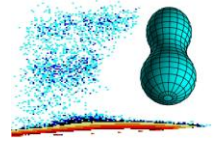


Measurement of prompt fission γ -rays with lanthanum halide scintillation detectors

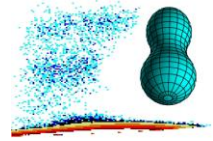
A. Oberstedt

This work was supported by the EFNUDAT programme of the European Commission
(agreement number 31027)



Outline

- Motivation
- Characterization of $\text{LaCl}_3(\text{Ce})$ detectors
 - intrinsic activity
 - energy resolution
 - intrinsic efficiency
 - timing resolution
- Experiment at IKI Budapest (cold neutrons)
- Results
- Summary and conclusions
- Outlook



Motivation

Assessment of γ -heating for design of Gen-IV reactors

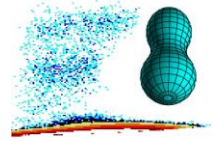
- about 10 % of total energy released in the core of a standard nuclear reactor by fission γ -rays
- about 40 % of those due to prompt γ -decay of fission products

Modelling requires uncertainty not larger than 7.5 % (1σ)

- but: present γ -ray emission data determined in early 1970's, underestimating γ -heating with 10 - 28 % for ^{235}U and ^{239}Pu

-> NEA Nuclear Data High Priority List:

- measurement of prompt γ -ray emission from $^{235}\text{U}(n,f)$ and $^{239}\text{Pu}(n,f)$!



Experimental task

Time-of-flight method to distinguish between γ -rays and neutrons (i.e. neutron-induced γ -rays)

-> requires good timing resolution

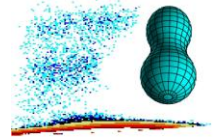
Back then:

- NaI detectors and ionization chambers
- $\tau \approx 3 - 5 \text{ ns}$ and $\tau \geq 1 \text{ ns}$

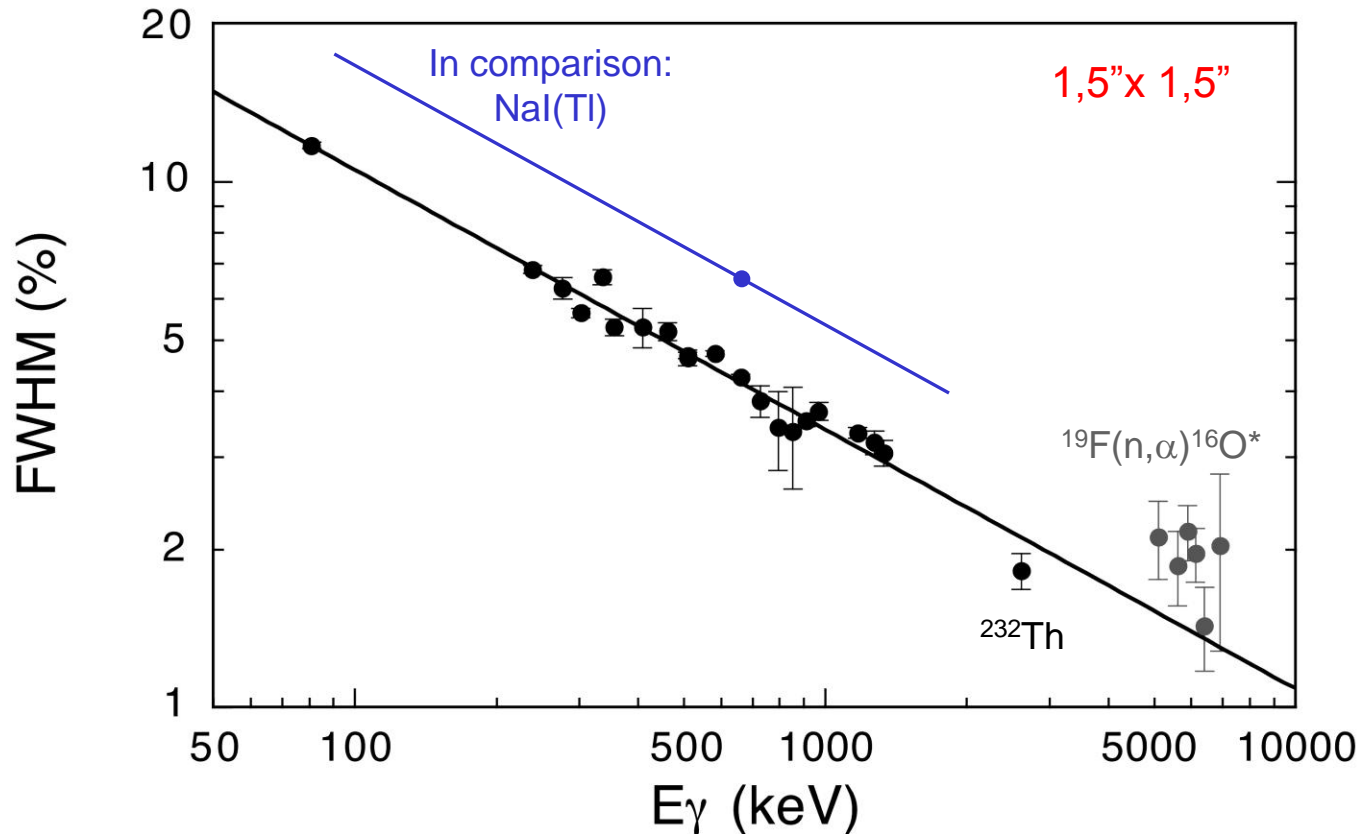
Today & tomorrow: new detectors offer new possibilities

- lanthanum halide crystals and pcCVD diamond detectors

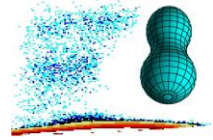
-> intended fission γ -ray measurements - a prerequisite to the assessment of γ -heating!



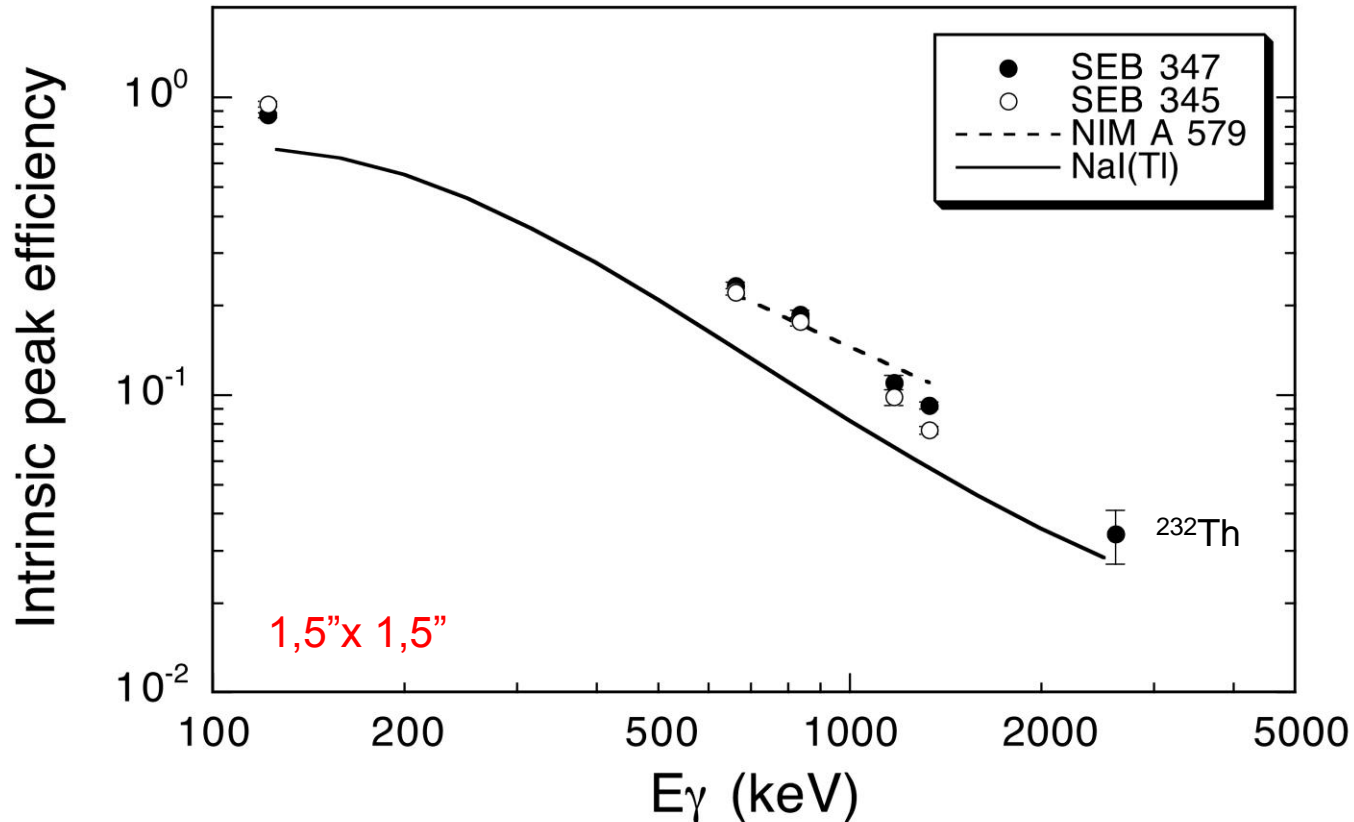
Energy resolution



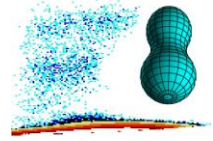
- expected $E^{-1/2}$ behaviour
- $\Delta E/E = 3.8 - 4.2 \%$ (FWHM) at 662 keV (^{137}Cs)



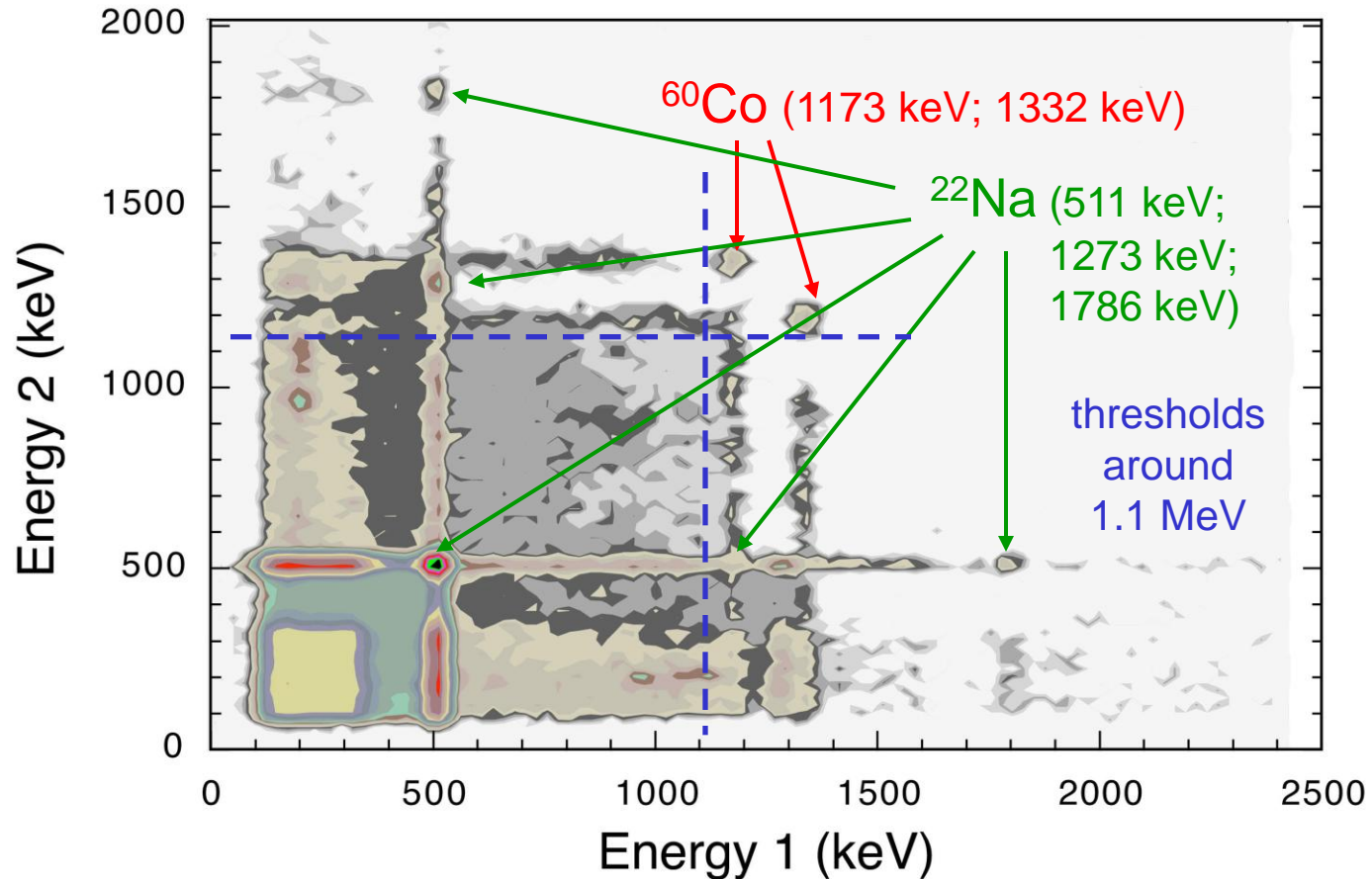
Intrinsic efficiency



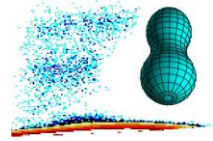
- in agreement with other LaCl_3 detectors (interpolated)
- 53 % better than NaI(Tl) detectors of same size



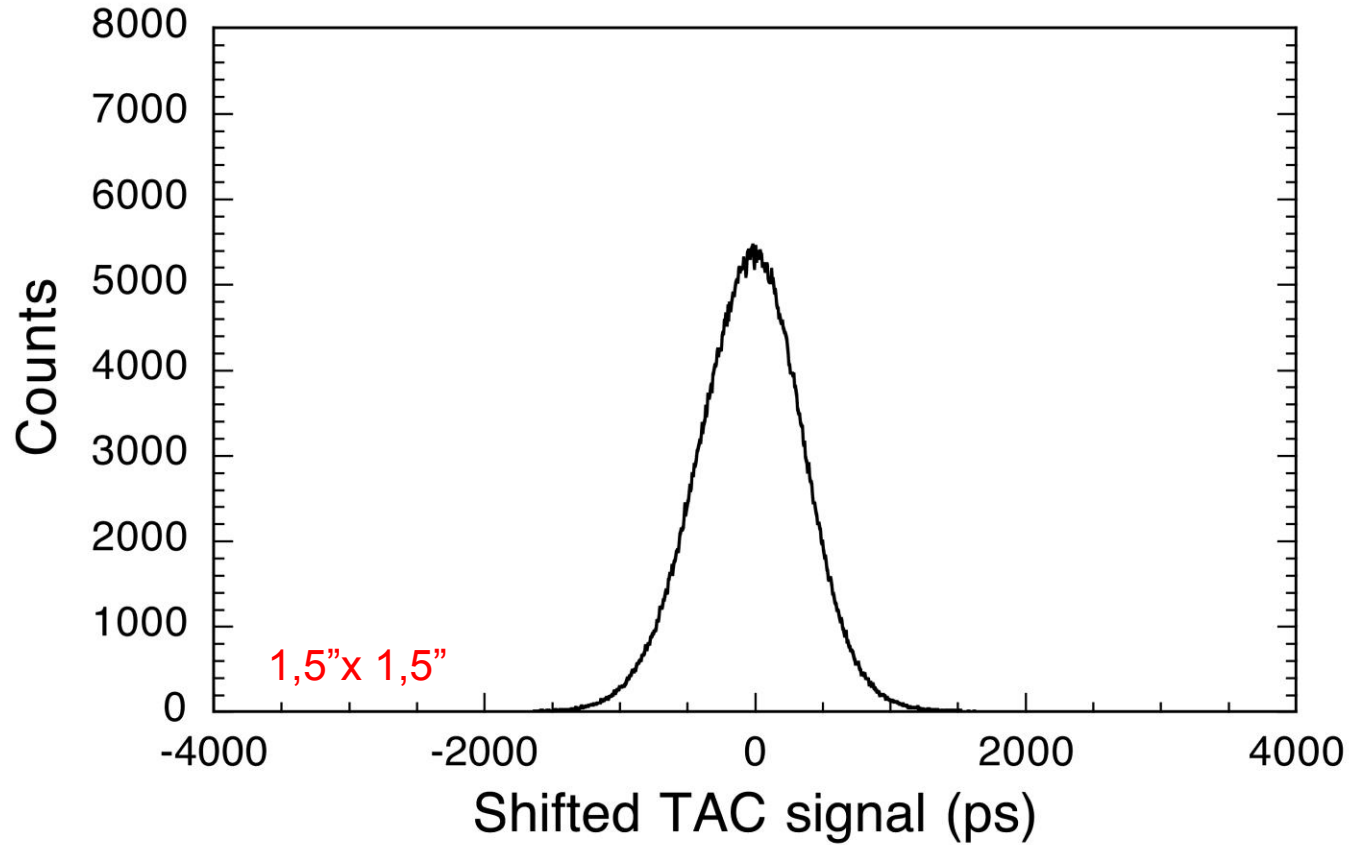
Timing - coincidence measurement

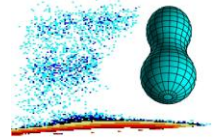


- two LaCl_3 detectors with multiple source (^{22}Na and ^{60}Co)

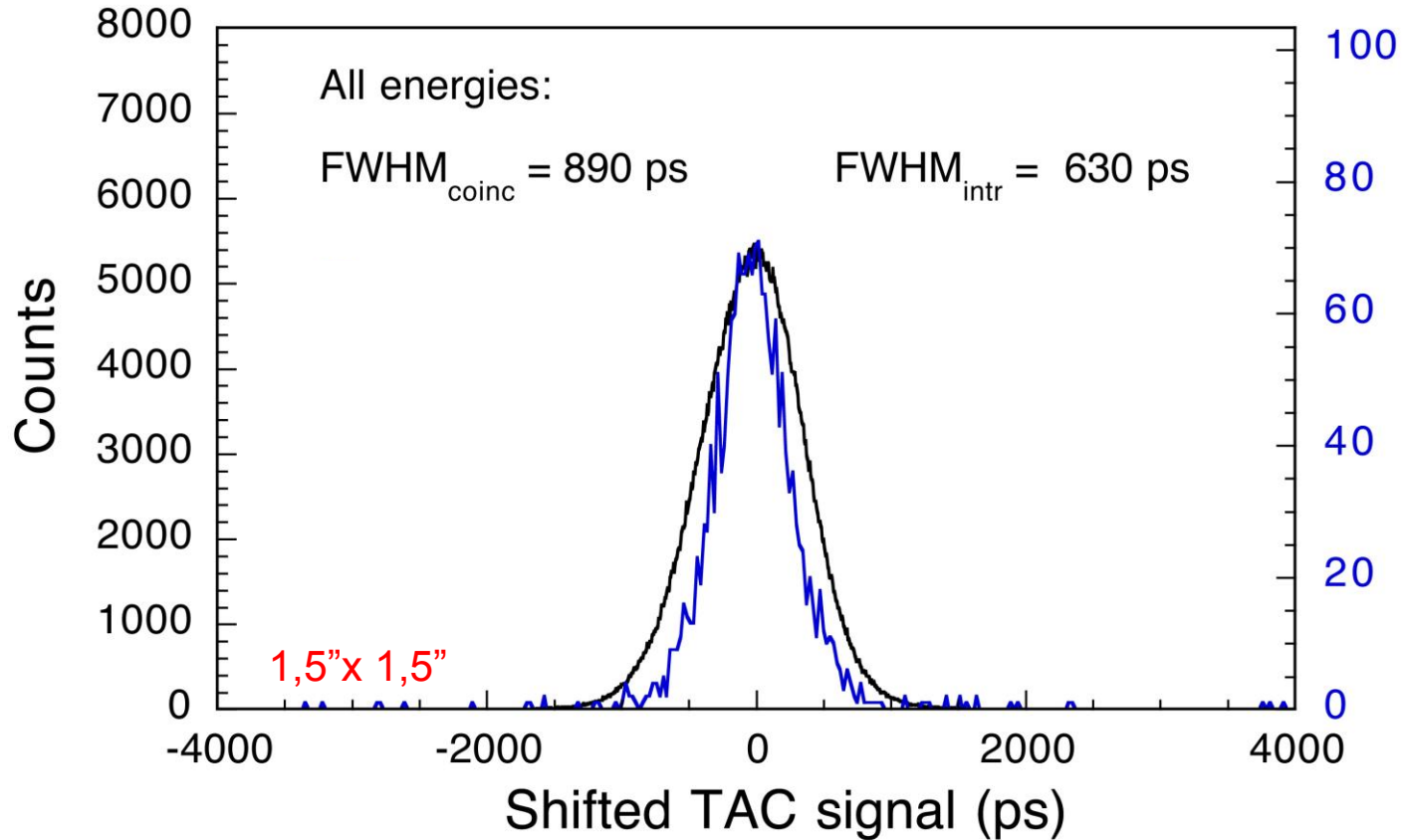


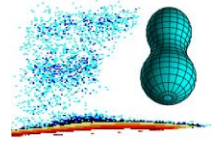
Timing - TAC spectrum





Timing - TAC spectrum





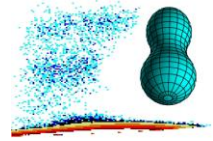
Fission-fragment and γ -spectrometry

Simultaneous measurement of:

- post-neutron fission fragment distributions of $^{235}\text{U} + n_{\text{th}}$
 - time-of-flight and kinetic energy
 - fission-fragment spectrometer VERDI (-> talk S. Oberstedt)
 - pcCVD diamond detectors as fission trigger
- prompt fission γ -rays
 - two $\text{LaCl}_3(\text{Ce})$ scintillation detectors (1,5" x 1,5", coaxial)
 - one $\text{LaCl}_3(\text{Ce})$ scintillation detector (3" x 3", coaxial)
 - one $\text{LaBr}_3(\text{Ce})$ scintillation detector (2" x 2", coaxial)
 - ^6Li shielding against thermal neutrons
 - coincidence with fission trigger (stop: fission - start: γ -ray)

Location:

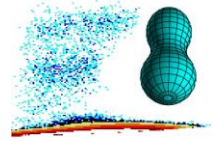
- 10 MW research reactor at IKI Budapest



Experimental details

Sample:	^{235}U (113 μg)
Thermal neutron flux:	$7 \cdot 10^7 \text{ cm}^{-2} \text{ s}^{-1}$
Fission rate:	$1.18 \cdot 10^4 \text{ s}^{-1}$
Fission fragment count rate:	12 s^{-1}
Fission γ count rate:	10 s^{-1}
Beam time:	2 weeks (10 days)
Expected number of counts	
– fission fragments:	$8.5 \cdot 10^6$
– γ -rays:	$\sim 3 \cdot 10^7$

Performed in February/March 2010



Experimental setup

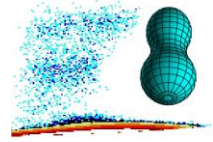
γ -detectors: LaBr_3 (2" x 2"), 2 x LaCl_3 (1,5" x 1,5"), LaCl_3 (3" x 3")



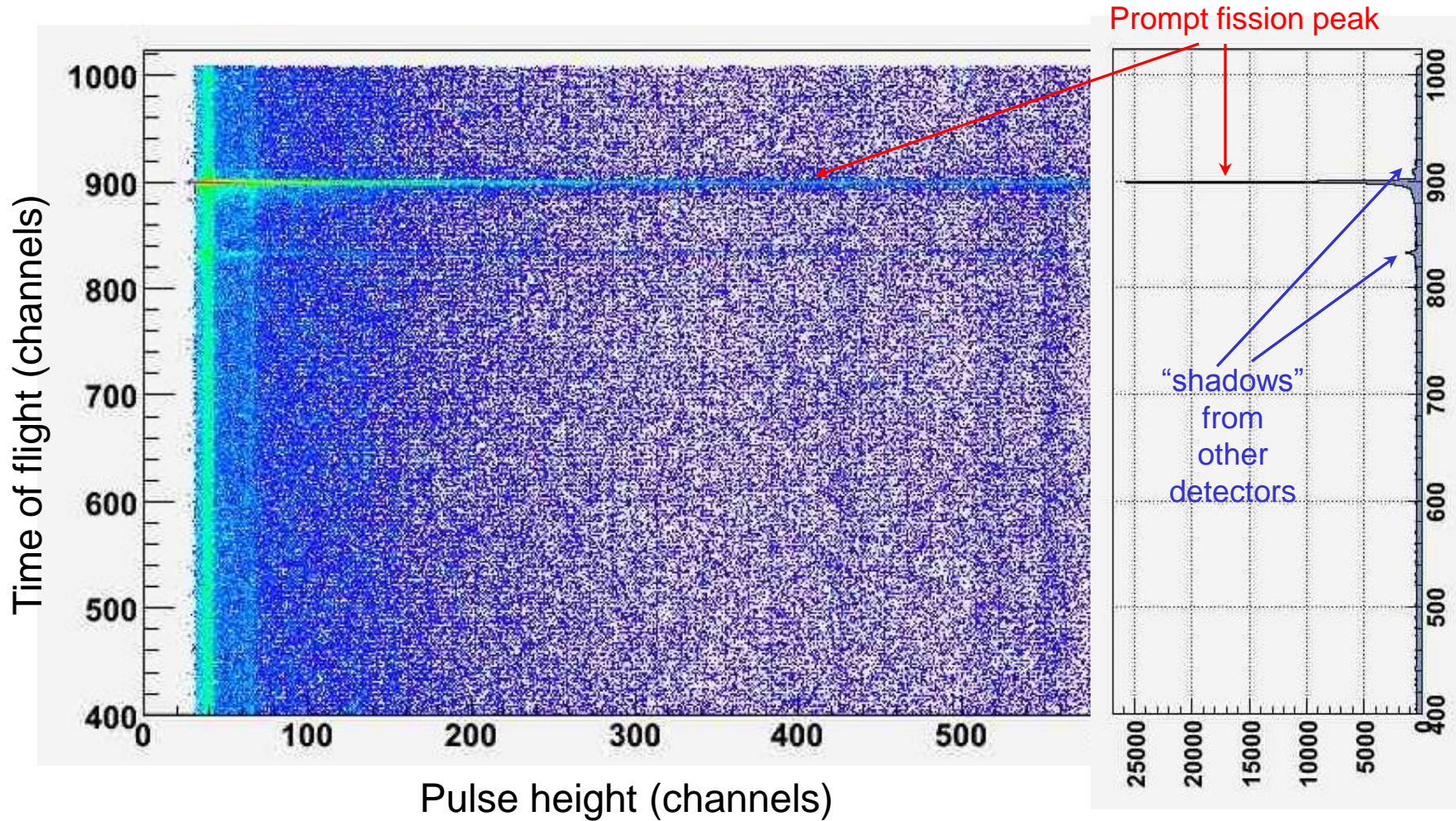
sample holder

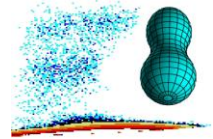
cold neutron beam

fission fragment spectrometer VERDI

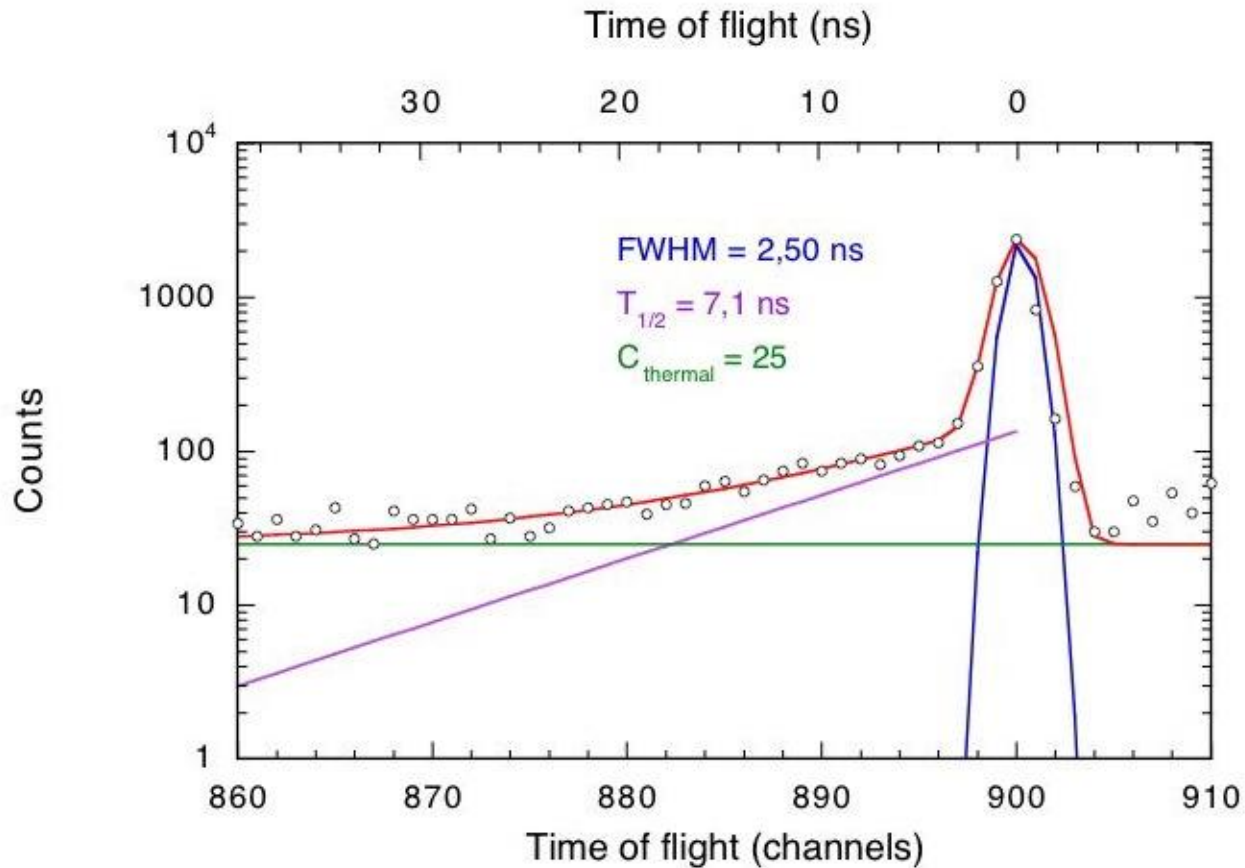


TAC signal vs. PH signal

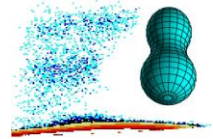




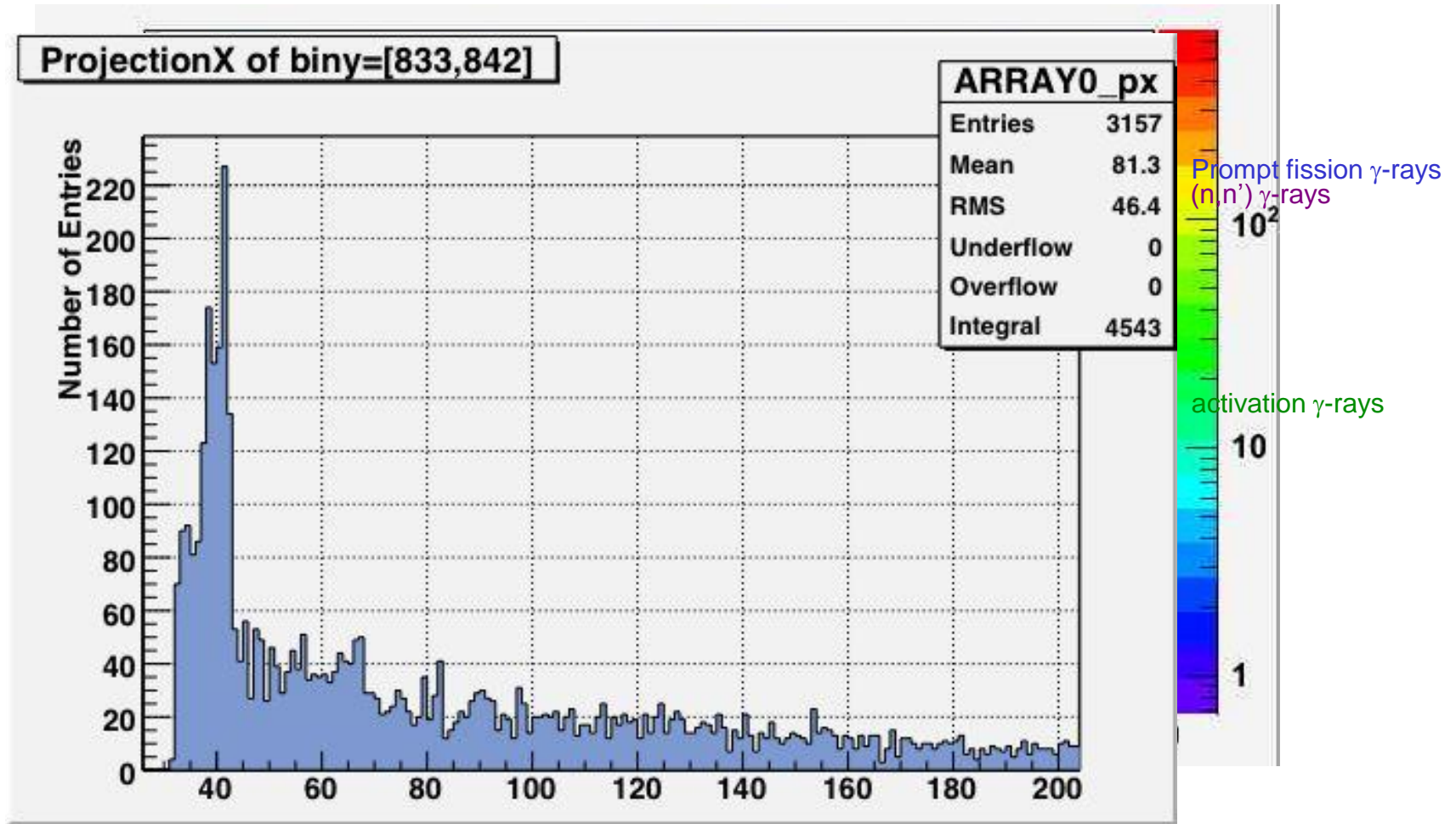
TAC spectrum (zoomed)

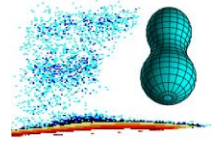


Prompt γ -peak + inelastic neutron scattering + neutron capture

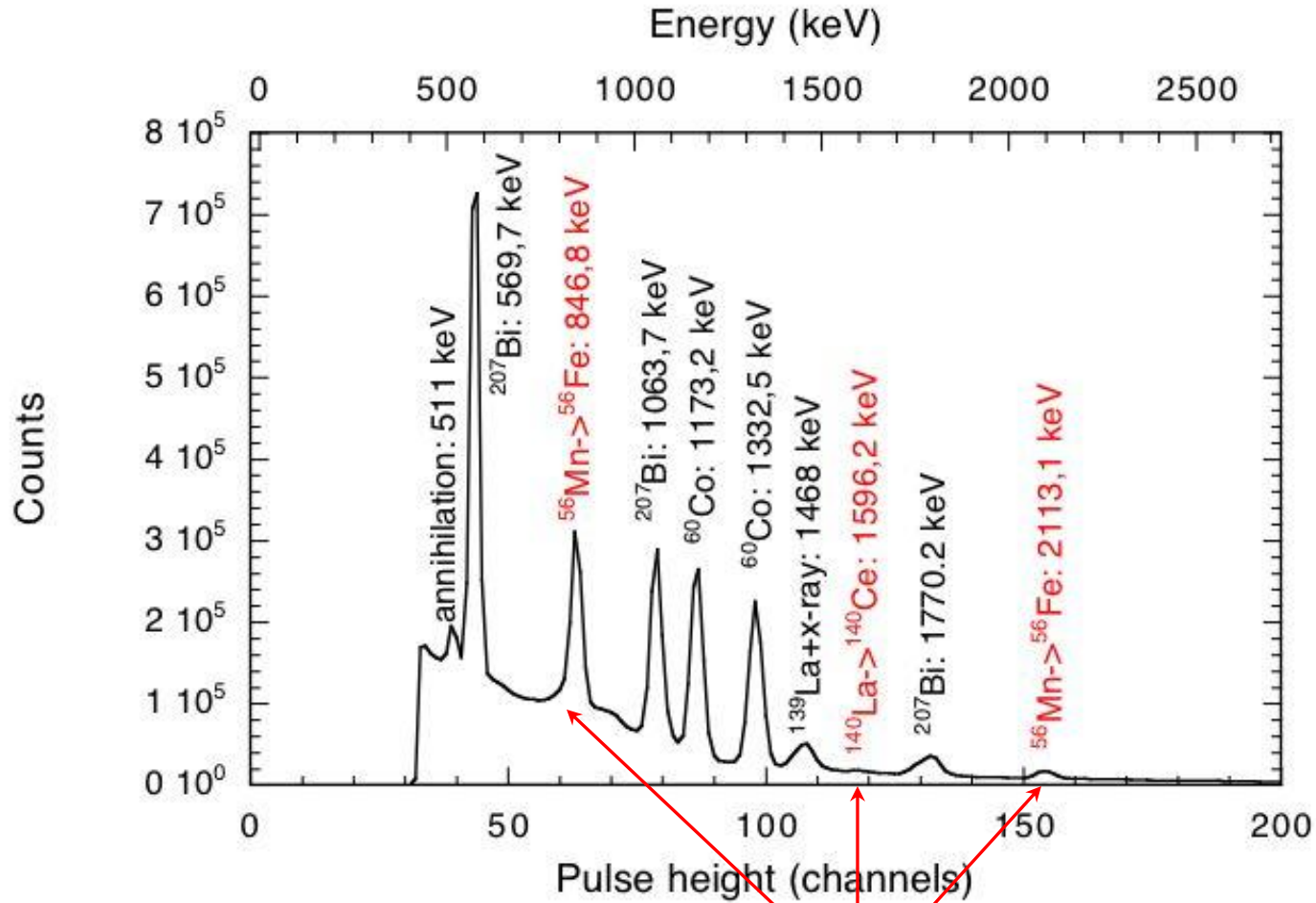


Characteristic spectra vs. TAC signature

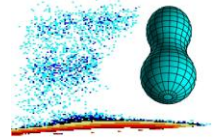




Energy calibration

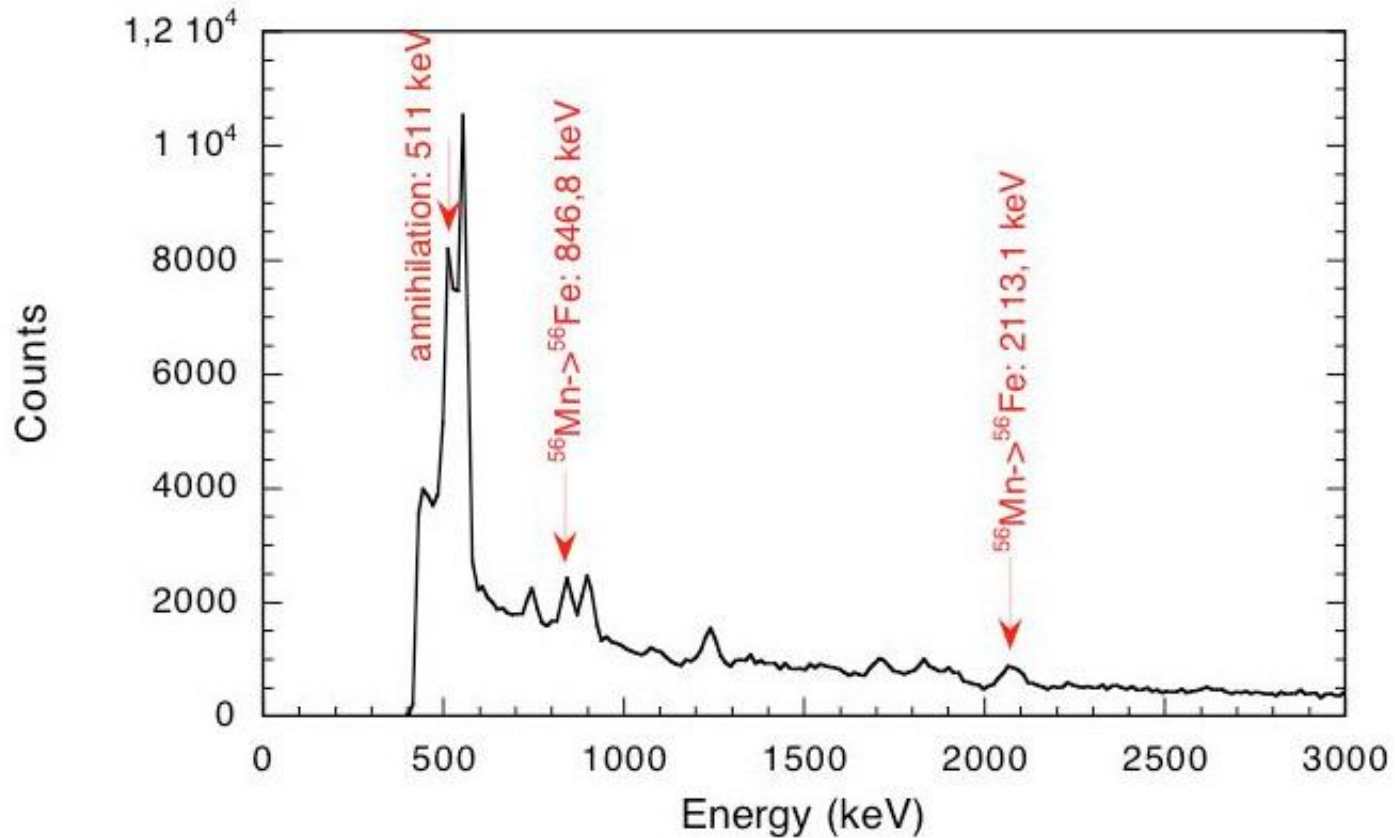


after irradiation!

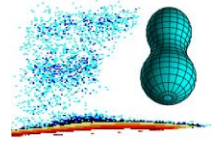


Preliminary results I

TAC regime of thermal neutron induced γ -rays: energy spectrum

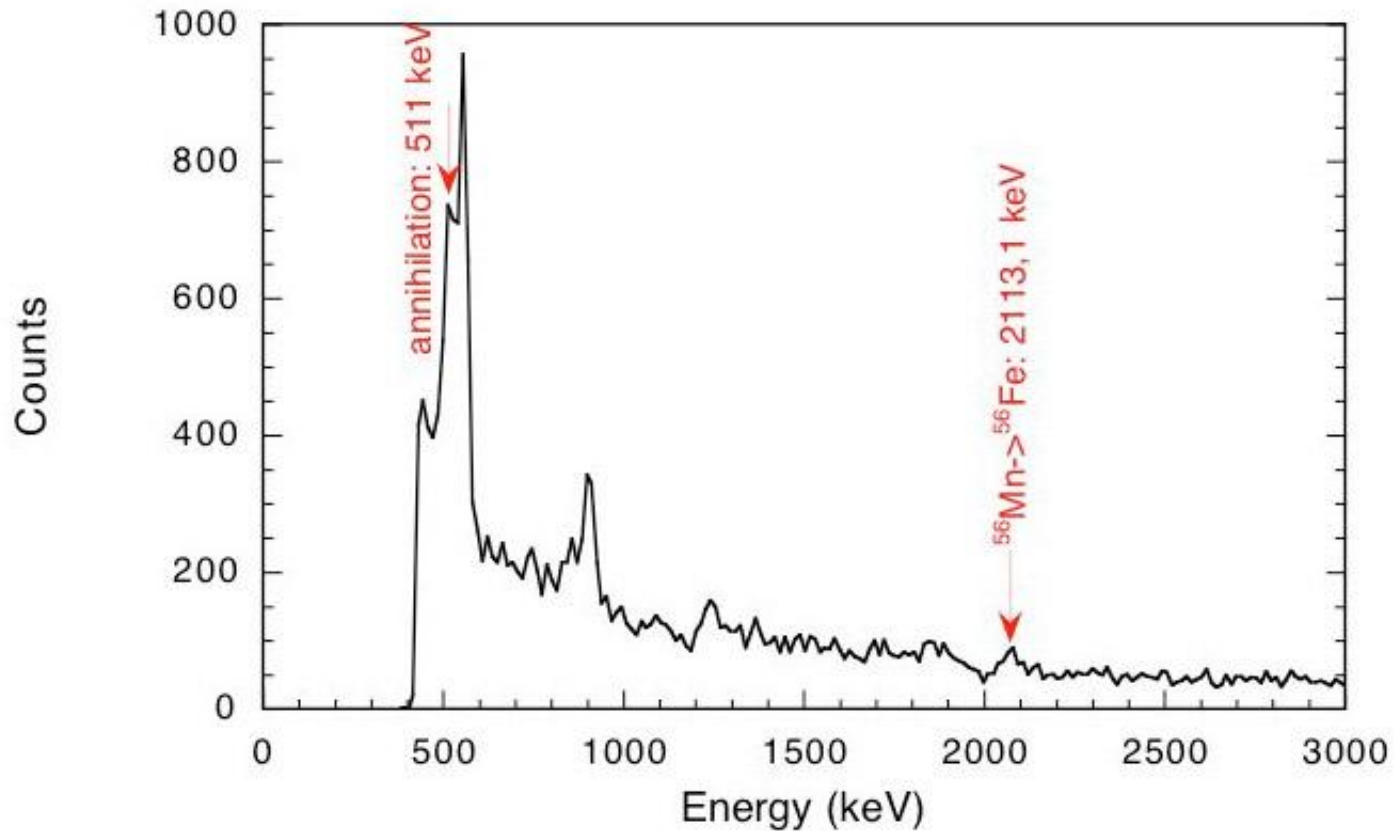


Activation of iron (VERDI): $^{56}\text{Fe}(n, p) \rightarrow ^{56}\text{Mn}$ ($T_{1/2} = 2.58$ h)!

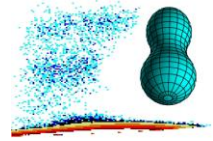


Preliminary results II

TAC regime of fast neutron induced γ -rays and isomers

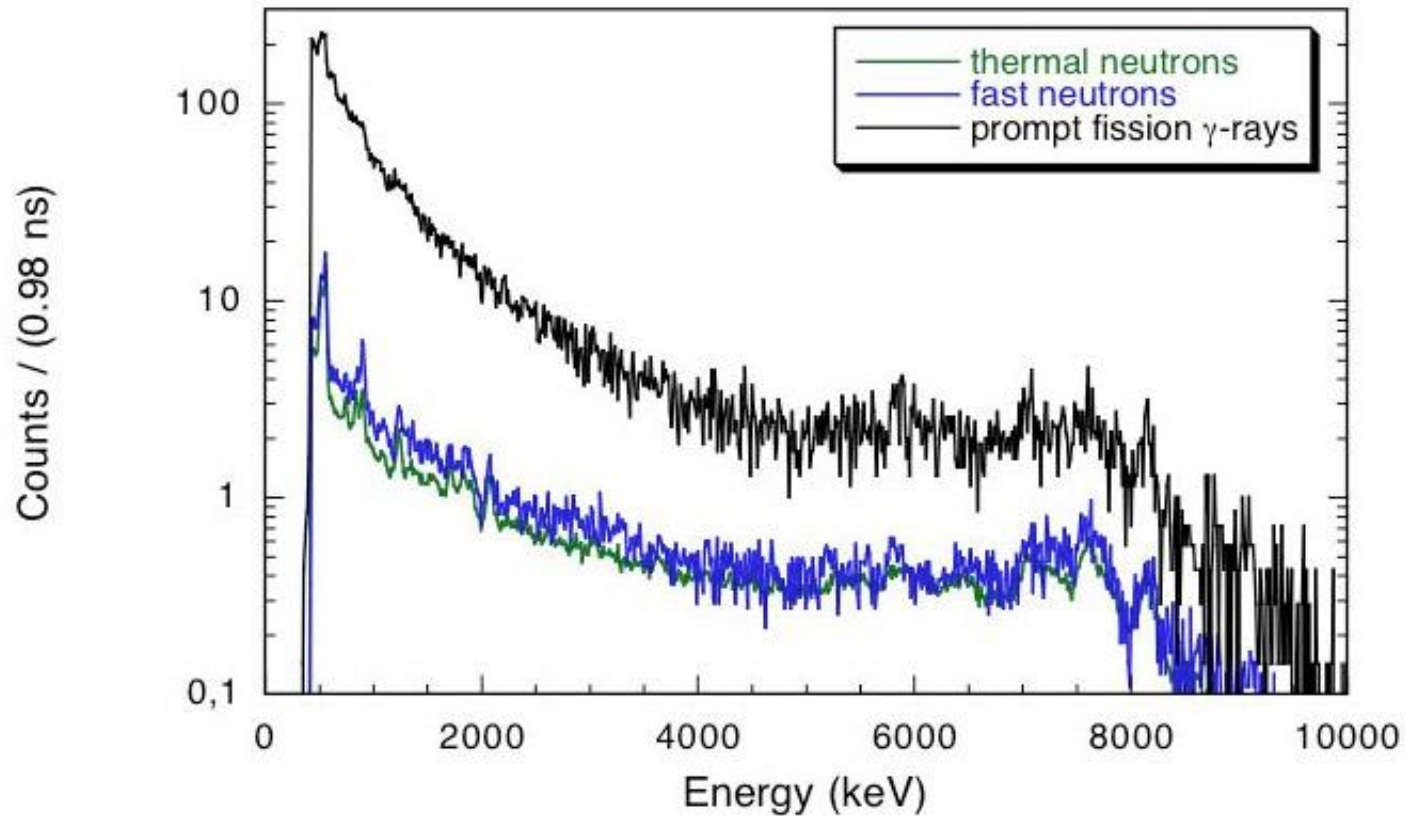


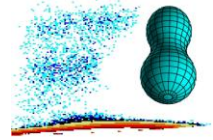
Observe: more peaks have still to be identified!



Preliminary results III

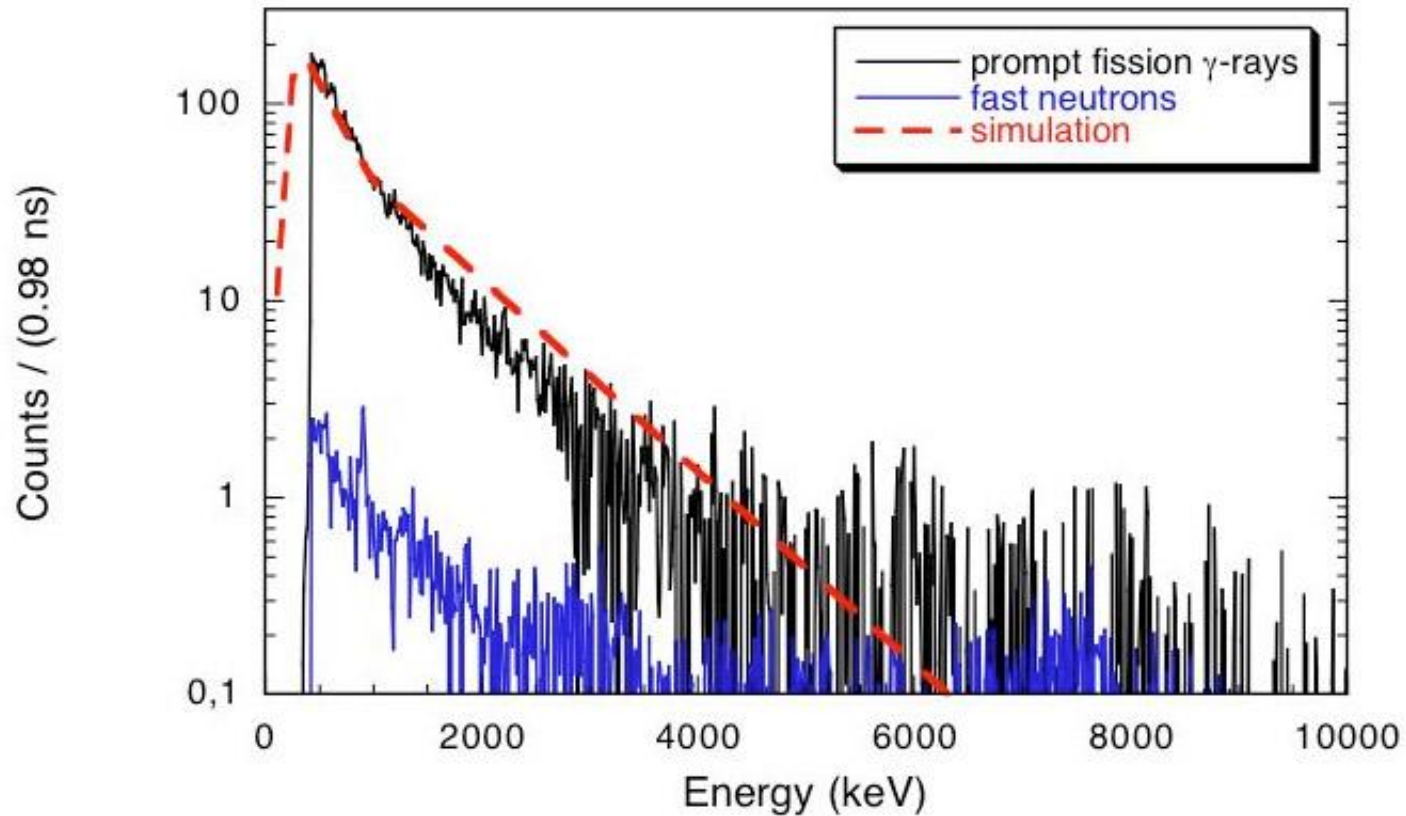
All TAC regimes: energy spectra - normalized & calibrated



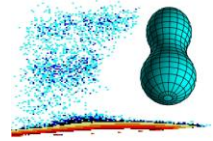


Preliminary results IV

Prompt fission γ -spectrum - background subtracted



To guide the eye: **simulation according to Verbeke et al., LLNL (2009)**

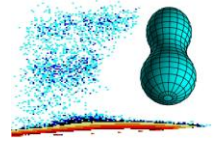


Background on simulation

Prompt fission γ -energy distribution

$$N(E) = \begin{cases} 38.13(E - 0.085)e^{1.648E} & E < 0.3 \text{ MeV} \\ 26.8e^{-2.30E} & 0.3 < E < 1.0 \text{ MeV} \\ 8.0e^{-1.10E} & 1.0 < E < 8.0 \text{ MeV} \end{cases}$$

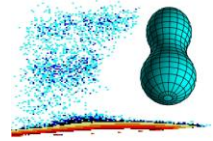
Obtained from measurements by Maienschein et al.,
Neutron Phys. Ann. Prog. Rep., ORNL (1958)



Summary and conclusions I

Characterization of $\text{LaCl}_3:\text{Ce}$ detectors (1,5" x 1,5"):

- Energy resolution $\Delta E/E \leq 4\%$ at 662 keV
 - 40 % better than NaI (TI) up to $E_\gamma \approx 7$ MeV
- Intrinsic peak efficiency determined
 - 50 % better than NaI (TI) of same size up to $E_\gamma \approx 2.6$ MeV
- Timing resolution $\tau_{\text{intr}} \approx 630$ ps (FWHM) for entire energy range (441 ps for ^{60}Co)
 - $\tau_{\text{intr}} \approx 3 - 5$ ns for NaI(TI)
 - previously published $\tau_{\text{intr}} \approx 300$ ps, but smaller detectors and higher Ce-concentration
- Considerable intrinsic activity, but controlled by coincidence
- Good linearity (residuals $< 0.3\%$ above 100 keV)
- Dynamical range up to ~ 17 MeV γ -rays



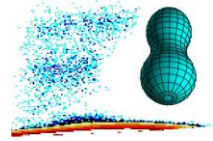
Summary and conclusions II

LaCl₃ scintillation detectors do indeed fulfill requirements for the measurement of prompt fission γ -rays, in particular in conjunction with **pcCVD diamond detectors** ($\tau \approx 300$ ps)

- excellent timing resolution
- improved n/ γ -discrimination (time-of-flight method)
- neutron sensitive, but no activation of LaCl₃ by fast neutrons

Prompt fission γ -rays from $^{235}\text{U}(n_{\text{th}}, f)$ measured:

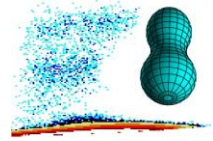
- γ -rays distinguished with respect to their origin (fission, neutron-induced, β -delayed ...)
- preliminary prompt fission γ -spectrum presented
- interactions between thermalized neutrons and detector material extremely rare, i.e. $\Phi(n_{\text{th}}) < 0,2 \text{ s}^{-1} \text{ cm}^{-2}$



Outlook

To come:

- data analysis to be completed (3 more detectors)
- response functions have to be determined and applied
- **LaBr₃:Ce (2" x 2")** detectors to be characterized
- new **LaCl₃:Ce (1.5" x 2")** detectors to be characterized
- effect of **BGO** anti-Compton shield to be investigated
- new experiment contemplated: detection of fission γ -rays during measurement of ²⁵²Cf(sf) at IRMM with VERDI
- ...



Collaborators



Örebro University

R. Billnert, A. Göök, J. Karlsson, A. Oberstedt



JRC - IRMM Geel

R. Borcea, F.-J. Hamsch, S. Oberstedt



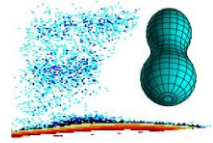
IKI Budapest

T. Belgya, Z. Kis, L. Szentmiklosi, K. Takács



CIEMAT Madrid

D. Cano-Ott, T. Martinez-Perez



Thank you!



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DEPARTMENT OF SCIENCE

