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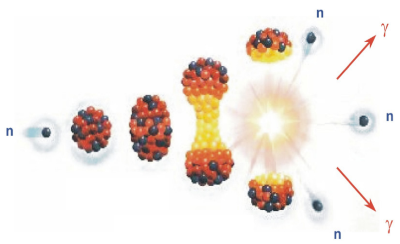
Institute for Reference Materials and Measurements



Correlation measurements of fission-fragment properties

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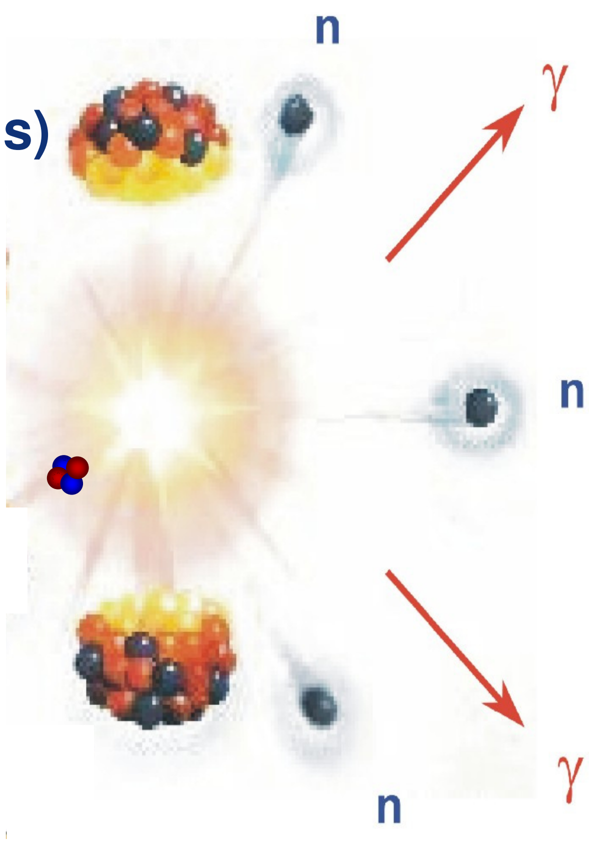
- Motivation
- Correlation measurements
- VERDI – the TOF spectrometer
- Characterization of $LaCl_3$:Ce detectors
- First (correlation) experiment @ IKI
- Conclusion & Outlook

Motivation

prompt neutrons (10^{-18} s)

prompt γ-rays (10^{-16} s)

fission fragments (10^{-21} 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)

Motivation

✓ **Reliable predictions on fission product yields relevant in modern nuclear applications (GEN-IV, ADS…)**

- Radio-toxicity of the nuclear waste
- γ-ray heating
- Decay heat calculations
- Delayed neutron yields relevant during reactor operation

- **Prediction of fission-fragment mass and kinetic energy distributions of minor actinides (^{237}Np, ^{241,243}Am, $^{242-248}$Cm)**
- **Delayed neutron emission pre-cursor yields**

- **Emission spectrum and multiplicity (as a function of fragment mass) of prompt γ-rays and neutrons**

⇨ **Experimental data as input for nuclear reaction models**

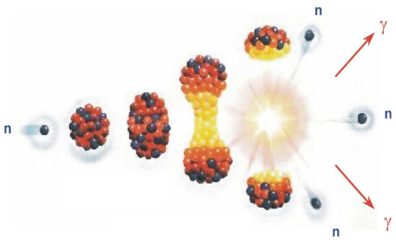
Correlation measurements

- **Prediction 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

⇨ **OECD NEA Nuclear Data High Priority List:**

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

Correlation measurements



- **Fission fragment (FF) yield Y(A, E_{kin}; TKE)**
- **Prompt fission γ-ray/neutron spectra; ε(A)**
- **Prompt fission γ-ray/neutron multiplicity ν(A, TKE)**

⇨ **High geometrical efficiency and decent mass resolution**

Correlation measurements

- **Compact (Frisch-grid) ionization chamber (ε > 50%)**
- **Diamond-based fission fragment spectrometers**
  - ✓ 2E detectors based on (large area/segmented) silicon detectors (ε < 20%)
  - ✓ Compact 2E detectors using segmented sCVDD detectors (ε < 20%)
  - ✓ Double (v, E) spectrometer using p/sCVD diamond detectors (ε < 1%)
- **Ultra-fast diamond-based fission event triggers for spectral neutron and γ-ray measurements**

✓ Simultaneous measurement of kinetic energy and velocity of both fission fragments, 2(v, E)♥

- 2v $\rightarrow$ pre-neutron masses, A_i^* (i = l, h), TKE
- v, E $\rightarrow$ post-neutron masses, A_i, $E_{k,i}$ (i = l, h)

➢ $\nu_i(A_i^*)$ from the difference $A_i^* - A_i \rightarrow$ **TXE(A_i)**

➢ **prompt (and delayed) decay modes of FF**

♥ Cosi Fan Tutte (ILL)

VERDI – the concept

- spectrometer efficiency $\varepsilon \approx 0.005 - 0.01$ ♥

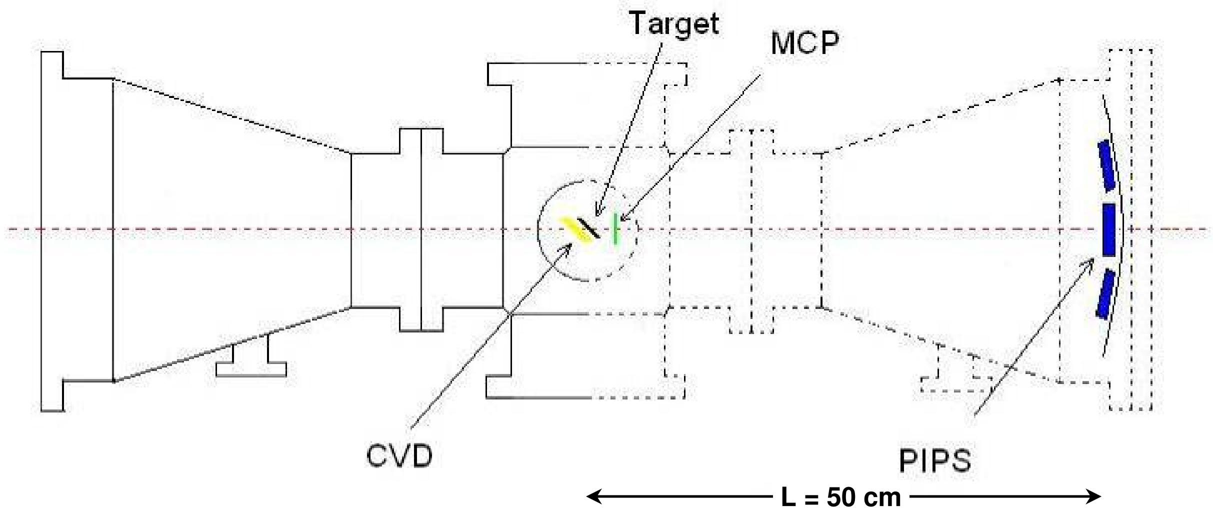
- high resolution energy detector ($\Delta E/E = 0.006$)
- high precision (transmission) time pick-up with $\tau < 200$ ps @ L = 50cm

- for a mass resolution of $\Delta A < 2$

- radiation hardness of the fission time pick-up

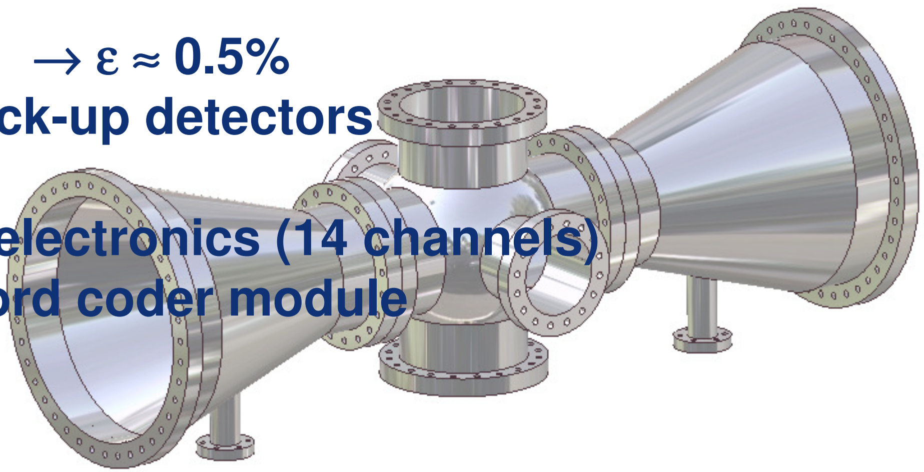
♥ Cosi Fan Tutte ($\varepsilon \approx 5\times10^{-5}$)

VERDI - the design

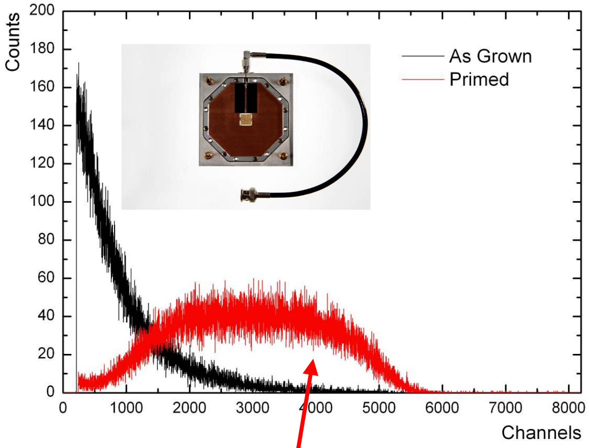


- ✓ 2 x 19 PIPS detectors (450 mm^2) $\rightarrow \varepsilon \approx 0.5\%$
- ✓ <u>pCVDD</u> or MCP ultra-fast time pick-up detectors

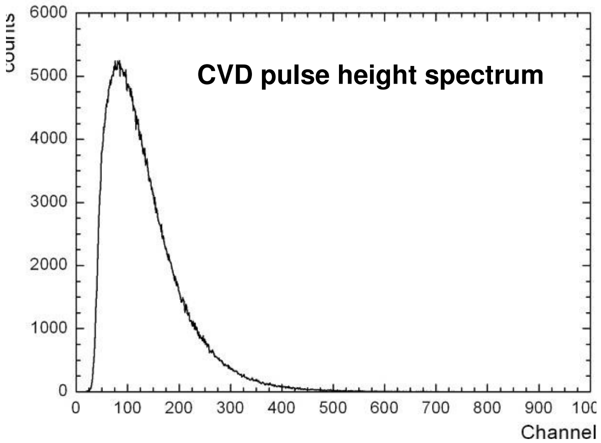
- ✓ set-up can be handled with NIM electronics (14 channels)
- ✓ development of an ASR + tag-word coder module

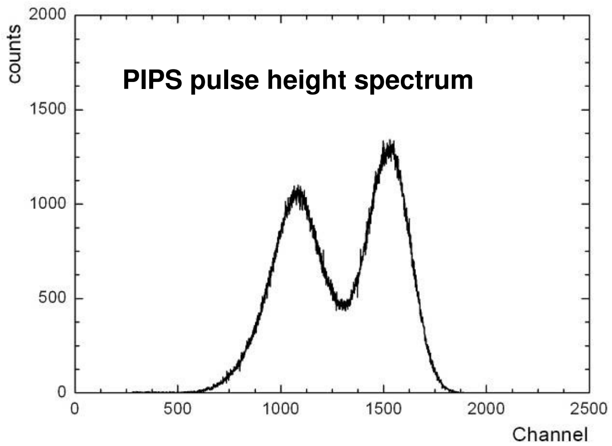


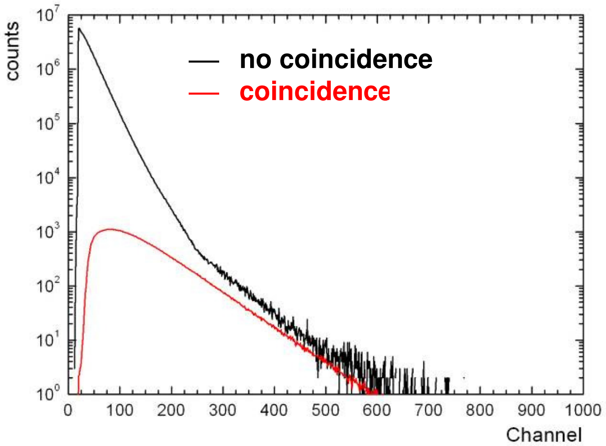
pCVD diamond: ultra-fast fission trigger

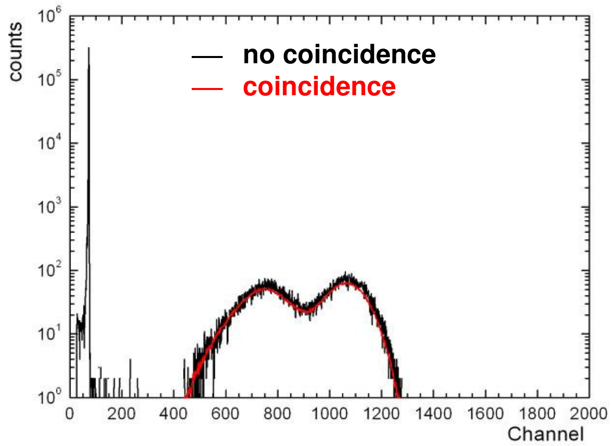


irradiated with a $^{90}Sr/^{90}Y$ β-source (3MBq, 72h) keep away from ambient light !!!

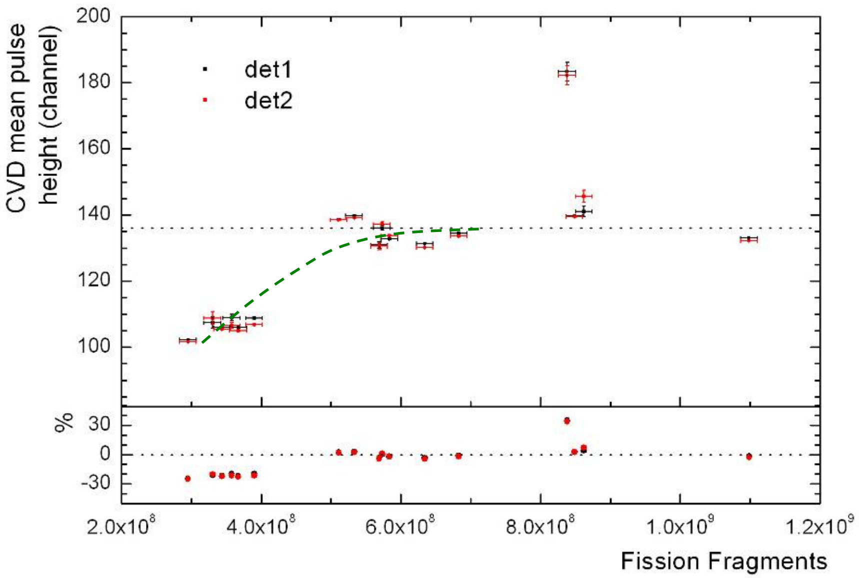


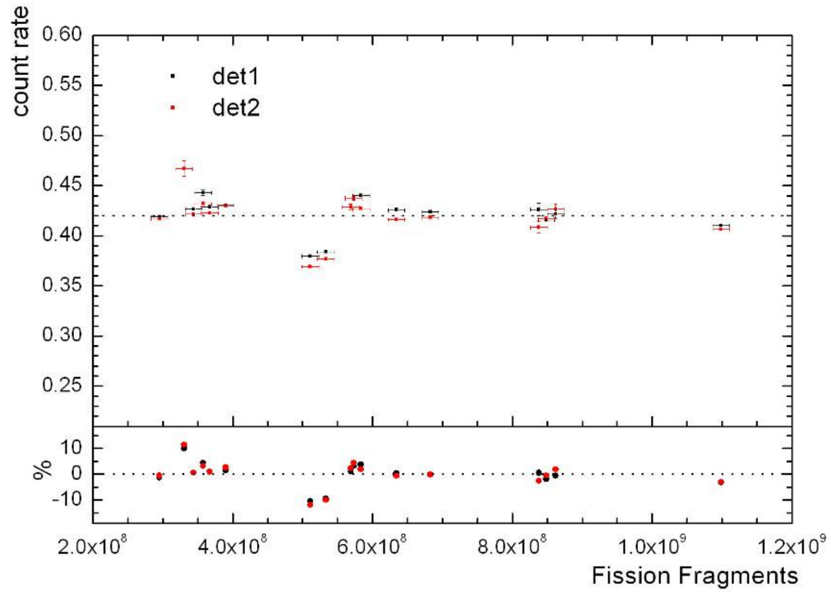






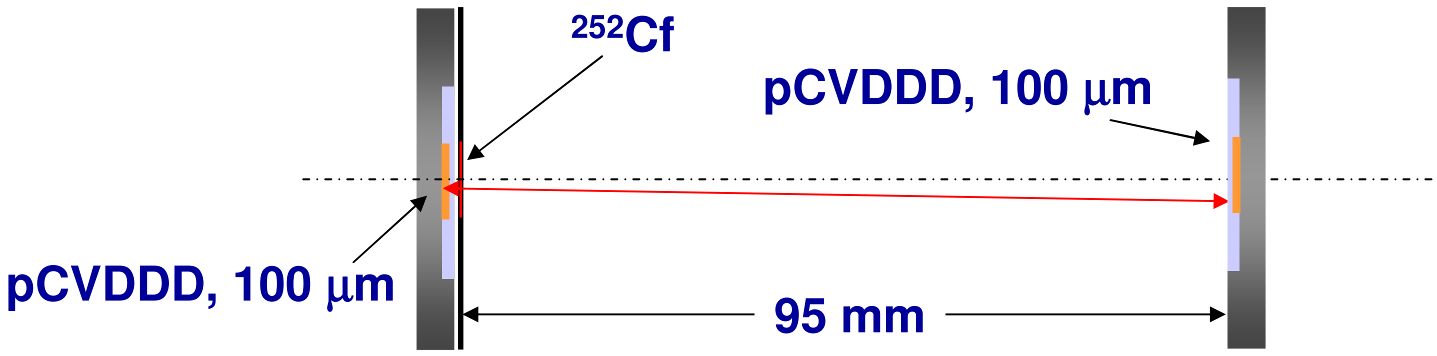
pCVDDD – pulse-height stability

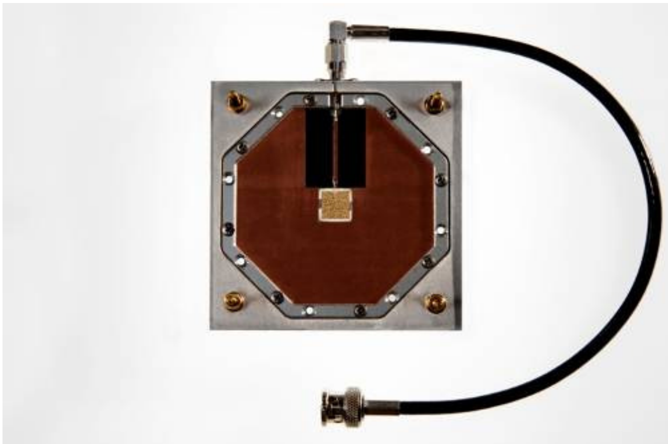




- ✓ **Pulse height stability against radiation damage up to a fission-fragment dose of at least 1.2×10^9**
- ✓ **Including an α-particle dose of 4×10^{10} and a fast neutron dose of about 5×10^9**

pcCVDDD - Intrinsic timing resolution

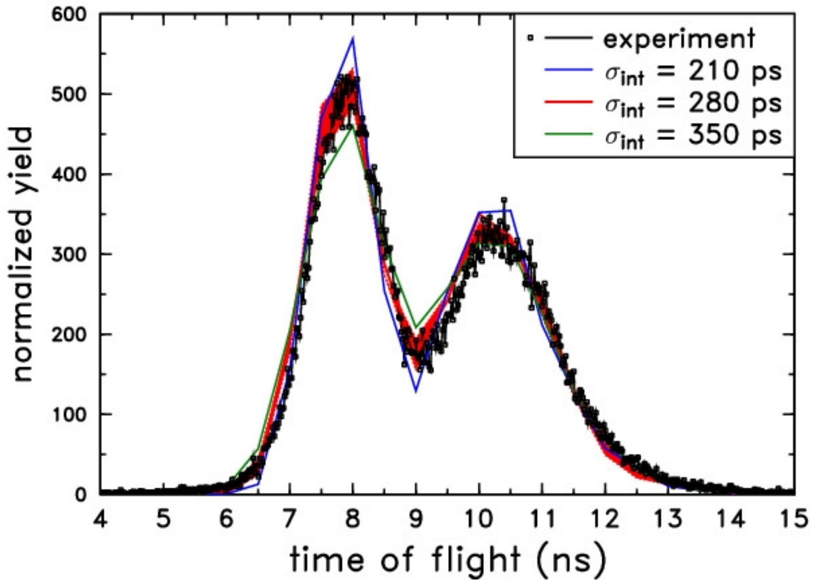


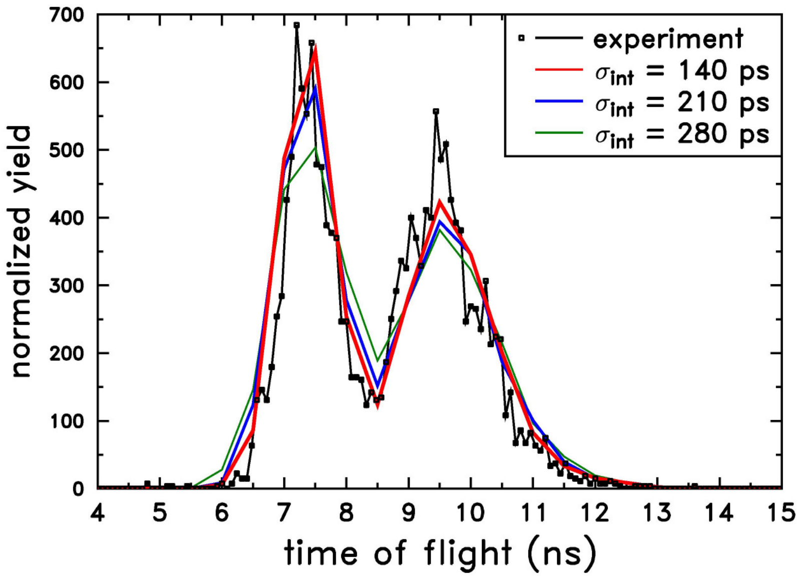


pCVDD material

- ✓ size: $1 \times 1\ cm^2$
- ✓ thickness: 100 μm

pcCVDDD - intrinsic timing resolution



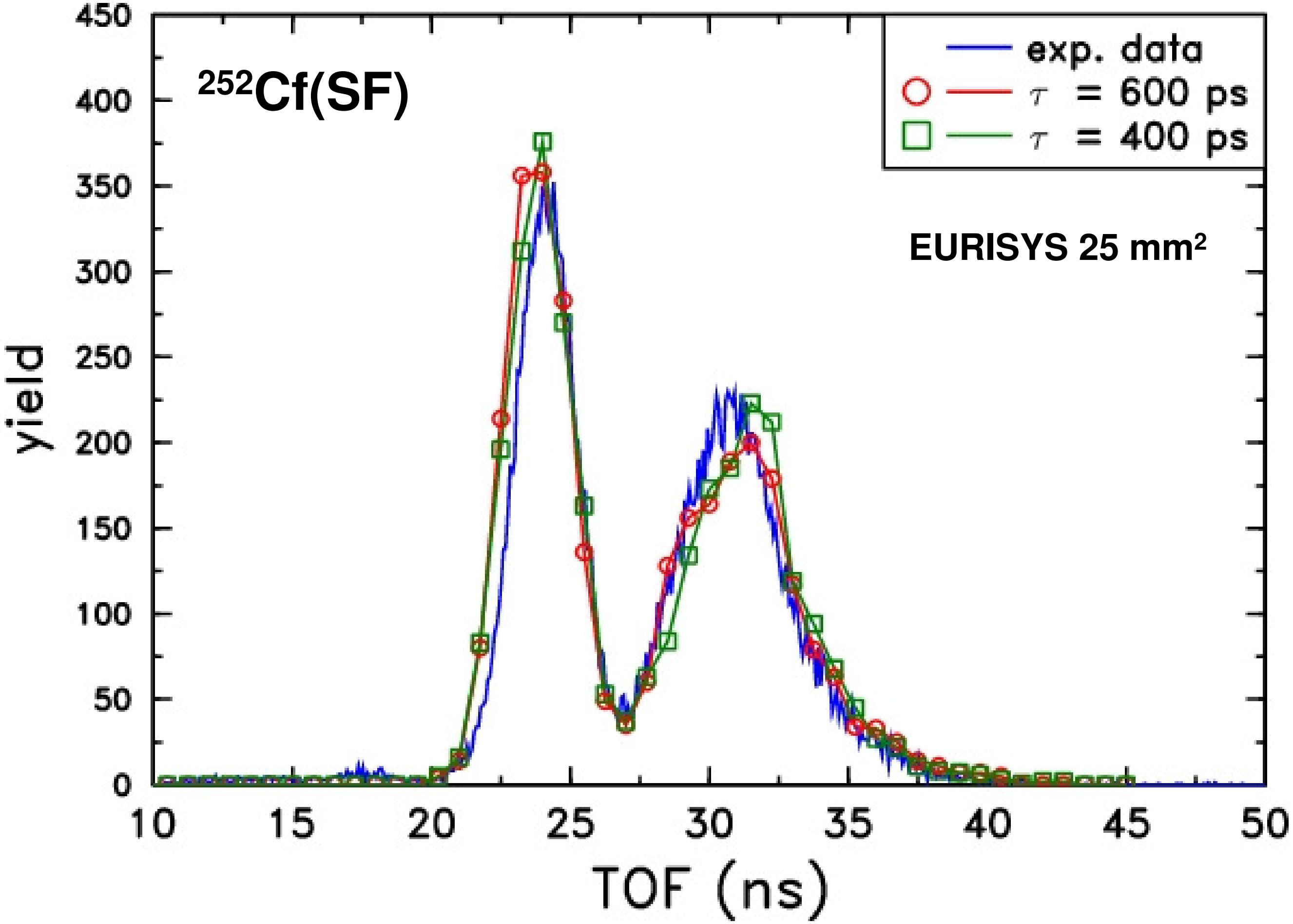


- **TUD pre-amplifier (QS) + TFA Ortec 474: rise-time 6 ns**

- **TUD pre-amplifier: current sensitive, rise-time < 4 ns**
- **Wave-form digitization (Tektronix: 1GHz, 10 GS/s)**

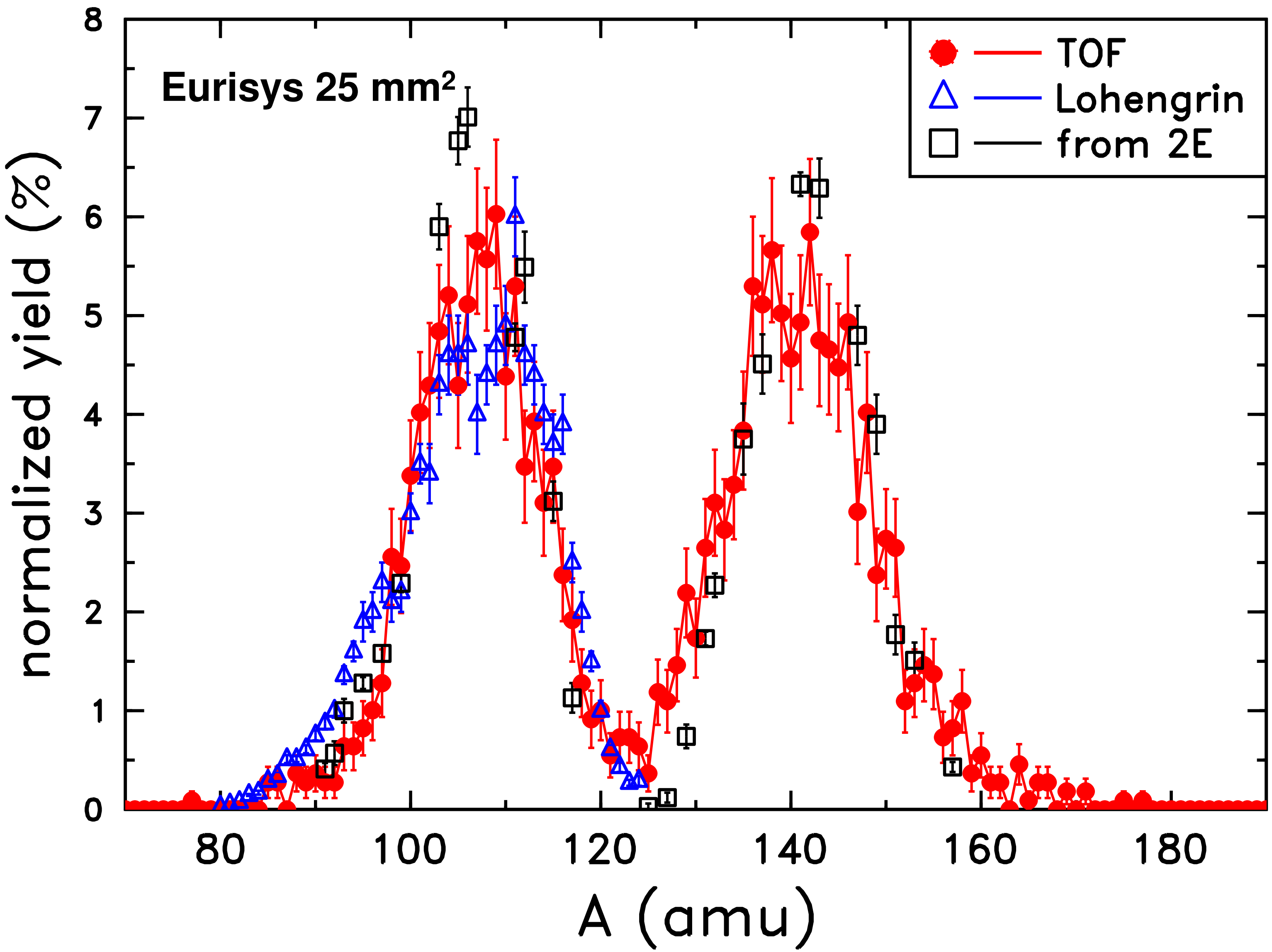
- **Timing resolution very good but worse than expected from HI experiments**
- **Don't forget! Here, we have a largely mixed particle source in A and E_{kin}**

VERDI - the timing resolution



- pCVDDD →TUD pre-amplifier (QS) + TFA Ortec 474: rise-time 6 ns
- PIPS → MSI 8 (SA, TFA + VT120)

VERDI – FF distributions



Improved electronics

- ✓ ORTEC 1GHz / GSI 2.5GHz pre-amplifiers
- ✓ Signal boost with ORTEC VT120 (improved S/N)
- ✓ Timing-optimized energy (stop) detector from CANBERRA with a size of 450 mm^2

- ➢ pCVDDD → Ortec 9306 (1GHz) +
- ➢ VT120: rise-time ≤1.2 ns
- ➢ PIPS → MSI 8 (SA, TFA + VT120)

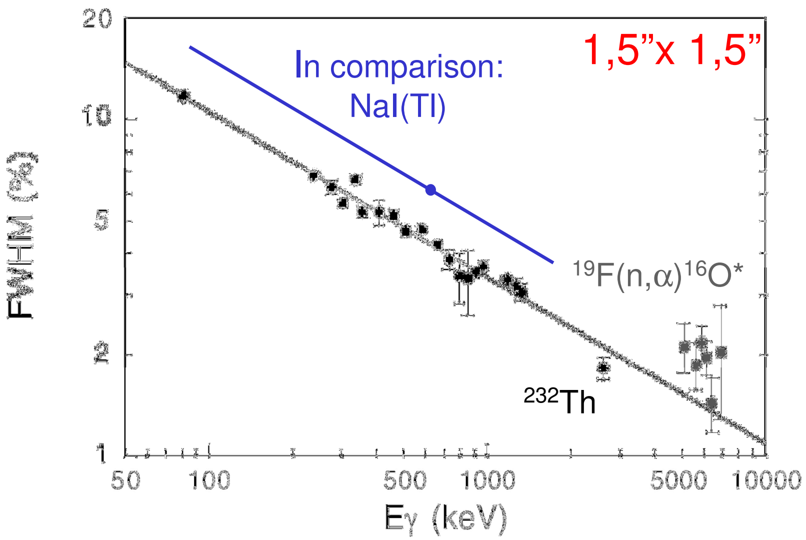
⇨ **VERDI** timing resolution certainly better than 400 ps

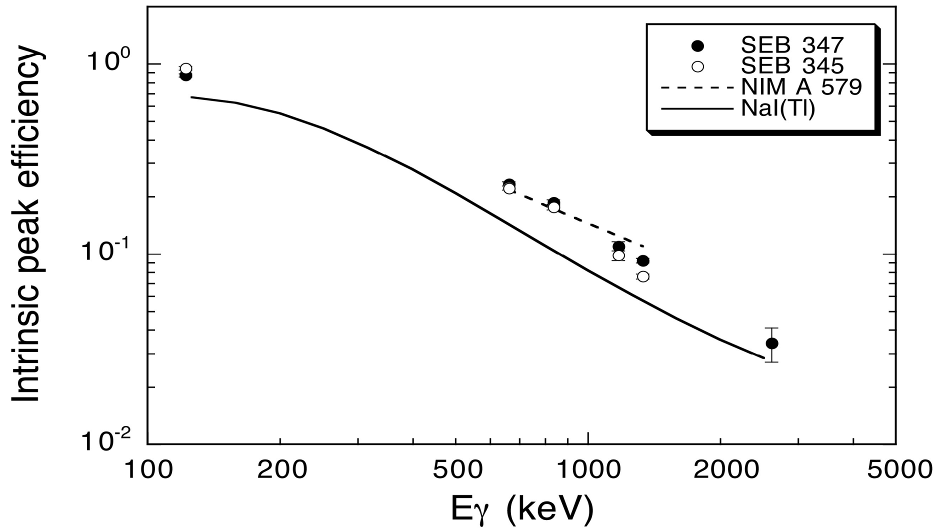
Prompt γ-ray emission in fission

- High intrinsic peak efficiency
- (high) energy resolution
- Excellent time-resolution to efficiently suppress the prompt neutron component

⇨ Lanthanum-halide crystal scintillation detectors

$LaCl_3$:Ce – characterization ♥





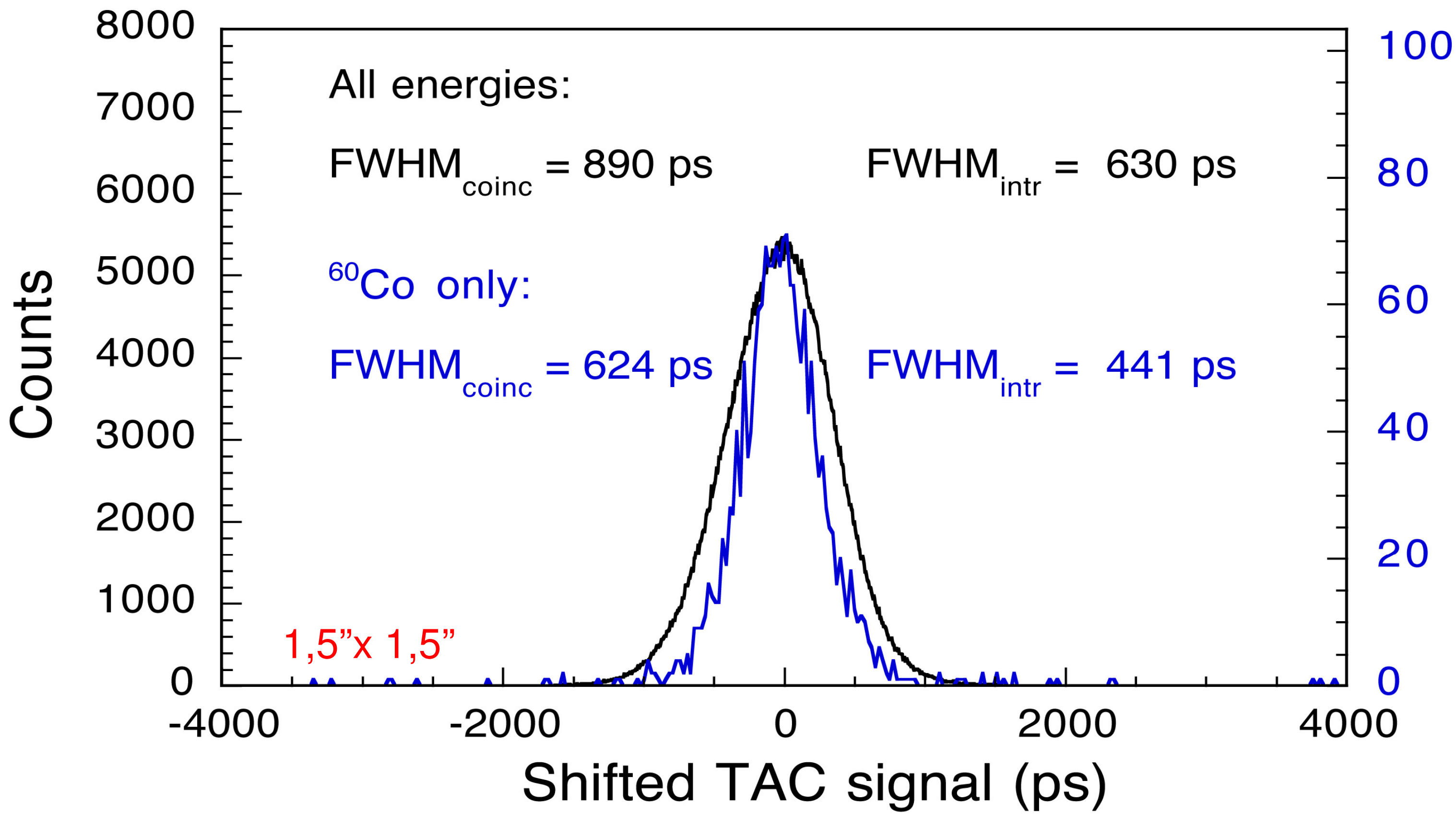
expected $E^{-1/2}$ behaviour
$\Delta E/E$ = 3.8 - 4.2 % (FWHM) at 662 keV (^{137}Cs)

in agreement with other $LaCl_3$ detectors (interpolated)

53 % better than NaI(Tl) detectors of same size

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

$LaCl_3$:Ce – characterization



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Test experiment at CEA/DAM Ile de France ♥

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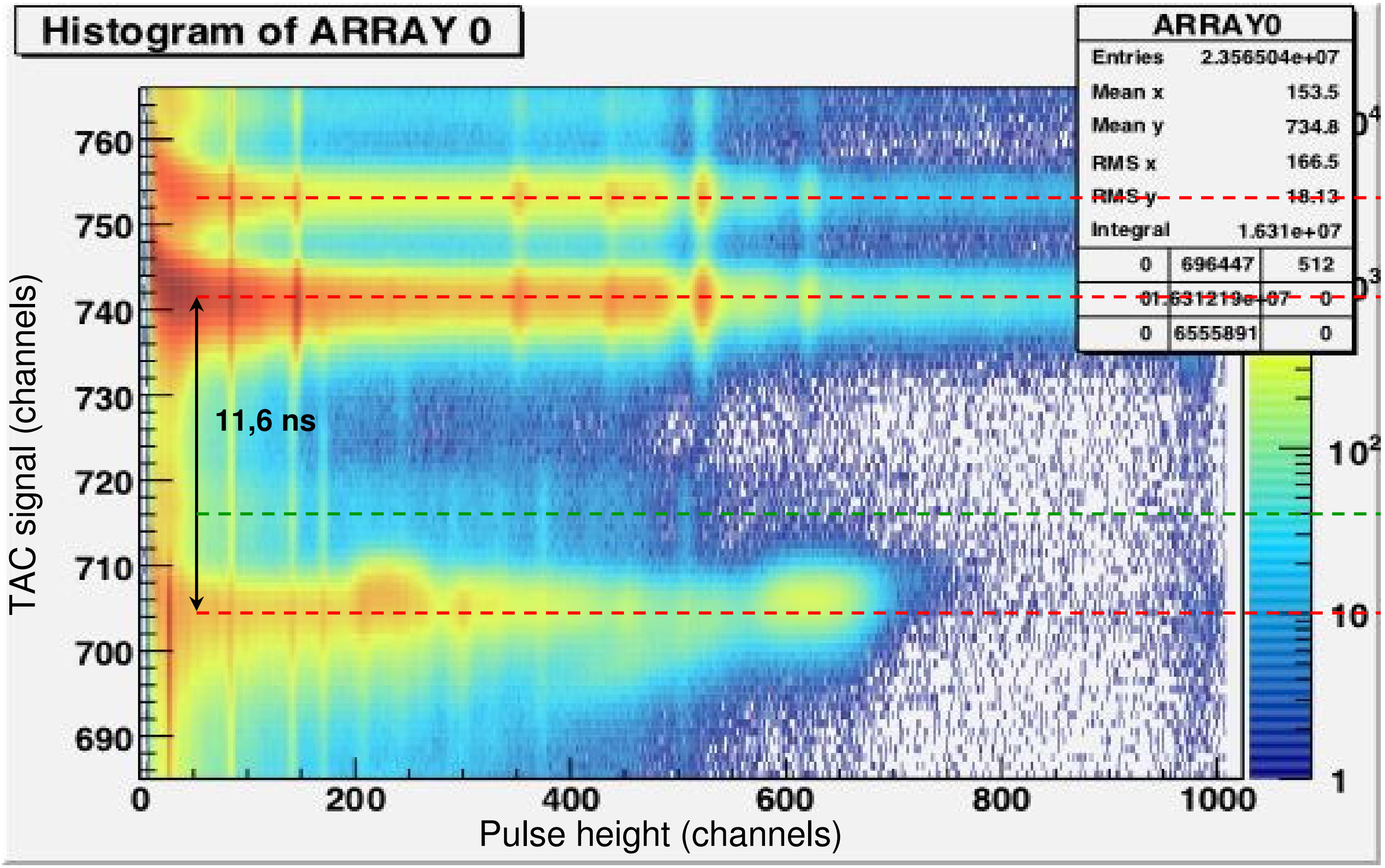
Beam: E_n = 5 MeV from the reaction D(d,n)^{3}He (1 mg/cm^2 TiD) with E_d = 1.86 MeV $I_d \approx$ 1.8 μA

Pulse: τ = 3 ns and Δt = 400 ns



Target: ^{209}Bi

γ's from scattering off beam line

prompt γ-flash from n-production

γ's from target

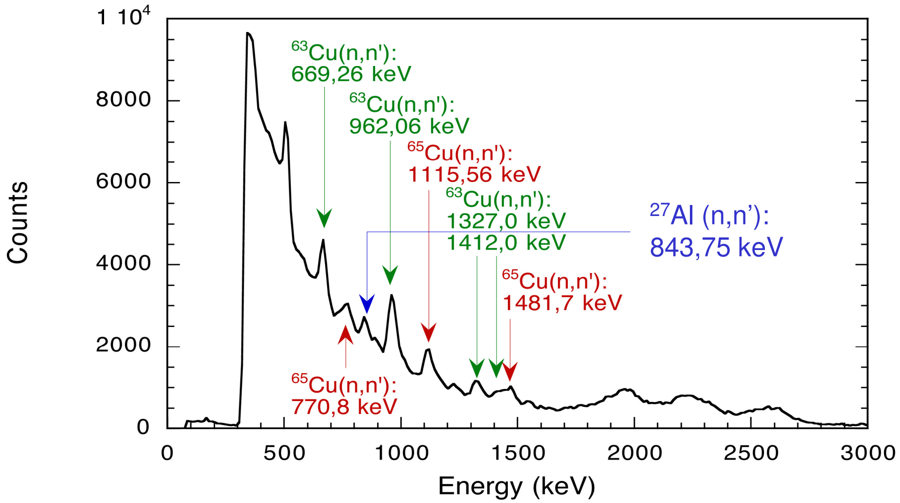
5 MeV neutrons on detector and γ's from (n,n'), (n,α), (n, γ)

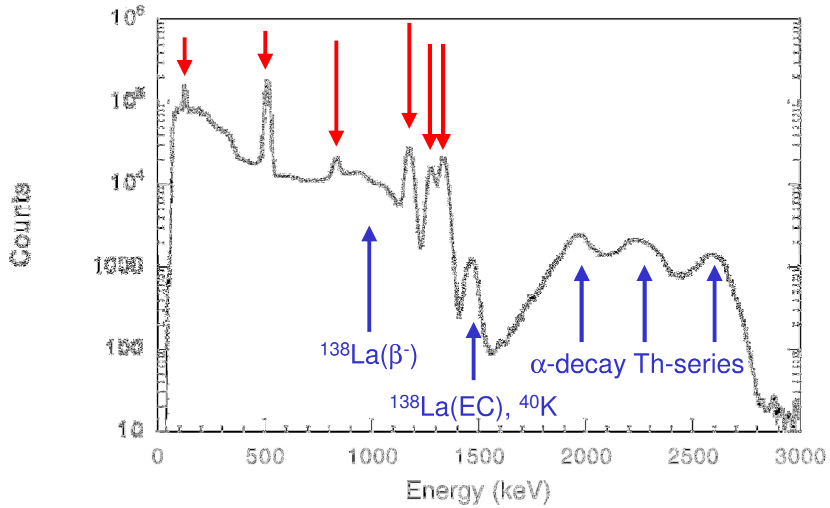
♥ This work was supported by the EFNUDAT programme of the European Commission (agreement number 31027)

Test experiment at CEA/DAM Ile de France

calibration spectrum after irradiation,

intrinsic activity, but no further γ-lines, i. e. no activation





prompt γ-rays from shielding material (^{63,65}Cu) … and detector caps (^{27}Al)

Energy	Source
122 keV	^{57}Co
511 keV	annihilation
835 keV	^{54}Mn
1173 keV	^{60}Co
1275 keV	^{22}Na
1332 keV	^{60}Co

The experiment(s) @ KFKI ♥

✓ Single (ν, E) experiment: $^{235}U(n_{th}, f) \Rightarrow Y(A, E_k)$

- with a 1×1 cm^2 pcCVDDD
- ^{235}U (113 μg)
- $\Phi_{n,th} \approx 5 \times 10^7/s/cm^2$
- **$c_{th} > 2$ FF/s or 10^6 FF/(120 h) per detector**
- **10 energy detectors → $\varepsilon = 0.0026$**

✓ Fission γ-ray spectral measurement

- pCVDDD serves as ultra-fast fission event trigger
- Use of 4 lanthanum halide detectors of various size
  - one Lanthanum-chloride (3" × 3", CIEMAT)
  - two Lanthanum-chloride (1.5" × 1.5")
  - one lanthanum bromide (2" × 2")
  - fission γ-ray count rate > 10/s or > 5×10^6/(120 h) per detector (1.5" × 1.5")

✓ *Integral measurement of the prompt neutron spectrum*

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

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The experiment(s) @ KFKI

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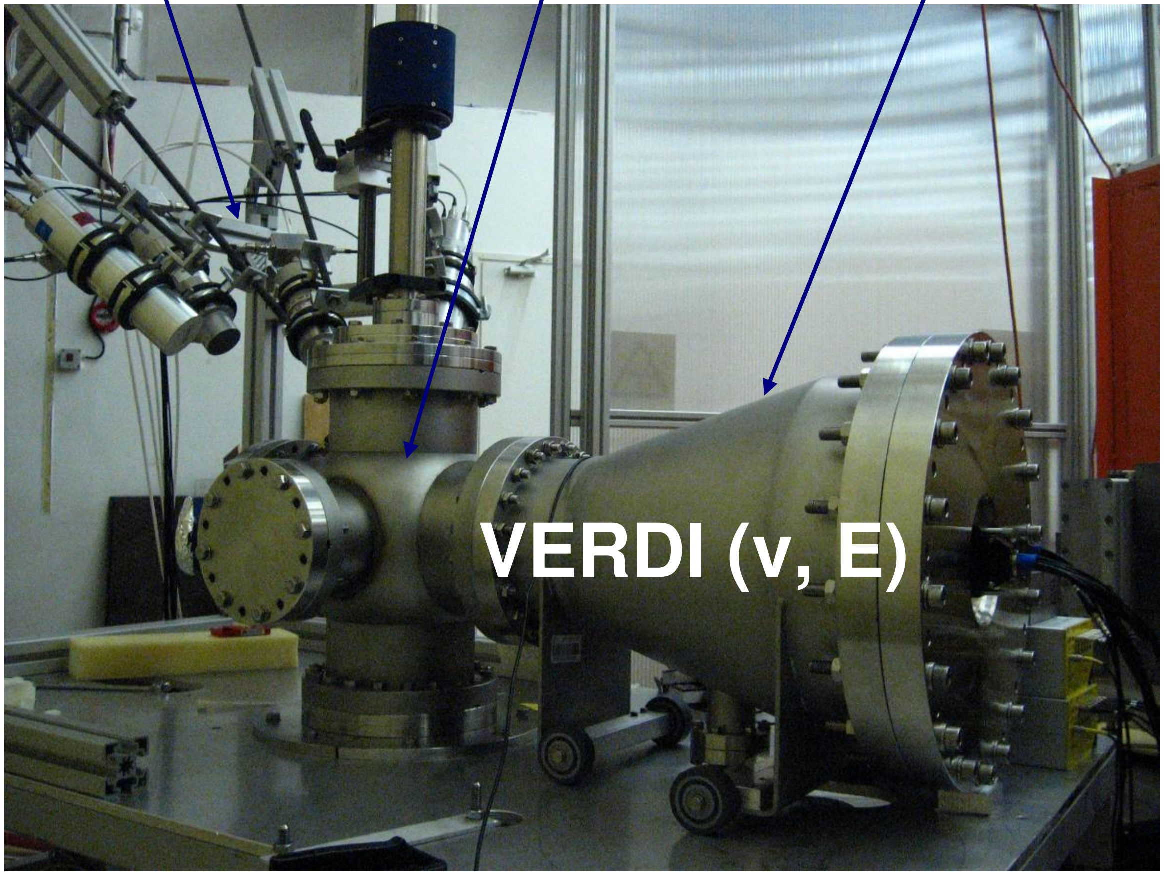
γ-ray detectors

sample holder/diamond

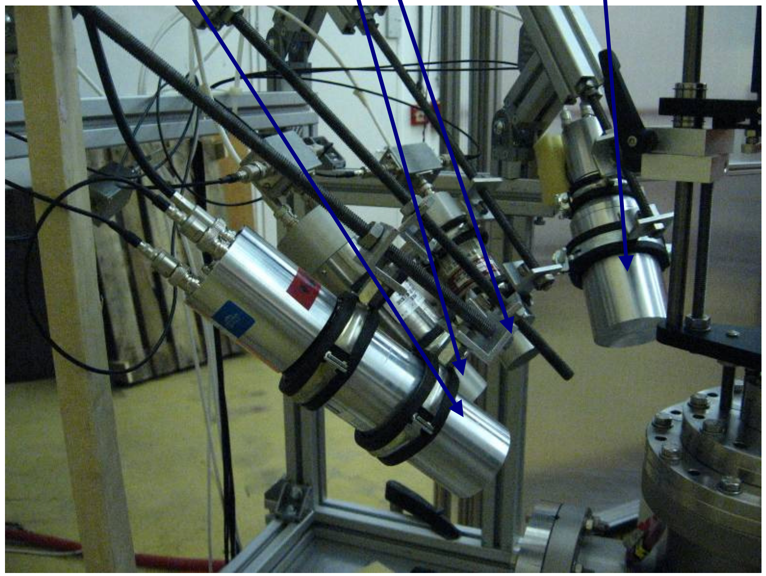
TOF section



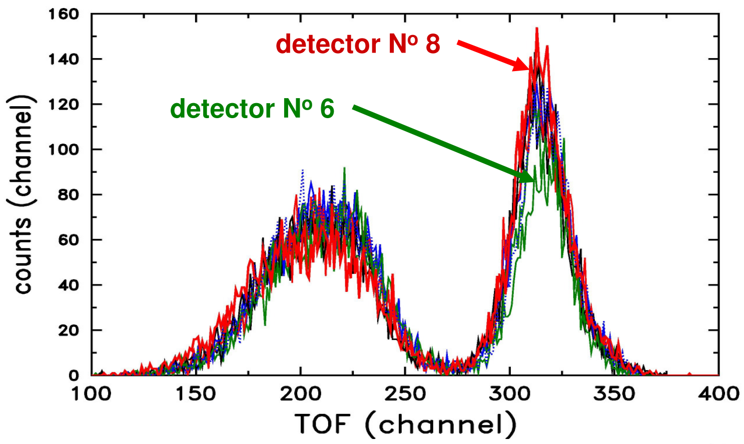
$LaCl_3$ 1.5”× 1.5”

$LaBr_3$ 2”× 2”

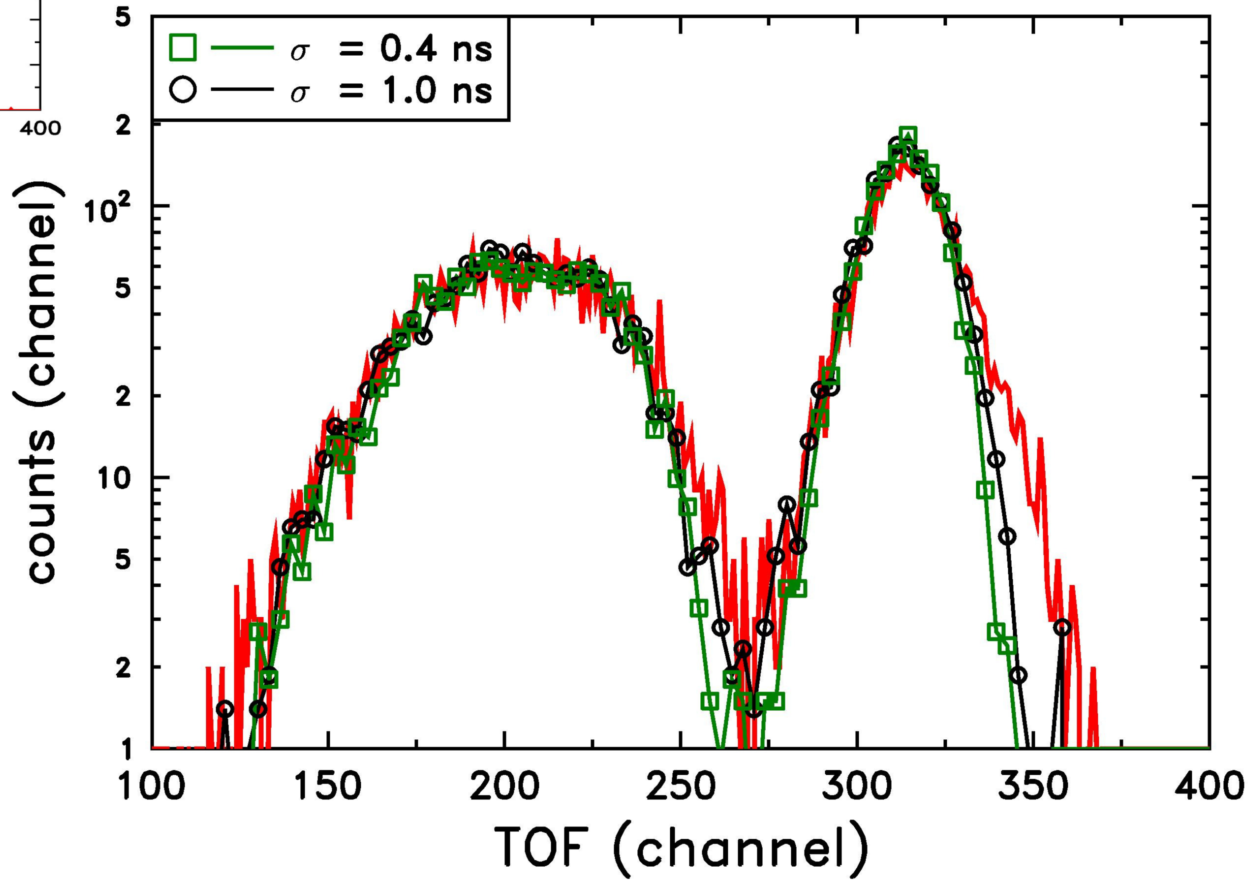
$LaCl_3$ 3”× 3”

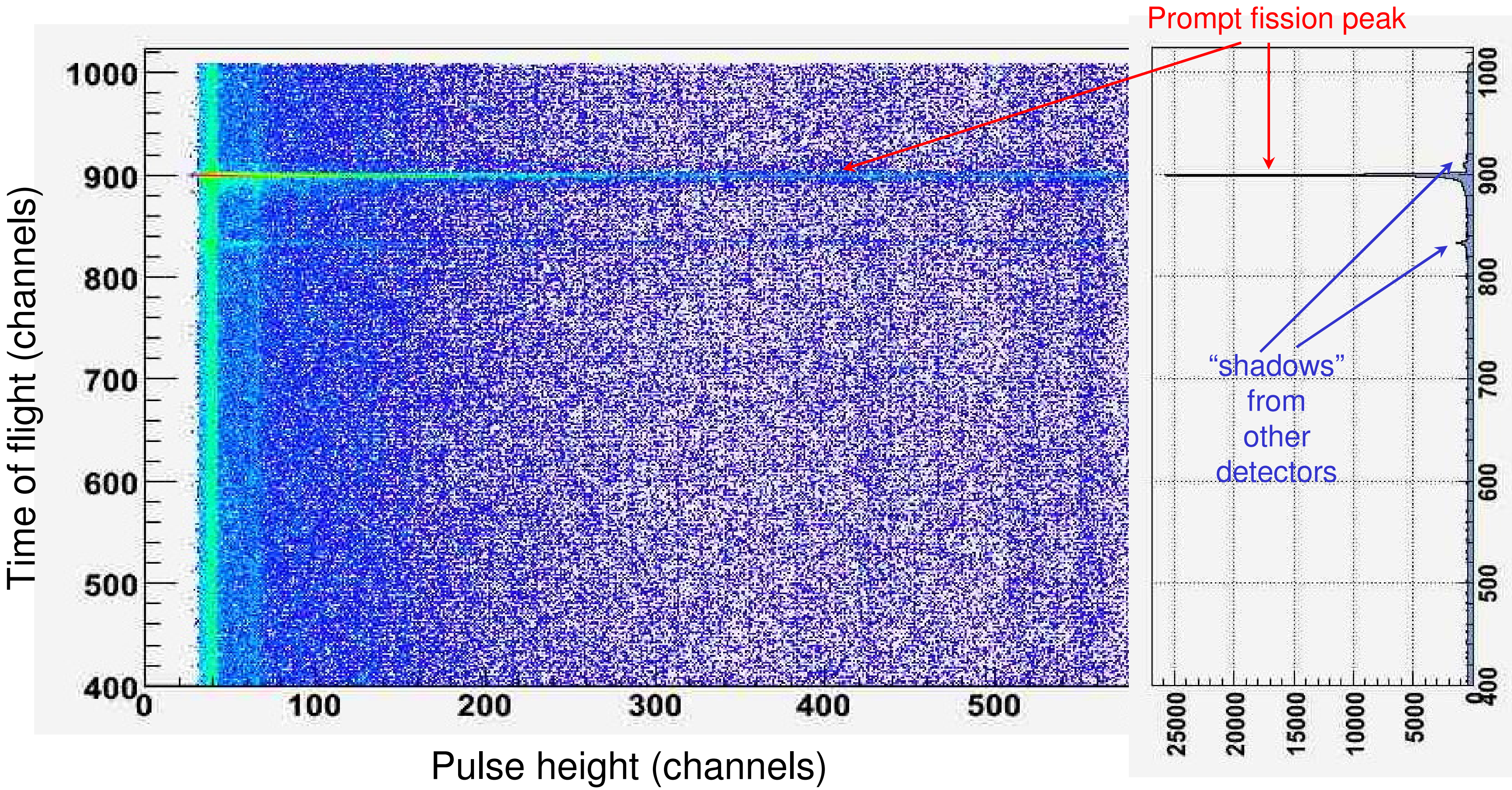


First spectra from a *few h* - run

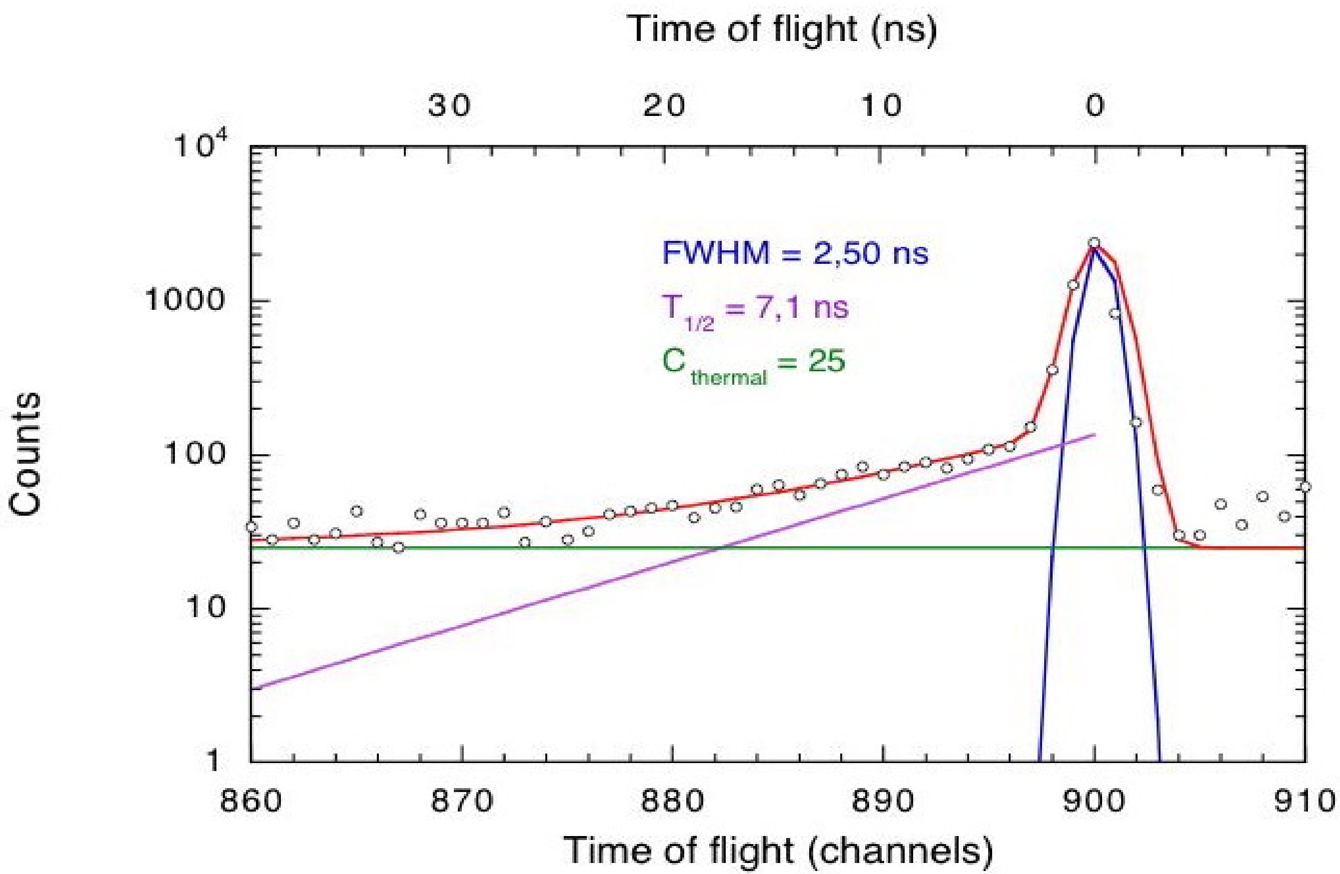


Measurement with 10 PIPS simultaneously

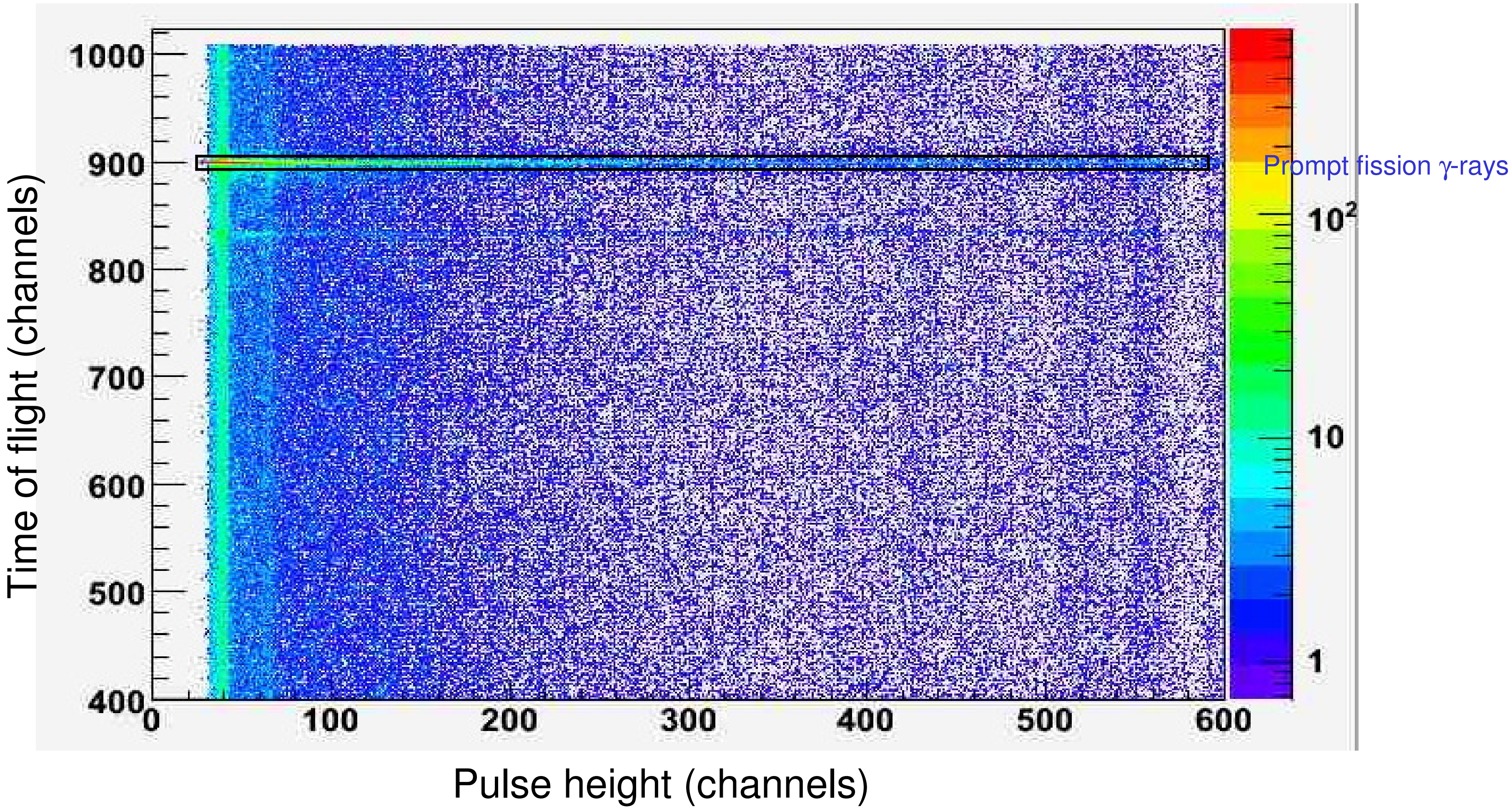


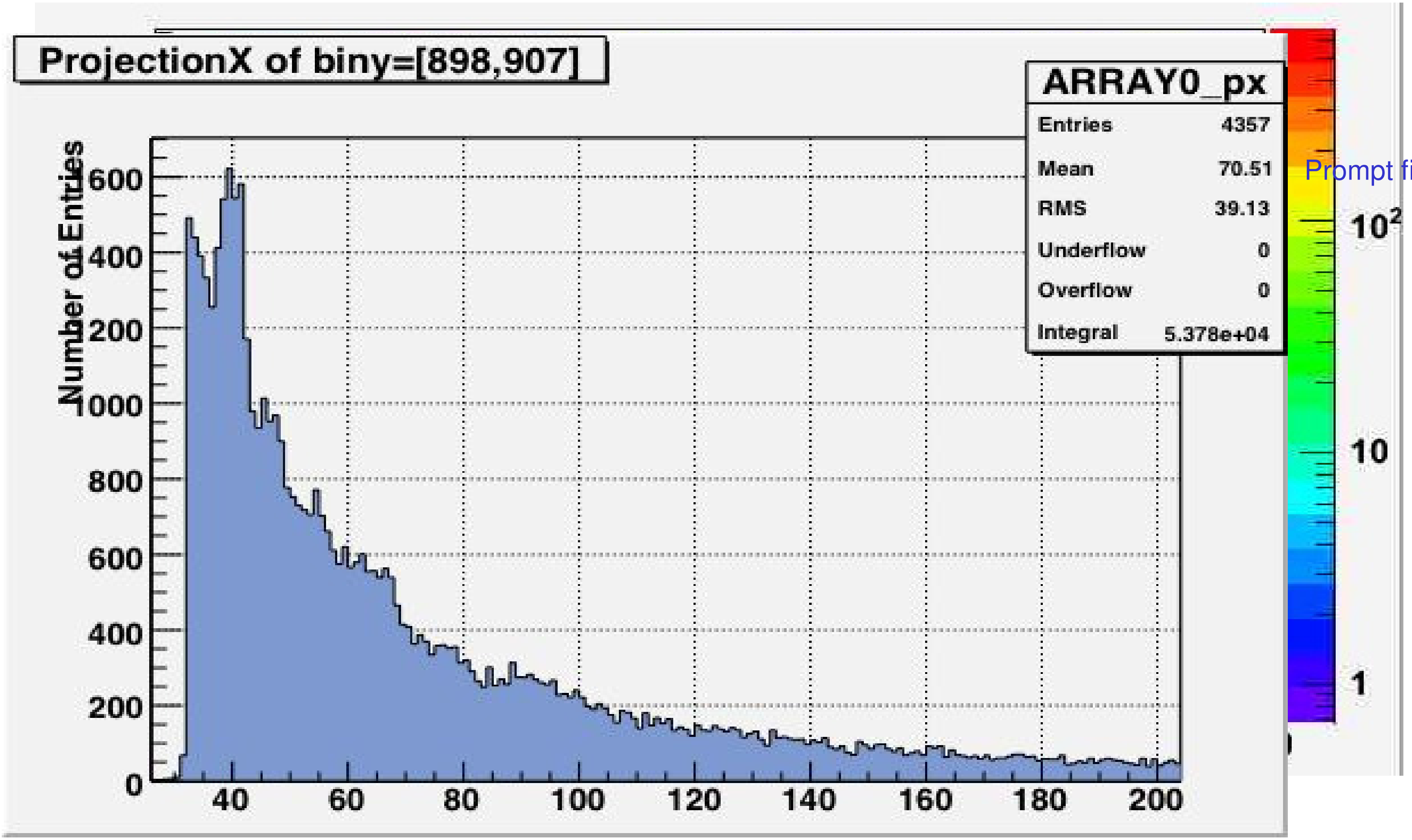


Prompt fission peak
Time of flight (channels)
1000
900
800
700
600
500
400
0
100
200
300
400
500
Pulse height (channels)
“shadows” from other detectors
25000
20000
15000
10000
5000
0

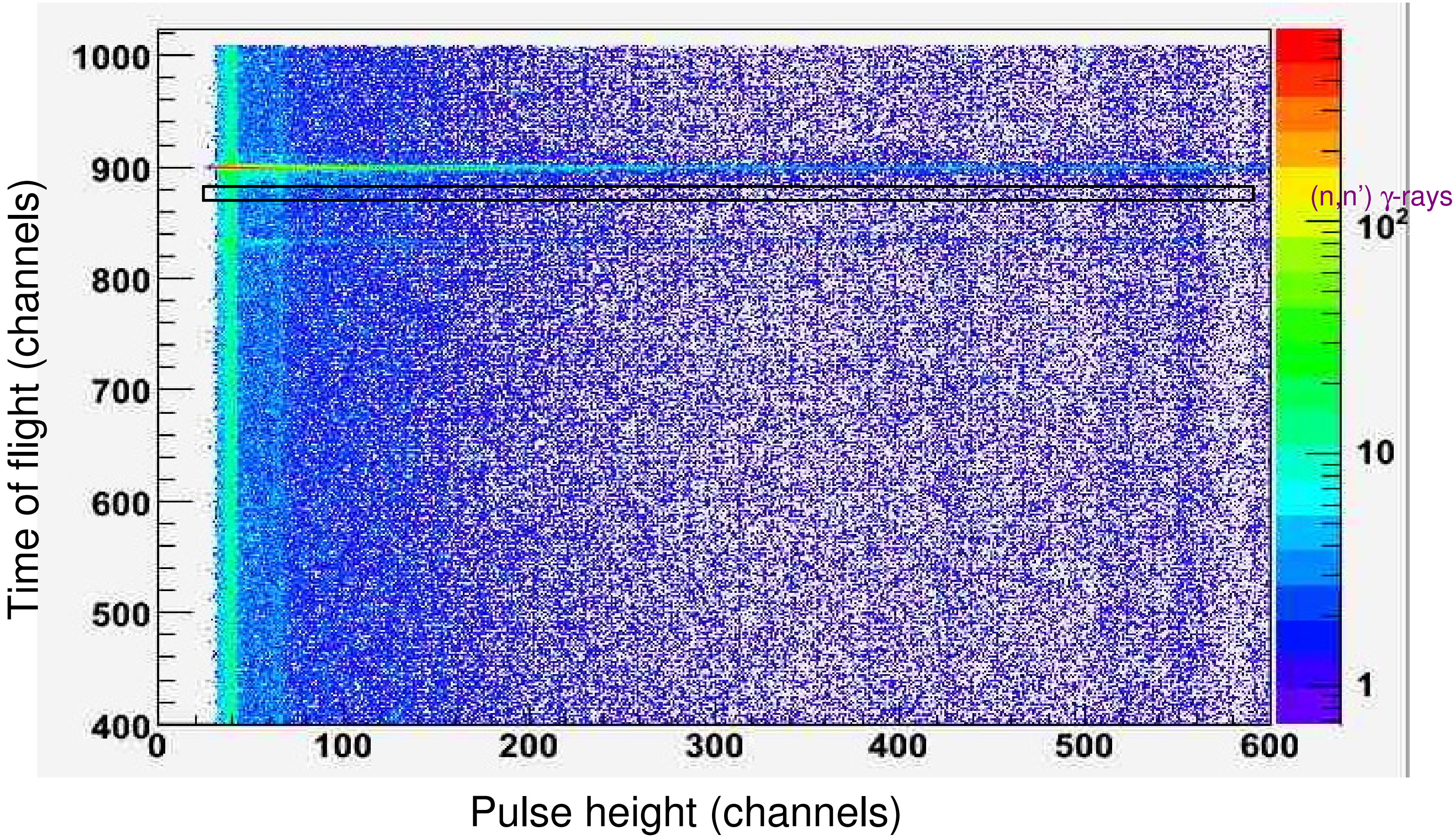


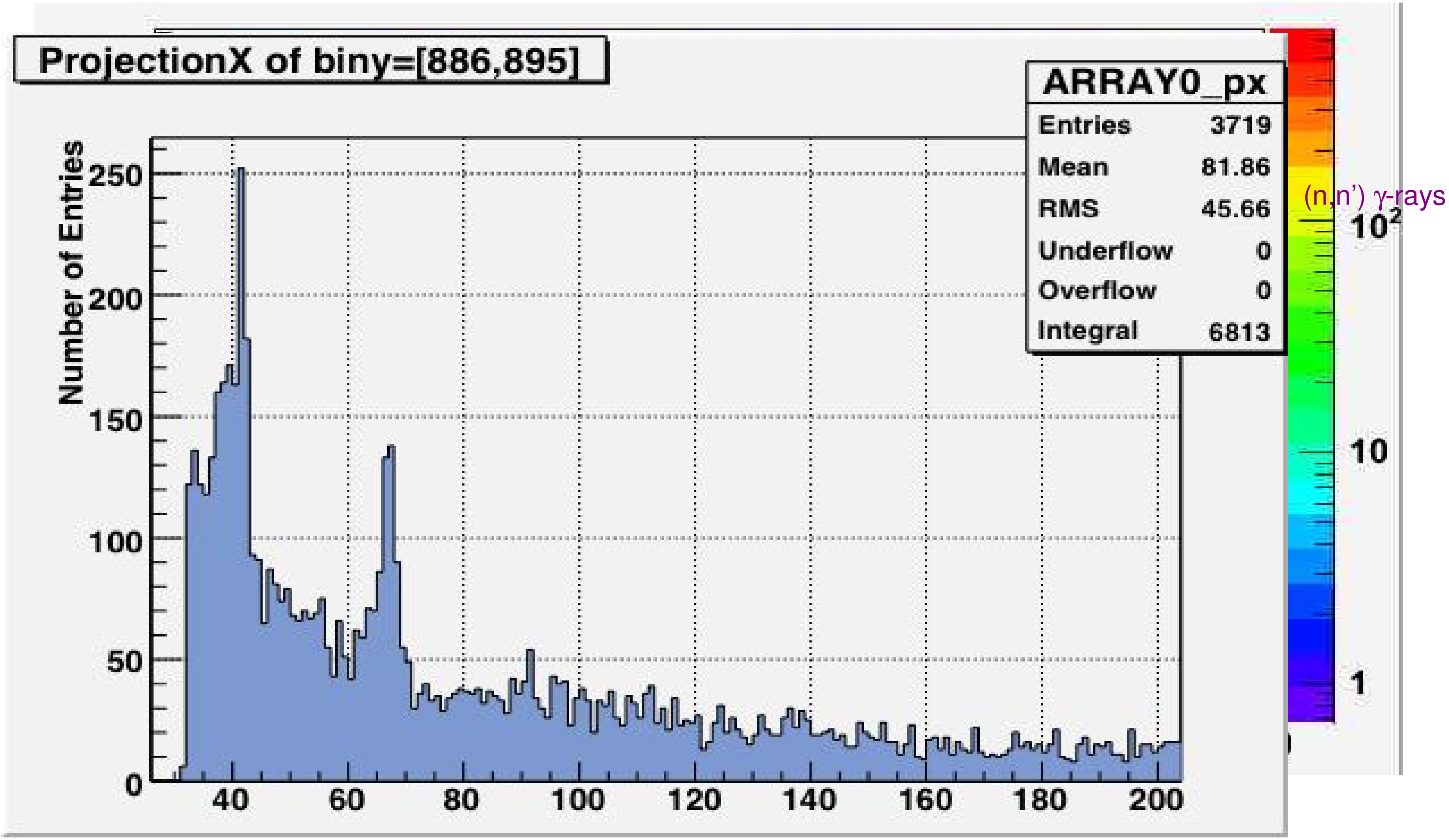
Prompt γ-peak + inelastic prompt neutron scattering + thermal neutron capture

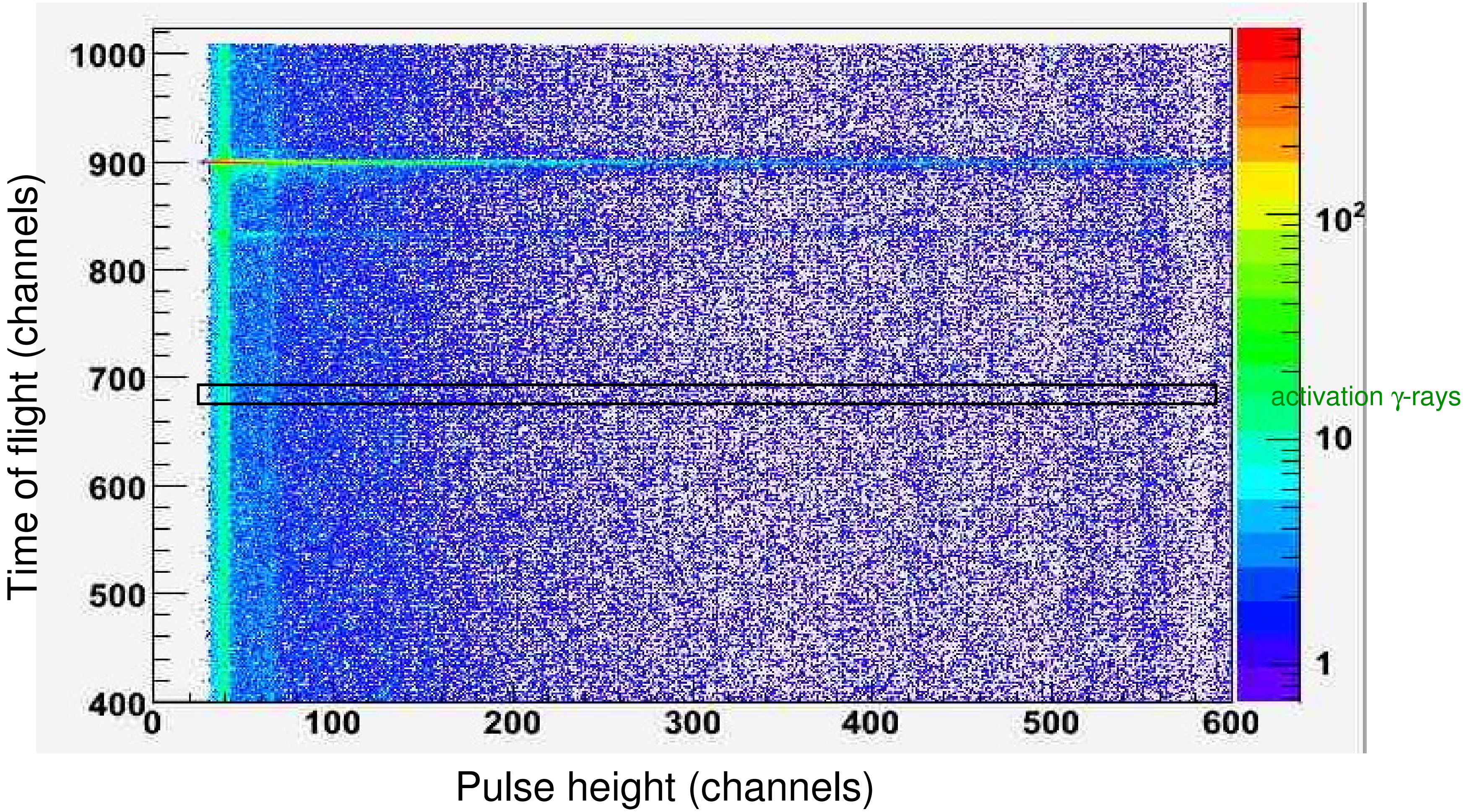

Prompt fission γ-rays
Time of flight (channels)
Pulse height (channels)
1000
900
800
700
600
500
400
0
100
200
300
400
500
600
10²
10
1

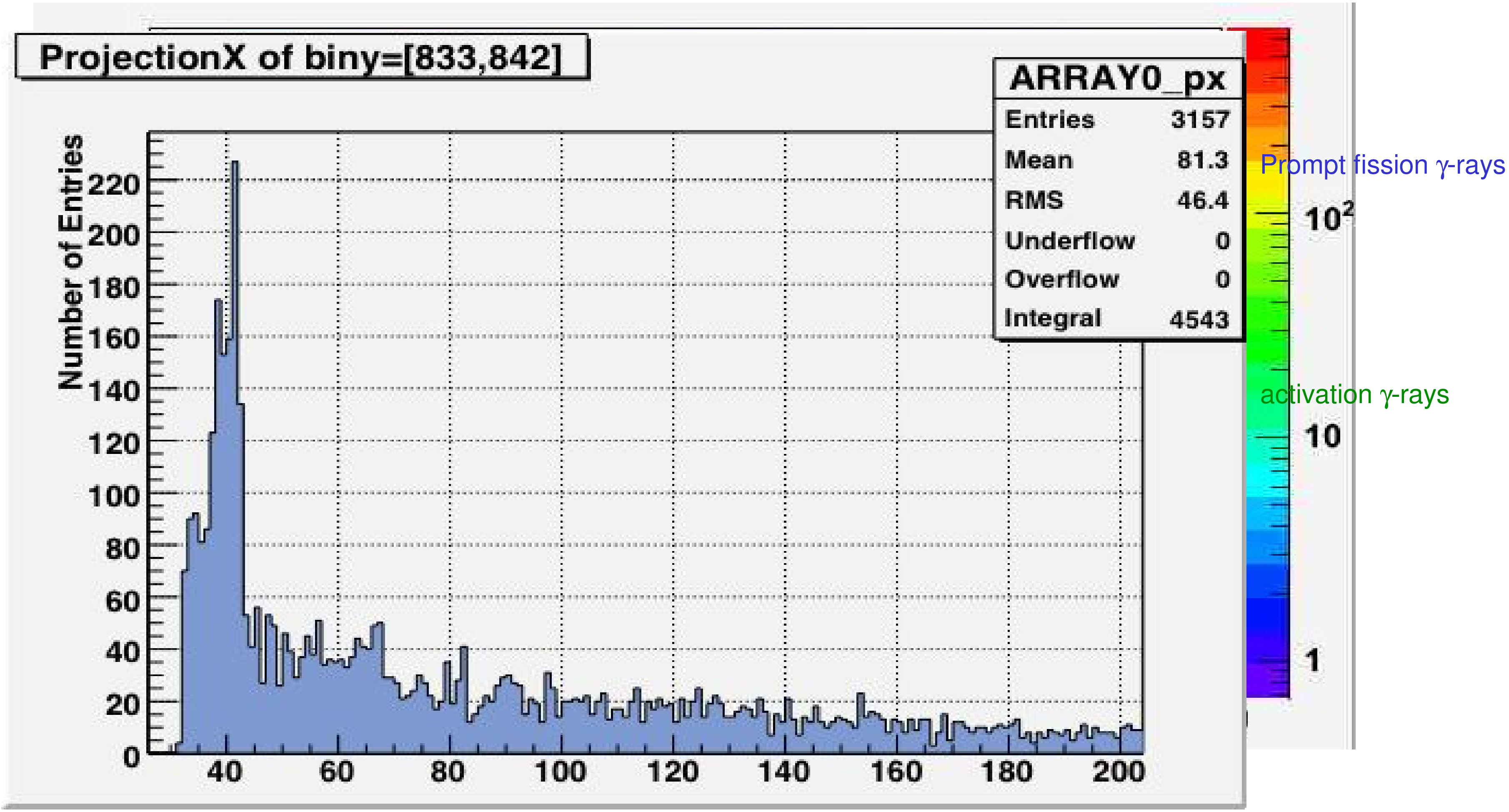


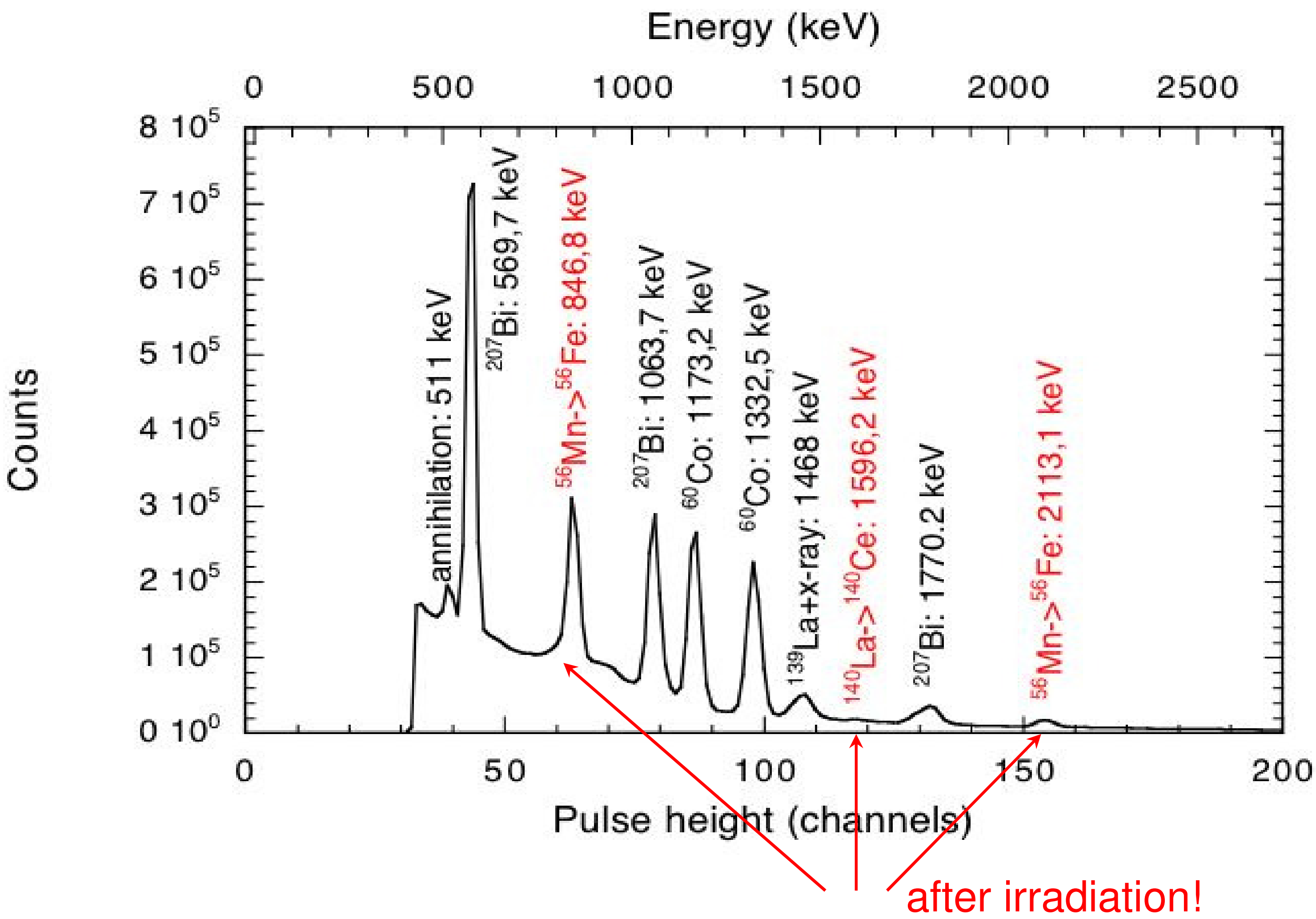
Prompt fission γ-rays


1000
900
800
700
600
500
400
0
100
200
300
400
500
600
10^2
10
1
(n,n') γ-rays
Time of flight (channels)
Pulse height (channels)

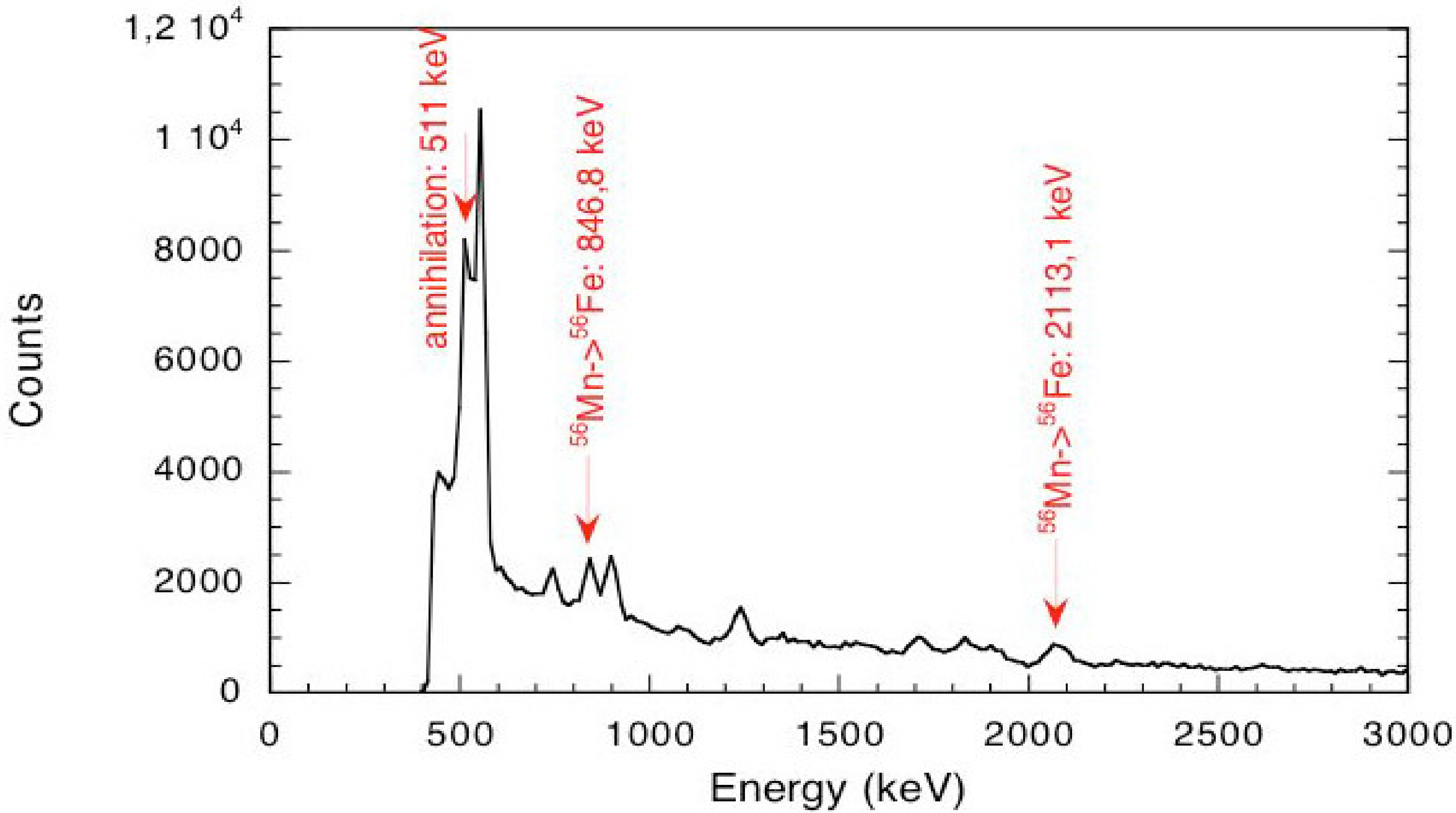

ProjectionX of biny=[886,895]
ARRAY0_px
Entries 3719
Mean 81.86
RMS 45.66
Underflow 0
Overflow 0
Integral 6813
Number of Entries
250
200
150
100
50
0
40
60
80
100
120
140
160
180
200
(n,n') γ-rays
10^2
10
1


1000
900
800
700
600
500
400
0
100
200
300
400
500
600
10^2
10
1
activation γ-rays
Time of flight (channels)
Pulse height (channels)


ProjectionX of biny=[833,842]
ARRAY0_px
Entries 3157
Mean 81.3
RMS 46.4
Underflow 0
Overflow 0
Integral 4543
Number of Entries
220
200
180
160
140
120
100
80
60
40
20
0
40
60
80
100
120
140
160
180
200
10^2
10
1
Prompt fission γ-rays
activation γ-rays

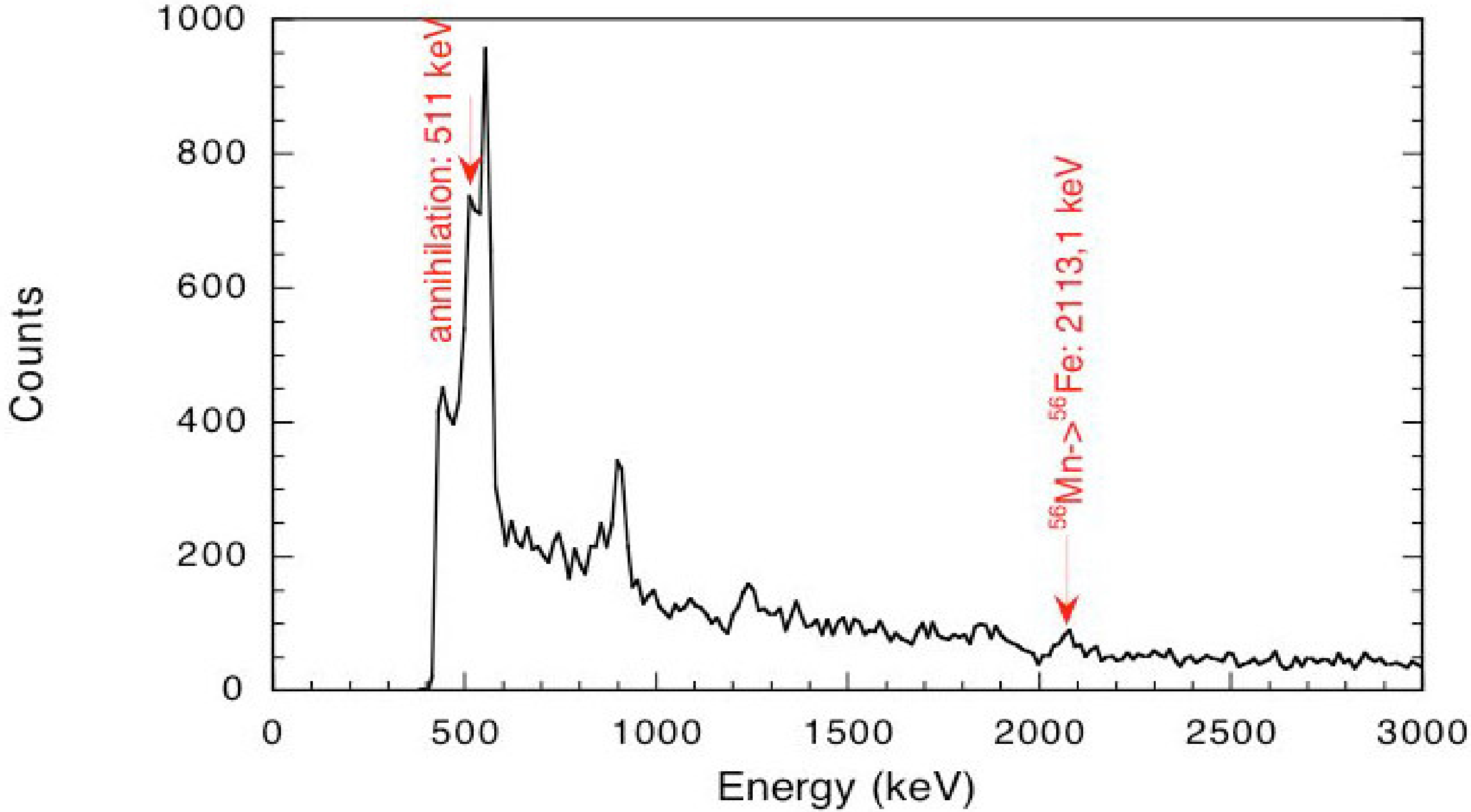

Energy (keV)
0
500
1000
1500
2000
2500
8 10^5
7 10^5
6 10^5
5 10^5
4 10^5
3 10^5
2 10^5
1 10^5
0 10^0
Counts
annihilation: 511 keV
207Bi: 569,7 keV
56Mn->56Fe: 846,8 keV
207Bi: 1063,7 keV
60Co: 1173,2 keV
60Co: 1332,5 keV
139La+x-ray: 1468 keV
140La->140Ce: 1596,2 keV
207Bi: 1770.2 keV
56Mn->56Fe: 2113,1 keV
0
50
100
150
200
Pulse height (channels)
after irradiation!

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



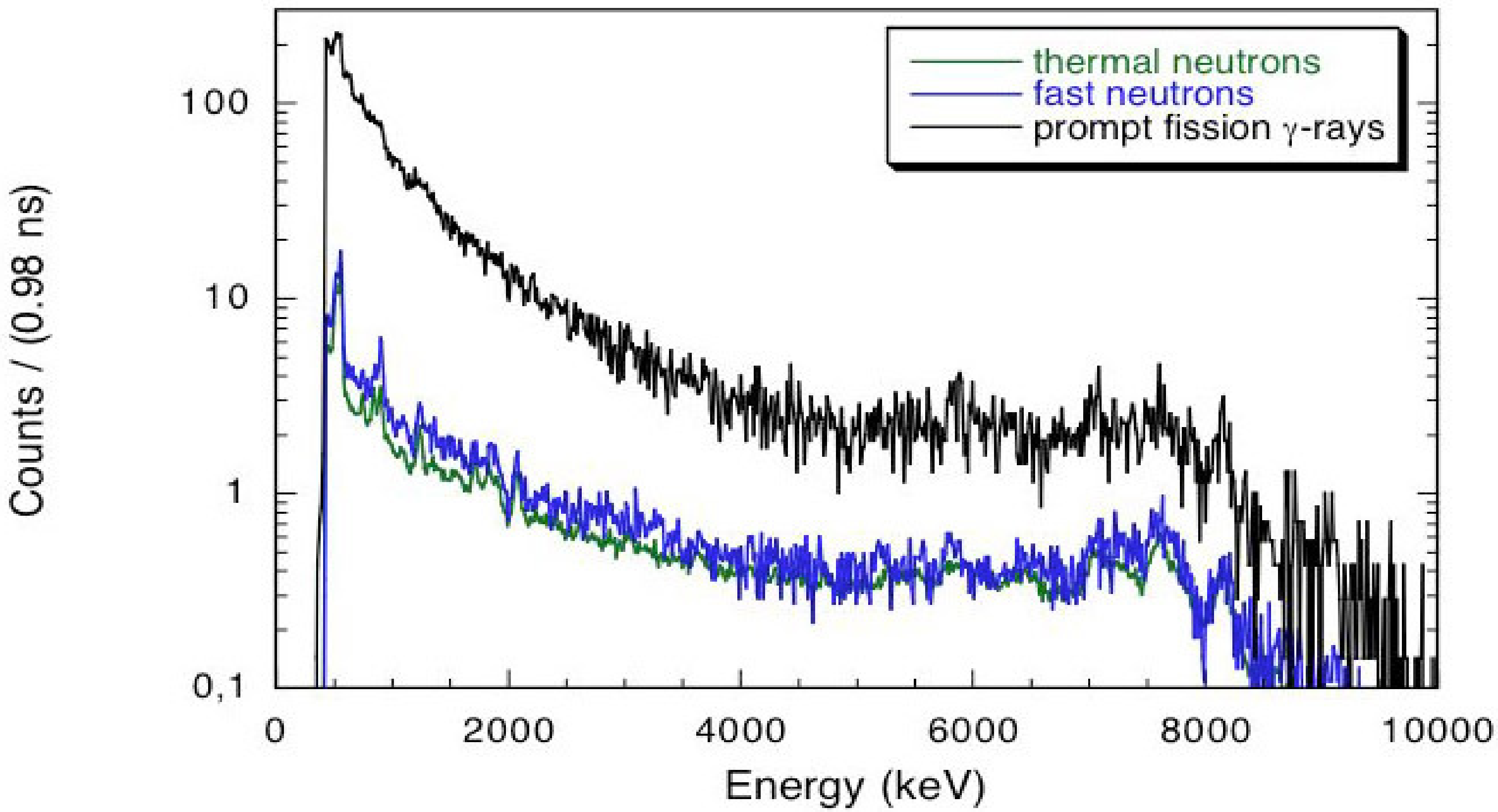
Activation of iron (VERDI): $^{56}Fe(n, p) \rightarrow {}^{56}Mn$ ($T_{1/2}$ = 2.58 h)!

TAC regime of fast neutron induced γ-rays and isomers

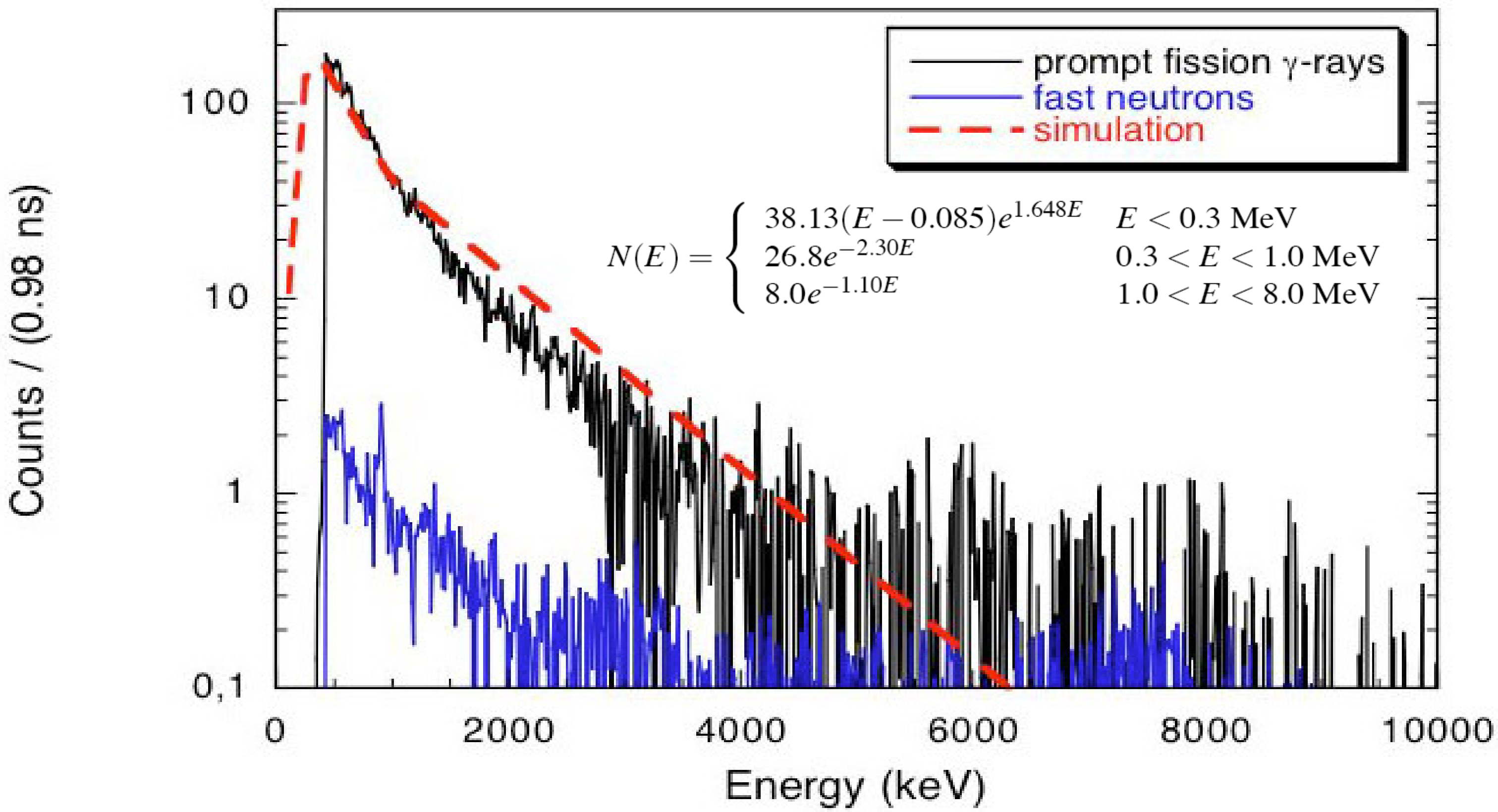


Observe: more peaks have still to be identified!

All TAC regimes: energy spectra - normalized & calibrated



Prompt fission γ-spectrum - background subtracted



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

Conclusions

- ✓ The **VERDI** TOF spectrometer for correlation measurements of fission fragment properties under development is already in a serious testing phase
- ✓ The first TOF section of the **VERDI** spectrometer operational with up to 19 energy detectors
- ⇨ TOF resolution better than or equal to 400 ps ⇨ ΔA = 2 - 3 u
- ⇨ With modern broadband pre-amplifiers and… ΔA = 1 – 2 u is in reach

- ✓ Artificial diamond detectors provide an ultra-fast fission trigger (τ < 280 ps)
- ✓ With broadband pre amplifiers and/or waveform digitization (τ < 180 ps)
- ✓ Radiation hard for characteristic neutron and fission-fragment doses

- ✓ **$LaCl_3$ scintillation detectors** do indeed fulfill requirements for the measurement of prompt fission γ-rays, in particular in conjunction with pcCVD diamond detectors
- ✓ Good energy resolution allowing for online calibration
- ✓ γ-rays distinguished with respect to their origin (fission, neutron-induced, β-delayed …)
- ⇨ Preliminary prompt fission γ-spectrum presented

“correlated” fission count rates

OUTLOOK

E_n	Φ_n	σ_f	c_{ff}	$c_{\gamma p}$
	($/cm^2/s$)	(b)	(/s)	(/s/det)
thermal	5×10^7	583	7.5×10^4	35
	5×10^{10}		7.5×10^7	3.5×10^4
25 keV	10^7	2	50	0.035
	10^8		500	0.35
14 MeV	10^6/MeV	1	< 5/MeV	< 1 /MeV *
BS@ 8MeV	5/s/50μA			18*

spectroscopic sample mass = 1 mg
* Unit = 1/h
efficiency (abs) = 10^{-4}

To be multiplied by fission detector efficiency = 0.5 – 60%

Spectral measurements

OUTLOOK

E_n	Φ_n	σ_f	C_{ff}	$c_{\gamma p}$
	$(/cm^2/s)$	(b)	(/s)	(/s/det)
25 keV	10^6	2	5×10^3	35
	10^7		5×10^4	350
14 MeV	10^6/MeV	1	5×10^3/MeV	350/MeV
BS@ 8MeV	5×10^3 /s/50μA			0.5

spectroscopic sample mass = 1 g
efficiency (abs) = 10^{-4}

Nothing without a good team!

MONNET technical team:
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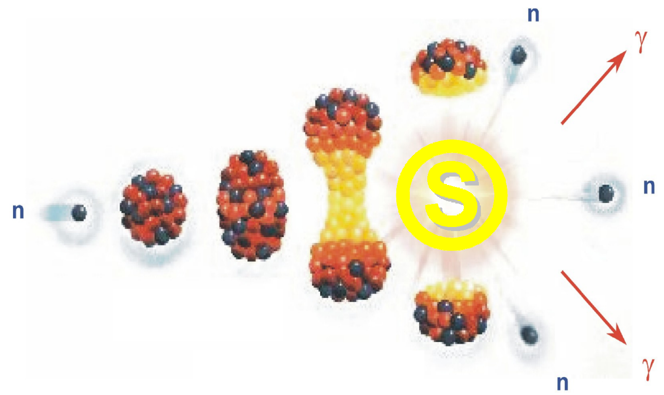

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