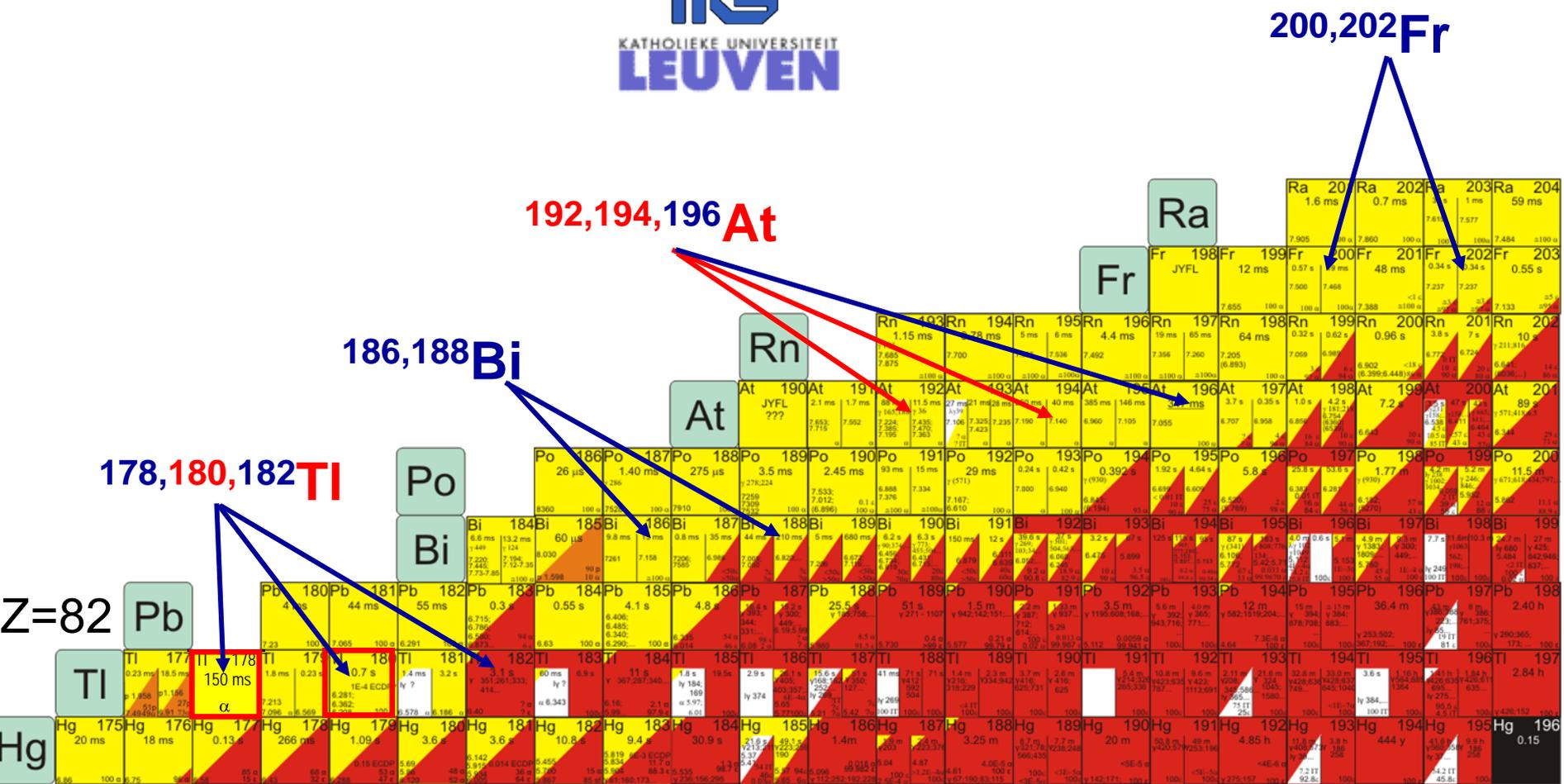


Electron-Capture Delayed Fission (ECDF) in the Pb region

A Andrei Andreyev



Collaboration



Andrei Andreyev
Nick Bree
Thomas Cocolios
Jan Diriken
Jytte Elseviers
Mark Huyse
Paul Van den Bergh
Piet Van Duppen
Martin Venhart



Stanislav Antalic



Katsuhisa Nishio



Jarno Van De Walle
RILIS & ISOLDE



Robert Page

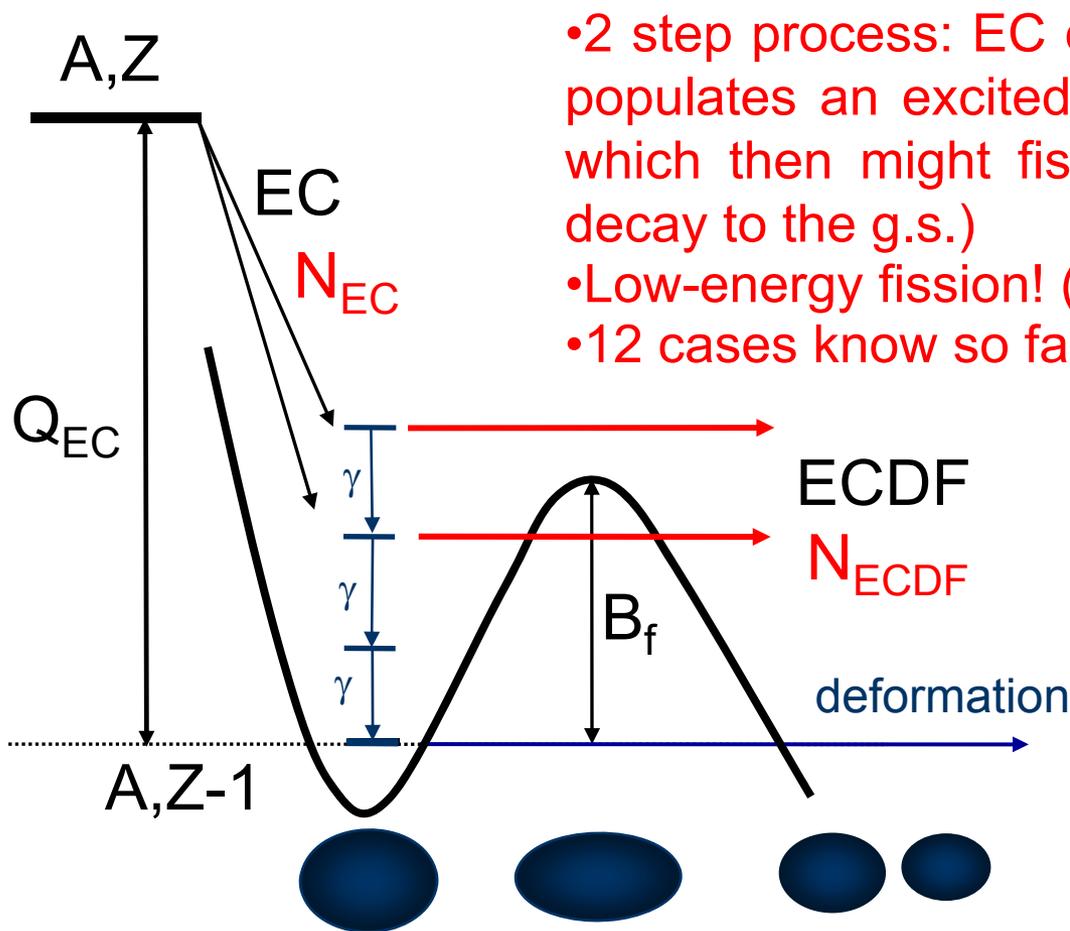
U. Koster (*ILL, Grenoble, France*)
S. Franchoo (*IPN, Orsay, France*)
S. Vermote, C. Wagemans (*University of Gent, Belgium*)
M. Veselský (*Slovak Academy of Sciences, Bratislava, Slovakia*)
I. Tsekhanovich (*Manchester University, UK*)

Outlook

- Electron-Capture Delayed Fission- what it is and why?
- Earlier ECDF studies in the U and Pb regions
- ECDF of $^{192,194}\text{At}$ at SHIP: cold fission of $^{192,194}\text{Po}$
- ECDF of ^{180}Tl at ISOLDE: **first ever fission study at ISOLDE?**
- Plans

Electron-Capture Delayed Fission (ECDF, $T_{1/2}(ff)=T_{1/2}(EC)$)

Discovery: parent isotopes $^{232,234}\text{Am}$ (1966, Dubna)



- 2 step process: EC decay of a parent (A,Z) nucleus populates an excited state in the (A,Z-1) daughter, which then might fission (in competition with the γ decay to the g.s.)
- Low-energy fission! ($E^* \sim 5-10$ MeV)
- 12 cases known so far (neutron-def. Uranium region)

EC-delayed branch

$$P_{\text{ECDF}} = \frac{N_{\text{ECDF}}}{N_{\text{EC}}}$$

P_{ECDF} depends strongly on:

- Q_{EC} of the parent: the higher Q_{EC} , the larger the P_{ECDF}
- B_{fis} of the daughter: the lower B_{fis} , the larger the P_{ECDF}
- Actually, $Q_{\text{EC}} - B_{\text{fis}}$ is important

ECDF Probability: Feeding Part & Decay Part

$$P_{\text{ECDF}} = \frac{N_{\text{ECDF}}}{N_{\text{EC}}} = \frac{\int_0^{Q_{\text{EC}}} (Q_{\text{EC}} - E)^2 \times S_{\beta}(E) \frac{\Gamma_f(E)}{\Gamma_{\text{tot}}(E)} dE}{\int_0^{Q_{\text{EC}}} (Q_{\text{EC}} - E)^2 \times S_{\beta}(E) dE}$$

$(Q_{\text{EC}} - E)^2$ – Phase factor for EC decay

$S_{\beta}(E)$ – β -strength function (nuclear matrix element)

$\frac{\Gamma_f}{\Gamma_{\text{tot}}} = \frac{\Gamma_f}{\Gamma_f + \Gamma_{\gamma}}$ -ratio of the fission and total widths of excited levels in daughter (Γ_n is not important for neutron-deficient nuclei)

$\Gamma_{\gamma} = \frac{9.7 \times 10^{-7} \times T^4 \times \exp(E/T)}{2\pi\rho}$, ρ – level density, T - temperature

$\Gamma_f = \frac{1}{2\pi\rho} \left\{ 1 + \exp\left[\frac{2\pi(B_f - E)}{h\omega_f}\right] \right\}^{-1}$ -inverted parabola approximation
D.L. Hill and J.A. Wheeler

Measurement of P_{ECDF} allows to deduce Fission Barrier B_f

β -delayed Fission and r-process Nucleosynthesis

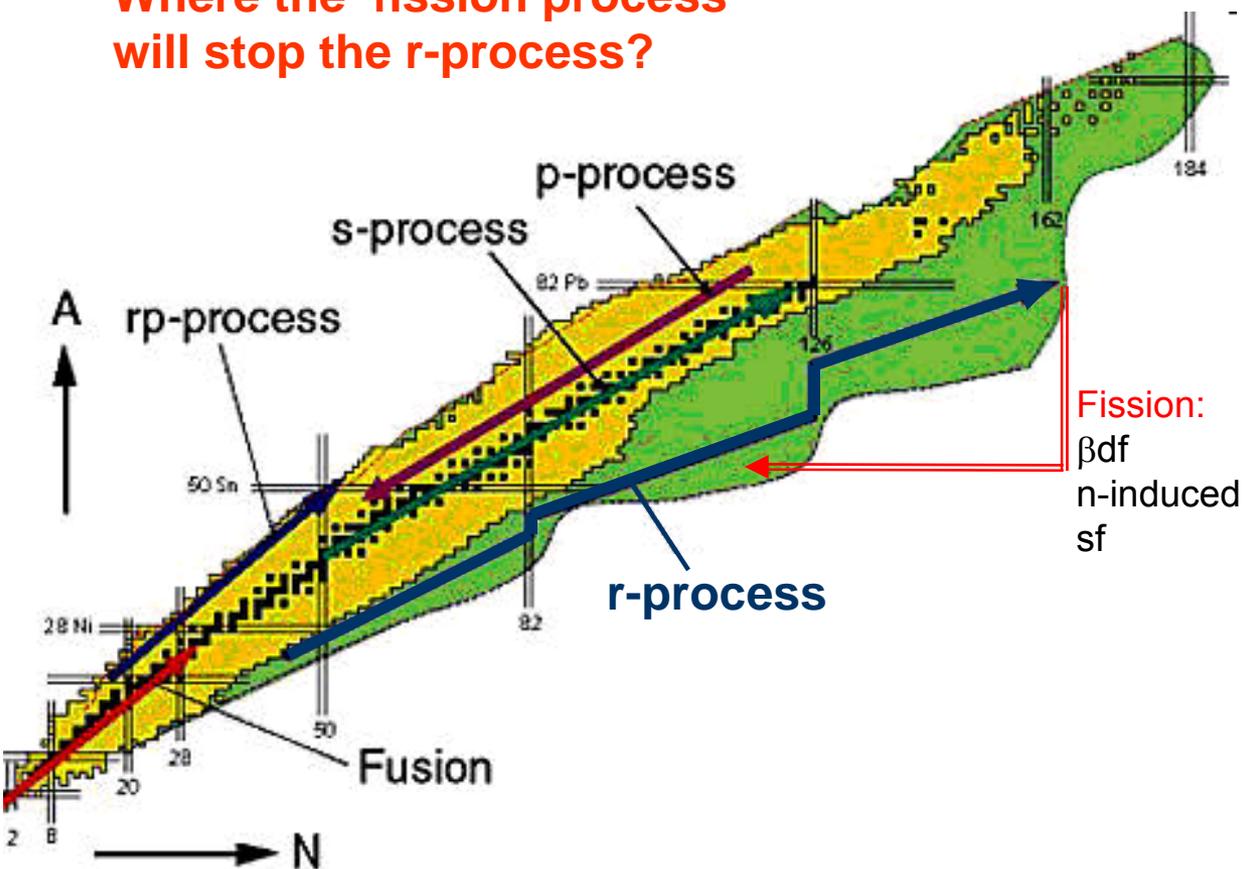
End point of r-process?

I. Panov et al, NPA747 (2005) 633

Trans-Uranium elements?

Cowan et al, Phys. Rep. 208 (1991) 267

Where the fission process will stop the r-process?



Fission cycling: **Fission products can serve as seed for the r-process!**

I. Panov et al, NPAA747 (2005) 633

r-process to the region with $A > 250$

r-process

Fission: β df n-induced, sf

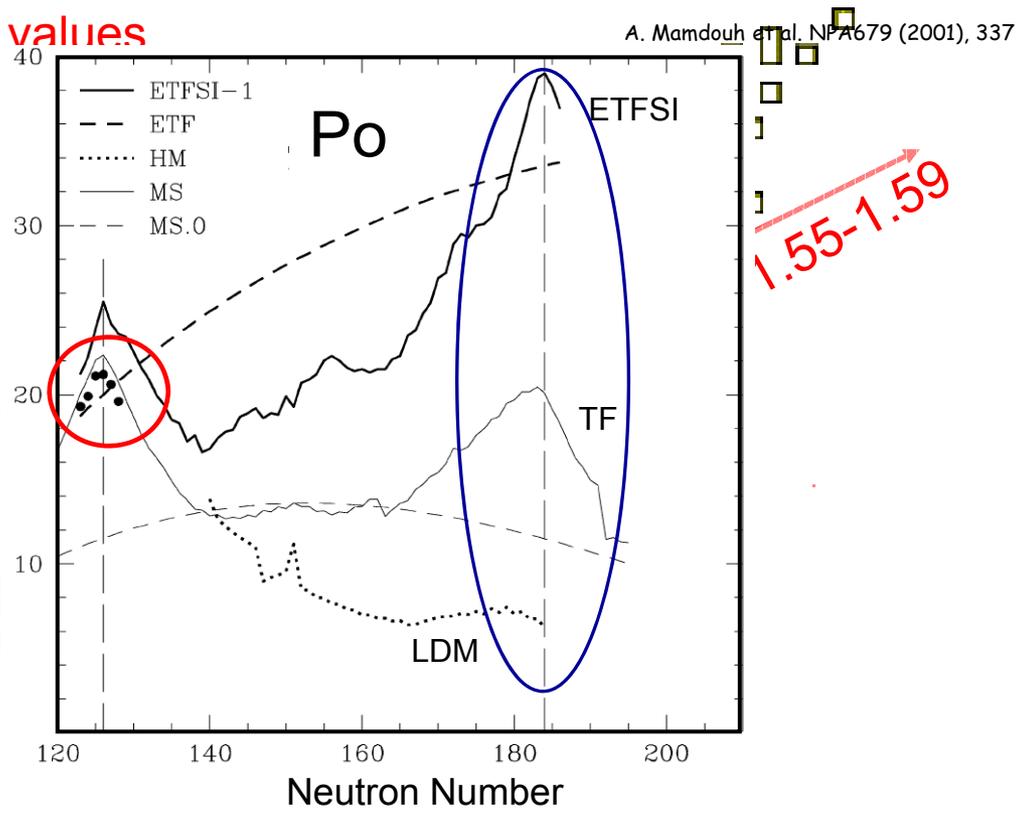
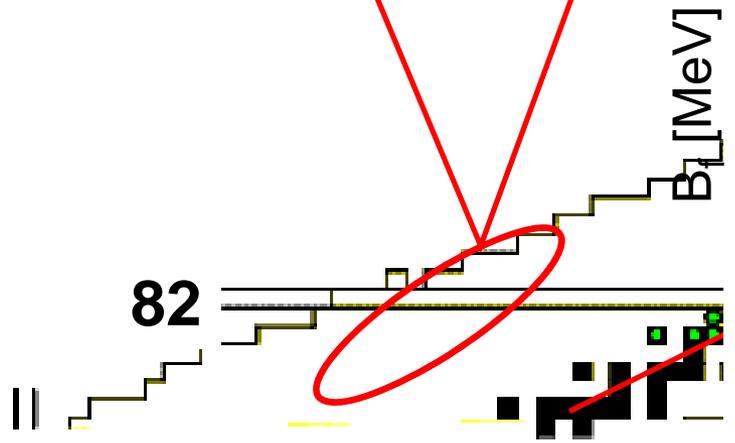
Fission $A \rightarrow A/2 \sim 125$

Need fission data for very heavy exotic nuclei!

Why ECDF studies?

- Experimentally fission barrier are known **only in the vicinity of the β -stability line** (e.g. $N/Z(^{238}\text{U})=1.59$)
- Theoretical models for B_f have been 'tuned' by using these data
- Large discrepancies between different models for n-def. and n-rich nuclei
- Must study isospin dependence of B_f values**

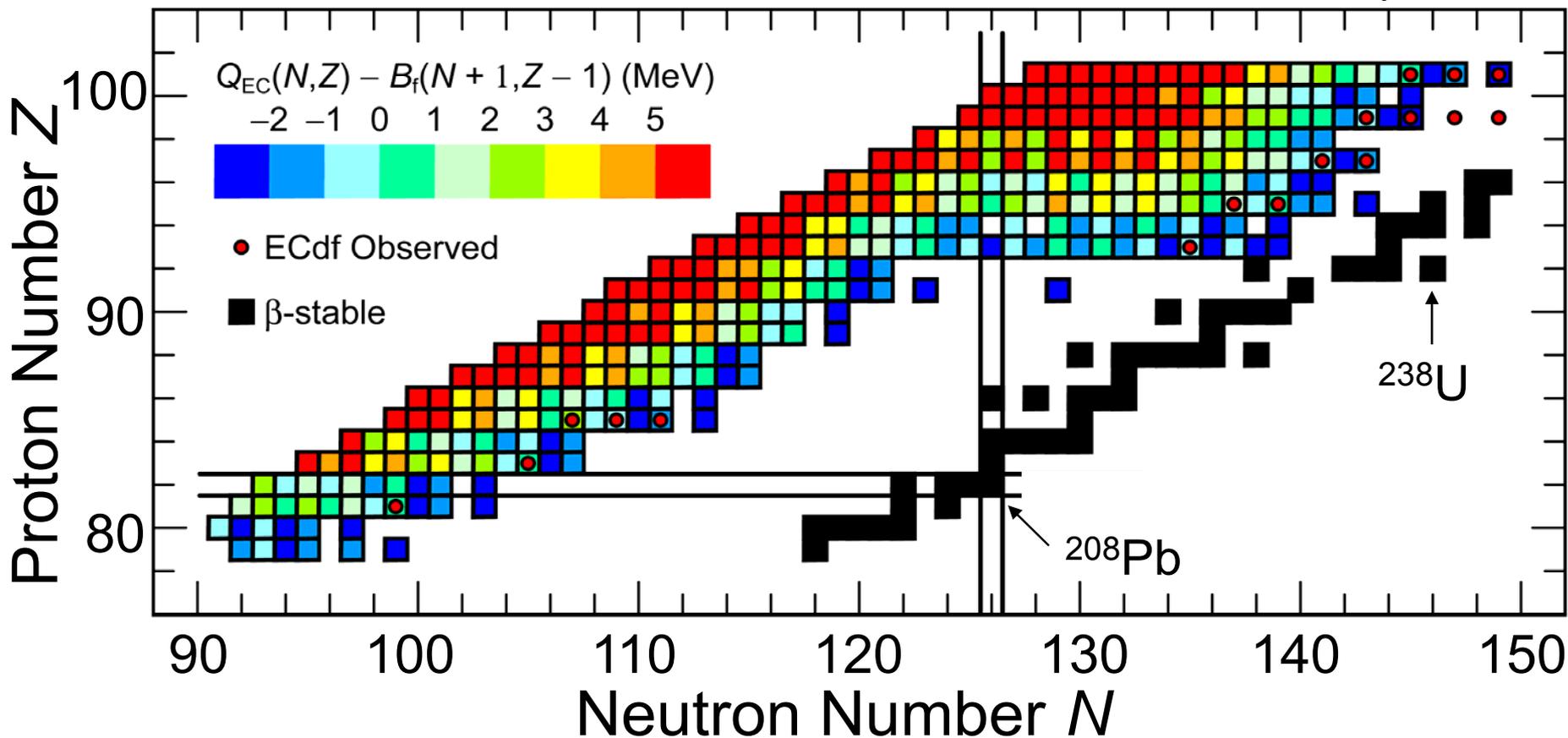
Region of our interest: $A \sim 180-200$
 $N/Z \sim 1.22-1.3$



- ECDF gives access to nuclei with very exotic N/Z ratios: e.g. $N/Z(^{178}\text{Hg})=1.225$
- Unexpected properties, e.g. mass-distributions, cold fission...?

Calculated Energy Window for EC-delayed Fission

Courtesy P. Möller

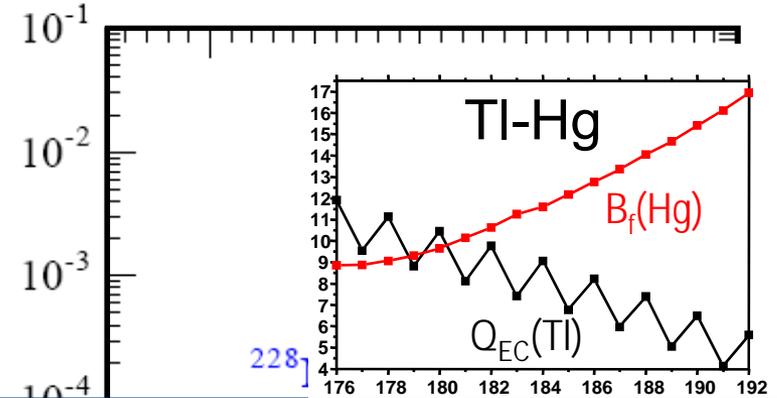
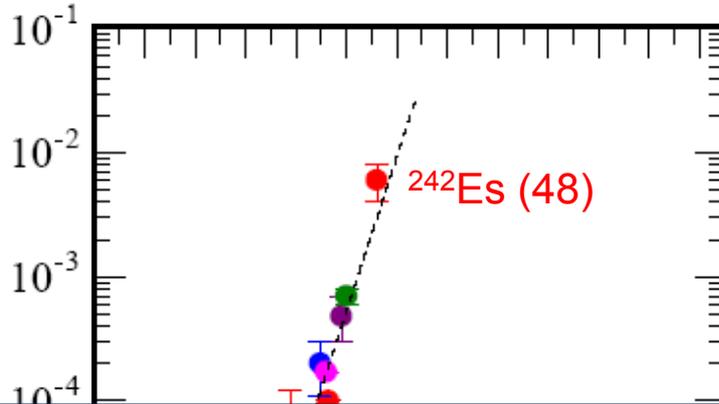


Q_{EC} values: P. Möller et al., ADNDT masses (1995), based on FRDM (1992) 8

B_f values: P. Möller et al., submitted to PRC (2008)

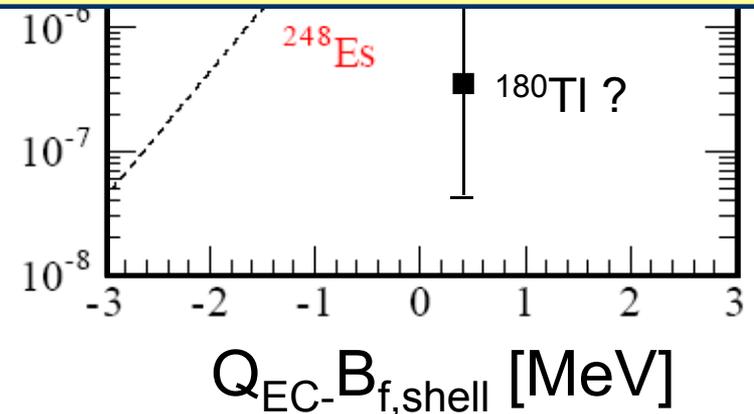
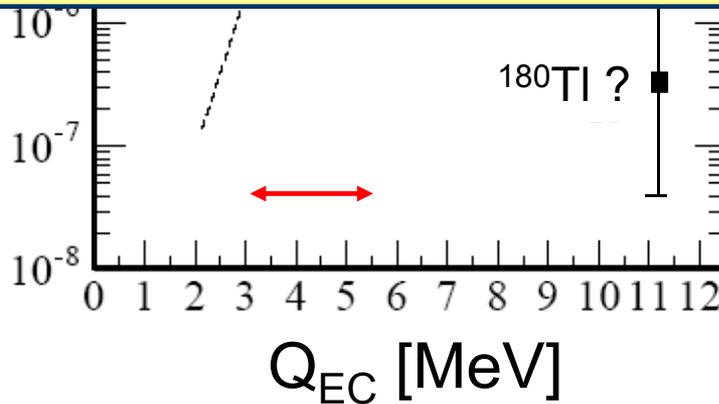
ECDF in trans-U region

- 12 known ECDF cases in trans-Uranium region (all odd-odd!)
- Relatively low Q_{EC} and B_f values (3-5 MeV)

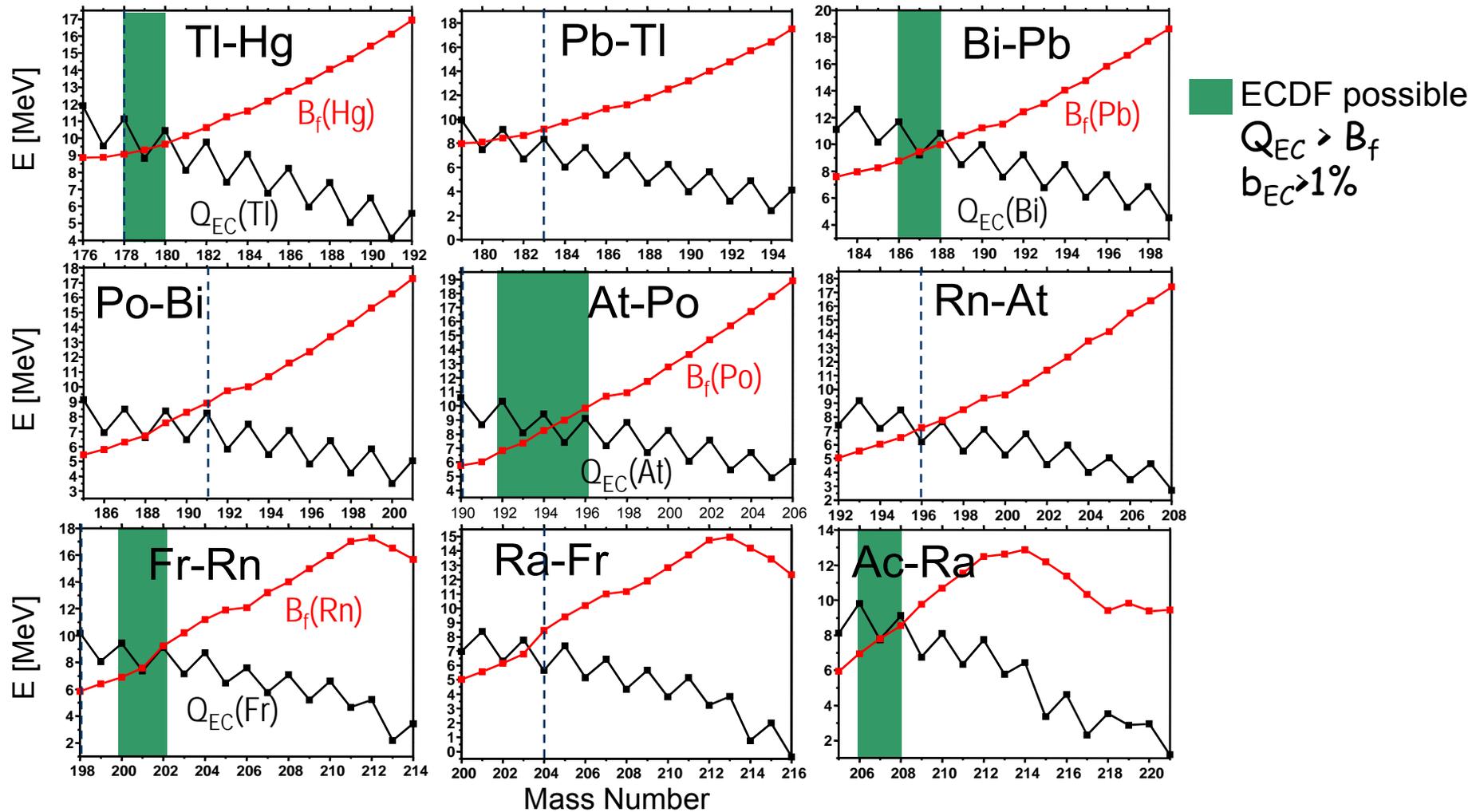


Why odd-odd EC-decaying parents?

- Larger Q_{EC} in comparison with e-e and o-e neighbors (odd-even effect)
- Even-even daughters are more fissile (specialization energy)



Q_{EC} vs B_f values in Pb region (look for cases $Q_{EC} > B_f$)



Red lines: Thomas-Fermi Fission Barriers, W.D. Myers, W. Swiatecki Phys. Rev. C60 (1999) 014606

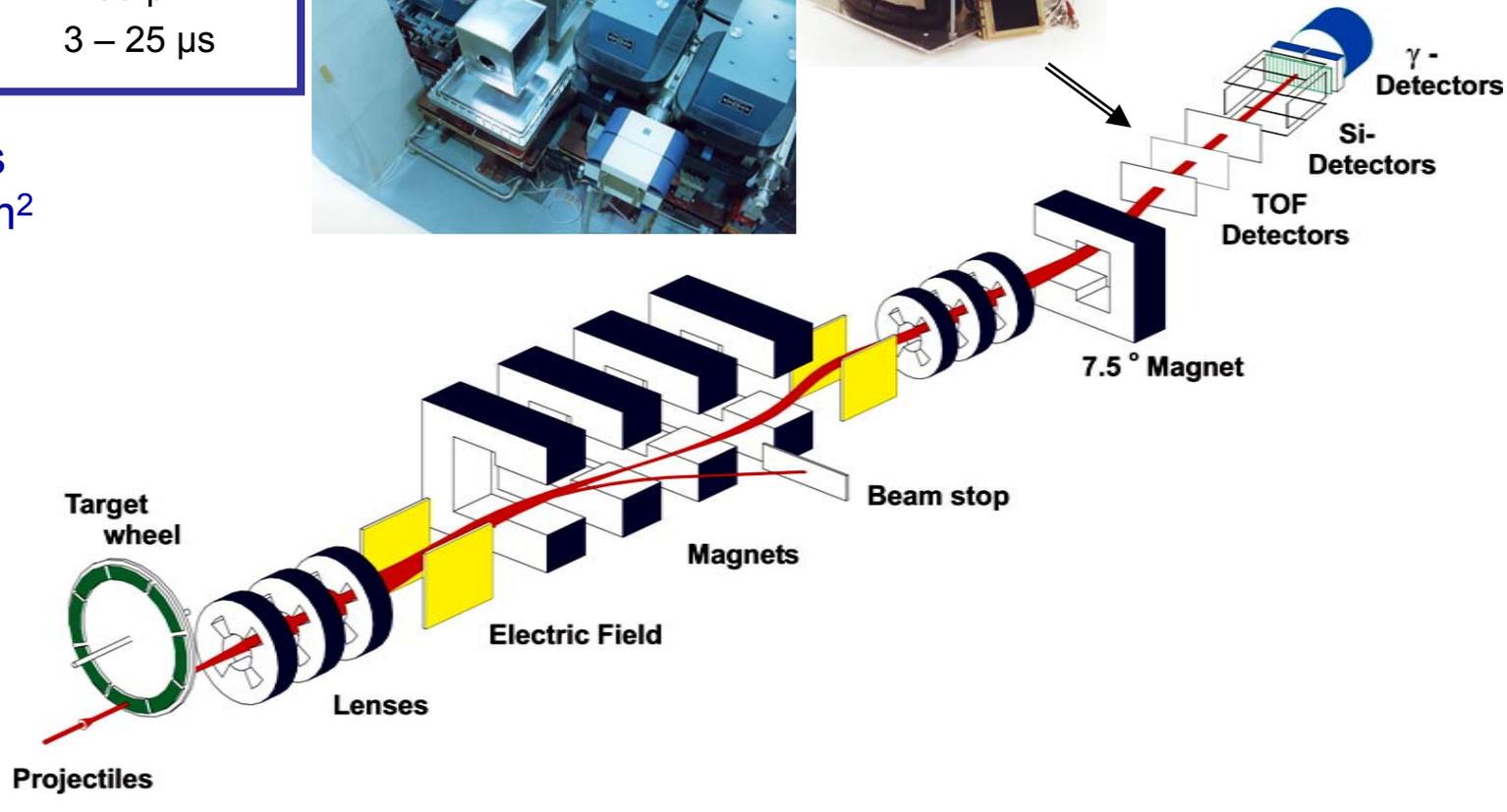
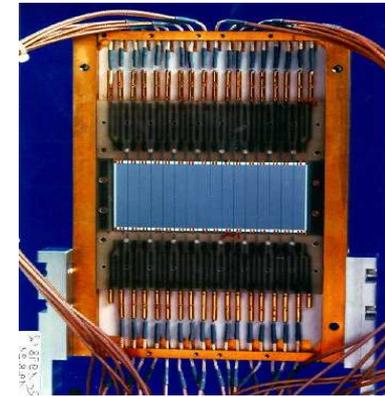
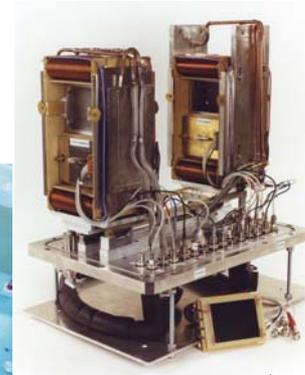
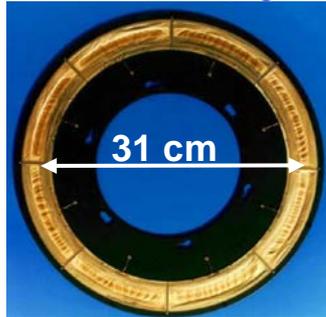
Black lines: Q_{EC} -values, P. Moller et al. At. Data and Nucl. Data table 59 (1995) 185

Velocity Filter SHIP (GSI, Darmstadt)

SHIP:

Separation time:	1 – 2 μs
Transmission:	20 – 50 %
Background:	10 – 50 Hz
Det. E. resolution:	18 – 25 keV
Det. Pos. resolution:	150 μm
Dead time:	3 – 25 μs

Rotating targets
 $\sim 400\text{-}600 \mu\text{g}/\text{cm}^2$

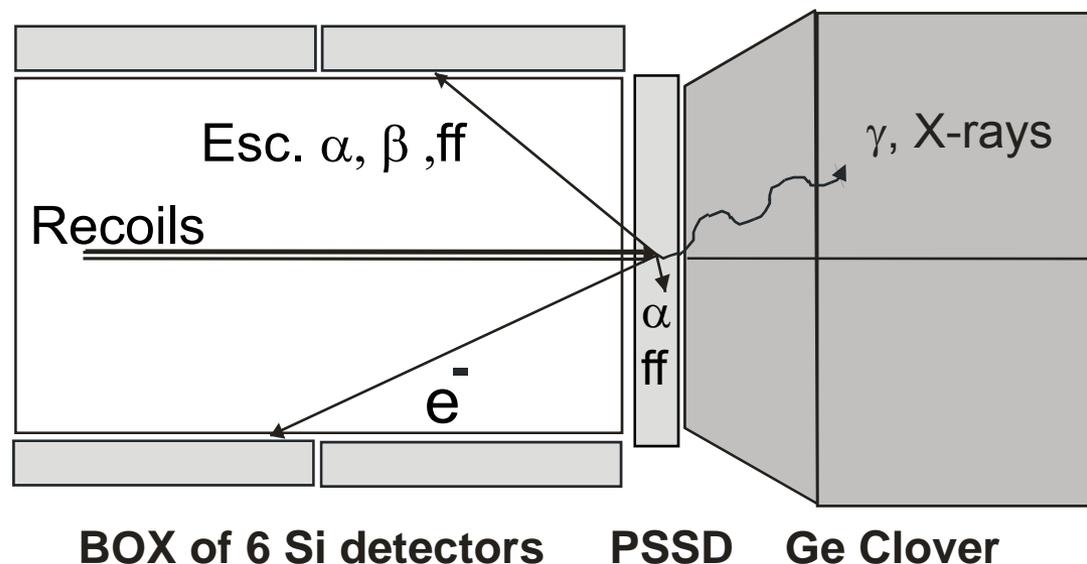
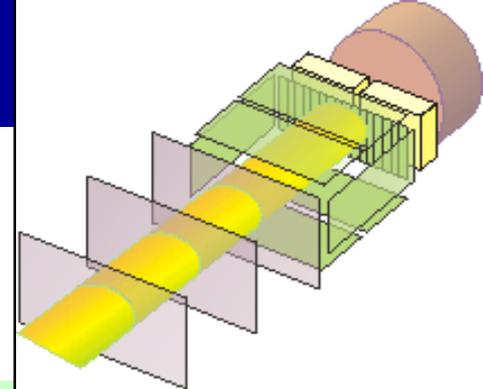


1 μA of $^{52}\text{Cr}, ^{58}\text{Ni}$
 $\sim 6 \times 10^{12}$ pps

SHIP Detection System

Measure **efficiently** all possible decays:

- particle decay (α , β , protons, fission) $E=0.1-250$ MeV
- gamma decay $E=10-4000$ keV
- internal conversion electrons $E=50-500$ keV



•3 Time-Of-Flight detectors

• **STOP detector** – 16 position sensitive Si strips (35×80 mm), pos. resolution FWHM=150 μ m, energy resolution 14 keV

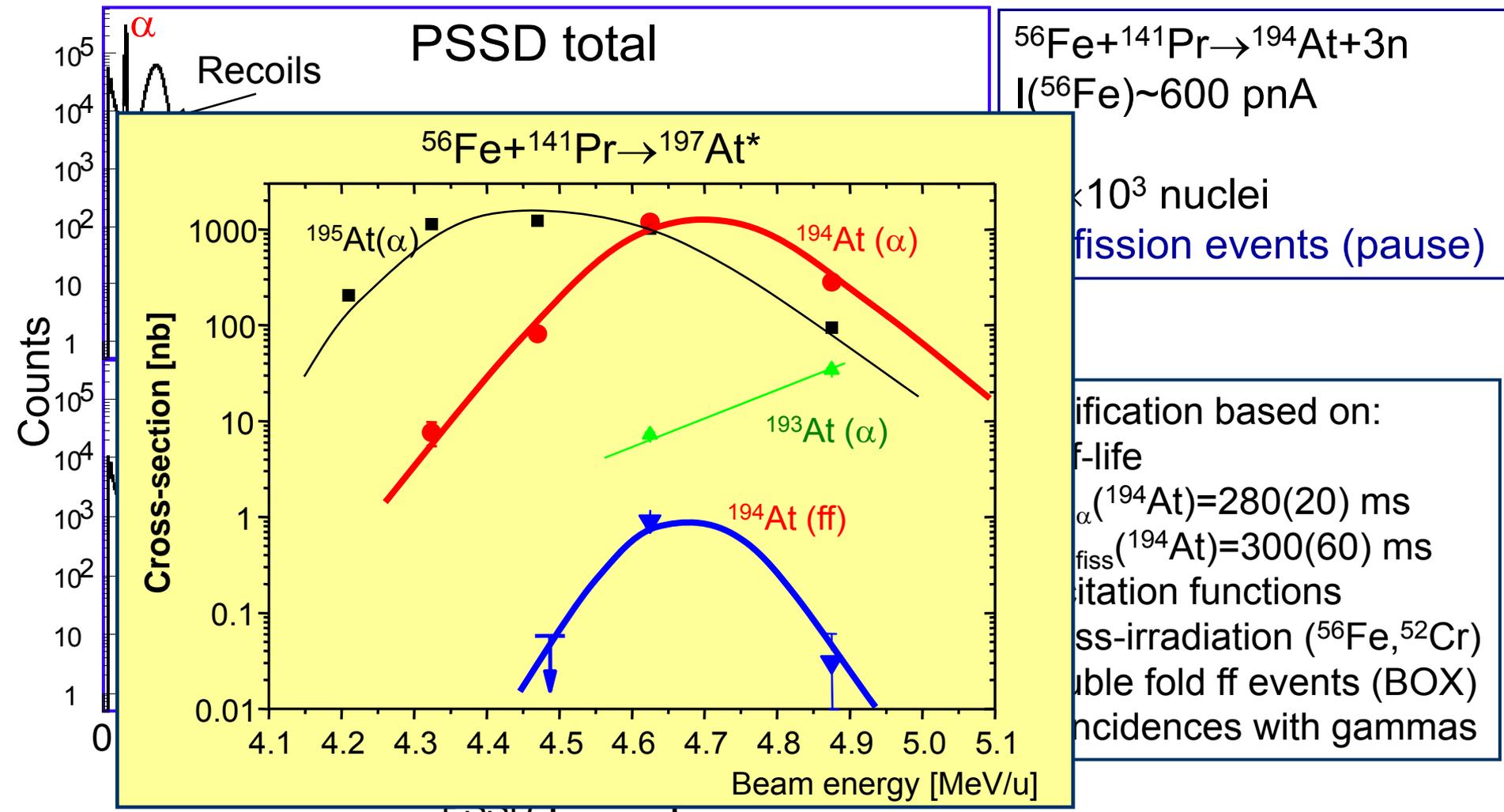
• **6 BOX Si detectors** – for β and escaping α particles with a solid angle 80% of 2π

• **GAMMA detectors** – large-volume Clover detector for x rays or γ rays in coincidence with α 's

• **VETO detector** – reduces background

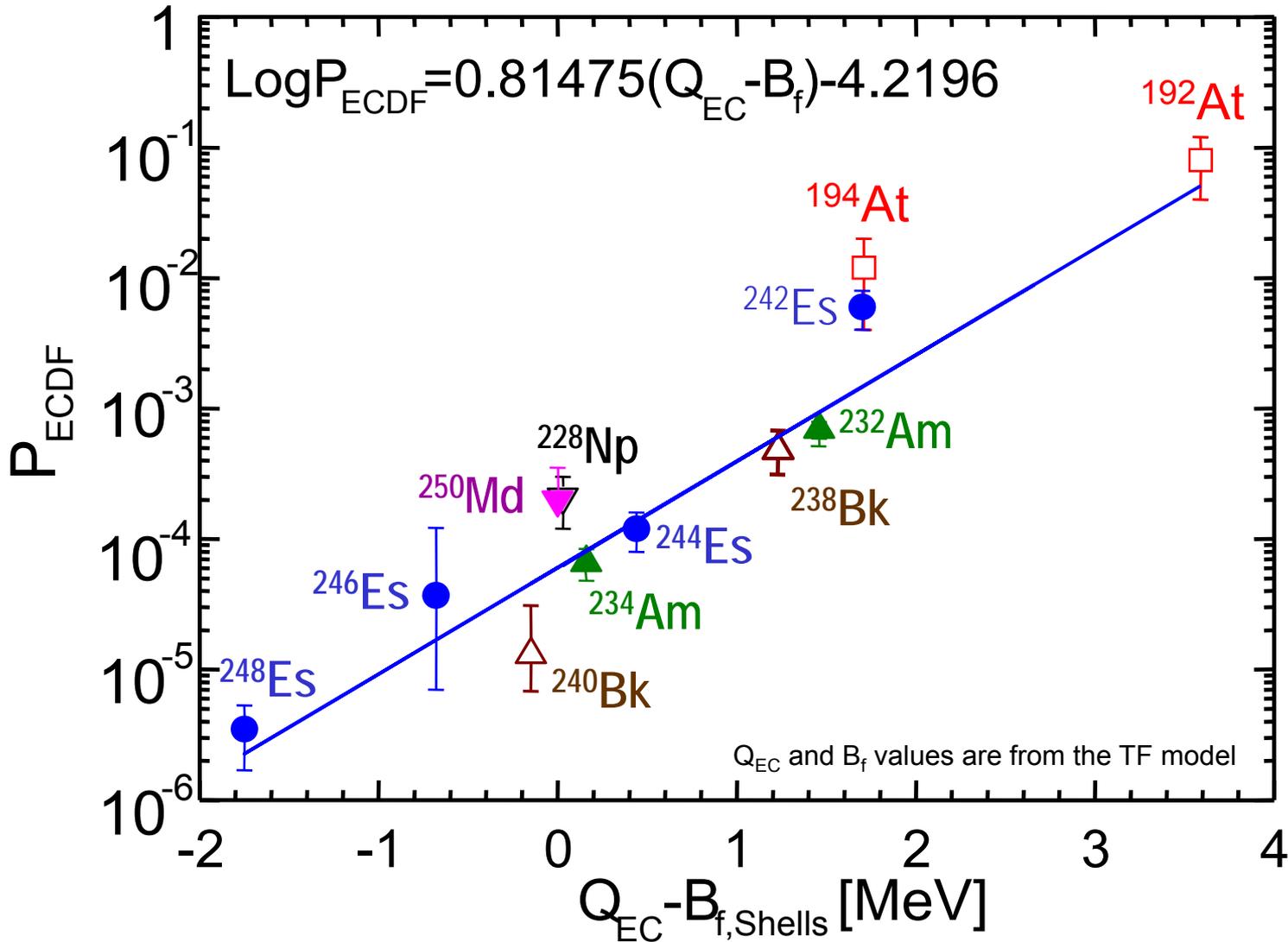
Unambiguous identification of ECDF in ^{194}At , SHIP(GSI)

A.Andreyev et al, paper in preparation



First observed in the $^{52}\text{Cr} + ^{144}\text{Sm} \rightarrow ^{194}\text{At} + pn$ reaction (SHIP, 2006)
 16 fissions in pause

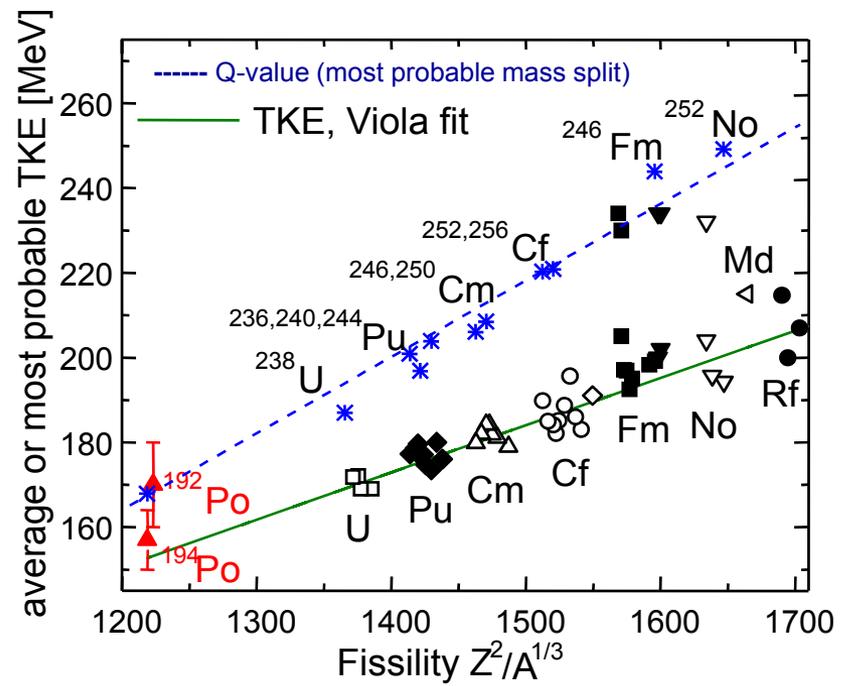
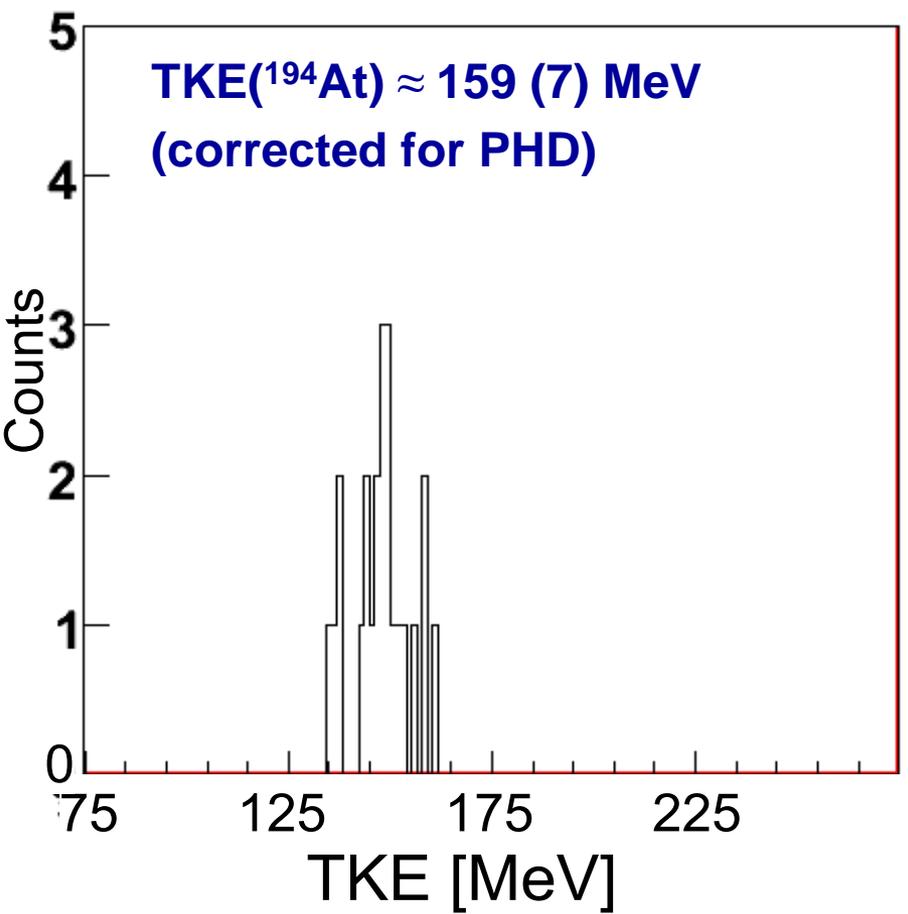
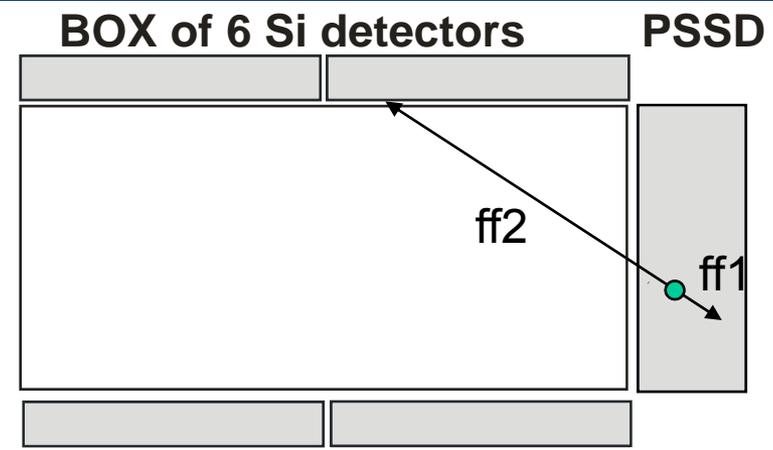
P_{ECDF} for $^{192,194}\text{At}$



Largest P_{ECDF} values ever obtained!

Total Kinetic Energy in the EC-delayed fission of ^{194}At

TKE: Add up the energies of 2ff from the PSSD and BOX detectors



Cold fission of $^{192,194}\text{Po}$!?

FIRST FISSION STUDY AT ISOLDE!?

December 1938:

O. Hahn and F. Strassmann : discovery of nuclear fission

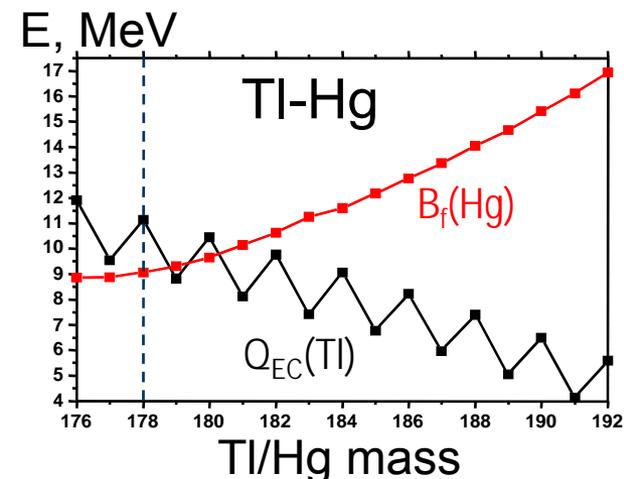
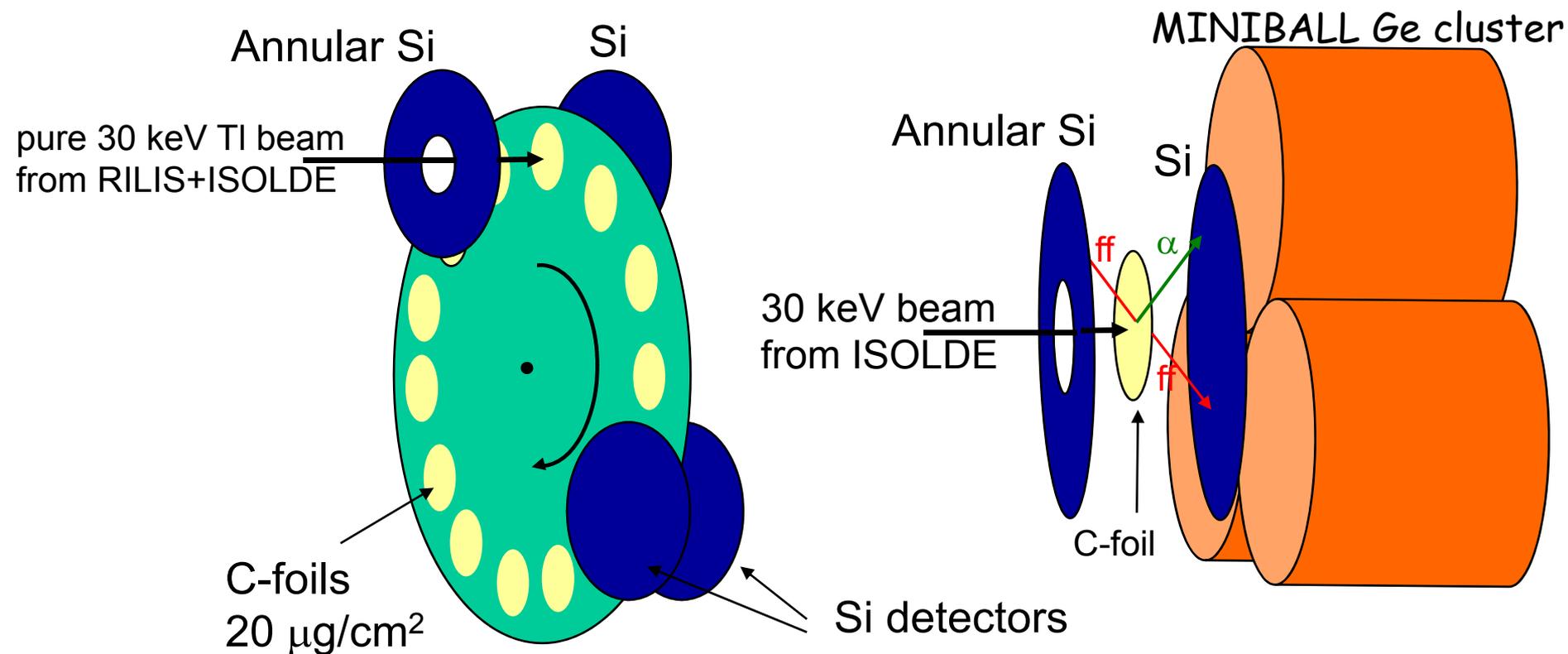
From 16th October 1967 on: ISOLDE operation

40 years later....

June 2008: **first ever fission study at ISOLDE!?** :

IS466 experiment

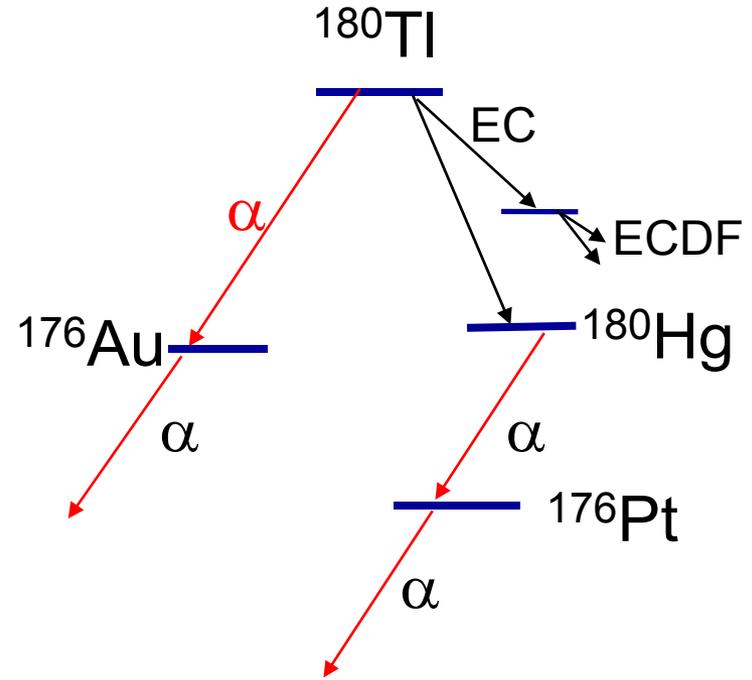
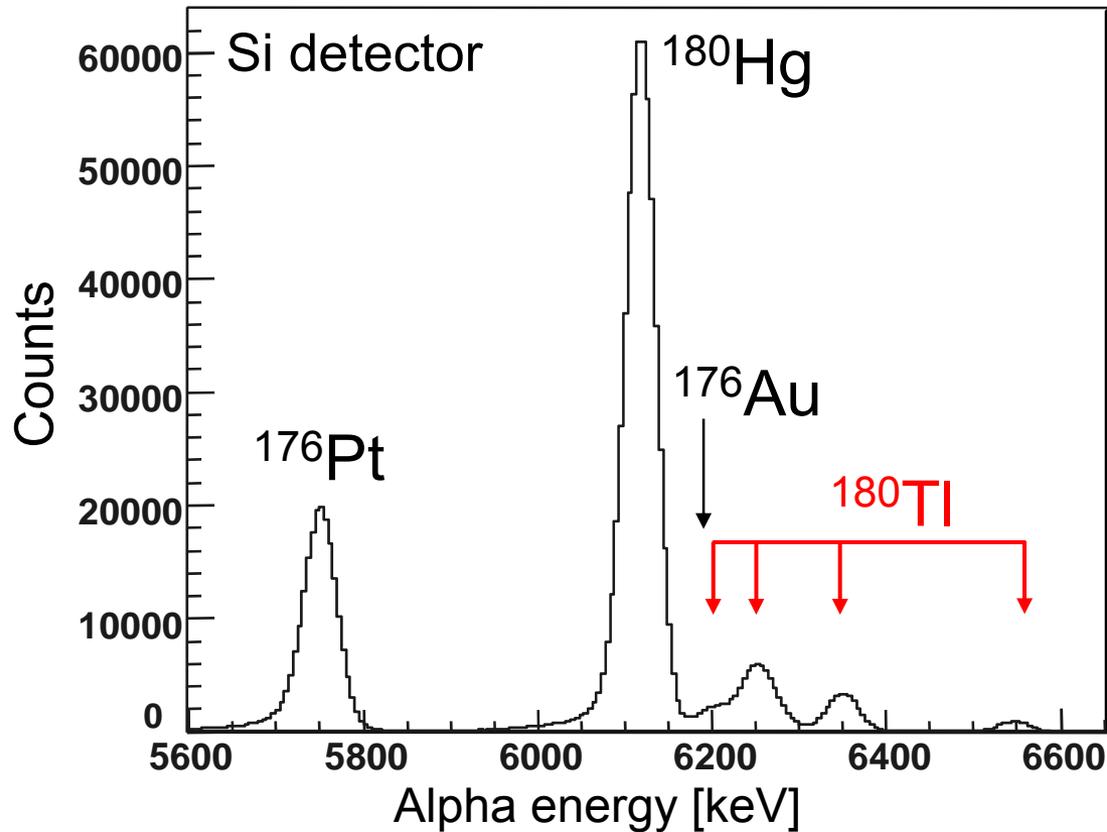
IS466: ECDF of ^{180}Tl isotope at ISOLDE (31 May–6 June 2008)



Setup: Si detectors from both sides of the C-foil

- Simple setup & DAQ: 4 PIPS (1 of them – annular)
- Large geometrical efficiency (up to 80%)
- 2 fold fission fragment coincidences
- ff-gamma coincidences
- Digital electronics (5 DGF modules)

IS466: EC and α decay of ^{180}Tl



- **Very clean spectra: only ^{180}Tl and its decay products! (first on-line use of SSL)**
- **Negligible direct ^{180}Hg production ($\rightarrow b_{\text{EC}}(^{180}\text{Tl})$)**

- $\times 100$ times statistics than in previous works
- New detailed decay scheme of ^{180}Tl
- Detailed α - γ data (~ 10 new γ lines in ^{176}Au)
- Correct low-energy level scheme of ^{176}Au

Due to negligible direct production of ^{180}Hg

$$P_{\text{ECDF}}(^{180}\text{Tl}) = \frac{N_{\text{fission}}(^{180}\text{Hg})}{N_{\alpha}(^{180}\text{Hg}) \times b_{\text{EC}}(^{180}\text{Tl})}$$

$$P_{\text{ECDF}}(^{180}\text{Tl}) = 5(1) \times 10^{-5} \quad (P_{\text{ECDF}} = 3 \times 10^{-(7 \pm 1)})$$

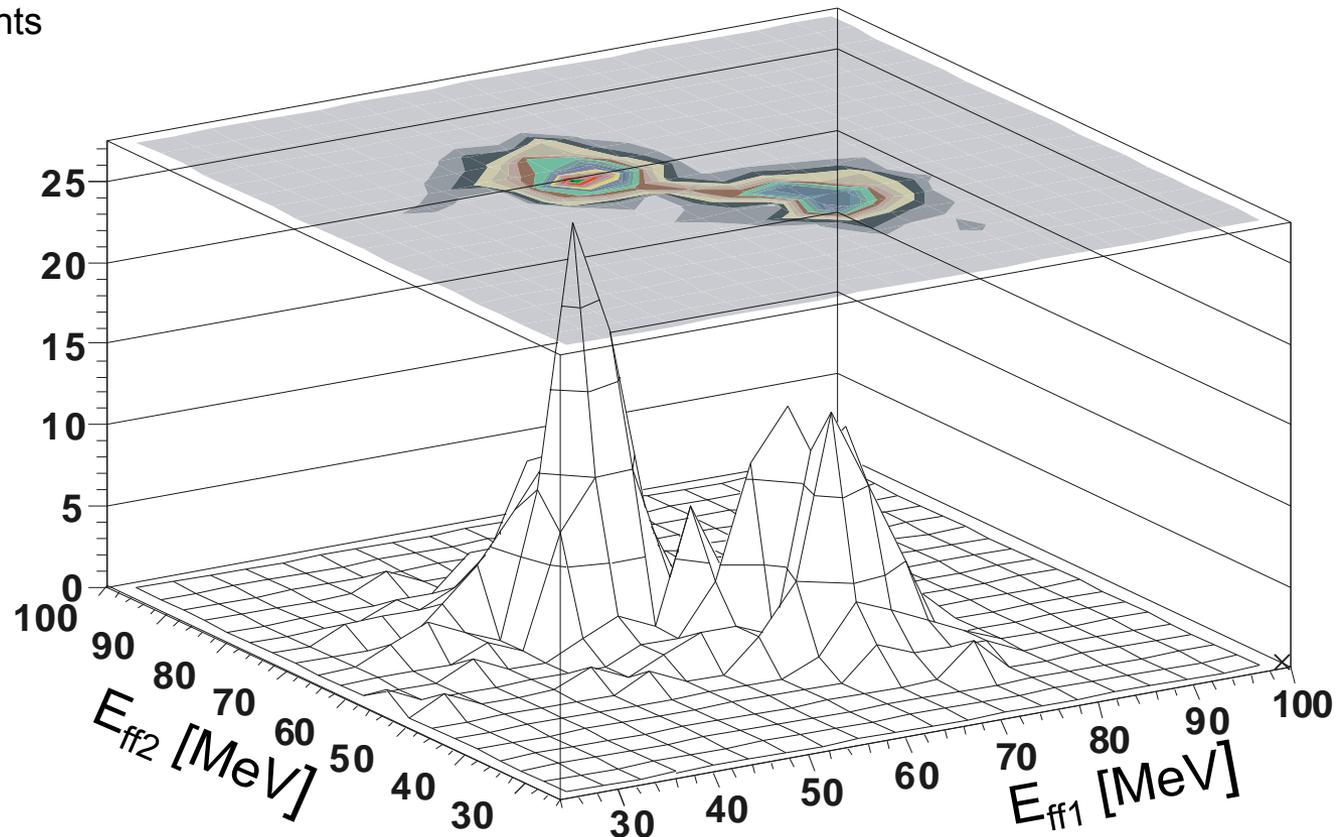
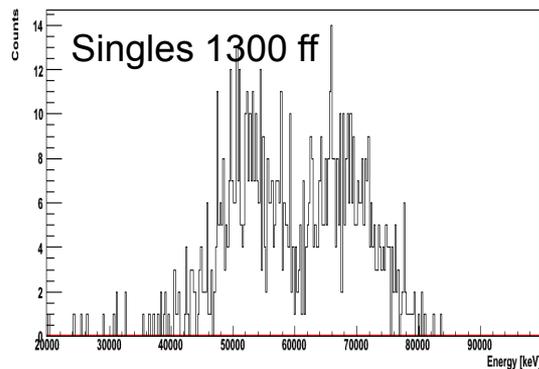
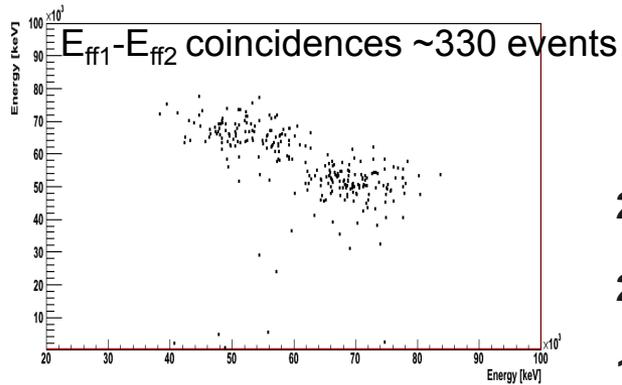
IS466: ECDF of ^{180}Tl

Before the IS466 experiment: How ^{180}Hg ($Z=80$, $N=100$, $N/Z=1.25$) fissions?

SYMMETRICAL mass split in two semi-magic ^{90}Zr ($Z=40$, $N=50$, $N/Z=1.25$)?

IS466:

NO! ASYMMETRICAL energy (thus, mass) split!



From comparison of TKE and Q-values \rightarrow Cold fission of ^{180}Hg !

Future ECDF studies in the Pb region

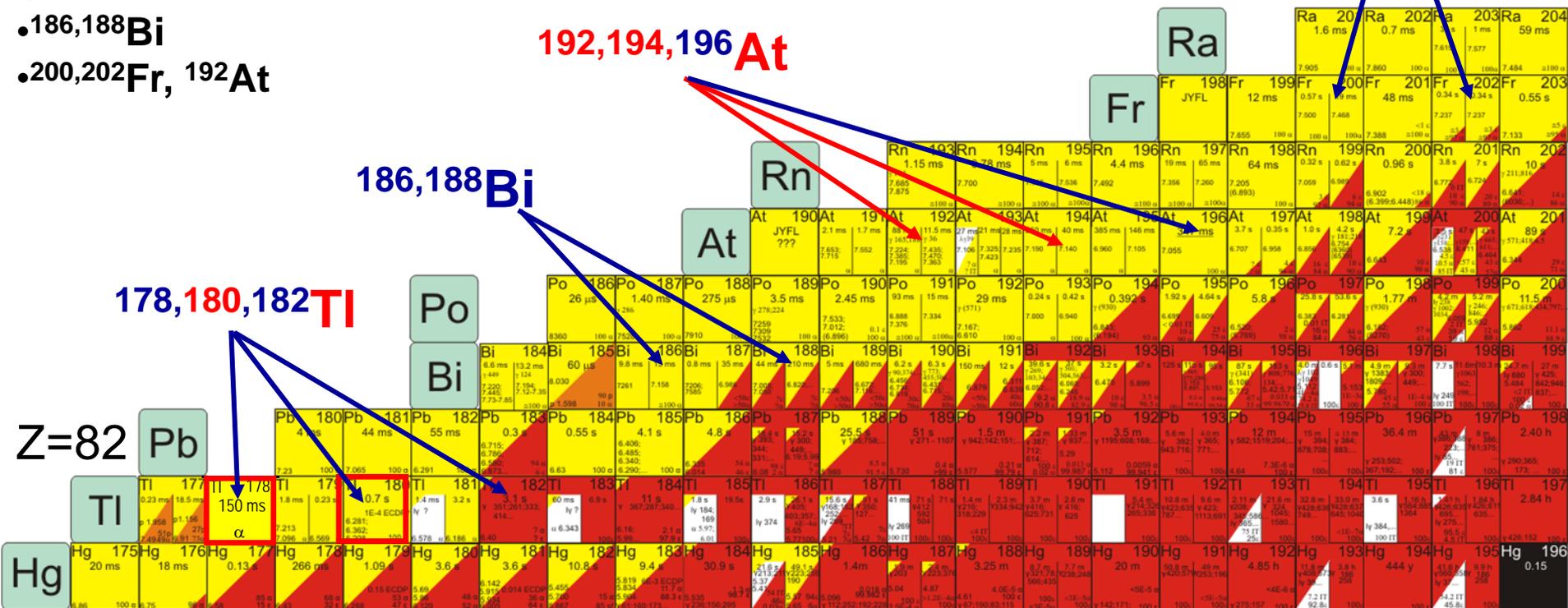
Identification of new ECDF nuclei and detailed studies (e.g. B_f , TKE...)

ISOLDE:

- ^{180}Tl – Beta strength function measurements with TAS
- ^{180}Tl – HFS scan with RILIS : search for 2 isomeric states
- ^{180}Tl – mass measurement at ISOLTRAP
- $^{178,182}\text{Tl}$ – ECDF experiments at ISOLDE

SHIP:

- $^{186,188}\text{Bi}$
- $^{200,202}\text{Fr}$, ^{192}At



Thank you!

Total Kinetic Energy in the EC-delayed fission of ^{194}At

TKE: Add up the energies of 2ff from the PSSD and BOX detectors

$^{252}\text{Cf}(\text{sf})$:

$$Q[^{252}\text{Cf} - (^{142}\text{Ba} + ^{110}\text{Mo})] = 219 \text{ MeV}$$

$$\langle \text{TKE} \rangle = 185 \text{ MeV}$$

$$\Delta(Q - \text{TKE}) = 34 \text{ MeV}$$

$^{194}\text{Po}(\text{ECDF})$:

$$Q[^{194}\text{Po} - (^{98}\text{Mo} + ^{96}\text{Mo})] = 166 \text{ MeV}$$

$$\langle \text{TKE} \rangle = 157(7) \text{ MeV}$$

$$\Delta(Q - \text{TKE}) = 9(7) \text{ MeV (+E* after EC)}$$

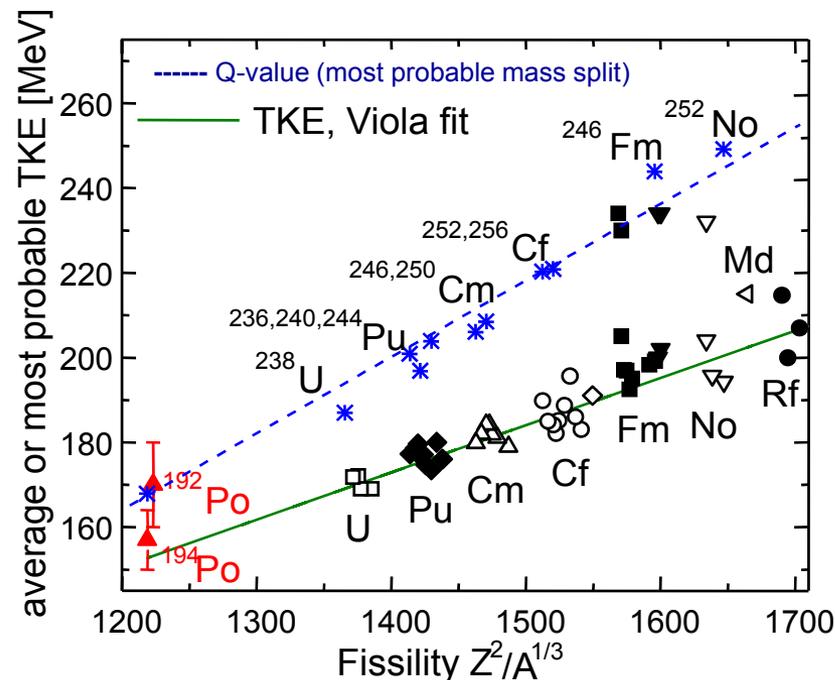
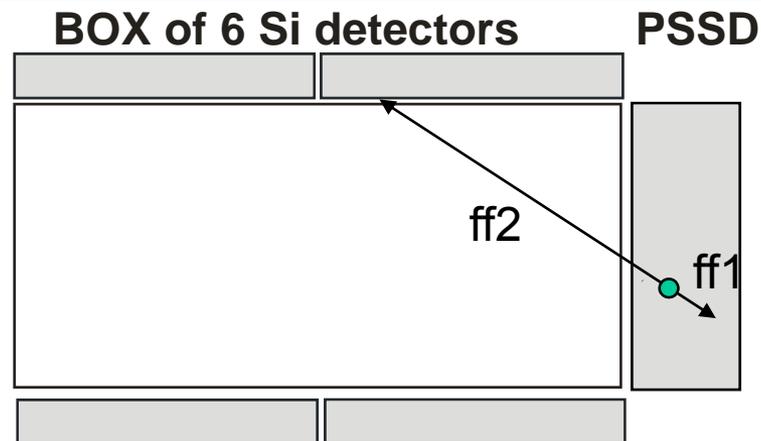
$^{192}\text{Po}(\text{ECDF})$:

$$Q[^{192}\text{Po} - (^{96}\text{Mo} + ^{96}\text{Mo})] = 165 \text{ MeV}$$

$$\langle \text{TKE} \rangle = 170(10) \text{ MeV}$$

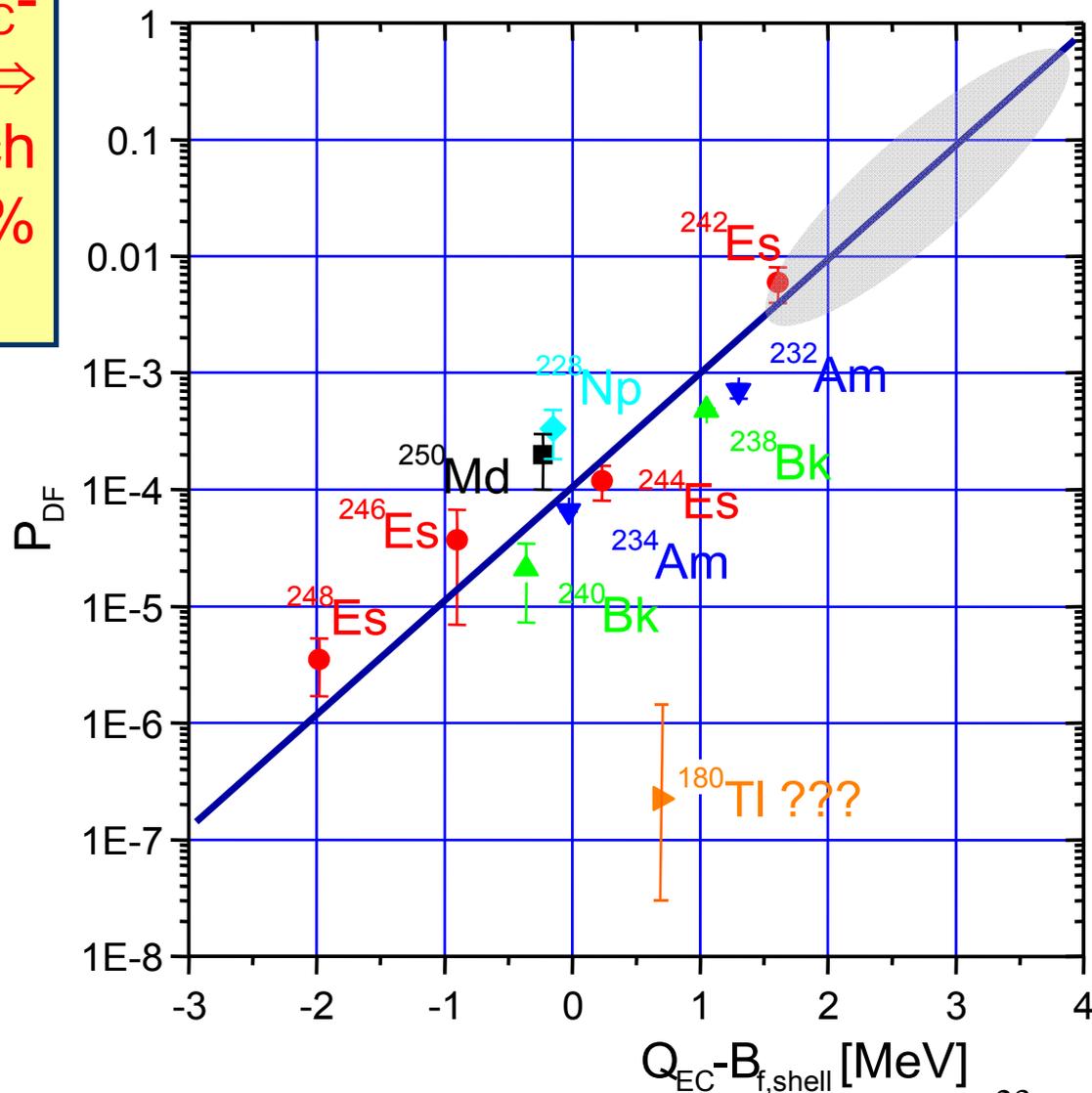
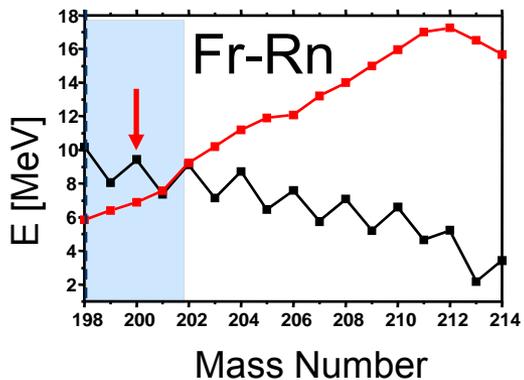
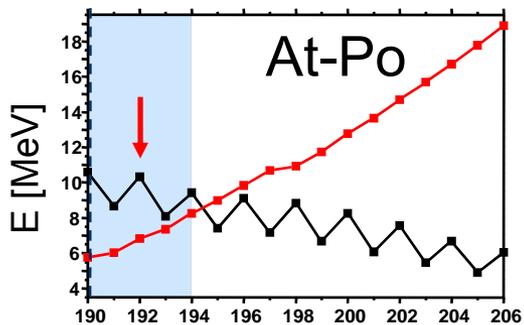
$$\Delta(Q - \text{TKE}) = -5(10) \text{ MeV (+E* after EC)}$$

Cold fission of $^{192,194}\text{Po}$? (no neutron emission during fission!)



ECDF probability in the Pb region

Large Q_{EC} and Q_{EC^-}
 $B_f = 3-4$ MeV \Rightarrow
 Possibility to reach
 nuclei with 10-100%
 ECDF probability!



Low-Energy ECDF data ($^{232,234}\text{Am}$ as examples)

H. L. Hall et al., PRC42 (1990) 1480

- Fission barriers and their isospin dependence
- Energy distributions of fission fragments, thus TKE
- Mass distributions of fission fragments
- Gamma multiplicities for fission fragments
- Neutron multiplicities for fission fragments

ALL THIS FOR VERY EXOTIC NUCLEI WHICH DO NOT FISSION SPONTANEOUSLY!

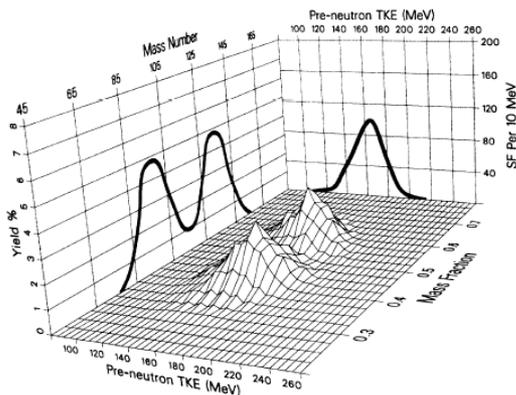


FIG. 2. Preneutron emission total-kinetic-energy (TKE) distribution of the ^{232}Am ϵDF mode and preneutron emission mass-yield distribution.

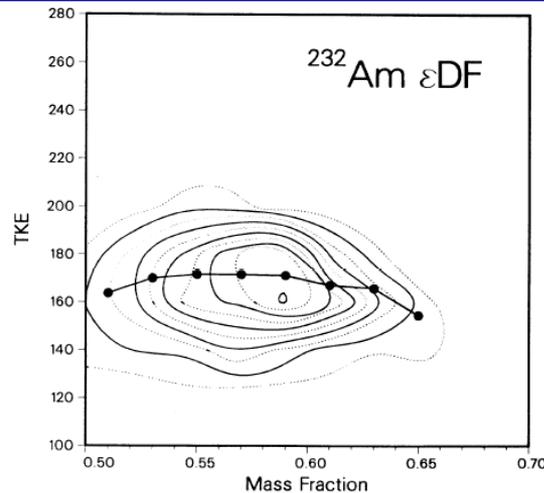
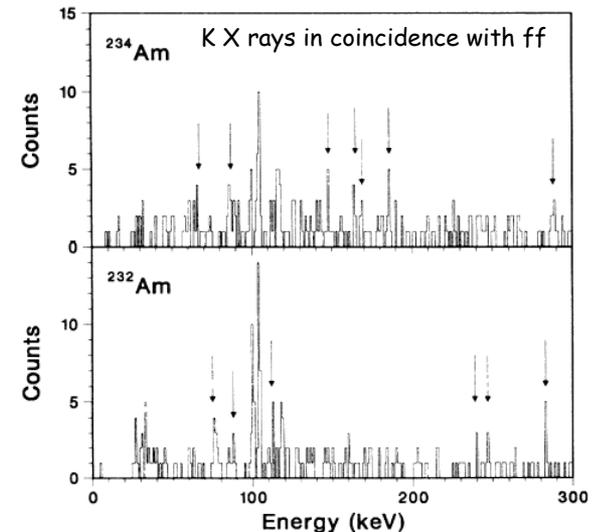
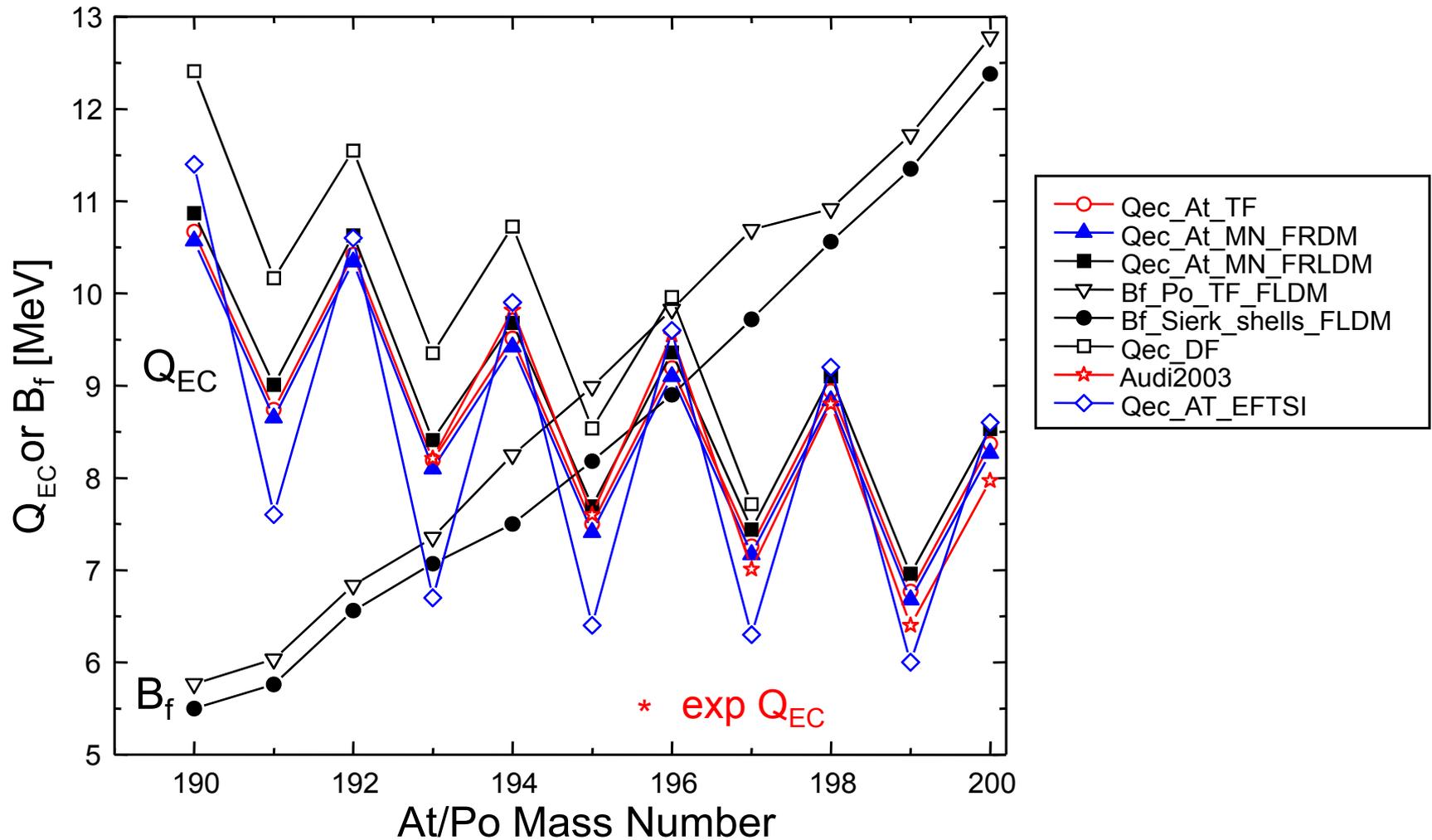


FIG. 4. Total kinetic energy and average total kinetic energy of ^{232}Am as a function of mass fraction. The solid points are average TKE values at each mass fraction. The contours represent the mass yield (normalized to 200%) as a function of TKE and mass fraction. The transition between a solid and dashed contour represents a change of 0.5% in the mass yield.

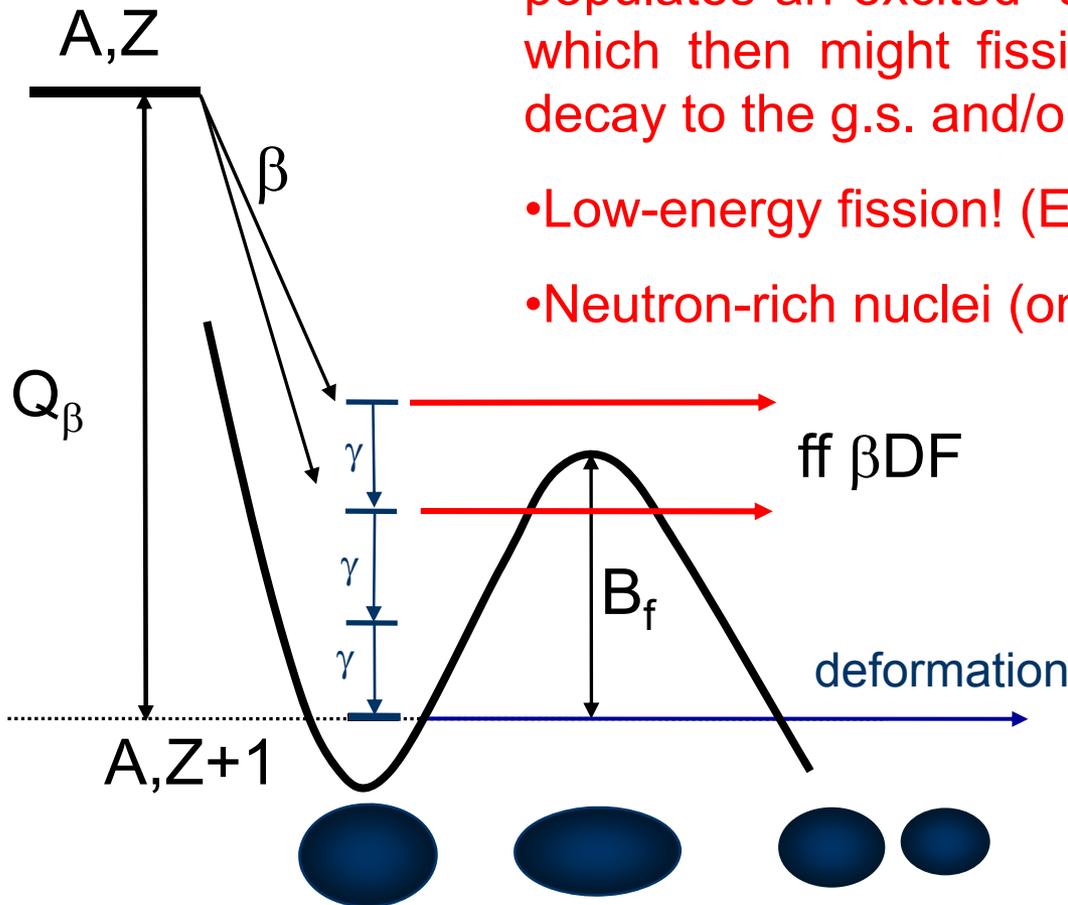


$Q_{EC}(At)$ and $B_f(Po)$ in different models



β -Delayed Fission (β DF, $T_{1/2}(ff)=T_{1/2}(\beta)$)

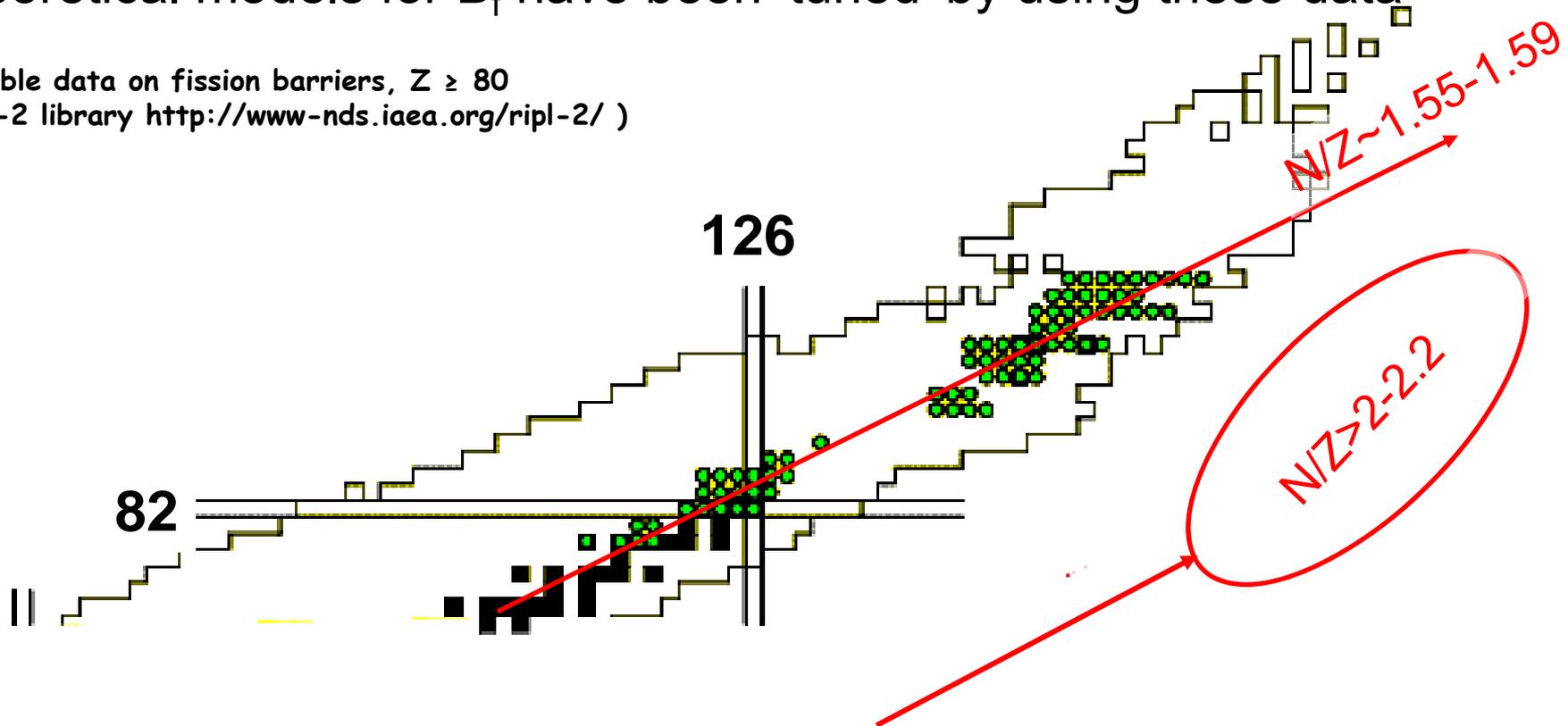
- 2 step process: β decay of a parent (A,Z) nucleus populates an excited state in the (A,Z+1) daughter, which then might fission (in competition with the γ decay to the g.s. and/or neutron emission)
- Low-energy fission! ($E^* \sim 5-10$ MeV)
- Neutron-rich nuclei (only 5 candidates known so far)



Example: Fission barriers - what do we know about them?

- Experimentally fission barriers B_f are known **only in the vicinity of the beta stability line** (e.g. $N/Z(^{238}\text{U})=1.59$)
- Theoretical models for B_f have been 'tuned' by using these data

Available data on fission barriers, $Z \geq 80$
(RIPL-2 library <http://www-nds.iaea.org/ripl-2/>)



- For the r-process calculations we need fission data far away from stability: e.g. ^{260}Po or ^{270}U ($N/Z > 2!$) – **they might not be accessible in the Lab at all! – Use calculations?**

Fission Barrier Calculations for the r-process nuclei

Full symbols – experimental data

- Unfortunately, so exotic nuclei are not presently accessible by available techniques!

- That is why the underlying mechanisms and properties of beta-delayed fission (and of low-energy fission in general) have to be investigated by using alternative approaches and in other regions of the Nuclear Chart.

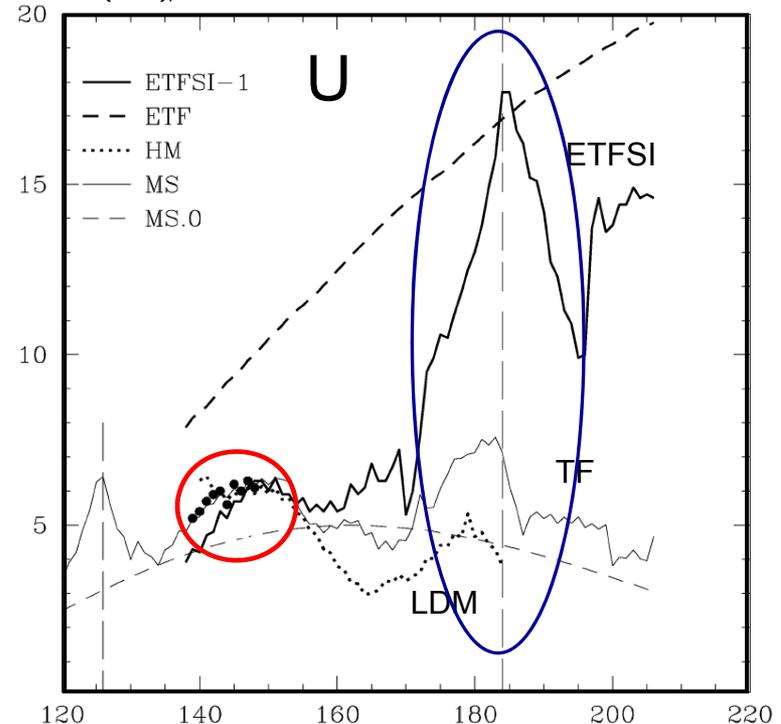
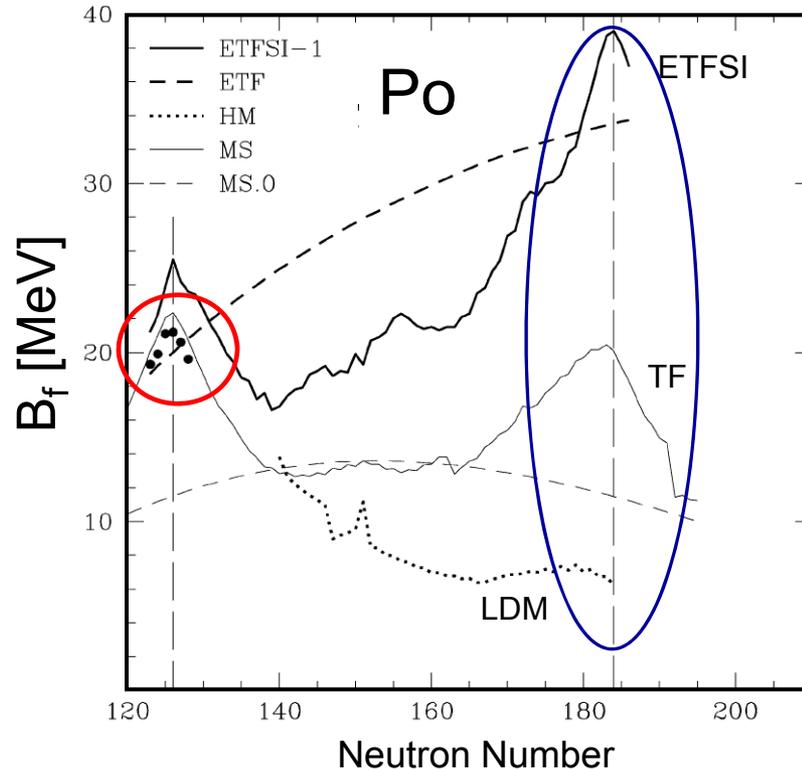
- According to semi-empirical estimates, the neutron-deficient nuclei in the U and Pb regions provide such a possibility via the ECDF decay

- Need measured fission data far of stability to tune fission models

Fission Barrier Calculations for the r-process nuclei

Full symbols – experimental data
Lines – calculations (LDM,TF, ETFSI)

A. Mamdouh et al. NPA679 (2001), 337



- Good agreement between $B_{f,cal}$ and $B_{f,exp}$ for nuclei close to stability
- Large disagreement far of stability (both on n-def. and n-rich sides)
- Need **measured** fission data far of stability to 'tune' fission models

(Speculations?) Double-Magic Fission?

One of the goals of ISOLDE proposal is ECDF of ^{178}Tl

How its daughter ^{178}Hg ($N/Z=1.225$) would fission?

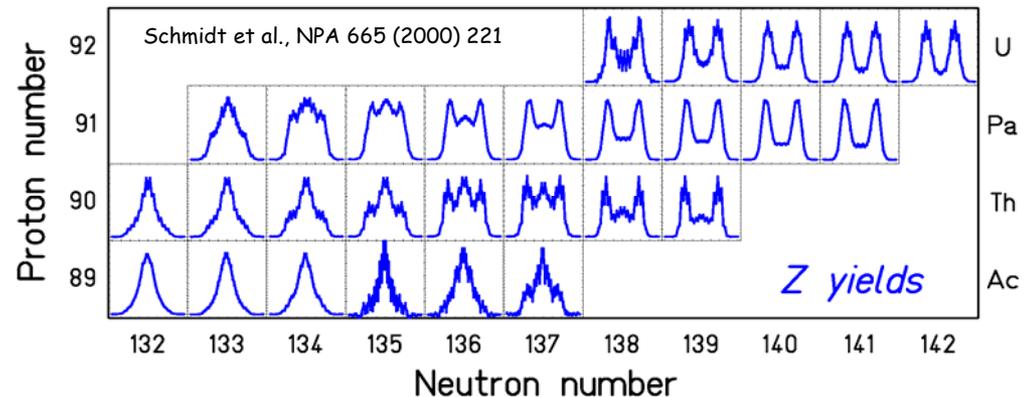
Would it give 'double-magic' fission – two double-magic ff's?

very neutron-rich ^{78}Ni ($Z=28, N=50$ $N/Z=1.79$)

very neutron-deficient ^{100}Sn ($Z=50, N=50$ $N/Z=1$)

P. Moller (private communication): **Most probably – NOT as a main channel**

- Most probable mass splits $\sim 90/\sim 90$ ($^{90}\text{Zr}/^{90}\text{Zr}$ $N/Z=1.25$)
- **but what about very asymmetrical split at the wings? :**



ECDF Probability: Fission/gamma competition

$$P_{\text{ECDF}} = \frac{N_{\text{ECDF}}}{N_{\text{EC}}} \sim \frac{\int_0^{Q_{\text{EC}}} (Q_{\text{EC}} - E)^2 \times S_{\beta}(E) \times \frac{\Gamma_f(E, B_f)}{\Gamma_{\text{tot}}(E)} dE}{\int_0^{Q_{\text{EC}}} (Q_{\text{EC}} - E)^2 \times S_{\beta}(E) dE}$$

$$\frac{\Gamma_f}{\Gamma_{\text{tot}}} = \frac{\Gamma_f}{\Gamma_f + \Gamma_{\gamma}} \quad \text{-ratio of the fission and total widths of excited levels in daughter}$$

(Γ_n is not important for neutron-deficient nuclei)

$$\Gamma_{\gamma} = \frac{9.7 \times 10^{-7} \times T^4 \times \exp(E/T)}{2\pi\rho}, \quad \rho - \text{level density, } T - \text{temperature}$$

$$\Gamma_f = \frac{1}{2\pi\rho} \left\{ 1 + \exp\left[\frac{2\pi(B_f - E)}{h\omega_f} \right] \right\}^{-1} \quad \text{-inverted parabola approximation}$$

D.L. Hill and J.A. Wheeler

Measurement of P_{ECDF} allows to deduce Fission Barrier B_f

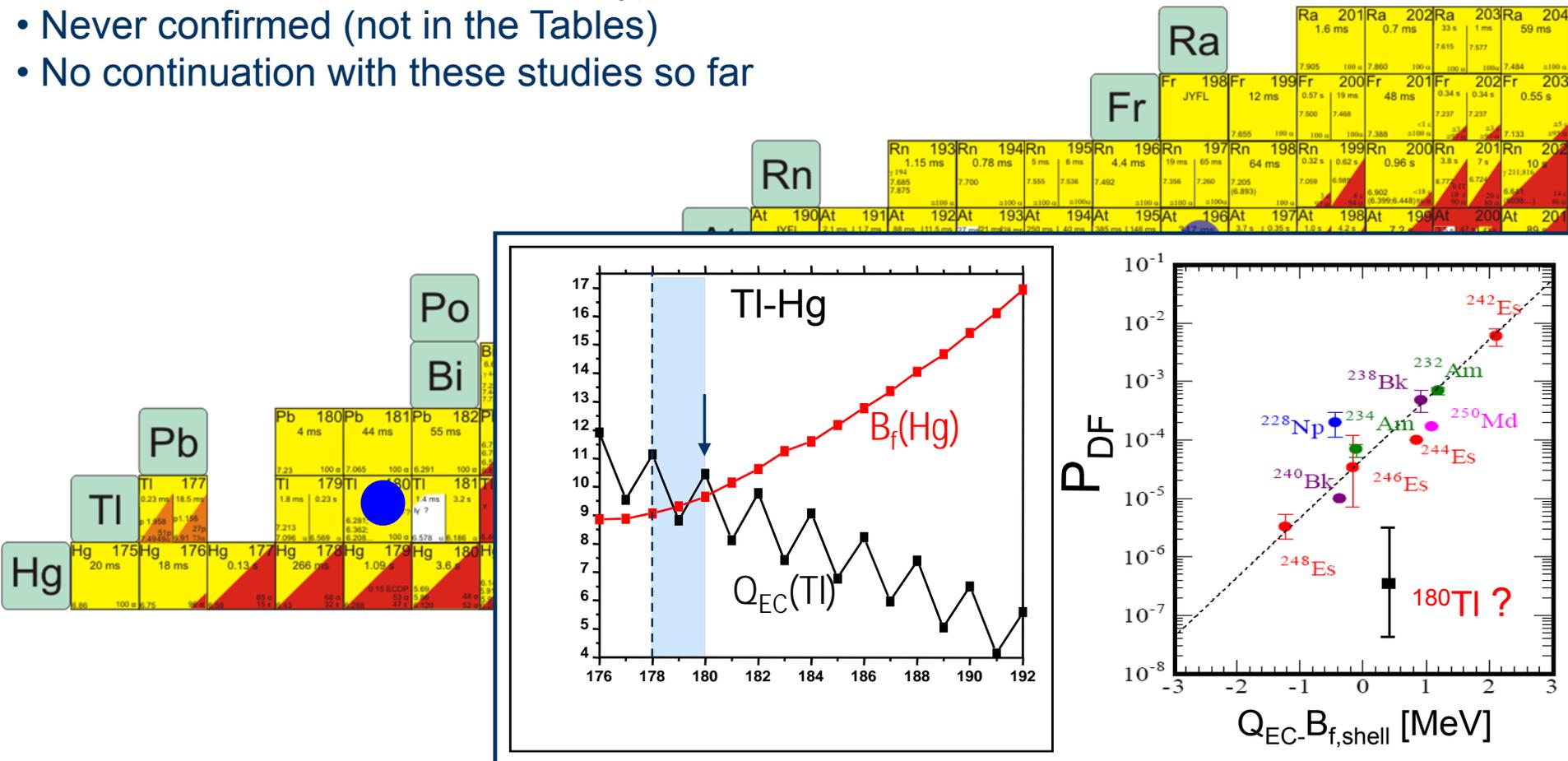
e.g. H.V. Klapdor et al., Z.Phys.A292, 1979,249; D. Habs et. al. Z.Phys. A285 (1978), 53

Previous Studies in the Pb region (Dubna)

Yu. A. Lazarev et al. Europhys. Lett. 4 (1987) 893; and Inst. Phys. Conf. Ser. No132 (1992) 739

“Most probable” candidates: ^{180}Tl ($P_{\text{ECDF}}=3 \times 10^{-(7 \pm 1)}$), ^{188}Bi , ^{196}At (no P_{ECDF} data)

- Irradiations inside the cyclotron (no A,Z selection for products)
- Rotating wheel system, thick effective targets (2 mg/cm²)
- Cross-irradiations, apparent $\sigma_{\text{fis}} \sim 15\text{-}50$ pb
- Mica detectors (fission tracks only)
- Never confirmed (not in the Tables)
- No continuation with these studies so far



An example: Fission Barrier of ^{232}Pu

D. Habs et. al. Z.Phys. A285 (1978), 53

$$P_{\text{ECDF}} \sim \frac{\int_0^{Q_{\text{EC}}} (Q_{\text{EC}} - E)^2 \times S_{\beta}(E) \frac{\Gamma_f(E)}{\Gamma_{\text{tot}}(E)} dE}{\int_0^{Q_{\text{EC}}} (Q_{\text{EC}} - E)^2 \times S_{\beta}(E) dE}$$

$$\Gamma_f = \frac{1}{2\pi\rho} \left\{ 1 + \exp\left[\frac{2\pi(B_f - E)}{h\omega_f} \right] \right\}^{-1}$$

Measured $P_{\text{ECDF}}(^{232}\text{Am}) = (1.3^{+4}_{-0.8}) \times 10^{-2} \Rightarrow B_{\text{fis}}(^{232}\text{Pu}) = 5.3(4) \text{ MeV}$

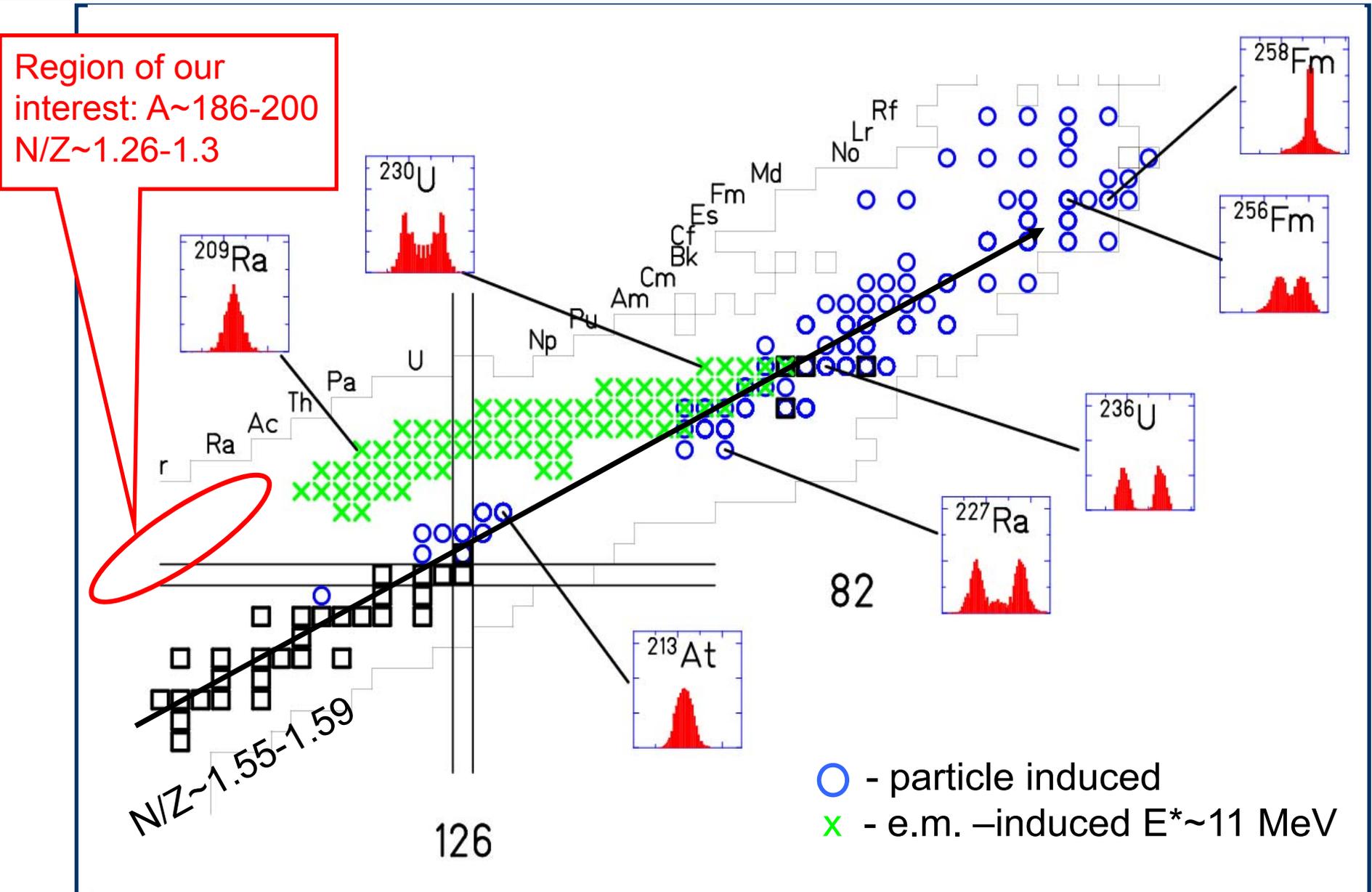
Assuming uncertainty of: $Q_{\text{EC}} = \pm 200 \text{ keV}$

$h\omega_f = \pm 100 \text{ keV}$

a factor of 3 in P_{ECDF}

B_f precision of $\sim 7.5\%$ - well comparable to direct methods!

Experimental information on fission - Low energy



Electromagnetically-Induced Fission In-flight (FRS, GSI)

A. Grewe et al. NPA614 (1997), 400

Fission from $E^* \sim 12$ MeV (E1 GDR)

For comparison:

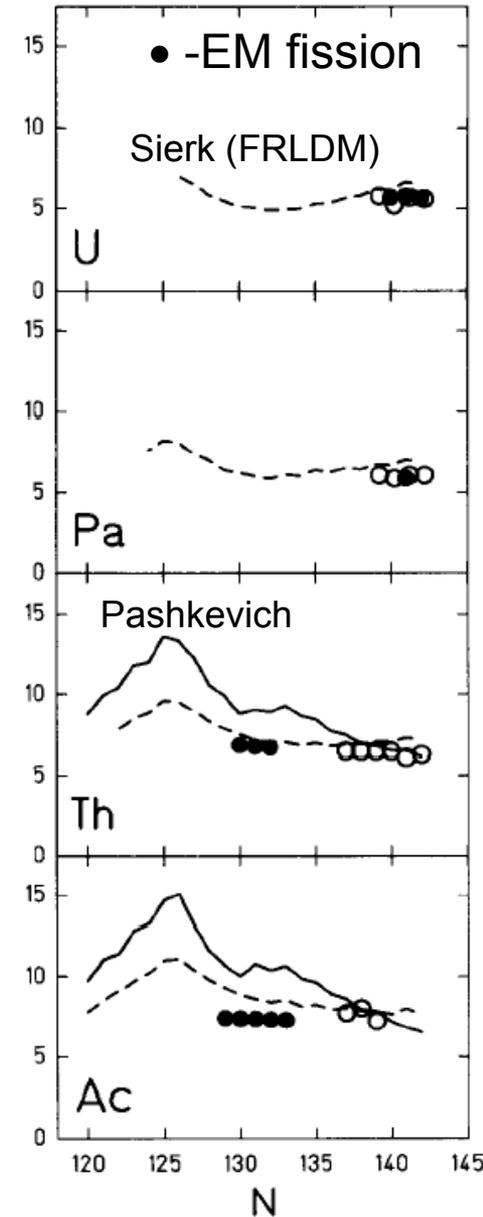
$$B_f(^{234}\text{U}) \sim 6 \text{ MeV}$$

$$B_f(^{220}\text{Th}) \sim 7.5 \text{ MeV}$$

$$B_f(^{218}\text{Ac}) \sim 7.5 \text{ MeV}$$

Thus, still fission from quite above the barrier!

In contrast, β df is near (or sub)-barrier effect!



- Primary beam at FRS
- 1 g/cm² Cu primary target
- Separated RIBs from FRS
- Pb secondary target
- $\sigma(\text{El.fission}) \sim 2.1 \text{ b } (^{234}\text{U})$

