Characteristics of prompt fission γ-ray emission – advances in measurements, evaluations and predictions

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Outline

- Historical background – the 1970s
- Evaluation of PFGS characteristics
- Renaissance – the 2010s
- New evaluation – systematics
- Fast neutron induced fission
- Predictions for $^{238}U(n, f)$
- Recent results
- Predictions for $^{235}U(n, f)$
- Conclusions
- Outlook

Historical background
Experiments

- First comprehensive studies of prompt fission γ-ray spectra (PFGS) in the 1970s on:
  - n_{th} + ^{233}U
  - n_{th} + ^{235}U
  - n_{th} + ^{239}Pu
  - ^{252}Cf (sf)

- Measured PFGS characteristics:
  - $E_{\gamma,tot}$ = average total γ-energy/fission
  - ε_{γ} = average γ-energy/photon
  - $\overline{\nu}_{\gamma}$ = average γ-multiplicity

Historical background

Evaluations

1972: Nifenecker et al. (NPA 189 (1972) 285)

- $E_{\gamma,tot}(\overline{\nu}_n) = 0.75\,\overline{\nu}_n + 2.0$

2001: Valentine (ANE 28 (2001) 191)

- $E_{\gamma,tot}(\overline{\nu}_n,A,Z) = \varphi(A,Z)\,\overline{\nu}_n + 4.0$ (MeV)

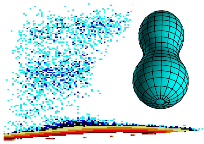
  with

  $\varphi(A,Z) = 2.51(\pm 0.01) - 1.13 \cdot 10^{-5}(\pm 7.2 \cdot 10^{-8})\, Z^2 A^{1/2}$

- $\varepsilon_\gamma(A,Z) = -1.33(\pm 0.05) - 119.6 \cdot 10^{-5}(\pm 2.5)\, Z^{1/3} / A$

- $\overline{\nu}_\gamma(\overline{\nu}_n,A,Z) = E_{\gamma,tot}(\overline{\nu}_n,A,Z) \,/\, \varepsilon_\gamma(A,Z)$

Note: A and Z dependences are purely empirical!



Renaissance
New efforts

- In the 2010s: new PFGS measurements and calculations motivated by NEA high priority request lists for
  - n_{th} + ^{235}U and n_{th} + ^{239}Pu
- Investigated processes:
  - ^{235}U(n_{th}, f), ^{239,241}Pu(n_{th}, f), ^{252}Cf(sf)
- Experimental groups:
  - LANL DANCE
  - IRMM/Chalmers/KFKI + others
- Theoretical groups (Monte Carlo Hauser Feshbach):
  - CEA Cadarache (Serot, Litaize, Regnier)
  - LANL (Talou et al.)
  - K.-H. Schmidt and others

New results - overview
Our work

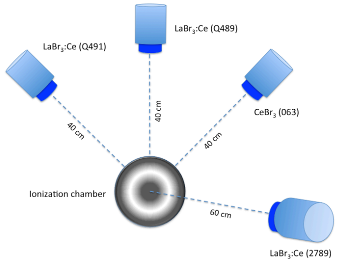
- Experiments at IRMM and KFKI Budapest:
  - ^{252}Cf(sf) — R. Billnert et al., PRC 87 (2013)
  - ^{235}U(n_{th}, f) — A. Oberstedt et al., PRC 87 (2013)
  - ^{241}Pu(n_{th}, f) — S. Oberstedt et al., PRC 90 (2014)

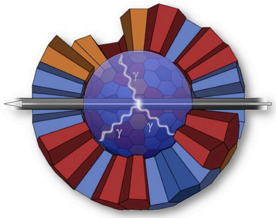
- Measured:
  - prompt fission γ-ray spectrum (PFGS)
- Determined:
  - average multiplicity
  - mean energy per photon
  - total photon energy
- Deduced:
  - multiplicity distribution

Experimental techniques

IRMM/KFKI　　　　　　DANCE

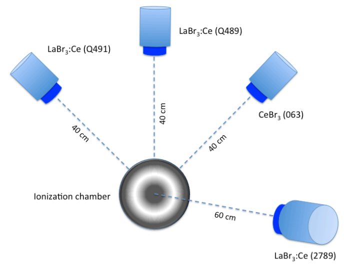
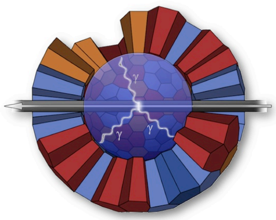
Experimental setup:





	IRMM/KFKI	DANCE
fission trigger:	IC	PPAC
photons:	e.g. $LaBr_3$	BaF_2 array

Experimental techniques

	IRMM/KFKI	DANCE
Experimental setup:		
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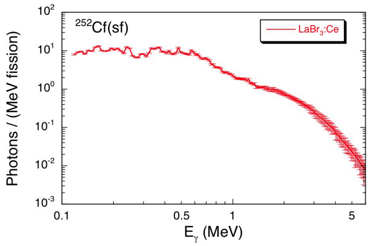
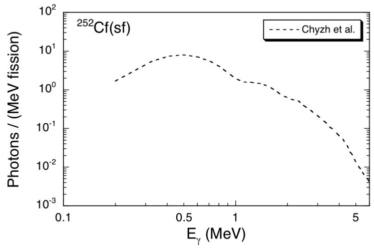
photons:	e.g. $LaBr_3$	BaF_2 array

PFGS: $E_{\gamma,tot}$, $\overline{\nu}_\gamma$, ε_γ



measured

↔



measured

ε_γ

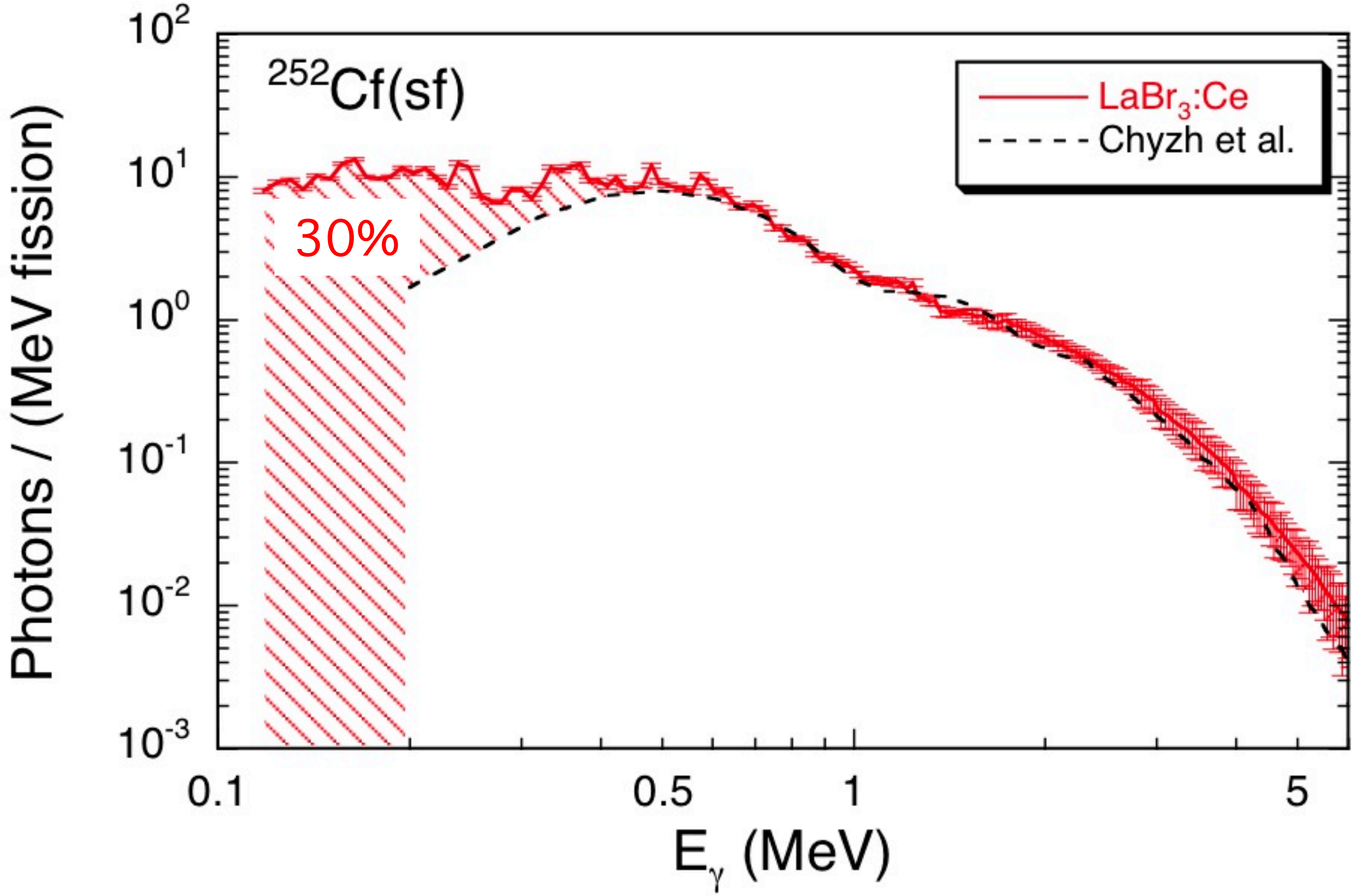
Experimental techniques

IRMM/KFKI DANCE

Experimental setup:

fission trigger:
photons:

PFGS:



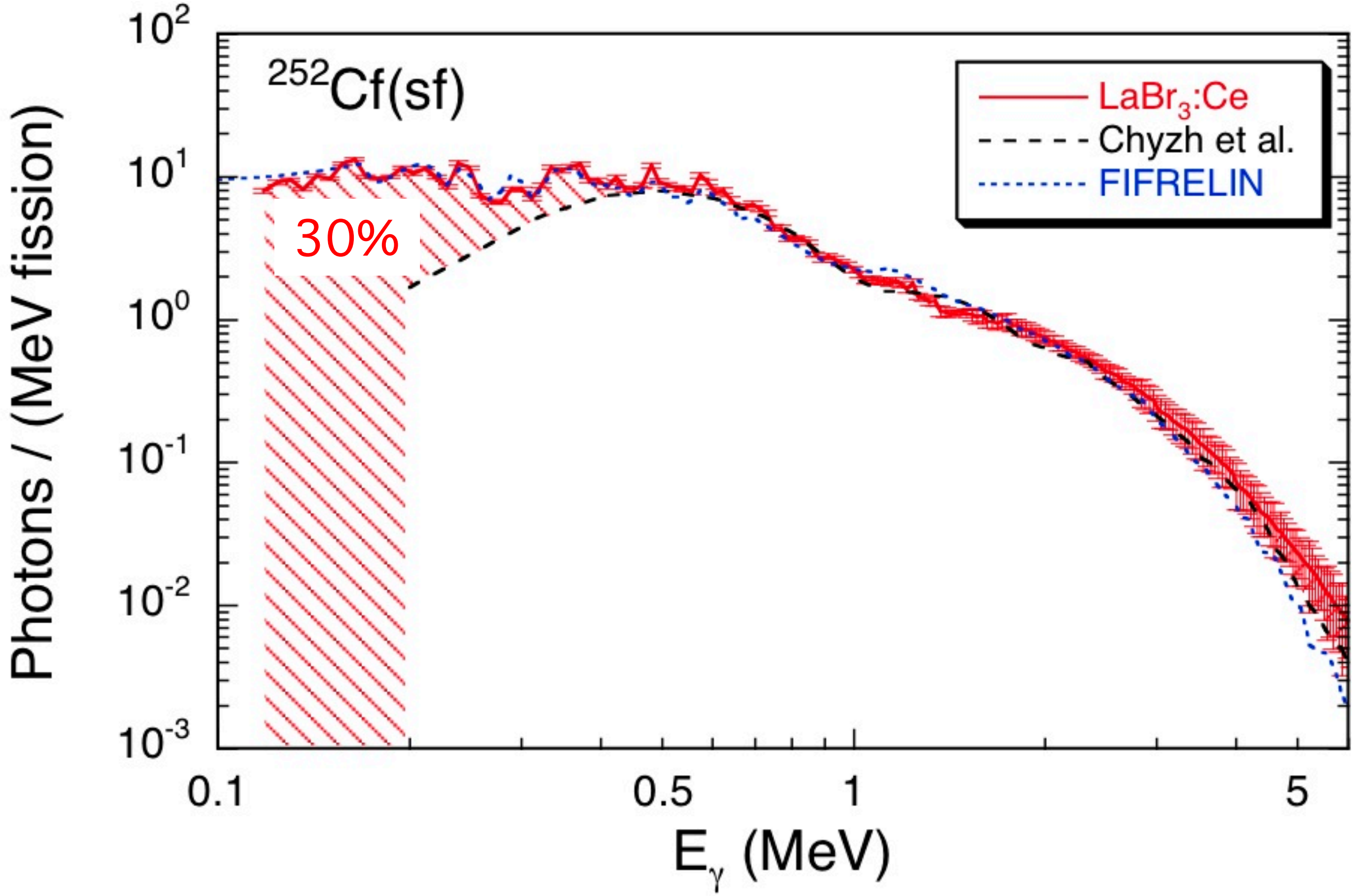
Experimental techniques

IRMM/KFKI DANCE

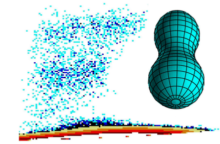
Experimental setup:

fission trigger:
photons:

PFGS:



Experimental techniques



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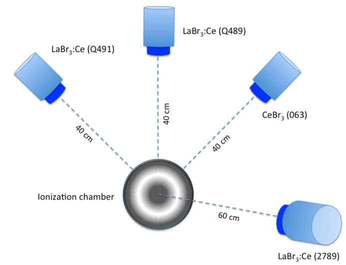
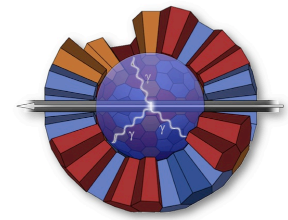
Experimental setup:		
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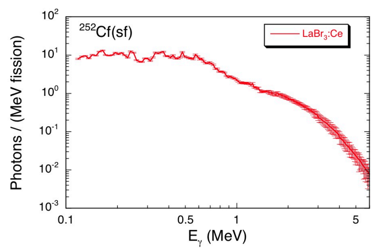
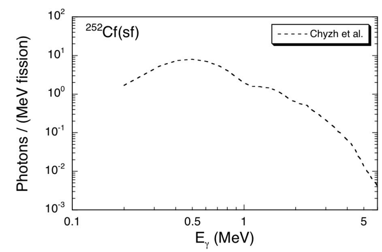
PFGS: $E_{\gamma,tot}$, $\overline{\nu}_\gamma$, ε_γ



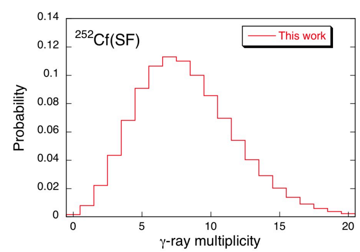
measured



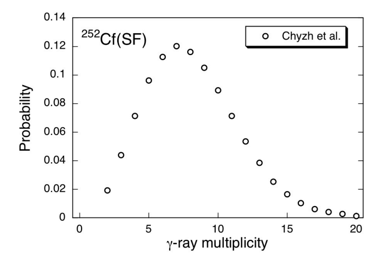
measured

ε_γ

multiplicity distribution:



deduced



measured

$\overline{\nu}_\gamma$

Experimental techniques

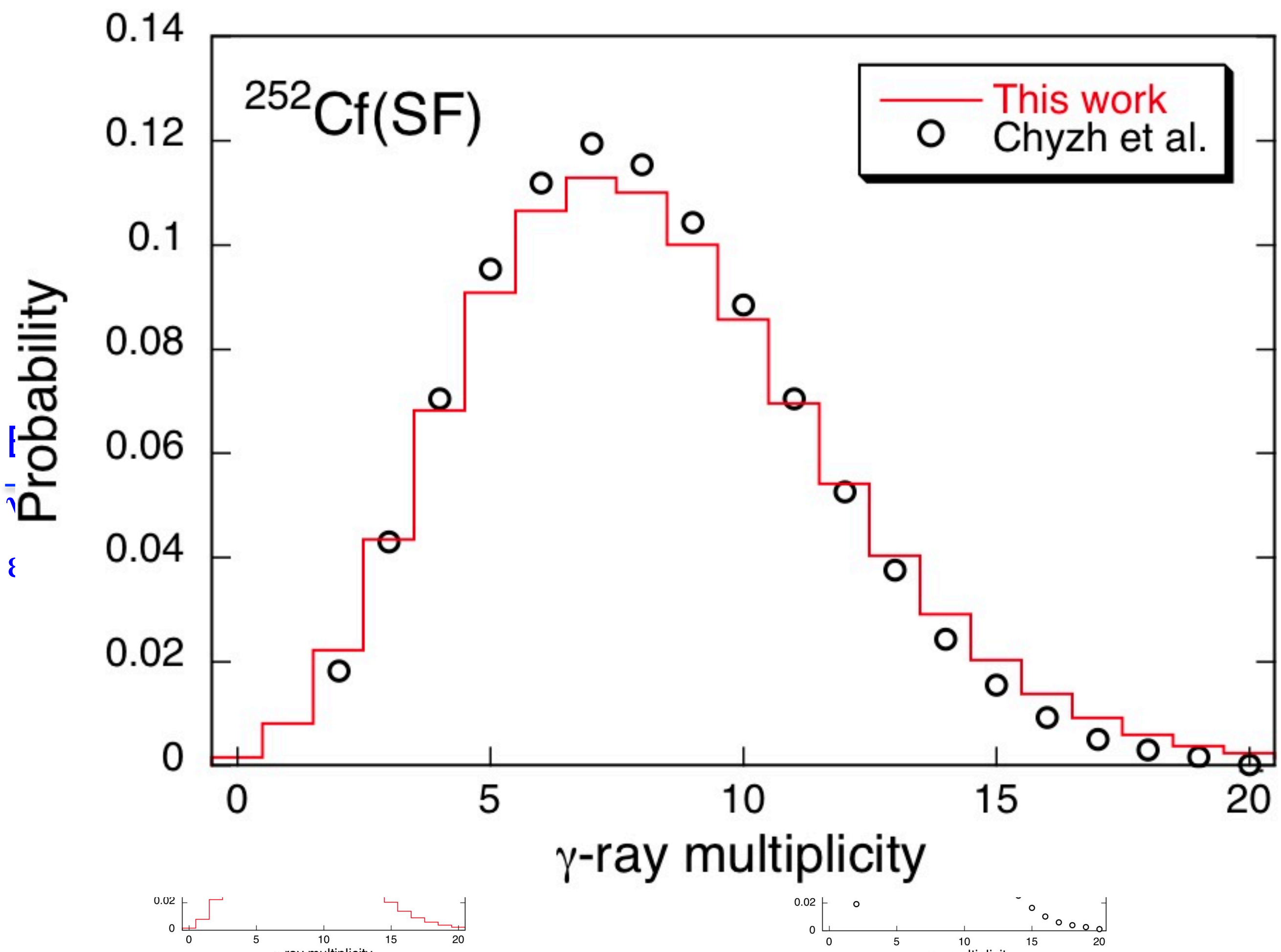
IRMM/KFKI　　DANCE

Experimental setup:

fission trigger:
photons:

PFGS:

multiplicity distribution:

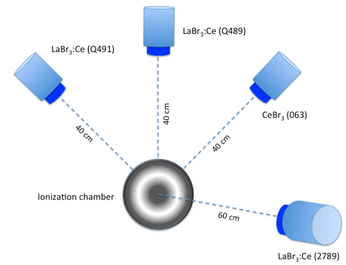


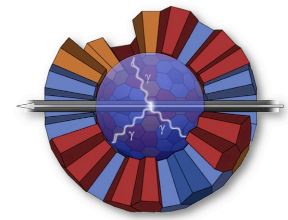
deduced　　measured

Experimental techniques

IRMM/KFKI — DANCE

Experimental setup:

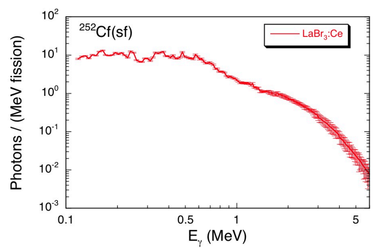




fission trigger: IC — PPAC

photons: e.g. $LaBr_3$ — BaF_2 array

PFGS: $E_{\gamma,tot}$, $\overline{\nu}_\gamma$, ε_γ



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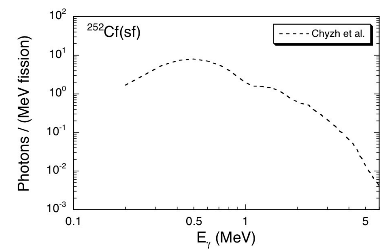
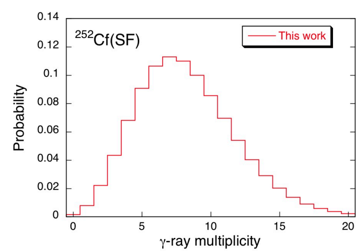


measured — measured

multiplicity distribution:



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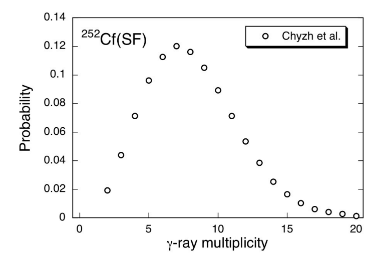


deduced — measured

ε_γ, $\overline{\nu}_\gamma$ → $E_{\gamma,tot}$

Comparison of results

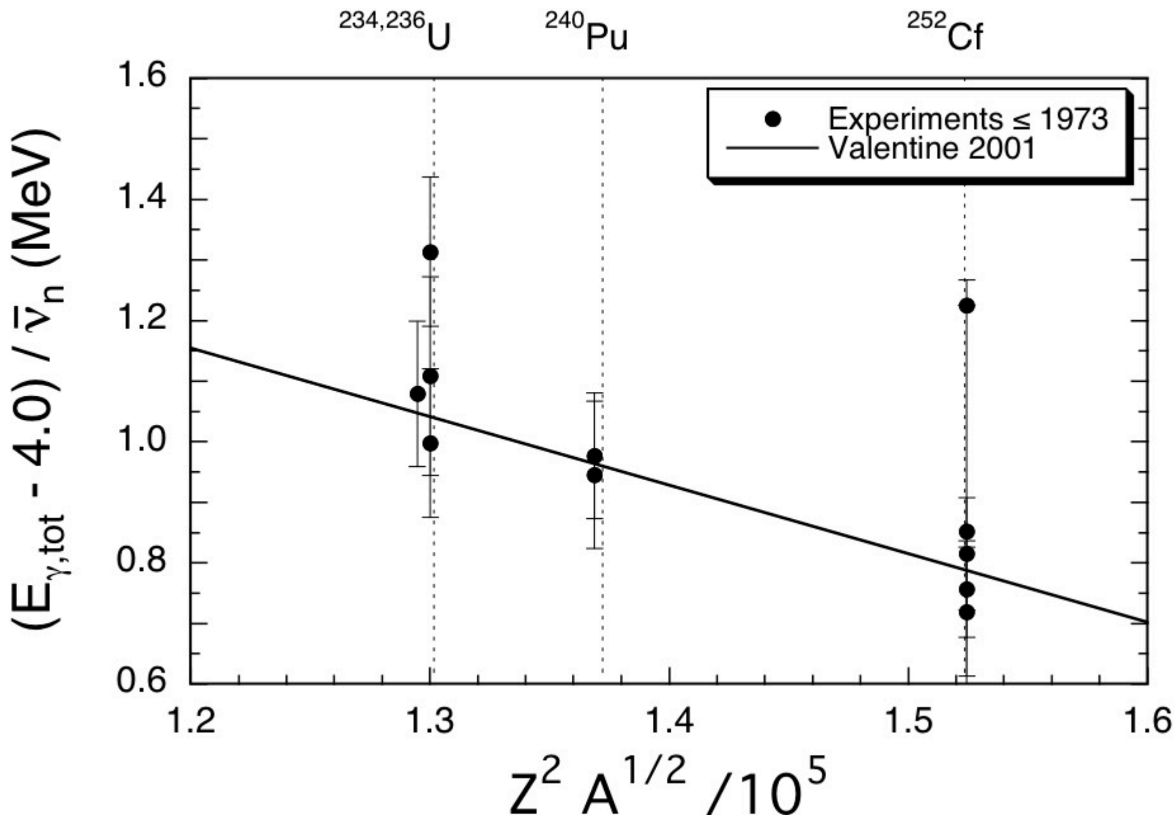
General impression for the fissioning systems
^{252}Cf(sf), ^{235}U(n_{th}, f) and ^{241}Pu(n_{th}, f)

- $E_{\gamma,tot}$ and ε_γ : good agreement between our results and those from the early 1970s, while values from DANCE are higher

- $\bar{\nu}_\gamma$: our results agree well with the 1970s results, but the DANCE values are somewhat lower

- Impact of our new results on evaluation according to Valentine?

New evaluation
PFGS average total energy per fission



Observe: $\bar{\nu}_n$ taken from experiments

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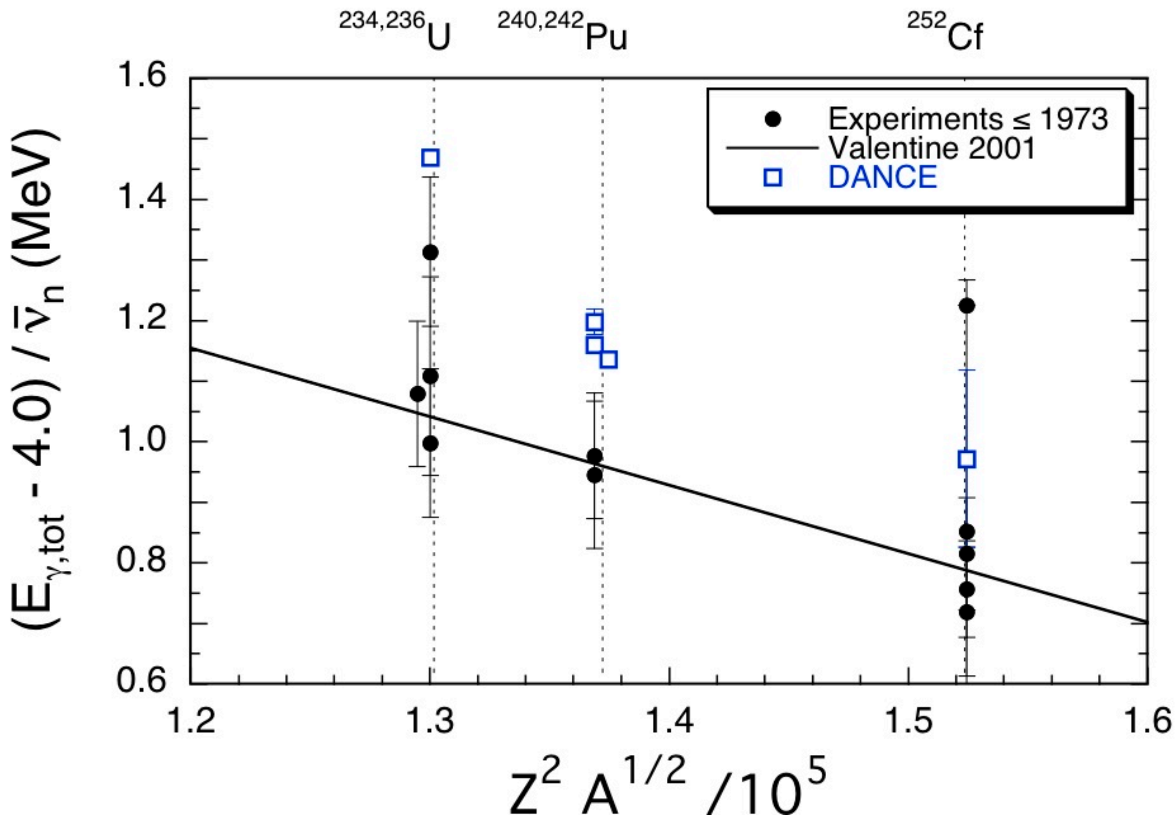
PFGS average total energy per fission



Observe: $\bar{\nu}_n$ taken from experiments
and ENDF/B-VII.1

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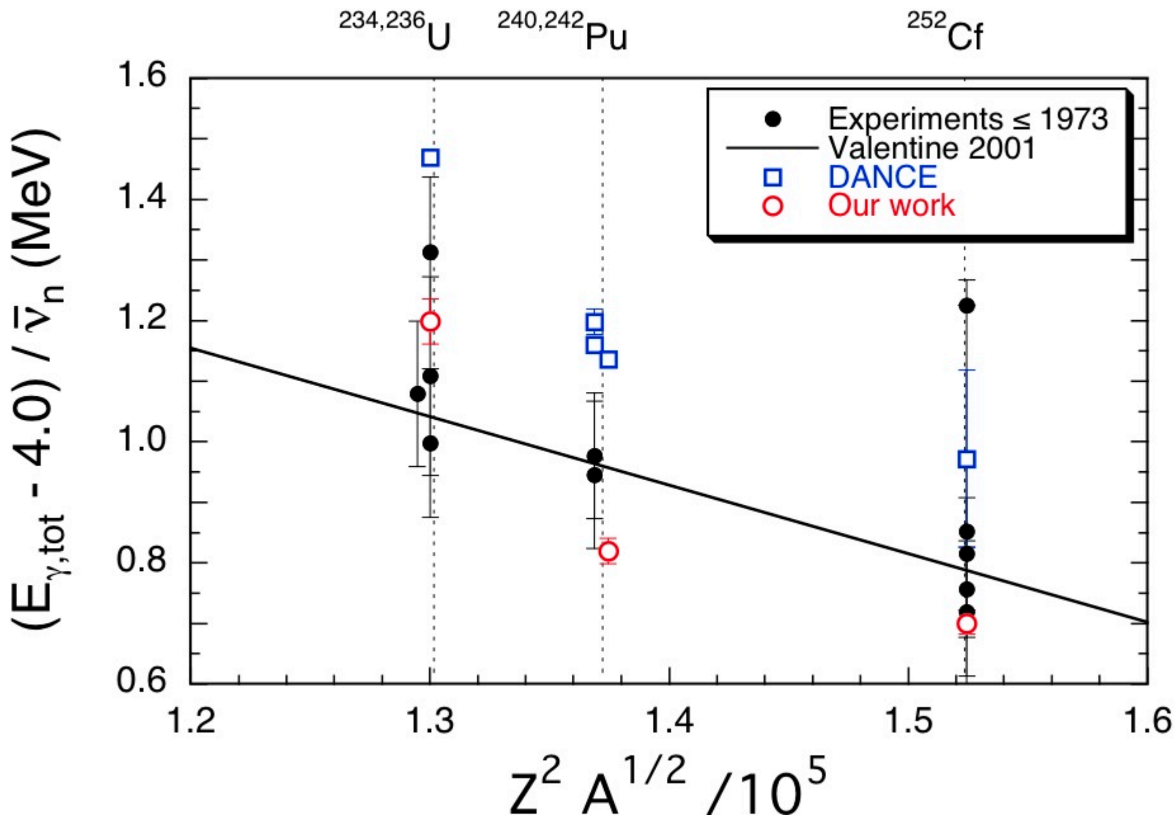

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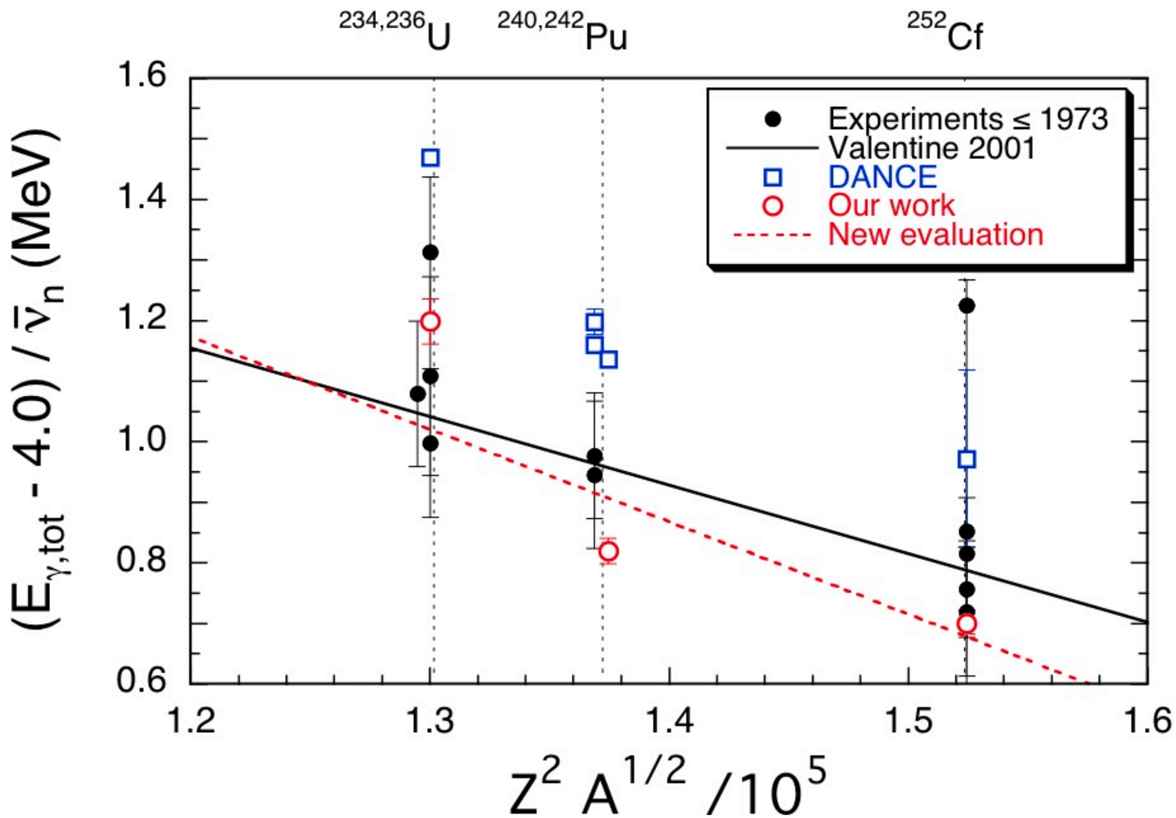

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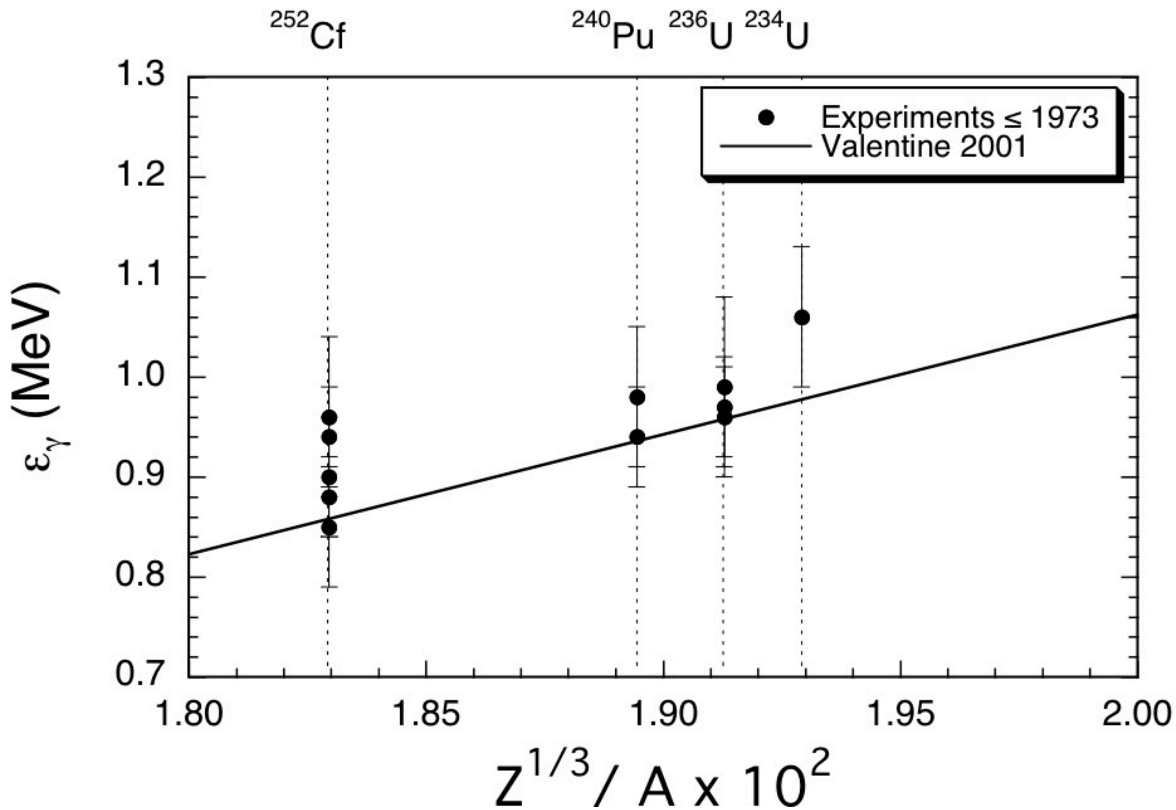
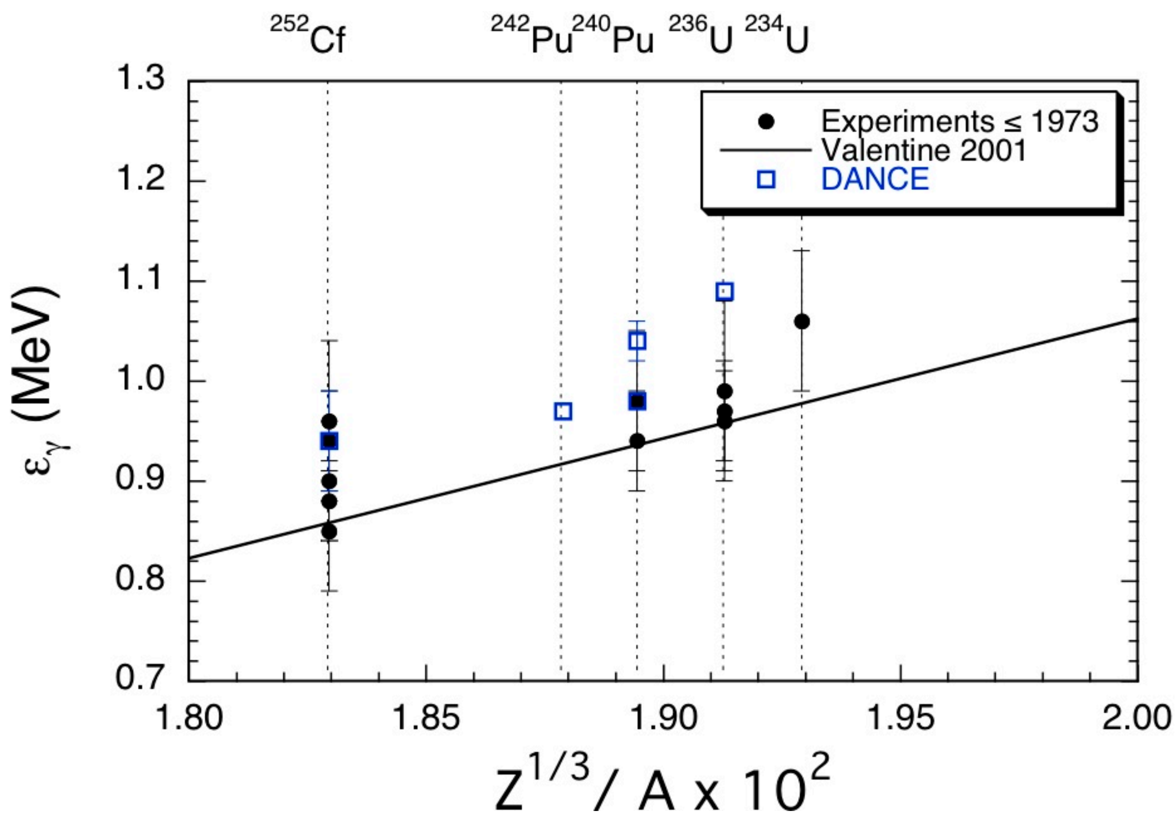
New evaluation
PFGS mean energy per photon



New evaluation
PFGS mean energy per photon



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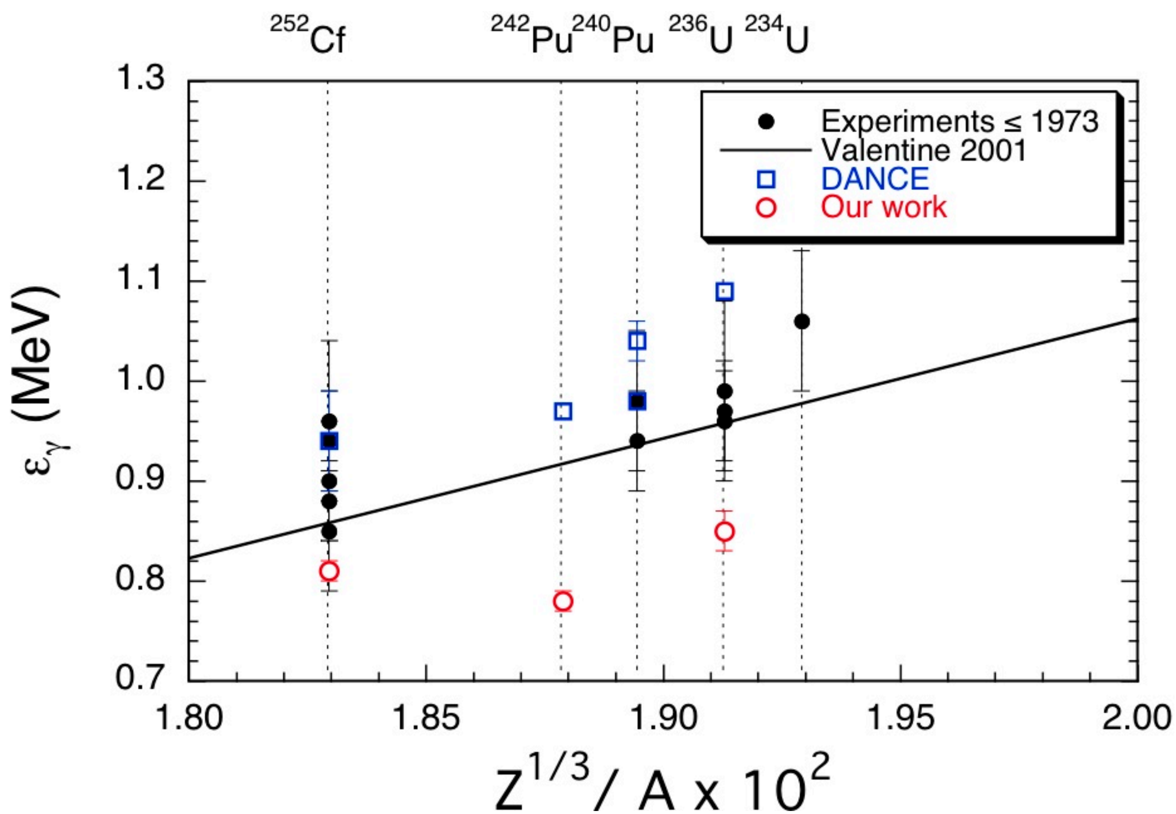

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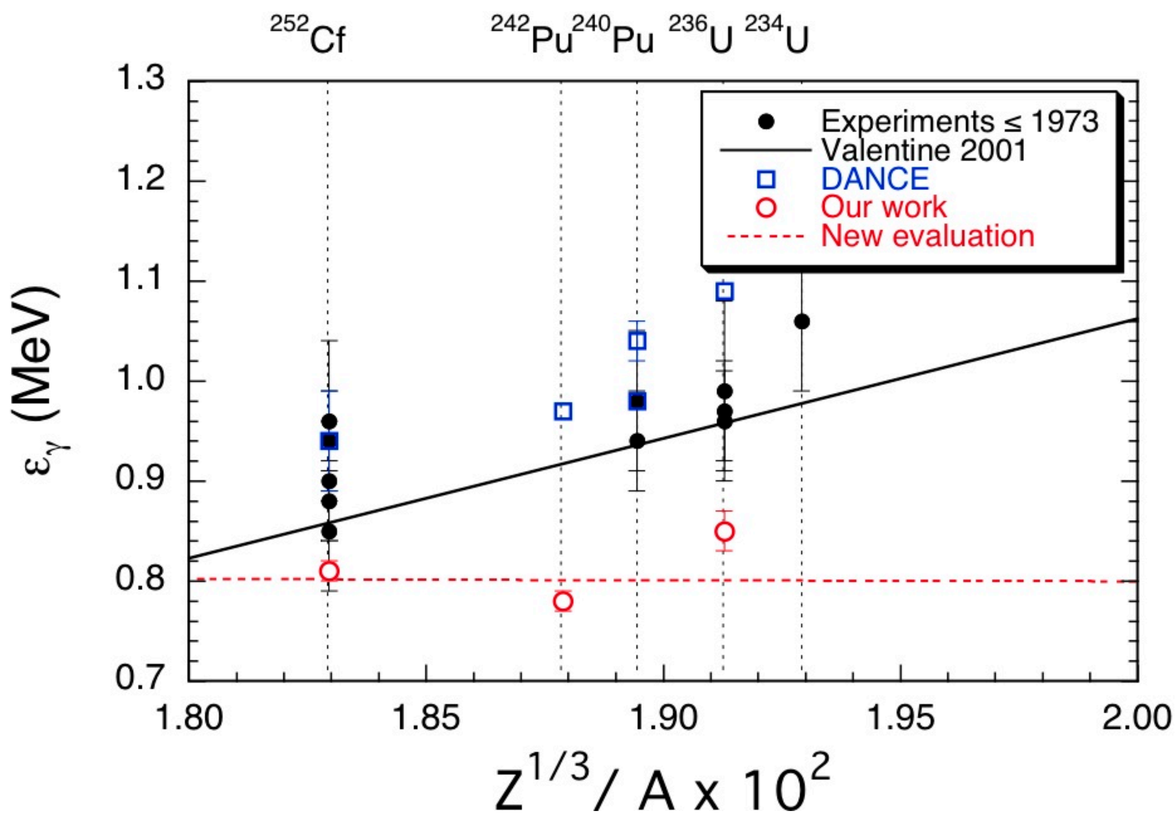
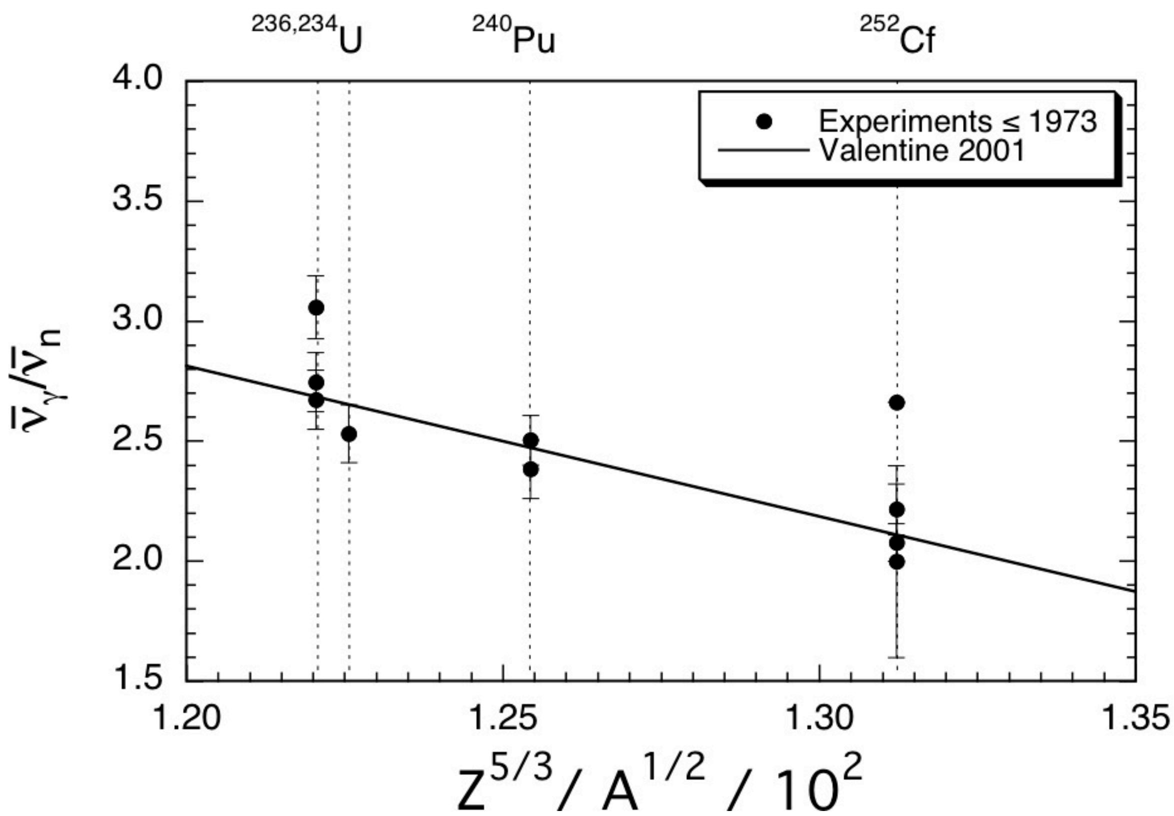
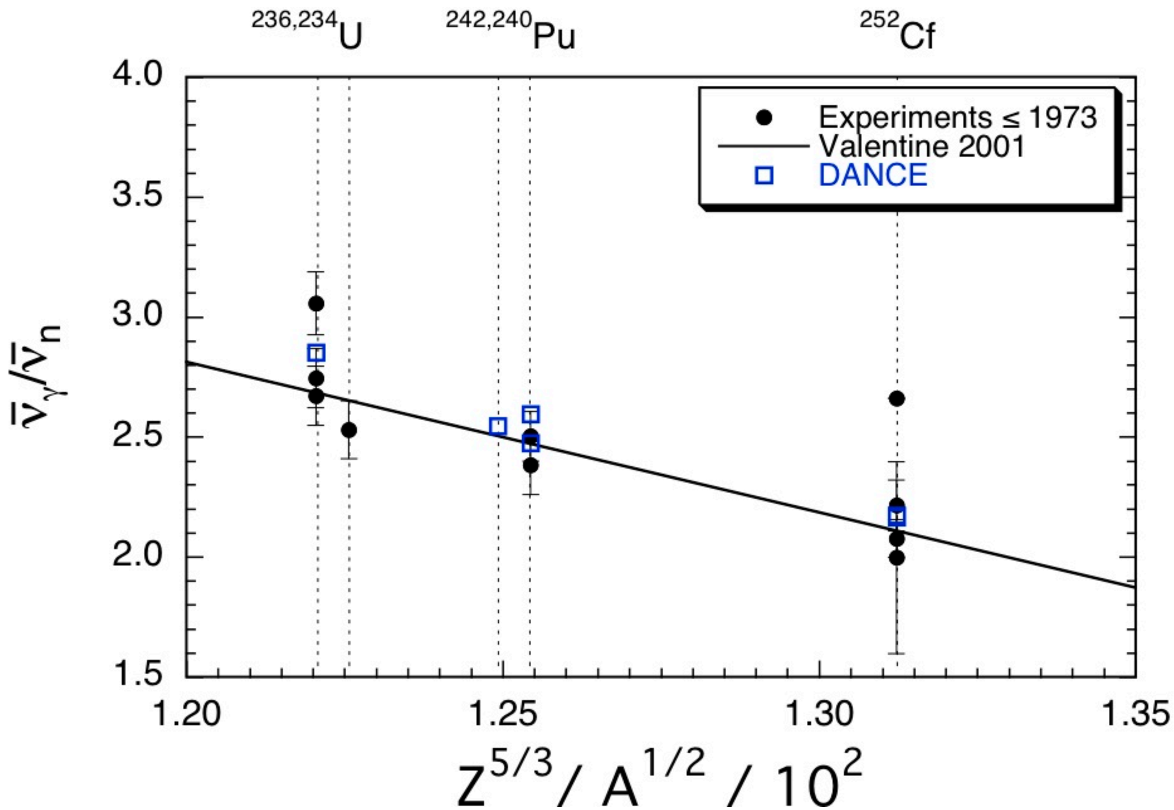


New evaluation
PFGS average multiplicity



Observe: $\bar{\nu}_n$ taken from experiments

New evaluation
PFGS average multiplicity



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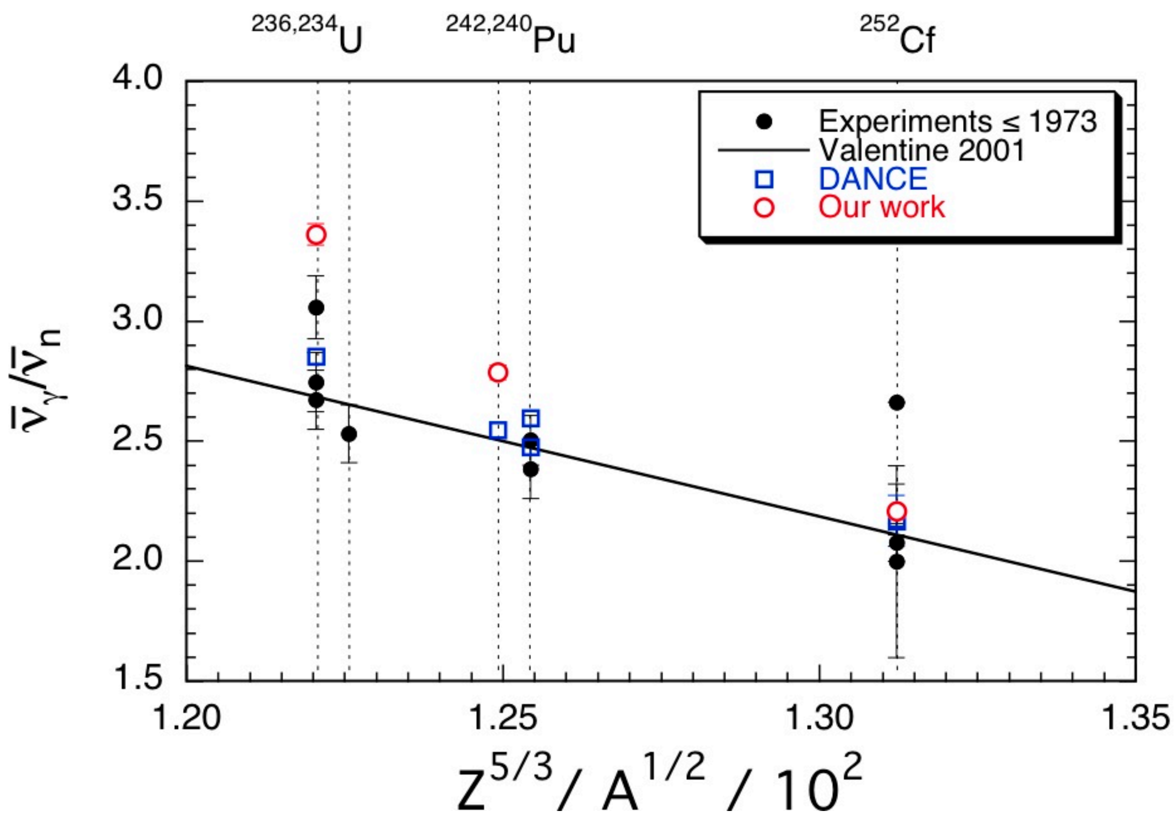

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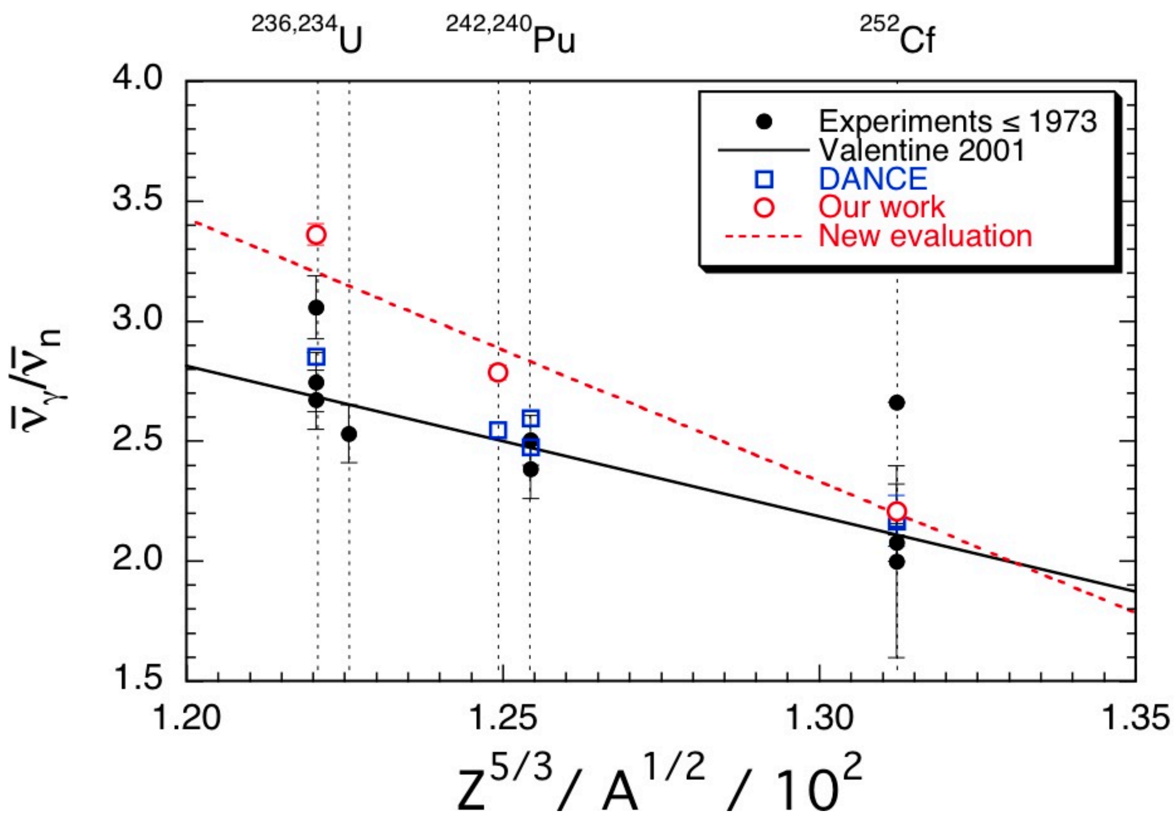

Observe: $\bar{\nu}_n$ taken from experiments.
and ENDF/B-VII.1

Summary – so far

- PFGS characteristics as function of A and Z of the compound system
- Agreement between new experimental results and "Valentine's evaluation" is rather good
- Parameters might need an adjustment
- Estimate (interpolations/extrapolation) of PFGS characteristics is possible for nuclei, which are not accessible experimentally
- However: only valid for thermal neutron induced and spontaneous fission?
- Attempt: fast neutron induced fission!
- Example below: ^{238}U(n, f)!

Fast neutron induced fission

Motivation

Why n + ^{238}U PFGS?

- Important nuclide for fast reactors
- one of six isotopes in the focus of the CIELO pilot project

- First – preliminary – experimental results from CEA in Bruyères-le-Châtel are available
  - at E_n = 1.7 and 15.6 MeV (BGO)
  - as well as E_n = 1.7 and 5.2 MeV (BGO), new!

    (Laborie et al., private communication)

- New experiment performed recently at IPN Orsay
  - LICORNE facility, covering energy range between

    E_n = 0.7 and 4 MeV
  - First preliminary results obtained

Fast neutron induced fission

Multiple chance fission

n + ^{238}U @ E_n → ^{239}U* → fission + neutrons

↓

(n' + ^{237}U @ E_n') n + ^{238}U* → fission + neutrons

↓

(n'' + ^{236}U @ E_n'') n + ^{237}U* → fission + neutrons

↓

(n''' + ^{235}U @ E_n''') n + ^{236}U* → fission + neutrons

↓

etc.

$$\bar{\nu} = \bar{\nu}_{pre} + \bar{\nu}_{ff}$$ with

$\bar{\nu}_{pre}$ = pre-fission neutrons

$\bar{\nu}_{ff}$ = prompt neutrons from fission fragments

Fast neutron induced fission

Multiple chance fission

n + ^{238}U @ E_n → ^{239}U* → fission + neutrons

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(n' + ^{237}U @ E_n') n + ^{238}U* → fission + neutrons

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↓

(n''' + ^{235}U @ E_n''') n + ^{236}U* → fission + neutrons

↓

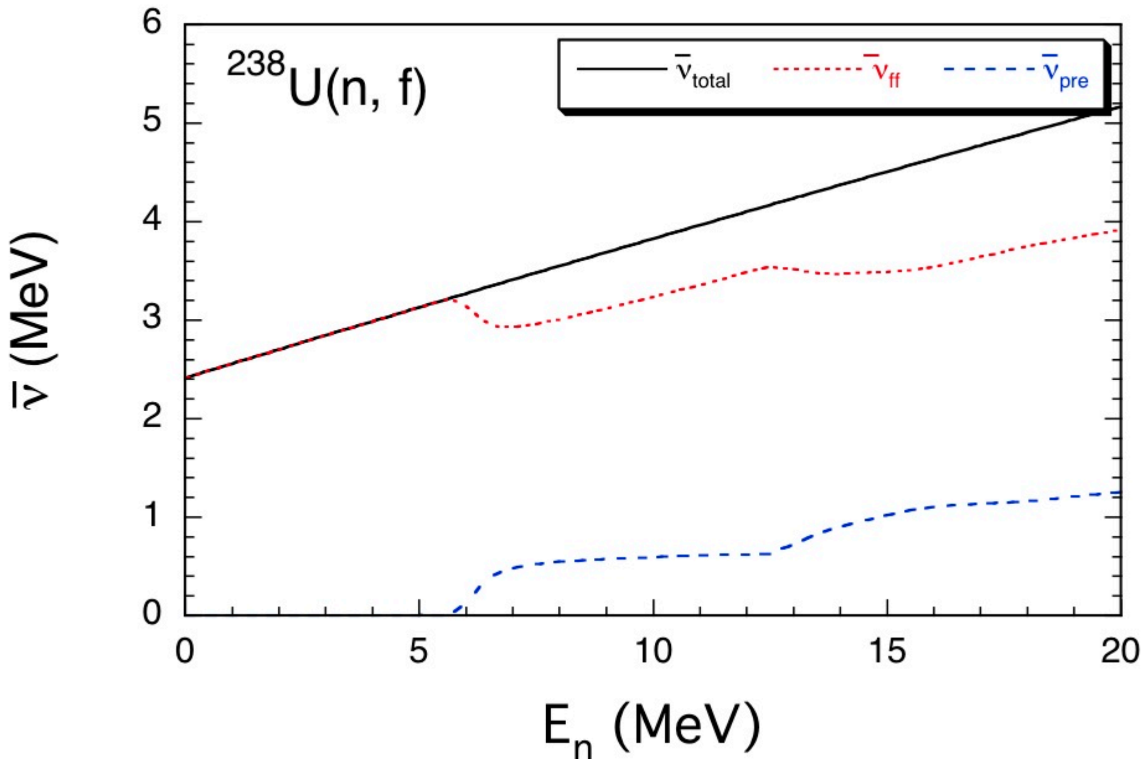
etc.

$\bar{\nu} = \bar{\nu}_{pre} + \bar{\nu}_{ff}$ with

$\bar{\nu}_{pre}$ = pre-fission neutrons

$\bar{\nu}_{ff}$ = prompt neutrons from fission fragments, only these compete with PFG emission!

^{238}U(n, f) PFGS characteristics
Prompt fission neutron multiplicity



- prompt fission neutrons from ENDF/B-VII.1
- pre-fission neutrons subtracted
- prompt neutrons from fragments for PFGS characteristics – only those may be related to prompt fission γ-ray emission!

^{238}U(n, f) PFGS characteristics
Calculating energy dependence

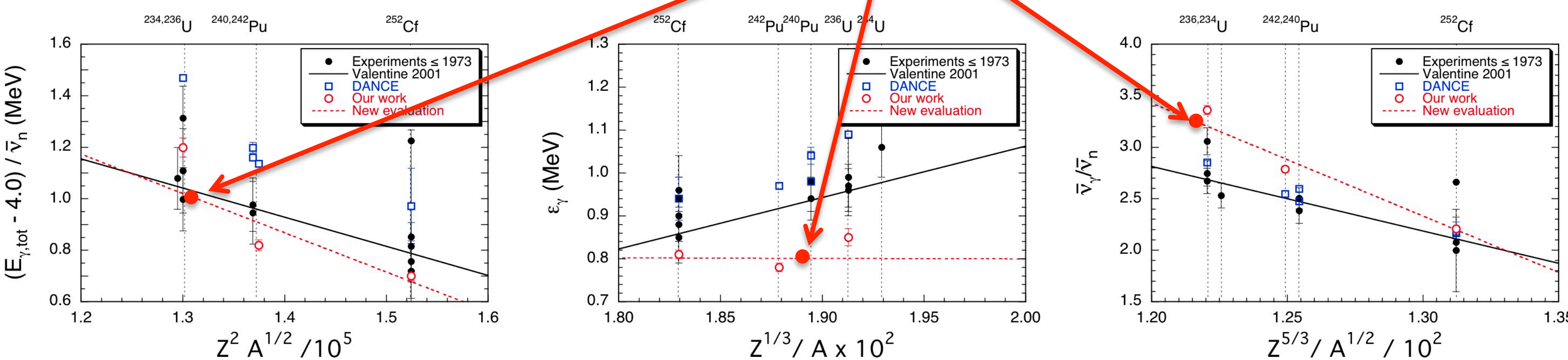
- from PFGS systematics as function of A and Z

$$\left.\begin{array}{l} (E_{\gamma,tot} - 4)/\overline{\nu}_n \\ \varepsilon_\gamma \\ \overline{\nu}_\gamma \;/\overline{\nu}_n \end{array}\right\} \text{ for } n + {}^{238}\text{U}$$

^{238}U(n, f) PFGS characteristics
Calculating energy dependence

- from PFGS systematics as function of A and Z

$$\left.\begin{array}{l} (E_{\gamma,tot} - 4)/\overline{\nu}_n \\ \varepsilon_\gamma \\ \overline{\nu}_\gamma \;\; /\overline{\nu}_n \end{array}\right\} \text{ for } n + {}^{238}U$$



^{238}U(n, f) PFGS characteristics
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- using $\overline{\nu}_n(E_n) = \overline{\nu}_{ff}(E_n)$

^{238}U(n, f) PFGS characteristics
Calculating energy dependence

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$$\left.\begin{array}{l}(E_{\gamma,tot} - 4)/\bar{\nu}_n \\ \varepsilon_\gamma \\ \bar{\nu}_\gamma \;\; /\bar{\nu}_n\end{array}\right\} \text{for } n + {}^{238}\text{U}$$

- using $\bar{\nu}_n(E_n) = \bar{\nu}_{ff}(E_n)$
- assuming: only energy dependence = $\bar{\nu}_{ff}(E_n)$

^{238}U(n, f) PFGS characteristics
Calculating energy dependence

- from PFGS systematics as function of A and Z

  $(E_{\gamma,tot} - 4)/\overline{\nu}_n$

  ε_γ } for n + ^{238}U

  $\overline{\nu}_\gamma / \overline{\nu}_n$

- using $\overline{\nu}_n(E_n) = \overline{\nu}_{ff}(E_n)$
- assuming: only energy dependence = $\overline{\nu}_{ff}(E_n)$
- calculating $E_{\gamma,tot}(E_n)$

  $\overline{\nu}_\gamma(E_n)$

  $\varepsilon_\gamma(E_n) = E_{\gamma,tot}(E_n)/\overline{\nu}_\gamma(E_n)$

^{238}U(n, f) PFGS characteristics
Calculating energy dependence

- from PFGS systematics as function of A and Z

  $(E_{\gamma,tot} - 4)/\overline{\nu}_n$

  ε_γ

  $\overline{\nu}_\gamma \ /\overline{\nu}_n$

  } for n + ^{238}U

- using $\overline{\nu}_n(E_n) = \overline{\nu}_{ff}(E_n)$
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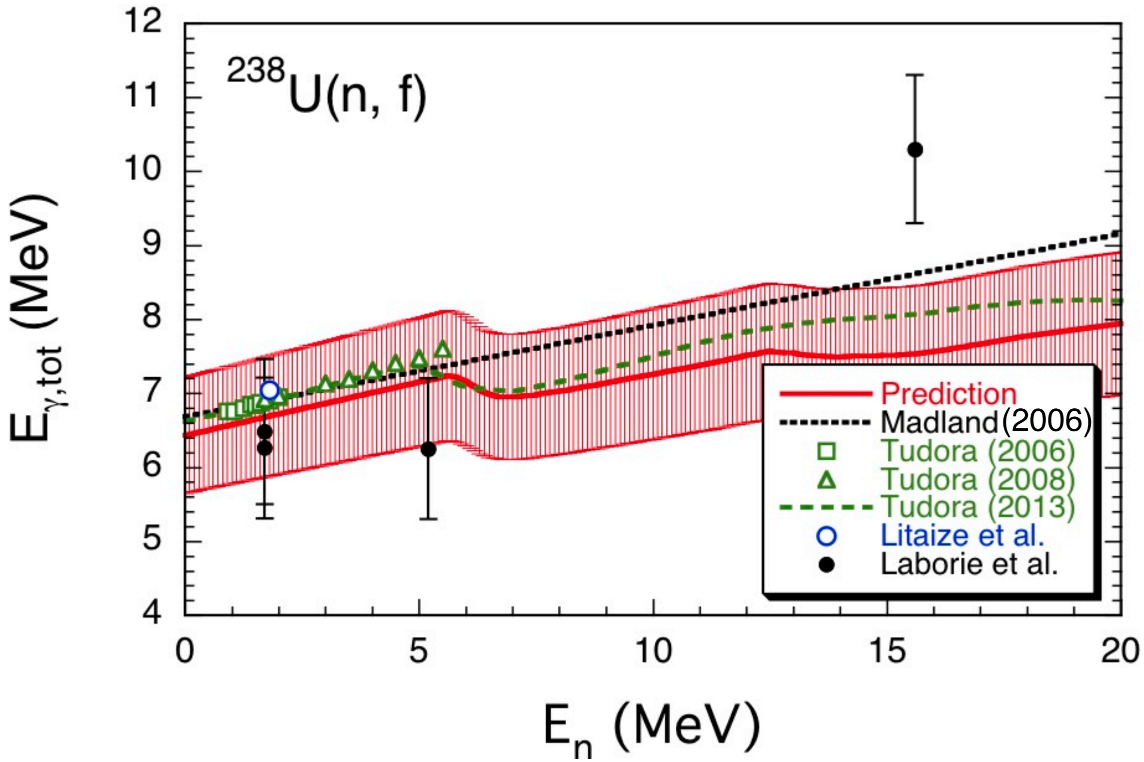
- calculating $E_{\gamma,tot}(E_n)$

  $\overline{\nu}_\gamma(E_n)$

  $\varepsilon_\gamma(E_n) = E_{\gamma,tot}(E_n)/\overline{\nu}_\gamma(E_n)$
- comparison with model calculations
- comparison with preliminary experimental results

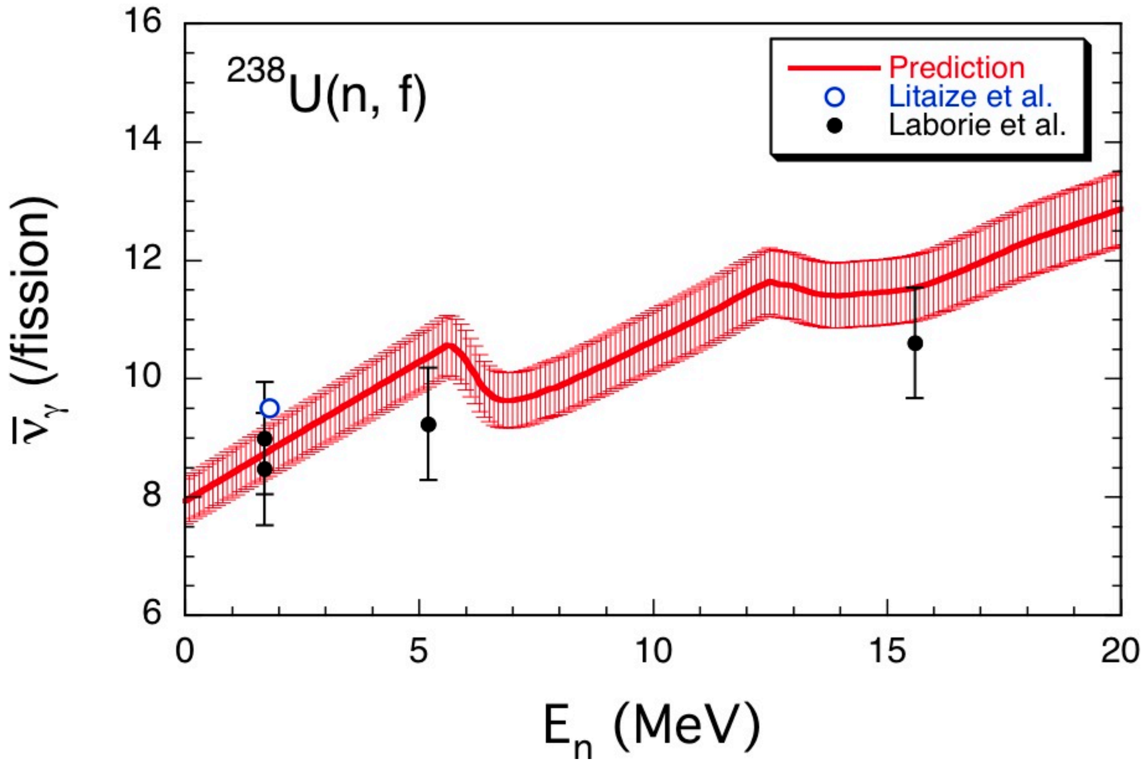
^{238}U(n, f) PFGS characteristics
Average total energy per fission



- Tudora: Point-by-Point model
- Litaize et al.: FIFRELIN code (ND 2013, to appear in NDS)
- Laborie et al.: preliminary results (2014)

$^{238}U(n, f)$ PFGS characteristics

Average γ-ray multiplicity



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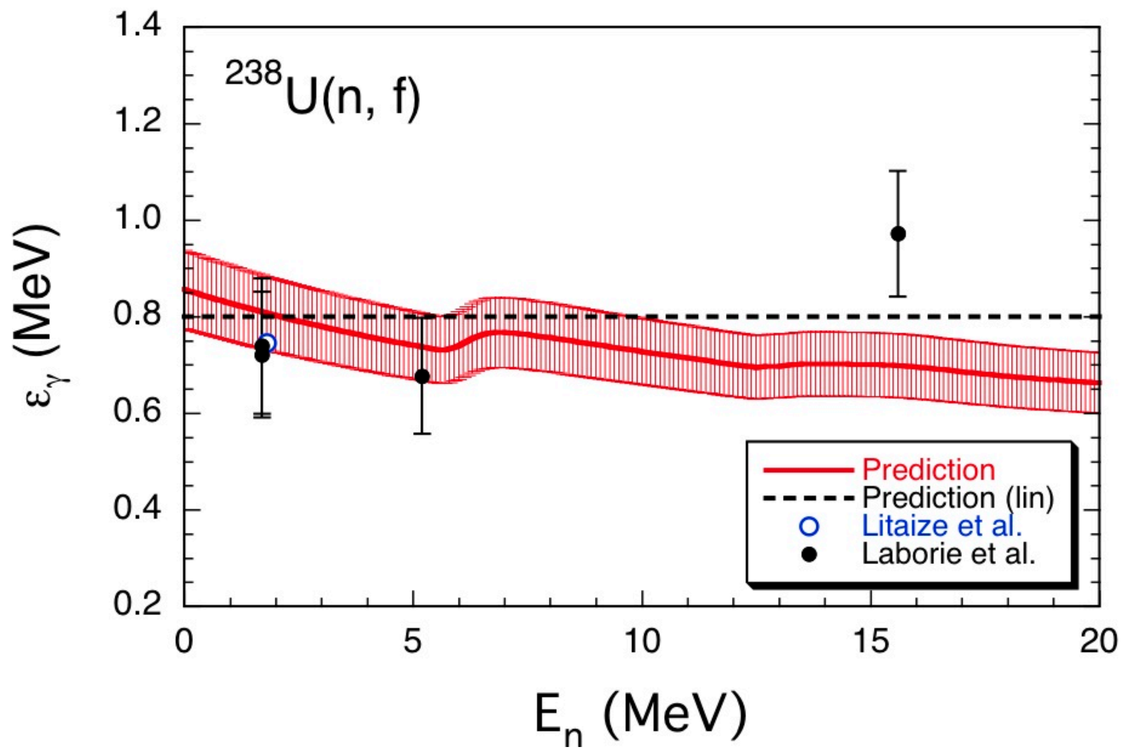
$^{238}U(n, f)$ PFGS characteristics
Mean energy per photon



- Litaize et al.: FIFRELIN code (calculated by $E_{\gamma,tot} / \overline{\nu}_{\gamma}$)
- Laborie et al.: preliminary results (2014)

Next summary …

- Predictions of PFGS characteristics as function of incident neutron energy have been presented for $^{238}U(n, f)$!
- Investigated energy range: E_n = 0 ... 20 MeV
- Good agreement with both preliminary experimental results and model calculations for
  - $E_{\gamma,tot}(E_n)$
  - $\varepsilon_\gamma(E_n)$
  - $\nu_\gamma(E_n)$

Recently …

New experiments and results

Recently …
New experiments and results

- $^{235}U(n, f)$ at E_n = 1.5 MeV, LICORNE (2013)
  - 14 BaF_2 detectors
  - 3 $LaBr_3$:Ce detectors (1 week)

Recently …

New experiments and results

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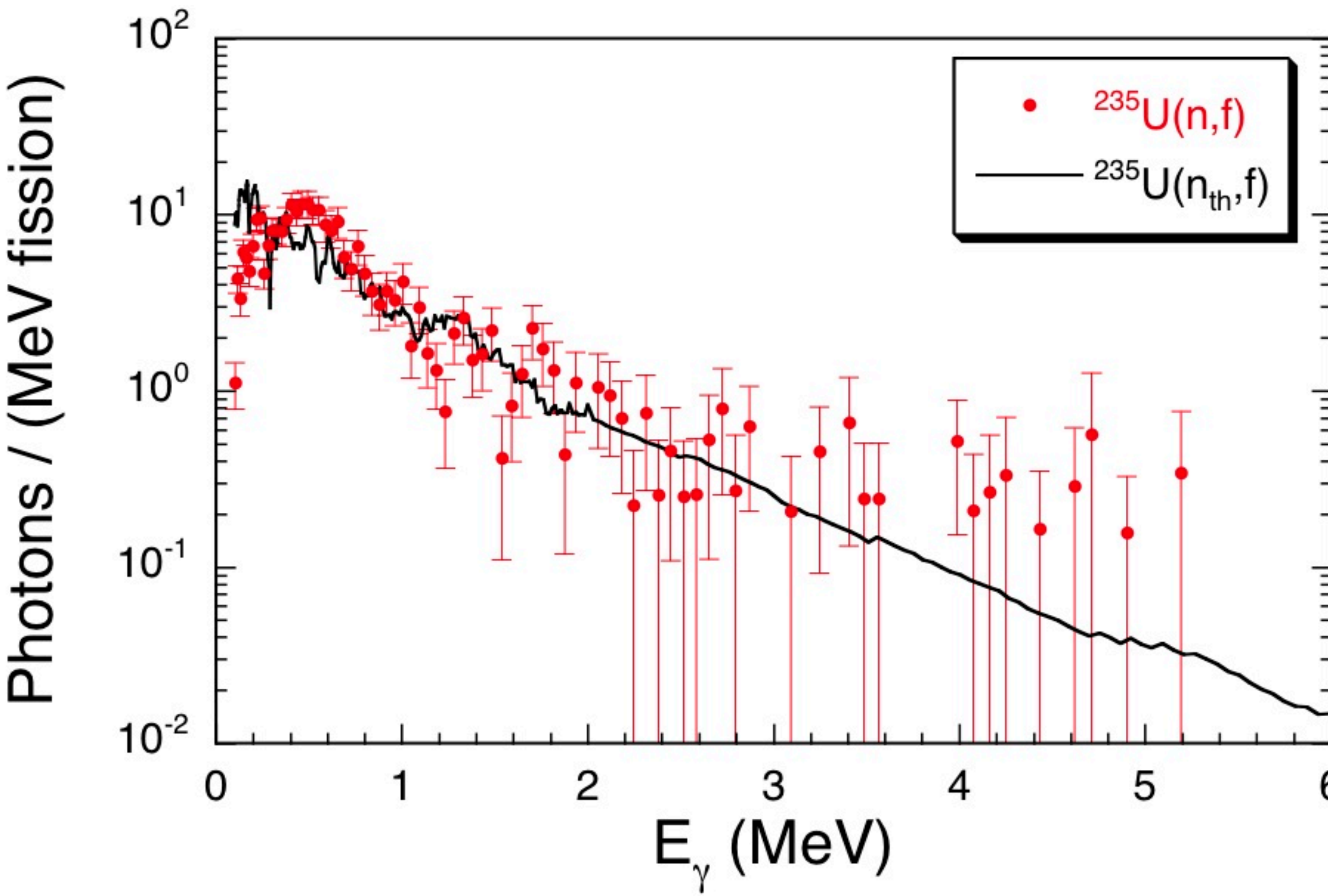
Recently …
New experiments and results



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- $^{235}U(n, f)$ at $E_n = 1.5$ MeV, LICORNE (2013)
  - 14 BaF_2 detectors
  - 3 $LaBr_3$:Ce detectors (1 week) –

preliminary:

$E_{\gamma,tot}$ = (7.4±0.7) MeV

$\overline{\nu}_\gamma$ = (8.7±0.4)

ε_γ = (0.85±0.07) MeV

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Predictions! <---

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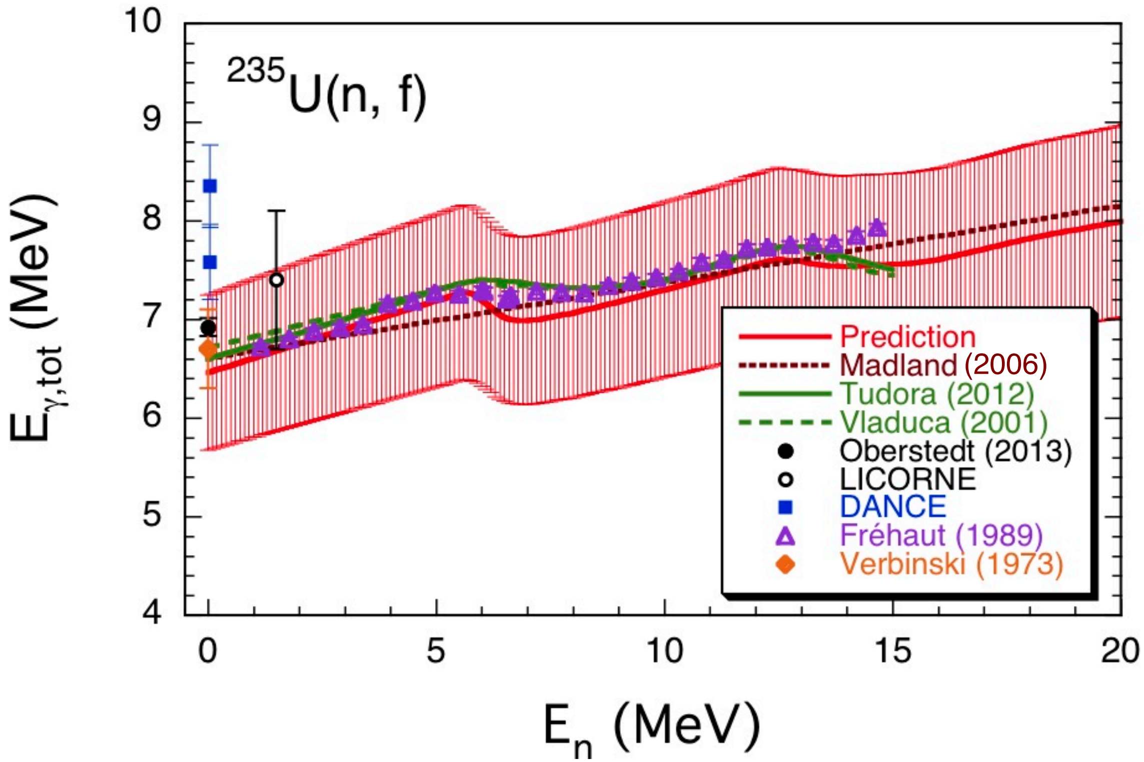
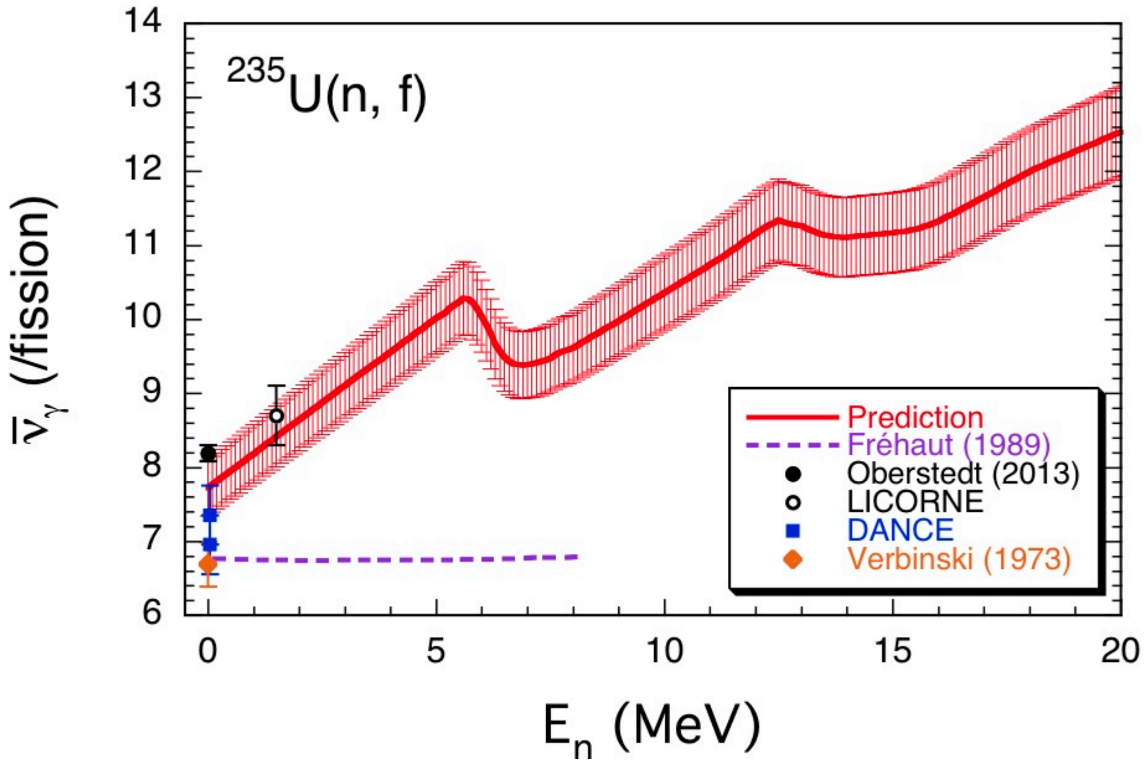
$^{235}U(n, f)$ PFGS characteristics
Average total energy per fission



- Tudora et al.: Phys. Procedia 31 (2012)
- LICORNE: preliminary results (2014)
- DANCE: Chyzh et al. (2013) and (2014)

^{235}U(n, f) PFGS characteristics

Average γ-ray multiplicity



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^{235}U(n, f) PFGS characteristics
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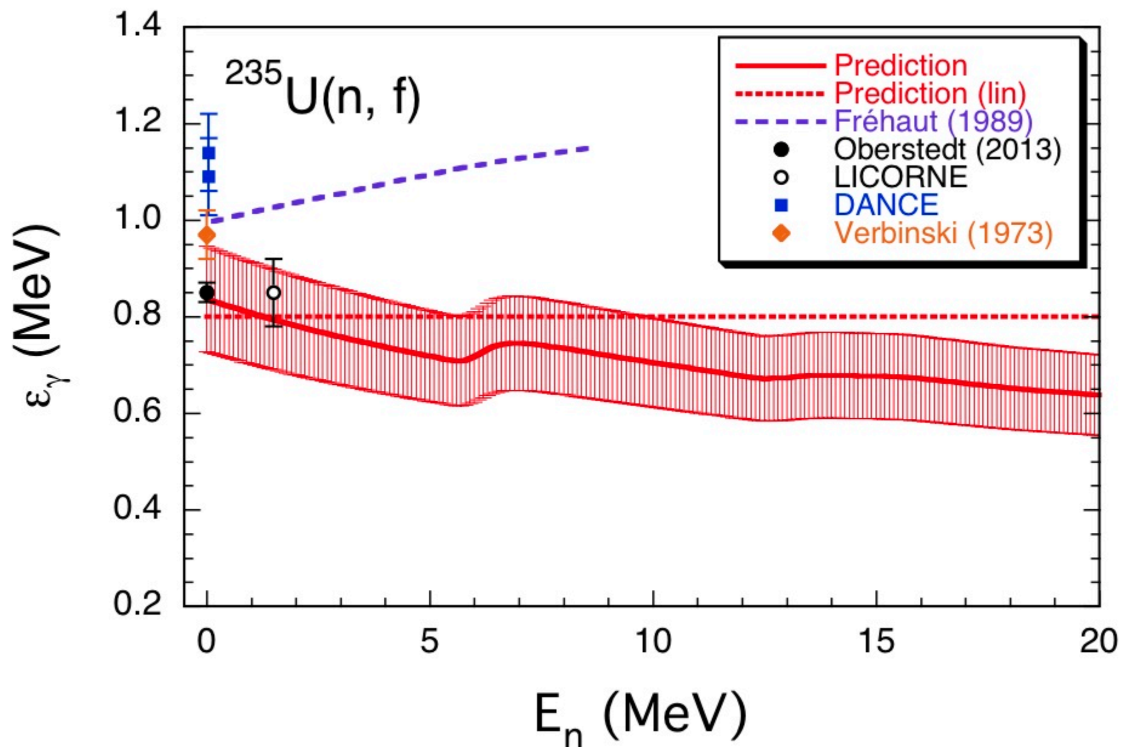

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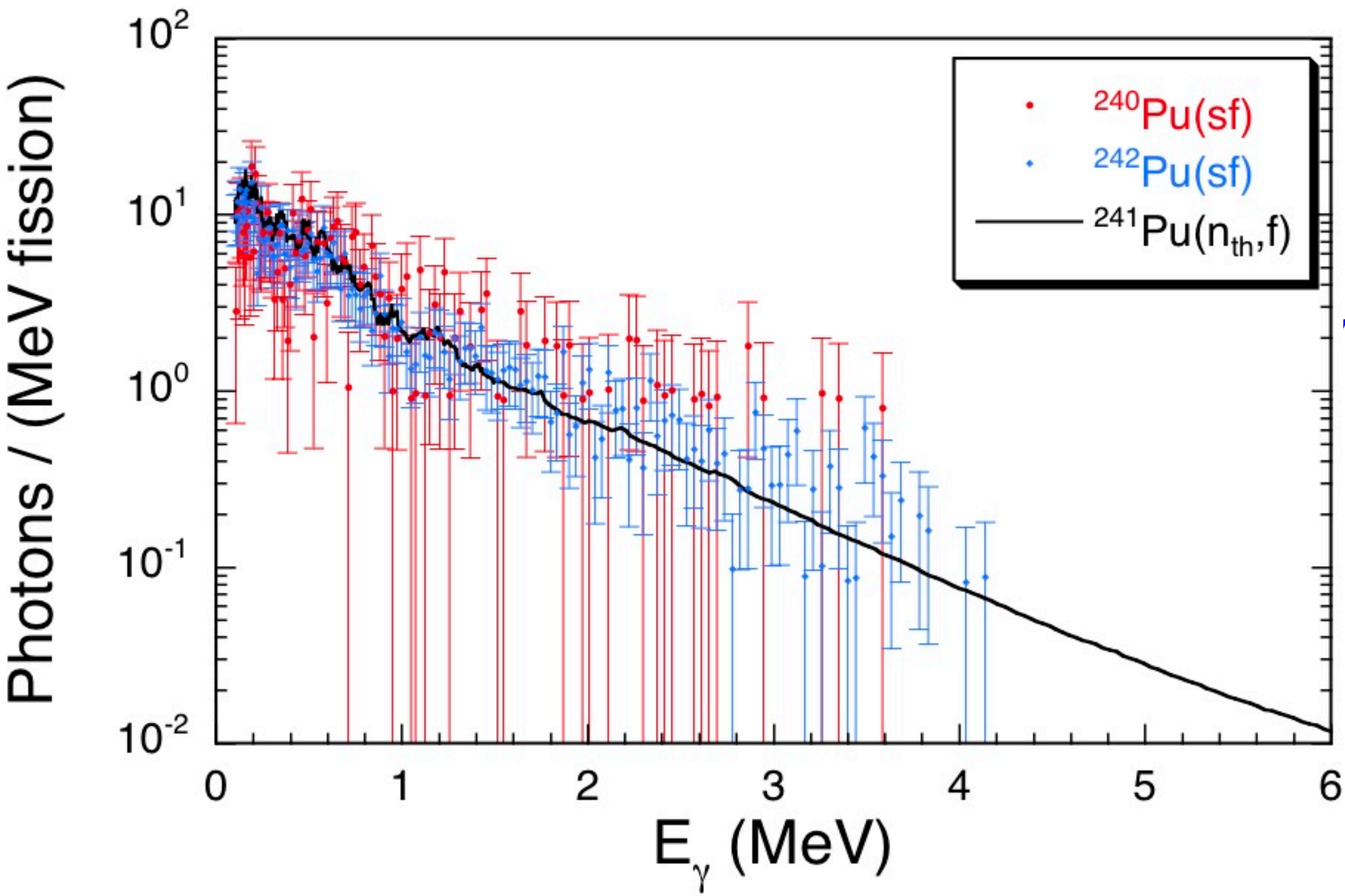

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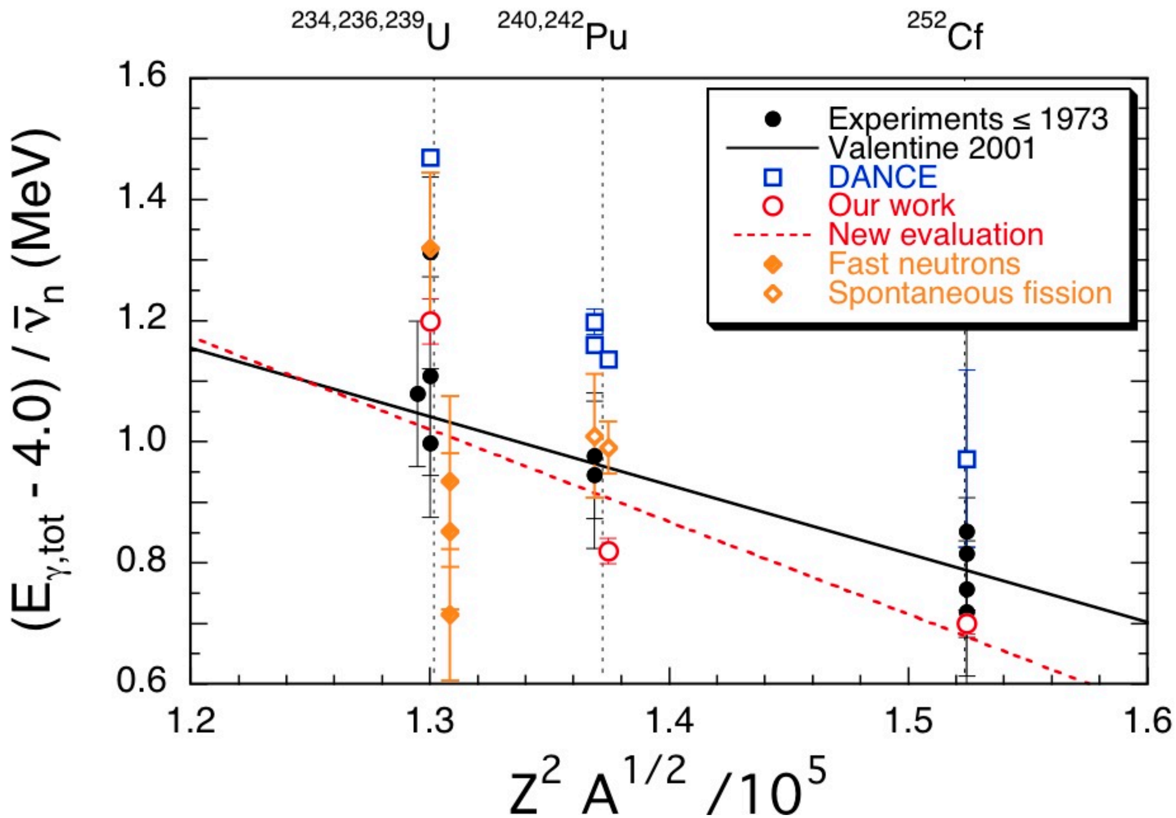
	^{240}Pu(sf)	^{242}Pu(sf)
$E_{\gamma,tot}$ =	(6.9±0.7) MeV	(6.9±0.3) MeV
$\overline{\nu}_\gamma$ =	(7.7±0.5)	(7.7±0.4)
ε_γ =	(0.9±0.1) MeV	(0.89±0.06) MeV

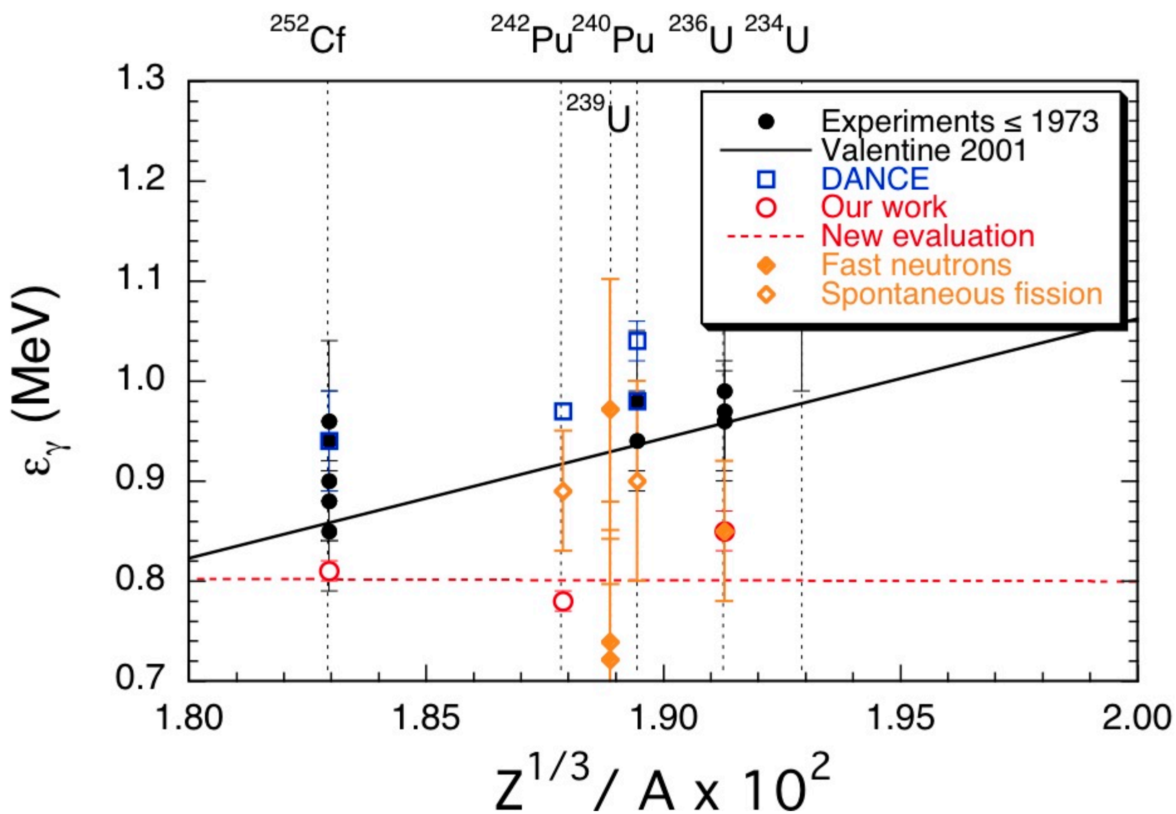
New evaluation

PFGS average total energy per fission

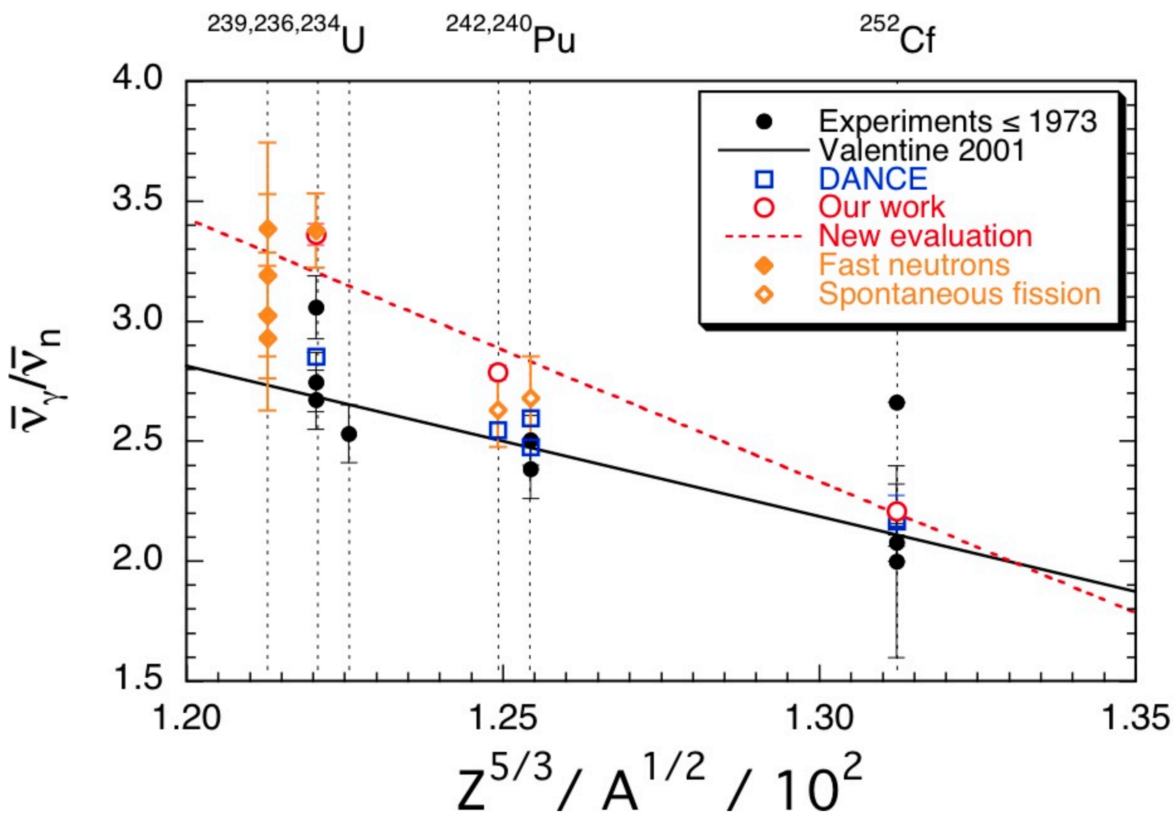


Observe: $\bar{\nu}_n$ taken from ENDF/B-VII.1.

New evaluation
PFGS mean energy per photon



New evaluation
PFGS average multiplicity



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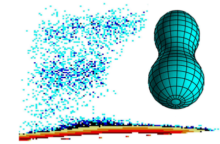

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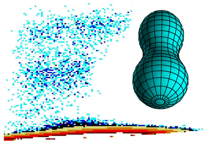
$\overline{\nu}_\gamma$ =	(7.7±0.5)	(7.7±0.4)
ε_γ =	(0.9±0.1) MeV	(0.89±0.06) MeV

- ^{235}U,$^{239,241}Pu(n, f)$ and $^{252}Cf(sf)$, DANCE (2014)
  - 160 BaF_2 detectors



Conclusions

- Systematics of PFGS characteristics as function of A and Z of the compound system makes sense, not only for thermal neutron induced and spontaneous fission
- Original parameters from Valentine's description need an adjustment
- Empirical A and Z dependence must be verified
- More experimental data needed
- Predictions for fast neutron induced fission of n + ^{238}U and n + ^{235}U presented
- Good agreement of our predictions with both calculations and preliminary experimental results!

Outlook

- From LICORNE experiment (E_n = 1.5 MeV):
  - final analysis of PFGS for ^{235,238}U(n, f) and ^{232}Th(n, f)
- Laborie et al.:
  - final analysis of PFGS for ^{238}U(n, f)
- IRMM:
  - final analysis of PFGS for ^{240,242}Pu(sf)

- New experiment at KFKI Budapest:
  - PFGS from ^{239}Pu(n_{th}, f) – spring 2015 (planned)
- New experiment with LICORNE at IPN Orsay:
  - PFGS from ^{239}Pu(n, f) – summer 2015 (planned)

- Updated systematics!
- New predictions!

The collaborators

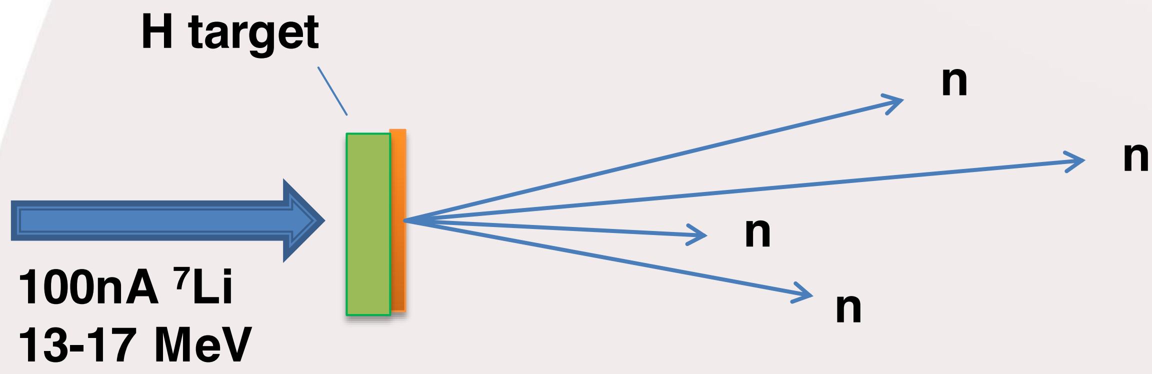
R. Billnert, A. Oberstedt, S. Oberstedt

with invaluable support from

T. Belgya, R. Borcea, T. Bryś, C. Chaves, Th. Gamboni, W. Geerts, A. Göök, C. Guerrero, F.-J. Hambsch, Z. Kis, M. Lebois, T. Martinez, L. Szentmiklosi, K. Takács, M. Vidali, J. Wilson and others

Parts of this work were supported by both the ERINDA programme (agreement number 269499) and the EFNUDAT programme (agreement number 31027) of the European Commission

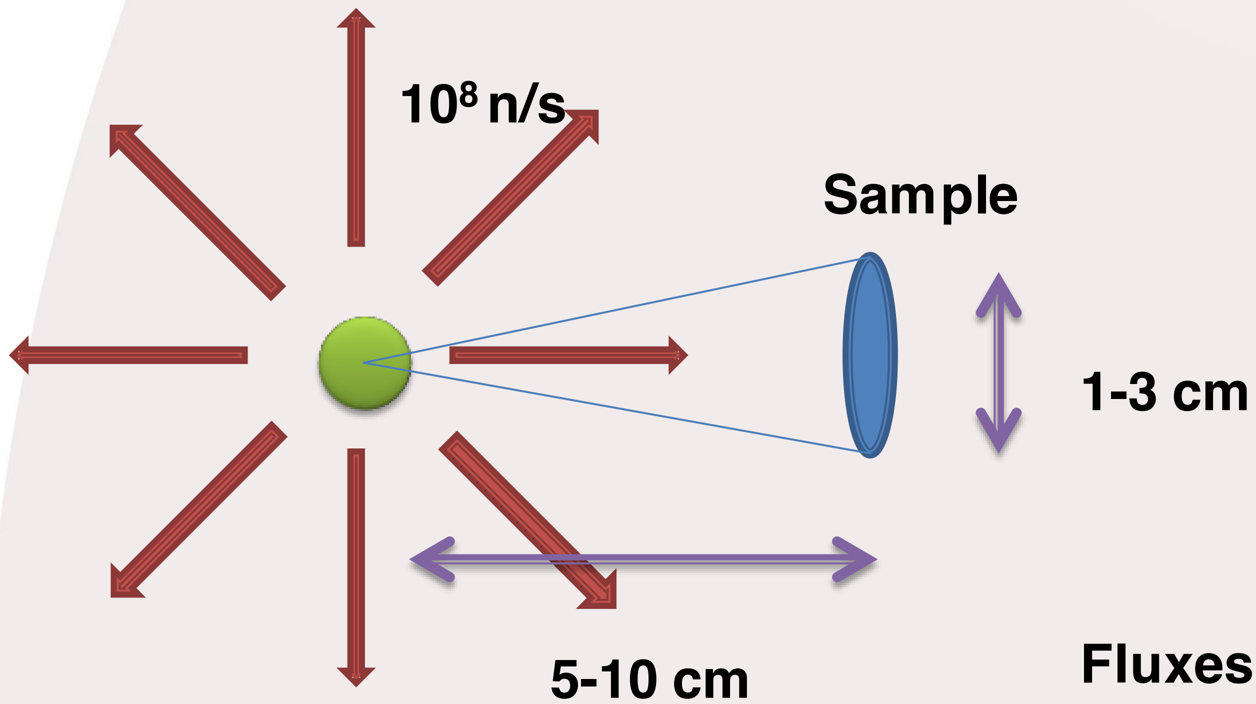
Thank you!



Lithium Inverse Cinematiques ORsay NEutron source

- **p(^{7}Li,^{7}Be)n reaction in inverse kinematics**

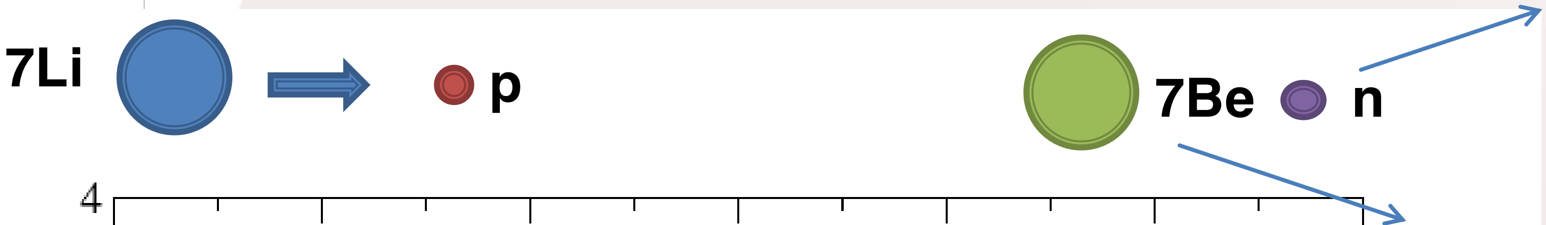
MONO-ENERGETIC NEUTRON SOURCES

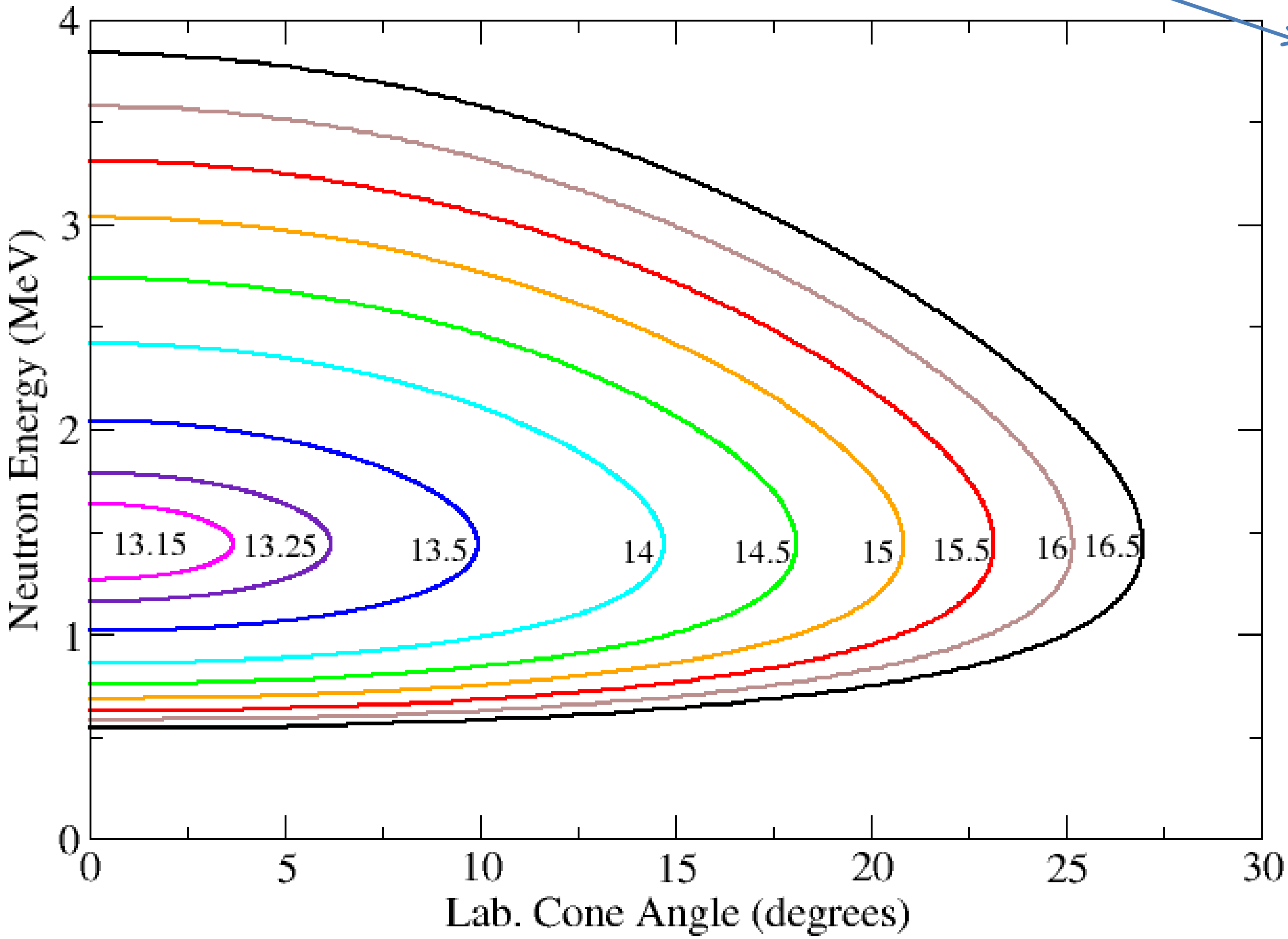


Fluxes typically
10^6 n/cm^2/s

- Typically over 99% of neutrons ''wasted"
- Wasted neutrons contribute to the room background
- Placement of gamma detectors impossible without heavy shielding

INVERSE KINEMATICS

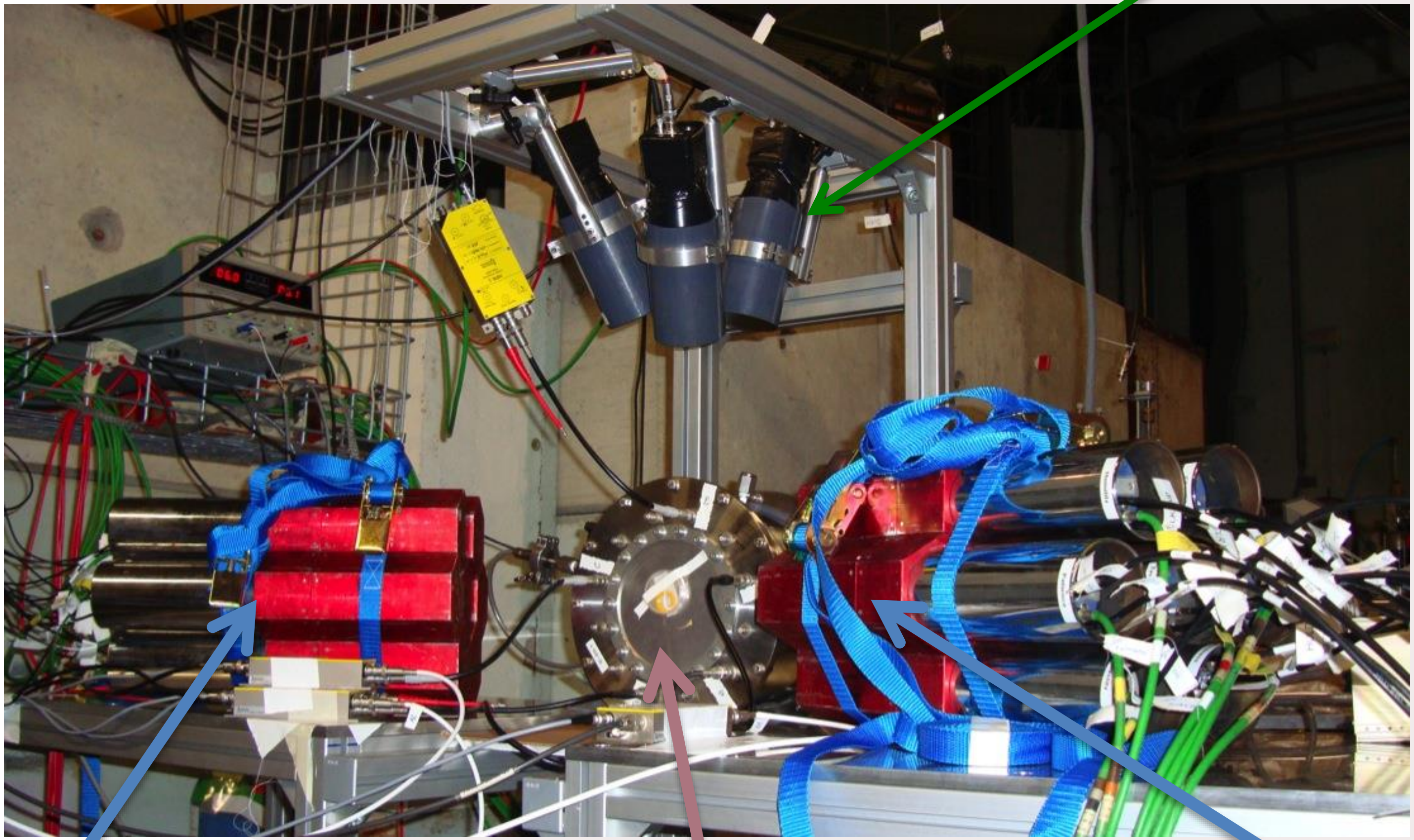




EXPERIMENTAL SETUP: MEASUREMENT OF PROMPT γ FOR FISSILE NUCLEI (JULY 2013)

IPN
INSTITUT DE PHYSIQUE NUCLÉAIRE
ORSAY

$LaBr_3$ from IPN & IRMM
(δt = 300 ps; δE = <3% @ .662 MeV)



BaF_2 from
Château de Cristal
(δt = 600 ps; δE = 10% @ 1.3 MeV)

Ionisation Chamber
$^{235-238}U$ target
10 mg (300 µg/cm^2); ø = 8 cm;
(δt = 700 ps; δE = 500 keV; ε = 100%)

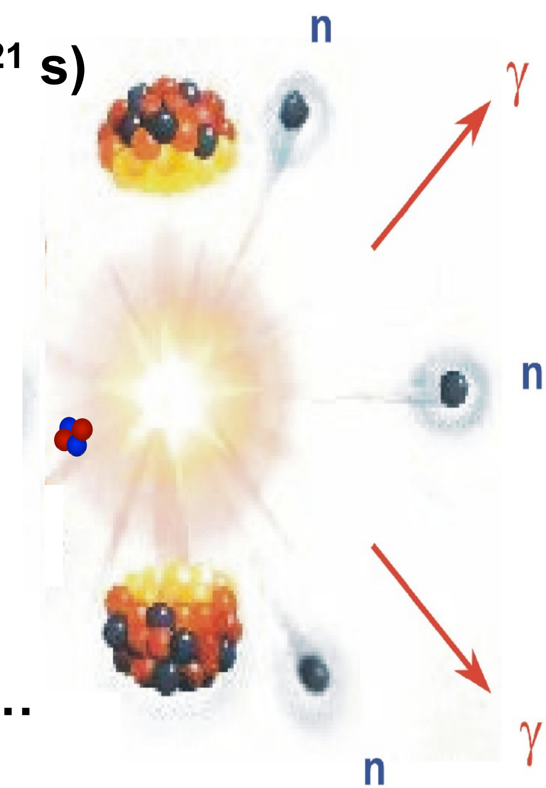
BaF_2 from
Château de Cristal
(δt = 600 ps; δE = 10% @ 1.3 MeV)

Prompt energy release in fission

prompt neutrons (< 10^{-18} s)

prompt γ-rays (10^{-16} s)

fission fragments (10^{-21} s)

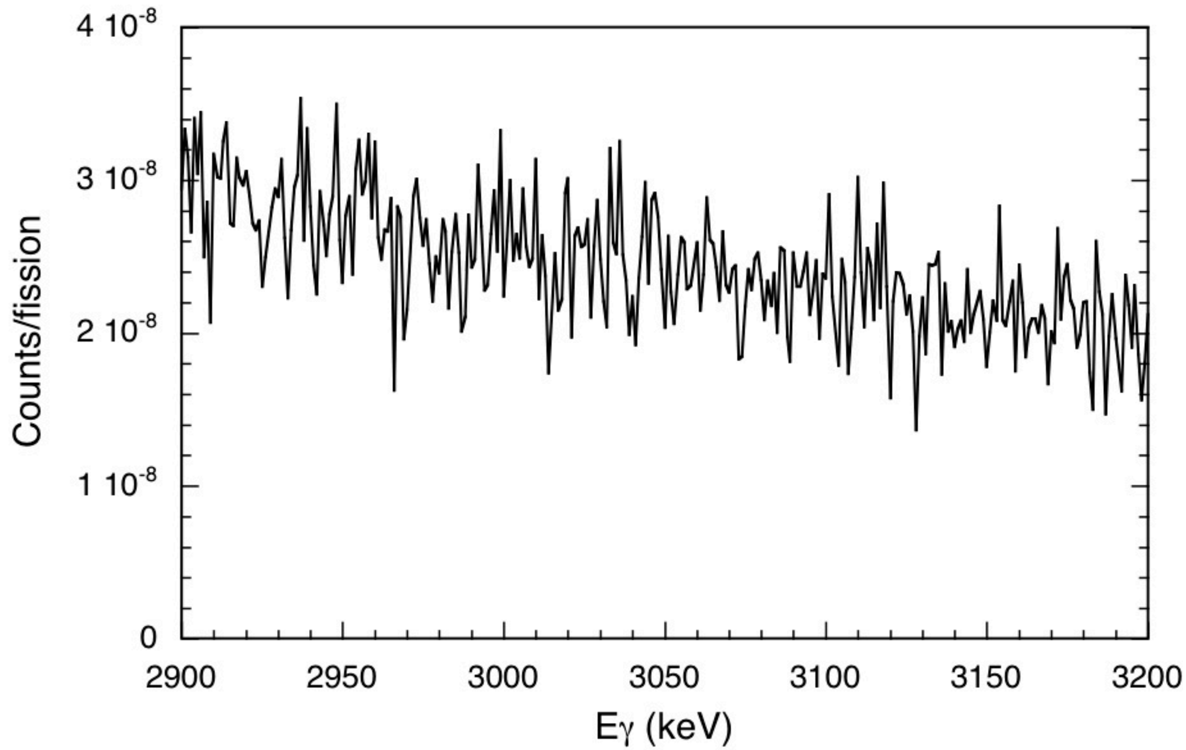


ternary α, t, d, ^{10}Be…

Prompt energy release:

- **kinetic energy of fission fragments**
- **prompt fission neutron emission**
- **prompt fission γ-ray emission**

Unfolding response function
(an illustration)

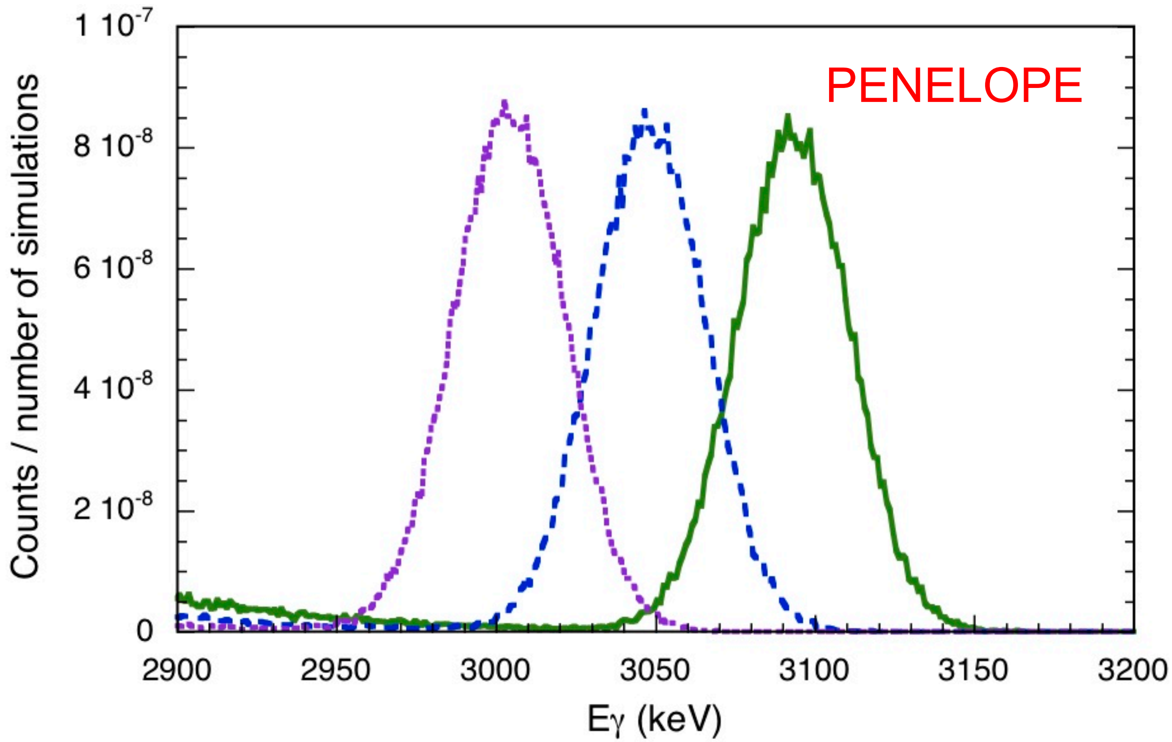


Measured ^{252}Cf(sf) prompt fission γ-ray energy spectrum

→ e.g. zooming into region around 3 MeV

Unfolding response function

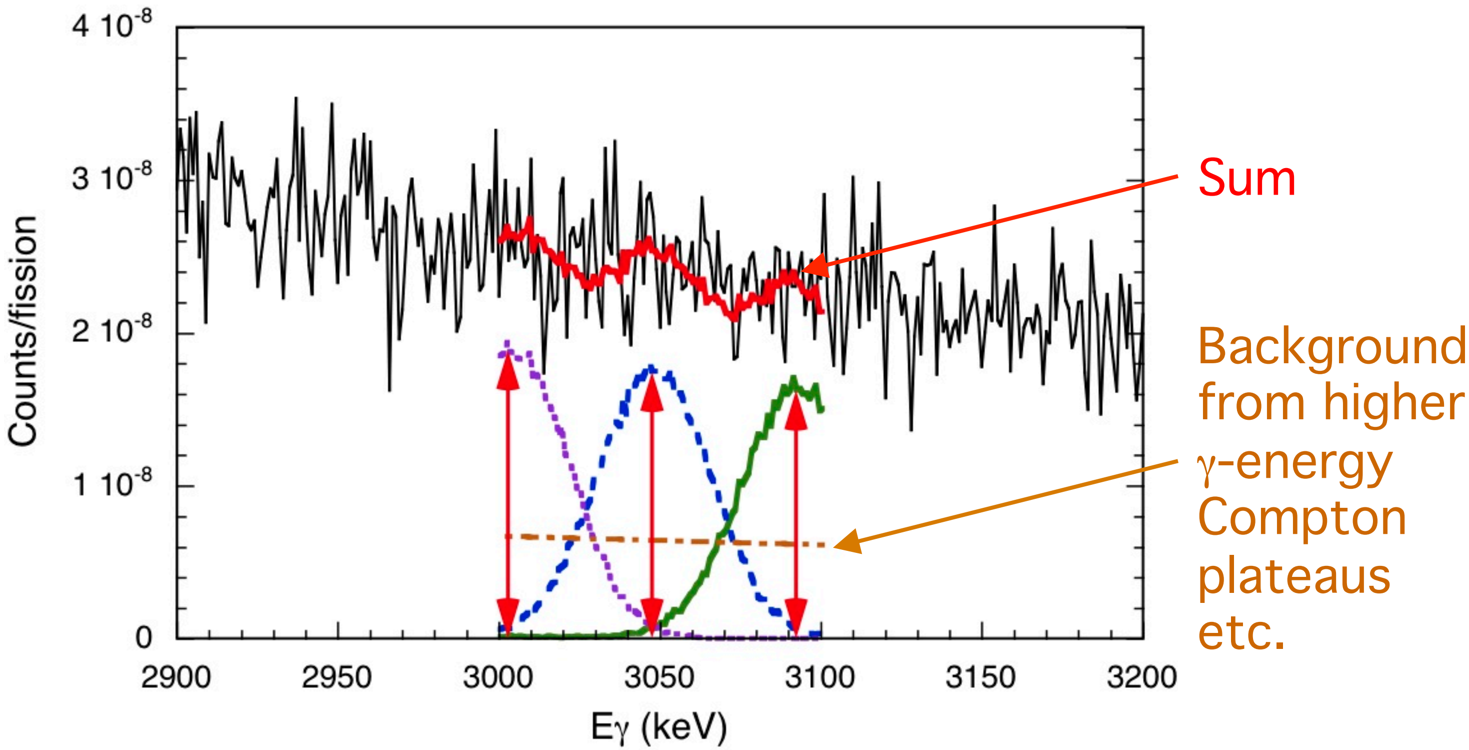
(an illustration)



Simulating response function for mono-energetic γ-rays,

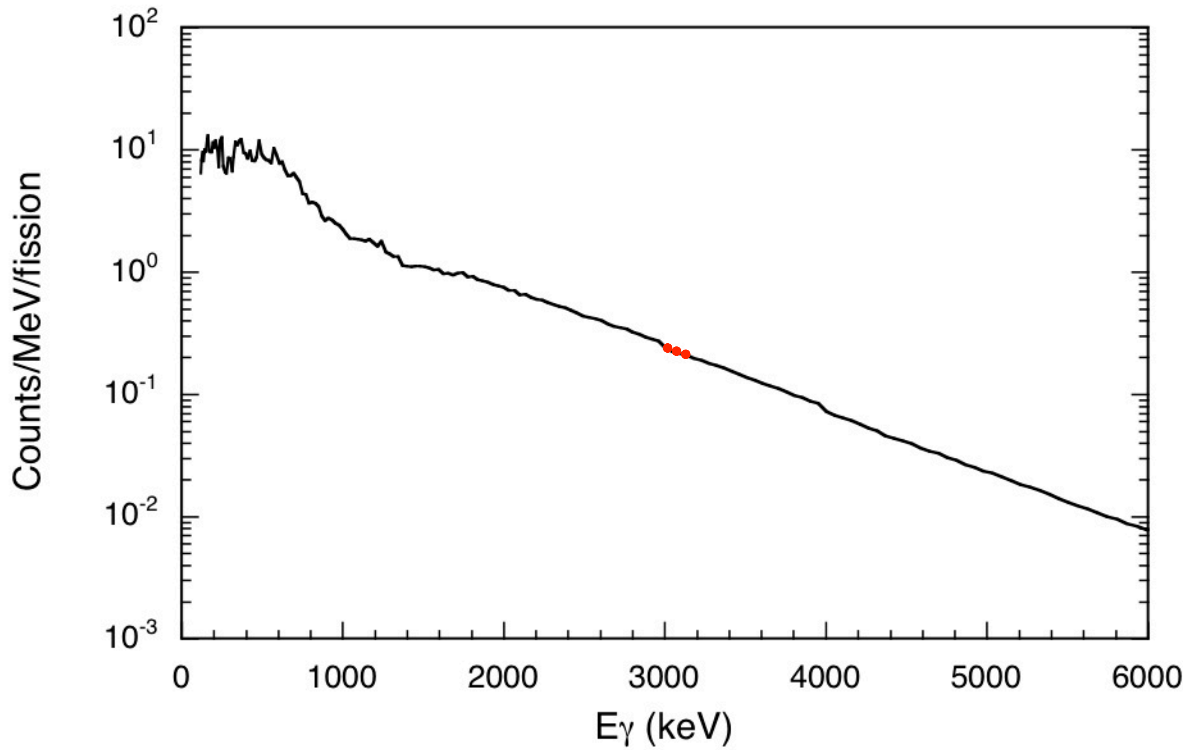
distance: FWHM from energy resolution measurements

Unfolding response function
(an illustration)



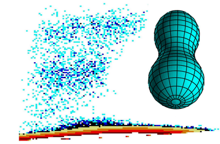
Adjusting simulated spectra to measured γ-ray spectrum and determining the scaling factors

Unfolding response function
(an illustration)



Properly normalized scaling factors

→ emission spectrum!



Fast neutron induced fission

Prerequisites

According to Valentine:

- $E_{\gamma,tot}$ depending linearly on $\overline{\nu}_n$, ε_γ independent
- $\overline{\nu}_\gamma$ approximately proportional to $\overline{\nu}_n$
- Knowledge of $\overline{\nu}_n(E_n)$ important

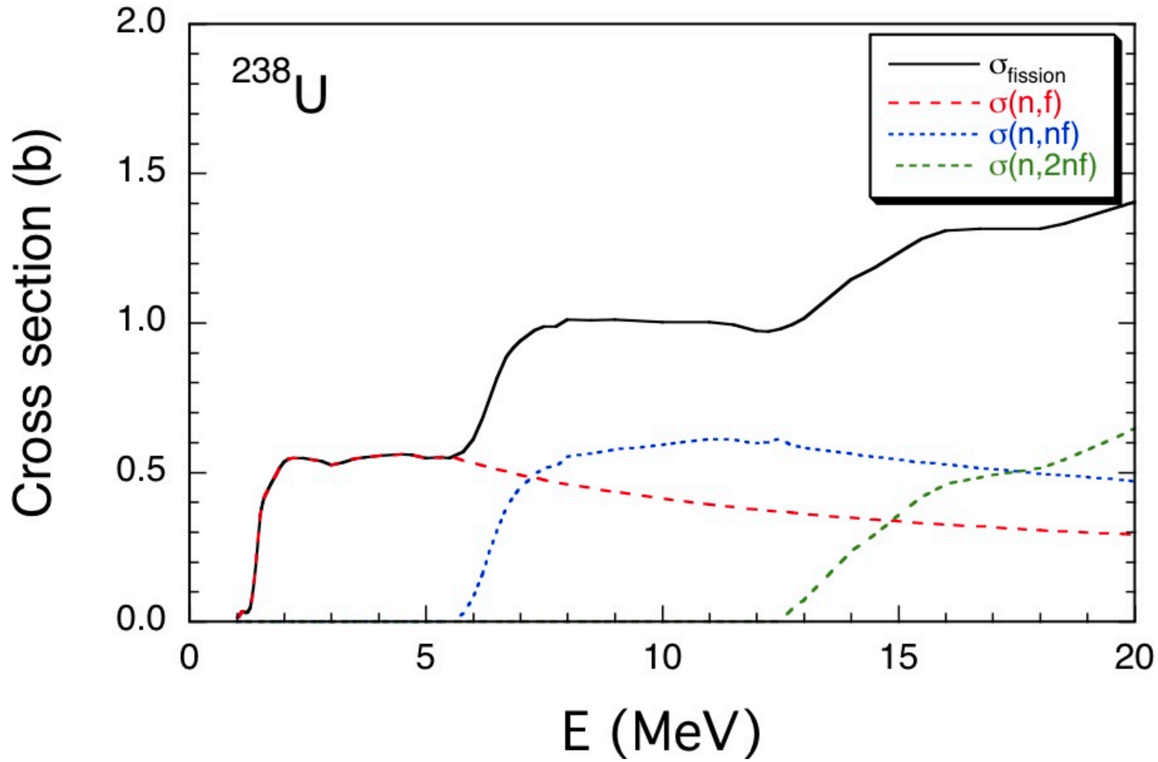
In case of multiple chance fission:

- (n, f) cross section has to be known (ENDF/B-VII.1)
- Contributions from different fission channels have to be taken into account
- $\overline{\nu}_n(E_n)$ for all fissioning systems (ENDF/B-VII.1)

^{238}U(n, f) PFGS characteristics

Fission cross section

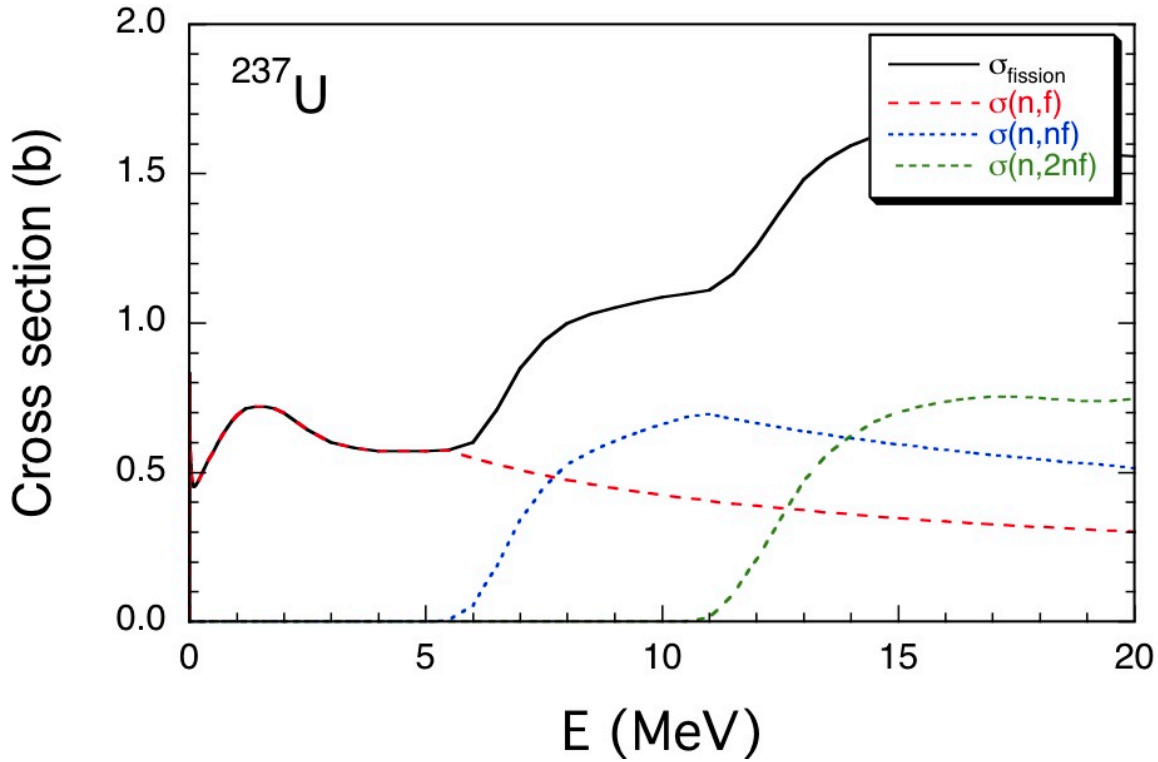
ENDB/B-VII.1



Contributions from different fission channels.

^{238}U(n, f) PFGS characteristics
Fission cross section

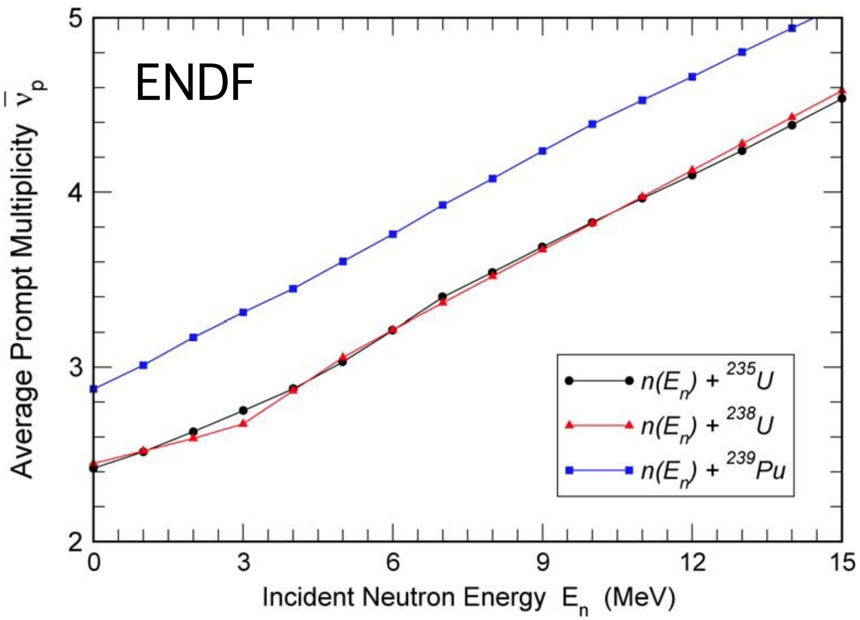
ENDB/B-VII.1



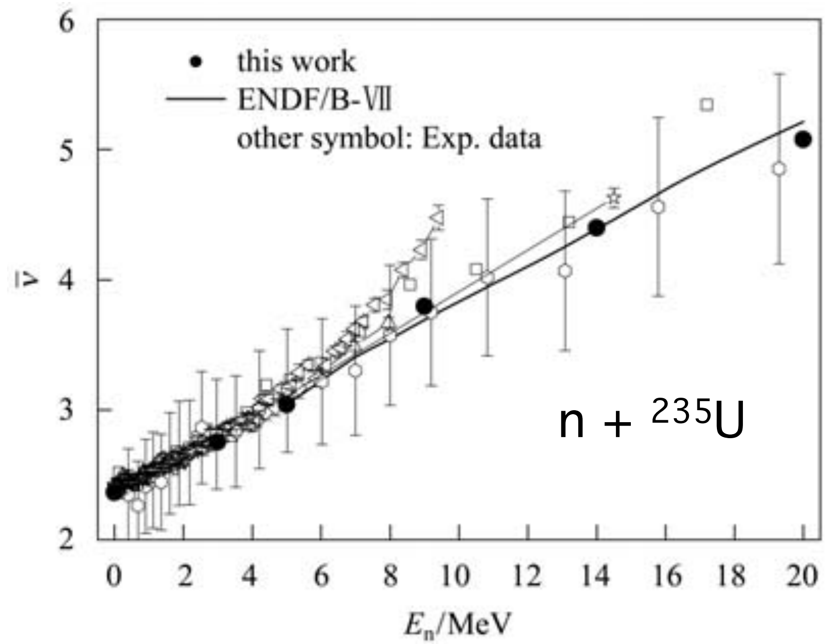
Accordingly for ^{237}U(n, f) ...

^{238}U(n, f) PFGS characteristics

Prompt fission neutron multiplicity



Madland, NPA 772 (2006) 113

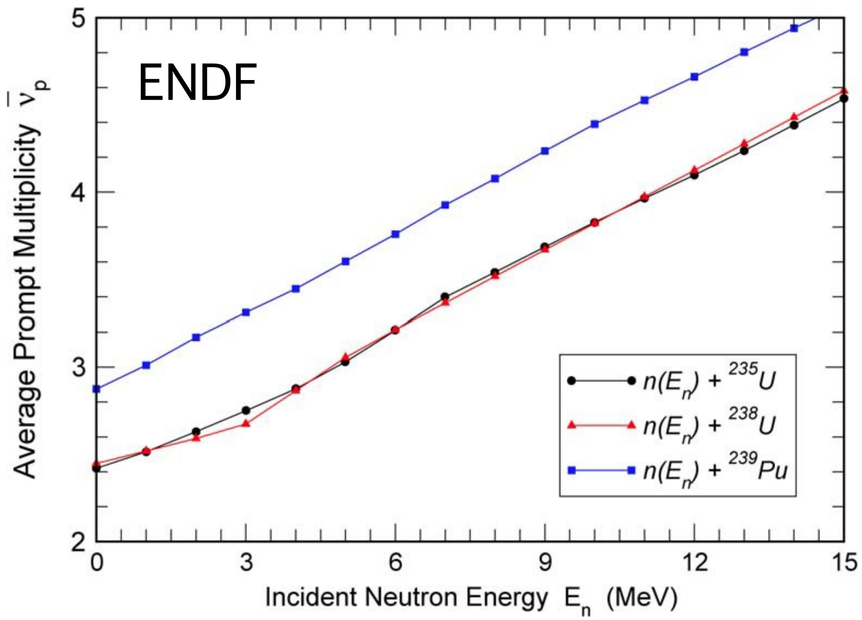


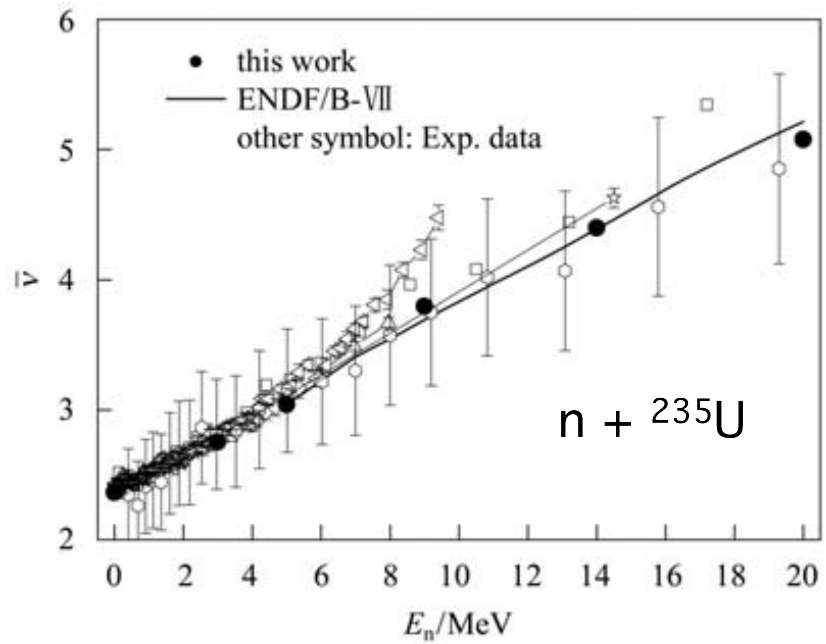
Chen & Liu, CPC 35,4 (2011) 341

Chen & Liu: prompt fission neutrons = evaporation neutrons (pre-fission) + prompt neutrons from fission fragments!

^{238}U(n, f) PFGS characteristics

Prompt fission neutron multiplicity





Madland, NPA 772 (2006) 113

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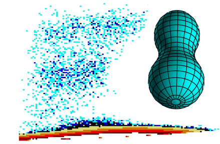
Chen & Liu: prompt fission neutrons =
evaporation neutrons (pre-fission) +
prompt neutrons from fission fragments!

Claim: only the latter may be related to PFGS!



^{238}U(n, f) PFGS characteristics
Prompt fission neutron multiplicity

Energetics:

$$E_x^{A_{CN}} = S_n^{A_{CN}} + E_n; \qquad \langle E_n \rangle = \frac{3}{2}T = \frac{3}{2}\sqrt{\frac{7.524MeV \cdot E_x^{A_{CN}}}{A_{CN}}} \quad *)$$

$$E_x^{A_{CN}-1} = S_n^{A_{CN}-1} + E'_n = E_x^{A_{CN}} - S_n^{A_{CN}} - \langle E_n \rangle; \qquad \langle E'_n \rangle = \frac{3}{2}T' = \frac{3}{2}\sqrt{\frac{7.524MeV \cdot E_x^{A_{CN}-1}}{A_{CN}-1}}$$

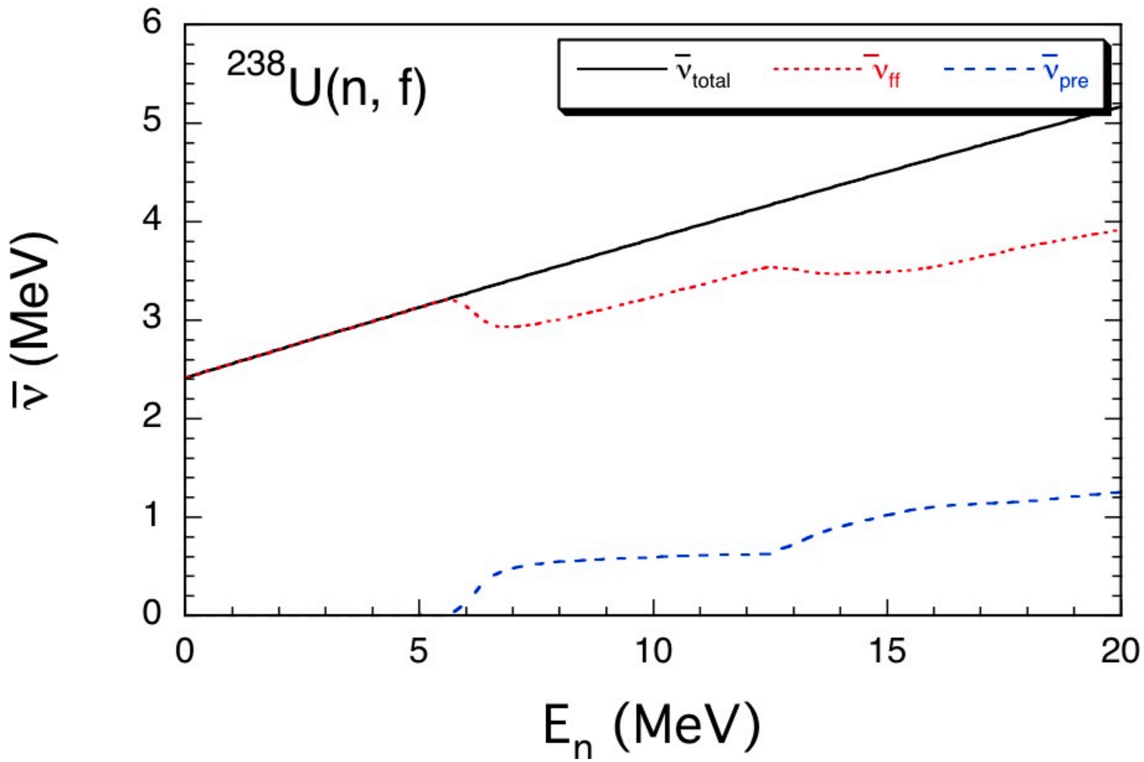
$$E_x^{A_{CN}-2} = S_n^{A_{CN}-2} + E''_n = E_x^{A_{CN}-1} - S_n^{A_{CN}-1} - \langle E'_n \rangle; \qquad \langle E''_n \rangle = \frac{3}{2}T'' = \frac{3}{2}\sqrt{\frac{7.524MeV \cdot E_x^{A_{CN}-2}}{A_{CN}-2}}$$

etc.

*) According to Chen & Liu: $E_x^{A_{CN}} = \dfrac{A_{CN}}{7.524MeV}T^2$

^{238}U(n, f) PFGS characteristics

Prompt fission neutron multiplicity



- prompt fission neutrons from ENDF/B-VII.1
- pre-fission neutrons subtracted
- prompt neutrons from fragments for PFGS characteristics