Calculation of fission fragment mass distributions by using a semi-empirical method

Jongwha Lee[1,2], C. -S. Gil[1], Y.-O. Lee[1], T.-S. Park[2], and S. W. Hong[2,3]

[1] KAERI
[2] Sungkyunkwan University
[3] IBS

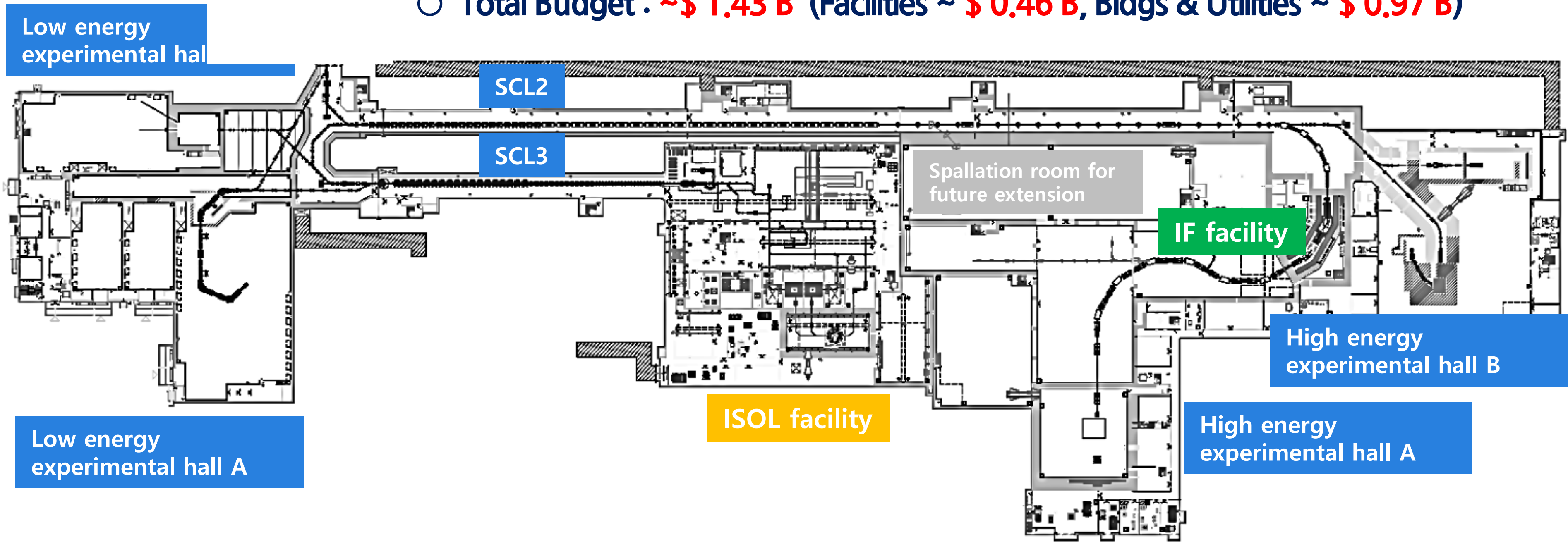
Korea Atomic Energy Research Institute
成均館大學校 SUNGKYUNKWAN UNIVERSITY
기초과학연구원 Institute for Basic Science

RAON: Rare Isotope accelerator complex for ON-line experiment

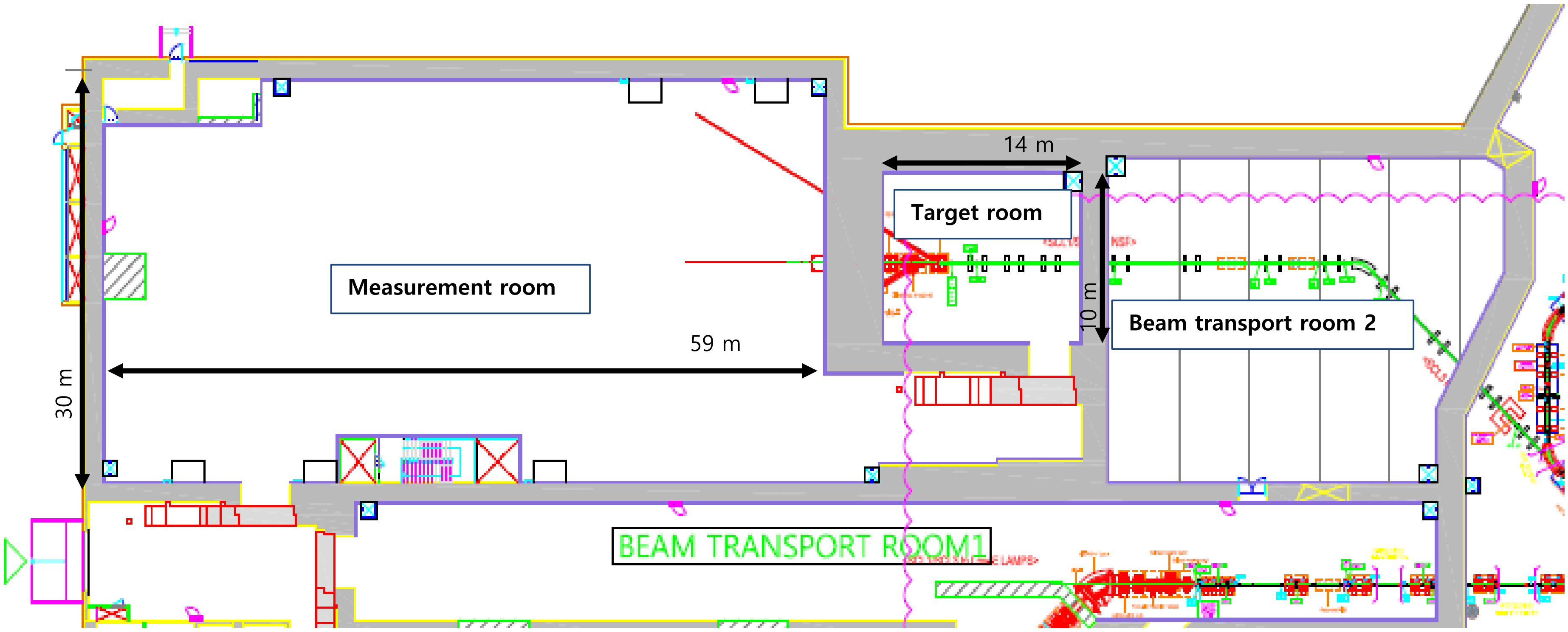
- ❖ **high intensity RI beams by ISOL and IF**
  - ISOL: direct fission of ^{238}U by 70 MeV proton
  - IF: 200 MeV/u ^{238}U (intensity: 8.3 pμA)
- ❖ **high quality neutron-rich beams**
  - e.g., ^{132}Sn with up to 250 MeV/u, up to 10^9 particles per second
- ❖ **More exotic RI beam production by combination of ISOL and IF (ISOLIF)**

○ **Project period : 2011.12 - 2021.12**

○ **Total Budget : ~$ 1.43 B (Facilities ~ $ 0.46 B, Bldgs & Utilities ~ $ 0.97 B)**



NDPS: Nuclear Data Production System

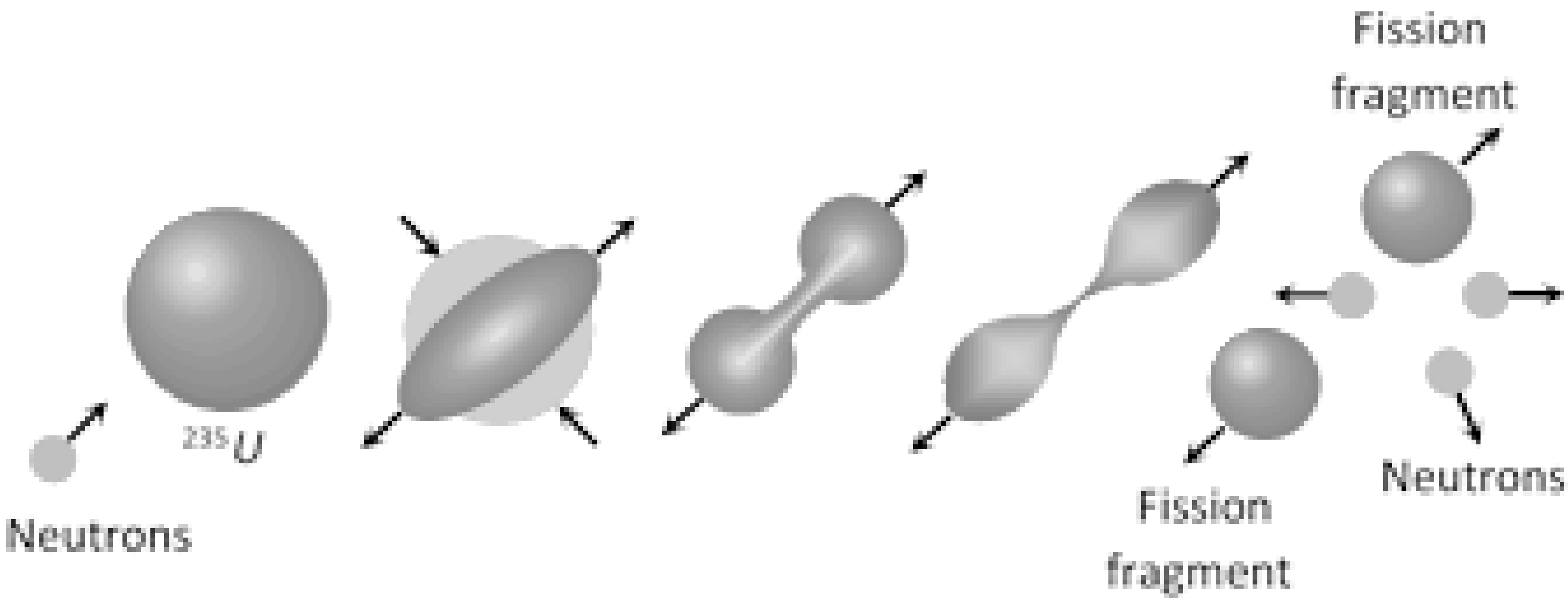


Nuclear fission is one of the candidates for early experiments at NDPS.

Nuclear fission

- 1938: Discovery of fission by Otto Hahn and Fritz Strassmann : fission of ^{235}U
  (O. Hahn & F. Strassmann, Naturwissenschaften 27, 1115 (1939))

- 1939: Explained and **coined** by Lise Meitner and Otto Frisch : Liquid Drop Model
  (L. Meitner & O. R. Frisch, Nature 143, 239 (1939))



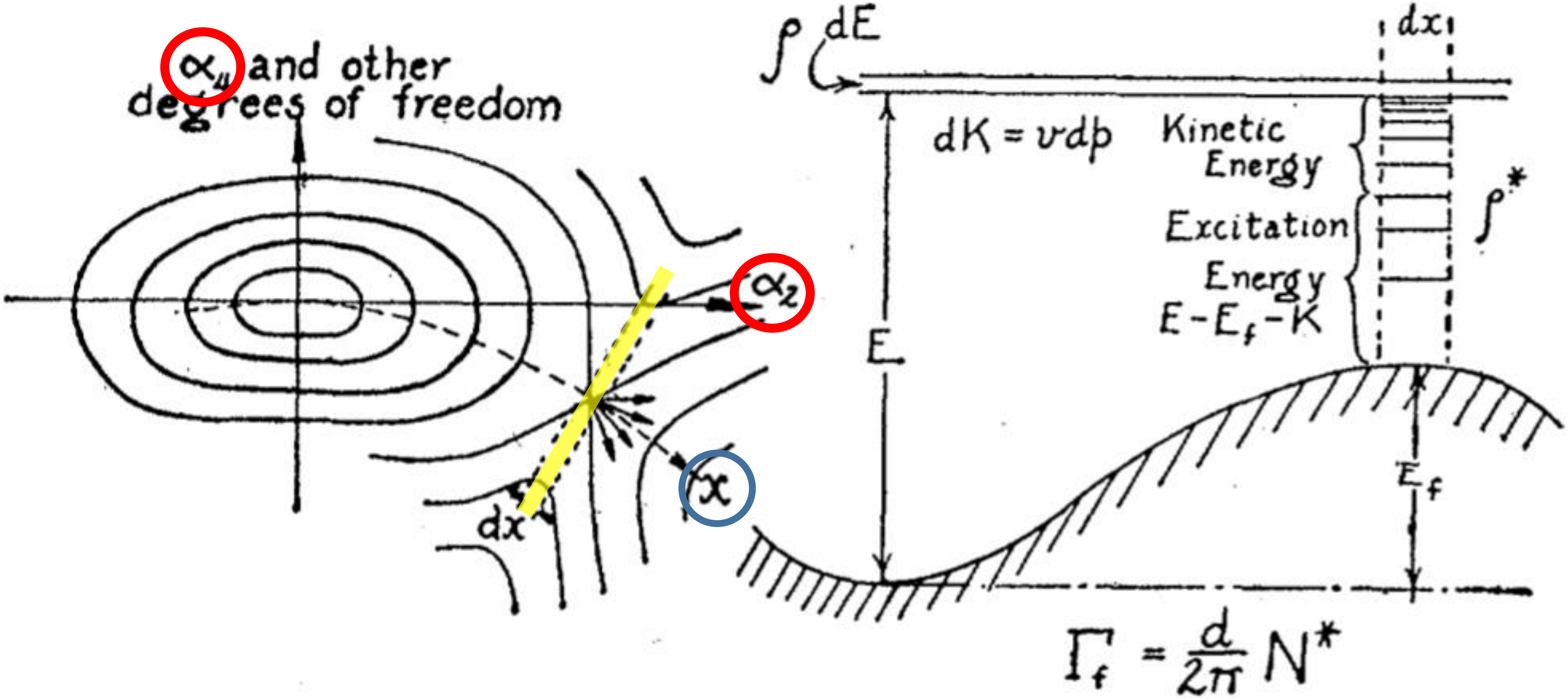
https://www.studyadda.com/notes/jee-main-advanced/physics/nuclear-physics/nuclear-fission/8656

Nuclear fission

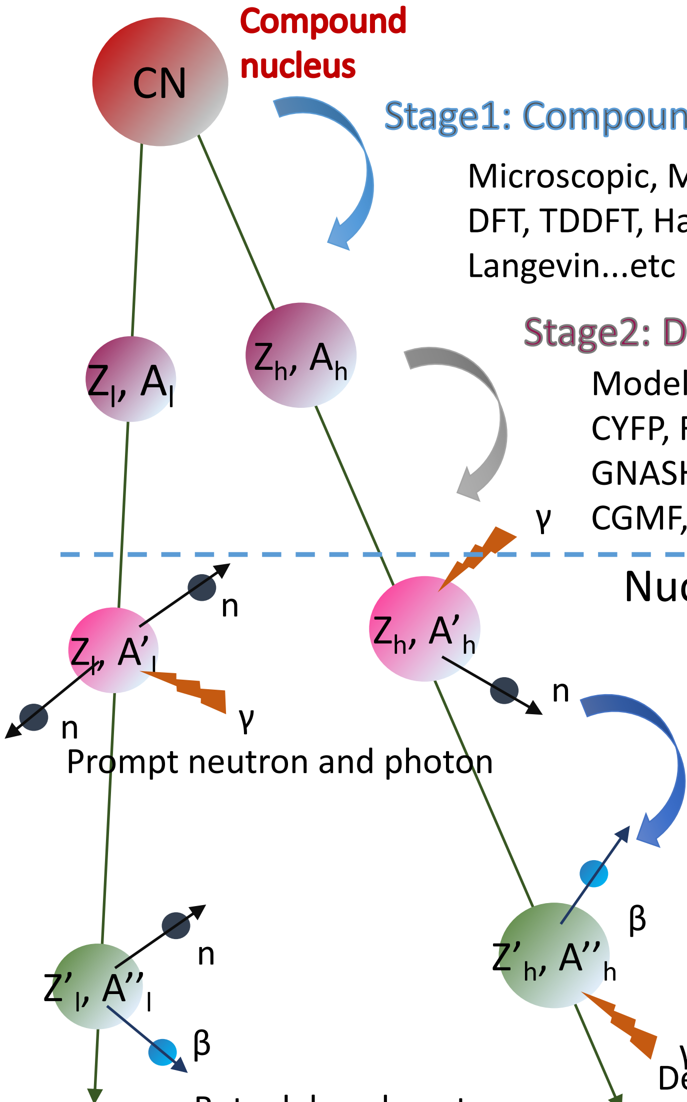
- 1939: N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1939) (Liquid drop model)

  fission barrier, saddle point, competition btw fission and neutron escape,

  fission probability determined by microcanonical ensemble, etc



Nuclear fission and decay processes



Stage1: Compound nucleus formation to scission

Microscopic, Macroscopic approaches:
DFT, TDDFT, Hartree-Fock-Bogoliubov, AMD, Langevin...etc

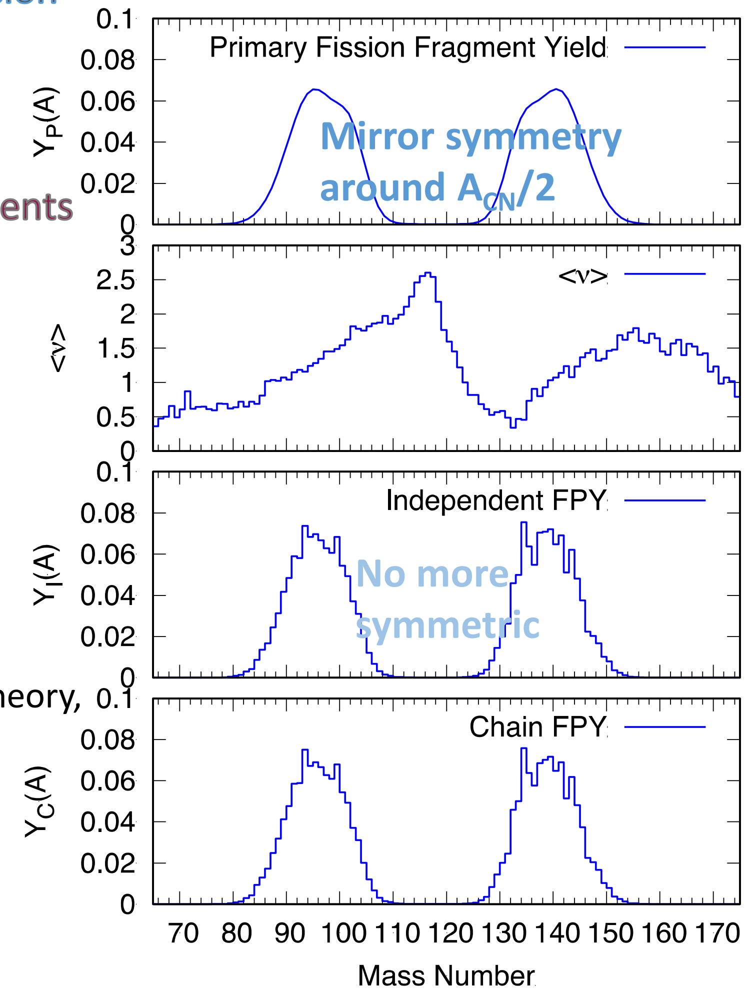
Stage2: De-excitation of fission fragments

Models and codes:
CYFP, FREYA, GEF, FIFRELYN, GNASH, EMPIRE, TALYS, CCONE, CGMF, CoH/BeoH, ..etc

Nuclear data library

Stage3: Beta decay

Bateman equation, Gross theory,
Summation calculation
Decay Data



Slide of Satoshi Chiba (Tokyo Tech)

Fission models

- Models of fission
  - Microscopic fission models (TD-GCM, etc)

    J.F. Berger et al., Nucl Phys A 428 (1984) 23c
  - Stochastic approaches (Langevin equation)

    Y. Abe, et al., J. Physique 47 (1986) C4-329, Usang et al, Scientific Reports, 9, 1525 (2019)
  - Empirical models

    A.R.de L. Mussgrove et al., IAEA-169, Vol. 2 (1974) pp. 163-200
  - Semi-empirical models (GEF)

    K.-H. Schmidt et al., Nucl. Data Sheets 131 (2016) 107

- Advantage of semi-empirical model
  - Low computing cost
  - Good accuracy
  - Predictive power

GEF (General Fission) model

Karl-Heinz Schmidt, CNRS/IN2P3,
Beatriz Jurado, CNRS/IN2P3,
Charlotte Amouroux CEA, DSM-Saclay

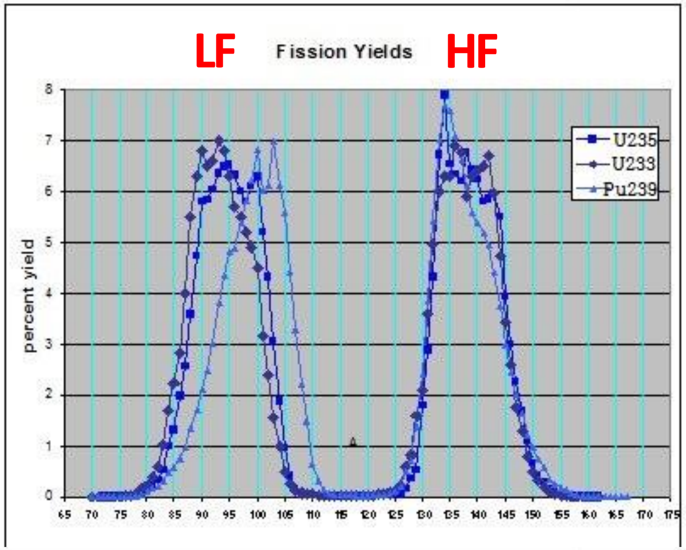
The GEF model explains the complex appearance of fission observables by universal principles of theoretical models and considerations on the basis of fundamental laws of physics and mathematics.

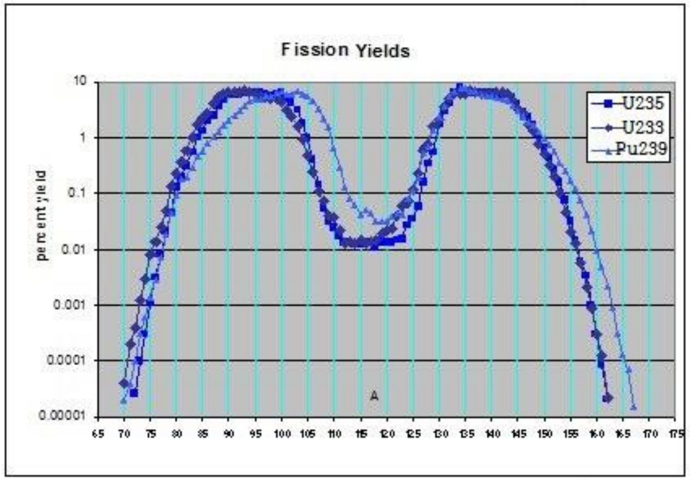
The GEF code calculates fission-fragment yields and associated quantities (e.g. prompt neutron and gamma) for a large range of nuclei and excitation energy.

Fission observables can be calculated with a precision that comply with the needs for applications in nuclear technology.

Some general features of fission product yields (FPY)

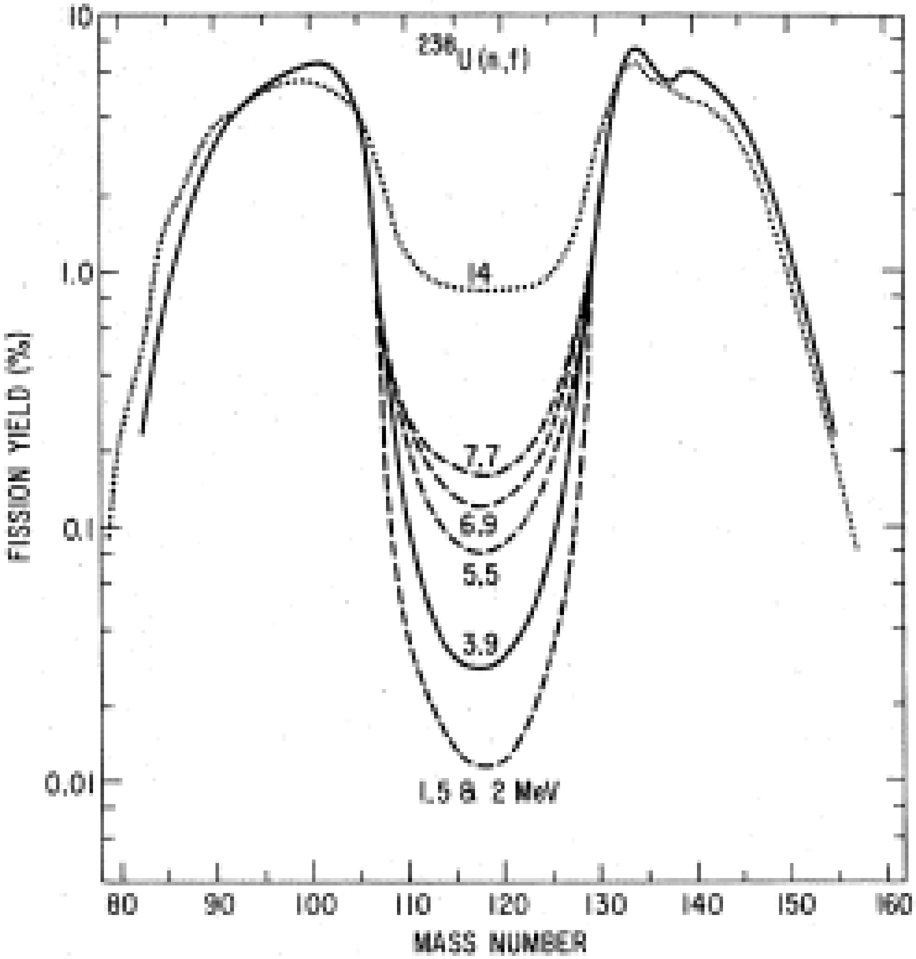
Thermal neutron induced fission of ^{235}U, ^{233}U and ^{239}Pu





www.unclear2nuclear.com

E^* dependence of fission fragment mass distribution (^{238}U)



S.Nagy et al., Phys. Rev. C 17 (1978) 163

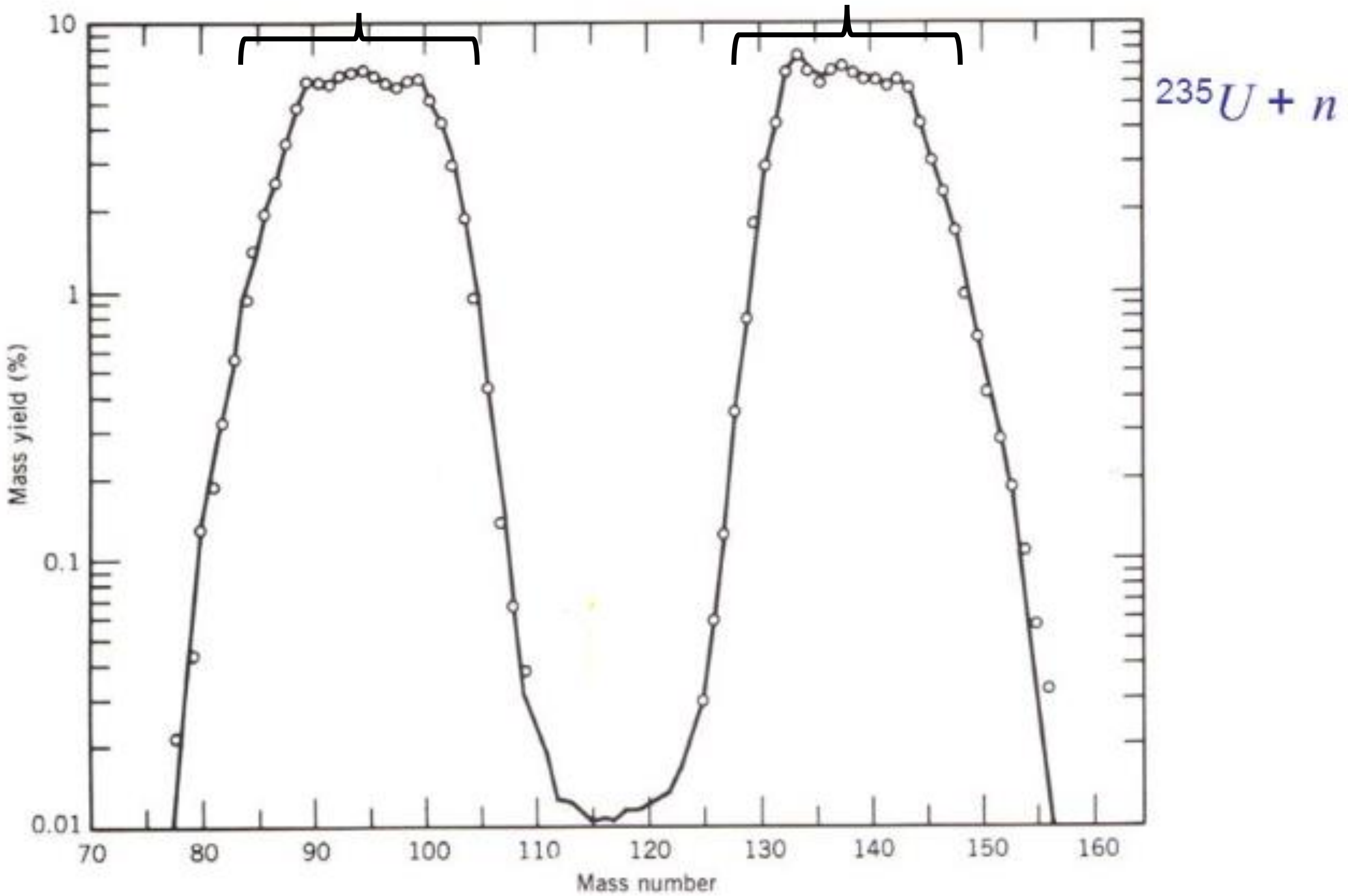
Asymmetric fission fragments mass distribution

LF : Light Fragments

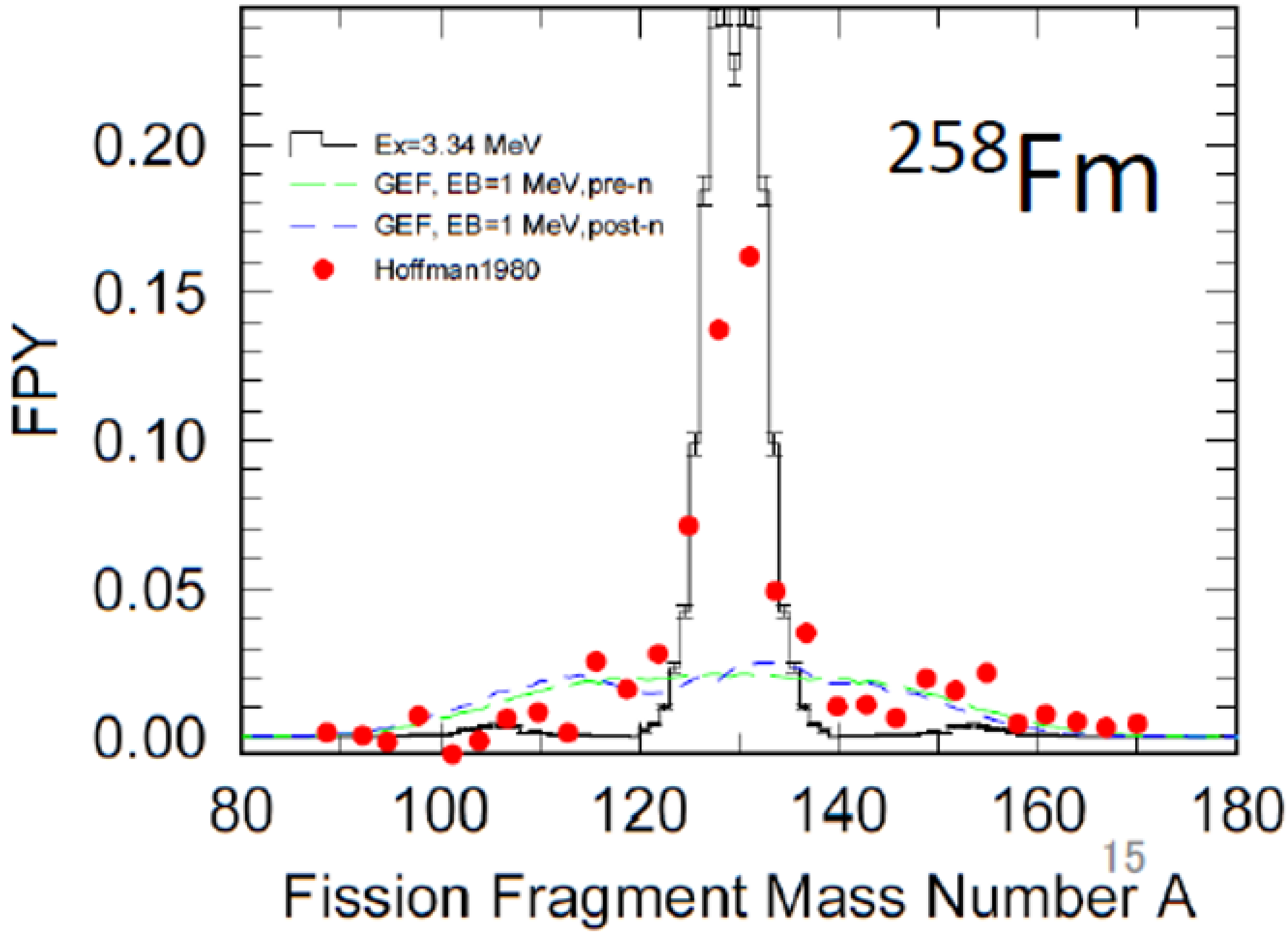
Kr, Rb, Sr, Y, Zr, Nb, Mo, Tc, …

HF : Heavy Fragments

I, Xe, Cs, Ba, La, Ce, Pr, …



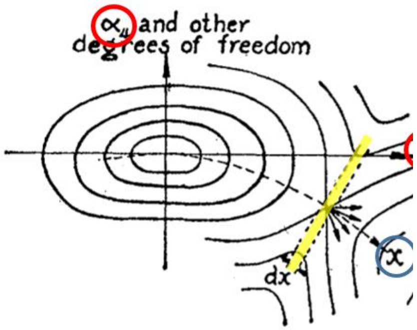
If a compound nucleus splits into two equal mass fragments, **Symmetric Fission**

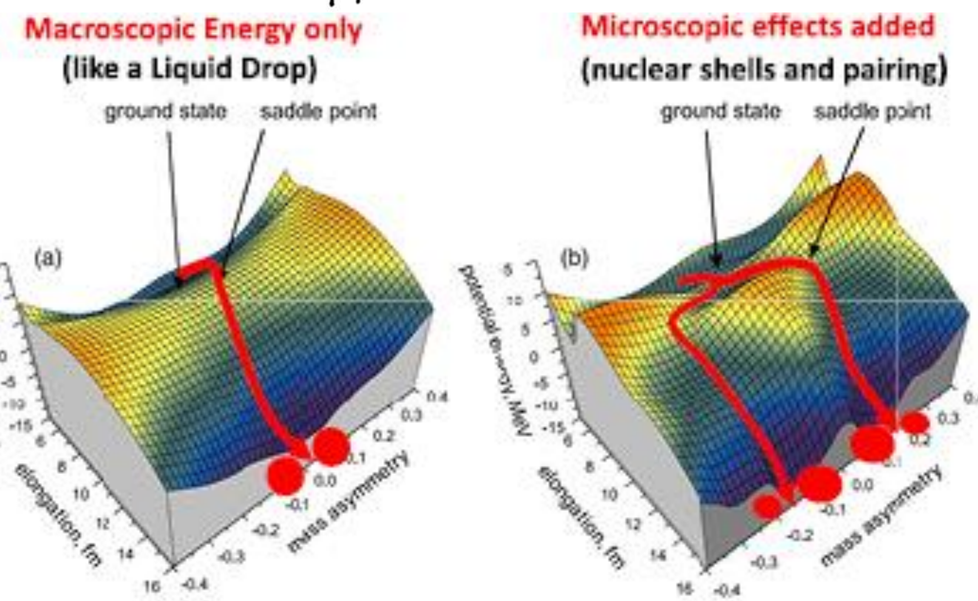


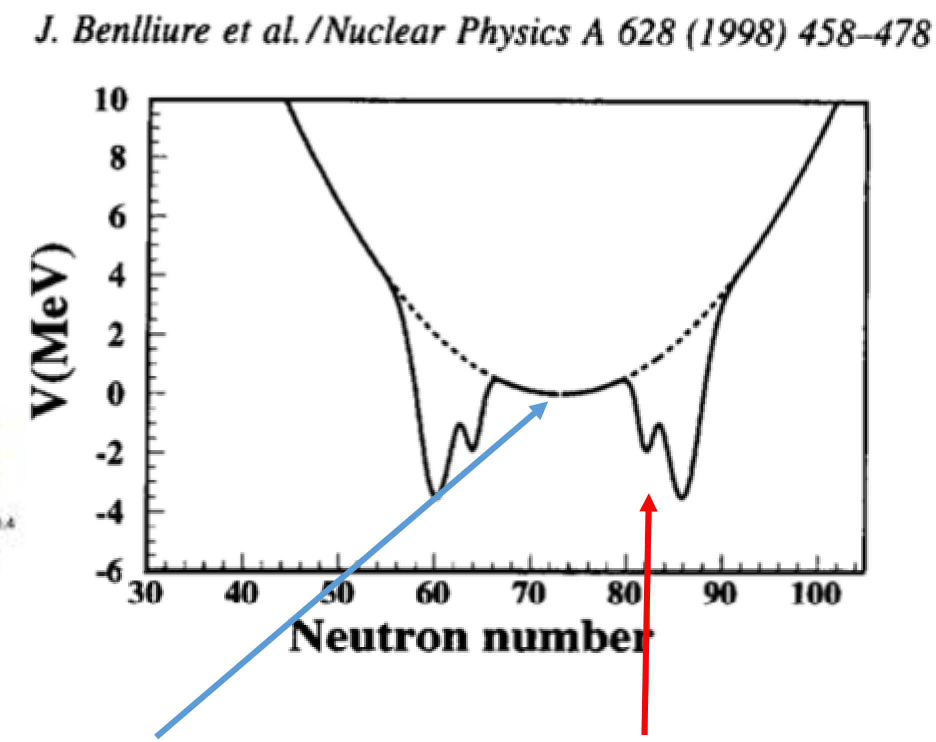
Macroscopic + microscopic potential energy

- 1939: N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1939) : **Liquid drop model**
- 1967: V. M. Strutinsky, Nucl. Phys. A95, 420 (1967), Nucl. Phys. A122, 1 (1968)

  Calculate **shell-correction energies** by using deformed single-particle energies



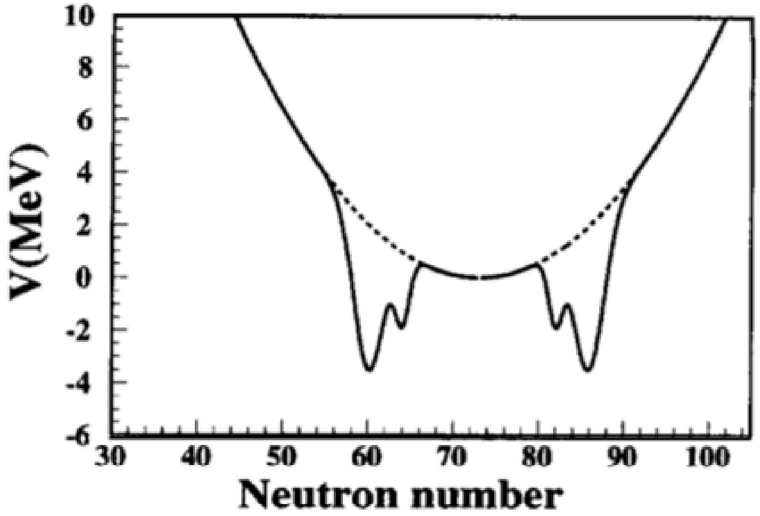




- Fission barrier energy: Macroscopic liquid drop energy + microscopic shell energy

Fission barrier energy: Macroscopic liquid drop energy + microscopic shell energy

$$V(N) = V_{mac}(N) + V_{sh,1}(N) + V_{sh,1}(N_{CN} - N) + V_{sh,2}(N) + V_{sh,2}(N_{CN} - N)$$



Fission fragments Yields

$$Y(A) \sim \exp(2\sqrt{a \cdot (E^* - U(A))})$$

H. A. Bethe, Phys Rev, 50, p. 332 (1936)

$$Y_{mac}(E_0^*, N) \approx \exp\left(2\sqrt{\tilde{a}\, E_{mac}(E_0^*, N)}\right),$$

$$Y_{sh,i}(E_0^*, N) \approx \exp\left(2\sqrt{\tilde{a}\, E_{sh,i}(E_0^*, N)}\right)$$

$$Y_{mac}(E_0^*, N) \approx \exp\left(\frac{-(N_{CN}/2 - N)^2}{\sigma_{mac}^2}\right)$$

$$Y_{sh,i}(E_0^*, N) \approx \exp\left(\frac{-(N_{sh,i} - N)^2}{\sigma_{sh,i}^2}\right)$$

: Five Gaussian method

J. Benlliurea et al, Nuclear Physics A 628 (1998) 458-478

Our semi-empirical model

- Take advantages of theoretical model and empirical model

- Strategy
  - Simplify the fission process
  - Replace the complicated calculation into empirical model
    - Shape of the fission barrier
    - Lump fission dynamics from saddle to scission as model parameters
    - Parameter fitting

Our model

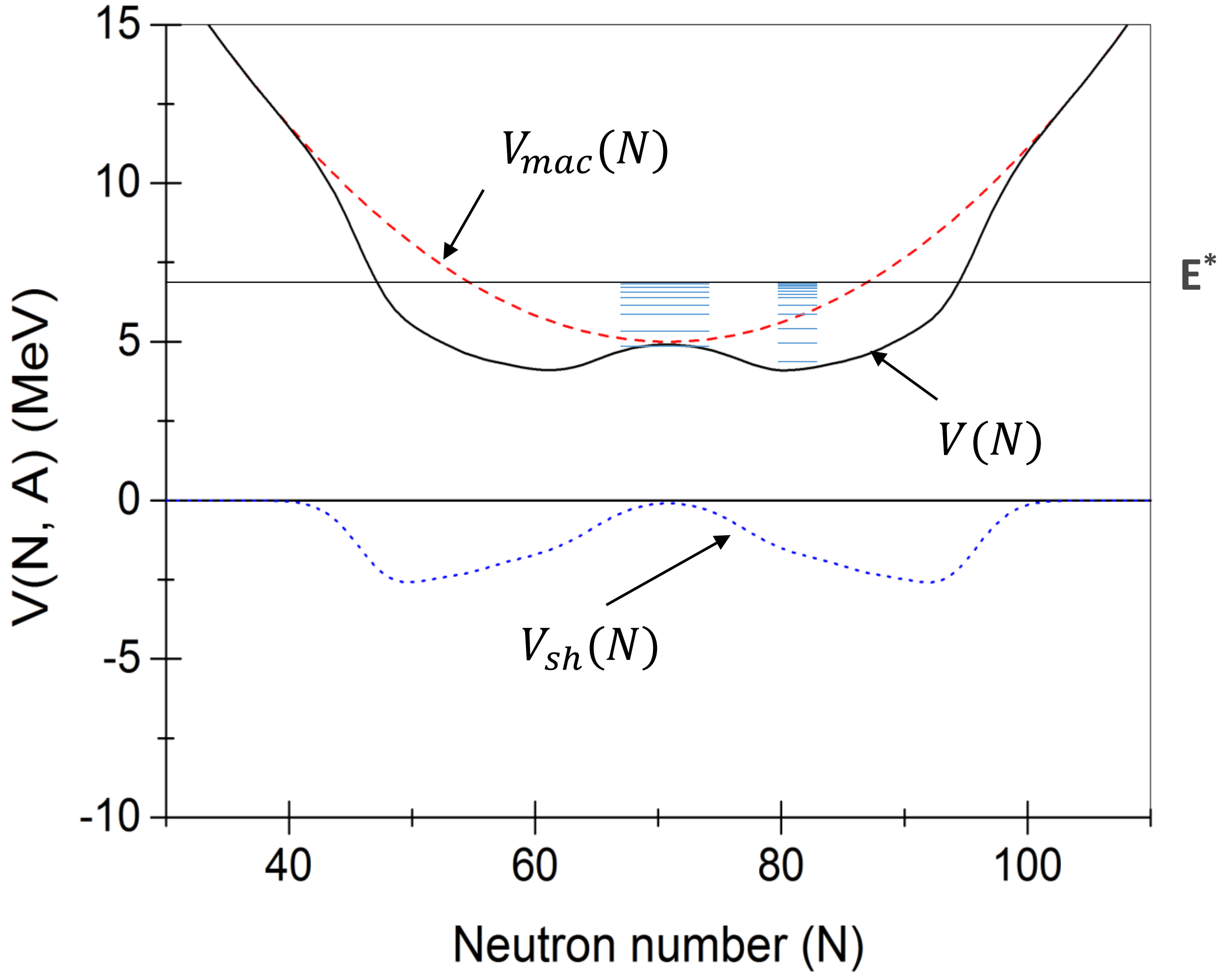
- Compound nuclei are considered as micro-canonical ensemble

- Assume the mass yield distribution is determined by the level density at fission barrier
  - From the functional form of level density of Fermi-gas model
    $\rho(E^*) \propto \exp\left(2\sqrt{\tilde{a}E_{int}}\right)$, fission yield can be expressed as
    $$Y(N,A;E) \approx \exp\left(2\sqrt{\tilde{a}\left(E^* - V(N,A)\right)}\right)$$
  - Internal excitation energy at fission barrier $E_{int} = E^* - V(N,A)$

- Shell structure of fission products play important roles in determining mass distribution
  - $N_{in} = 82$, $N_{out} \sim 90$
  - Express V as a function of N (neutron number)
  - Unchanged charge distribution

$$\begin{aligned} V(N) = {} & V_{\rm mac}(N) \\ & + V_{\rm sh,1}(N) + V_{\rm sh,1}(N_{\rm CN} - N) \\ & + V_{\rm sh,2}(N) + V_{\rm sh,2}(N_{\rm CN} - N) \end{aligned}$$

Fission barrier



Fission barrier for n + ^{235}U at the thermal energy of neutron

Fission barrier

- Fission barrier : $V(N,A) = V_{mac}(N) + V_{sh}(N)\exp(-\gamma\epsilon(N))$
- Macro potential

$$V_{mac}(N) = C_{mac}\left(N - \frac{\tilde{N}_{CN}}{2}\right)^2 + V_0$$

- Shell correction

$$V_{sh}(N) = C_{in}\left[\exp\left(\frac{(N-N_{in})^2}{\sigma_{in}^2}\right) + \exp\left(\frac{\left(N-\overline{N}_{in}\right)^2}{\sigma_{in}^2}\right)\right] + C_{out}\left[\exp\left(\frac{(N-N_{out})^2}{\sigma_{out}^2}\right) + \exp\left(\frac{\left(N-\overline{N}_{out}\right)^2}{\sigma_{in}^2}\right)\right]$$

Damping term

$$\epsilon(N) = E^* - [V_{mac}(N) + V_{sh}(N)]$$

where

$$\tilde{N}_{CN} \equiv N_{CN} - \nu \text{ and } \overline{N}_j \equiv \tilde{N}_{CN} - N_j \; (j = in, out)$$

Parameters

- Ten parameters in total
  $C_{mac}, C_{in}, C_{out}, N_{in}, N_{out}, \sigma_{in}, \sigma_{out}, V_0, \gamma$ and $\breve{a}$

- The curvature of macro potential

$$C_{mac} = \left(\frac{8}{N_{CN}^2}\right) 10^{7.16993-0.26602\left(\frac{Z_{CN}^2}{A_{CN}}\right)+0.00283\left(\frac{Z_{CN}^2}{A_{CN}}\right)^2}$$

  from J. Benlliure et al., Nucl. Phys. A 628 (1998) 458

- Effect of spherical neutron shell: $N_{in} = 82$

- Damping factor for shell effects: $\gamma = 0.06$
  from T. von Egidy and D. Bucurescu, Phys Rev. C 72 (2005) 044311

- Constant term of macro potential: $V_0 = 5$ MeV
  Determined by considering the fission barrier height of U isotopes
  from W.D. Myers and W.J. Swiatecki, Nucl. Phys. 81 (1966) 1

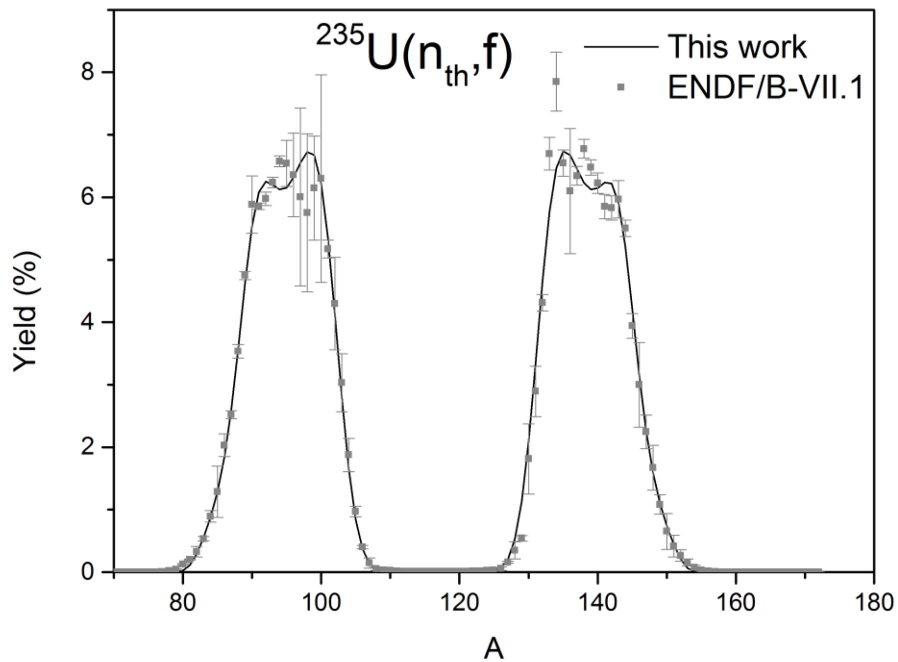
6 adjustable parameters

$C_{in}, C_{out}, N_{out}, \sigma_{in}, \sigma_{out}$ and $\tilde{a}$

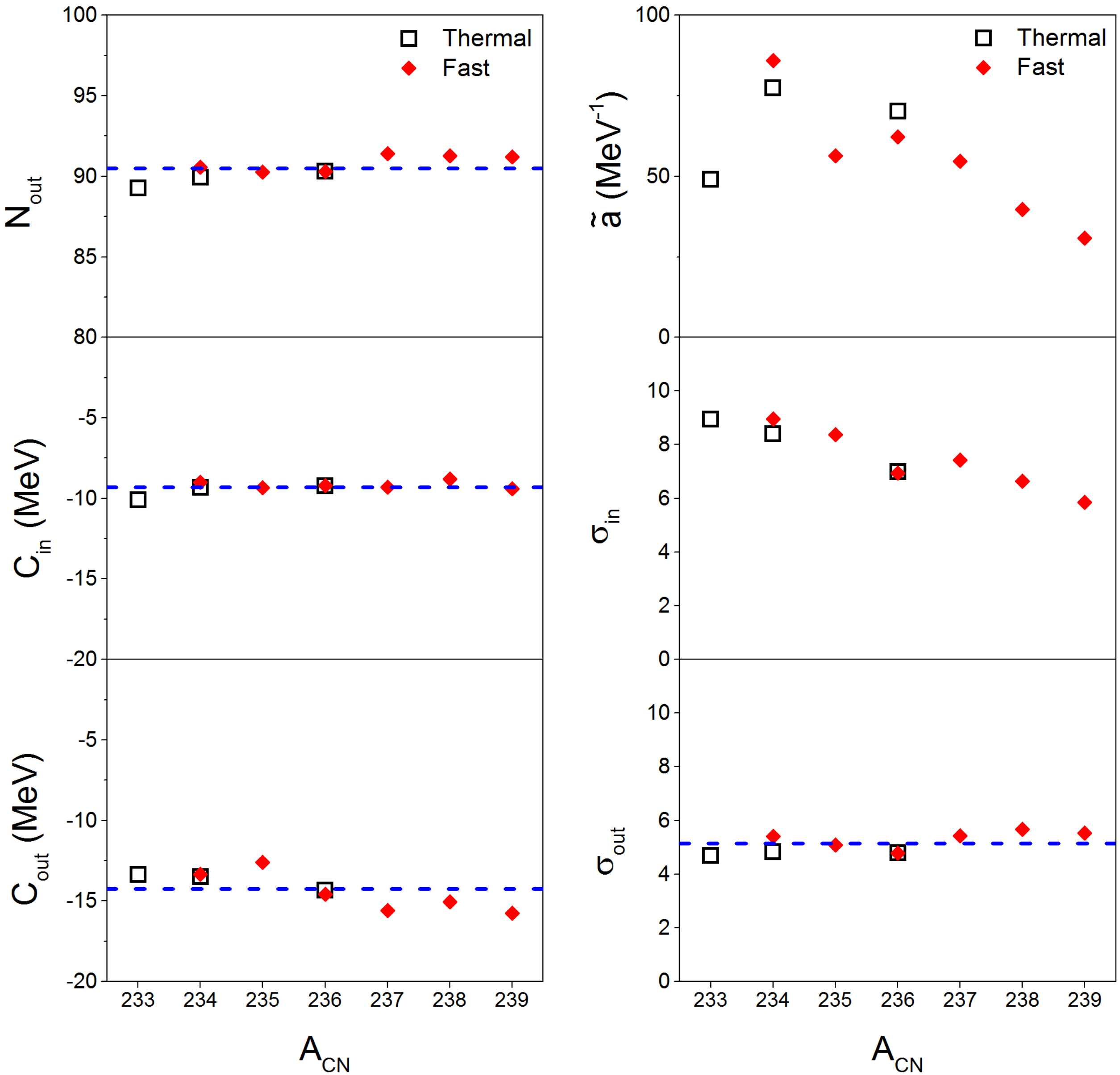
- Least-square fit by minimizing $< Y^2 >$
  - $< Y^2 > \equiv \frac{1}{n}\sum_{k=1}^{n}(Y_k - \bar{Y}_k)^2$
  - Uncertainties are ignored

  cf) $\chi^2 \equiv \frac{1}{n}\sum_{k=1}^{n}\left(\frac{Y_k - \bar{Y}_k}{\Delta_k}\right)^2$

- Nine cases of ENDF data are considered for parameter fitting
  - Thermal neutron ($E_n = 0.0253$ eV) induced fission of ^{232}U, ^{233}U and ^{235}U
  - Fast neutron ($E_n = 500$ keV) induced fission of ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{237}U and ^{238}U

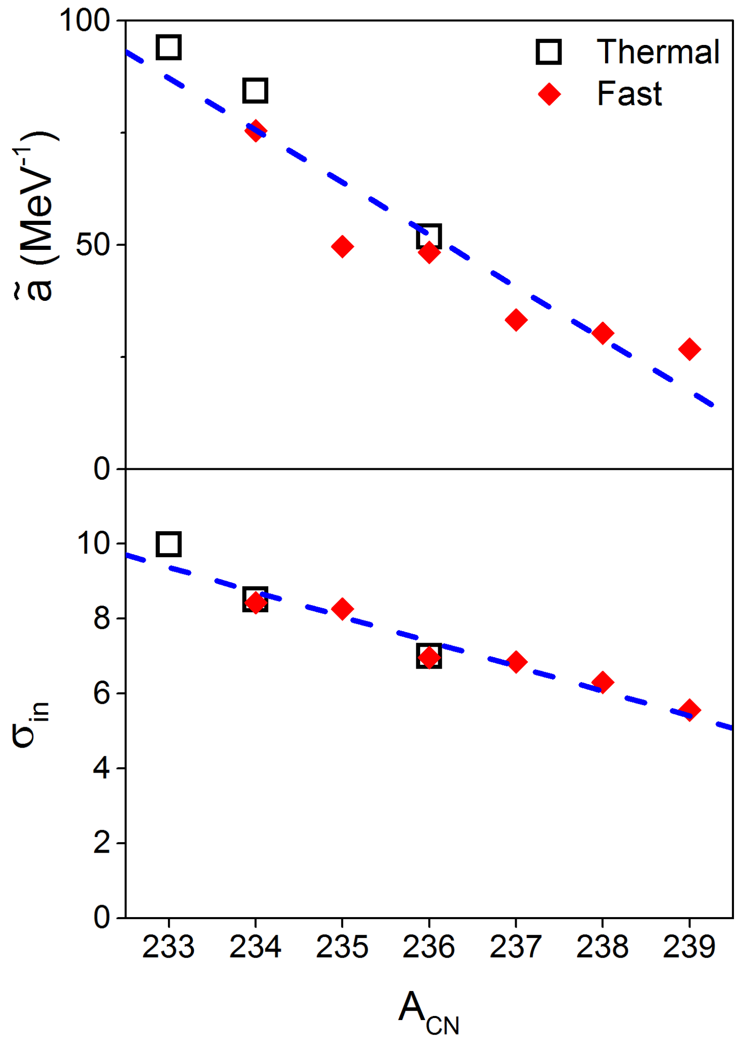


Resulting parameters (uranium isotopes)



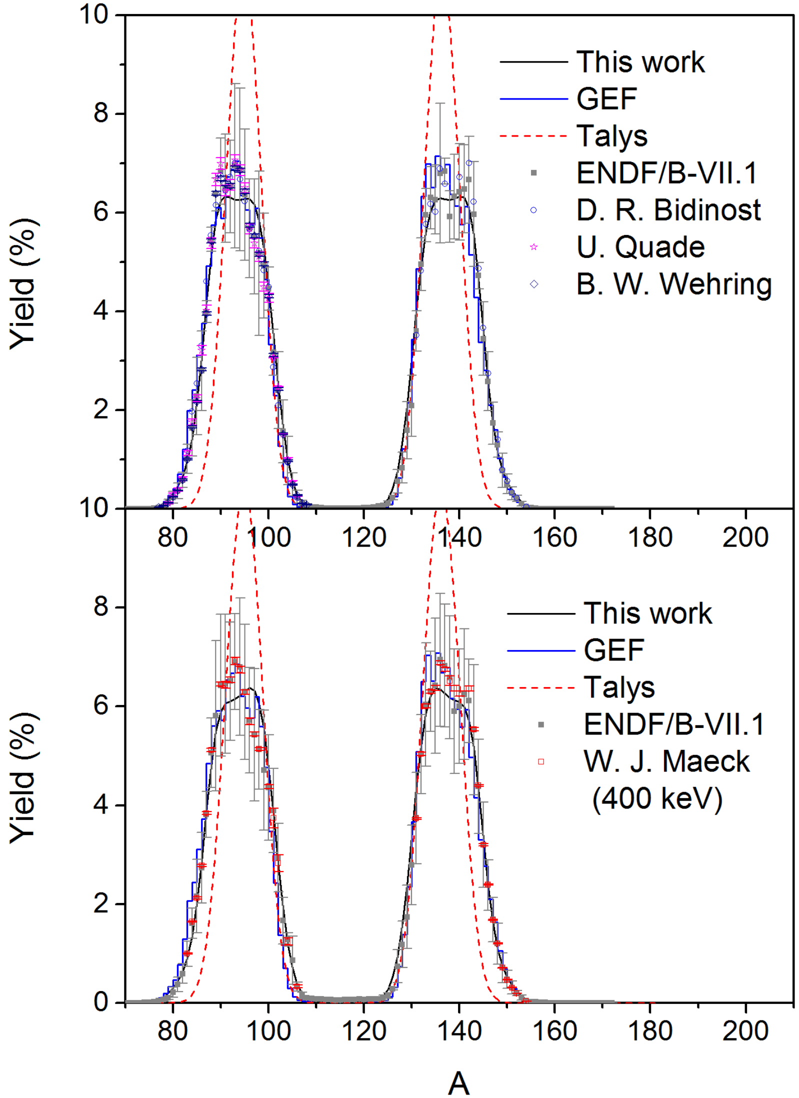
- $N_{out}, C_{in}, C_{out}, \sigma_{out}$ are almost constant while $\tilde{a}$ and σ_{in} vary with A_{CN}
- Parameter fitting was done in two steps. (First, fix four parameters and then fit the rest.)

Resulting parameters

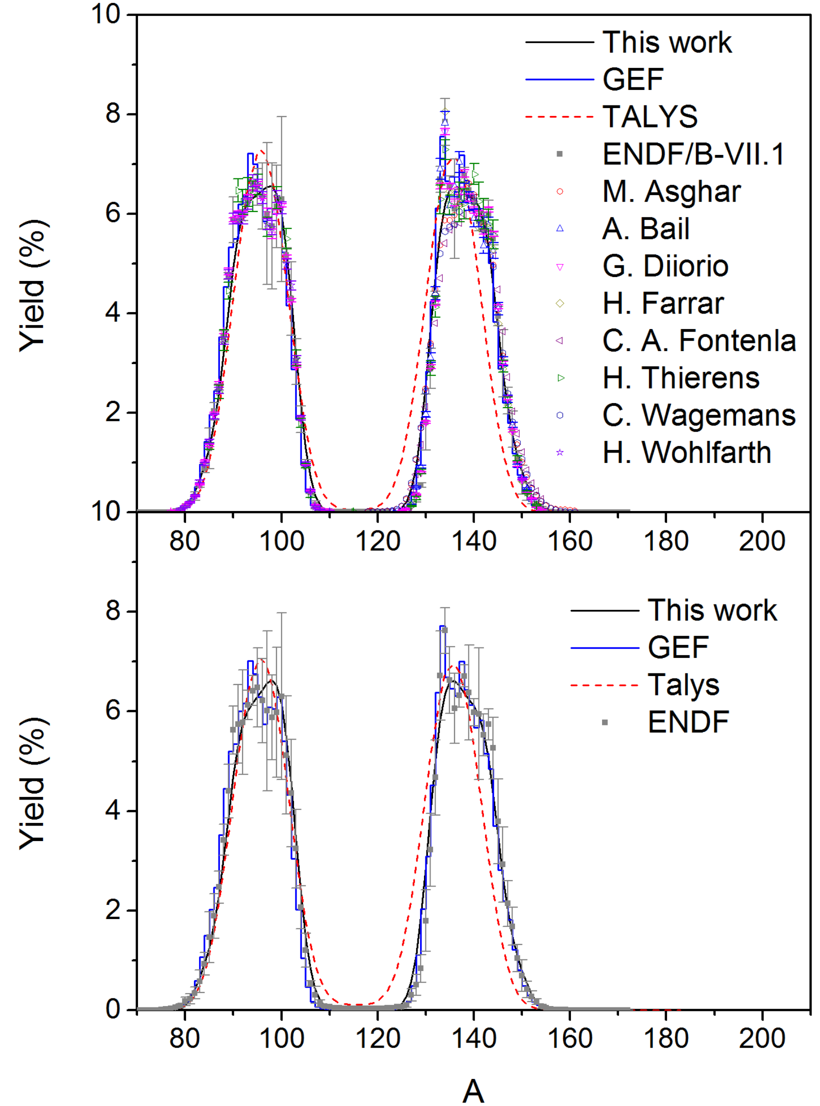


N_{out}	90.50
C_{in} (MeV)	-9.31
C_{out} (MeV)	-14.24
$\tilde{a}$ (MeV^{-1})	$-11.64A_{CN} + 2799$
σ_{in}	$-0.66A_{CN} + 163$
σ_{out}	5.13

Calculation results (uranium isotopes)



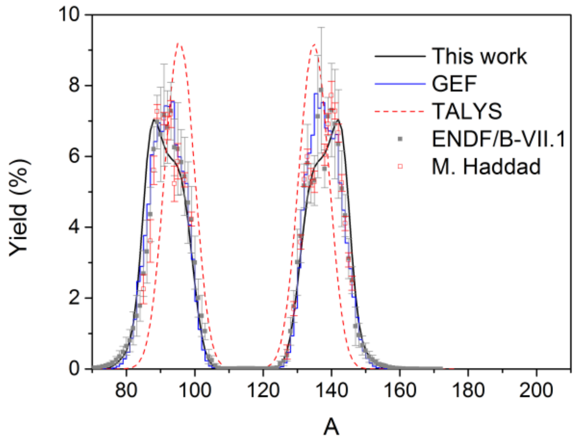
$^{233}U(n_{th},f)$, E_n=500 keV



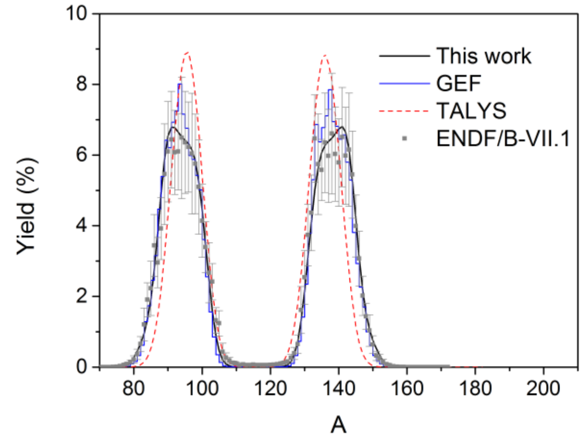
$^{235}U(n_{th},f)$, E_n=500 keV

Calculation results (uranium isotopes)

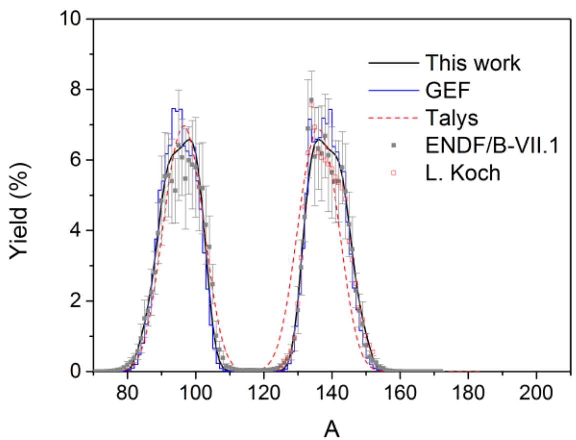
$^{232}U(n_{th},f)$



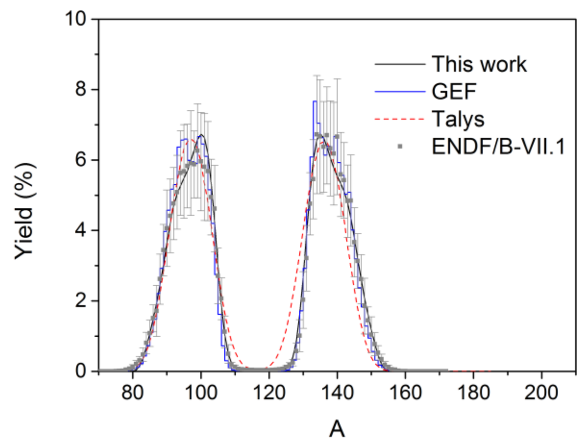
$^{234}U(n,f)$, E_n=500 keV



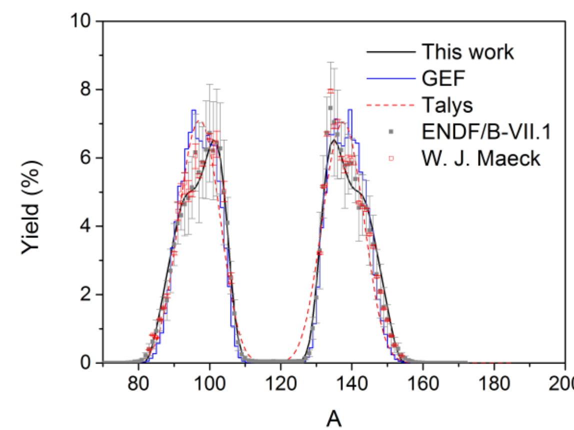
$^{236}U(n,f)$, E_n=500 keV



$^{237}U(n,f)$, E_n=500 keV



$^{238}U(n,f)$, E_n=500 keV



Comparison with other models

- To check the degree of agreement with ENDF/B-VII.1
  - $< Y^2 > \equiv \frac{1}{n}\sum_{k=1}^{n}(Y_k - \bar{Y_k})^2$
  - $\chi^2 \equiv \frac{1}{n}\sum_{k=1}^{n}\left(\frac{Y_k - \bar{Y}_k}{\Delta_k}\right)^2$

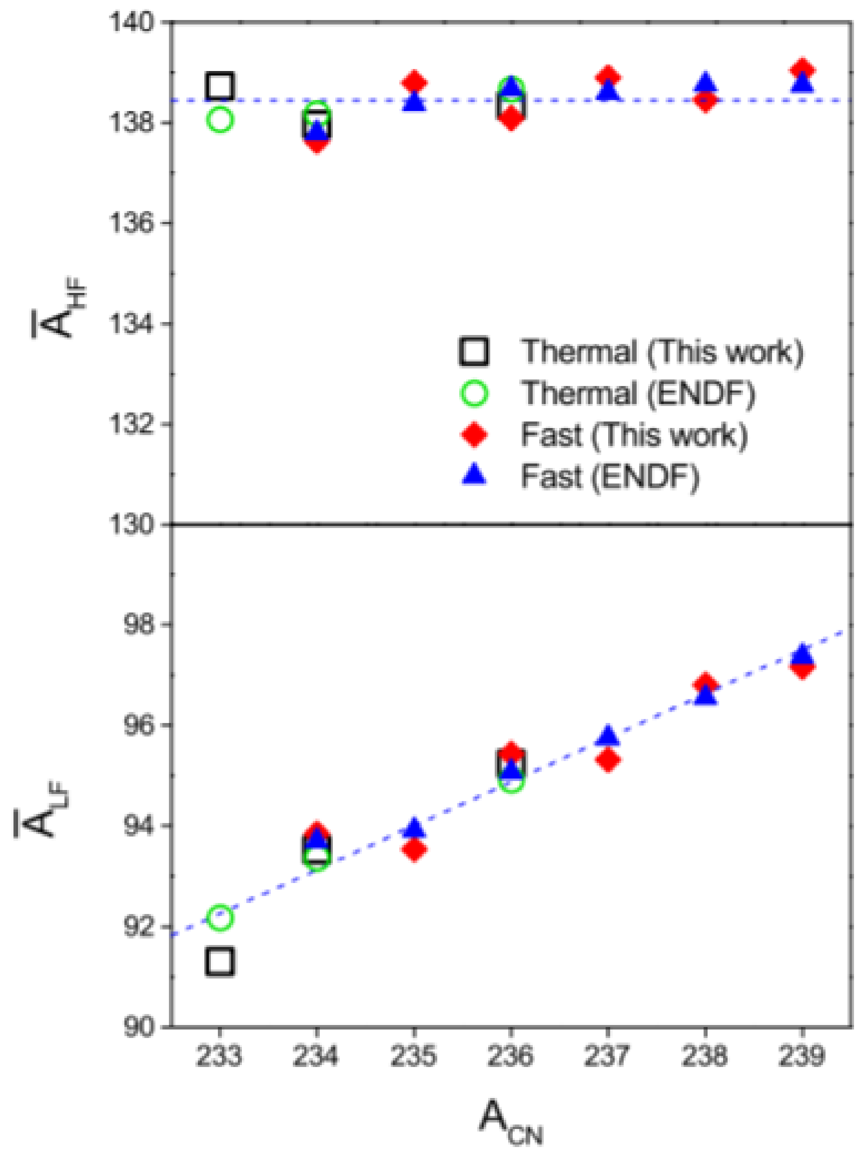
- Compared $< Y^2 >$ and χ^2 with GEF and TALYS
  - GEF: Describe the fission observables (ex. fission fragment yields, angular momentum distribution, neutron multiplicity, etc.) using semi-empirical model
  - TALYS: Software for the simulations of nuclear reactions. FPY are provided.

$\langle \Delta Y^2 \rangle$ and χ^2 (Uranium)

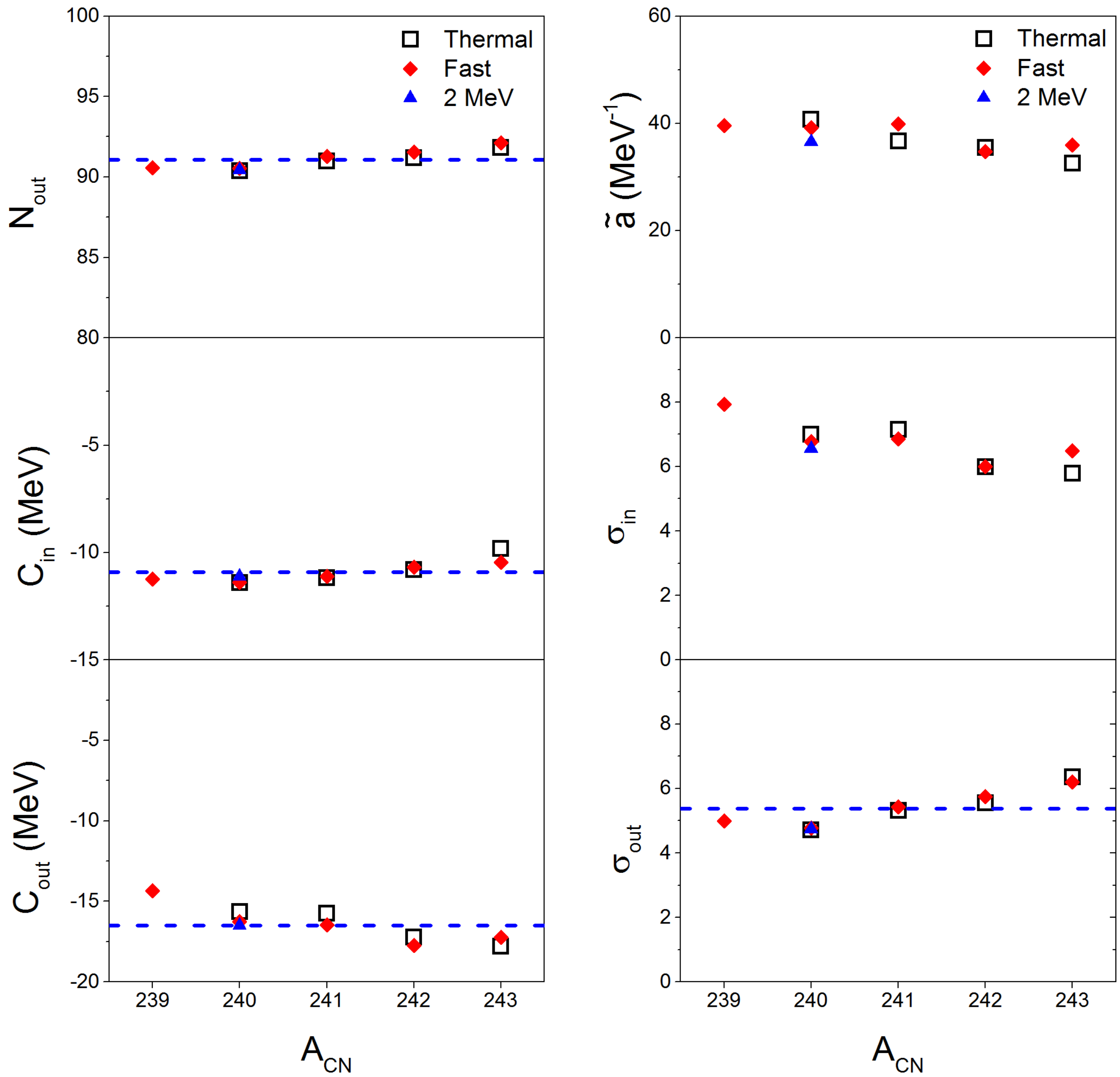
Incident neutron	Target	$\langle \Delta Y^2 \rangle$			χ^2		
		TALYS	GEF	This model	TALYS	GEF	This model
Thermal (0.0253 eV)	^{232}U	3.11	0.19	0.48	49.64	5.23	2.85
	^{233}U	3.05	0.09	0.07	16.65	1.40	4.44
	^{235}U	1.02	0.04	0.14	1153.19	4.34	54.01
Fast (500 keV)	^{233}U	2.44	0.09	0.08	6.37	1.64	8.12
	^{234}U	1.80	0.28	0.13	4.74	1.98	1.91
	^{235}U	0.88	0.03	0.13	101.11	0.58	7.83
	^{236}U	0.82	0.35	0.19	22.46	2.01	1.90
	^{237}U	0.59	0.34	0.09	50.35	1.42	1.17
	^{238}U	0.57	0.40	0.11	48.87	2.33	1.52
Average		**1.59**	**0.20**	**0.16**	**161.49**	**2.33**	**9.31**

Characteristic feature of fission well described

$$\bar{A}_{HF} = \left(\sum_{A\in HF} A\,Y(A)\right) \Big/ \left(\sum_{A\in HF} Y(A)\right)$$



Resulting parameters (plutonium isotopes)



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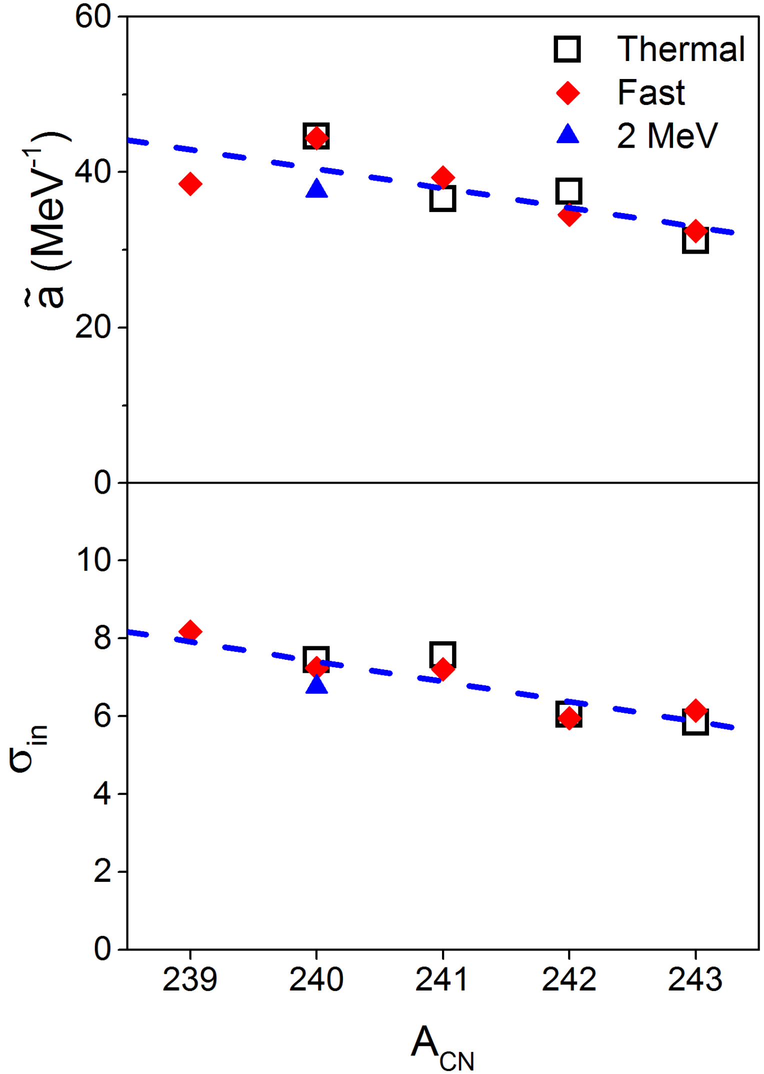
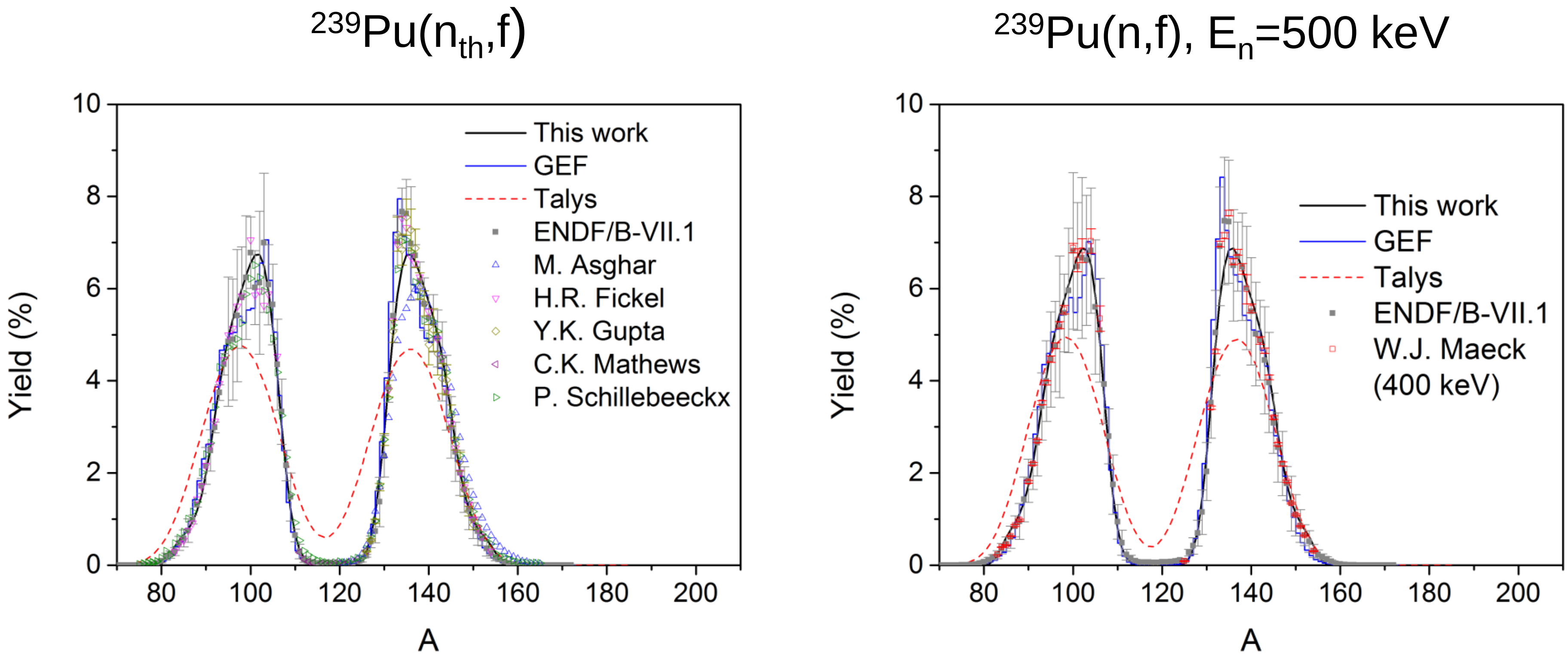


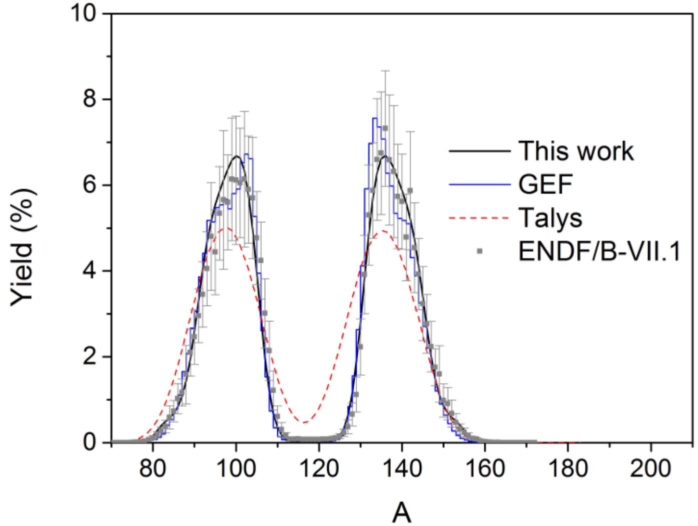
N_{out}	91.08
C_{in} (MeV)	-10.92
C_{out} (MeV)	-16.49
$\tilde{a}$ (MeV^{-1})	$-2.49A_{CN} + 637$
σ_{in}	$-0.51A_{CN} + 130$
σ_{out}	5.38

Calculation results (plutonium isotopes)

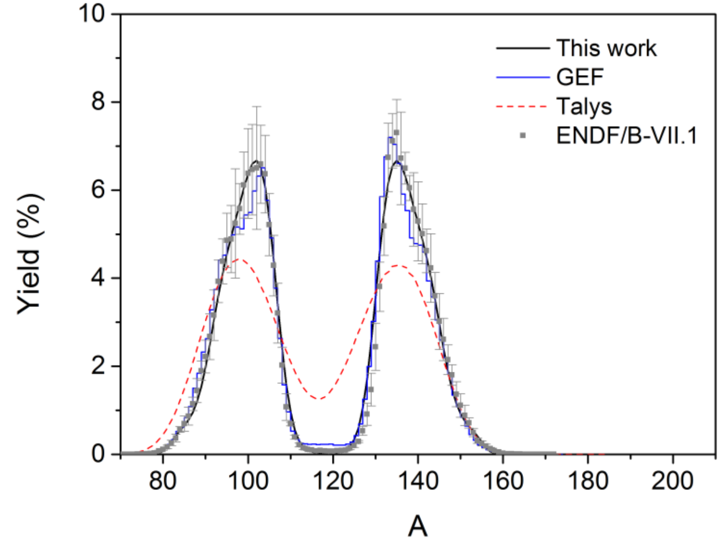


Calculation results (plutonium isotopes)

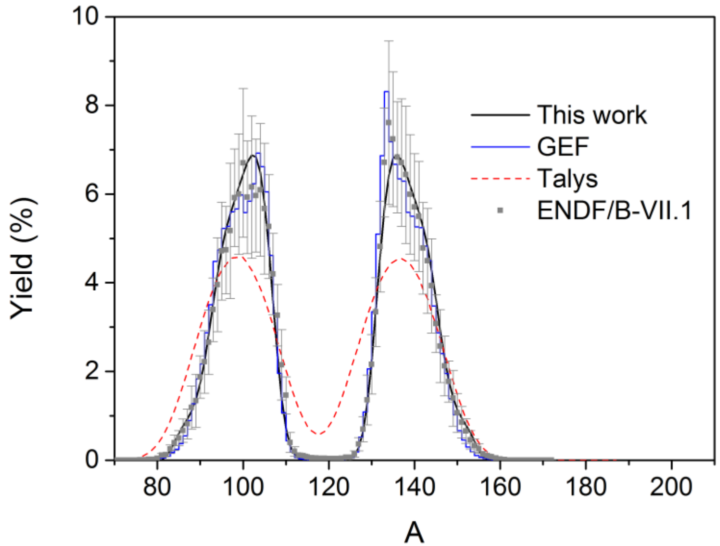
$^{238}Pu(n,f)$, E_n=500 keV



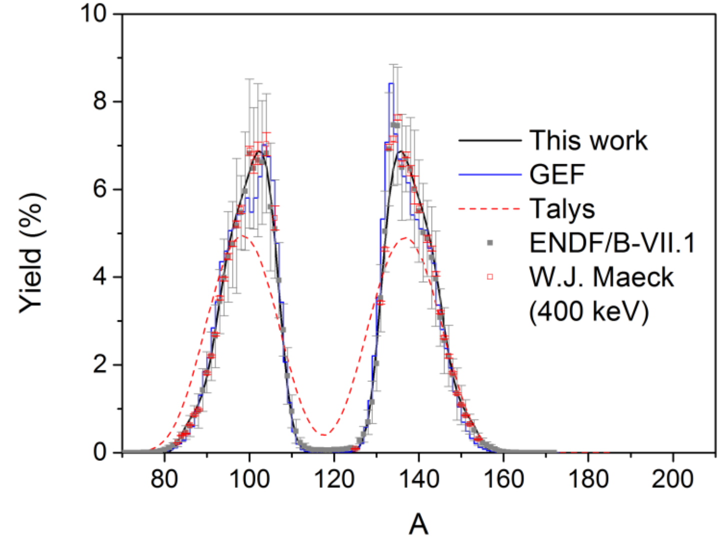
$^{239}Pu(n,f)$, E_n=2 MeV



$^{240}Pu(n_{th},f)$

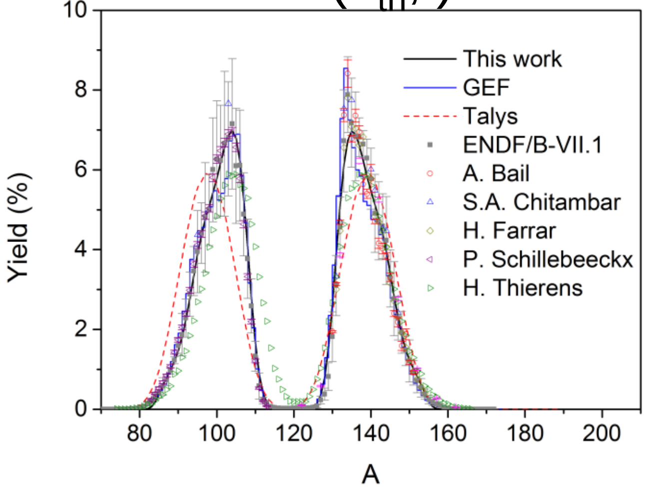


$^{240}Pu(n,f)$, E_n=500 keV

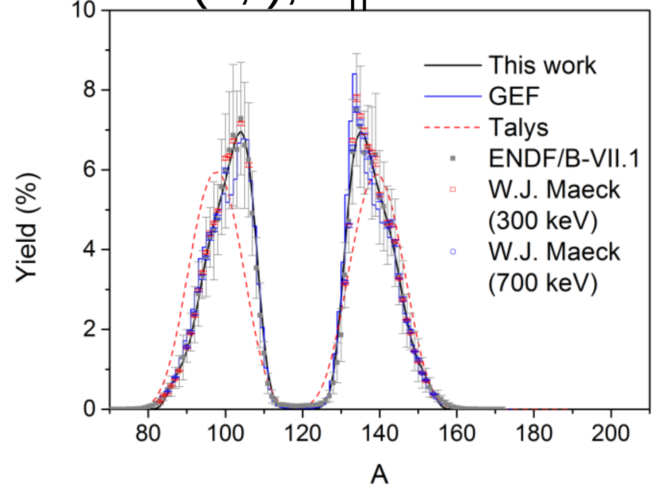


Calculation results (plutonium isotopes)

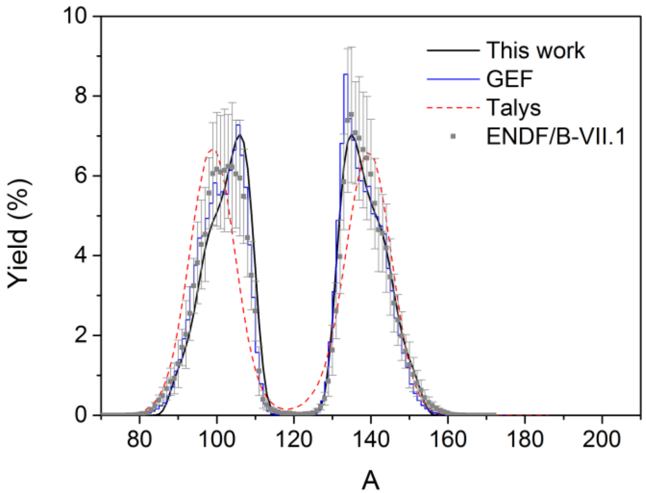
$^{241}Pu(n_{th},f)$



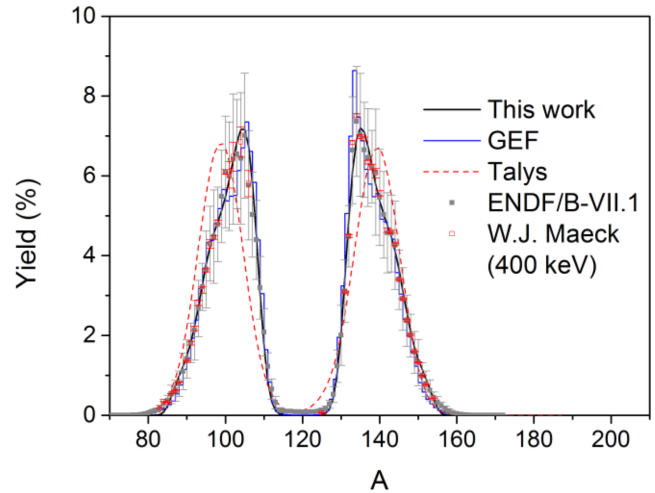
$^{241}Pu(n,f)$, E_n=500 keV



$^{242}Pu(n_{th},f)$



$^{242}Pu(n,f)$, E_n=500 keV



<ΔY²> and χ² (Plutonium)

Incident neutron	Target	$\langle \Delta Y^2 \rangle$			χ^2		
		TALYS	GEF	This model	TALYS	GEF	This model
Thermal (0.0253 eV)	^{239}Pu	1.66	0.09	0.09	657.11	0.92	1.50
	^{240}Pu	1.66	0.09	0.12	608.46	0.96	1.25
	^{241}Pu	1.51	0.18	0.08	52.37	0.67	1.44
	^{242}Pu	1.21	0.20	0.34	51.36	1.15	1.85
Fast (500 keV)	^{238}Pu	1.30	0.12	0.16	153.63	0.85	1.37
	^{239}Pu	1.84	0.10	0.05	1002.99	1.27	3.47
	^{240}Pu	1.42	0.13	0.07	129.42	1.11	1.23
	^{241}Pu	1.49	0.18	0.07	6.18	0.68	1.32
	^{242}Pu	1.30	0.22	0.07	5.58	1.33	1.57
2 MeV	^{239}Pu	1.81	0.10	0.08	2055.98	24.52	8.60
Average		**1.52**	**0.14**	**0.11**	**472.31**	**3.35**	**2.36**

Consistent parameters for U and Pu

	U	Pu
N_{out}	90.50	91.08
C_{in} (MeV)	-9.31	-10.92
C_{out} (MeV)	-14.24	-16.49
$\tilde{a}$ (MeV^{-1})	-11.64A_{CN} + 2799	-2.49A_{CN} + 637
σ_{in}	-0.66A_{CN} + 163	-0.51A_{CN} + 130
σ_{out}	5.13	5.38
γ	0.06	
V_0 (MeV)	5	
N_{in}	82	
C_{mac}	$\left(\frac{8}{N_{CN}^2}\right) 10^{7.16993-0.26602\left(\frac{Z_{CN}^2}{A_{CN}}\right)+0.00283\left(\frac{Z_{CN}^2}{A_{CN}}\right)^2}$	

Calculation of “pre”-neutron & “post”-neutron FPY data

- Post-neutron FPY data
  - Fairly complete datasets are available (cf. ENDF)
  - There are less pre-neutron emission FPY data

- Pre-neutron emission FPY data :
  - observables directly related to fission
  - Post-neutron FPY data contains neutron evaporation of nascent fragments

- Our model can be used for both pre- and post- FPY data

- Consistent way of calculation both pre- and post- data in one model

Apply the model to pre-neutron FPY data

Macro potential

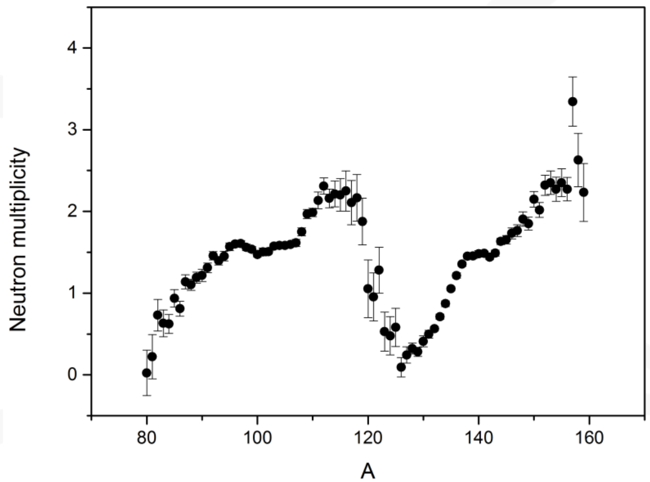
$$V_{mac}(N) = C_{mac}\left(N - \frac{\tilde{N}_{CN}}{2}\right)^2 + V_0$$

Shell correction

$$V_{sh}(N) = C_{in}\left[\exp\left(\frac{(N-N_{in})^2}{\sigma_{in}^2}\right) + \exp\left(\frac{(N-\overline{N}_{in})^2}{\sigma_{in}^2}\right)\right]$$
$$+ C_{out}\left[\exp\left(\frac{(N-N_{out})^2}{\sigma_{out}^2}\right) + \exp\left(\frac{(N-\overline{N}_{out})^2}{\sigma_{in}^2}\right)\right]$$

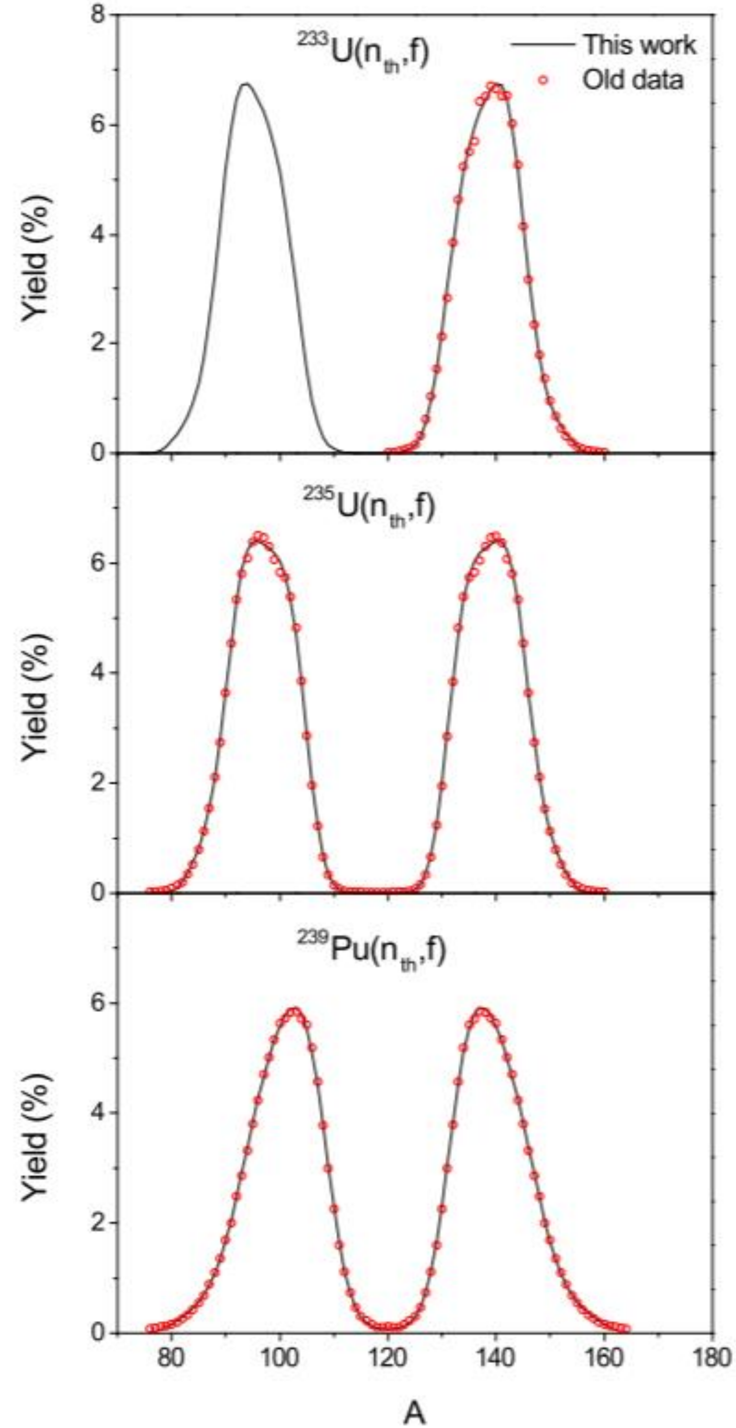
where

$\tilde{N}_{CN} \equiv N_{CN} - \nu$ and $\overline{N}_j \equiv \tilde{N}_{CN} - N_j$ $(j = in, out)$

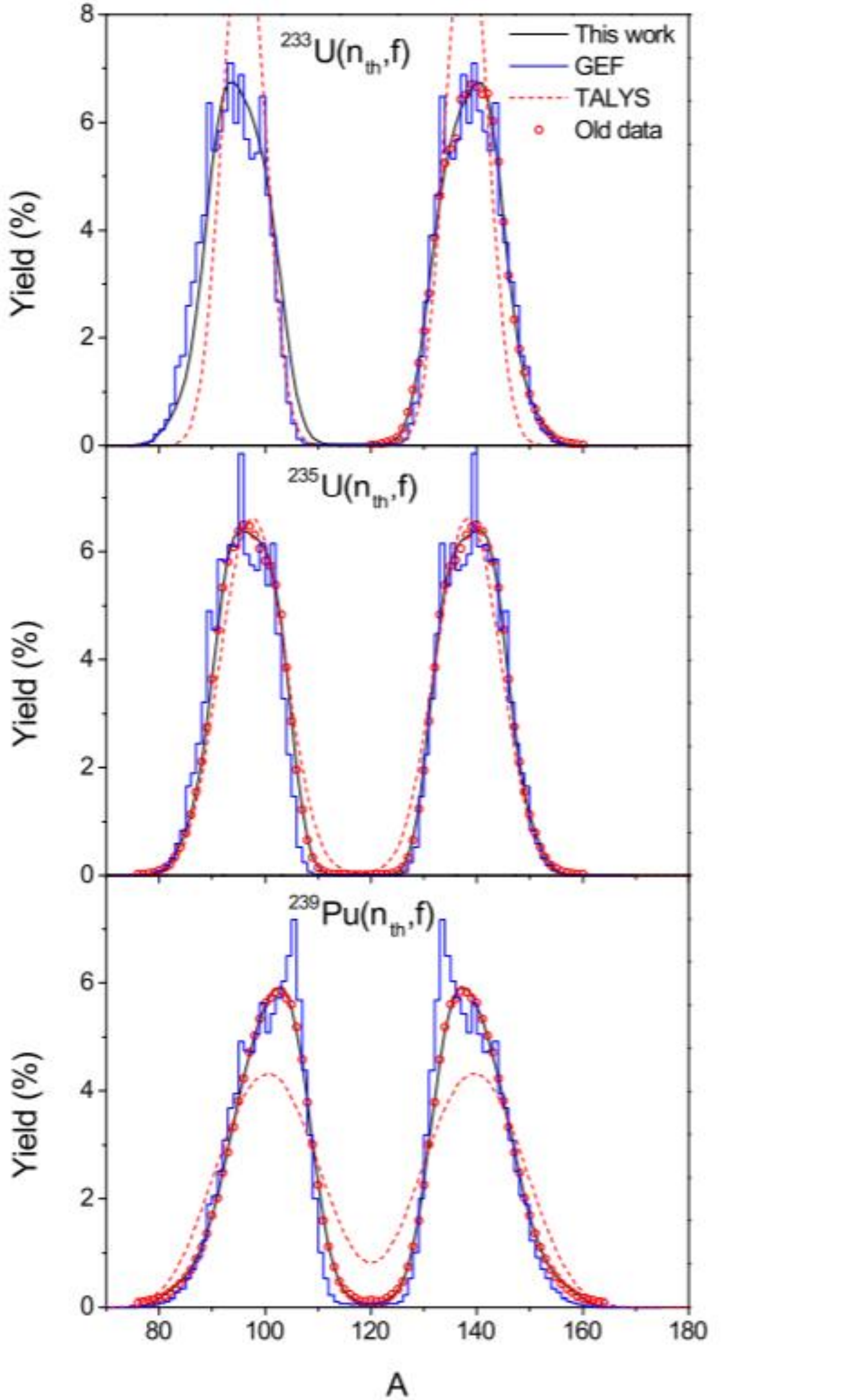


- Neutron multiplicity (ν) was treated at fission barrier as an average value
- If data exist for pre- and post- FPY and ν (N), replace the average by the measured ν (N).

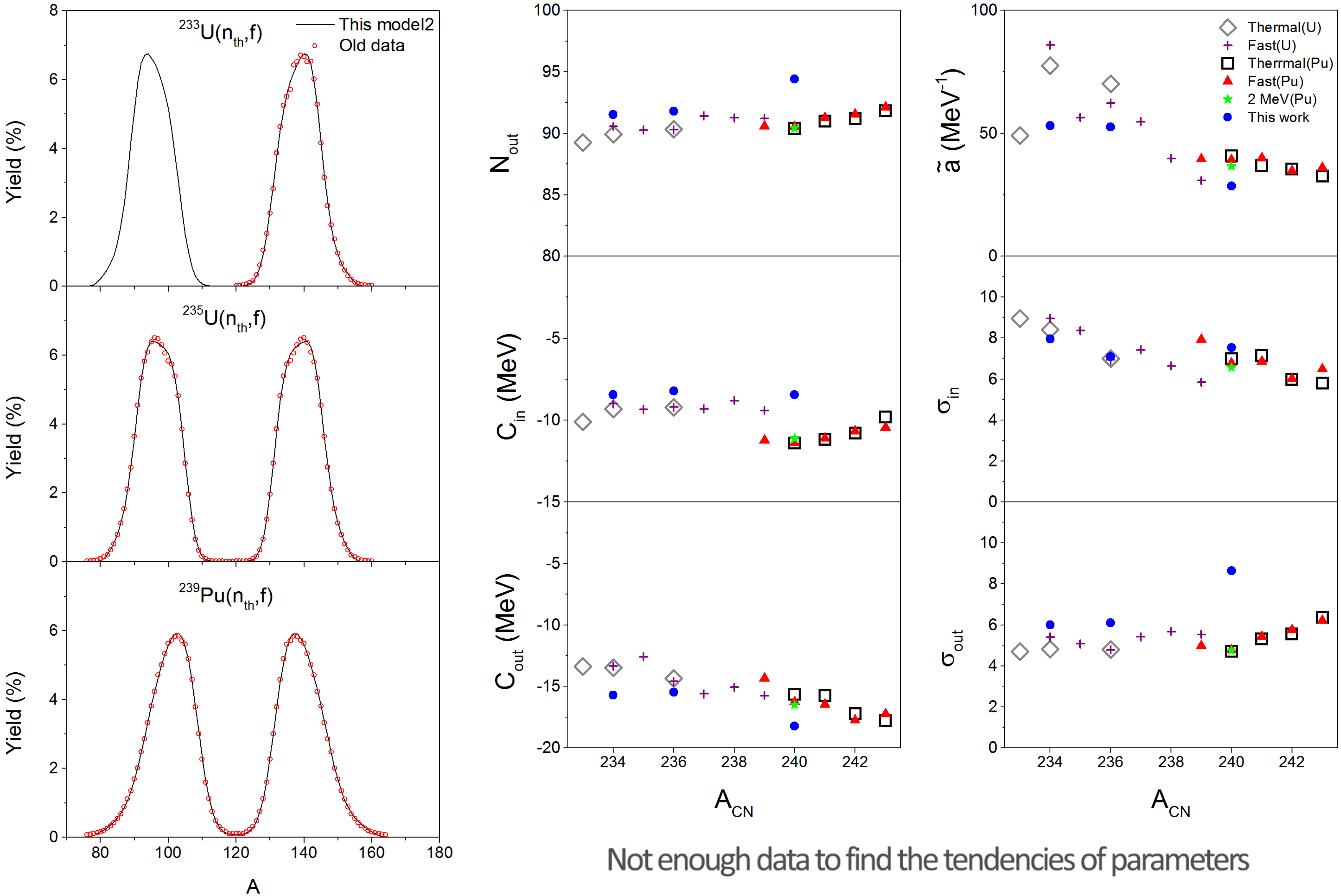
Calculation results for pre-neutron emission FPY data



Calculation results for pre-neutron emission FPY data

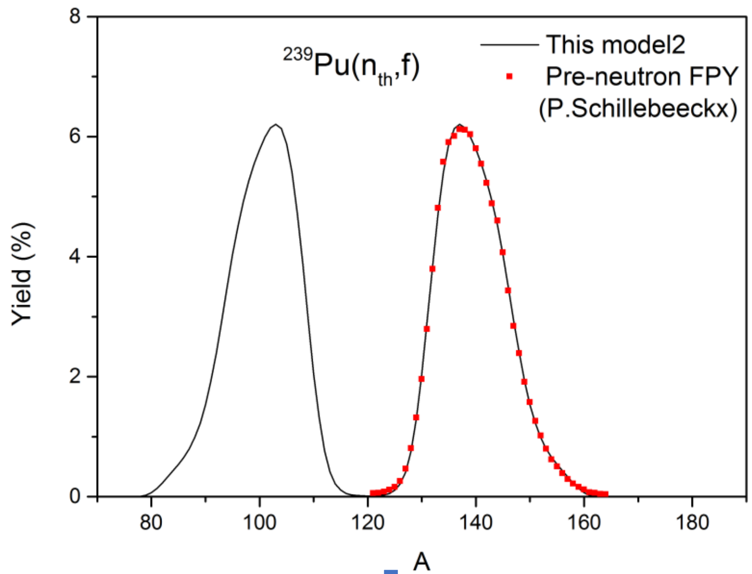


Parameters for pre-neutron FPY data

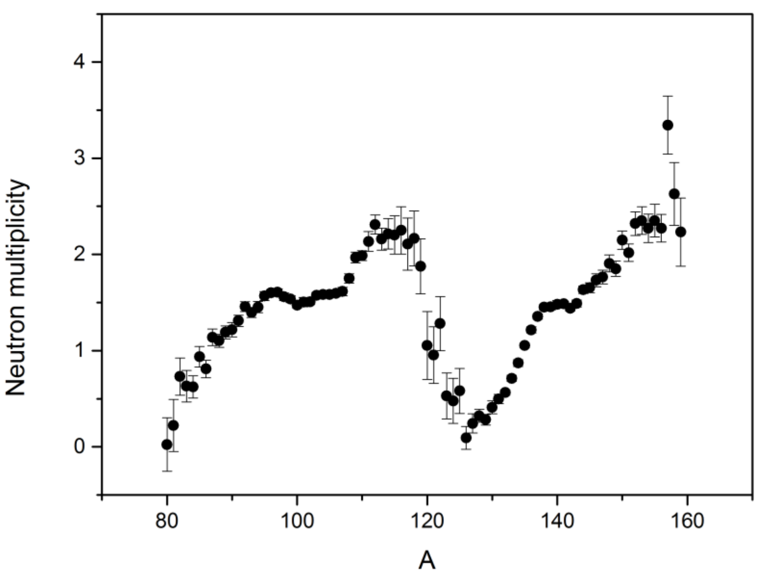


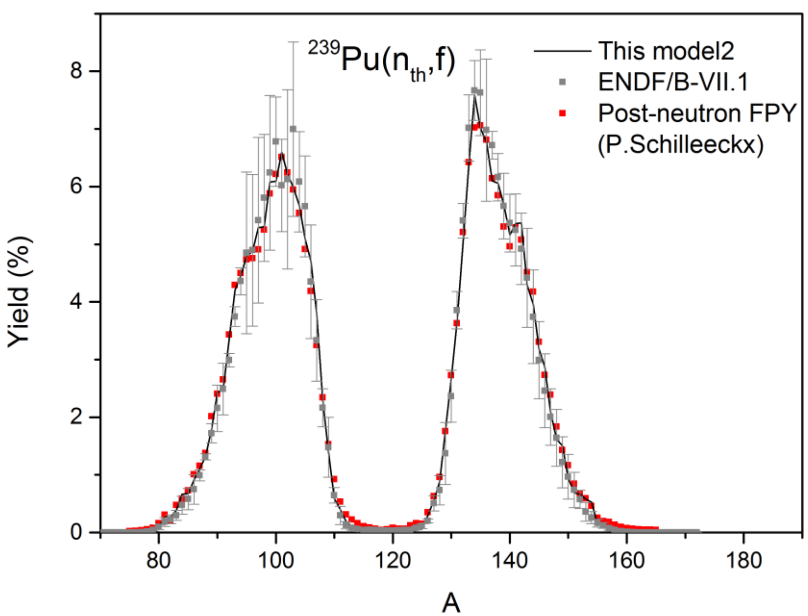
Not enough data to find the tendencies of parameters

Post-neutron FPY data by using neutron multiplicity

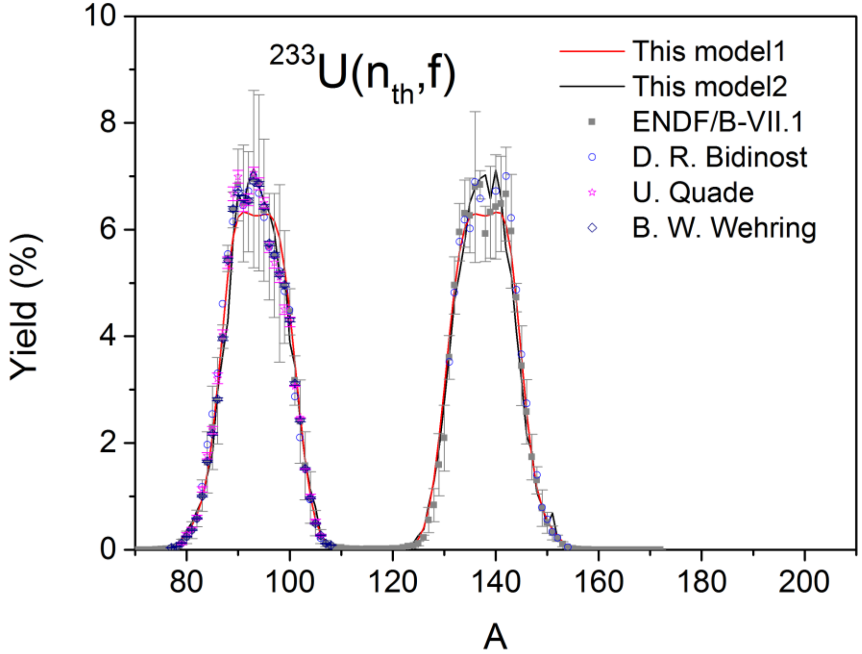


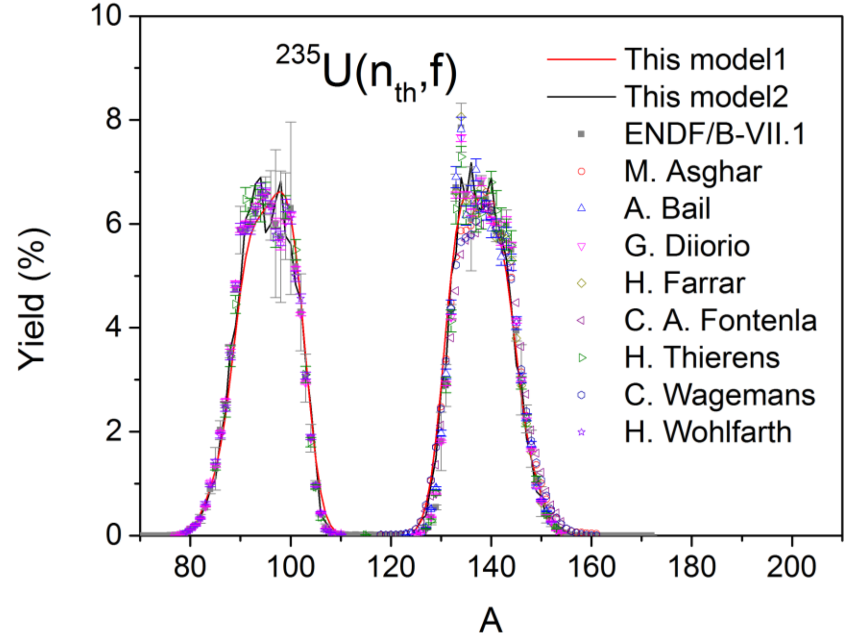
- Apply our model to pre-neutron FPY data
- Calculate post-neutron FPY using experimental ν (N)

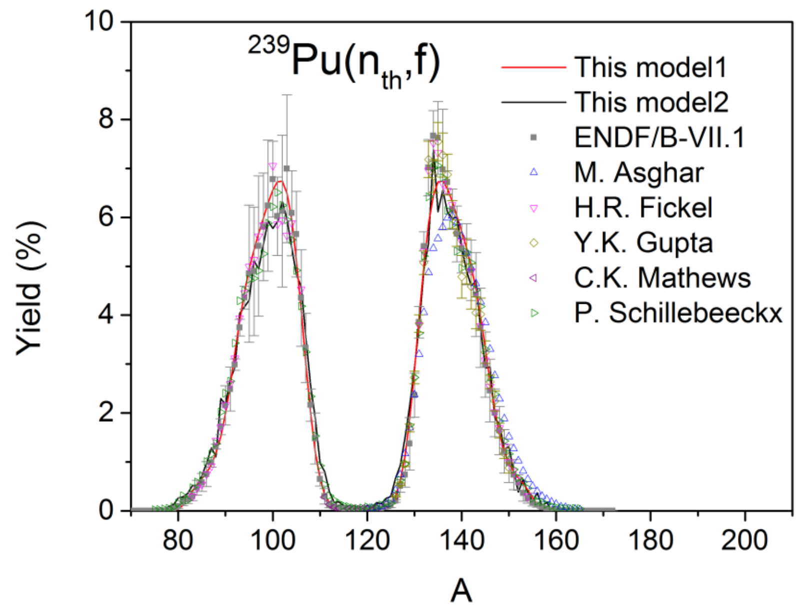




Post-neutron FPY with the same parameters







Conclusion

- We developed a simple semi-empirical model for FPY, which has just 10 parameters
- Our simple model reproduces the overall shape of the mass distribution of uranium and plutonium of ENDF data
- **Consistent description of pre- and post-neutron FPY data.**