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Joint Research Centre

Experimental Measurements of Prompt Fission Neutrons and γ-rays

F.-J. Hambsch

FIESTA2017 School and Workshop, Santa Fe, NM, Sept. 18 – 22, 2017

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Table of content

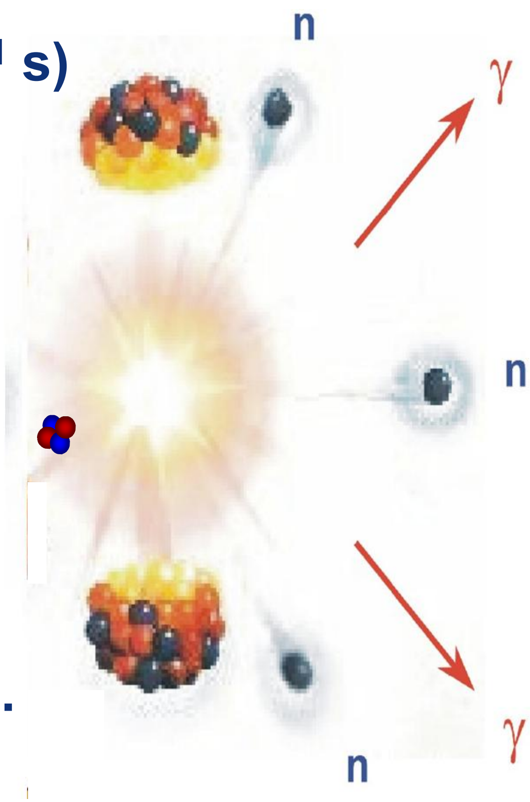
- General aspects of neutron and γ-emission
- Prompt neutron measurements
- Prompt γ-ray measurements
- Conclusions

The fission process

prompt neutrons (10^{-18} s)

fission fragments (10^{-21} s)

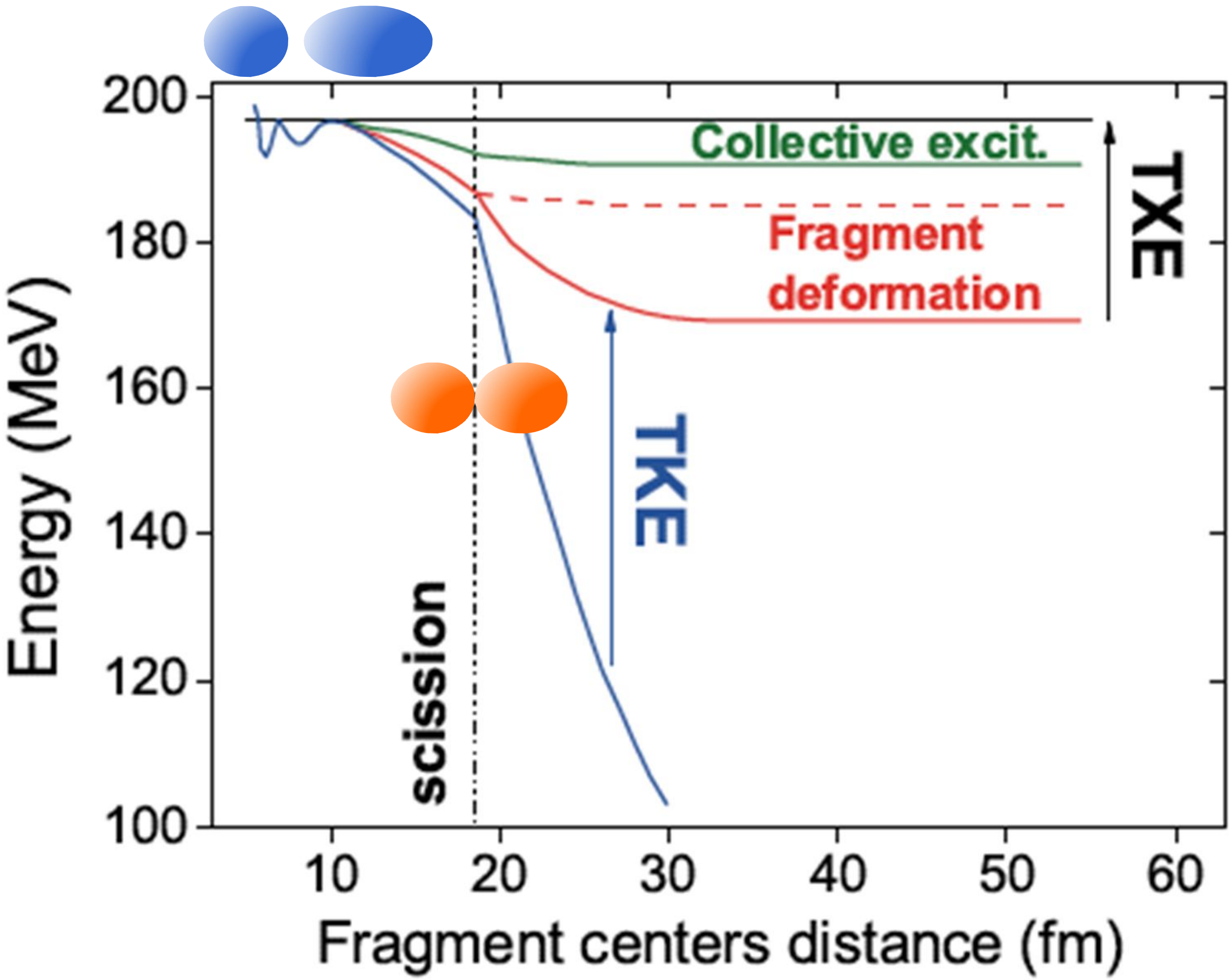
prompt γ-rays (10^{-16} s)



ternary α, t, d, ^{10}Be…

kinetic energy prompt γ-rays	heat
prompt neutrons (delayed neutrons)	chain reaction
ternary α, t, d	gas production in the fuel (waste)
fission fragments	decay heat, toxicity (waste)

The fission process



Compared to eV in chemical reactions

Fission fragment de-excitation

- Excitation energy of the fragments is dissipated through particle emission, here essentially neutrons and γ-rays

- On average 2 – 4 neutrons are released

- The exact value depends on the isotope and the excitation energy of the compound nucleus e.g. for ^{235}U ~2.4)

- The average energy of a neutron in the LS is around 2 MeV showing a white spectrum

- On average 6 – 10 γ-rays are emitted too, with a mean total energy release of about 7 – 9 MeV

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Fission fragment de-excitation

- **Observable quantities:**
  - **Spectral characteristics (neutrons and γ-rays)**
    - **Average multiplicity** (/fission)
    - **Average total energy** (/fission)
    - **Average photon energy** (/fission)
  - **Correlations with fission fragment characteristics**
    - **ν(A*, TKE), $<E_{tot}>$(A*, TKE),**
  - **Correlation of prompt γ-ray data with PFN**

How to measure neutrons and γ-rays

- Prompt neutron measurements

How to measure neutrons

- Suitable detectors

- Any material with a high capture cross section
  for neutrons (H, He, Li, B, Gd)

  - He-3 counters
  - Boron (BF_3) counters
  - Lithium-glass detectors
  - Liquid scintillator detectors (mainly containing H)

Choice of material (large interaction cross section)

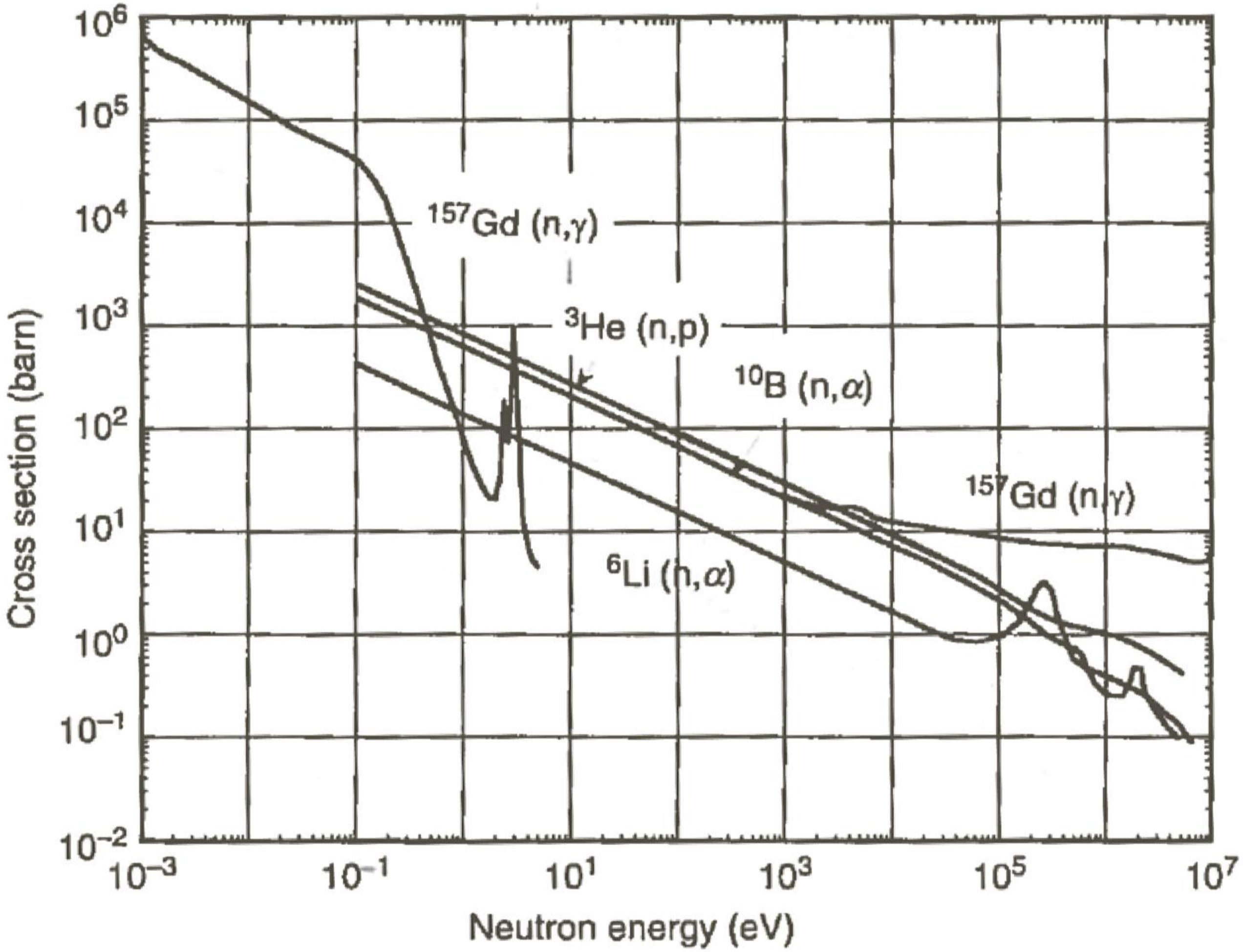

Figure 3.30 Neutron cross sections for several isotopes. Data from Garber and Kinsey (1976).

F. Sauli, Gaseous Radiation Detectors, Cambridge U. Press, 2014

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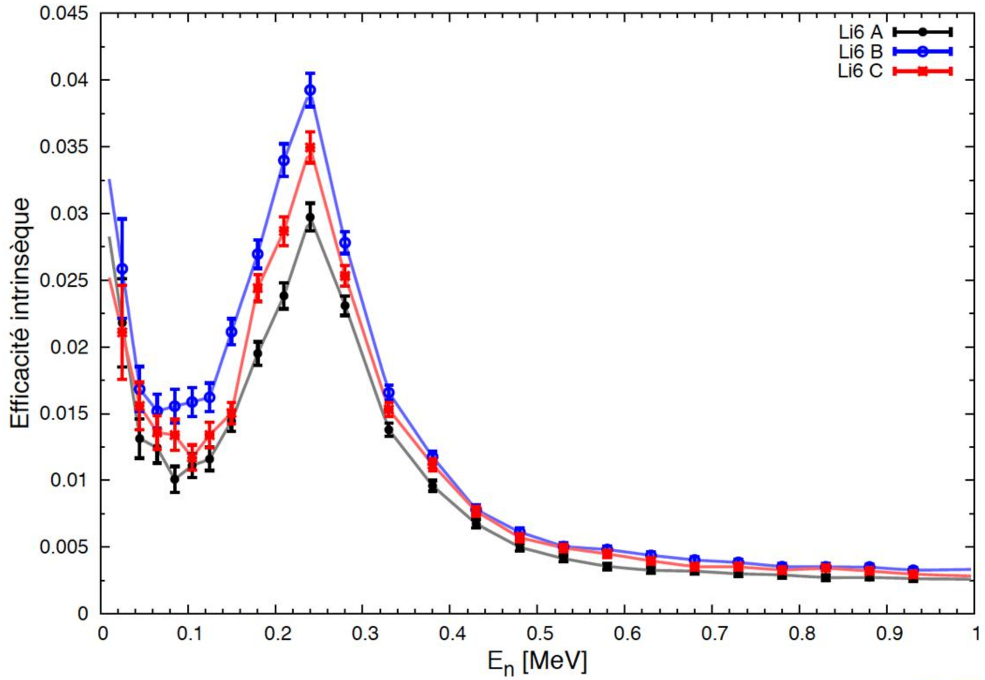
How to measure neutrons

- Lithium-glass detectors:
  - Enriched in ^{6}Li: $^{6}Li(n,t)\alpha$
  - Exothermic reaction: Q = 4.8 MeV

- CONs:
  - Relatively low detection efficiency
  - Bad timing resolution -> need for small crystals



PhD thesis, A. Sardet, Université Paris Sud (2015)

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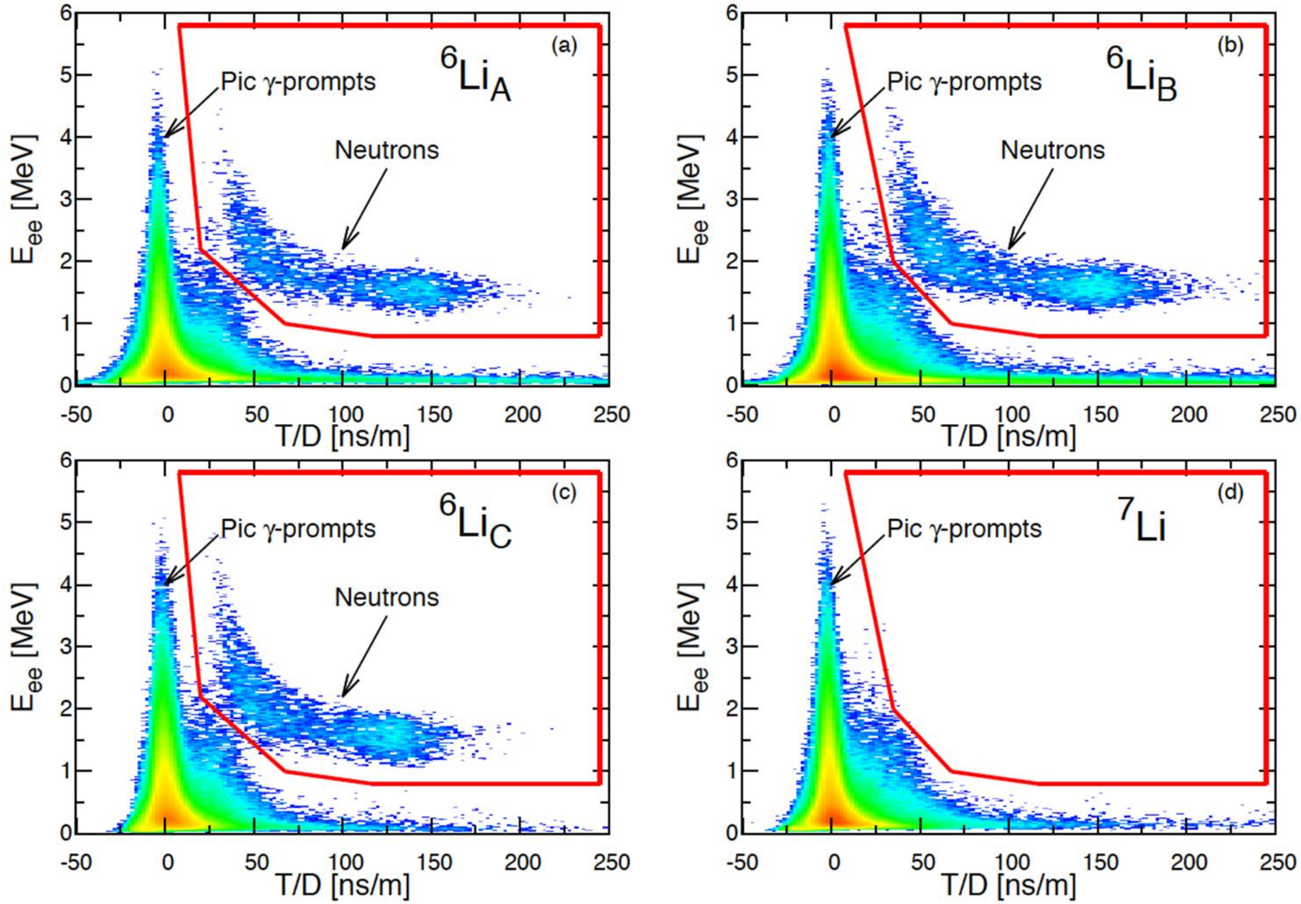
How to measure neutrons

➢ **Lithium-glass detectors:**

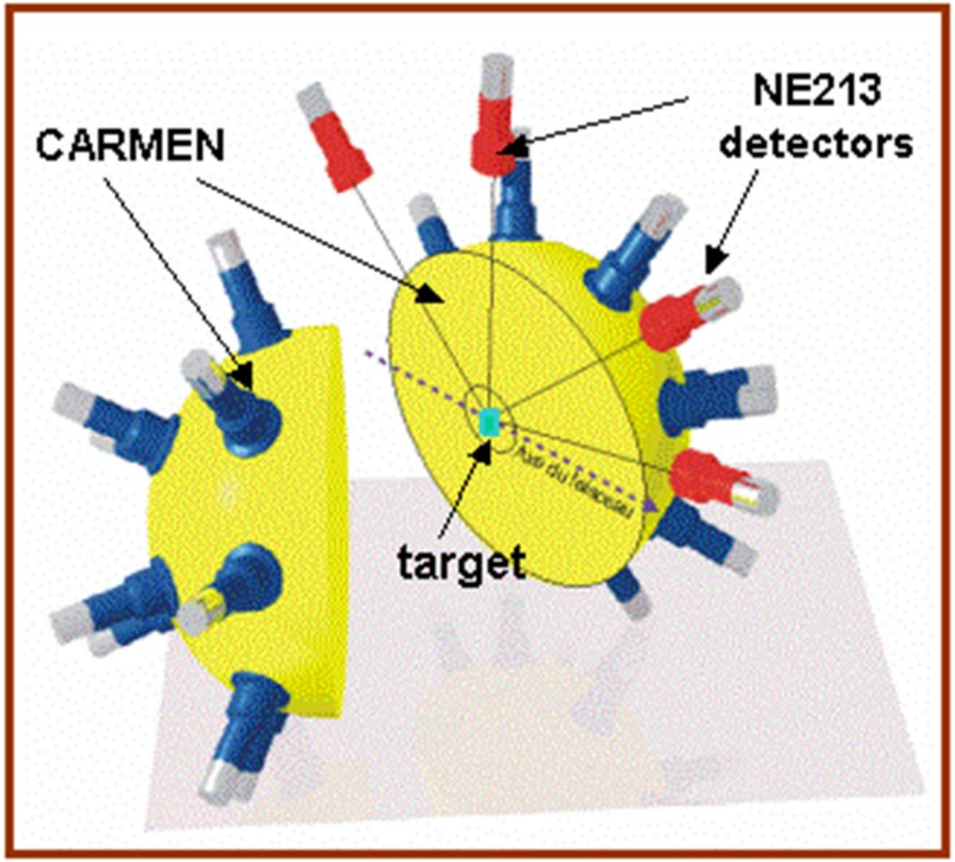


Determine background

PhD thesis, A. Sardet, Université Paris Sud (2015)

How to measure neutrons

- **Detectors based on liquid scintillator(s):**
  - **Gd loaded scintillation tanks**
  - **NE213 equivalent scintillators (pulse shape discrimination)**



Liquid scintillation tank

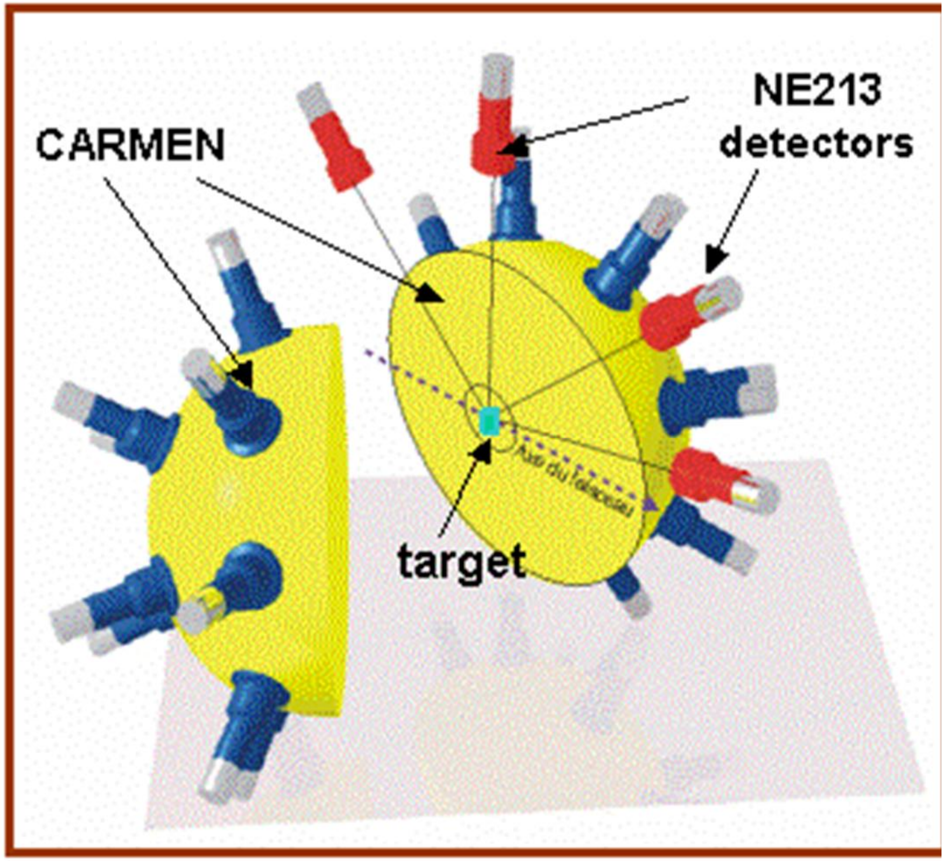
- **Pros:**
- **High efficiency (4π geometry)**
- **Large thermal neutron capture cross section of Gd isotopes**
- **Release of a high energy gamma cascade**

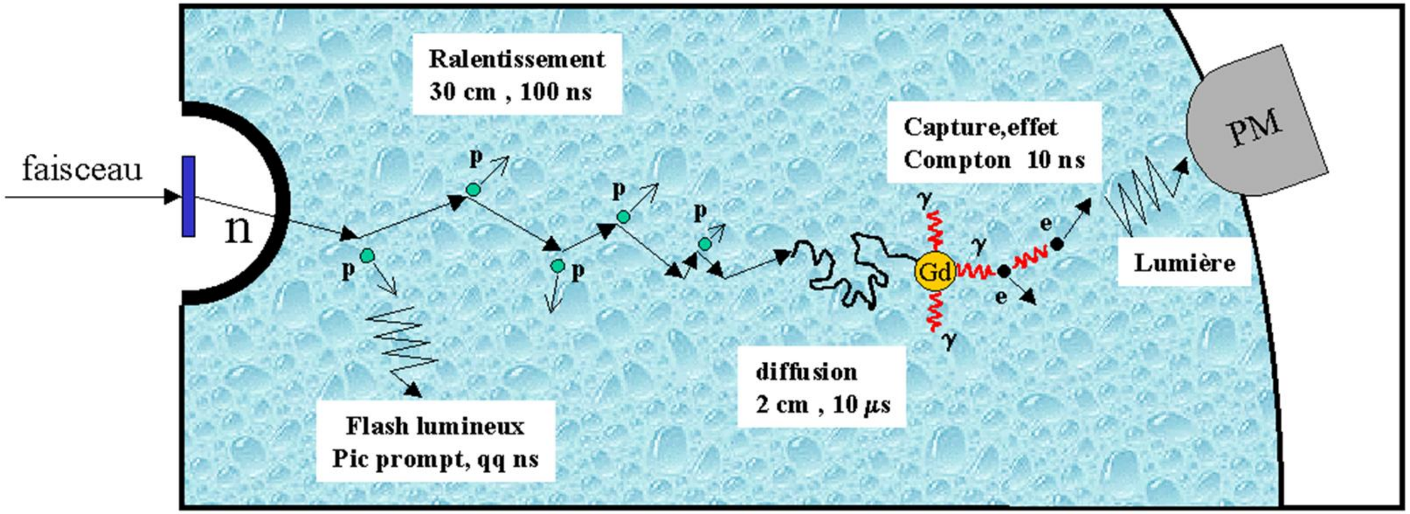
- **Cons:**
- **High toxicity (xylene- or dioxane-based) scintillator**
- **Low flash point -> highly flammable**
- **NO pulse shape discrimination**

Liquid scintillation tank (CEA-Arpajon)

- 85 % eff for fission neutrons
- Two hemispheres (r=60cm)
- 950l (C_9H_{12}) with 0.5% Gd
- 24 phototubes

- Can be combined with other Detectors (e.g. NE213)





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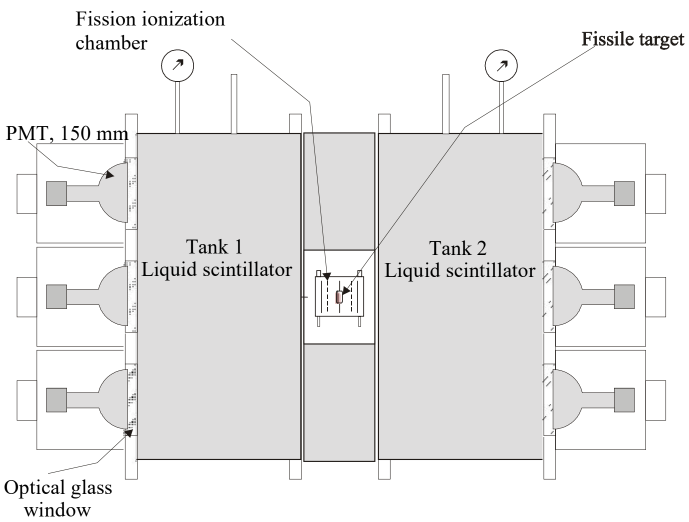
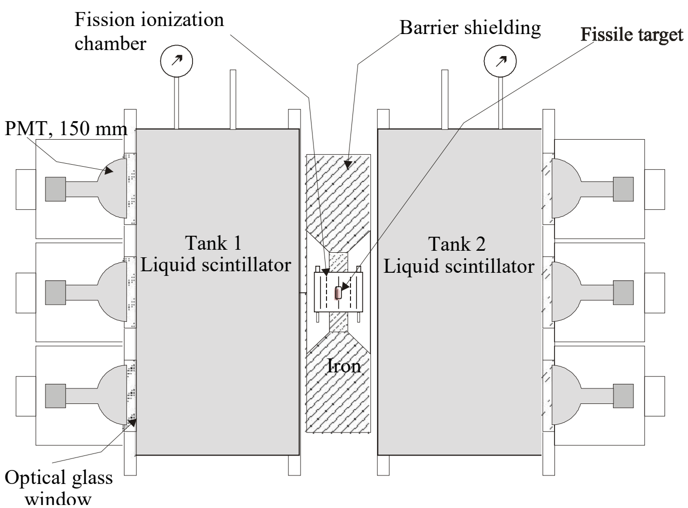
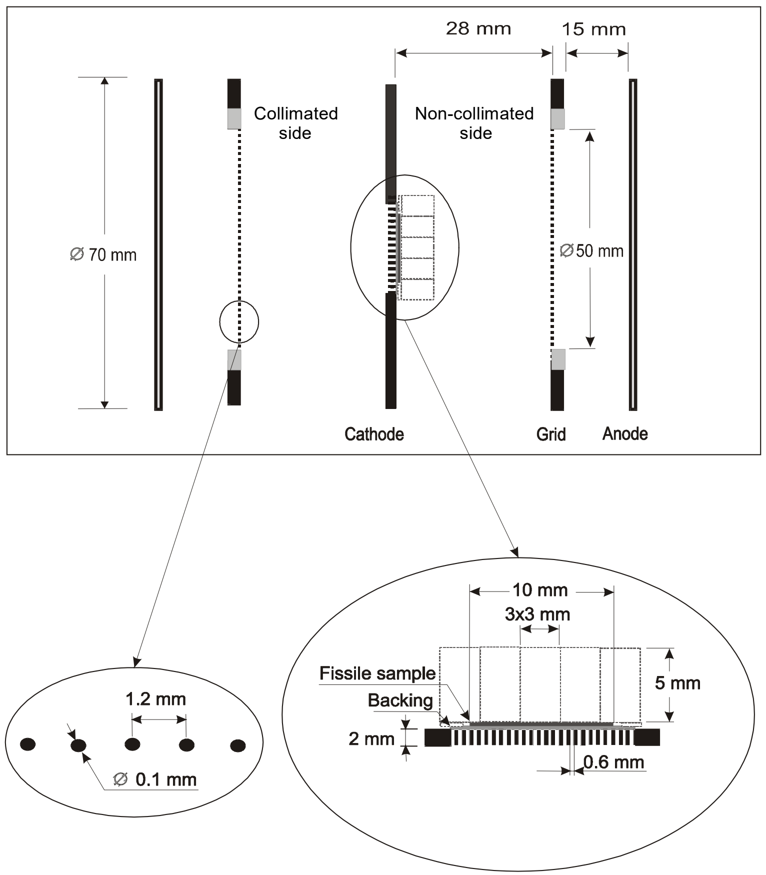
Liquid scintillation tank (used at PNPI, Gatchina, Russia)

4π-geometry



2 x 2π- geometry





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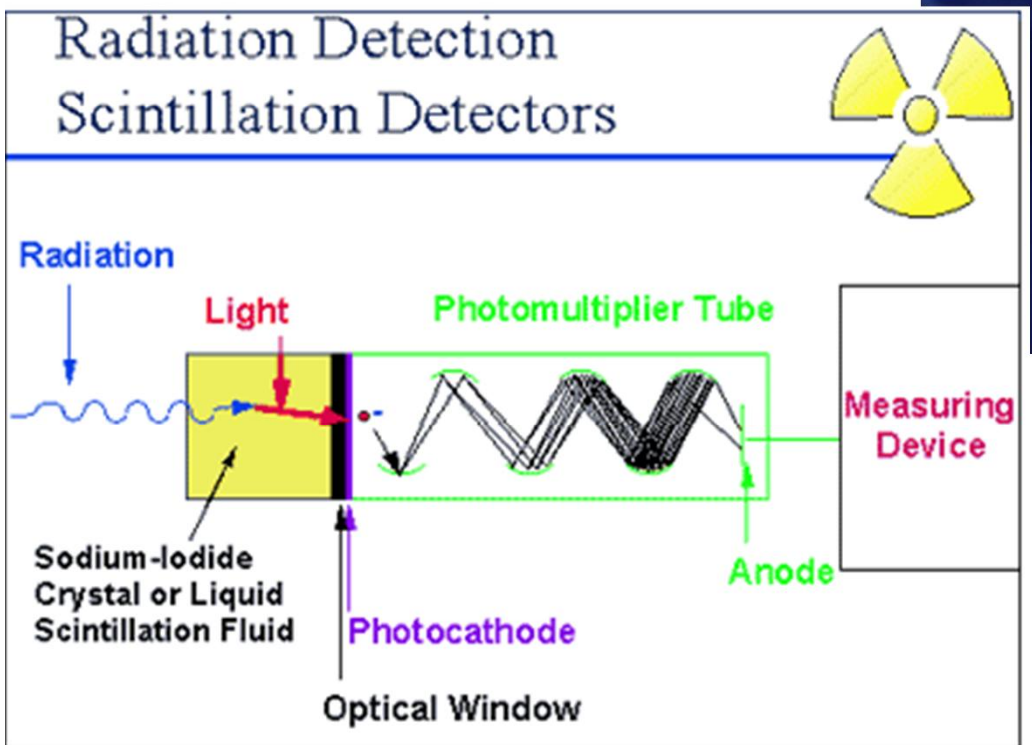
NE213 equivalent scintillation detectors and photomultipliers

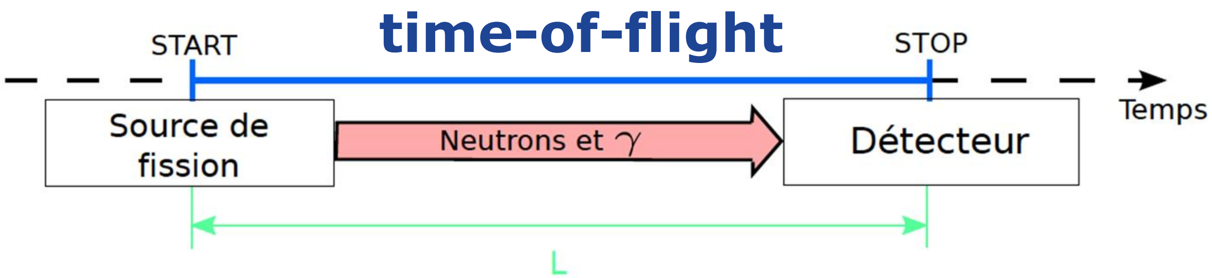


Electron amplification orders of mag.

Source: WWW

How to measure neutrons

- **Prompt neutron measurements (white spectrum)**



- **Relativistic equation**

$$E_n = (\gamma - 1)m_n c^2 = \left(\frac{1}{\sqrt{1 - \frac{L^2}{\Delta t^2} \frac{1}{c^2}}} - 1 \right) m_n c^2$$

  - m_n = 939.56533 MeV/c^2
  - c = 0.299792458 m/ns
  - Δt : time of flight (TOF)

$$\left(\frac{\sigma_E}{E}\right)^2 = 2\left[\left(\frac{\sigma_L}{L}\right)^2 + \left(\frac{\sigma_{\Delta t}}{\Delta t}\right)^2\right]$$

PhD thesis, A. Sardet, Université Paris Sud (2015)

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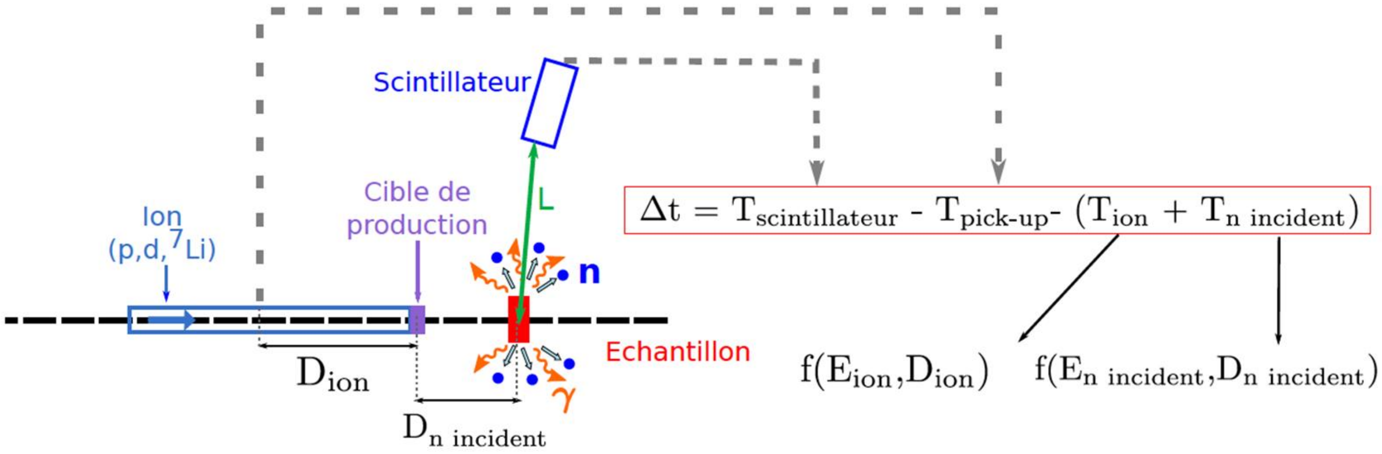
How to measure neutrons

- **Measurement with passive sample**



- **Use of massive targets (several g)**
- **Pulsed neutron beam (usually low beam currents; 1 – 2 μA)**
- **In general leads to a sufficiently high event rate**

- **Resolution depends on beam pulse**
- **Minimum neutron energy depend on incident neutron energy**
- **Multiple scattering in the sample**

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How to measure neutrons

- The measurement environment (direct reaction):

- Limited number of detectors
- Contributions from neutron scattering
- Simulation by means of MCNP or Geant4

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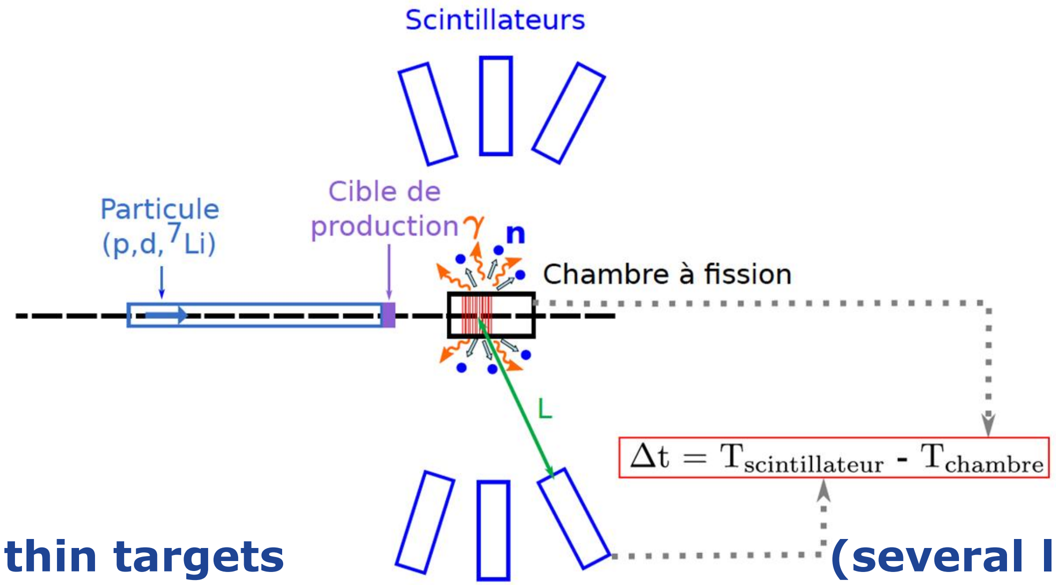
How to measure neutrons

- **Measurement with an active sample**



- **Use of thin targets (several layers)**
- **Continuous neutron beam (high beam currents; > 20 μA)**
- **Allows to measure neutrons below the beam energy**
- **Allows measuring at different energies with changing particle beam**

- **Multiple scattering in the detector to be taken care of**

PhD thesis, A. Sardet, Université Paris Sud (2015)

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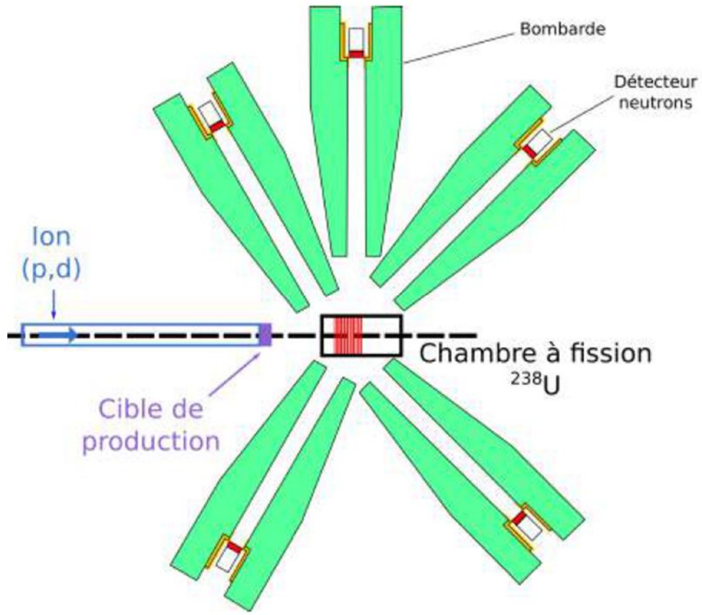
How to measure neutrons

- The measurement environment (direct reaction):

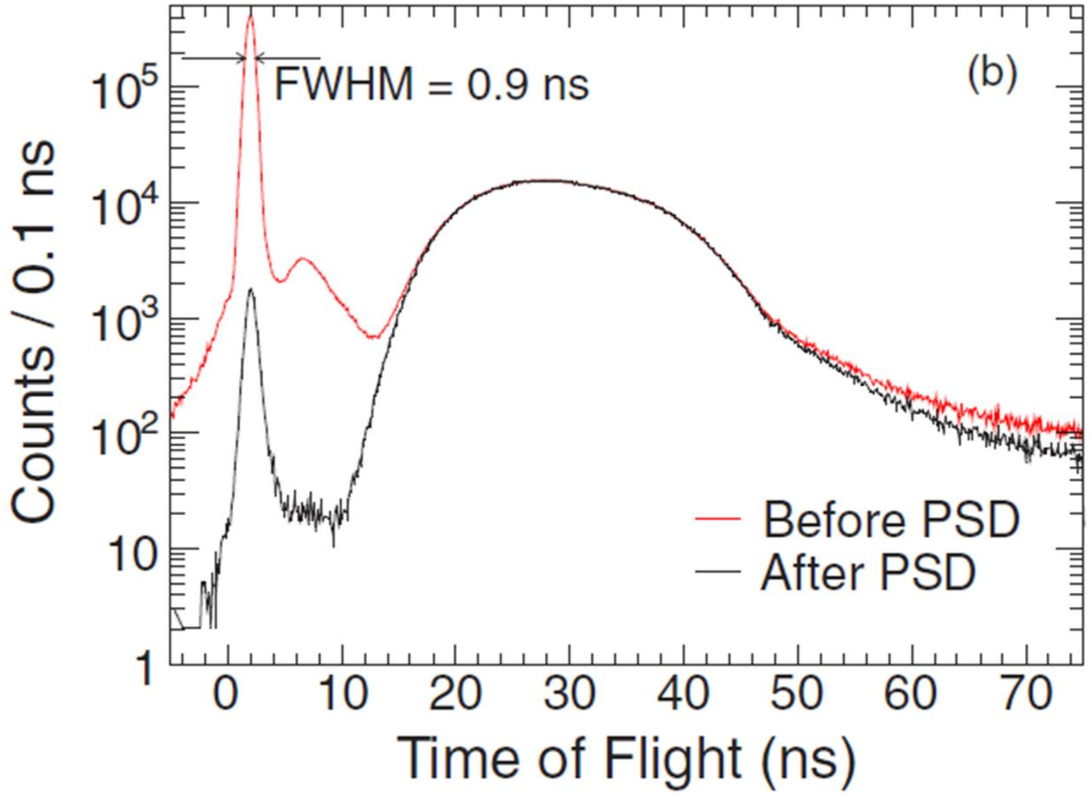


- Limited number of detectors
- Contributions from neutron scattering…

PhD thesis, A. Sardet, Université Paris Sud (2015)

How to measure neutrons

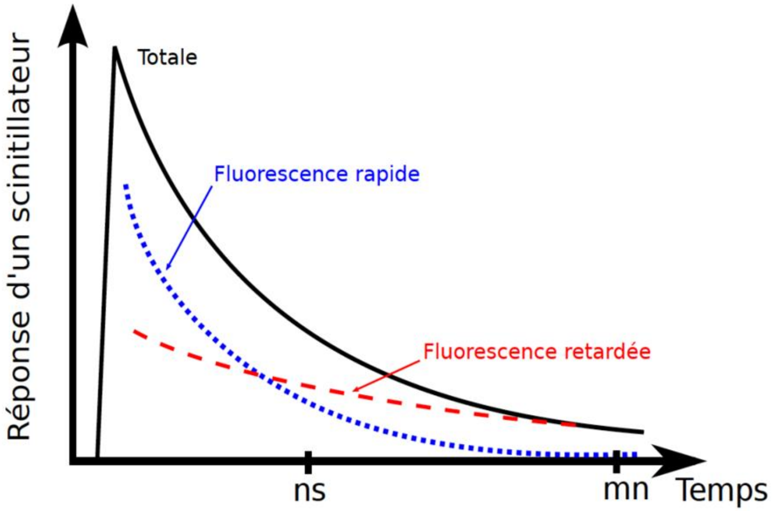
- **Detectors based on liquid scintillator(s):**
  - **Very fast detectors (e.g. IC): σ_t < 1 ns**
  - **Neutron – γ separation by means of TOF**

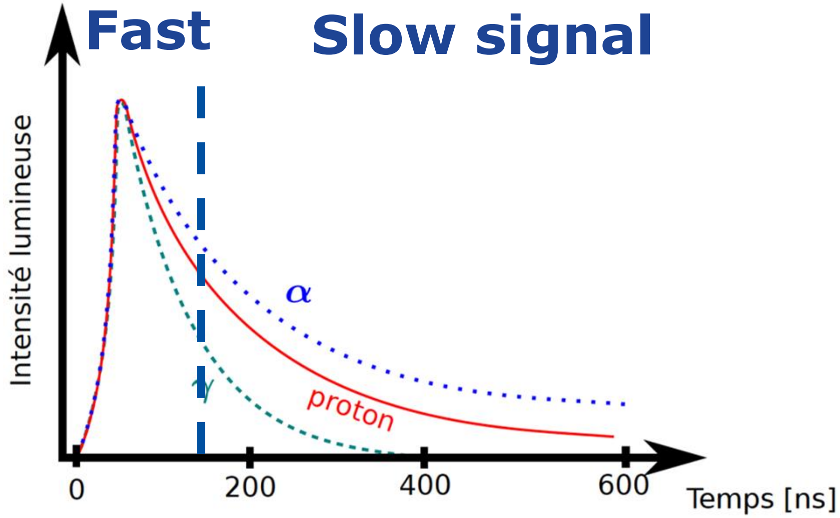


A. Göök et al., PRC90 (2014) 064611

How to measure neutrons

- Detectors based on NE213 liquid scintillator(s):
  - Allow pulse shape discrimination
  - Electrons and recoil protons excite different fluorescent levels
  - Detector signal shows different fall times





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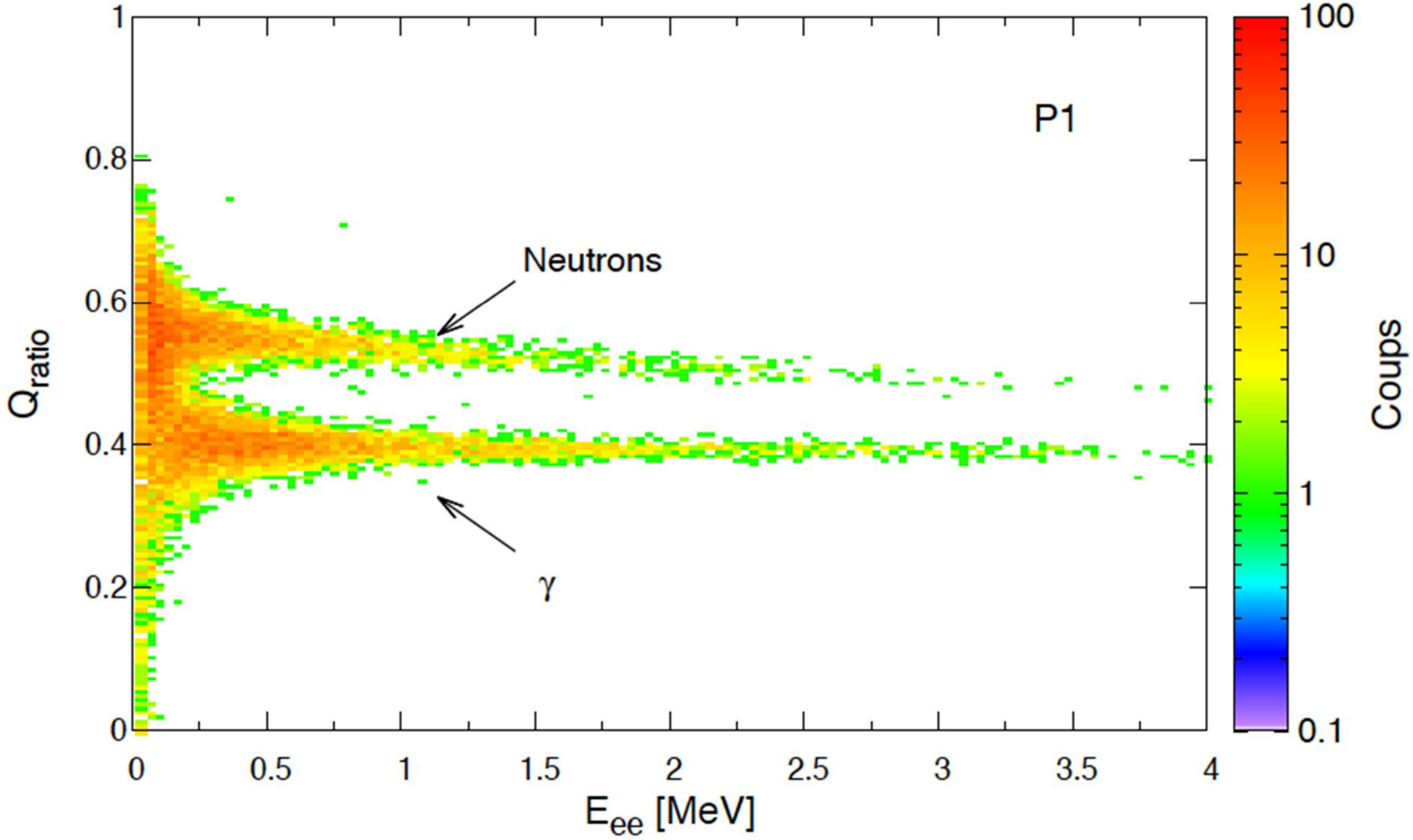
How to measure neutrons



PhD thesis, A. Sardet, Université Paris Sud (2015)

Detector calibration

- Selection of γ-rays sources for calibration

Source	E_γ [keV]	E_{ee} [keV]	Type	Résolution B1 [%]
^{133}Ba	81	81	FE	59,98 ± 0,2
^{133}Ba	356	207	CE	29,27 ± 3,2
^{22}Na	511	340,7	CE	20,7 ± 2,3
^{207}Bi	569	393,3	CE	20,2 ± 2,0
^{137}Cs	662	477,65	CE	18,2 ± 1,6
^{207}Bi	1063	857,7	CE	14,8 ± 0,9
^{22}Na	1 275	1 061,7	CE	12,8 ± 0,7
^{207}Bi	1770	1 546,9	CE	10,0 ± 0,6
AmBe	4 430	4 196	CE	8,5 ± 0,4
Pu^{13}C	6 130	5 883	CE	7,8 ± 0,3

- Use of mono-energetic neutron beams for calibration

CE: Compton Edge

FE: Photo peak

PhD thesis, A. Sardet, Université Paris Sud (2015)

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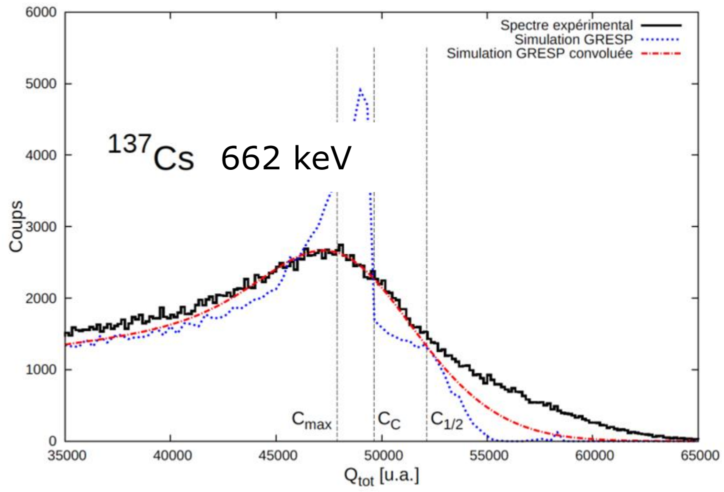
Detector calibration

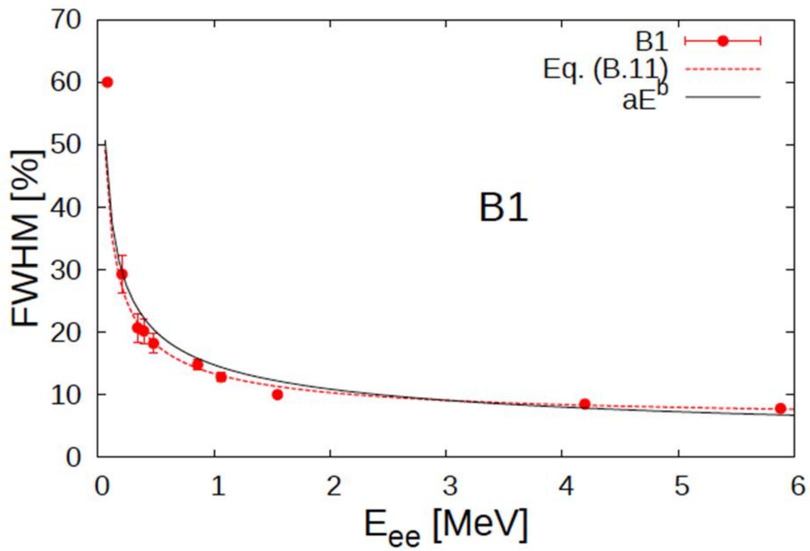
➢ **Response of LS detectors to neutrons and γ-rays:**



$$\frac{\Delta E}{E} = 1.5\frac{C_{1/2} - C_{max}}{C_{1/2}}$$

Simulation with GRESP

$$\frac{\Delta E}{E} = \sqrt{\alpha^2 + \frac{\beta^2}{E} + \left(\frac{\gamma}{E}\right)^2}$$



A. Sardet, et al., NIM A792 (2015) 74

PhD thesis, A. Sardet, Université Paris Sud (2015)

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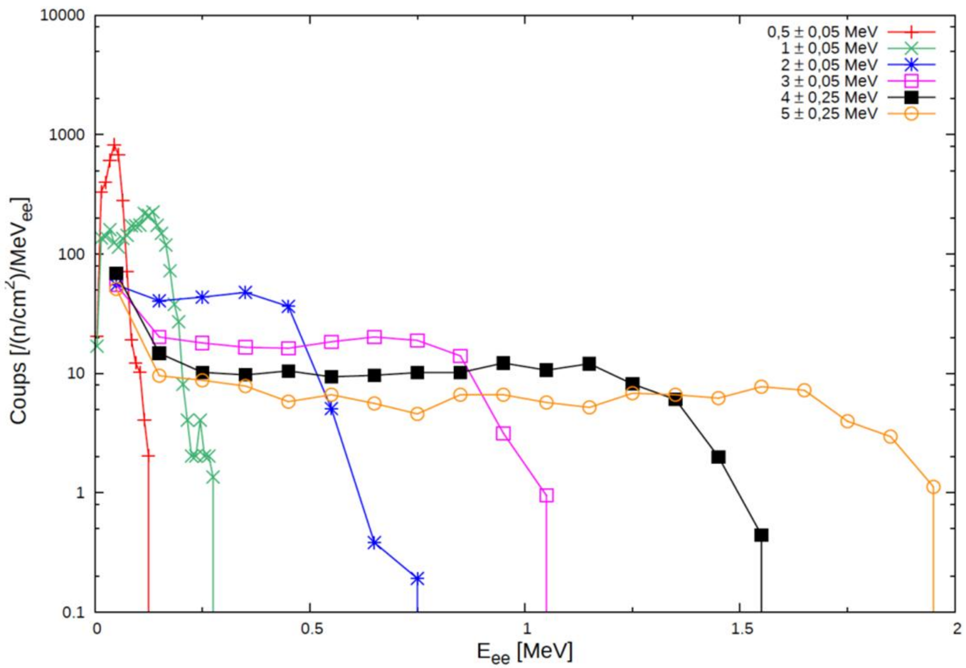
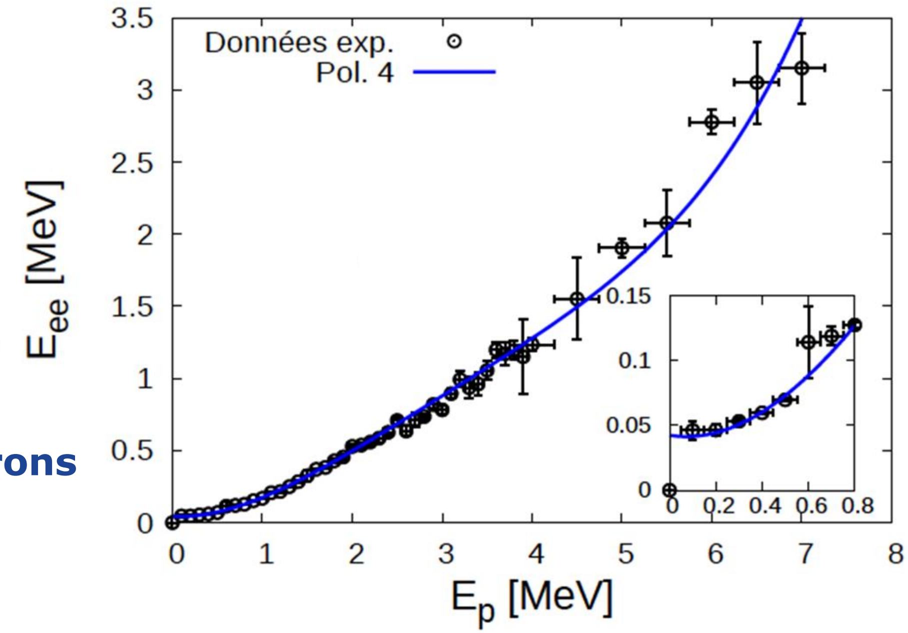
Detector calibration

- **Response of LS detectors to neutrons and γ-rays:**





- **Response for mono-energetic neutrons**
- **Selection from the TOF information**
- **Calibrated neutron beam**

A. Sardet, et al., NIM A792 (2015) 74

PhD thesis, A. Sardet, Université Paris Sud (2015)

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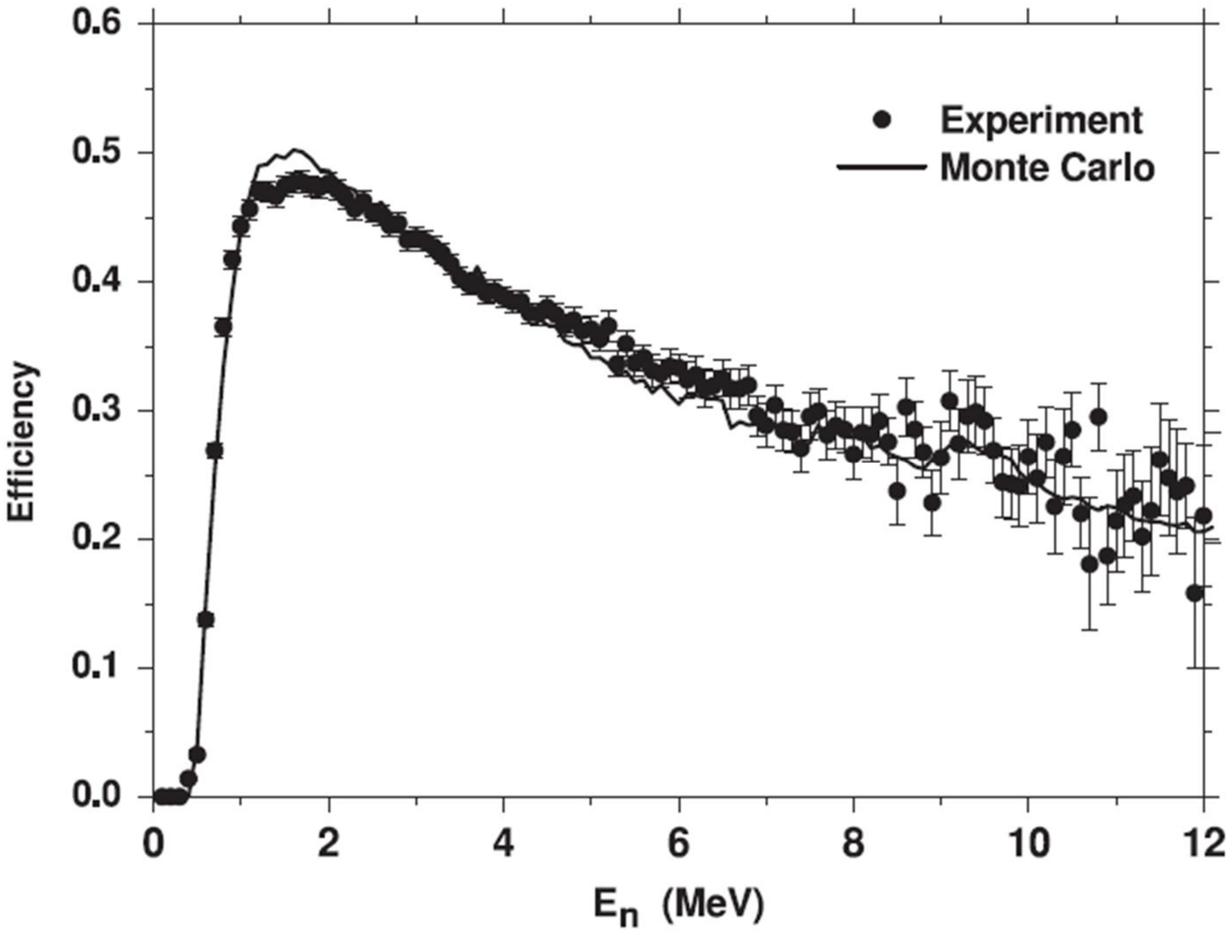
Efficiency curve

➢ **Response of LS detectors to neutrons and γ-rays:**



N. V. Kornilov et al. NIMA599 (2009) 226

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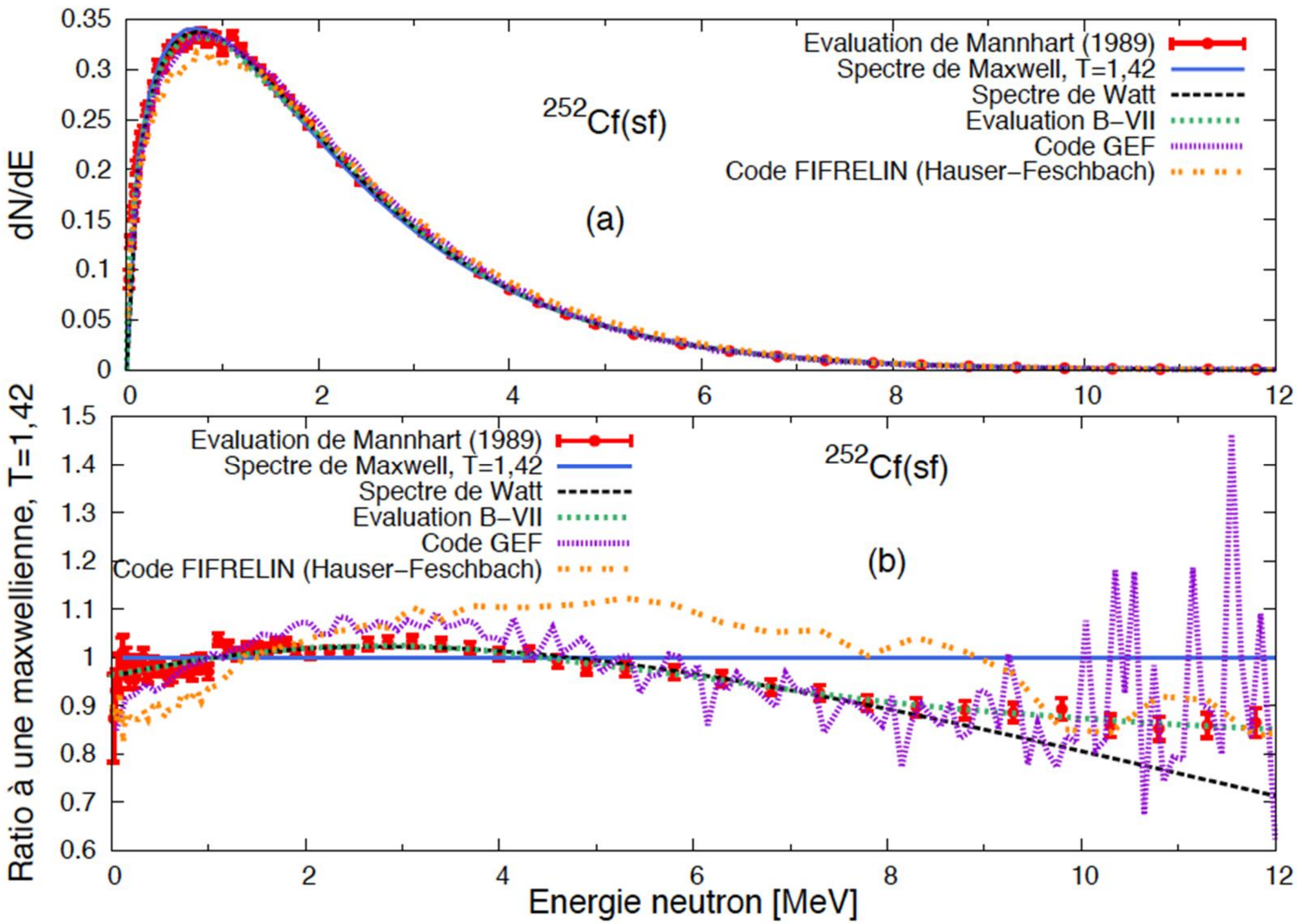
How to measure neutrons

- **And finally: a prompt fission neutron spectrum**

$$dN/dE \sim Y\ \Omega\ \varepsilon\ \nu\ S(E)\ \ C_n$$

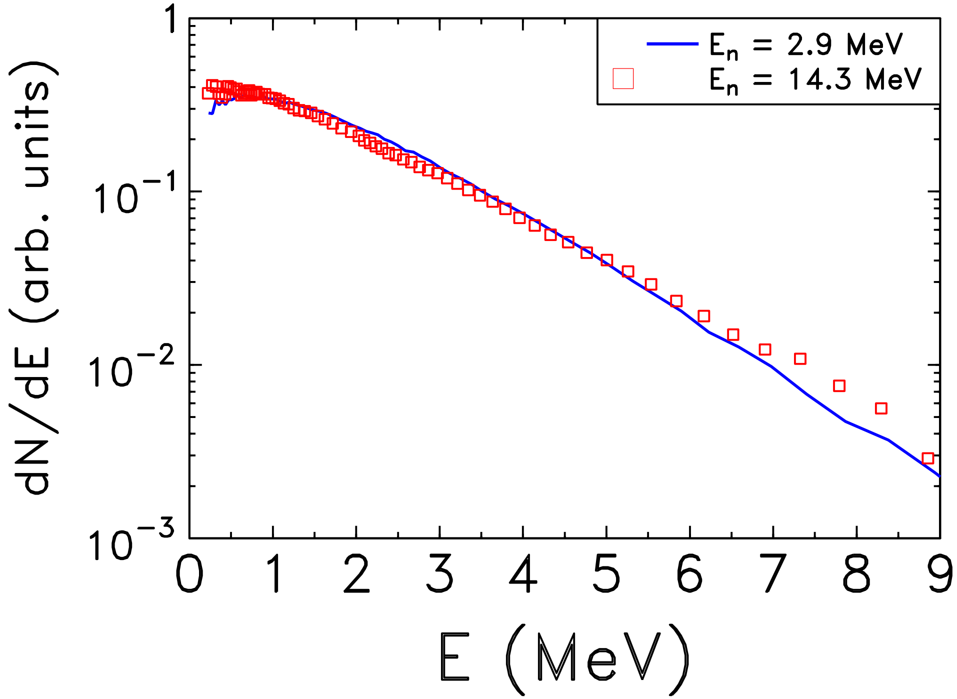
Y : Fission yield
Ω : Solid angle
ε : Efficiency
ν : n-multiplicity
S(E) : Spectrum from ToF
C_n : correction factors

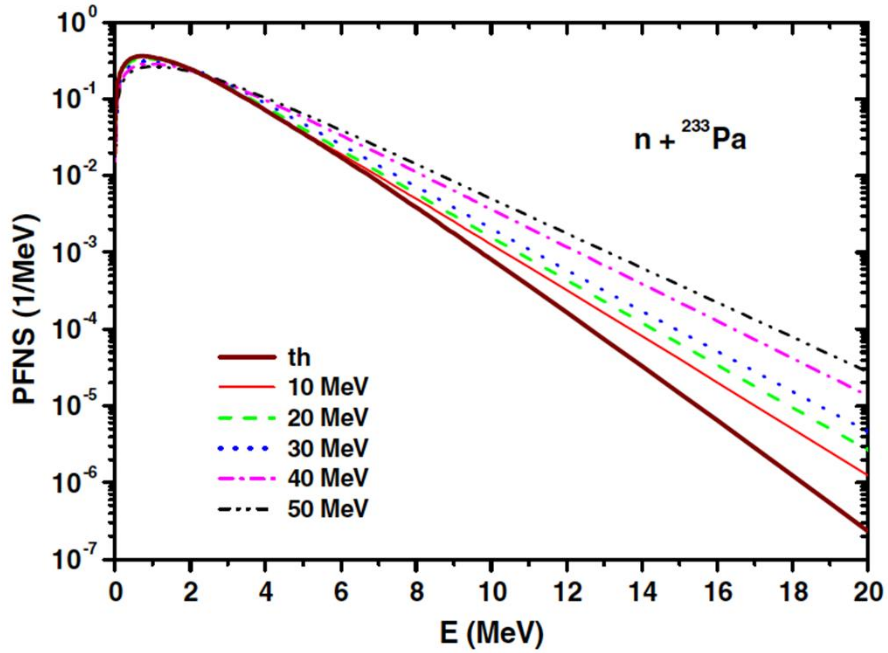


T depends on fissioning system

How to measure neutrons

➢ **Energy dependence of PFN emission**



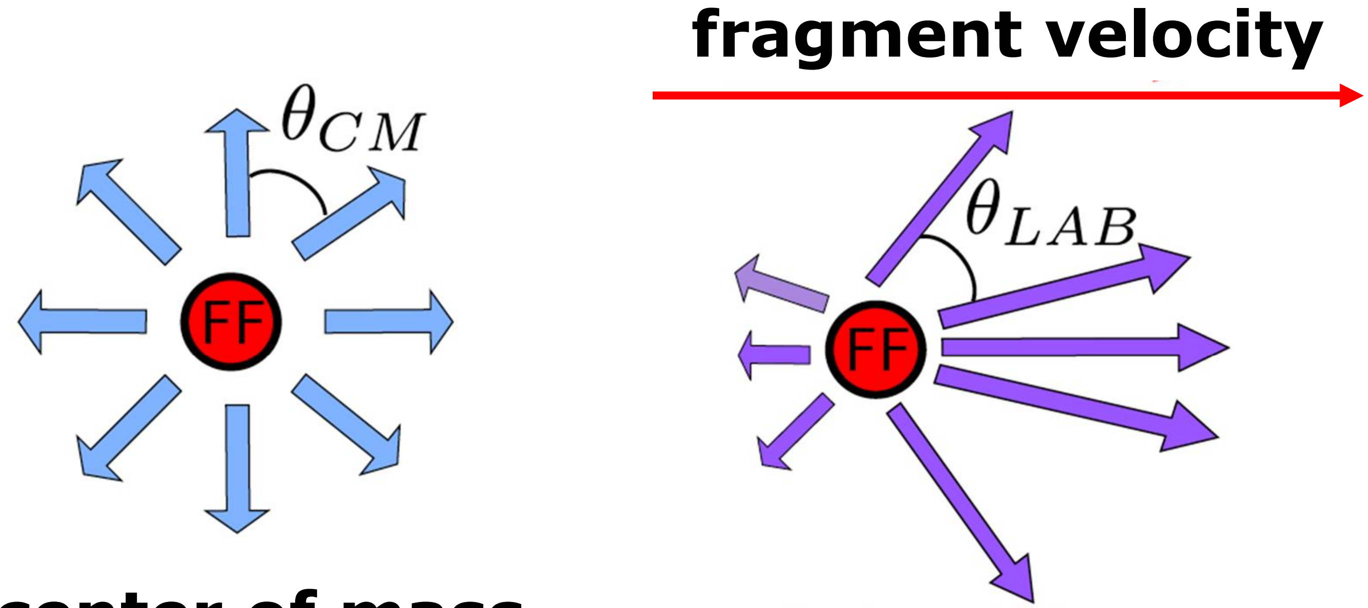


Boikov et al., EXFOR: 41110

A. Tudora et al. et al., ANE 35 (2008) 1131

How to measure neutrons

- **Prompt neutron measurements**



center of mass frame

To extract physics

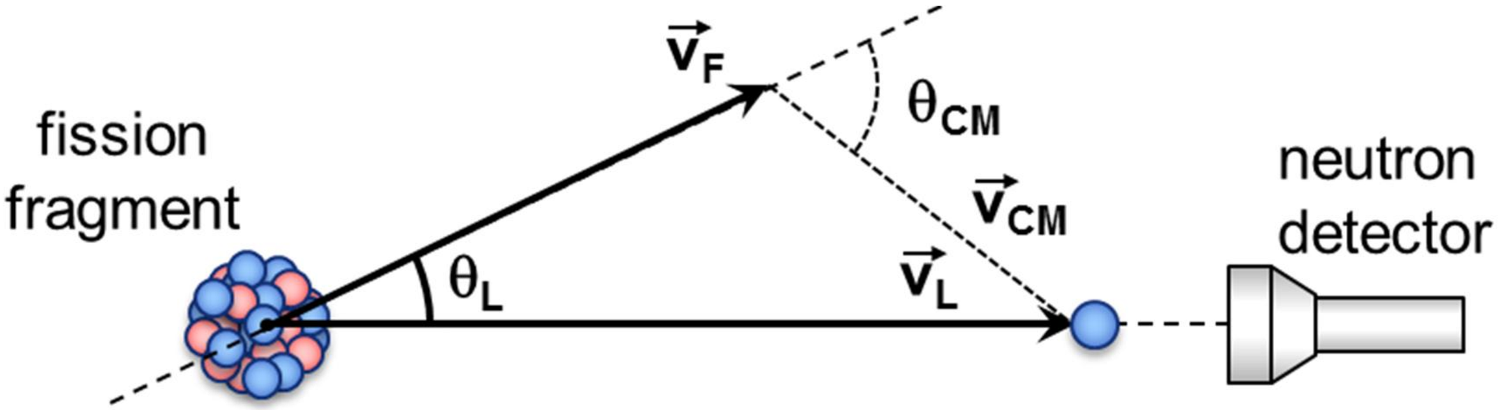
laboratory frame

What you measure, relevant for application

Transformation from the LS to CMS

How to measure neutrons

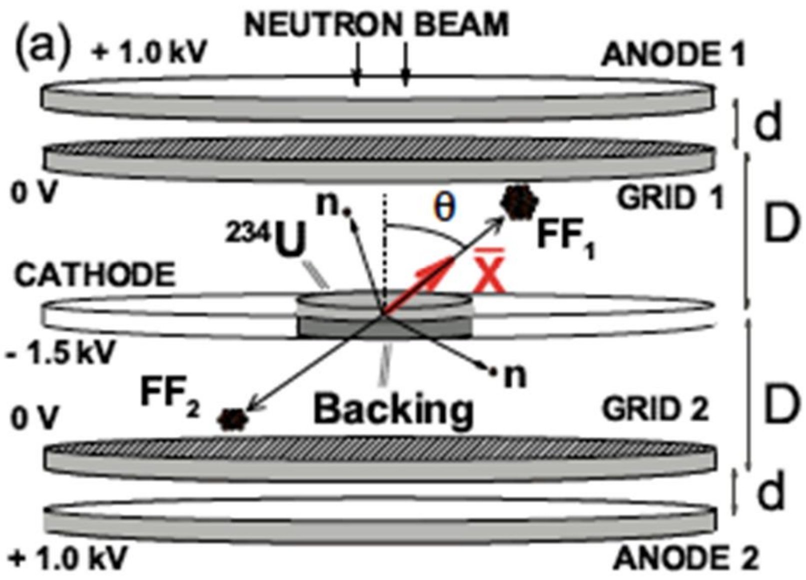
- Emission of neutrons from **fully accelerated fragments**
  - Obtain basic **kinematic information** in laboratory-frame
  - **Reconstruct emission process** in fission fragment rest-frame

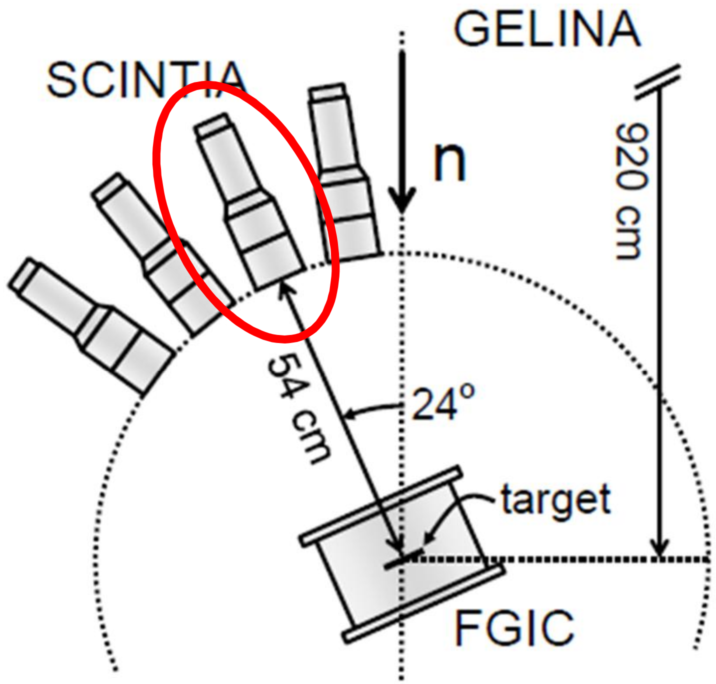


- Unbiased selection of events: $\cos\theta_{CM} \geq 0$

How to measure neutrons

- The angle between the fission axis and the neutron vector is needed:



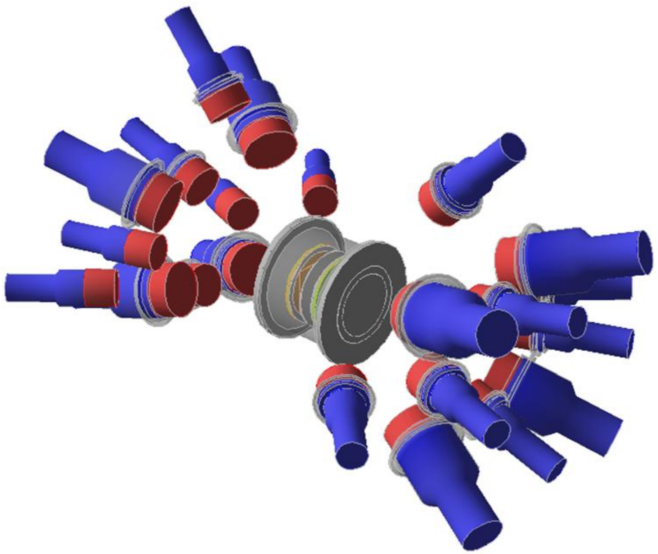


Grid gives polar angle θ

How to measure neutrons

➢ **SCINTIA array (JRC-Geel)**

- ✓ **Array of 22 neutron detectors**
  - ✓ SCIONIX LS301 (different sizes)
  - ✓ P-Therphenyl (Φ=8.5 cm, h=6.8 cm)
- ✓ **Double Frisch-grid position sensitive IC**



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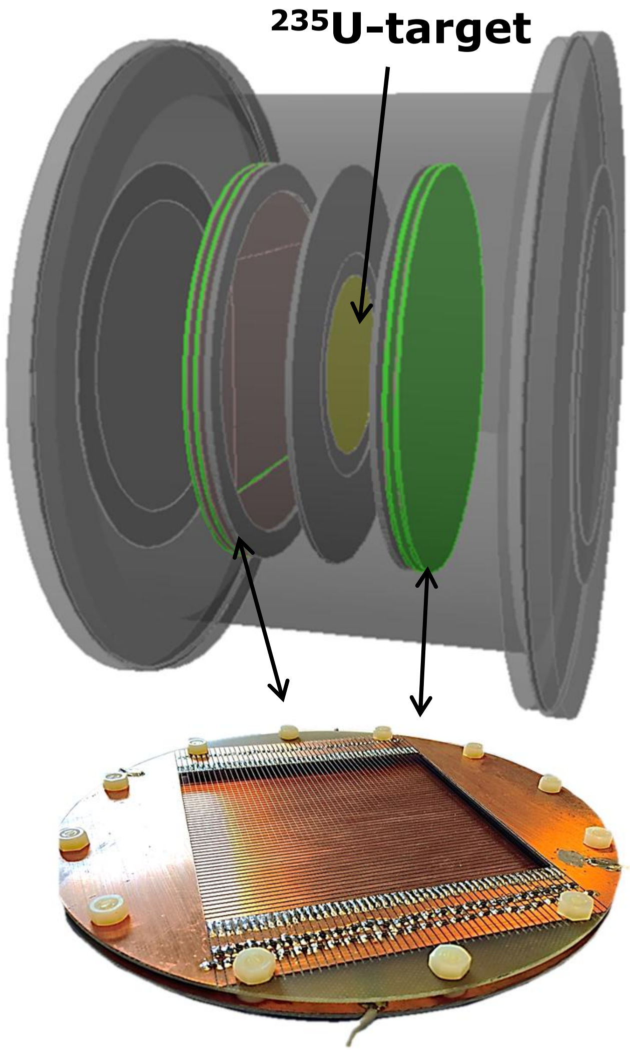
Fission fragment detector

Twin Ionization Chamber

- ✓ Energies and Masses of fission fragments
- ✓ Large Geometrical Efficiency
- ✓ Timing resolution ~1 ns (FWHM)
- ✓ Polar angle θ of fission axis relative to the chamber axis
- – Need for azimuthal angle ϕ



Position Sensitive Electrodes

- ➢ Replaces anodes
  - ➢ wire plane + strip anode

  Projection of fission-axis on the electrode – plane

- ✓ Fission axis orientation in 3D

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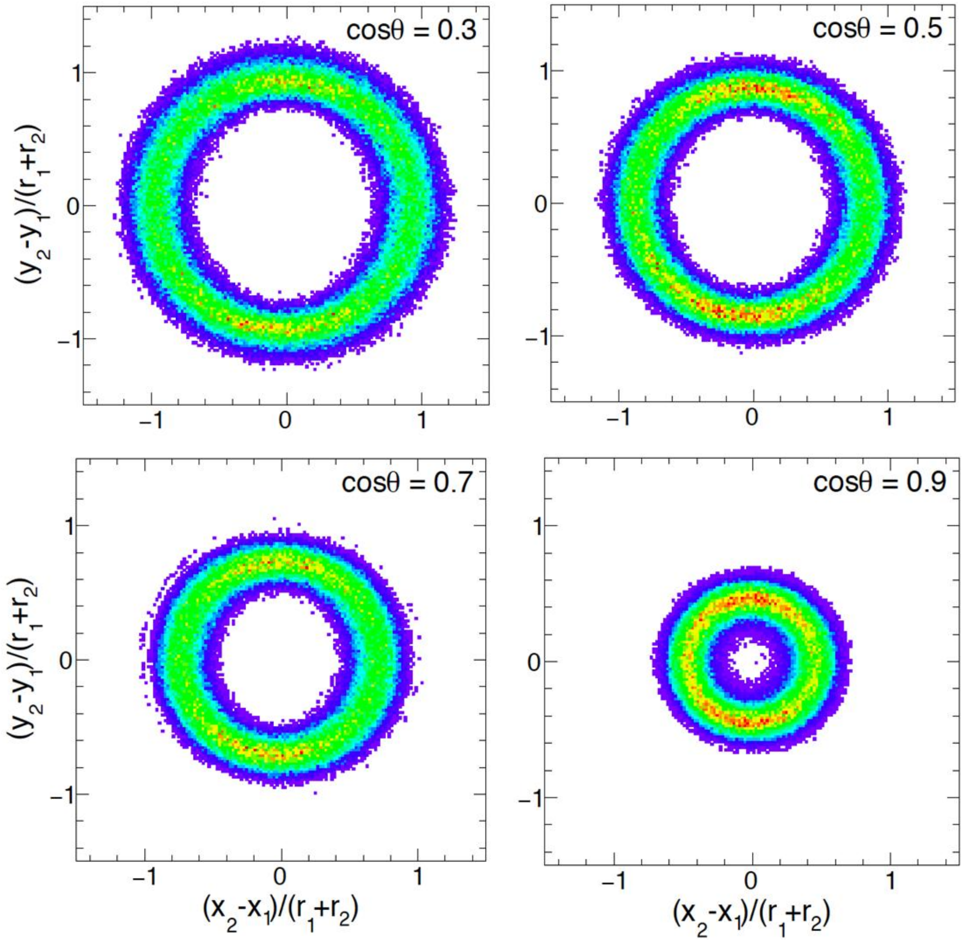
How to measure neutrons

Azimuthal angle of fission axis around chamber symmetry axis is determined from charge division read-out of position sensitive anodes



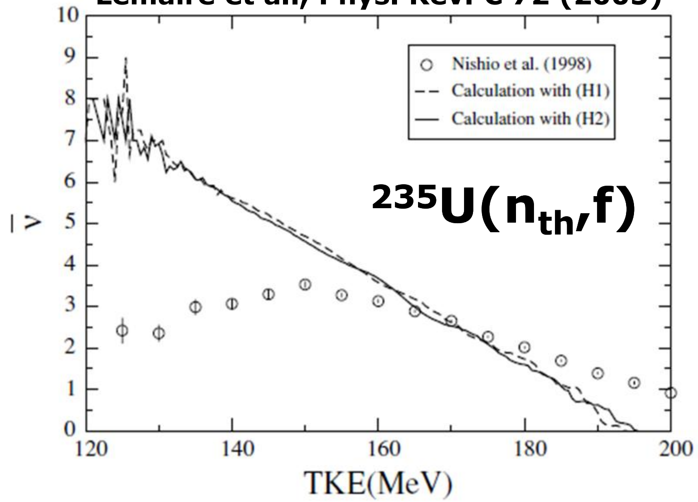
Difference in x-coordinates and y-coordinates of the Bragg-peak for the fission fragments detected on opposite chamber sides.

A. Göök et al., NIMA 830 (2016) 366

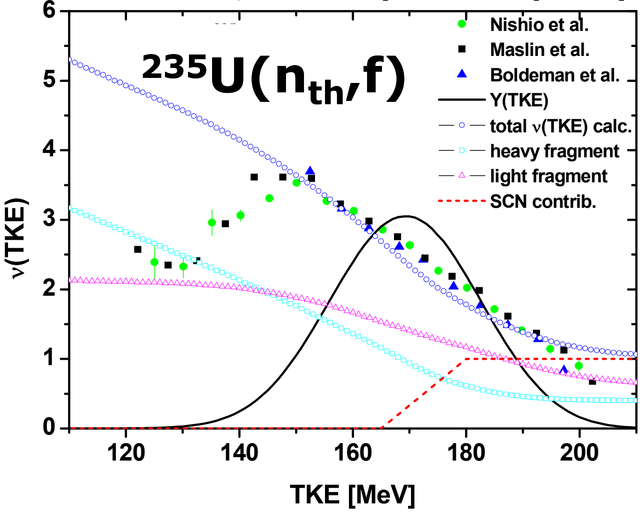
Motivation

PFN multiplicity correlations with fragment observables

Lemaire et al., Phys. Rev. C 72 (2005)



Kornilov et al., Nucl. Phys. A 780 (2007)



Based on energy balance in fission

- **Detailed modelling (CGMF, Fifrelin, Freya…)**
  - **successfully reproducing correlations in the case ^{235}U(n,f)**
    - » **difficulties: in particular ν(TKE)**

Lemaire et al. (2005)

*"…a **dramatic deviation** between calculation and experiment on ν is observed **at low TKE** that would indicate the presence of **additional opened channels**"*

Kornilov et al. (2007)

*"The **incorporation of** the **SCN emission** leads to a much **better agreement** between theoretical and experimental data for ν(TKE) in the high energy range. However, the assumption of **SCN emission at high TKE should be confirmed with direct experimental data**"*

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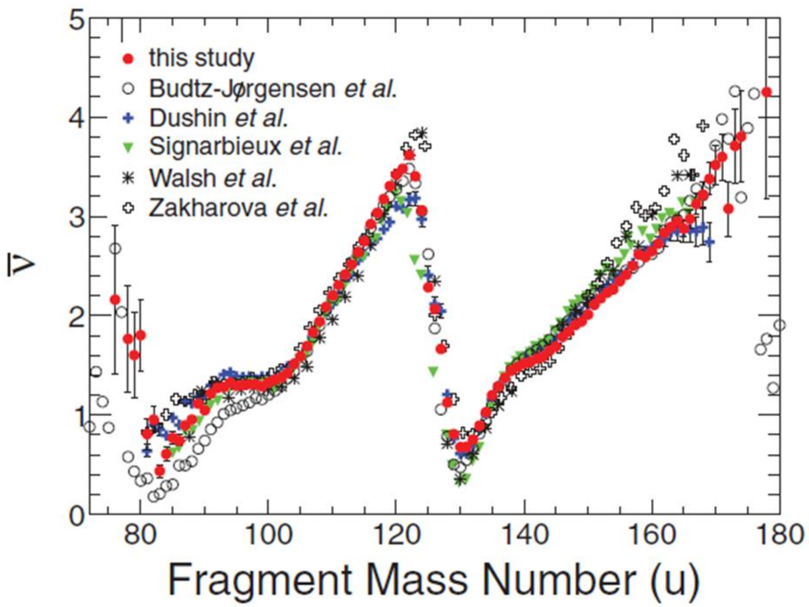
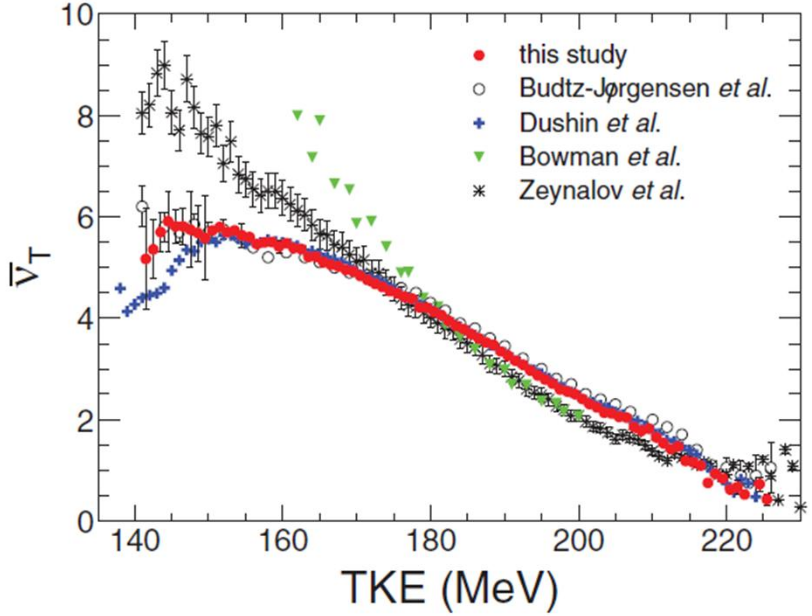
Validation of method ^{252}Cf(sf) and single NE213 detetcor

PHYSICAL REVIEW C **90**, 064611 (2014)





- **Results show consistency with literature data**
- **Specifically with methods that do not suffer from neutron energy detection threshold**

  (Dushin et al.) Gd-loaded 4π scintillator tank

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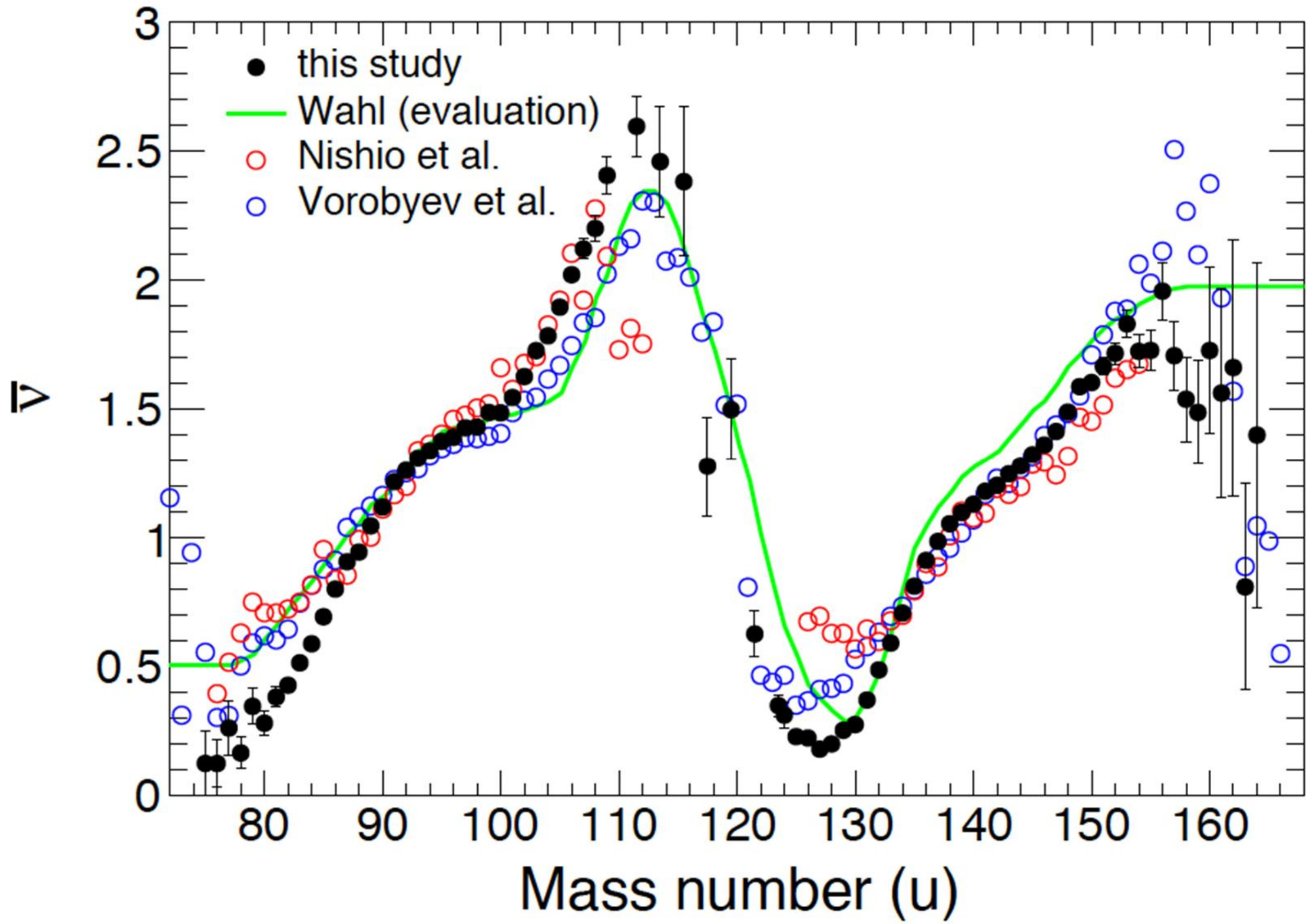
Multiplicity vs. Fragment Mass of $^{235}U(n,f)$



$E_n \in$ [0.3 eV, 60 keV]

Pronounced minima around A_L=80 and A_H=130

Shoulders around A_L=100 and A_H=140

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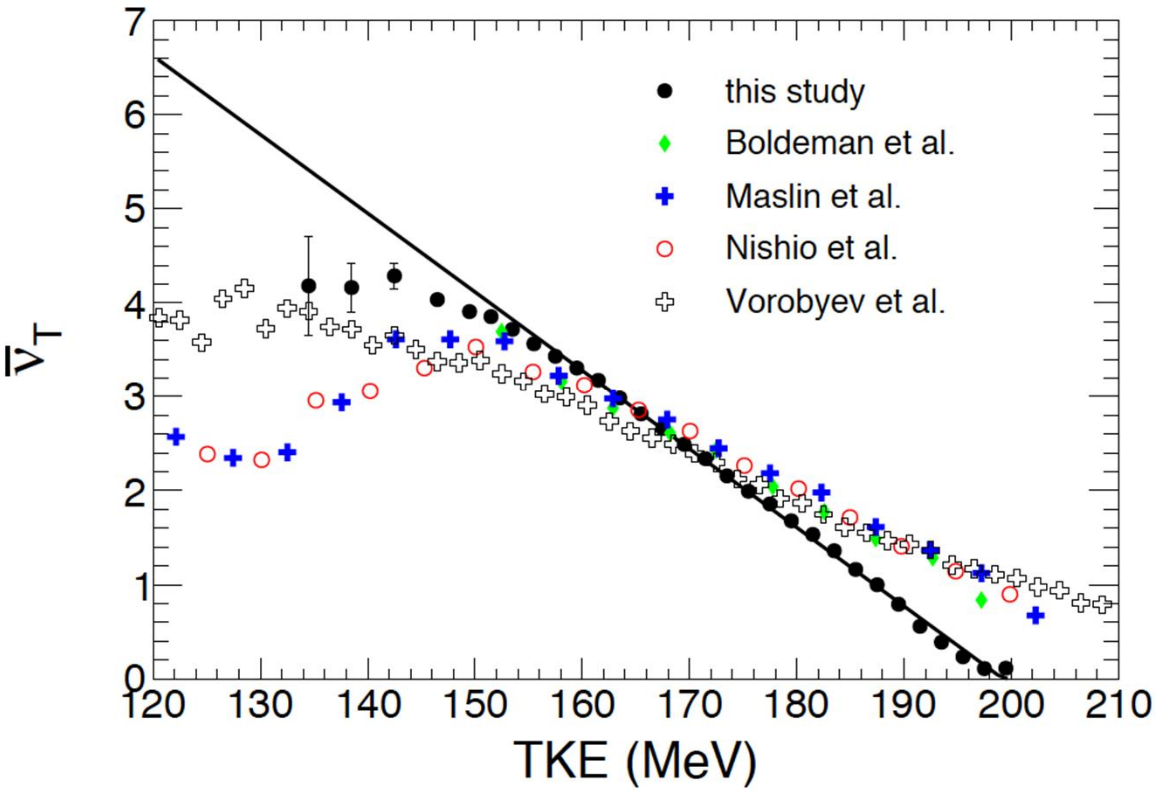
Multiplicity vs. Fragment TKE of $^{235}U(n,f)$



Close to linear dependence

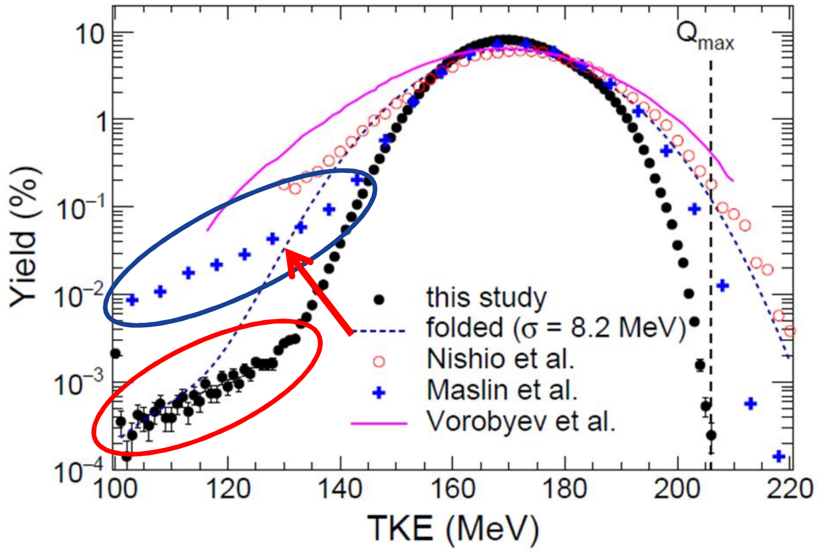
$$-\frac{dTKE}{d\bar{\upsilon}_T} = 12.0\ \mathrm{MeV/n}$$

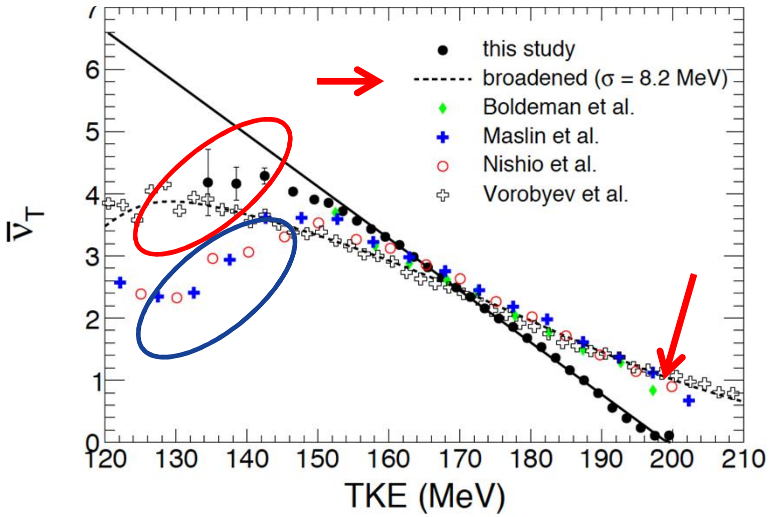
The slope is much steeper than earlier studies

$$-\frac{dTKE}{d\bar{\upsilon}_T} = 16.7 - 18.5\ \mathrm{MeV/n}$$

The difference cannot be explained by difference in incident neutron energy

Multiplicity vs. Fragment TKE





- Wide TKE-distributions
- Significant Yield at TKE>Q_{max}

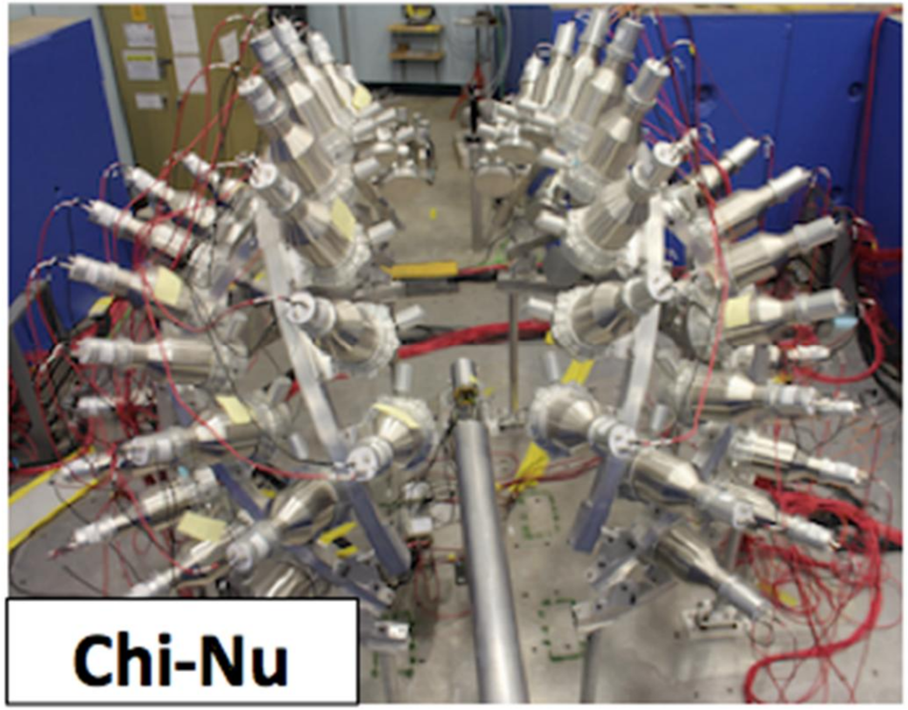
⇒ Resolution broadening

- ➢ Decreased slope
- ➢ Increased neutron yield at Q_{max}

Tailing of TKE distribution

- Energy degraded scattered fission fragments
- Neutron yield should approach average nubar
- ➢ Drop in nubar at low TKE
- ➢ Present also in our data

How to measure neutrons



- Chi-nu array (LANL)

- ✓ Array of 54 neutron detectors
- ✓ Multi plate IC
- ✓ Measure PFNS as a fct of E_n

How to measure neutrons and γ-rays

- **Prompt γ-ray measurements**

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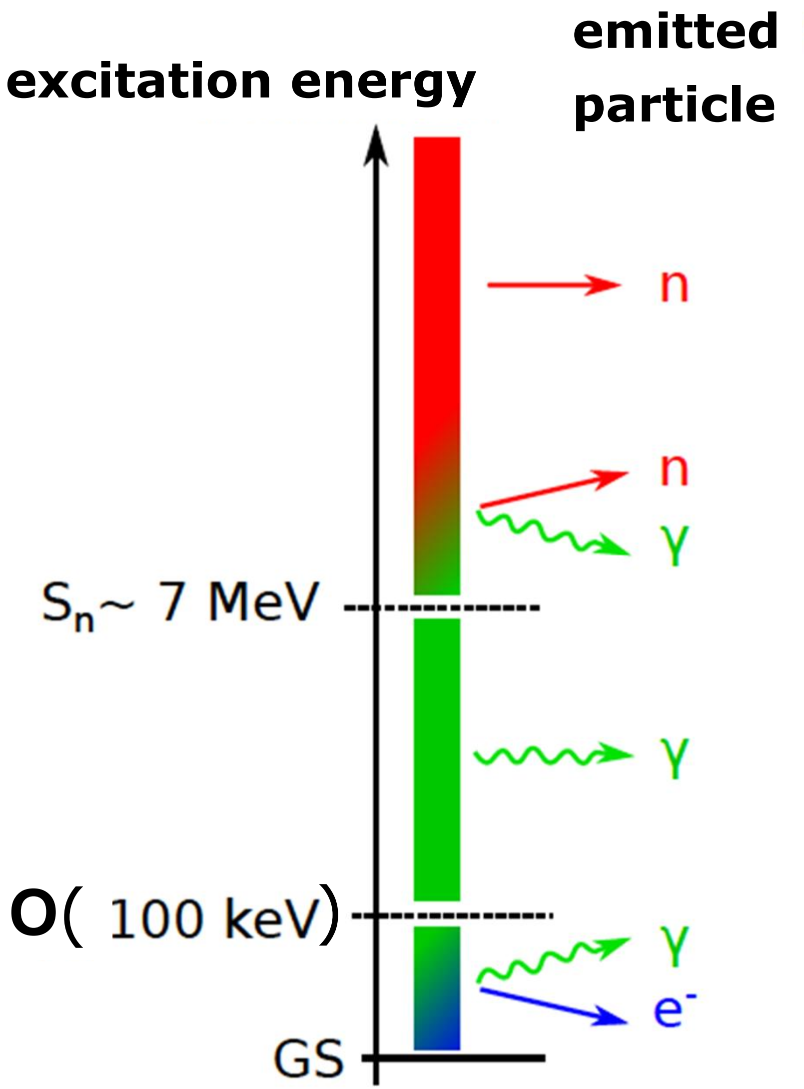
How to measure prompt fission γ-rays



D. Regnier, PhD-thesis (2013)

How to measure prompt fission γ-rays

- **Separation of γ-rays from prompt neutrons**
  - **Time-of-flight method**
  - **Excellent timing resolution of the combined γ-ray and fission detector**
  - **Determines the geometrical efficiency of your instrument**
- **Best possible energy resolution**

How to measure prompt fission γ-rays

- **Fission detectors: FGIC, Si-detectors, single-crystal diamond detectors (σ_t < 100 ps)**

- **Choice of suitable γ-ray detectors:**

  – **High purity germanium detectors**

  ✓ **Excellent energy resolution, bad timing resolution**

  – **Fragments moving → Doppler broadened γ-peak**

  – **Very neutron sensitive**

How to measure prompt fission γ-rays

➢ **Choice of suitable γ-ray detectors:**

- ~~**High purity germanium detectors**~~
- ✓ ~~**Excellent energy resolution, bad timing resolution**~~
- ~~**Fragments moving → Doppler broadened γ-peak**~~
- ~~**Very neutron sensitive**~~

- **Scintillation detectors**
- **Limited energy resolution**
- ✓ **In general much better timing resolution**
- ✓ **Higher efficiency, larger sizes available**

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How to measure prompt fission γ-rays

➢ Scintillation detectors:

In the 1970s sodium iodine (NaI) was used

- ✓ **Timing resolution of the order of 5 – 7 ns**
- ✓ **Energy resolution 7% @ 662 keV**
- ✓ **TOF distance 1m or larger**
- – **Limited geometrical efficiency**

Since a few years new detectors have emerged

- ✓ **Lanthanum halide detectors**

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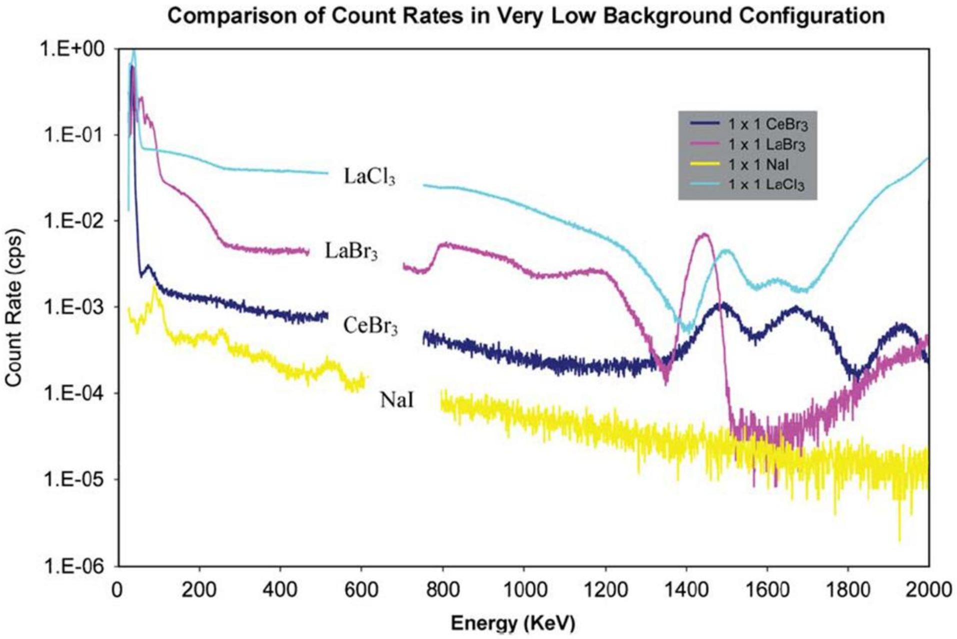
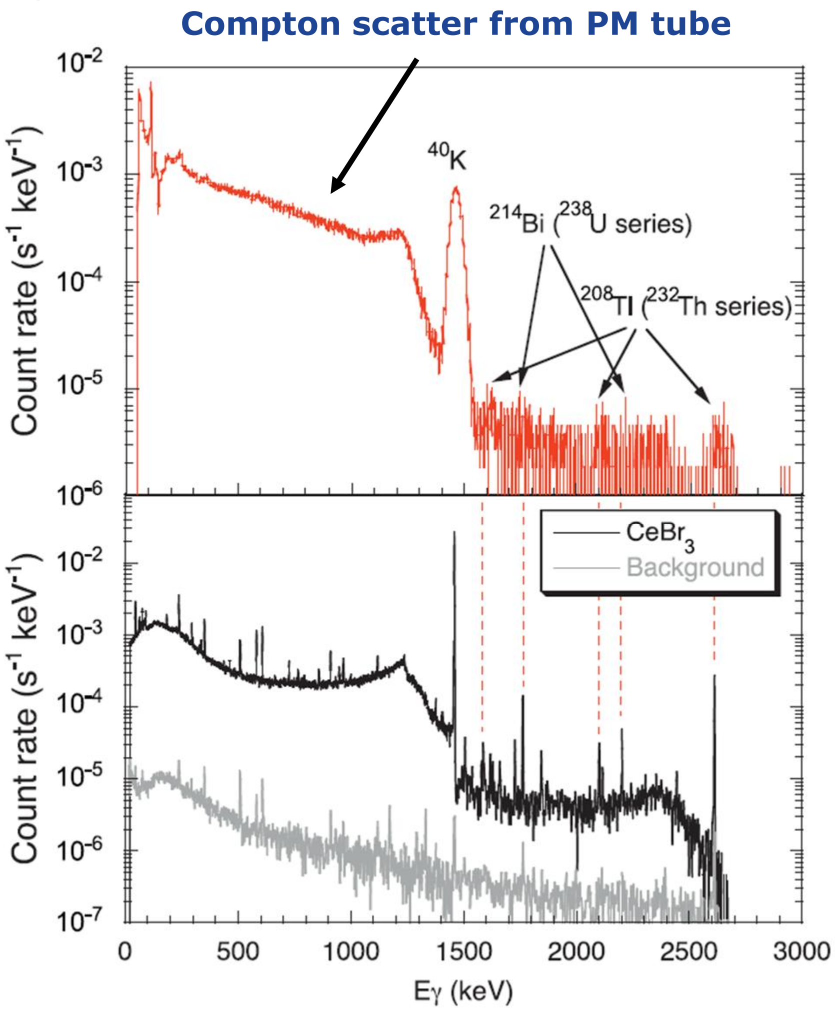
How to measure prompt fission γ-rays

- **Lanthanide halide detectors:**
  - **Intrinsic activity**



P. Guss et al. NIMA 608 (2009) 297

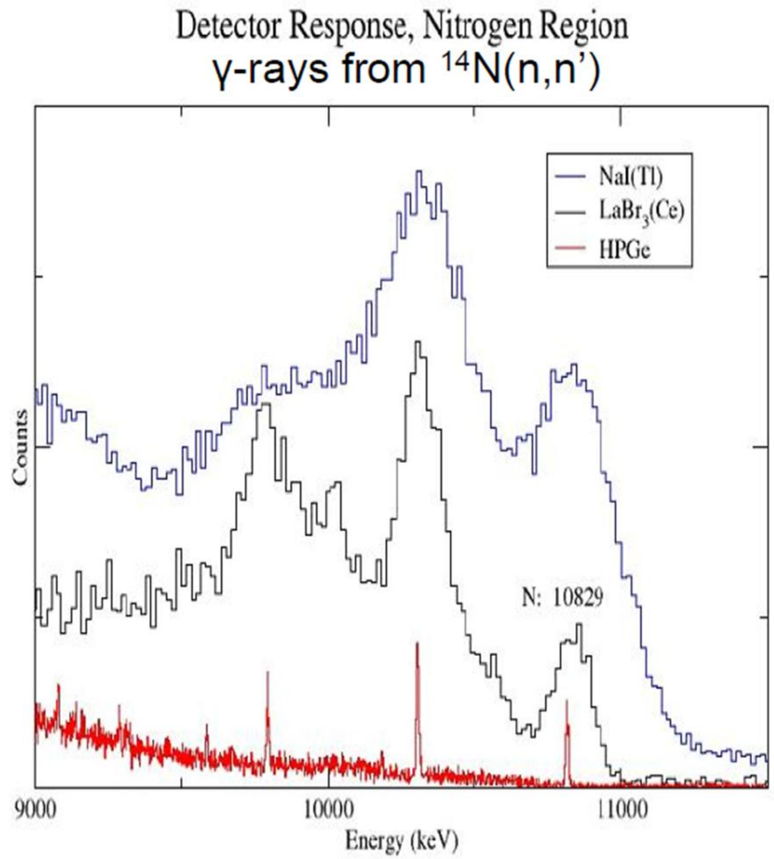


13 days measured in HADES

R. Billnert et al. NIMA 647 (2011) 94

How to measure prompt fission γ-rays

- **Lanthanide halide detectors:**
  - **$CeBr_3$ detector**



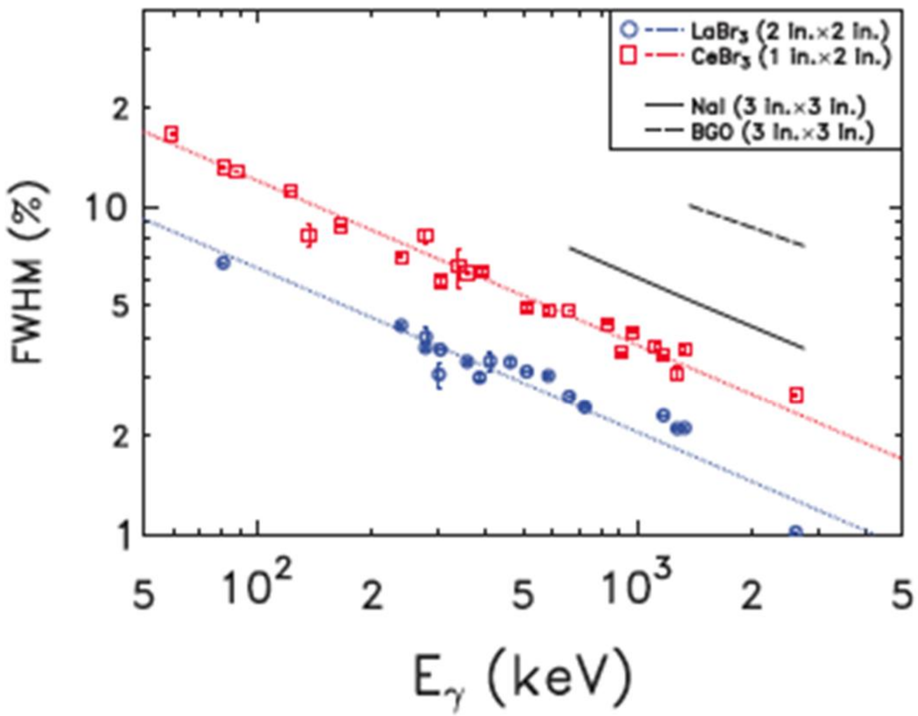
P. Guss et al. NIMA 608 (2009) 297

P. Guss, 2011 SPIE conf presentation

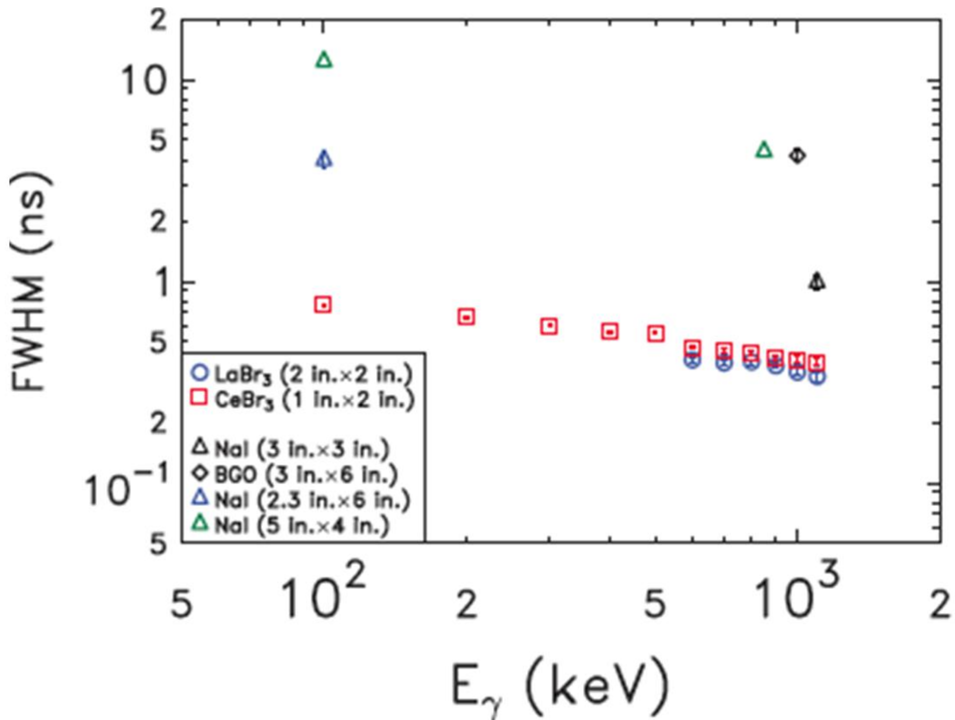
How to measure prompt fission γ-rays

➢ Lanthanide halide detectors:

- Cerium-doped lanthanum chloride ($LaCl_3$:Ce)
- Cerium-doped lanthanum bromide ($LaBr_3$:Ce)
- Cerium bromide ($CeBr_3$)
- BGO: Bismuth Germanium Oxide



Energy resolution



Timing resolution

R. Billnert et al., PRC87 (2013) 024601

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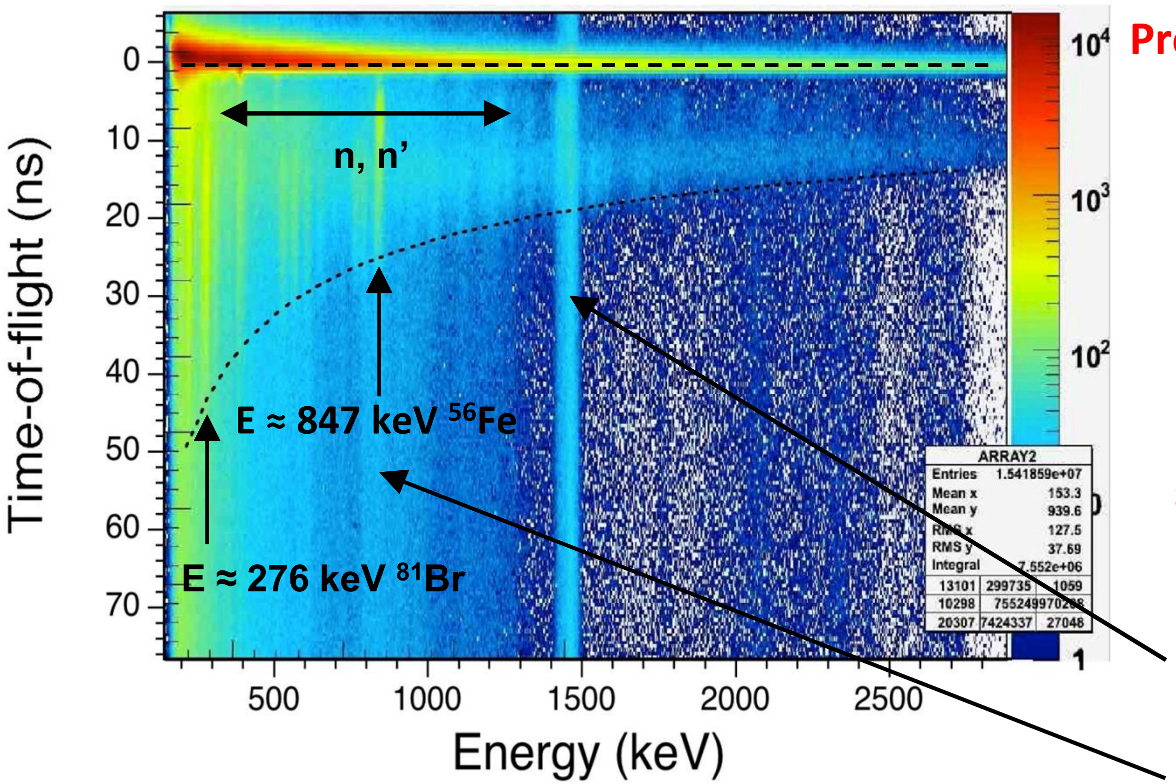
How to measure prompt fission γ-rays



Prompt fission γ-rays

γ-decay after inelastic neutron scattering

Intrinsic and external background

γ-rays in coincidence with fission fragments

A. Oberstedt, R. Billnert et al., NIM A708 (2013) 7

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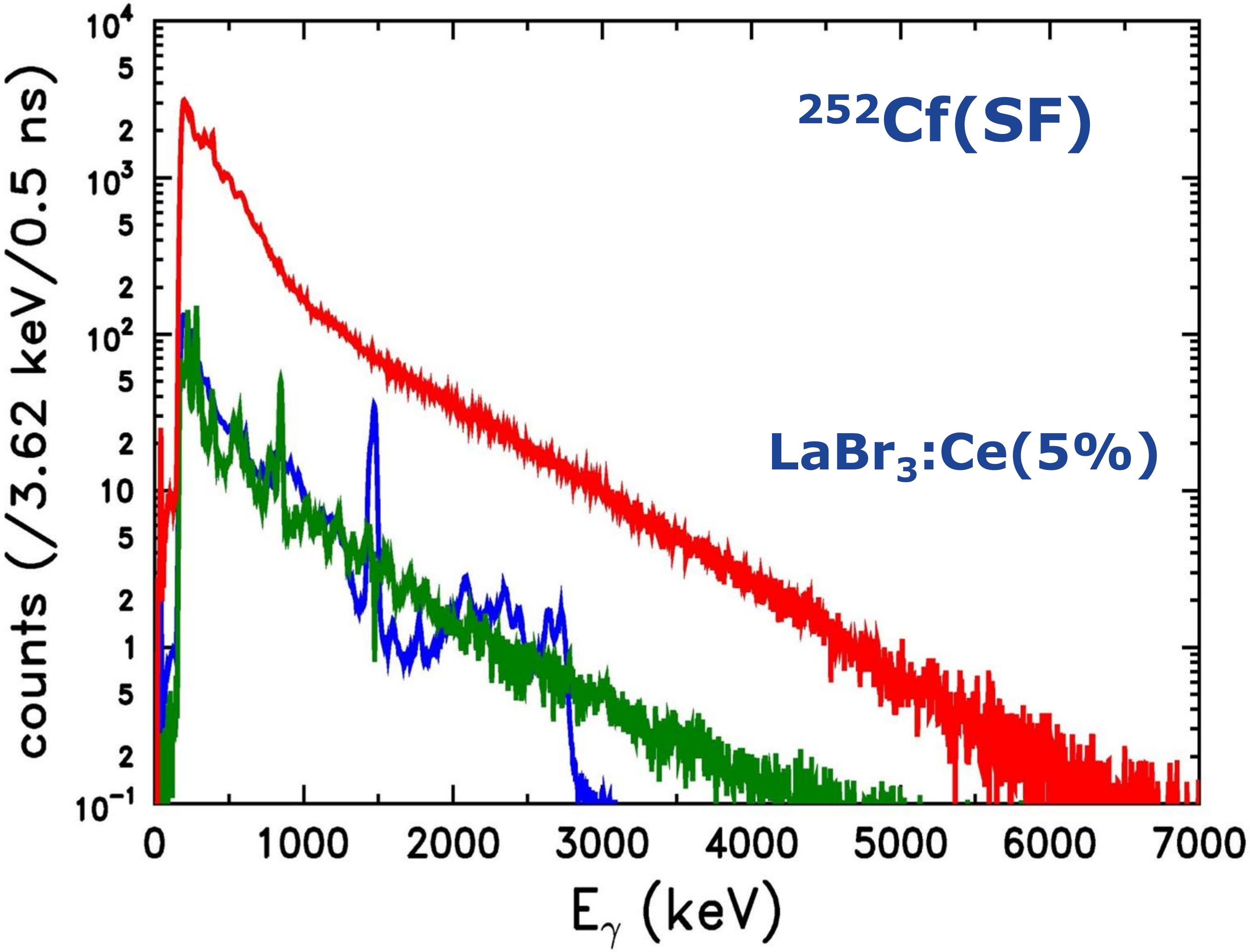
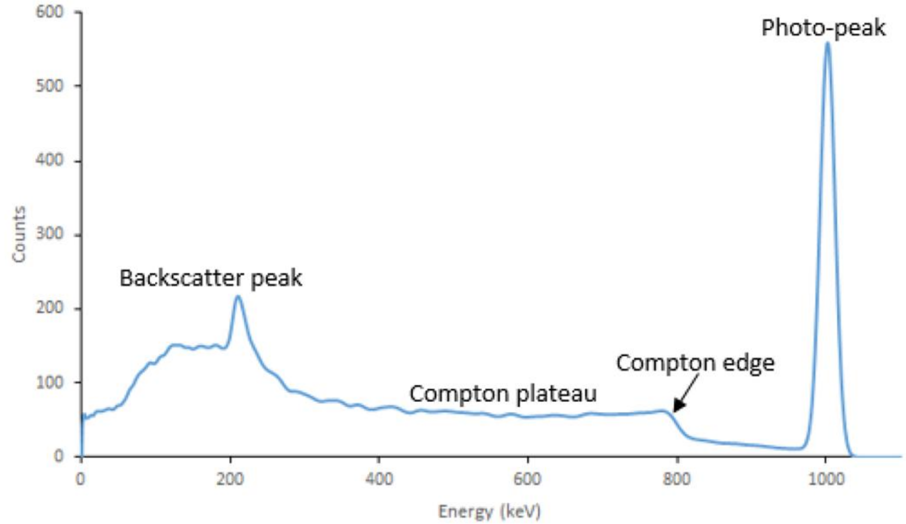
How to measure prompt fission γ-rays



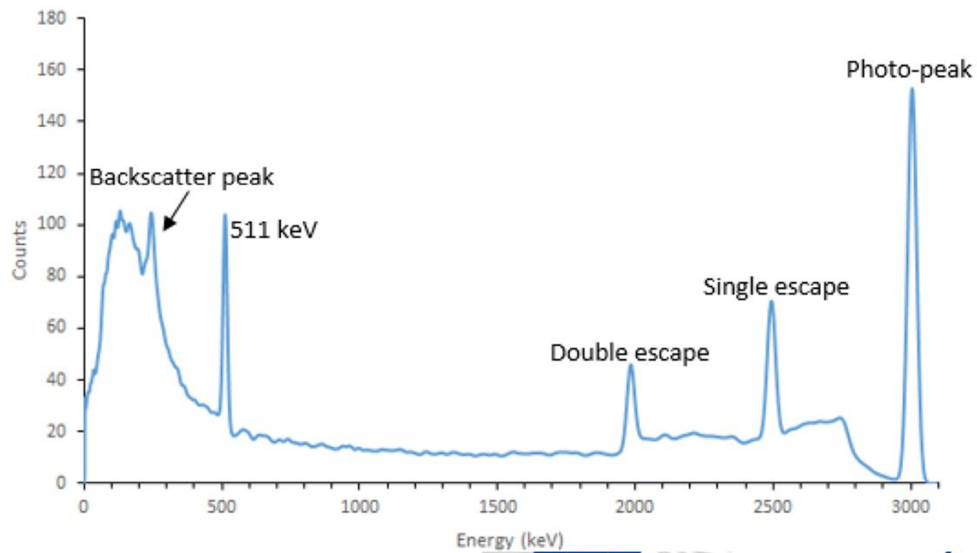
Red: PFGS, Green: Isomer decay, inelastic scattering, Blue: intrinsic bgrd

How to measure prompt fission γ-rays

➢ **Detector response to different γ-ray energies**



Simulated spectrum for 1 MeV photons hitting a 51 mm × 51 mm $LaBr_3$:Ce detector

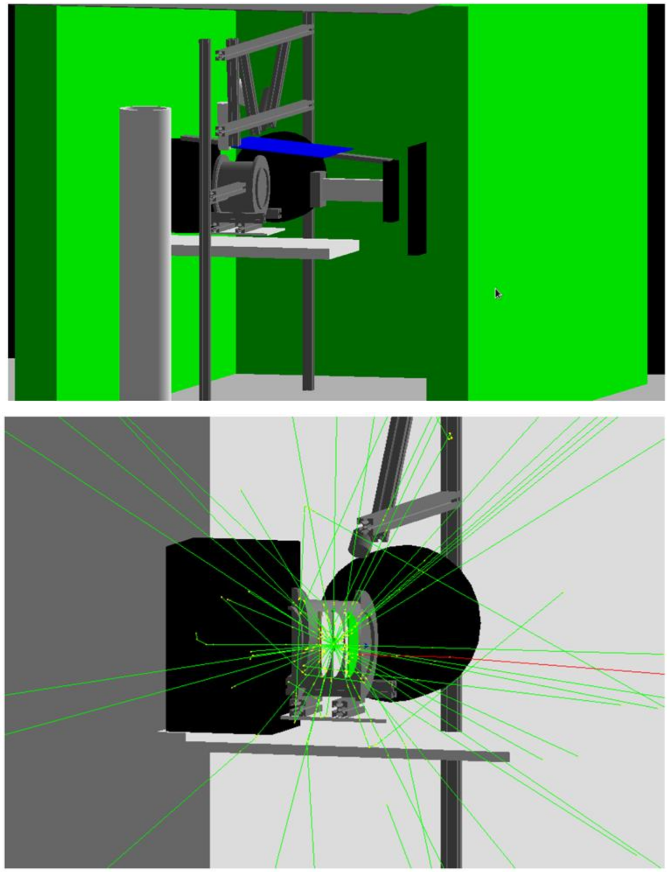


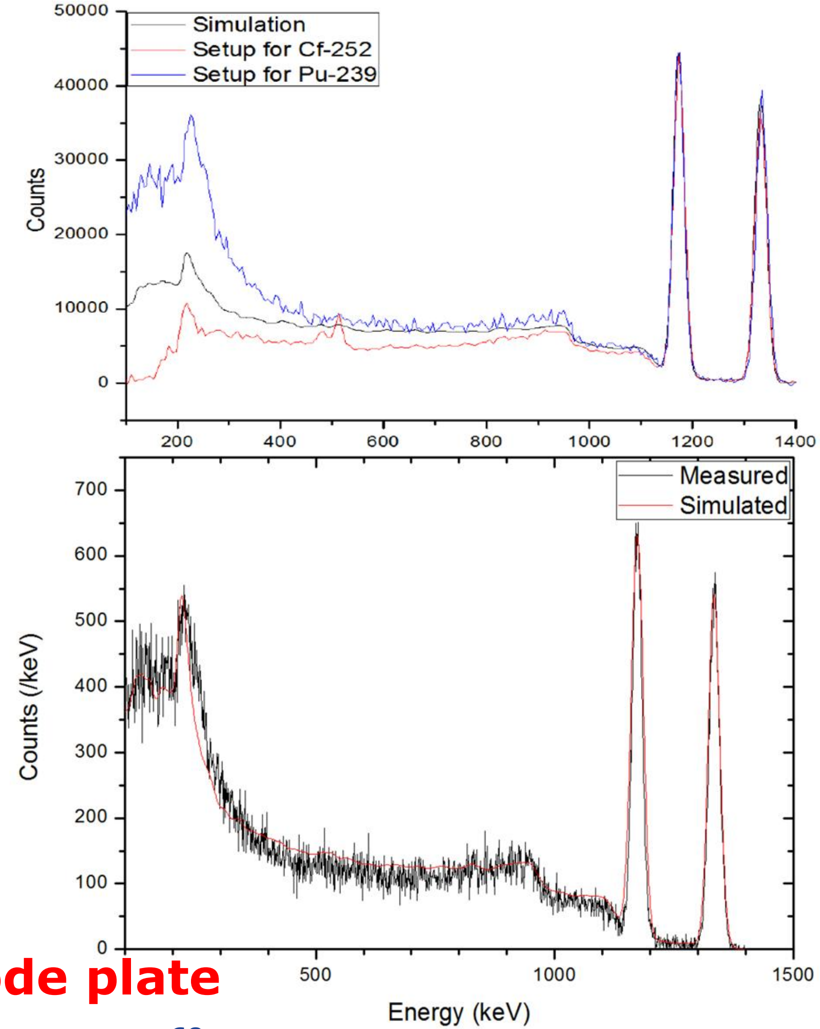
Simulated spectrum for 3 MeV photons hitting a 51 mm × 51 mm $LaBr_3$:Ce detector

PhD thesis of Angélique Gatera

How to measure prompt fission γ-rays

- **Optimized response function simulations (GEANT4) to better reproduce the backscatter peak**





Unexpected importance of cathode plate

PhD thesis of Angélique Gatera

^{60}Co source

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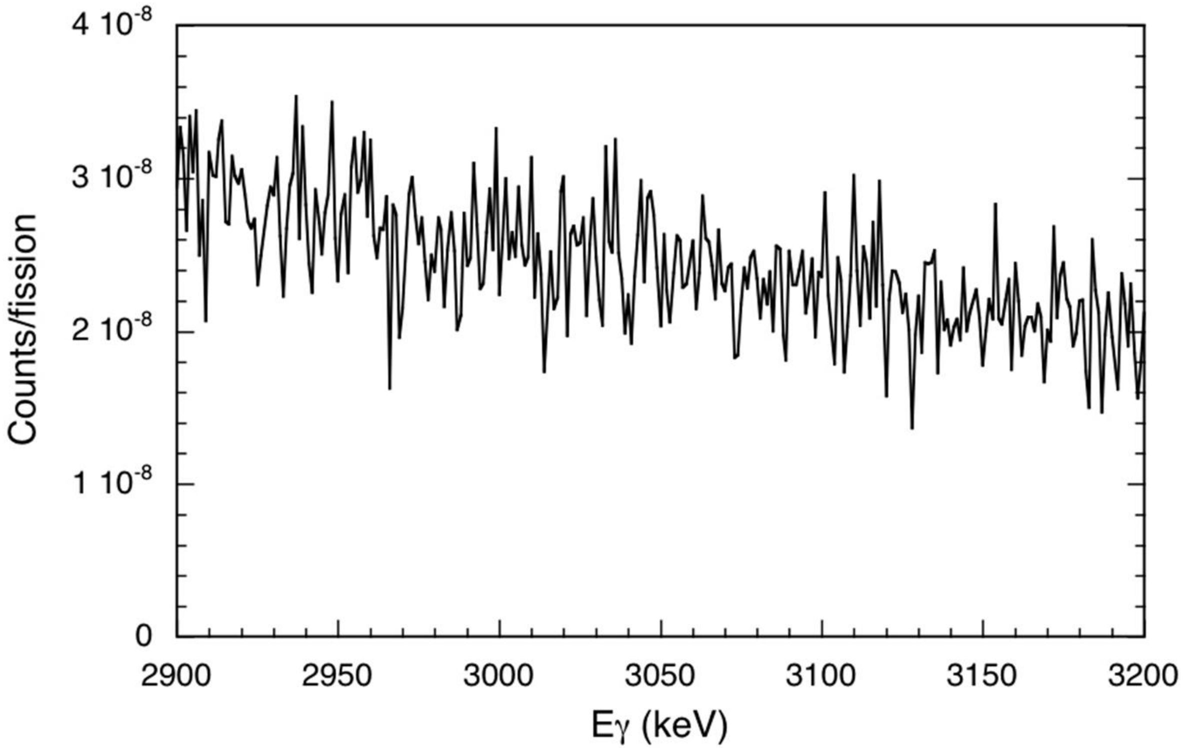
How to measure prompt fission γ-rays

➢ Unfolding the detector response



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

→ e.g. zooming into region around 3 MeV

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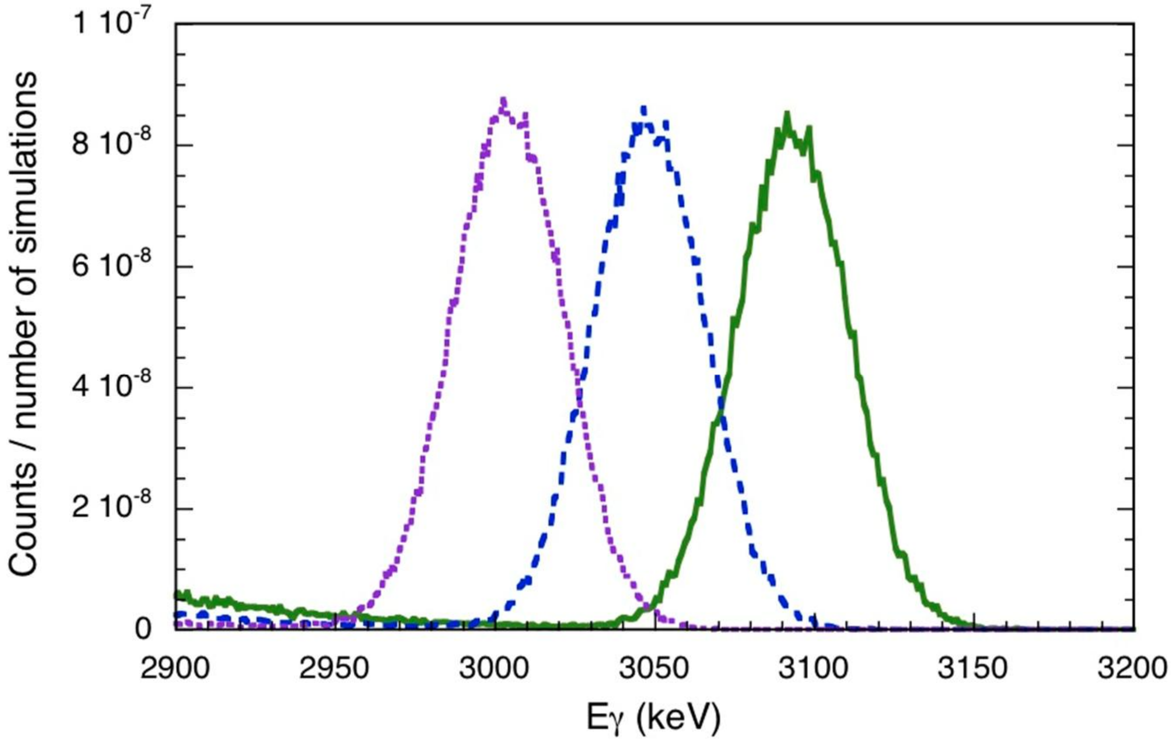
How to measure prompt fission γ-rays

➢ Unfolding the detector response



Simulating response function for mono-energetic γ-rays,

distance: FWHM from energy resolution measurements

100s of γ-ray peaks

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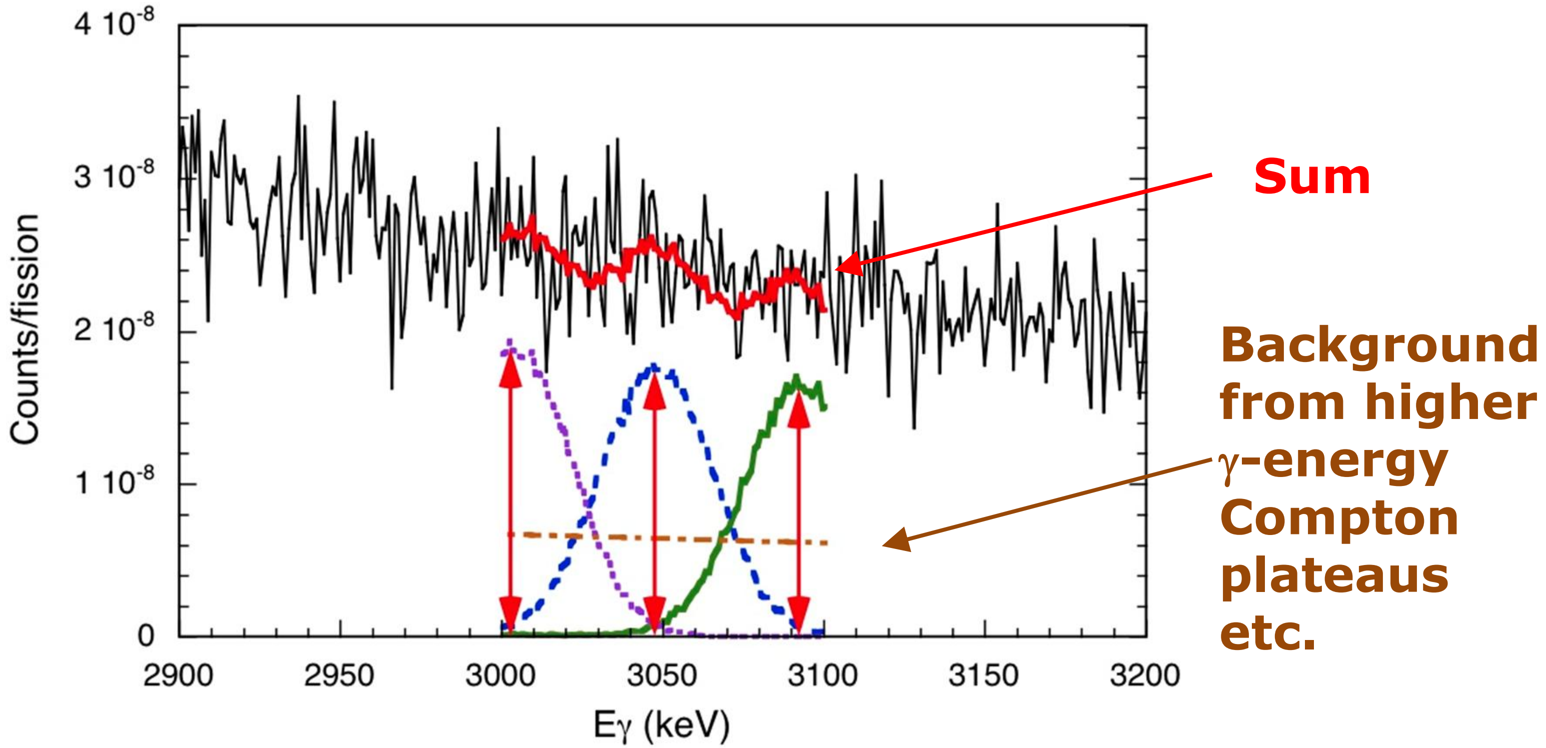
How to measure prompt fission γ-rays

➢ Unfolding the detector response



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

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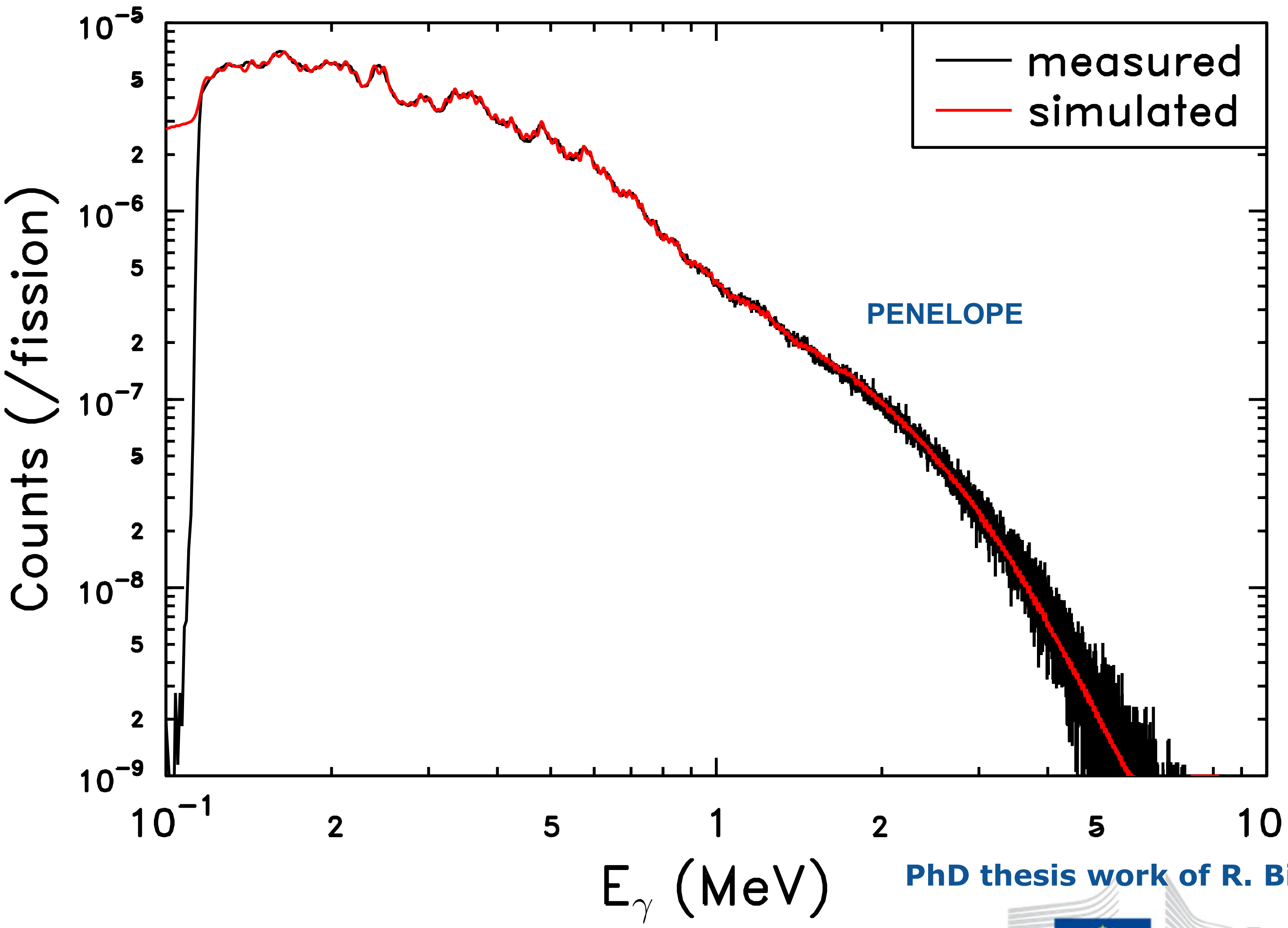
How to measure prompt fission γ-rays

➢ Unfolding the detector response



PhD thesis work of R. Billnert

R. Billnert, et al., Phys. Rev. C87, 024601 (2013)

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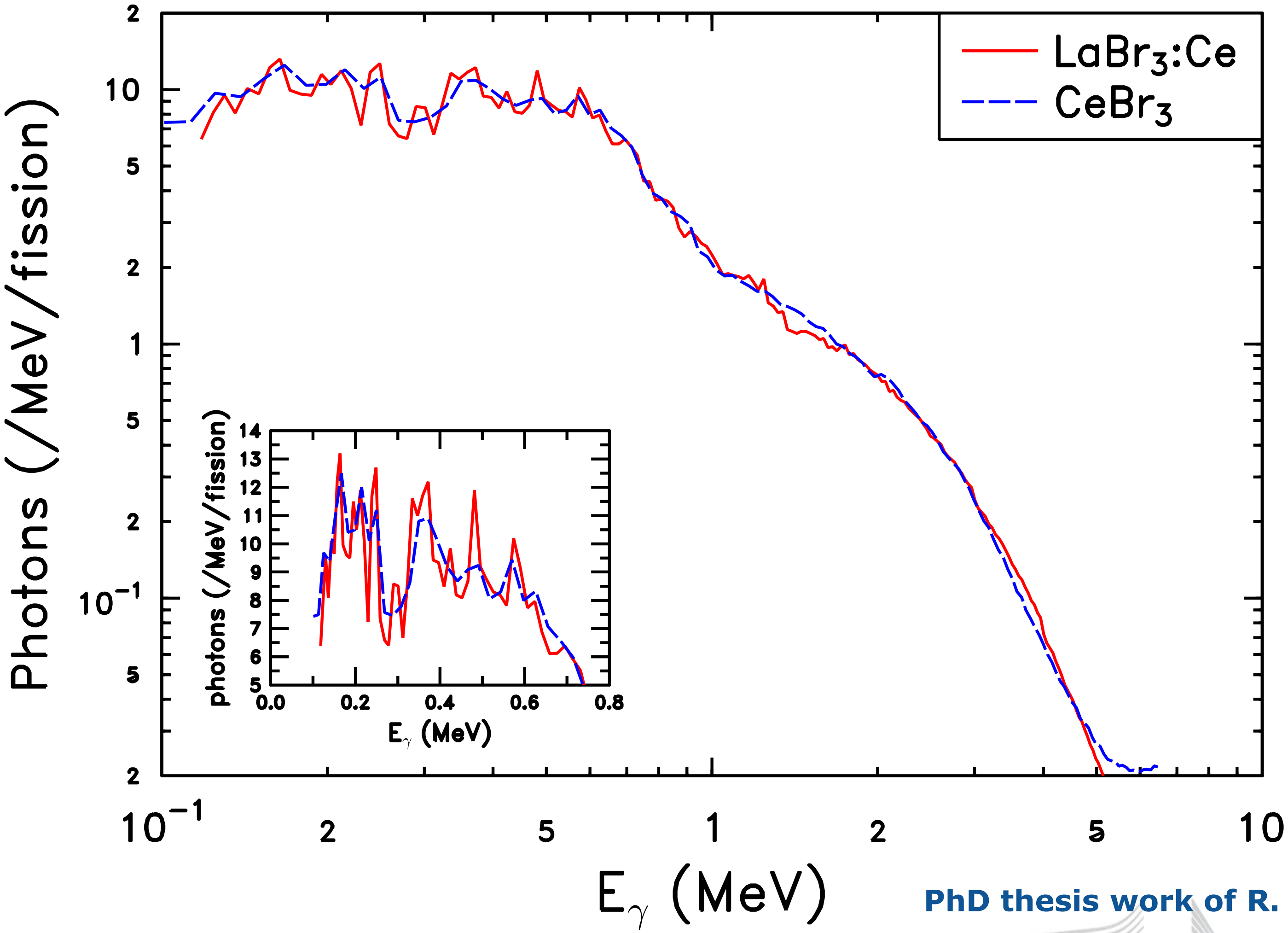
How to measure prompt fission γ-rays

➢ **Unfolded emission spectrum**



PhD thesis work of R. Billnert

R. Billnert, et al., Phys. Rev. C87, 024601 (2013)

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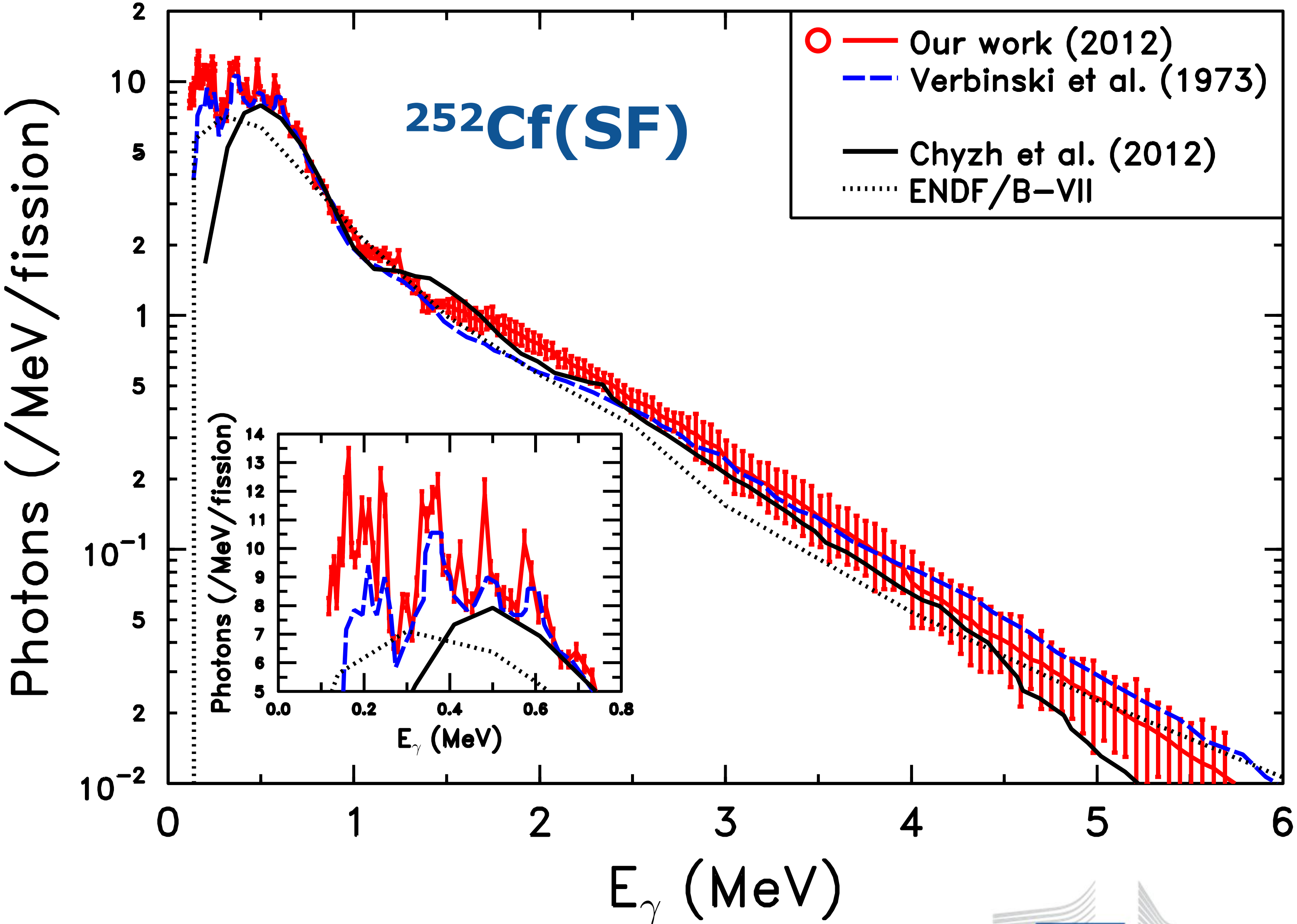
How to measure prompt fission γ-rays



R. Billnert et al., Phys. Rev. C87, 024601 (2013)

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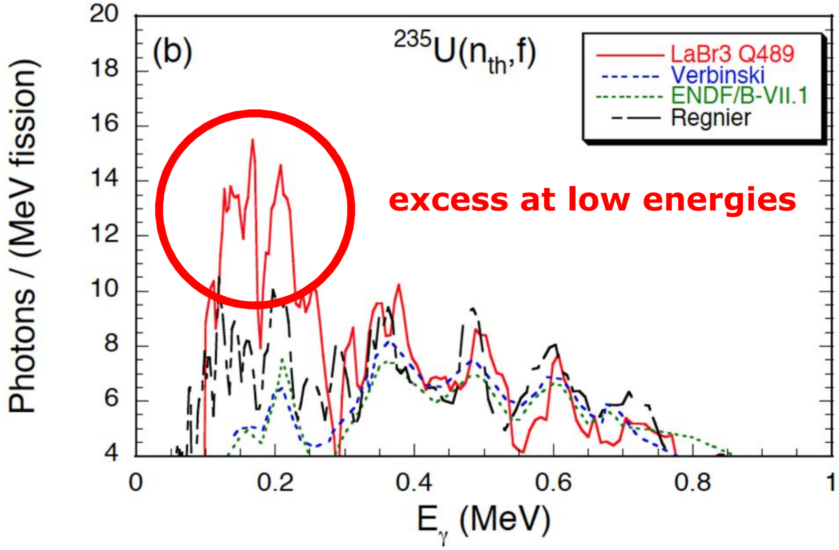
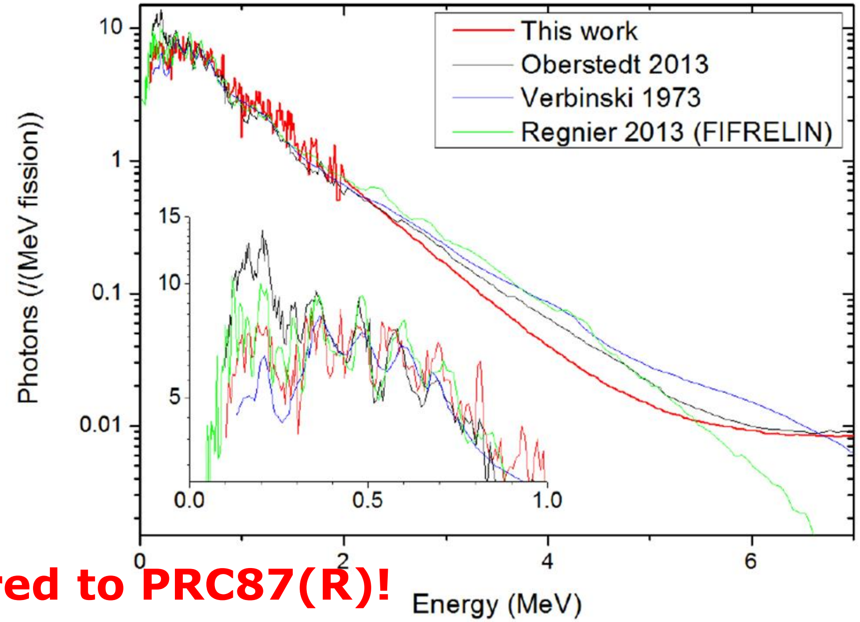
New detector response calculations



Situation as published in PRC87(R)

- M_γ = (8.19 ± 0.11) / fission
- $E_{\gamma tot}$ = (6.92 ± 0.09) MeV / fission

Situation after re-measuring at the GELINA neutron TOF facility



✓ M_γ = (7.4 ± 0.2) / fission
✓ $E_{\gamma tot}$ = (6.4 ± 0.2) MeV / fission

✓ **Decrease of 10% and 8% compared to PRC87(R)!**

PhD thesis of Angélique Gatera

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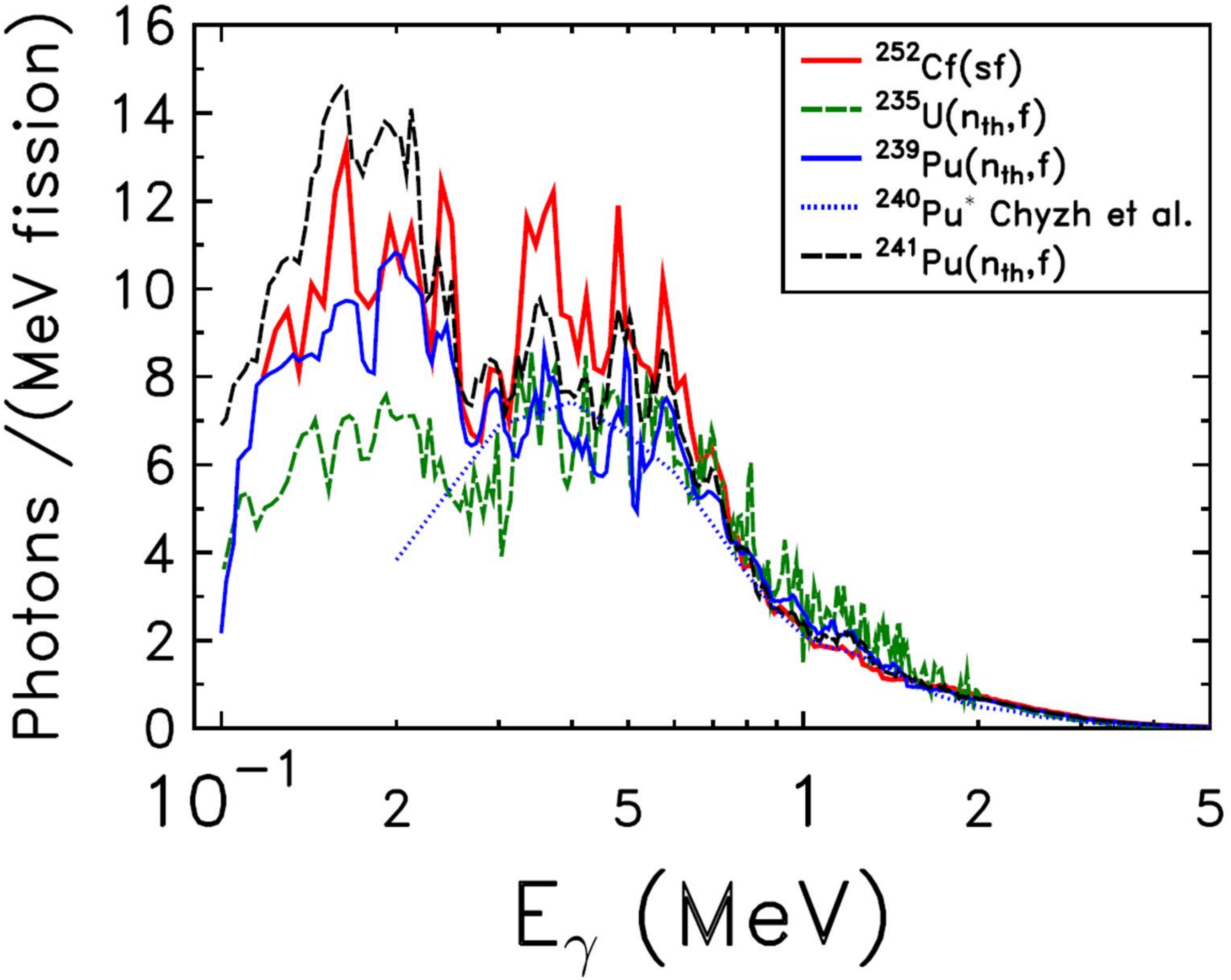
How to measure prompt fission γ-rays



^{241}Pu needs recheck of low energy contribution

PhD thesis work of A. Gatera

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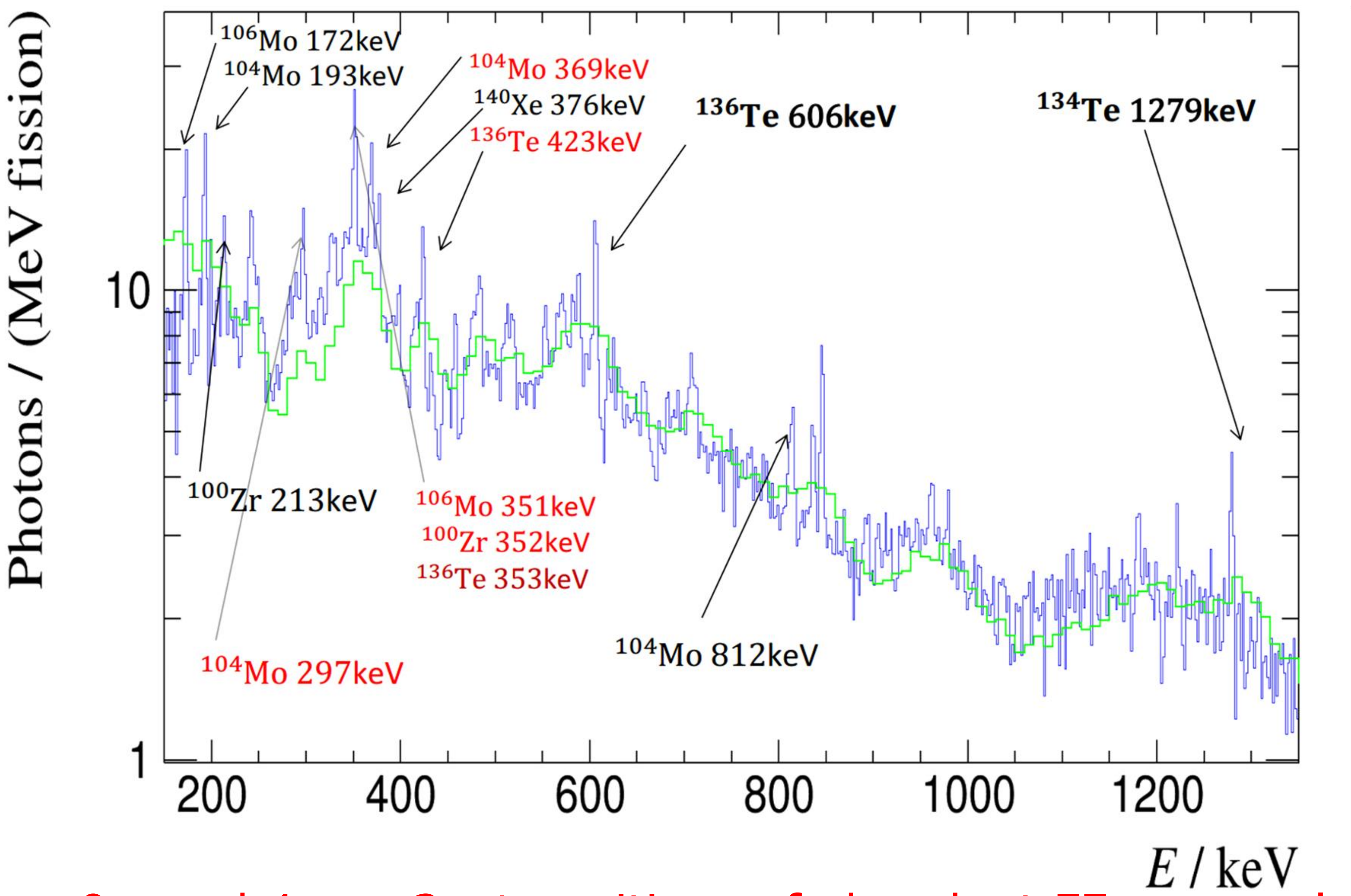
How to measure prompt fission γ-rays

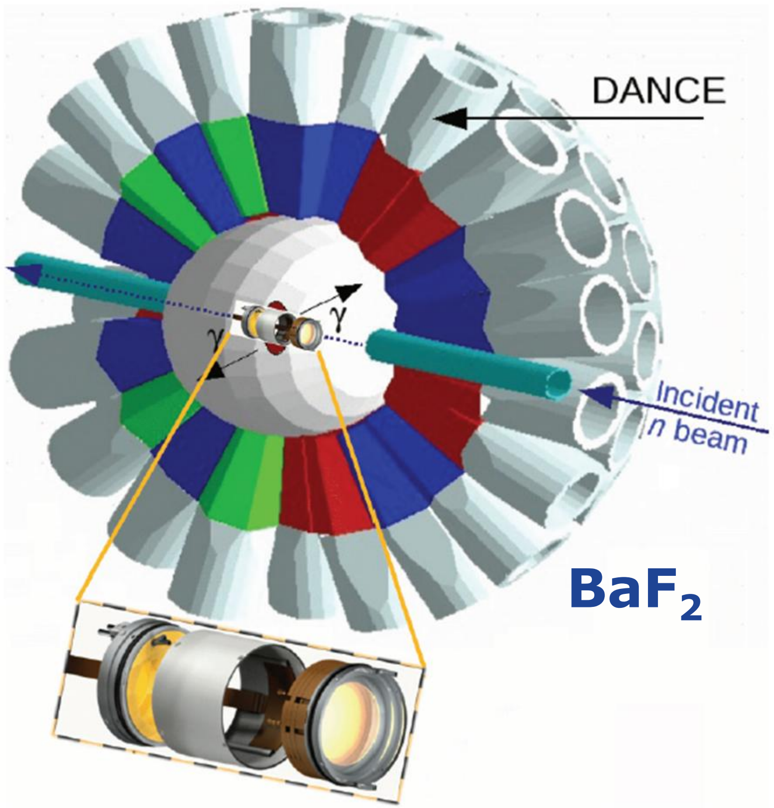


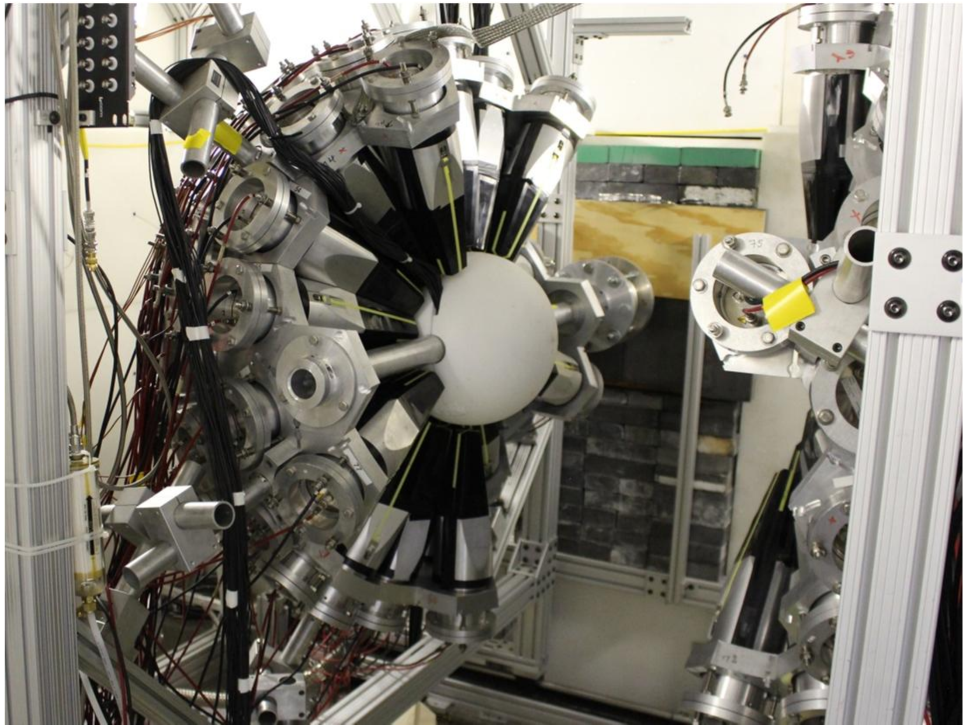
2+ → 0+ and 4+ → 2+ transitions of abundant FF are resolved

Use of combination of $LaBr_3$ and HPGe (nELBE)

How to measure prompt fission γ-rays

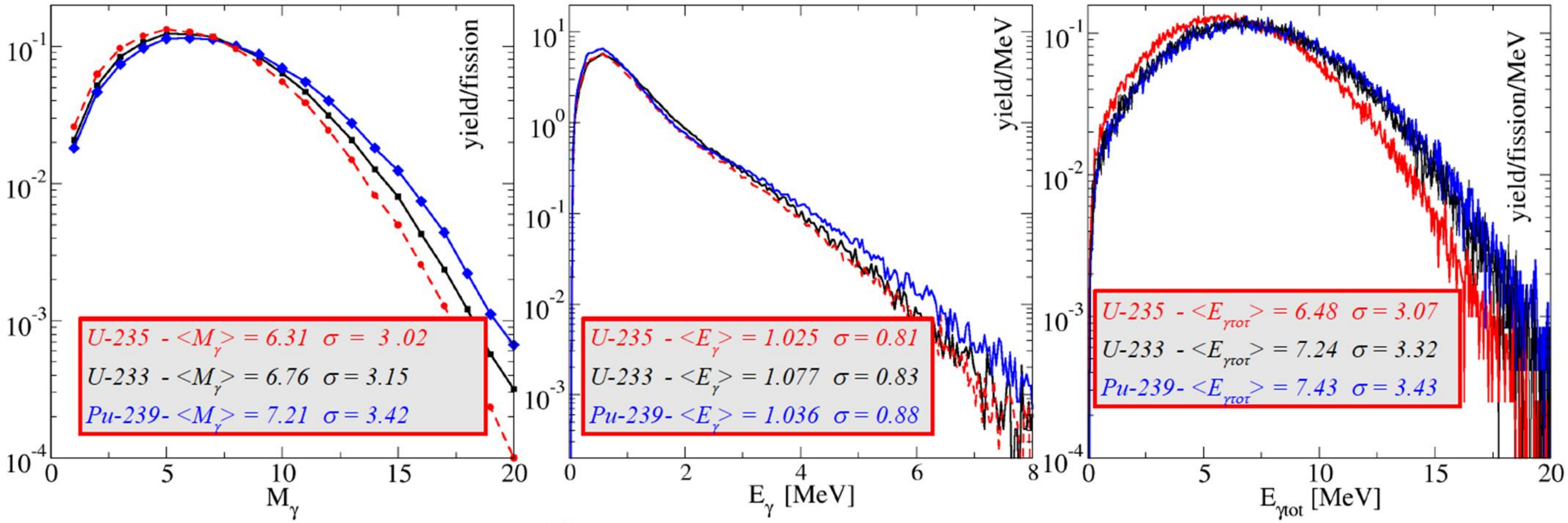
- **Other devices**
  - **DANCE (LANL), 4π array BaF_2 detector array)**

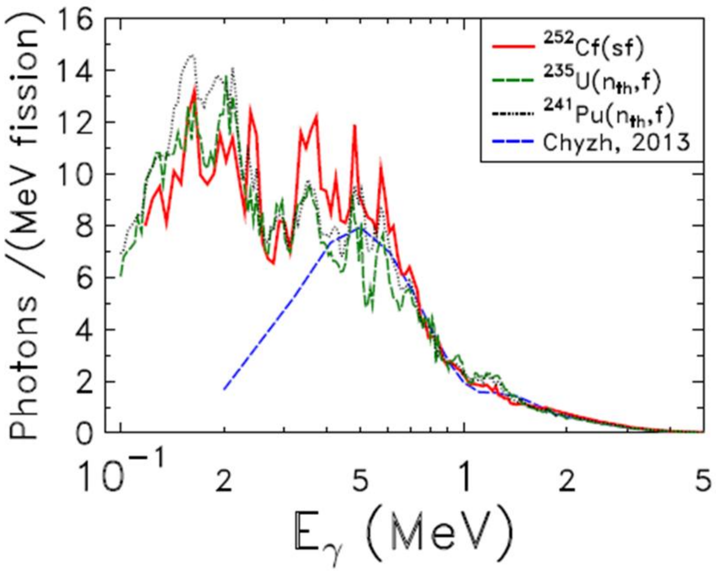




How to measure prompt fission γ-rays

M. Jandel, GAMMA-2 Workshop, Sremski Karlovci, RS





S. Oberstedt et al., Rad. Phys. Chem (2017) in press

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Conclusions

- Fission is a complicated process
- Prompt neutron and γ-ray emission gives insight into the physics of the process
- Improved detection systems allow to get better quality data
- Importance of correlations of particle emission and fission fragment properties
- Importance of simulation of the experiment
- Be honest and show all relevant information

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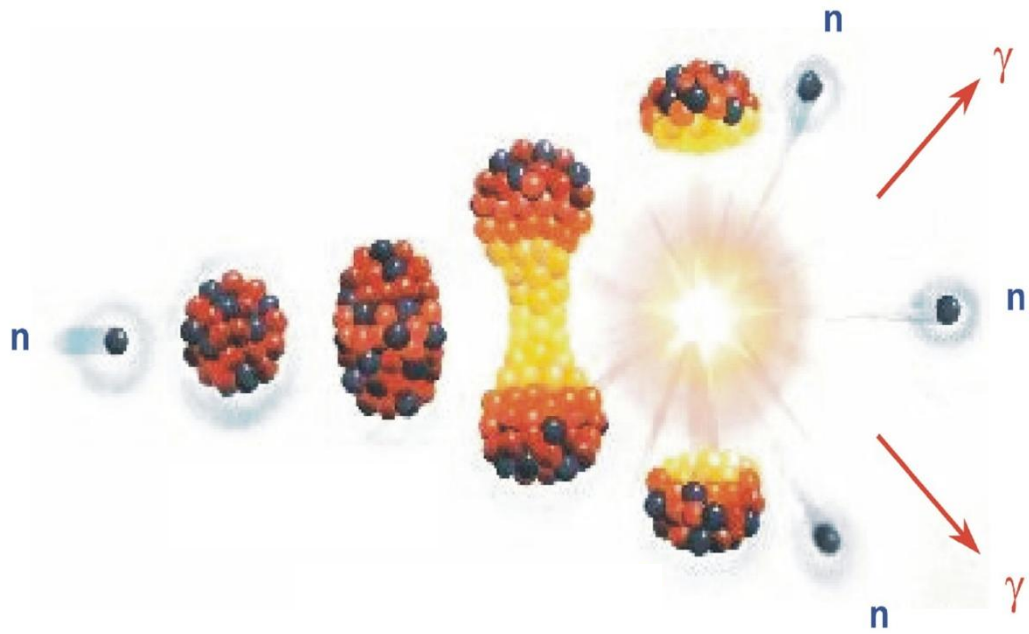
Thank you very much for Your attention



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