Fission-yield data

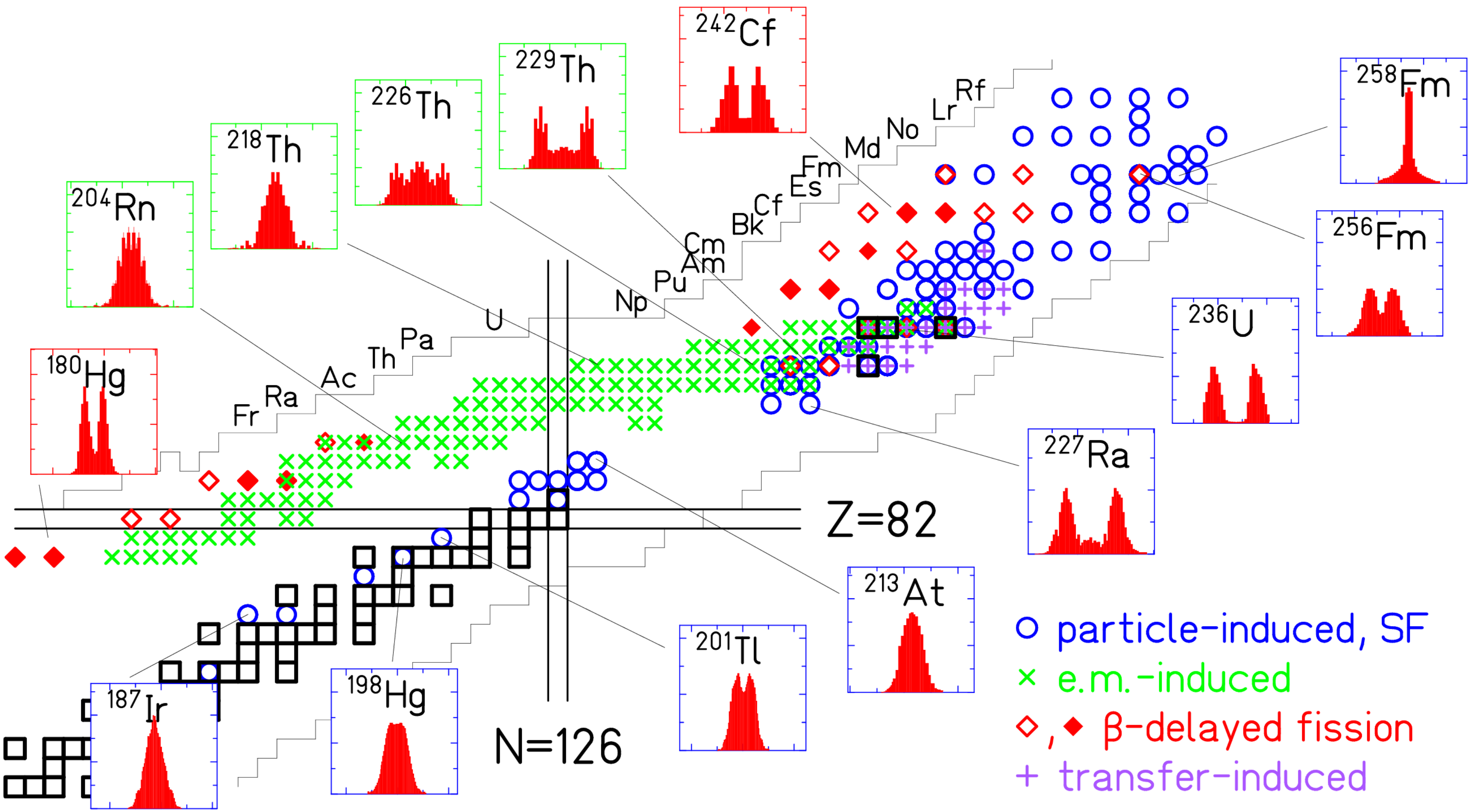
Karl-Heinz Schmidt

Topical day | From nuclear data to a reliable estimate of spent fuel decay heat
October 26, 2017 | SCK•CEN Lakehouse, Mol, Belgium

Lay out

- Introduction
- Stages of the fission process
- Detection methods
- Excitation mechanisms
- Characteristics of FY
- Evaluation
- Correlations
- Conclusion

Overview on FY (A and Z distributions)



Many systems investigated by different methods. Strong variations found. Only few around 235U are relevant for nuclear technology.

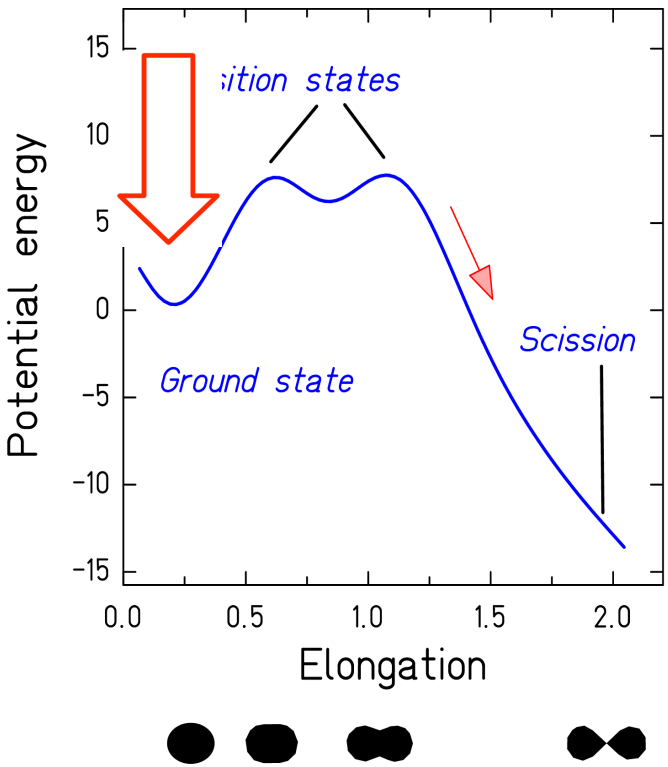
A naïve scenario of nuclear fission

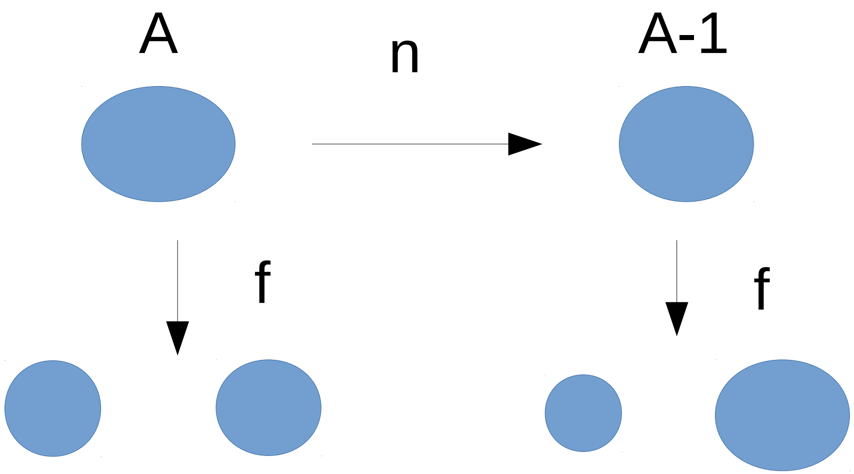
- The nucleus is divided in two parts
  $A1 + A2 = ACN$
- Protons and neutrons are divided in the same portion:
  $Z1/N1 = Z2/N2 = ZCN/NCN$
  $Z1 + Z2 = ZCN$, $N1 + N2 = NCN$
- The shape of the mass distribution is characteristic for the fissioning system and its excitation energy and determines the fission yields.

Reality is much more complex. To understand the complexity one must follow the stages of the fission process.

Stages of the fission process

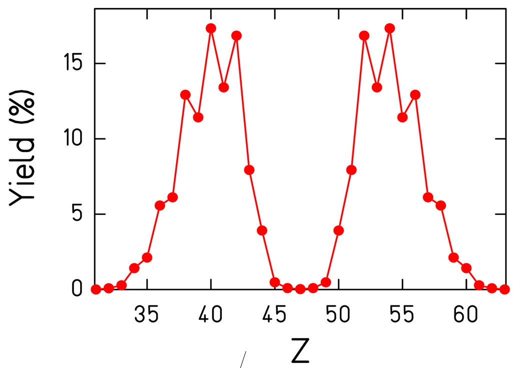
Stages of the fission process 1: Pre-saddle particle emission

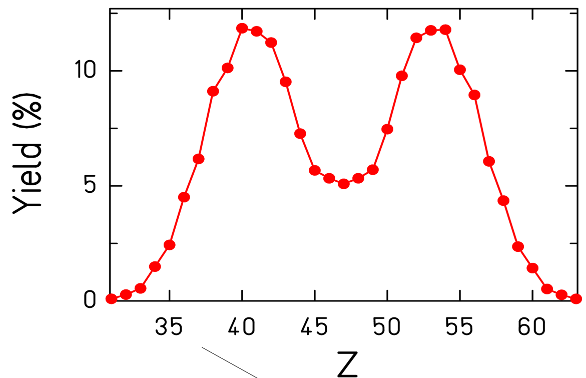




- Competition between particle emission and fission → multi-chance fission
- FY from several fissioning nuclei with different E*

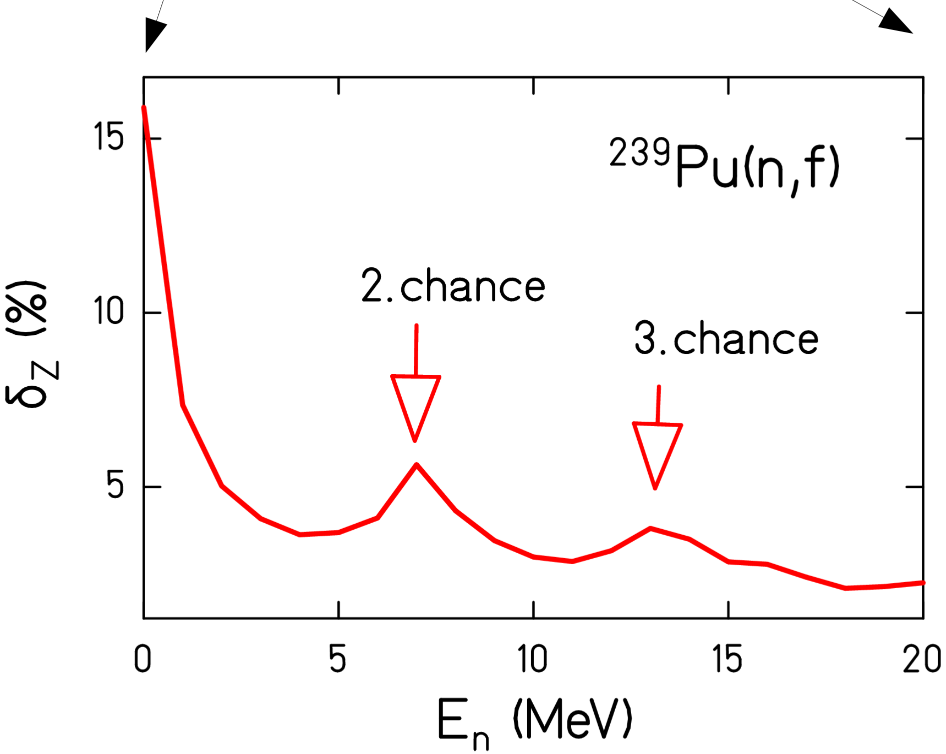
Signature of multi-chance fission: Even-odd effect in FF Z distribution





GEF calculations

(Experimental data not yet available)

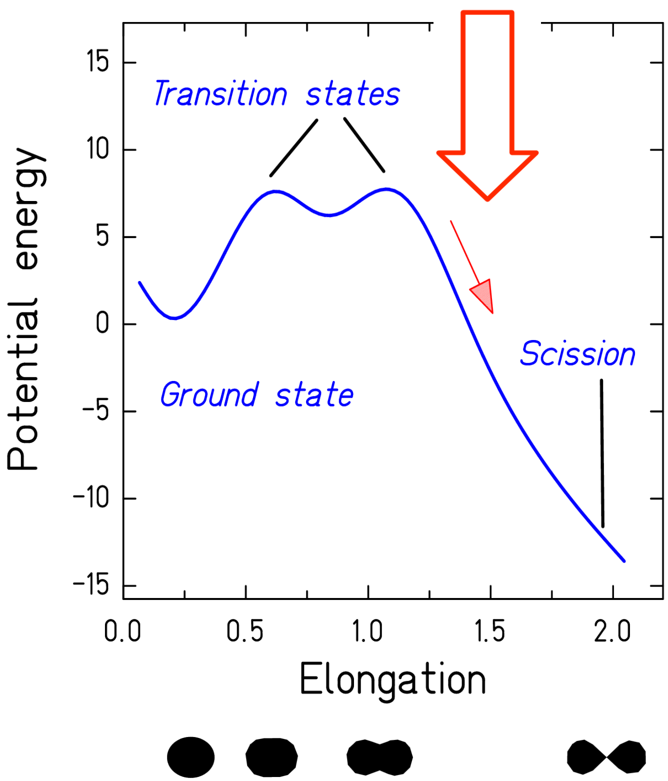


(δ_z is a measure of the even-odd staggering.)

Reduced <E*> leads to increased even-odd effect at the onset of 2nd chance and 3rd chance fission.

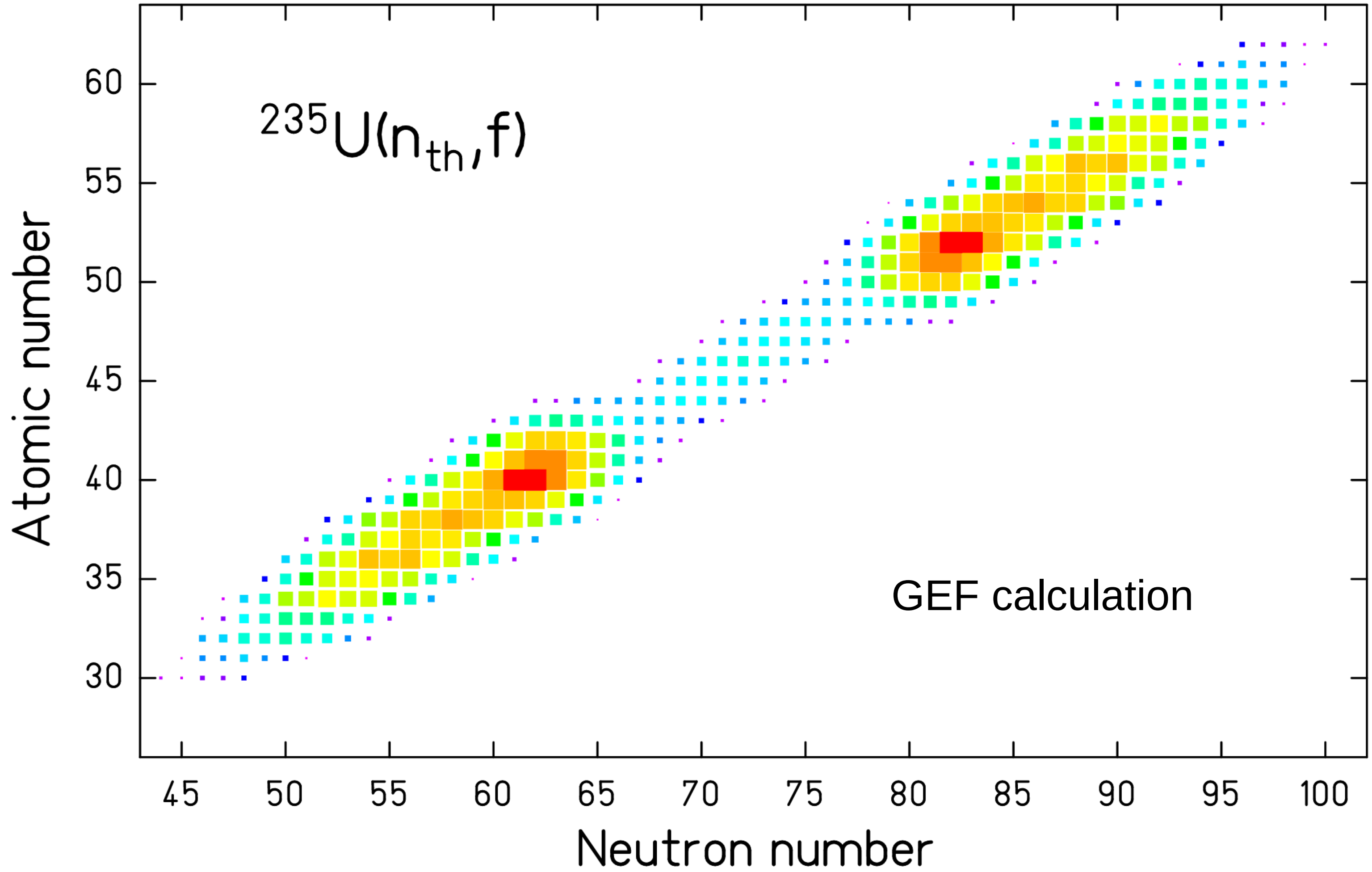
(GEF calculation.)

Stages of the fission process 2: Saddle to scission



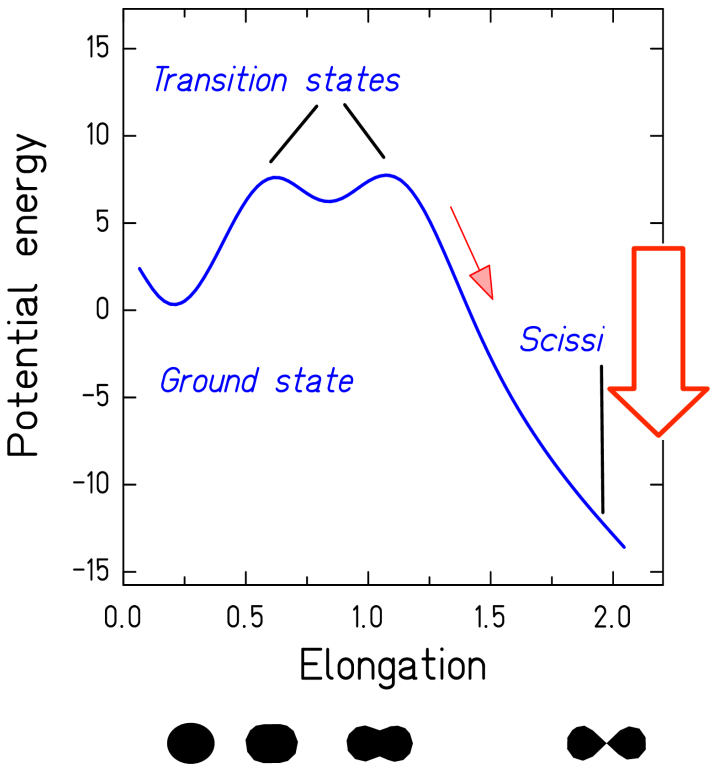
- Formation of fragment sizes.
- Exchange of protons and neutrons between the nascent fragments.
- Dissipation, pair breaking.
- Exchange of E* between the nascent fragments.
- Pre-scission (or scission) neutron emission: Loss of mass and E* during the fission process.
- “Pre-neutron” FY (before neutron emission from the separated fragments).
- Only Z yields are observable.

FY on the nuclide chart



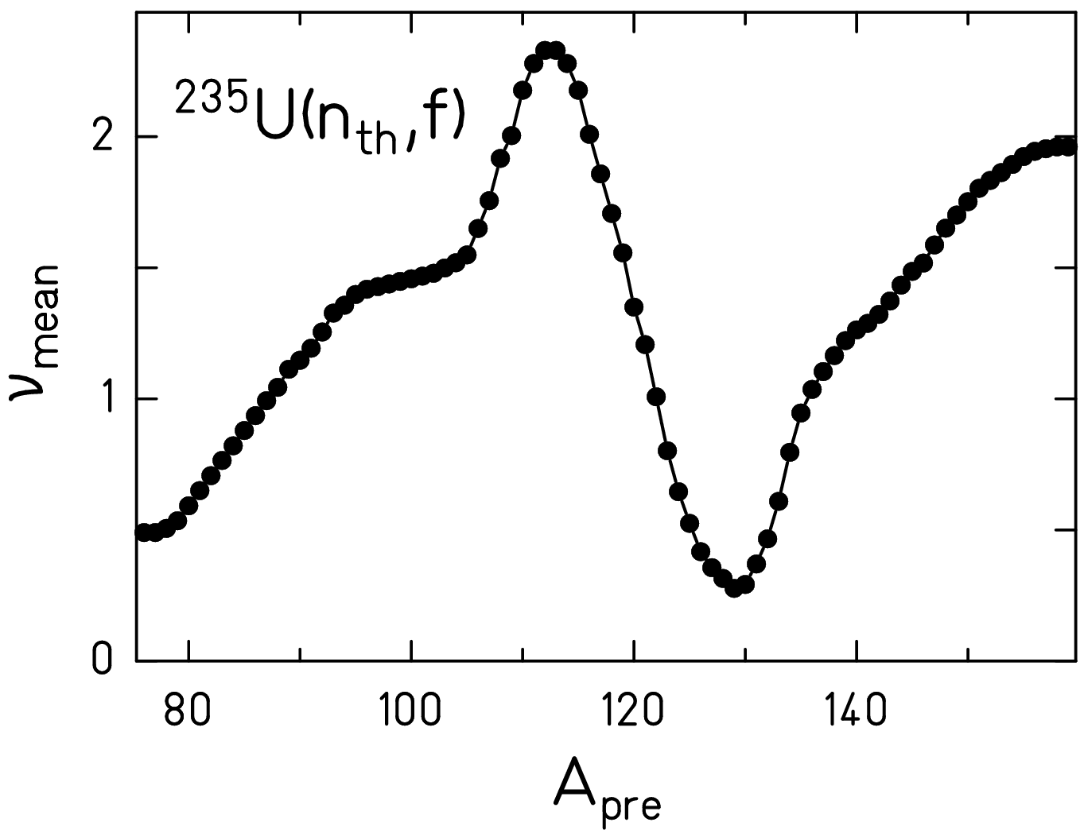
Yields before prompt-neutron emission

Stages of the fission process 3: Emission of prompt neutrons and gammas



- Neutron (and gamma) emission from the separated fragments
- "Post-neutron" FY (after emission of "prompt" neutrons from the separated fragments) = independent yields

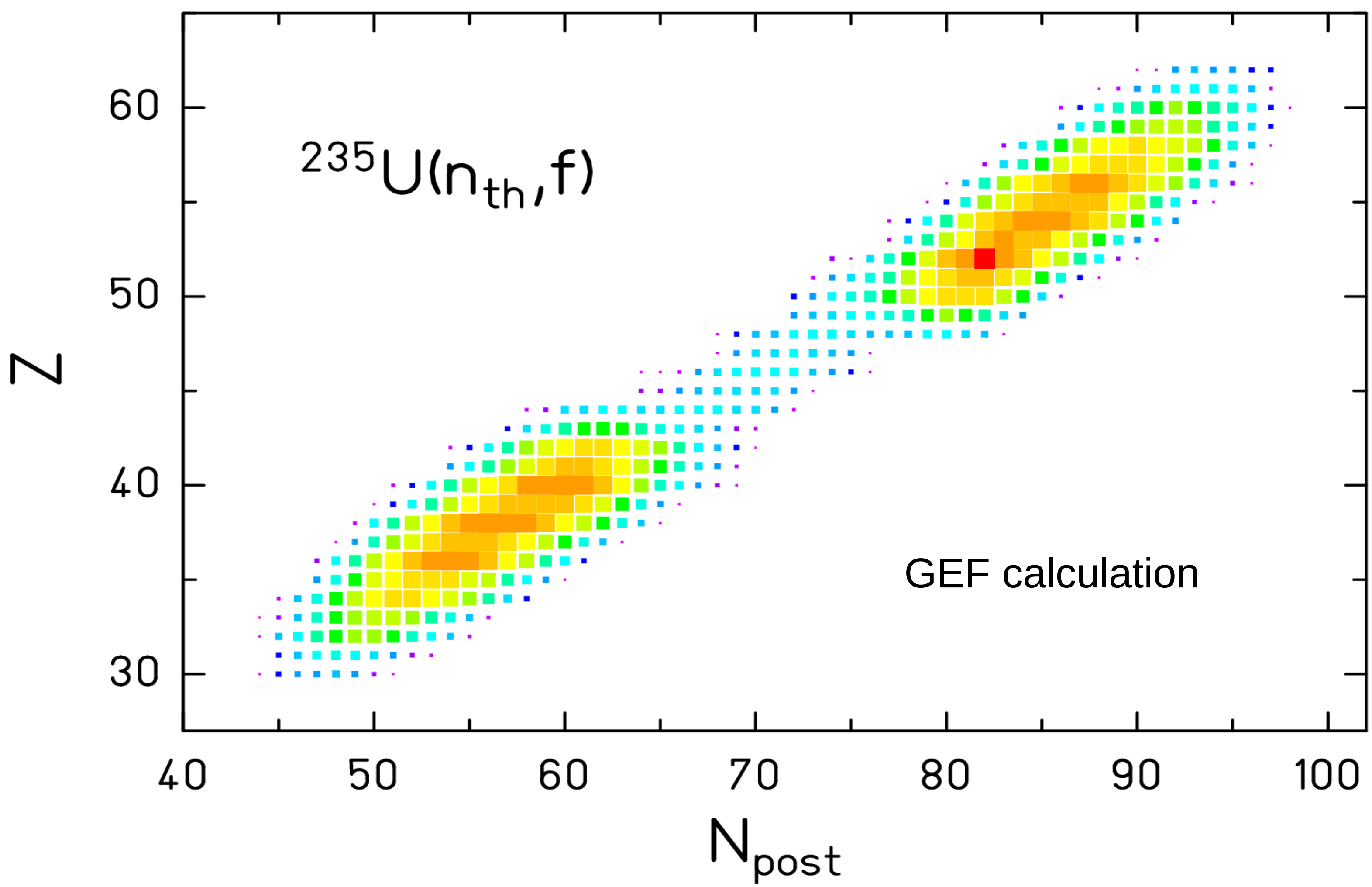
Mass-dependent neutron multiplicities



Data from Wahl*s systematic

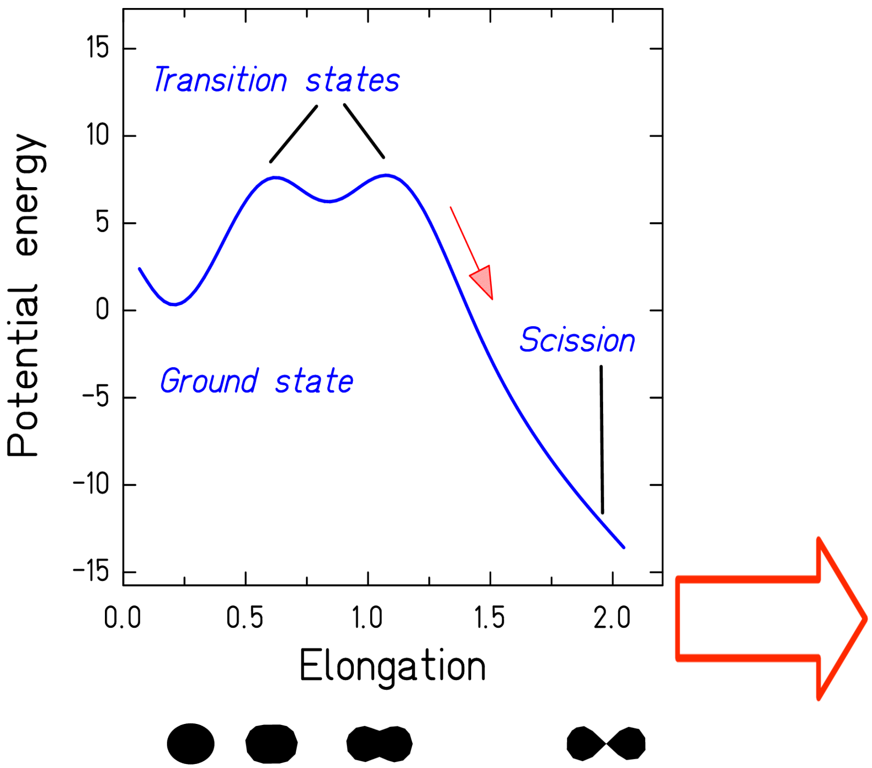
Strong variation of prompt-neutron multiplicity over the FF mass range

FY on the nuclide chart



Yields after prompt-neutron emission

Stages of the fission process 4: beta decay and beta-delayed processes



- Beta decay, partly into excited states. Emission of anti-neutrinos.
- Delayed gamma emission
- Delayed neutron emission
- Cumulative yields

Beta-delayed processes

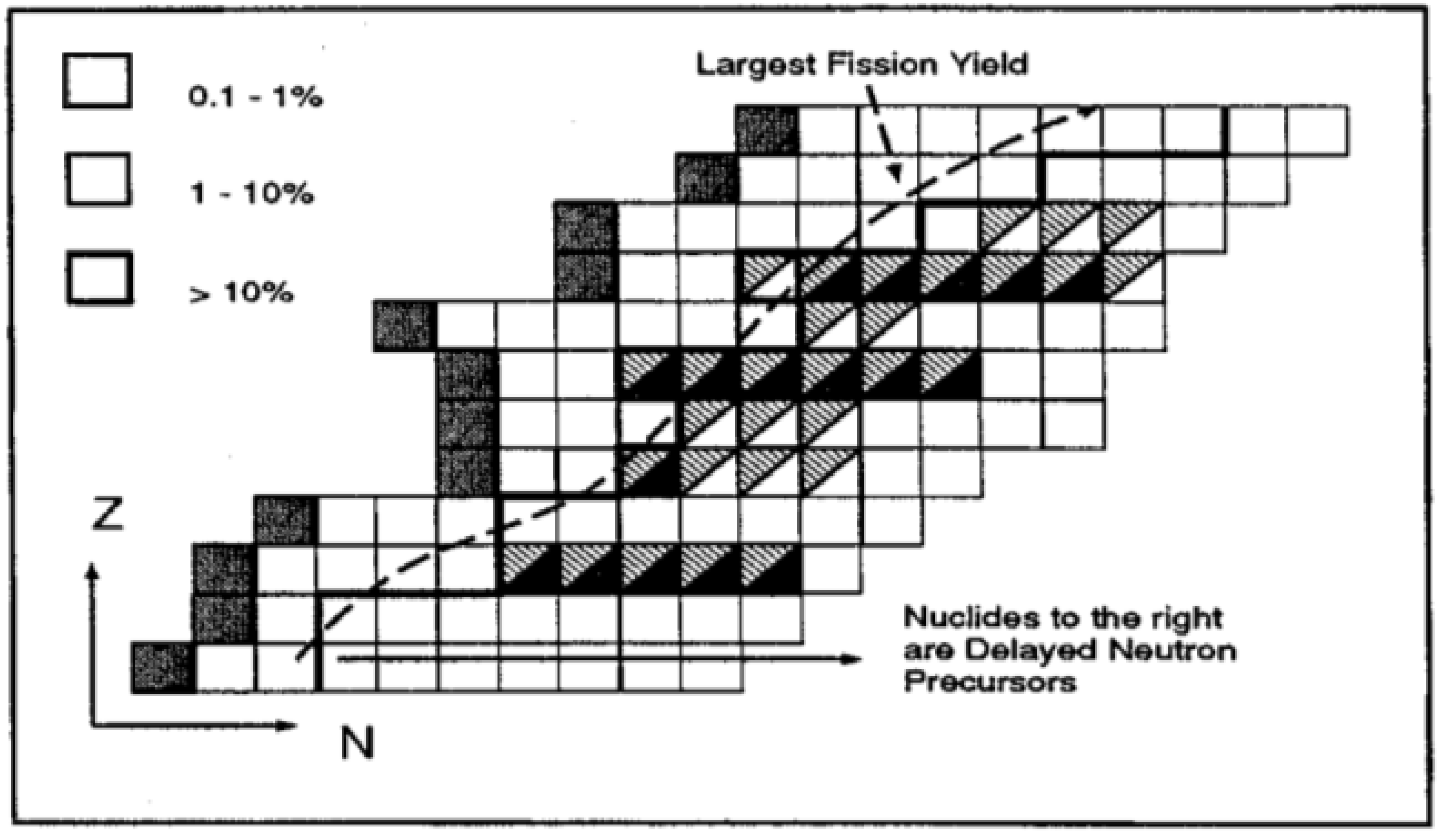


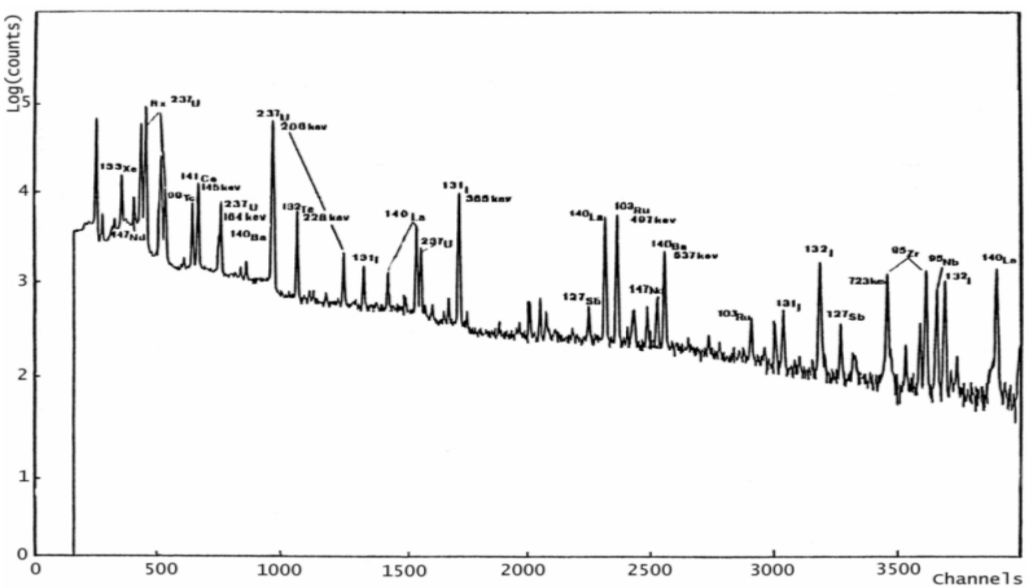
Figure from NEACRP-L-323 (1990)

- Most fission fragments are beta-unstable.
- Daughter nuclei emit delayed gamma radiation.
- For very neutron-rich (most short-lived) fragments, the Q value is higher than Sn.
  → Emission of delayed neutrons.

Detection methods

Radiochemical methods: Technique

- Irradiation of a sample in the neutron flux of a reactor or by an accelerator beam together with a monitor sample for normalization.
- Remove the radioactive sample.
- Chemical separation (to reduce the gamma background).
- Recording gamma spectra.
- Attributing gamma lines to specific nuclides and determine number of atoms (gamma efficiency of detector, spectroscopic properties of the nuclide).



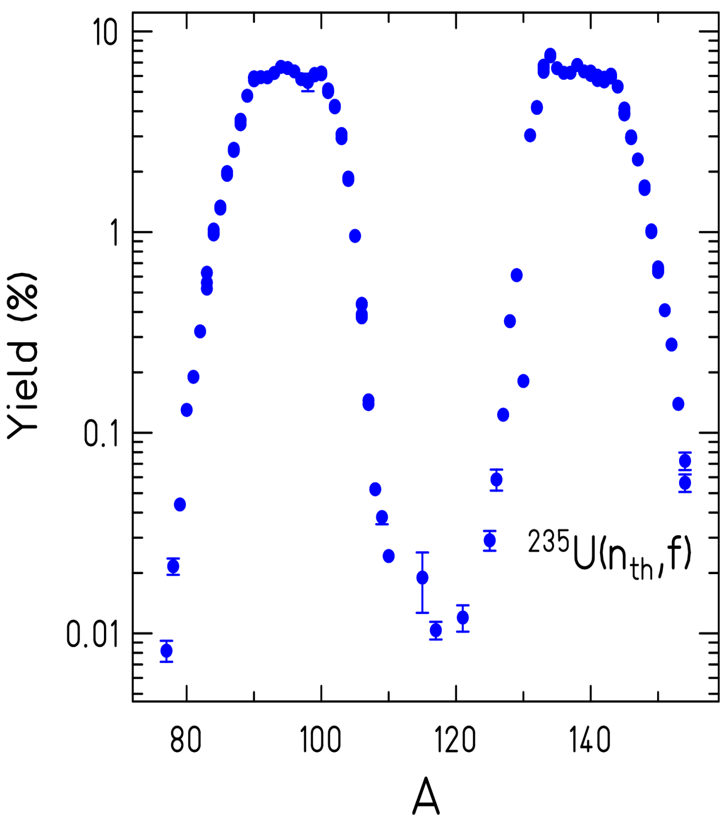
Typical gamma spectrum with identified gamma lines for 238U(n,f), En = 14 MeV. From Laurec et al., NDS 111 (2010) 2965

Radiochemical methods: Strengths and weaknesses

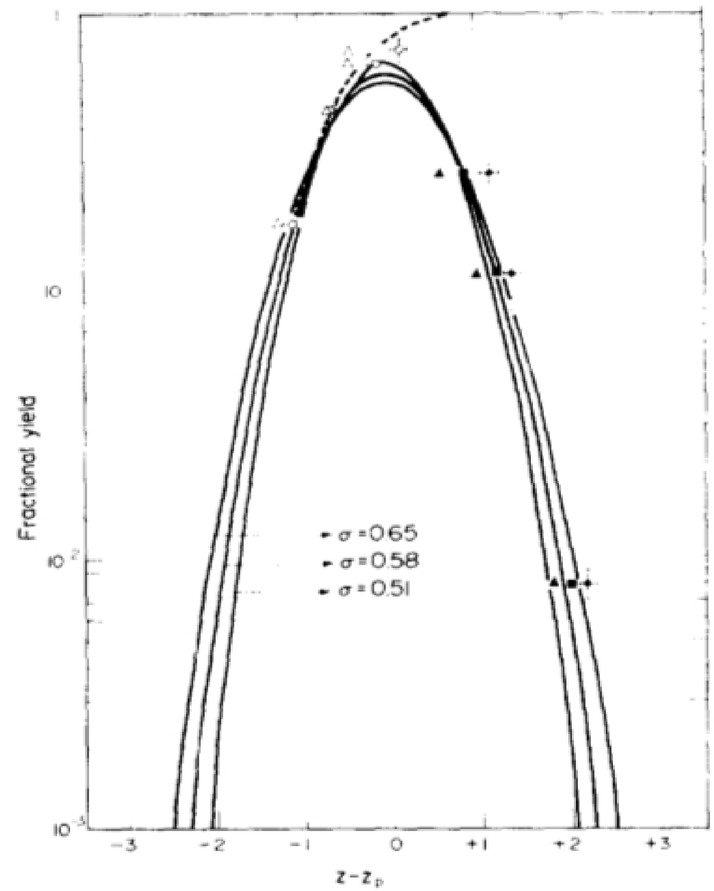
- Unambiguous identification (Z and A) (+)
- Limited choice of suitable targets (-)
- Unable to measure short-lived nuclides, cumulative yields are measured (-)
- Uncertainties by possible deficiencies of spectroscopic data (-)

Radiochemical methods: Results

Mass distributions



Isobaric Z distributions



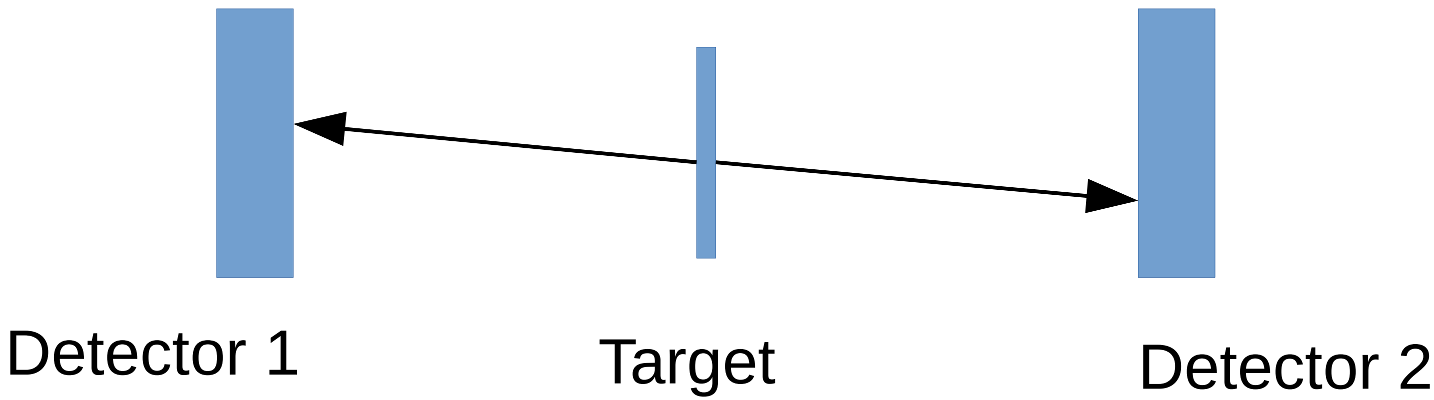
Open symbols: measured cumulative yields

Full symbols: estimated independent yields

Mass yields well measured, because β decay preserves A but incomplete.

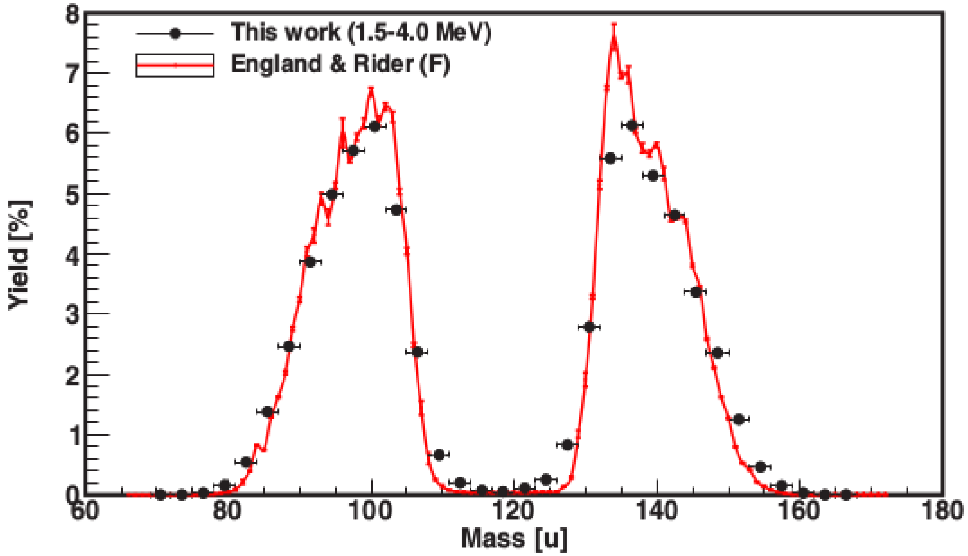
Attempts to estimate independent yields from measured cumulative yields.

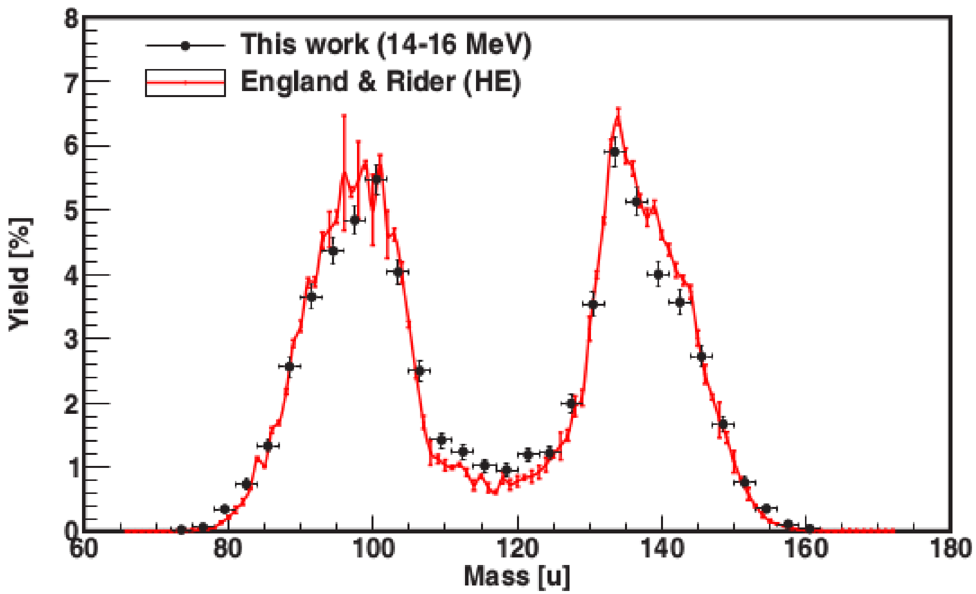
Double ToF, double E etc.



- Mass ratio from momentum conservation
- Full continuous mass distribution
- Limited resolution (typical 4 mass units)
- Disturbed by scattering effects and calibration issues
- Not directly usable for nuclear technology

Mass distribution from double-E





238U(n,f)

Comparison of double-E data (black symbols) with evaluated data based on radiochecmial data (red line).

Limited resolution of the double-E method.

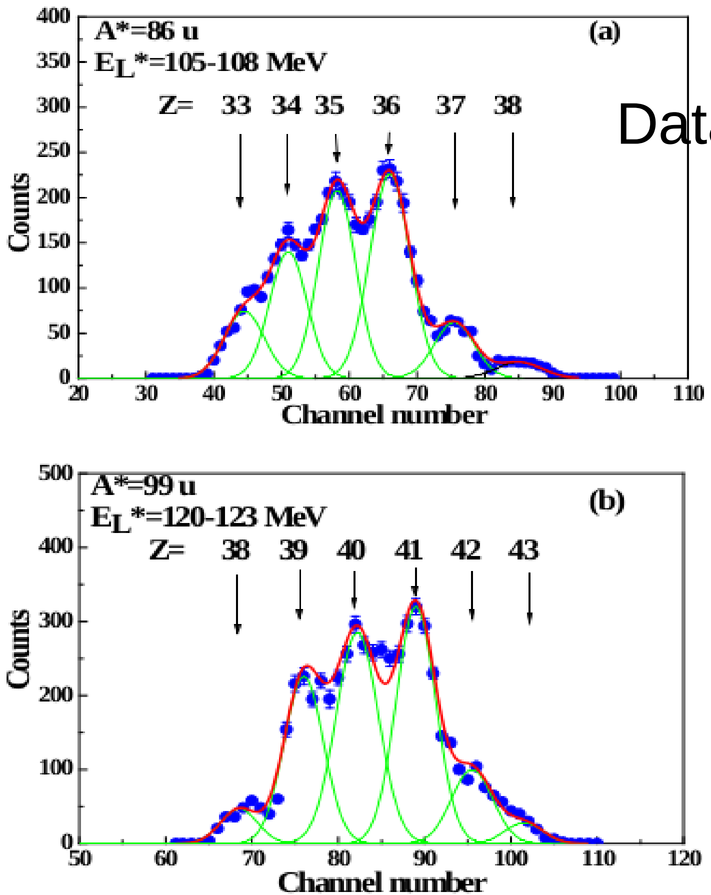
Figure from Duke et al., Phys. Rev. C 94 (2016) 054604

Z resolution in direct kinematics

- Cosi fan tutte (2 x ToF-ΔE-E)
- Lohengrin (separator + ΔE/β-delayed gammas)
- Double ΔE-E

New!

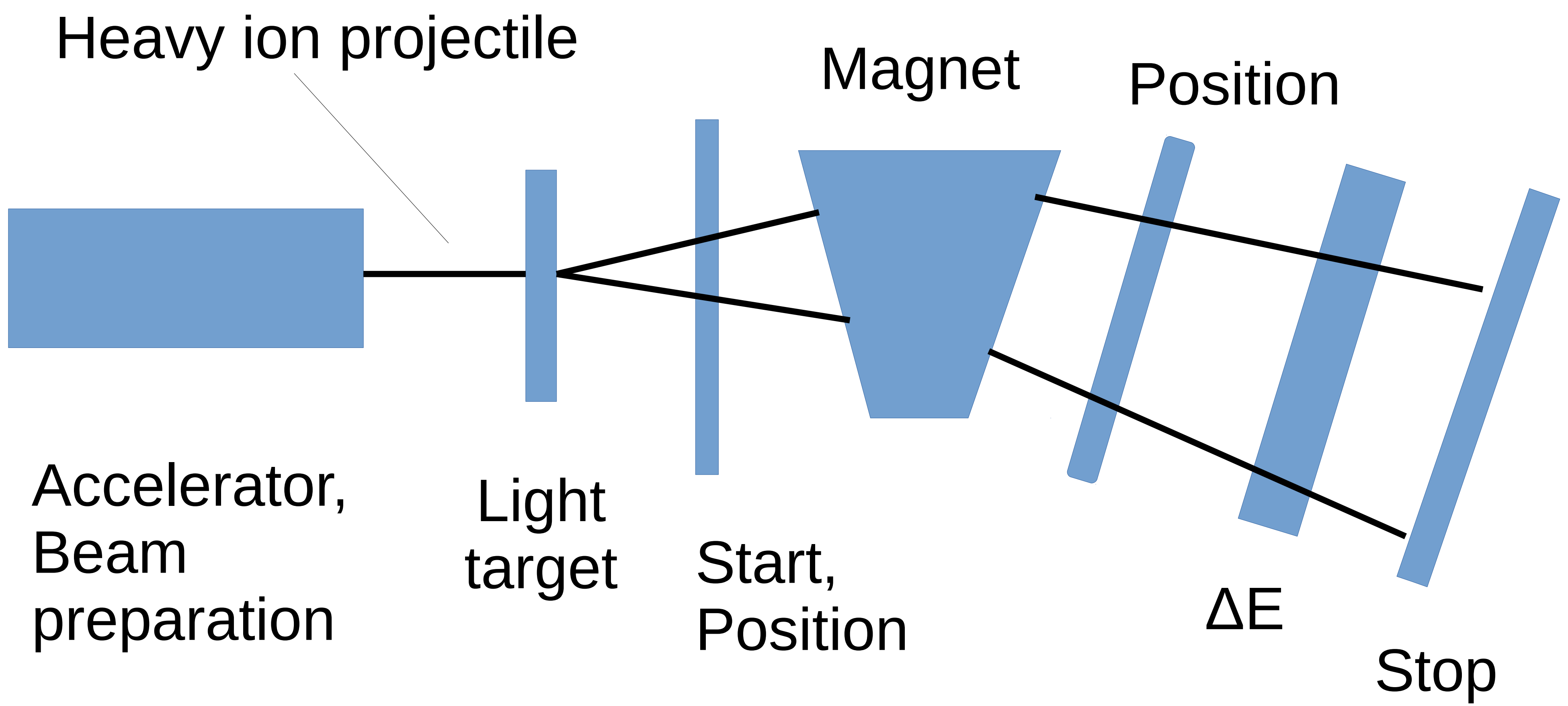
Data from Wang et al. (arXiv)



- Z are resolved.
- Limited to light fragments at high Ekin.

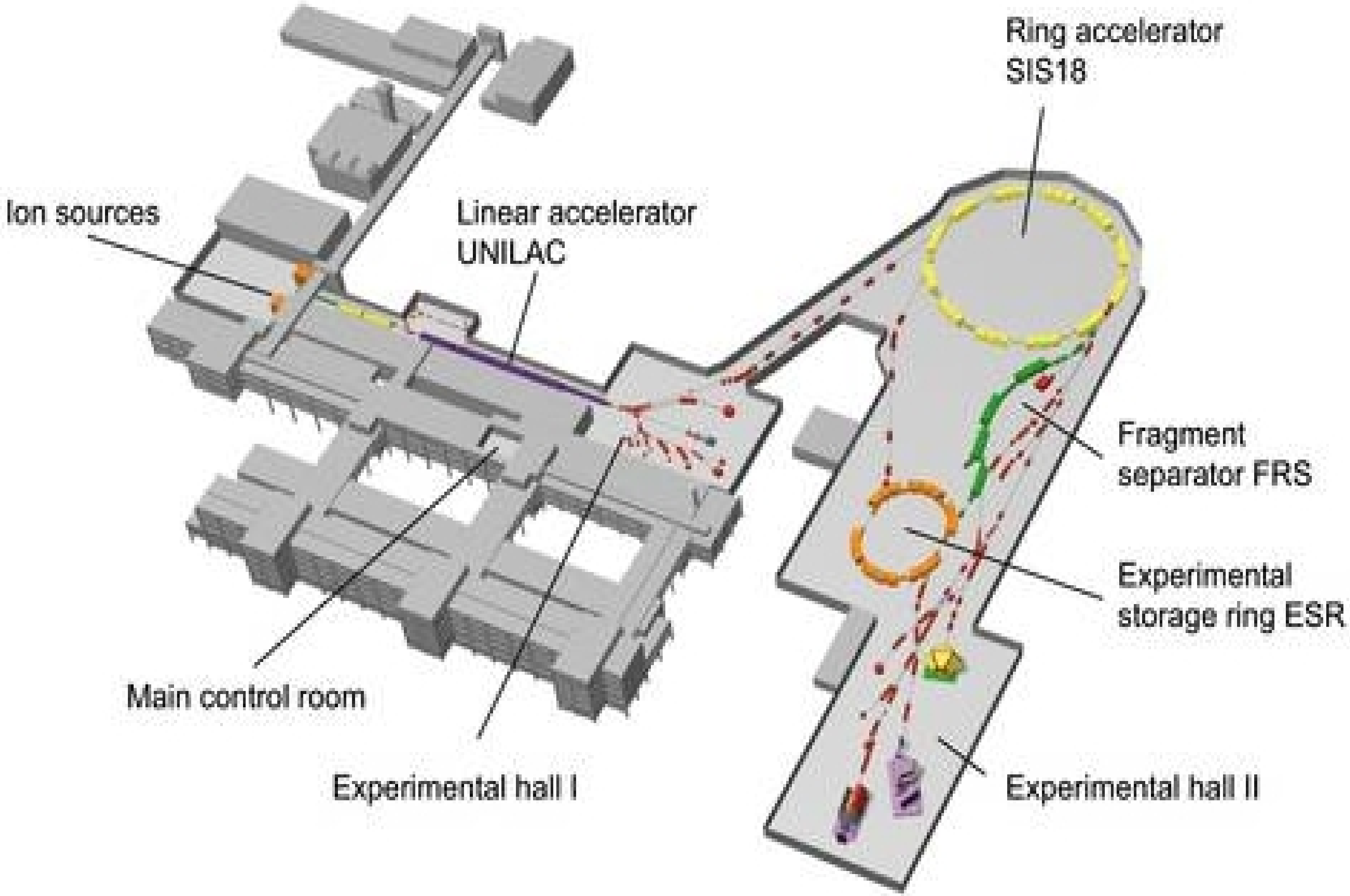
- Lohengrin + ΔE provides unique data for (nth,f) (resolution in A and Z).

ΔE-Tof-Bρ method in inverse kinematics



Principle of the method.
Provides excellent resolution in A and Z.

Fission experiments with relativistic secondary beams (1 A GeV) at GSI, Darmstadt (Germany)



The SOFIA experiment: set-up and resolution

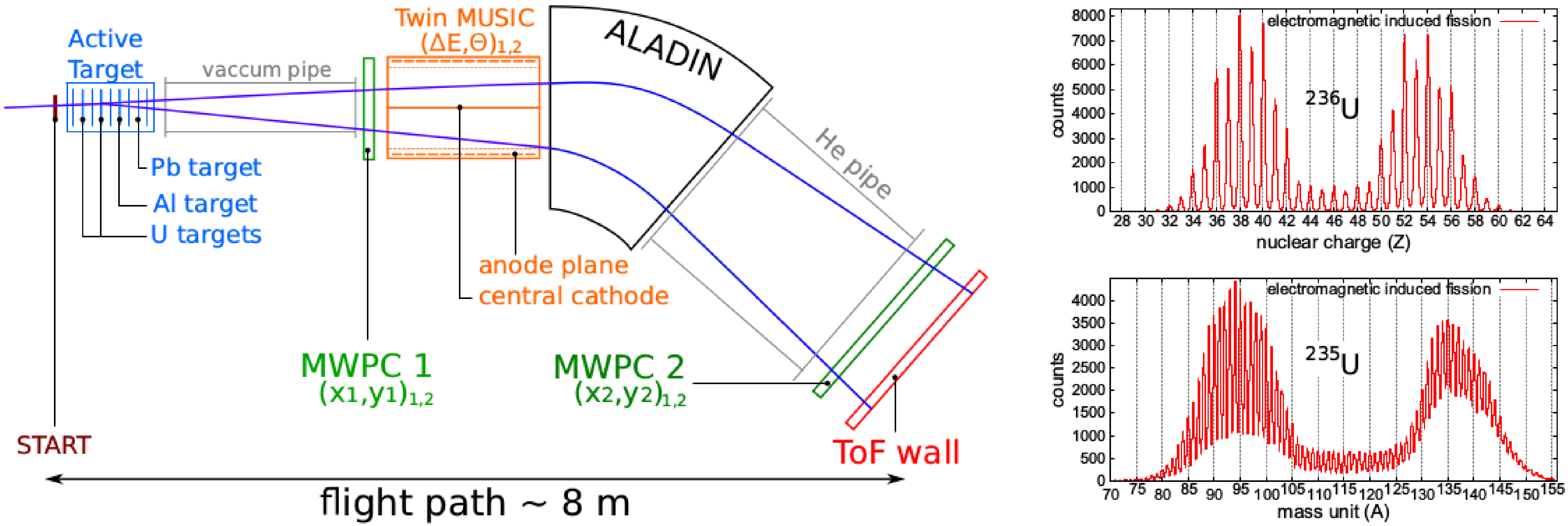
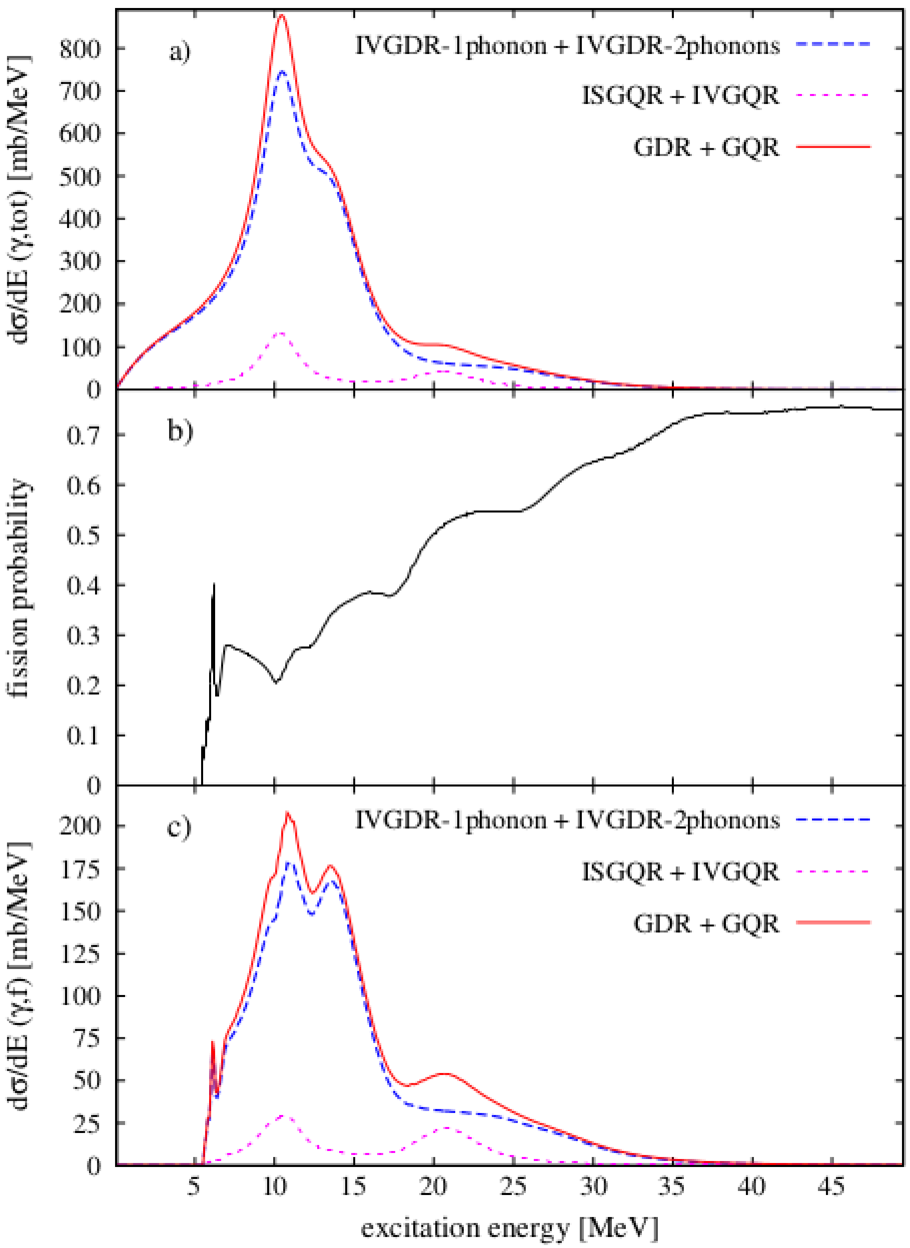


Figure from EPJ Web of Conferences 111 (2016) 08001

Excellent resolution in Z and A, measurement of both fragments in coincidence.

Full FY distribution can be measured in one shot!!!

The SOFIA experiment: Coulomb excitation

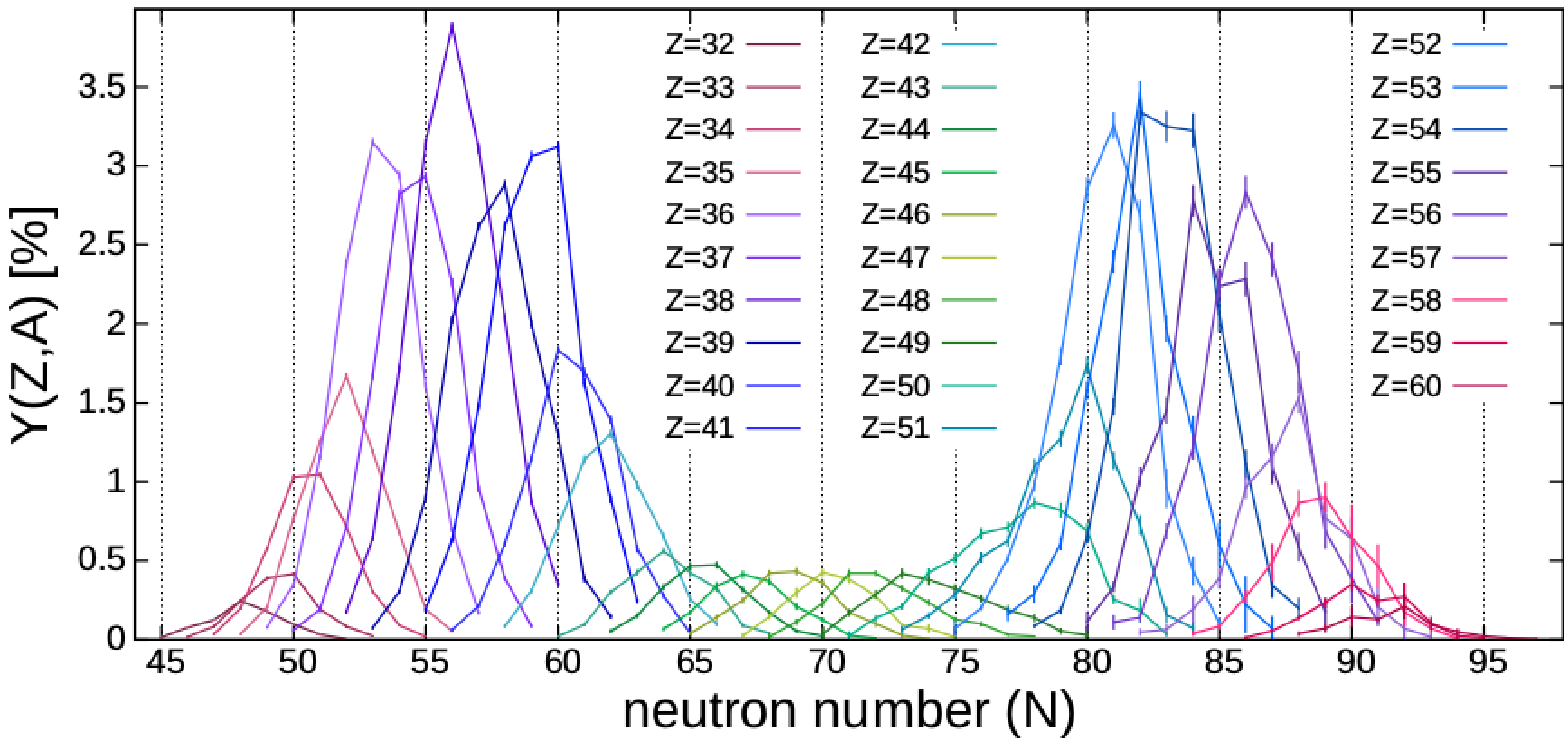


Drawback:

Fission is induced by Coulomb excitation in a uranium target:

The excitation-energy cannot be varied, and the excitation-energy distribution is broad.

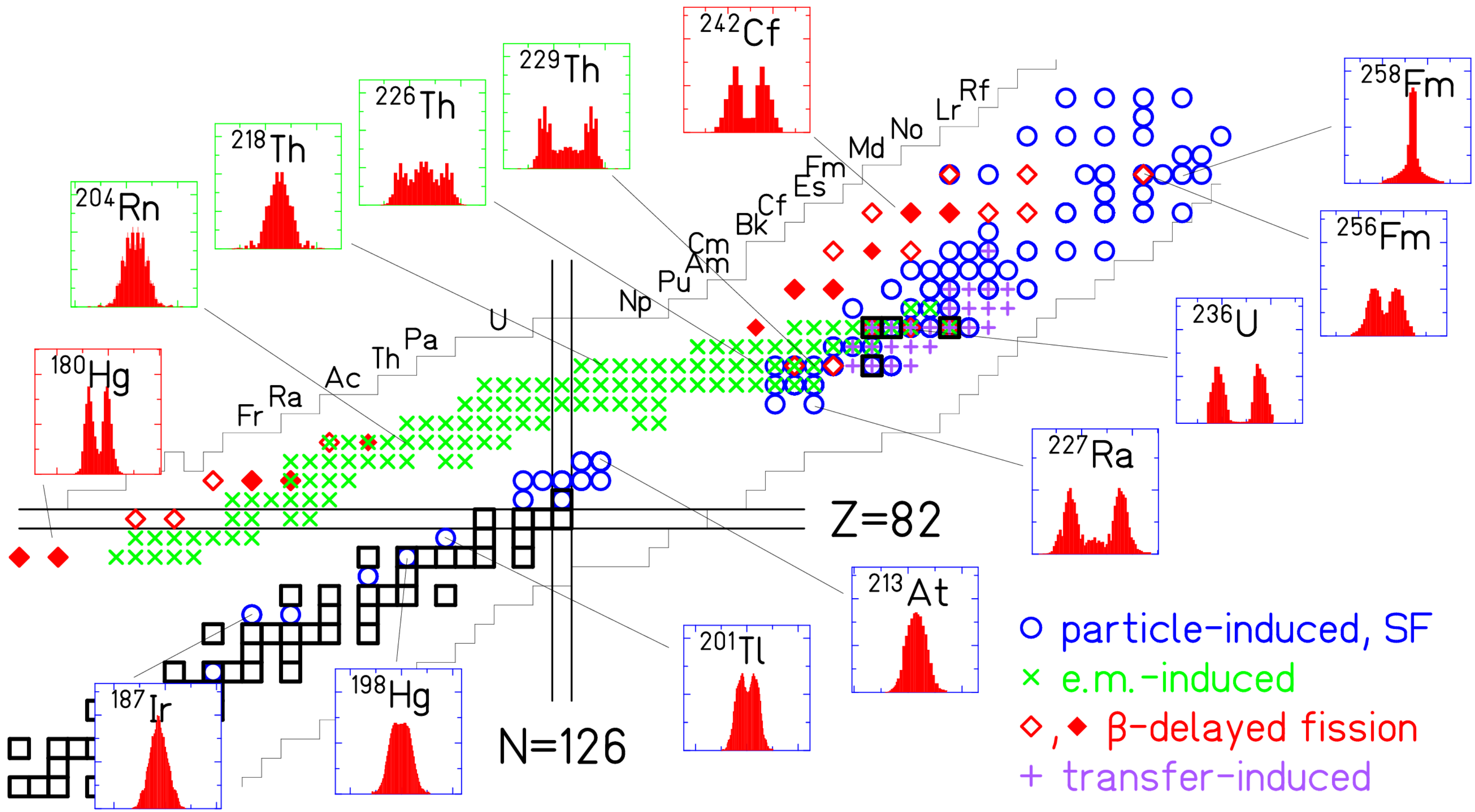
FY from SOFIA



Coverage of all fission fragments.
FY data with high precision and accuracy.

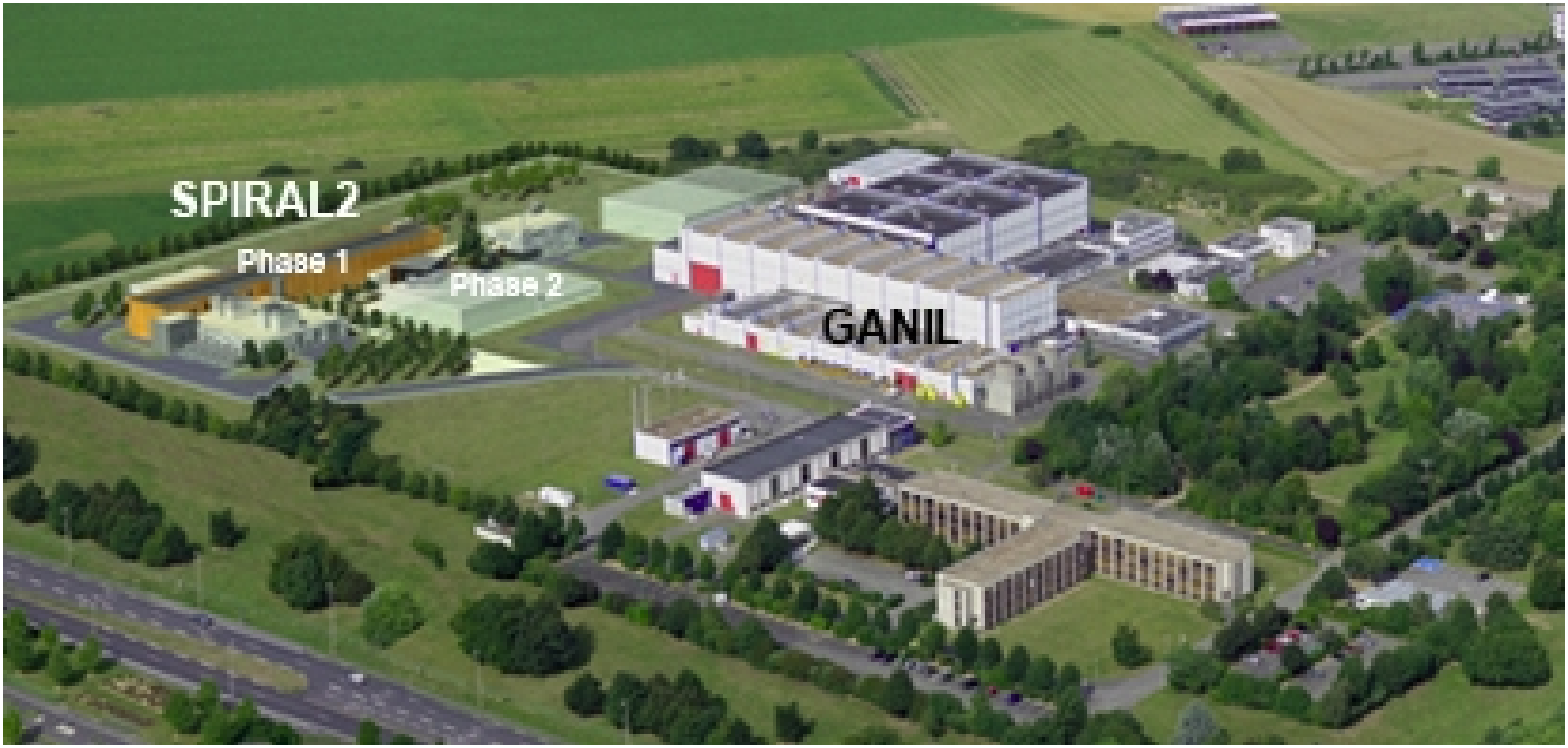
Figure from Chatillon, EPJ Web of Conferences 111 (2016) 08001

Overview on FY (A and Z distributions)



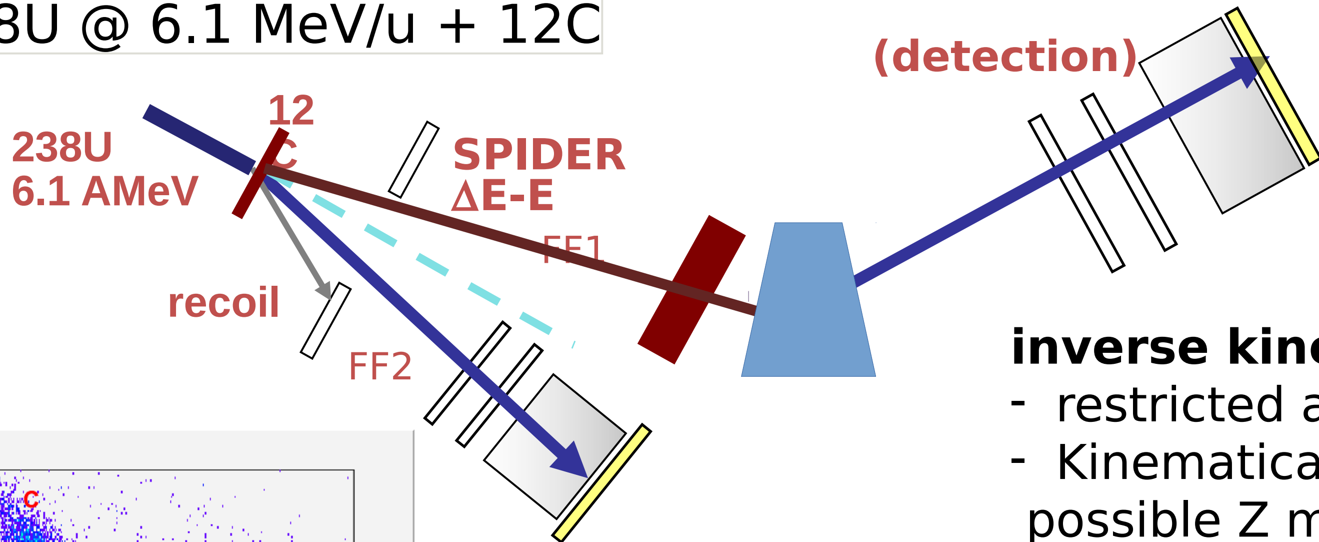
Many systems investigated by different methods. Strong variations found. Only few around 235U are relevant for nuclear technology.

Fission experiments with 238U (6 A MeV) at GANIL, Caen (France)

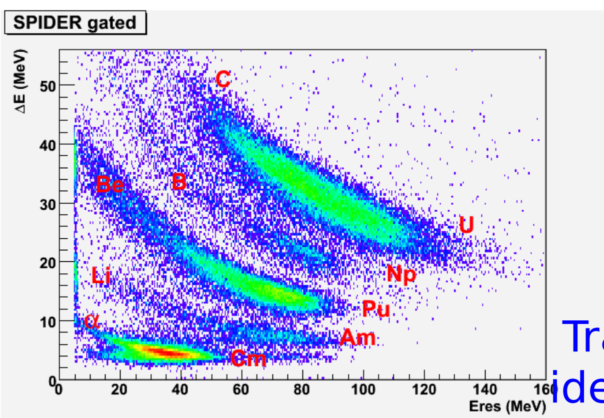


Transfer-induced fission in inverse kinematics

238U @ 6.1 MeV/u + 12C

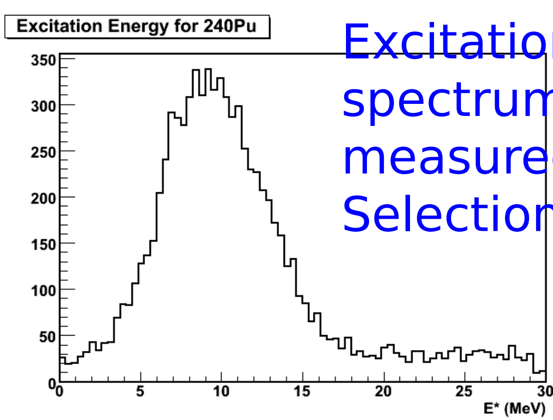


inverse kinematics
- restricted angular distribution
- Kinematical push that makes possible Z measurement
- Spectrometer to resolve masses

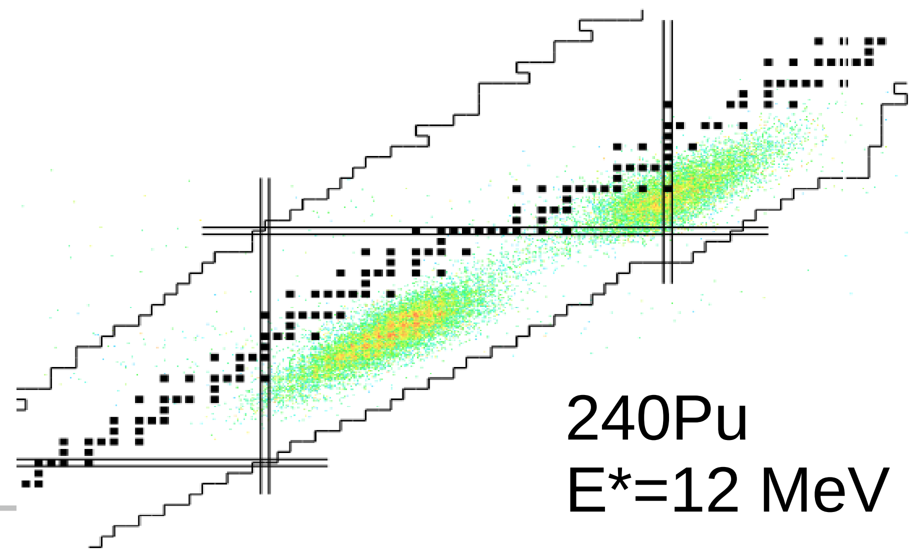


Transfer channels identified in SPIDER.

Corresponding isotopic and mass yields



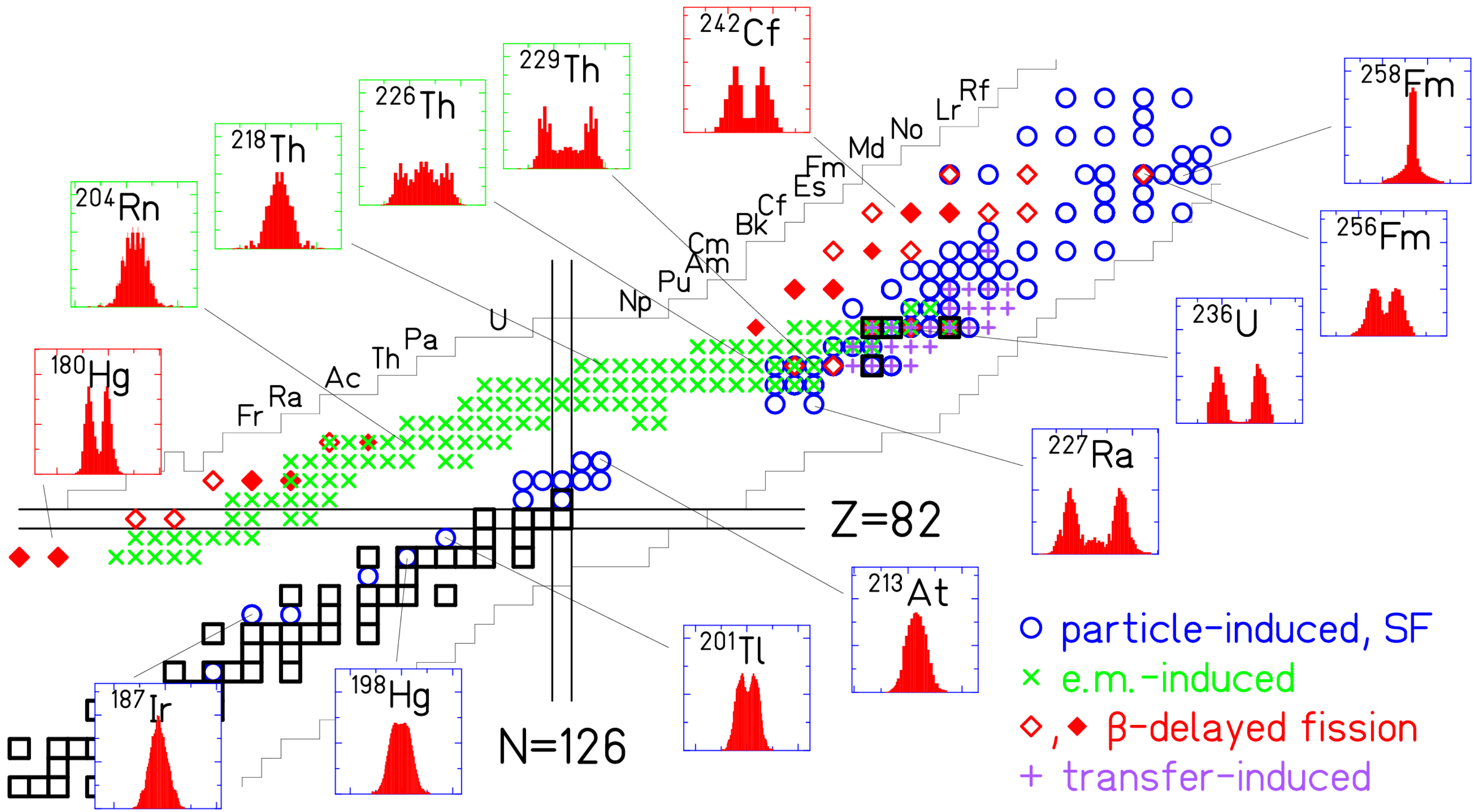
Excitation energy spectrum measured. Selection of E*!



240Pu
E*=12 MeV

Fanny Farget, 2013

Overview on FY (A and Z distributions)



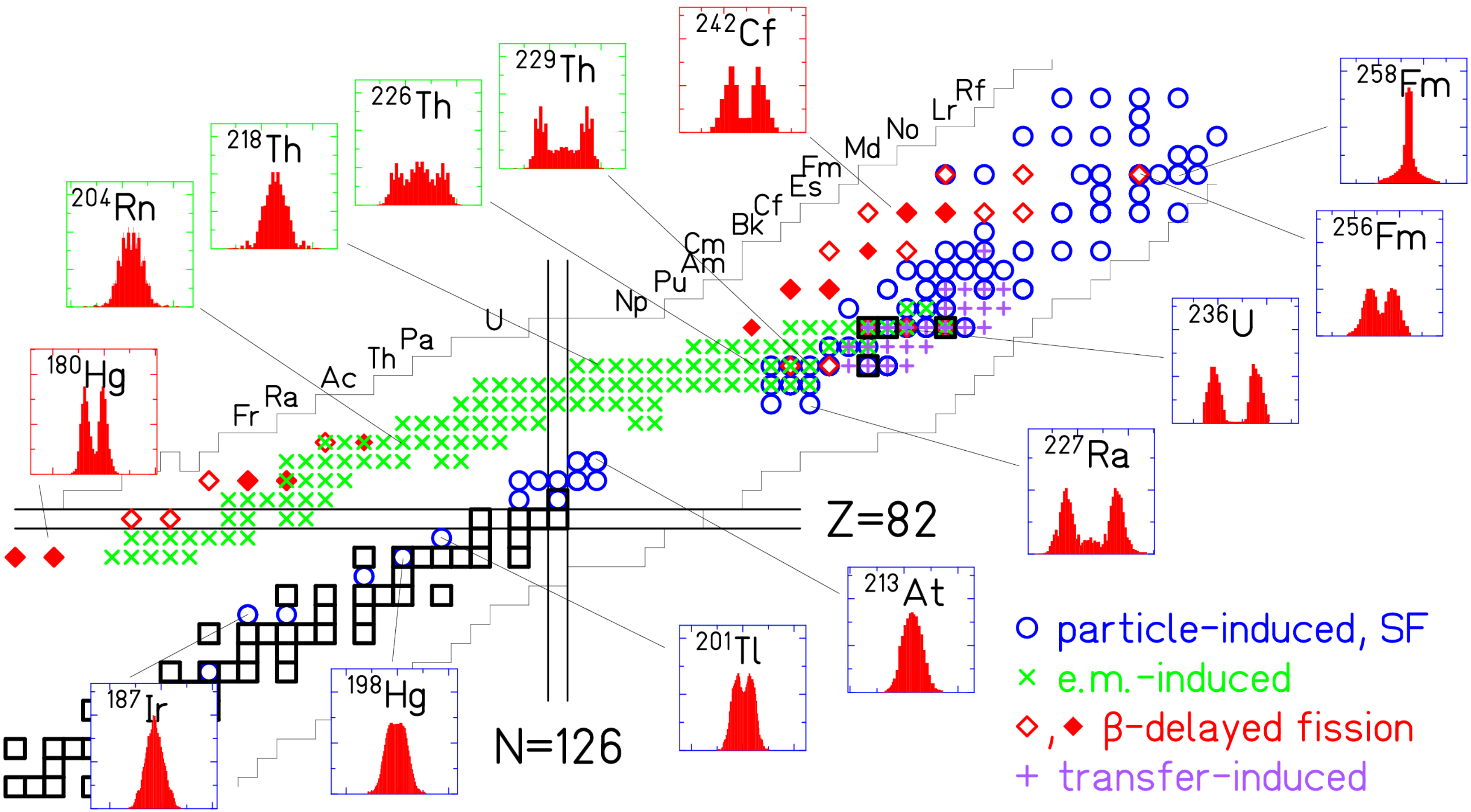
Many systems investigated by different methods. Strong variations found. Only few around 235U are relevant for nuclear technology.

Excitation mechanisms

Excitation mechanisms

- Neutron-induced fission
  - Relevant for fission reactors
  - Mostly thermal, fast, 14-MeV (main source for FY evaluations)
  - Few suitable targets
- Proton-induced fission
  - Easily tunable energy
- LCP or C/O-induced (fusion/transfer)
  - Several systems around target/beam accessible
  - E* can be selected
- Heavy-ion fusion: CN-fission/spontaneous fission
  - Exotic, very heavy systems
- Beta-delayed fission
  - Limited to few, neutron-deficient systems
- Coulomb-induced fission
  - Bremsstrahlung, high-Z targets → often broad E* distribution

Overview on FY (A and Z distributions)



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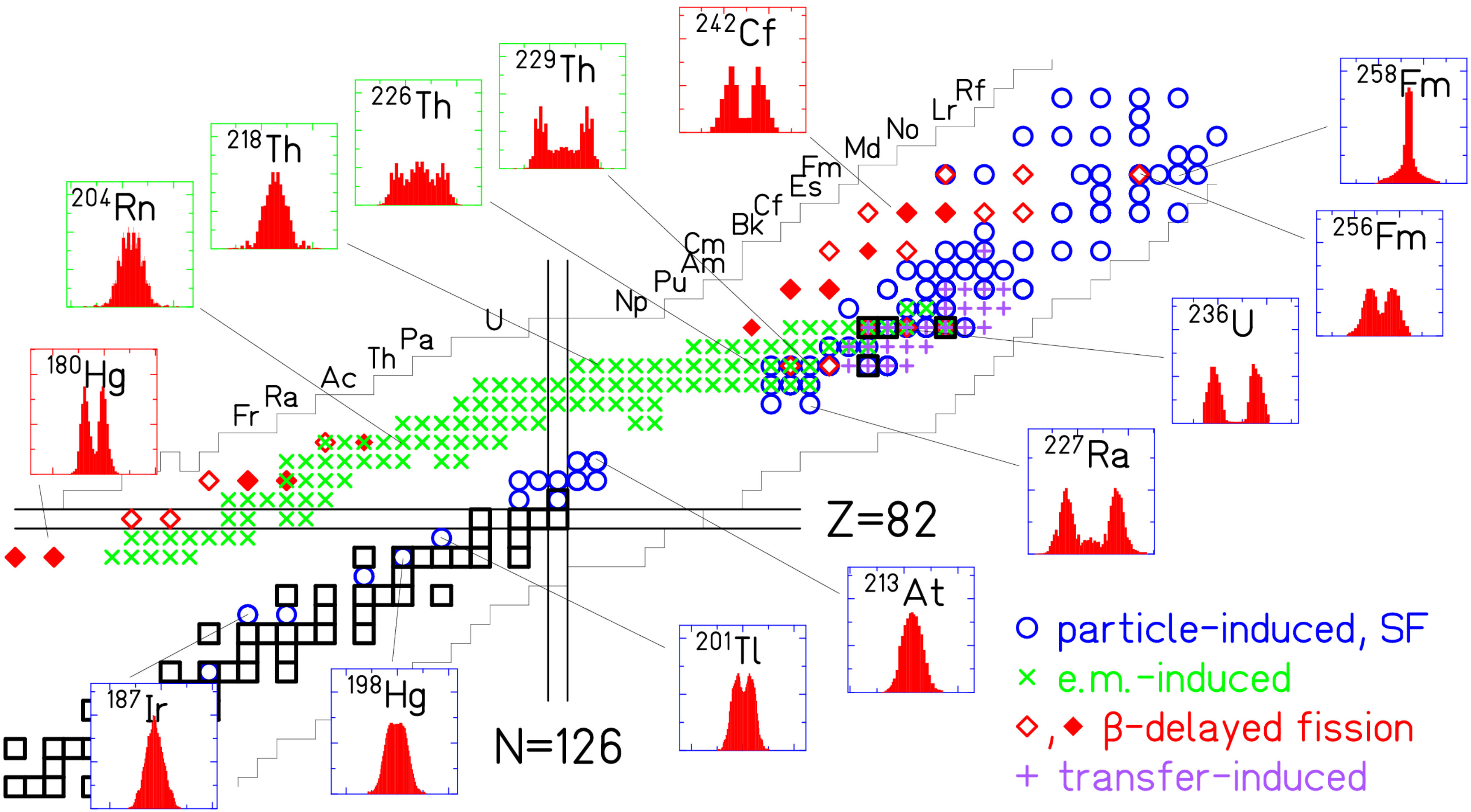
Characteristics of FY

Overview on FY (A and Z distributions)



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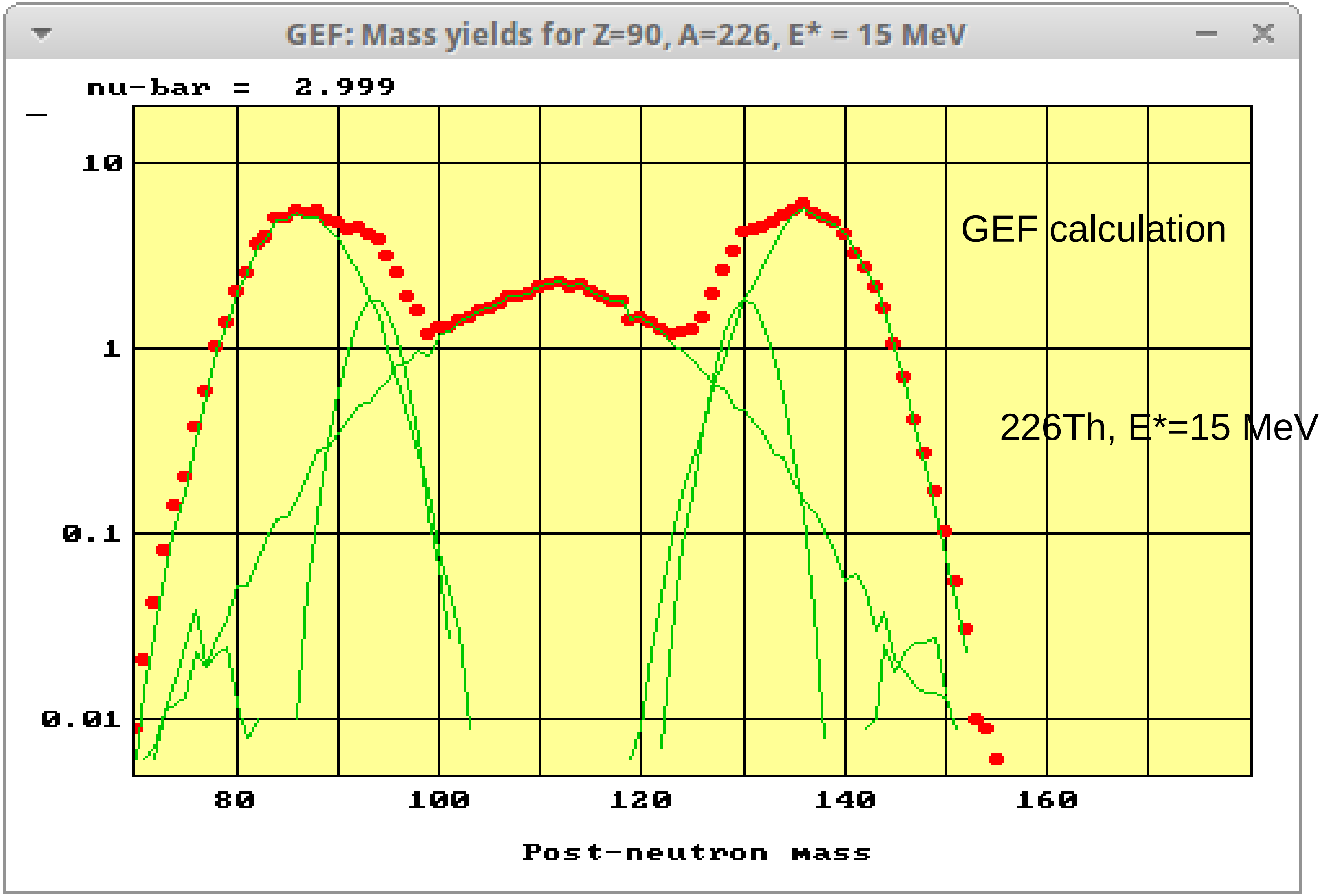
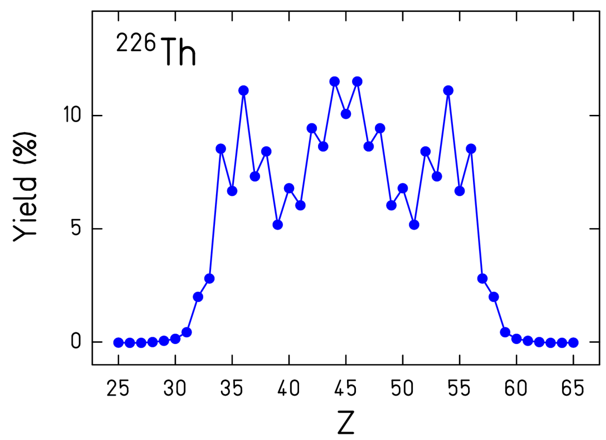
Fission channels

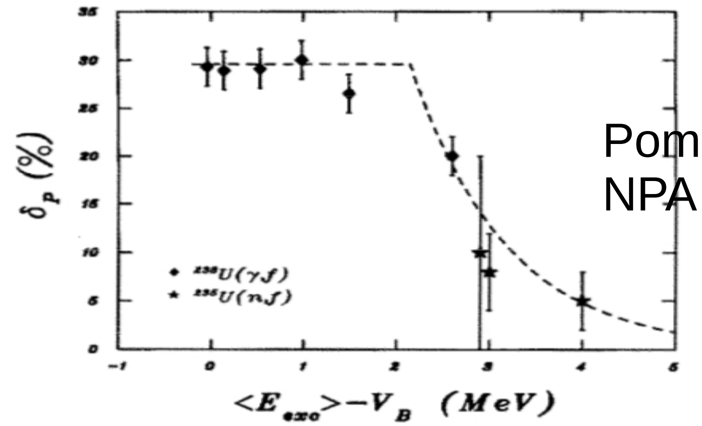


Gaussian-like components in the mass distribution

Even-odd effect in Z yields

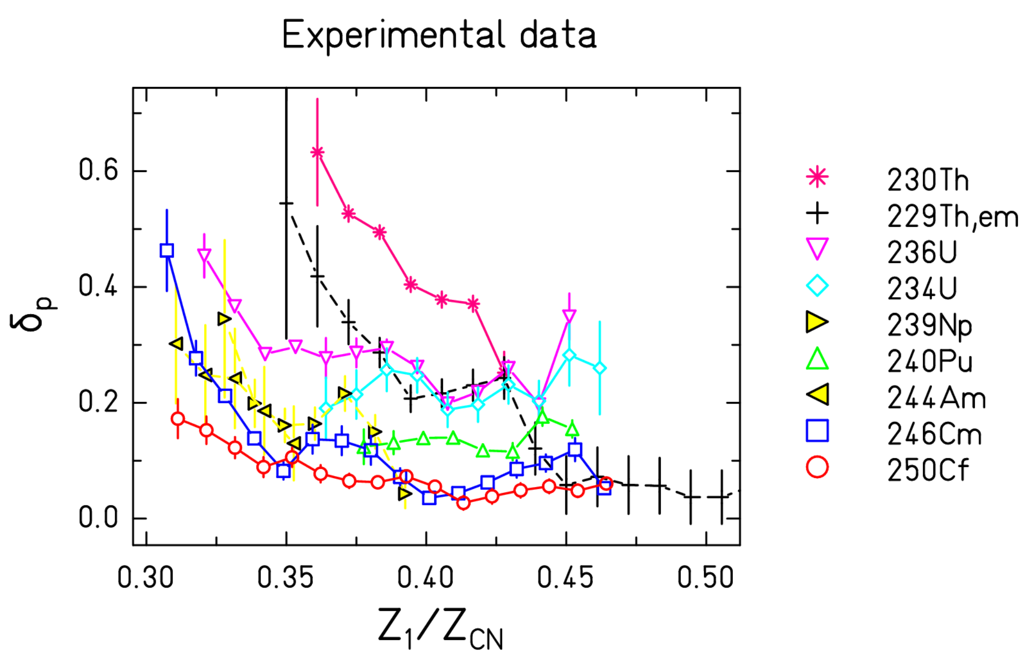


Even-odd staggering in Z yields

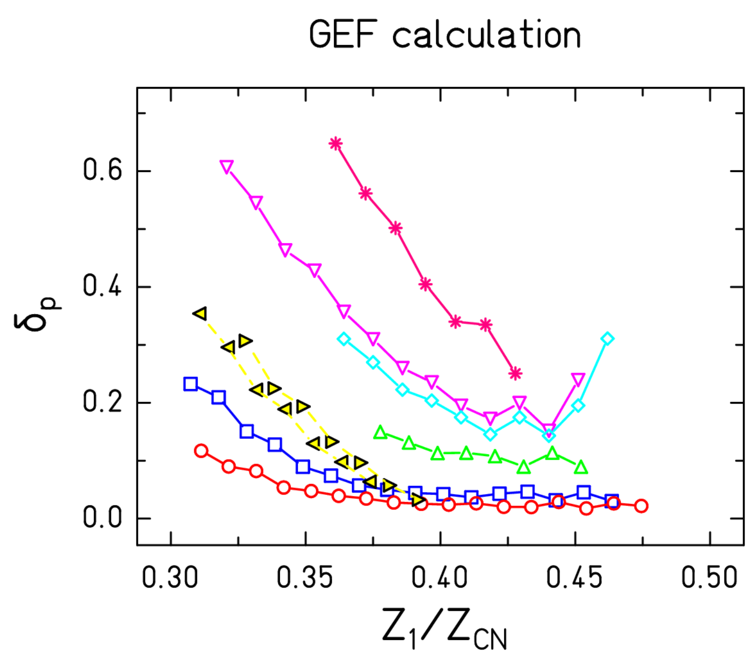


Pomme et al., NPA 560 689

E* dependence

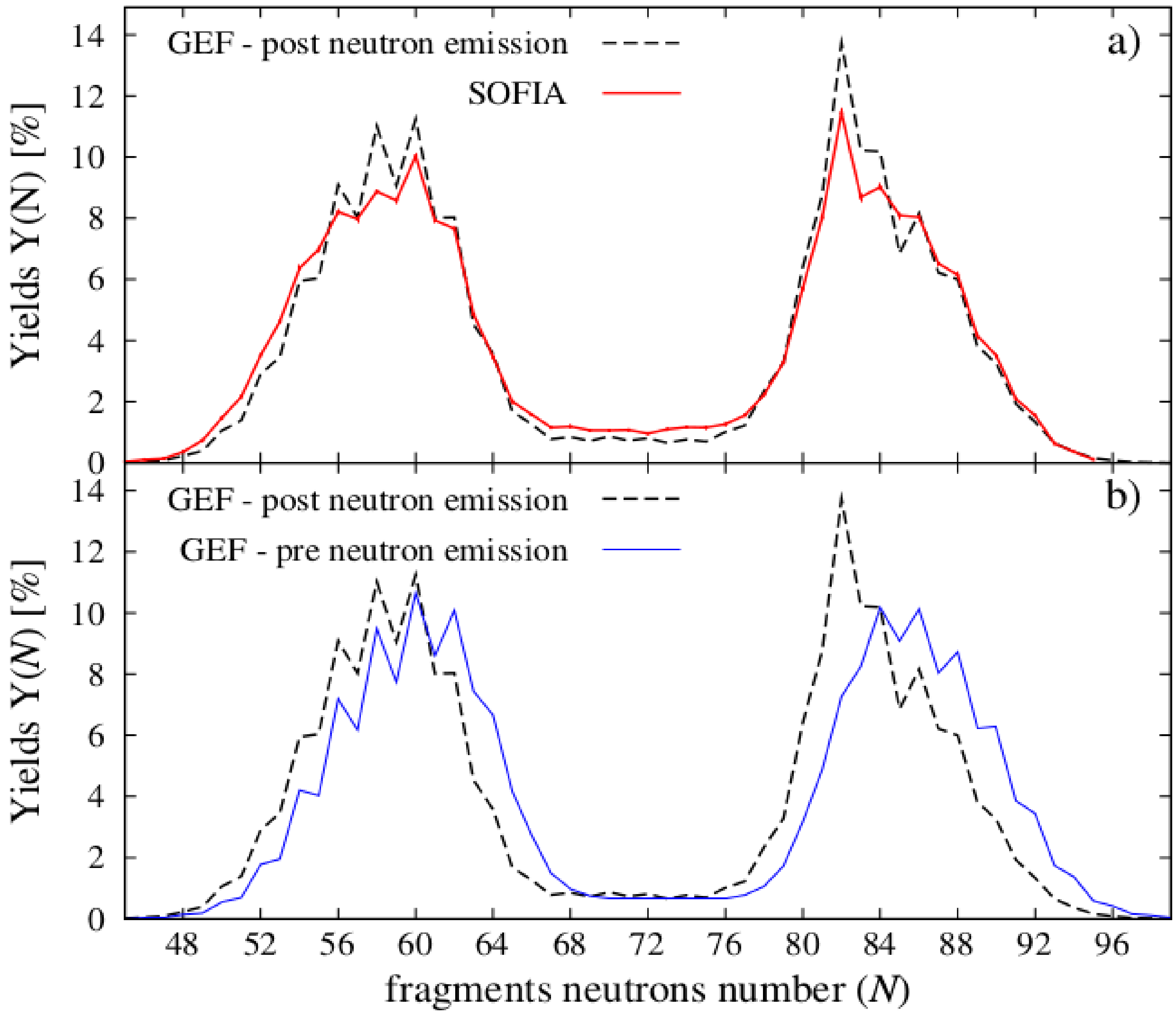


Increase with asymmetry



Reproduction by the GEF model

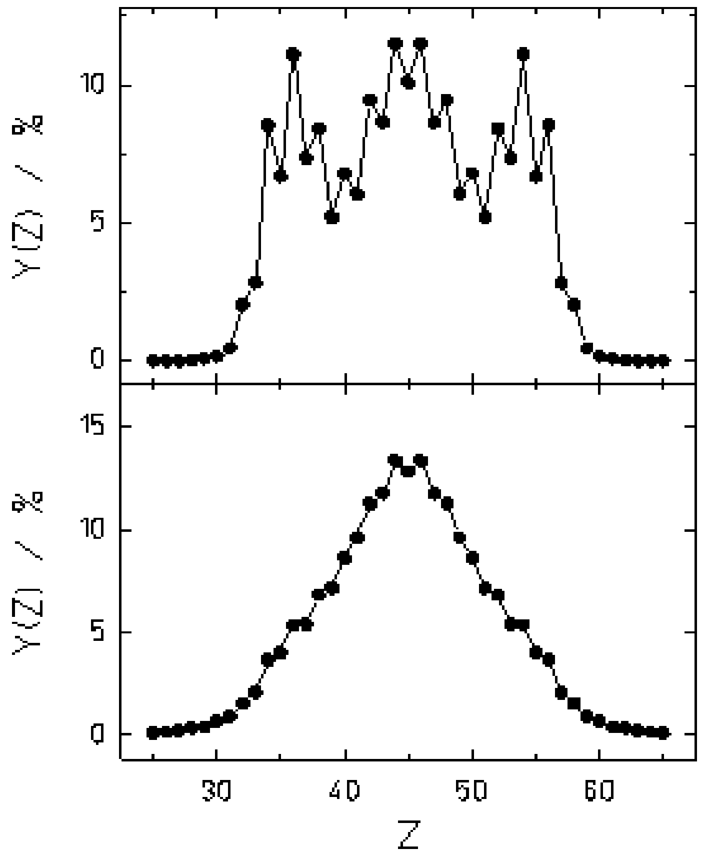
Fine structure in N yields



From Pellereau et al.
PRC 95 (2017) 054603

Fine structure in N yields before (not observable) and after prompt-neutron emission.
Post-neutron fine structure does not depend on E*!

Evolution with E*



Fission of ^{226}Th

← Fission at low excitation energy (Coulomb fission, E* = 14 MeV)

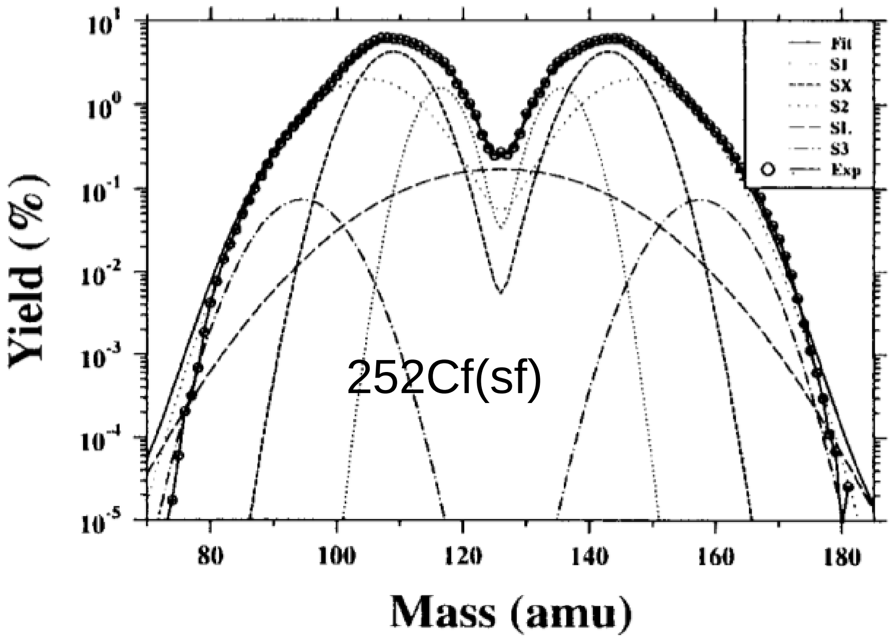
← Fission at high excitation energy (after nuclear interactions, E* > 30 MeV)

Even-odd effect and fission channels disappear → Gaussian-like shape without structure

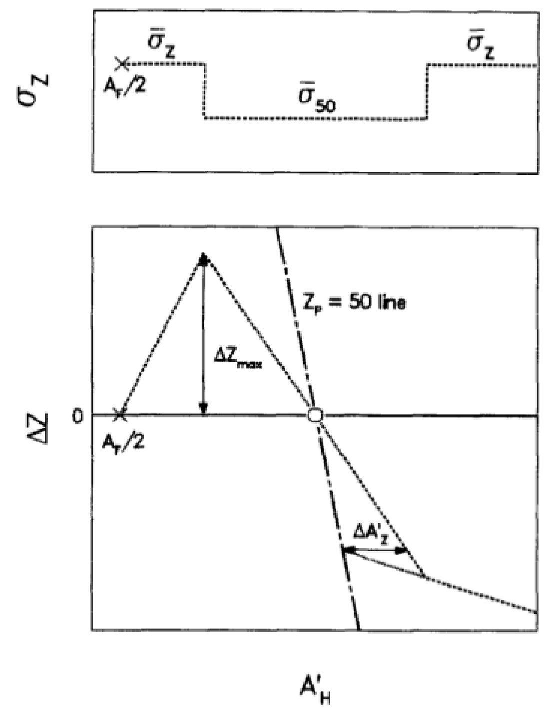
Systematics and evaluations

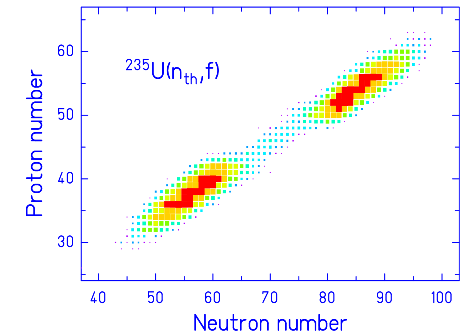
Systematics (e.g. Wahl)

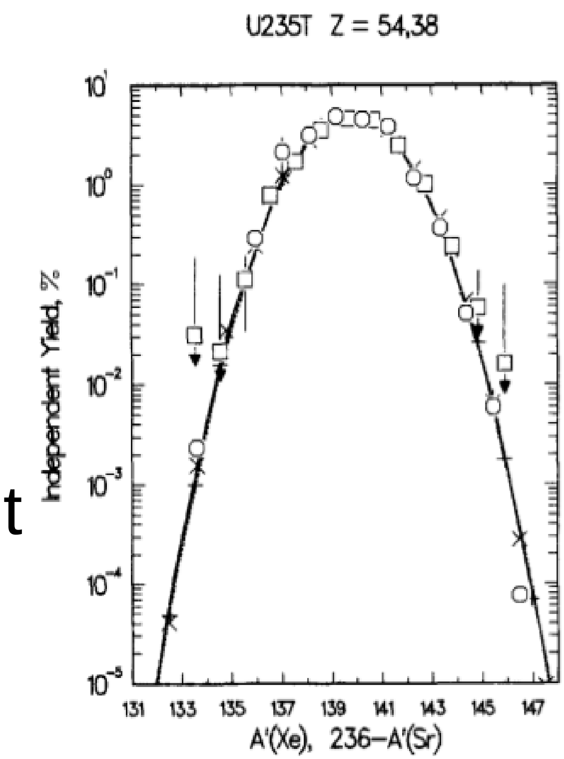
1988

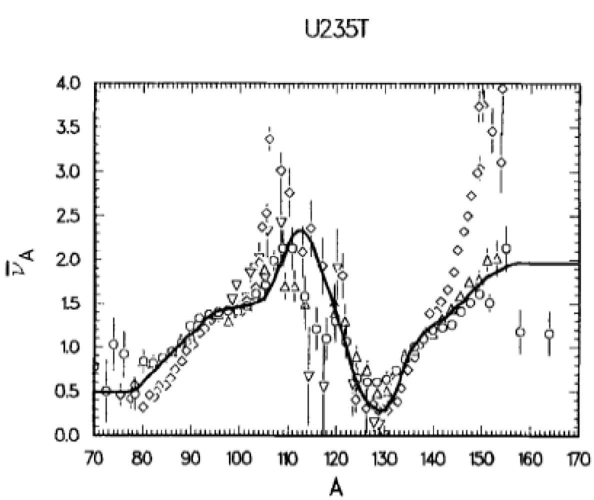


235U(nth,f)









Neutron yields

Analytical functions fitted to measured data. Good quality in case of sufficient data. Still used for applications. Little predictive power.

Evaluation

- **What is evaluation?**
  - Assesses experimental data and their uncertainties.
  - Reconciles discrepant data .
  - Fills in missing data from systematics (e.g. Wahl) and theories.
  - Enforces consistency by normalization (Σ(FY) = 200 %).
  - Enforces consistency by symmetrization (light and heavy fragment).
  - Introduces relations between independent and cumulative yields.
  - Considers constraints by integral and other data.
- **Who does it?**
  - National and international nuclear-reaction data centers under the auspices of the IAEA.
  - ENDF:(England and Rider, ...), JEFF: (Mills), JENDL, BROND, CENDL.
- **How is it published?**
  - Nuclear-data files provided by the Nuclear-Data Banks and the IAEA.
  - Traditionally published in ENDF-6 format (punch-card format).
  - GND (General Nuclear Data) format being developed (XML-based).

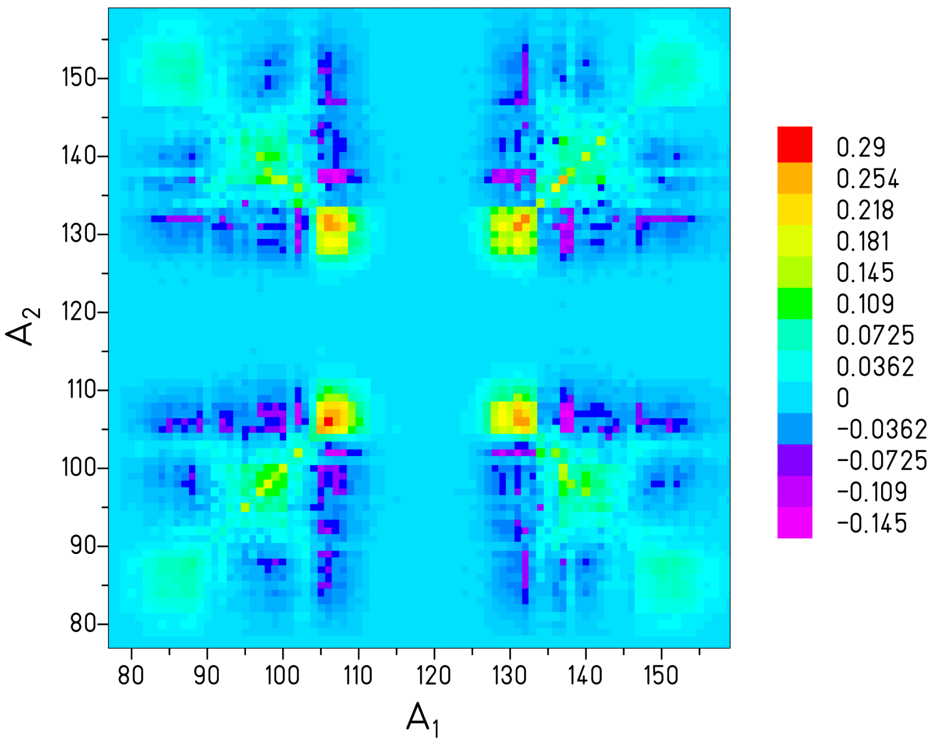
Uncertainties and covariances

Uncertainties and covariances

Complex calculations using FY require uncertainties and correlations.

Uncertainties and correlations of different nature are provided by experiment, trivial consistencies and models.

Work on correlations (covariances) is intense but still in development.



Typical example:

Covariances between the FF mass yields from GEF.

Conclusion

- The FY show the footprints of the stages of the fission process. Many are measured, some predicted by models (e.g. GEF).
- Different kind of FY: pre-neutron, post-neutron, cumulative.
- Post-neutron (independent) and cumulative yields are evaluated and published in nuclear-data tables (ENDF, JEFF etc.).
- Most evaluated FY data still rely on neutron-induced fission and radiochemical methods. Data from advanced approaches need new analysis and evaluation methods.
- Full identification of all fragments in Z and A has been achieved by inverse-kinematics methods with high technical expense.
- Choice of available systems made big progress, but there are still important restrictions.
- Knowledge on evolution of FY with E* is fragmentary.
- Work with fission-yield data requires knowledge in many fields like fission physics, detector technology, assessment, uncertainty quantification, correlations, and integral tests.