The physics behind fission-yield models

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Training course | From nuclear data to a reliable estimate of spent fuel decay heat
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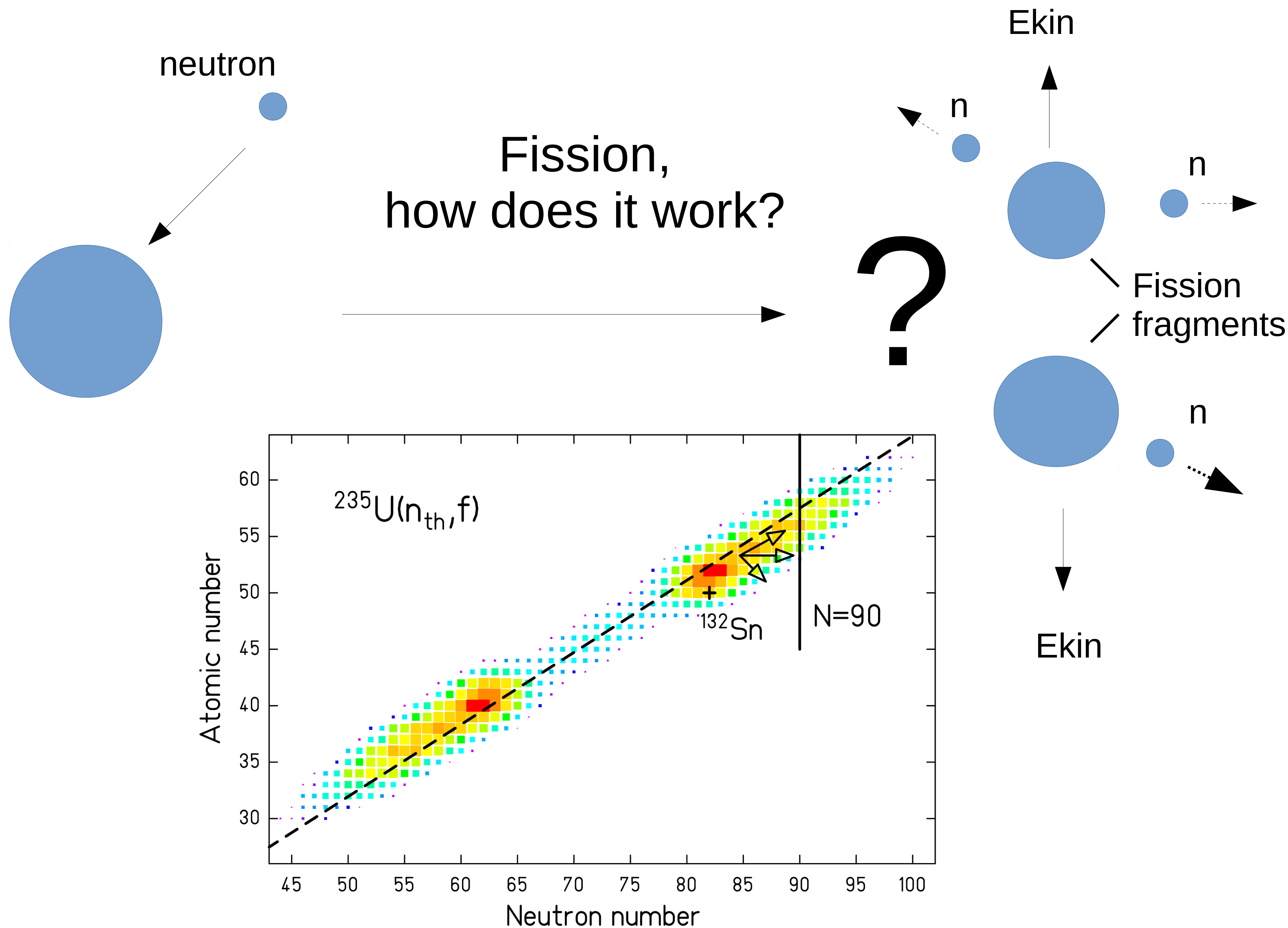
- Prompt neutron emission (shift in N, fine structure in N)
- Multi-chance fission
- Accuracy of fission yields, covariances
- Beta-delayed phenomena (neutrons, cumulative yields)
- Related quantities (decay heat, anti-neutrinos etc.)

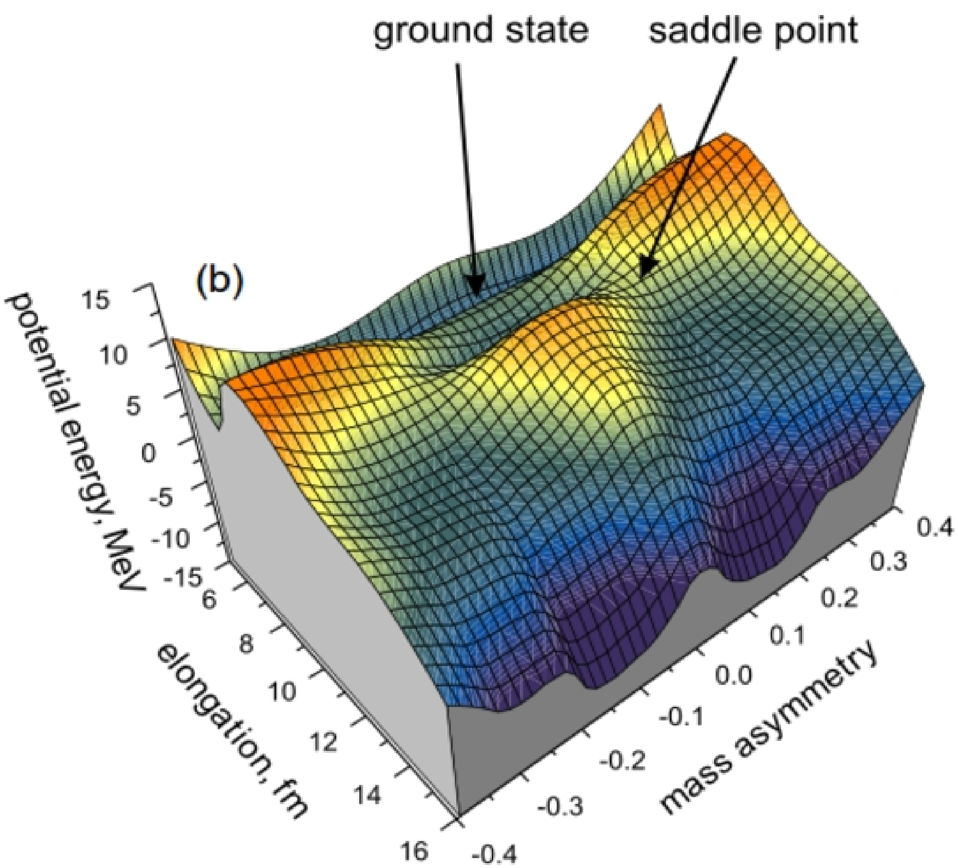
Introduction

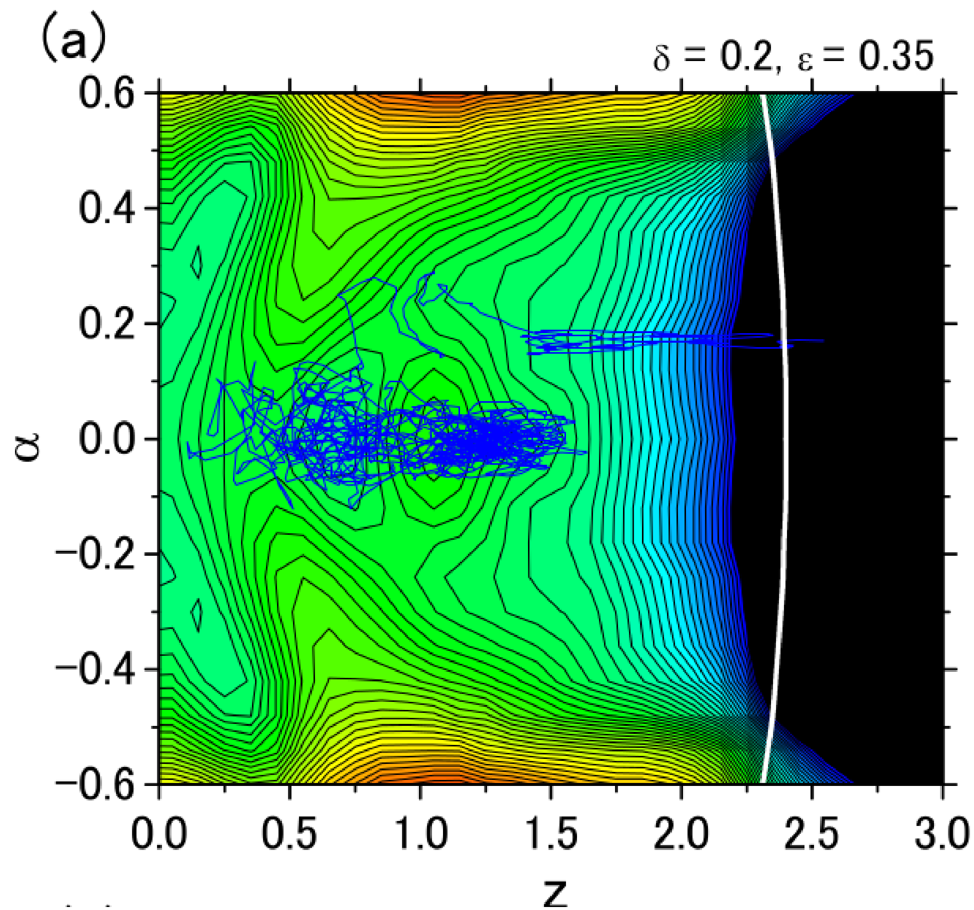


Classification of fission models

Model	Main features	Main results	Limitations
TDDFT (TDHF) (Bulgac 2016, Tanimura 2017)	Quantum-mechanical transport model, self-consistent, no shape restriction	Collective excitations, long fission times	1 most probable event in 550 min on Cray XK7
TDGCM+GOA (Goutte 2005, Regnier 2016)	Quantum-mechanical, self-consistent	Rough reproduction of FF mass and TKE distributions	Adiabaticity and other approximations, Limited accuracy, very high computing demand
Stochastic (Langevin equations) (Sierk 2017)	Classical transport equation towards statistical equilibrium	Systematic calculations of mass distributions	Limited accuracy, high computing demand
GEF (Schmidt 2016)	Semi-empirical, general concepts and laws, very fast code	Covers almost all fission quantities, and systems, predictive power, high accuracy	Accuracy is limited by uncertainties of experimental data
Parametrization (Wahl, Katakura)	Empirical systematics, FF yields, prompt neutrons	Evaluation and completion of experimental data	Restricted by available experimental data

Illustrative example: Stochastic model





Potential energy in landscape of nuclear shapes (238U)

Karpov 2008

Shape evolution (trajectories) with forces from potential, inertia and dissipation
Aritomo 2015

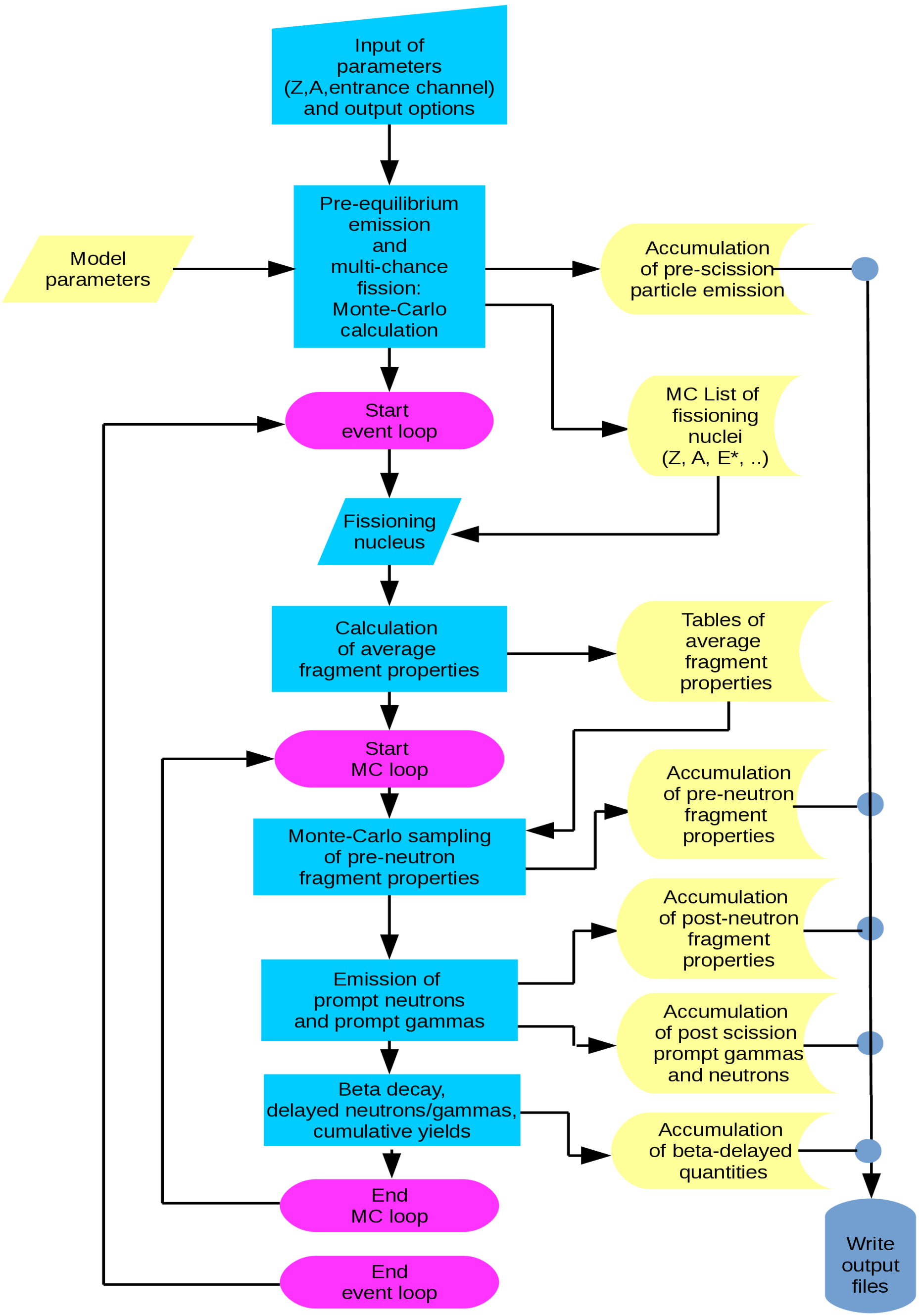
Limitations of stochastic models

- Insufficient flexibility due to limited number of shape parameters
  - Does not cover all observables (mostly A - TKE)
  - Permits only “simple” shapes
- Limited accuracy due to inherent uncertainties of quantitative results
- Practical restrictions by high computing demand

Philosophy of GEF

- Avoid complexities and uncertainties of microscopic calculations.
- Exploit laws and concepts (mathematics and physics) that establish regularities, correlations and general trends.
- Rather direct fit to experimental data.
- Comprehensive coverage of all fission quantities in a consistent way.
- Fast algorithms.

Structure of the GEF model code



<u>Input</u>:

- n-induced fission
  - Target nucleus
  - Incident-neutron energy

or

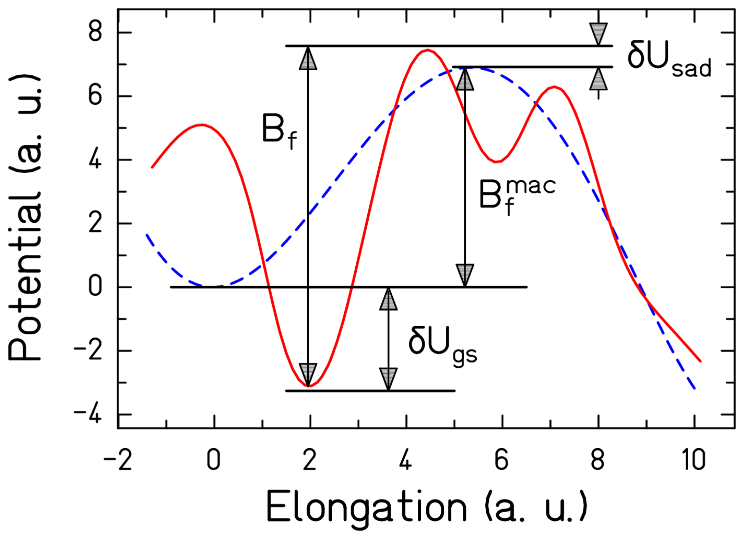
- Z, A, E*, l of CN

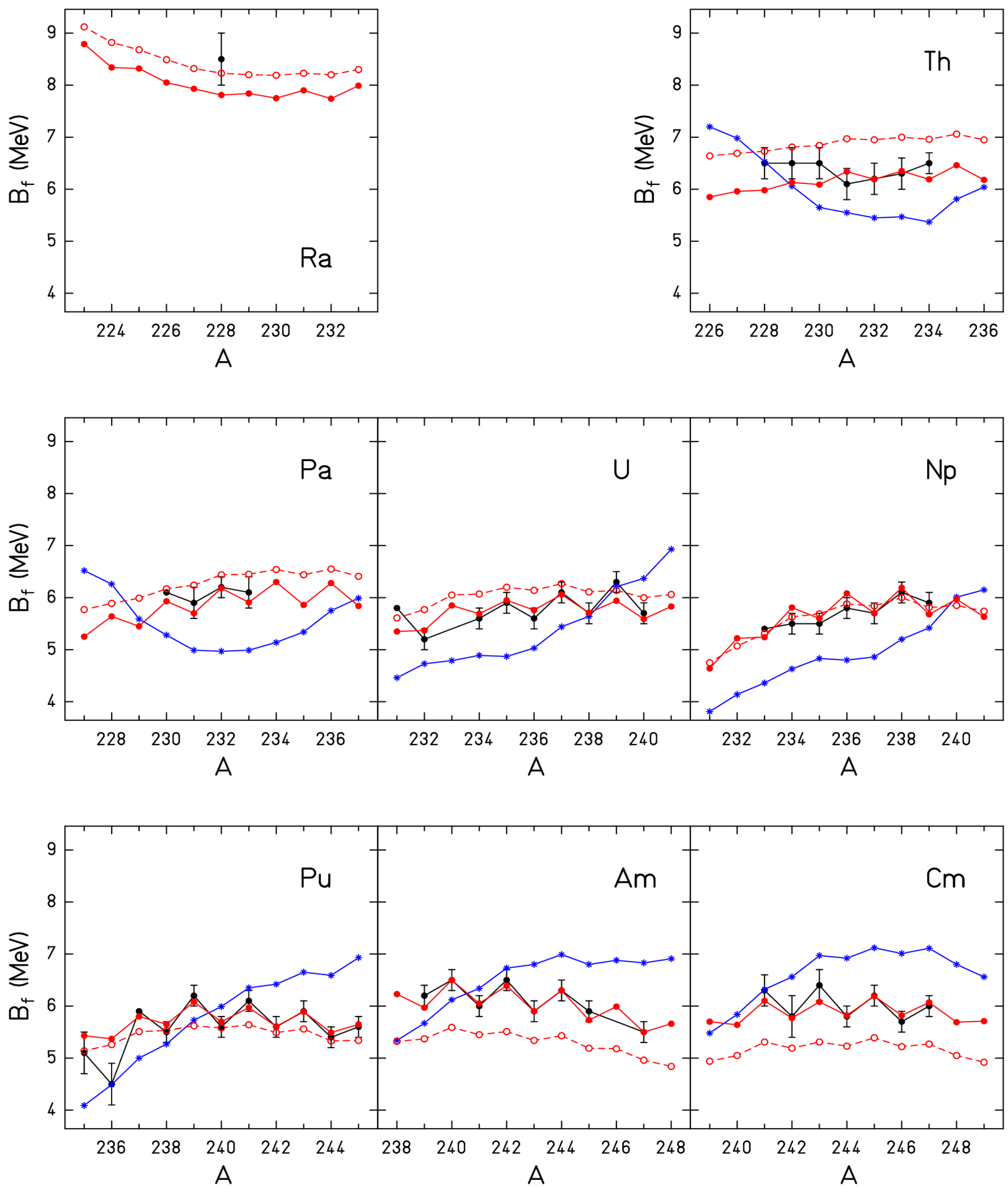
<u>Output</u>:

- Z1, A1, Z2, A2,, l1,l2, pre-and post-neutron, isomeric ratios, TKE, prompt neutrons and gammas
- Event generator, covariances, ENDF files, random files
- Beta-delayed processes

GEF covers the whole fission process!

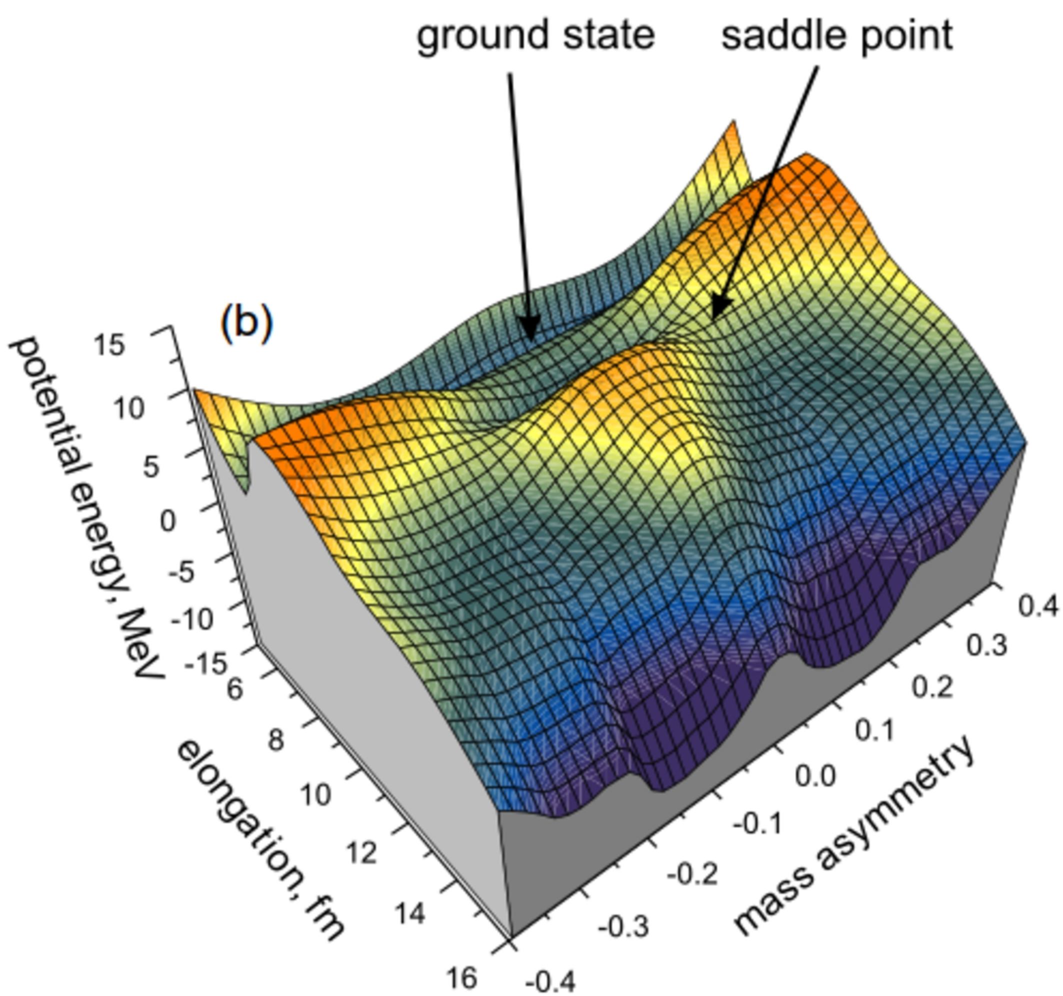
Fission barriers





Bf constructed by the topograpic theorem of Myers and Swiatecki:
1. Neglect shell-correction energy at saddle (use mac. model),
2. Use experimental g.s. masses.
Eventually:
3. Increase pairing and adjust global trend in Z.
Compared with mic-mac model (Möller 2009) and empirical data (Bjornholm, Lynn 1980).

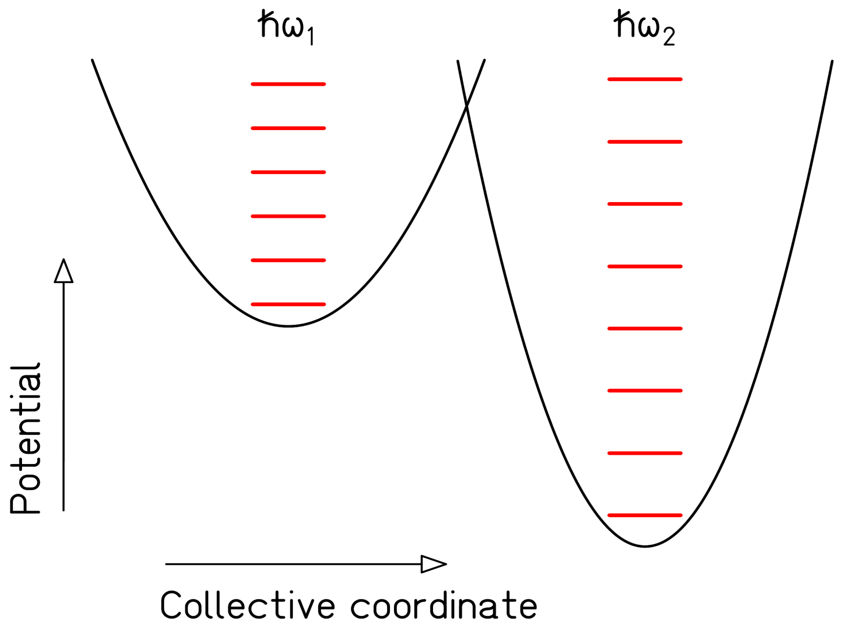
Concept of normal modes



Potential of 238U (Karpov 2008).

Concept used in GEF:
The fission process is characterized by a motion towards fission with oscillations (harmonic oscillators) in many other degrees of freedom (normal modes: mass asymmetry, charge polarization etc.) on top. The population and the excitation of these oscillators determine the distribution of FF yields.

Normal modes: Population of fission valleys



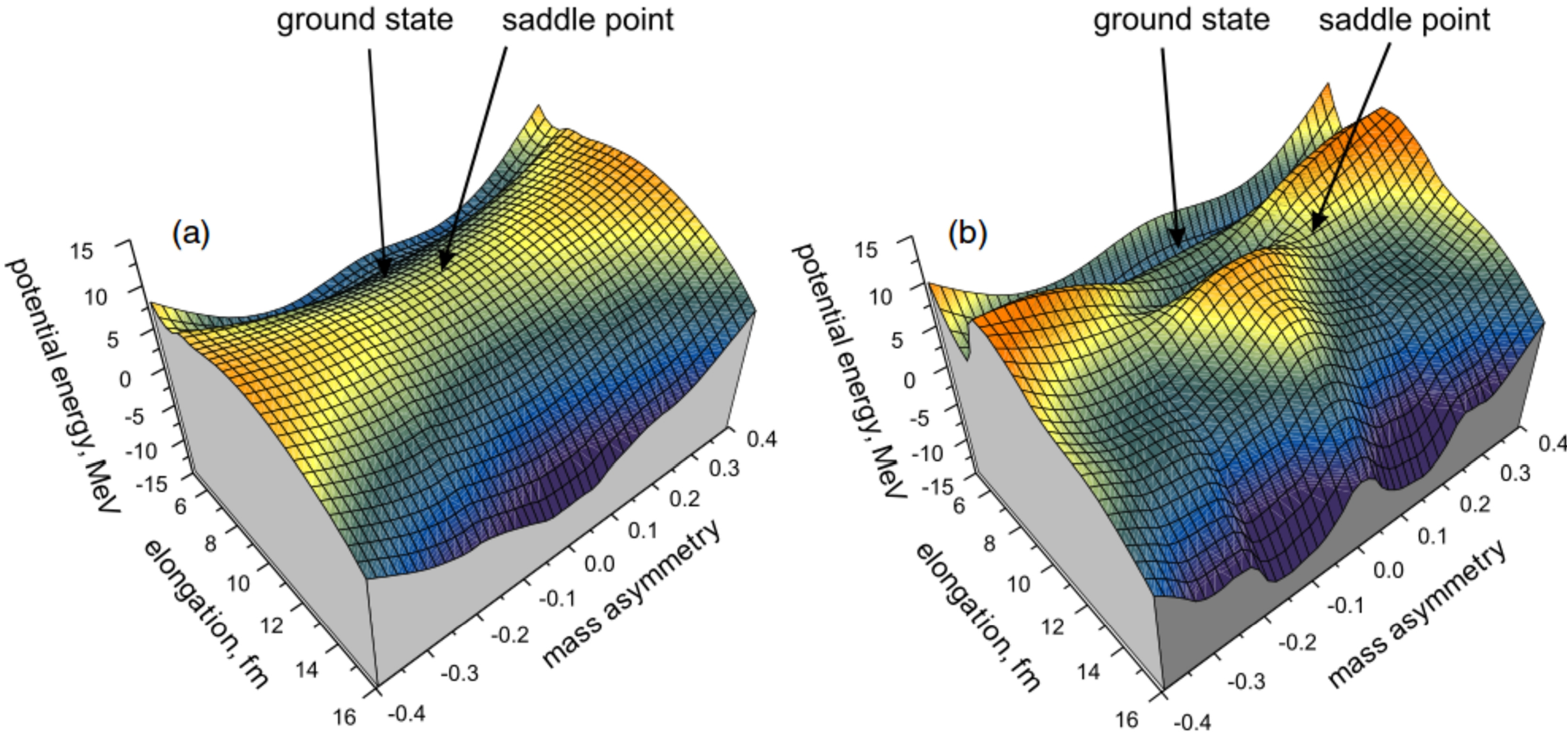
Population of fission valleys (fission channels) in statistical equilibrium right after the second barrier:

$$\frac{Y_1}{Y_2} = \exp(-\Delta E / T)$$

Variance of the corresponding collective coordinate at scission is given by the equation of the quantum oscillator:

$$\sigma^2 = \frac{\hbar\omega}{2C} \coth\left(\frac{\hbar\omega}{2T}\right)$$

Importance of fragment shells: Illustration



Macroscopic potential of 238U

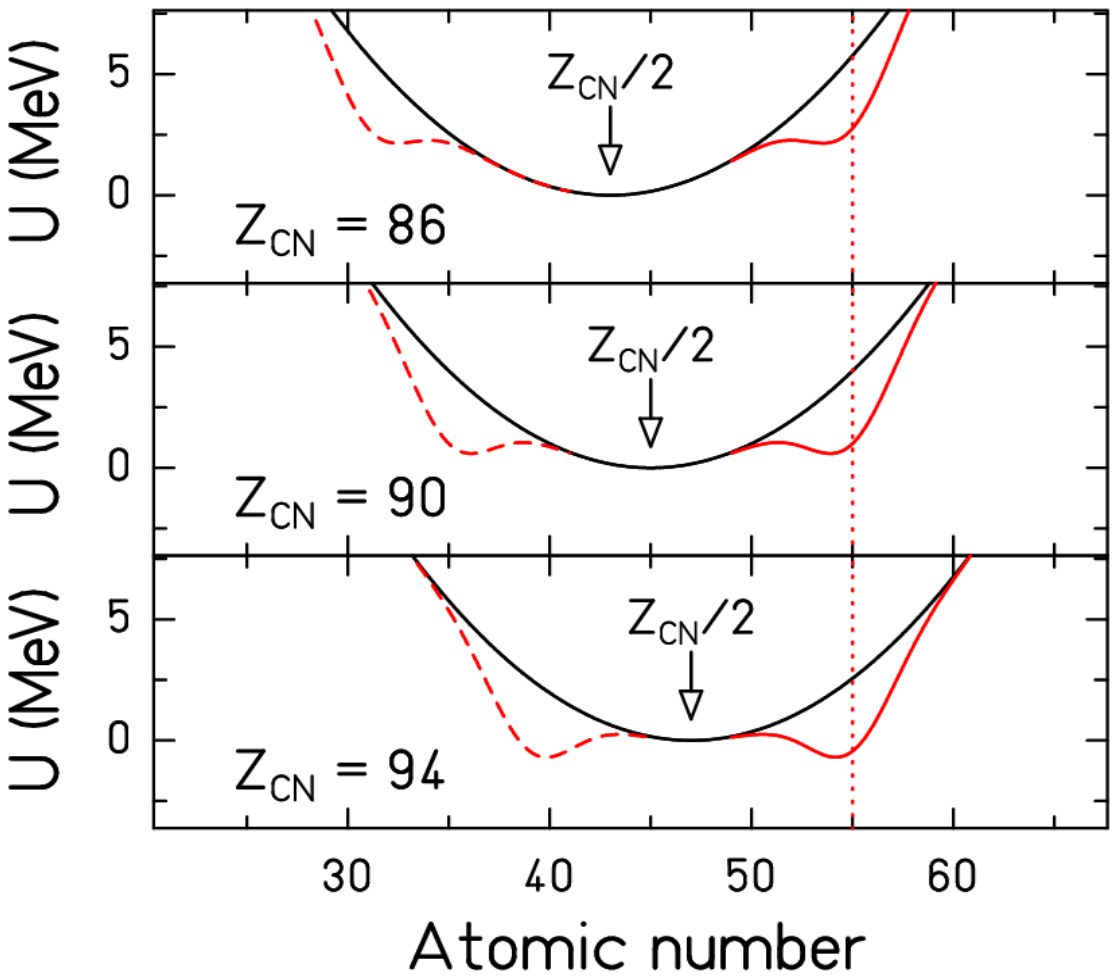
Mac-mic potential of 238U

Calculations of Mosel and Schmitt (1971!) revealed that shells behind the 2nd saddle are essentially the superposition of fragment shells. → The same fragment shells are responsible for the structures in the FF yields of different systems!

Exploiting the fragment shells in GEF

- Macroscopic features of the FF yields are specific to the fissioning nucleus (ACN, ZCN).
- Microscopic features of the FF yields are specific to the fragments, independent from the fissioning nucleus (A1,Z1, A2, Z2).
- This creates regularities in the variation of FF yields for different systems and different E*
- This simplifies the adjustment of the GEF model to the experimental data enormously.

Variation of fission channels



Relative yields of the fission channels are given by the interplay of macroscopic and microscopic features of the potential energy in asymmetry.

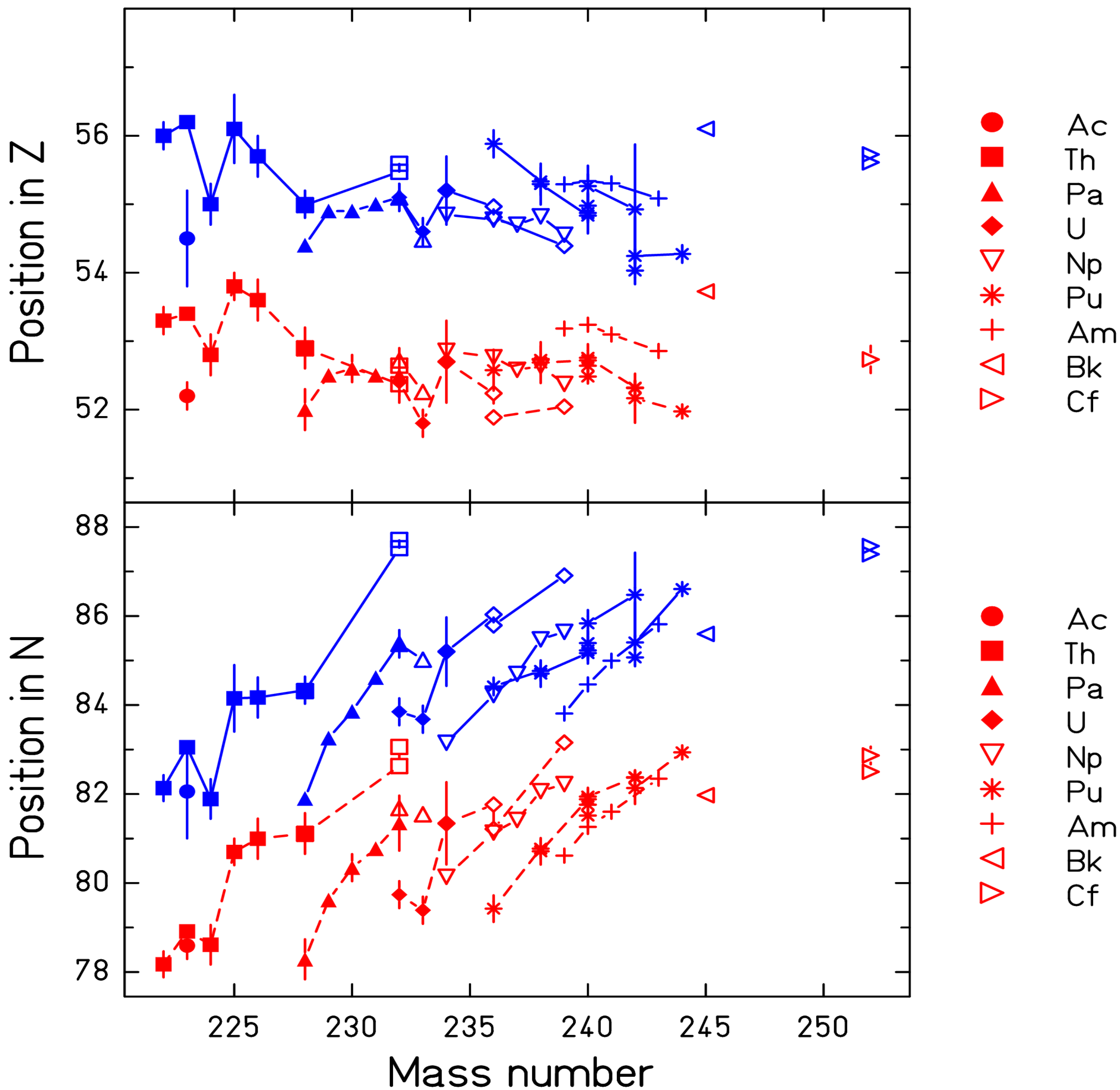
Dynamic effects

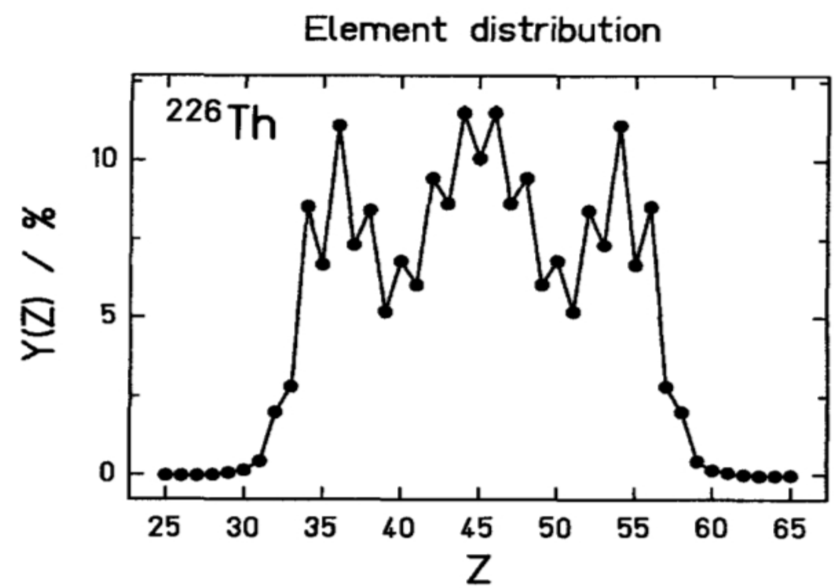
The mass distribution does not directly reflect the potential at scission, because the system needs some time to adjust to the potential due to inertia and friction → memory effect (Adeev, Pashkevich 1989).

Every collective degree of freedom has a specific characteristic time constant.

Therefore, the effective potential-energy properties in GEF correspond (approximately) to the potential at the characteristic time before reaching scission.

Empirical fragment shells deduced

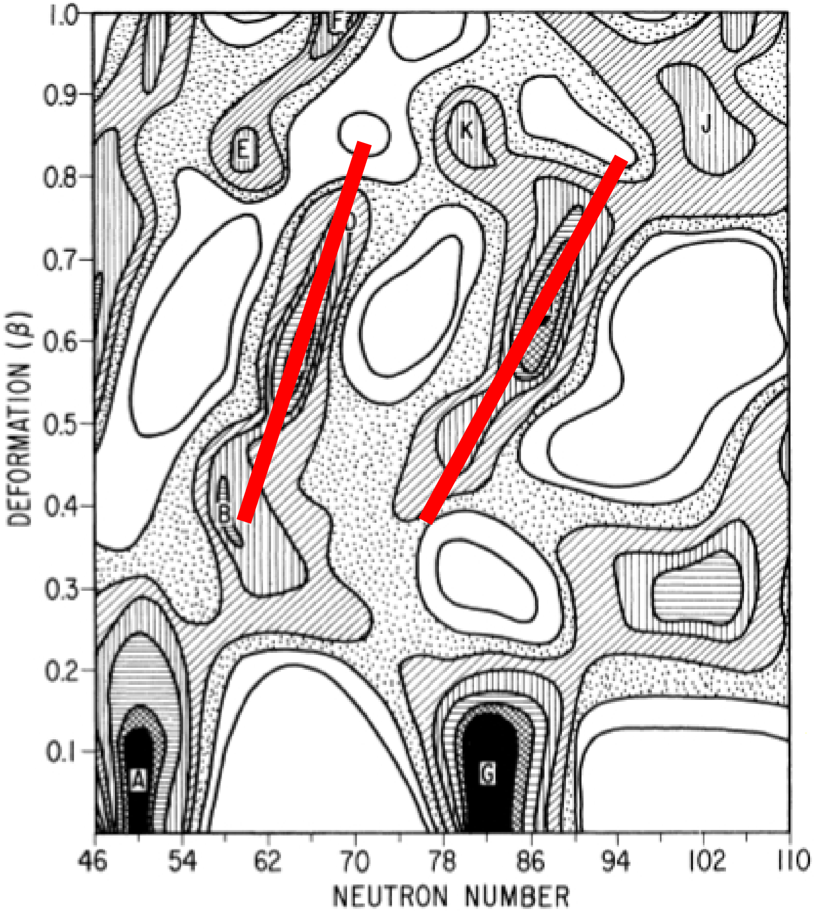




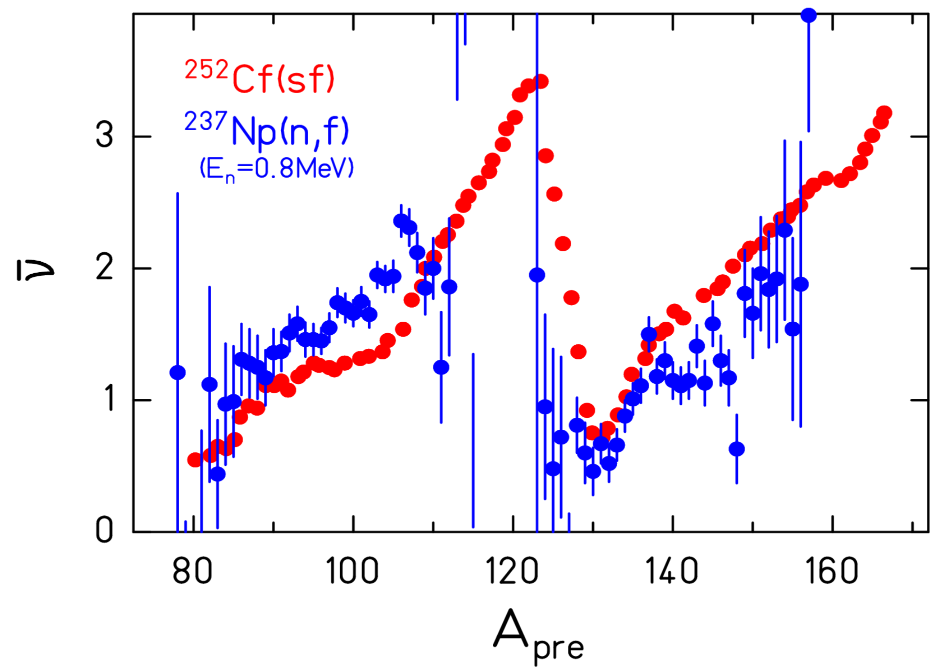
The properties of the fragment shells (position etc) are extracted from measured FF Z or A distributions.

(Böckstiegel et al. 2008))

Shapes of the fragment shells



Wilkins et al., Phys. Rev. C 14 (1976) 1832



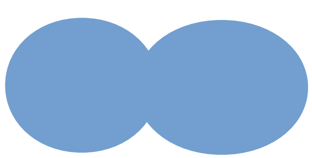
Naqvi et al, 1986 / Zeynalova et al., 2012

General systematics of deformed shells:
Correlation particle number ↔ deformation
(Additional influence of mac. potential.)

Saw-tooth behaviour reflects fragment deformation at scission.

Statistical mechanics

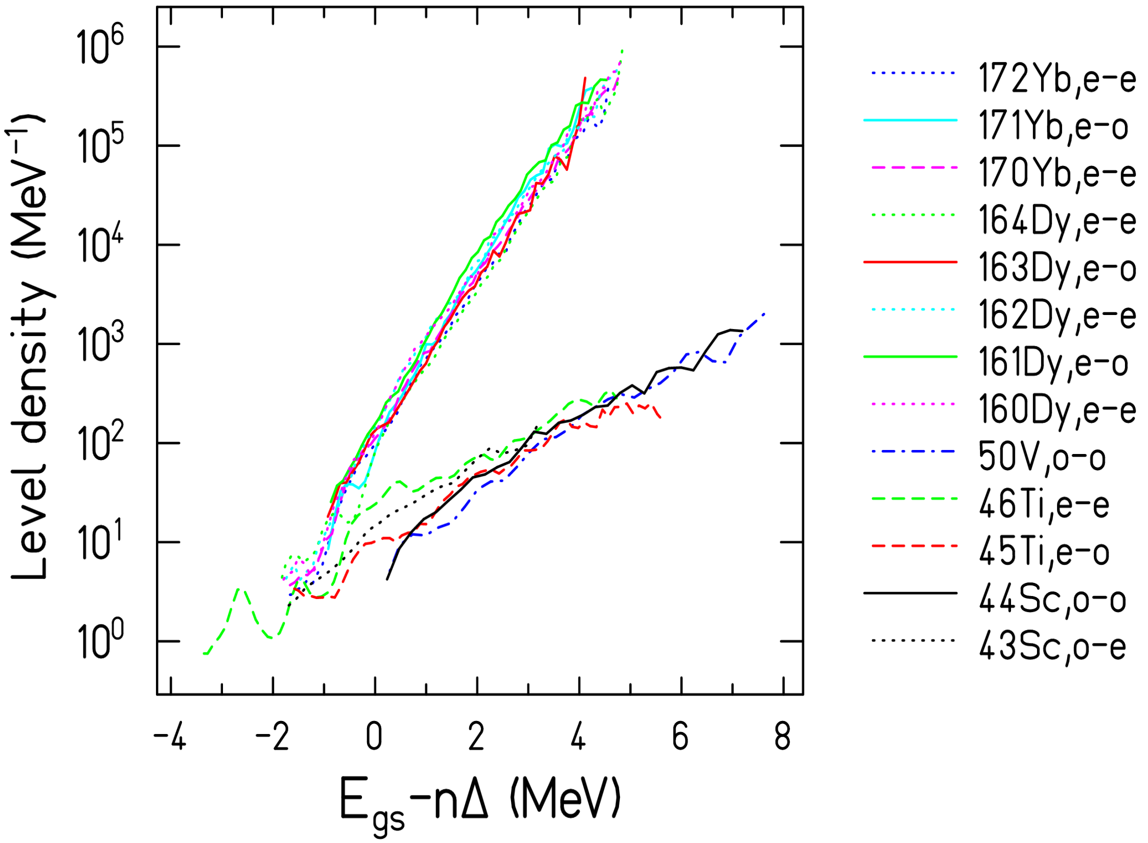
- The importance of fragment shells beyond the second barrier implies also that the two fragments get their own thermodynamical properties.
- The system well before scission consists of two nuclei with their own temperatures.
- Statistical mechanics requires that the two temperatures tend to equilibrate.

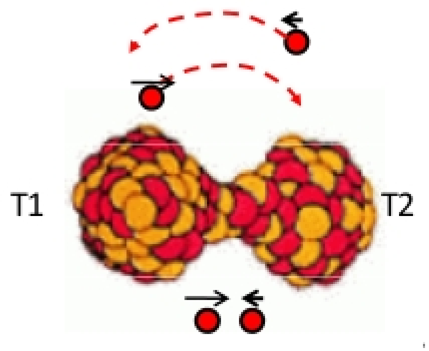


T1 > T2

In the superfluid regime ($E < 10$ MeV): $T \sim A^{-2/3}$ (Independent from E*!)
Two thermostats in contact.

New results on level densities demonstrate constant-temperature behaviour





Nascent fragments:

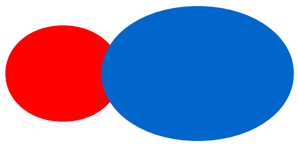
Two thermostats in contact.
→ Energy sorting

Guttormsen et al. 2013

Schmidt, Jurado,
PRL 104 (2010) 212501

Constant nuclear temperature at low E*.

Energy sorting



$T \sim A^{-2/3}$

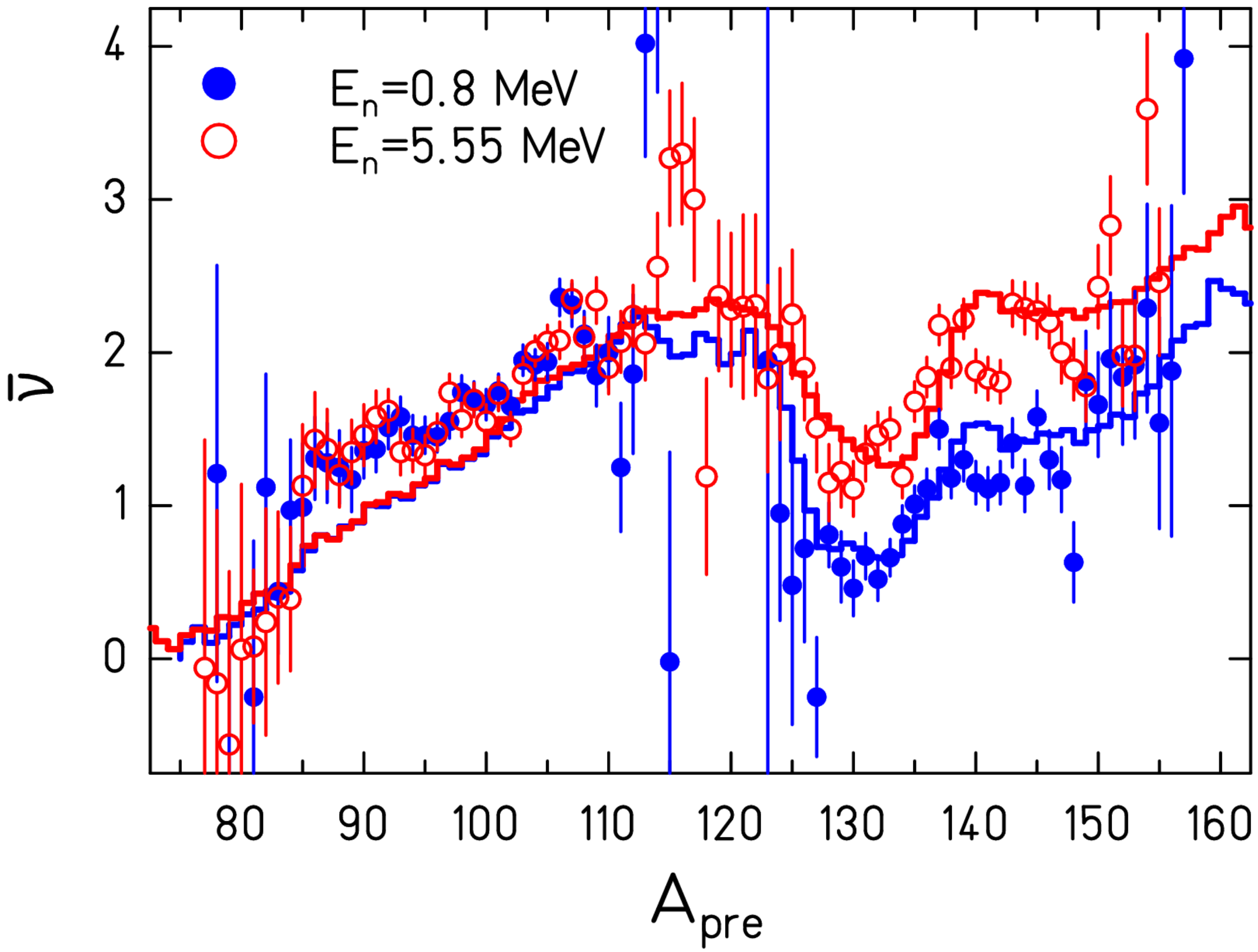
T1 > T2

Second law of Thermodynamics:

Heat (E*) flows from the hotter to the colder object (nascent fragment), until all E* has moved to the heavy fragment.

Important consequences for the prompt-neutron multiplicities:

Energy sorting: experimental evidence

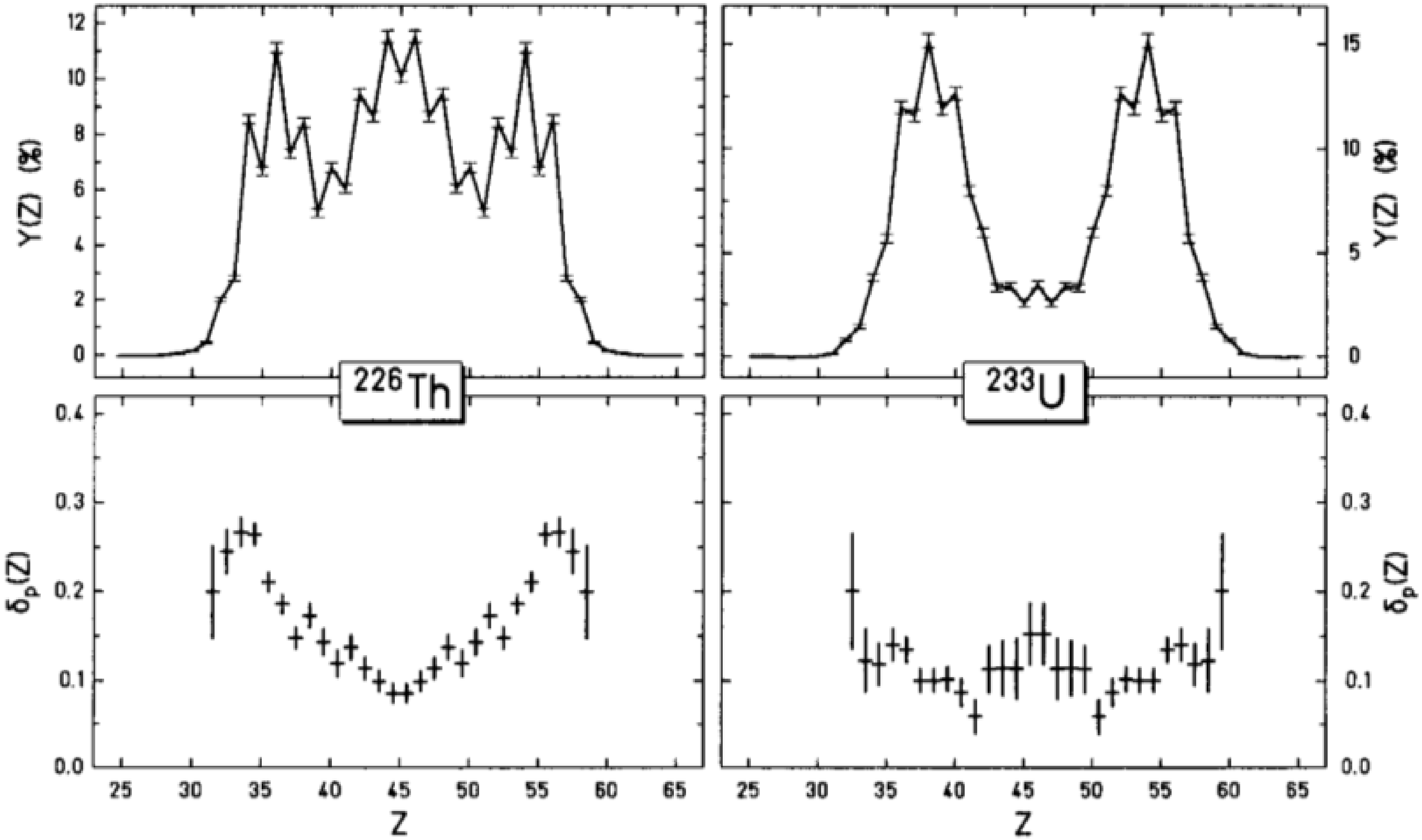


Data points: Naqvi 1986

Lines: GEF

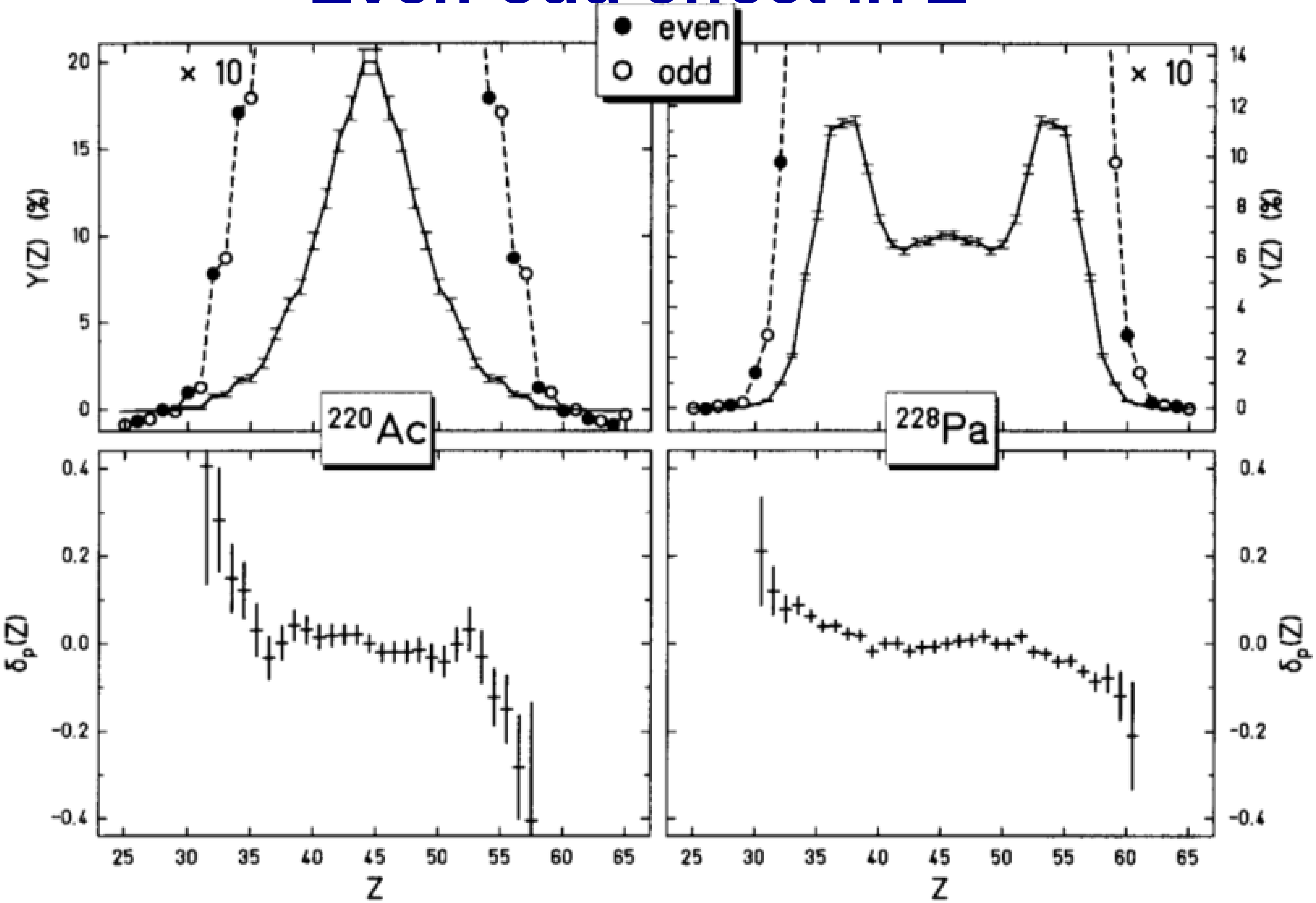
Energy increase by about 5 MeV → Increased prompt-neutron yield in the heavy fragment, only

Even-odd effect in Z



Even-Z fissioning nuclei: There is a chance that the system separates with the protons fully paired. The effect is enhanced at asymmetry. (Steinhäuser et al., 1998)

Even-odd effect in Z



Odd-Z fissioning nuclei: There is a chance that the light fragment has fully paired protons after scission (Steinhäuser et al. 1998). Consistent with the asymmetry-enhanced even-odd effect for even-Z systems.

Impact on the fission yields

Increasing E*:

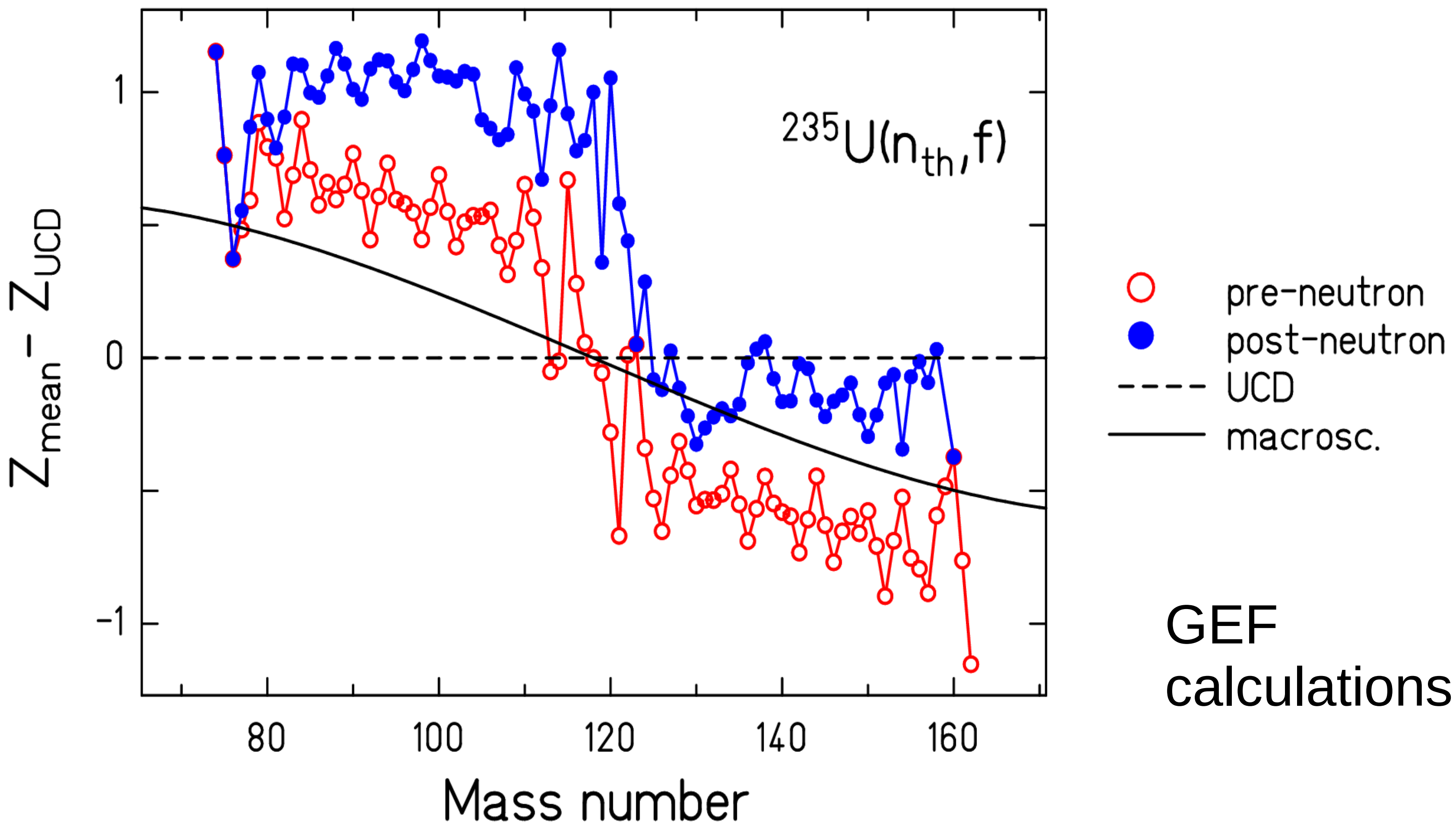
- Shift to neutron-deficient isotopes in the heavy fragment $^{+}$)
- No change in the isotopic distribution in the light fragment^{+})

$^{+}$) unpublished results of the SOFIA experiment

- Even-odd effect in Z governed by the light fragment (because it has the lower E*)

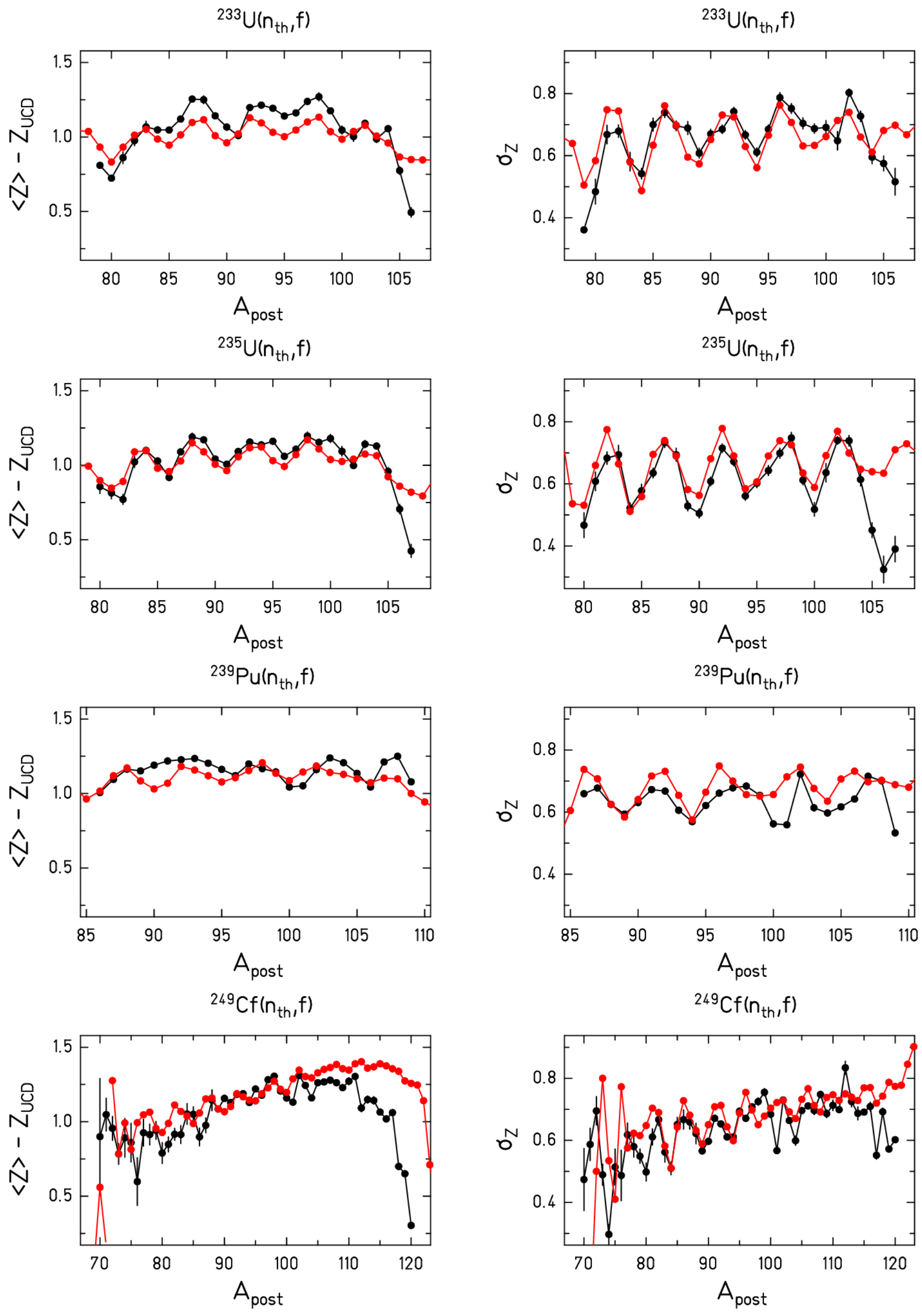
These features are ignored in most estimations and applications of nuclear data.

Influence of neutron emission



GEF calculations

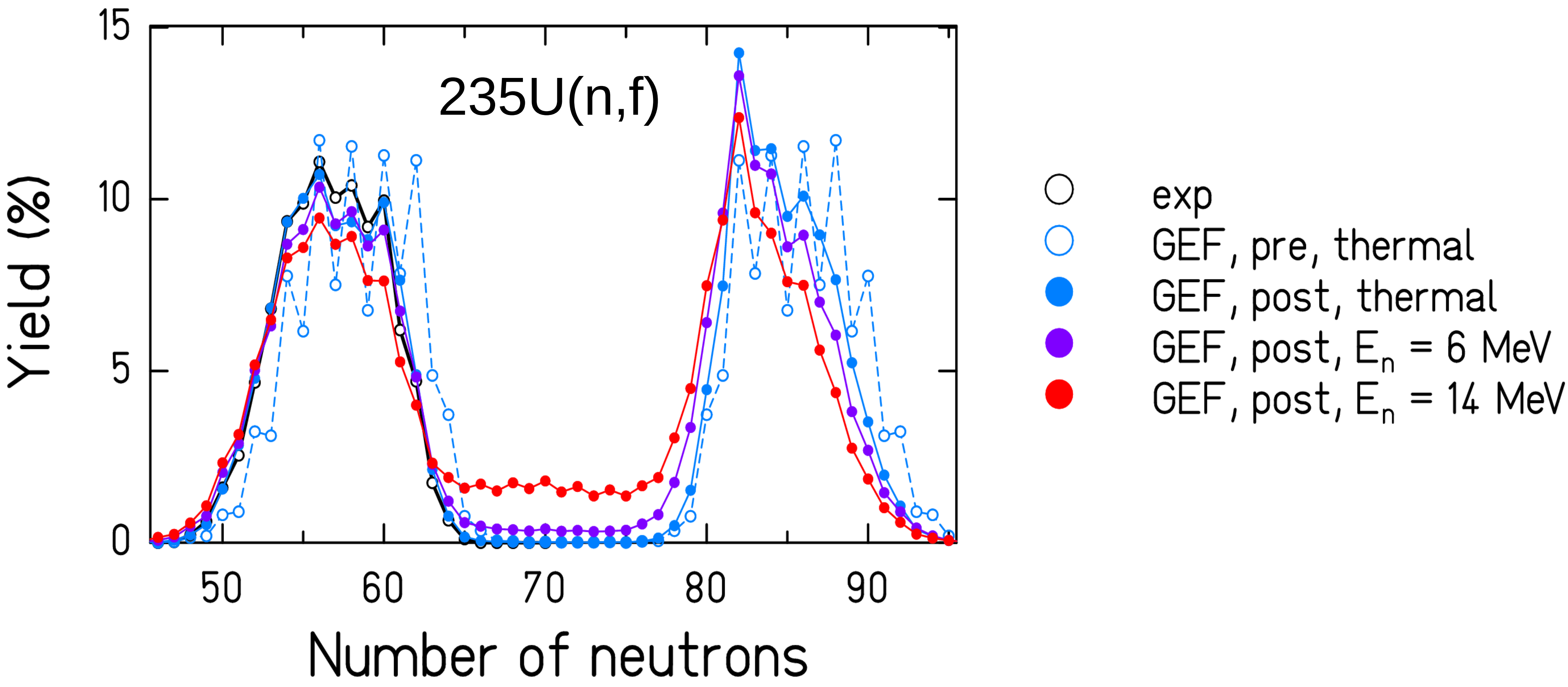
Isobaric sequences



Influence of charge polarization at scission and prompt-neutron emission.

Data compared with **GEF calculations**

Fine structure in FF neutron number



Even-odd effect in neutron number of fragments (post-neutron) is created by evaporation. (Does not depend on E*!)

By influence of pairing on binding energies and level densities:
M. V. Ricciardi et al., Nucl. Phys. A 733 (2004) 299

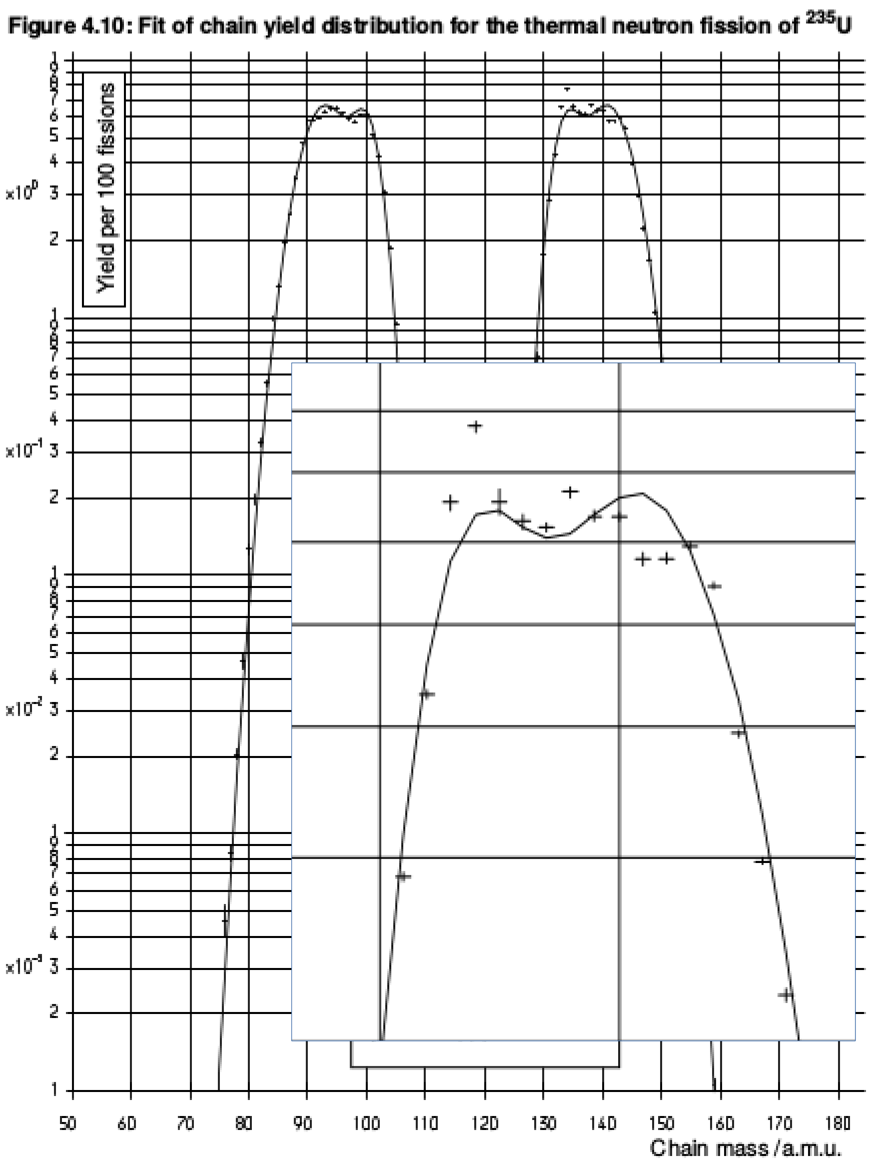
Characteristics of neutron emission

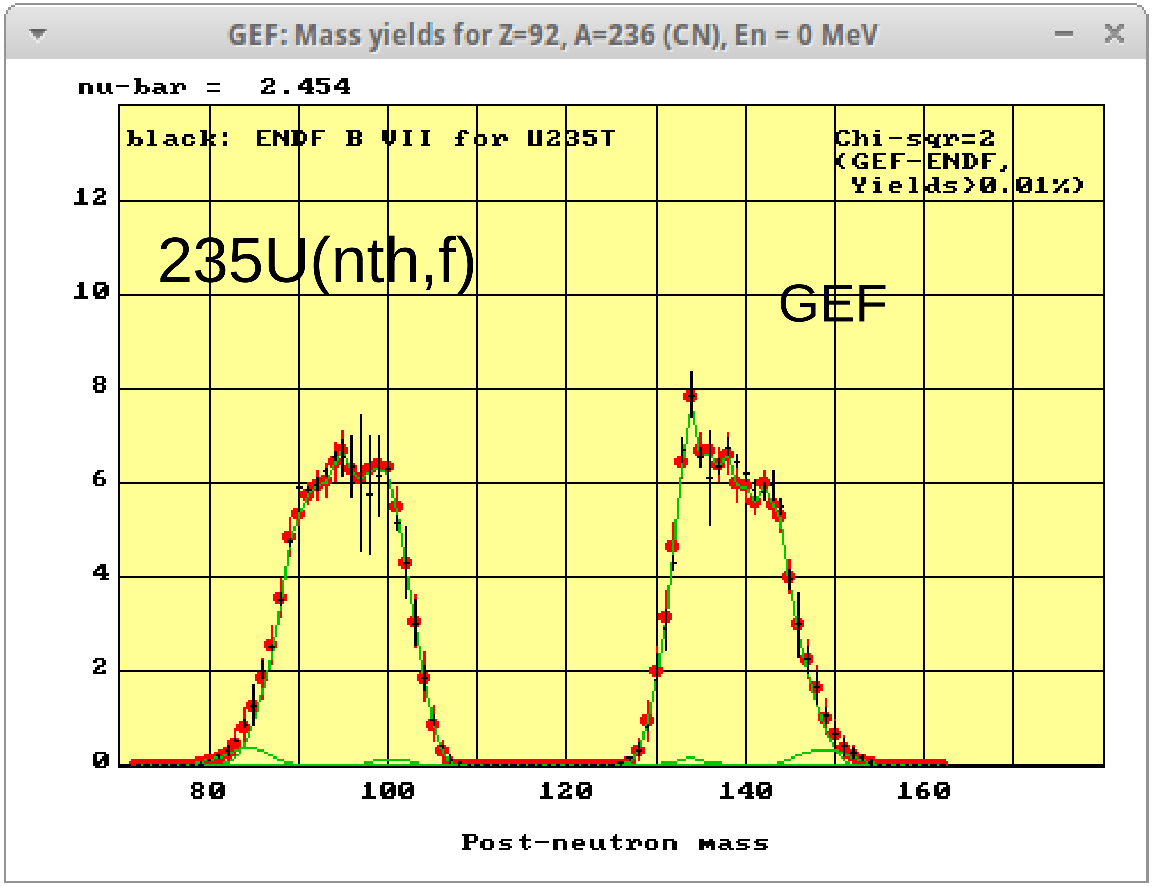
		GEF	Exp.	GEF	Exp.	GEF	Exp.
System	En/MeV	<E>/MeV	<E>/MeV	ν_prompt	ν_prompt	ν_delayed	ν_delayed
233U(n,f)	thermal	2.02(1)	2.030(13)	2.36(1)	2.4884(40)	0.77(9) %	0.74(4) %
	5	2.06(1)		3.10(2)		0.79(16) %	
235U(n,f)	thermal	2.00(1)	2.000(10)	2.42(2)	2.4169(36)	1.60(10) %	1.62(8) %
	5	2.06(1)		3.18(2)		1.48(12) %	
238U(n,f)	5	2.01(1)		3.05(2)		3.51(14) %	
237Np(n,f)	thermal	2.02(1)		2.38(6)	2.52(5)	1.47(7) %	1.07(10) %
	5	2.08(1)		3.12(2)		1.05(5) %	
238Np(n,f)	thermal	2.02(1)		2.57(6)	2.77(5)	1.82(15) %	
	5	2.09(1)		3.36(3)		1.40(7) %	
239Pu(n,f)	thermal	2.08(1)	2.073(10)	2.80(4)	2.876(5)	0.68(4) %	0.650(30)%
	5	2.13(1)		3.57(5)		0.61(3) %	
241Pu(n,f)	thermal	2.06(1)		2.88(5)	2.931(6)	1.42(5) %	1.57(15) %
	5	2.12(2)		3.70(4)		1.16(5) %	
241Am(n,f)	thermal	2.87(2)				0.58(6) %	0.44(5) %
252Cf(sf)	------	2.16(2)		3.76(2)	3.759(5)	0.76(12)%	0.86(10)%

GEF uncertainties only from fission. / “exp” from Mills thesis, 1995; WPEG6; Waldo; Capote
Generally good agreement, GEF uncertainties comparable with exp. / larger deviations in red

235U(nth,f)

Structure effects



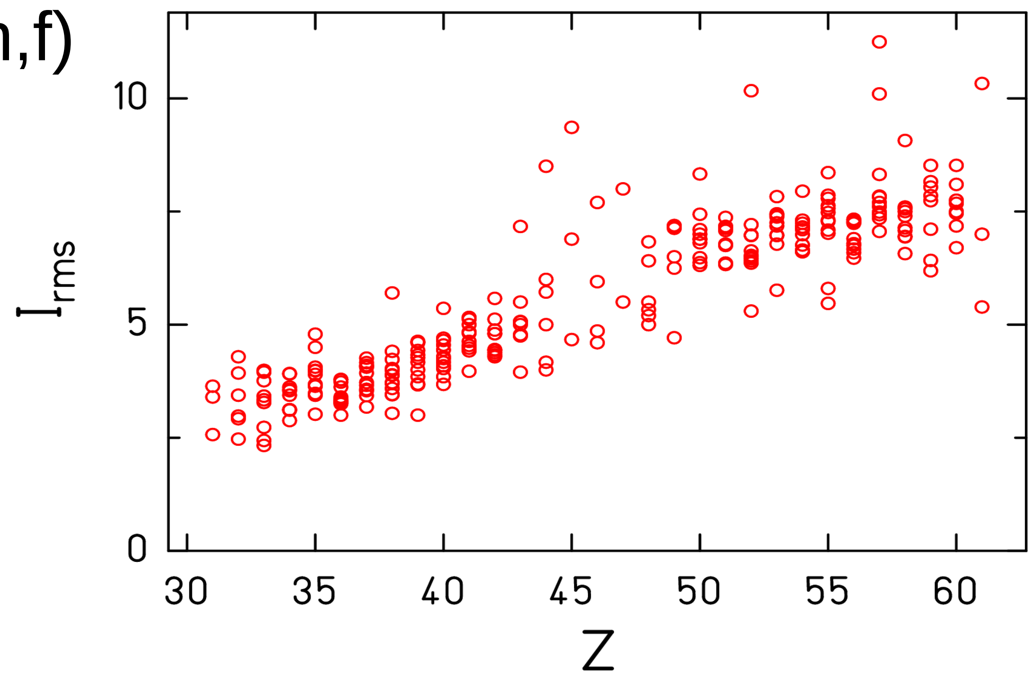


5-Gaussian model (Mills, 1995)

Fine structure in mass distribution is well reproduced by GEF. (New Fit. Strong influence of neutron evaporation. Experimental masses are important!)

Fragment angular momentum

235U(nth,f)



GEF calculations

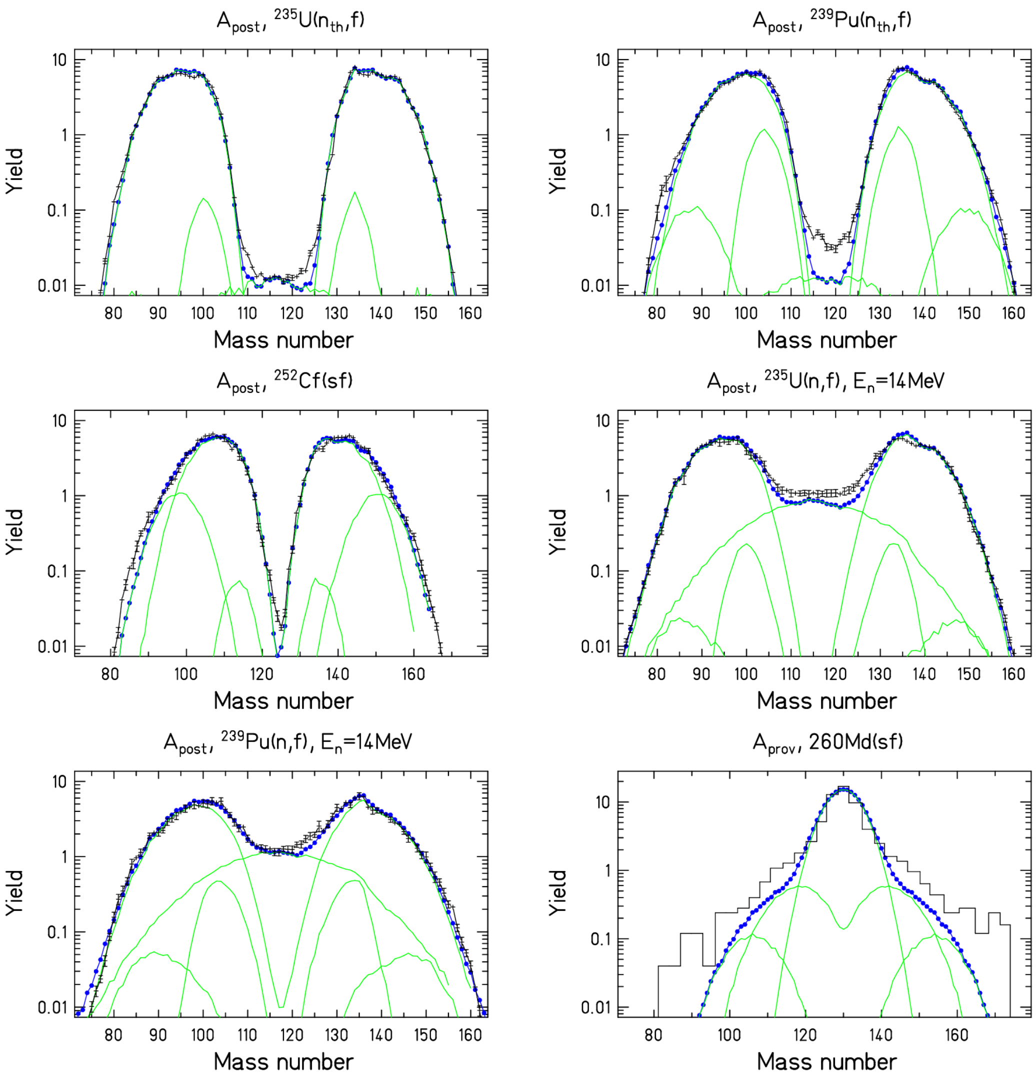
in good agreement with measured **isomeric ratios**

Description with an effective temperature + I of unpaired nucleons.
(The theoretical idea: “Pumping” from q.m. uncertainty of orbital angular momentum (Kadmensky) appears to be more convincing, but a quantitative model is not available.)

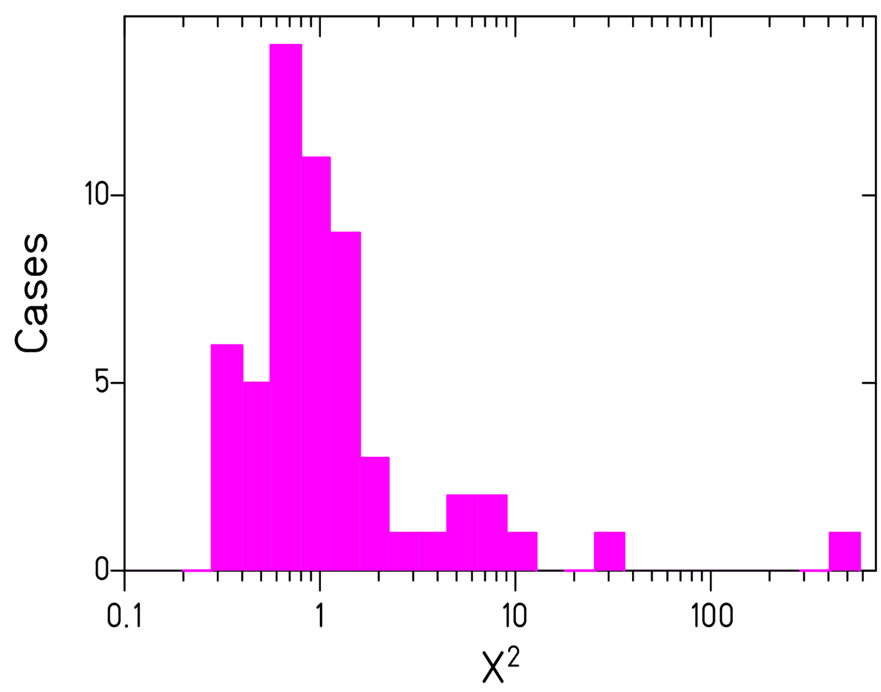
Fragment angular momentum

- stores collective energy at scission (less TKE)
- feeds contributions of rotational transitions to prompt gamma spectrum

Quality of mass yields from GEF



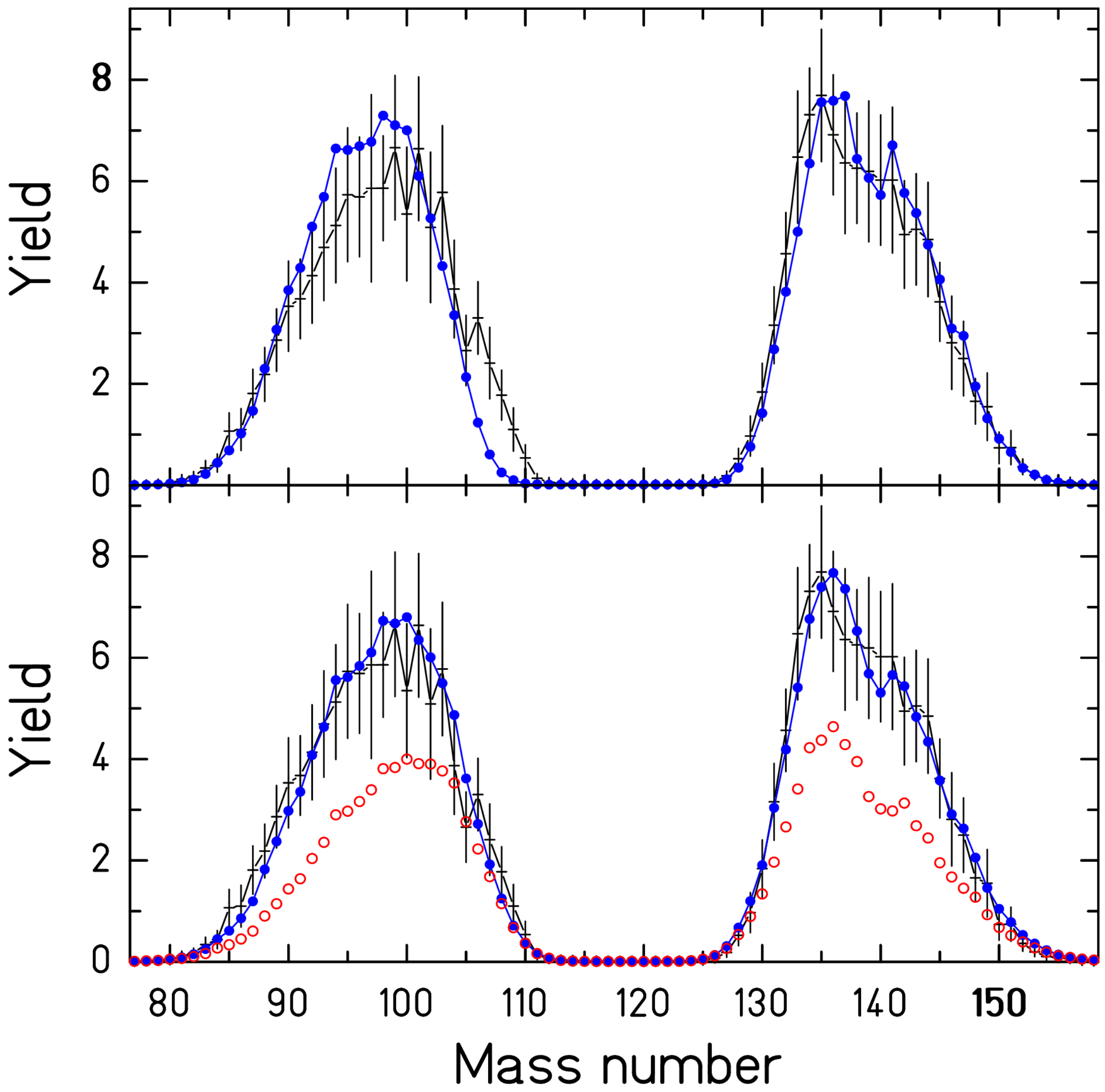
Chi-squared deviations per system



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

Almost all large deviations caused by erroneous evaluation (evidenced by GEF)!

237Np(nth,f), the contributions



Black: Evaluation ENDF/B-VII

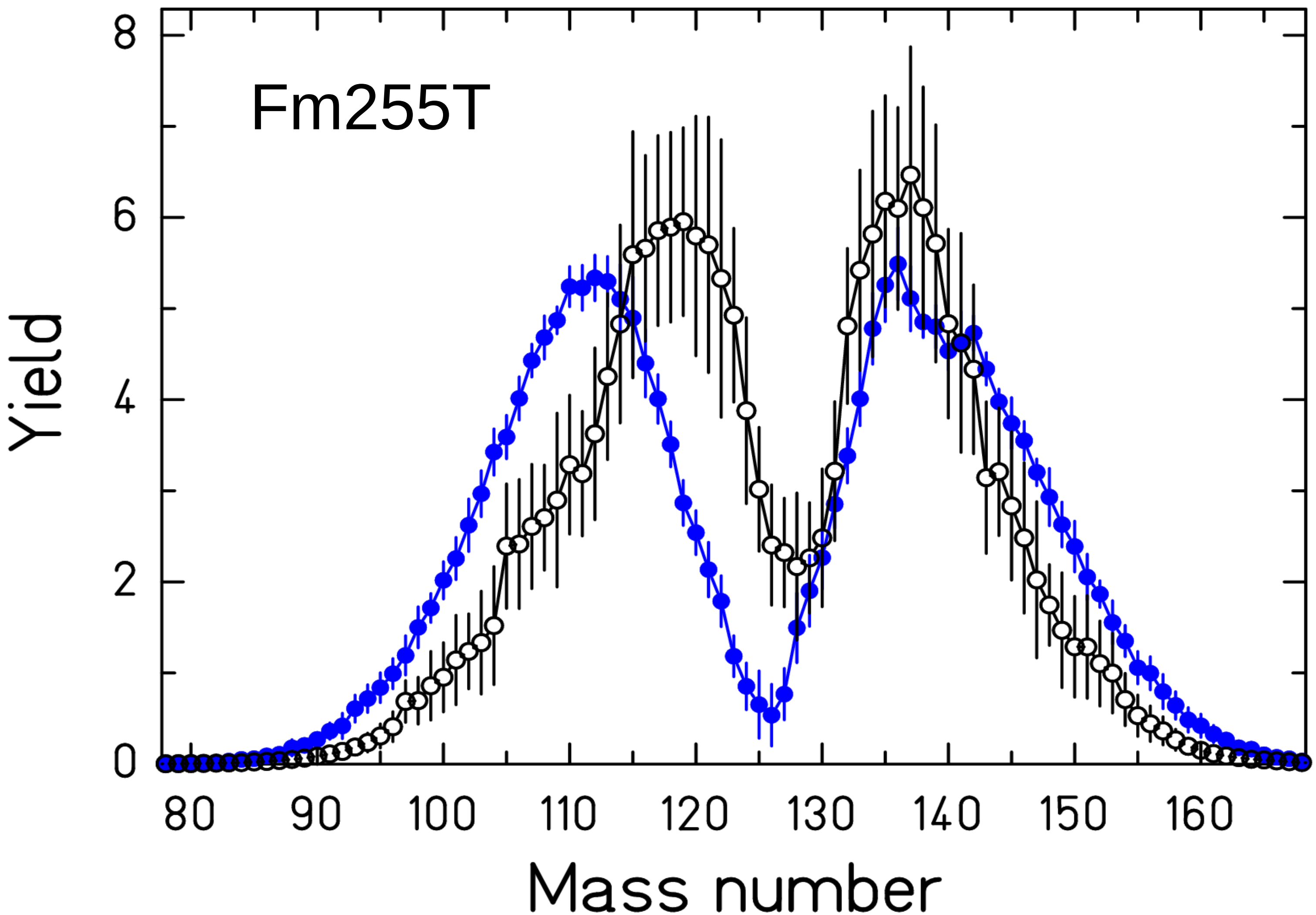
Blue: GEF (pure 237Np)

Blue: GEF (total)

Red: GEF (Contribution from 239Pu)

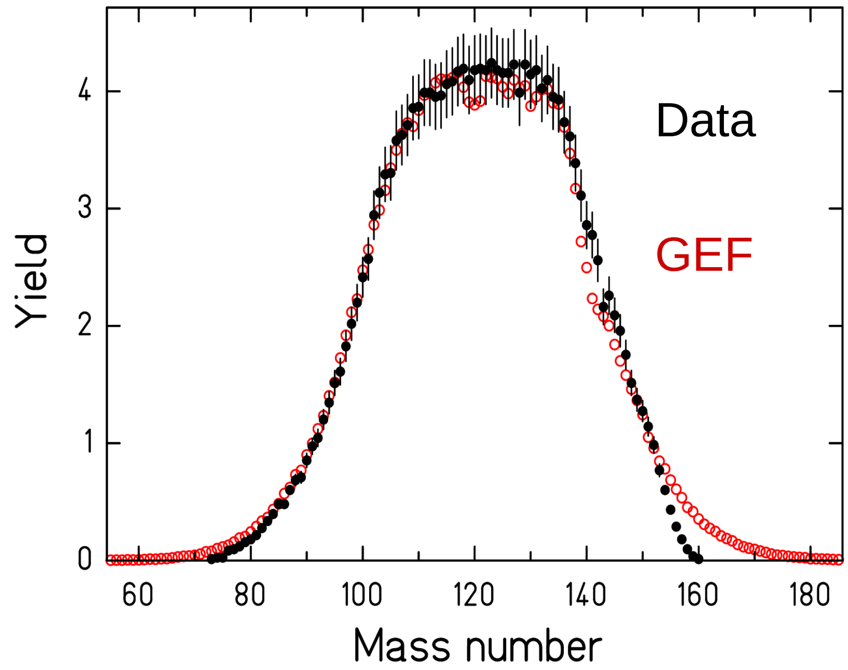
Also noticeable in the prompt-fission multiplicity.

Other erroneous data in ENDF/B-VII

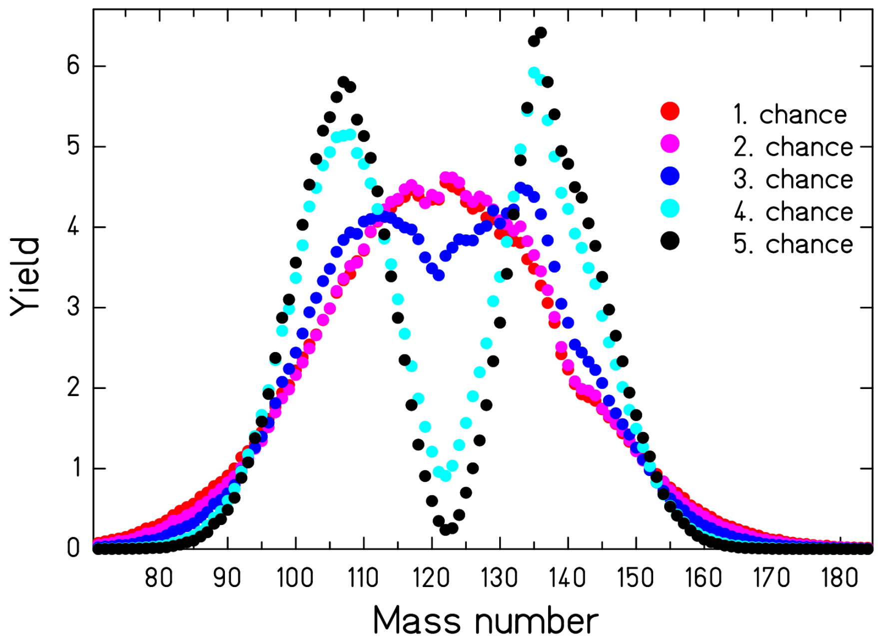


Evaluation / **GEF**

Multi-chance fission



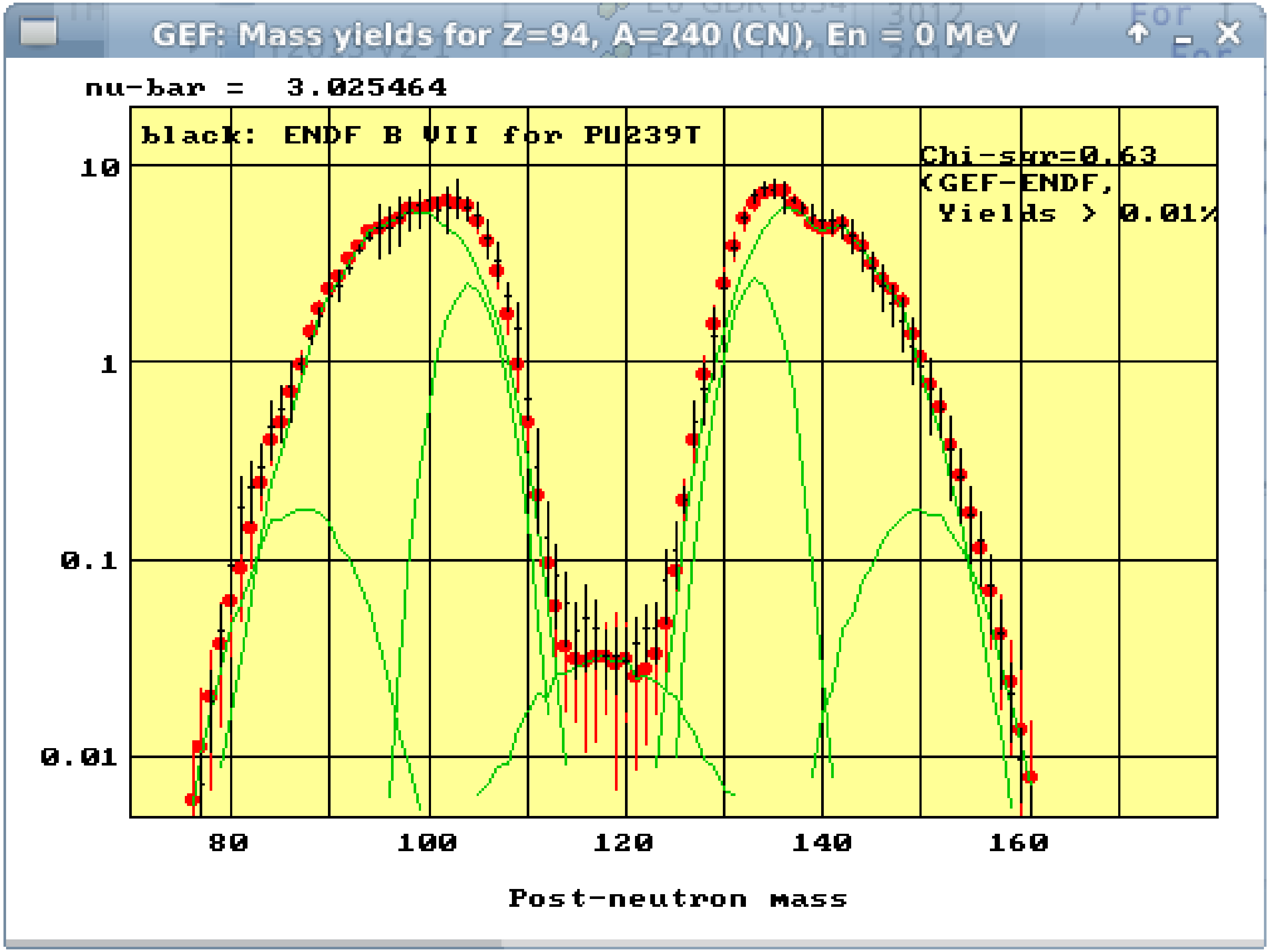
250Cf, E* = 45 MeV
VAMOS experiment



GEF: Contribution of fission chances

GEF: The final FF distribution is the sum of the different fission chances.

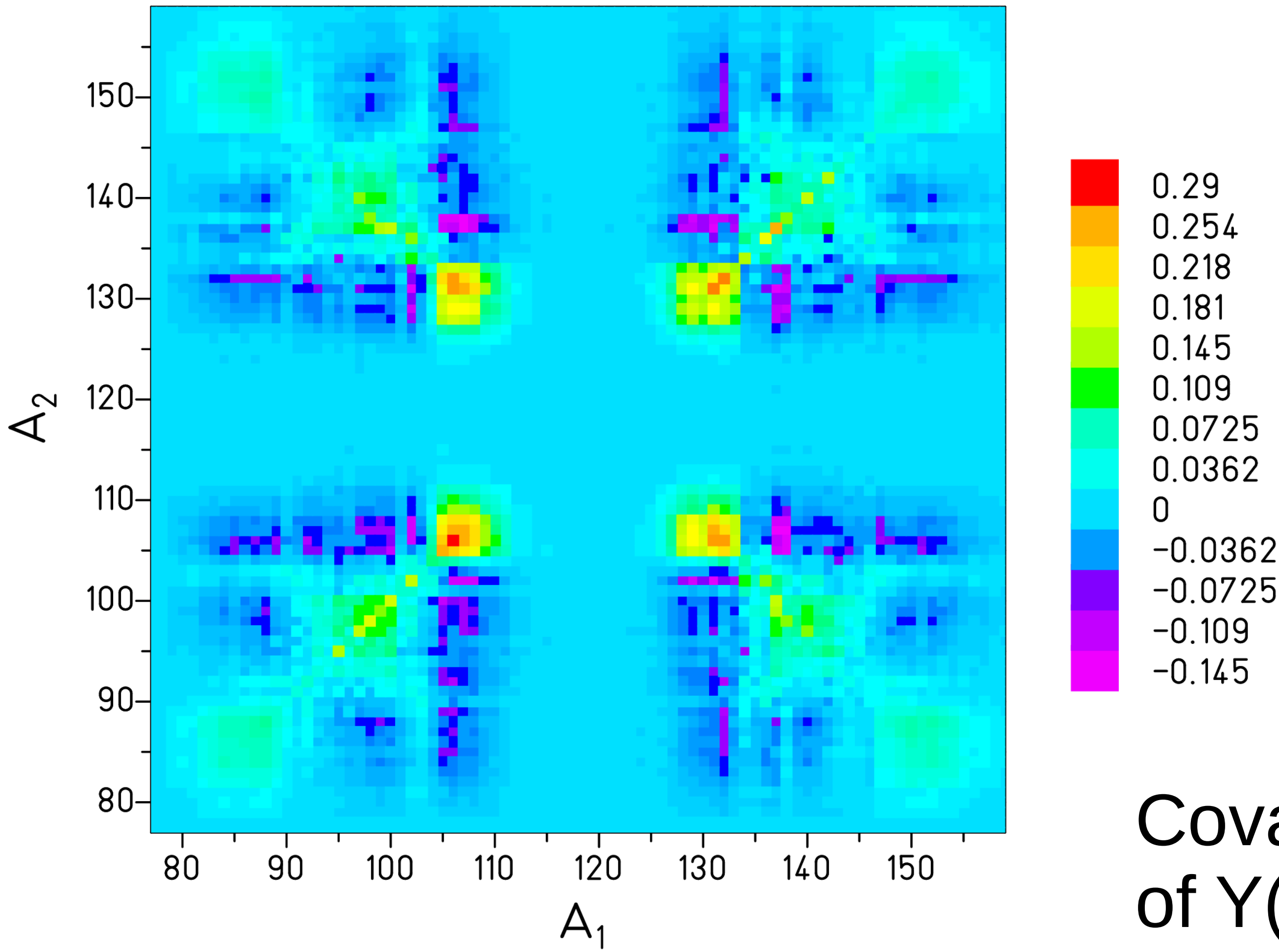
Uncertainties of the model



Mass yields from GEF with estimated uncertainties.

GEF calculations with perturbed parameters.

Covariances from GEF

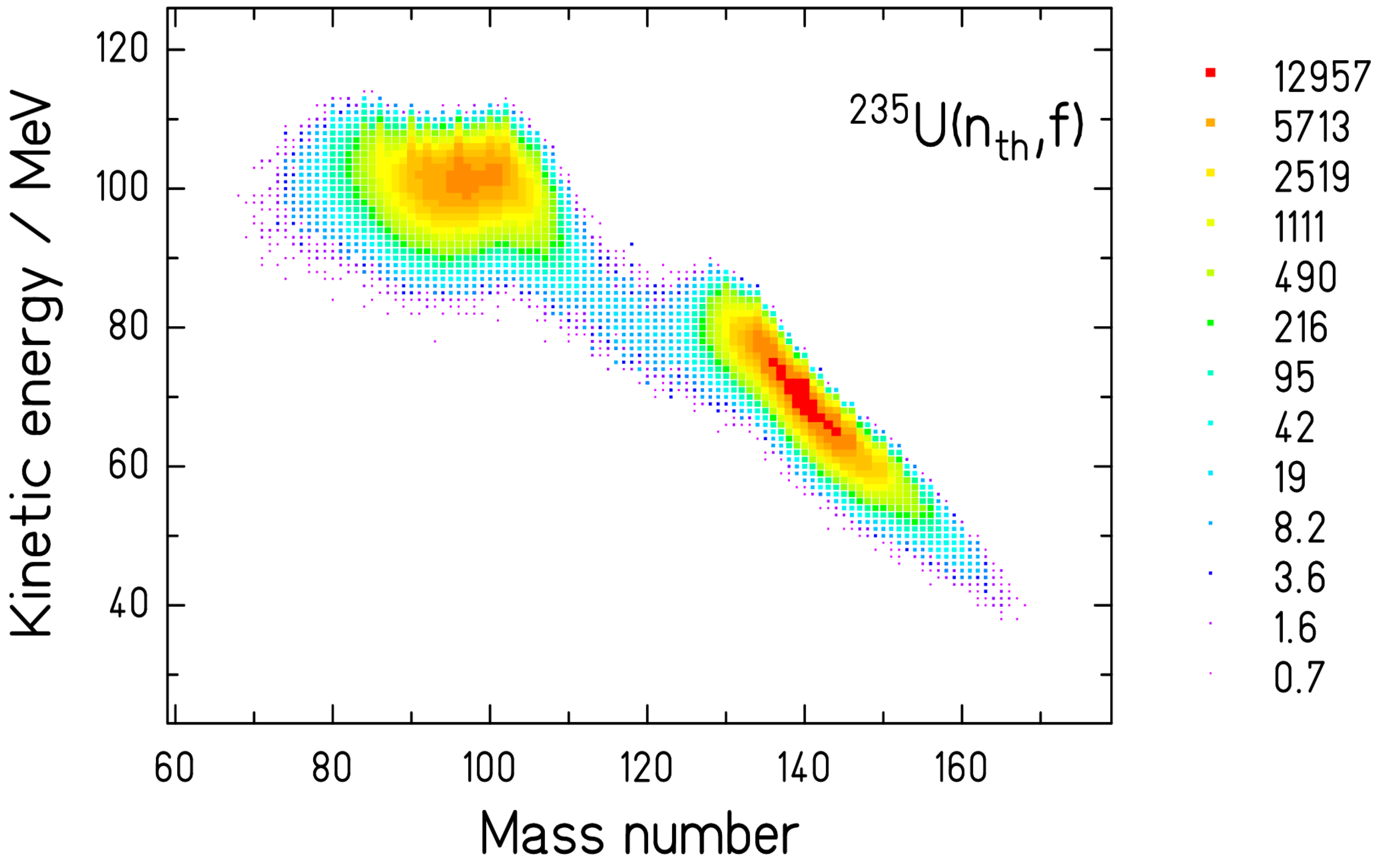


239Pu(nth,f)

Covariance matrix of Y(A) from GEF.

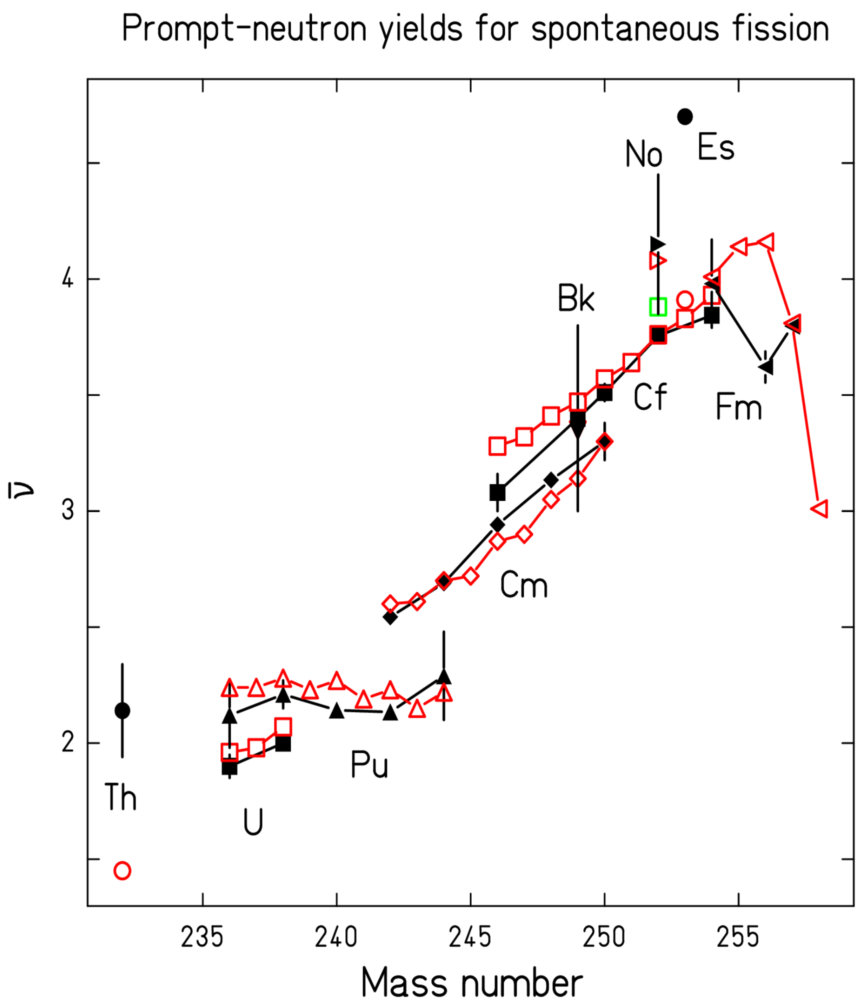
Covariances available for any pair of fission observables or between different systems

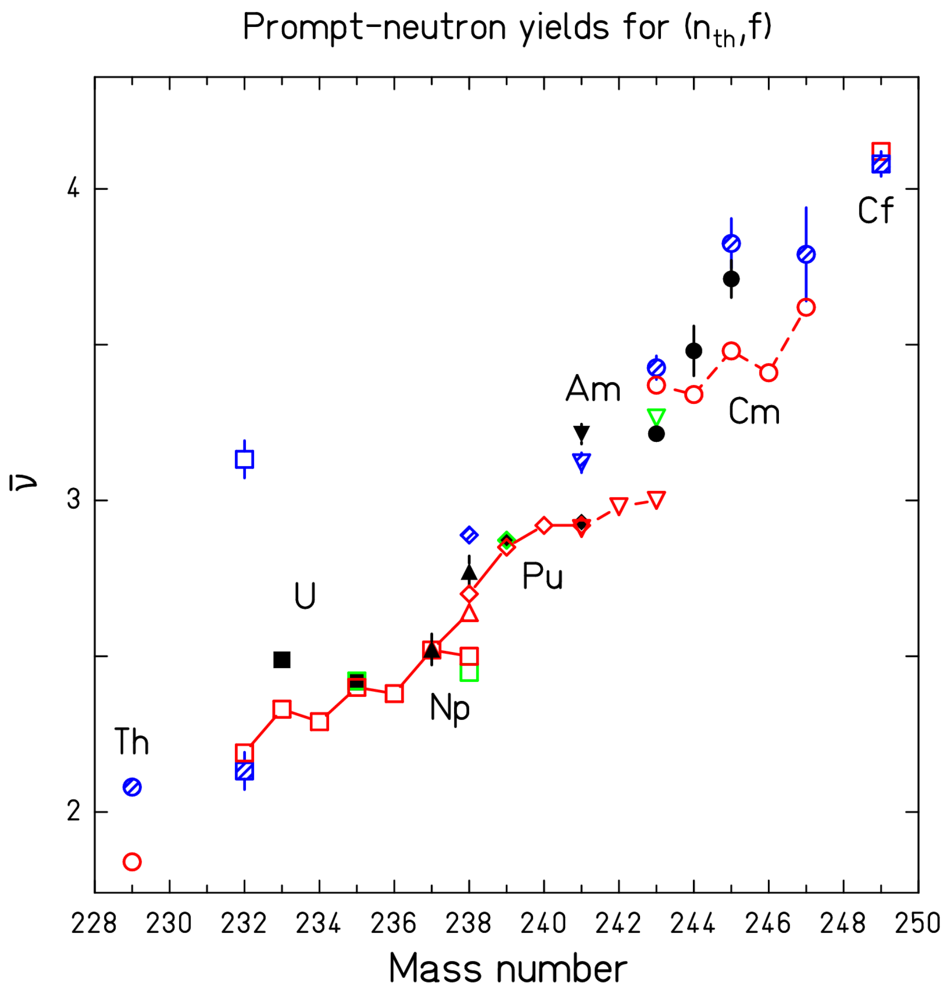
GEF: A - TKE



TKE follows from Q value and energy conservation.

Total prompt-neutron multiplicities



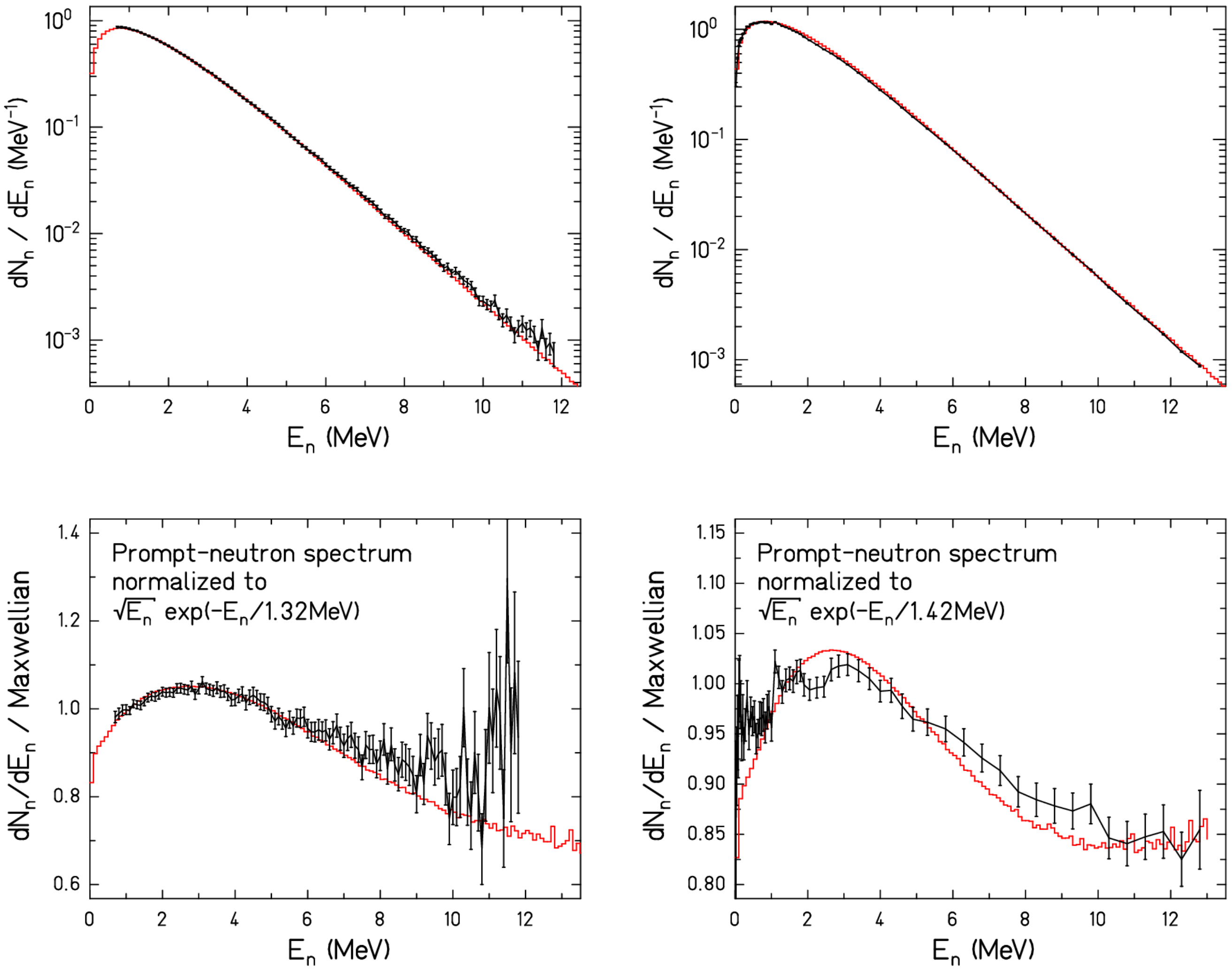


rms deviation: 0.1 units

rms deviation: 0.2 units

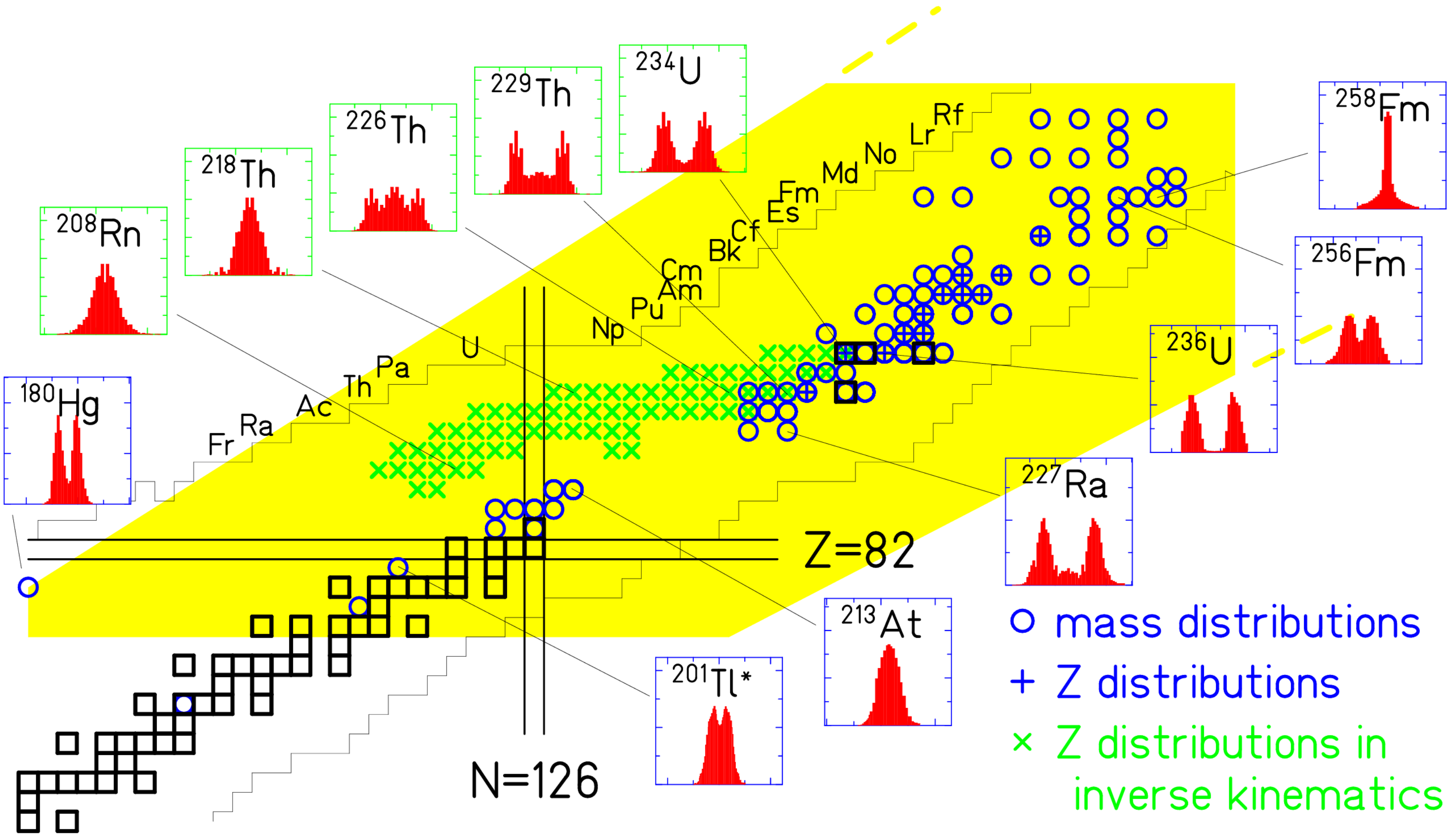
(experimental problems?)

Energy spectra of prompt neutrons



Clue: Modified composite Gilbert-Cameron nuclear level density.
(Increased condensation energy, collective enhancement)
K.-H. Schmidt, B. Jurado, Phys. Rev. C 86 (2012) 044322

Application range of GEF



All systems (spontaneous fission up to E*=100 MeV) with a unique parameter set, ≈ 30 to 50 parameters relevant for FY.

Theoretical ideas exploited in GEF

- "Nuclear properties according to the Thomas-Fermi model" (Topographic theorem), W. D. Myers, W. J. Swiatecki, Nucl. Phys. A 601 (1996) 141
- "Structure of the potential energy surface at large deformations" (Early manifestation of fragment shells), U. Mosel, D. Scharnweber, Phys. Rev. Lett. 25 (1970) 678
- "Theory of macroscopic fission dynamics" (Dynamical freezing), G. D. Adeev, V. V. Pashkevich, Nucl. Phys. A 502 (1989) 405

Specific theoretical developments for GEF

- "Assessment of saddle-point-mass predictions for astrophysical applications", A Kelic, K.-H. Schmidt, Phys. Lett. B 634 (2006) 362
- "On the topographical properties of fission barriers", A. V. Karpov, A. Kelic, K.-H. Schmidt, J. Phys. G: Nucl. Part. Phys. 35 (2008) 035104
- "Experimental evidence for the separability of compound-nucleus and fragment properties in fission", K.-H. Schmidt, A. Kelic, M. V. Ricciardi, Europh. Lett. 83 (2008) 32001
- "Entropy-driven excitation-energy sorting in superfluid fission dynamics", K.-H. Schmidt, B. Jurado, Phys. Rev. Lett. 104 (2010) 212501
- "Thermodynamics of nuclei in thermal contact", K.-H. Schmidt, B. Jurado, Phys. Rev. C 83 (2011) 014607
- "Final excitation energy of fission fragments", K.-H. Schmidt, B. Jurado, Phys. Rev. C 83 (2011) 061601(R)
- "Inconsistencies in the description of pairing effects in nuclear level densities", K.-H. Schmidt, B. Jurado, Phys. Rev. C 86 (2012) 044322
- "General laws of quantum and statistical mechanics governing fission", K.-H. Schmidt, B. Jurado, FIAS Interdisciplinary Science Series (2014) 121
- "Influence of complete energy sorting on the characteristics of the odd-even effect in fission-fragment element distributions", B. Jurado, K.-H. Schmidt, J. Phys. G 42 (2015) 055101
- "Revealing hidden regularities with a general approach to fission", K.-H. Schmidt, B. Jurado, Eur. Phys. J. A 51 (2015) 176
- "General description of fission observables: GEF model code", K.-H. Schmidt, B. Jurado, C. Amouroux, C. Schmitt, Nucl. Data Sheets 131 (2016) 107

Summary

- GEF: Description of the fission process on an “intermediate” level with empirical adjustment.
- Solid theoretical frame, efficient and powerful.
- High precision, good predictive power over a large range of nuclei.
- Suited to detect erroneous data.
- Freely available code (10^6 events in ≈ 1 minute).
- Covariances, ENDF tables of FY, random files provided.
- See more complete presentation of the GEF code in: "General description of fission observables: GEF model code", K.-H. Schmidt, B. Jurado, C. Amouroux, C. Schmitt, Nucl. Data Sheets 131 (2016) 107.

Other references

- "Induced fission of 240Pu within a real-time microscopic framework"
  A. Bulgac, P. Magierski, K. J. Roche, I. Stetcu, Phys. Rev. Lett. 116 (2016) 122504
- "Microscopic phase-space exploration modeling of 258Fm spontaneous fission"
  Yusuke Tanimura, Denis Lacroix, Sakir Ayik, Phys. Rev. Lett. 118 (2017) 152501
- "Microscopic approach of fission dynamics applied to fragment kinetic energy and mass distributions in 238U"
  H. Goutte, J. F. Berger, P. Casoli, D. Gogny, Phys. Rev. C 71 (2005) 024316
- "Fission fragment charge and mass distributions in Pu239(n,f) in the adiabatic nuclear energy density functional"
  D. Regnier, N. Dubray, N. Schunck, M. Verriere, Phys. Rev. C 93 (2016) 054611
- "Langevin model of low-energy fission"
  A. J. Sierk, Phys. Rev. C 96 (2017) 034603
- "A new set of parameters for 5 Gaussian fission yield systematics"
  J. Katakura, Proc. ICNC (2003) 143
- "Nuclear properties according to the Thomas-Fermi model"
  W. D. Myers, W. J. Swiatecki, Nucl. Phys. A 601 (1996) 141
- "Heavy-element fission barriers"
  Peter Moeller, Arnold J. Sierk, Takatoshi Ichikawa, Akira Iwamoto, Ragnar Bengtsson, Henrik Uhrenholt, Sven Aberg, Phys. Rev. C 79 (2009) 064304
- "The double-humped fission barrier"
  S. Bjoernholm, J. E. Lynn, Rev. Mod. Phys. 52 (1980) 725
- "On the topographical properties of fission barriers"
  A. V. Karpov, A. Kelic, K.-H. Schmidt, J. Phys. G: Nucl. Part. Phys. 35 (2008) 035104
- "Independent fission yields studied based on Langevin approach"
  Y. Aritomo, S. Chiba, K. Nishio, Progr. Nucl. Energy 85 (2015) 568
- "Fragment-shell influences in nuclear fission",
  U. Mosel, H. W. Schmitt, Phys. Rev. C 4 (1971) 2185
- "Nuclear-fission studies with relativistic secondary beams: analysis of fission channels"
  C. Boeckstiegel, S. Steinhaeuser, K.-H. Schmidt, H.-G. Clerc, A. Grewe, A. Heinz, M. de Jong, A. R. Junghans, J. Mueller, B. Voss, Nucl. Phys. A 802 (2008) 12
- "Scission-point model of nuclear fission based on deformed-shell effects"
  B. D. Wilkins, E. P. Steinberg, R. R. Chasman, Phys. Rev. C 14 (1976) 1832
- "Fission fragment properties in fast-neutron-induced fission of 237Np"
  A. A. Naqvi, F. Kaeppeler, F. Dickmann, R. Mueller, Phys. Rev. C 34 (1986) 218
- "Odd-even effects observed in the fission of nuclei with unpaired protons"
  S. Steinhaeuser, J. Benlliure, C. Boeckstiegel, H.-G. Clerc, A. Heinz, A. Grewe, M. de Jong, A. R. Junghans, J. Mueller, M. Pfuetzner, K.-H. Schmidt, Nucl. Phys. A 634 (1998) 89
- "Constant-temperature level densities in the quasicontinuum of Th and U isotopes"
  M. Guttormsen, B. Jurado, J. N. Wilson, M. Aiche, L. A. Bernstein, Q. Ducasse, F. Giacoppo, A. Goergen, F. Gunsing, T. W. Hagen, A. C. Larsen, M. Lebois, B. Leniau, T. Renstroem, S. J. Rose, S. Siem, T. Tornyi, G. M. Tveten, M. Wiedeking, Phys. Rev. C 88 (2013) 024307
- "Complex nuclear-structure phenomena revealed from the nuclide production in fragmentation reactions",
  M. V. Ricciardi, A. V. Ignatyuk, A. Kelic, P. Napolitani, F. Rejmund, K.-H. Schmidt, O. Yordanov, Nucl. Phys. A 733 (2004) 299
- R. W. Mills, PhD thesis, 1995
- "Angular and spin distributions of primary fission fragments"
  S. G. Kadmensky, D. E. Lyubashevsky, L. V. Titova, Bull. Russ. Academy Sciences: Physics 75 (2011) 989