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

J.L. Ullmann
(For the DANCE collaboration)

LANSCE-NS
Los Alamos National Laboratory
Los Alamos, New Mexico USA

Workshop on Fission Experiments and Theoretical Advances
(FIESTA)

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Collaborators and Acknowledgements

S. Mosby, M. Jandel, T.A. Bredeweg, A. Couture,
R.C. Haight, J.M. O'Donnell, D.J. Vieira, J.B. Wilhelmy,
A. Hayes-Sterbenz, P. Talou, I. Stetcu, T. Kawano
Los Alamos National Laboratory

C.-Y. Wu, A. Chyzh, J.A. Becker, J. Gostic, R. Henderson, E. Kwan
Lawrence Livermore National Laboratory

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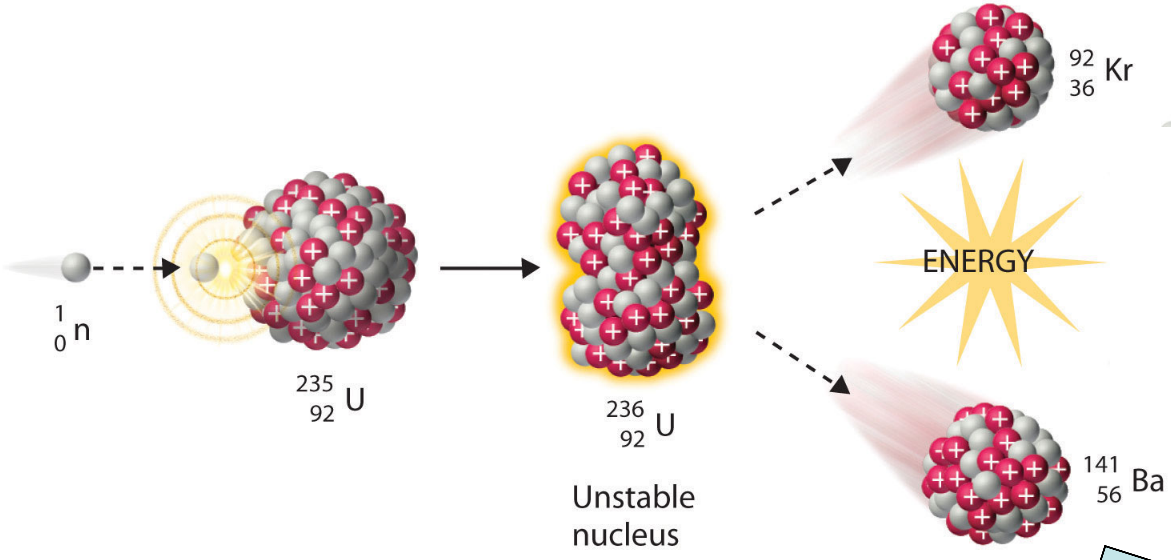
Fission Physics

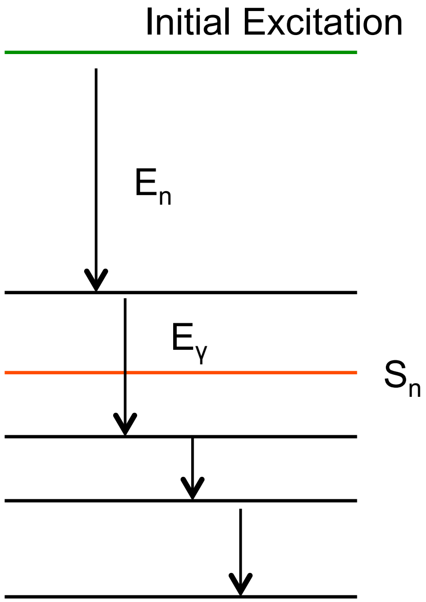


$^{239}Pu(n,f)$

$\nu_n = 2.9$

$\nu_\gamma = 7.2$

(Thermal)

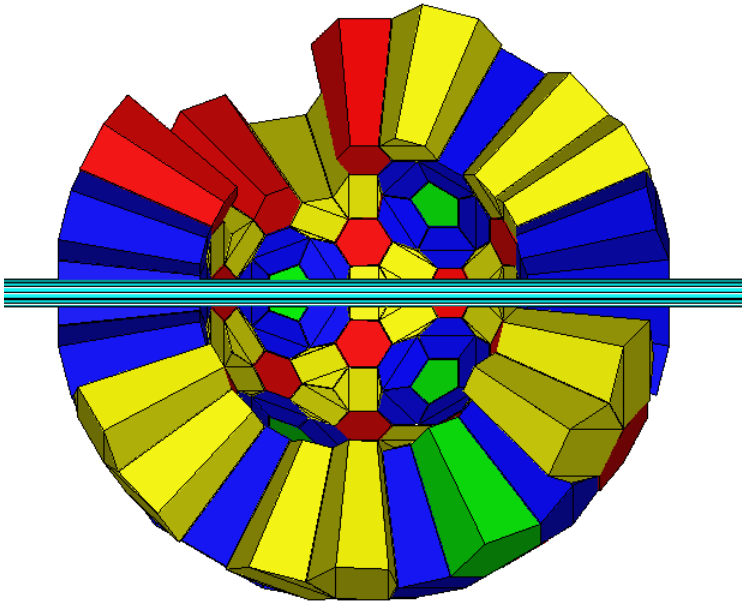


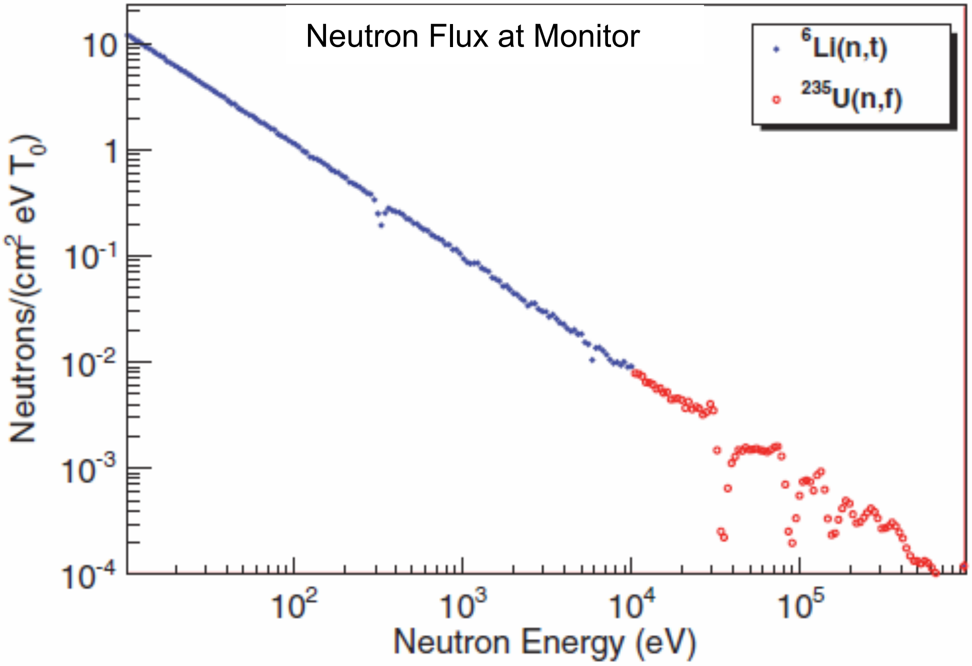
Initial fragment – high spin, excitation
Neutron decay – removes excitation
Gamma decay – (E1) removes spin and energy
Gammas – from fragment decay
- neutron rich (? Structure)
- Fragment mass distribution changes with neutron energy

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 gamma-ray calorimeter



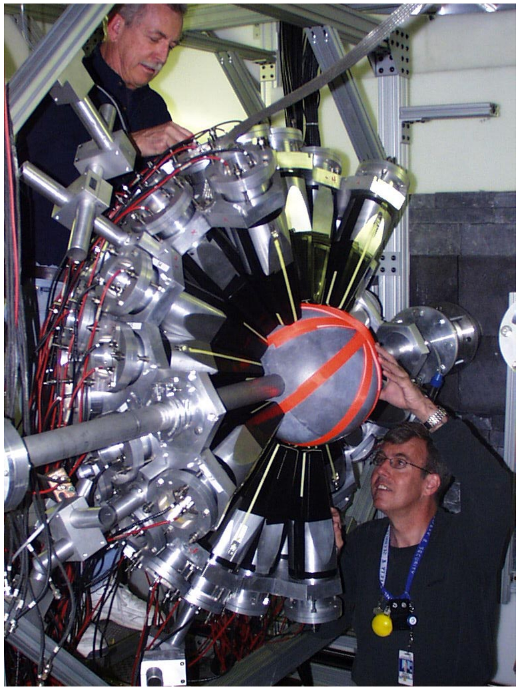


(20 spills (To)/sec)

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

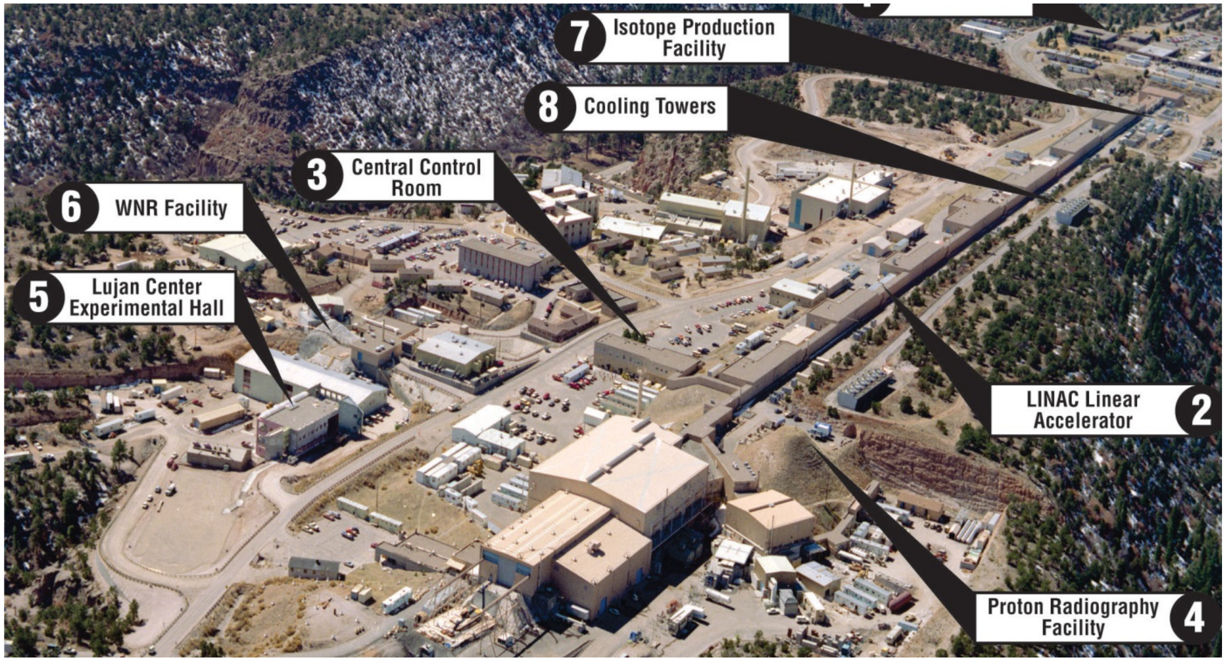
- ^{6}LiH inner sphere to absorb scattered neutrons
- Internal conversion plus absorption in LiH may affect low-energy gamma spectrum

DANCE and LANSCE



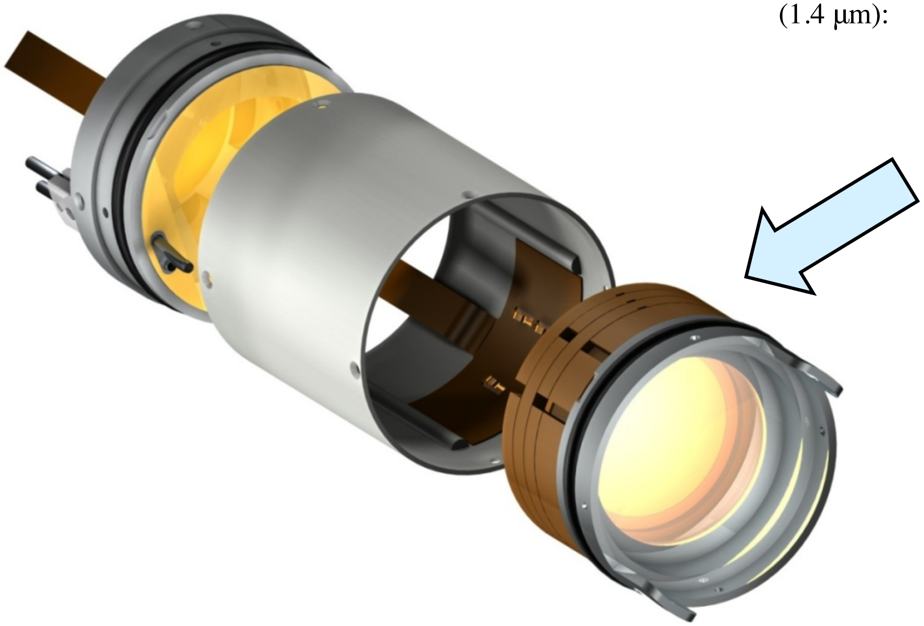
DANCE ball
(Open)
^{6}LiH sphere in center

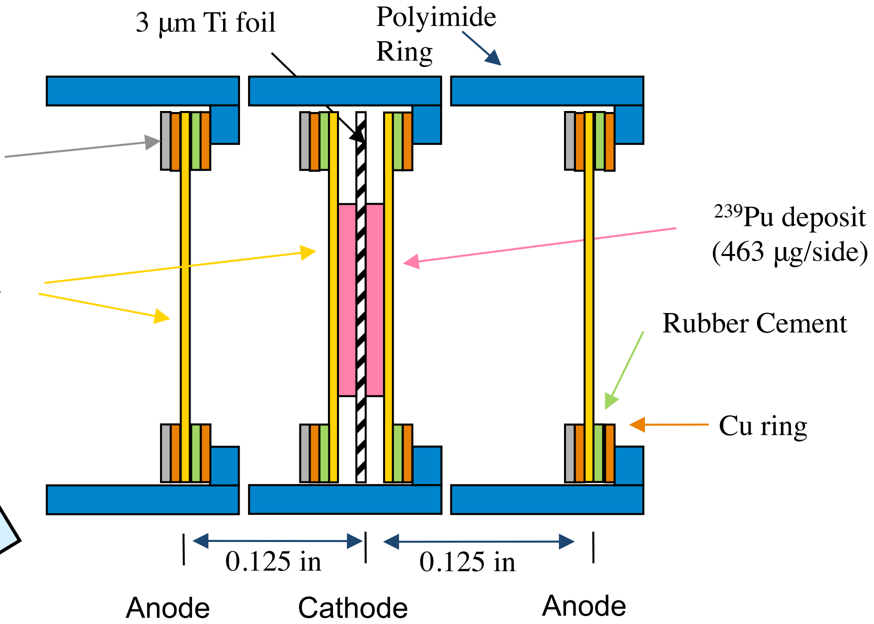
(Los Alamos Neutron Science Center (LANSCE)



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

Fission efficiency ~ 70%

DANCE response correction crucial

Single gamma efficiency ε = 0.85
7 gammas => $\varepsilon = (0.85)^7 = 0.40$

Methods of response correction

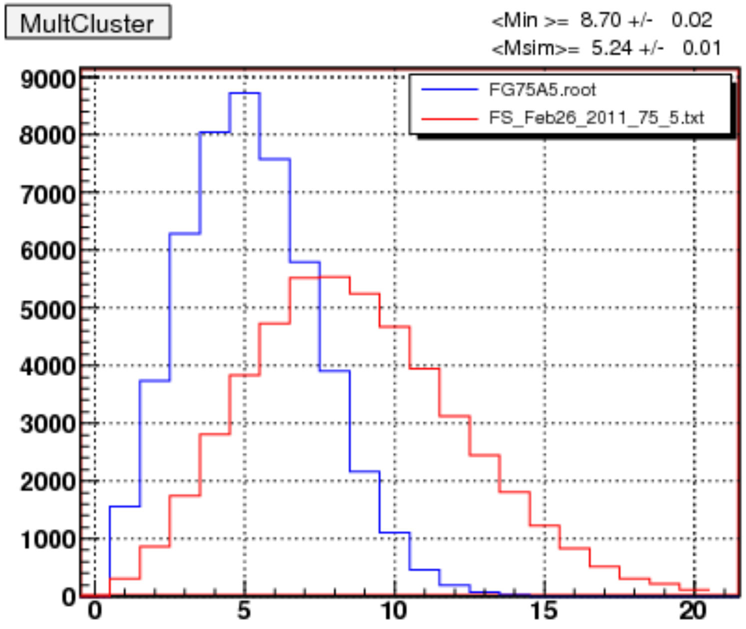
"Spectrum stripping" – calc response, subtract starting at
highest energy (eg. R. Billnert, et al., Phys Rev C **87**, 024601 (2013)

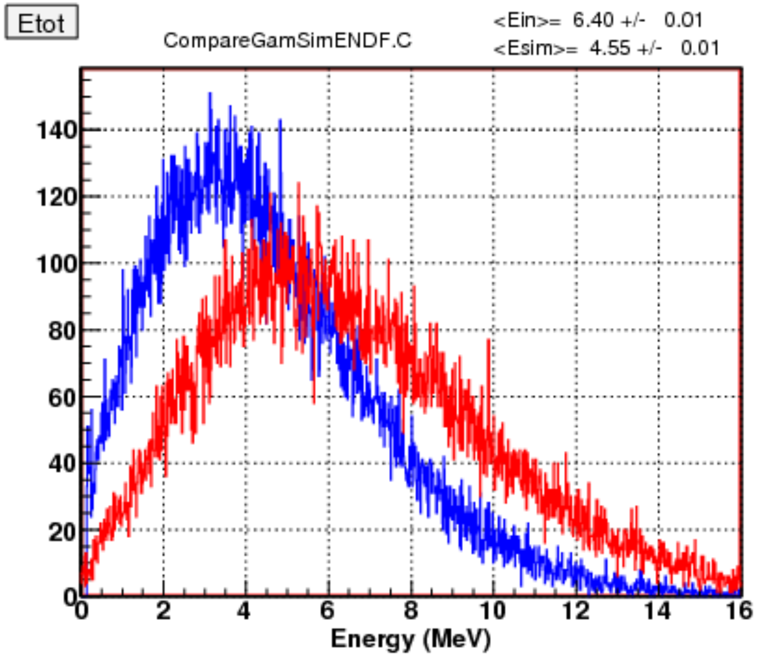
"Inverse Methods" – solve **O** = **R** **I** for input spectrum **I**
1-dimensional or 2-dimensional **O** and **I**
eg. A. Chyzh, et al., Phys. Rev. C **90**, 014602 (2014)
1D: Eγ, Mult each unfolded
2D: Unfold E_{tot} vs Mult Matrix

"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!)





$\varepsilon = 0.85$ (Geometric)

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

Parameterized fission spectra

Multiplicity

- Sum over two distributions

$$M_\gamma = M_1 + M_2$$

- Assume Multiplicity ~ Spin distributions $P(J) \sim (2J+1)e^{-J(J+1)/B^{\wedge}2}$

(Wilhelmy, Phys Rev C 5, 2041 (1972))

Gamma energy distribution

- $P(\varepsilon) \sim T(\varepsilon)\, \rho(\varepsilon)$ $T(\varepsilon) \sim A\, \varepsilon^3$ (E1), $\rho(\varepsilon) \sim Be^{a(Eo-\varepsilon)}$
- 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
- Parameterization – not a physics model

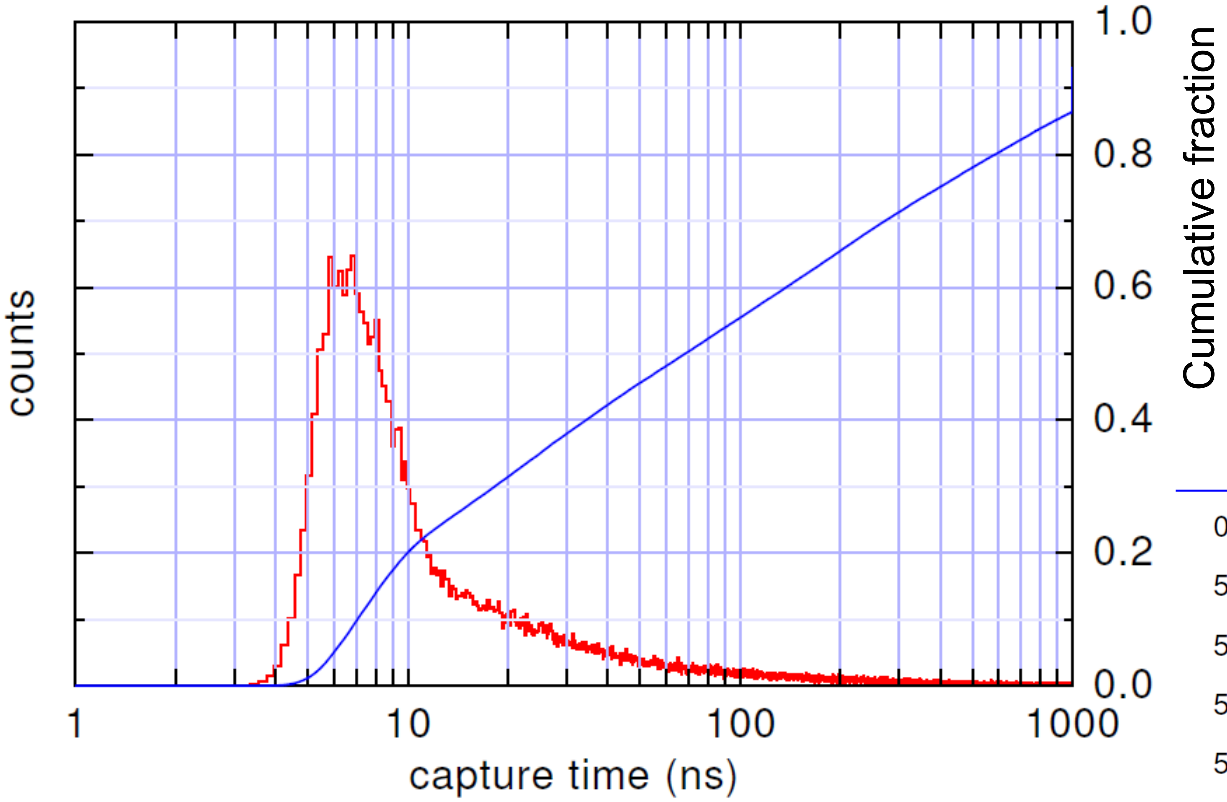
Procedure for fitting parameters

- 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
  - M_{cr} chosen to avoid overlapping clusters
  - M_{cr} easily simulated by GEANT
  - E_{tot} (Tresh = 150 and 400 keV)
  - M_{cr} (150 keV Threshold)
  - E_γ (150 keV threshold)
  - More weight to higher threshold
- Vary parameters at random to find minimum χ^2

  Iterated "Simulated Annealing" technique
  Vary parameters over range $1\pm\delta$
  $P(\Delta) = e^{-\Delta/T}$

Detour: Fission neutron response

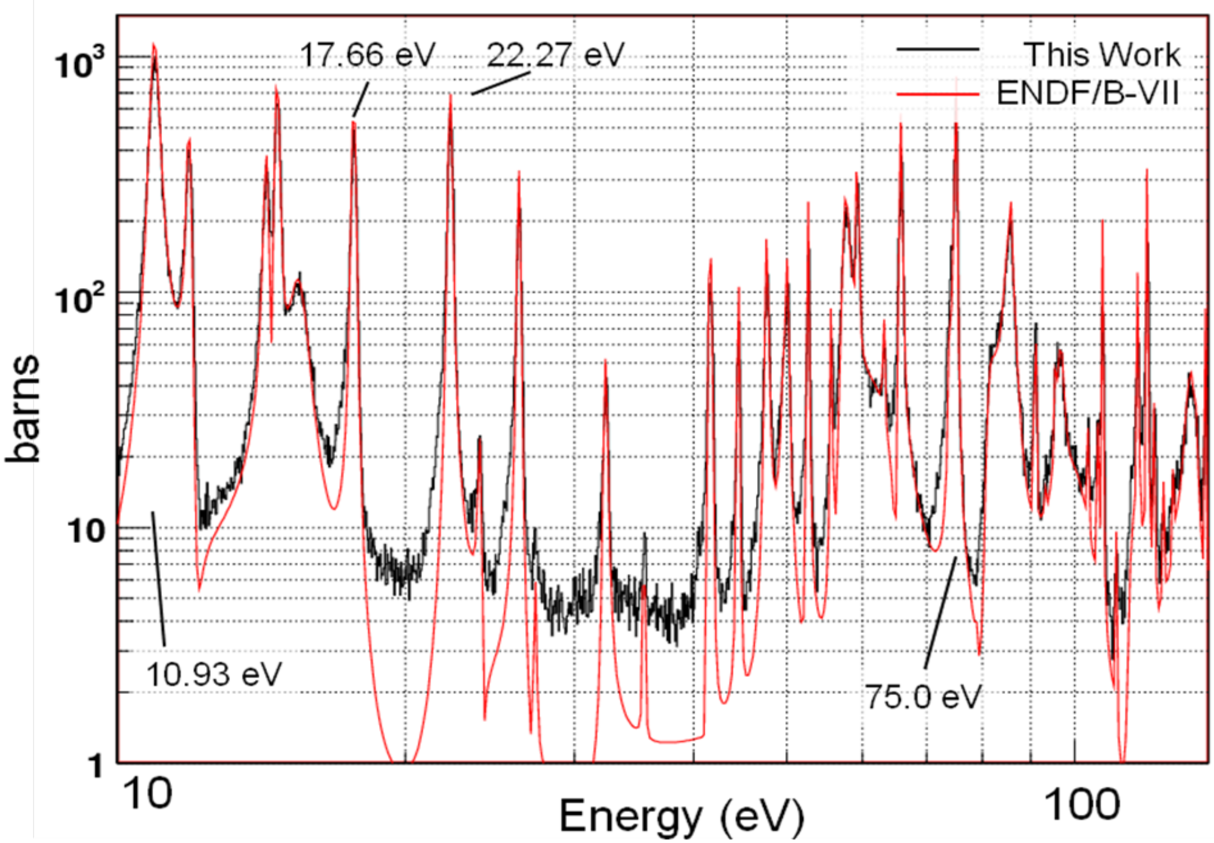
DANCE response to fission-spectrum neutrons



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%

^{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

Best-fit parameters

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

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

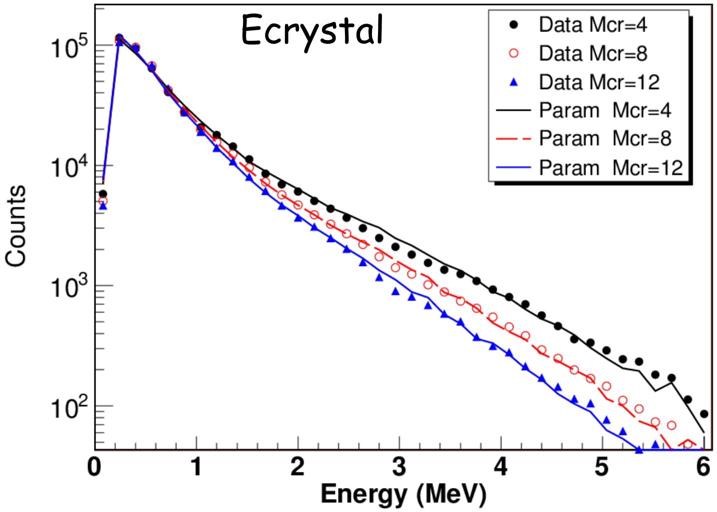
$$P(M_2) = \left(2M_2 + 1\right)e^{-M_2(M_2+1)/B_2^2}$$

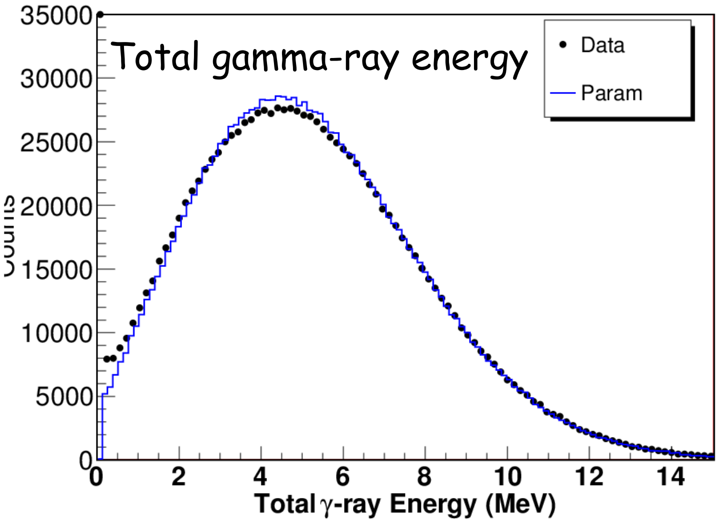
$$P_1(\varepsilon_\gamma) = \varepsilon^2 e^{-(a_1+M_\gamma b_1)\varepsilon}$$

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

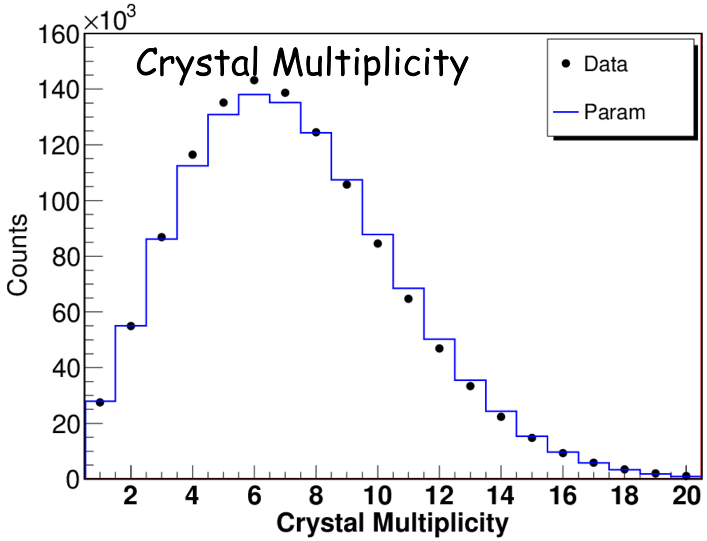
$B_1 = 6.66 \pm 0.15$
$B_2 = 2.54 \pm 0.12$
$a_1 = 3.80 \pm 0.12$
$a_2 = 1.53 \pm 0.14$
$b_1 = 0.0428 \pm 0.0019$
$b_2 = 0.0522 \pm 0.0034$

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, Phys. Rev. C 90, 024617 (2014)
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:

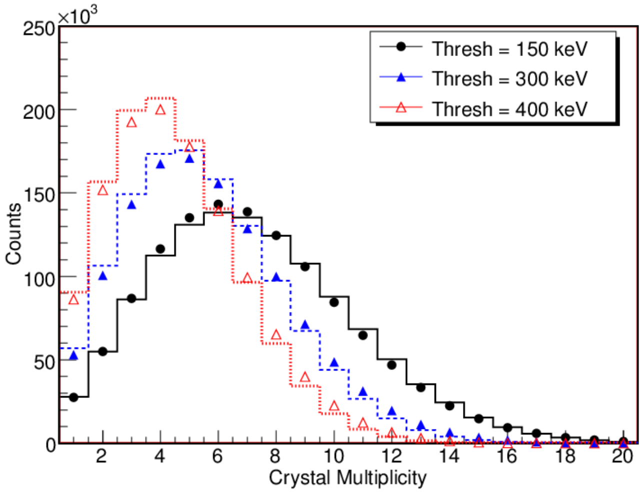
$$\mathrm{P(J)} = (2\mathrm{J}+1)e^{-J(J+1)/2B^2}$$

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

I_o = ground-state moment of inertia
T = fragment temperature
A = adjustable parameter

Results – Gamma-ray Multiplicity

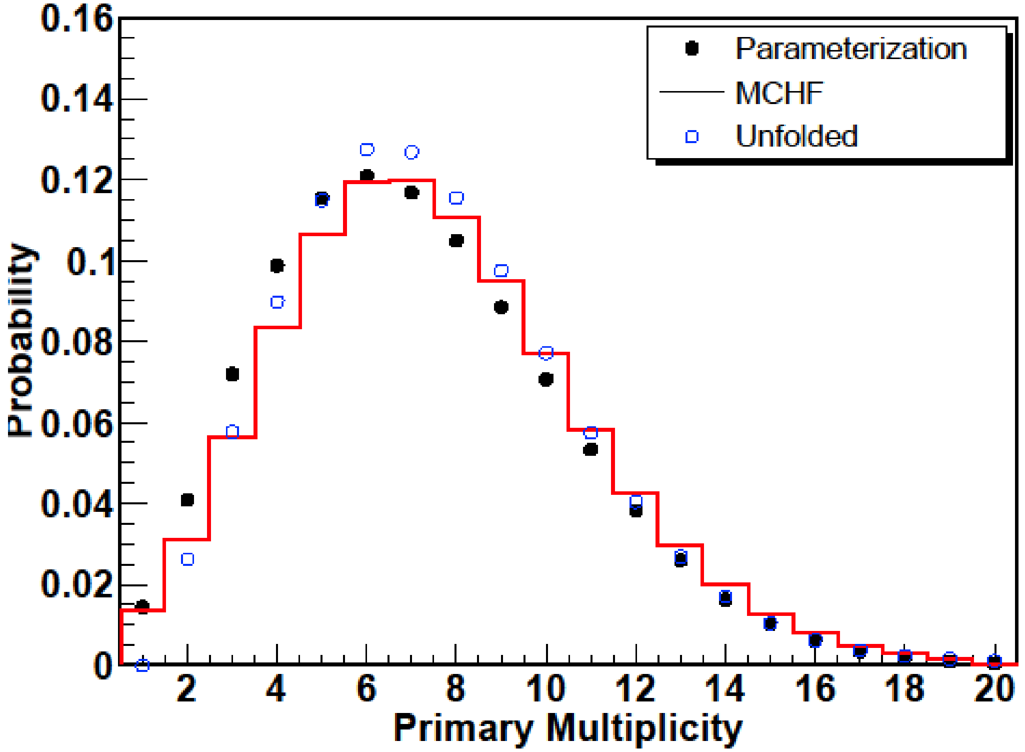
^{239}Pu(n,f) Measured Crystal Multiplicity very sensitive to threshold



Average Raw Crystal Multiplicity

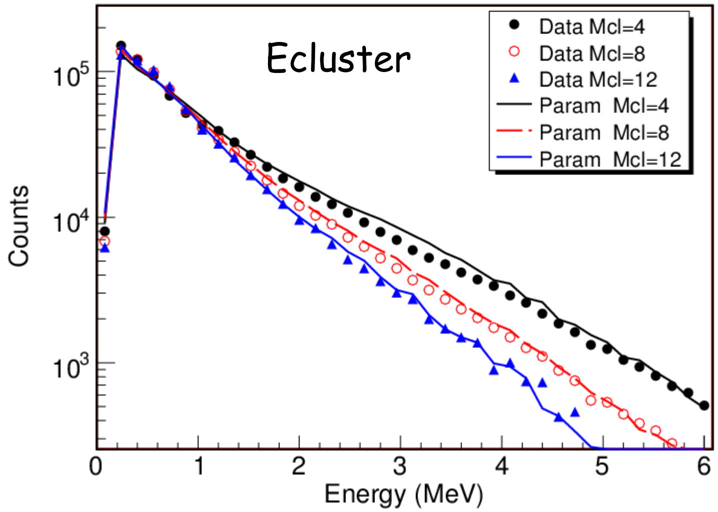
Thresh	<Mcl>
150 keV	7.2
300	5.7
400	4.7

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

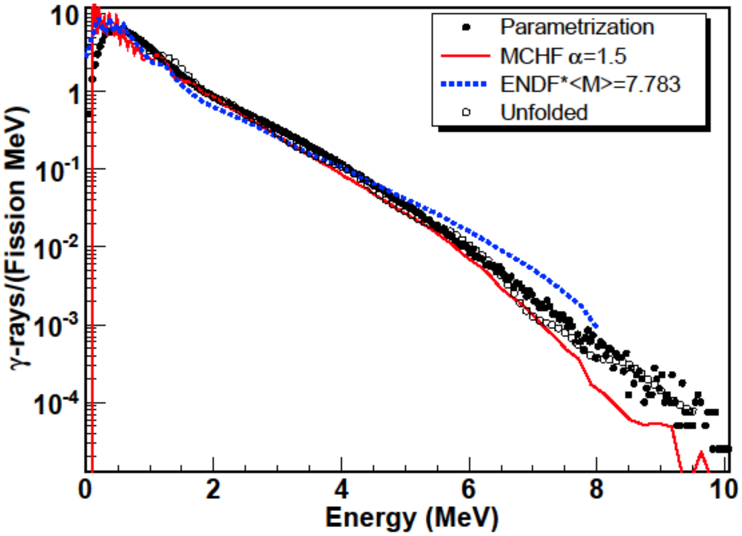


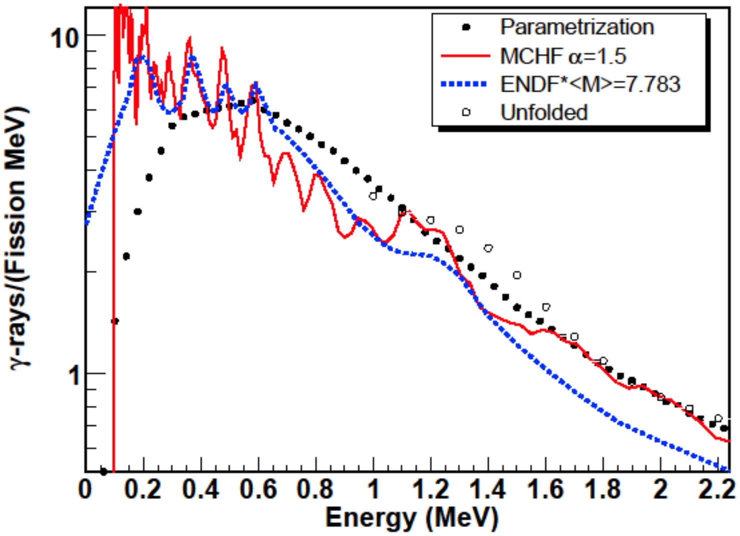
- 150 keV Threshold
- Unfolded – 1D from Chyzh, Phys. Rev. C 87 034620 (2014)
- MCHF from Stetcu, LA-UR-14-23128 (α = 1.5)

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



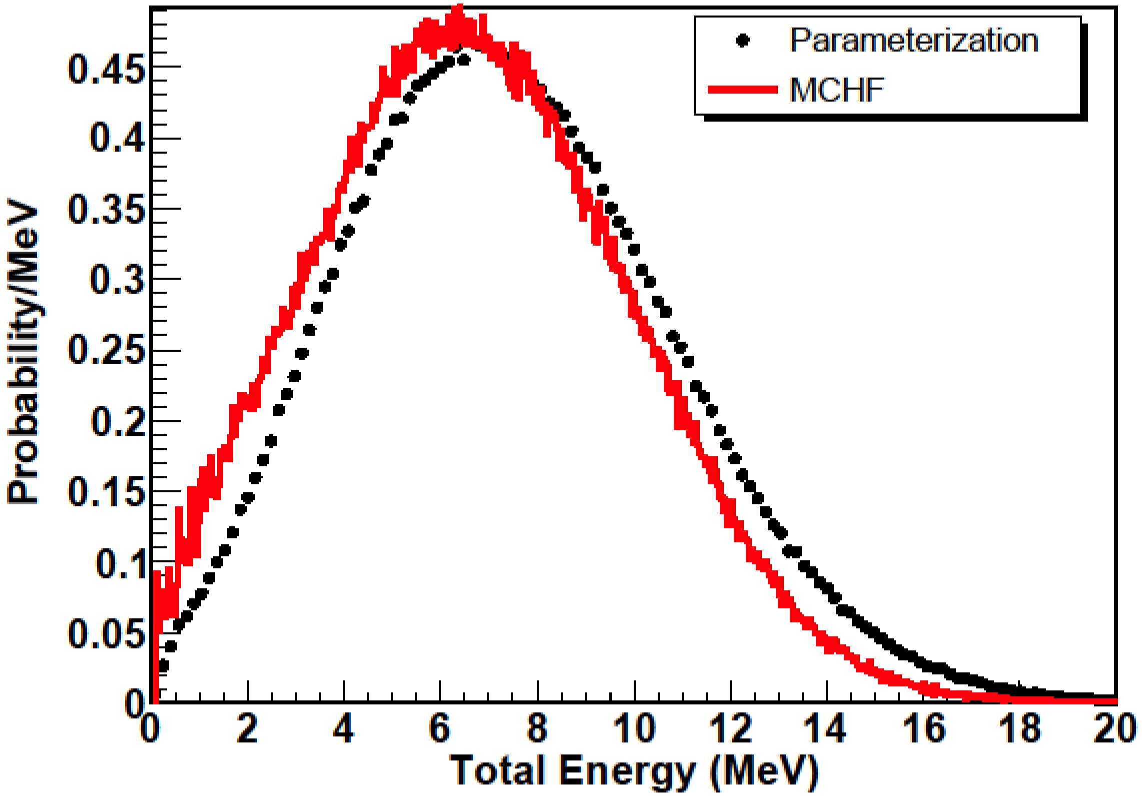
Measured (before response correction) cluster energy for Cluster Multiplicities 4,8,12





- 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)

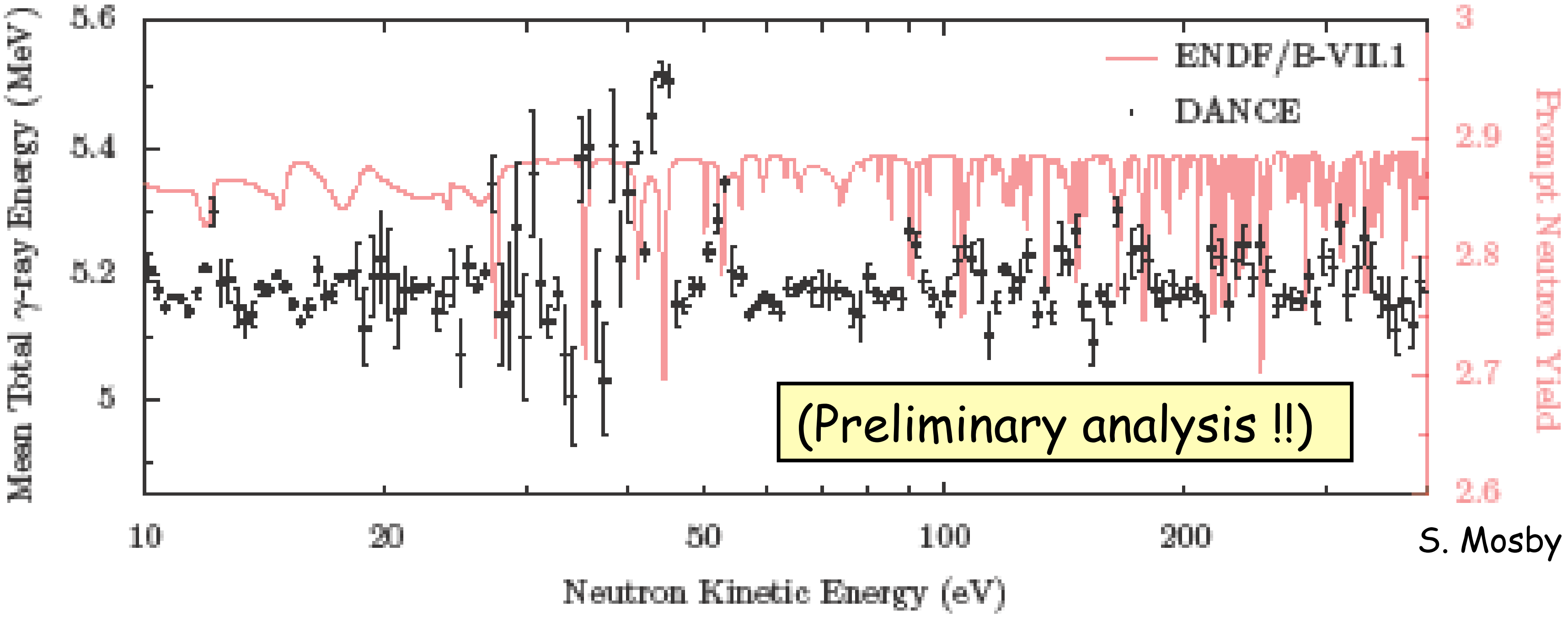
^{239}Pu(n,f) Average γ Multiplicity and $E_{tot,\gamma}$

	<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)



(Preliminary analysis !!)

S. Mosby

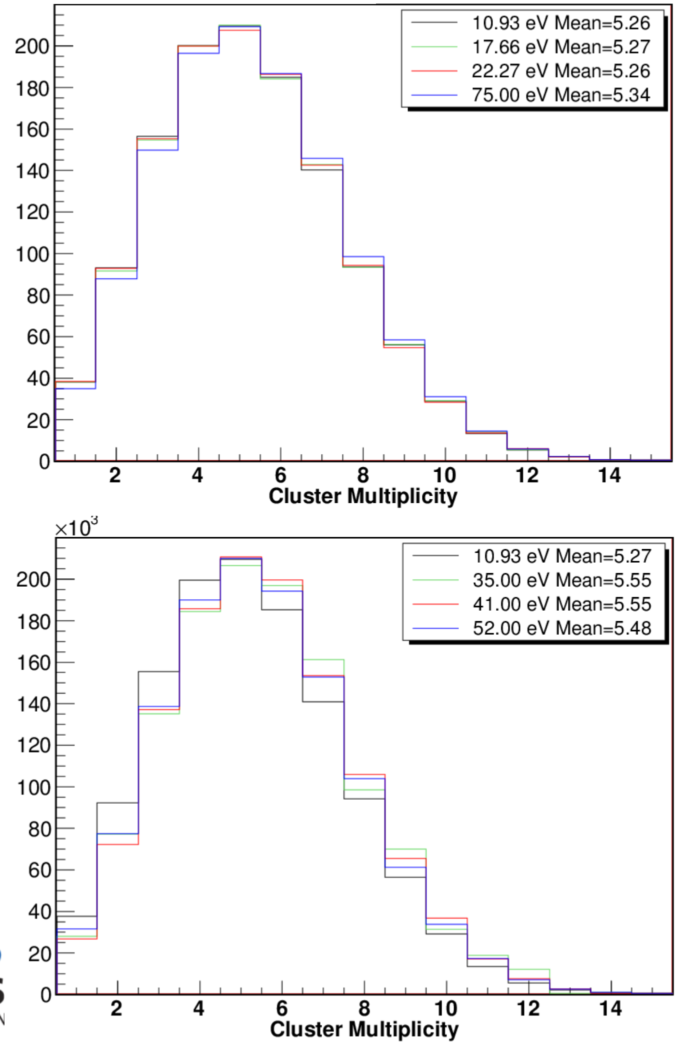
$\langle \nu_n \rangle$ dips at several weak L=0 resonances

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

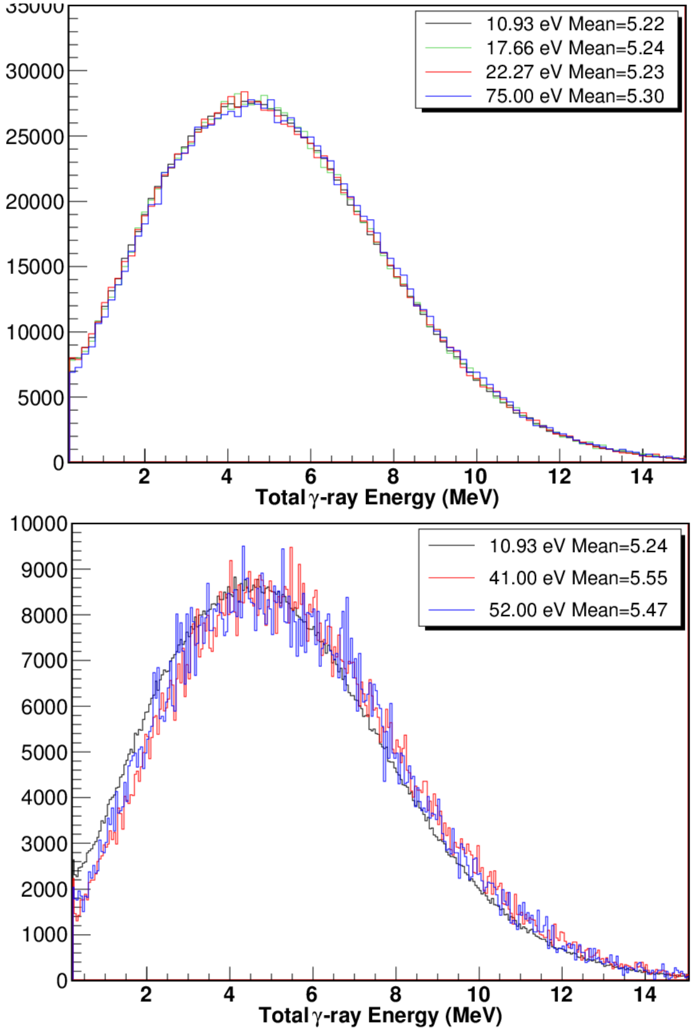
Neutron multiplicity measurements
E. Fort, Nucl. Sci. Eng. **99**, 375 (1988)
Gamma multiplicity measurements
Yu. Ryabov Nucl. Phys. **A216**, 395 (1973)

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!"**

Extra Slides

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

<Eγ> Changes with neutron energy

- Fragment mass distribution changes with neutron energy
- Madland formula reflects changing products and J (excitation)
- $E_{tot} = 6.741 + 0.117\ T_n(\mathrm{MeV}) - 0.0002\ T_n^2$ 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)

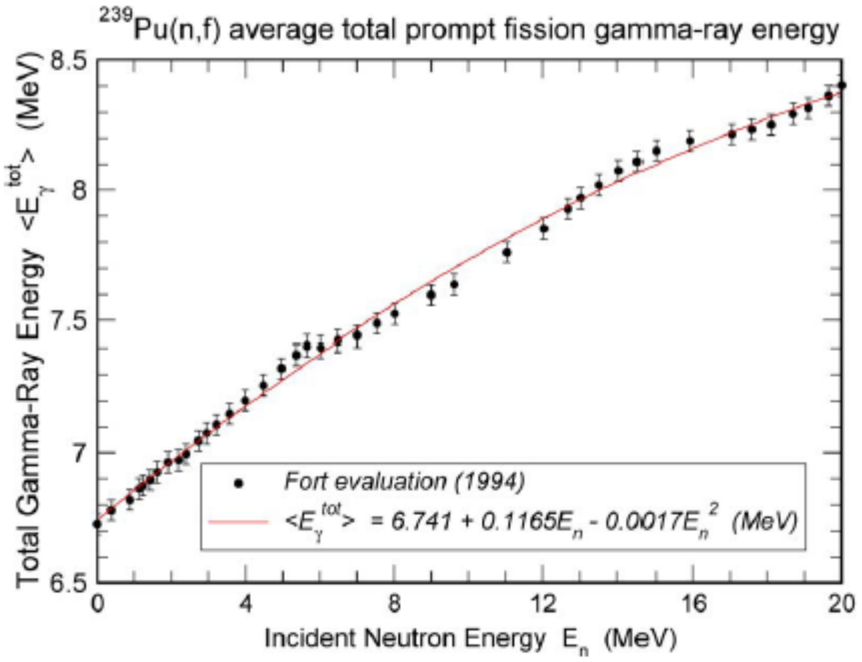
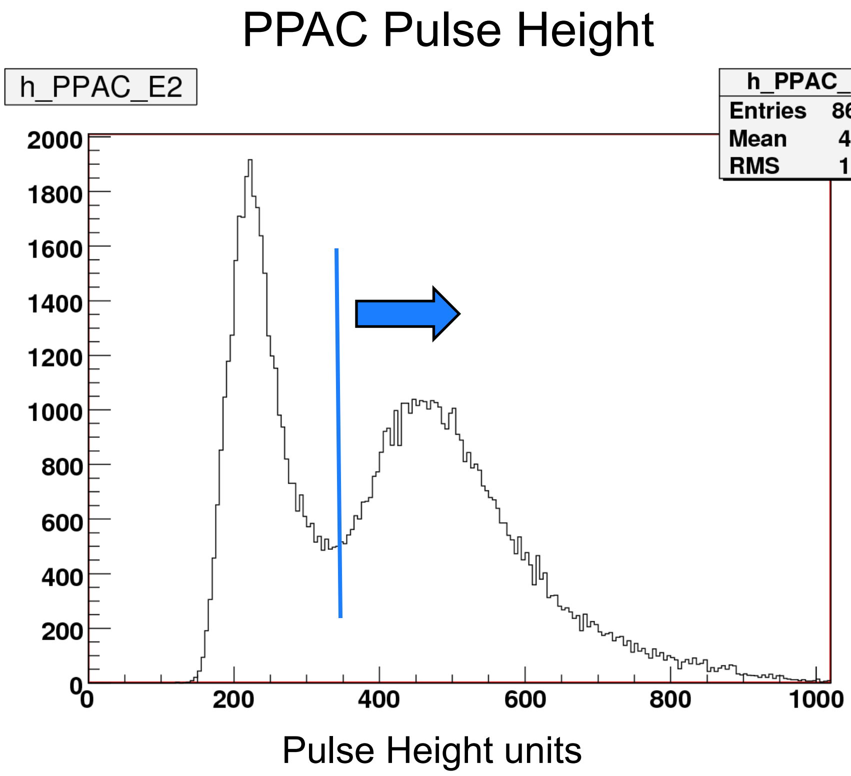
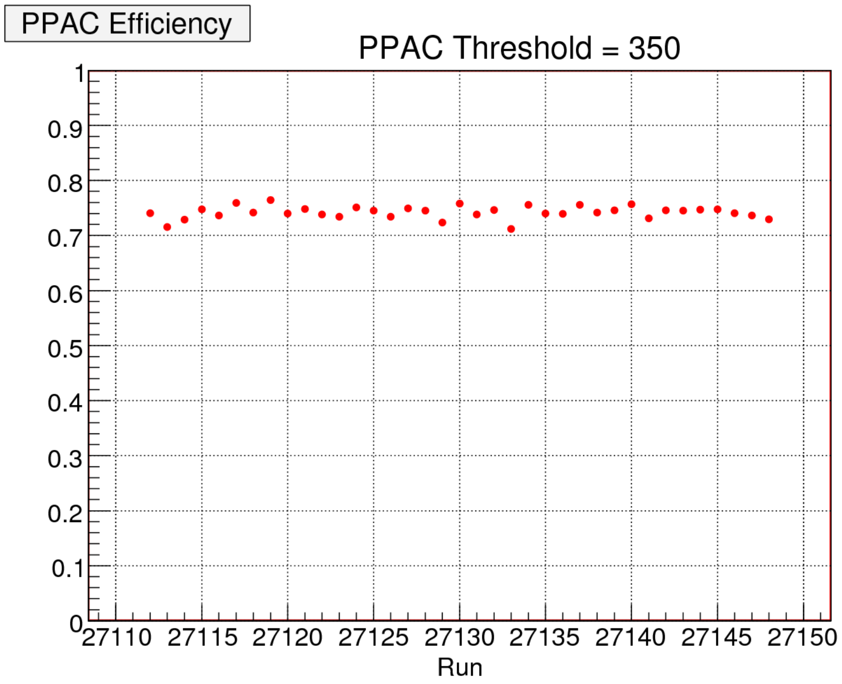


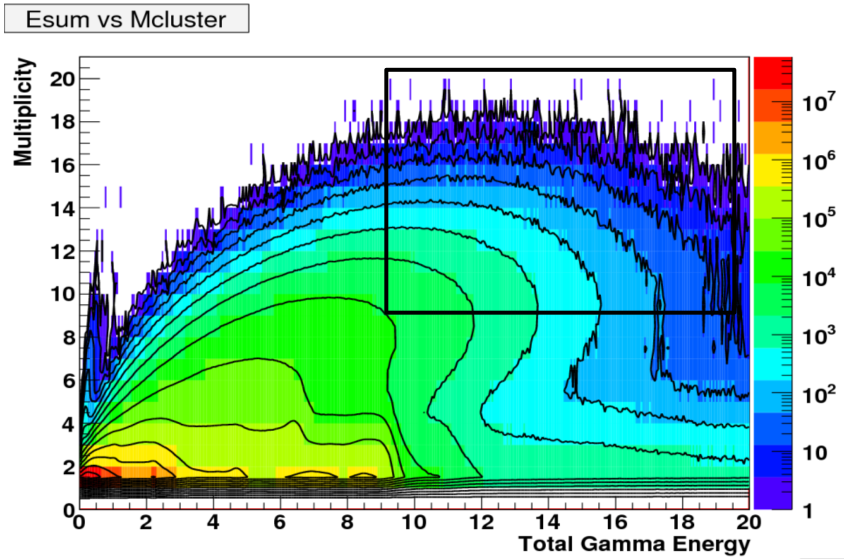
Fig. 16. Average total prompt fission gamma-ray energy $\langle E_\gamma^{tot} \rangle$ for the $n(E_n) + {}^{239}$Pu system.

PPAC Performance

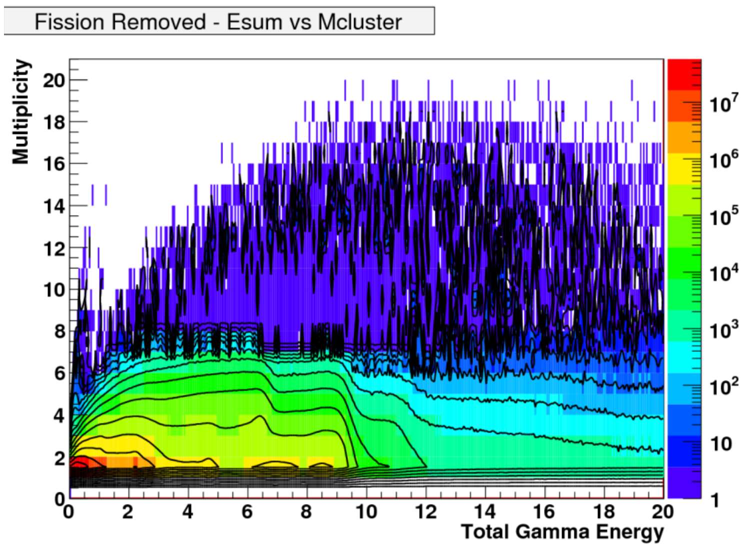


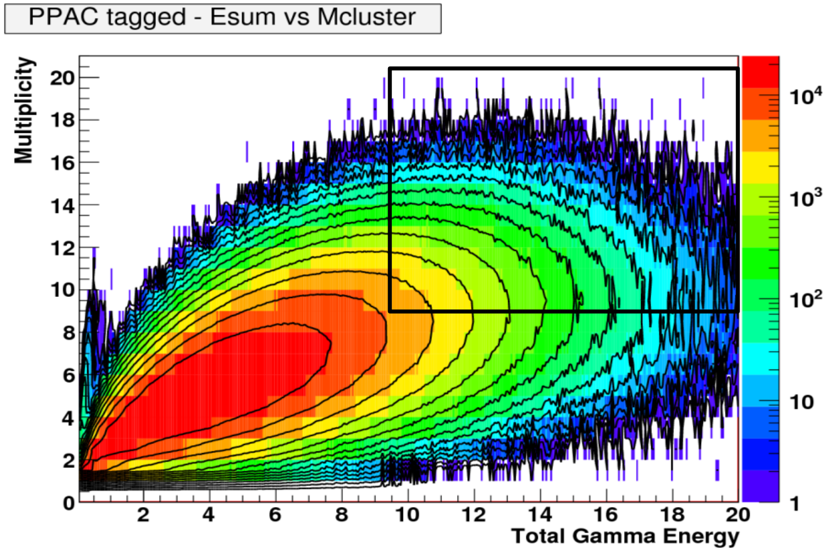


Measure Mult (ν_γ), E_{gam}, E_{tot} and Neutron energy


Esum vs Mcluster

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


Fission Removed - Esum vs Mcluster


PPAC tagged - Esum vs Mcluster

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

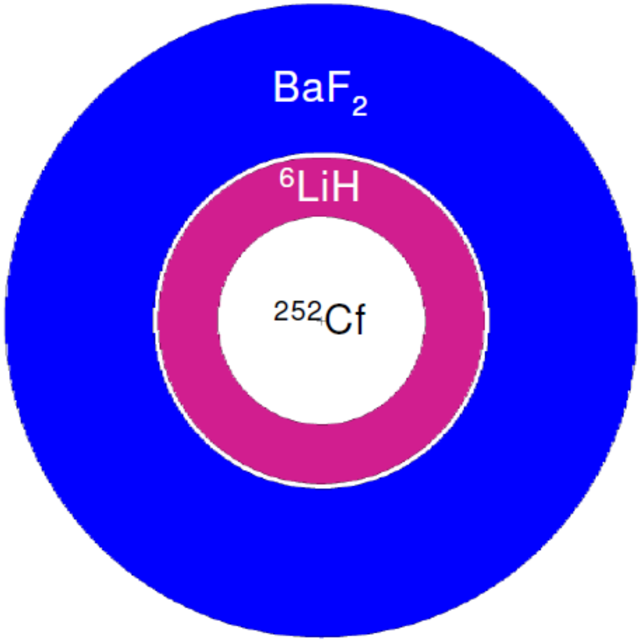
Detour: Fission neutron response

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)

Detector Response Function

Multiplicity

- Sum over two distributions

$$M_\gamma = M_1 + M_2$$

- Spin distributions $P(J) \sim (2J+1)e^{-J(J+1)/B\hat{}2}$

(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" $\rho(\varepsilon) \sim Be^{aEx} = Be^{a(Eo-\varepsilon)}$
  - $P(\varepsilon) \sim \varepsilon^3 e^{-a\varepsilon}$
- Lemaire calculation: $P(\varepsilon) \sim \varepsilon^2 e^{-\beta\varepsilon}$

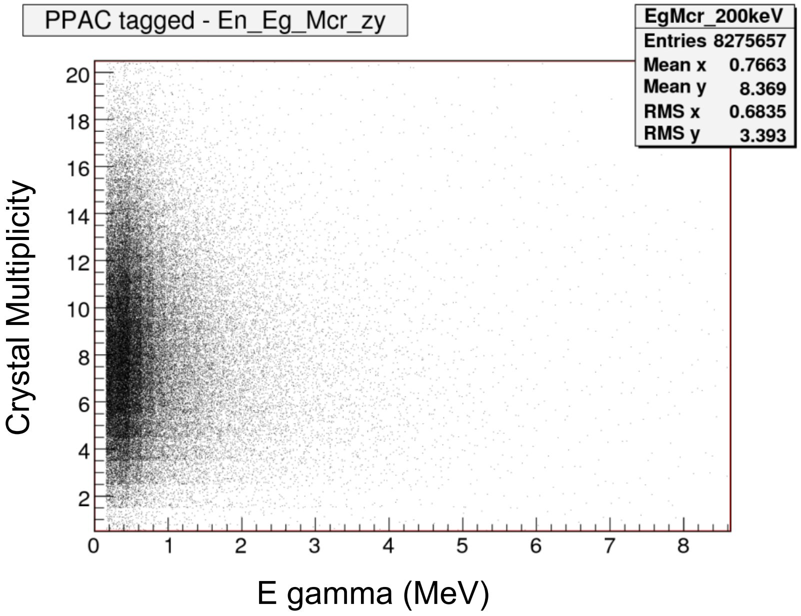
  (S. Lemaire et al., Phys. Rev. C **73**, 014602 (2006))
- Best fit: :

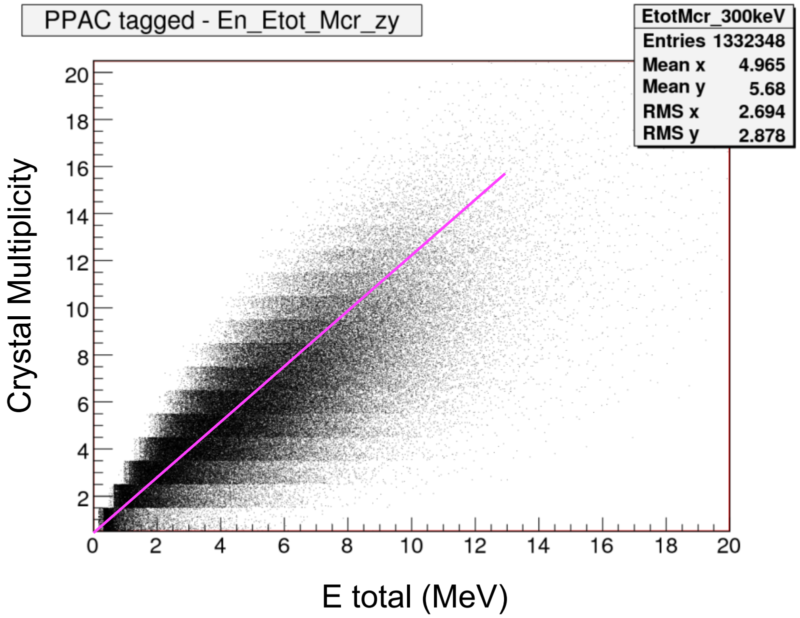
  $P_1(\varepsilon) \sim \varepsilon^2 e^{-(a1+M\gamma b1)\varepsilon}$

  $P_2(\varepsilon) \sim \varepsilon^3 e^{-(a2+M\gamma b2)\varepsilon}$ (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

Multiplicity dependence of Etot and Egam

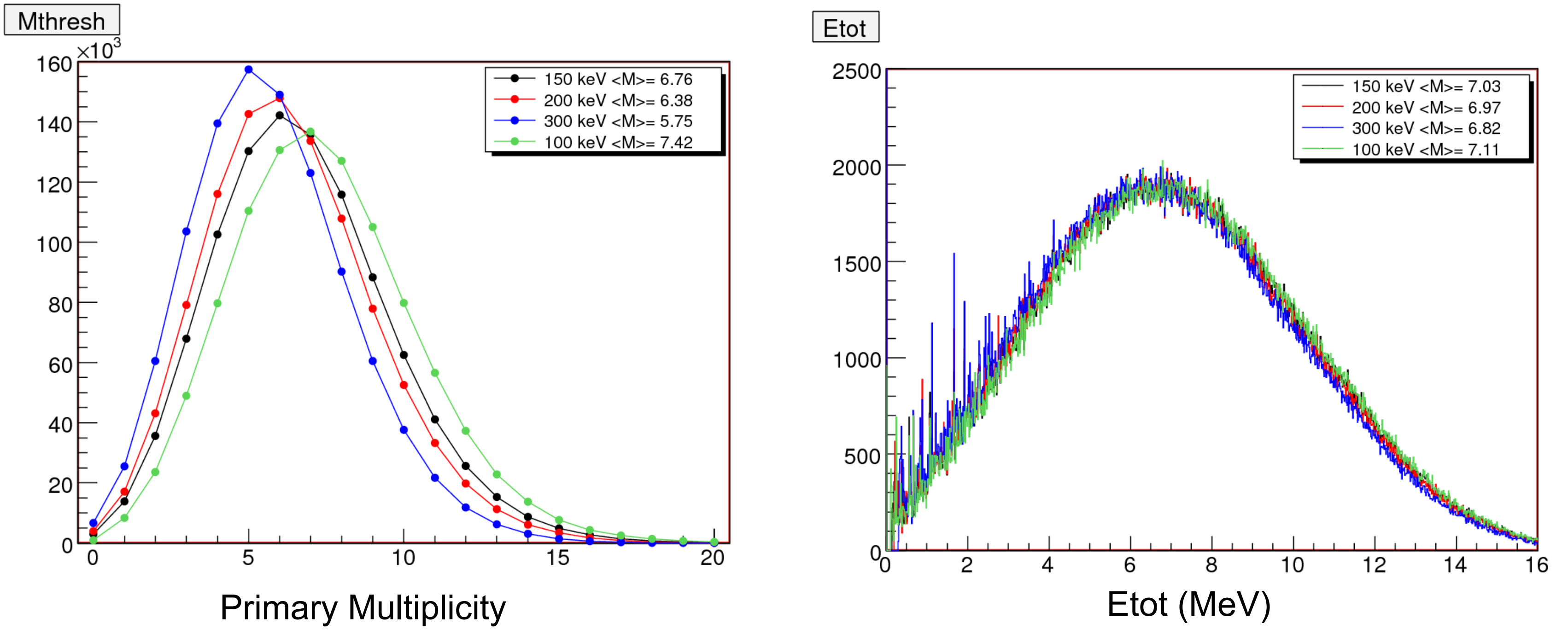




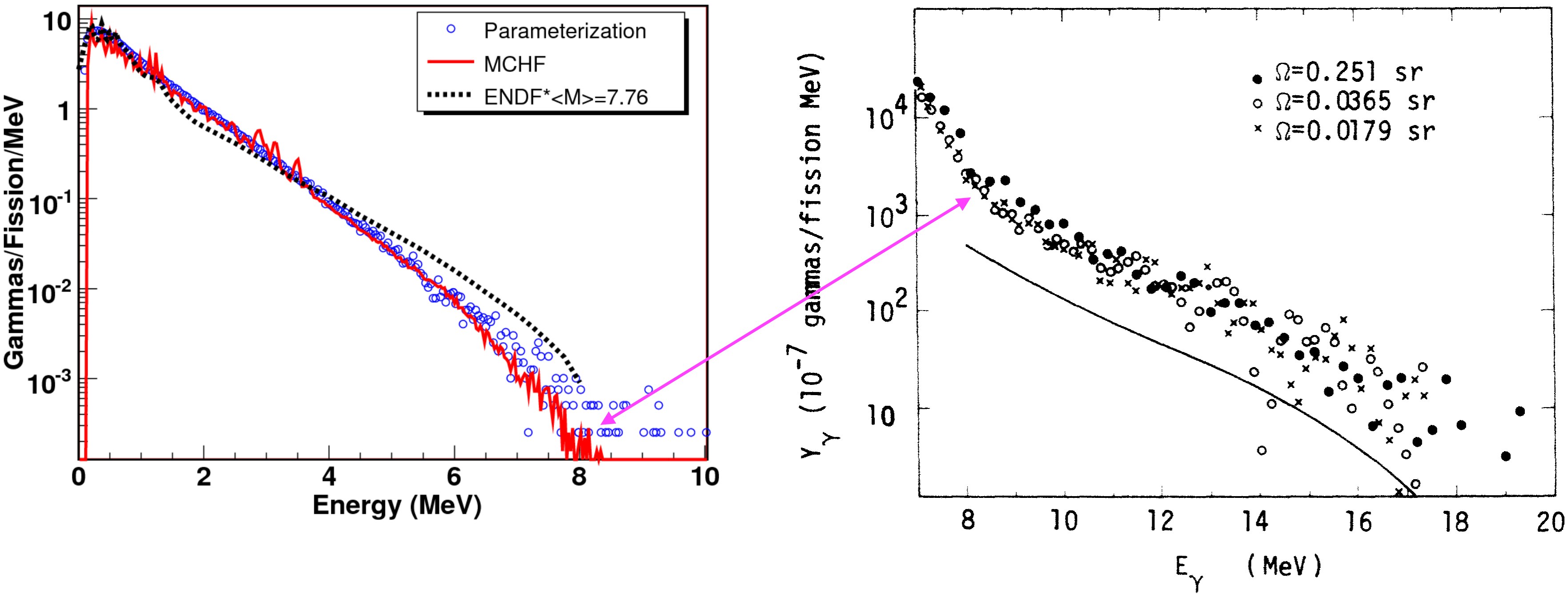
Egam: no strong multiplicity dependence
Etot: Etot roughly proportional to multiplicity

Multiplicity sensitive to DANCE threshold

Multiplicity, total energy with different thresholds determined from MCHF calculation



High-energy gammas



F.S. Dietrich, J.C. Browne, W.J. O'Connell, and M.J. Kay, Phys. Rev. C **10**, 795 (1974)

Changes to Analysis 29-Aug-12

- Parameterization
- Method fitting: "Metropolis Algorithm" + Simulated Annealing
- Parameters fitted for Chi-square minimization
    - 7 spectra (P9VA2MD)
        - Overestimates high-energy effect
    - 4 Spectra (P9VA2ME)
- Uncertainties in <E>, <M>
    - New method of minimization implies cannot easily use previous technique for estimating uncertainties
    - Use % Std of 14 best-fit (lowest Chi2) iterations
    - BUT – use Value of best-fit iteration!
- Effect of threshold on measured multiplicity

Previous Measurements

F. Pleasonton, Nucl. Phys. A**213**, 413 (1973) (Thermal)

V.V. Verbinski, H. Weber, and R.E. Sund, Phys. Rev C **7**,1173 (1973) (Thermal)

Other measurements – incomplete Gamma energy range

- "Unfolding" of measured spectrum critical to results
- Pleasonton also determined fission product ID from Doppler shift.

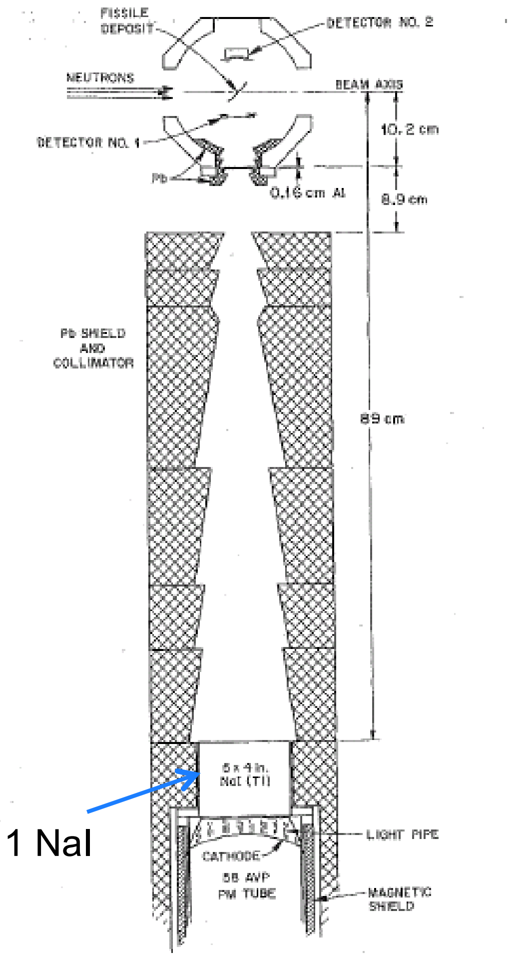
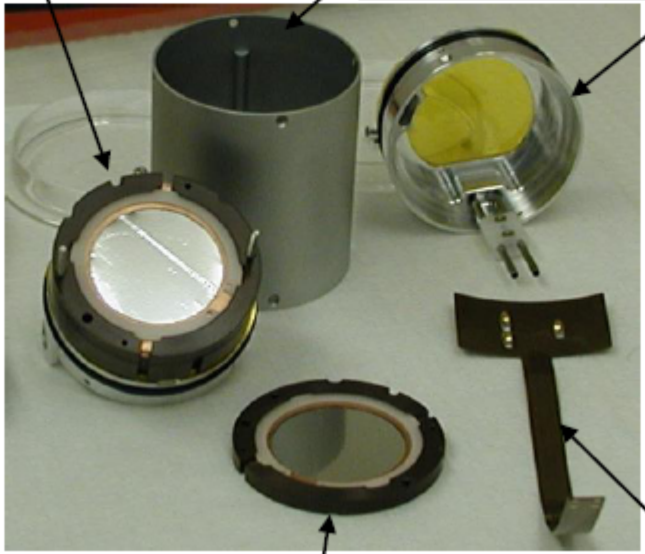


Fig. 1. Scale drawing of the experimental arrangements. The γ-rays were attenuated only by the silicon wafer of detector no. 1, the thin Al window on the vacuum chamber, and the Al cover on the NaI(Tl) crystal. Scattering of the γ-rays into the crystal was minimized by the interior design of the Pb collimator.

PPAC Assembly



Front cover with assembled polyimide rings

Counter container

Back cover with the Kapton window and gas feedthrough

Fig 4. Various counter parts before the final assembling.

Flexible cable for the anode signal transmission

The assembled polyimide ring for one of two anodes