New measurements of the $^{239}Pu(n,\gamma)$ cross section

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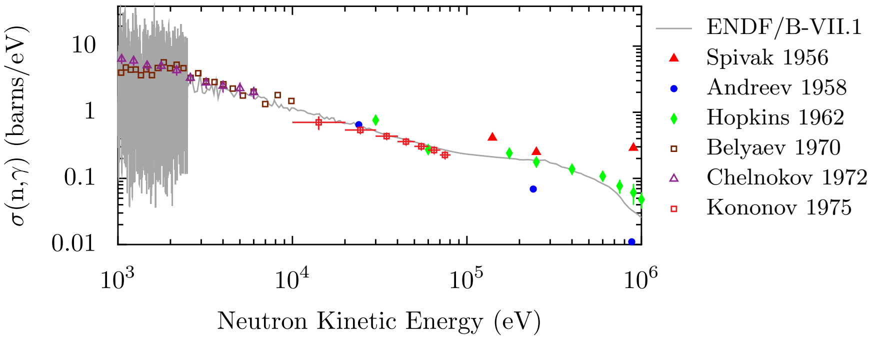
Introduction to ^{239}Pu(n,γ)

- The Advanced Reactor Concepts (ARC) program envisions reactors with a fast neutron spectrum, and a sensitivity study has been performed to characterize what systems need measuring and what uncertainties are needed
- ^{239}Pu(n,γ) is important for defense programs, providing a channel to destroy ^{239}Pu
- Accurate measurements of ^{239}Pu(n,γ) are needed for both programs, particularly above 1 keV
- Desire: 10% or better above 10 keV[1,2]

[1] M. Chadwick, private communication, 2012
[2] G. Aliberti *et al*, Annals of Nuclear Energy, 2006

Previous Measurements

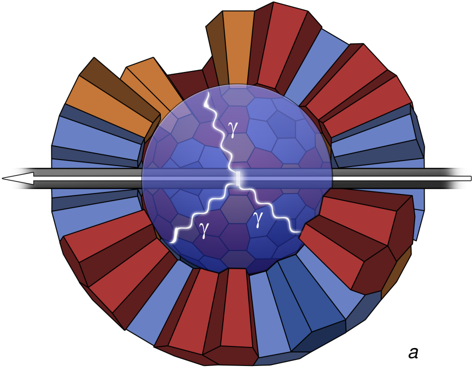


- Data rather sparse and widely spread in keV region
- Old methods have technological limitations - less information to work with
- Exploit new experimental method to improve on these measurements (successfully performed same measurement on ^{235}U)[3]

[3] M. Jandel *et al*, PRL 109, 202506 (2012)

A Detector for Advanced Neutron Capture Experiments

- 160 BaF_2 crystals w/ 4 crystal geometries
- 320 channels of digital DAQ
- 85% Efficiency - calorimeter
- Radioactive / Rare targets (5 μg target in January)
- γ-ray energy / multiplicity information for sopisticated data reduction
- Capture identified by unique Q-value
- ^{6}LiH sphere reduces scattered neutron background



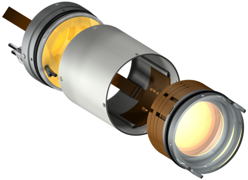
[a]

[a] M. Weigand, 2013

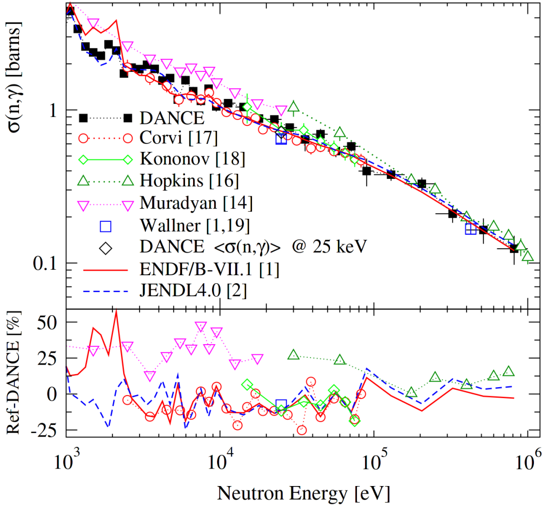
Experimental Details

- Four run conditions used to obtain cross section across all relevant energies:
  1. Thin target (1 mg ^{239}Pu) experiment with fission-tagger
  2. Thick target (50 mg ^{239}Pu) experiment without fission-tagger (3/2013)
  3. ^{208}Pb run to characterize scattered neutron background (3/2013)
  4. ^{239}Pu delayed fission characterization (12/2013)
- Target, flux related uncertainties removed by measuring fission, capture simultaneously (ratio measurement)

- Fission tagging PPAC provided by LLNL collaborators
- Sample serves as central cathode
- Small size accommodates DANCE beamline

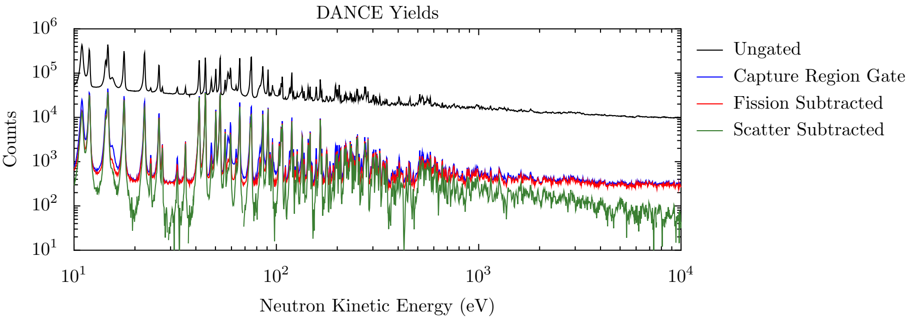


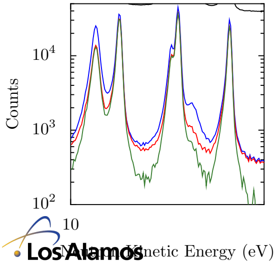
Method works on ^{235}U



- Same basic procedure published for ^{235}U
- M. Jandel *et al*, PRL 109, 202506 (2012)
- Uncertainties:
  - 2.3% at 1 keV
  - 4% at 10 keV
  - 9% at 100 keV
  - 16% at 500 keV
- Expect similar results for ^{239}Pu

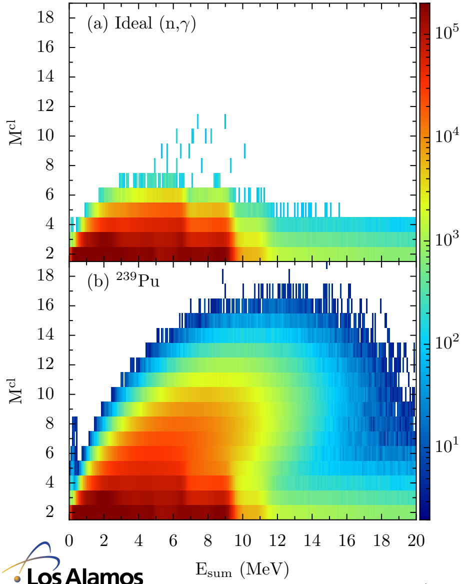
Data Analysis Overview

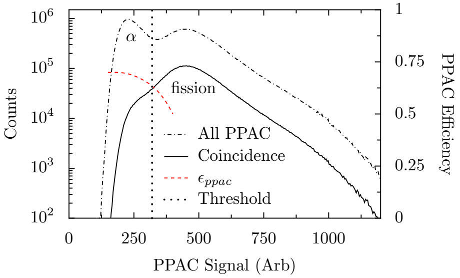




- Fission produces unwanted signals in DANCE
- Scattered neutrons moderate in DANCE crystals, produce light
- Primary challenge for this analysis: appropriately characterize, subtract off these backgrounds
- Old experiments provide limited disentanglement

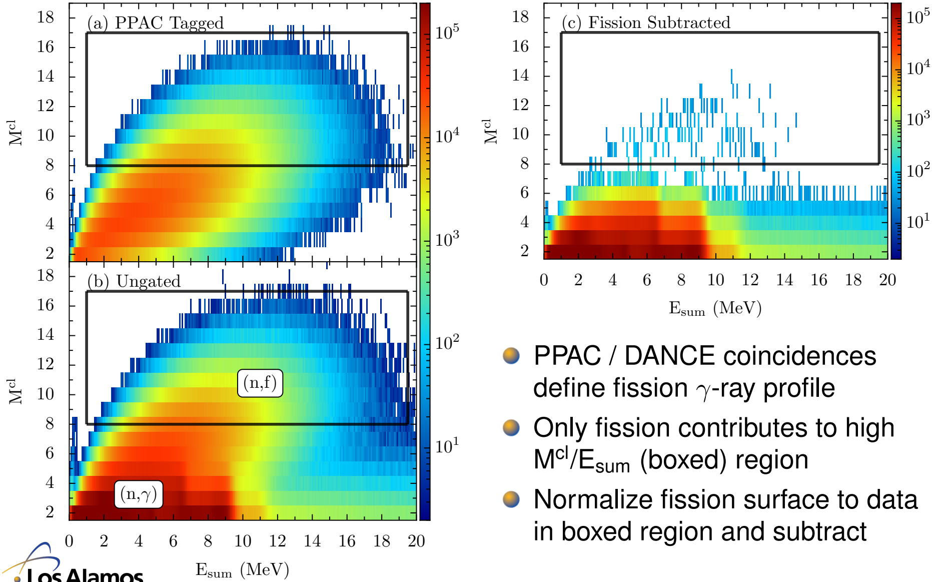
Prompt Fission γ-ray Background / PPAC





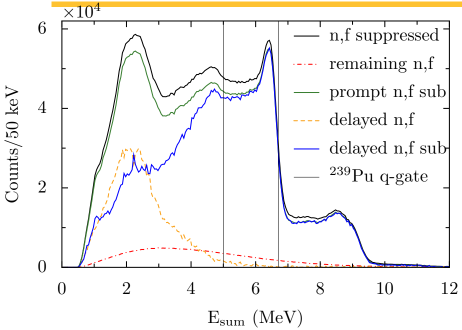
- Ideal (n,γ) spectrum occupies low multiplicity region
- ^{239}Pu γ spectrum contaminated by fission
- PPAC: tag fission by charged particles
- PPAC coinc with DANCE improves confidence

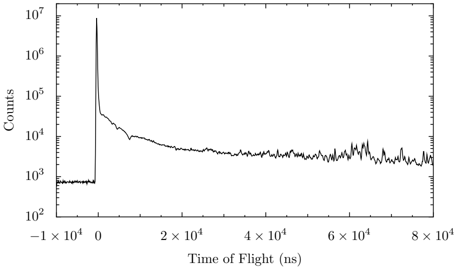
Prompt Fission γ-ray Background Subtraction



- PPAC / DANCE coincidences define fission γ-ray profile
- Only fission contributes to high M^{cl}/E_{sum} (boxed) region
- Normalize fission surface to data in boxed region and subtract

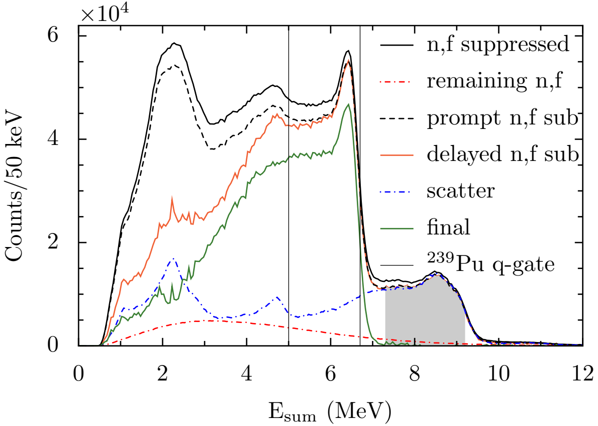
Delayed Fission γ-ray Background Subtraction





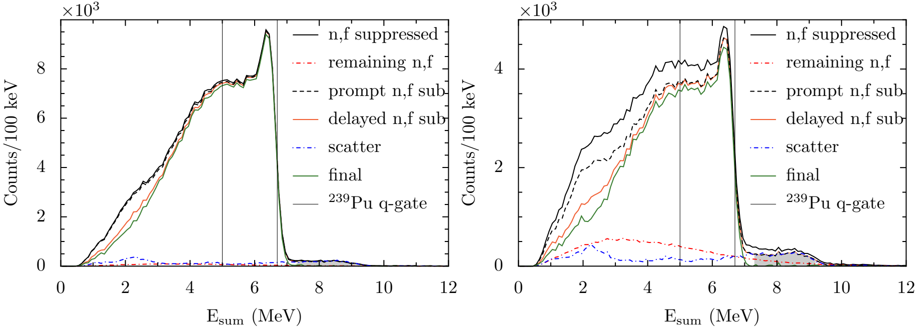
- Long lived fission products grow in and cause background that is constant in time
- "Look-back window" lets us see events prior to beam T0
- Background environment thus characterized - dominates spectrum below 3 MeV

Scattered Background



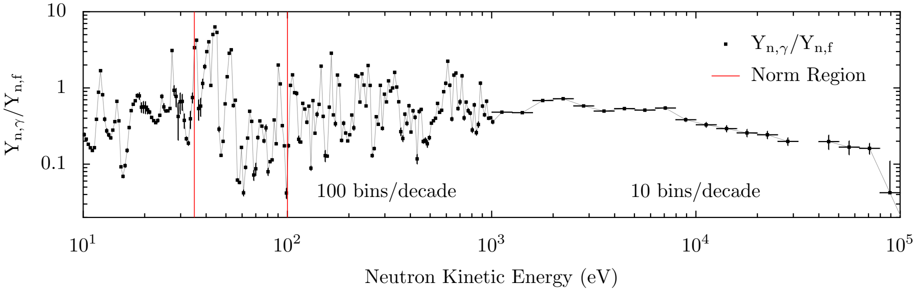
- Data taken with blank PPAC taken (no ^{239}Pu) to characterize scattering background - method depends on calorimetry
- Characterized as function of energy, normalized in high E_{sum} region
- Calorimetry allows gate on ^{239}Pu(n,γ) Q-value to select capture events

Subtraction Results On Resonance



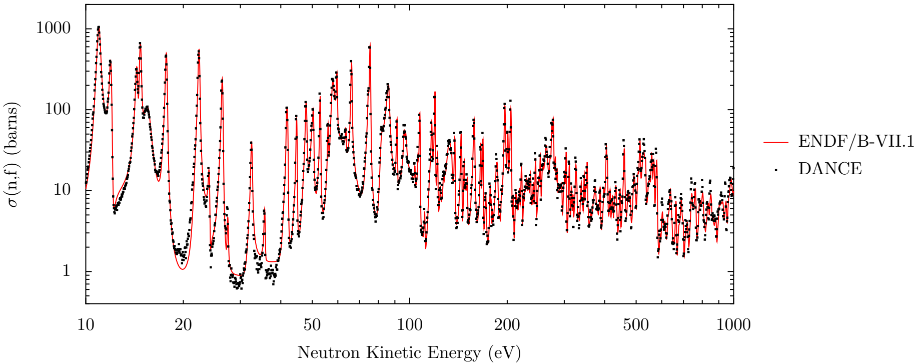
- On resolved resonances, backgrounds are fairly small but can change significantly
- Background subtraction as function of energy **important**

Cross Section Calculation



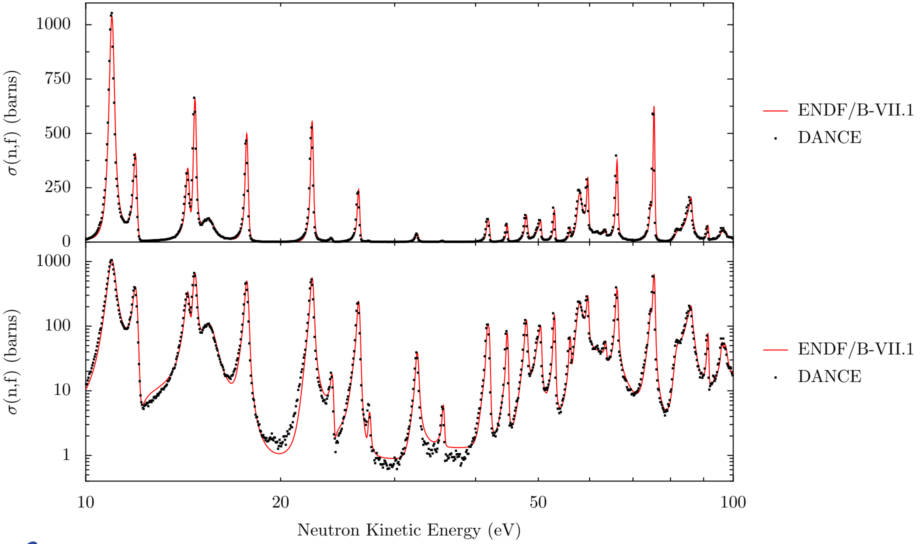
- $\sigma(\mathrm{n},\gamma) = A_{\mathrm{n}\gamma}\sigma_{\mathrm{n}f}\frac{Y_{cap}}{Y_{\mathrm{n,f}}}$ (ratio to fission measurement)
- $A_{\mathrm{n}\gamma} = \frac{I_{n\gamma}}{I_{nf}}\frac{Y_{integral\ \mathrm{n,f}}}{Y_{integral\ \mathrm{n},\gamma}}$ (scaling factor from ENDF integral cross sections)
- Signal/Background limits the 1 mg sample dataset to 1 keV
- Target Mass $\sim$ count rate - put 50 mg sample in Spring 2013

Fission Cross Section Tests

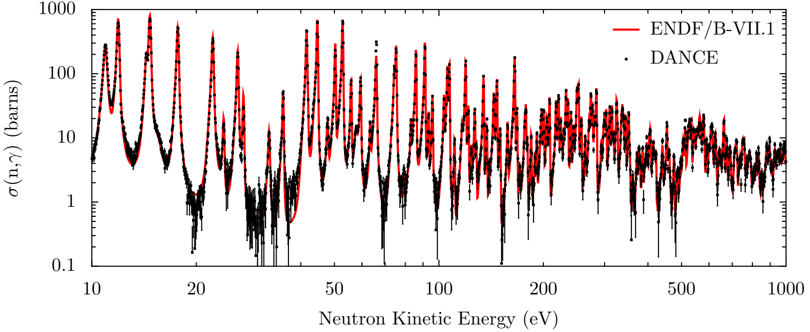


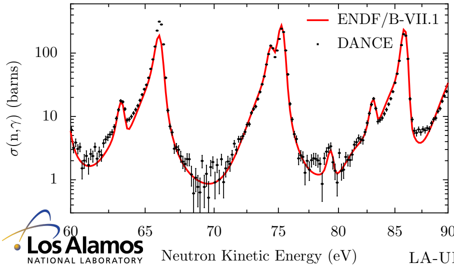
- Comparison to fission cross section serves as a check
- Good agreement with evaluations lends confidence to analysis

Fission 10 - 100 eV



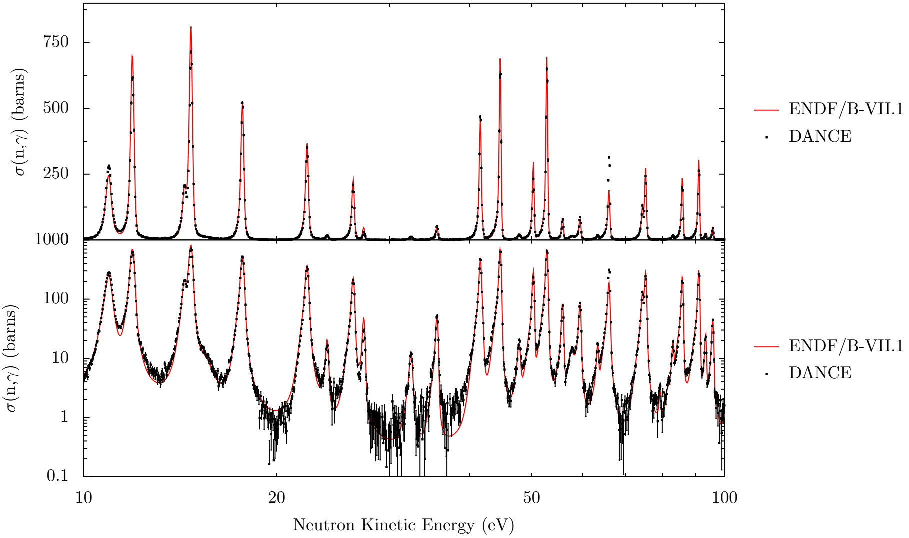
1 mg Sample Results



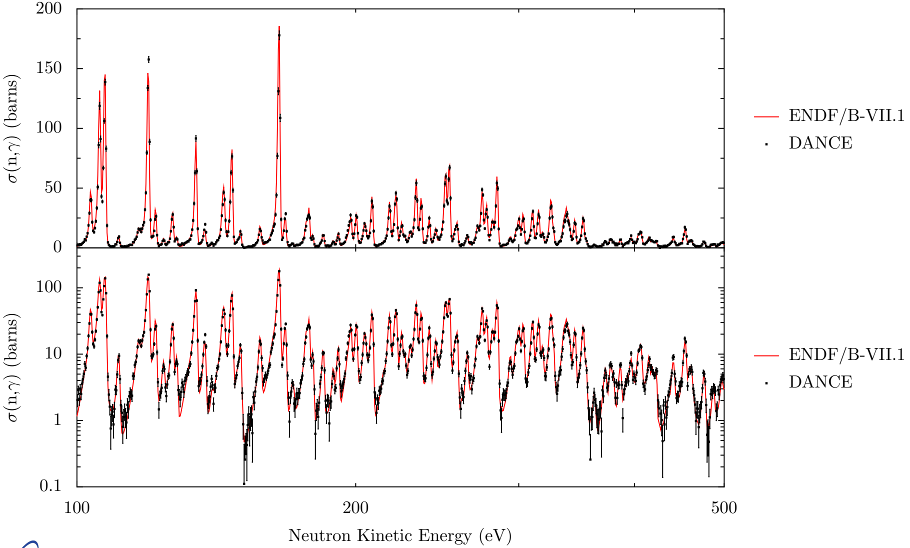


- Overall structure is similar to existing evaluation
- Individual resonance parameters appear to be different in some cases
- S. Mosby *et al*, PRC 89, 034610 (2014)

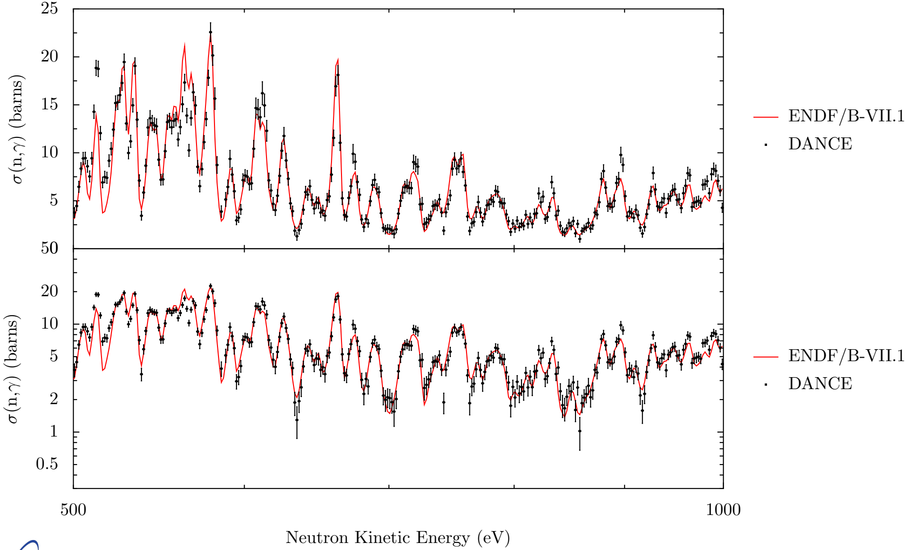
Capture 10 - 100 eV



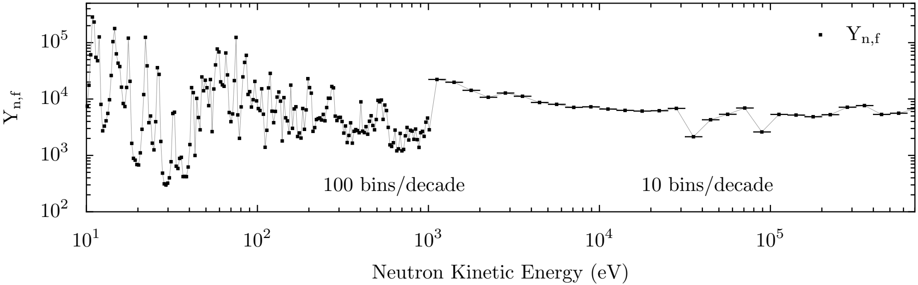
Capture 100 - 500 eV

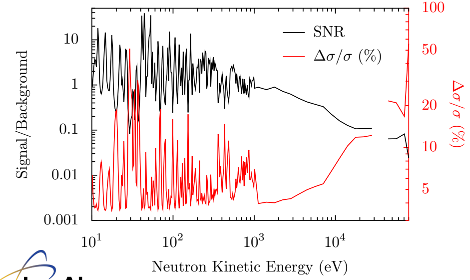


Capture 500 - 1000 eV



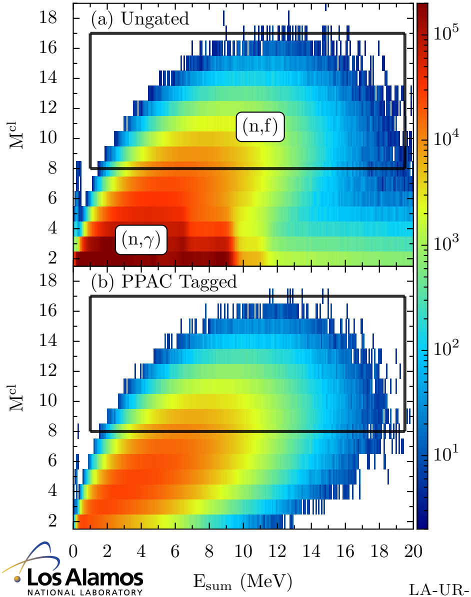
Extension to Higher Energies





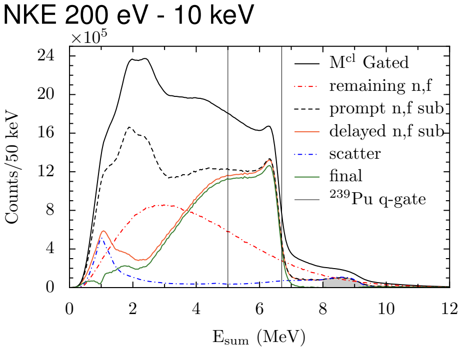
- ^{239}Pu(n,f) PPAC coinc data extend to much higher energy than statistics-limited (n,γ)
- Thick sample precludes use of PPAC - different treatment
- (n,f) γ lineshape is known from thin-sample PPAC coinc and can be used for subtraction

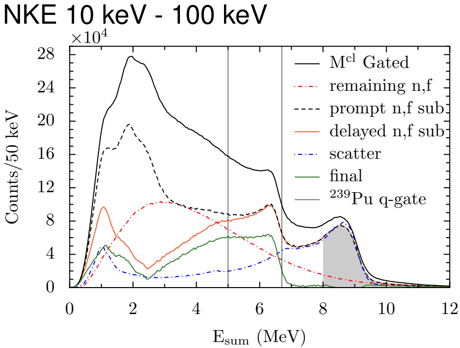
Thick Target (n,f) Treatment



- High M^{cl} region only has (n,f)
- With (n,f) lineshape known from present data, thick-target background can be calculated as $N_{fis} = \epsilon_{fis} Y_{mcl>8}$
- ϵ_{fis} is "fission efficiency" scaling factor from high M^{cl} region to Q-value gate

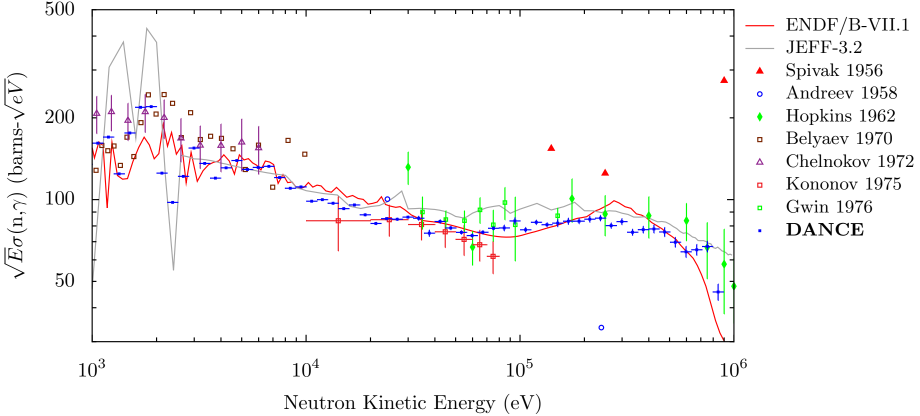
Background at High Neutron Energy





- Signal-to-noise crosses through 1 in 10 keV - 100 keV energy decade
- Fission is dominant background - **absolutely critical** to get this right

Thick Target Preliminary Results



- Closer consistency with ENDF at highest energies
- Looking into details of the keV region

Conclusions

- Improved measurements of ^{239}Pu(n,γ) cross section are important to nuclear energy and defense programs
- The DANCE calorimeter setup is capable of making this measurement and disentangling the relevant backgrounds - demonstrated for ^{235}U
- Improved statistics from a thick target, combined with the fission γ spectrum measured from the PPAC coinc data extend this measurement into the 100s of keV

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J. M. O'Donnell (LANSCE-NS)
J. Ullmann (LANSCE-NS)
C. Y. Wu (LLNL)

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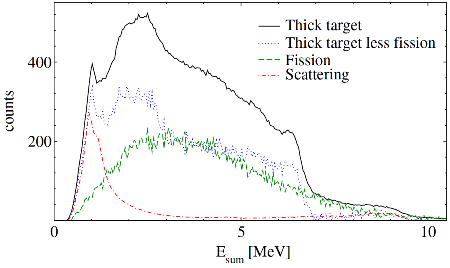
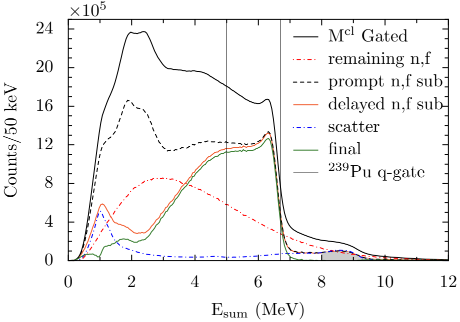
Background Comparison with ^{235}U Publication



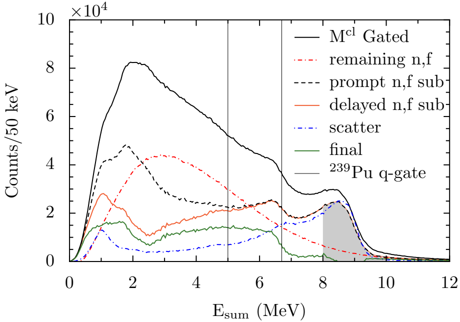


- ^{235}U 200 eV - 10 keV
- 26 mg/cm^2 sample
- Q-gate 5.7 - 6.7 MeV

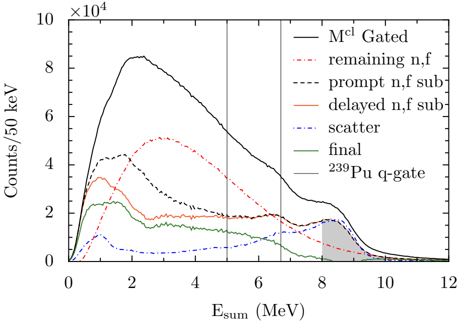
- ^{239}Pu 200 eV - 10 keV
- 44 mg/cm^2 sample
- ∼200 mg/cm^2 Ni plate
- Q-gate 5.0 - 6.7 MeV

The 100 keV to 1 MeV Region *(Very Rough)*

NKE 100 keV - 300 keV

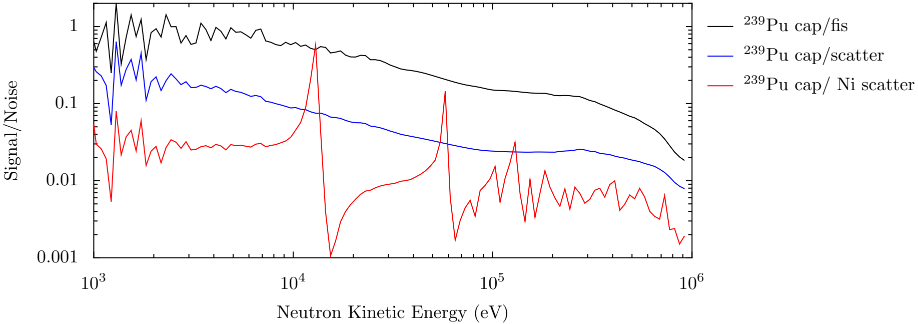


NKE 300 keV - 1 MeV



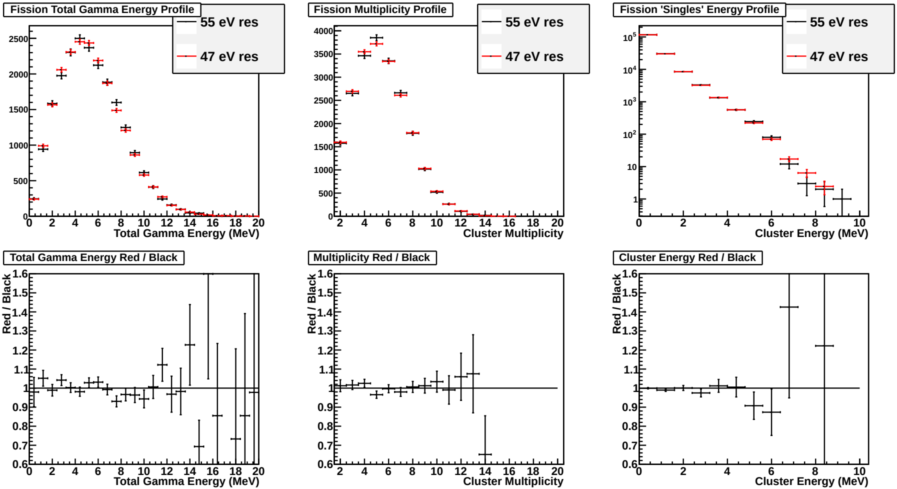
- We have counts all the way out!
- Incident neutron energy smears Q-value - shift Q-gate accordingly
- Fission dominates backgrounds - **absolutely critical** to get this right!
  - In progress: matching of detector thresholds to improve bg lineshapes
- ...fission rejection w/ massive targets would help (NEUANCE?)
- $\sim$200mg/cm^2 Ni backing is next most significant background

Why Nickel Plating Matters (for 50 mg target)



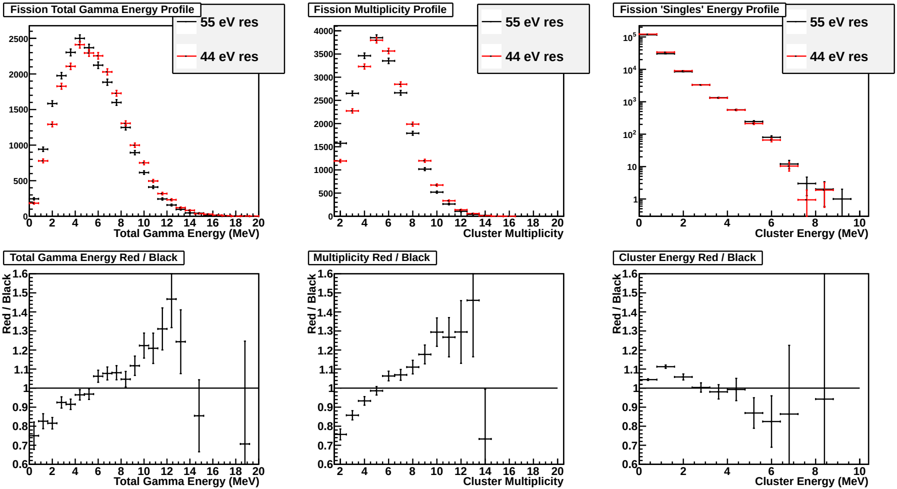
- Ratios calculated from cross section folded with illuminated mass
- No adjustment for relative detection efficiency / gating
- Ni plating had 5x areal density of ^{239}Pu deposit
- Neutrons scattering from Ni adds a lot to the scatter background

Interesting Observations (1)



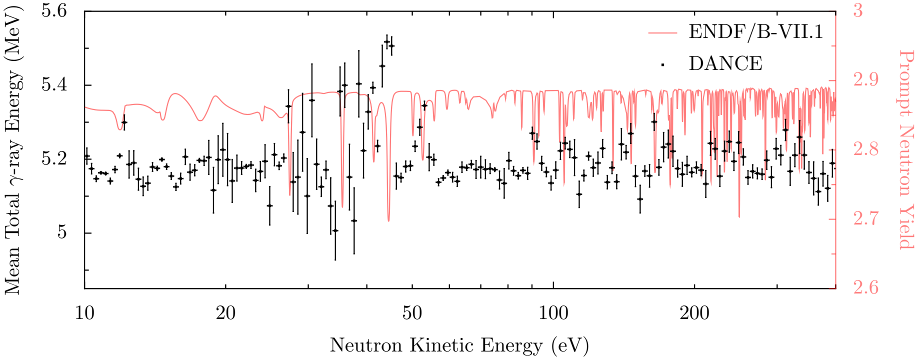
- This is what you'd expect for PFGS...

Interesting Observations (2)



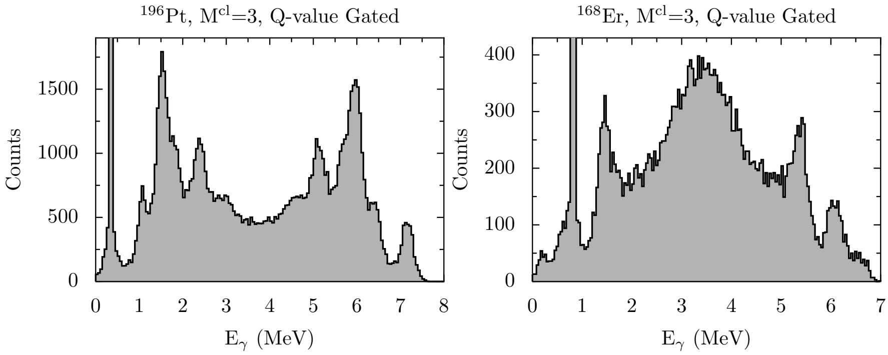
- ...and this is what we get!

Interesting Observations (3)



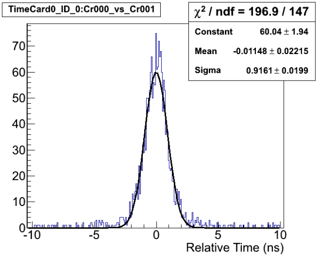
- ...and the behavior is anti-correlated with neutron emission
- Evidence for ^{239}Pu(n,γf) e.g. Lynn 1965?
- Qualitative behavior reported by Shackleton in 1972
- Further analysis, conversations with T-2 underway...

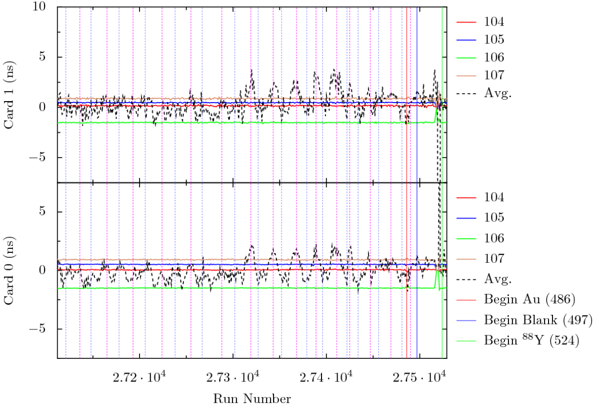
γ-ray Cascade Information



- γ-ray cascades resulting from capture can provide structure information
- Segmentation, efficiency of DANCE allows us to see this structure
- Different nuclei are different!

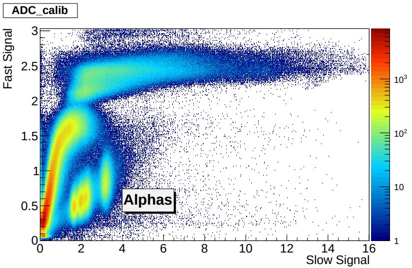
DANCE Timing Properties

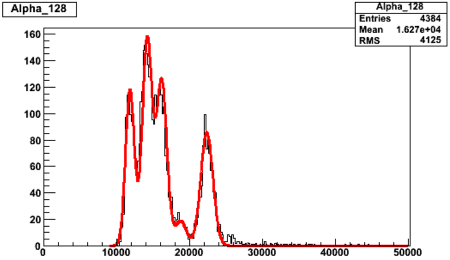




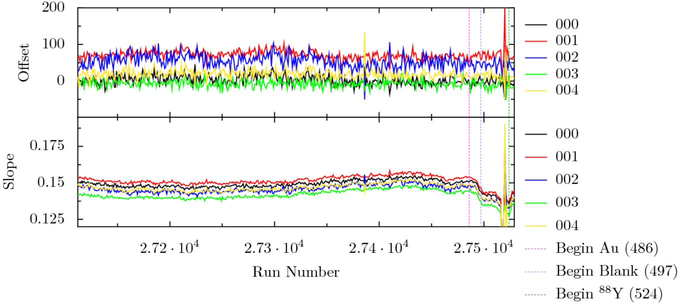
- Crystal-to-crystal timing allows narrow coinc gates ($<$10 ns)
- PPAC requires wider gate - 12 ns
- Temp dependence of digitizers in vs. time plots (red=18:30, blue=04:00)

Energy Calibrations / Drift Corrections

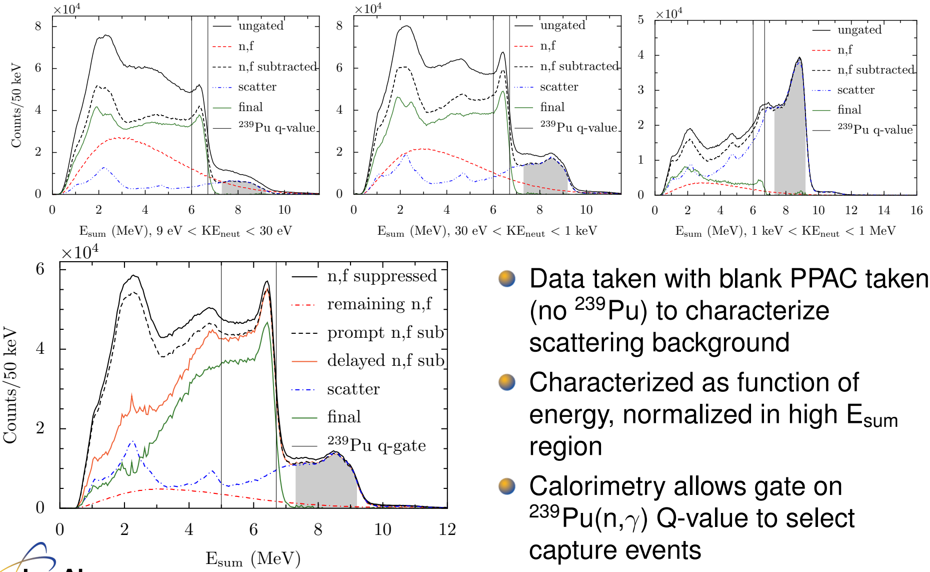




- Natural α background seen with PSD
- Use for energy calibration
- Run by run gain tracking



Scattered Background



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