Gamma-Ray Output Spectra from ^{239}Pu Fission

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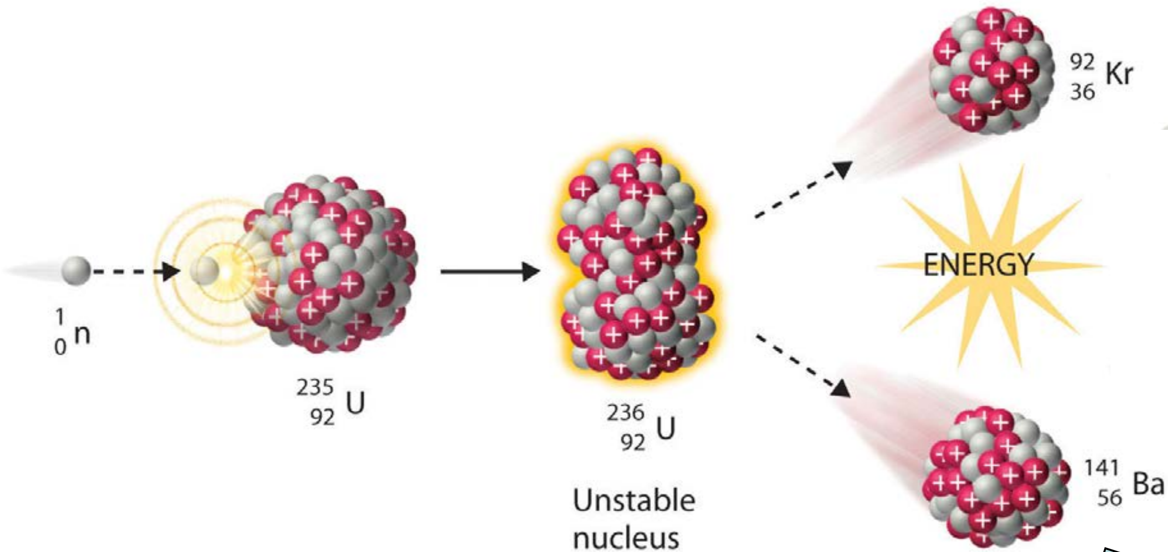
Why study fission gammas?

- Applications – need to know distribution of gamma-ray multiplicity, gamma energy, total gamma energy
  - Reactors: heating, decay heat
  - Non proliferation
  - Illicit nuclear materials (portal monitors)
- Physics of fission
  - Fission products – high-spin, neutron-rich
  - Decay properties constrain models

Fission Physics

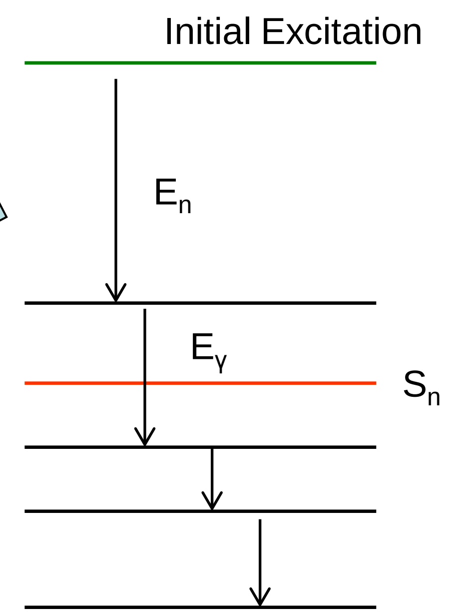


$^{239}Pu(n,f)$

$\nu_n = 2.9$

$\nu_\gamma = 7.2$

(Thermal)



Initialfragmrnt – high spin, excitation
Neutron decay – removes excitation
Gamma decay – (E1) removes spin and energy
Gammas – from fragment decay
- neutron rich (? Structure)
- Fragment mass distribution changes with neutron energy

<Eγ> Changes with neutron energy

- Fragment mass distribution changes with neutron energy
- Madland formula reflects changing products and J (excitation)
- $E_{tot} = 6.741 + 0.117\ T_n(\text{MeV}) - 0.0002\ T_n^2$ MeV
- Low energies resonances – no significant change in gamma properties (Thermal and 100 keV should have similar $\langle E_\gamma \rangle$)

D.G. Madland, Nucl. Phys. **A772**,113 (2006)

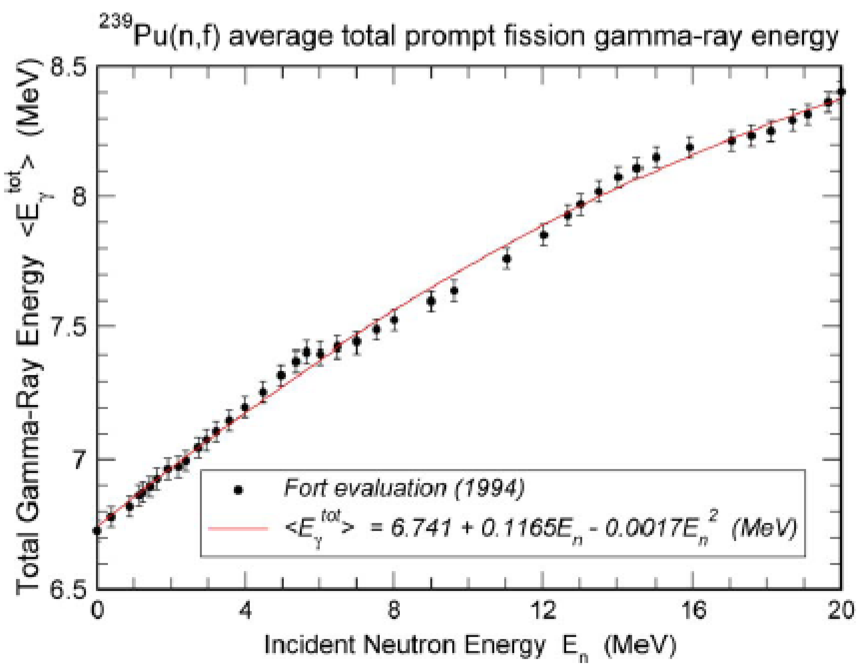
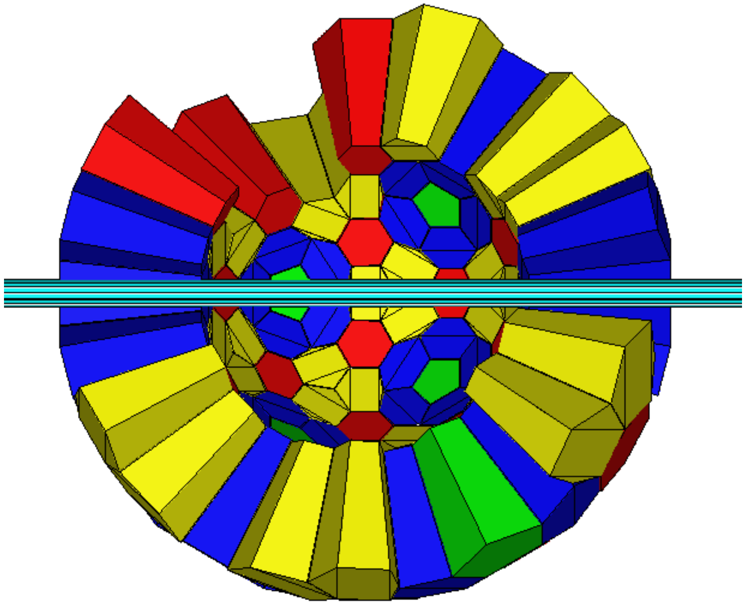


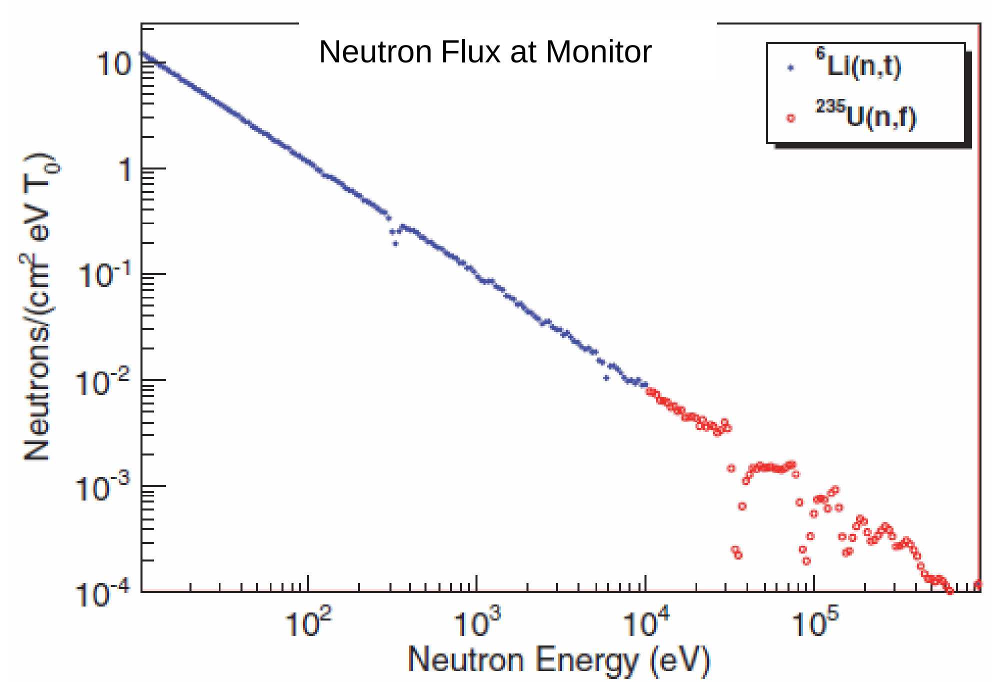
Fig. 16. Average total prompt fission gamma-ray energy $\langle E_\gamma^{tot} \rangle$ for the $n(E_n) + {}^{239}$Pu system.

Gamma-ray output from ^{239}Pu(n,fission)

- Prompt gamma ray emission from fission not well studied
  - Only 1 published spectrum for ^{239}Pu(n,f) – at thermal
  - (V.V. Verbinski, Phys. Rev. C 7, 1173 (1973))
  - Other measurements – but do cover wide gamma energy range
- Experiment at LANSCE moderated white neutron source
  - Need fission tagging – use LLNL/LANL PPAC
  - Gammas detected using Detector for Advanced Neutron Capture Experiments (DANCE) "4π" detector
  - Gammas ± 20 ns from fission event
  - Direct measurement of multiplicity and total energy distribution

DANCE calorimeter



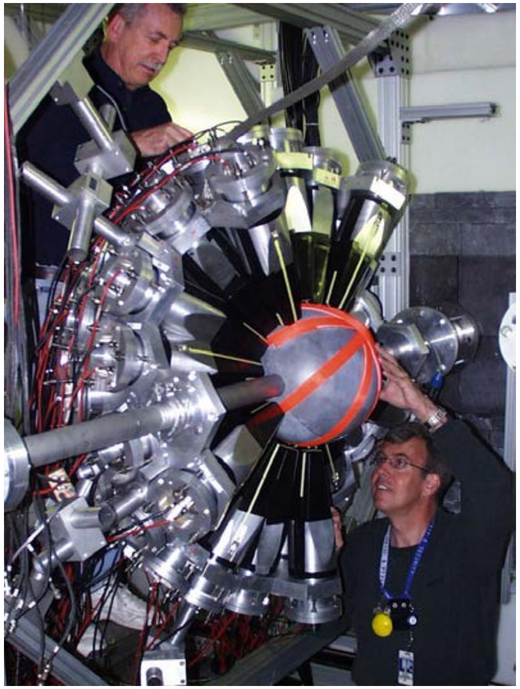


(20 spills (To)/sec)

- 160 BaF_2 crystals – each 0.75 liter
- Inner radius = 17 cm, crystal depth = 15 cm

- ^{6}LiH inner sphere to absorb scattered neutrons
- Internal conversion plus absorption in LiH may affect low-energy gamma spectrum

DANCE and LANSCE

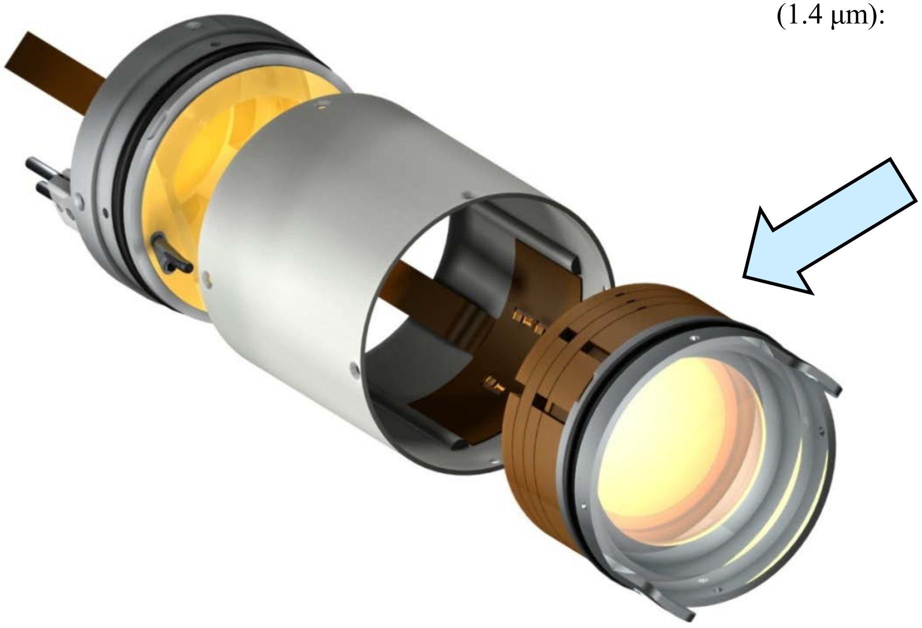


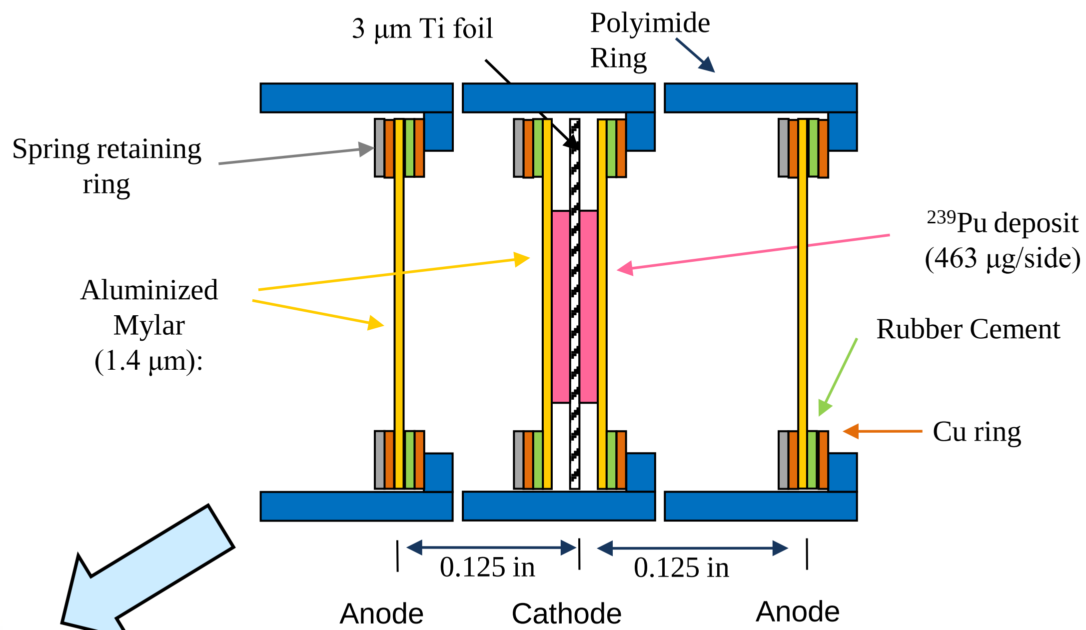
DANCE ball
(Open)
^{6}LiH sphere in center

(Los Alamos Neutron Science Center (LANSCE)

Fission tagging using PPAC

LLNL / LANL / MSI PPAC
4.37 cm dia X 4.77 cm long



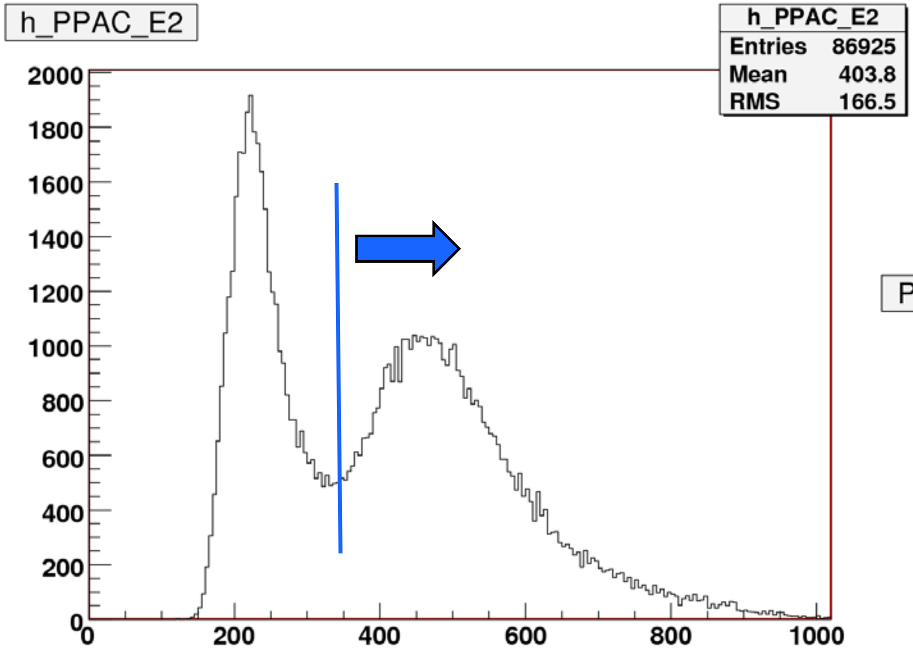


PPAC Target Assembly

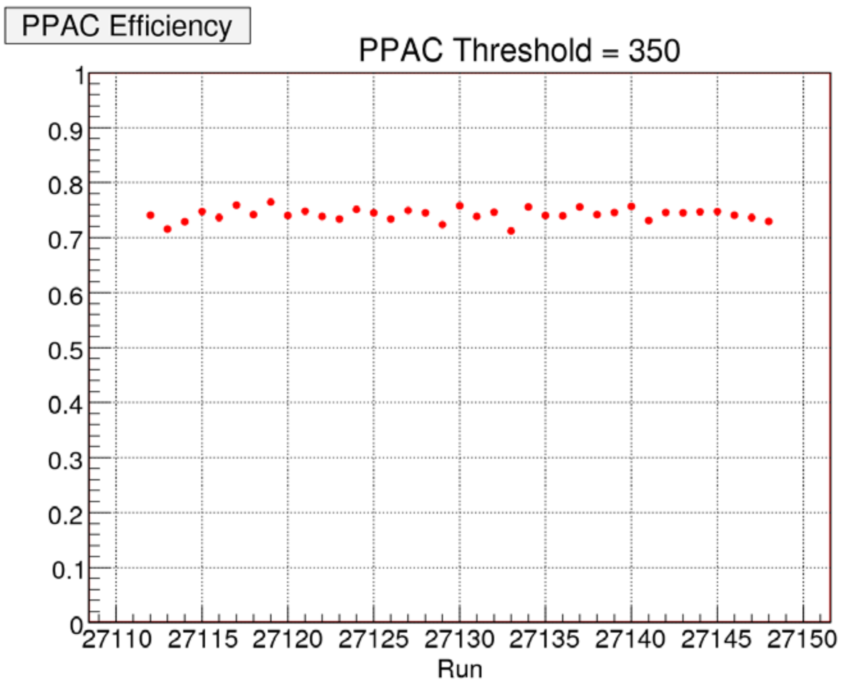
PPAC Params
Gas = Isobutane
P = 4.5 Torr
Flow = 4 cc/m
^{239}Pu (total) = 2.43 mg/cm^2
(0.7 cm dia deposit)
99.967% enriched

PPAC Performance

PPAC Pulse Height



Pulse Height units



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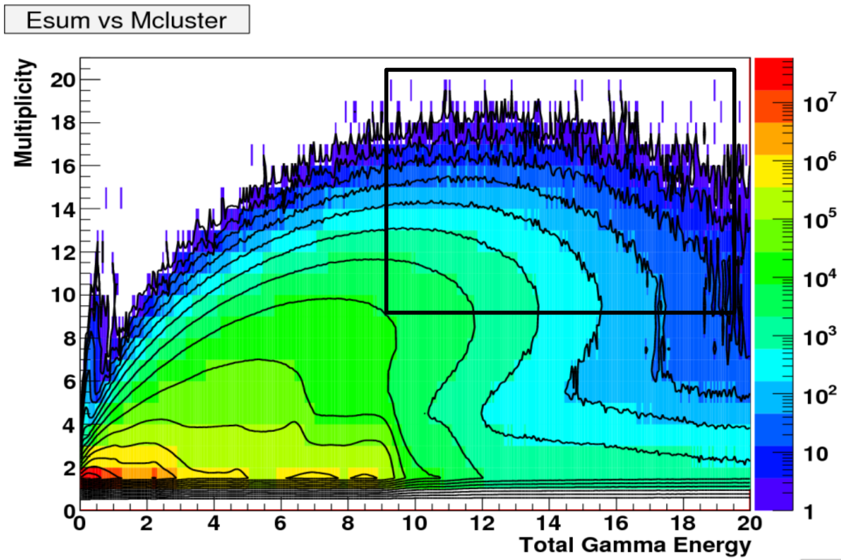
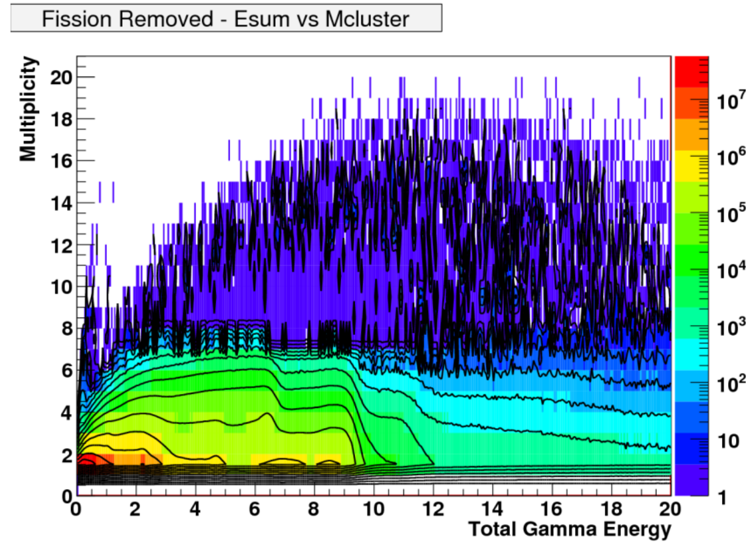
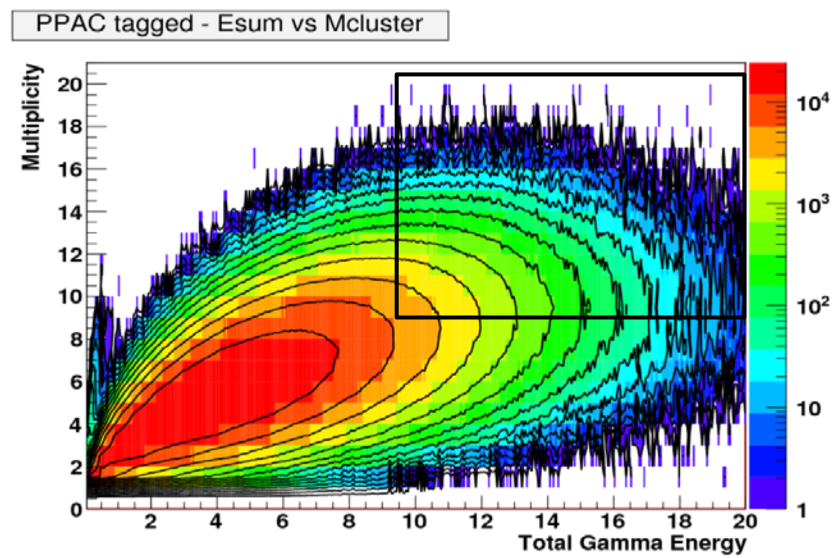
Measure Mult (ν_γ), E_{gam}, E_{tot} and Neutron energy





No fission tag
(Capture + (1-ε) Fission)



DANCE response correction crucial

Single gamma efficiency ε = 0.85

7 gammas => $\varepsilon = (0.85)^7 = 0.40$

Methods of response correction

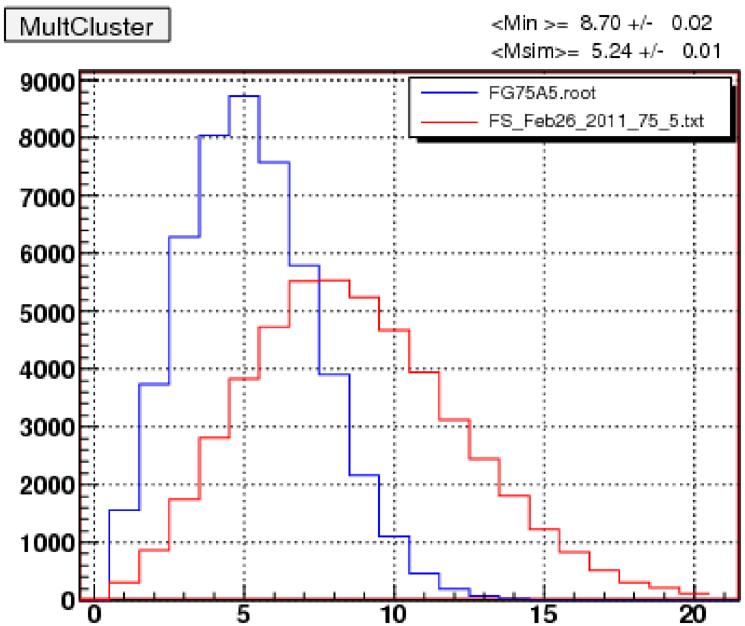
"Spectrum stripping" – calc response, subtract starting at
highest energy (eg. R. Billnert, et al., Phys Rev C **87**, 024601 (2013)

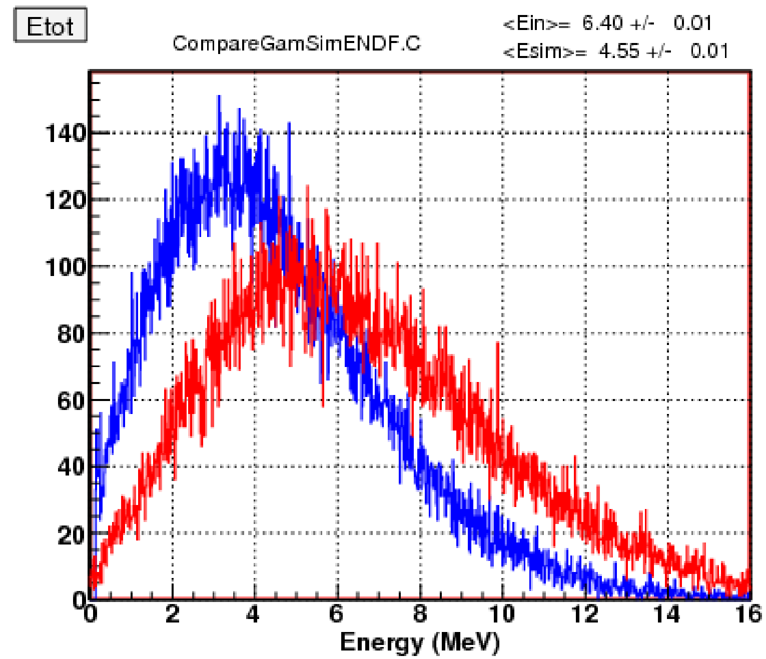
"Inverse Methods" – solve **O** = **R** **I** for input spectrum **I**
1-dimensional or 2-dimensional **O** and **I**
eg. A. Chyzh, et al., Phys. Rev. C **90**, 014602 (2014)
1D: Eγ, Mult each unfolded
2D: Unfold E_{tot} vs Mult Matrix

"Forward Methods" – Assume spectra, simulate response and compare
Iterate spectra until fit
Ultimate – use a real physics model with parameters
Experimental approach – don't depend on physics model
Parameterize data analytically
NOT (!!) a physics model (but may be motivated by physics)

Detector Response Correction

(Preliminary results shown in this figure!)





ε = 0.85 (Geometric)

$\varepsilon = (0.85)^7$ for 7 gamma rays = 0.40

Response Correction / Unfolding

- Philosophy – experimenters want to measure something,
- Not just fit parameters to a theory
- Forward method – not a physics model
  - Parameters motivated by physics
  - but really only parameters
  - Represent data

Detector Response Function

Multiplicity

- Sum over two distributions

  $M_\gamma = M_1 + M_2$
- Spin distributions $P(J) \sim (2J+1)e^{-J(J+1)/B^2}$

  (Wilhelmy, Phys Rev C 5, 2041 (1972))
- Assume P(M) = P(J) (Number of gammas = spin)

  (Good for E1, M1 roughly)
- 2 fission products => $P(M\gamma) = P(M_1) + P(M_2)$

B's are fitted – M's (J's) are random variables

Detector Response Function

Gamma energy distribution

- $P(\varepsilon) \sim T(\varepsilon)\,\rho(\varepsilon)$
  - For E1 transitions: $T(\varepsilon) \sim A\varepsilon^3$
  - "Constant Temperature" $\rho(\varepsilon) \sim Be^{\alpha Ex} = Be^{\alpha(Eo-\varepsilon)}$
  - $P(\varepsilon) \sim \varepsilon^3 e^{-\alpha\varepsilon}$
- Lemaire calculation: $P(\varepsilon) \sim \varepsilon^2 e^{-\beta\varepsilon}$

  (S. Lemaire et al., Phys. Rev. C **73**, 014602 (2006))
- Best fit: :

  $P_1(\varepsilon) \sim \varepsilon^2 e^{-(a1+M\gamma b1)\varepsilon}$

  $P_2(\varepsilon) \sim \varepsilon^3 e^{-(a2+M\gamma b2)\varepsilon}$ (Fit params: a_1, a_2, b_1, b_2)
- Observed Gamma spectrum is sum over many fission products
  - Different excitation energies, temperatures, multipolarity

Details:

J.L. Ullmann, et al., Phys. Rev C **87**, 044607 (2013)

M. Jandel et al., Los Alamos Report LA-UR-12-24975

Procedure for fitting parameters

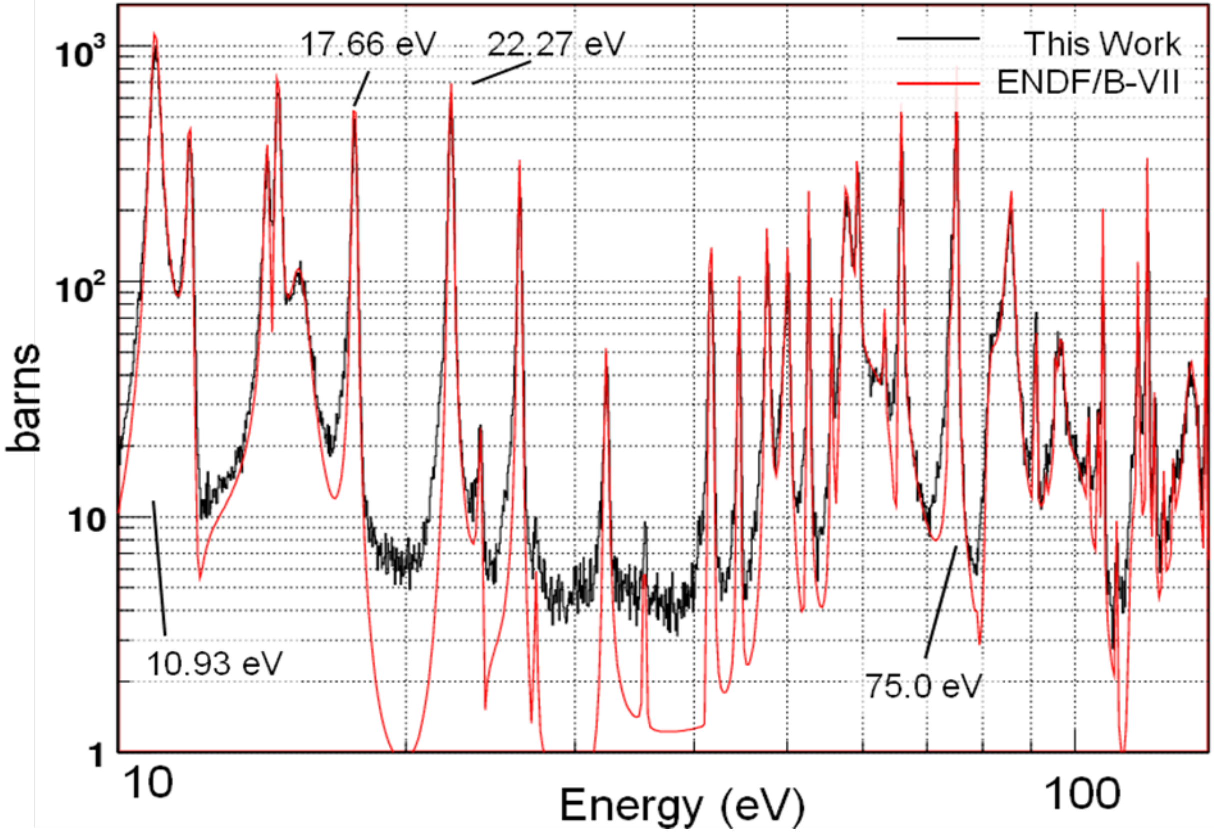
- Generate event consisting of $M_\gamma = M_1 + M_2$ gamma rays with energy ε_i from $P(M_{1,2})$ and $P_{1,2}(\varepsilon_\gamma)$
- Transport all gammas from event through GEANT4 model of DANCE to produce "experimental" values
- Compare to measured values – calculate χ^2
    - 2D arrays of ε_γ vs crystal multiplicity, ε_{tot} vs M_{cr}
    - M_{cr} chosen to avoid overlapping clusters
    - M_{cr} easily simulated by GEANT
    - Arrays with 4 different thresholds (150,200,300,400 keV)
    - More weight to higher threshold
    - Total degrees of freedom: 750
- Vary parameters at random to find minimum χ^2

  Iterated "Simulated Annealing" technique

  Vary parameters over range $1\pm\delta$

  $P(\Delta) = e^{-\Delta/T}$

^{239}Pu(n,fission) Cross Section



Current results gated on **10.93 + 11.89** eV 1^{+} resonances
Thermal, 7.82, 22.26, 75.0 1^{+} and 32.33 0^{+} resonances have similar spectra

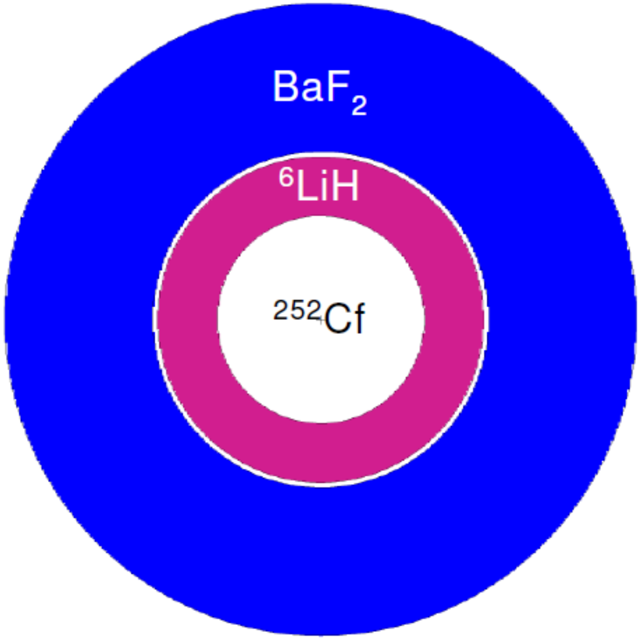
Detour: Fission neutron response

Fission neutrons – Ave energy ~ 2 MeV

BUT – High energy tail ! (Maxwellian)

Fission neutron effects – MCNP by TNT

Transport ^{252}Cf fission neutron spectrum into DANCE



DIMENSIONS:

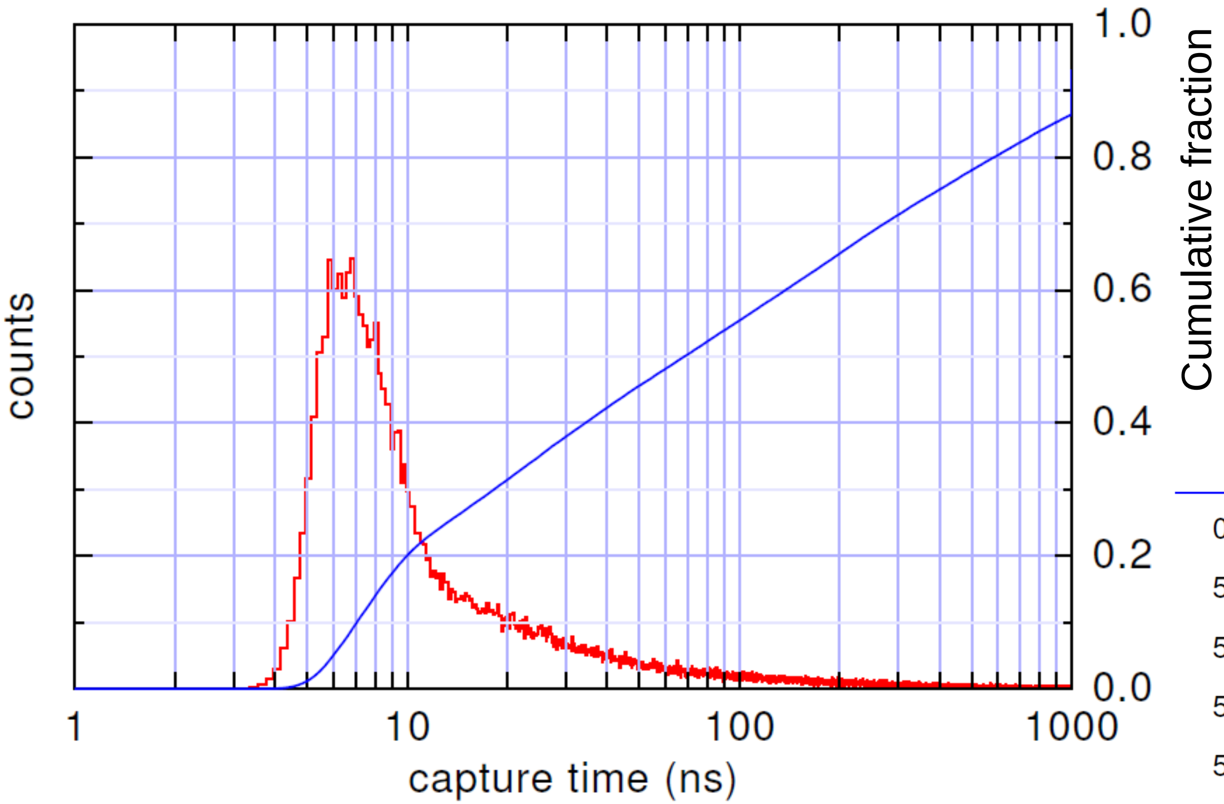
^{6}LiH sphere	=	10.50 cm (id)
		16.51 cm (od)
		0.85 g/cm^3
BaF_2 sphere	=	17.00 cm (id)
		32.00 cm (od)
		4.88 g/cm^3

^{252}Cf point source at center

(MCNP Calculations by T. Taddeucci)

Detour: Fission neutron response

DANCE response to fission-spectrum neutrons



Z	A	captures
09	19	29606
56	134	11590
56	135	62166
56	136	10779
56	137	10153
56	138	6403
total captures		130697 = 1.307%
total neutrons		10000000

Total Efficiency for neutrons
1.3%

Best-fit parameters

$$P(M_\gamma) = P(M_1) + P(M_2)$$

$$P(M_1) = (2M_1 + 1)e^{-M_1(M_1+1)/B_1^2}$$

$$P(M_2) = (2M_2 + 1)e^{-M_2(M_2+1)/B_2^2}$$

$$P_1(\varepsilon_\gamma) = \varepsilon^2 e^{-(a_1 + M_\gamma b_1)\varepsilon}$$

$$P_2(\varepsilon_\gamma) = \varepsilon^3 e^{-(a_2 + M_\gamma b_2)\varepsilon}$$

$B_1 = 6.66 \pm 0.15$

$B_2 = 2.54 \pm 0.12$

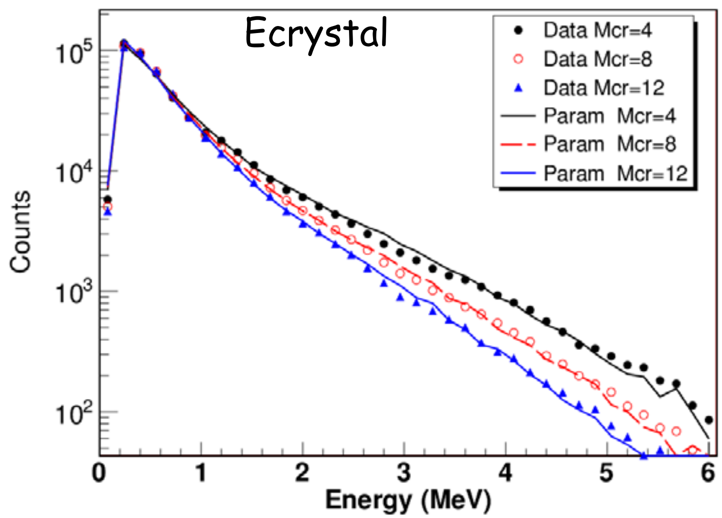
$a_1 = 3.80 \pm 0.12$

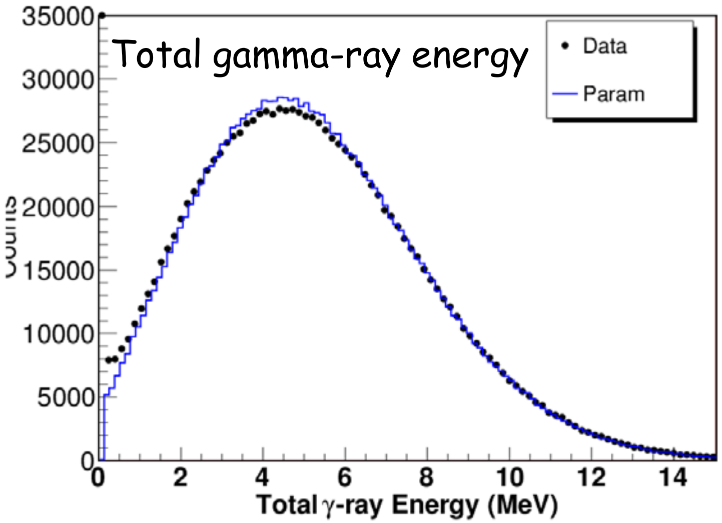
$a_2 = 1.53 \pm 0.14$

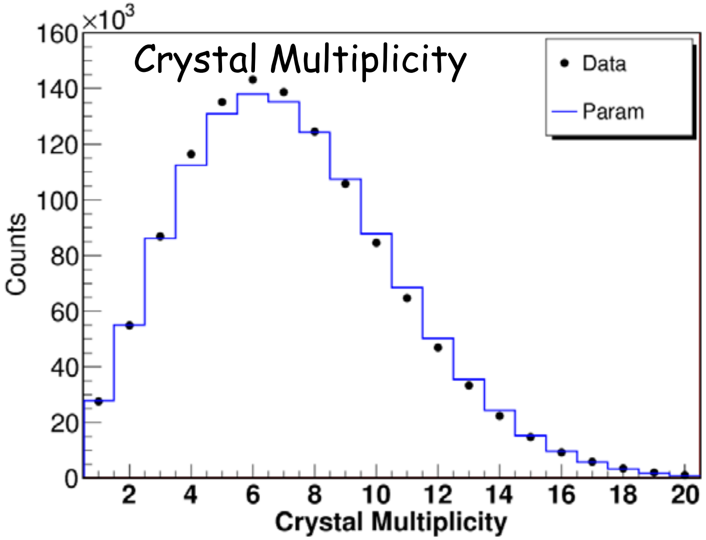
$b_1 = 0.0428 \pm 0.0019$

$b_2 = 0.0522 \pm 0.0034$

Raw Data vs Parametrized fit







(All spectra for 150 keV E_{cr} threshold)

Theoretical Calculations

Monte-Carlo Hauser-Feshbach Model

I. Stetcu, P. Talou, T. Kawano, M. Jandel, to be published

T. Kawano, P. Talou, M.B. Chadwick, T. Watanabe, J. Nucl. Sci. Tech. 47, 462 (2010)

N. Becker, P. Talou, T. Kawano, Y. Danon, I. Stetcu, Phys. Rev.C 87, 014627 (2013)

- Fragment mass distribution semi-empirical
- Radiative strength functions from RIPL-3
- Level density was Gilbert-Cameron formalism
- Spin Distribution:

$$\mathrm{P(J)} = (2\mathrm{J}+1)e^{-J(J+1)/2B^2}$$

$$B = \alpha \frac{I_o T}{\hbar^2}$$

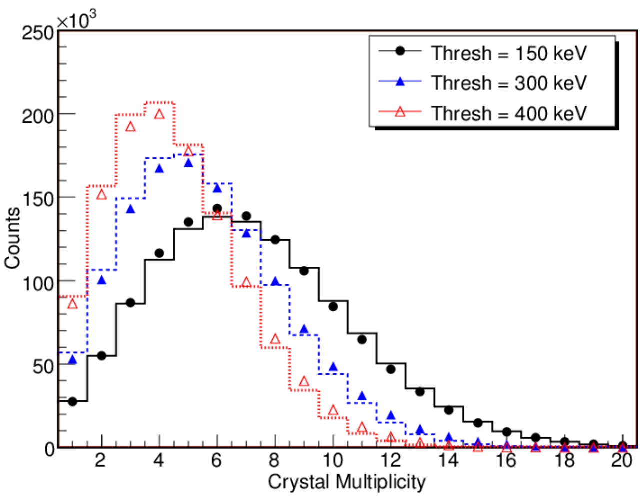
I_o = ground-state moment of inertia

T = fragment temperature

A = adjustable parameter

Results – Gamma-ray Multiplicity

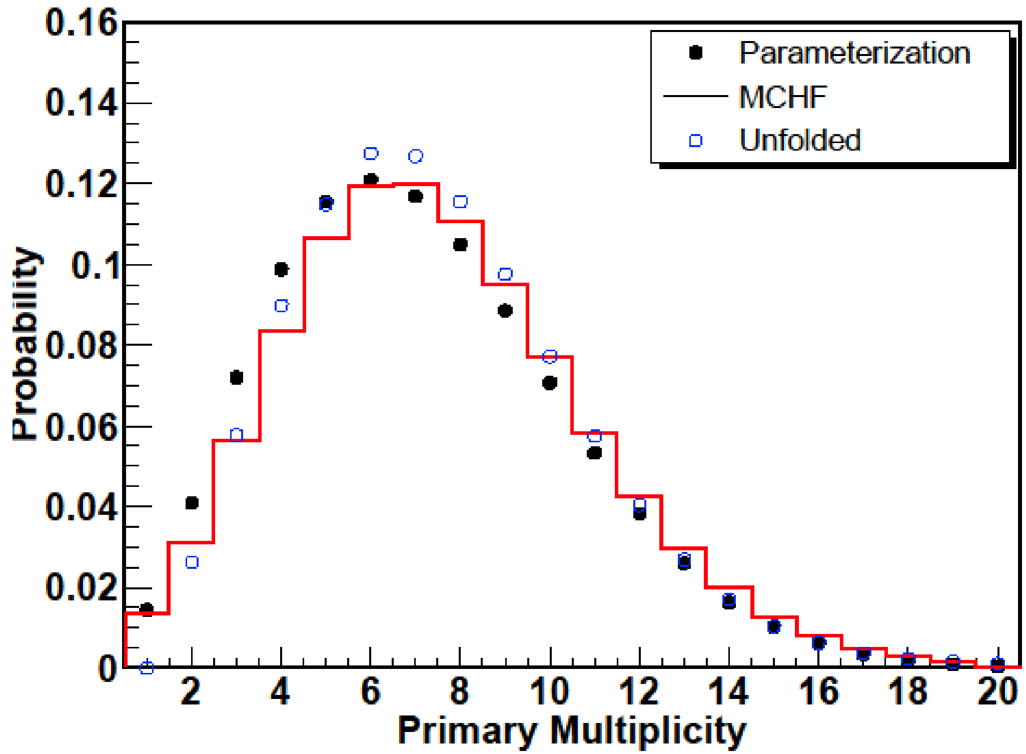
^{239}Pu(n,f) Measured Crystal Multiplicity very sensitive to threshold



Average Raw Crystal Multiplicity

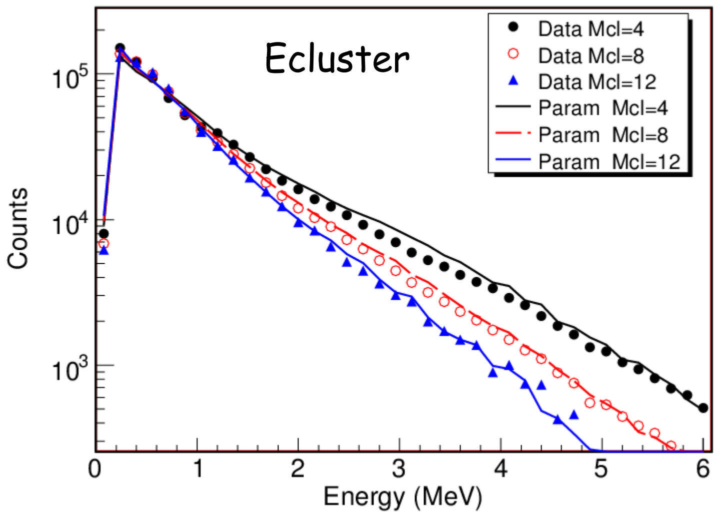
Thresh	<Mcl>
150 keV	7.2
300	5.7
400	4.7

^{239}Pu(n,f) Corrected γ Multiplicity

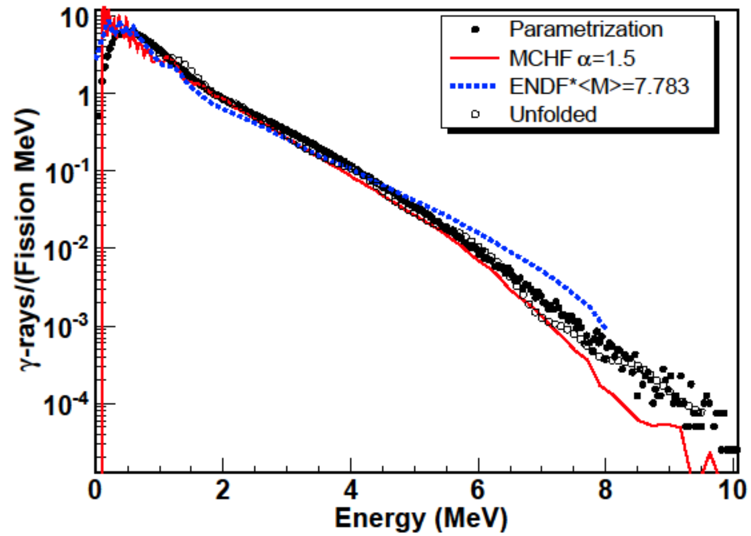


- 150 keV Threshold
- Unfolded – 1D from Chyzh, Phys. Rev. C 87 034620 (2014)
- MCHF from Stetcu, LA-UR-14-23128 (α = 1.5)

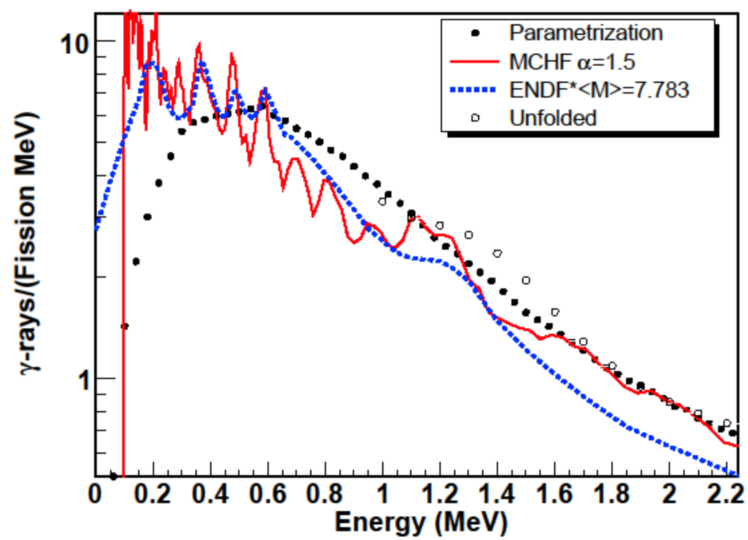
Results: ^{239}Pu(n,f) Gamma Energy



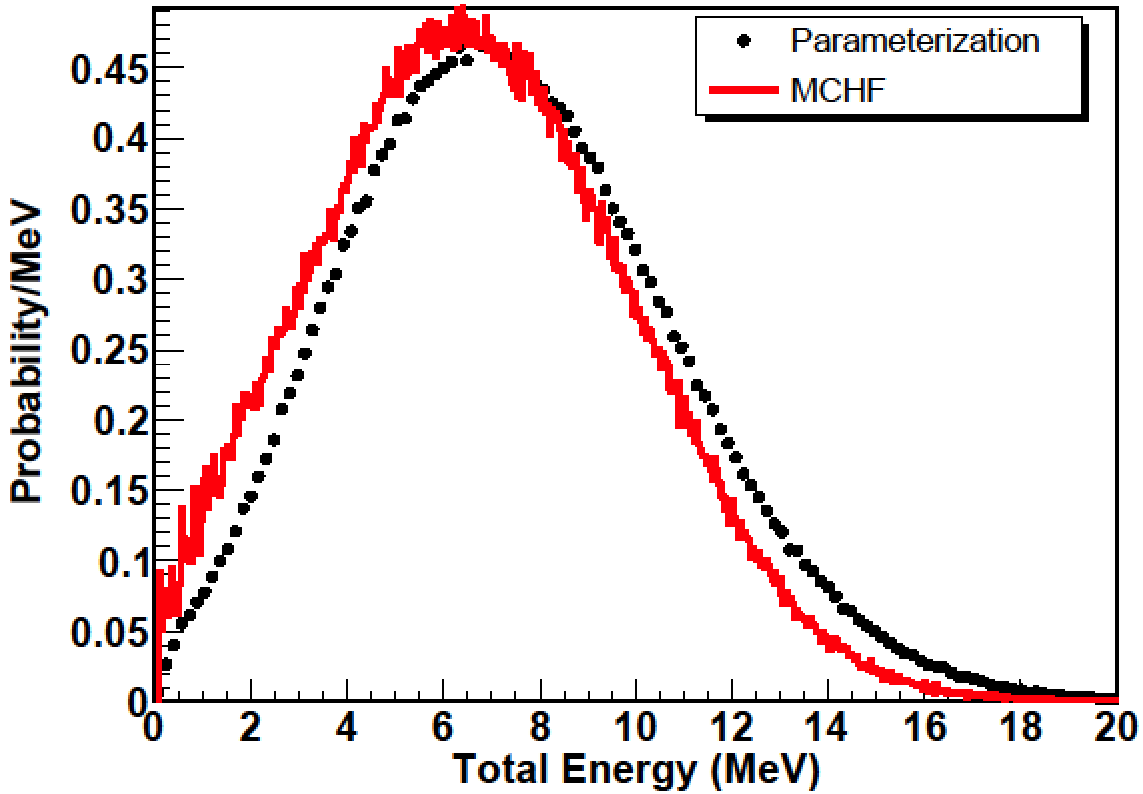
Measured (before response correction) cluster energy for Cluster Multiplicities 4,8,12



- Response-corrected gamma energies (all multiplicities)
- MCHF Calculation (Stetcu)
- Unfolded 1D (Chyzh)



Results: ^{239}Pu(n,f) Total gamma-ray energy



- Response-corrected gamma energies (all multiplicities)
- MCHF Calculation (Stetcu, α = 1.5)

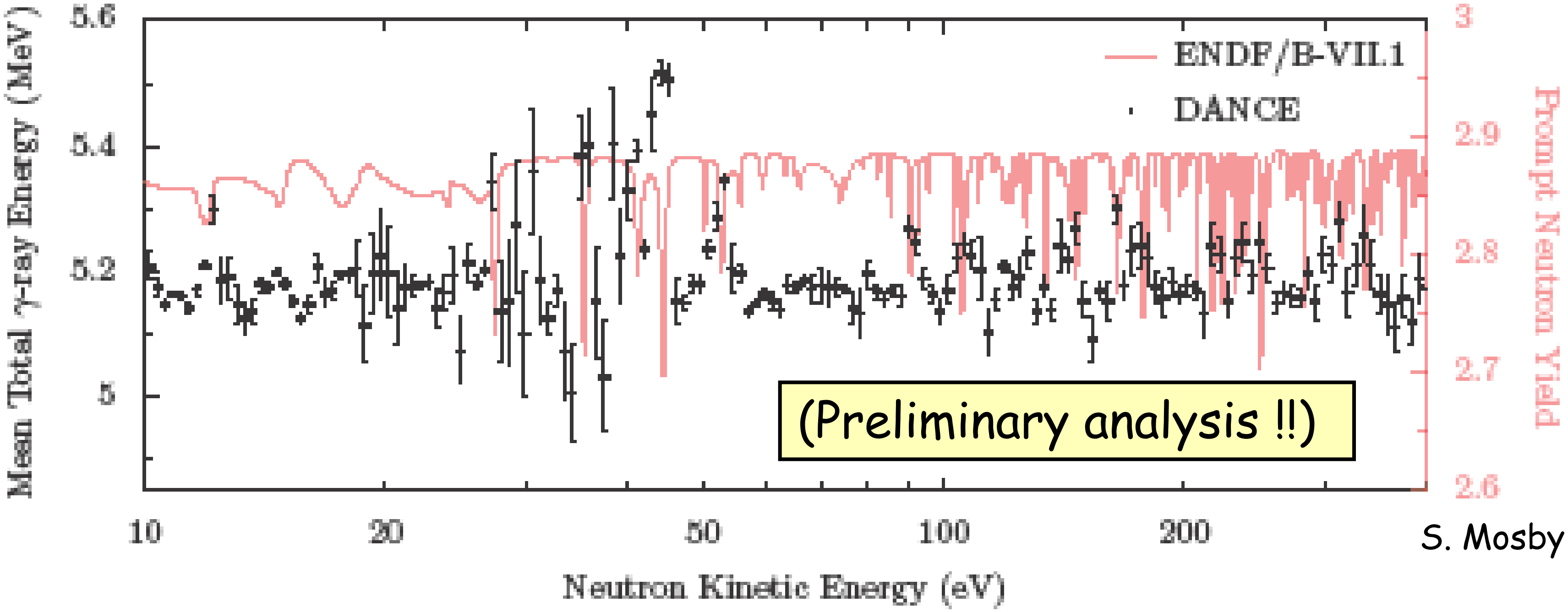
^{239}Pu(n,f) Average γ Multiplicity and $E_{tot,\gamma}$

	<M>	$\langle E_{tot,\gamma} \rangle$
This Work (10.93 eV)	7.15 ± 0.09	7.46 ± 0.06
Pleasonton (thermal) (Nucl. Phys. A 213, 413 (1973))	6.88 ± 0.35	6.73 ± 0.35
Verbinski (thermal) (Phys. Rev. C 7, 1173 (1973))	7.23	6.81
MCHF (Stetcu/Talou) (Thermal, 140 keV thresh. α=1.5) LA-UR-14-23128	7.05	6.74
Madland Summary		6.74
Chyzh, “1D” Unfolding (Phys. Rev. C 87, 034620 (2013))	7.50	7.30
Chyzh “2D” Unfolding (Phys. Rev. C 90, 014602 (2014))	7.93	7.94

- Uncertainty in <Etot> ~ Fitting uncertainty, determined as σ of 14 iterations with lowest χ^2
- <M> very sensitive to detection threshold!

Unexpected detail: (n,γf)?

Resonances we studied looked to be quite similar – but is that universally true? (S. Mosby – ^{239}Pu(n,γ) analysis)



S. Mosby

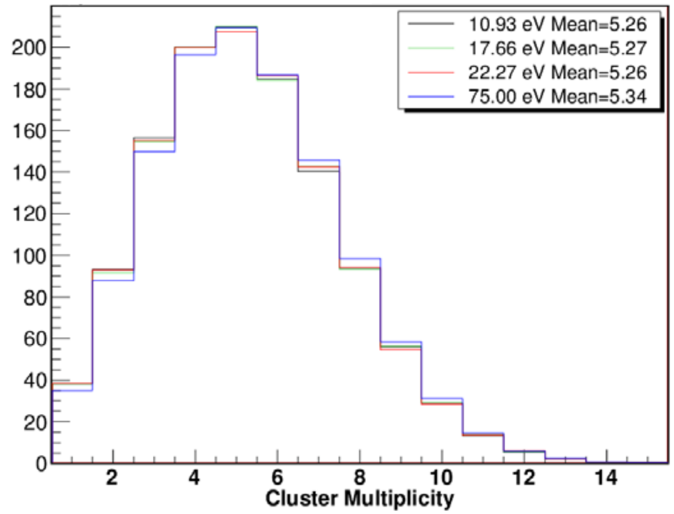
$\langle \nu_n \rangle$ dips at several weak L=0 resonances

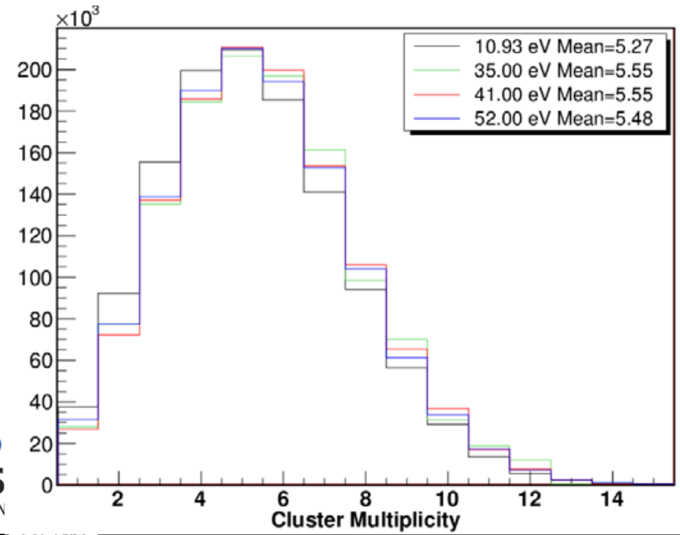
35.5 eV 1+
41 1+
44.5 1+
52.7 1+

Neutron multiplicity measurements
E. Fort, Nucl. Sci. Eng. **99**, 375 (1988)
Gamma multiplicity measurements
Yu. Ryabov Nucl. Phys. **A216**, 395 (1973)

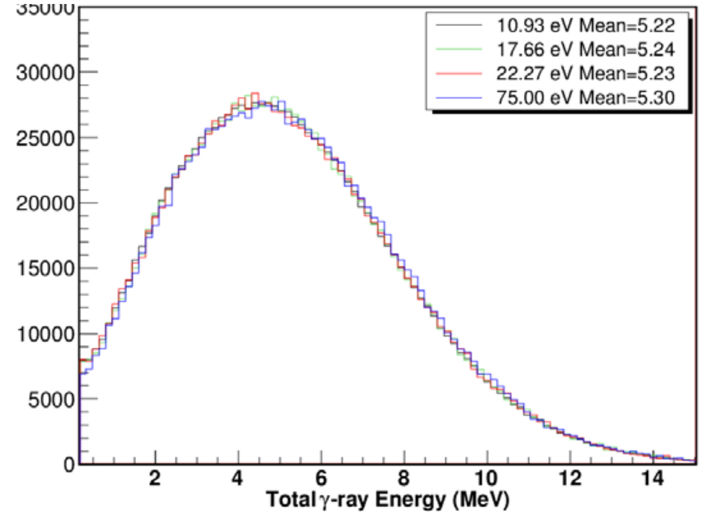
Resonance properties – Raw data

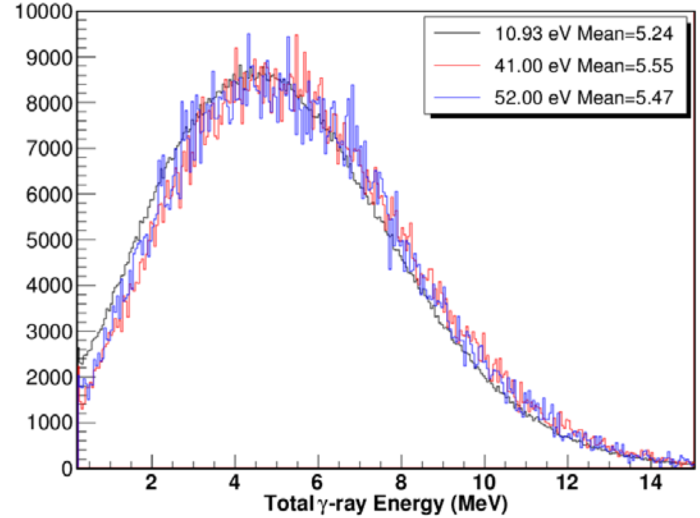
Gamma multiplicity





Total Gamma Energy





Summary and Conclusions

- Measurements of *distribution* of multiplicity, gamma energy, and total gamma energy ("forward modeling" parameterization of data) – Needs high-segmentation, 4π capability !!
- Average multiplicity in agreement with previous measurements – sensitive to thresholds
- Average Etot ~ 10% higher than previous
- Theoretical modelling reproducing ^{239}Pu data, but with adjustable parameters
- Still some puzzles; (n,γf)?
- **"More work to be done!"**