

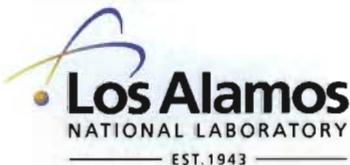
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*Title:* Two Detector Arrays for Fast Neutrons at LANSCE

*Author(s):* R. C. Haight, H. Y. Lee, T. N. Taddeucci, J. M. O'Donnell, B. A. Perdue, N. Fotiades, M. Devlin, J. L. Ullmann, A. Laptev, T. Bredeweg, M. Jandel, R. O Nelson, S. A. Wender, C.-Y. Wu, E. Kwan, A. Chyzh, R. Henderson, J. Gostic

*Intended for:* Presentation at the International Workshop on Fast Neutron Detectors and Applications, Ein Gedi, Israel  
November 6-11, 2011



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Vu-graphs to be presented at the  
International Workshop on Fast Neutron Detectors and Applications  
Ein Gedi, Israel  
November 6-11, 2011

## Two Detector Arrays for Fast Neutrons at LANSCE

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N. Fotiades, M. Devlin, J. L. Ullmann, A. Laptev, T. Bredeweg,  
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Livermore, CA 94551, USA

### ABSTRACT

The neutron spectrum from neutron-induced fission needs to be known in designing new fast reactors, predicting criticality for safety analyses, and developing techniques for global security application. The experimental data base of fission neutron spectra is very incomplete and most present evaluated libraries are based on the approach of the Los Alamos Model. To validate these models and to provide improved data for applications, a program is underway to measure the fission neutron spectrum for a wide range of incident neutron energies using the spallation source of fast neutrons at the Weapons Neutron Research (WNR) facility at the Los Alamos Neutron Science Center (LANSCE). In a double time-of-flight experiment, fission neutrons are detected by arrays of neutron detectors to increase the solid angle and also to investigate possible angular dependence of the fission neutrons. The challenge is to measure the spectrum from low energies, down to 100 keV or so, to energies over 10 MeV, where the evaporation-like spectrum decreases by 3 orders of magnitude from its peak around 1 MeV. For these measurements, we are developing two arrays of neutron detectors, one based on liquid organic scintillators and the other on  $^{6}\text{Li}$ -glass detectors. The range of fission neutrons detected by organic liquid scintillators extends from about 600 keV to well over 10 MeV, with the lower limit being defined by the limit of pulse-shape discrimination. The  $^{6}\text{Li}$ -glass detectors have a range from very low energies to about 1 MeV, where their efficiency then becomes small. Various considerations and tests are in progress to understand the important contributing factors to designing these two arrays and they include selection and characterization of photomultiplier tubes (PM), the performance of relatively thin (1.25 cm)  $^{6}\text{Li}$ -glass scintillators on 12.5 cm diameter PM tubes, use of 17.5 cm diameter liquid scintillators with 12.5 cm PM tubes, measurements of detector efficiencies with tagged neutrons from the WNR/LANSCE neutron beam, and efficiency calibration with  $^{252}\text{Cf}$  spontaneous fission neutrons. In addition, significant modeling is underway to assess contributions from room-return neutrons and detector cross-talk. A new flight path is being constructed to reduce the effect of room-return neutrons. A data acquisition system based on wave-form digitizers is being developed to extract the maximum amount of information from the signals with the minimum amount of dead time. Design considerations and test results will be presented in this talk.

## Two Detector Arrays for Fast Neutrons at LANSCE

R. C. Haight, H. Y. Lee, T. N. Taddeucci, J. M. O'Donnell, B. A. Perdue,  
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Lawrence Livermore National Laboratory

International Workshop on Fast Neutron Detectors  
and Applications – FNDA 2011

Ein Gedi, Israel  
November 6-10, 2011



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### Outline – two detector arrays at LANSCE

- Motivation
  - Spectra of Neutrons from neutron-induced fission
  - Accuracy required
- Measurements at LANSCE
  - Double Time-of-flight
  - Previous results (brief)
- Detector arrays
  - Liquid scintillator
  - $^{6}\text{Li}$ -glass
- Detector calibration approaches
  - $^{252}\text{Cf}$
  - Ohio U
  - Neutron Tagging
  - Other methods ("known" monoenergetic sources)
- Factors influencing accuracy
  - Time of flight
  - Distance

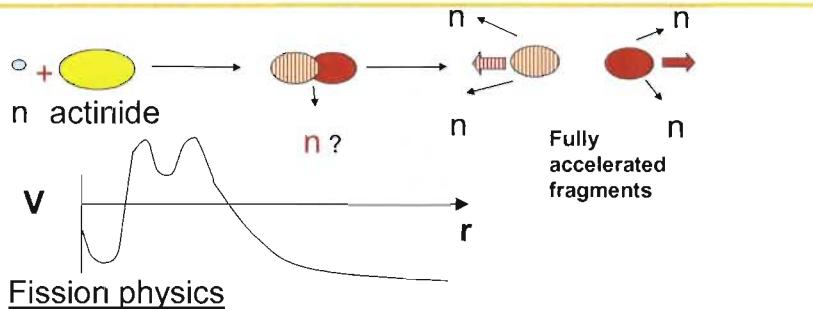


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## Fission physics model is used to predict neutron spectrum from fission



### Fission physics

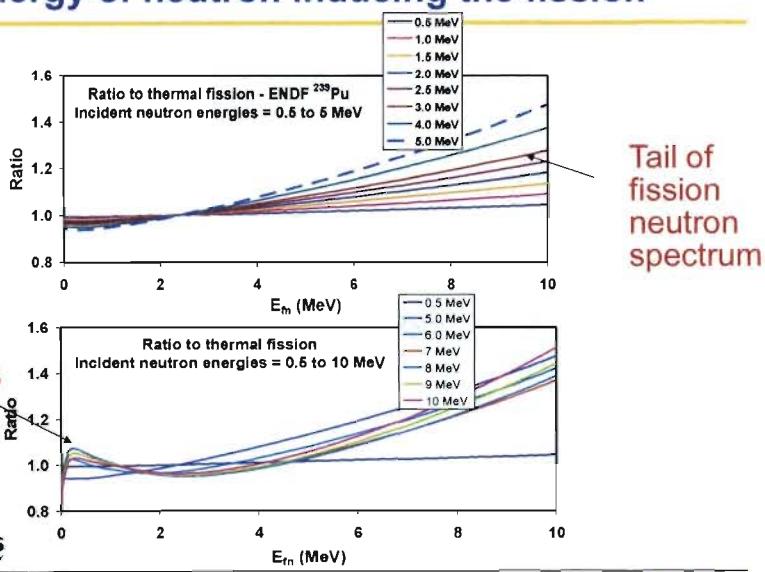
- Mass splits ( $Z, A$ )
- Total kinetic energy
- Nuclear level densities
- Temperatures (excitation energies) of fragments
- Spectrum changes with energy of incident neutron:
  - Temperature of fragments; distribution of  $Z, A$  of fragments, etc.
- No “Pre-scission” neutrons



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## Fission neutron spectrum is predicted to change with energy of neutron inducing the fission



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## Present theoretical approaches to fission

- Los Alamos model – improvement by Monte Carlo sampling of fission products
- “Event-by-event” simulation
- Quantum mechanical tunneling of many-particle wave function (Hartree Fock Bogoliubov)



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## Data for incident neutrons $> 100$ keV are very sparse

$^{239}\text{Pu}(n,f)$   
– fission neutrons

Data from Staples  
(U.Mass-Lowell)

Solid curve is  
from Madland  
and Nix

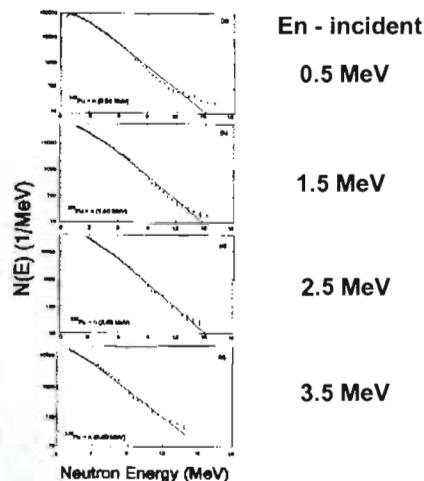


Fig. 7. The relative fission neutron yield for  $^{239}\text{Pu}$  in the nuclear energy ranges of (a) 0.30, (b) 1.39, (c)



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## For better visibility of the 4 orders of magnitude:

- Take ratio of data to Maxwellian

$$N(E) \sim \sqrt{E} \exp(-E/T)$$

with  $T = 1.30 \text{ MeV}$

This form is taken for convenience only !



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## Present data show significant discrepancies both at low emitted energies and at $E_{n'} > 6 \text{ MeV}$

Low energy neutron emission data are inconsistent

Data for  $E_{n-in} > \text{thermal}$  and  $E_{n-out} > 0.5 \text{ MeV}$  are inconsistent

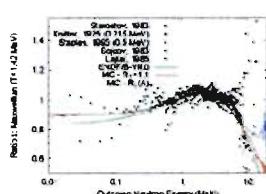
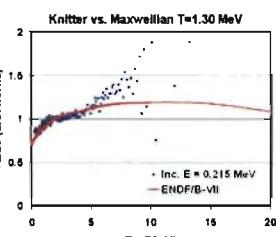
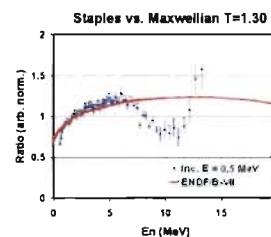


FIG. 11. (Color online) Same as Fig. 10 but plotted as ratio to Maxwellian at temperature  $T=1.30 \text{ MeV}$ .



Advanced Monte Carlo Modeling of Prompt Fission Neutrons for Thermal and Fast Neutron-Induced Fission Reaction on Pu-239, P. Talou, B. Becker, T. Kawano, M. B. Chadwick, and Y. Danon, submitted to PRC

Accuracy desired: ~ 5% in shape of spectrum from 100 keV to 12 MeV



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## How we have done it



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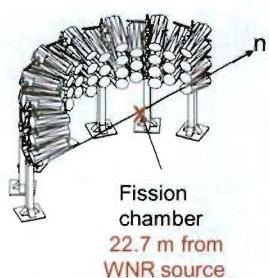
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**LANSCE experiments: made with CEA physicists and CEA fission chambers; data taken and analyzed at LANL and later by CEA**

FIGARO ( $n, xn+\gamma$ )

- 20 liquid scintillator neutron detectors
- 2 gamma-ray detectors



Double time-of-flight experiment



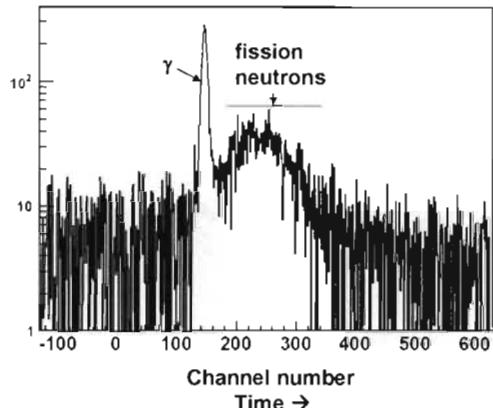
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## Time difference spectrum from fission shows neutron spectrum



Important features:

- Timing to separate gamma rays from fast neutrons
- Backgrounds

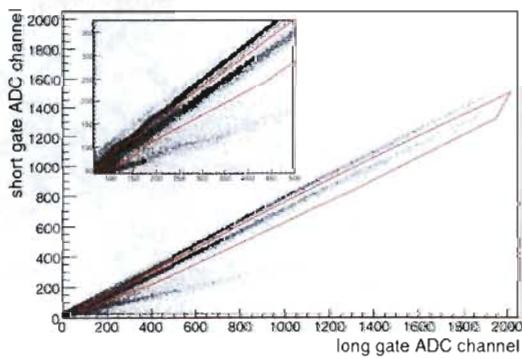
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## Liquid scintillators are used to separate neutrons from gamma rays

- Pulse shape discrimination – current integration for short and long component of pulse



Limitation:  
 $E_n > 0.7$  MeV

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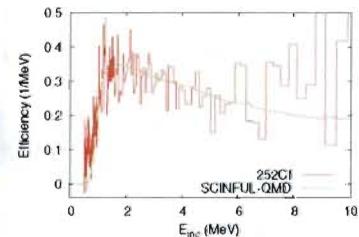
## Detector efficiencies were determined by calculation normalized to $^{252}\text{Cf}$ standard source

PROMPT FISSION NEUTRON SPECTRA FROM FISSION ...

FIG. 3. (Color online) The FIGARO array, consisting of 20 EJ301 neutron detectors and a BaF<sub>2</sub> gamma-ray detector.

*n-p* interactions as opposed to lower LET from fast electrons produced by  $\alpha$  rays to produce signals of different shape

PHYSICAL REVIEW C 83, 034604 (2011)

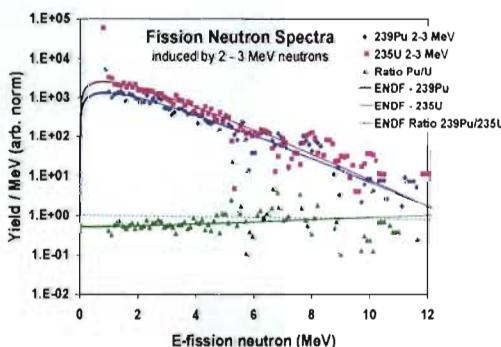
FIG. 4. (Color online) An example of efficiency of one EJ301 neutron detector. The histogram is the result of the  $^{252}\text{Cf}$  experiment, and the dotted curve is the SCINFUL-QMD calculation. Statistical uncertainties account for fluctuations in the experimental data.

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## We measured the shapes of fission neutron spectra and ratios of spectra for $^{235}\text{U}(\text{n},\text{f})$ and $^{239}\text{Pu}(\text{n},\text{f})$

Incident  $E_n = 2$  to 3 MeV



Spectra show no significant difference from ENDF in 1-7 MeV region

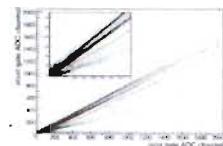
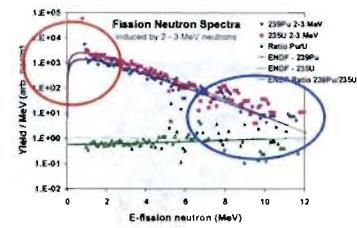


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## Program of fission neutron output measurements is designed to improve these data

- Measure fission neutrons below 0.7 MeV
  - Need better n-gamma discrimination
- Measure fission neutrons better above 8 MeV
  - Better timing on fission chamber
  - More efficient neutron detectors (larger solid angle for detection)
- Reduce background from accidental coincidences
  - Reduce mass of backing foils in fission chamber



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## What we are doing

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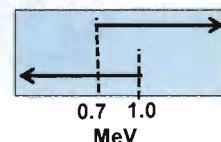
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## Present approach

- Parallel plate avalanche detector (LLNL)
  - Much better timing than CEA fission ion chambers
  - Less mass → less neutron scattering
- Two types of neutron detectors
  - For  $E_n > 0.7$  MeV -- EJ309 liquid scintillator; 7" diameter 2" thick (bigger and somewhat better than those used previously)
    - pulse shape discriminations (PSD) (n vs. gamma)
    - larger array (~ 50 detectors)
  - For  $E_n < 1.0$  MeV –  $^6\text{Li}$ - glass
- Gamma-ray output (LLNL) with liquid scintillator
- Data acquisition (DAQ) with waveform digitizers
  - More information on waveform – PSD
- WNR source
  - New flight path (15-degrees – left) in new building
  - Reduced room scattering



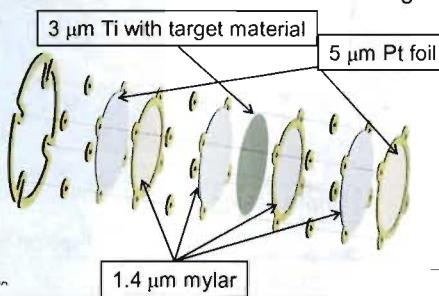
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## Fission counter design and fabrication

1. The counter has 20 parallel-plate avalanche counters, combined into 10 cells. Each cell has ~ 10 mg target material deposited over 4 cm diameter spot on both sides of a 3  $\mu\text{m}$  thick Ti foil.
2. The target foil is covered by the 1.4  $\mu\text{m}$  aluminized mylar glued to the G-10 ring, which constitutes the cathode.
3. Two anodes are for each cell and made of 1.4  $\mu\text{m}$  aluminized mylar and 5  $\mu\text{m}$  Pt foil.
4. The output signal is processed by a newly LLNL designed amplifier with a gain of 300 and bandwidth of 500 MHz to measure the timing and pulse height simultaneously.

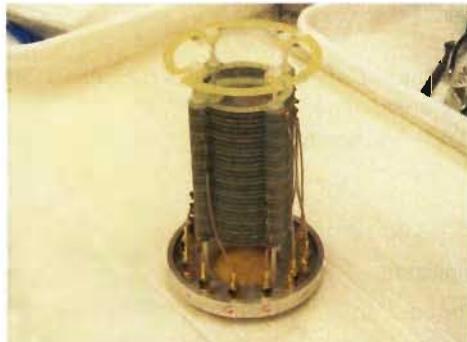


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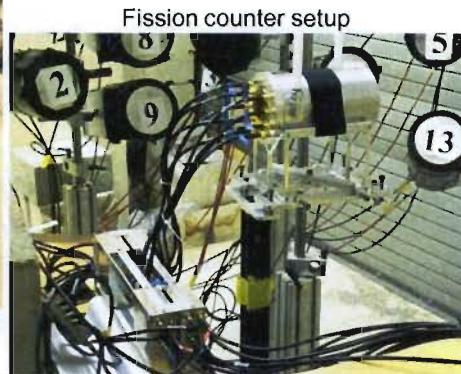
## Fission counter assembly

- Two  $^{235}\text{U}$  and one  $^{252}\text{Cf}$  fission counters have been assembled in addition to a blank one. All function well.



Fully assembled  $^{235}\text{U}$  counter with a total mass of 113 mg

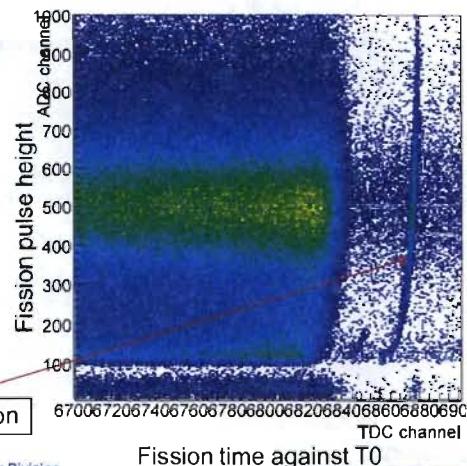
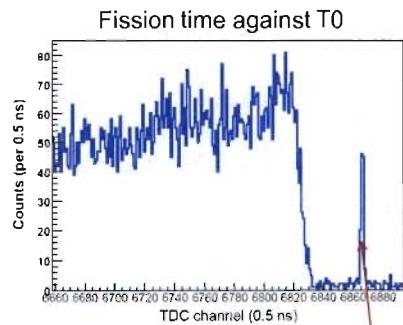
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## Fission counter performance

- Excellent timing  $\sim 1$  ns is achieved and pulse height information is adequate for the  $^{235}\text{U}$  counter.

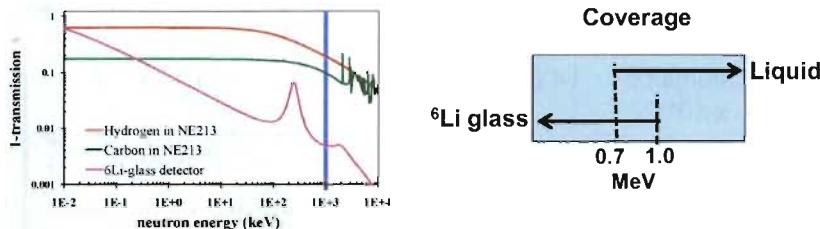


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## Neutron detectors

- Liquid organic scintillators with PSD for fission neutrons > 0.7 MeV
  - 17.8 cm diameter x 5 cm thick scintillator on 12.5 cm diameter PMT
- $^{6}\text{Li}$ -glass scintillators for fission neutrons < 1.0 MeV
  - 10 cm diameter x 0.18 cm thick GS20 on 12.5 cm PMT



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## Liquid organic scintillators

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## Choice of scintillators for detecting neutrons with $E_n > 0.7$ MeV

- Liquid organic scintillators → pulse shape discrimination

Scintillator/ Property	EJ301 = BC501A = NE213	EJ309 = BC599-17	EJ309/EJ301
Density (g/cc)	0.874	0.959	1.097
H/C ratio	1.212	1.25	1.031
H atoms/cc -- $\times 10^{22}$	4.82	5.46	1.133
C atoms/cc - $\times 10^{22}$	3.98	4.37	1.098
Decay time (short) ns	3.2	3.5	



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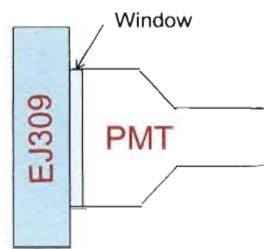
## Liquid Scintillators

17.8 cm diameter x 5 cm thick



Scionix

Elien



Q: Is light collection a problem?



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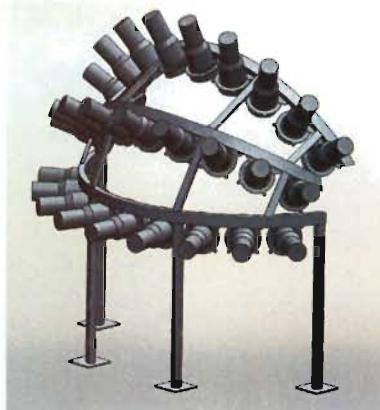
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## Physical supporting structure

Half of the structure supports 27 detectors

Detector "cross talk" calculations are underway

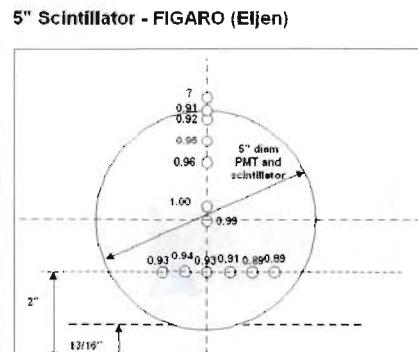
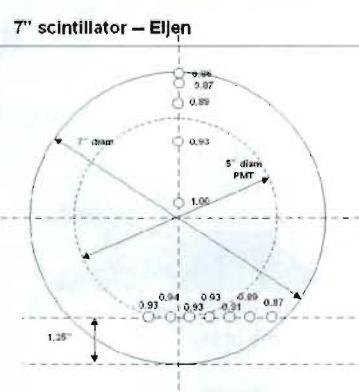


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## Response of liquid scintillators vs position



From tests with thinner scintillators, problem seems to be in uniformity of PMT response vs position

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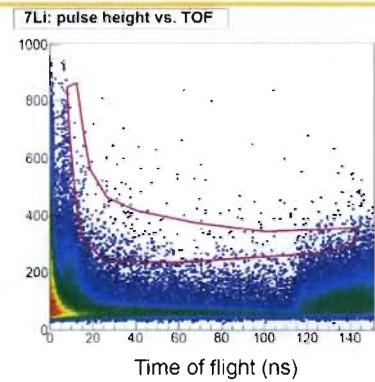
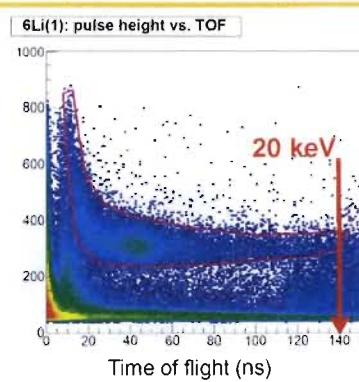
## 6Li-glass scintillators



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## Comparison of ${}^6\text{Li}$ -glass and ${}^7\text{Li}$ -glass detectors



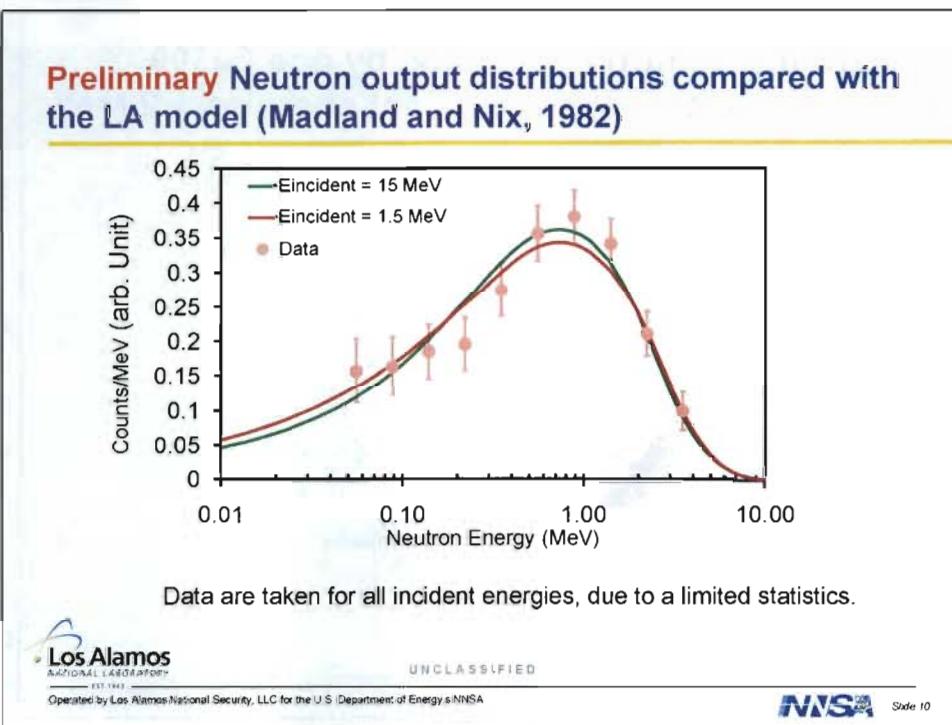
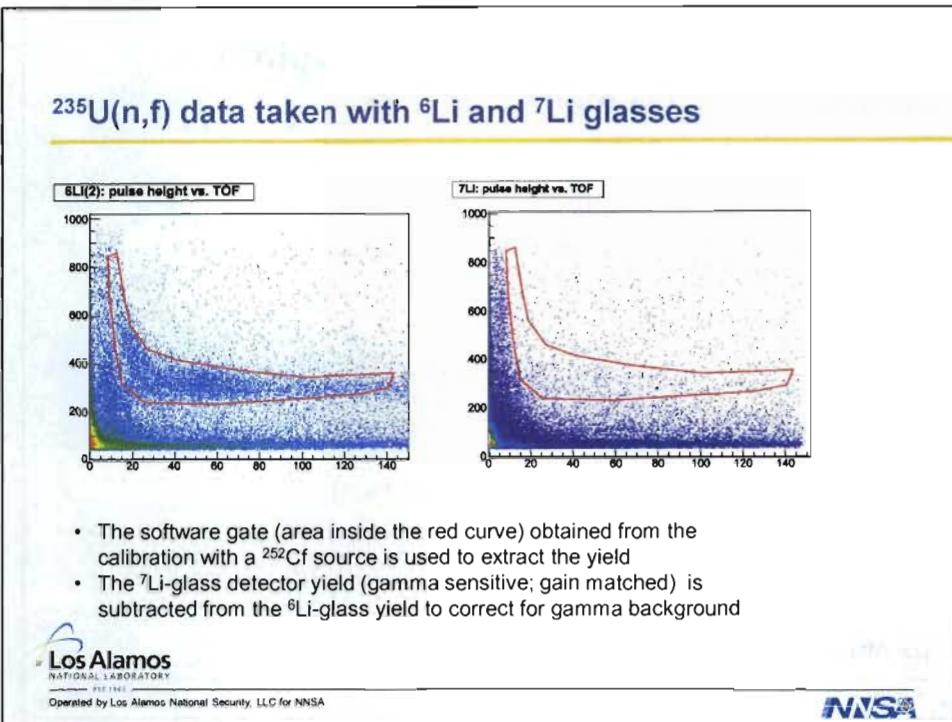
Software cut in pulse height vs. TOF is used to identify low energy neutrons.



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## How to achieve 5% accuracy for neutron emission spectra measurements

- Statistical uncertainties
  - Simulations
- Systematic uncertainties
  - Neutron detector efficiency
  - Time measurements
  - Flight path distance measurement

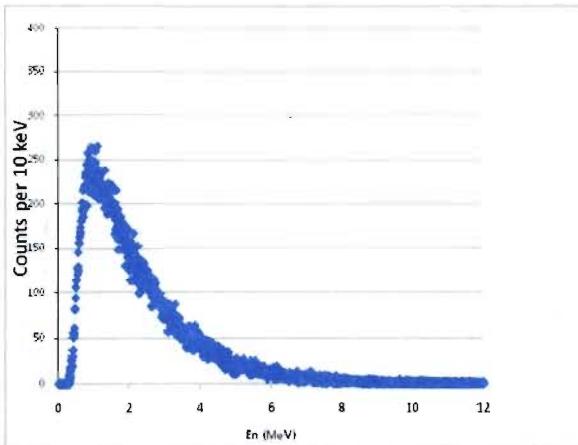


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## Spectrum of neutrons detected by one EJ309 detector in one week for incident energies 1-2 MeV



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## Possible systematic errors

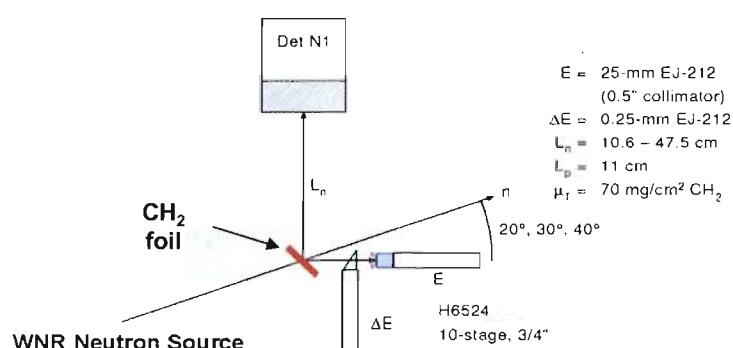
- Systematic uncertainties
  - Neutron detector efficiency
  - Time measurements
  - Flight path distance measurement



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## Geometry for tagged neutron efficiency calibration

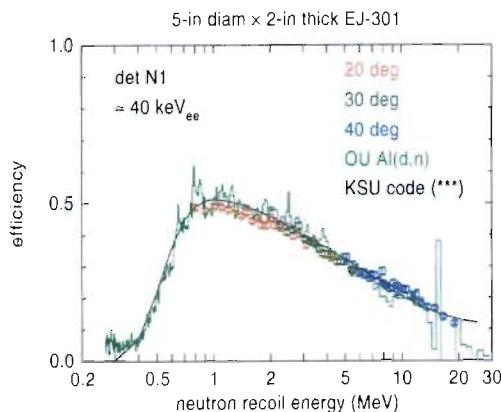


After method of Ch. Weber, I. Fabry, V. Huhn, A. Siepe, and W. von Witsch (Bonn), Nucl. Instr. Meth. Phys. Res. A488, 307 (2002).

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## Preliminary results for neutron detector efficiency

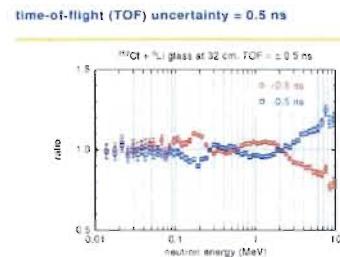
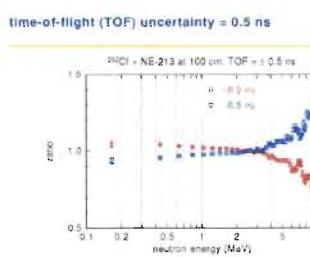


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## Effect of error in timing



→ Need timing uncertainty (centroid) of ~0.1 ns for high energy neutrons

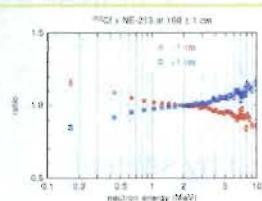
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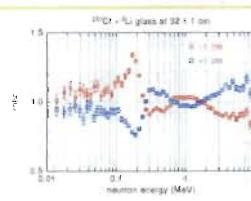
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## Effect of error in flight path length

Flight path uncertainty = 1 cm



Flight path uncertainty = 1 cm



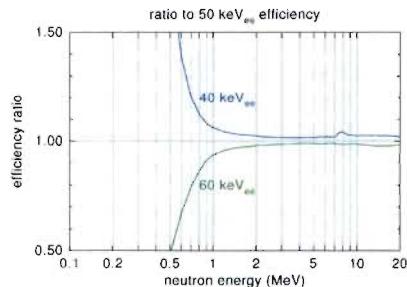
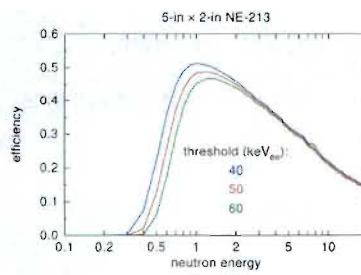
→ Need flight path uncertainty  
(to front face of detector) to  
about 2 mm

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## Effect of error in threshold



→ Need to know threshold: e.g.  
at 50 keVee, need to know  
threshold to about 5 keVee

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## Summary

- Two arrays of neutron detectors are being constructed at LANSCE to measure the fission neutron spectrum as a function of incident neutron energy
  - Liquid organic scintillators for  $E_n > 0.7$  MeV
  - $^6\text{Li}$ -glass scintillators for  $E_n < 1.0$  MeV
- Neutron detector efficiency is measured by several methods
  - $^{252}\text{Cf}$
  - Tagged neutrons
  - Ohio “standard” spectrum
- Possible systematic uncertainties are being studied



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## Many participants in the experimental work

- LANL
  - John O'Donnell, **Hye Young Lee**, Terry Taddeucci, Matthew Devlin, Ronald Nelson, Nikolaos Fotiades, John Ullmann, **Alexander Laptev**, **Brent Perdue**, Todd Bredeweg, Marian Jandel, David Vieira, Morgan White, Robert Haight
- LLNL
  - Ching-Yen Wu, **Elaine Kwan**, John Becker, **Andrii Chyzh**, Roger Henderson, Julie Gostic
- CEA
  - Thierry Granier, Gilbert Belier, Julien Taieb, Audrey Chatillon, Benoit Laurent

red = postdoc



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Thank you for your attention !



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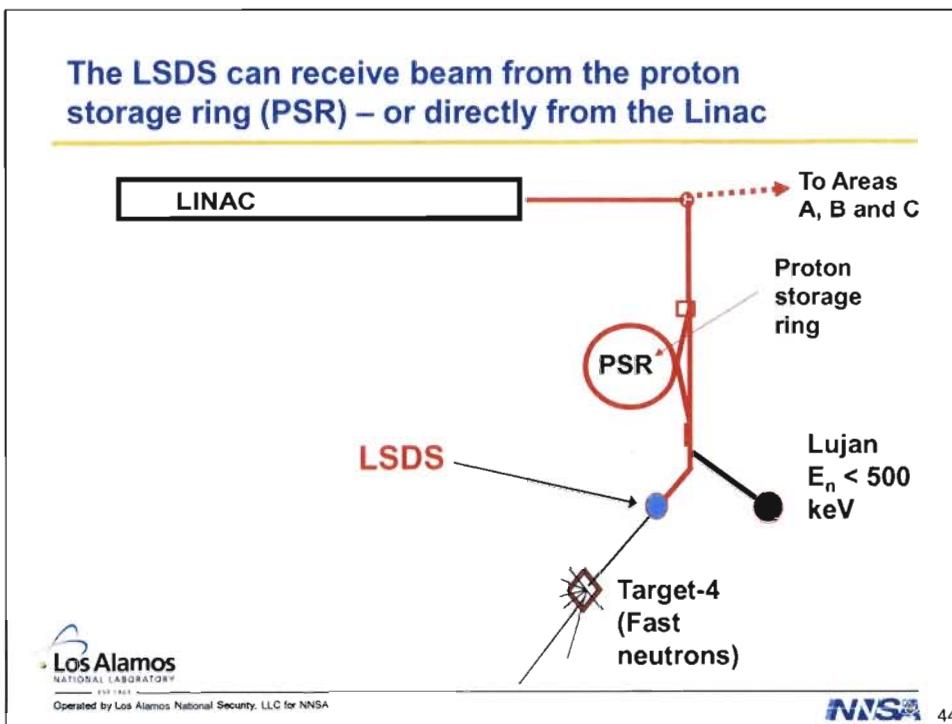
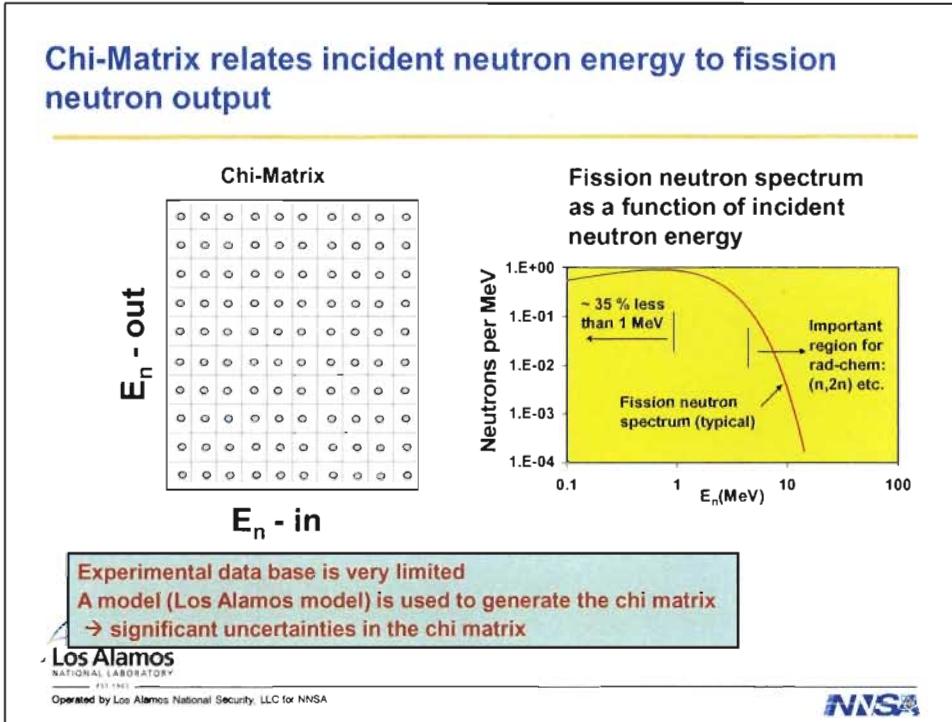
Supplementary Vugraphs



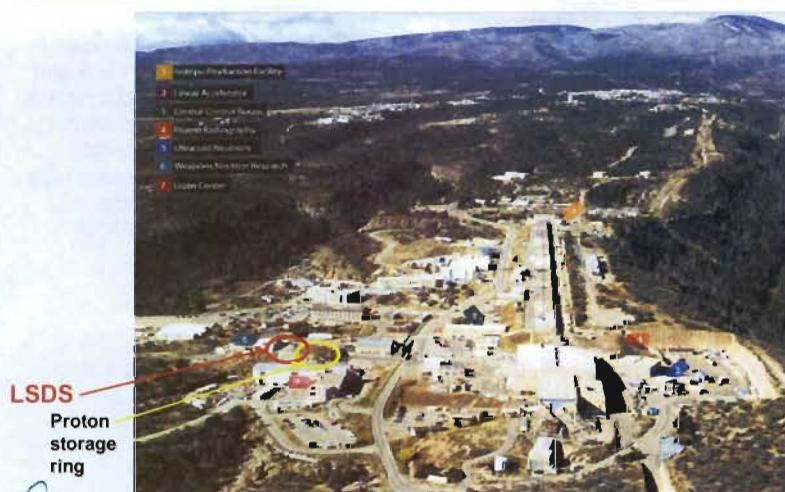
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## Los Alamos Neutron Science Center



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## Simulation of counting rate with liquid scintillators

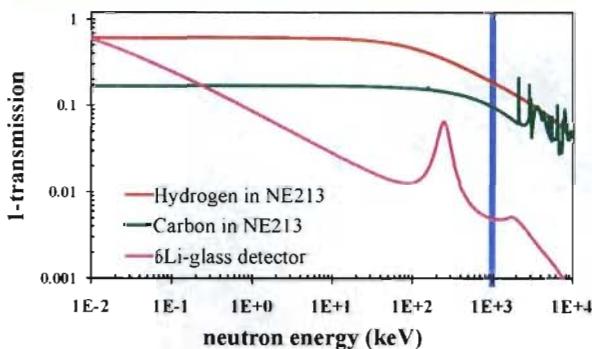
- WNR source
  - New flight path (15-degrees – left) in new building
  - 1.8 microamperes with 1.8 microsecond spacing
    - Lowest incident neutron energy = 0.8 MeV
  - Factor of 2 de-rate for availability, neutron attenuation or ??
- Parallel plate avalanche detector (LLNL)
  - 100 mg of  $^{239}\text{Pu}$
  - Assume Maxwellian fission neutron spectrum with  $T=1.30$  MeV
    - Isotropic angular distribution
- Two types of neutron detectors
  - For  $E_n > 0.7$  MeV – 7" diameter 2" thick @ 1 m; eff. from Terry T.
  - For  $E_n < 1.0$  MeV –  $^{6}\text{Li}$ - glass 1.2 cm thick @ 40 cm
- ENDF/B-VII fission cross section and nu-bar

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## Efficiency comparison between liquid scintillators and $^{6}\text{Li}$ -glass detectors



Although the efficiencies in liquid scintillators (red and green lines) are still high at  $E_n < 1 \text{ MeV}$  (blue divider), it is difficult to separate neutrons from gamma rays using a pulse shape discrimination method.

The pulse height is generated from the  $^{6}\text{Li}(n,\alpha)^{3}\text{H}$  reaction, where the  $\alpha$  and triton products have a kinetic energy of about 4.8 MeV.

$^{6}\text{Li}$ -glass detector has an usable efficiency below 1 MeV and could differentiate neutrons from most gamma rays in this region.

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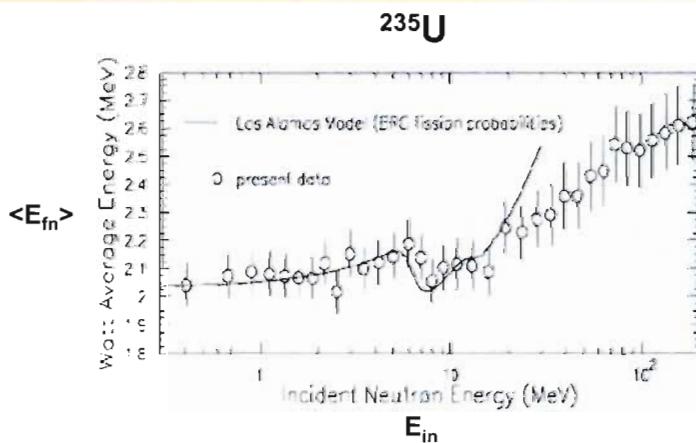
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Slide 3

## Previously we reported average fission neutron energies for $^{235}\text{U}$ (n,f) and $^{238}\text{U}$ (n,f)



Ref: T. Ethvignot et al., Phys. Rev. Lett. 92, 052701 (2005)

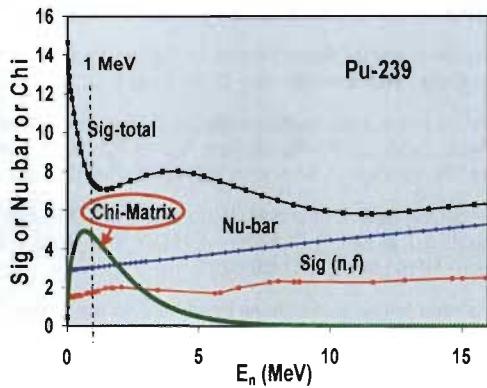
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## Reactivity depends on total cross section, fission cross section, and fission neutrons (nu-bar and Chi-Matrix)

- Total cross section → distance to first interaction
- Fission cross section and nu-bar increase with neutron energy → "hotter" fission neutron spectrum means more reactivity
- Average fission neutron energy (velocity) → dynamics
- Prompt diagnostics – NUEX
- Radchem diagnostics, esp.  $(n,2n)$  and  $(n,\gamma)$



Need all 3 for dynamic calculations:  $\sigma_f$ ,  $v$ ,  $X$



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## Publications and presentations (1)

- $^{238}\text{U}$ : T. Ethvignot, et al. Physics Letters B575 (2003), 221.
- $^{235}\text{U}$  and  $^{238}\text{U}$ : T. Ethvignot, et al. Phys. Rev. Lett. 94, 052701 (2005).
- $^{237}\text{Np}$  and  $^{238}\text{U}$ : J. Taieb et al., Int. Conf. on Nuclear Data for Science and Technology, Nice, France, April 23-27, 2007.
- $^{235}\text{U}$  and  $^{239}\text{Pu}$ : R. C. Haight et al., LA-UR-08-2585, April 16, 2008.
- $^{235}\text{U}$  and  $^{239}\text{Pu}$ : S. Noda, R. C. Haight, R. O. Nelson, M. Devlin, J. M. O'Donnell, A. Chatillon, T. Granier, G. Belier, J. Taieb, T. Kawano and P. Talou, "Measurement and analysis of prompt fission neutron spectra from 1 to 8 MeV in neutron-induced fission of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  using the double time-of-flight technique," Phys. Rev. C (2011) (in press)
- $^{239}\text{Pu}$ : A. Chatillon, G. Bélier, Th. Granier, B. Laurent, J. Taïeb, S. Noda, R.C. Haight, M. Devlin, R.O. Nelson, J.M. O'Donnell, "Energy measurement of prompt fission neutrons in  $^{239}\text{Pu}(n,f)$  for incident neutron energies from 1 to 200 MeV," ed. F. Cerutti and A. Ferrari, 12th International Conference on Nuclear Reaction Mechanisms, Villa Monastero, Varenna, Italy, 15 - 19 Jun 2009, pp.239-244, CERN-Proceedings-2010-001 (2010).



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## Publication and presentations (2)

- "Low-mass fission counter for the fission neutron spectrum measurement", C.Y. Wu, R. Henderson, J. Gostic, R.C. Haight, and H.Y. Lee, Oct. 2010 (LLNL-TR-461044)
- "Fission chamber development for the fission neutron measurement", C.Y. Wu, Science Campaign Year-End Review, LLNL, Sep 1, 2010 (LLNL-PRES-452543)
- "Gamma rays from neutron-induced fission of  $^{239}\text{Pu}$  and  $^{235}\text{U}$ ", E. Kwan, C.Y. Wu, A. Chyžh, J. Gostic, R. Henderson, R.C. Haight, H.Y. Lee, R.O. Nelson, J.M. O'Donnell, and T.N. Taddeucci, Mar 2011 (LLNL-PROP-472793)
- "Electrodeposition of U and Pu on thin C and Ti substrates", R.A. Henderson, J.M. Gostic, J.T. Burke, S.E. Fisher, and C.Y. Wu, Nucl. Instr. and Meth. A (2011), doi:10.1016/j.nima.2011.06.023
- Expected future publications in addition to the fission neutron data:
  - Fission  $\gamma$ 's for  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{252}\text{Cf}$
  - Fission counter if it works well for  $^{239}\text{Pu}$

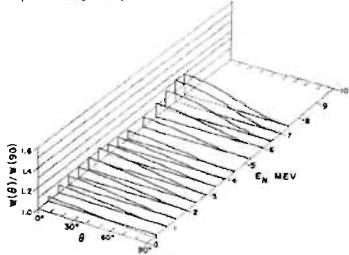


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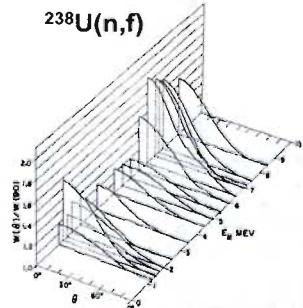


## Angular distribution of the fission fragments introduces another factor

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



$^{238}\text{U}(n,f)$



→ "Excess" of fission neutrons at 0 and 180 degrees

Ref: Simmons and Henkel,  
Phys. Rev. 120, 198 (1960)

→ We want to measure the angular distribution of the fission neutrons, hence another reason for the arrays.



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