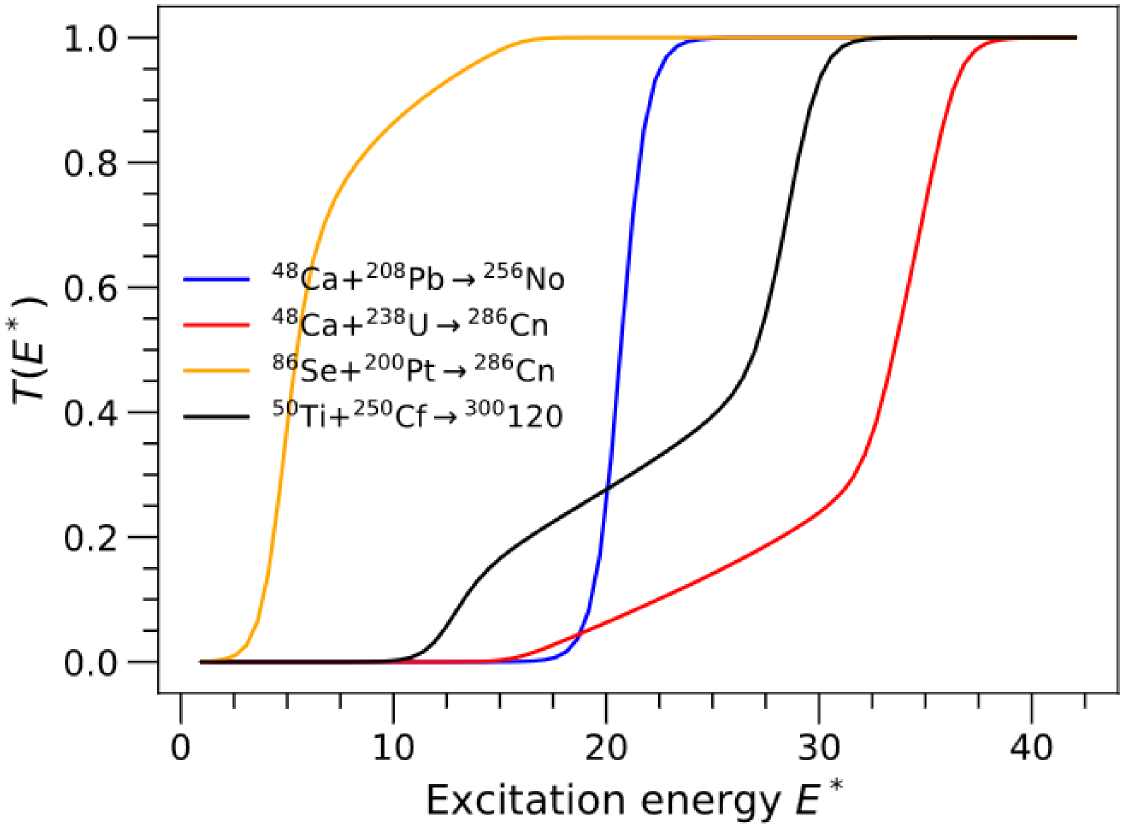
Method results

	First system			Second system			Third system		
Guess:	Even probability [s]			Reuse 1^{st} solution [s]			Linear interp.[s]		
Size (N)	IRAM			IRAM			IRAM		
	ItMat		LinSys	ItMat		LinSys	ItMat		LinSys
Energy spacing of the systems in the three columns below is $0.1\,\mathrm{MeV}$									
4.0×10^4	7.3	0.9	0.9	2	0.4	0.95	1.85	0.51	0.95
9.0×10^4	29	3.2	3.7	9.4	1.5	2.7	7.8	2.7	3.2
1.6×10^5	67	11.6	8	26	6.3	8	19.7	6.3	7.8
3.6×10^5	208	42	29	101	19	28	73	30	27
1.0×10^6	813	204	126	545	226	125	443	202	123
Energy spacing of the systems in the three columns below is $0.05\,\mathrm{MeV}$									
Size (N)	IRAM			IRAM			IRAM		
	ItMat		LinSys	ItMat		LinSys	ItMat		LinSys
3.6×10^5	232	54	25	65	17	26	31	13	26
1.0×10^6	940	230	126	370	88	123	181	89	124

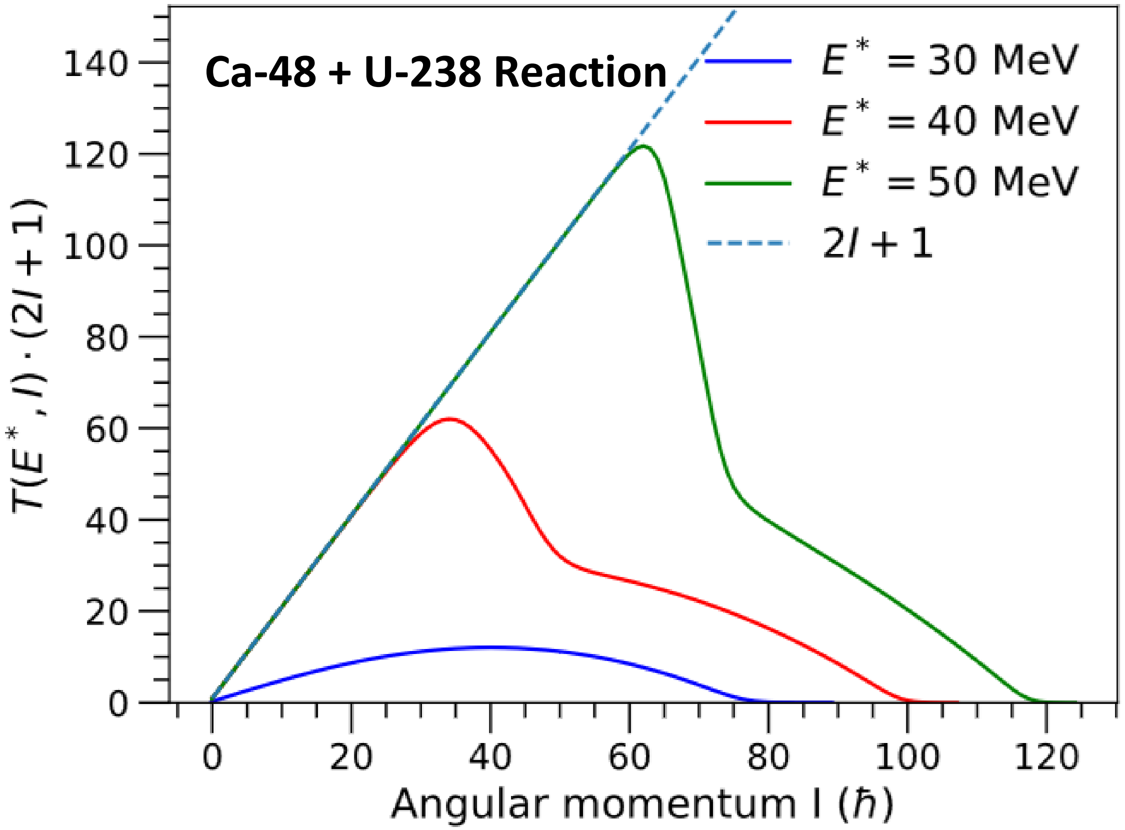
Method results



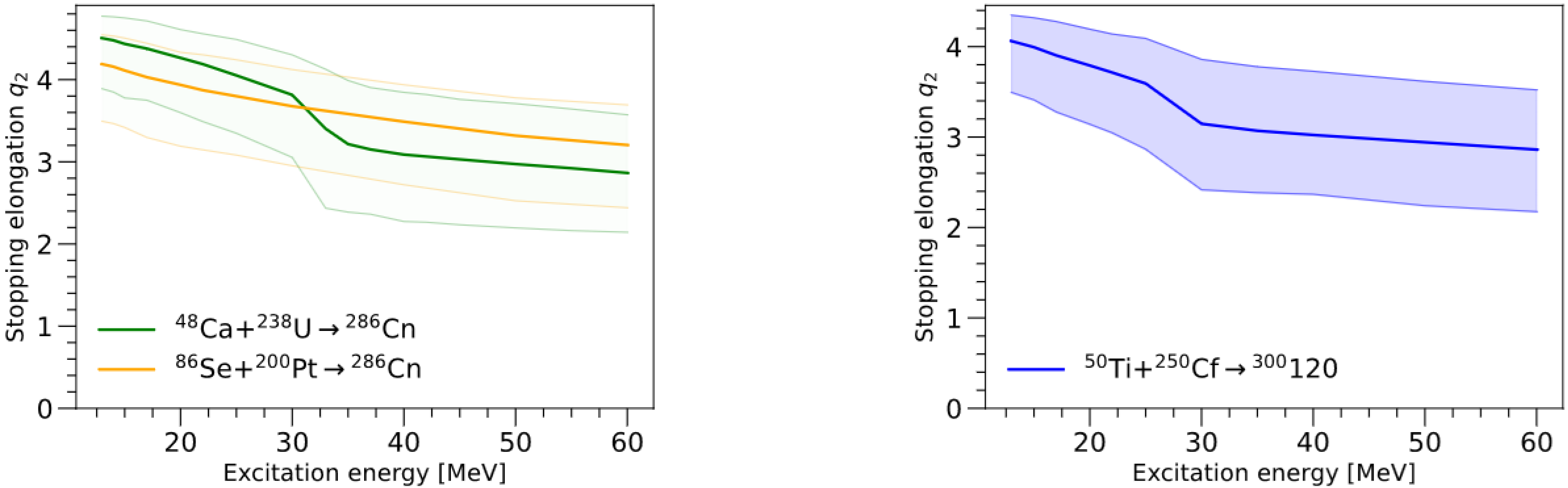
Third system		
Linear interp.[s]		
	IRAM	
	ItMat	LinSys
below is 0.1 MeV		
1.85	0.51	0.95
7.8	2.7	3.2
19.7	6.3	7.8
73	30	27
443	$\boxed{202}$	$\boxed{123}$
below is 0.05 MeV		
	IRAM	
	ItMat	LinSys
31	13	26
181	$\boxed{89}$	$\boxed{124}$

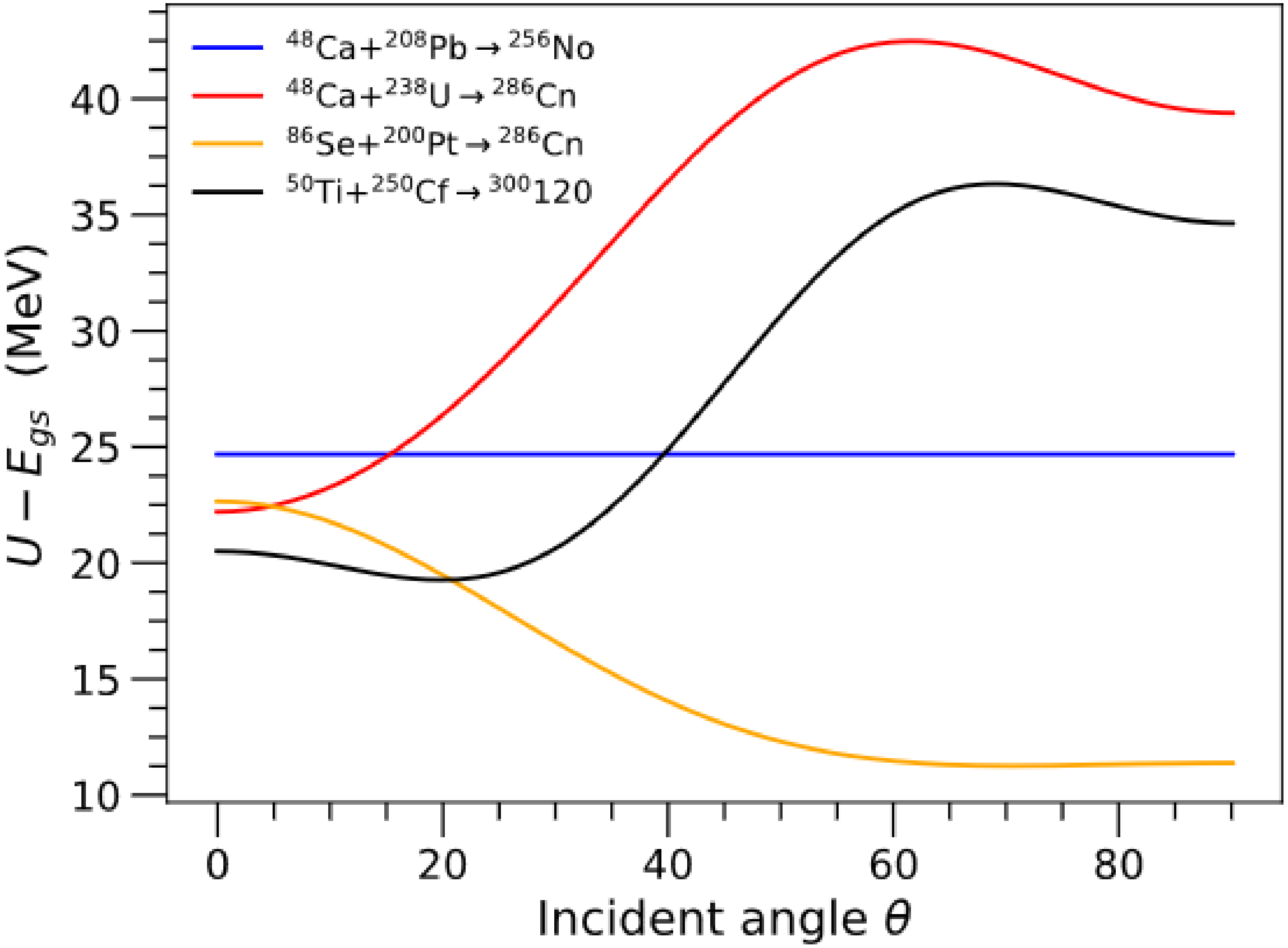


$T(E^*)$
Excitation energy E^*
$^{48}Ca+^{208}Pb \to ^{256}No$
$^{48}Ca+^{238}U \to ^{286}Cn$
$^{86}Se+^{200}Pt \to ^{286}Cn$
$^{50}Ti+^{250}Cf \to ^{300}120$

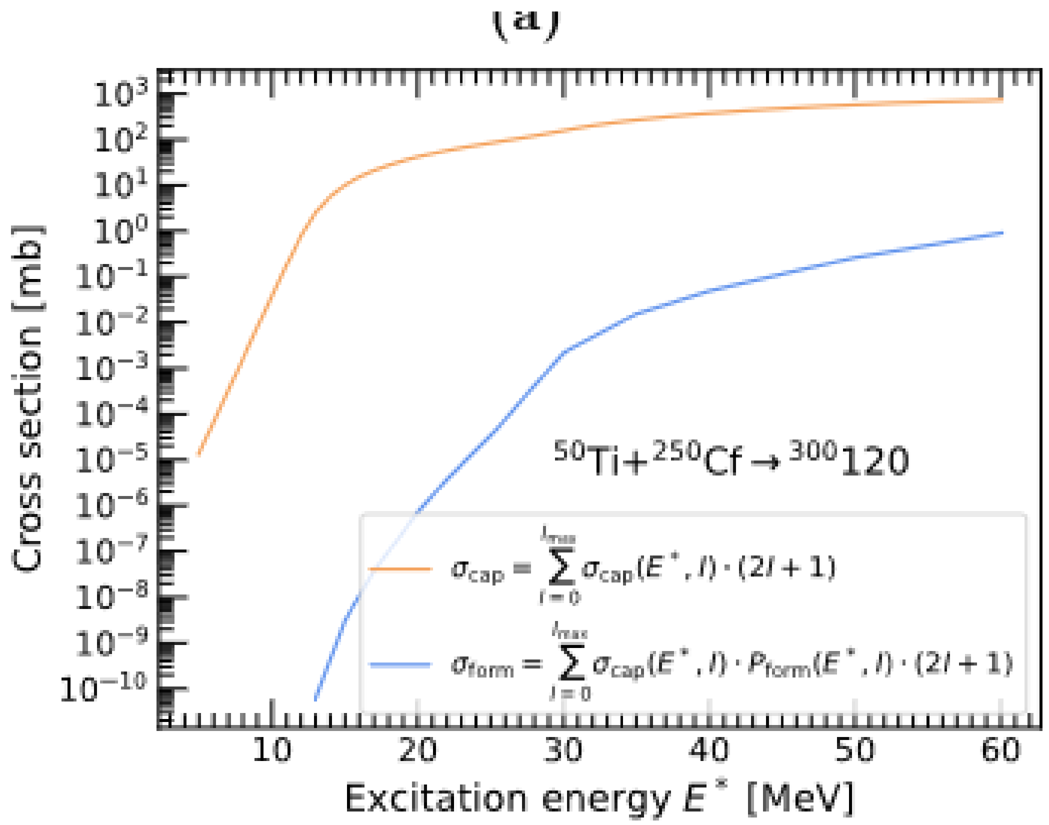


Ca-48 + U-238 Reaction
$T(E^*, I) \cdot (2I+1)$
Angular momentum I ($\hbar$)
$E^* = 30$ MeV
$E^* = 40$ MeV
$E^* = 50$ MeV
$2I+1$

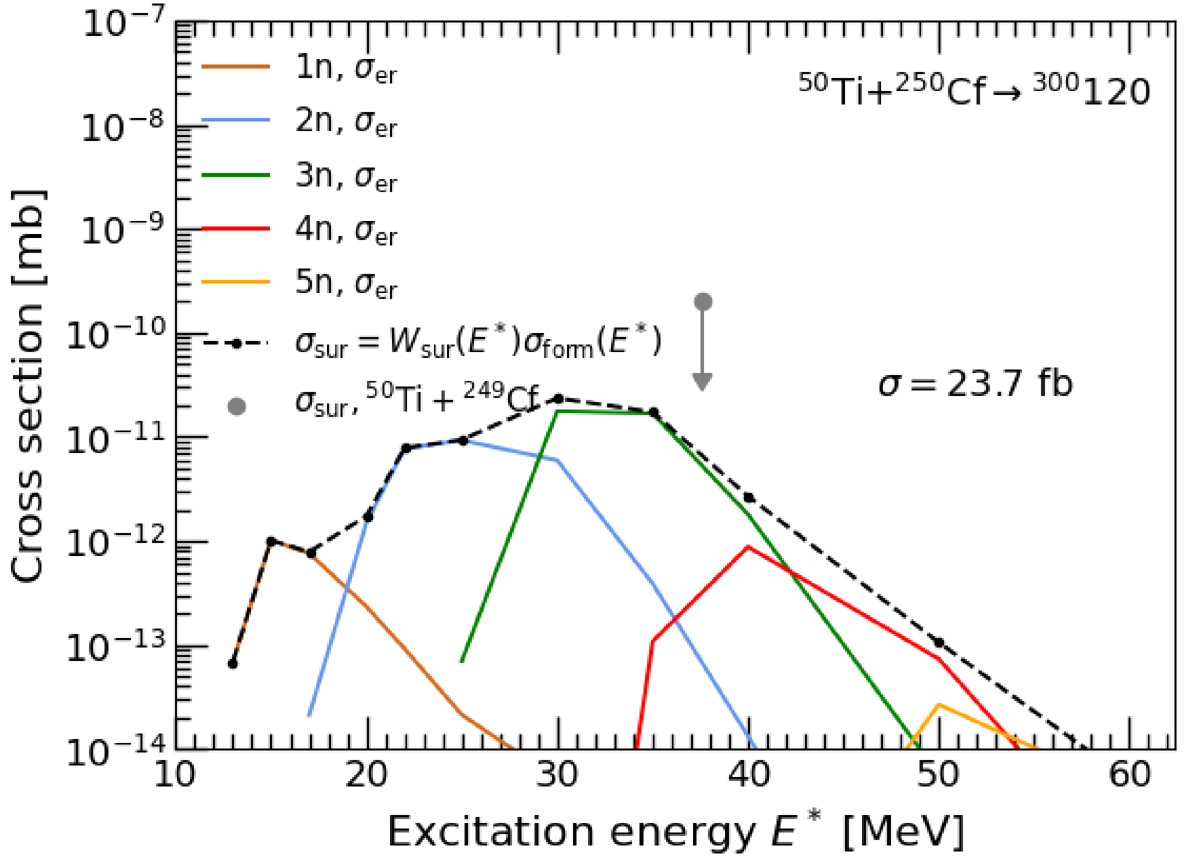

Stopping elongation q_2
Excitation energy [MeV]
^{48}Ca+^{238}U → ^{286}Cn
^{86}Se+^{200}Pt → ^{286}Cn
^{50}Ti+^{250}Cf → ^{300}120



^{48}Ca+^{208}Pb → ^{256}No
^{48}Ca+^{238}U → ^{286}Cn
^{86}Se+^{200}Pt → ^{286}Cn
^{50}Ti+^{250}Cf → ^{300}120
$U - E_{gs}$ (MeV)
Incident angle θ



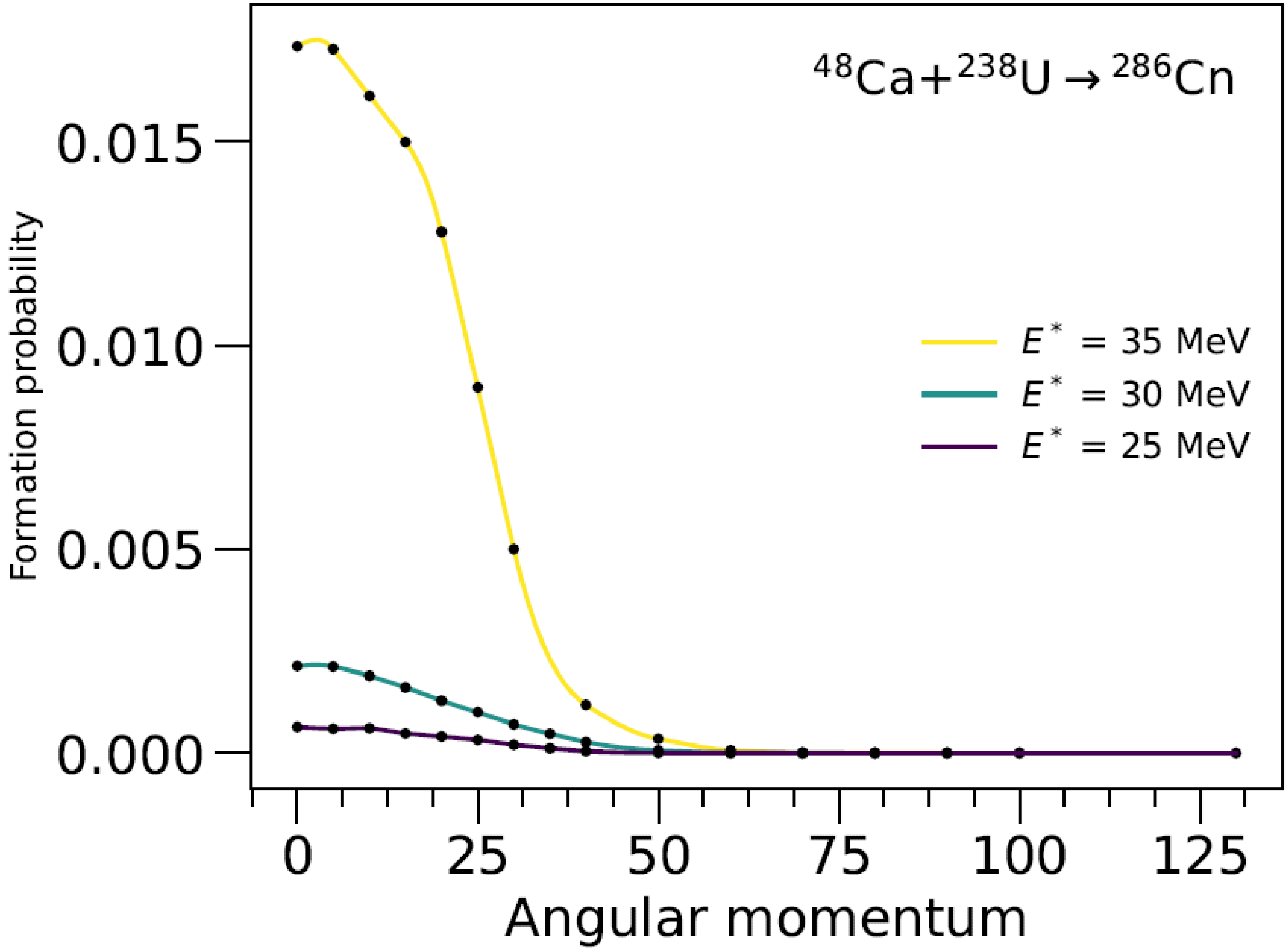
(a)
Cross section [mb]
Excitation energy E^* [MeV]
^{50}Ti+^{250}Cf$\rightarrow$^{300}120
$\sigma_{\rm cap}=\sum_{l=0}^{l_{\max}}\sigma_{\rm cap}(E^*,l)\cdot(2l+1)$
$\sigma_{\rm form}=\sum_{l=0}^{l_{\max}}\sigma_{\rm cap}(E^*,l)\cdot P_{\rm form}(E^*,l)\cdot(2l+1)$



Cross section [mb]
Excitation energy E^* [MeV]
^{50}Ti+^{250}Cf$\rightarrow$^{300}120
1n, $\sigma_{\rm er}$
2n, $\sigma_{\rm er}$
3n, $\sigma_{\rm er}$
4n, $\sigma_{\rm er}$
5n, $\sigma_{\rm er}$
$\sigma_{\rm sur}=W_{\rm sur}(E^*)\sigma_{\rm form}(E^*)$
$\sigma_{\rm sur}$, ^{50}Ti + ^{249}Cf
$\sigma = 23.7$ fb

$$\sigma_{\mathrm{fus}}(E^*) = \sum_{I=0}^{\infty}(2I+1)\sigma_{\mathrm{cap}}(E^*,I)P_{\mathrm{form}}(E^*,I)W_{\mathrm{sur}}(E^*,I)$$

Spline



Results

Survival probability

From $^{287}\mathrm{Fl}$ (Z=114)

Not $^{286}\mathrm{Cn}$ (Z=112)

Cn officially named 2010

Fl officially named 2012

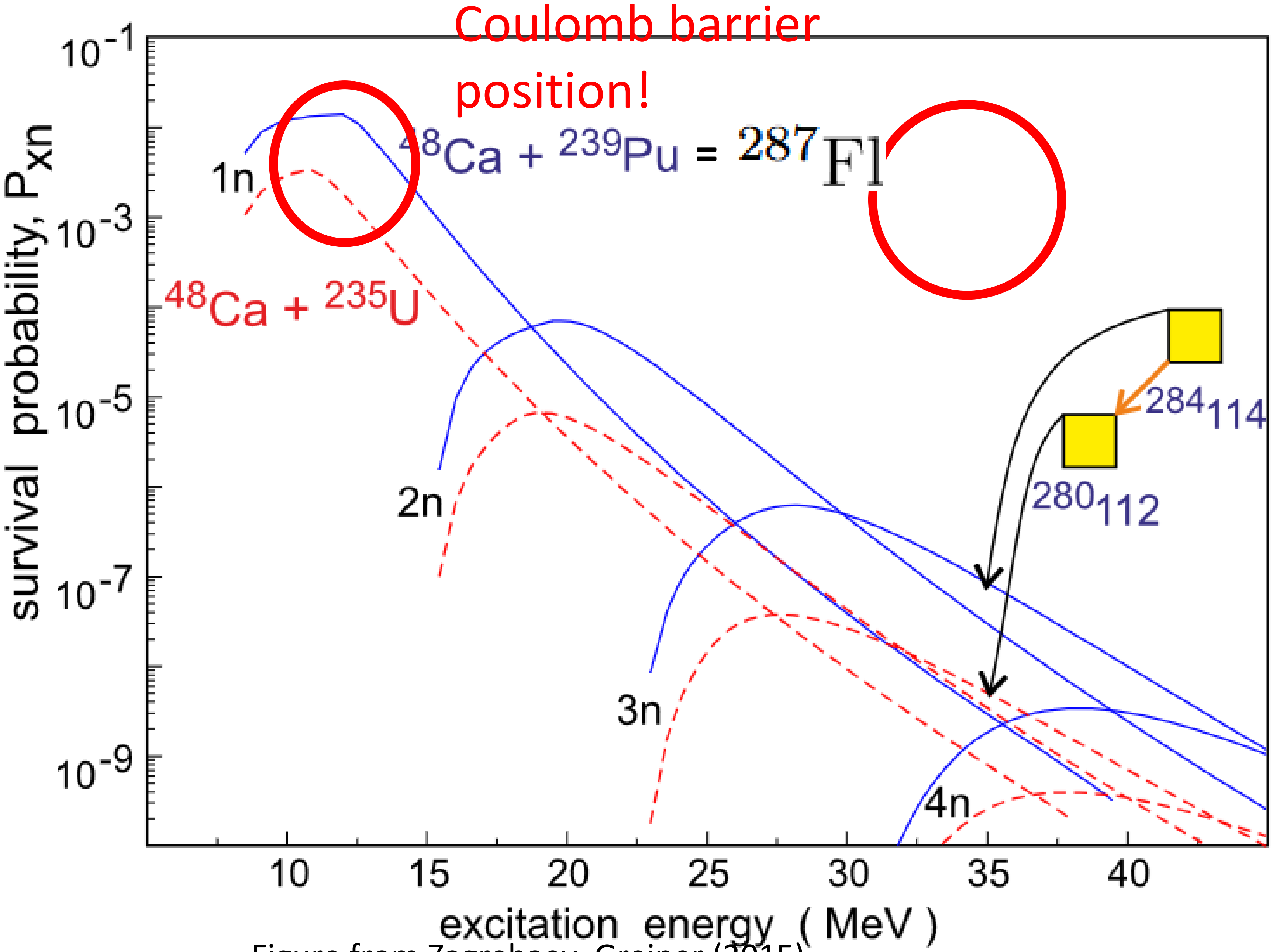


Figure from Zagrebaev, Greiner (2015)