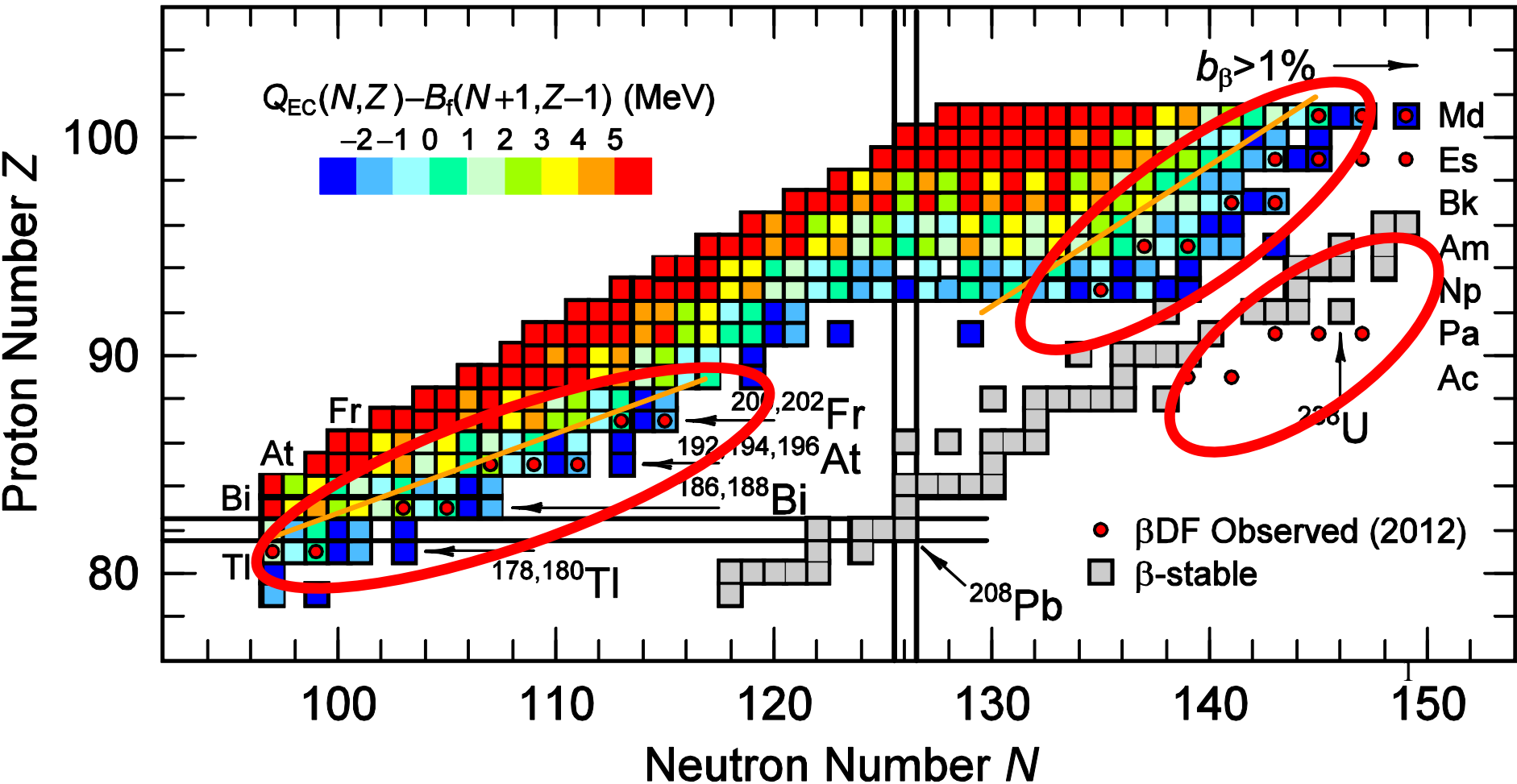


# Beta-delayed fission as a tool to study near and sub-barrier fission in extremely exotic nuclei

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 Japan Atomic Energy Agency (JAEA, Tokai, Japan)



# Collaboration

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# Outlook

- Brief (experimental) review on low-energy fission
- Low-energy fission in "new" regions of the Nuclear Chart
- Beta Delayed Fission ( $\beta$ DF) - what it is and why?
- $\beta$ DF  $^{194,196}\text{At}$ ,  $^{202}\text{Fr}$  at ISOLDE (CERN)
- $\beta$ DF  $^{194}\text{At}$  at SHIP (GSI)
- Further plans

# Outlook

- Many nuclear properties change far from stability line (e.g. disappearance of traditional magic numbers; appearance of new shell gaps; halos, skins...

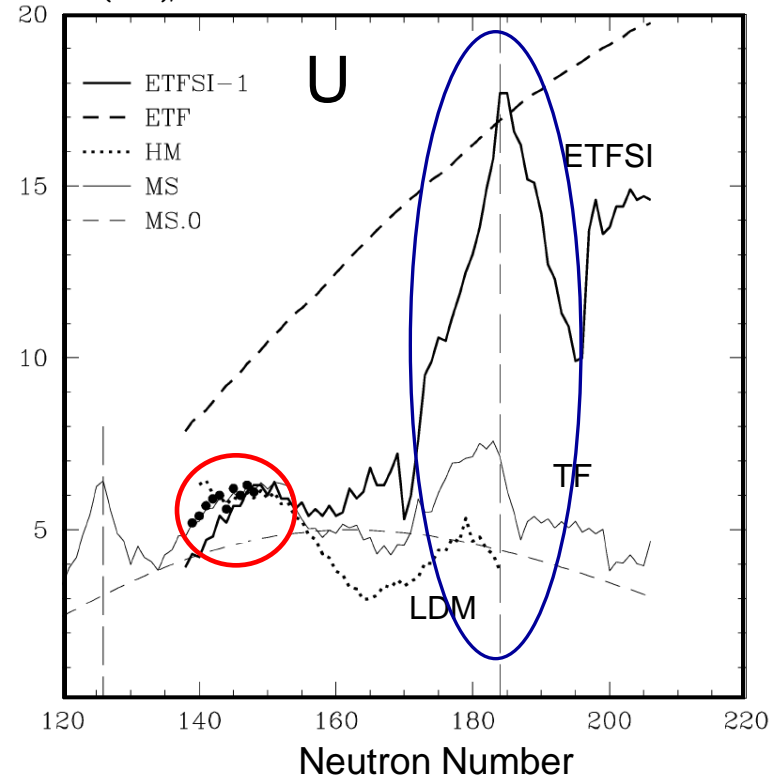
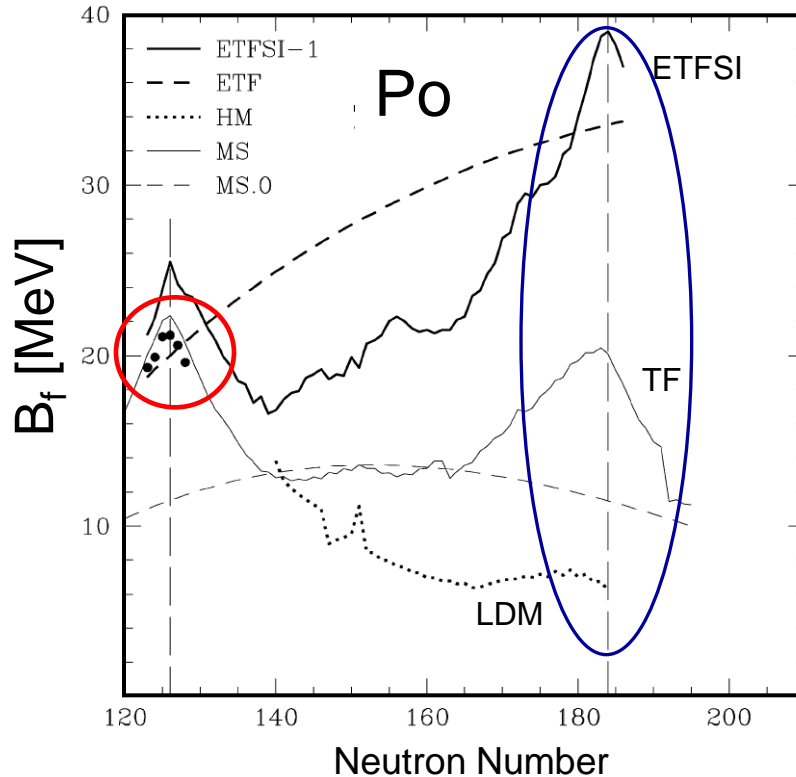
- What happens to fission far from stability, e.g. on the extremely proton-rich or neutron-rich side (relevant for r-process)?

- Not simple to answer, as to fission these nuclei at low excitation energy ( $E^* \sim B_f$ ) is a very challenging task as none of them fissions from g.s.

# 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 (especially on the n-rich sides)
- Need **measured** fission data far of stability to 'tune' fission models

A Detour:

What can one learn with a rate of 1 fission/h?

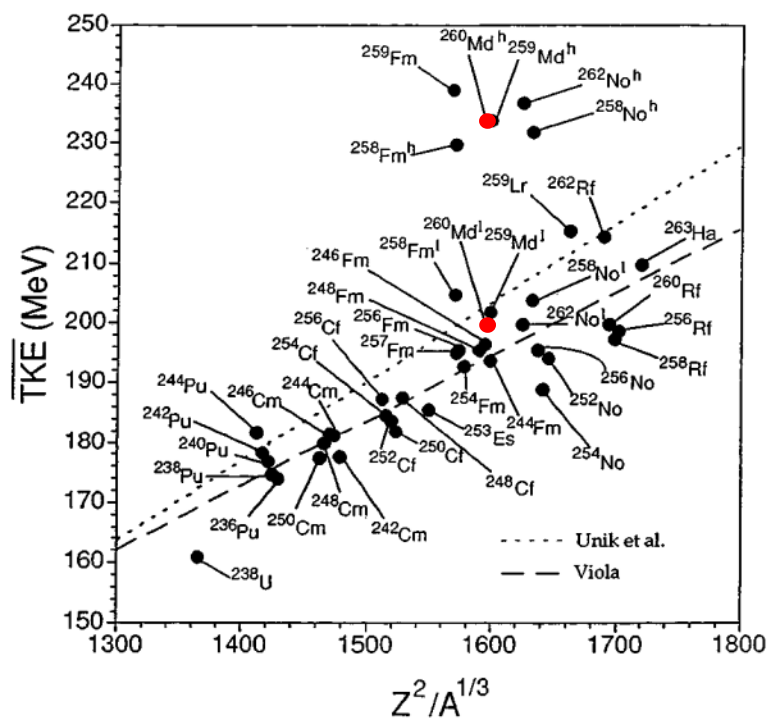
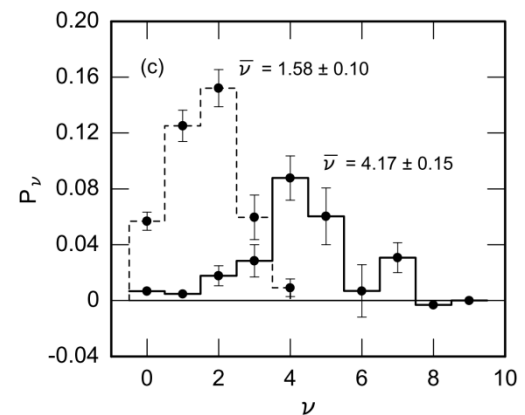
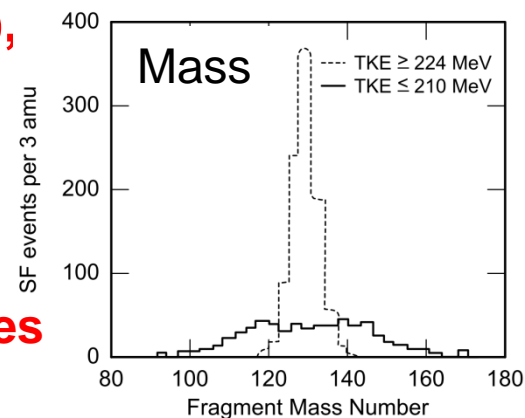
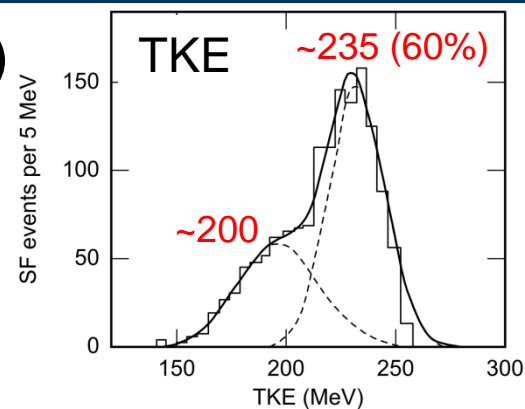
OR

What can one learn from ~100-1000 fission events?

# Bimodal Fission of $^{260}\text{Md}$ ( $T_{1/2} \sim 32$ d)

J. F. Wild et al., Phys. Rev. C4, 640, 1990

- $^{22}\text{Ne} + ^{254}\text{Es}$  ( $T_{1/2} = 276$  d)  $\rightarrow$   $^{260}\text{Md}$  ( $\alpha$  transfer reaction)
- 34 irradiations in a **2.5 months irradiation period**
- Collection of recoils on a foil
- Radiochemical separation of  **$\sim 3000$   $^{260}\text{Md}$  atoms**
- Deposited on a very thin foil
- **98 days of counting with Si and neutron detectors**
- **1207 singles fission fragments, 905 coincident (TKE),**
- **Neutron multiplicity (as a function of TKE)**

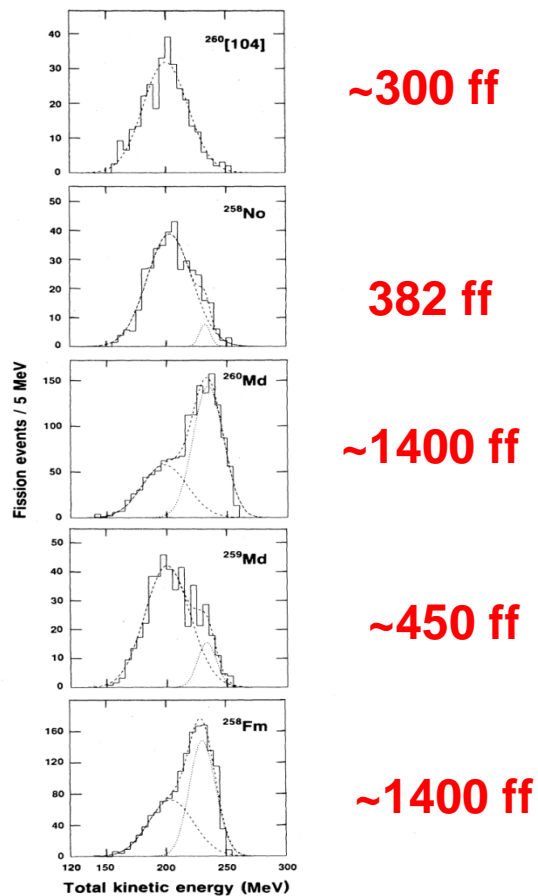


- **Two-humped TKE**
- **Two components in masses**
- **High TKE – low  $\nu$**
- **Low TKE – high  $\nu$**

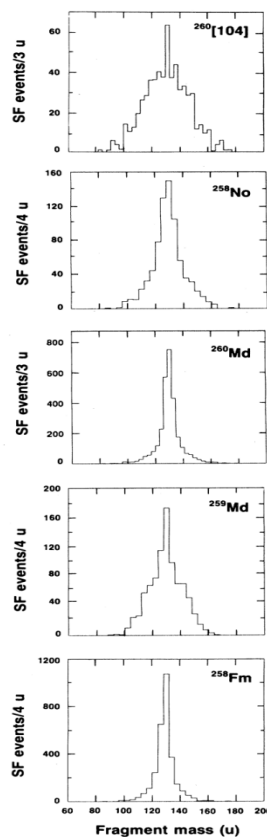
# Bimodal Fission

E. K. Hulet et, Phys. Rev. C40, 770 (1989)

## Total Kinetic Energy (TKE)



## Masses



## Mass components as a function of TKE

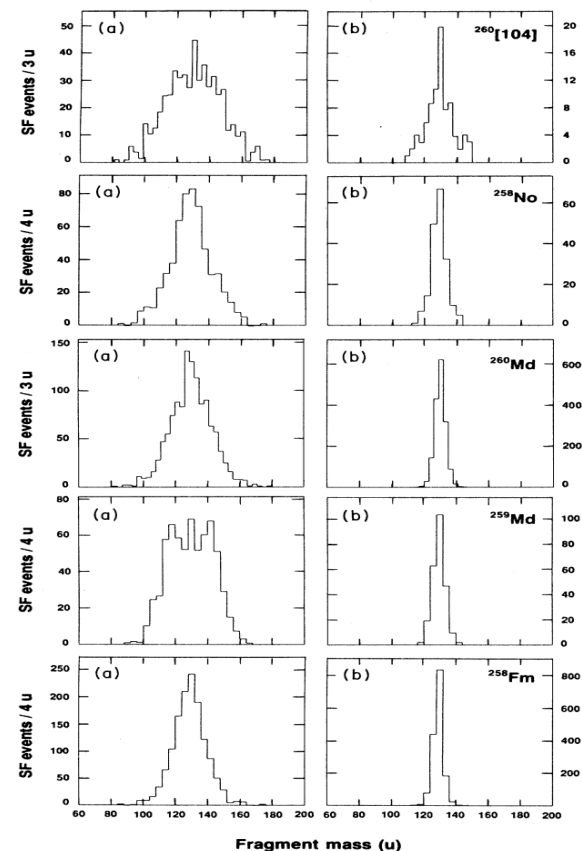


FIG. 7. Unfolding of the asymmetric TKE distributions of

FIG. 5. Provisional mass distributions (no neutron corrections) obtained from correlated fragment energies. The mass bins have been chosen to be slightly different for each nuclide.

FIG. 8. Mass distributions obtained by sorting fission events according to their total kinetic energies: (a) for events with

**Evidence for bimodal fission: strong deviation of TKE from a single Gaussian**  
**Detailed fission fragments energy measurements are "A MUST"**

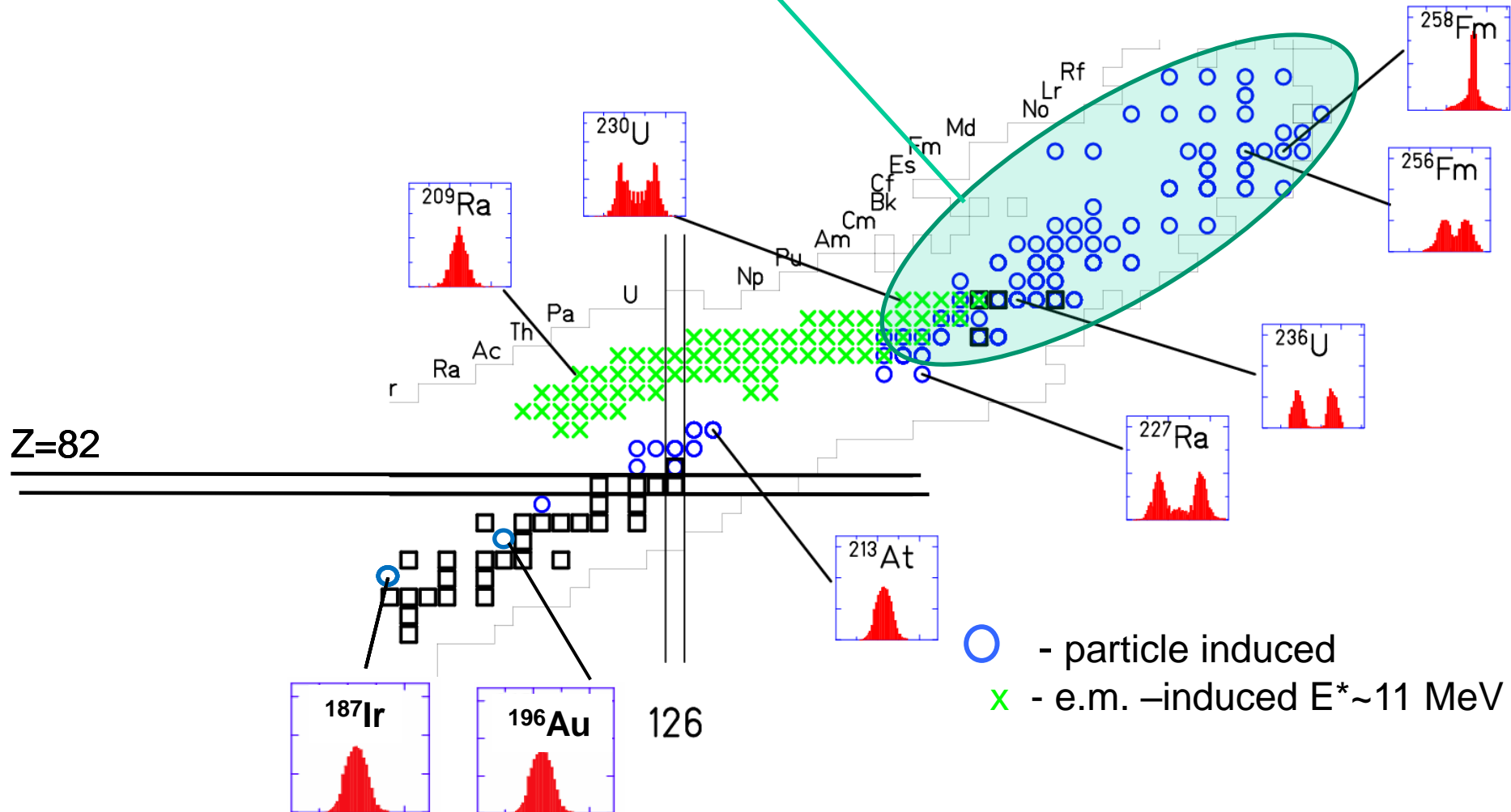
Y. Nagame and H. Nakahara, Radiochim. Acta 100, 605, 2012



# Experimental information on low-energy fission

## Nuclei with measured charge/mass split (RIPL-2 + GSI)

Heavy Actinides,  $N/Z \sim 1.56$ : **predominantly asymmetric**; spontaneous fission, fission isomers



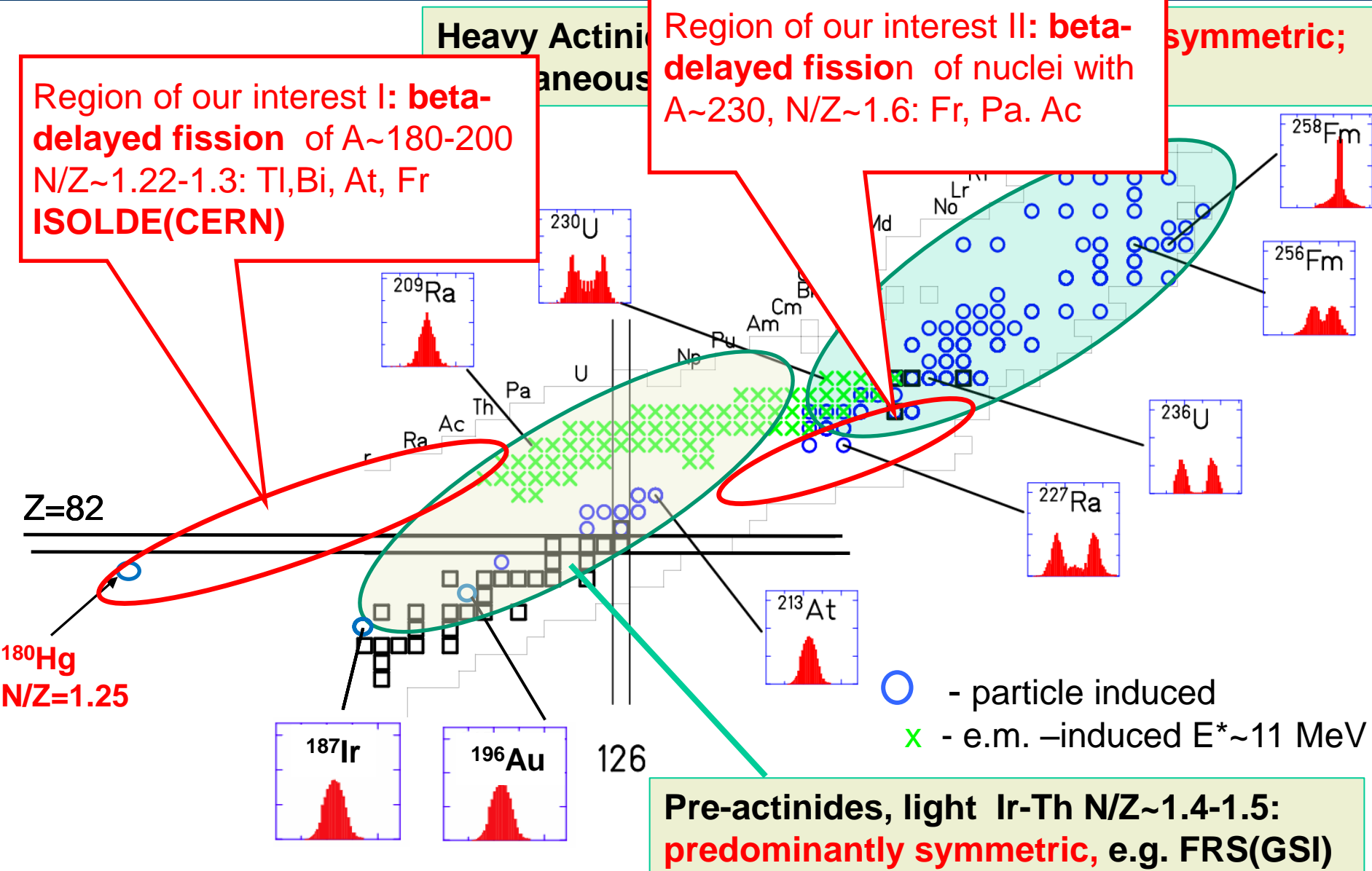
# Experimental information on low-energy fission

## Nuclei with measured charge/mass split (RIPL-2 + GSI)

Region of our interest I: **beta-delayed fission** of  $A \sim 180-200$   
 $N/Z \sim 1.22-1.3$ : Tl, Bi, At, Fr  
**ISOLDE(CERN)**

Region of our interest II: **beta-delayed fission** of nuclei with  
 $A \sim 230$ ,  $N/Z \sim 1.6$ : Fr, Pa, Ac

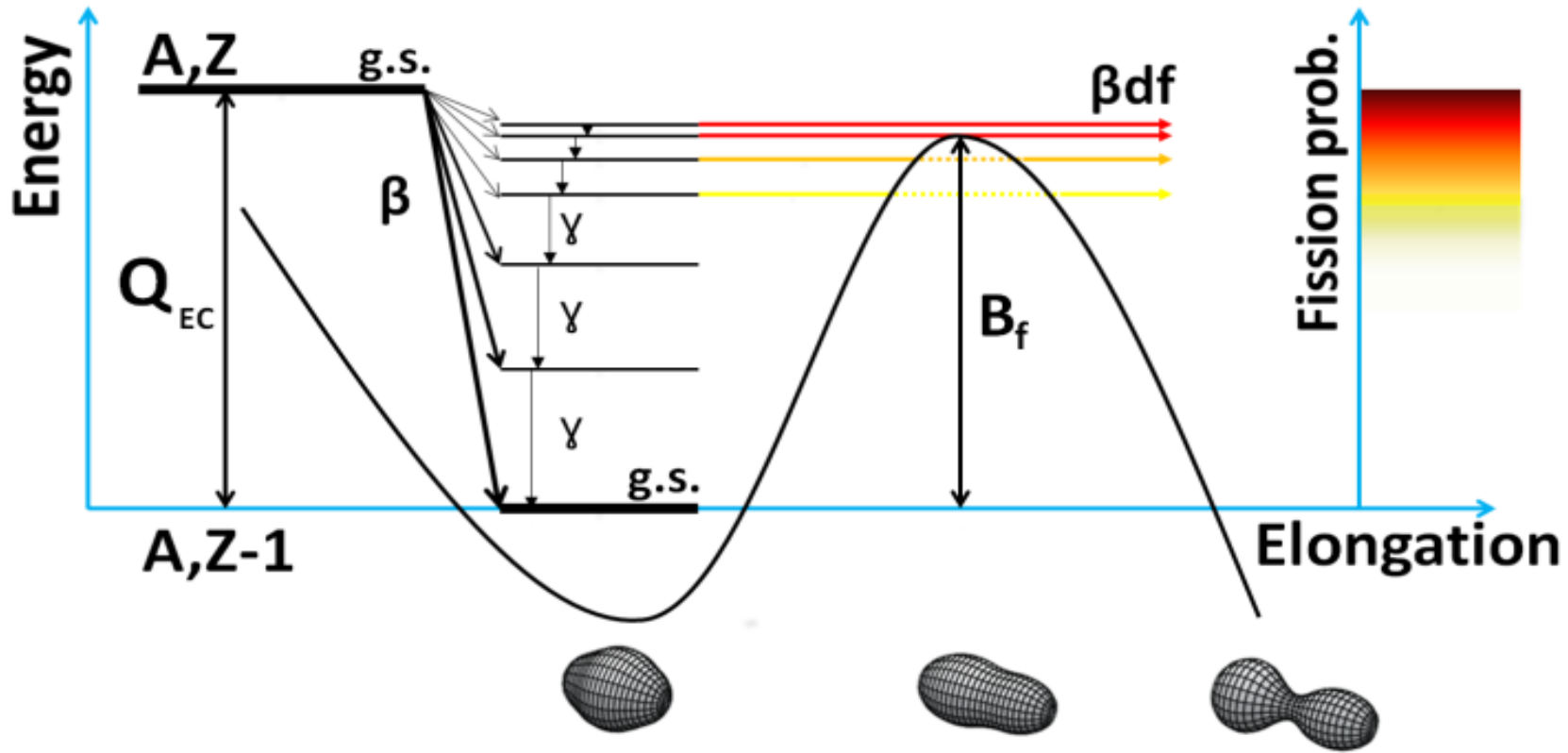
symmetric;



Pre-actinides, light Ir-Th  $N/Z \sim 1.4-1.5$ :  
**predominantly symmetric**, e.g. FRS(GSI)

# Beta-Delayed Fission

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

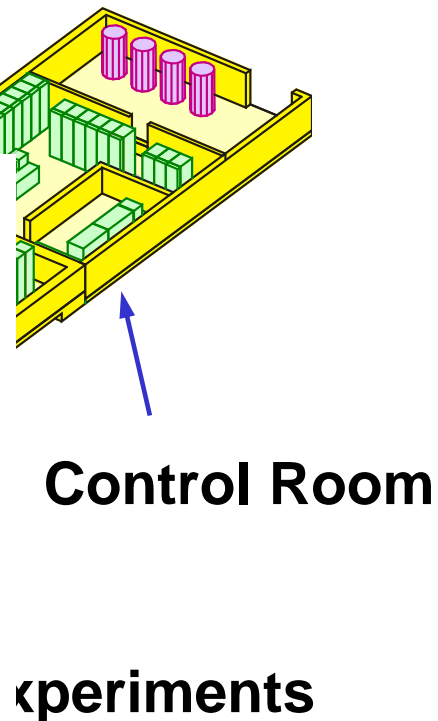
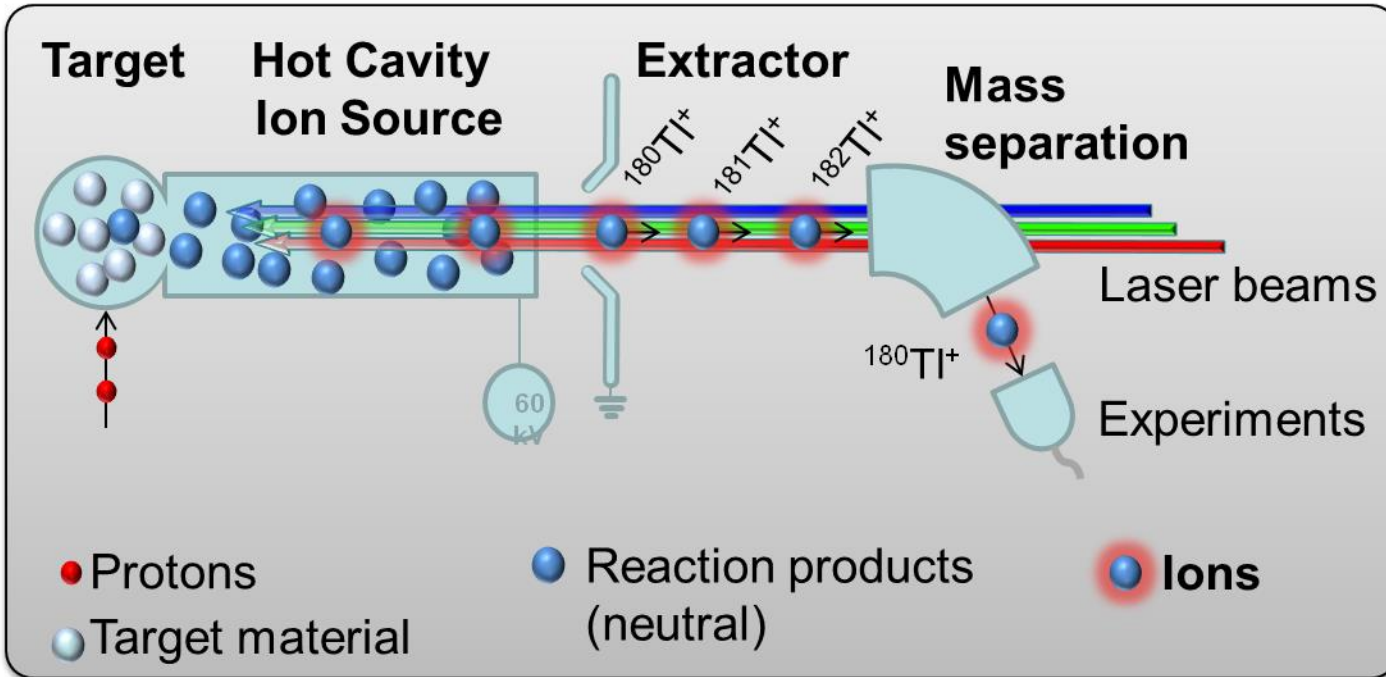
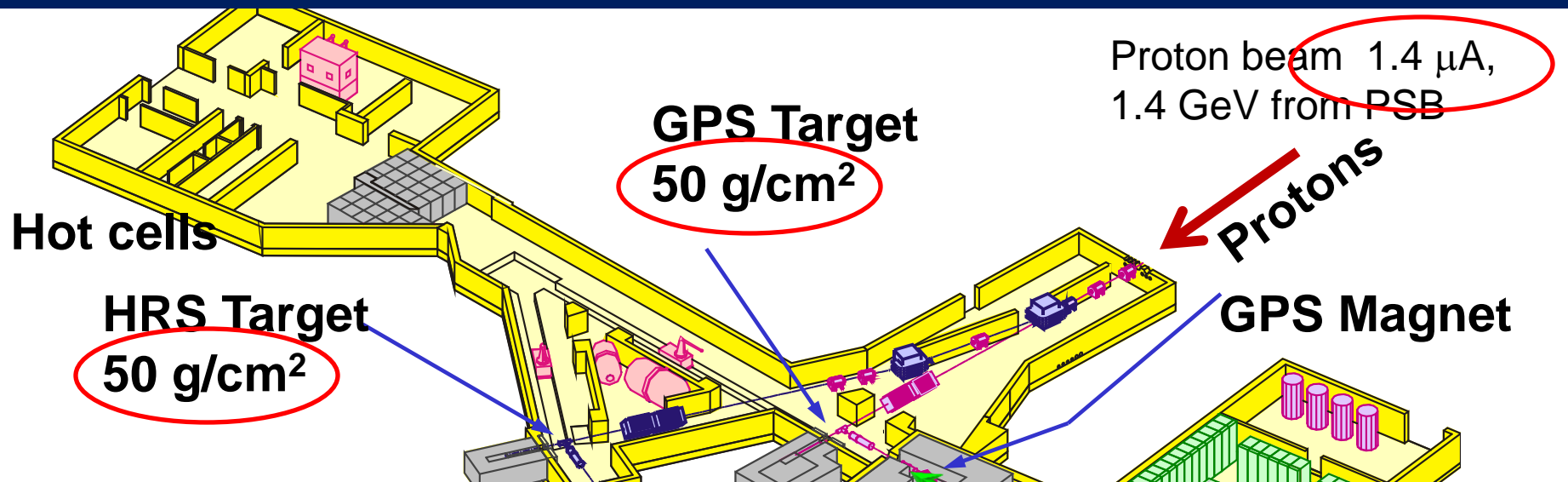


- Two step process:  $\beta$  decay followed by fission
- Low-energy fission ( $E^* \sim 3-12$  MeV, limited by  $Q_{EC}$ )  
e.g.  $^{180}\text{Tl}$ :  $Q_{EC} = 10.4$  MeV,  $B_{f,calc} = 9.8$  MeV
- Relatively low angular momentum of the state  
e.g.  $^{180}\text{Tl}$ :  $l = 4$  or  $5$  (some cases: up to  $10$ )

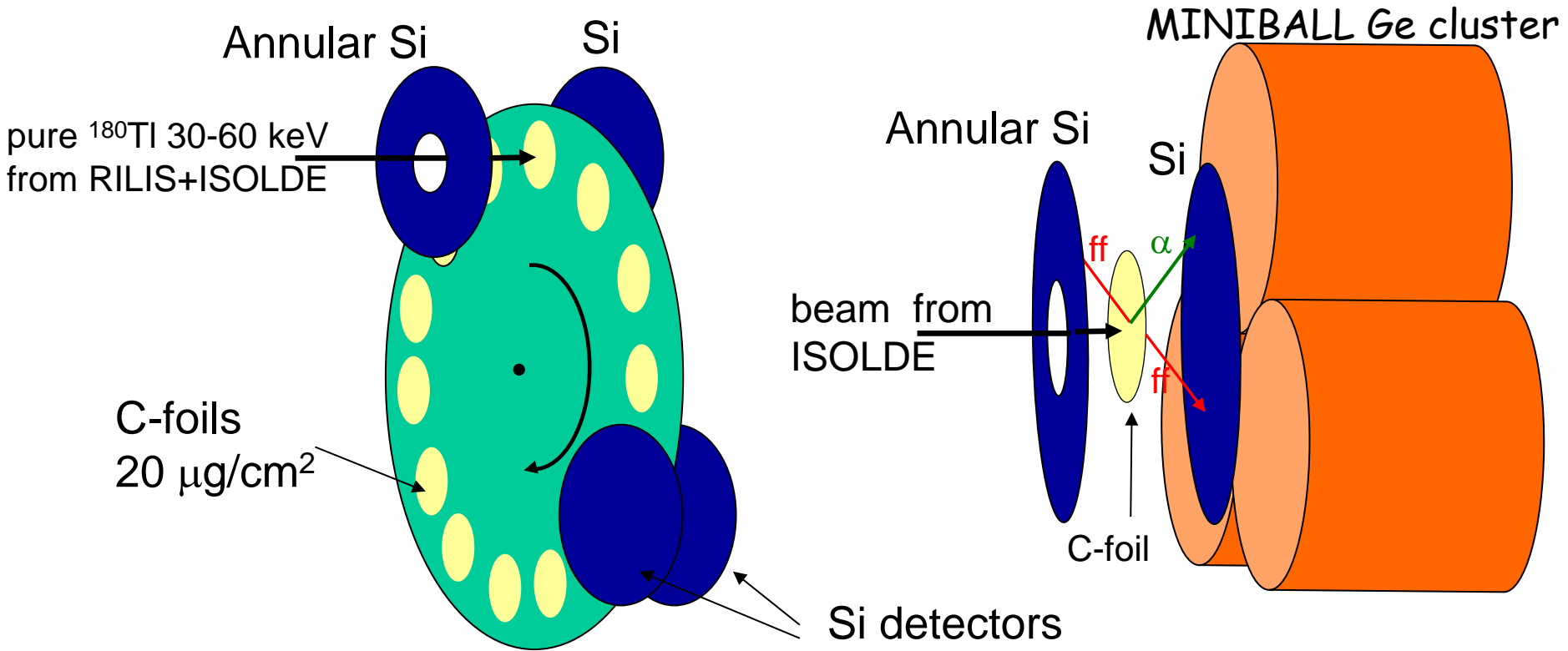
$\beta$ DF branch

$$P_{\beta DF} = \frac{N_{\beta DF}}{N_{\beta}}$$

# Mass Separator ISOLDE (CERN)

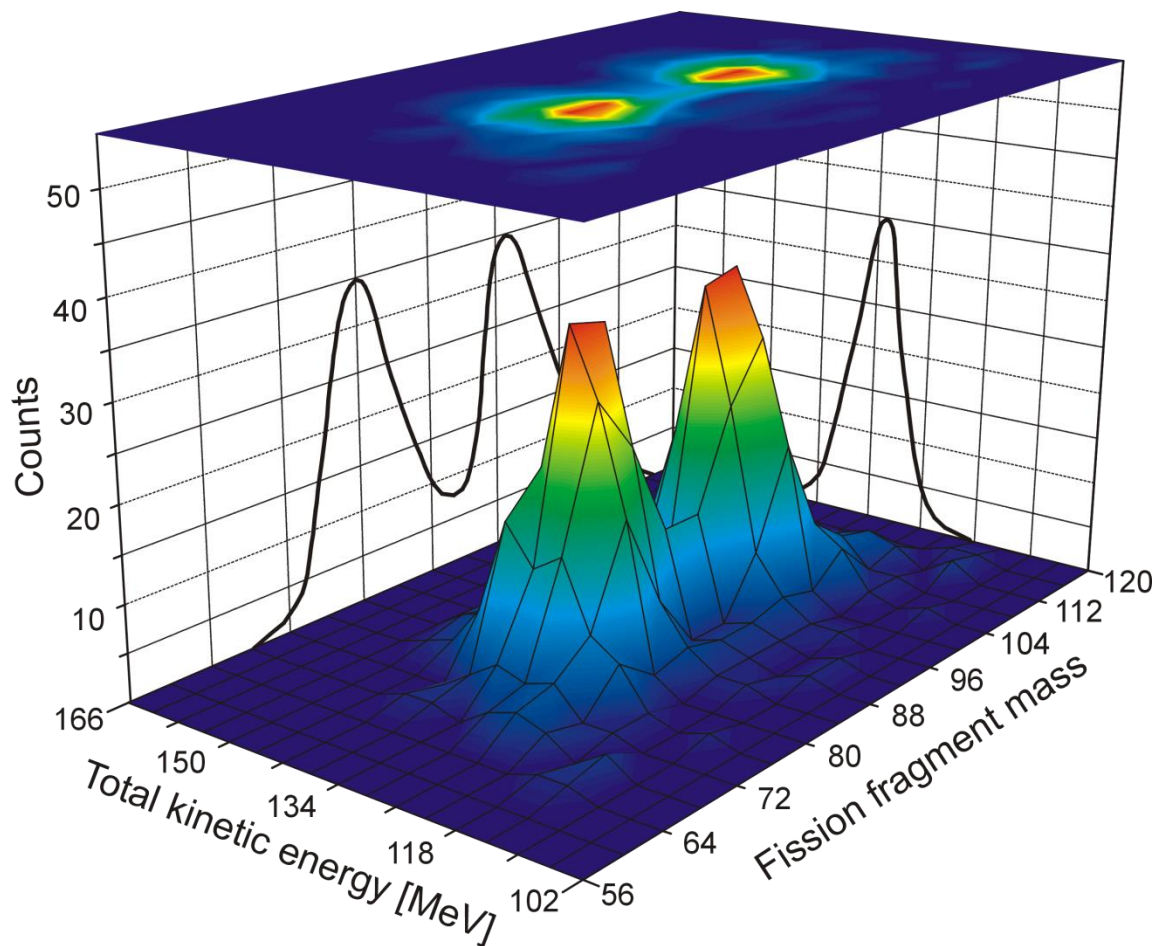
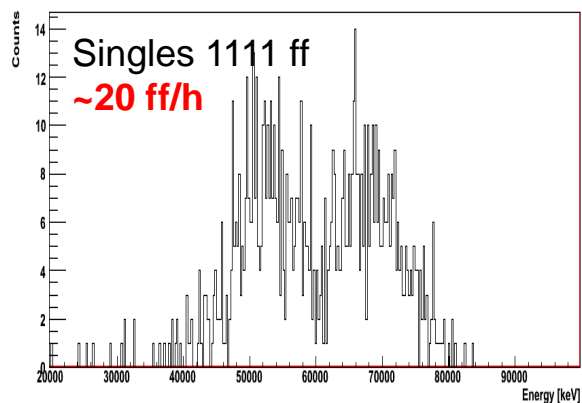
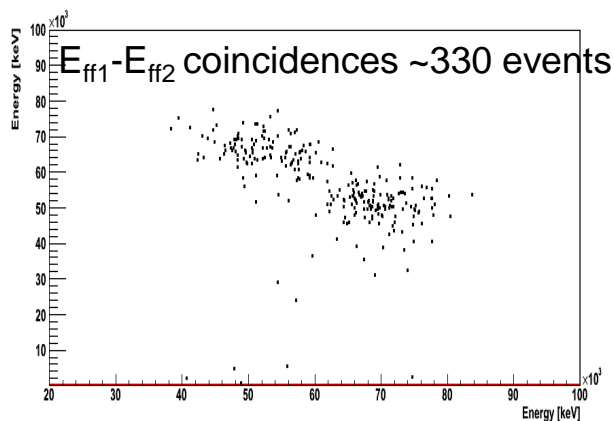


# Detection system for $\beta$ DF studies at ISOLDE



# Mass distribution of fission fragments from bDF of $^{180}\text{Tl}$

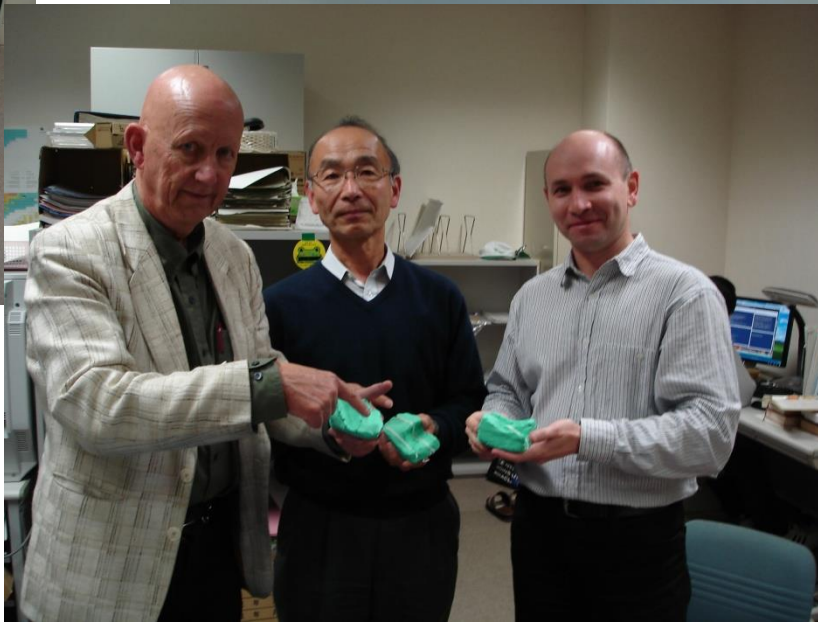
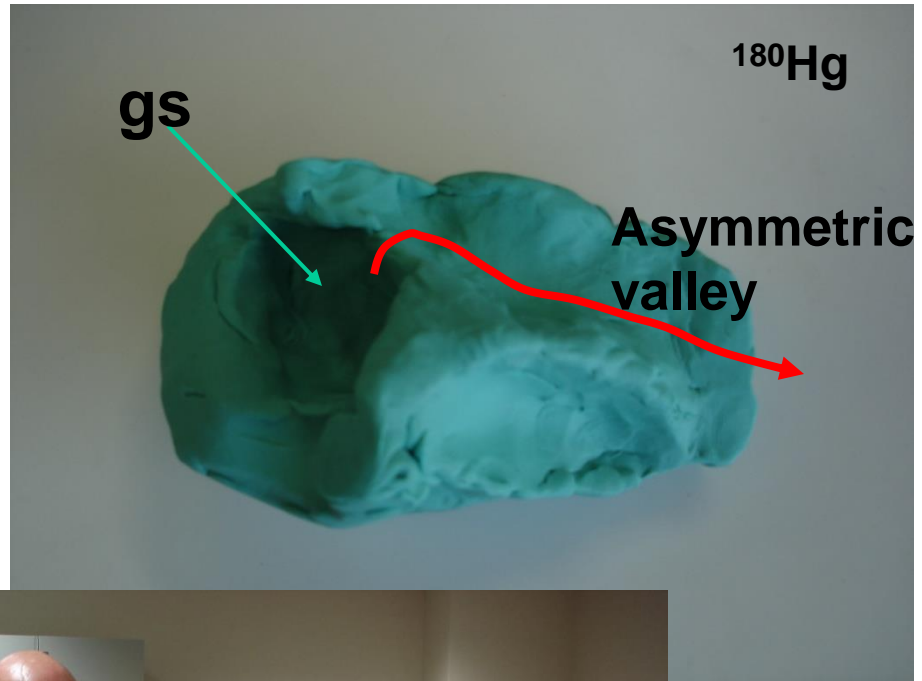
**ASYMMETRIC energy split! Thus asymmetric mass split:  $M_H=100(4)$  and  $M_L=80(4)$**



**The most probable fission fragments are  $^{100}\text{Ru}$  (N=56,Z=44) and  $^{80}\text{Kr}$  (N=44,Z=36)**

# CLDM (P. Möller et al., yet unpublished)

## CLDM: Clay Liquid Drop Model (circa 2008)



# New Type of Asymmetric Fission in Proton-Rich Nuclei

PRL **105**, 252502 (2010)

PHYSICAL REVIEW LETTERS

week ending  
17 DECEMBER 2010



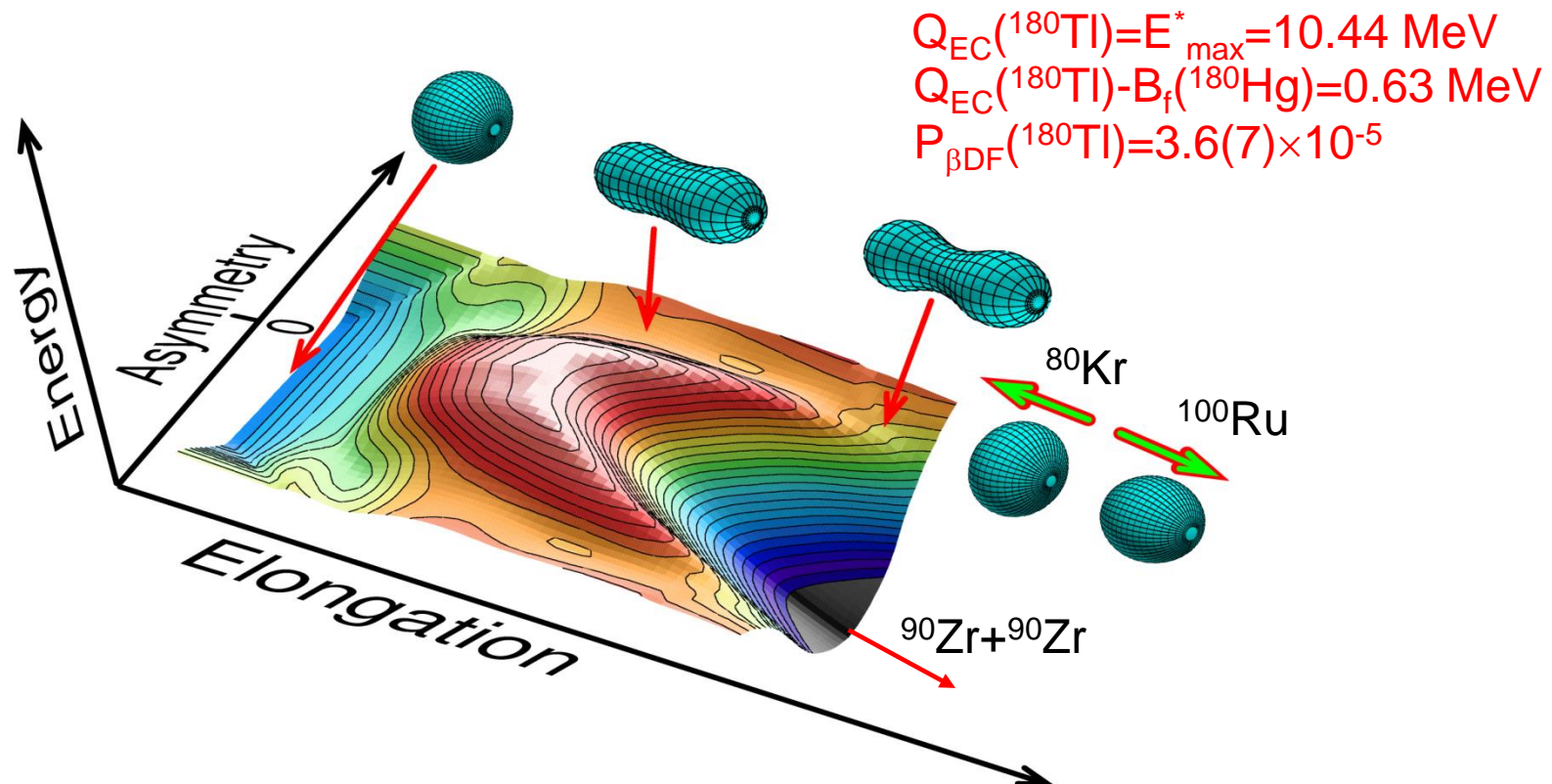
## New Type of Asymmetric Fission in Proton-Rich Nuclei **via $\beta$ DF of $^{180}\text{Tl}$**

A. N. Andreyev,<sup>1,2</sup> J. Elseviers,<sup>1</sup> M. Huyse,<sup>1</sup> P. Van Duppen,<sup>1</sup> S. Antalic,<sup>3</sup> A. Barzakh,<sup>4</sup> N. Bree,<sup>1</sup> T. E. Cocolios,<sup>1</sup> V. F. Comas,<sup>5</sup> J. Diriken,<sup>1</sup> D. Fedorov,<sup>4</sup> V. Fedosseev,<sup>6</sup> S. Franchoo,<sup>7</sup> J. A. Heredia,<sup>5</sup> O. Ivanov,<sup>1</sup> U. Köster,<sup>8</sup> B. A. Marsh,<sup>6</sup> K. Nishio,<sup>9</sup> R. D. Page,<sup>10</sup> N. Patronis,<sup>1,11</sup> M. Seliverstov,<sup>1,4</sup> I. Tsekhanovich,<sup>12,17</sup> P. Van den Bergh,<sup>1</sup> J. Van De Walle,<sup>6</sup> M. Venhart,<sup>1,3</sup> S. Vermote,<sup>13</sup> M. Veselsky,<sup>14</sup> C. Wagemans,<sup>13</sup> T. Ichikawa,<sup>15</sup> A. Iwamoto,<sup>9</sup> P. Möller,<sup>16</sup> and A. J. Sierk<sup>16</sup>

<sup>1</sup>*Instituut voor Kern- en Stralingsfysica, K.U. Leuven, University of Leuven, B-3001 Leuven, Belgium*

<sup>2</sup>*School of Engineering, University of the West of Scotland,*

*Paisley, PA1 2BE, United Kingdom, and the Scottish Universities Physics Alliance (SUPA)*



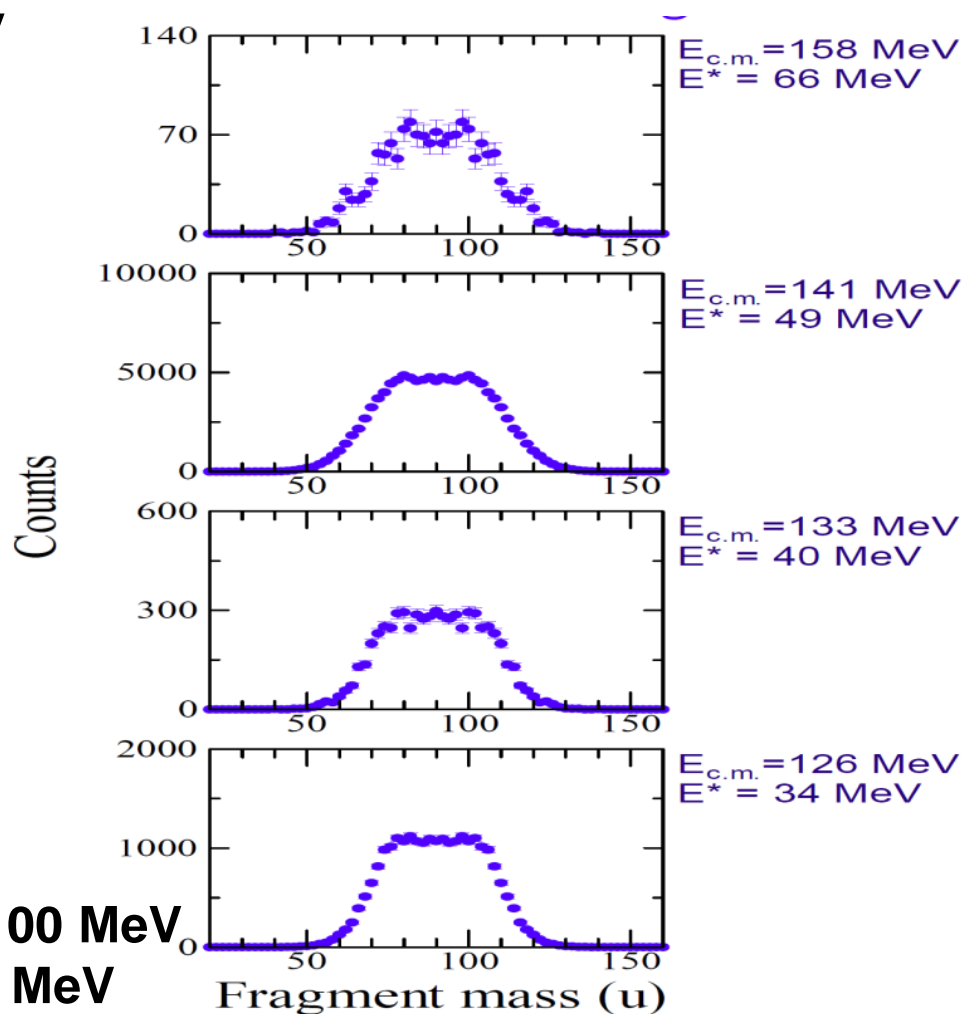
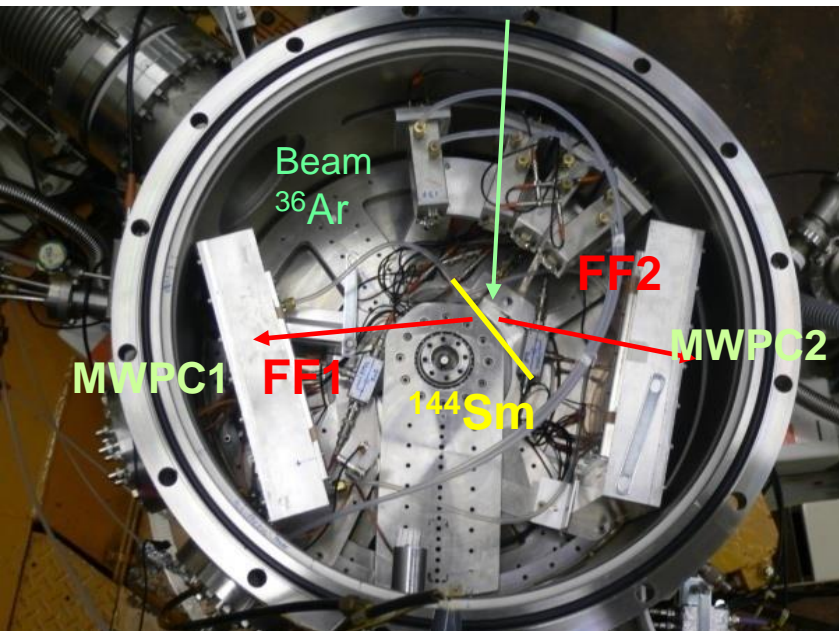
Calculations according to 5D fission model (P. Möller et al., Nature 409, 785 (2001))



# $^{180}\text{Hg}$ : More surprises?

## How does $^{180}\text{Hg}$ fission at higher excitation energies?

- $^{36}\text{Ar} + ^{144}\text{Sm} \rightarrow ^{180}\text{Hg}^*$   $E^* = 34\text{-}66\text{ MeV}$
- 2010-2012: JAEA, Tokai



- 2010-2012, JAEA
- $^{36,40}\text{Ar} + ^{144,154}\text{Sm} \rightarrow ^{180-194}\text{Hg}^*$   $E^* = 30\text{-}100\text{ MeV}$
- $^{36,40}\text{Ar} + ^{142}\text{Nd} \rightarrow ^{178,182}\text{Pt}^*$   $E^* = 30\text{-}100\text{ MeV}$

• Approved  $^{90}\text{Zr} + ^{90}\text{Zr} \rightarrow ^{180}\text{Hg}^*$  ( $E^* \sim 15\text{ MeV}$ )

Courtesy K. Nishio

**Even at  $E^* = 66\text{ MeV}$ : asymmetric mass split with  $A_1 \sim 100$ ,  $A_2 \sim 80$**

Supported by Reimei Foundation (JAEA)

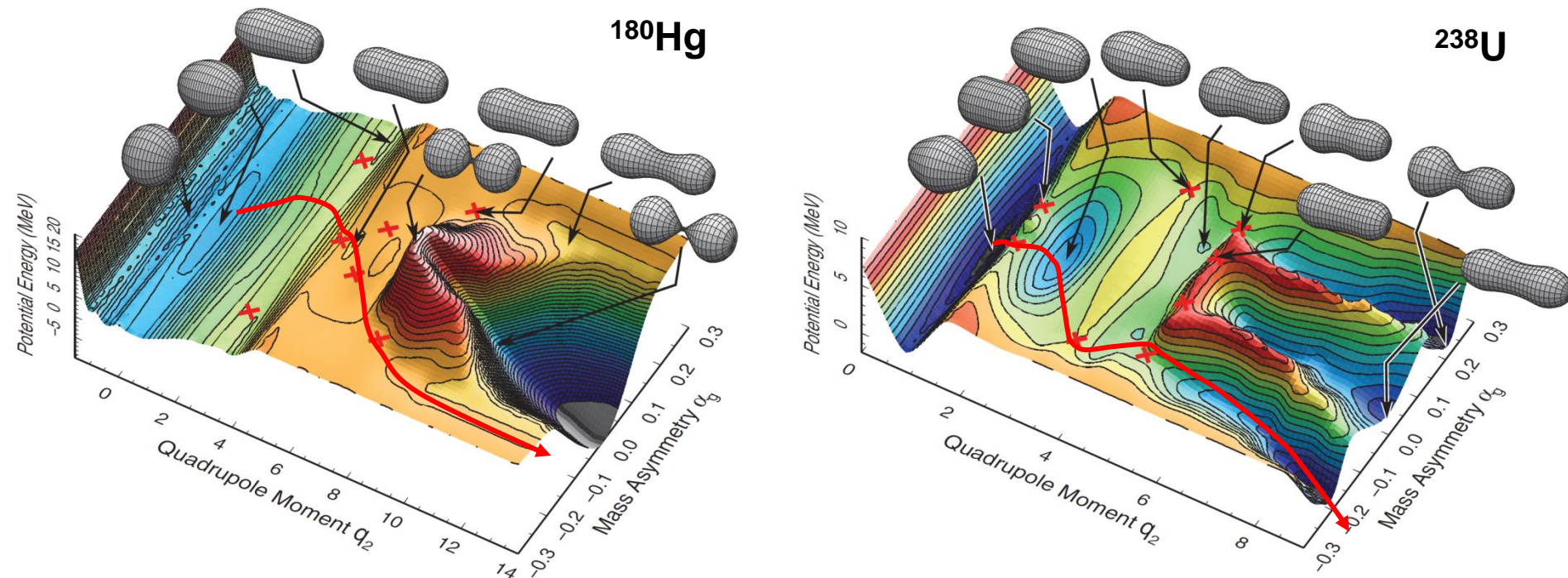
# Two types of asymmetry: what's the difference?

PHYSICAL REVIEW C **86**, 024610 (2012)

## Contrasting fission potential-energy structure of actinides and mercury isotopes

Takatoshi Ichikawa,<sup>1</sup> Akira Iwamoto,<sup>2</sup> Peter Möller,<sup>3</sup> and Arnold J. Sierk<sup>3</sup>

**Conclusions:** The mechanism of asymmetric fission must be very different in the lighter proton-rich mercury isotopes compared to the actinide region and is apparently unrelated to fragment shell structure. Isotopes lighter than  $^{192}\text{Hg}$  have the saddle point shielded from a deep symmetric valley by a significant ridge. The ridge vanishes for the heavier Hg isotopes, for which we would expect a qualitatively different asymmetry of the fragments.



# 'Self-consistent Scission-Point Model'

PHYSICAL REVIEW C **86**, 064601 (2012)

## Role of deformed shell effects on the mass asymmetry in nuclear fission of mercury isotopes

Stefano Panebianco, Jean-Luc Sida, Héloïse Goutte, and Jean-François Lemaître  
*IRFU/Service de Physique Nucléaire, CEA Centre de Saclay, F-91191 Gif-sur-Yvette, France*

Noël Dubray and Stéphane Hilaire  
*CEA, DAM, DIF, F-91297, Arpajon, France*  
 (Received 9 October 2012; published 3 December 2012)

$$\begin{aligned}
 E_{av}(Z_{1,2}, N_{1,2}, \beta_{1,2}, d) \\
 = E_{tot} - E_{HFB}(Z_1, N_1, \beta_1) - E_{HFB}(Z_2, N_2, \beta_2) \\
 - E_{nucl}(Z_{1,2}, N_{1,2}, \beta_{1,2}, d) - E_{Coul}(Z_{1,2}, N_{1,2}, \beta_{1,2}, d).
 \end{aligned}$$

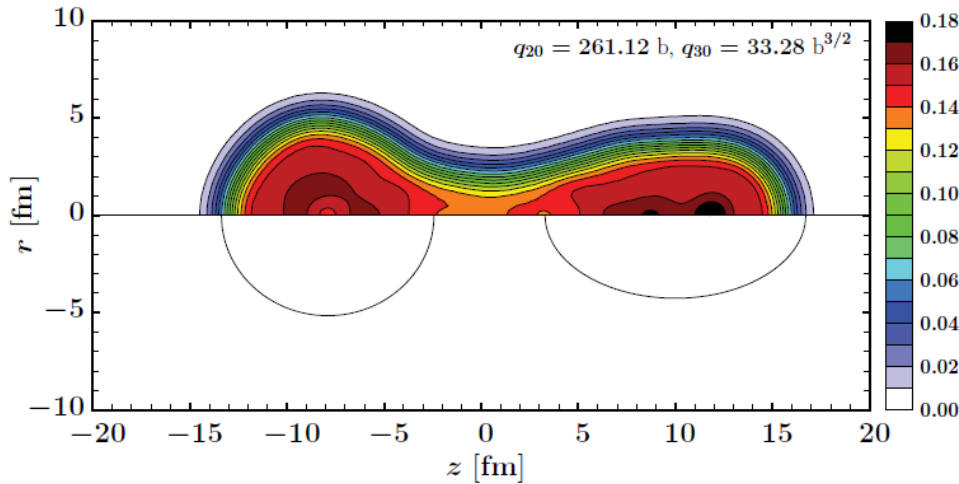


FIG. 4. (Color online) Total nuclear density for the most energetically favorable scission configuration in  $^{180}\text{Hg}$  fission, extracted from a self-consistent HFB calculation. In the lower part of the figure, two

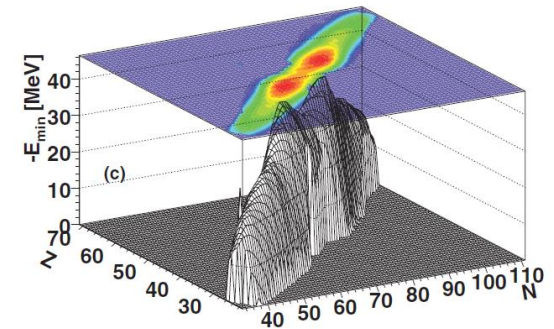
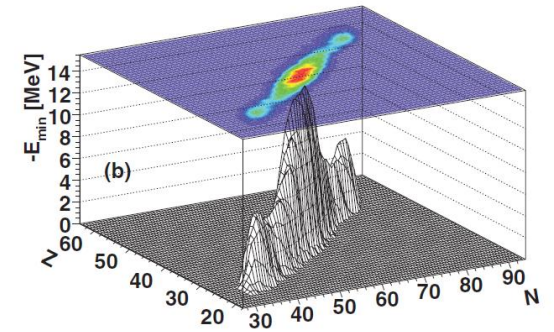
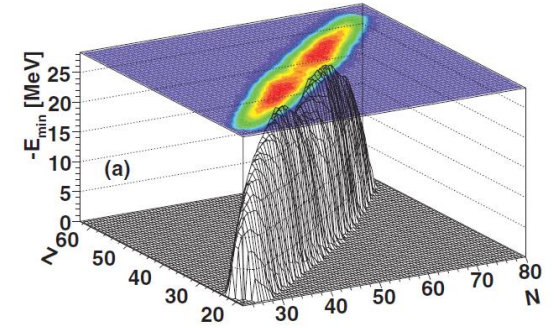


FIG. 2. (Color online) Minimum absolute available energy at scission calculated for all possible fragmentations in (a)  $^{180}\text{Hg}$  and (b)  $^{198}\text{Hg}$  fission at 10 MeV and in (c) the thermal  $n$ -induced fission of  $^{235}\text{U}$ .

# 'Mean-field HFB+Gogny D1S'

PHYSICAL REVIEW C **86**, 024601 (2012)

## Fission modes of mercury isotopes

M. Warda,<sup>1</sup> A. Staszczak,<sup>1,2,3</sup> and W. Nazarewicz<sup>2,3,4</sup>

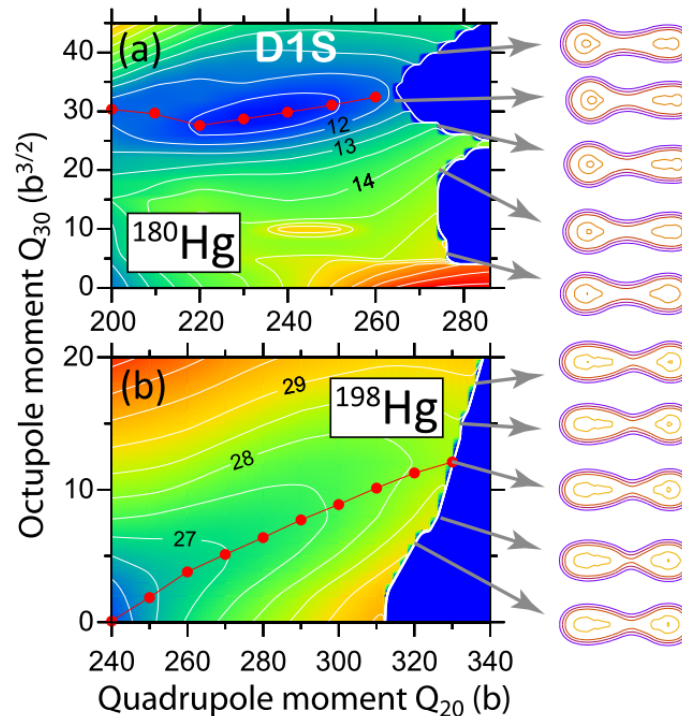
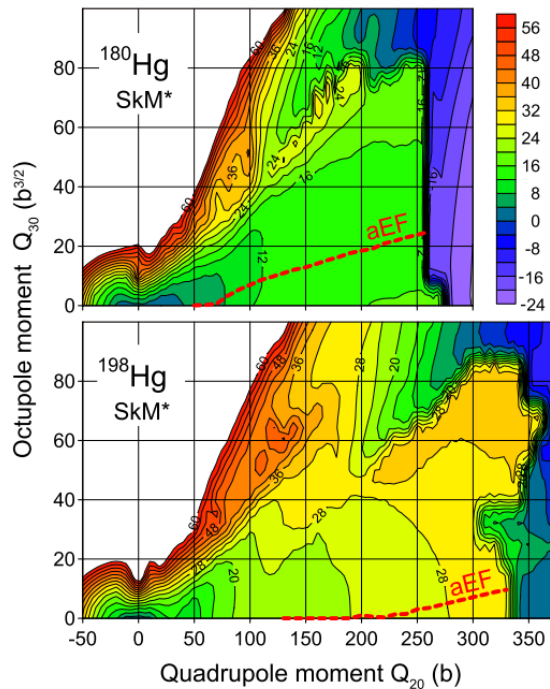


FIG. 2. (Color online) PES for  $^{180}\text{Hg}$  (top) and  $^{198}\text{Hg}$  (bottom) in the plane of collective coordinates  $Q_{20} - Q_{30}$  in HFB-SkM\*. The aEF fission pathway corresponding to asymmetric elongated fragments is marked. The difference between contour lines is 4 MeV. The effects due to triaxiality, known to impact inner fission barriers in the actinides, are negligible here.

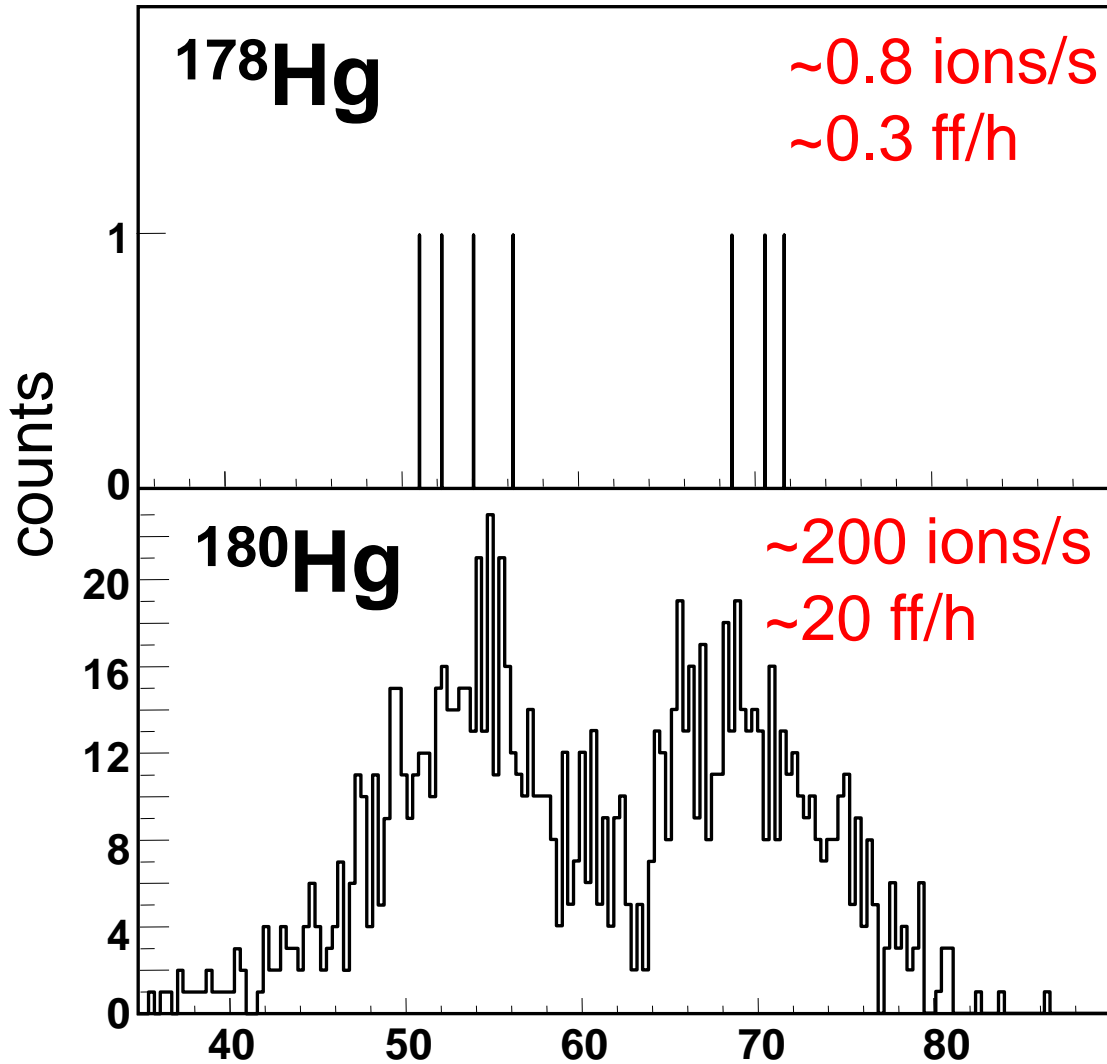
FIG. 3. (Color online) PES in HFB-D1S for  $^{180}\text{Hg}$  (top) and  $^{198}\text{Hg}$  (bottom) in the  $(Q_{20}, Q_{30})$  plane in the pre-scission region of aEF valley. The symmetric limit corresponds to  $Q_{30} = 0$ . The aEF valley and density profiles for pre-scission configurations are indicated. The difference between contour lines is 0.5 MeV. Note different  $Q_{30}$ -scales in  $^{180}\text{Hg}$  and  $^{198}\text{Hg}$  plots.

# $\beta$ DF of $^{178}\text{Tl}$ @ISOLDE

V. Liberati et al (PRC, 2013, in print)

$$Q_{\text{EC}}(^{178}\text{Tl}) = E_{\text{max}}^*(^{178}\text{Hg}) = 11.14 \text{ MeV}$$

$$Q_{\text{EC}}(^{178}\text{Tl}) - B_f(^{178}\text{Hg}) = 1.82 \text{ MeV}$$



At this level of statistics:  
also asymmetric fission  
of  $^{178}\text{Hg}$ , with mass split  
similar to  $^{180}\text{Hg}$

$$E_{\text{max}}^*(^{180}\text{Hg}) = 10.44 \text{ MeV}$$

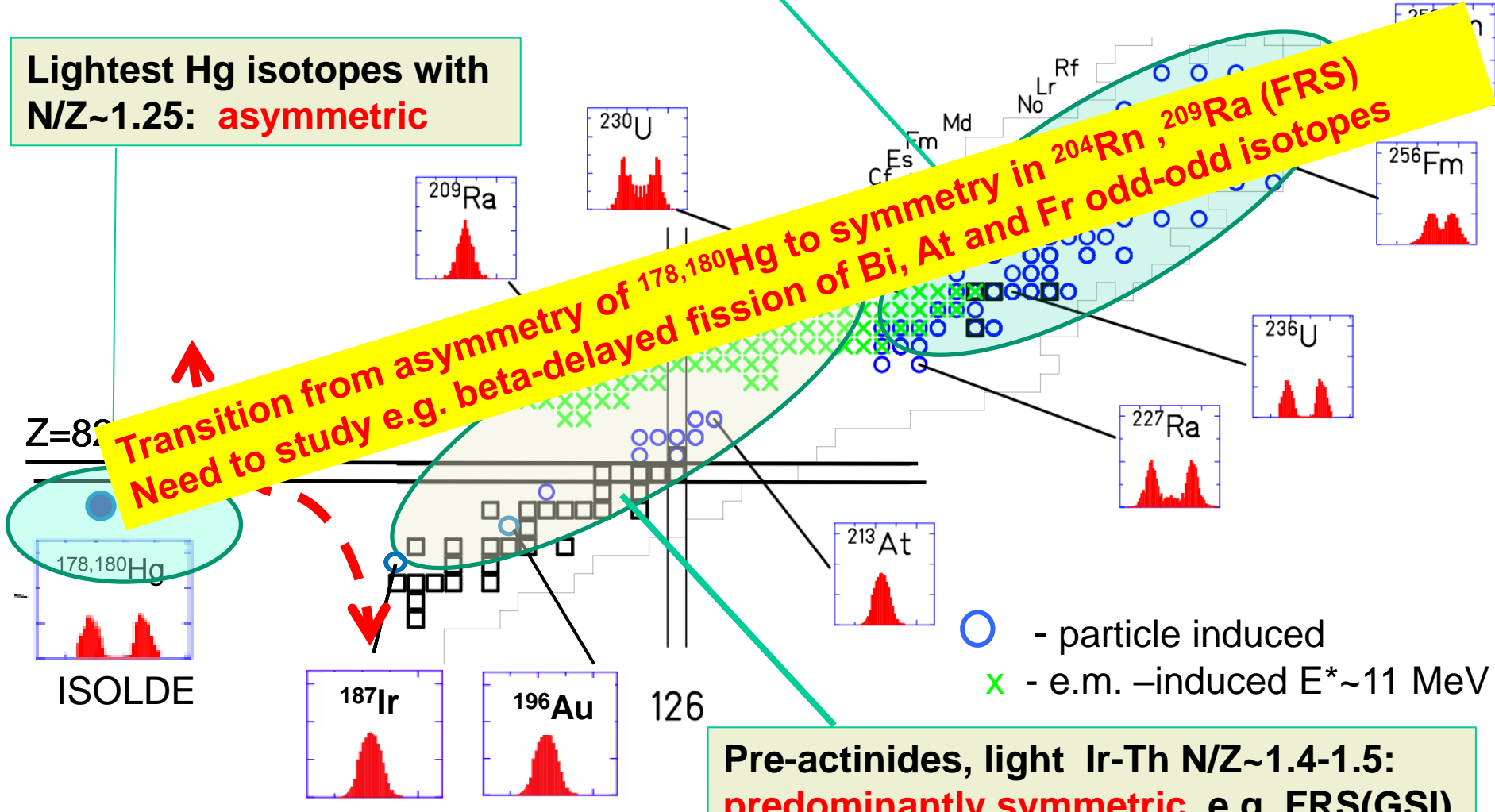
Fission Fragments Energy in Si detector [MeV]

# From Asymmetry to Symmetry

Heavy Actinides,  $N/Z \sim 1.56$ : **predominantly asymmetric**; spontaneous fission, fission isomers

Lightest Hg isotopes with  $N/Z \sim 1.25$ : **asymmetric**

**Transition from asymmetry of  $^{178,180}\text{Hg}$  to symmetry in  $^{204}\text{Rn}$ ,  $^{209}\text{Ra}$  (FRS)  
Need to study e.g. beta-delayed fission of Bi, At and Fr odd-odd isotopes**



○ - particle induced  
x - e.m. -induced  $E^* \sim 11$  MeV

Pre-actinides, light Ir-Th  $N/Z \sim 1.4-1.5$ : **predominantly symmetric**, e.g. FRS(GSI)

ISOLDE

Z=82

126

# Fission of Proton-rich nuclei with $A \sim 180-200$

Courtesy P. Moller (LANL) and J. Randrup (LBNL), 5<sup>th</sup> ASRC workshop on Fission, Tokai 2012

CERN-ISOLDE

JAEA tandem

Heavy-ion induced  
Fission Mercury  
chain of isotopes  
(High Excitation  
Energy)

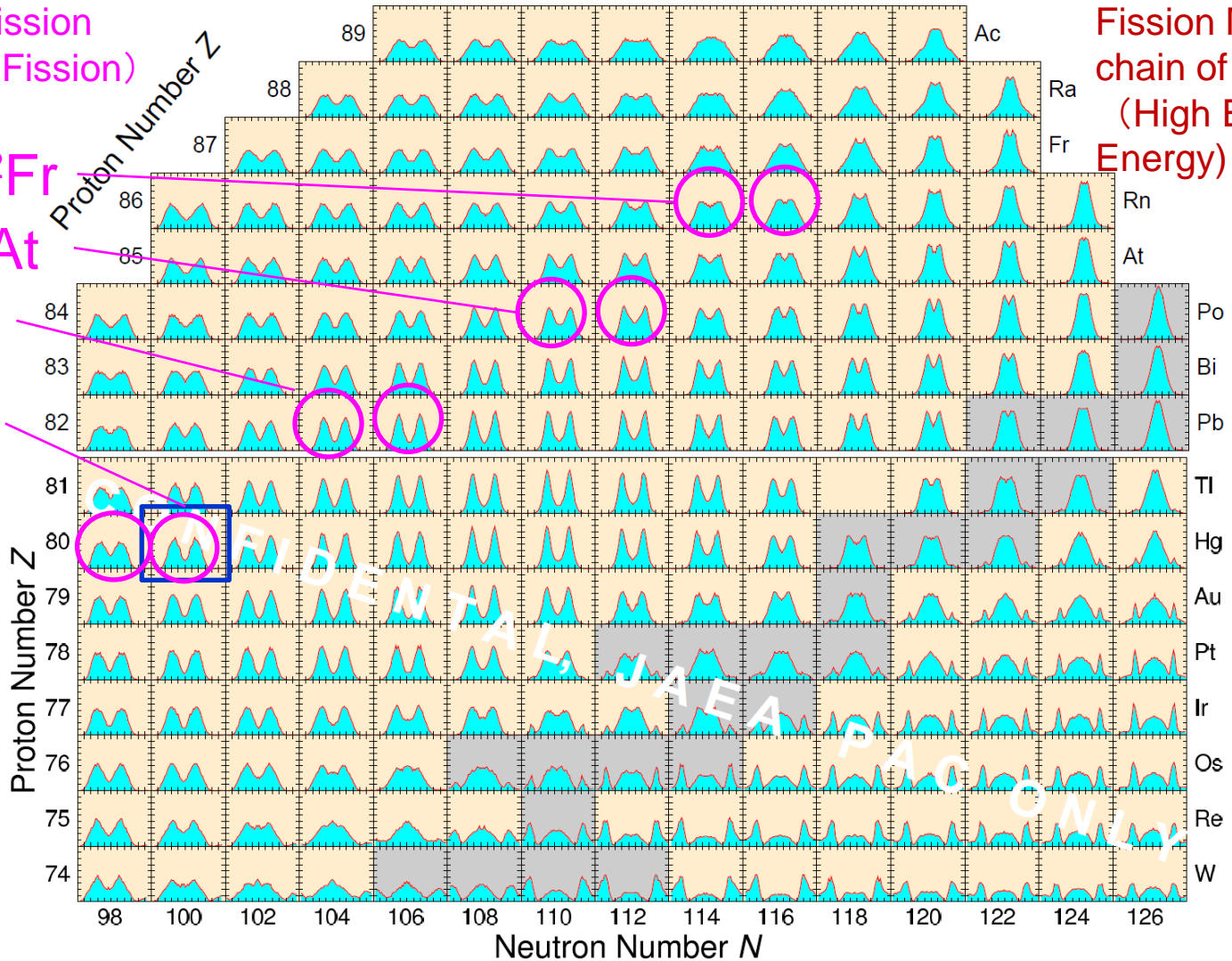
EC -delayed Fission  
(Low Energy Fission)

200,202Fr

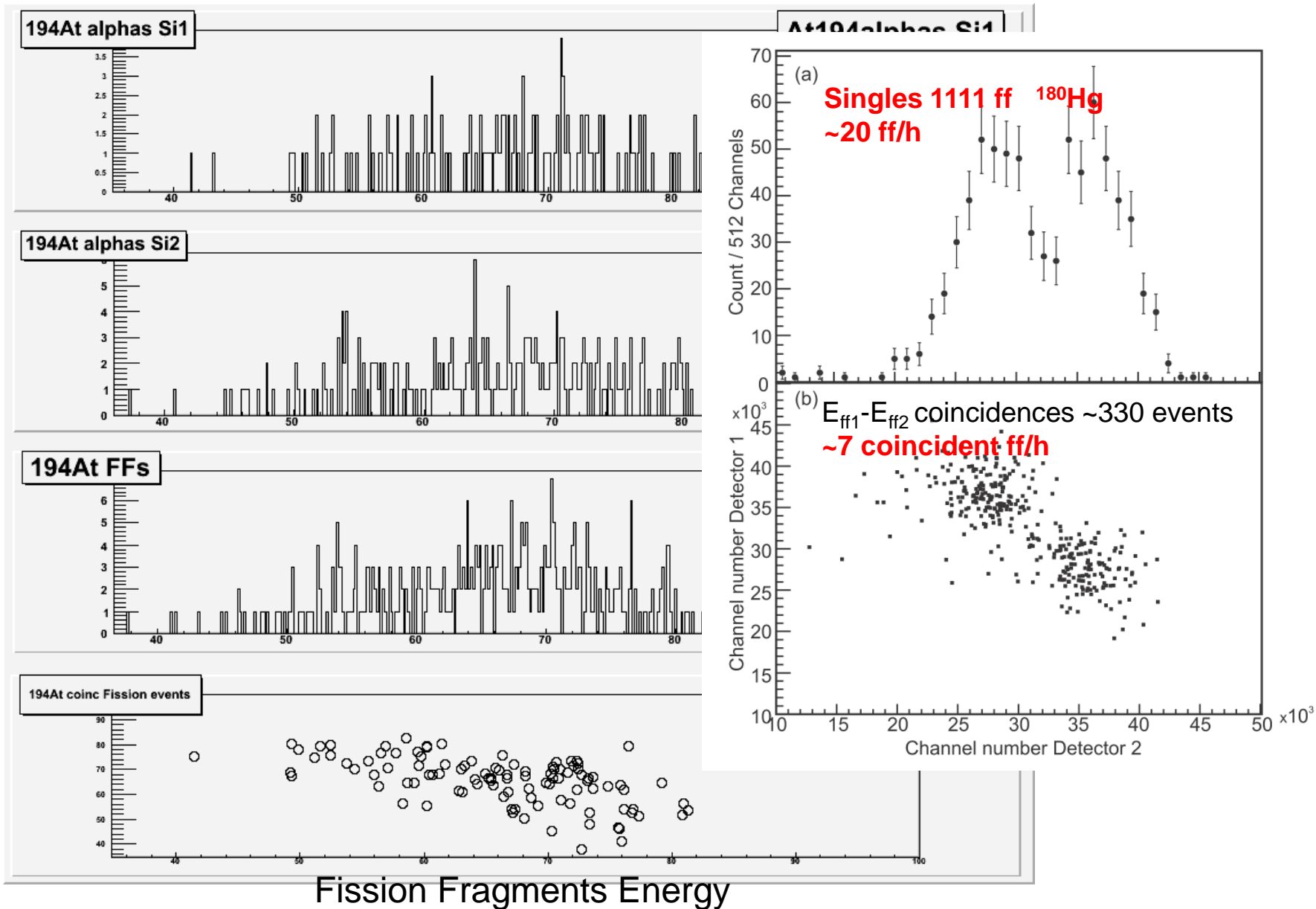
194,196At

186,188Bi

180,178Tl



# IS534 (ISOLDE) , 9-14 May 2012: Mass Distributions Measurements of $^{194,196}\text{Po}$ via $\beta\text{DF}$ of $^{194,196}\text{At}$





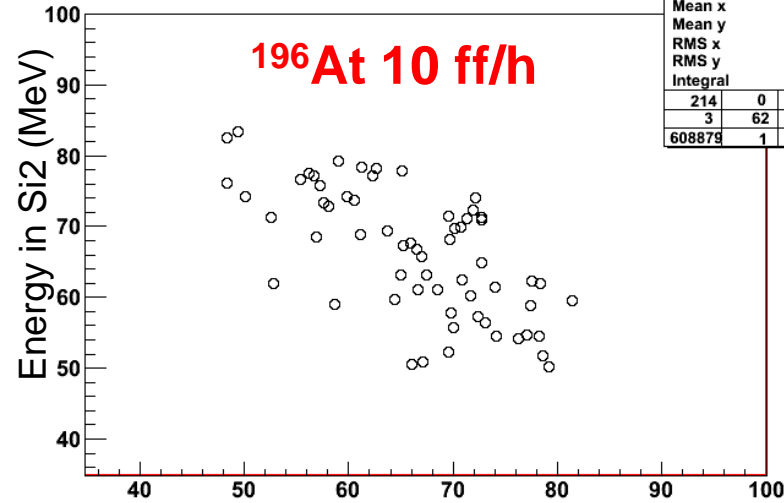
# IS534, 9-14 May 2012: Mass Distributions Measurements of $^{194,196}\text{Po}$ via $\beta\text{DF}$ of $^{194,196}\text{At}$

**$^{196}\text{At}$  coinc Fission events**

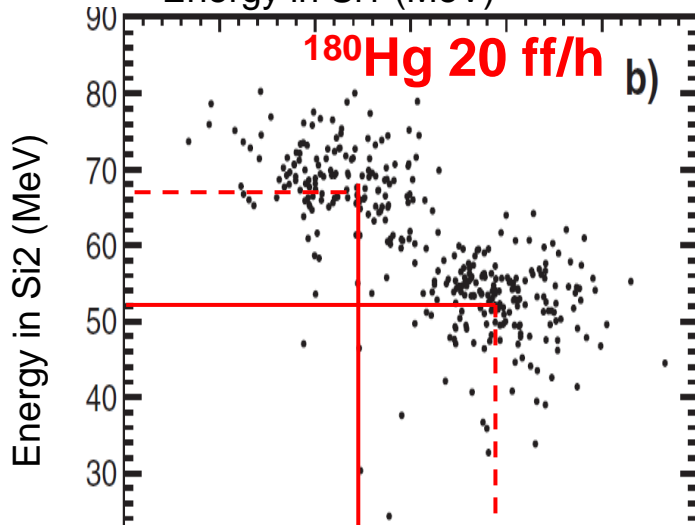
**At196coincidences**

|          |        |   |
|----------|--------|---|
| Entries  | 609160 |   |
| Mean x   | 66.24  |   |
| Mean y   | 66.39  |   |
| RMS x    | 8.372  |   |
| RMS y    | 8.842  |   |
| Integral | 62     |   |
| 214      | 0      | 0 |
| 3        | 62     | 0 |
| 608879   | 1      | 1 |

**$^{196}\text{At}$  10 ff/h**



**Energy in Si1 (MeV)**

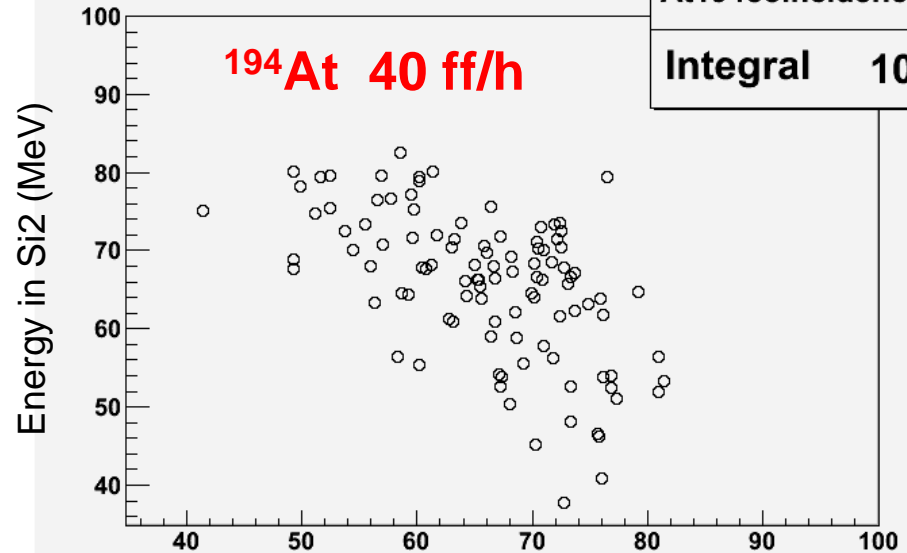


**$^{194}\text{At}$  coinc Fission events**

**At194coincidences**

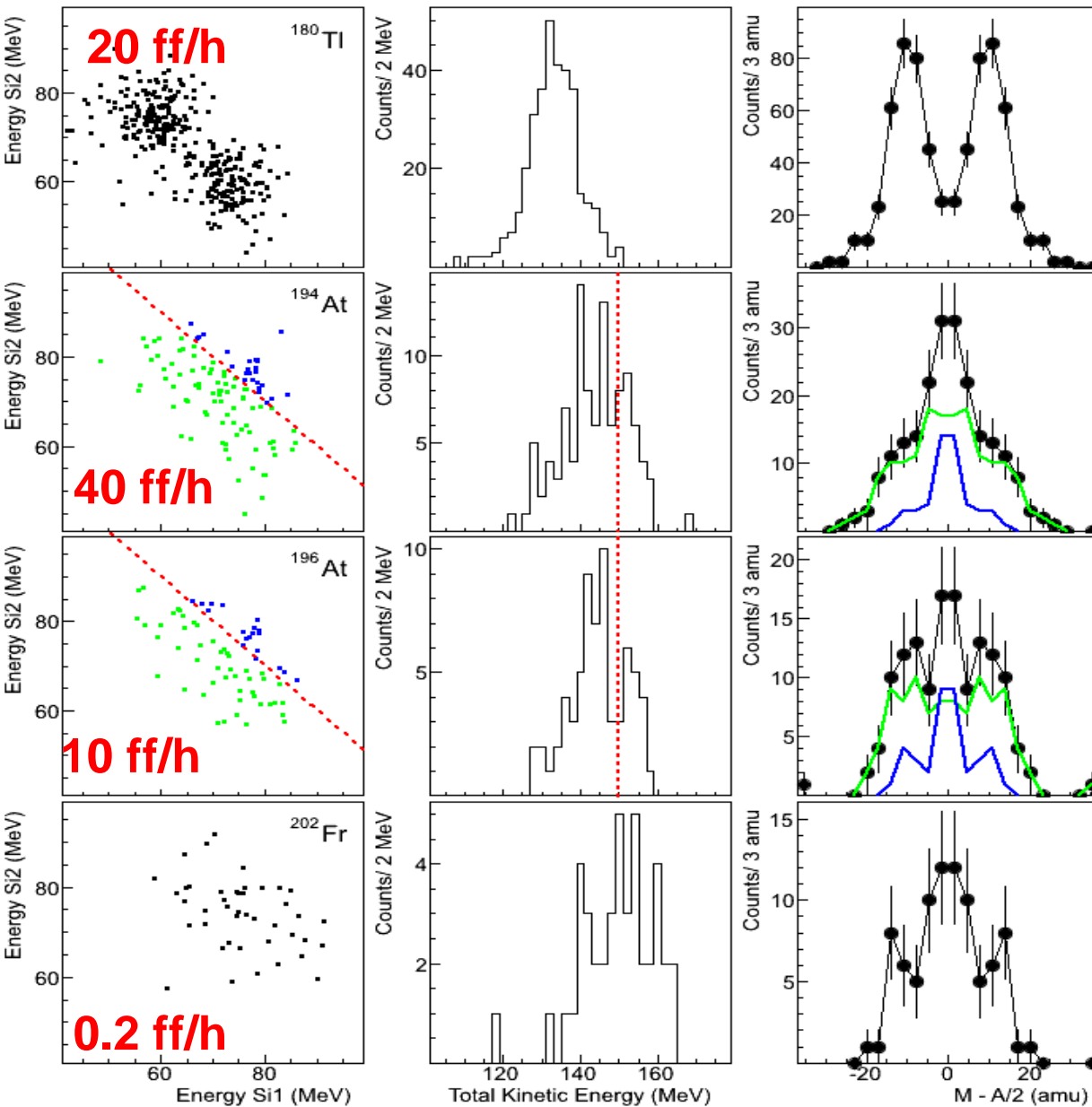
**Integral 104**

**$^{194}\text{At}$  40 ff/h**



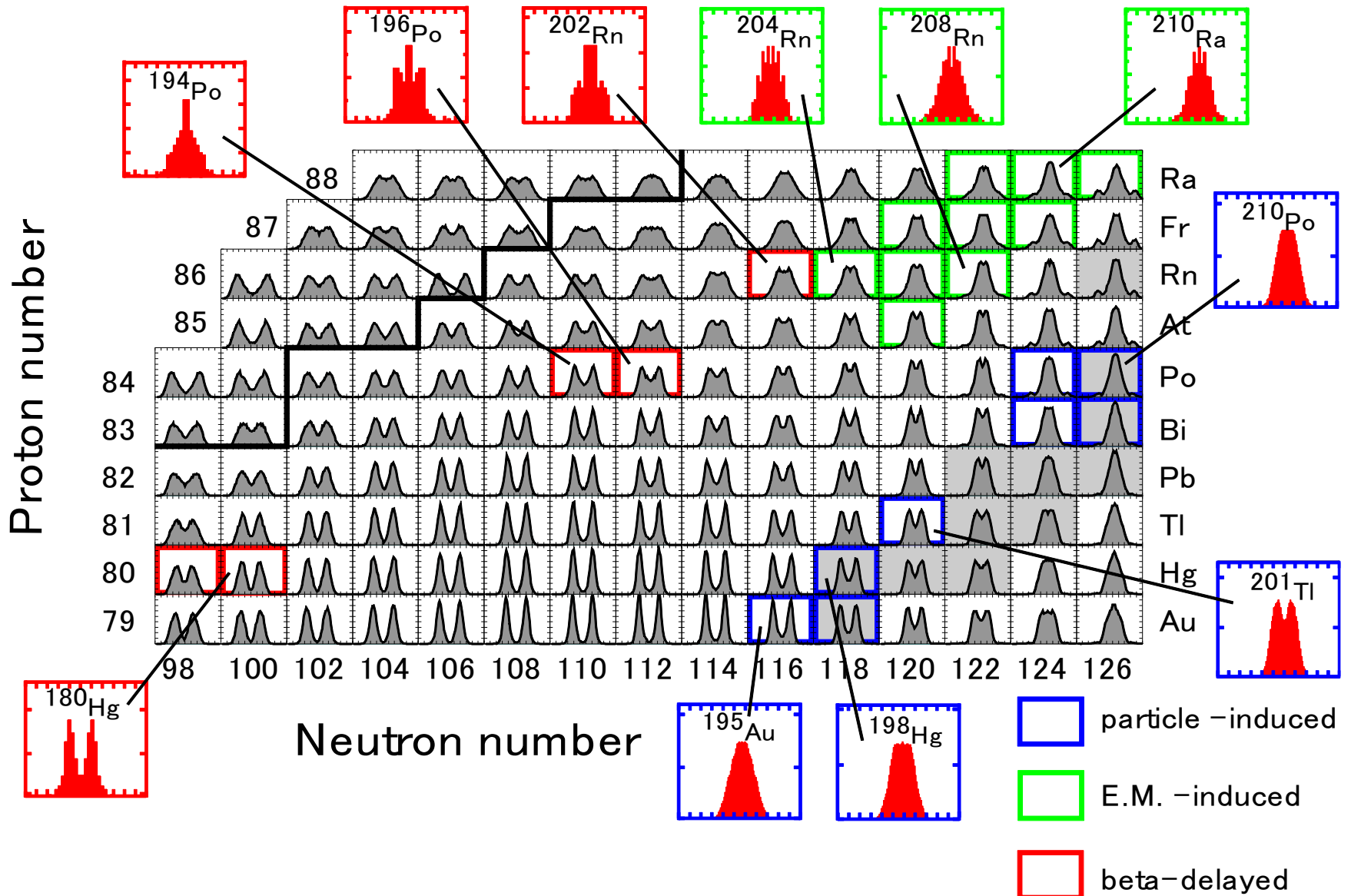
**Clear difference in energy (thus, mass) distribution between 2-peaked fission of  $^{180}\text{Hg}$  and a broad distribution in  $^{194,196}\text{Po}$**

# May and June 2012: Mass Distributions Measurements via $\beta$ DF of $^{194,196}\text{At}$ and $^{200,202}\text{Fr}$

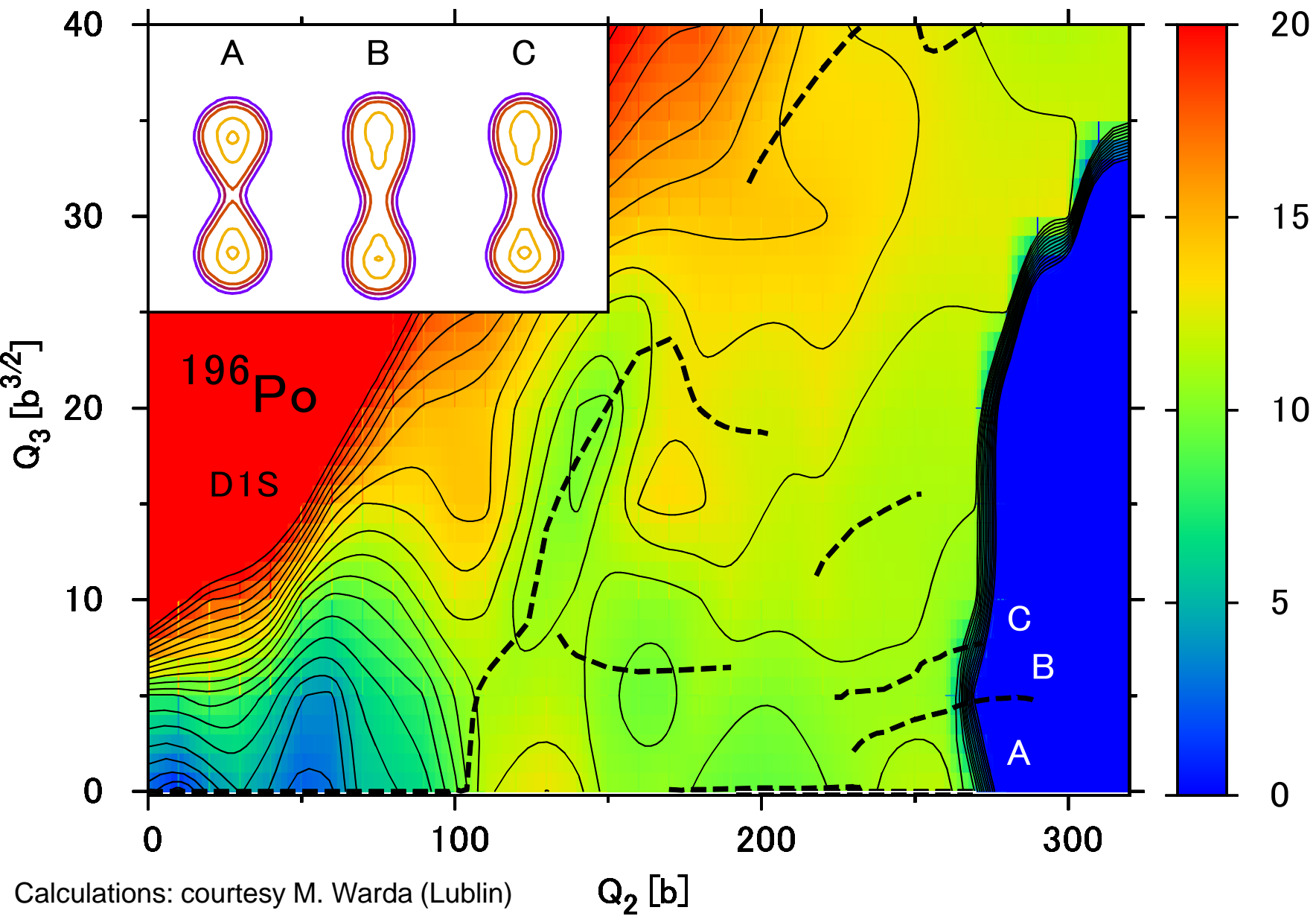


Courtesy L. Ghys (KU Leuven)

# Experiment vs Theory



# Experiment vs Theory ( $^{196}\text{At}$ )



# Mapping beta-delayed fission: from neutron-deficient to neutron-rich nuclei

Reviews of Modern Physics, 85, 1541 (2013)

## *Colloquium:* **Beta-delayed fission of atomic nuclei**

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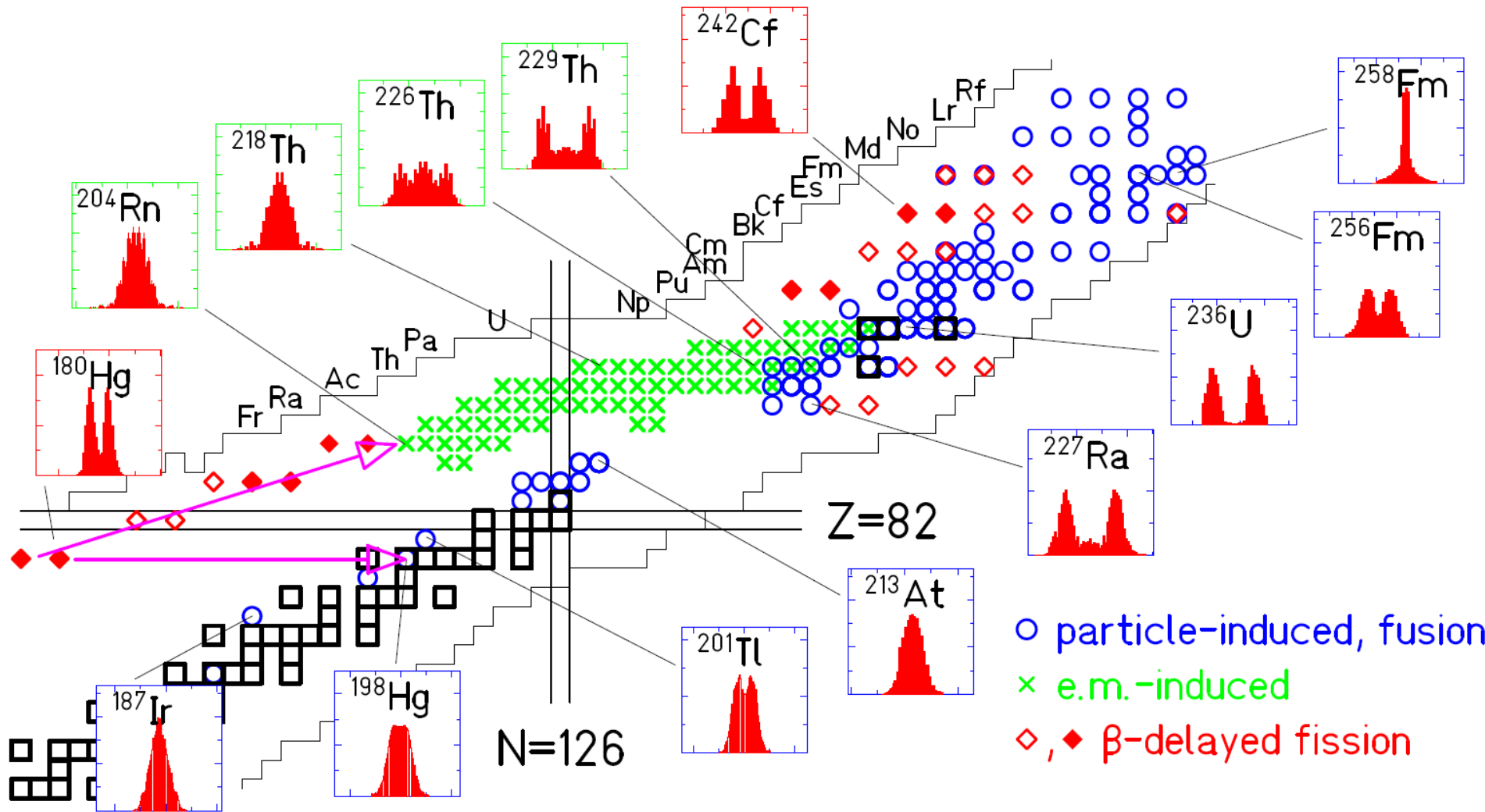
This Colloquium reviews the studies of exotic type of low-energy nuclear fission, the  $\beta$ -delayed fission ( $\beta$ DF). Emphasis is made on the new data from very neutron-deficient nuclei in the lead region, previously scarcely studied as far as fission is concerned. These

# Known Beta-delayed fission nuclei

| Isotope  | $T_{1/2}$                                | $Q_{EC-B_f}$<br>[MeV] | Production <sup>&amp;</sup> ,<br>Separation,<br>Detection | $P_{\beta DF}$                         | Observables*       | References                           |
|--|--|-----------------------|---|--|--------------------|--------------------------------------|
| <b><math>\beta^+/\text{EC}</math> -delayed fission in the neutron-deficient isotopes</b> |  |                       |   |  |                    |                                      |
| <sup>178</sup> Tl  | 252(20) ms                               | 1.82                  | SR,IS,W/M   | $1.5(6) \times 10^{-3}$                | Z,A,T,KE,TKE,MD,GF | (Liberati <i>et al.</i> , 2013)      |
| <sup>180</sup> Tl  | 1.09(1) s                                | 0.63                  | SR,IS,W/M   | $3.2(2) \times 10^{-5}$                | Z,A,T,KE,TKE,MD,GF | (Elseviers <i>et al.</i> , 2013)     |
|  | 0.97 <sup>+0.09</sup> <sub>-0.08</sub> s |                       | FE,NS,MF  | $\sim 3 \times 10^{-(7\pm 1)}$         | T,EXF              | (Lazarev <i>et al.</i> , 1987, 1992) |
| <sup>186m1,m2</sup> Bi   | 9.8(4), 14.8(8) ms <sup>#</sup>          | 2.09                  | FE,RS,Si/Ge   | $7.6 \times 10^{-2,e}$                 | T,EXF,KE,GF        | (Lane <i>et al.</i> , 2013)          |
| <sup>188m1,m2</sup> Bi   | $\sim 0.3$ s <sup>c</sup>                | 0.51                  | FE,NS,MF  | $3.4 \times 10^{-4,a,c}$               | T,EXF              | (Lazarev <i>et al.</i> , 1992)       |
|  | 265(10), 60(3) ms <sup>#</sup>           |                       | FE,RS,Si/Ge   | $(0.16-0.48) \times 10^{-2,f}$         | T,EXF,KE,GF        | (Lane <i>et al.</i> , 2013)          |
| <sup>192m1,m2</sup> At   | 88(6), 11.5(6) ms <sup>#</sup>           | 2.09                  | FE,RS,Si/Ge   | $(7-35) \times 10^{-2}$                | T,EXF,KE,GF        | (Andreyev <i>et al.</i> , 2013)      |
| <sup>194m1,m2</sup> At   | 310(8), 253(10) ms <sup>#</sup>          | -0.04                 | FE,RS,Si/Ge   | $\sim (0.8-1.6) \times 10^{-2}$        | T,EXF,KE,GF        | (Andreyev <i>et al.</i> , 2013)      |
|  |  |                       | SR,IS,W/M   |  | Z,A,T,KE,TKE,MD,GF | (Andreyev <i>et al.</i> , 2012)      |
| <sup>196</sup> At  | 0.23 <sup>+0.05</sup> <sub>-0.03</sub> s | -1.19                 | FE,NS,MF  | $8.8 \times 10^{-4,a}$                 | T,EXF              | (Lazarev <i>et al.</i> , 1992)       |
|  |  |                       | SR,IS,W/M   |  | Z,A,T,KE,TKE,MD,GF | (Andreyev <i>et al.</i> , 2012)      |
| <sup>200</sup> Fr  | 49(4) ms <sup>#</sup>                    | 0.82                  | SR,IS,W/M   |  | Z,A,T,KE,TKE,MD,GF | (Andreyev <i>et al.</i> , 2011)      |
| <sup>202m1,m2</sup> Fr   | 0.30(5), 0.29(5) s <sup>#</sup>          | -1.17                 | SR,IS,W/M   |  | Z,A,T,KE,TKE,MD,GF | (Andreyev <i>et al.</i> , 2011)      |
| <sup>228</sup> Np  | 61.4(14) s                               | -0.87                 | FE,RC,MG  | $2.0(9) \times 10^{-4}$                | Z,T,KE,TKE,MD,GF   | (Kreek <i>et al.</i> , 1994a)        |
|  | 60(5) s                                  |                       | FE,NS,MF  |  | T,EXF              | (Kuznetsov <i>et al.</i> , 1966)     |
| <sup>232</sup> Am  | 1.31(4) min                              | 1.65                  | FE,RC,MG  | $6.9(10) \times 10^{-4}$               | Z,T,KE,TKE,MD,GF   | (Hall <i>et al.</i> , 1990a)         |
|  | 55(7) s                                  |                       | FE,NS,Si  | $(1.3^{+4}_{-0.8}) \times 10^{-2}$     | T,KE               | (Habs <i>et al.</i> , 1978)          |
|  | 1.40(25) min                             |                       | FE,NS,MF  | $6.96 \times 10^{-2}$                  | T,EXF              | (Kuznetsov <i>et al.</i> , 1967)     |
| <sup>234</sup> Am  | 2.32(8) min                              | 0.29                  | FE,RC,MG  | $6.6(18) \times 10^{-5}$               | Z,T,KE,TKE,MD,GF   | (Hall <i>et al.</i> , 1989a, 1990b)  |
|  | 2.6(2) min                               |                       | FE,NS,MF  | $\sim 6.95 \times 10^{-5}$             | T,EXF              | (Kuznetsov <i>et al.</i> , 1967)     |
| <sup>238</sup> Bk  | 144(5) s                                 | -0.15                 | FE,RC,MG  | $4.8(20) \times 10^{-4}$               | Z,T,KE,TKE,MD,GF   | (Kreek <i>et al.</i> , 1994b)        |
| <sup>240</sup> Bk  | 4.2(8) min                               | -1.99                 | FE,NS,MF  | $(1.3^{+1.8}_{-0.7}) \times 10^{-5}$   | T                  | (Galeriu, 1983)                      |
|  | 5(2) min                                 |                       | FE,NS,MF  | $1 \times 10^{-5,b}$                   | T                  | (Gangrsky <i>et al.</i> , 1980)      |
| <sup>242</sup> Es  | 11(3) s                                  | -0.94                 | FE,RC,MG  | $0.6(2) \times 10^{-2}$                | Z,T,KE,TKE,MD      | (Shaughnessy <i>et al.</i> , 2000)   |
|  | 5-25 s                                   |                       | FE,RS,Si  | $1.4(8) \times 10^{-2}$                | T,KE               | (Hingmann <i>et al.</i> , 1984)      |
|  | 17.8(16) s                               |                       | FE,RS,Si  | $(1.3^{+1.2}_{-0.7}) \times 10^{-2}$   | T,KE               | (Antalic <i>et al.</i> , 2010)       |
| <sup>244</sup> Es  | 38(11) s                                 | -2.24                 | FE,RC,MG  | $1.2(4) \times 10^{-4}$                | Z,T,KE,TKE,MD      | (Shaughnessy <i>et al.</i> , 2002)   |
|  |  |                       | FE,NS,MF  | $1 \times 10^{-4,b}$                   | T                  | (Gangrsky <i>et al.</i> , 1980)      |
| <sup>246</sup> Es  | 7.7(5) min                               | -3.47                 | FE,RC,MG  | $(3.7^{+8.5}_{-3.0}) \times 10^{-5}$   | Z,T,KE             | (Shaughnessy <i>et al.</i> , 2001)   |
|  | 8 min                                    |                       | FE,NS,MF  | $3 \times 10^{-5,b}$                   | T                  | (Gangrsky <i>et al.</i> , 1980)      |
| <sup>248</sup> Es  | 23(3) min                                | -4.26                 | FE,RC,MG  | $3.5(18) \times 10^{-6}$               | Z,T,KE             | (Shaughnessy <i>et al.</i> , 2001)   |
|  |  |                       | FE,NS,MF  | $3 \times 10^{-7,b}$                   | T                  | (Gangrsky <i>et al.</i> , 1980)      |
| <sup>246m1,m2</sup> Md   | 0.9(2), 4.4(8) s                         | 0.14                  | FE,RS,Si  | $> 1 \times 10^{-1}$                   | T,KE               | (Antalic <i>et al.</i> , 2010)       |
|  | 1.0(4) s <sup>c</sup>                    |                       | FE,RS,Si  | $\sim 0.65 \times 10^{-1}$             | T,KE               | (Ninov <i>et al.</i> , 1996)         |
| <sup>250</sup> Md  | 52(6) s <sup>#</sup>                     | -2.64                 | FE,NS,MF  | $2 \times 10^{-4,b}$                   | T                  | (Gangrsky <i>et al.</i> , 1980)      |
| <b><math>\beta^-</math> -delayed fission in the neutron-rich isotopes</b>                |  |                       |   |  |                    |                                      |
| <sup>228</sup> Ac  | 6.15(2) h <sup>#</sup>                   | -4.45                 | LLP,RC,MF/Ge  | $5(2) \times 10^{-12}$                 |                    | (Yanbing <i>et al.</i> , 2006)       |
| <sup>230</sup> Ac  | 122(3) s <sup>#</sup>                    | -2.73                 | TR,RC,MF/Ge   | $1.19(40) \times 10^{-8}$              |                    | (Shuanggui <i>et al.</i> , 2001)     |
| <sup>256m</sup> Es   | 7.6 h <sup>#</sup>                       | -3.23                 | TR,RC,Si/Ge   | $2 \times 10^{-5}$                     | T,KE               | (Hall <i>et al.</i> , 1989b)         |
| <sup>234gs</sup> Pa  | 6.70(5) h <sup>#</sup>                   | -2.55                 | NI,NS,MF  | $3 \times 10^{-12,d}$                  | T                  | (Gangrsky <i>et al.</i> , 1978)      |
| <sup>234m</sup> Pa   | 1.159(11) min <sup>#</sup>               |                       | LLP,RC,MF   | $10^{-12,d}$                           | T                  | (Gangrsky <i>et al.</i> , 1978)      |
| <sup>236</sup> Pa  | 9.1(1) min <sup>#</sup>                  | -2.02                 | SR,RC,MF/Ge   | $\sim 10^{-9}$                         | T                  | (Batist <i>et al.</i> , 1977)        |
|  |  |                       | FE/GL,NS,MF   | $10^{-9,d}/3 \times 10^{-10,d}$        | T                  | (Gangrsky <i>et al.</i> , 1978)      |
| <sup>238</sup> Pa  | 2.3(1) min <sup>#</sup>                  | -2.14                 | NI,NS,MF  | $6 \times 10^{-7}, 1 \times 10^{-8,d}$ | T                  | (Gangrsky <i>et al.</i> , 1978)      |
|  |  |                       | NI,RC,MF  | $< 2.6 \times 10^{-8}$                 |                    | (Baas-May <i>et al.</i> , 1985)      |

**2013: <sup>230</sup>Am at GARIS(RIKEN)**

# Mapping 'Terra Incognita' in Low-Energy Fission



A. N. Andreyev, M. Huyse, P. Van Duppen, "Beta-delayed Fission in atomic nuclei",  
 Reviews of Modern Physics, 85, 1541 (2013)

**Thank you!**

**Strong support from Reimei Program of ASRC (JAEA) is very much appreciated**



# Beta-delayed fission in the neutron-rich Fr nuclei?

## Earlier Gatchina attempts

| Isotope             | $T_{1/2}$                | $Q_{EC} - B_f$<br>[MeV] | Production<br>Separation,<br>Detection | $P_{\beta DF}$<br>Upper Limit |
|---------------------|--------------------------|-------------------------|--|-------------------------------|
| $^{242}\text{Bk}^a$ | 7.0(13) min <sup>#</sup> | -3.49                   | FE,NS,MF                               | $< 3 \times 10^{-7}$          |
| $^{248}\text{Md}^a$ | 7(3) s <sup>#</sup>      | -1.45                   | FE,NS,MF                               | $< 5 \times 10^{-4}$          |
| $^{228}\text{Fr}^b$ | 38(1) s <sup>#</sup>     | -3.33                   | SR,IS,Si/Ge                            | $< 2 \times 10^{-7}$          |
| $^{230}\text{Fr}^b$ | 19.1(5) s <sup>#</sup>   | -2.05                   | SR,IS,Si/Ge                            | $< 3 \times 10^{-6}$          |
| $^{232}\text{Fr}^b$ | 5.5(6) s                 | -1.34                   | SR,IS,Si/Ge                            | $< 7 \times 10^{-4, c}$       |
| $^{232}\text{Ac}^b$ | 119(5) s                 | -1.75                   | SR,IS,Si                               | $< 10^{-6}$                   |

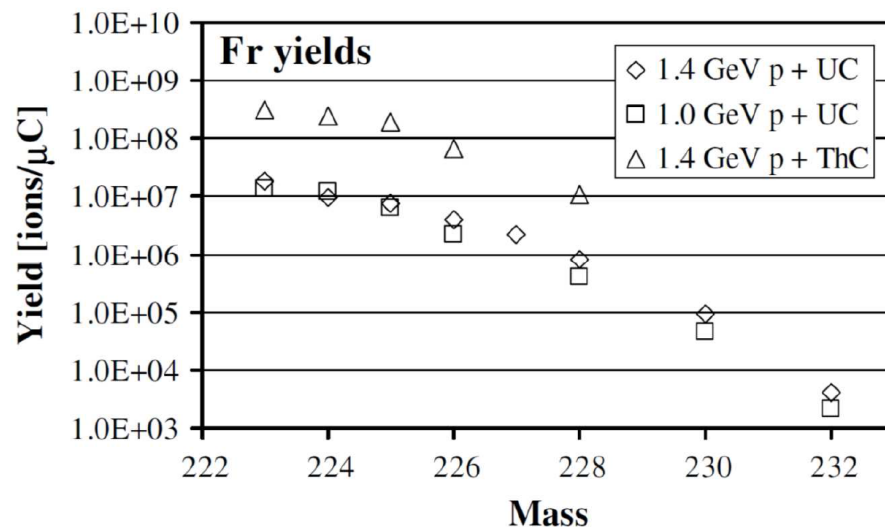
<sup>#</sup> Evaluated half-life value from (ENSDF, 2013).

a) Studied by (Gangrsky *et al.*, 1978).

b) Studied by (Mezilev *et al.*, 1990).

c) Different limits for different  $\beta$ - $\gamma$  transitions.

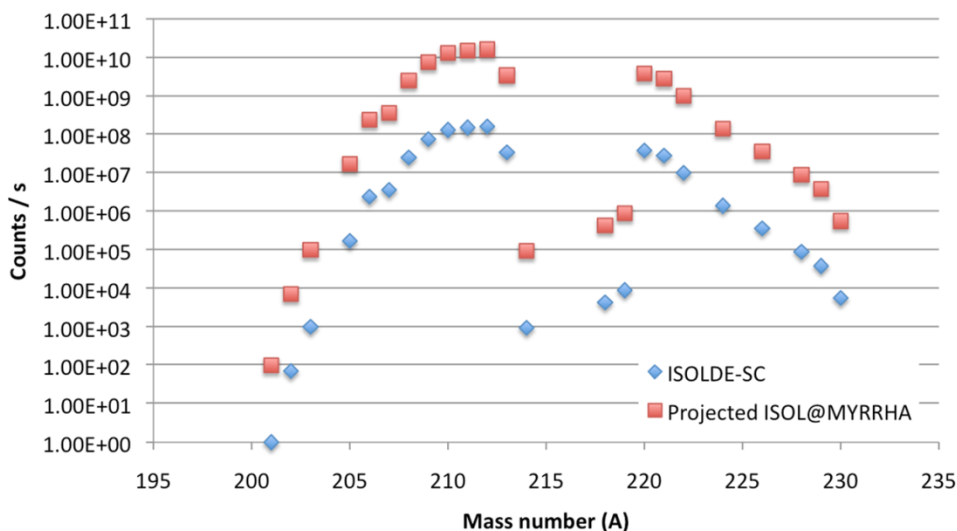
## ISOLDE yields (UC, ThC)



## ThC targets?

$\sim 10^{-8}$  branching in 1 day now

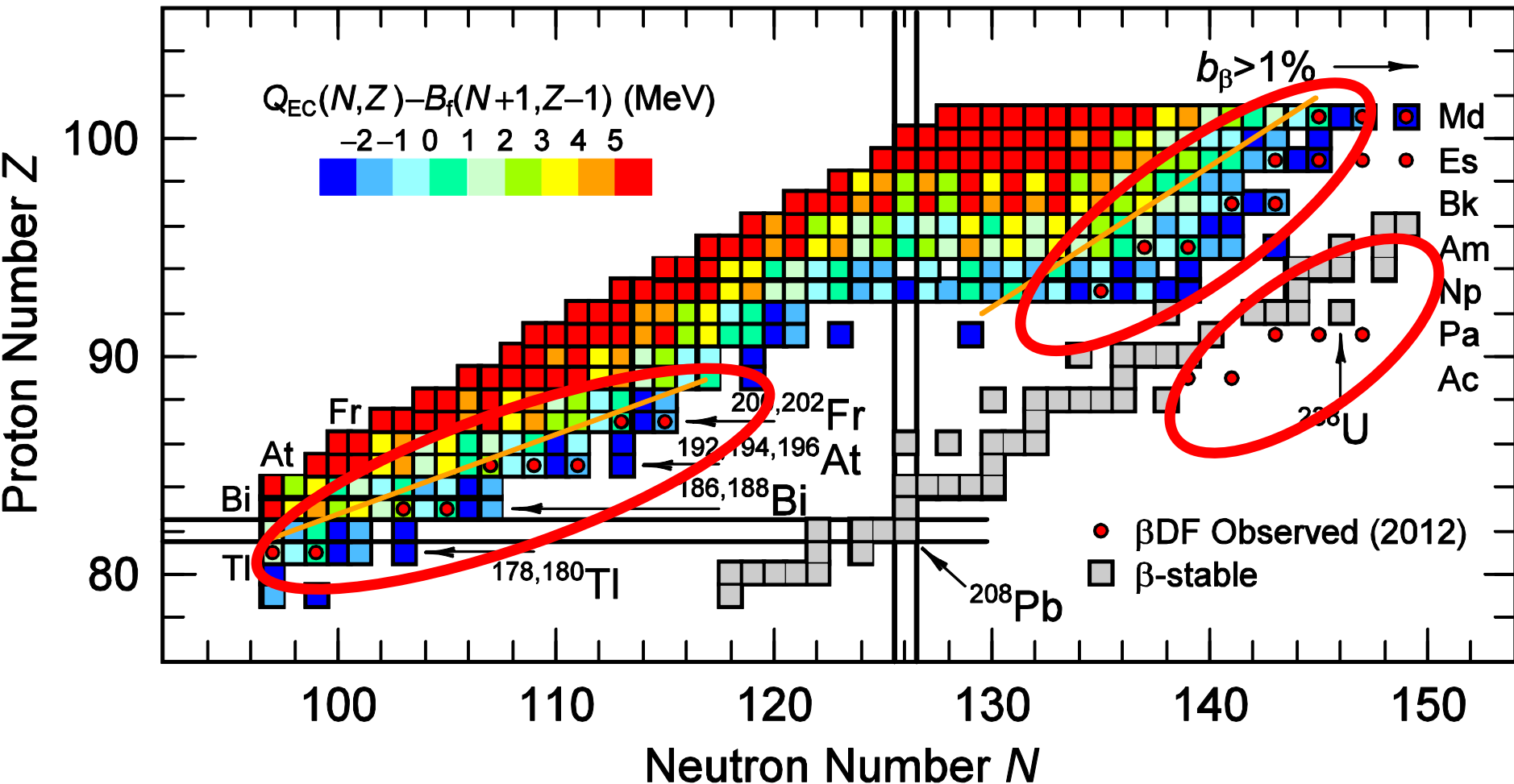
### Fr yields



Beta-delayed fission studies at  
recoil separators  
(e.g. at Lanzhou?)

# Why recoil separators to search for $\beta$ DF?

- Unlike lead region, heavy actinides cannot be accessed by projectile fragmentation (as at ISOLDE)
- The only method at present: complete-fusion reactions

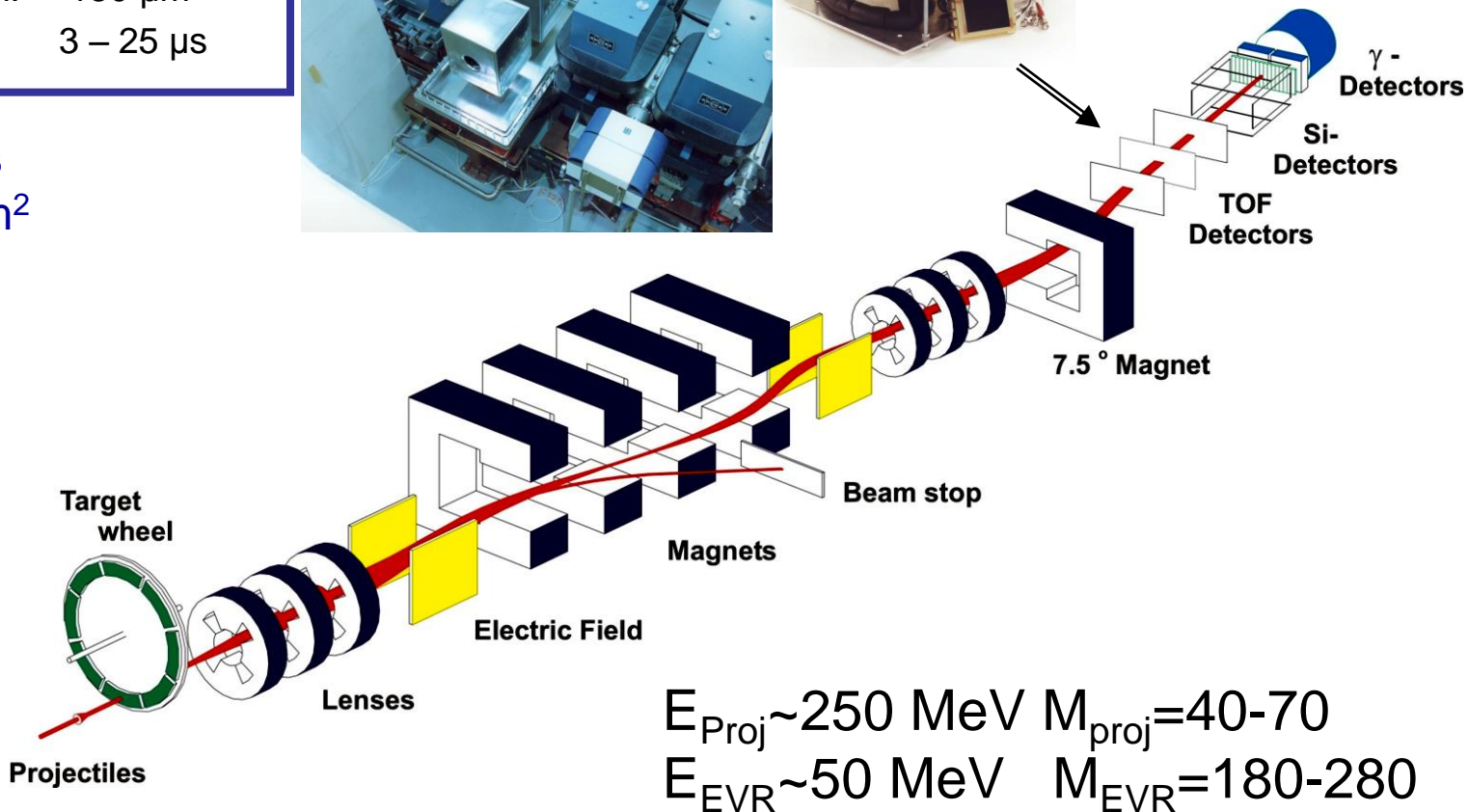
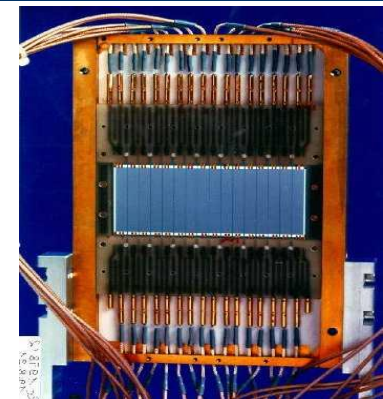
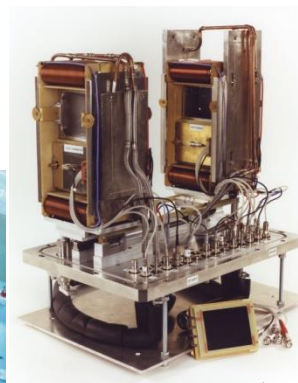
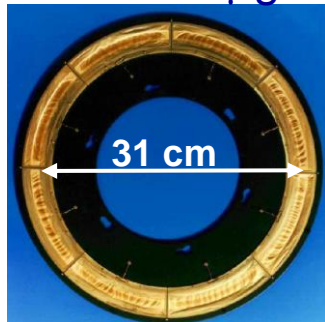


# An example: $\beta$ DF of $^{194}\text{At}$ at SHIP (GSI, Darmstadt)

## SHIP:

|                       |                      |
|-----------------------|----------------------|
| Separation time:      | 1 – 2 $\mu\text{s}$  |
| Transmission:         | 20 – 50 %            |
| Background:           | 10 – 50 Hz           |
| Det. E. resolution:   | 18 – 25 keV          |
| Det. Pos. resolution: | 150 $\mu\text{m}$    |
| Dead time:            | 3 – 25 $\mu\text{s}$ |

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

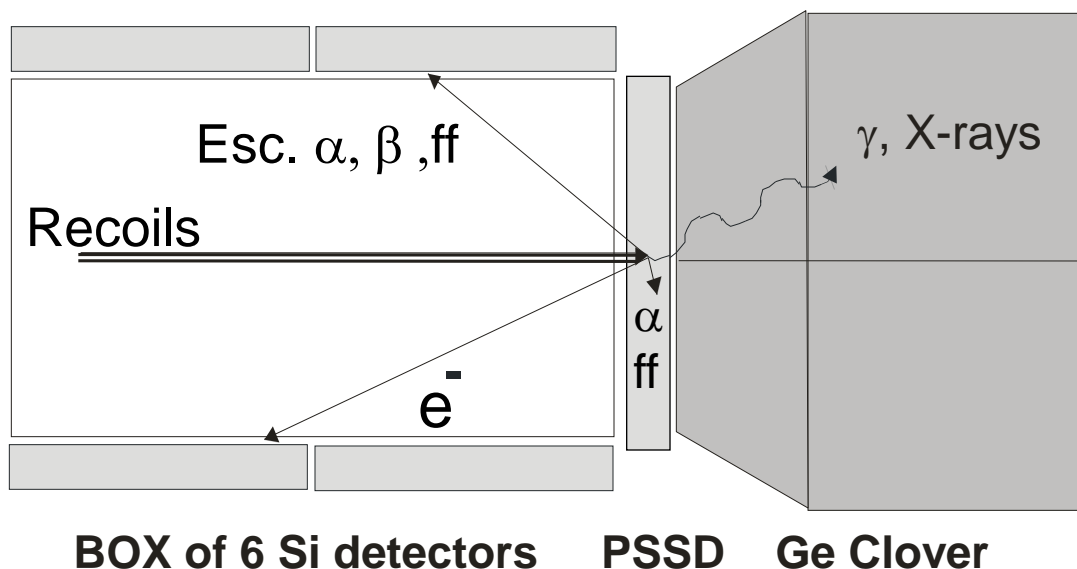
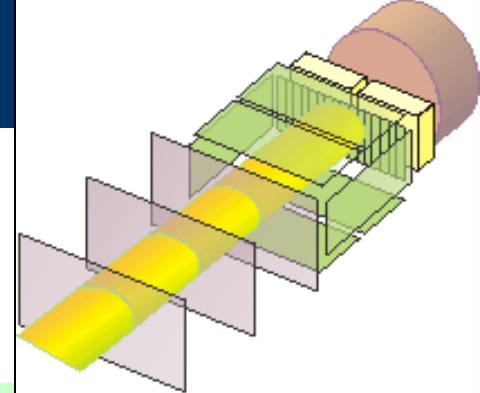


1  $\mu\text{A}$  of  $^{52}\text{Cr}, ^{58}\text{Ni}$   
 $\sim 6 \times 10^{12}$  pps

# SHIP Detection System

Measure **efficiently** all possible decays:

- particle decay ( $\alpha$ ,  $\beta$ , 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 (35×80mm), pos. resolution FWHM=150  $\mu$ m, energy resolution 14 keV

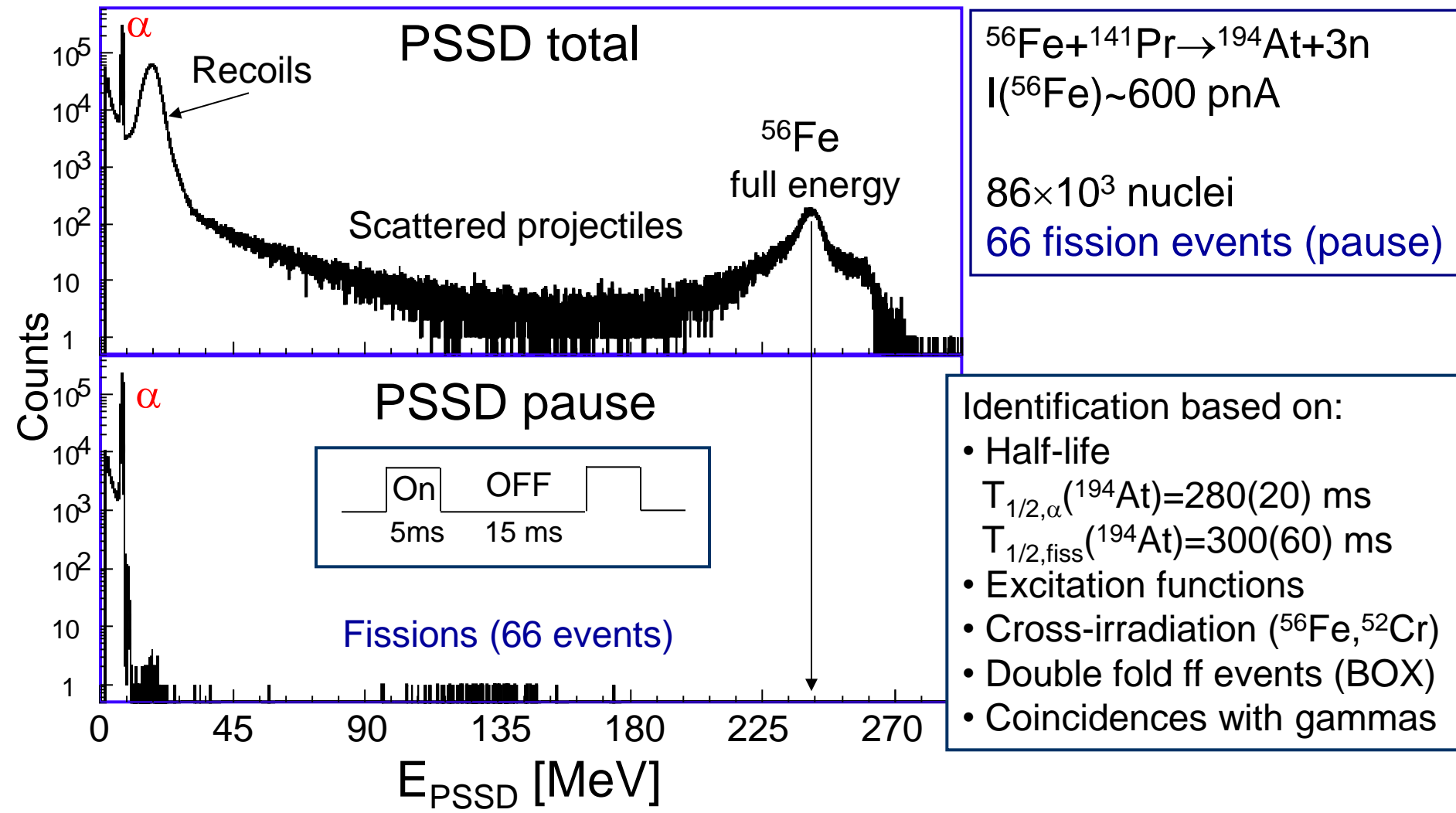
• **6 BOX Si detectors** – for  $\beta$  and escaping  $\alpha$  particles with a solid angle 80% of  $2\pi$

• **GAMMA detectors** – large-volume Clover detector for x rays or  $\gamma$  rays in coincidence with  $\alpha$ 's

• **VETO detector** – reduces background

# Identification of $\beta$ DF in $^{194}\text{At}$ , SHIP(GSI)

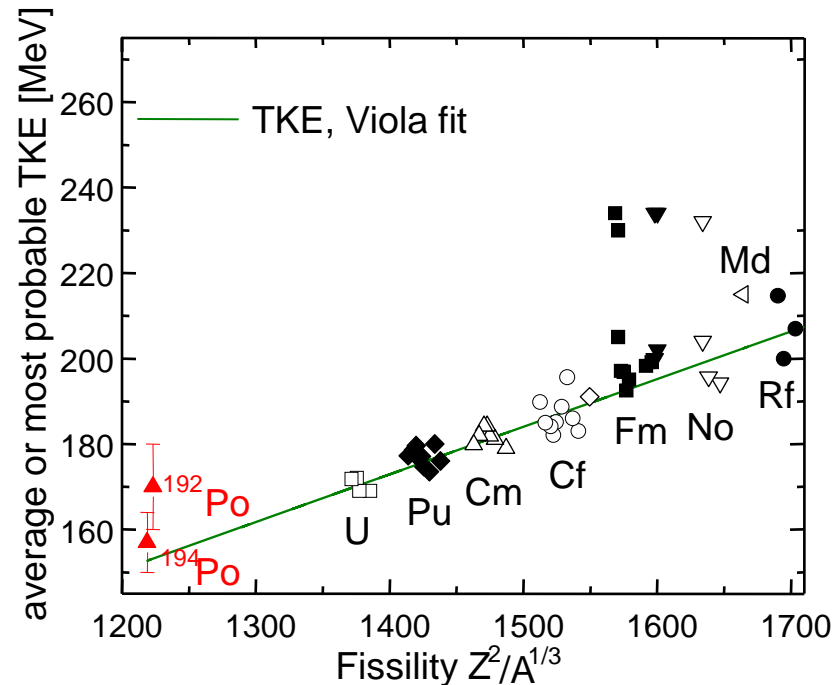
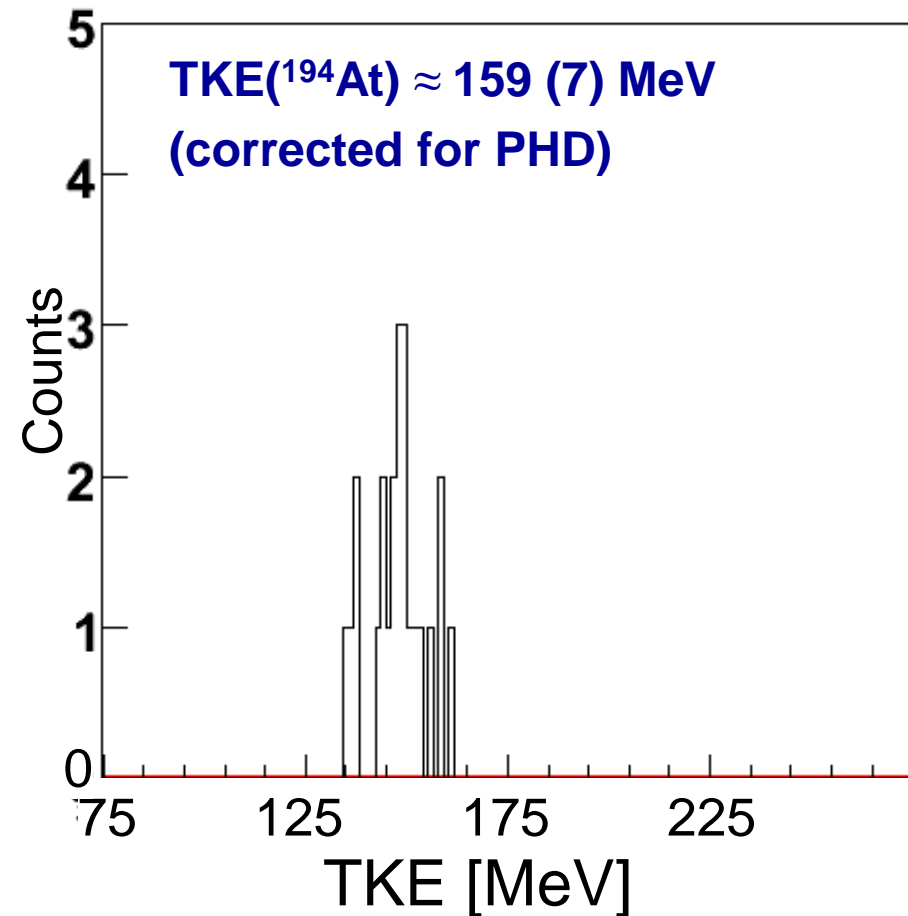
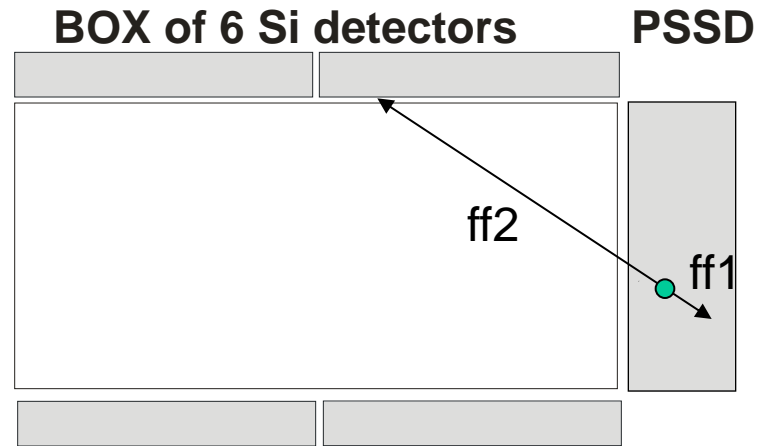
A. Andreyev et al, PRC, 87, 014317 (2013)

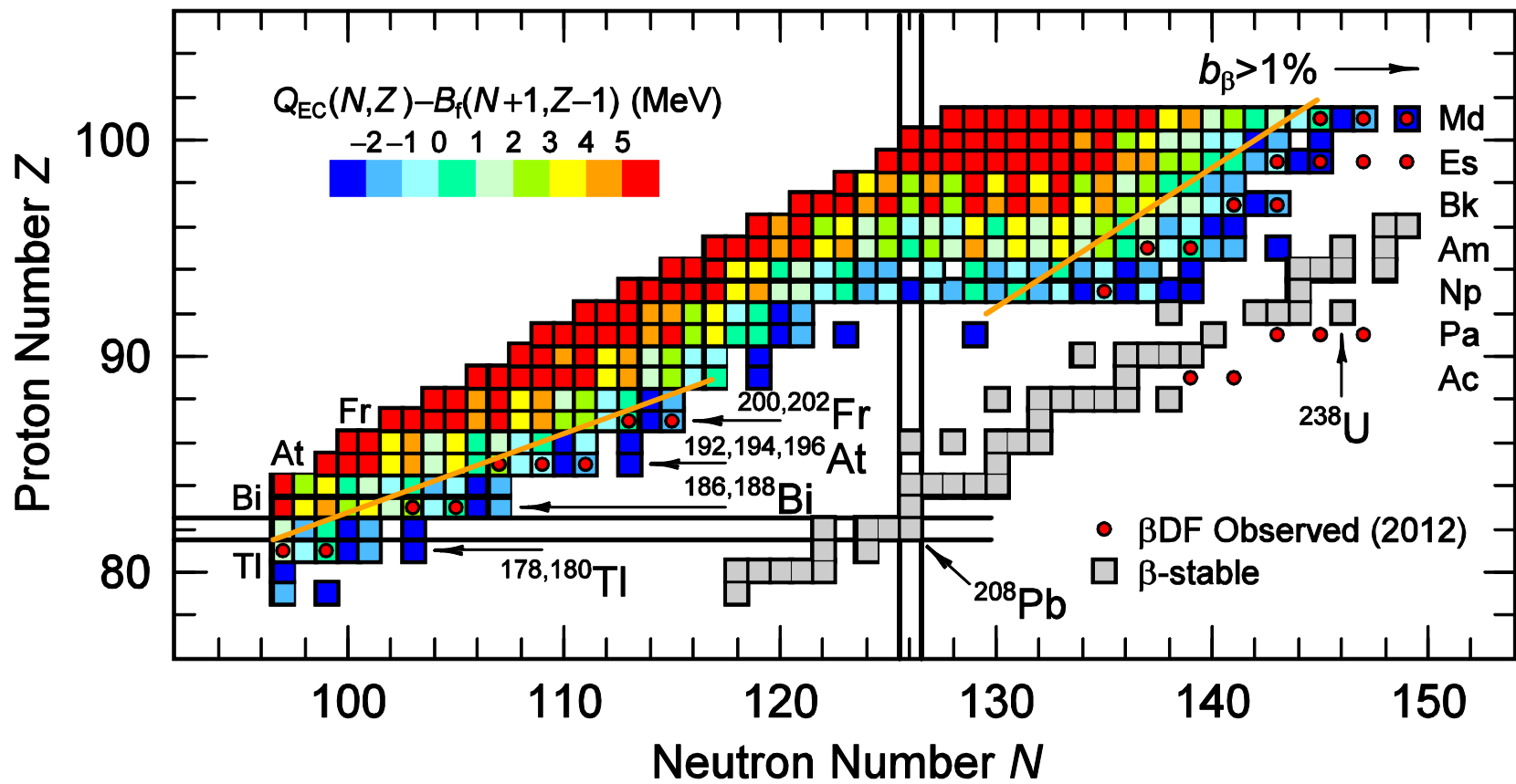


First observed in the  $^{52}\text{Cr} + ^{144}\text{Sm} \rightarrow ^{194}\text{At} + pn$  reaction (SHIP, 2006)  
16 fissions in pause

# Total Kinetic Energy in $\beta$ DF of $^{194}\text{At}$

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







# 'Brownian Metropolis Shape Motion'

based on J. Randrup and P. Moller, PRL 106, 132503 (2011)

Phys. Rev. C 85, 024306 (2012)

## Calculated fission yields of neutron-deficient mercury isotopes

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<sup>2</sup>Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

(Dated: November 21, 2011)

The recent unexpected discovery of asymmetric fission of  $^{180}\text{Hg}$  following the electron-capture decay of  $^{180}\text{Tl}$  has led to intense interest in experimentally mapping the fission-yield properties over more extended regions of the nuclear chart and compound-system energies. We present here a first calculation of fission-fragment yields for neutron-deficient Hg isotopes, using the recently developed Brownian Metropolis shape motion treatment. The results for  $^{180}\text{Hg}$  are in approximate agreement with the experimental data. For  $^{174}\text{Hg}$  the symmetric yield increases strongly with decreasing energy, an unusual feature, which would be interesting to verify experimentally.

PACS numbers: 25.85.-w, 24.10.Lx, 24.75.+i

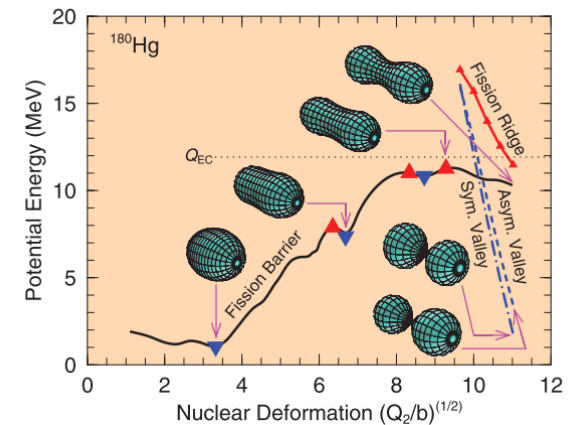
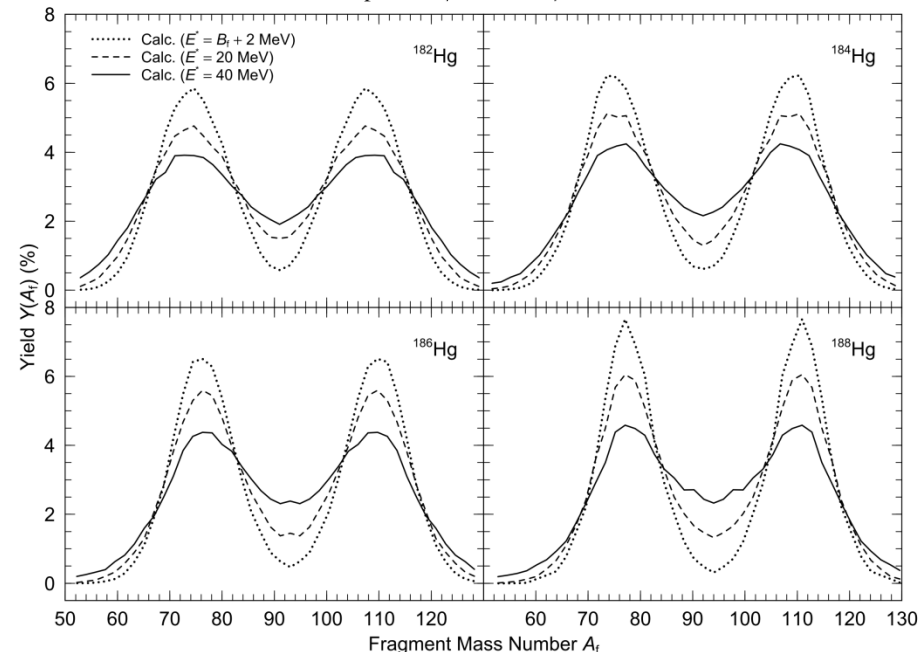
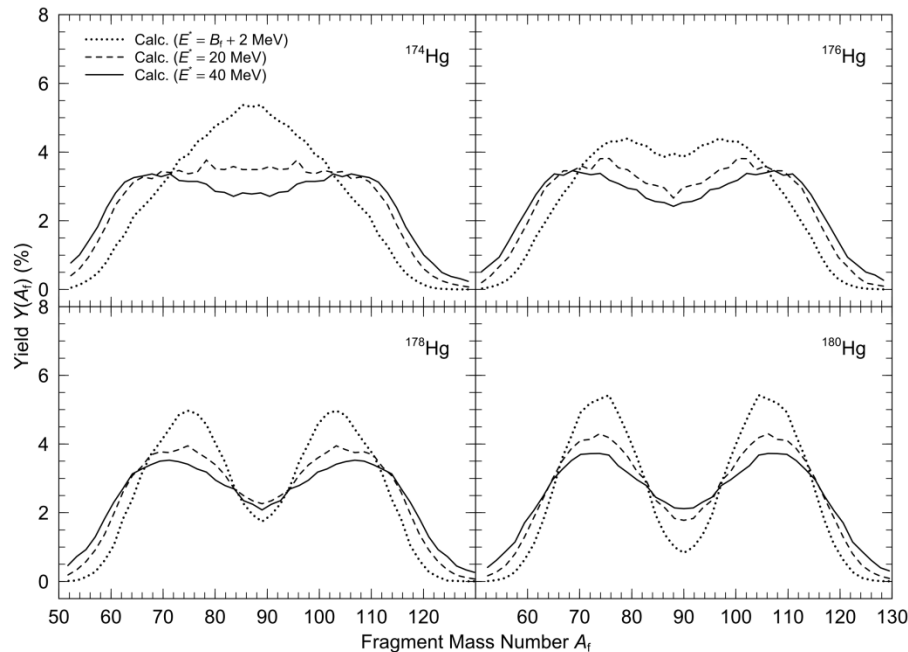


FIG. 4. (Color online) Minima, saddles, major valleys, and ridges in the 5D potential-energy surface of  $^{180}\text{Hg}$  (see text). At the last plotted point on the fission barrier,  $(Q_2/b)^{(1/2)} \approx 11$ , the asymmetry of the shape is  $A_H/A_L = 108/72$ .



# 'Improved Scission-Point Model'

PHYSICAL REVIEW C **86**, 044315 (2012)

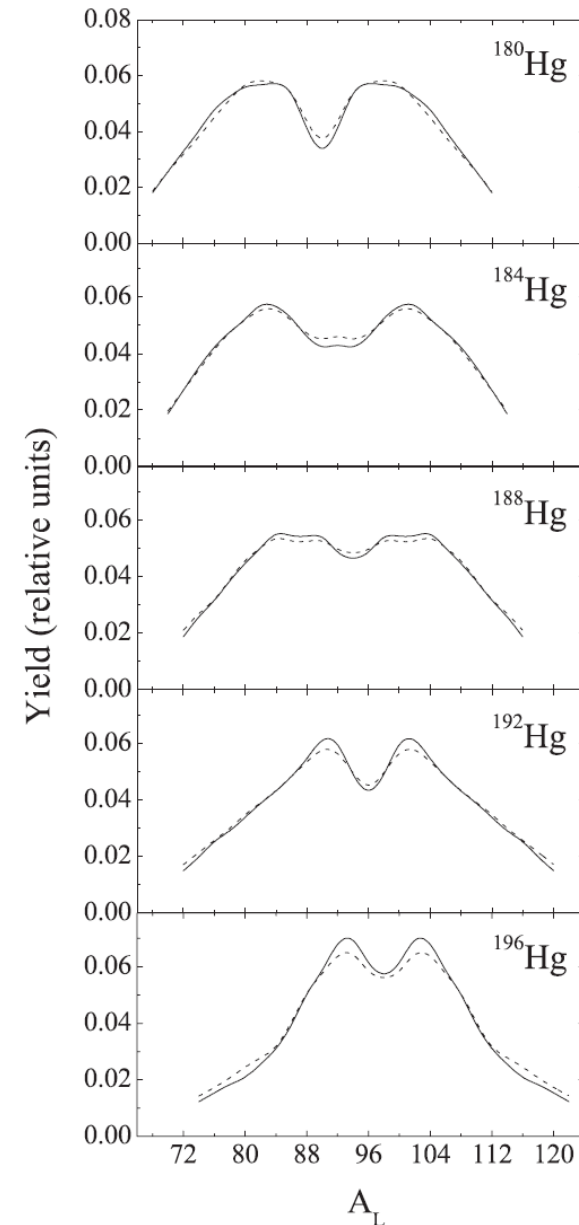
## Mass distributions for induced fission of different Hg isotopes

A. V. Andreev, G. G. Adamian, and N. V. Antonenko  
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(Received 20 June 2012; revised manuscript received 6 September 2012; published 11 October 2012)

With the improved scission-point model mass distributions are calculated for induced fission of different Hg isotopes with even mass numbers  $A = 180, 184, 188, 192, 196$ , and 198. The calculated mass distribution and mean total kinetic energy of fission fragments are in good agreement with the existing experimental data. The asymmetric mass distribution of fission fragments of  $^{180}\text{Hg}$  observed in the recent experiment is explained. The change in the shape of the mass distribution from asymmetric to more symmetric is revealed with increasing  $A$  of the fissioning  $^A\text{Hg}$  nucleus, and reactions are proposed to verify this prediction experimentally.

- Inter-fragment distance is not fixed and calculated.
- values of  $\sim 0.5$ -1 fm result (Wilkins – fixed at 1.4 fm)
- Mass symmetry/asymmetry doesn't change as a function of  $E^*$  (up to  $E^* \sim 60$  MeV) – good for future experiments



Also, J.-L. Sida et al. – private communication

# 'Mean-field HFB+Gogny D1S'

PHYSICAL REVIEW C **86**, 024601 (2012)

## Fission modes of mercury isotopes

M. Warda,<sup>1</sup> A. Staszczak,<sup>1,2,3</sup> and W. Nazarewicz<sup>2,3,4</sup>

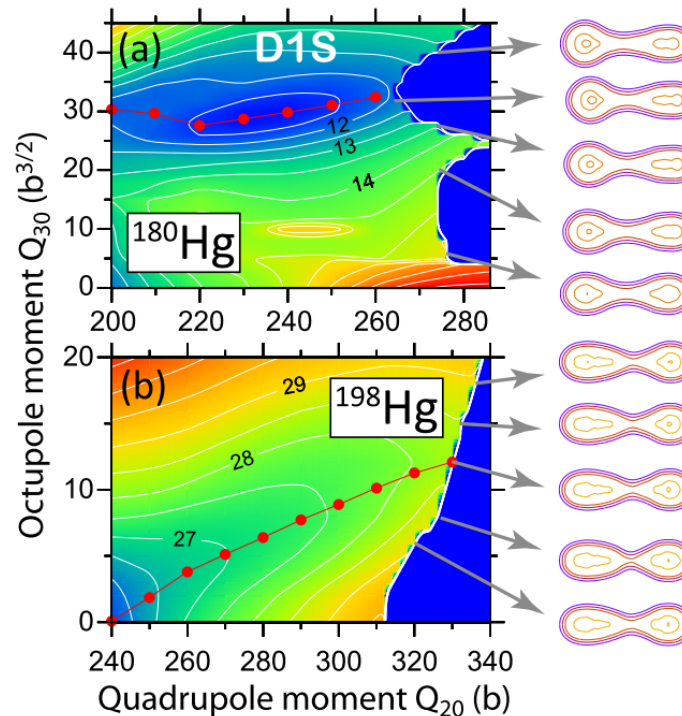
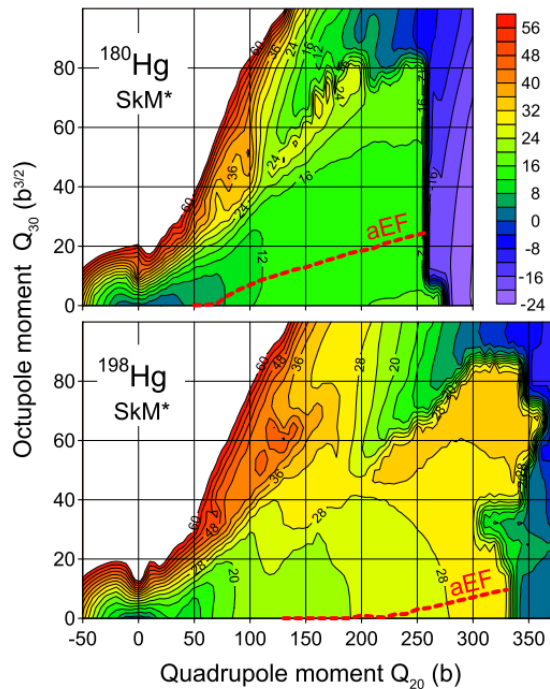
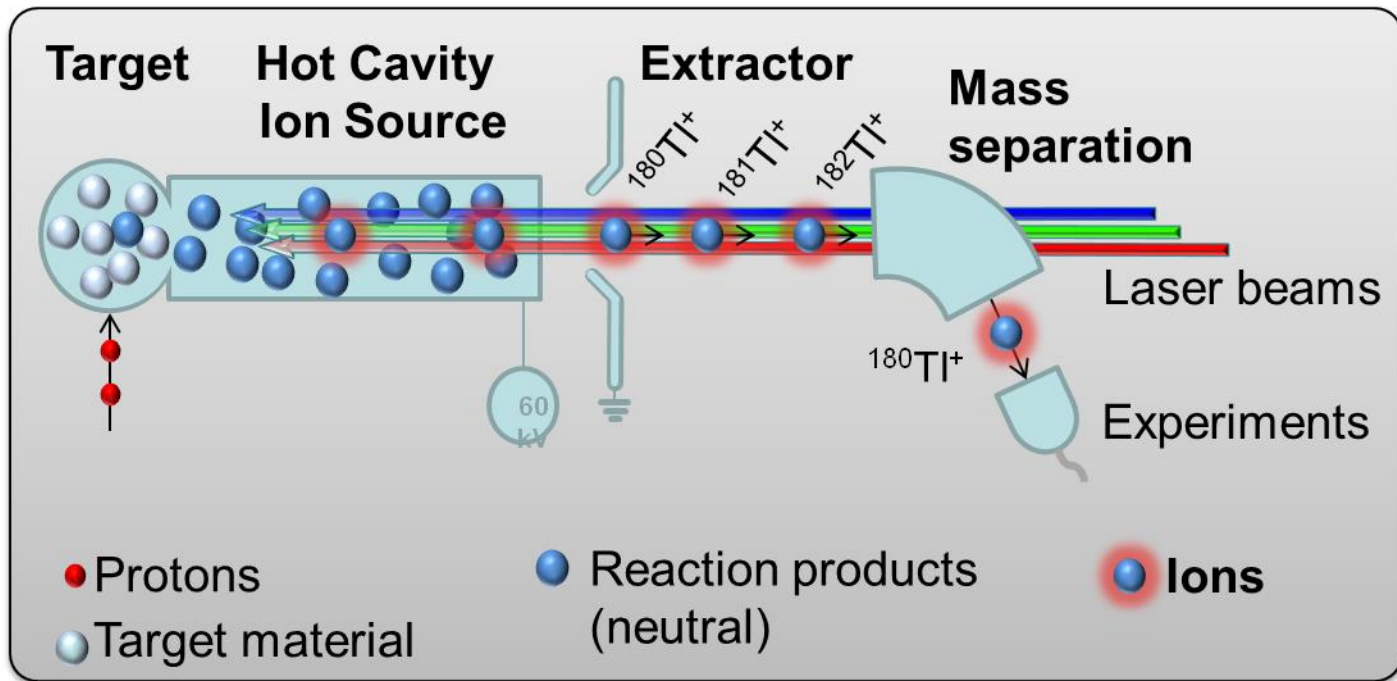


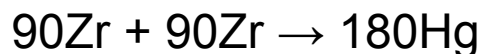
FIG. 2. (Color online) PES for  $^{180}\text{Hg}$  (top) and  $^{198}\text{Hg}$  (bottom) in the plane of collective coordinates  $Q_{20} - Q_{30}$  in HFB-SkM\*. The aEF fission pathway corresponding to asymmetric elongated fragments is marked. The difference between contour lines is 4 MeV. The effects due to triaxiality, known to impact inner fission barriers in the actinides, are negligible here.

FIG. 3. (Color online) PES in HFB-D1S for  $^{180}\text{Hg}$  (top) and  $^{198}\text{Hg}$  (bottom) in the  $(Q_{20}, Q_{30})$  plane in the pre-scission region of aEF valley. The symmetric limit corresponds to  $Q_{30} = 0$ . The aEF valley and density profiles for pre-scission configurations are indicated. The difference between contour lines is 0.5 MeV. Note different  $Q_{30}$ -scales in  $^{180}\text{Hg}$  and  $^{198}\text{Hg}$  plots.

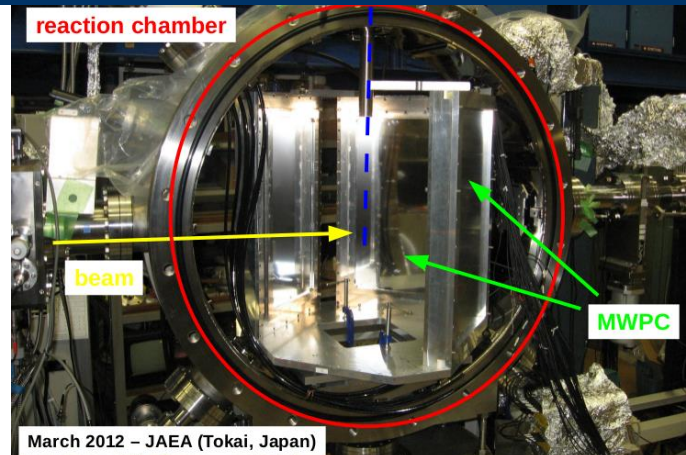


# Fusion-Fission Reactions in the Lead Region at JAEA's tandem (K.Nishio et al)

New experiment at JAEA (March-April 2012)

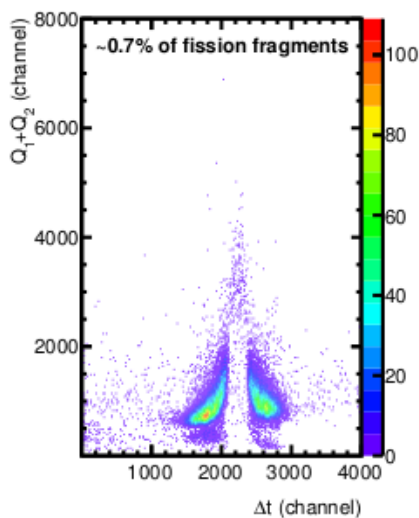


Ebeam from 160 to 235 MeV

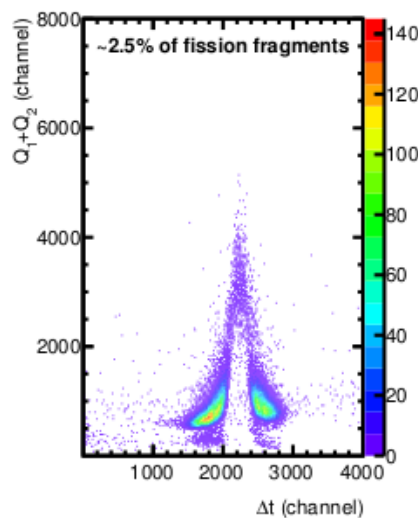


New reaction chamber, larger MWPC  
new electronics (time-stamping)

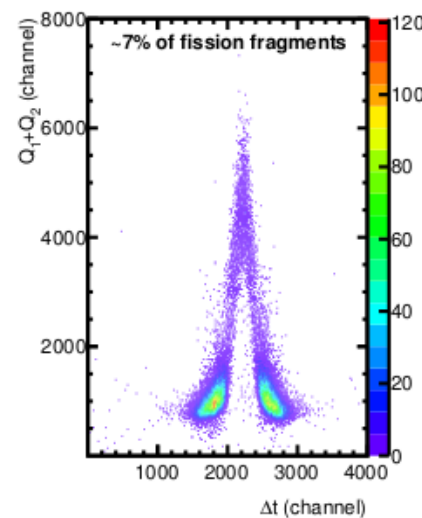
40Ar + 144Sm @ 180 MeV



40Ar + 144Sm @ 210 MeV



40Ar + 144Sm @ 235 MeV



Analysis in progress

# How exotic is $\beta$ DF?

## Neutron-deficient side:

Beta-delayed p, d, t,  $\alpha$ .. ,  $\beta$ 2p...emission ~160 cases  
M. Borge and B.Blank (2008)

## Neutron-rich side:

Beta-delayed neutron emission.. ,  $\beta$ 2n.. ~217 cases  
B. Pfeiffer et al., Prog. Nucl. Ener. 41 (2002) 39

## Beta-delayed fission:

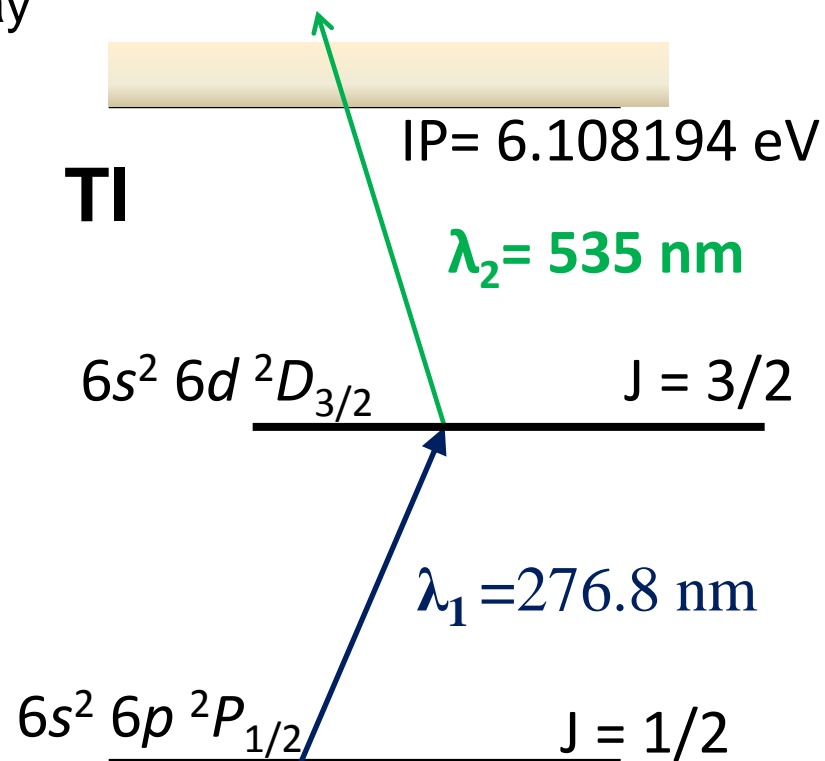
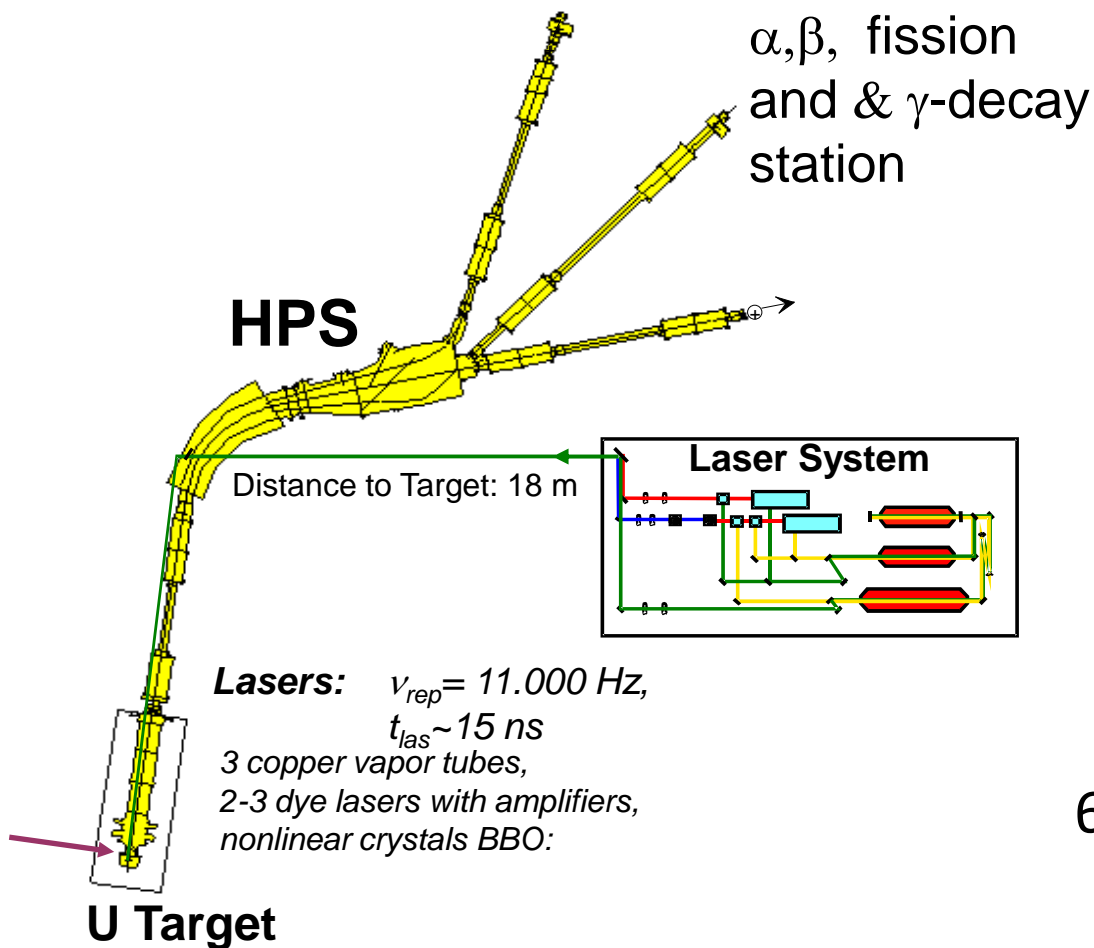
### neutron-deficient side:

12 cases before our studies, U region (up to ~2008)  
9 new cases in the Pb region (from ~2008 on)

several nuclei on the neutron-rich side (data unclear)

# RILIS: Resonance Ionization Laser Ion Source at ISOLDE

Procedure: decay measurements ( $\alpha$ , fission,  $\beta$ - $\gamma$  decays) of TI ions, resonantly ionized by RILIS and mass-separated by HRS@ISOLDE

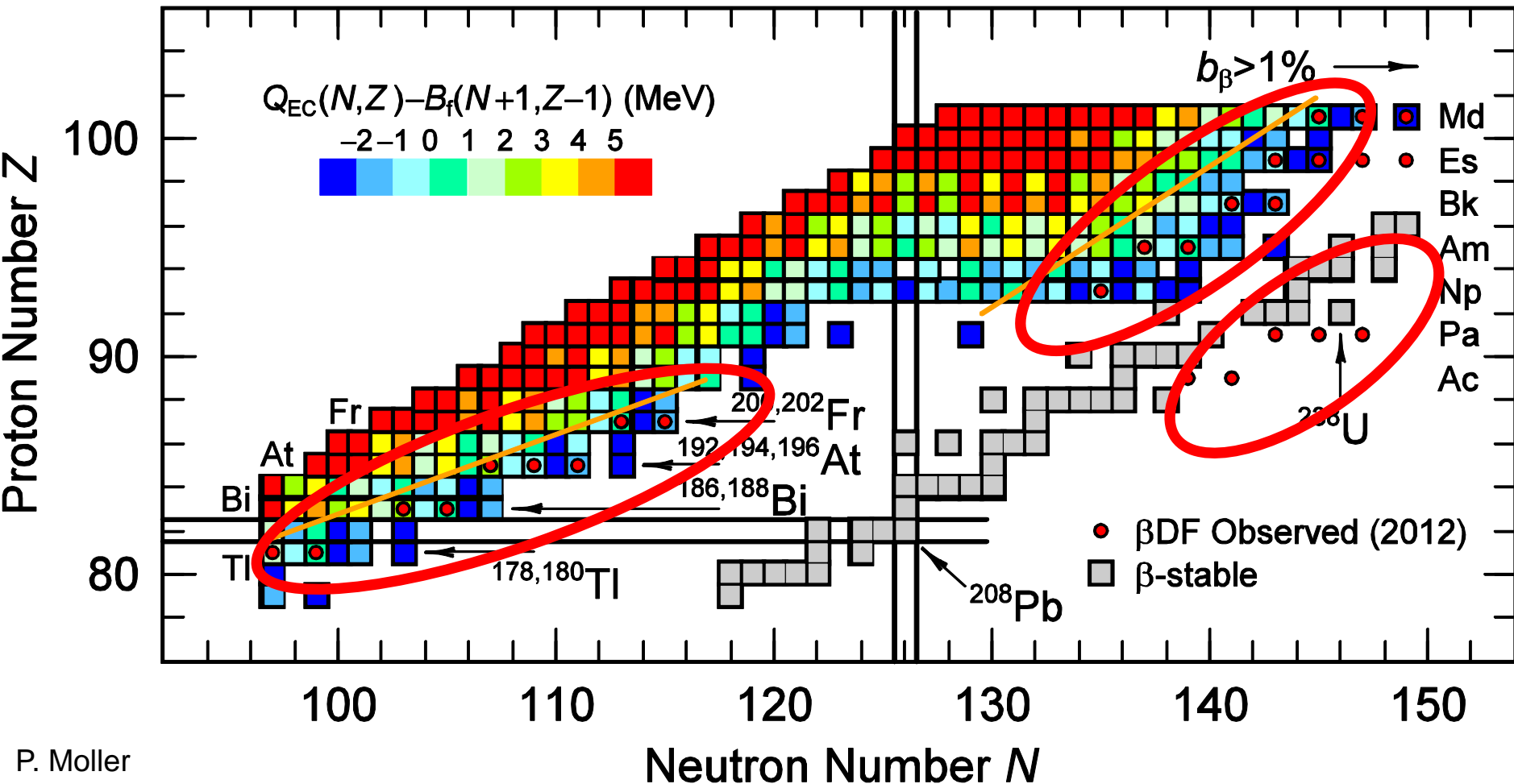


**Proton Beam 1.4 GeV,  $3 \cdot 10^{13}$  pps**  
2.4  $\mu$ s pulse length, 1.2 s period

# Three regions to search for $\beta$ DF

## Necessary conditions for $\beta$ DF to occur:

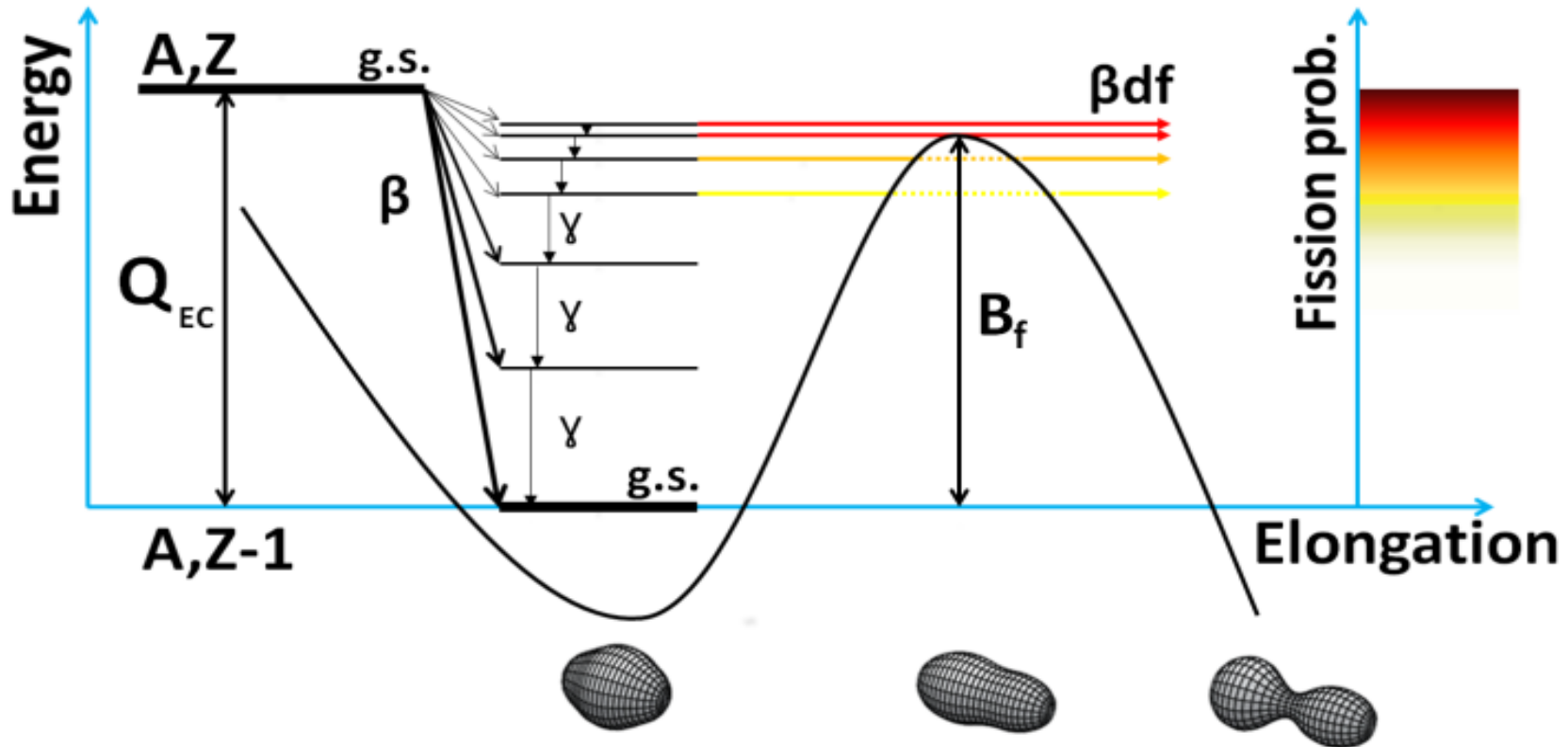
- $Q_{EC}(\text{Parent}) \sim B_f(\text{Daughter})$  [ $Q_{EC} - B_f > -2 \text{ MeV}$ ]
- Beta-branching ratio  $b_\beta > 0$





# Beta-Delayed Fission

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



$\beta$ DF branch

$$P_{\beta DF} = \frac{N_{\beta DF}}{N_{\beta}}$$

# $P_{\beta\text{DF}}$ Probability: Extraction of fission barriers?!

$$P_{\beta\text{df}} = \frac{\int_0^{Q_\beta} F(Q_\beta - E) S_\beta(E) \frac{\Gamma_f(E)}{\Gamma_f(E) + \Gamma_\gamma(E)} dE}{\int_0^{Q_\beta} F(Q_\beta - E) S_\beta(E) dE}$$

**Need to know  $S_\beta$ !?**  
**(next slide)**

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

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

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

D.L. Hill and J.A. Wheeler

**Measurement of  $P_{\beta\text{DF}}$  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

# $P_{\beta\text{DF}}$ Probability: Extraction of fission barriers?!

PHYSICAL REVIEW C 86, 024308 (2012)

## Fission-barrier heights of neutron-deficient mercury nuclei

M. Veselský,<sup>1,\*</sup> A. N. Andreyev,<sup>2</sup> S. Antalic,<sup>3</sup> M. Huyse,<sup>4</sup> P. Möller,<sup>5</sup> K. Nishio,<sup>6</sup> A. J. Sierk,<sup>5</sup>  
P. Van Duppen,<sup>4</sup> and M. Venhart<sup>1,4</sup>

$$P_{\beta\text{df}} = \frac{\int_0^{Q_\beta} F(Q_\beta - E) S_\beta(E) \frac{\Gamma_f(E)}{\Gamma_f(E) + \Gamma_\gamma(E)} dE}{\int_0^{Q_\beta} F(Q_\beta - E) S_\beta(E) dE}$$

(a)  $^{180}\text{Hg}$

(b)  $^{178}\text{Hg}$

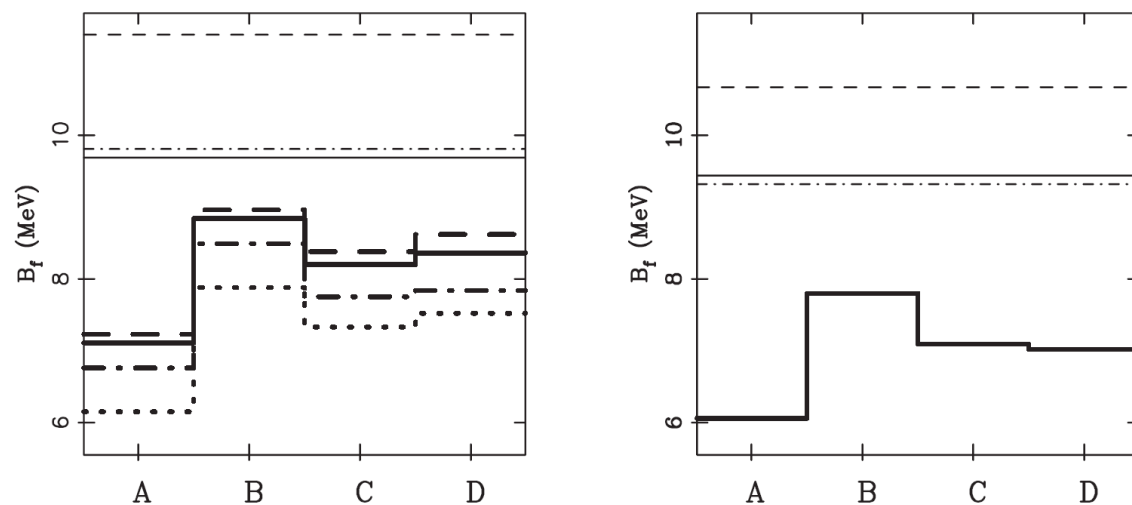
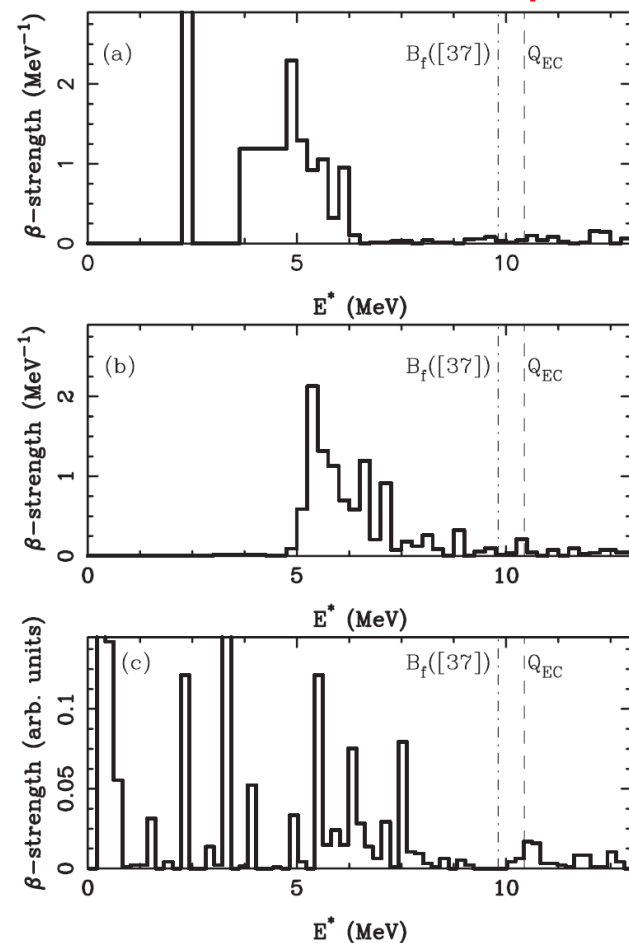


FIG. 3. (Color online) (a) Fission-barrier heights of  $^{180}\text{Hg}$  in four variants A–D. Four  $\beta$ -strength functions were used: calculated

**Need to know  $S_\beta$ !?**



**Clearly – model-dependent (but we tried many parametrizations)  
Conclusion: “experimental barriers” are always lower than calculated**

# Development of At beams

**The Problem:** Element At has no stable isotopes,  $T_{1/2}=8$  h ( $^{210}\text{At}$ )  
Only ~70 mg of At present in the 1<sup>st</sup> mile of Earth' core  
Must be produced and studied 'on-line' at an accelerator

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN-INTC-2010-010  
INTC-I-086

Letter of Intent to the ISOLDE and N-ToF Experiments Committee (INTC)

## Development of astatine ion beams with RILIS

A. Andreyev<sup>1</sup>, S. Antalic<sup>2</sup>, L.-E. Berg<sup>3</sup>, T. Cocolios<sup>4</sup>, V.N. Fedosseev<sup>5</sup>, T. Gottwald<sup>6</sup>,  
M. Huyse<sup>4</sup>, Yu. Kudryavtsev<sup>4</sup>, U. Köster<sup>7</sup>, W. Kurcewicz<sup>8</sup>, J. Lassen<sup>9</sup>, H. Mach<sup>10</sup>,  
B.A. Marsh<sup>5,11</sup>, R. Page<sup>12</sup>, D. Pauwels<sup>4</sup>, S. Räder, S. Rothe<sup>5,6</sup>, A.M. Sjödin<sup>5</sup>, T. Stora<sup>5</sup>,  
P. Van Duppen<sup>4</sup>, K. Wendt<sup>6</sup>

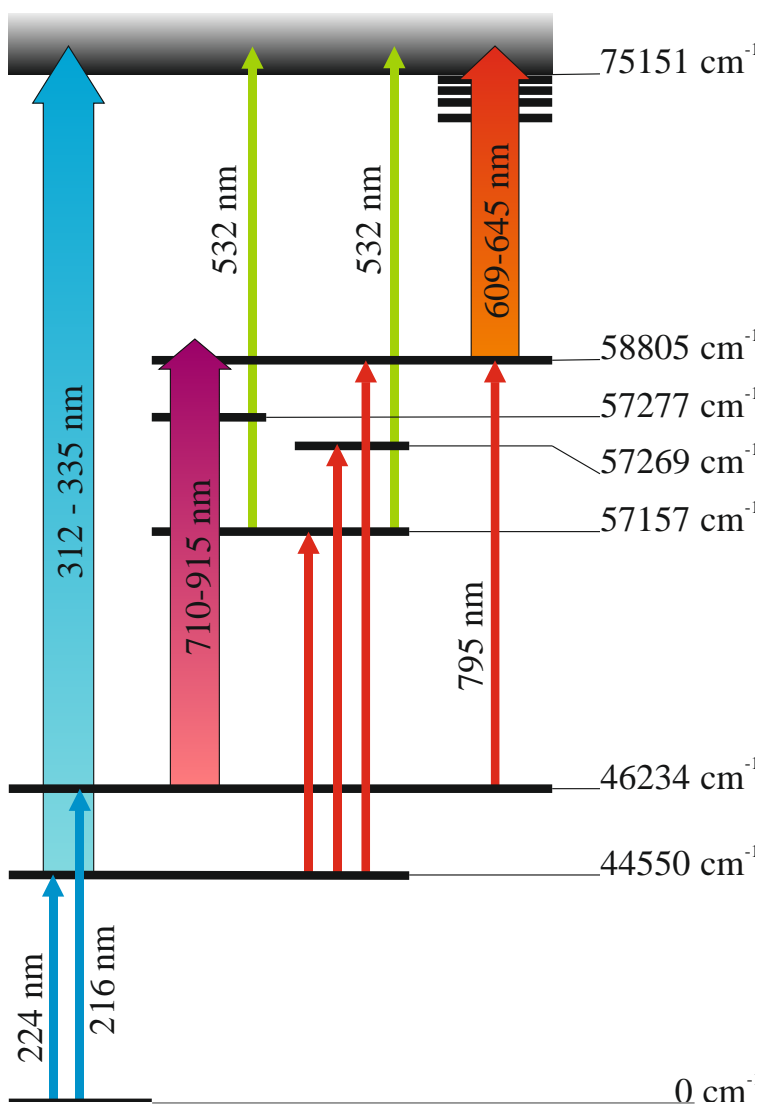
- 1: University of the West of Scotland, Paisley, UK
- 2: Comenius University, Bratislava, Slovakia
- 3: Royal Institute of Technology (KTH), Stockholm, Sweden
- 4: IKS, KU Leuven, Belgium
- 5: CERN, Geneva, Switzerland
- 6: Institut für Physik, Mainz University, Mainz, Germany
- 7: ILL, Grenoble, France
- 8: University of Warsaw, Poland
- 9: TRIUMF, Vancouver, Canada
- 10: Department of Nuclear and Particle Physics, Uppsala University, Sweden
- 11: The University of Manchester, Manchester, UK
- 12: University of Liverpool, UK

Spokespersons: A. Andreyev, V.N. Fedosseev  
Contact person: S. Rothe

**2010: Letter of Intent (CERN):**  
“Development of astatine ion  
beams with Resonance Ionization  
Laser Ion Source (RILIS)”

Spokespersons:  
A. Andreyev and V. Fedosseev

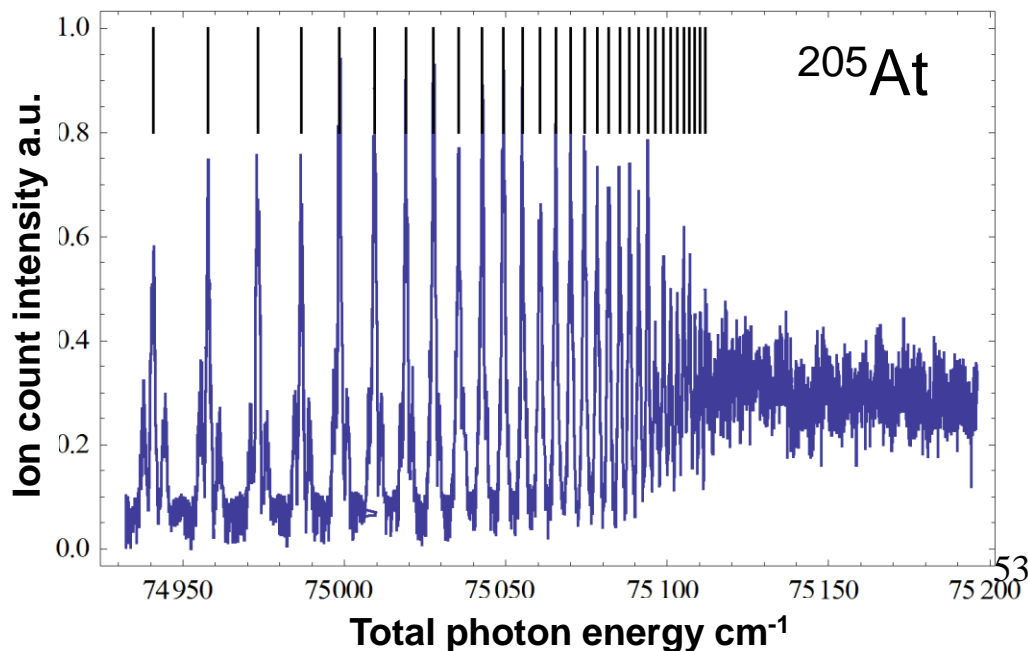
# First determination of the Ionization Potential for the radioactive element At



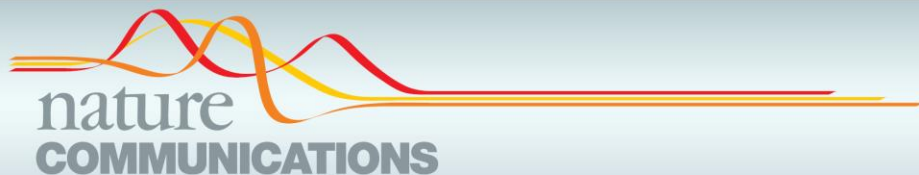
3 successful **on-line** development runs  
ISOLDE (Nov. 2010, May 2011)  
TRIUMF, Canada (Dec. 2010)

- Many new atomic levels found
- Transition strengths measured

• **Ionization potential measured** (scan of ionizing laser: converging Rydberg levels allow precise determination of the IP)



# First determination of the Ionization Potential for the radioactive element At



## ARTICLE

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OPEN

## Measurement of the first ionization potential of astatine by laser ionization spectroscopy

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