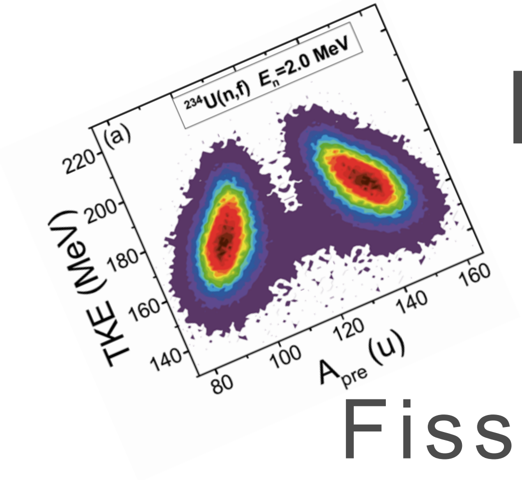
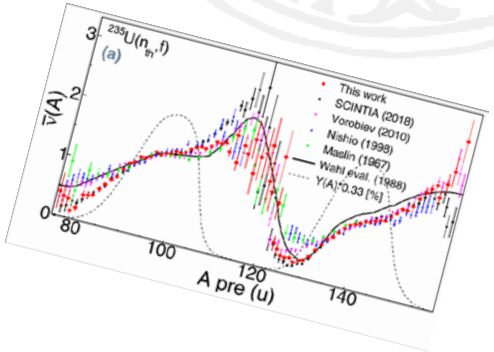


Experimental studies of the fission processes

Fission yields, neutron and γ-ray emission



Stephan Pomp
Department of physics and astronomy
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Overview

Evaluated nuclear data files (used in nuclear technology, astrophysics, medical applications, etc) are based on/guided by model calculations with, e.g., TALYS, GEF, FIFRELIN, etc.
The models are guided by **experimental data**; microscopic measurements and integral experiments.

We now look at measurement techniques for some observables in the fission reaction:

- **Mass yields**
- **Prompt neutron emission**
- **Prompt γ-ray emission**

For these experiments we **need:**

- Facilities providing, e.g., neutron beams
- Targets
- Detector setups (with suitable DAQ, analysis routines, etc.)

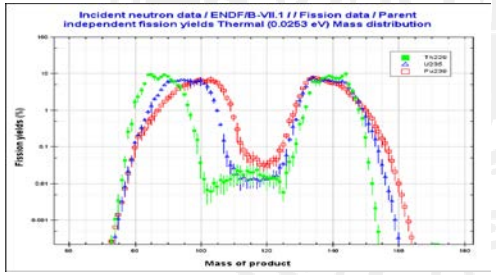
Result: Experimental data points, reported in EXFOR, retrievable with, e.g., JANIS (including uncertainties).

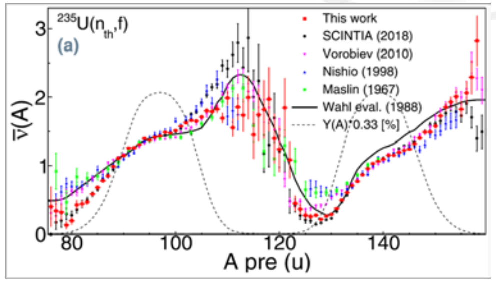
In this talk

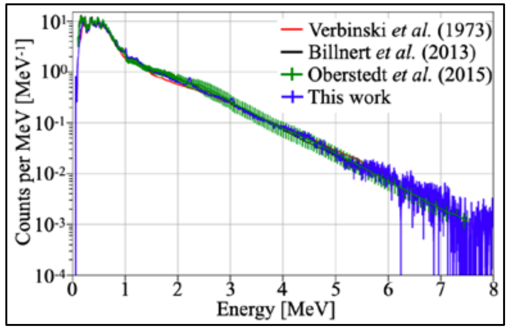
- Recap of fission observables and their interconnection
- Observations and trends and experimental methods regarding
  - Fission yields
  - Prompt neutron emission
  - Prompt γ-ray emission
- Note:

  (1) These observables are linked.

  (2) There are uncertainties and “old truths” sometimes change.







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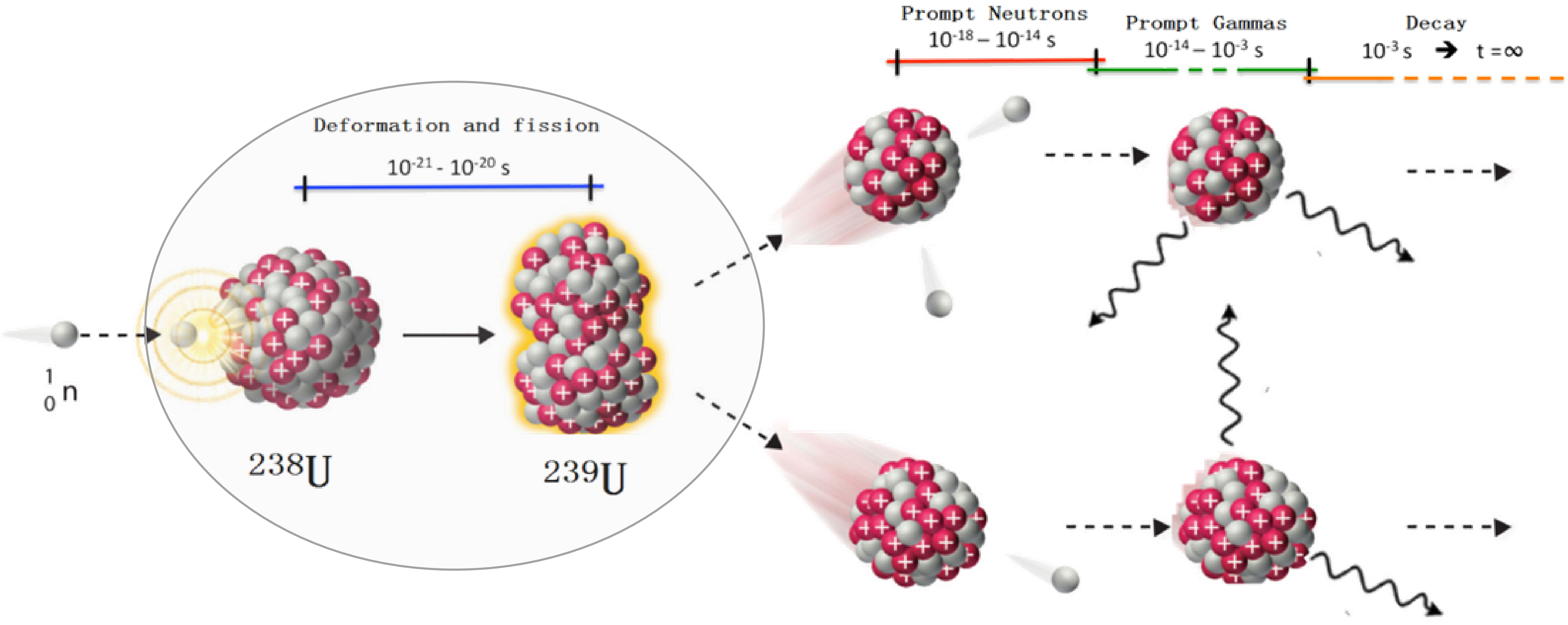
Recap: The fission process and timeline



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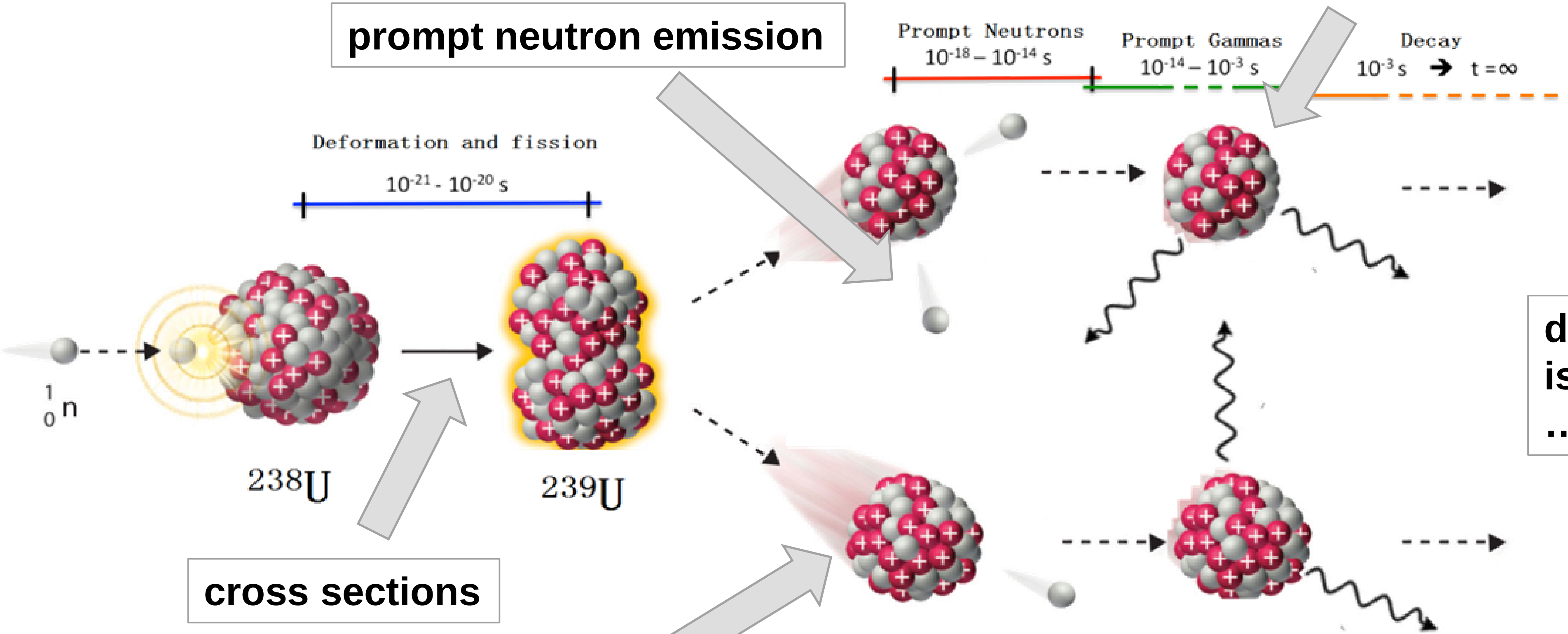
Fission observables



prompt neutron emission

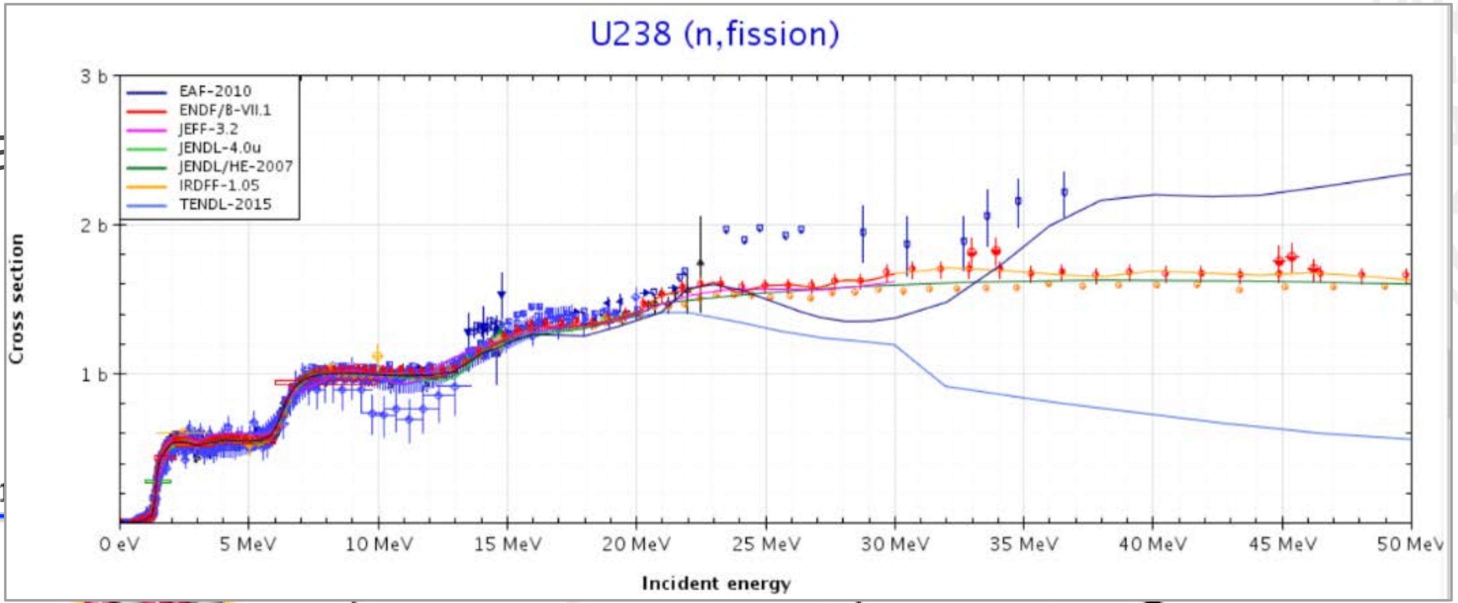
prompt gamma emission

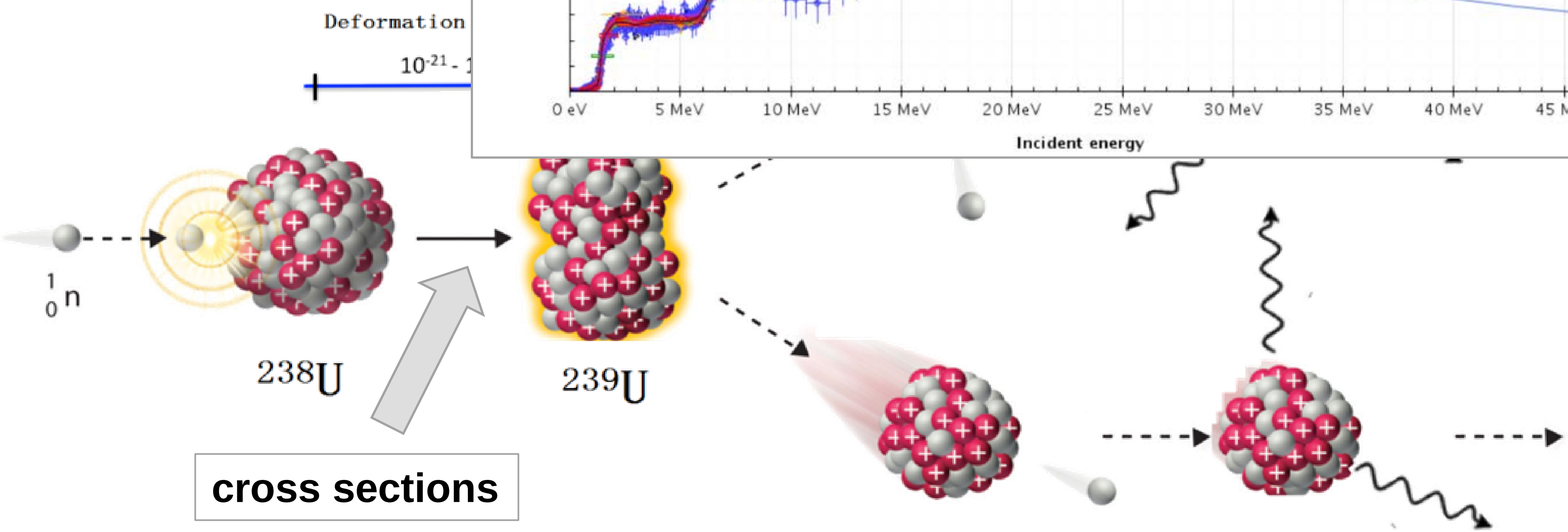
delayed neutrons
isomeric yield ratios
…

cross sections

fission yields
(A,Z,E)

Fission observa





cross sections

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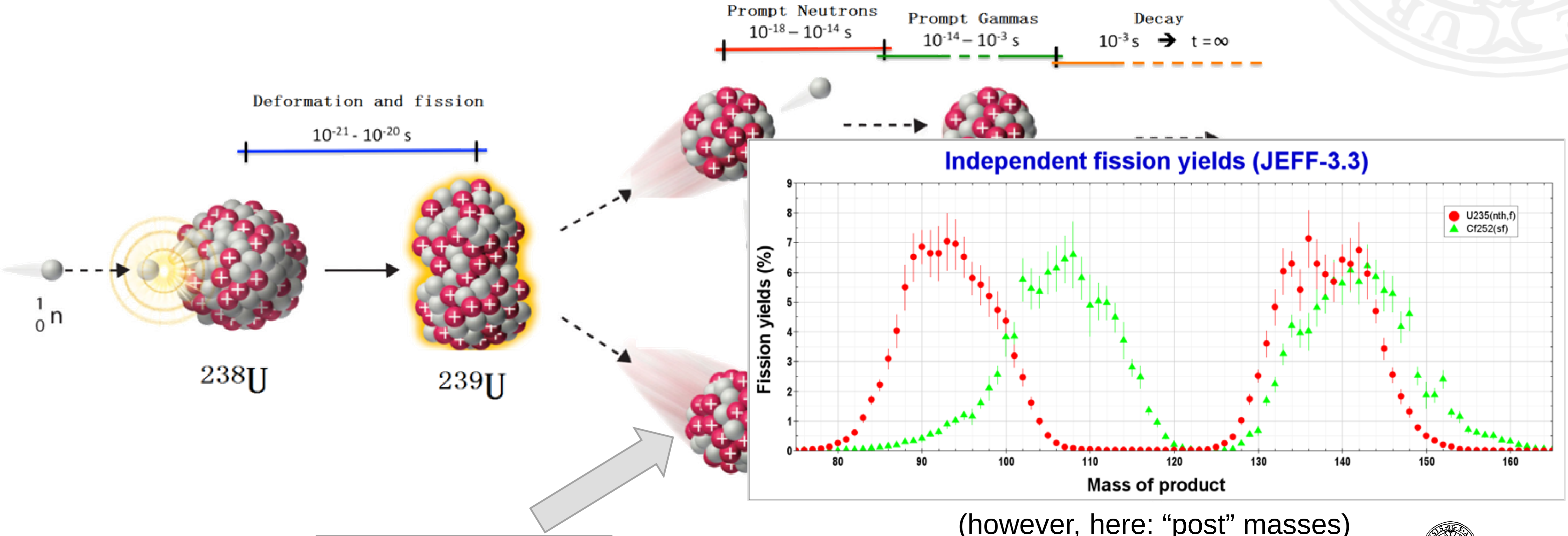
Fission observables



fission yields (A,Z,E)

(however, here: “post” masses)

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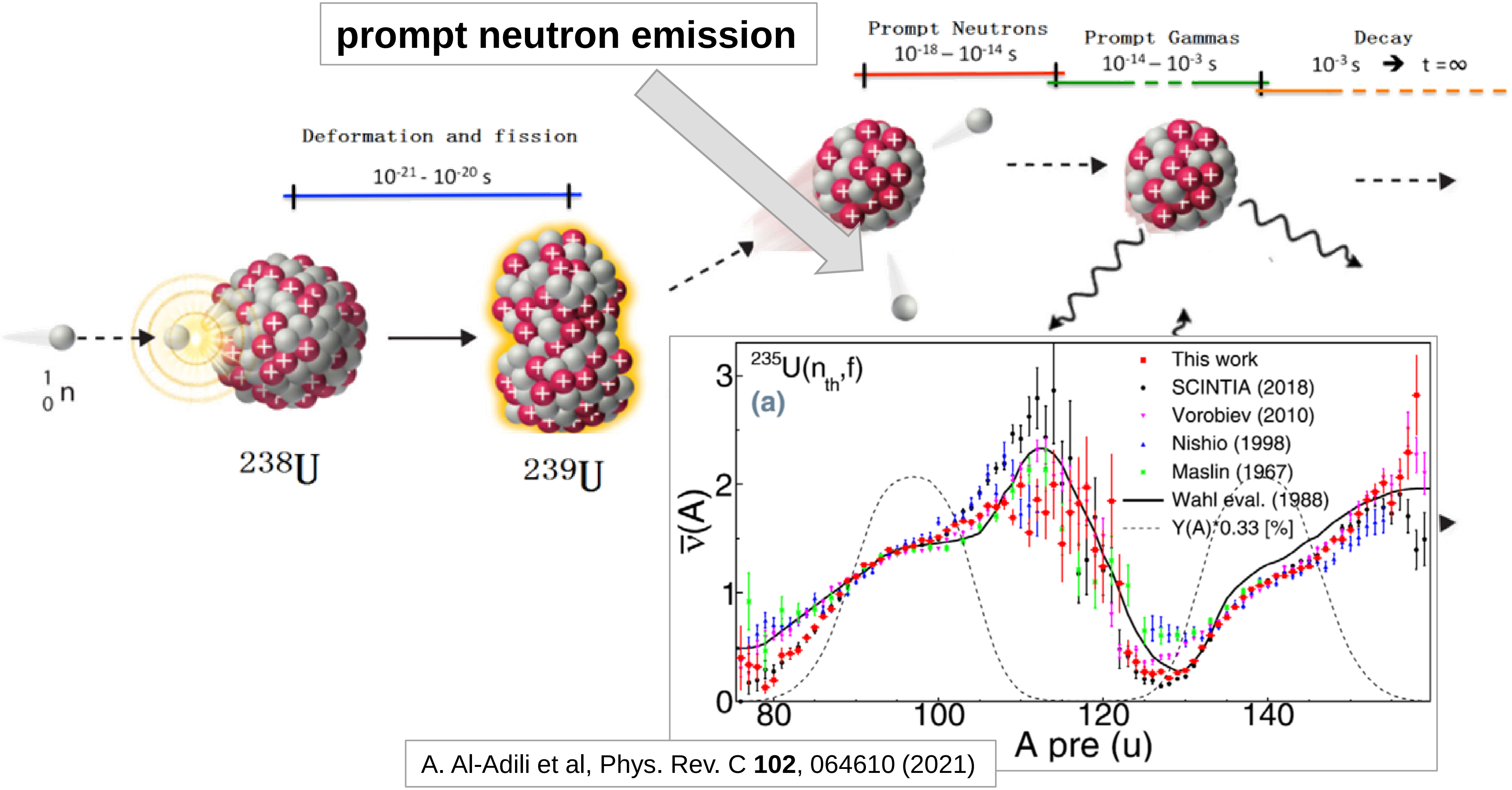
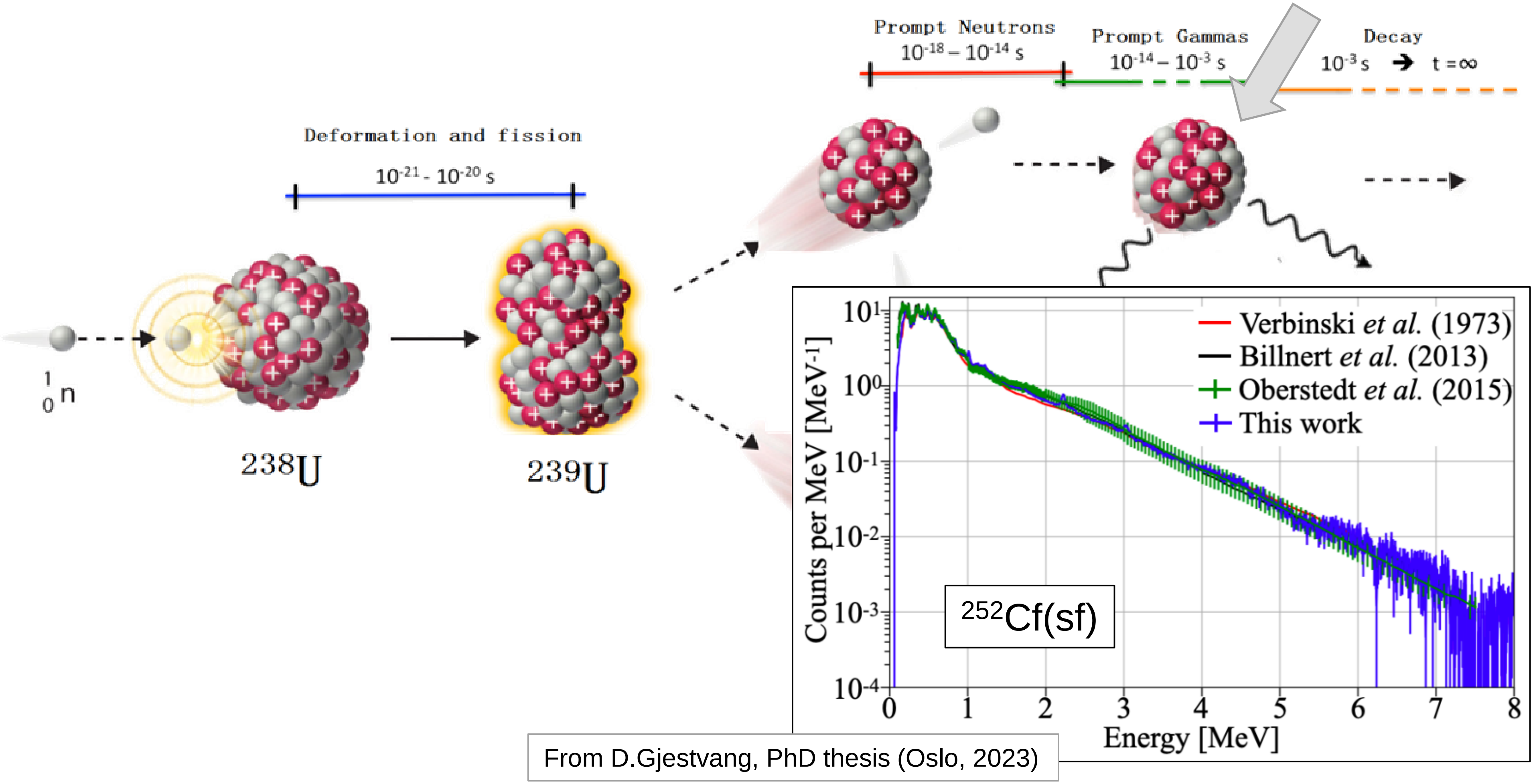
Fission observables



A. Al-Adili et al, Phys. Rev. C **102**, 064610 (2021)

Fission observables

prompt gamma emission



From D.Gjestvang, PhD thesis (Oslo, 2023)

Energetics in the fission process

- Q-value in case of neutron-induced fission:

Q = mass(target) + mass(neutron) + E(incident neutron) – mass(fragment)
= TKE + TXE

(TKE = Total Kinetic Energy; TXE = Total eXitation Energy)

Case of thermal fission of ^{235}U:

<Q>	$\approx$ 195 MeV	
<TKE>	$\approx$ 170 MeV	kinetic energy of FF
<TXE>	$\approx$ 25 MeV	available for n and γ emission

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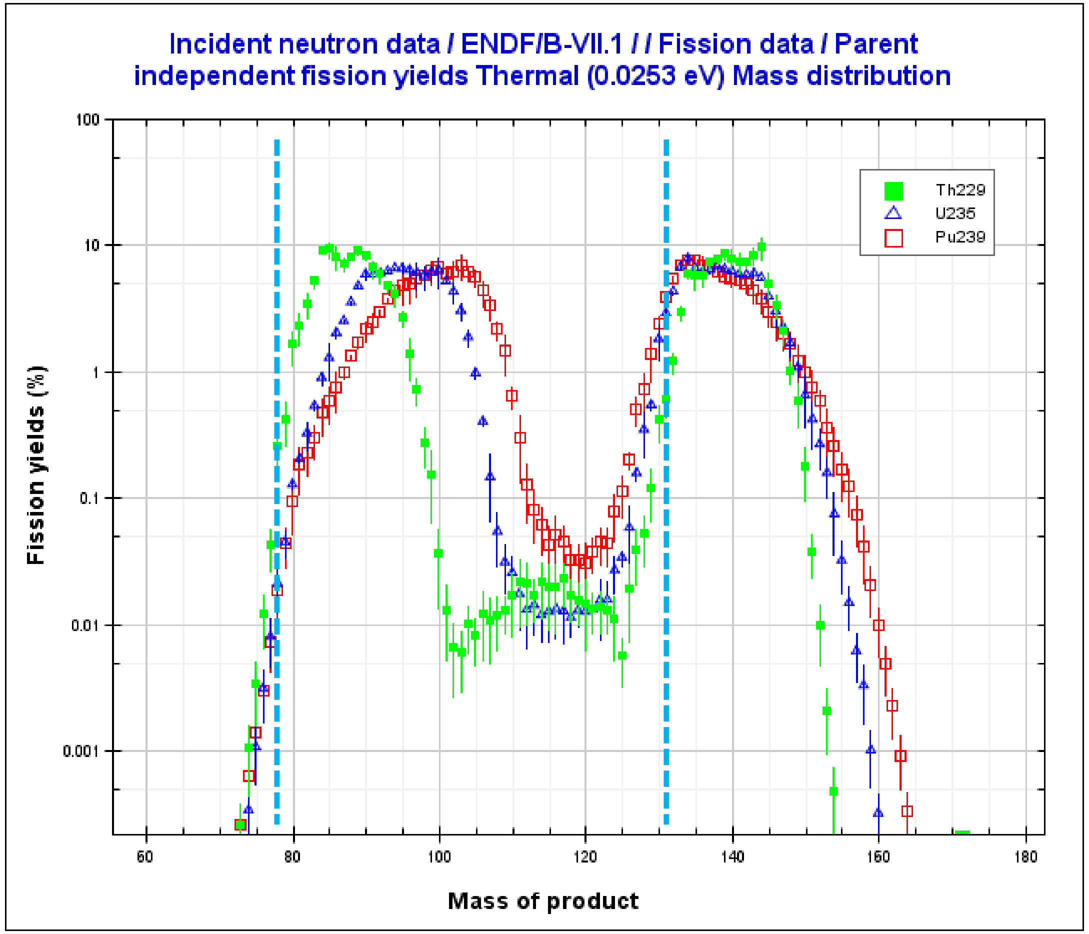
Fission yields – general observations



- Asymmetric distribution (in general).
- Light mass peak shifts as compound mass (mass of the fissioning system) increases.
- Heavy mass peak remains centred around A≈140.
- Cannot be understood from the liquid drop model.
- Attributed to the energetically favourable closed shells at (or around) mass 132 (with Z=50, N=82), as well as mass 78.

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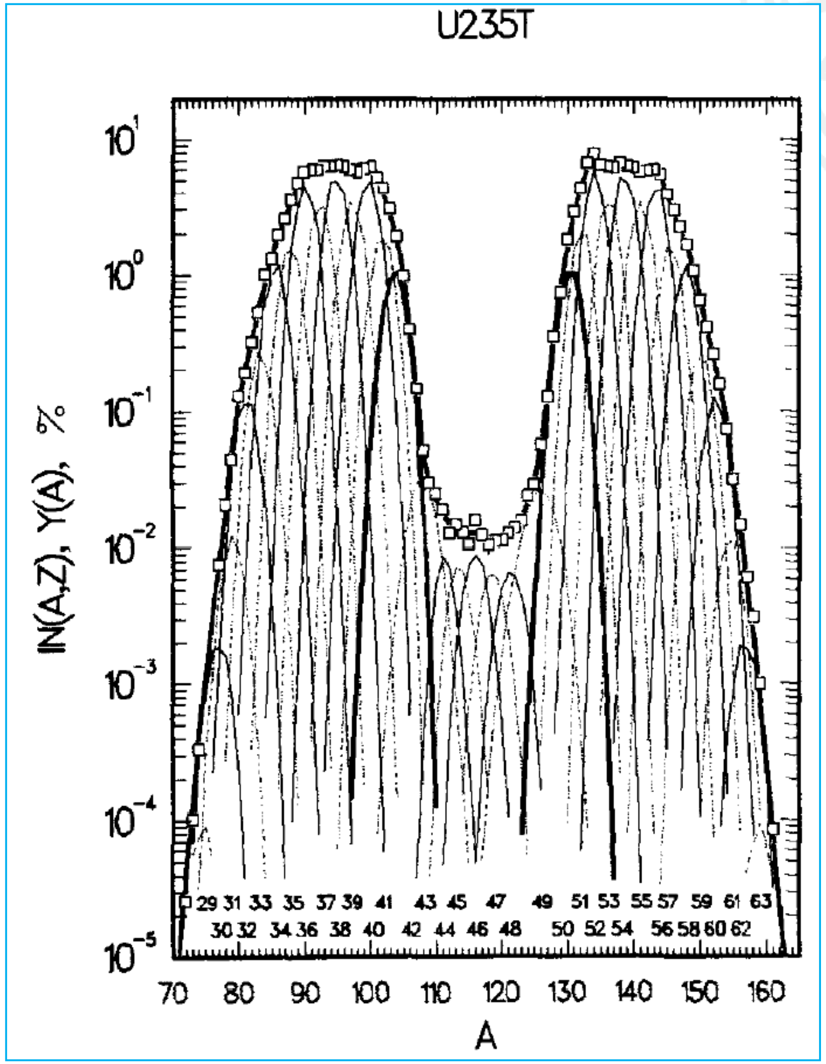
Wahl systematics

Review of (at the time) available experimental FY data and development of an empirical model for ^{233}U(n$_{th}$,f), ^{235}U(n$_{th}$,f), ^{239}Pu(n$_{th}$,f), ^{252}Cf(SF) :

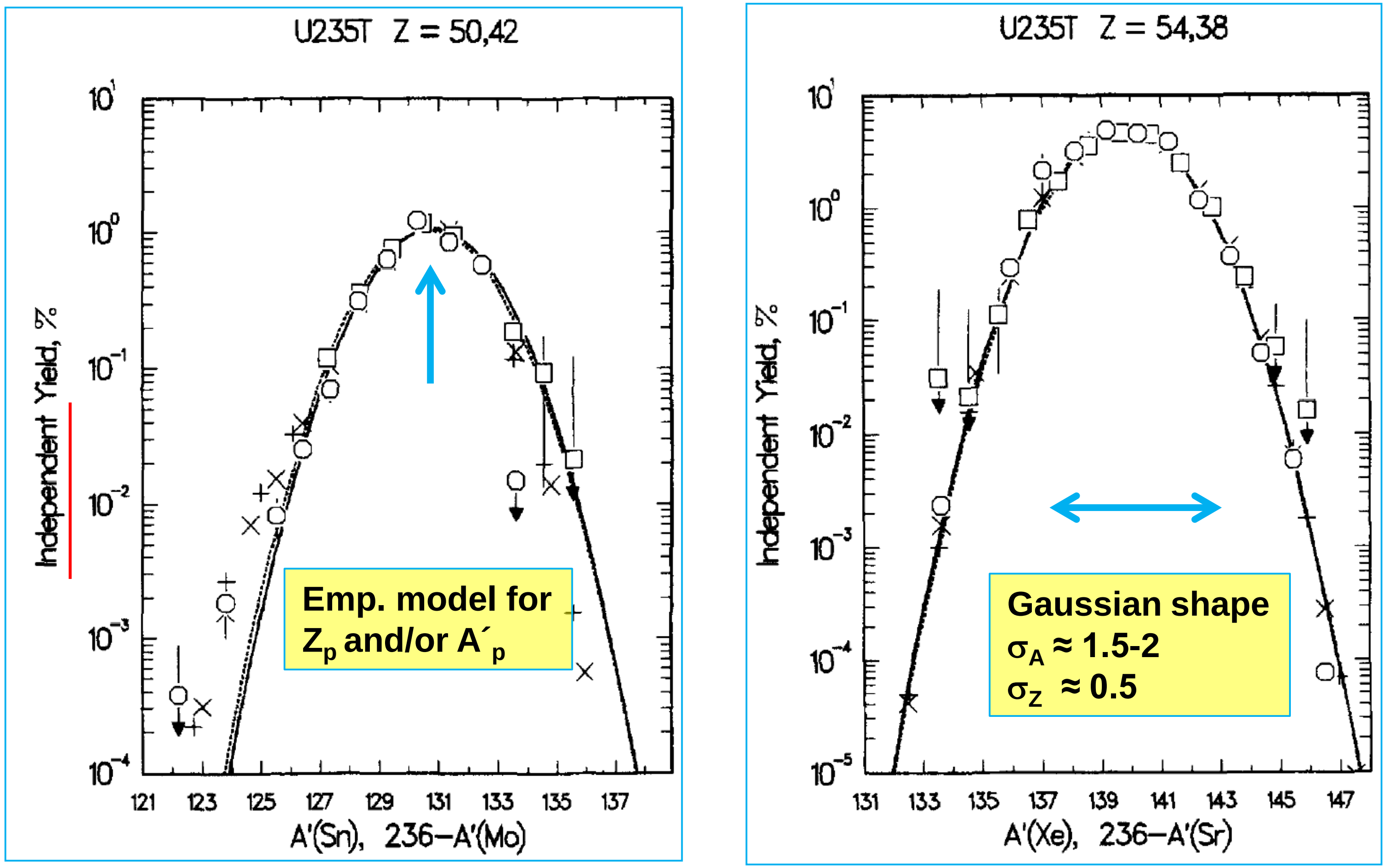
A.C. Wahl, At Data and Nucl Data Tables, 39 (1988) 1-156

Later extensions (C.Wahl, *LA-13928* (2002)) include, e.g., nubar(A).



Yields normalized to total yield of 200%.

Wahl systematics



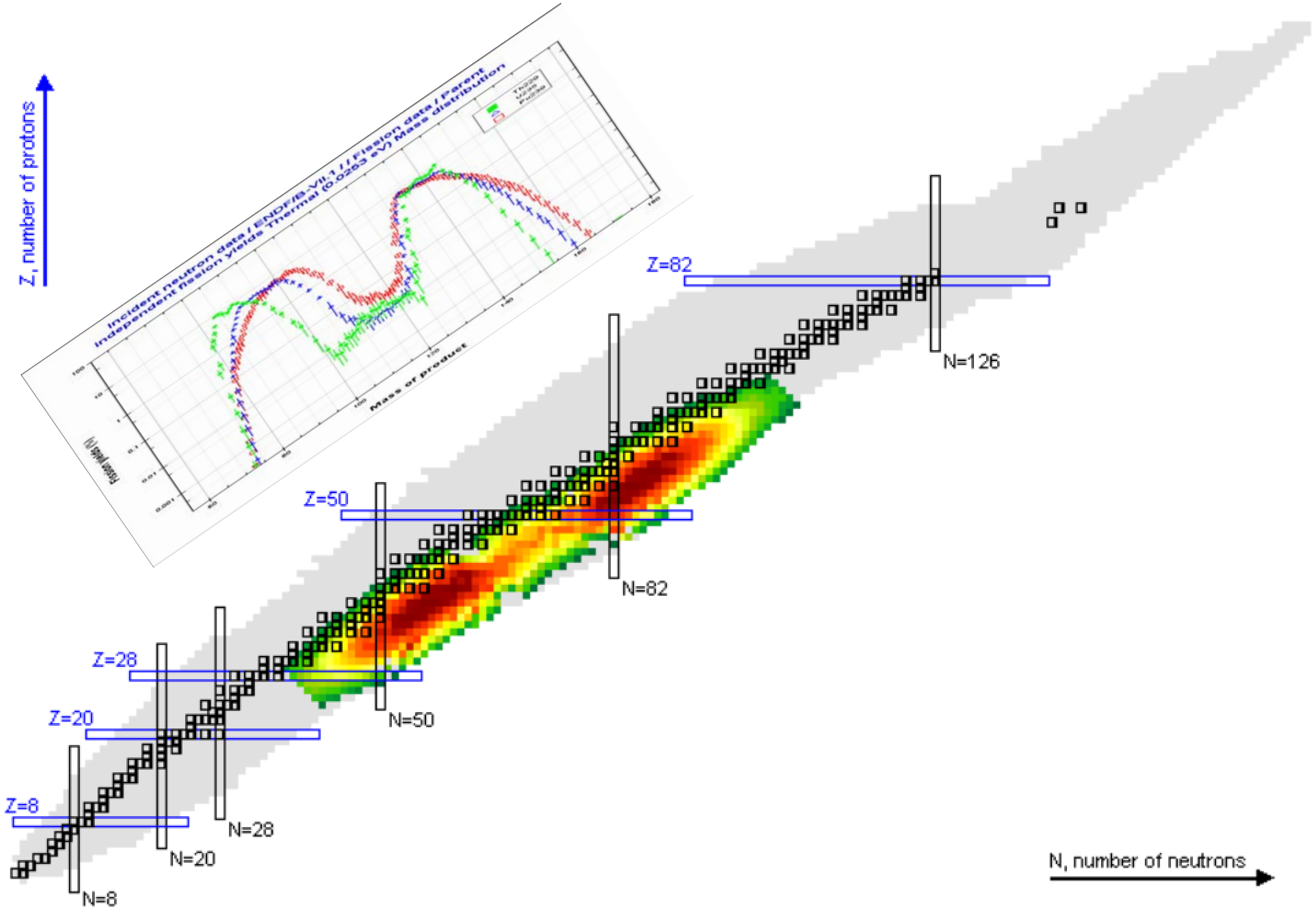
Emp. model for Z_p and/or A'_p

Gaussian shape
$\sigma_A \approx 1.5\text{-}2$
$\sigma_Z \approx 0.5$

Corrections for even-odd effects

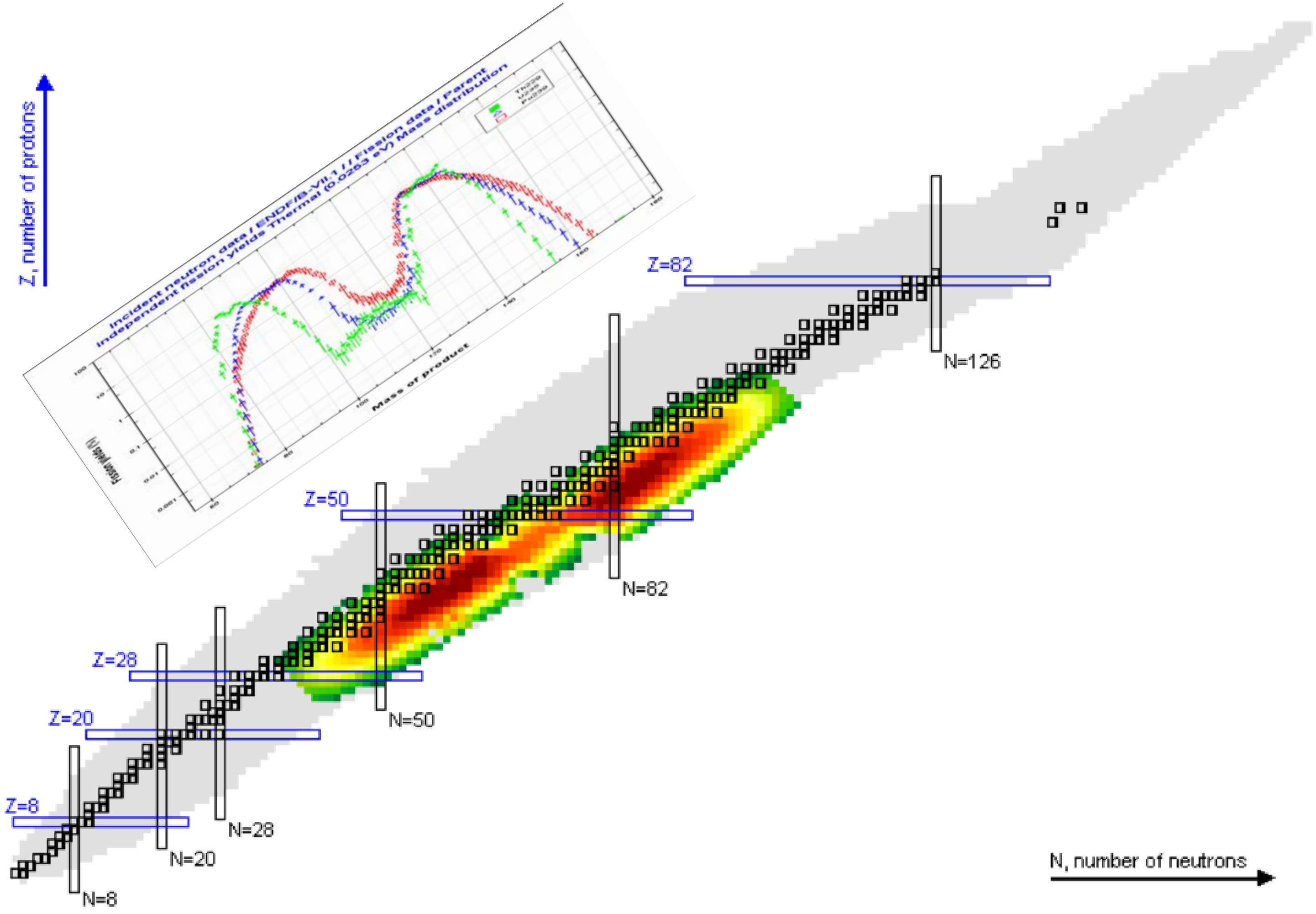
A.C. Wahl, At Data and Nucl Data Tables, 39 (1988) 1-156

Isotopic fission yields for $^{235}U(n_{th},f)$



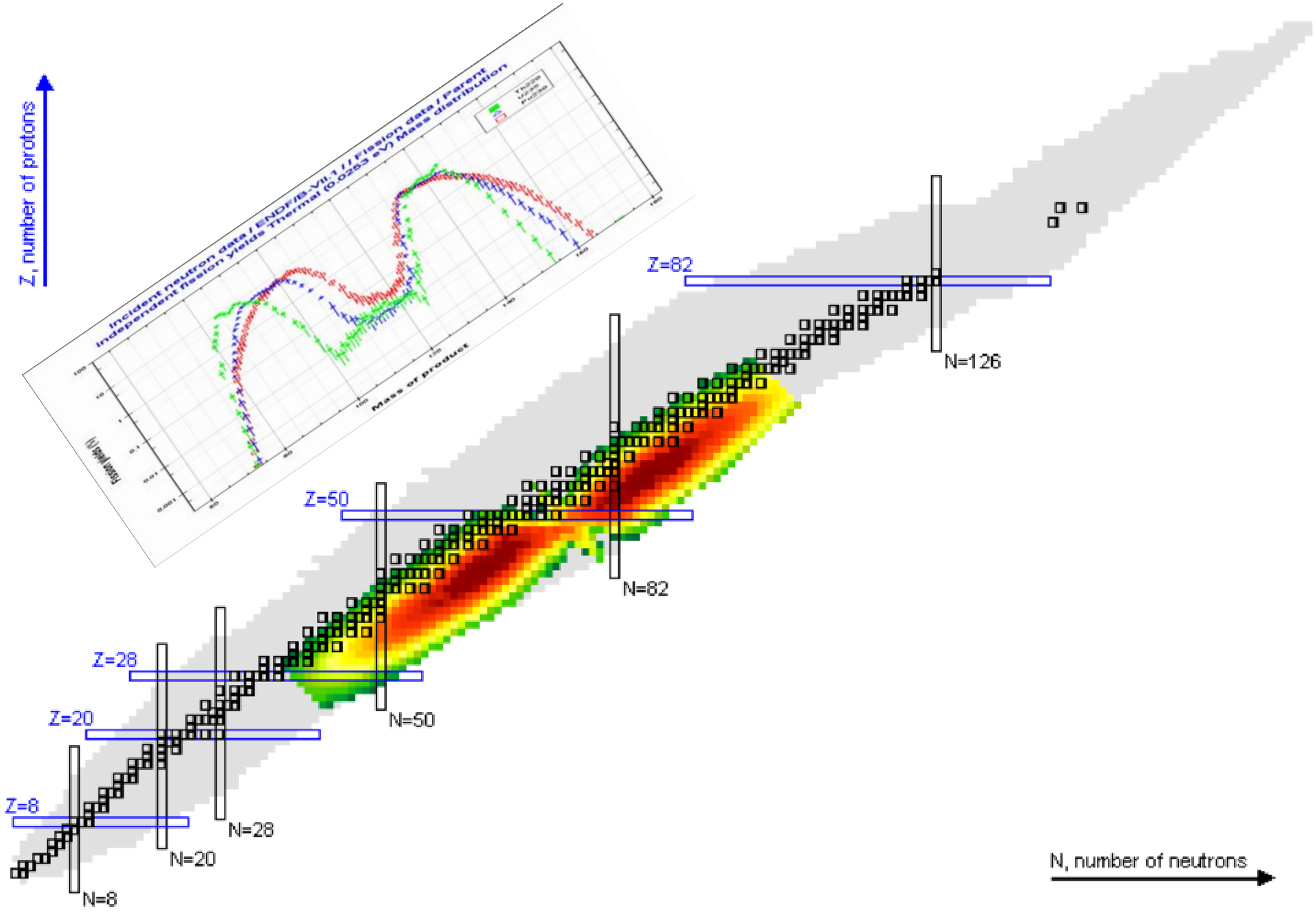
Data from https://www.nndc.bnl.gov

Isotopic fission yields for $^{239}\text{Pu}(n_{th},f)$



Data from https://www.nndc.bnl.gov

Isotopic fission yields for ^{252}Cf(sf)



Data from https://www.nndc.bnl.gov

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Dependence of yields on neutron energy

Yields generally asymmetric.

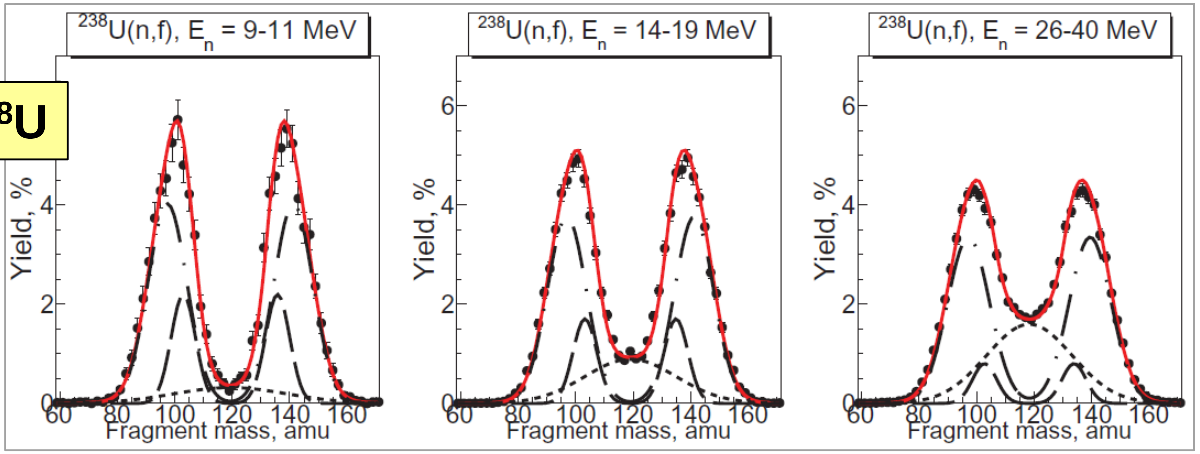
But towards higher energies the symmetric component increases (and distributions get wider).

I.e. shell effects less important at high excitation energies.

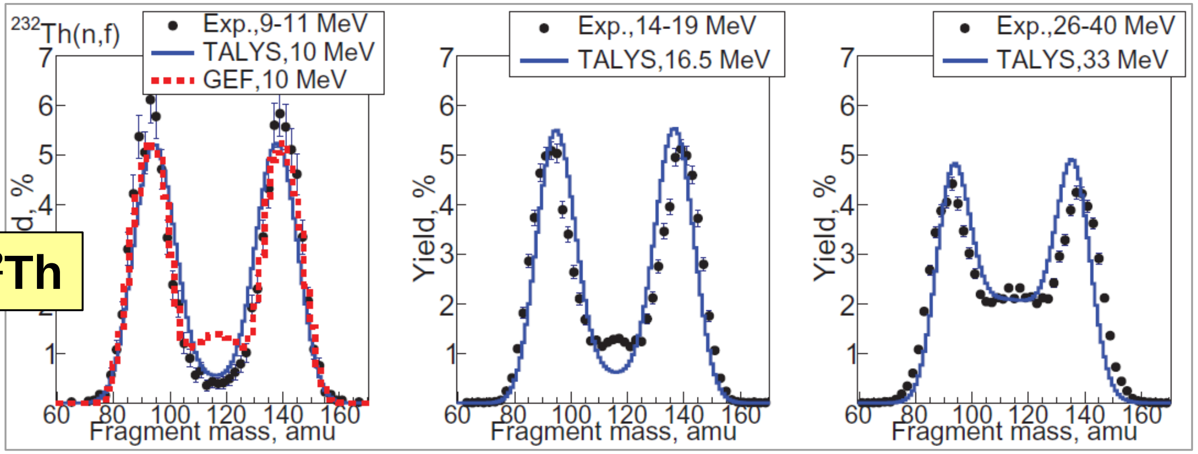
Figures show **exp. data** (from LLN) for two target nuclei, analysed in terms of **fission modes** (top) as well as comparison with **model calculations** (bottom) (2014!)

increasing energy of incident neutron →

^{238}U

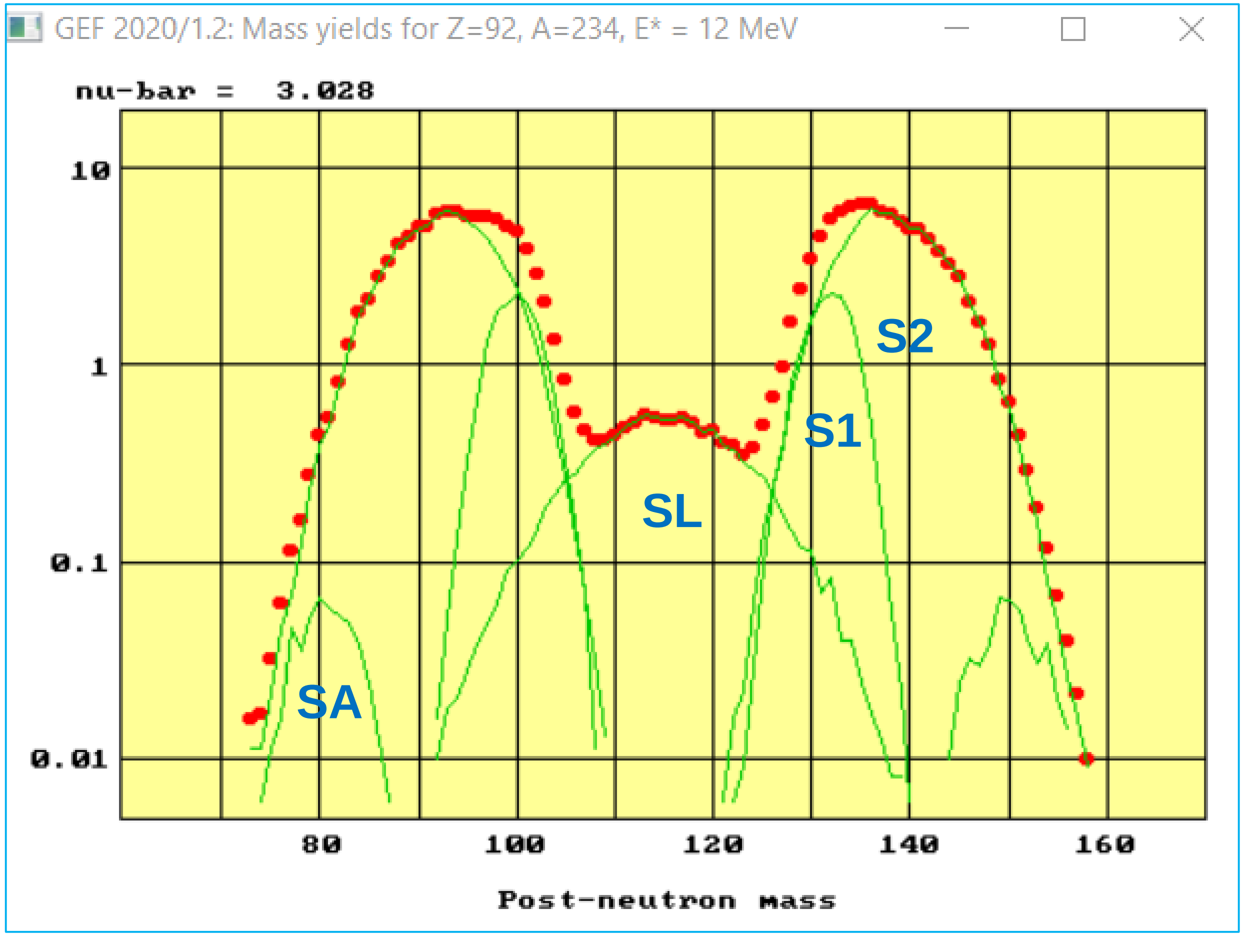


^{232}Th



V. Simutkin et al., Nucl. Data Sheets 119 (2014) 331.

Example calculation result using GEF (2020/1.2)



First chance fission of ^{234}U*

with 12 MeV excitation energy.

Monte Carlo code;

note that the code gives contributions from different fission modes.

Reference paper for GEF:
K.-H. Schmidt, et al., Nuclear Data Sheets 131 (2016) 107-221.
https://doi.org/10.1016/j.nds.2015.12.009

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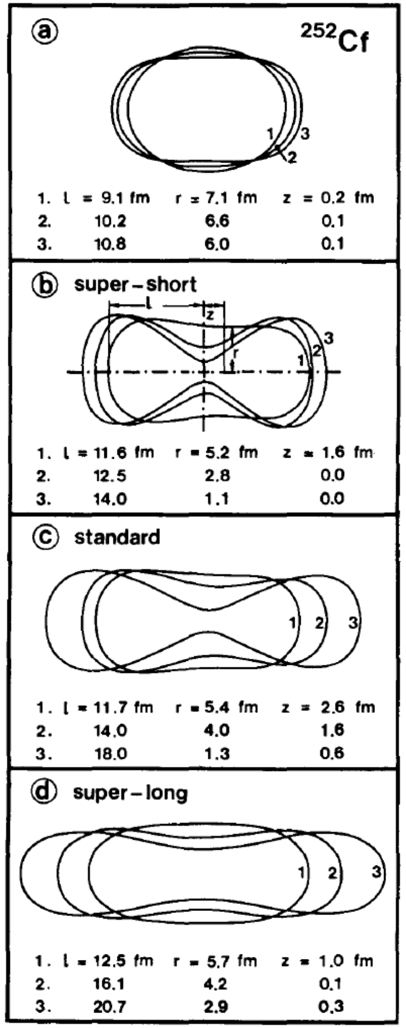
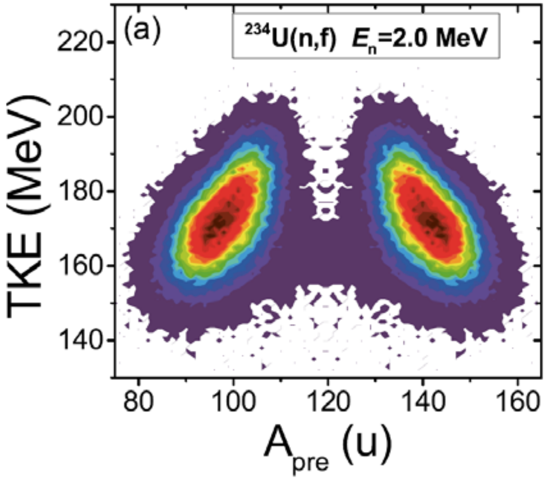
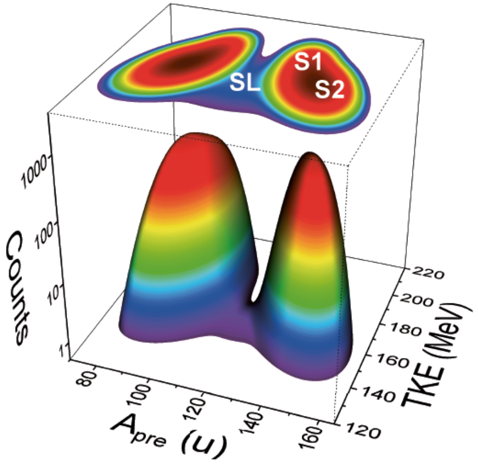
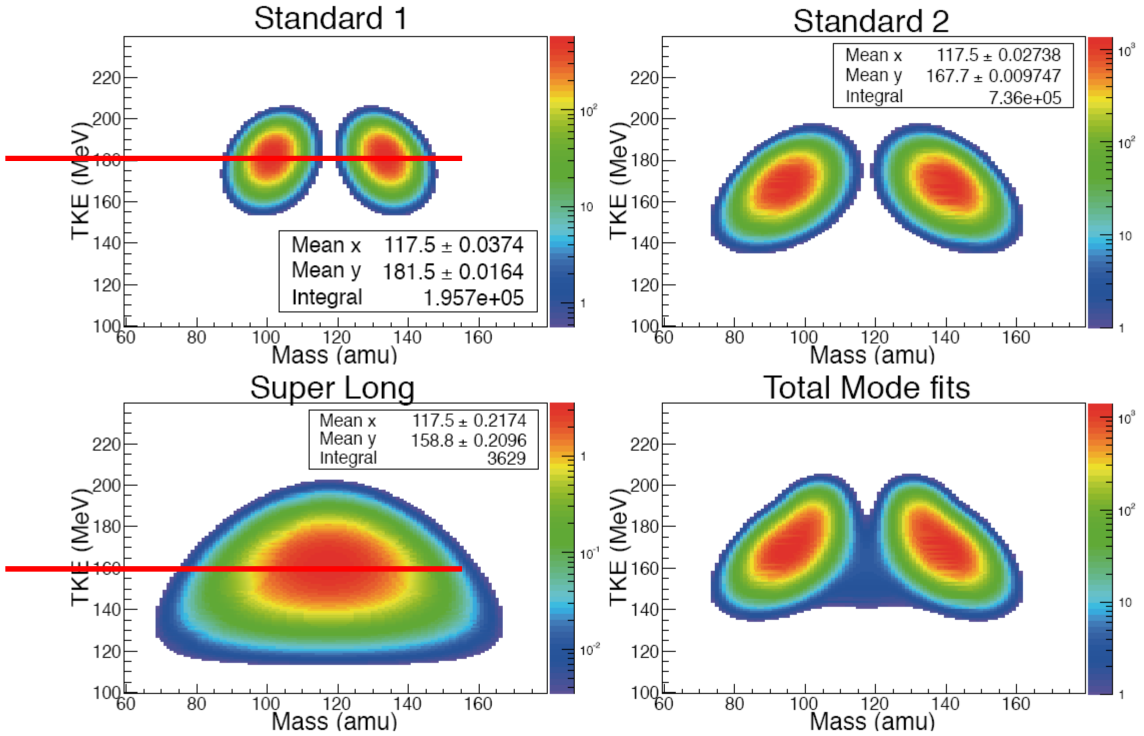
Fission modes a la Brosa



From: Brosa et al., *Phys. Rep.* **197** (1990) 167







From: Ali Al-Adili, PhD thesis, Uppsala, 2013

Across the nuclear chart

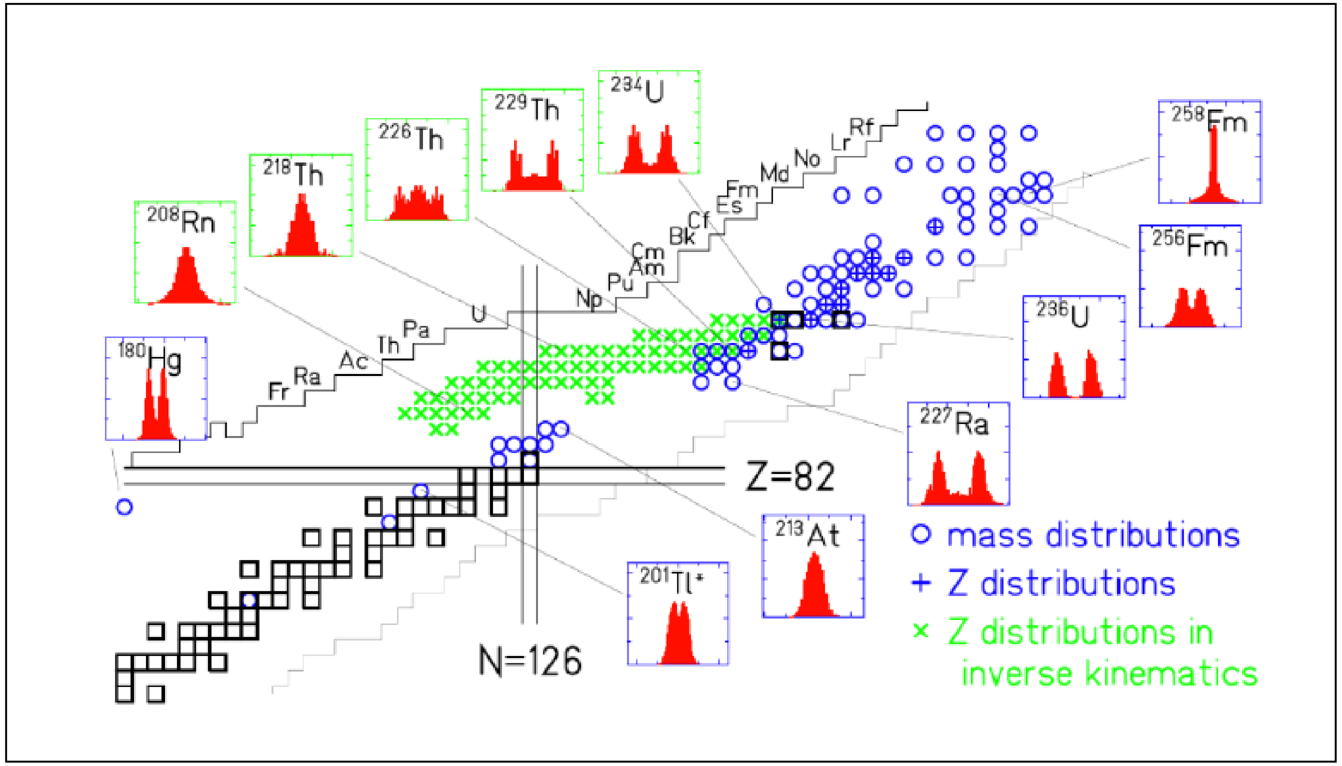


Figure from K.H. Schmidt et al., Nucl. Data Sheets **131** (2016) 107.

Looking beyond the region of the major actinides, we see a variety of fission yield distributions.

Driven by nuclear structure (of the fragments).

Sometimes there are surprises:
Fission of ^{180}Hg should be symmetric (centred around double magic ^{90}Zr) but is not!

(see A.Andreyev et al. Phys. Rev. Lett. 105, 252502 (2010))

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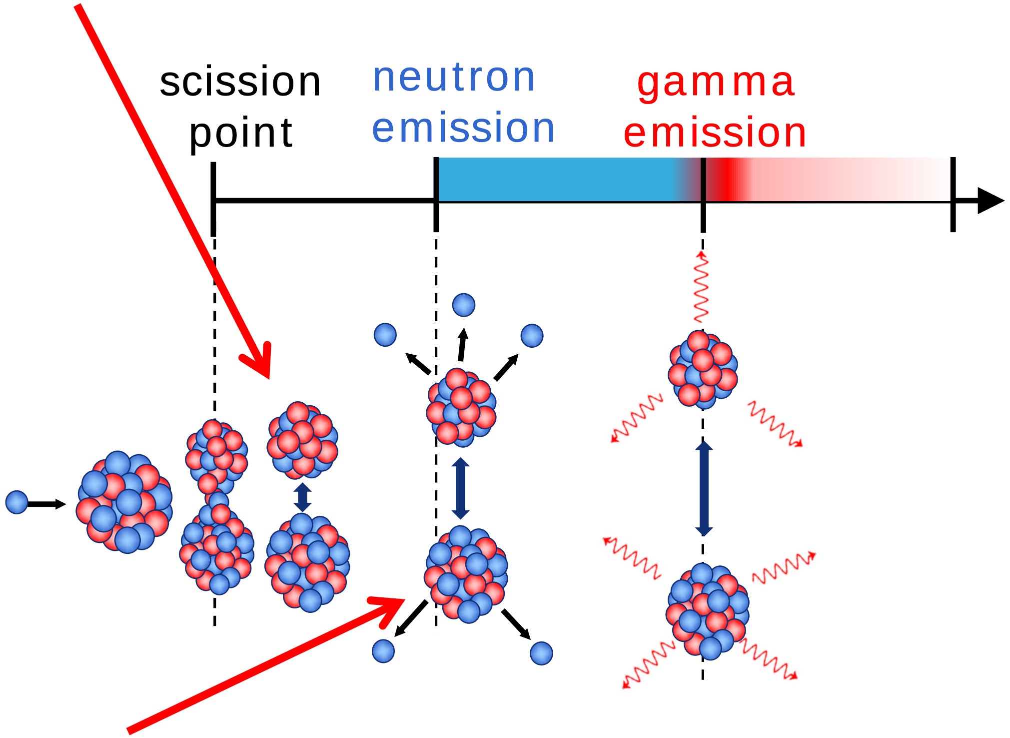
Yields: Definitions I

- **Fragment yields**: Yields prior to prompt neutron emission.



- **Product yields:** Yields posterior to prompt neutron emission.

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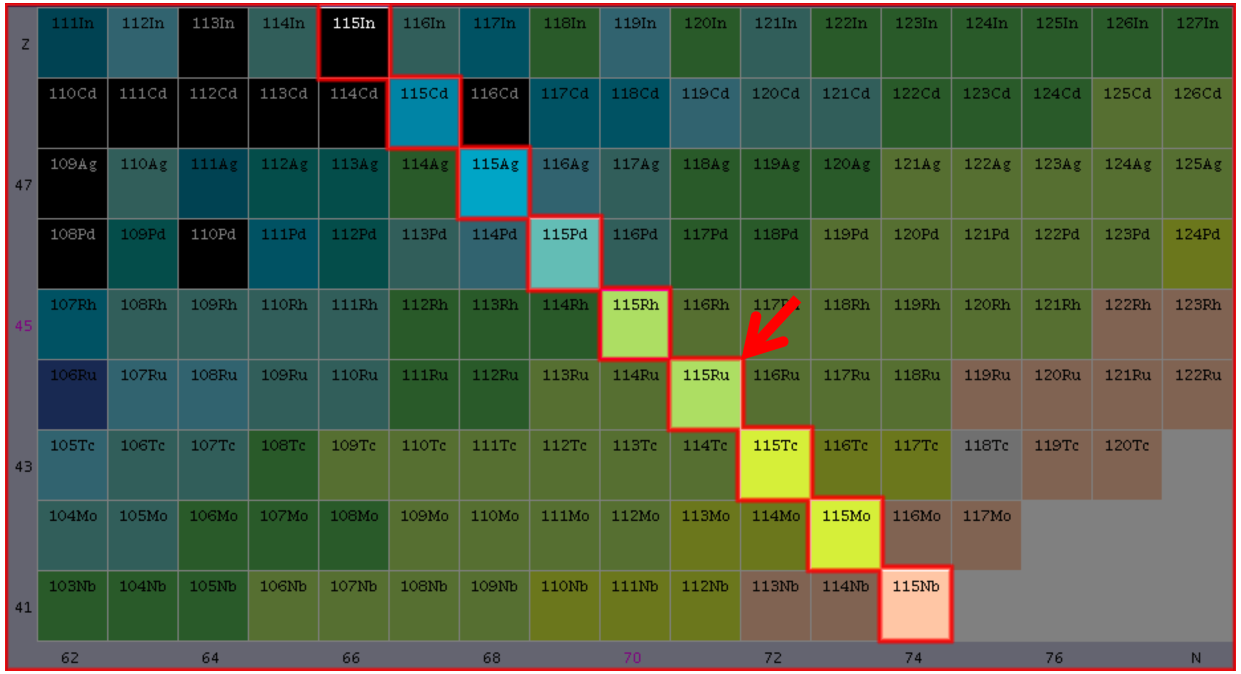
Yields: Definitions II

- **Independent fission yields:** number of atoms of a specific nuclide produced directly in the fission process (*i.e.*, not via radioactive decay of **precursors**).

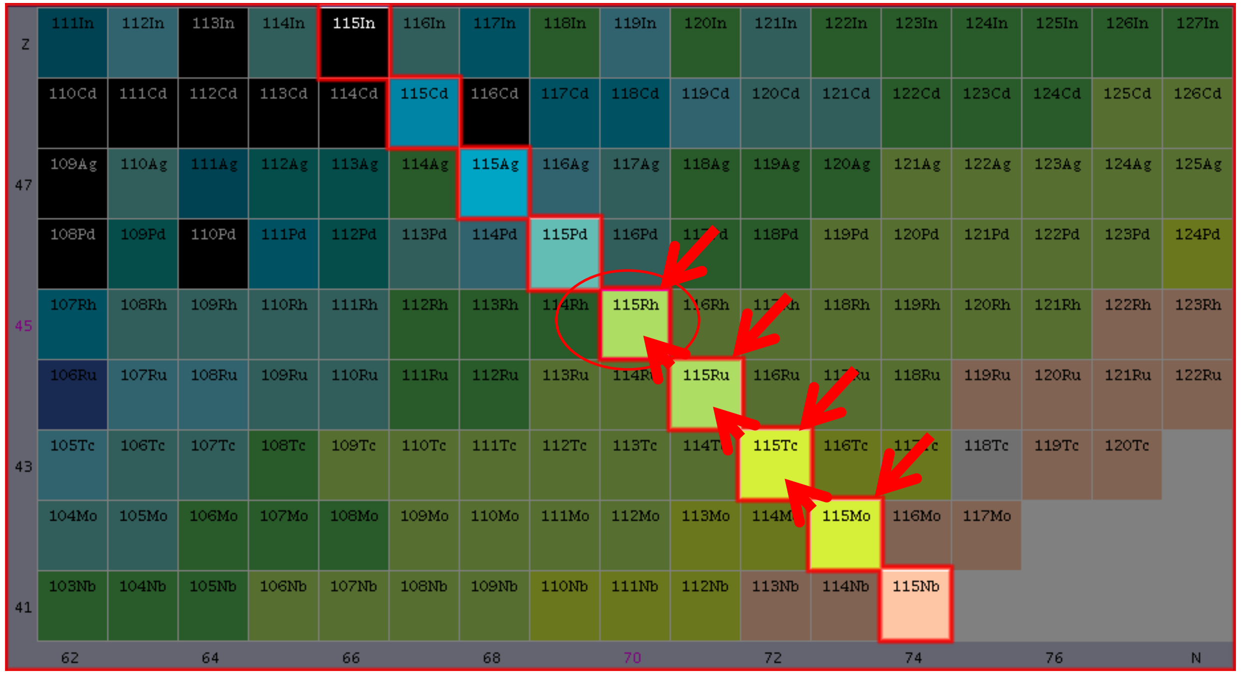


Z	111In	112In	113In	114In	115In	116In	117In	118In	119In	120In	121In	122In	123In	124In	125In	126In	127In
	110Cd	111Cd	112Cd	113Cd	114Cd	115Cd	116Cd	117Cd	118Cd	119Cd	120Cd	121Cd	122Cd	123Cd	124Cd	125Cd	126Cd
47	109Ag	110Ag	111Ag	112Ag	113Ag	114Ag	115Ag	116Ag	117Ag	118Ag	119Ag	120Ag	121Ag	122Ag	123Ag	124Ag	125Ag
	108Pd	109Pd	110Pd	111Pd	112Pd	113Pd	114Pd	115Pd	116Pd	117Pd	118Pd	119Pd	120Pd	121Pd	122Pd	123Pd	124Pd
45	107Rh	108Rh	109Rh	110Rh	111Rh	112Rh	113Rh	114Rh	115Rh	116Rh	117Rh	118Rh	119Rh	120Rh	121Rh	122Rh	123Rh
	106Ru	107Ru	108Ru	109Ru	110Ru	111Ru	112Ru	113Ru	114Ru	115Ru	116Ru	117Ru	118Ru	119Ru	120Ru	121Ru	122Ru
43	105Tc	106Tc	107Tc	108Tc	109Tc	110Tc	111Tc	112Tc	113Tc	114Tc	115Tc	116Tc	117Tc	118Tc	119Tc	120Tc	
	104Mo	105Mo	106Mo	107Mo	108Mo	109Mo	110Mo	111Mo	112Mo	113Mo	114Mo	115Mo	116Mo	117Mo			
41	103Nb	104Nb	105Nb	106Nb	107Nb	108Nb	109Nb	110Nb	111Nb	112Nb	113Nb	114Nb	115Nb				
	62		64		66		68		70		72		74		76		N

Source: IAEA-TECDOC-1168 (2000)

Yields: Definitions III

- **Cumulative fission yields**: total number of atoms of a specific nuclide produced (directly and via decay of precursors).



Z	111In	112In	113In	114In	115In	116In	117In	118In	119In	120In	121In	122In	123In	124In	125In	126In	127In
	110Cd	111Cd	112Cd	113Cd	114Cd	115Cd	116Cd	117Cd	118Cd	119Cd	120Cd	121Cd	122Cd	123Cd	124Cd	125Cd	126Cd
47	109Ag	110Ag	111Ag	112Ag	113Ag	114Ag	115Ag	116Ag	117Ag	118Ag	119Ag	120Ag	121Ag	122Ag	123Ag	124Ag	125Ag
	108Pd	109Pd	110Pd	111Pd	112Pd	113Pd	114Pd	115Pd	116Pd	117Pd	118Pd	119Pd	120Pd	121Pd	122Pd	123Pd	124Pd
45	107Rh	108Rh	109Rh	110Rh	111Rh	112Rh	113Rh	114Rh	115Rh	116Rh	117Rh	118Rh	119Rh	120Rh	121Rh	122Rh	123Rh
	106Ru	107Ru	108Ru	109Ru	110Ru	111Ru	112Ru	113Ru	114Ru	115Ru	116Ru	117Ru	118Ru	119Ru	120Ru	121Ru	122Ru
43	105Tc	106Tc	107Tc	108Tc	109Tc	110Tc	111Tc	112Tc	113Tc	114Tc	115Tc	116Tc	117Tc	118Tc	119Tc	120Tc	
	104Mo	105Mo	106Mo	107Mo	108Mo	109Mo	110Mo	111Mo	112Mo	113Mo	114Mo	115Mo	116Mo	117Mo			
41	103Nb	104Nb	105Nb	106Nb	107Nb	108Nb	109Nb	110Nb	111Nb	112Nb	113Nb	114Nb	115Nb				
	62		64		66		68		70		72		74		76		N

Source: IAEA-TECDOC-1168 (2000)

Yields: Definitions IV

- **Total chain yields:** defined as the sum of cumulative yield(s) of the last (stable or long-lived) chain member(s).

- **Mass number yields:** defined as the sum of all independent yields of a particular mass chain and are in this way distinguished from chain yields (delayed neutrons emission).

- Many modern methods to measure fission yields provide sets of truly independent yields which - at summation - will produce *mass number yields* rather than *chain yields*.

Source: IAEA-TECDOC-1168 (2000)

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Yields – what can we measure, what do we need?

- FY measurements give, in the best case, information about the independent fission **product** yield.
- Fission **fragment** yields can be inferred using information on prompt-neutron emission (relevant for studies of the fission process).
- Depending on the measurement technique, one might measure:
  - Cumulative yields for an isotope (measurement time <-> precursor lifetimes)
  - Mass yields (no resolution on Z)
- For **fission research** we want information *on isotopic fission fragment* yields.
- For **applications**, one might need:
  - Information on the yield for a chemical element (e.g. fission gas)
  - Isotopic yields for certain important isotopes, e.g.:
    - Neutron absorbers
    - Delayed-neutron emitters
    - Long-lived isotopes

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Experimental methods for measuring fission yields

With stopped fragments:

- Radiochemical separation
  - earliest method; only long-lived products; activity measured from β-decay or γ-decay (with low resolution) (Radiochemistry: M. Curie; in fission: e.g. O. Hahn et al.)
- Mass spectrometry
  - long-lived products only; sample evaporated and mass analyzed; precise technique but difficult to normalize
- γ-ray spectroscopy
  - isotope specific; also short-lived nuclides accessible; can be used with activation technique or fast on-line mass separation
- ISOL method (similar to mass spectrometry but “online” separation)
  - can be combined with Penning trap or γ-ray spectroscopy; can be difficult to normalize

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Experimental methods for measuring fission yields

Unstopped fragments:

- LOHENGRIN@ILL
  - reactor based
  - mass, charge, and energy are measured
  - only one fission product per fission, no corr.
  - combined with, e.g., γ-spectroscopy

Thermal flux at target: about $5 \cdot 10^{14}$ neutrons/s/cm^2

Fission product yields: down to

10^{-4} % for binary fission, and

10^{-6} % for ternary fission can be measured.

Mass resolving power: 0.3%.

Energy resolution: 1%.

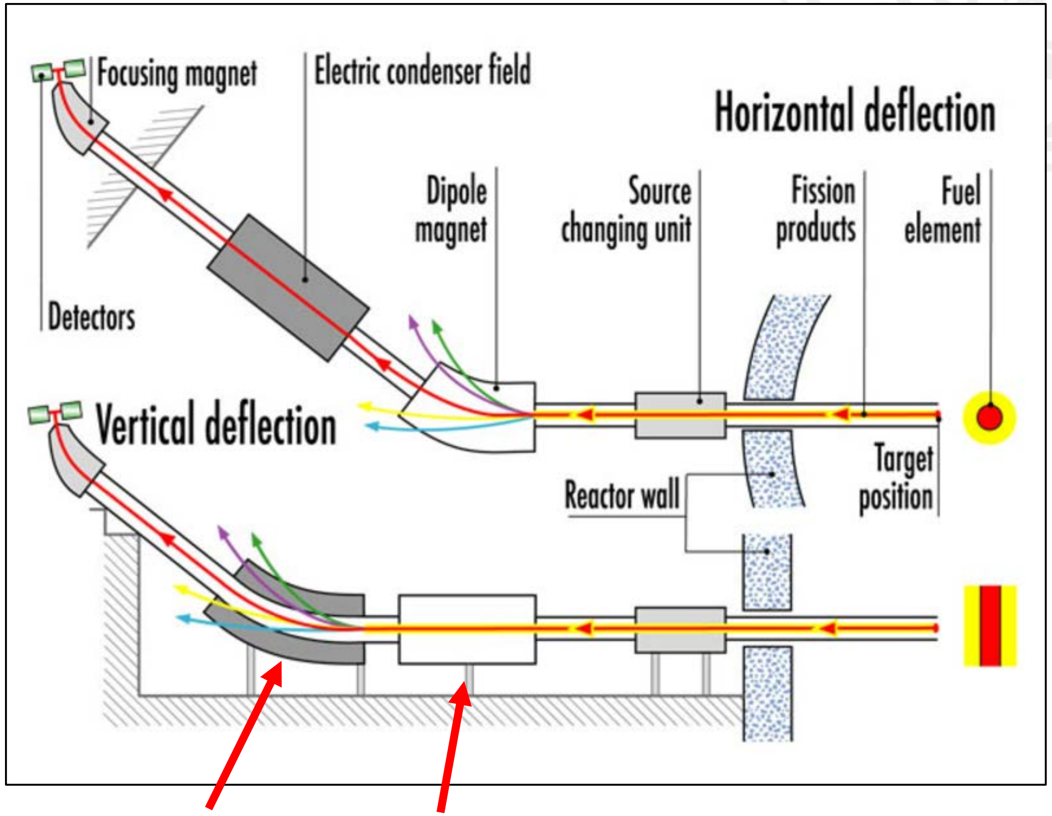

Image from https://www.ill.eu

Electric and magnetic field settings allow selection by A/q and E/q

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LOHENGRIN – example results

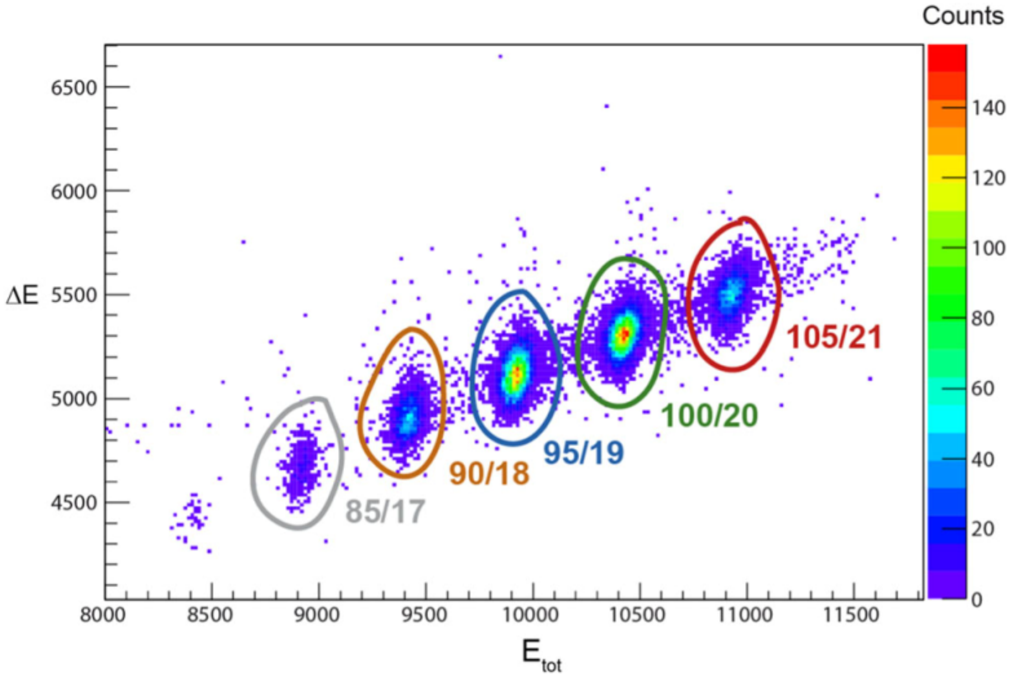

Fig. 3 (ΔE, E_{tot}) spectrum measured with the IC with the LOHENGRIN spectrometer set to select $\frac{A}{q} = 5$ and $\frac{E_k}{q} = 5$ ratios. The numbers displayed such as 100/20 indicates that the mass $A = 100$ is selected with the ionic charge $q = 20$ and a kinetic energy $E_k = 100$ MeV

A. Chebboubi, et al., Eur. Phys. J. A (2021) **57**:335.

Fission products are stopped in an ionisation chamber and ΔE-E is measured (see figure).

In the figure: A/q = 5 and E/q = 5.

> “Measurements of one mass yield require around 80 different settings. During a week up to 4500 different instrument settings are requested to perform yield measurements of about 20 masses and to monitor the target.”

Target burnup is an issue and must be monitored and corrected for.

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LOHENGRIN – example results

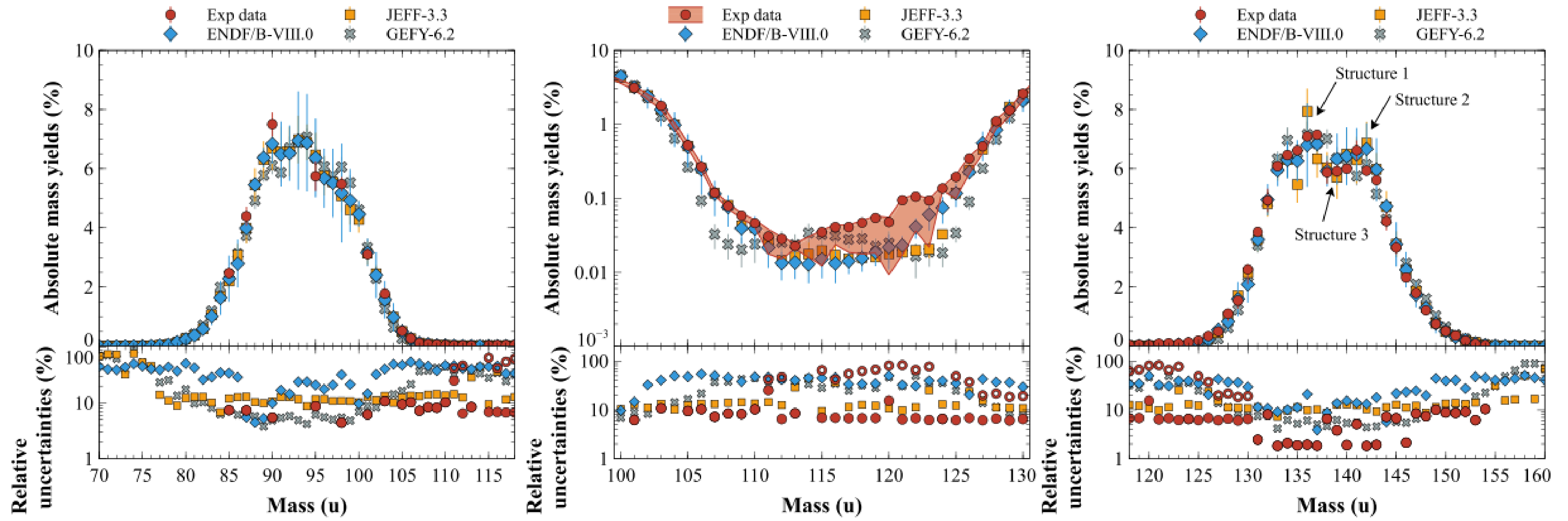

Fig. 13 Absolute fission product mass yields for ^{233}U(n_{th}, f) (red circle points) in comparison with ENDF/B-VIII.0 (blue diamond points), JEFF-3.3 (yellow square points) and GEFY-6.2 (grey cross points) libraries. ENDF/B-VIII.0 seems slightly in better agreement with our experimental data. Uncertainties of each data set are displayed in the lower part. The precision of LOHENGRIN data is of the order of 2% in the heavy mass peak and around 10% in the wings. In the symmetry region, the positive (red circle points) and negative (empty red circle) uncertainties are displayed. Lines are only to guide the eye

A. Chebboubi, et al., Eur. Phys. J. A (2021) **57**:335.

Experimental methods for measuring fission yields

Unstopped fragments:

- Inverse kinematics
  - accelerator based (multi-nucleon transfer or Coulomb excitation)
  - can access fission of exotic, very short-lived nuclei
  - e.g. SOFIA@GSI (both FF) and VAMOS@GANIL (one FF)
  - high mass resolution (isotopic yields)
  - Detector setup allows for identification of charge of fragment

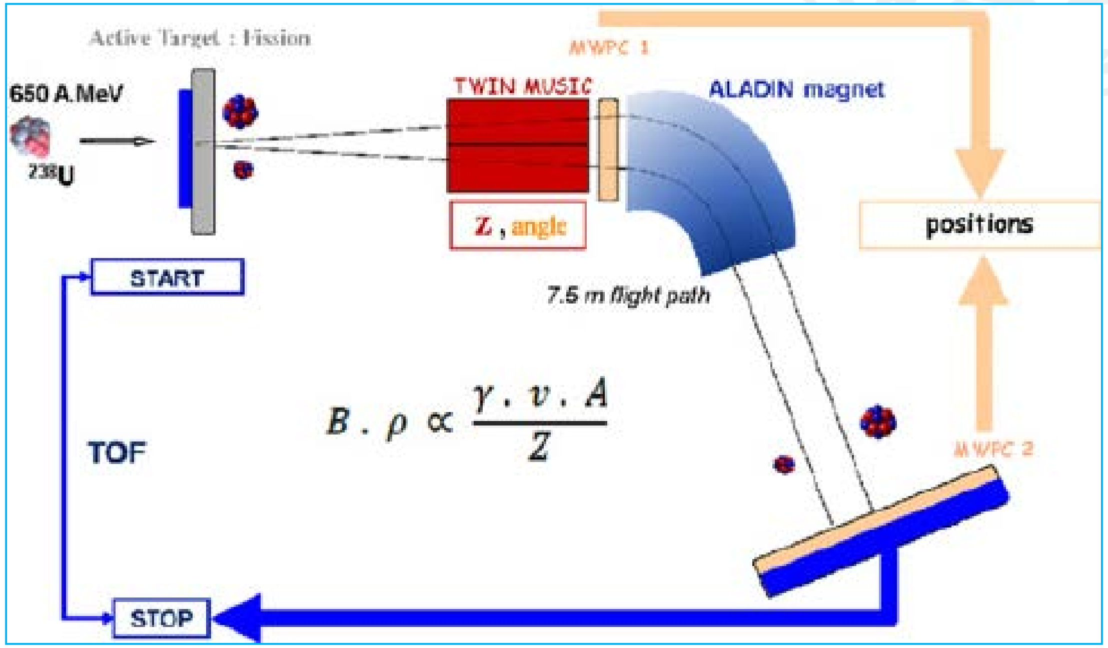

Figure: E. Pellereau et al., EPJ Web of Conf. **62**, 06005 (2013)

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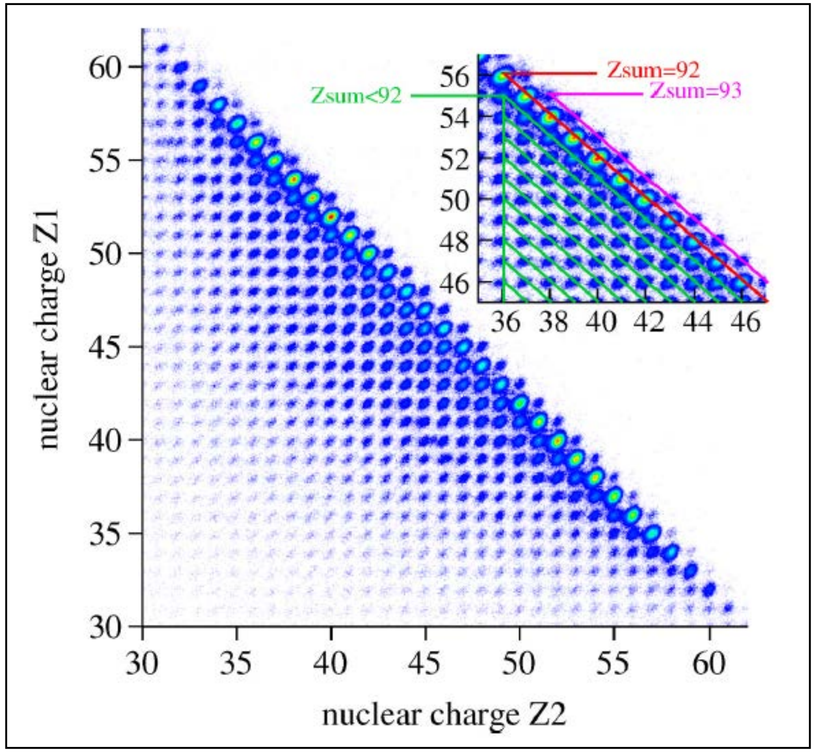
Example from SOFIA@GSI:

E. Pellereau et al. Phys. Rev. C **95**, 054603 (2017)



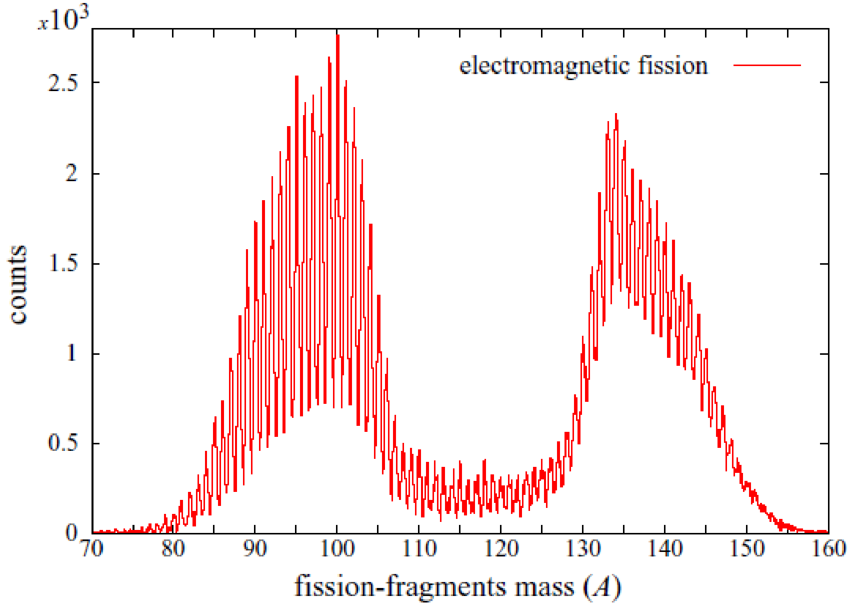


FIG. 11. Fission-fragment mass distribution for electromagnetically induced fission of ^{238}U.

Charges of correlated fission products and mass distribution from electromagnetically induced fission of ^{238}U. Beam: ^{238}U; Target: ^{12}C. Selecting Zsum = 92 gives fission of Uranium.

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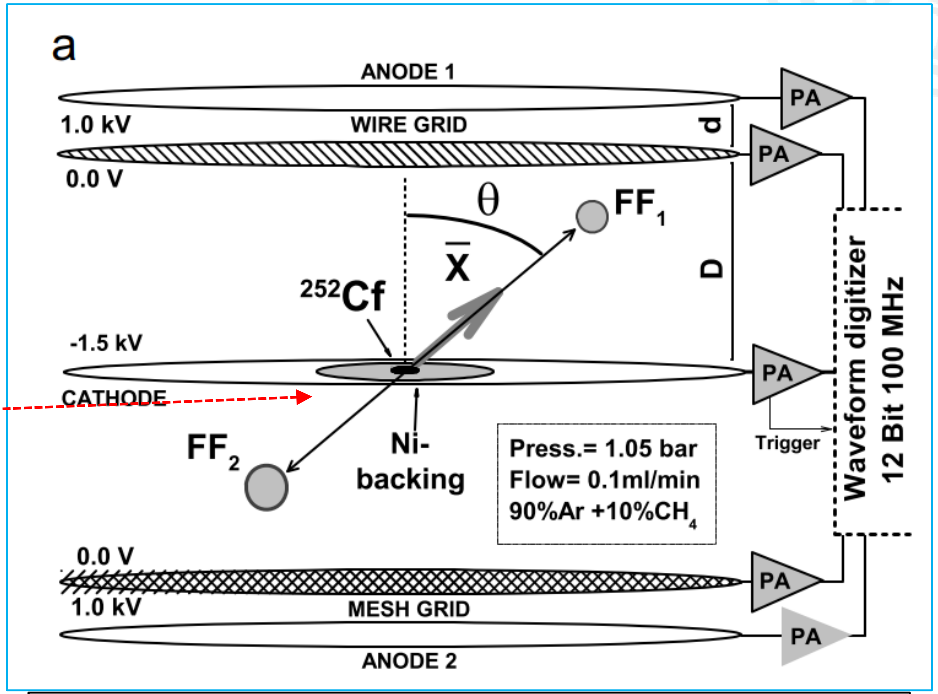
Experimental methods for measuring fission yields

Unstopped fragments:

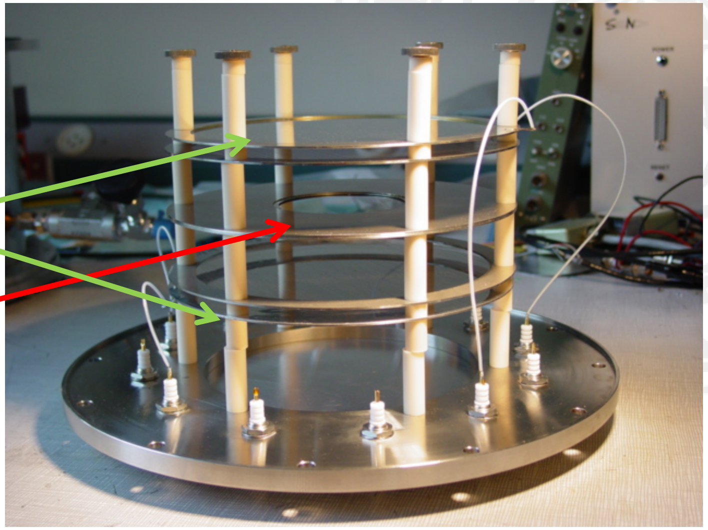
- More “small scale”, flexible experiments using
  - 2E method and 2v method
  - 2E-2v method
- Use of neutron beams (thermal, high energy) or sources (like ^{252}Cf)
- Need for suitable targets (sizeable amounts).
- Example: Ionization chambers like the “Twin Frisch-Grid Ionization Chamber” (TFGIC)
- Goal: measure kinetic energy and/or velocity of both fission product and extract information about fragment mass, angular distribution, TKE, etc.



A. Al-Adili et al. Nucl. Inst. Meth. A **671**, 103 (2012)

Unstopped fragments: 2E-method

Inside of a FGIC:



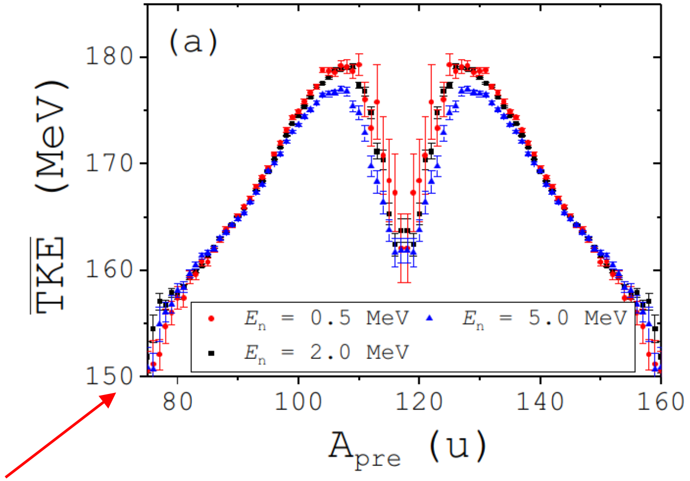
Frisch-Grids and Anodes

Target (Cathode, backing)

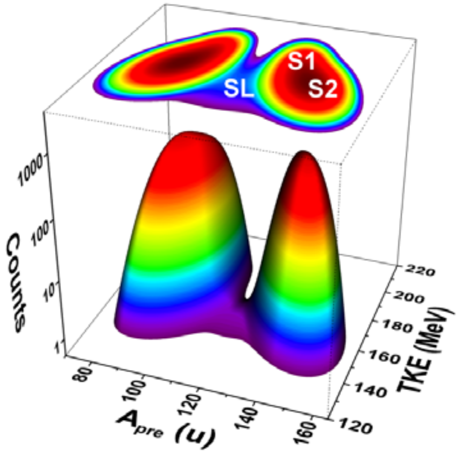
Used with e.g. a Frisch-Grid Ionization Chamber.

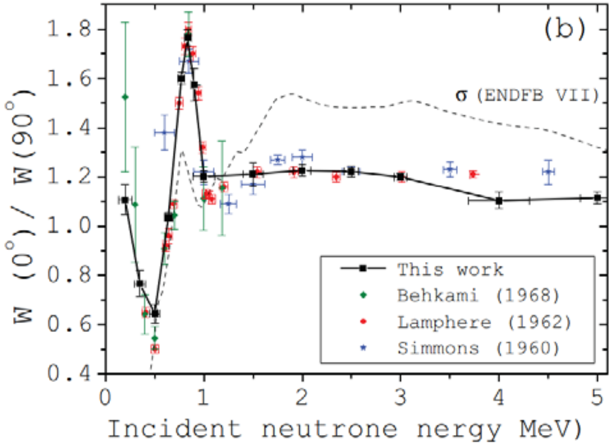
Frequently and over a long time used at JRC-Geel.

Example of results:



Not a mass distribution ☺





A. Al-Adili et al. Phys. Rev. C **93**, 034603 (2016)

How it works: 2E-method

- If the **kinetic energies** E_1^{pre} and E_2^{pre} of the *two* fission fragments are measured (like via a pulse-height defect corrected signal in a ionization chamber), the fragment masses can be obtained from momentum conservation (in the c.m. system).

$$\boxed{\boldsymbol{m_{1,2}^{pre} = m_{CN} \cdot \frac{E_{2,1}^{pre}}{E_1^{pre} + E_2^{pre}}}}$$

It's largely ok to ignore relativistic effects …

Problem:

Measured: $E_{1,2}^{post}$ $\longrightarrow$ need to make an *assumption* on the energy change due to neutron emission.

2E-method

Use **iterative procedure** assuming kinetic energy scales with mass of fragment/product to get better value for the fragments energy:

neutron multiplicity as function of fragment mass
(input from some model)

$$\boldsymbol{m_{1,2}^{post}(i+1) = m_{1,2}^{pre}(i) - \overset{\downarrow}{\nu}_{1,2}\left(m_{1,2}^{pre}\right)}$$

$$\boldsymbol{E_{1,2}^{pre}(i+1) = E_{1,2}^{post} \cdot \left(\frac{m_{1,2}^{pre}(i)}{m_{1,2}^{post}(i+1)}\right)}$$

$$\boldsymbol{m_{1,2}^{pre}(i+1) = m_{CN} \cdot \frac{E_{2,1}^{pre}(i+1)}{E_{1}^{pre}(i+1) + E_{2}^{pre}(i+1)}}$$

Do until changes are small

2v-method

- Similar to the previous method but instead of the energies, velocities are measured.
- Assuming we have the fragment (pre neutron-emission) velocities, **momentum conservation** gives:

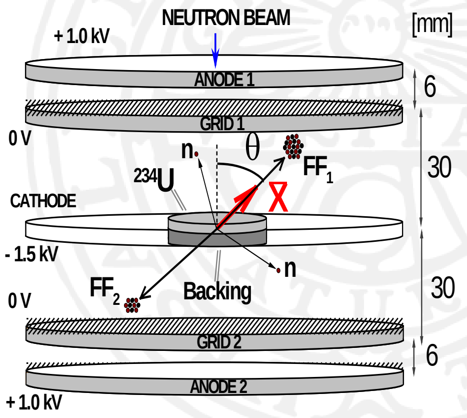
$$\boldsymbol{m_{1,2}^{pre} = m_{CN} \cdot \frac{v_{2,1}^{pre}}{v_1^{pre} + v_2^{pre}}}$$

How to?
Use, e.g. PPACs (Parallel Plate Avalanche Chambers) as start and stop detectors.
Challenge: energy losses …

And same problem as in 2E-method:

Measured: $v_{1,2}^{post}$ –→ need to *assume* that velocity is unchanged by neutron emission and use an iterative procedure …

Experiments – 2E with FGIC (as mentioned above)

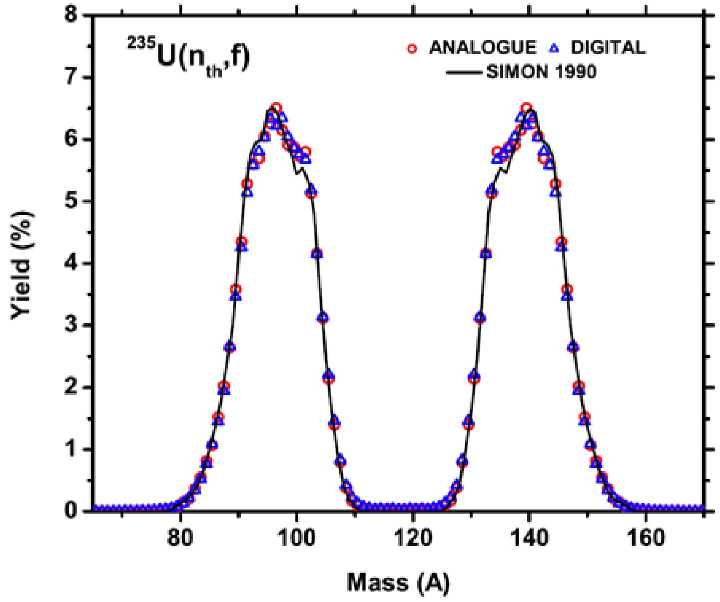


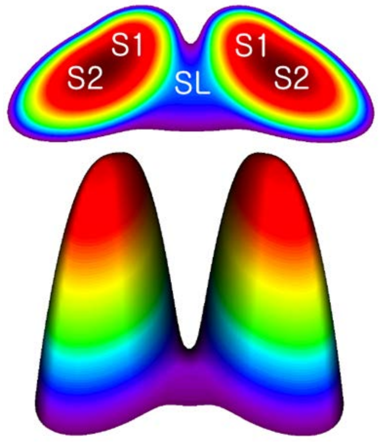
Plus:

- Large geometrical efficiency (~4π).
- Several observables can be measured (TKE, FY, σ_f, ang. dist., …).
- Relatively simple operation and analysis (using momentum conservation and assumption on the compound mass).

Minus:

- Rather poor mass resolution (4-6 u). This is ”corrected” via unfolding.
- Assumption on prompt-neutron emission needed.





Mass vs TKE

See, e.g., A. Al-Adili et al.,
Phys. Rev. C **93**, 034603, (2016)

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2E-2v-method – of course we can combine the two

Measurement of energy and velocity of both products.

Post neutron-emission masses are readily obtained:

Hence we can get number of emitted neutrons.
But we need to put in some work to find out form which fragment they are emitted.

$$m_{1,2}^{post} = \frac{2 \cdot E_{1,2}^{post}}{\left(\mathrm{v}_{1,2}^{post}\right)^2}$$

Like in 2v-method:

assuming that, on average, velocities are unchanged by neutron emission,
pre neutron-emission masses are obtained (momentum conservation):

$$m_{1,2}^{pre} = m_{CN} \cdot \frac{\mathrm{v}_{2,1}^{pre}}{\mathrm{v}_{1}^{pre} + \mathrm{v}_{2}^{pre}}$$

Not really true … See:
K. Jansson, et al., Eur. Phys. J. A **54**, 114 (2018).
https://doi.org/10.1140/epja/i2018-12544-0

2E-2v-method

Now comes *the thing*:

$\overline{\nu}(\boldsymbol{m_{1,2}^{pre}})$ can be obtained (!):

$$\boxed{\overline{\boldsymbol{\nu}}_{\mathbf{1,2}} = \frac{\boldsymbol{m_{1,2}^{pre}} - \boldsymbol{m_{1,2}^{post}}}{\boldsymbol{m}_{\mathbf{n}}}}$$

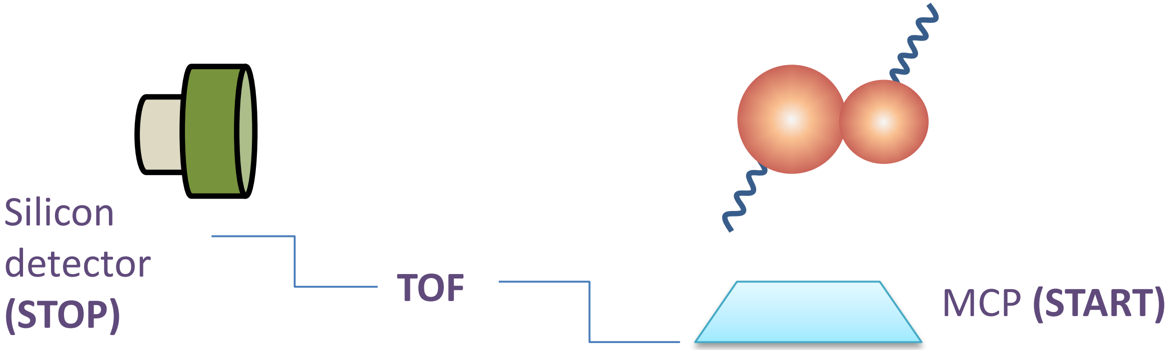
(event-by-event)

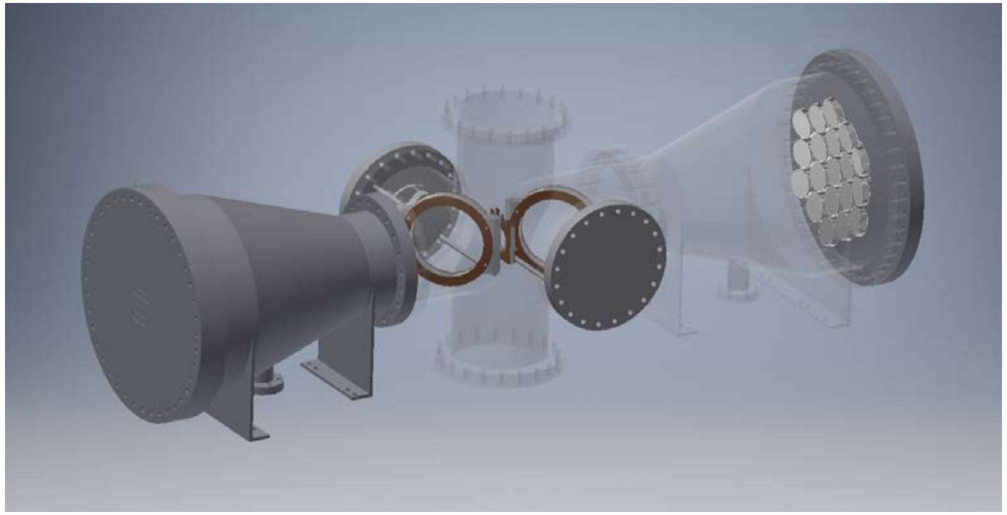
and also:

$$\boxed{\boldsymbol{E_{1,2}^{pre}} = \frac{\mathbf{1}}{\mathbf{2}}\,\boldsymbol{m_{1,2}^{pre}}\,\left(\mathbf{v}_{\mathbf{1,2}}^{\boldsymbol{post}}\right)^{\mathbf{2}}}$$

I.e.: ideally, you “measure” prompt-neutron multiplicity
without the need to actually measure (count) the neutrons ☺

2E-2v with VERDI: Velocity foR Direct particle Identification





Energy

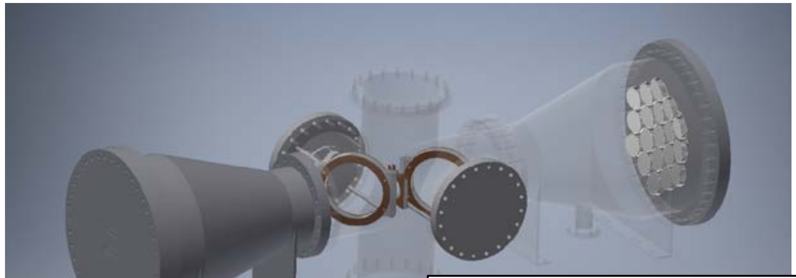
2 arrays of Si detectors,

Velocity (Time-of-Flight)

Start: Electrons emitted from target detected by Micro Channel Plate (MCP)

Stop: Si detector

2E-2v with VERDI



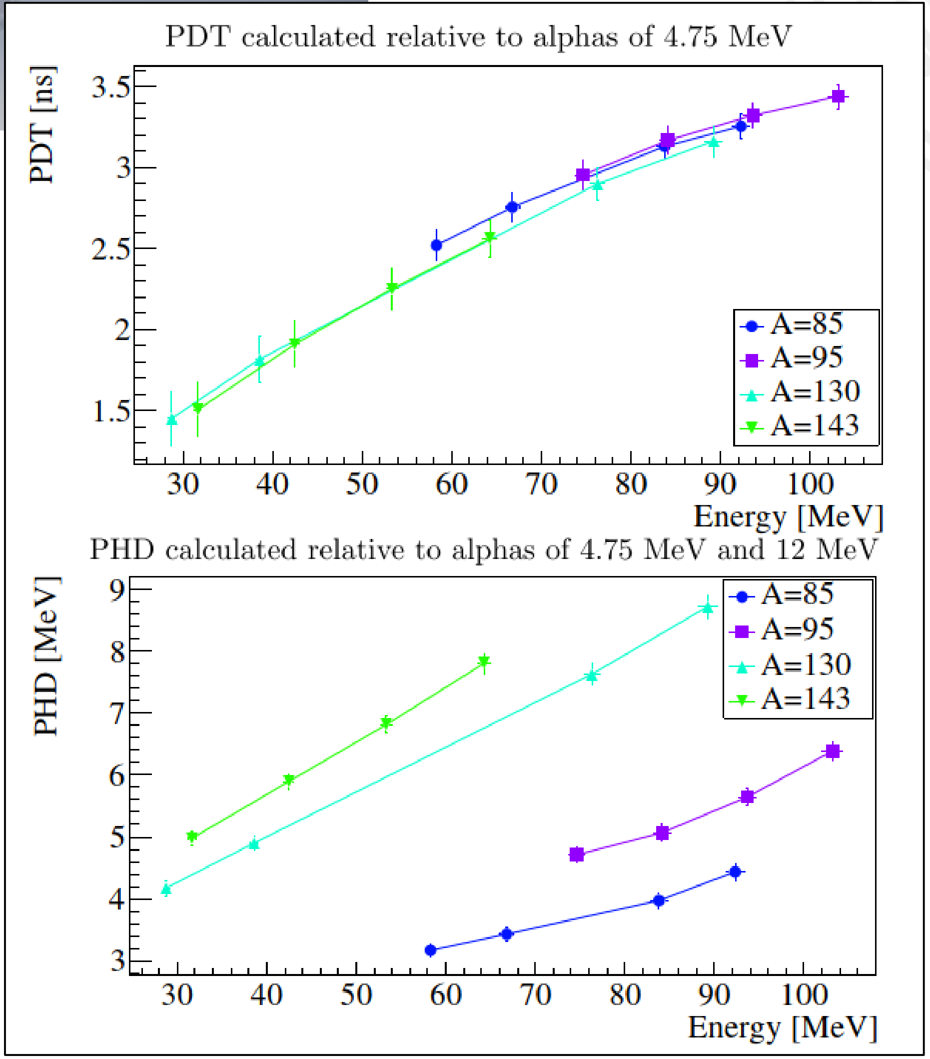
Plus:

- Good mass resolution (1-2 u) possible.
- No assumption on neutron emission needed. -> $\overline{\nu}(A)$ can be obtained!

Minus:

- <1% geometrical efficiency.
- Rather complicated/folded analysis.
- To reach goal very good understanding of Si detector response is needed!
- To this end: Plasma delay time (PDT) and pulse height defect (PHD) must be characterized.

Some, still preliminary, results from a Si detector study with LOHENGRIN at ILL



A.M. Gómez et al, EPJ Web of Conf **284**, 04012 (2023)

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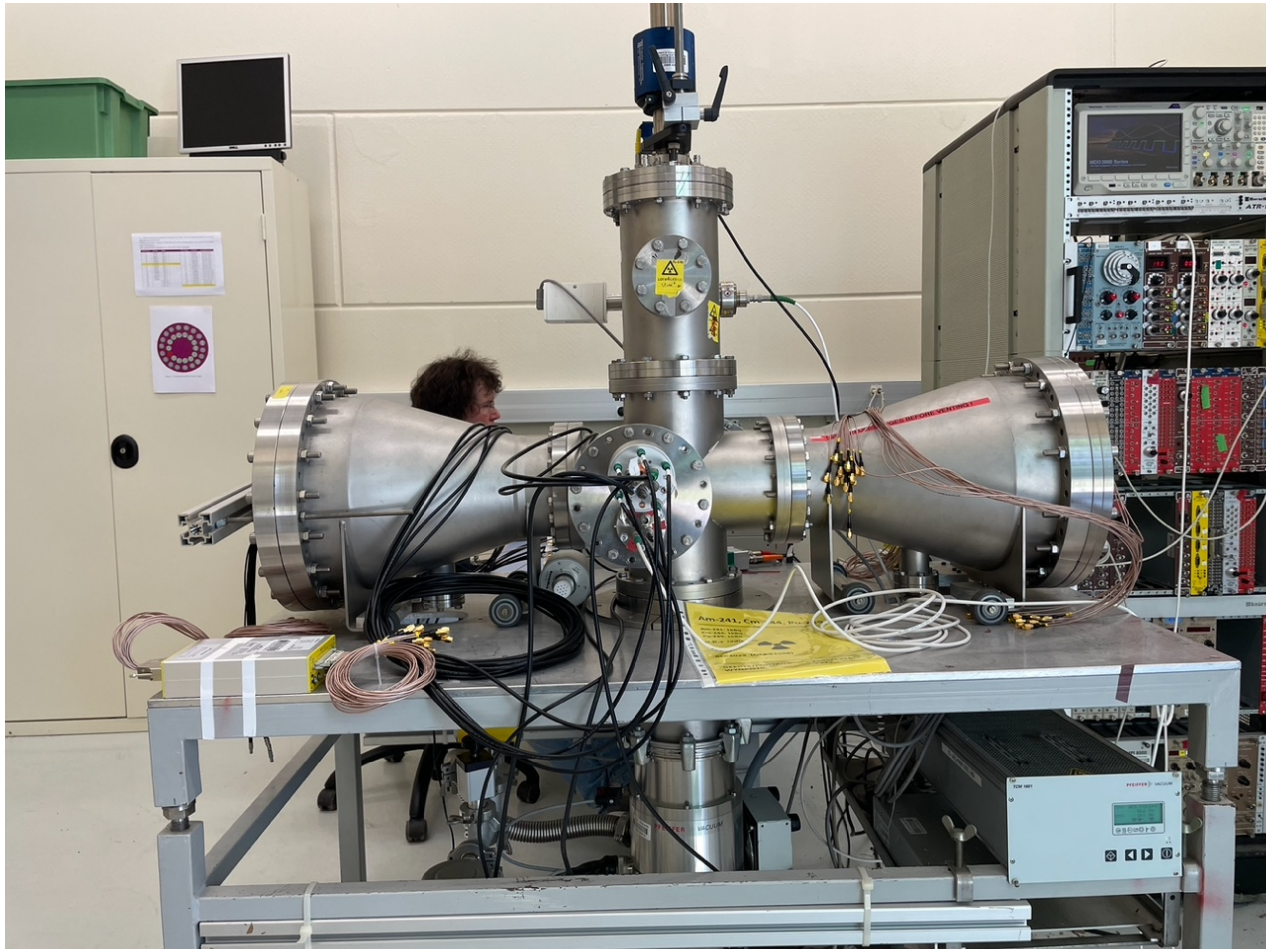
VERDI IRL (JRC Geel, Belgium)


Picture courtesy Ali Al-Adili

This is still a small scale experiment.

It can be
- handled by a small group of scientists
- relatively easily moved to different suitable facilities.

Example of a state-of-the-art experiment.

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Slide courtesy Diane Doré

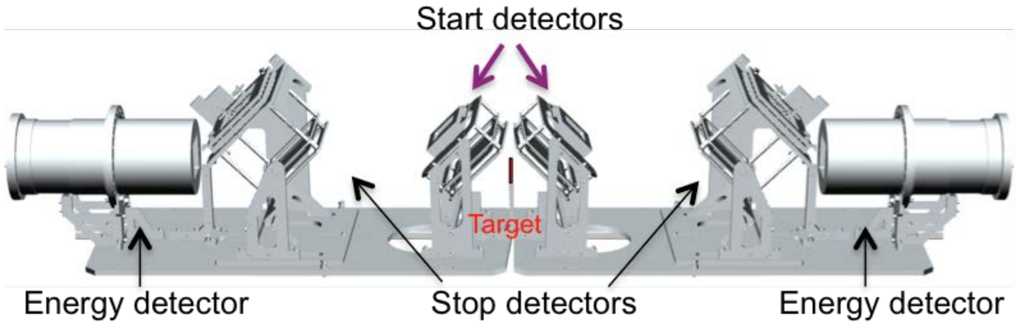
FALSTAFF
Four arm cLover for the Study of Actinide Fission Fragments

- Spectrometer for fission fragment detection in coincidence
  - Kinetic energy
  - Masses BEFORE and AFTER evaporation (-> ν(A))
  - Charge

- Mass **before** evaporation → 2V method
  TOF : *Good time resolution (σ) <150 ps*

- **Large solid angle (~1% of 4π)**
- **Good position resolution (1.2 mm)**

- Mass **after** evaporation → EV method
  Energy & TOF
  - *Good energy resolution (~1%)*
  - *Charge identification (dE profile)*



So far: only one of the arms constructed.
Currently at GANIL, France

Other ongoing developments and an example from history

- **SPIDER** (Los Alamos National Lab): see, e.g., P. Gastis et al., Nucl. Inst. Meth. A **1037**, 166853 (2022)
- **STEFF** (Univ. of Manchester): see, e.g., A. Ryan, Ph.D. thesis, University of Manchester (2017)

Historically:

COSI VAN TUTTE:

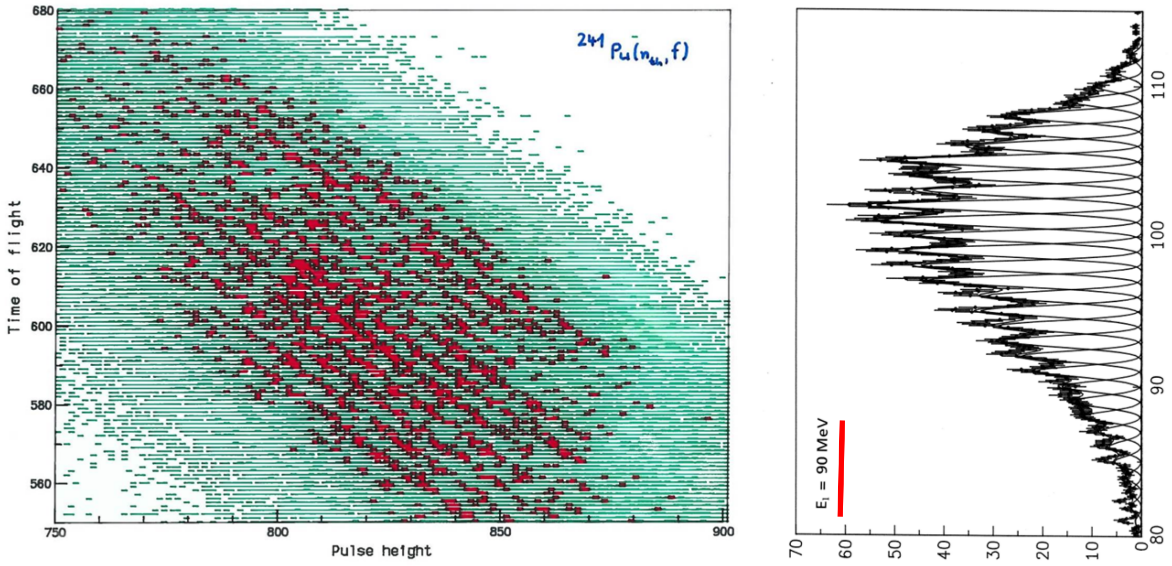
A. Oed et al. NIM **219** (1984) 569

E-v method, one fragment only,

hence post-masses,

very good time resolution,

$\Delta m/m = 0.6\%$ achieved.



Figures courtesy P. Schillebeeckx (PhD thesis)

Stopped fragments: Penning Trap - IGISOL

Now something “completely” different.

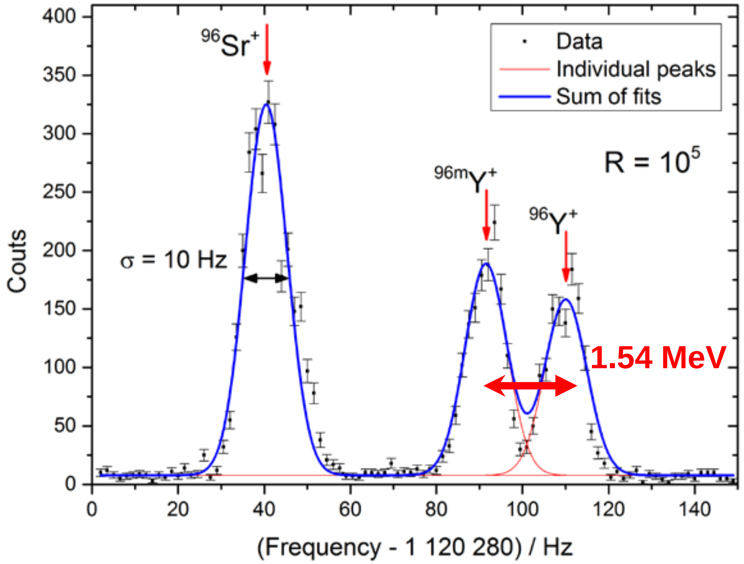
One can use **mass measurement techniques** to identify independent fission yields:

- isotope separation online technique (ISOL),
- Followed by, e.g., Penning traps or a Multi-Reflection Time-Of-Flight spectrometer (MR-TOF).

Precis mass measurements:

- mass resolving power greater than 10^6 achieved and mass differences <100 keV are thus resolved
- possibility to measure yields (and even isomeric yields!) by direct ion counting

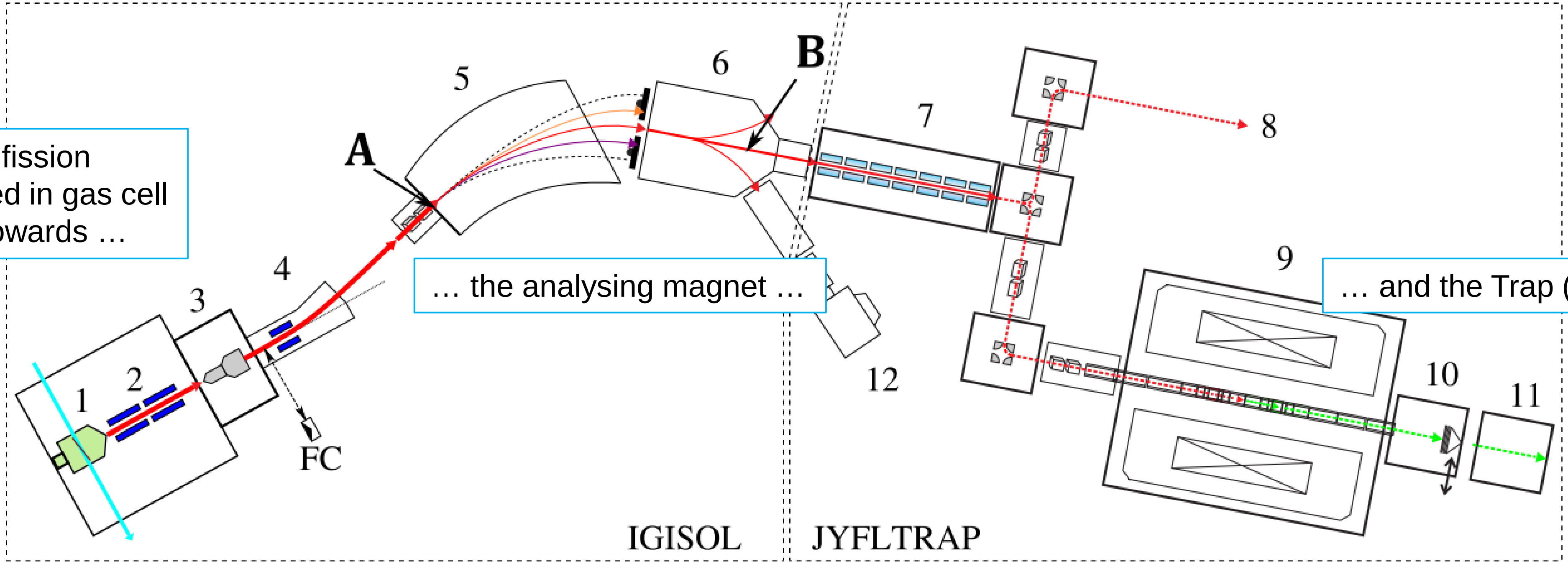
Figure: IGISOL + JYFLTRAP in Jyväskylä, Finland



H. Penttilä et al., Eur. Phys. J. A **44**, 147 (2010)

Fission yields from Penning traps

(p,f), (n,f), (α,f): fission products stopped in gas cell and extracted towards …



… the analysing magnet …

… and the Trap (9) + MCP (10).

Fig. 1. Schematic layout of IGISOL-4, ground floor. (1) ion guide, (2) sextupole ion guide (SPIG), (3) extraction electrodes, (4) bending electrodes, (5) 55° dipole magnet, (6) switchyard, (7) radiofrequency cooler-buncher, (8) beam line towards a collinear laser spectroscopy setup, (9) Penning trap (JYFLTRAP), (10) microchannel plate (MCP) detector, (11) post-trap spectroscopy setup, (12) β-γ spectroscopy setup. FC is the Faraday cup used to measure the extracted ion guide current in this work. Positions A and B are explained in the text.

D. Gorelov et al., Nucl. Inst. Meth. B **376**, 46 (2016)

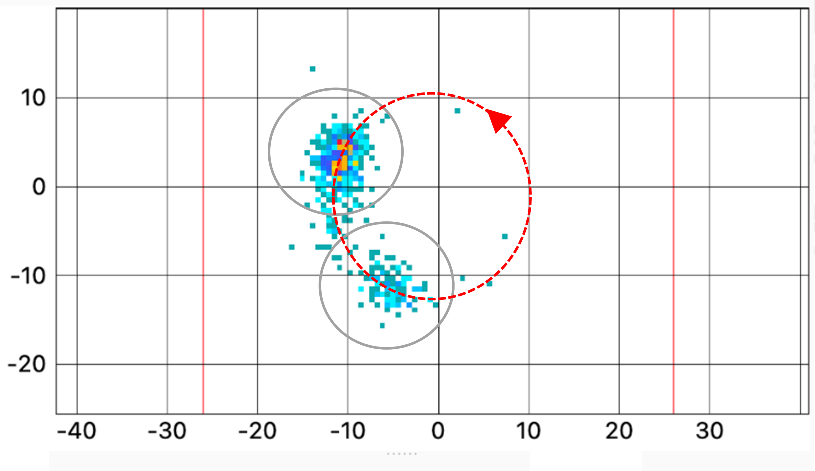
Fission yields from Penning traps

Plus:

- High mass resolving power gives isotopic fission yields (without need of measuring the charge).
- Even isomers can be resolved (especially with PI-ICR)
- Direct ion-counting (no γ-spectrometry needed)

Minus:

- Time from fragment production to detection can be several 100 ms
- If precursor is short lived: cumulative yield
- No correlation to other observables (TKE, etc)
- For fission yield: normalization needed (different chemistry of products)



Ground state (bottom) and isomer (top) in ^{129}**Sn**. Raw data from a measurement in March 2023 using the PI-ICR technique. Mass difference: **35 keV** (!).

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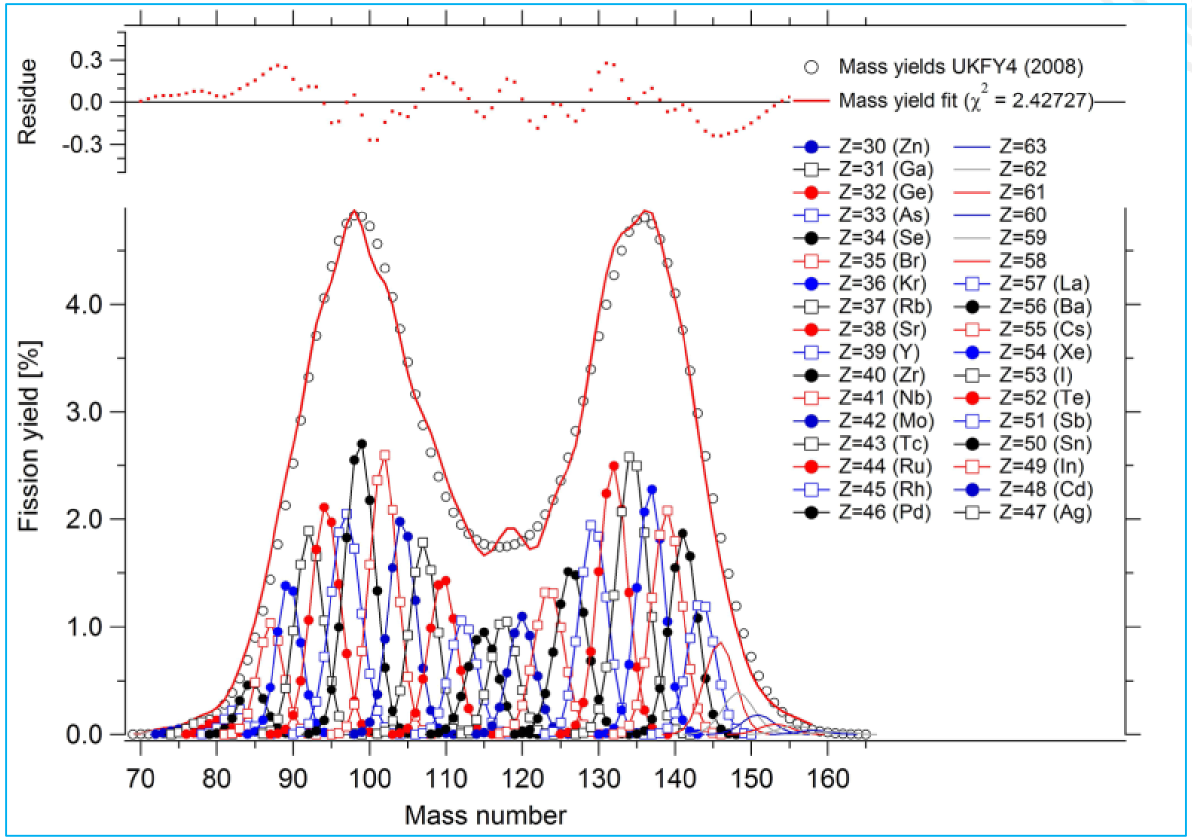
Independent isotopic yields from $^{nat}U(p,f)$ at 25 MeV

Isotopic yield distribution as obtained at IGISOL after adjustment to the yield distribution from the UKFY4 evaluation by R. Mills



H. Penttilä et al., Eur. Phys. J. A **52**, 104 (2016)

MR-TOF – Example from the CSC @ GSI, Darmstadt

Y. Waschitz et al., EPJ Web of Conf. **284**, 04005 (2023)

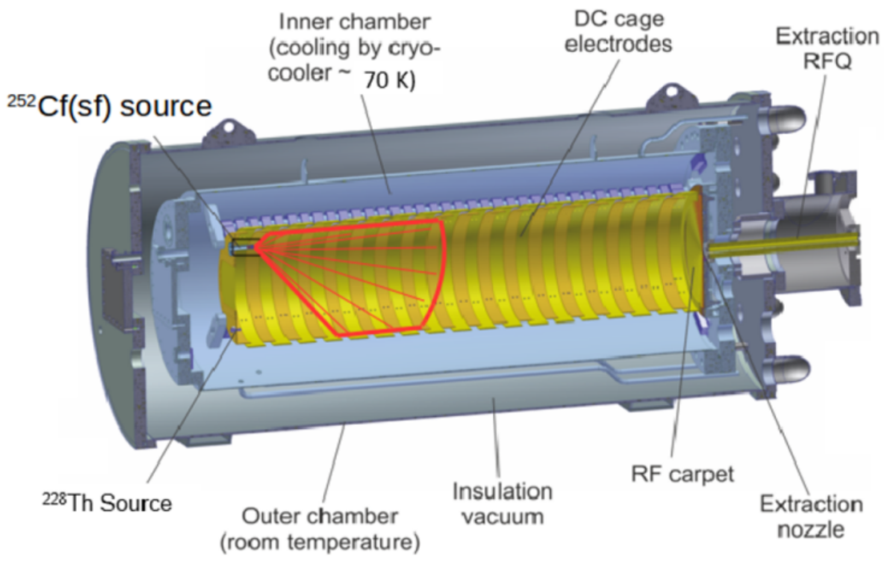
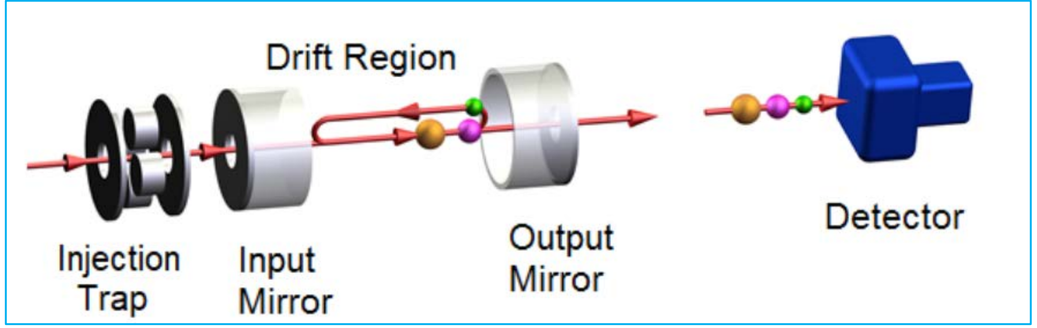
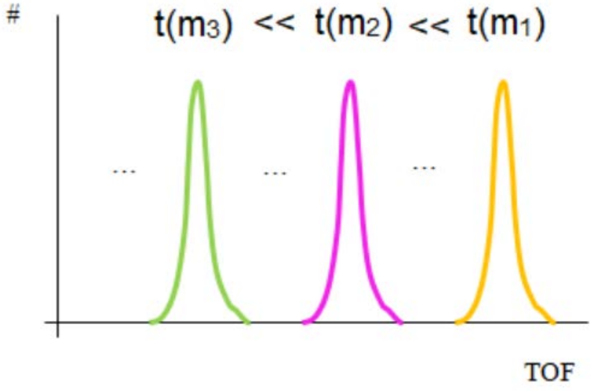


Figure 1. Cross section of the CSC with the internal long DC cage that is optimized for thermalizing relativistic ions. The ^{252}Cf SF source is installed at the upstream side of the CSC, 9 cm off-axis. The red lines mark the approximate range of the emitted FPs in the buffer gas.

Fission product from internal source (or induced via external beam on target) are stopped in the cryogenic stopping cell (CSC) and extracted towards an MR-TOF system.





Figures: PhD thesis
S. Ayet San Andrés,
Giessen 2018.

MR-TOF – Example from the CSC @ GSI, Darmstadt

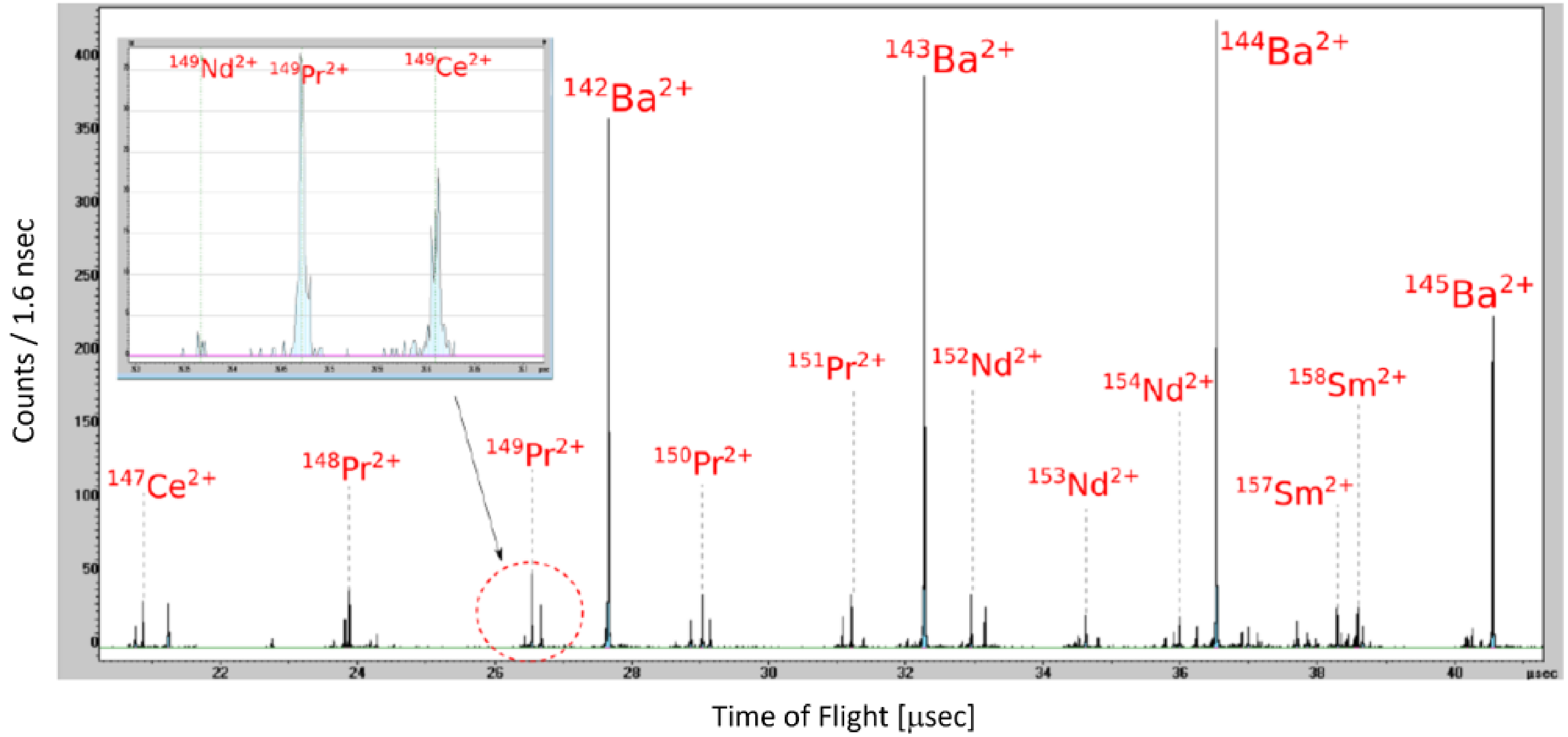


Figure 2. Typical time-of-flight spectrum during this experiment. All identified FPs are doubly charged and marked on the plot. The MRP is 320,000. The inset shows the clear separation between three A=149 isobars.

Y. Waschitz et al., EPJ Web of Conf. **284**, 04005 (2023)

Data from a recent measurement of ^{252}Cf(sf).

Mass resolving power: 320 000. Penning traps still better.

But: MR-TOF systems have achieved 10^6 in MRP.

Advantage vs. Penning Trap: Extraction from gas cell similar but mass measurement much faster (<ms).

Next up ....

Neutron and gamma emission in fission

A nice write-up on neutron and gamma emission in fission is provided by Friedrich Gönnenwein:
https://t2.lanl.gov/fiesta2014/school.shtml

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First: remember that …

- Neutron and γ emission is driven by

  the excitation energy of a fragment (incl rotational energy)

- Hence: low TKE -> high TXE -> more neutrons (and gammas)

  and vice versa

- Fission shape matters!

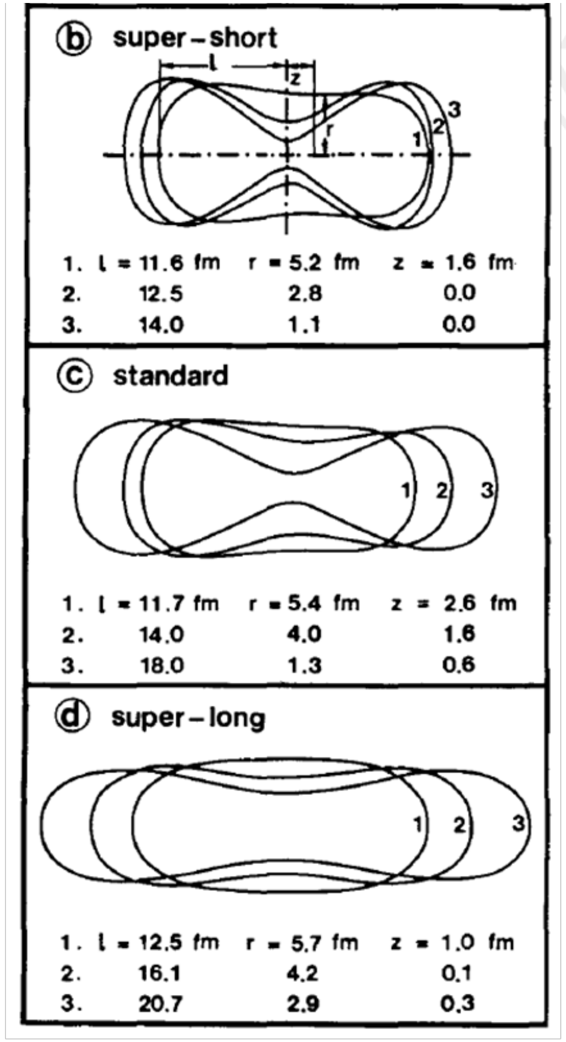
  E.g. super-long mode (important close to symmetry):

  Due to relatively large distance between fragments at

  scission the TKE is low.

  On the other hand:

  Elongated fragments -> large deformation -> high TXE



From: Brosa et al., *Phys. Rep.* **197** (1990) 167

(see presentation by O. Litaize for details)

Typical TXE (numbers fit best to thermal fission of U-235)

- On average we have (for thermal fission) about 25 MeV in the form of fragment excitation energy

- Where does this energy go? Some rough numbers:
  - The average total energy to **prompt gamma emission** is about 7 MeV (we will look at that later).
  - This leaves 18 MeV for neutron emission.
  - In the case of thermal fission of ^{235}U, the average **neutron separation energy** of the fragments is around 5.6 MeV.
  - Average **kinetic energy** of prompt neutrons is about 2 MeV (from PFNS).
  - That means **a typical neutron removes 7.6 MeV** (5.6 MeV + 2 MeV).
  - With 18 MeV available we **expect around 2.4 (prompt) neutrons per fission**.

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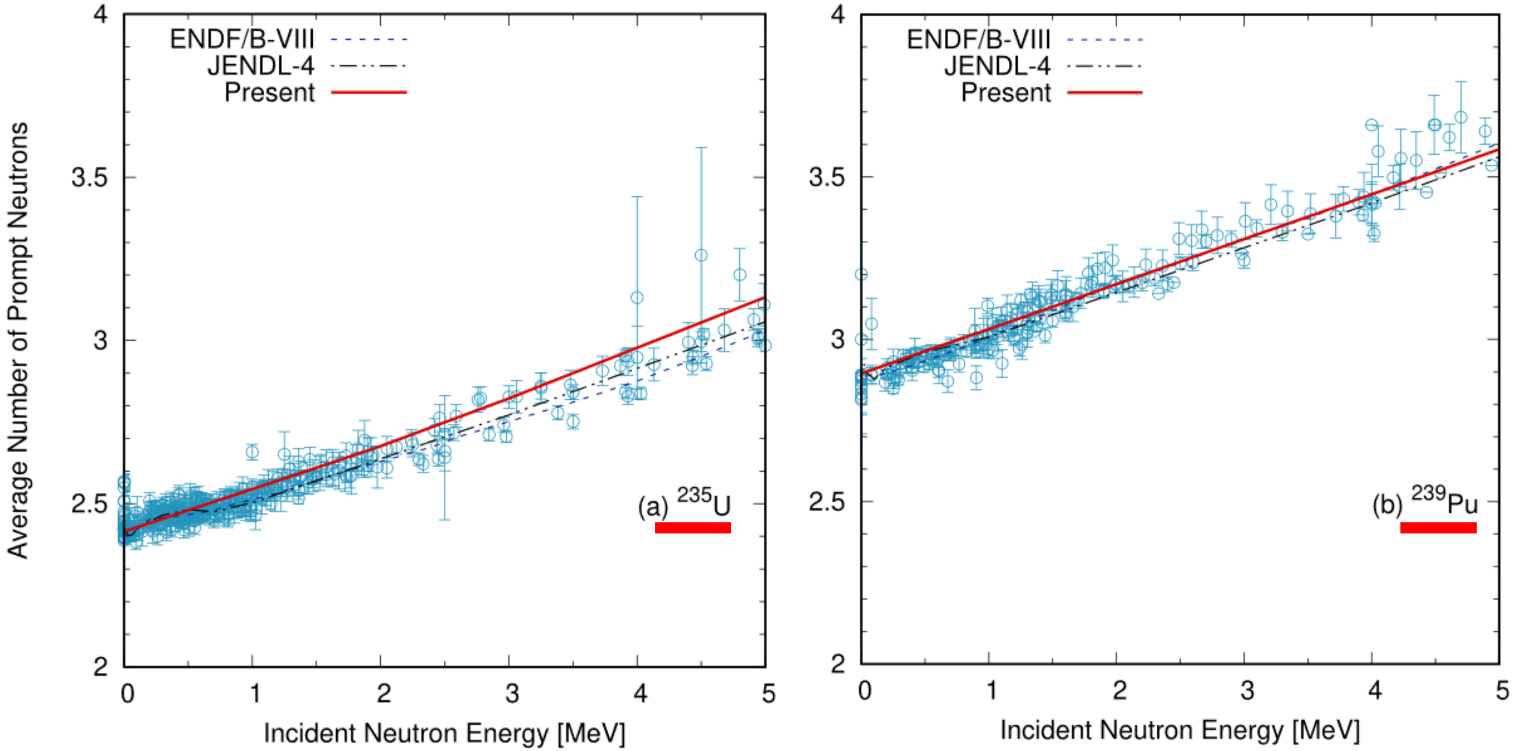
So how does it look? Neutron multiplicity (nubar)

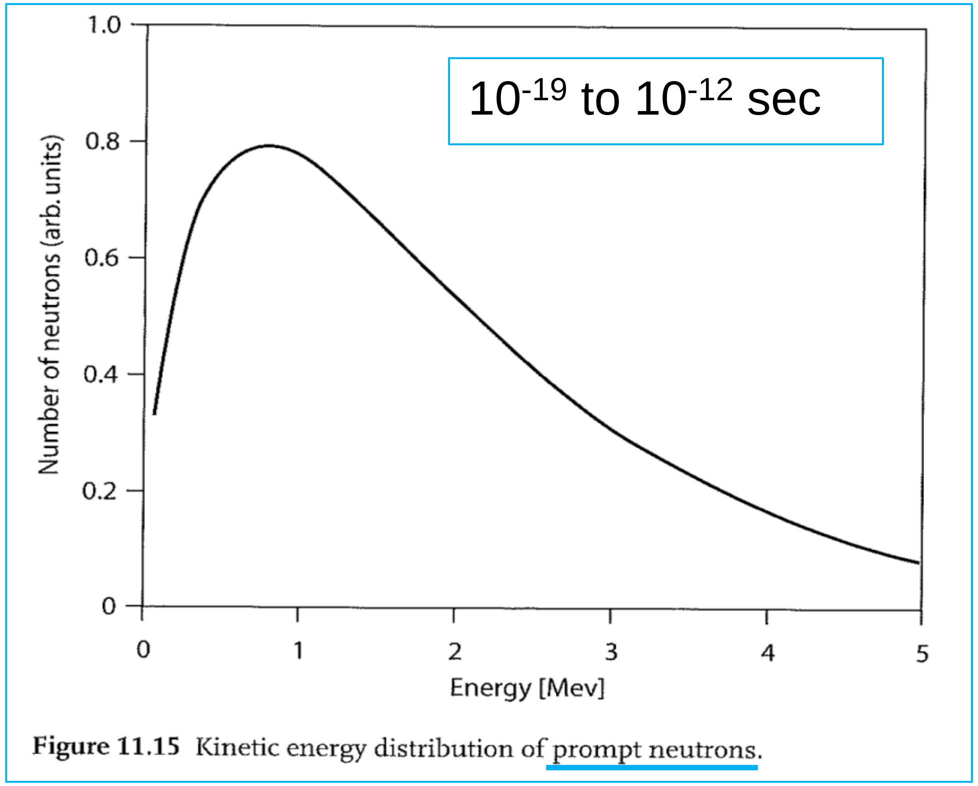


Multiplicity ($\bar{\nu}$) as function of *incoming* neutron energy.

Figures from S. Okumura, et al., Journal of Nuclear Science and Technology, 59:1, 96-109 (2022)
DOI: 10.1080/00223131.2021.1954103
https://arxiv.org/pdf/2102.01015.pdf

So yes (surprise), we get around this number for nubar (2.5-ish) for thermal fission.
Higher incident neutron energy (“high-energy” fission): more excitation energy, more neutrons …

Neutrons emission: kinetic energy spectra (PFNS)



10^{-19} to 10^{-12} sec

Figure 11.15 Kinetic energy distribution of prompt neutrons.

Energy distribution of **prompt** neutrons.
Peak around 1 MeV, average about 2 MeV.

Indicated is also the **timescale** for prompt neutron emission.

To good approximation neutrons are emitted from the fully accelerated fragments.

The shape is, to good approximation, a Maxwell spectrum, for evaporation of neutrons from a “hot” nucleus with a certain temperature (about 1.4 MeV)

Figure from C. Bertulani: Nuclear physics in a nutshell, Princeton University Press 2007

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What do experiments say?

For the case of ^{235}**U(n$_{th}$,f)**:

Overall, good agreement between measurements.

At high energies (low statistics!) some discrepancies seem to appear.

Deviations from a Maxwellian distribution can be seen.

It is found that the **average neutron energy** is very close to 2.00 MeV,
This is the value recommended of the IAEA.
Trkov et al., Nucl. Data Sheets 123, 8 (2015)

Göök et al., PRC **98**, 044615 (2018)

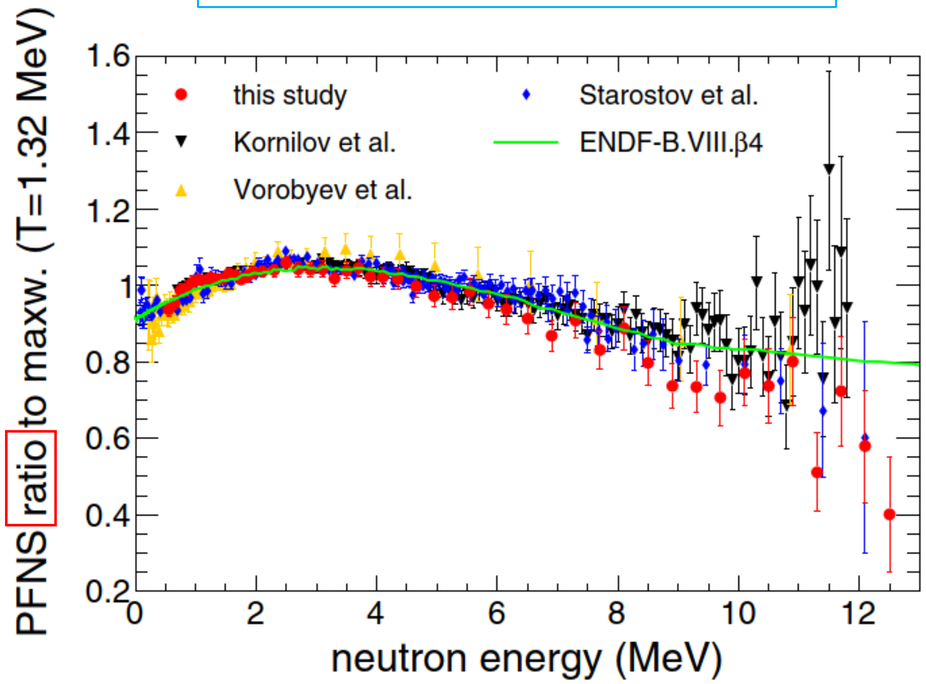


FIG. 8. The laboratory prompt fission neutron spectrum presented as a ratio to a Maxwellian distribution. Data from this study are compared to data from Refs. [33–36] and evaluated data from Ref. [31].

Two recent PFNS measurements for ^{235}U(n_{th},f):

Göök et al., PRC **98**, 044615 (2018)

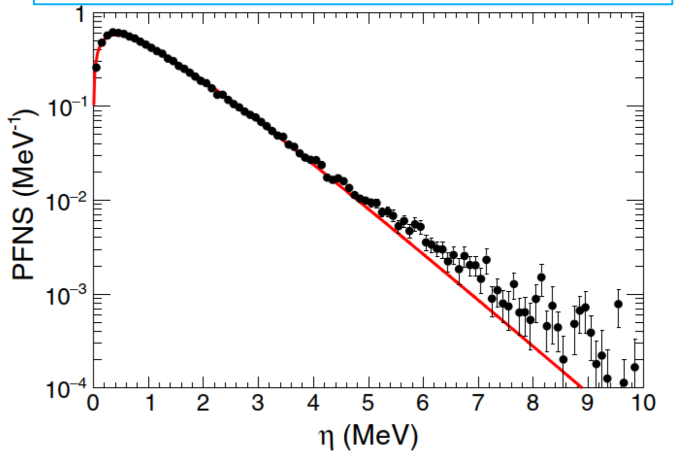
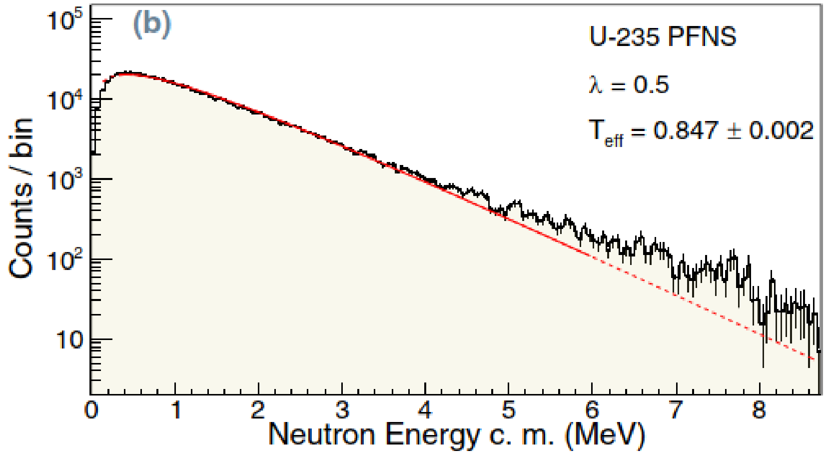
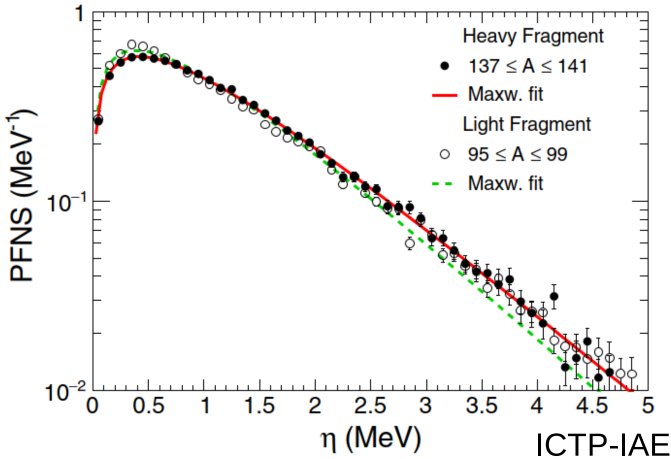


FIG. 9. Integral prompt fission neutron spectrum in the c.m. frame. The full red line represents a Maxwellian spectrum with fitted temperature (0.831 ± 0.005) MeV.

Al-Adili et al., PRC **102**, 064610 (2020)





Measured at JRC in Geel with two different setups.

Reasonable agreement between both measurements.

Note: different PFNS for light/heavy fragment.
Effect of nuclear structure, energy sharing, …

Delayed neutron emission

- ✓ Multiplicity (nubar)
- ✓ Energy distribution (PFNS)
- ❖ Delayed neutrons!

Delayed neutron emission depends on the decay scheme for a fission product (post *prompt*-neutron emission).

The delayed neutron fraction depends, therefore, on yields ☺, and differs with the fissioning system, the incident neutron energy, etc.

Delayed neutron fractions (JEFF-3.3):

^{235}U(n_{th},f):	0.668%	(total: 2.4253, delayed: 0.0162)
^{239}Pu(n_{th},f):	0.227%	(total: 2.8683, delayed: 0.0065)
^{241}Am(n_{th},f):	0.139%	(total: 3.1018, delayed: 0.0043)

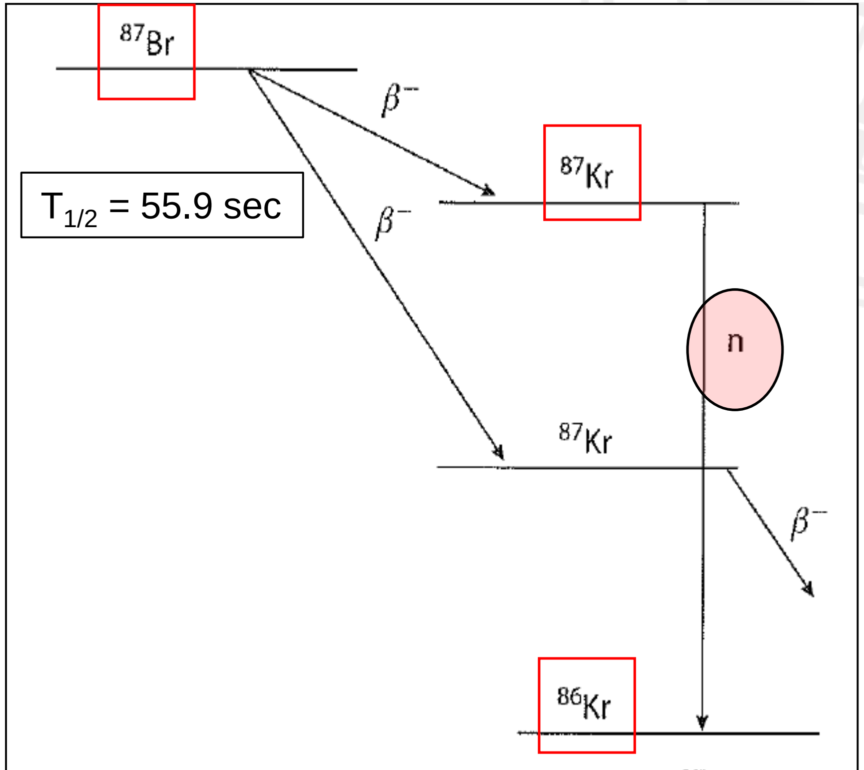
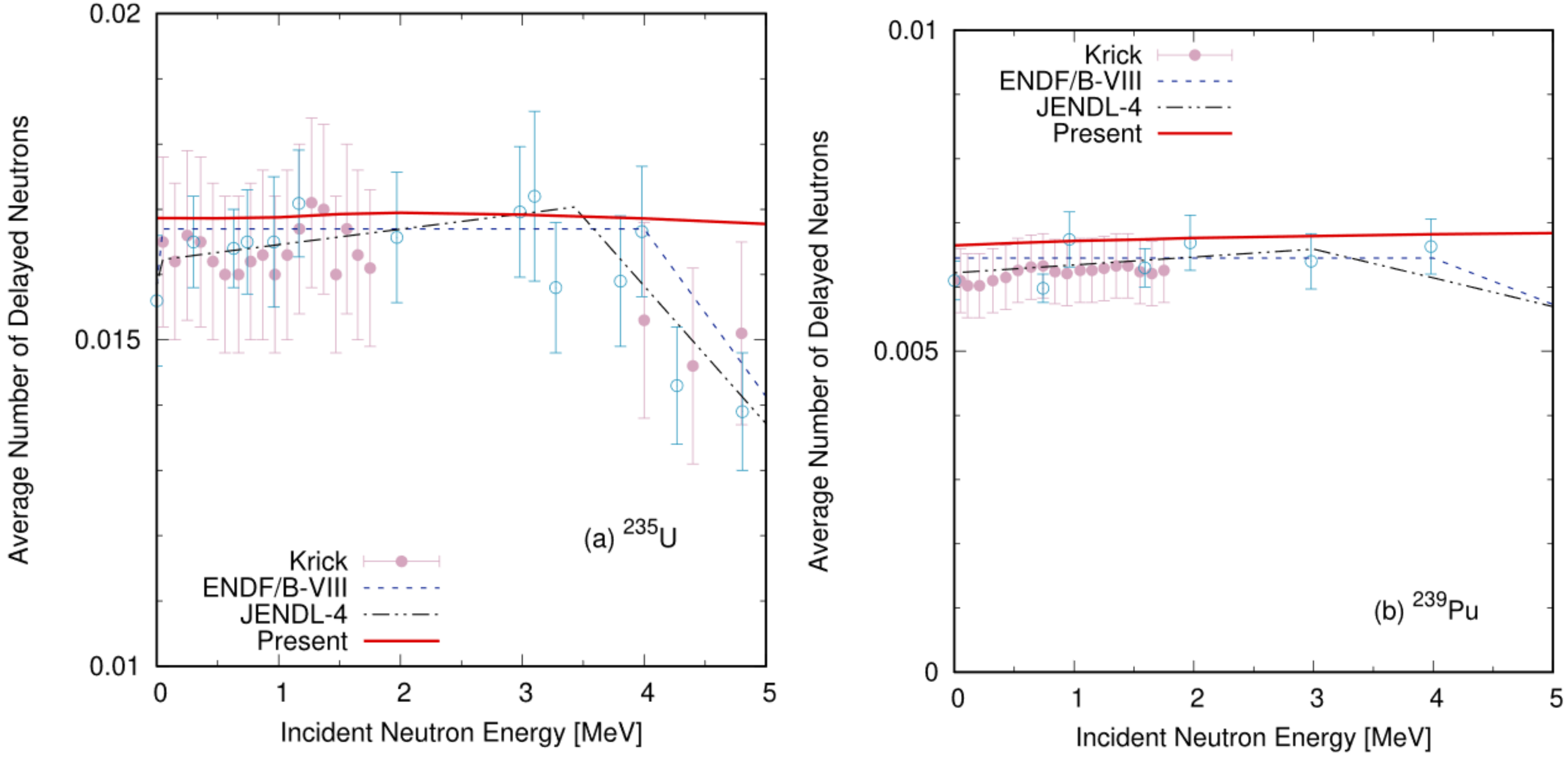


Figure 11.14 Emission of a delayed neutron in ^{87}Br decay.

Figure from C. Bertulani: Nuclear physics in a nutshell, Princeton University Press 2007

Delayed neutron emission – changes with incident energy



Figures from S. Okumura, et al.,
Journal of Nuclear Science and Technology,
59:1, 96-109 (2022)
DOI: 10.1080/00223131.2021.1954103
https://arxiv.org/pdf/2102.01015.pdf

Prompt neutron emission: the details …

Fission mode -> fragment shape -> neutron emission

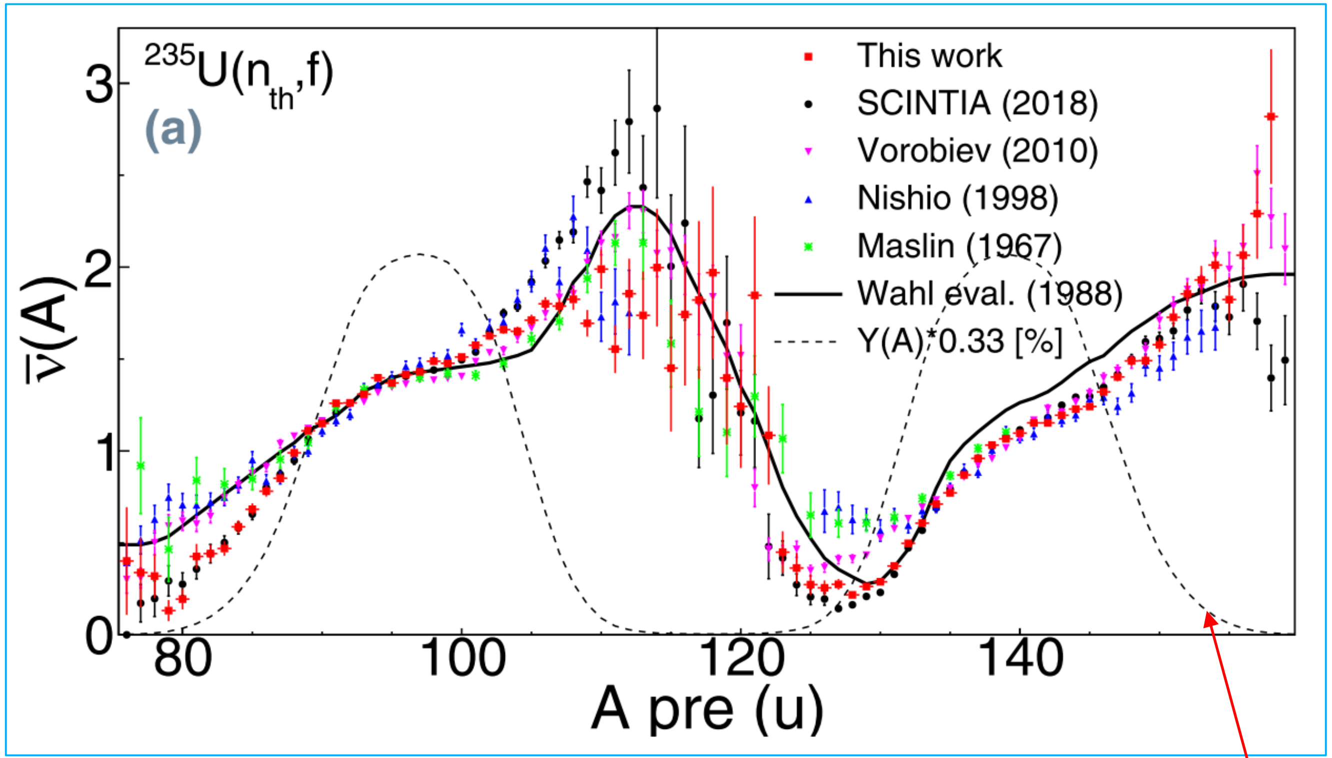


Figure from Al-Adili et al., PRC **102**, 064610 (2020)

Yield distribution for reference

The figure shows a measurement of the famous “sawtooth”, linking (average) neutron multiplicity to fragment mass (pre-neutron emission).

While the *total* neutron multiplicity from fission is pretty well known, the neutron multiplicity per fragments mass is less well-known.

The figure illustrates this …

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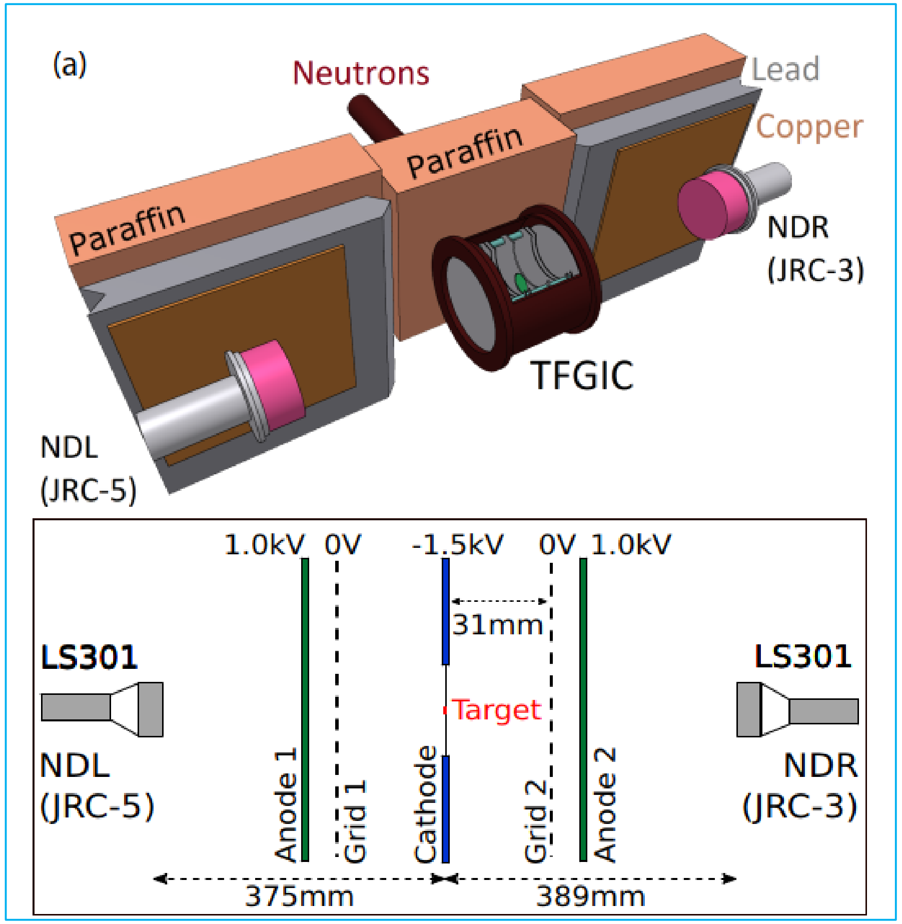
Neutron emission – an experiment

Al-Adili et al., PRC **102**, 064610 (2020)



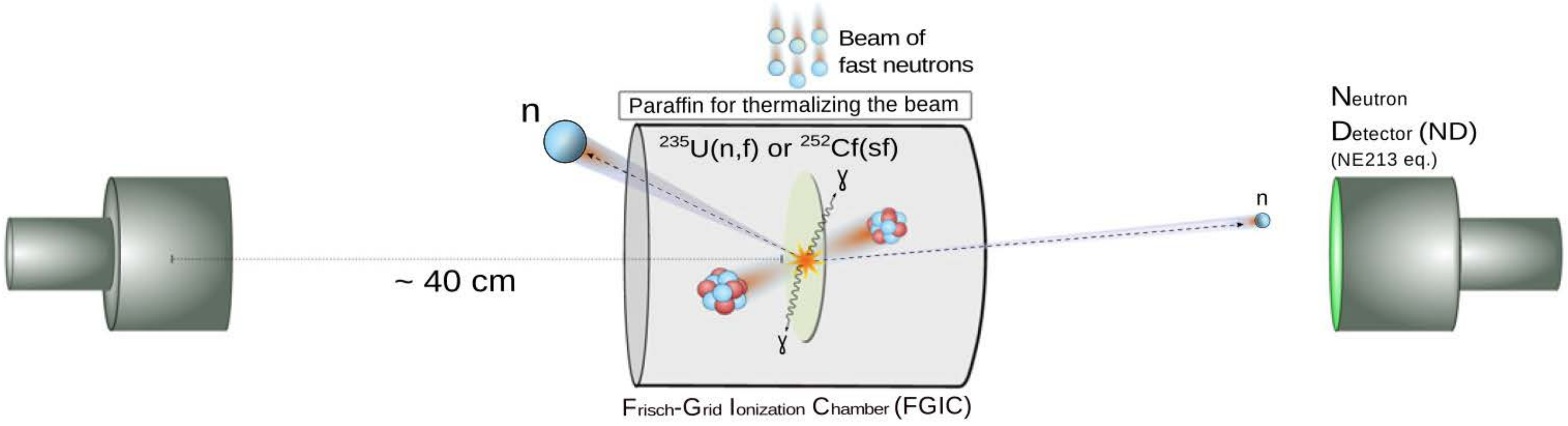
Neutrons with E_n = 0.5 MeV produced via ^{7}Li(p,n) at the (former) Van de Graaf accelerator of JRC Geel.

The neutrons are thermalized with 12 cm paraffin.

The setup needed **extensive simulations** (FLUKA) to optimize the shielding;

Wanted:
- good neutron flux at target position
- low flux at the position of the neutron detectors
- reasonable count rate

Neutron emission – an experiment



A. Al-Adili et al, EPJ Web of Conf **146**, 04056 (2017)

Fission trigger from the TFGIC (Twin Frisch-Grid Ionization Chamber).

The TFGIC measures fragments masses and TKE with the **2E-technique.**

Two **liquid scintillator detectors** (ND) register the neutrons in coincidence with a fission event in the TFGIC.

The ND can **discriminate background** γ-radiation and thus identify and count neutrons.

Neutron energy obtained from **time-of-flight** (TOF).

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Neutron emission – an experiment

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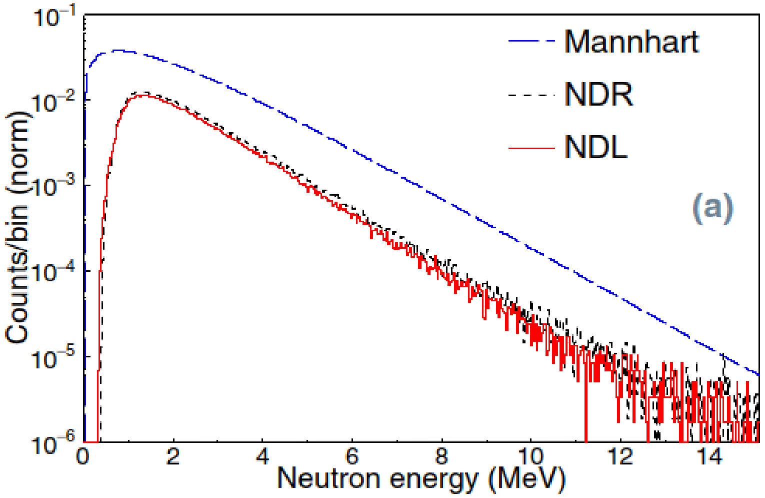
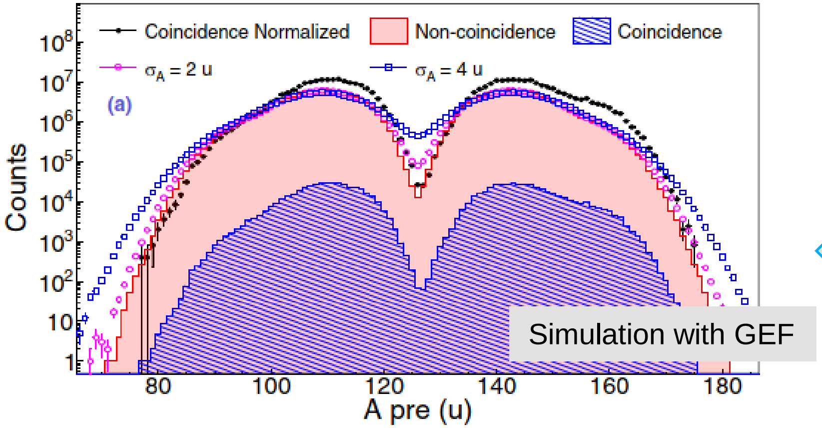




Simulation with GEF

Advanced data analysis considering, e.g.:

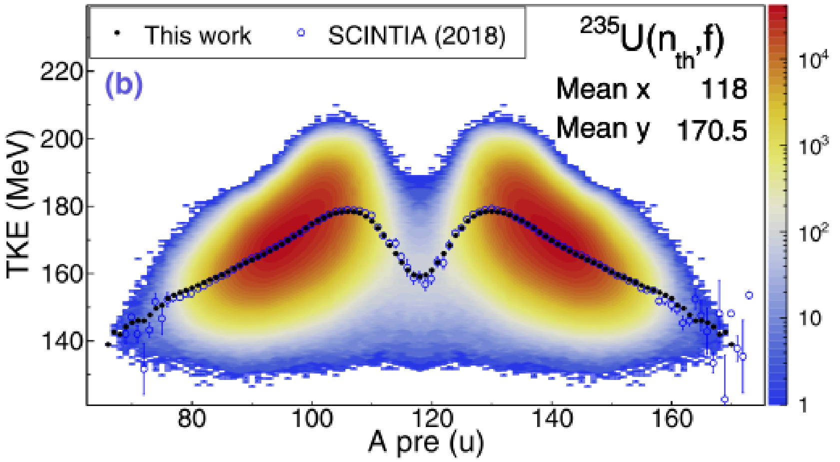
- **neutron detection efficiency**: comparison of measured distribution from ^{252}Cf(sf) with a reference PFNS spectrum (Mannhart evaluation).

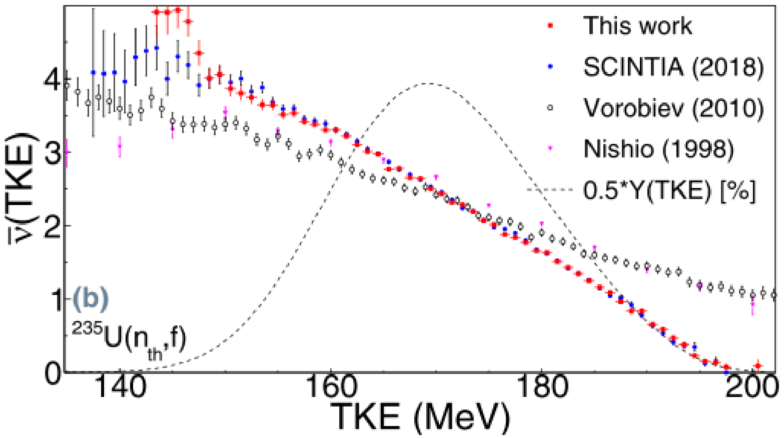
- **kinematics:** Neutrons are emitted from moving fragments and receive a boost in forward direction in the lab system. This helps in associating a registered neutron with a fragment mass (as measured in the TFGIC).

- **coincidence vs. non-coincidence:** the ratio of mass distribution measured with the TFGIC with and without registering a neutron in a NDR yields the neutron multiplicity as a function of fragment mass.

Neutron emission – an experiment

Al-Adili et al., PRC **102**, 064610 (2020)





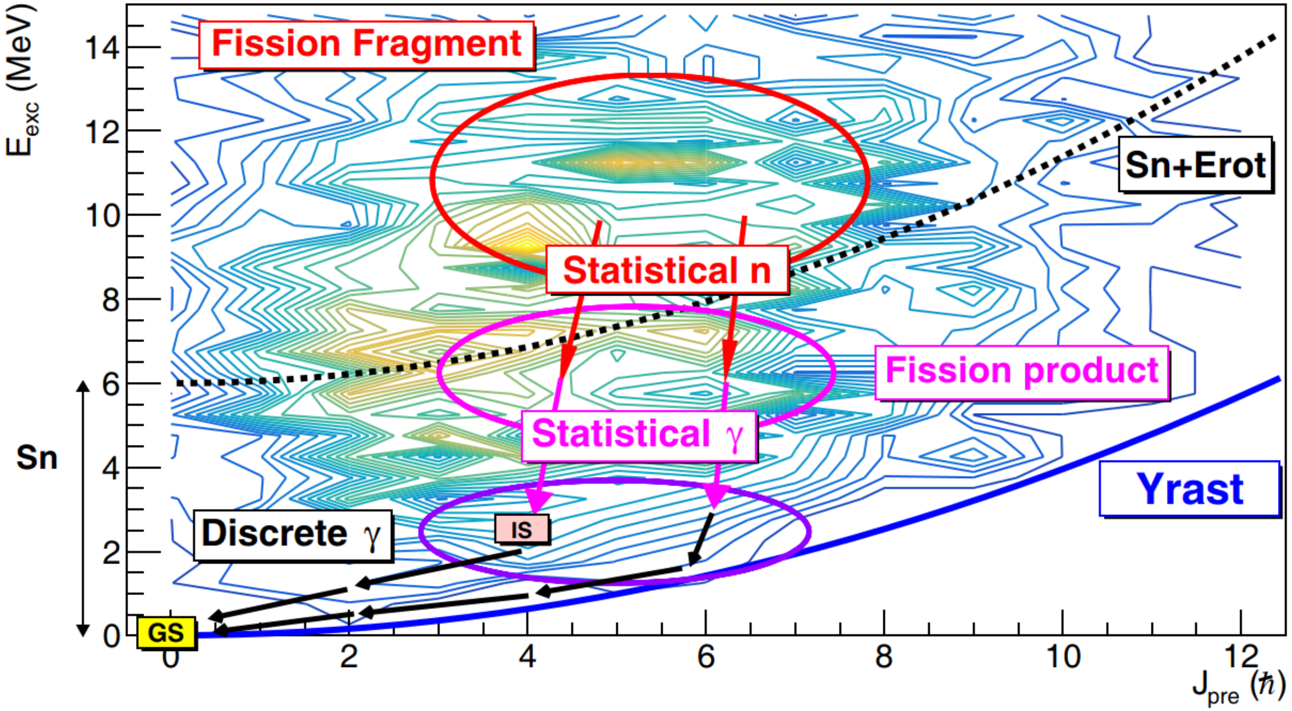
Some results from this experiment have already been shown in previous slides.

Here are some more results:

TKE distribution as function of fragment mass and its average. Data are also compared to data from the SCINTIA setup (Göök et al. 2018) and show good agreement.

Neutron multiplicity vs TKE compared to literature data. Most interesting: the slope: one extra neutron “costs” 12.0 ± 0.1 MeV in TKE.

And, finally …



Contour plot of excitation energy versus spin for ^{136}Xe in ^{235}U(n_{th},f) as calculated with the GEF code.

Figure from Al-Adili et al, Eur. Phys. J. A **55**, 61 (2019).

Once the fragments excitation energy is below the **neutron separation energy**, S_n, the only way to remove energy (and angular momentum) is γ-ray emission.

Above S_n, neutron emission dominates.

Typical S_n for fission products are 5.5-6 MeV.

Total γ-ray energies are typically 7 MeV.

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Prompt γ-ray emission

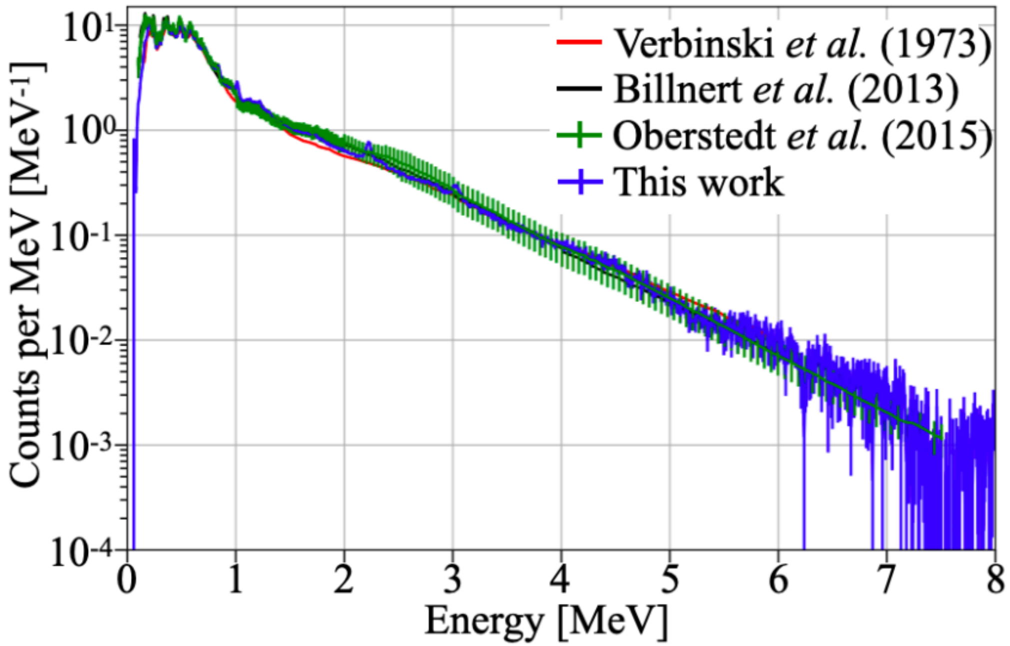

Figure 3.2: New measured PFG spectrum from ^{252}Cf(sf) compared to previous measurements from Verbinski *et al.* (1973) [28], Billnert *et al.* (2013) [37], and Oberstedt *et al.* (2015) [38].

From: PhD thesis Dorthea Gjestvang (Oslo, 2023)

Typical example of a γ-ray spectrum from fission (prompt).

Rather high-energy γ-rays are possible!

But most are below 1 MeV.

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Average quantities for prompt γ-rays

<u>Example</u>: fission of **^{241}Pu***.

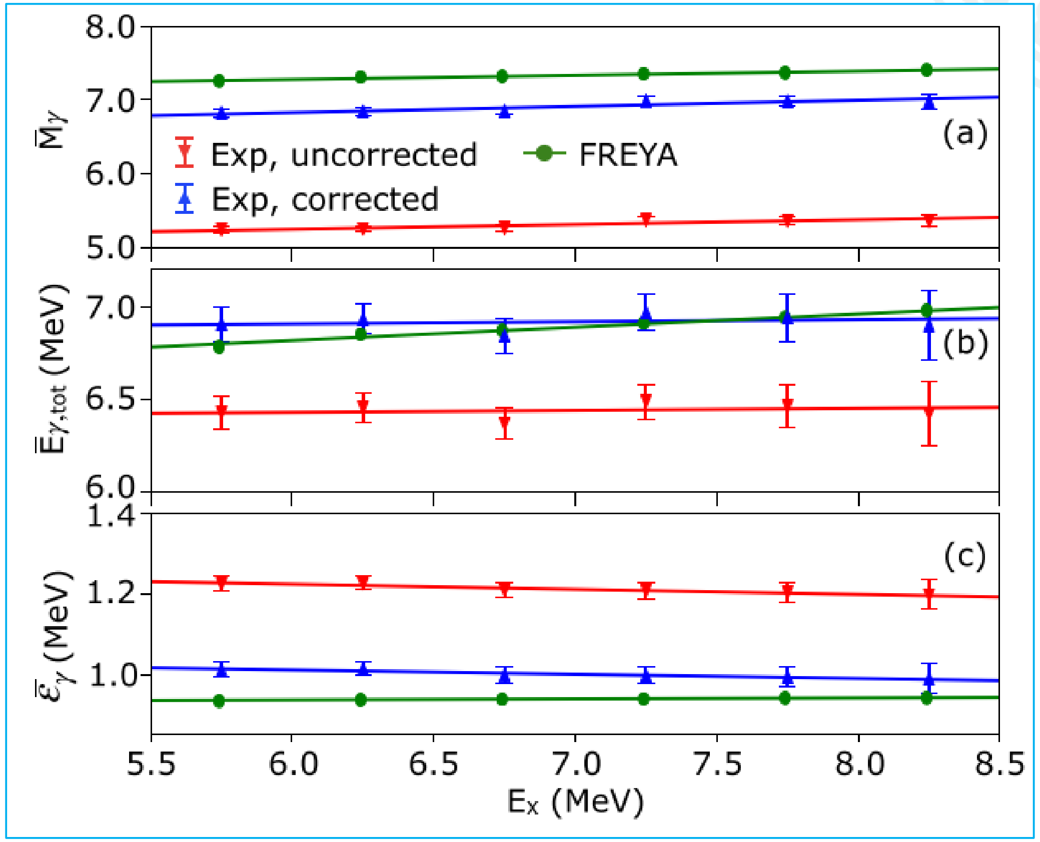
No or very weak dependence on the excitation energy of the fissioning nucleus is found for the (average) quantities:

- multiplicity (“nubar for γ-rays”), M_γ
- total γ-ray energy, $E_{\gamma,tot}$
- average γ-ray energy, ε_γ

Experiment: blue
FREYA code: green

<u>Consistency check (roughly):</u>

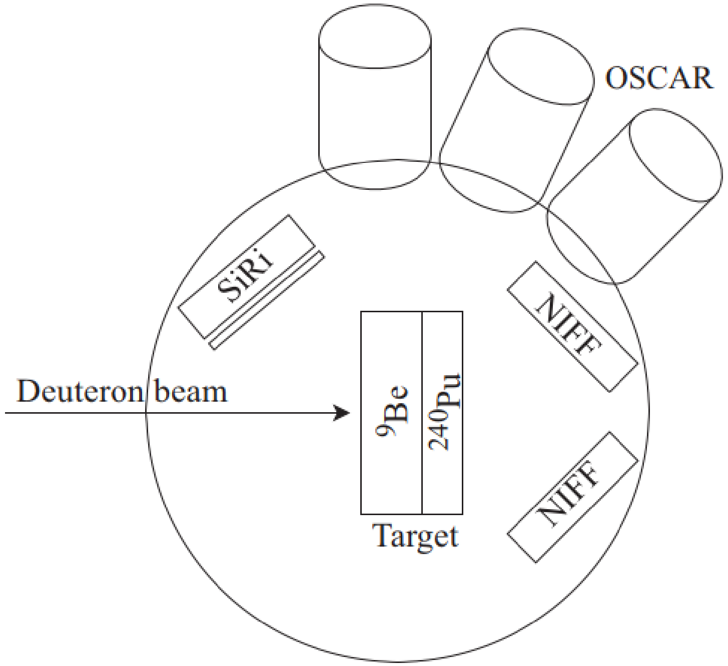
M_γ: 7
ε_γ: 1 MeV
$E_{\gamma,tot}$: 7 MeV (= 7 x 1 MeV)

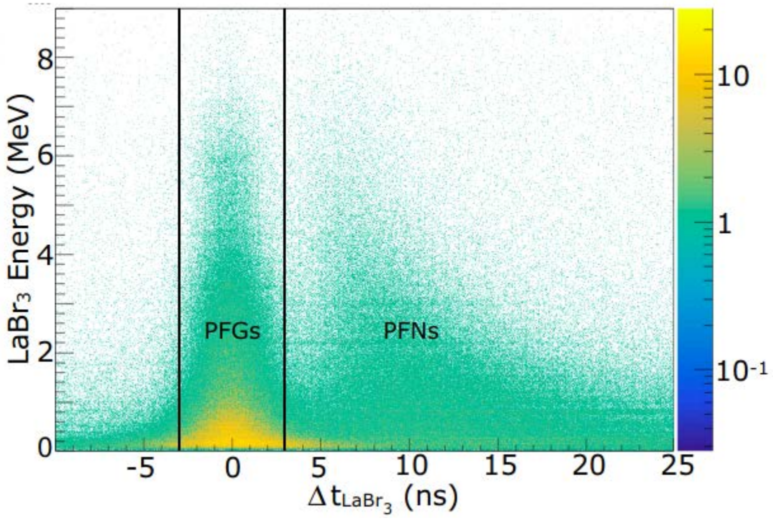


D. Gjestvang et al., Phys. Rev. C **103**, 034609 (2021)

Experiments – Example I

- In the case shown, the experiment was carried out at the Oslo cyclotron using a deuteron beam (!).
- The reaction is thus **^{240}Pu(d,pf)** and it is **^{241}Pu*** that fissions.
- NIFF: registers fission fragments and is used as tag for fission events.
- SiRi: Silicon detectors measure the proton energy which allows for calculating the excitation energy of **^{241}Pu***.
- OSCAR: array up to 30 $LaBr_3$:Ce scintillator detectors for measuring the γ-rays.
- Time between proton signal (in SiRi) and gamma ray in OSCAR is used to discriminate prompt neutrons (which are slower than γ-rays).
- Unfolding of the γ-spectra to correct for detector response.





D. Gjestvang et al., Phys. Rev. C **103**, 034609 (2021)

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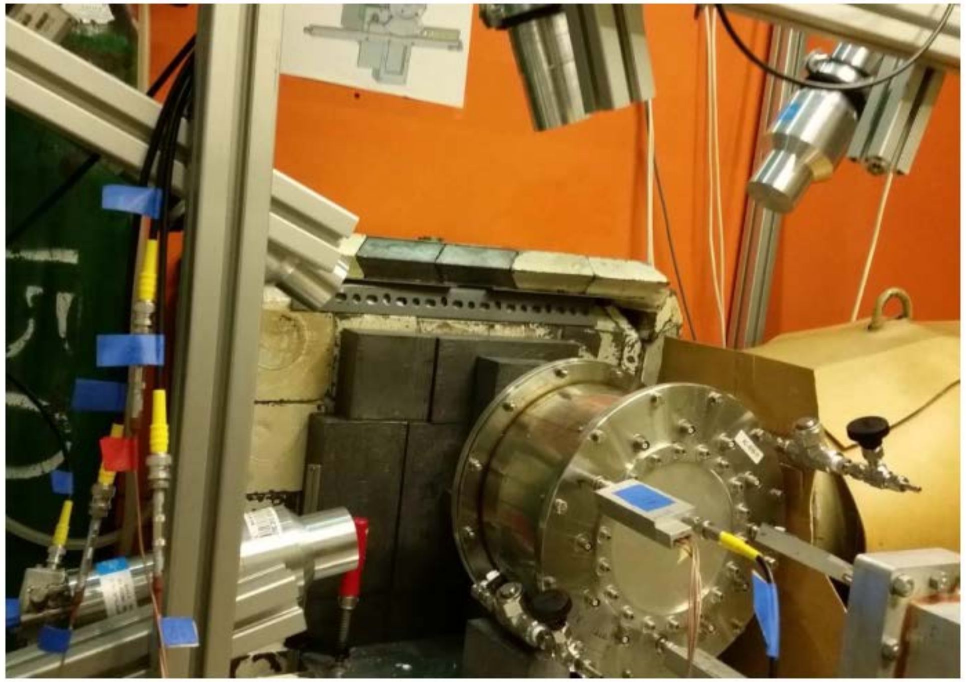
Experiments – Example II

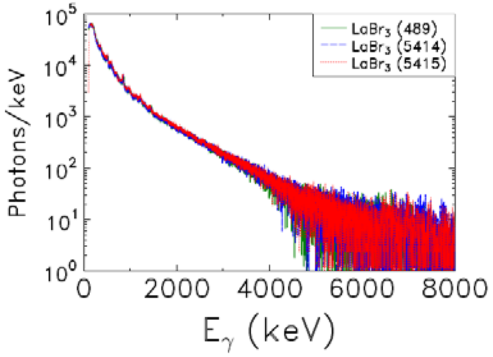
Measurement of prompt γ-rays from ^{239}**Pu(n$_{th}$,f)**:

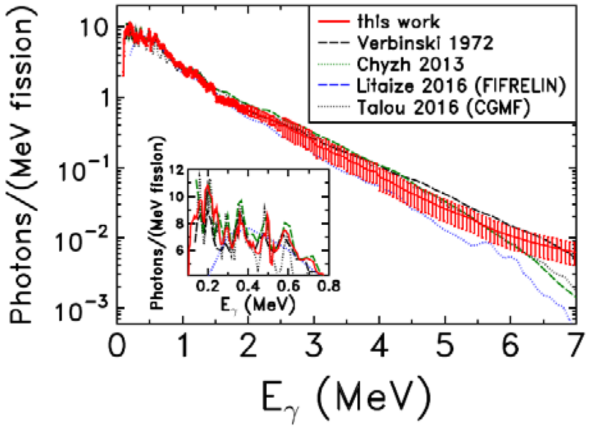
- Beam: Thermal neutrons from the Budapest 10 MW research reactor
- Detectors: Four LaBr$_3$:Ce and one FGIC
- Target: High-purity (99.97%) ^{239}Pu target; 430 µg



A. Gatera et al., Phys. Rev. C **95**, 064609 (2017)

Again: Use of TOF and unfolding of γ–spectra (etc.)





$\overline{M}_\gamma$: 7.35 ± 0.11

ε_γ: 0.85 ± 0.02 MeV

$E_{\gamma,tot}$: 6.42 ± 0.11 MeV

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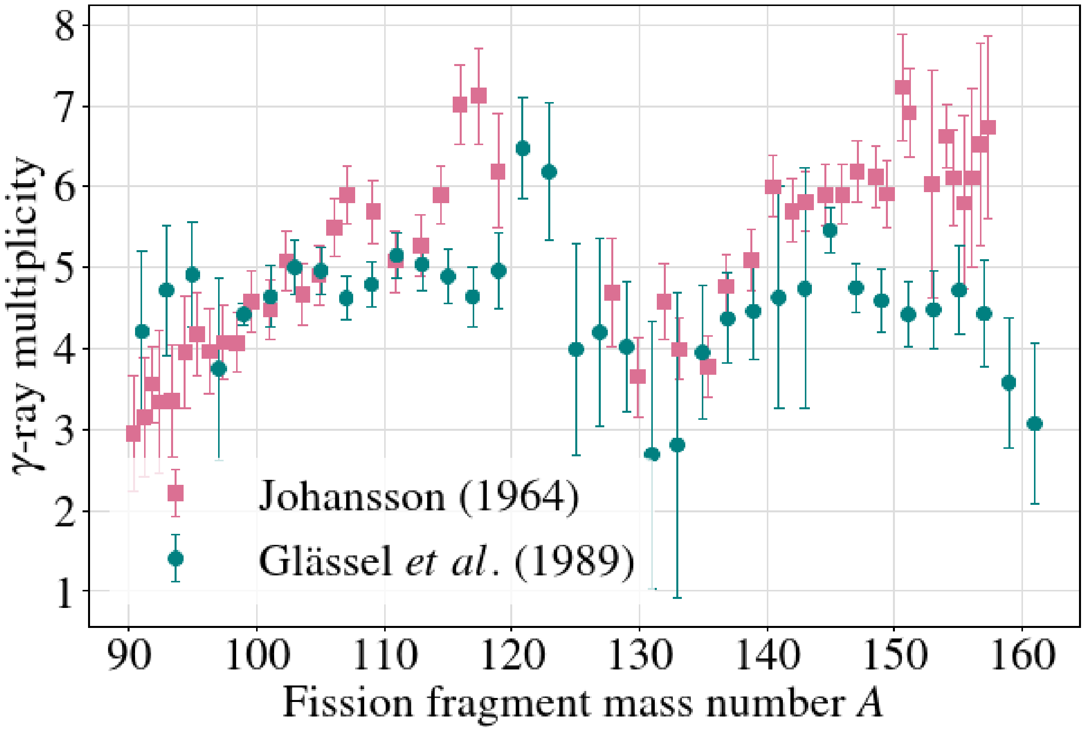
How about multiplicity as function of fragment mass?

- Its a sawtooth
- No its not, its flat
- Or is it?

Early measurements (Johansson 1964) suggested a sawtooth in $M_\gamma(A)$.

Later this was contradicted by Glässel (1989).

Considering the similarity of S_n the latter seems rather reasonable. But …



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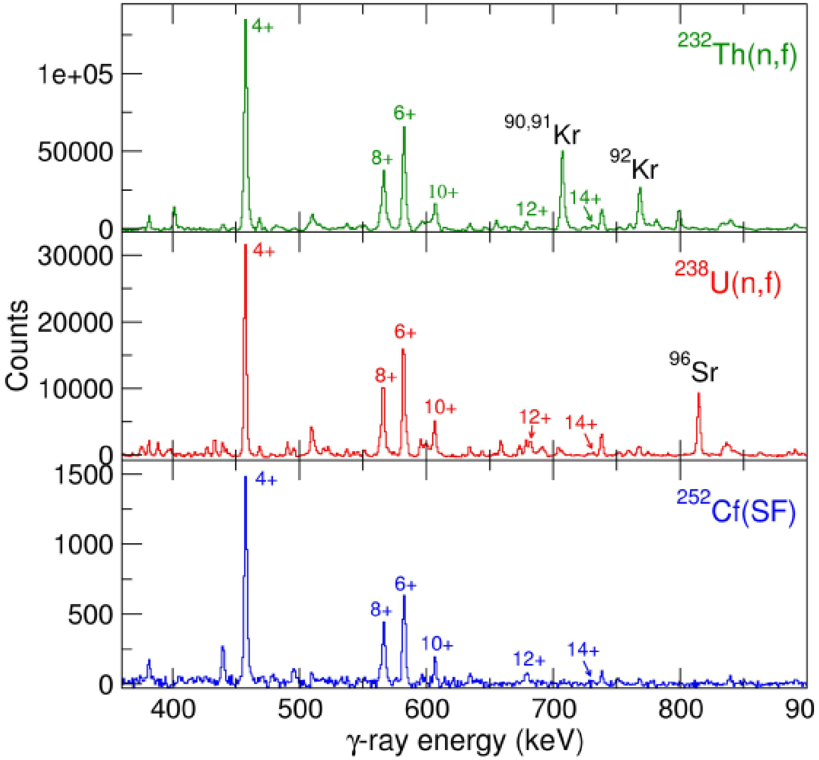
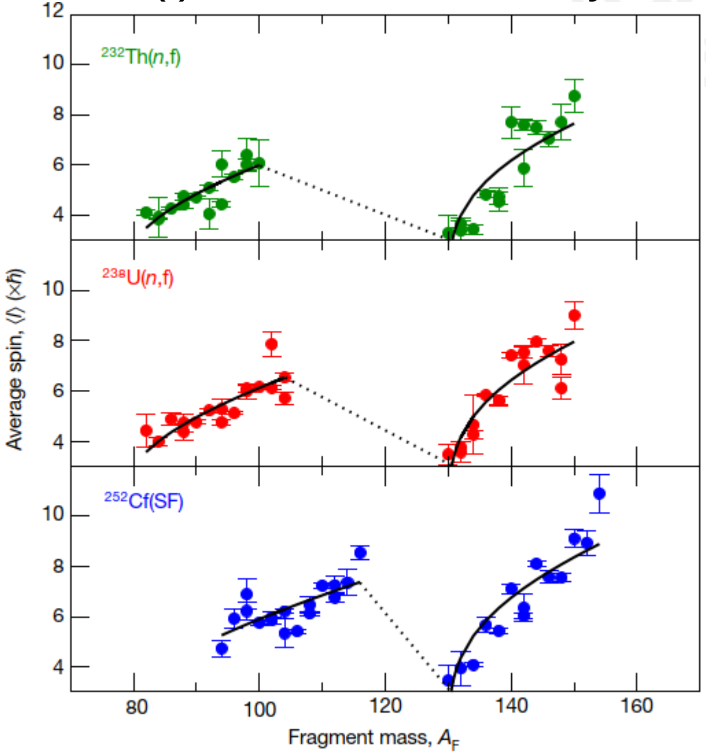
γ-spectrometry for fragment spin extraction



γ-spectrometry allows product identification and extraction of the fragment spin (!)

Same (!) behaviour for all systems



Wilson et al., Nature **590**, 566 (2021)

So: It seems the sawtooth is everywhere

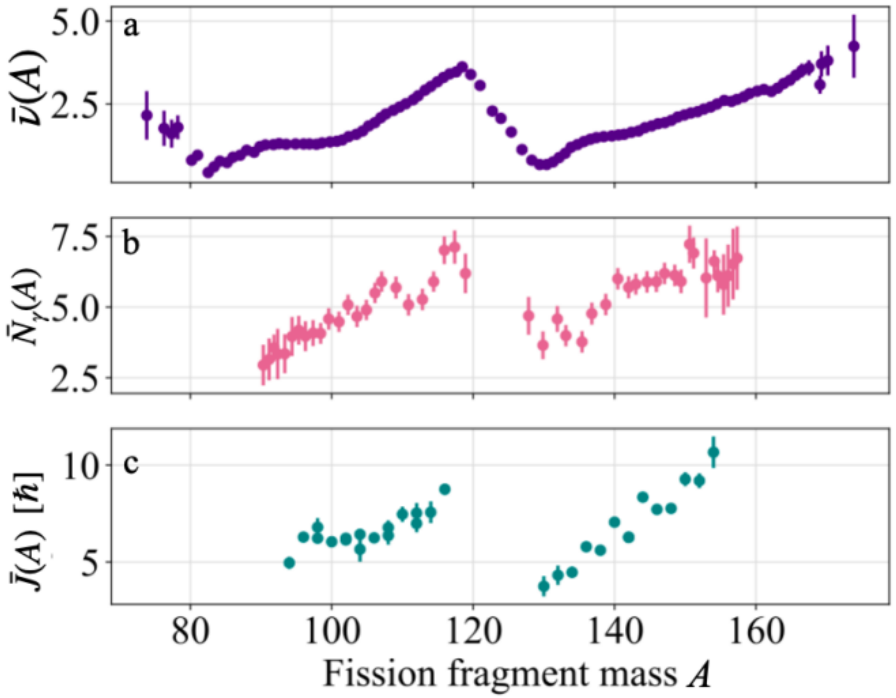


Figure 6.1: Sawtooth patterns observed in various observables as a function of fragment mass following ^{252}Cf(sf). In (a) the average neutron multiplicity $\bar{\nu}(A)$ from Göök *et al.* [17] is plotted, (b) shows the average γ-ray multiplicity $\bar{N}_\gamma(A)$ from Johansson [46], and (c) is the average angular momentum $\bar{J}(A)$ from Paper II.

The nubar sawtooth (here: Göök 2014).

Recent measurements (Wilson 2021) found a sawtooth for fragment spin (at least for even-even nuclei).

Since angular momentum is removed by y-emission,
it seems likely that suggest that M_γ also shows a sawtooth.

Glässel: issues defining emitting FF?

From PhD thesis of
Dorthea Gjestvang, Oslo 2023

The way forward? The revival of γ-spectrometry in fission

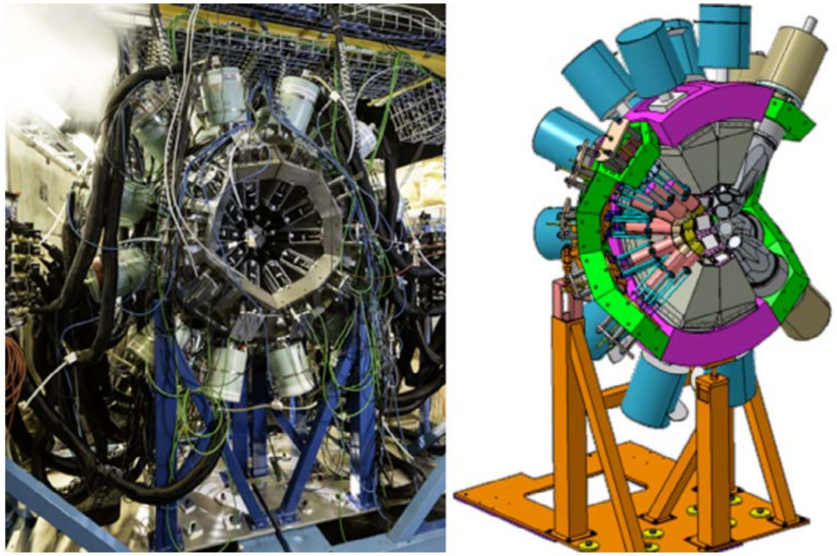


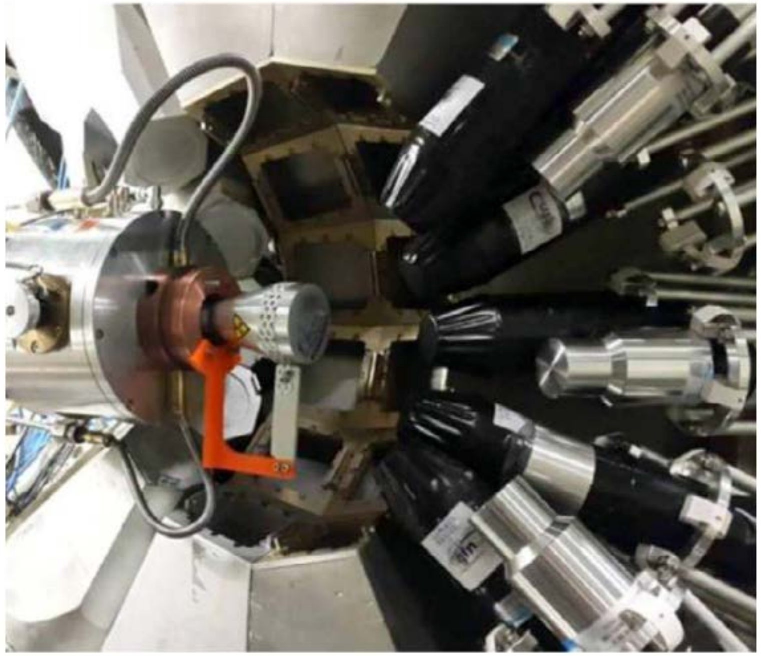
Fig. 1. Photograph of the ν-ball array installed in the experimental area of the ALTO facility (left panel) and a technical drawing of one ν-ball hemisphere (right panel).

Combine:

Innovative neutron source LICORNE @ ALTO (inverse kinetics)

γ-spectrometer with “4π” coverage and fully digital DAQ.

Target inside fission chamber.



Pictures: M. Lebois et al.,
Nucl. Inst. Meth. A **960** (2020), 163580

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Summary and concluding remarks

- We looked a **measurement techniques** for yields, and prompt neutron and gamma emission.
- Fission may be **spontaneous or induced in different ways**:
  - (n,f), (p,f) …
  - Surrogate: (d,pf), …
  - MNT, Coulex
- **Fragments** may be **stopped** (e.g. ISOL methods) or **unstopped** (Ionization chamber, etc).
- We need good, well-characterized **targets** (or **sources**), and suitable **accelerators**, **reactors** , ….
- Depending on the method, observables may or may not be measured in **correlation.**
- Often a range of **assumptions** (more or less correct) need to be invoked in the analysis.
- In general: careful **correction** to detector response needed and modern techniques (e.g. digital DAQ) open for new insights.
- We have performed experimental studies of fission for 80+ years now but **we are not done** …

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