Safeguards: Modeling of the Detection and Characterization of Nuclear Materials

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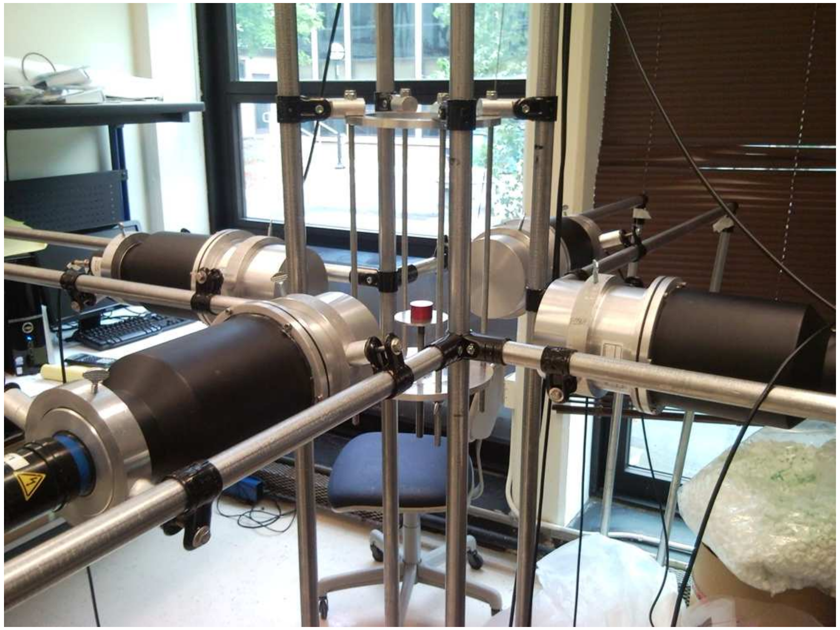


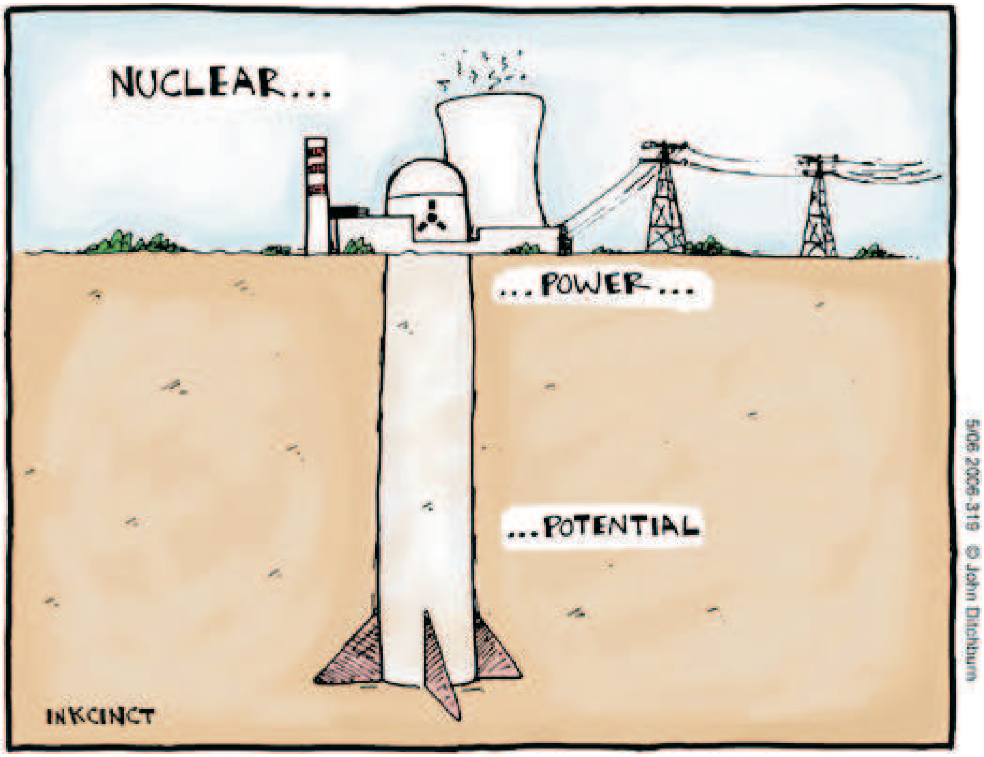
Overview

Safeguards

- ✔ Safeguards is a very broad area ranging from nuclear physics to political regulations.
- ✔ Three main tools are used for describing the physics of nuclear materials:
  - ✘ Analytical models.
  - ✘ Simulations.
  - ✘ Experiments.
- ✔ All of which have strengths and weaknesses.
- ✔ The work shown today is partly based on analytical methods for describing the physical processes governing the multiplication and detection of neutrons and gamma rays. The results have often been compared to simulations, and experiments in form of measurements have also been performed to gain additional insight.

Potential Perils?

The motivation of the work is the double-edged sword that nuclear materials presents: It is a way to provide carbon emission-free energy, while at the same time producing waste which needs careful management.



The Task

- ✔ The subject of this talk is the non-intrusive investigation of nuclear materials.

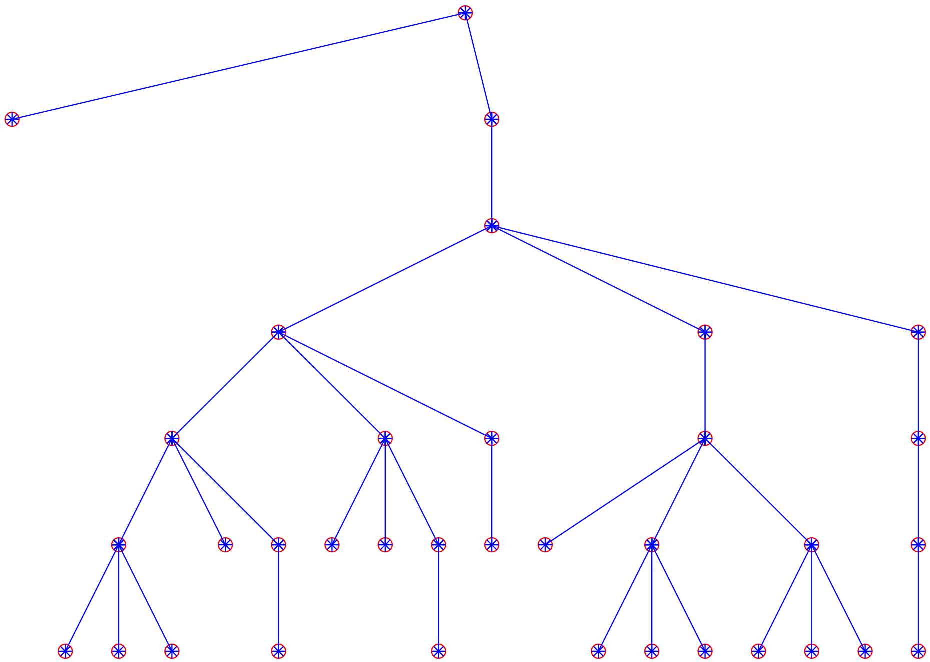
- ✔ Detection, identification and quantification of nuclear materials from the properties of the detected radiation.

Number Distributions

Branching Processes and Fissile Material

In fissile material the life of neutrons is a branching process in which neutrons can generate additional neutrons and reactions.

- ✔ Correlated events.
- ✔ Microscopic physics generating macroscopic effects.



Probability Distributions

- ✔ Neutrons in a fissile sample can undergo a number of different processes.
  - ✘ Fission.
  - ✘ Capture.
  - ✘ (n, xn)-reactions.
  - ✘ Escape the sample.
- ✔ Using the associated reaction probabilities, which depend on sample characteristics, probability balance equations can be formulated.
- ✔ Generating functions and master equations then provide the tools needed to describe the behavior of particles in the sample.

The Theory

$$p_1(n) = (1-\mathrm{p})\delta_{n,1} + \mathrm{p}\sum_{k=1}^{\infty} p_i(k) \sum_{\{n_1+n_2+\ldots+n_k=n\}} \prod_{i=1}^{k} p_1(n_i) \quad . \tag{1}$$

converted using generating functions to simple master equations:

$$h(z) = (1-\mathrm{p})z + \mathrm{p}q_i[h(z)], \tag{2}$$

$$H(z) = q_s[h(z)]. \tag{3}$$

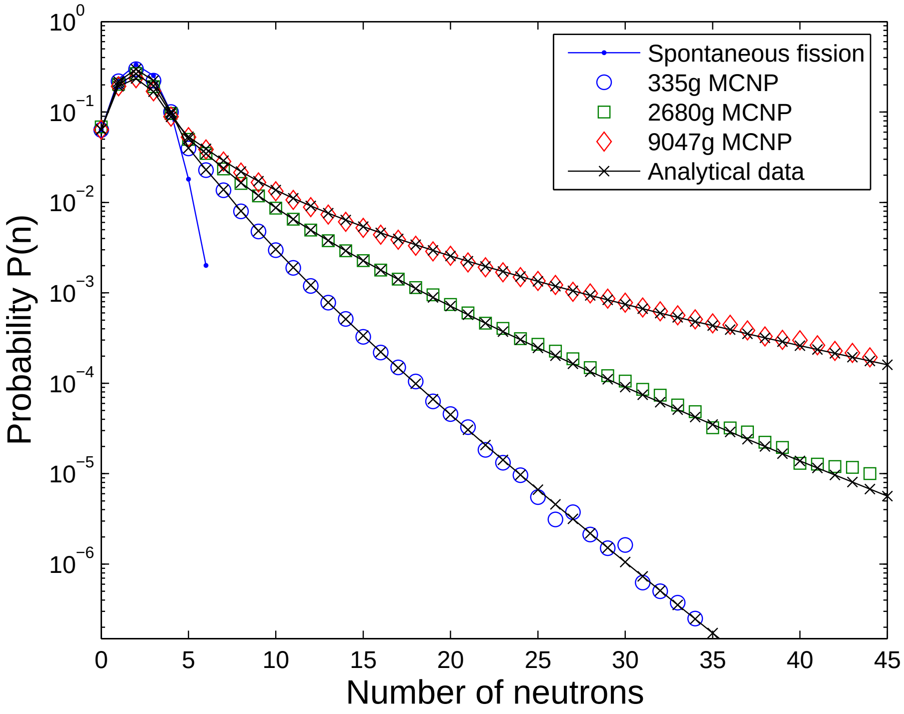
giving probability terms such as:

$$P(2) = \frac{1}{2}\left(\frac{1-p}{1-p\overline{\nu_i}}\right)^2 \left[\overline{\nu_s(\nu_s-1)} + \frac{p}{1-p\overline{\nu_i}}\overline{\nu_s}\,\overline{\nu_i(\nu_i-1)}\right]. \tag{4}$$

through:

$$p_1(n) = \frac{1}{n!}\frac{d^n h(z)}{dz^n}\bigg|_{z=0} \quad \text{and} \quad P(n) = \frac{1}{n!}\frac{d^n H(z)}{dz^n}\bigg|_{z=0}. \tag{5}$$

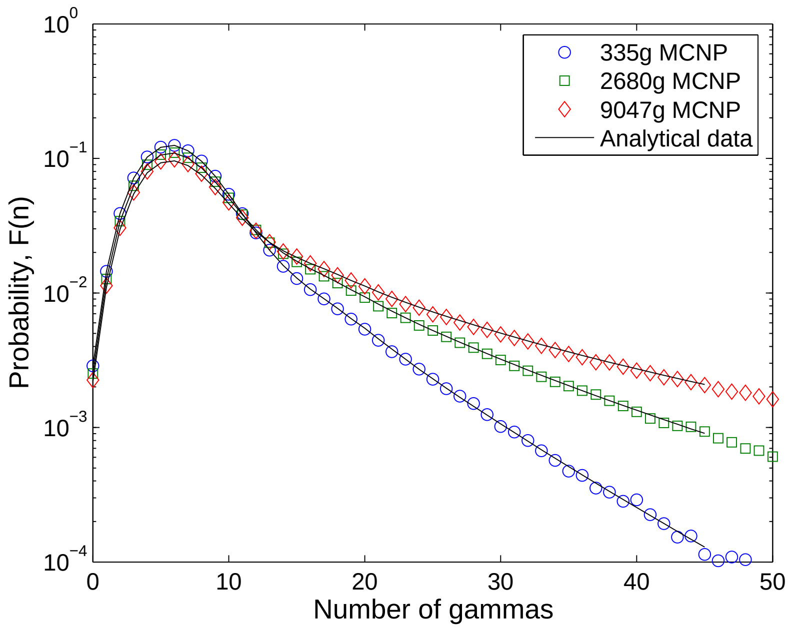
Neutrons



- ✔ Probabilities of having n neutrons generated when starting with a source event, as calculated from expressions derived from master equations.
- ✔ Dependence on mass shown for 20 wt% ^{240}Pu and 80 wt% ^{239}Pu metallic samples.
- ✔ Change compared to non-multiplying case.
- ✔ Excellent agreement with MCNP-PoliMi.

Gamma Rays

Gamma rays have no self multiplication, instead they are generated as a by-product of the branching process of neutrons.

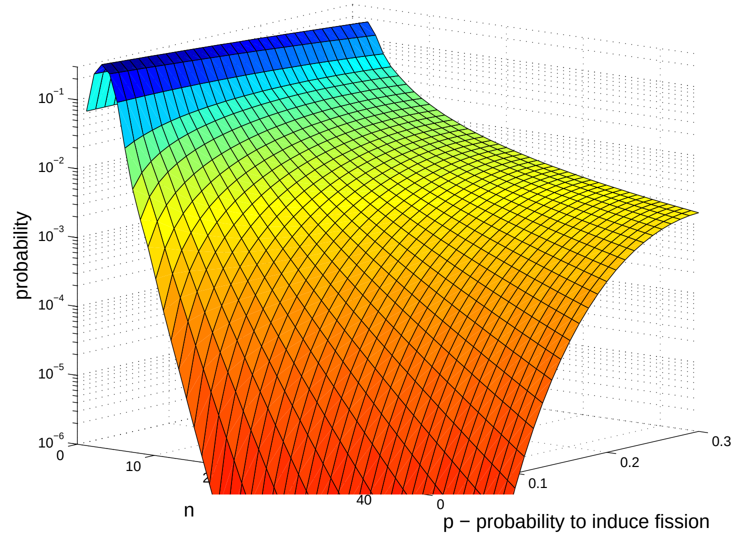


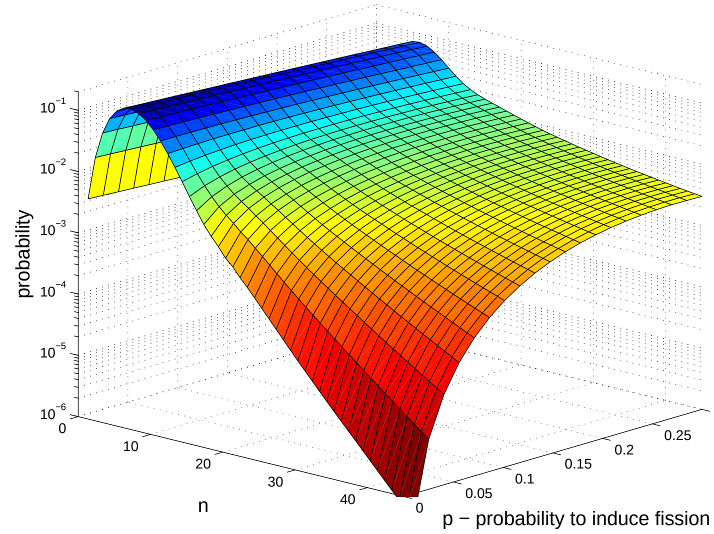
- ✔ Dependence on mass shown.
- ✔ Wider distribution compared to neutrons.
- ✔ Contains the information needed for calculating the moments.

The models are extended to also include absorption and detection.

Parameter Dependence

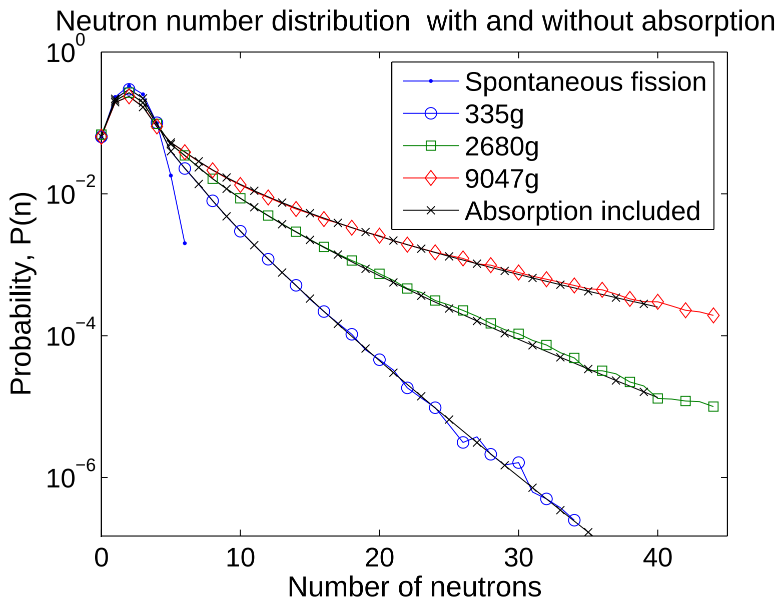
- ✔ The analytical formulae can be analyzed in symbolic handling programs such as Mathematica.
- ✔ Parameter dependencies can easily be investigated, in this case the reaction probability p is varied.

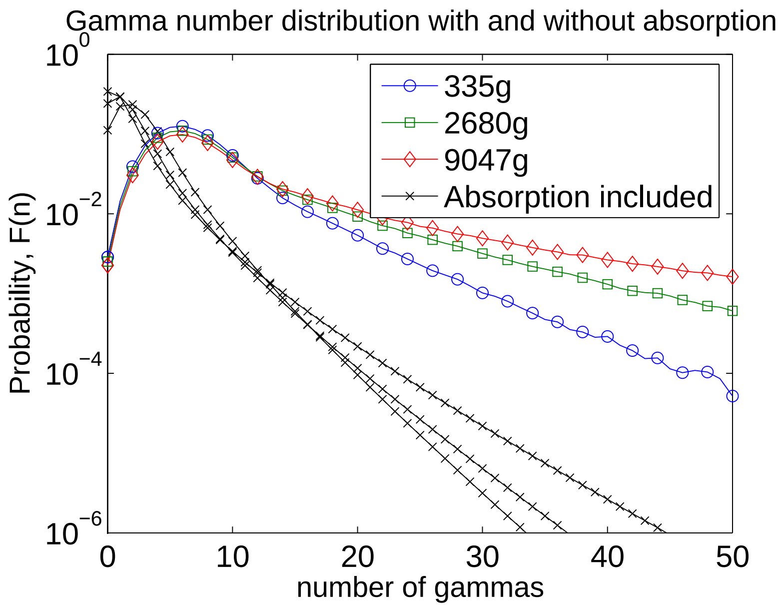




Emitted Distributions

Including absorption in the material has two effects for neutrons and photons.





- ✔ Self shielding is evident only for the photons.
- ✔ The photon distribution is furthermore changed because of the neutron absorption!
- ✔ Photons have a higher initial multiplicity but neutrons show a larger probability to reach high mutiplets.

Multiplicity Theory

Multiplicity Counting

- ✔ Neutron multiplicity counting is used extensively for materials control and accountability (MC&A).
- ✔ The system has four parameters: neutron leakage multiplication, alpha ratio, fission rate and neutron detection efficiency.
- ✔ Three multiplicity rates are normally measured: singles, doubles and triples.
- ✔ The systems are usually based on ^{3}He-detectors, which are sensitive to slow neutrons.
- ✔ The analysis is based on the same type of master equations previously mentioned.
- ✔ Use of scintillation detectors opens of possibilities of detecting not only neutrons.

Possibilities: Neutrons

Neutrons:

- ✔ Three measured multiplicity rates: S, D and T;
- ✔ Four unknowns: F, $\mathbf{M} \equiv \frac{1-\mathrm{p}}{1-\mathrm{p}\nu_{i1}}$, α and ε_n

where

- ✔ F = sample fission rate,
- ✔ p = first collision probability,
- ✔ $\alpha = \frac{Q_\alpha}{Q_f \nu_{sf,1}}$ = the fraction of (α, n) neutrons in the spontaneous source,
- ✔ ε_n = neutron detection efficiency.

Neutron Rates

The following rates can be derived:

$$S = F\varepsilon_n(1+\alpha\nu_{s,1})\frac{\mathbf{M}\nu_{s,1}(1+\alpha)}{(1+\alpha\nu_{s,1})} = F\varepsilon_n\mathbf{M}\nu_{s,1}(1+\alpha), \tag{6}$$

$$D = \varepsilon_n^2 C_2 = \frac{F\varepsilon_n^2\mathbf{M}^2}{2}\left[\nu_{s,2} + \left(\frac{\mathbf{M}-1}{\nu_{i1}-1}\right)\nu_{s,1}(1+\alpha)\nu_{i2}\right], \tag{7}$$

$$T = \varepsilon_n^3 C_3 =$$

$$= \frac{F\varepsilon_n^3\mathbf{M}^3}{6}\left\{\nu_{s,3} + \left(\frac{\mathbf{M}-1}{\nu_{i1}-1}\right)[3\nu_{s,2}\nu_{i2} + \nu_{s,1}(1+\alpha)\nu_{i3}]\right.$$

$$\left. + 3\left(\frac{\mathbf{M}-1}{\nu_{i1}-1}\right)^2 \nu_{s,1}(1+\alpha)\nu_{i2}^2\right\}. \tag{8}$$

Possibilities: Gamma rays

Gamma rays:

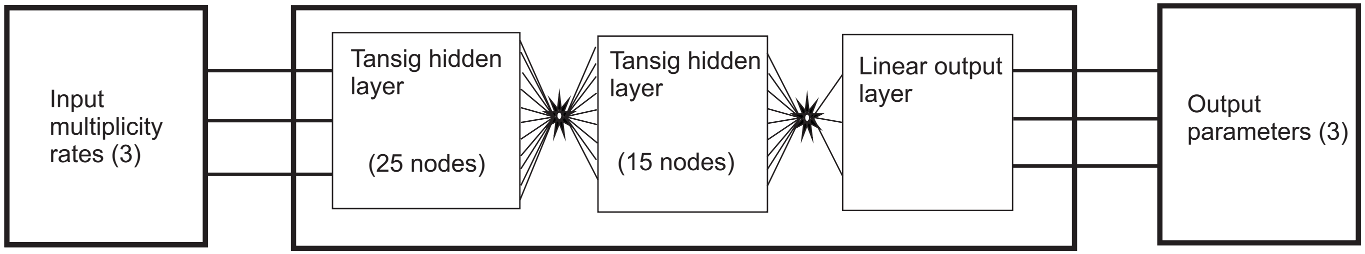
- ✔ Three measured multiplicity rates: S_γ, D_γ and T_γ;
- ✔ Three more unknowns: $\mathbf{M}_\gamma$, γ and ε_γ

where

- ✔ $\mathbf{M}_\gamma$ = gamma leakage multiplication,
- ✔ γ = fraction of single gamma rays in the spontaneous gamma source,
- ✔ ε_γ = gamma detection efficiency.

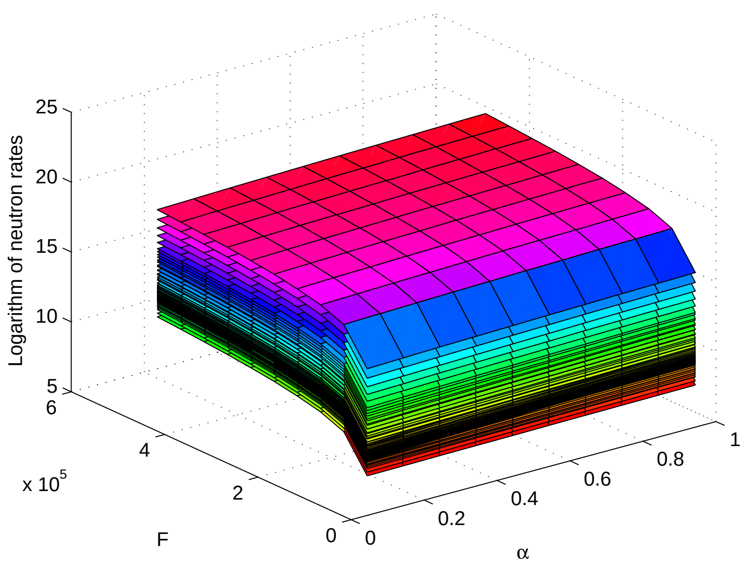
Extensions and Neural Networks

- ✔ The formalism was extended to take into account not only gamma rays, but also mixed particle multiples such as $nn\gamma$, $n\gamma$.
- ✔ The extra multiples raise the total number of measurables to 9 while there is only 7 unknowns of which two are detection efficiencies.
- ✔ The analysis grows more and more complicated which motivated the usage of artificial neural networks for inverting the solutions (unfolding).
- ✔ Parameter unfolding using the information in an over-determined system. (Work done in collaboration with Senada Avdic).



Method Validation

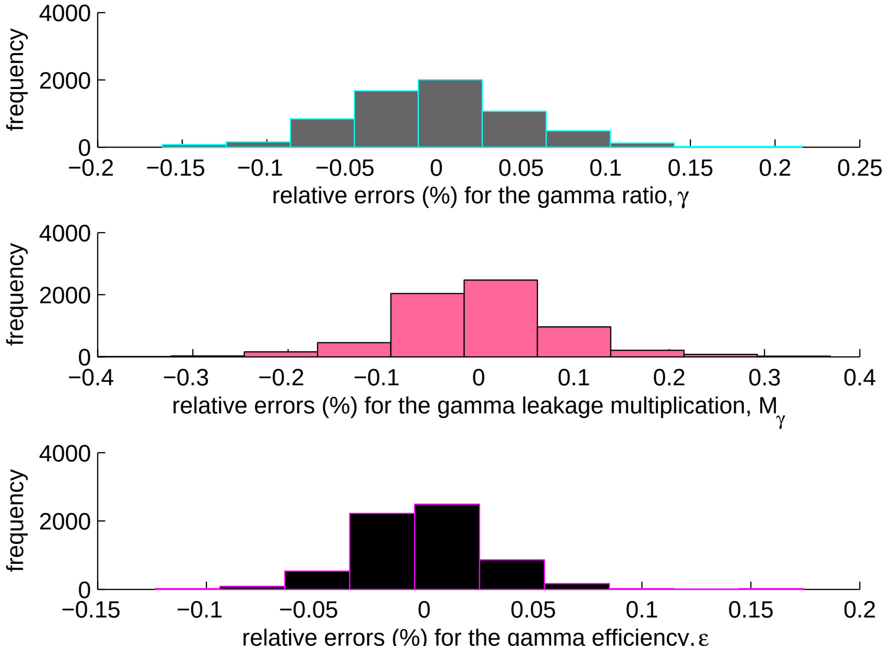
	fission rate (F)	α	p
max. abs. rel. error (%)	0.0001	0.0021	0.0001
mean error of training (%)	-1.98e-9	-4.14e-7	4.96e-9
standard deviation of training (%)	7.56e-6	2.11e-4	3.24e-5
mean error of test data (%)	1.28e-6	-7.15e-6	2.56e-6
standard deviation of test data (%)	9.34e-6	1.42e-4	3.29e-5



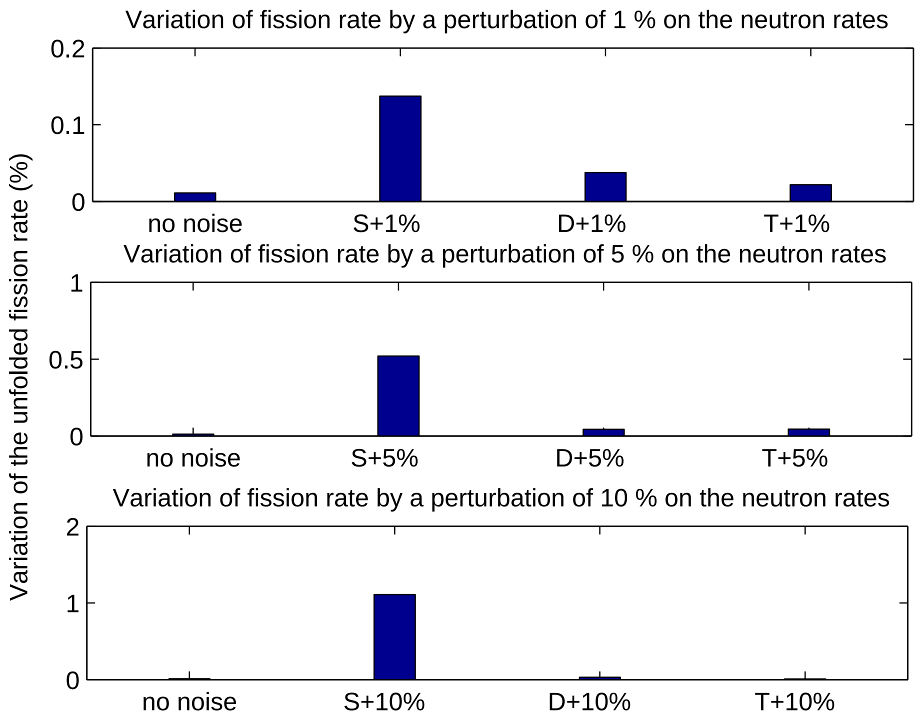
- ✔ The neural network was validated using neutron multiplicities.
- ✔ The accuracy of the unfolded parameters, especially the fission rate (F), which is directly linked to the sample mass, is very encouraging.

ANN Results

- ✔ The multiples up to third order give a total of 9 measurables.
- ✔ Additional parameters such as gamma ray detection efficiency, gamma ratio and gamma leakage multiplicity, needs to be unfolded.



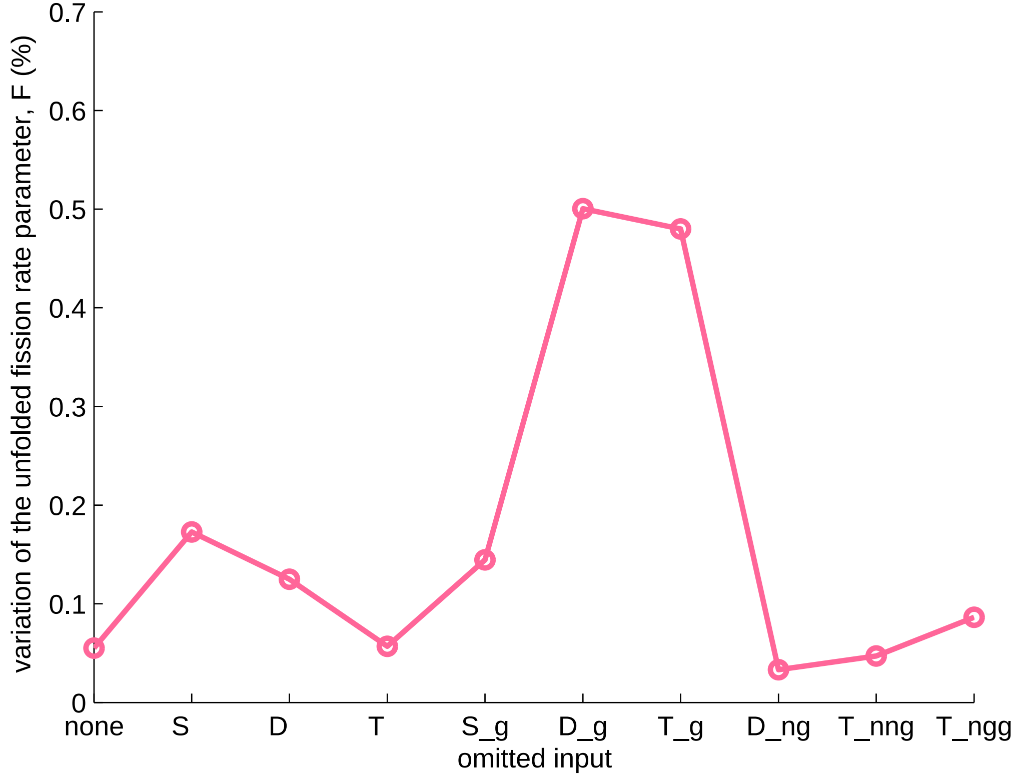
Sensitivity analysis



- ✔ Sensitivity analysis was performed by adding noise to the training data.
- ✔ The higher moments which are harder to measure, turned out to be the *least* sensitive to noise/statistical uncertainty, which is beneficial.

Sensitivity analysis

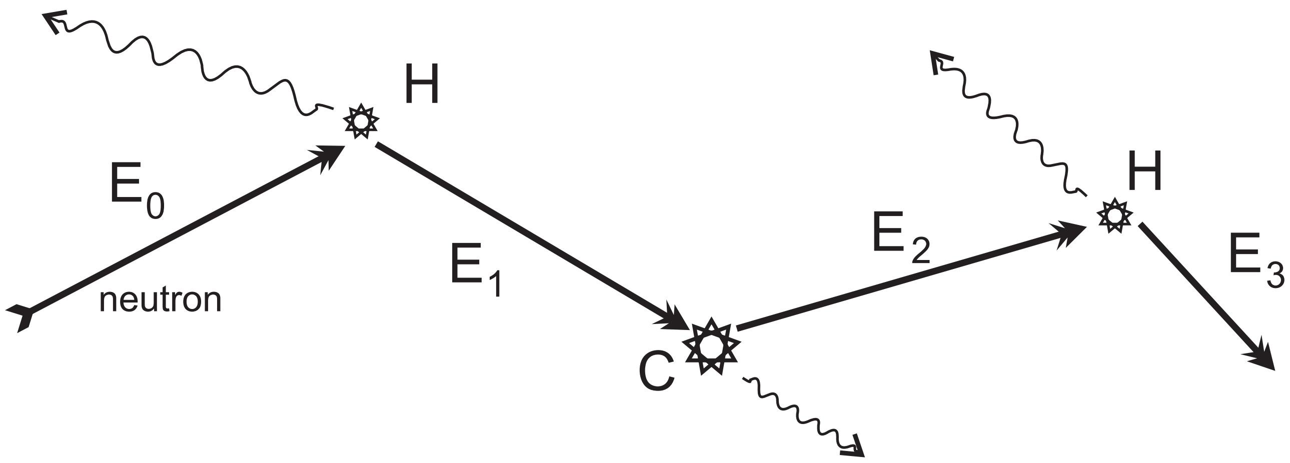
✔ Omitting of input parameters show which observables that are of most importance and which could be omitted due to redundancies.



Scintillation Light Pulses

Neutron Scattering

- ✔ The light pulses generated in a scintillation detector by fast neutrons, depend on the scattering history.
- ✔ Energy is transferred in the collisions and transformed into light depending on the nuclei involved in the scattering.
- ✔ The order and type of nuclei are very important to the generated light pulse.



Convolution Integrals

New types of mathematical tools needed. It is not a branching process but rather interdependent processes:

$$f_{HCH}(L,E_0) = \int_0^{L-l_1} \int_0^{L} f_H\left[L-(l_1+l_2), E_0-(T_h(l_1)+T_c(l_2))\right] \times$$

$$f_C\left[l_2, E_0-T_h(l_1)\right] f_H[l_1,E_0] \times W\left[E_0-T_h(l_1)-T_c(l_2)-T_h(L-(l_1+l_2))\right] dl_1 dl_2.$$

using:

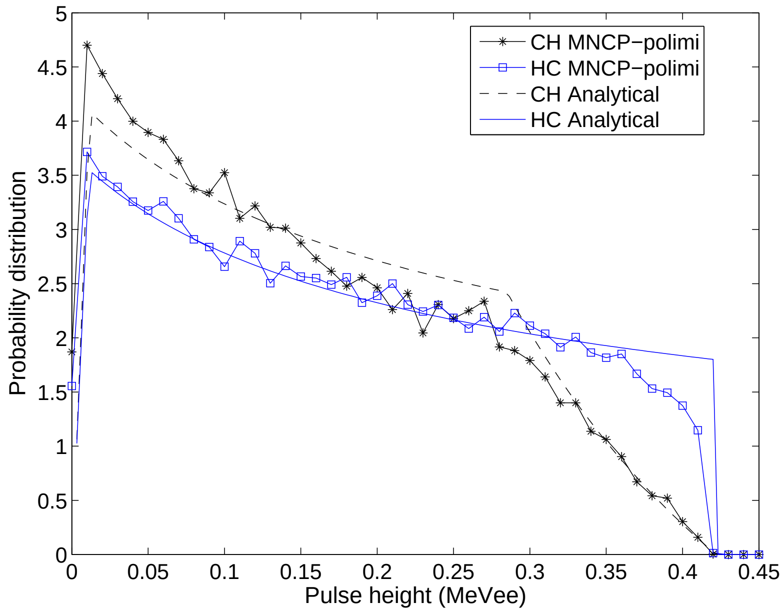
$$f_{1h}(L,E_0) = \frac{1}{E_0\sqrt{b^2+4aL}},$$

$$f_{1c}(L,E_0) = \frac{\theta(L_{max,c}-L)}{c\,(1-\alpha)\,E_0}.$$

$$T_h(L) = \frac{\sqrt{b^2+4aL}-b}{2a} \quad , \quad T_c(L) = L/c$$

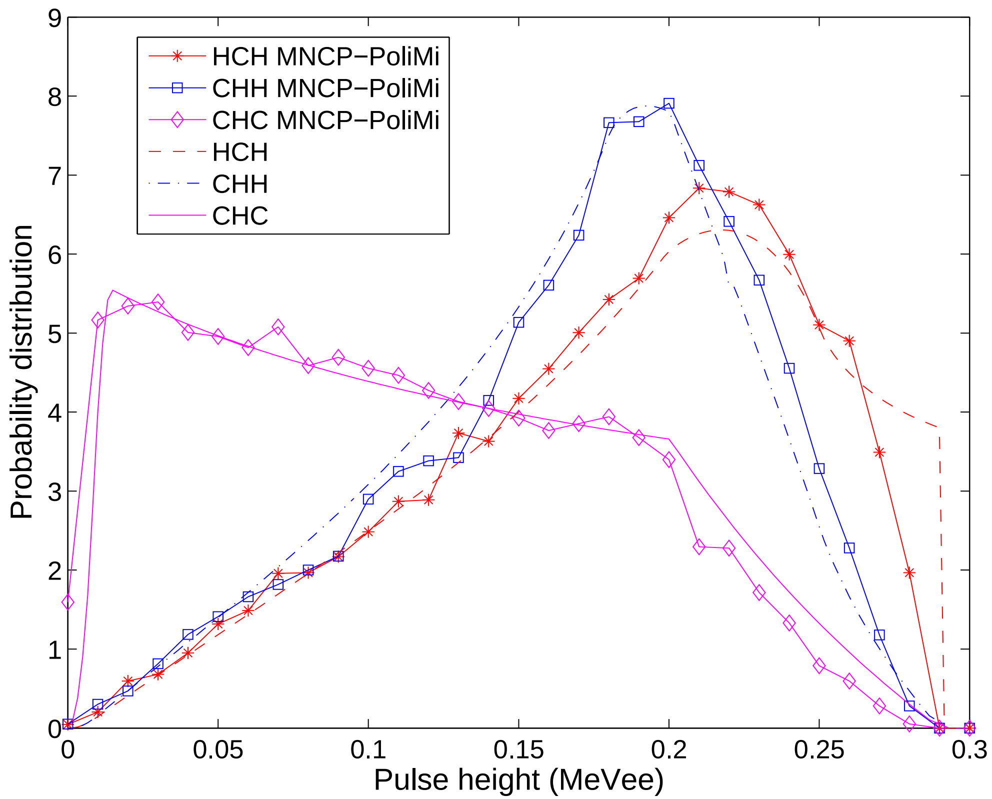
Collision Histories

- ✔ Monte Carlo simulations were used as a comparison with good agreement.
- ✔ Limited energy transfer for neutrons scattering on carbon (C), changes the resulting light distribution clearly.
- ✔ Neutron energy: 2MeV.



Collision Histories

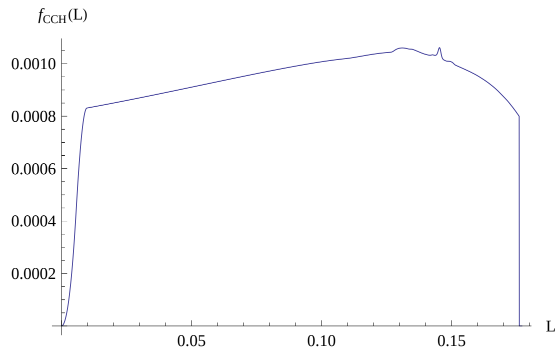
The light pulse amplitude distribution was calculated for individual collisions histories for mono-energetic neutrons of 1.5 MeV.



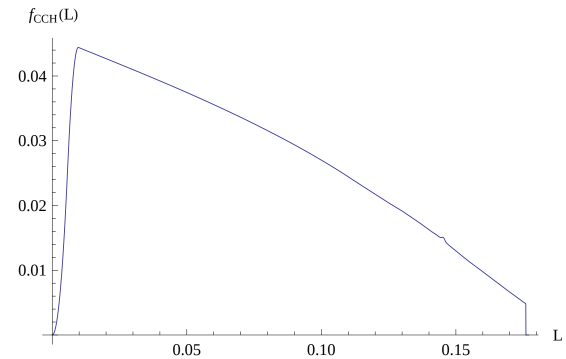
- ✔ Even for collisions histories with the same type of nuclei, the light pulse distribution is very different.
- ✔ Comparisons with MCNP-PoliMi show good agreement.
- ✔ The results from individual collisions histories can be used to understand the shape of the full light pulse distribution.

Size Dependence

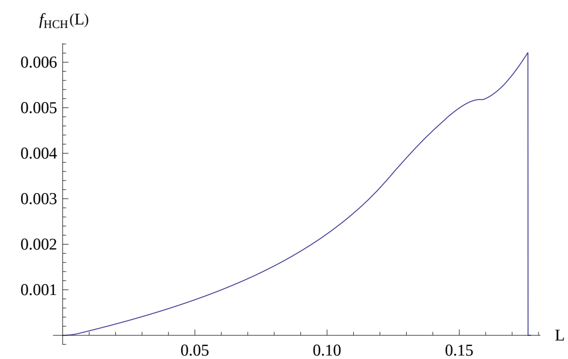
The differences depending on detector size and collision history are accurately highlighted.



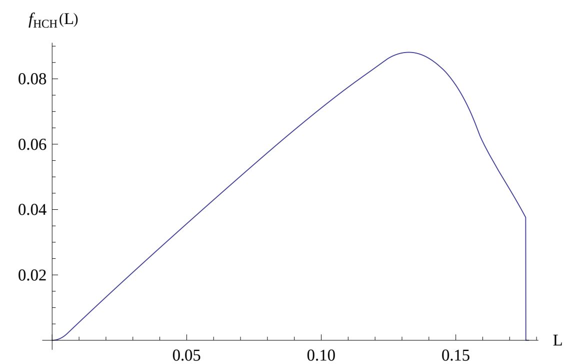
(a) CCH, 1 cm.



(b) CCH, 10 cm.



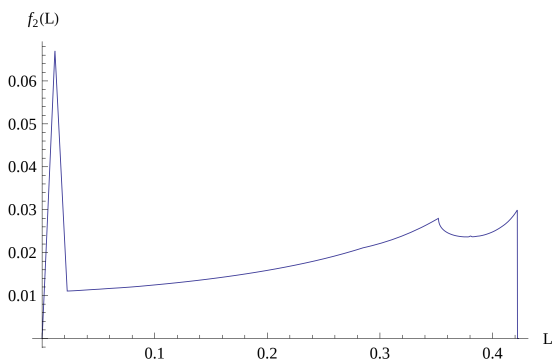
(c) HCH, 1 cm.



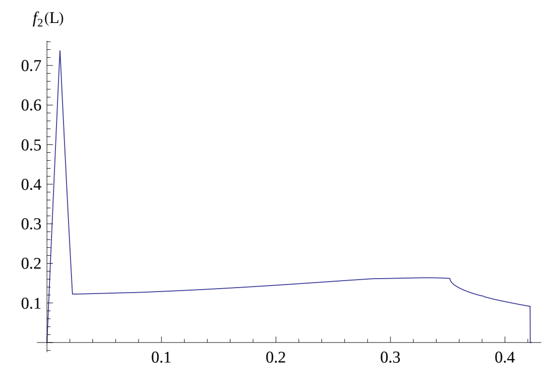
(d) HCH, 10 cm.

Size Dependence

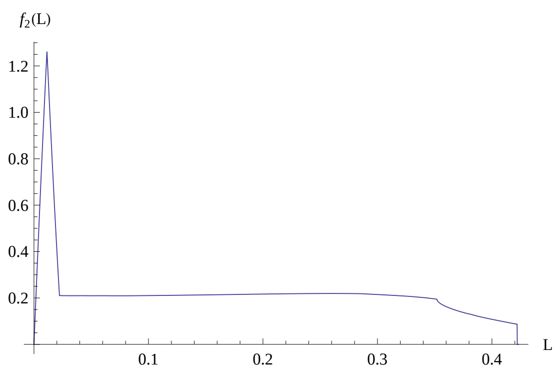
Size dependence is noticeable also in combined light pulse distributions (below: All neutrons that underwent two collisions).



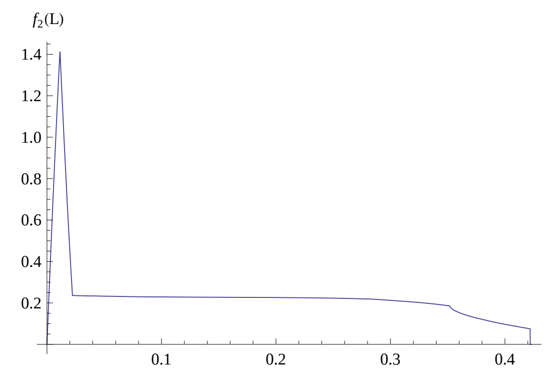
1 cm



5 cm



10 cm

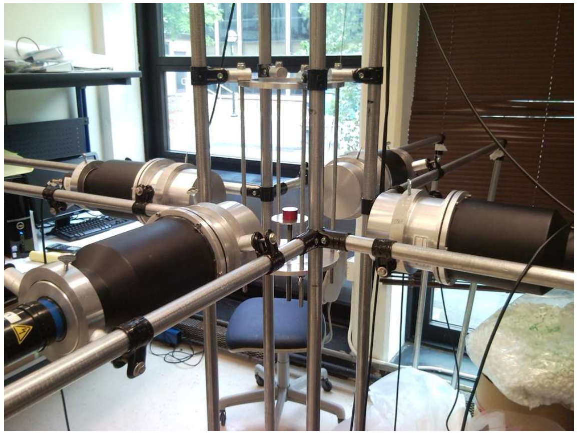


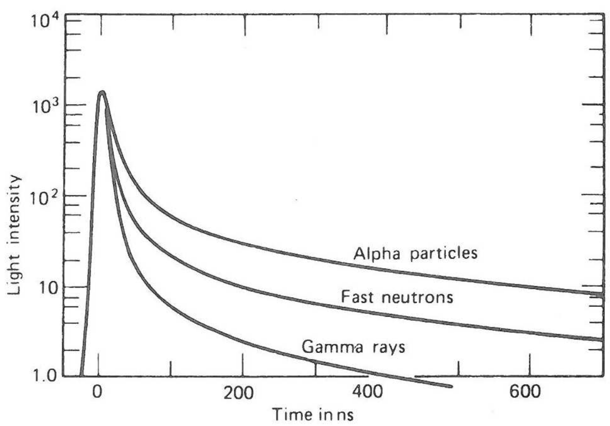
15 cm

Correlation Measurements

Event Correlations

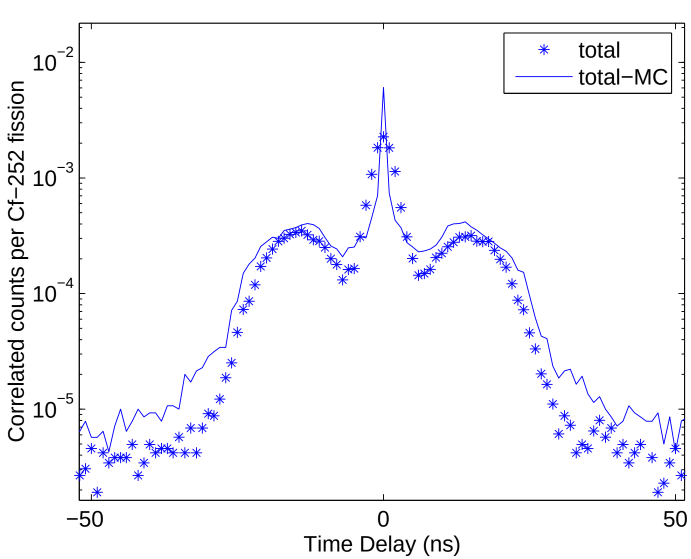
- ✔ Fissions generate a number of neutrons and gamma rays in each event.
- ✔ Simultaneously detecting multiple particles is therefore a good indicator of fissile material.
- ✔ Correlation measurements could also give additional information about sample parameters in a similar way to multiplicity measurements.
- ✔ Detecting both types of radiation has good benefits.

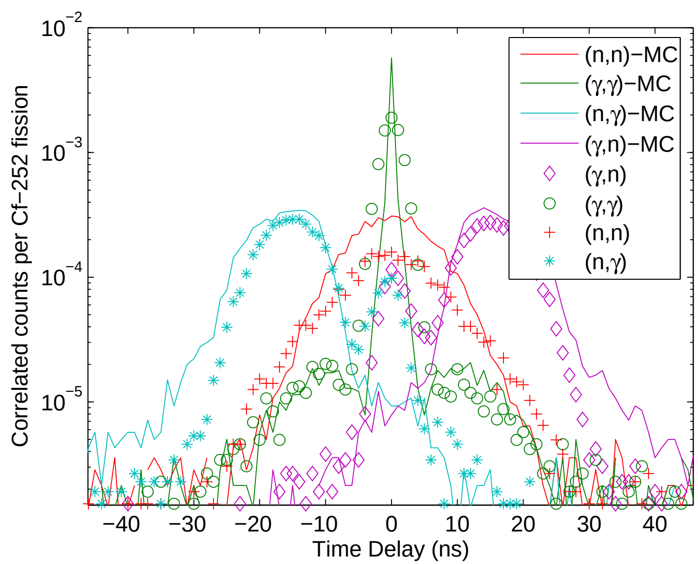




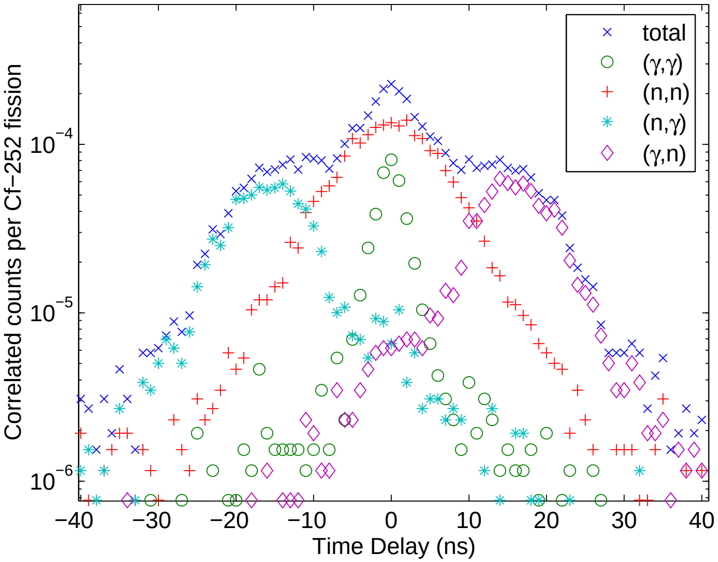
Cross-Correlations

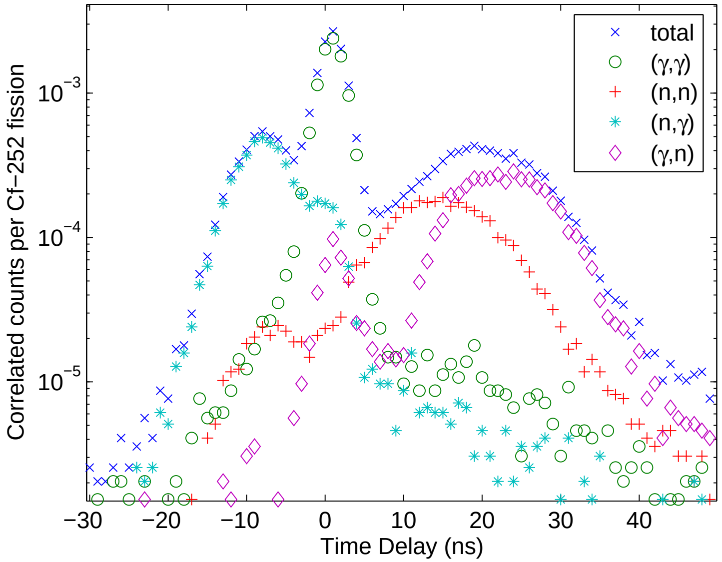
- ✔ Cross-correlation (CC) measurements utilize two correlated pulses to give a sample-geometry signature.
- ✔ It requires very fast detectors and systems, due to the inherent speed of the gamma rays and neutrons.
- ✔ The accuracy of the CC functions depends on the pulse shape discrimination (PSD) and other considerations.





- ✔ With 2.2 cm lead between the source and the detectors the gamma rays are clearly attenuated.
- ✔ Asymmetric detector setup (or source placement) show the effect of geometry for the measured CC-functions.

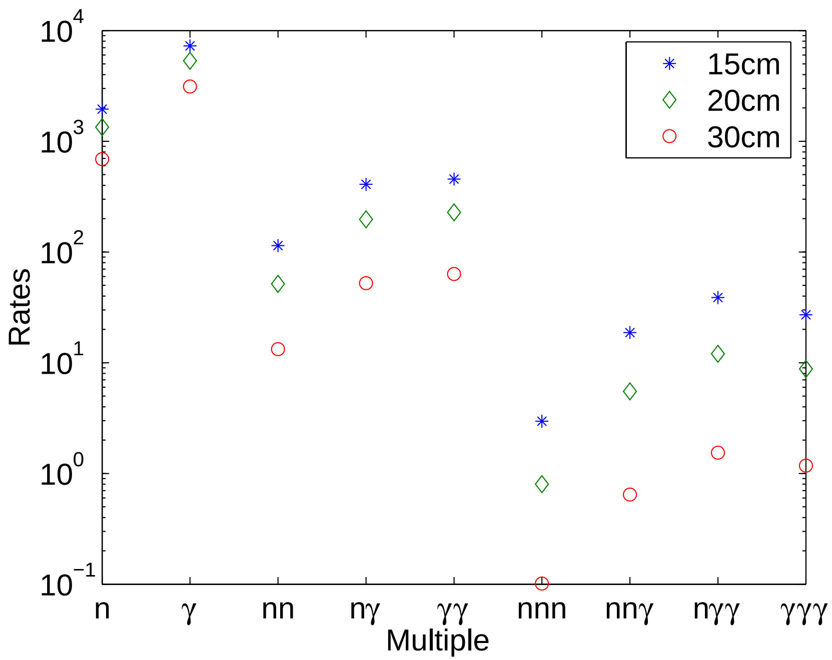




Mixed Multiplicity Counting

✔ As seen from multiplicity theory, successful detection of multiples of both neutron and photons might be used to improve pure neutron multiplicity counting.

+ No moderation needed, more measurables.
– Low detection efficiency, requires PSD or two types of detectors.



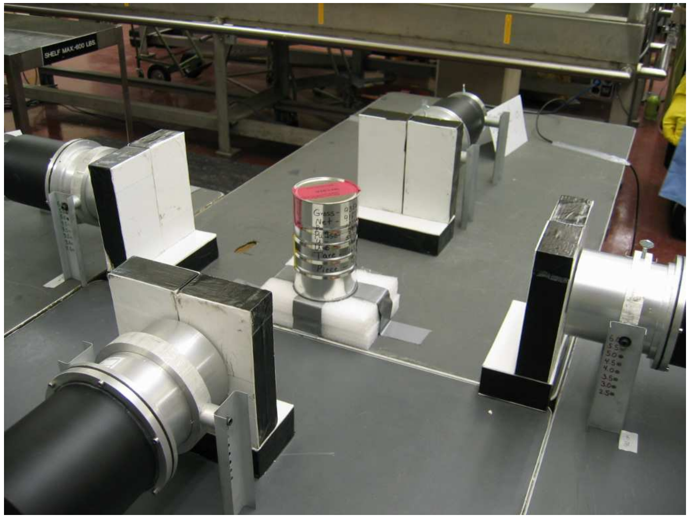
Detection rates

Count rates (s^{-1}) and statistical errors:						
	30cm	$\pm 1\sigma$	20cm	$\pm 1\sigma$	15cm	$\pm 1\sigma$
R_n	690.4	0.63	1338.6	0.95	1947.4	1.36
R_γ	3119.9	1.34	5335.6	1.90	7256.6	2.62
R_{nn}	13.3	0.09	51.5	0.19	114.3	0.33
$R_{n\gamma}$	52.3	0.17	197.3	0.37	409.3	0.62
$R_{\gamma\gamma}$	63.4	0.19	227.7	0.39	455.0	0.66
R_{nnn}	0.1	0.01	0.8	0.02	3.0	0.05
$R_{nn\gamma}$	0.6	0.02	5.5	0.06	18.7	0.13
$R_{n\gamma\gamma}$	1.5	0.03	12.0	0.09	38.7	0.19
$R_{\gamma\gamma\gamma}$	1.2	0.03	8.8	0.08	27.0	0.16
	Measurement time (s):					
T	1735.8		1475.3		1054.6	

A 20 μCi ^{252}Cf source was used yielding 19500 fissions/s.
Only triples require a somewhat longer measurement time to achieve good statistics.

MOX-Fuel

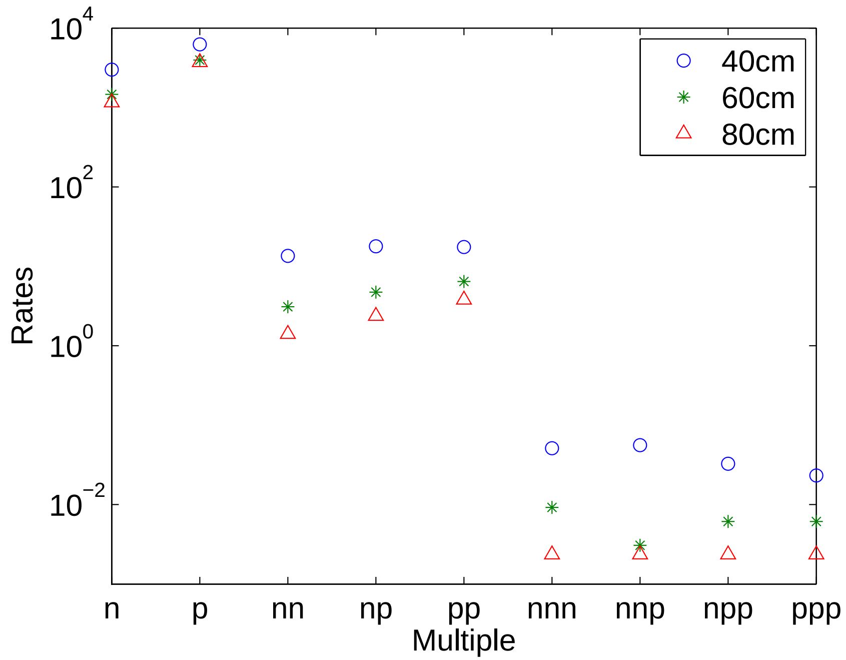
- ✔ At INL, measurements were performed with MOX fuel.
- ✔ Due to the very strong intrinsic gamma sources, lead shielding was used, which could also simulate the reduction in count rates that is to be expected in shielded samples.
- ✔ Up to 100 fuel pins were contained in a single sample.



Acknowledgement goes to Dr. Chichester and his colleagues from INL for organizing the measurements and providing the MOX samples.

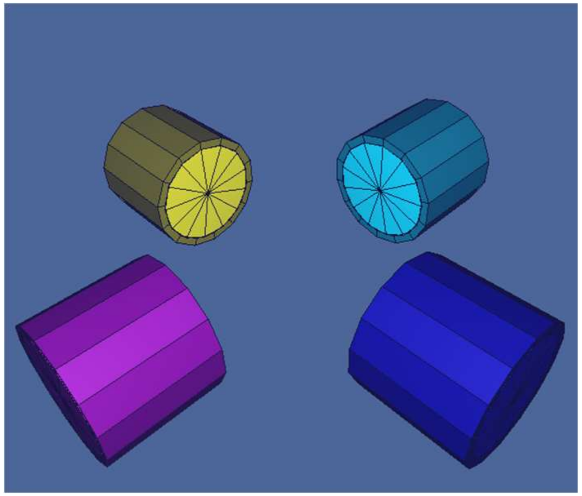
Measured MOX Rates

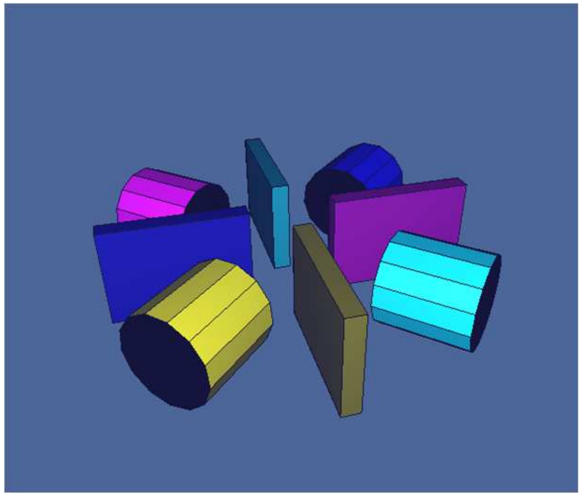
- ✔ For MOX samples measured at various distances, the importance of geometrical detector efficiency is once again demonstrated.
- ✔ due to the shielding the higher order multiplet rates are heavily reduced.

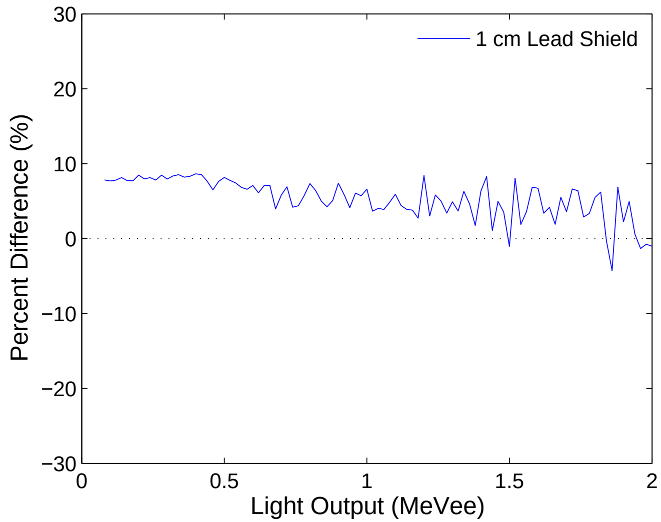


Simulation of shielding

As cross-talk increases with the proximity of the detectors, simulations were made to investigate if shielding could reduce the phenomenon.



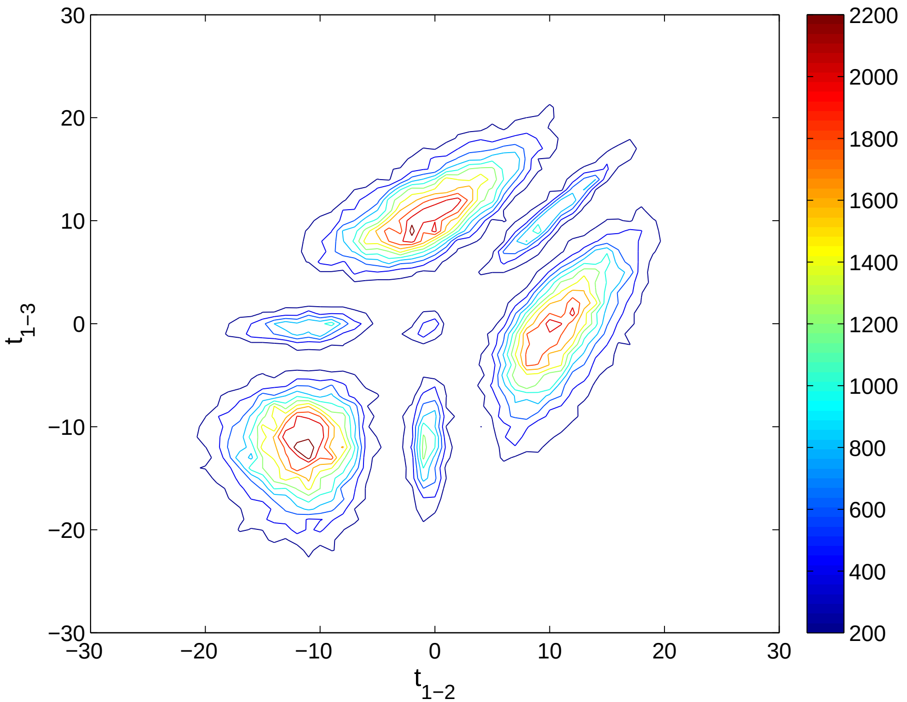


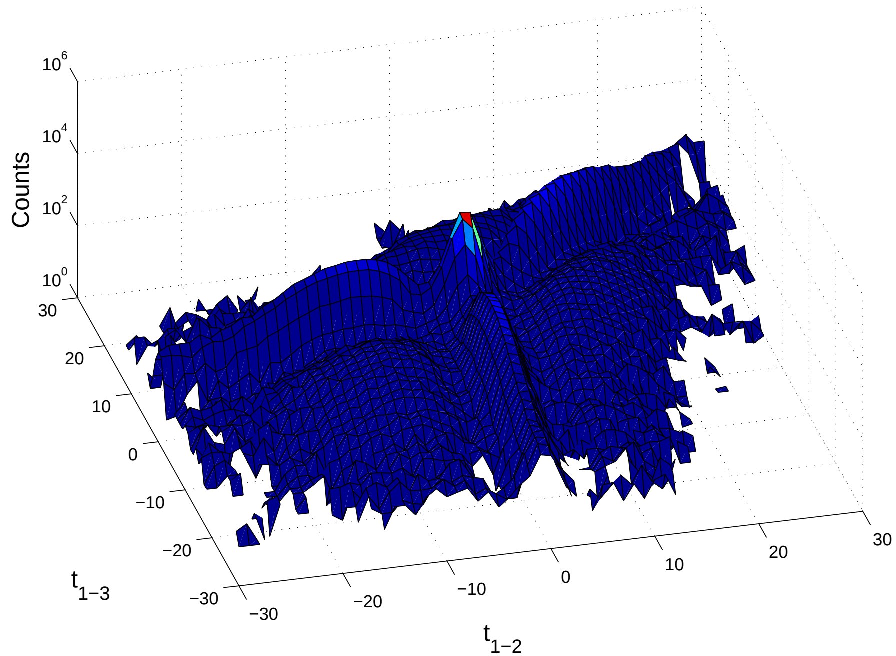


- ✔ Both lead (Pb) and polyethylene (PE) shielding was used surrounding the detector volumes.
- ✔ It was found that both high and low-Z shielding only increased the count rates, due to scattering in the shields.

Bi-correlations

- ✔ Bi-correlations are easy to get as a bi-product of higher order multiplicity
- ✔ bi-correlation of $nn\gamma$ (left), all bi-correlations (right).

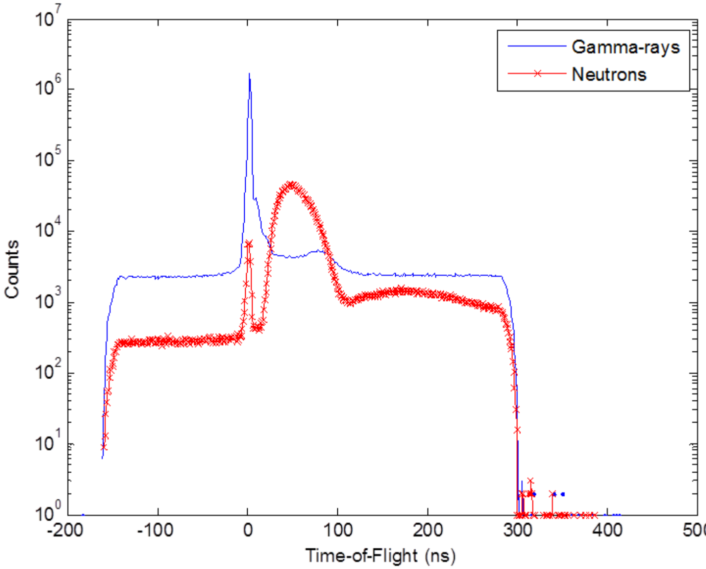




TOF Measurements for Scintillator Characterization

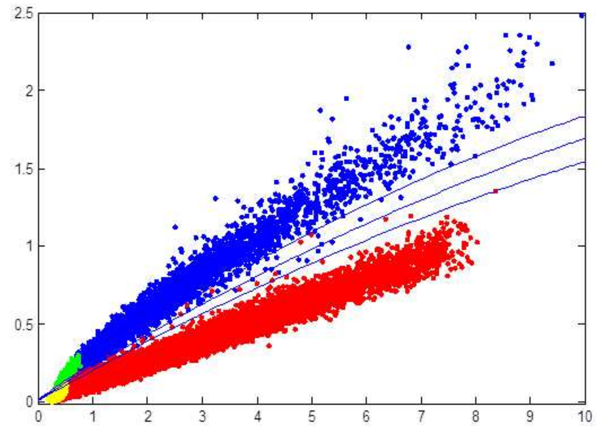
TOF setups

- ✔ Using simple laboratory time-of-flight (TOF) setups one can obtain data for scintillation detectors which is vital for adequate simulation of such detectors.
- ✔ The light response as well as the full response matrix and efficiency are a few important parameters which all depend on geometry, scintillator substance, and parts such as PMT.

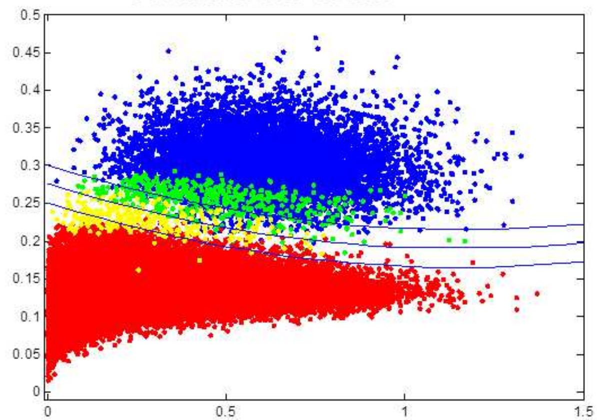


PSD methods

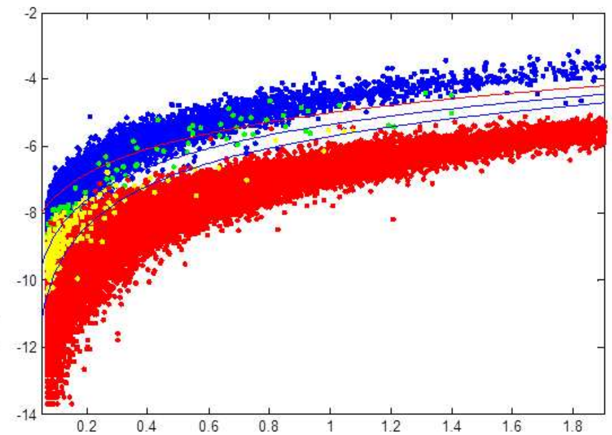
✔ Good pulse shape discrimination (PSD) is of vital importance for scintillation measurements and code validation.



- Yellow and Green pulses are low voltage pulses
- 1.77% of pulses inside of confidence lines



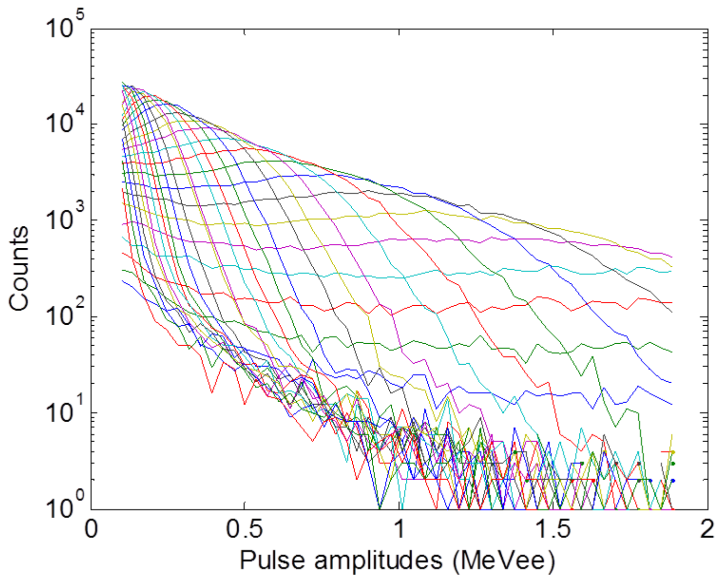
- Yellow and Green pulses are inside of method 1's confidence lines.
- 1% of pulses in method 1 and method 2's confidence lines

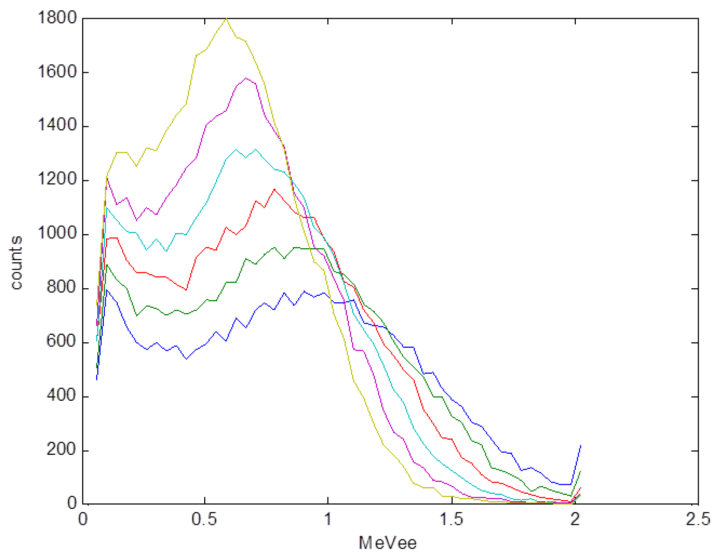


- Yellow and Green pulses are inside of method 1 and method 2's confidence lines.
- .87% of pulses in all three methods confidence lines

Energy Response

- ✔ With highly accurate time response the TOF spectrum can be divided into "quasi" mono-energetic bins.
- ✔ The extent of each pulse height distribution correspond to maximal energy transfer on hydrogen due to the non-linear response of the scintillation material.

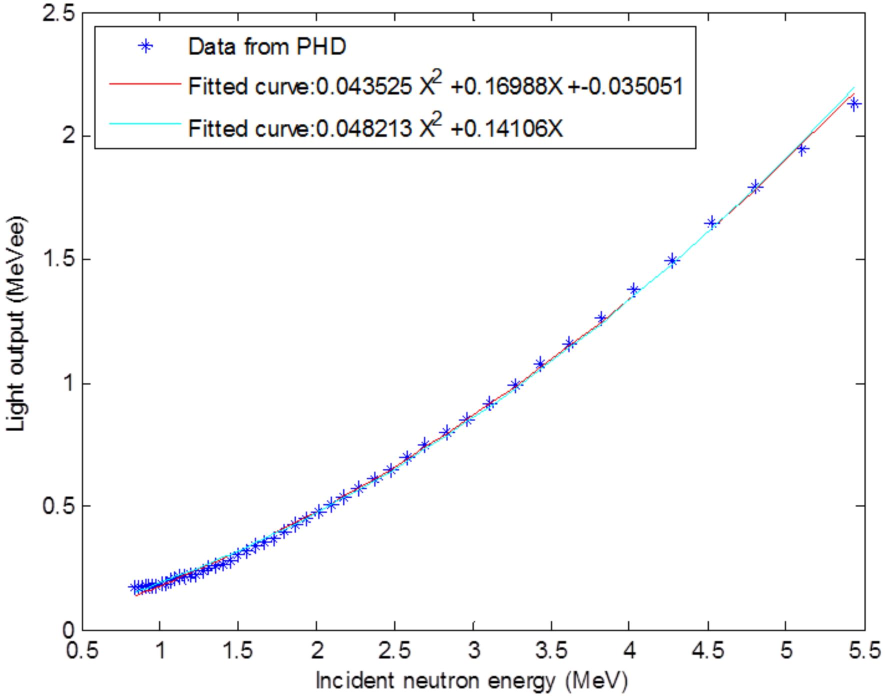


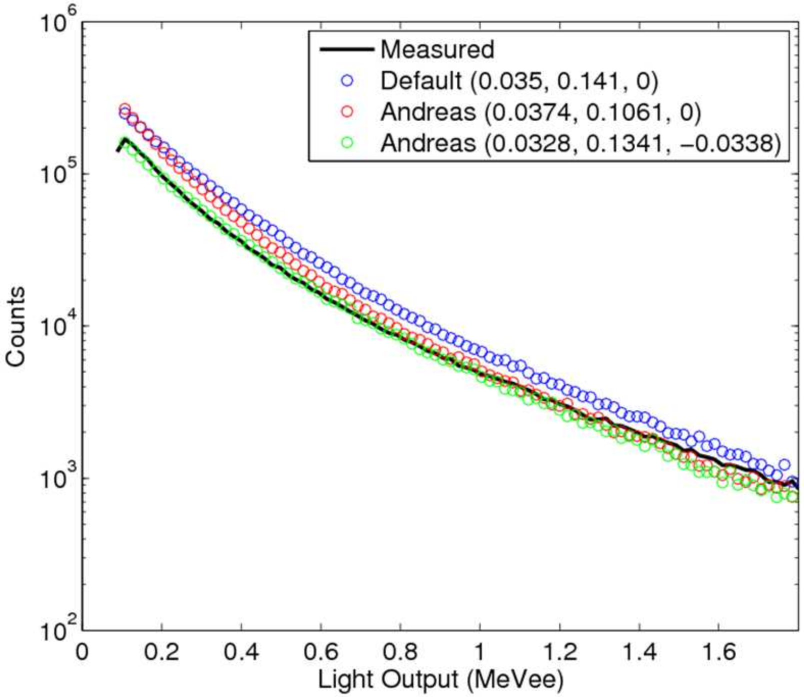


The characteristic light generation for collisions on carbon (very small) can not be accessed with the same simple setup.

Light Output Function

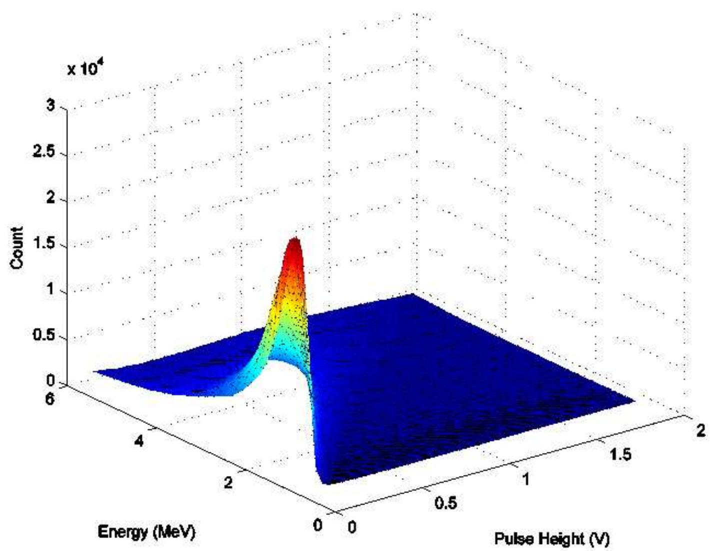
- ✔ For each energy a PHD generates a data point for light generation at that transferred energy.
- ✔ An empiric curve can then be fitted to the data and used as a conversion for any energy transfer to its corresponding light output in keVee for that specific detector.

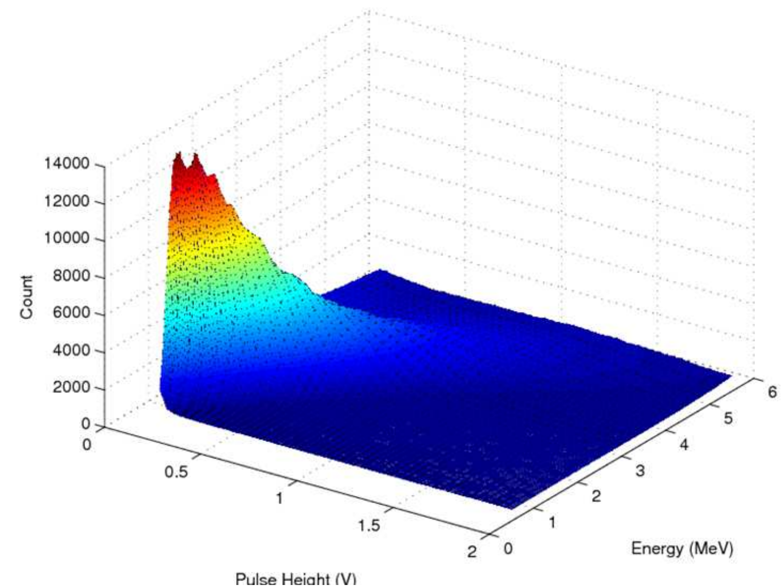




Response matrix

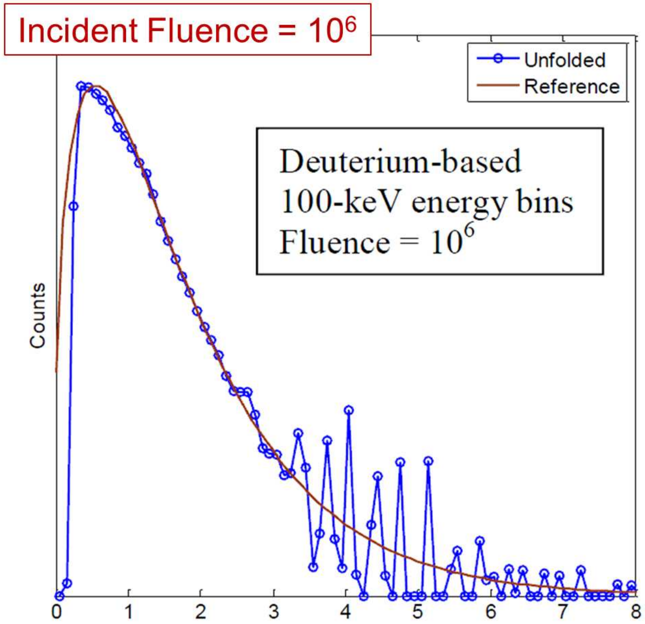
- ✔ A full response matrix can also be generated in the same way, to be used for spectrum unfolding.
- ✔ This type of data can be favorably measured at a well characterized beam facility with reduced background such as Ohio University's tandem Van Der Graaf generator.

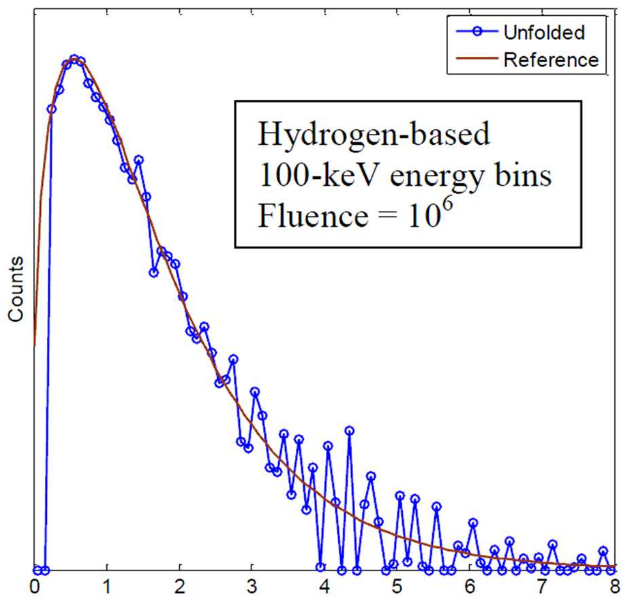




Deuterated scintillators

- ✔ Theoretically deuterated scintillators would be better suited for spectrum unfolding since the neutrons prefer direct backscattering with 8/9th energy transfer.
- ✔ The response of the scintillator will then have a stronger relationship between transferred energy and light (especially at small geometries or high energies).





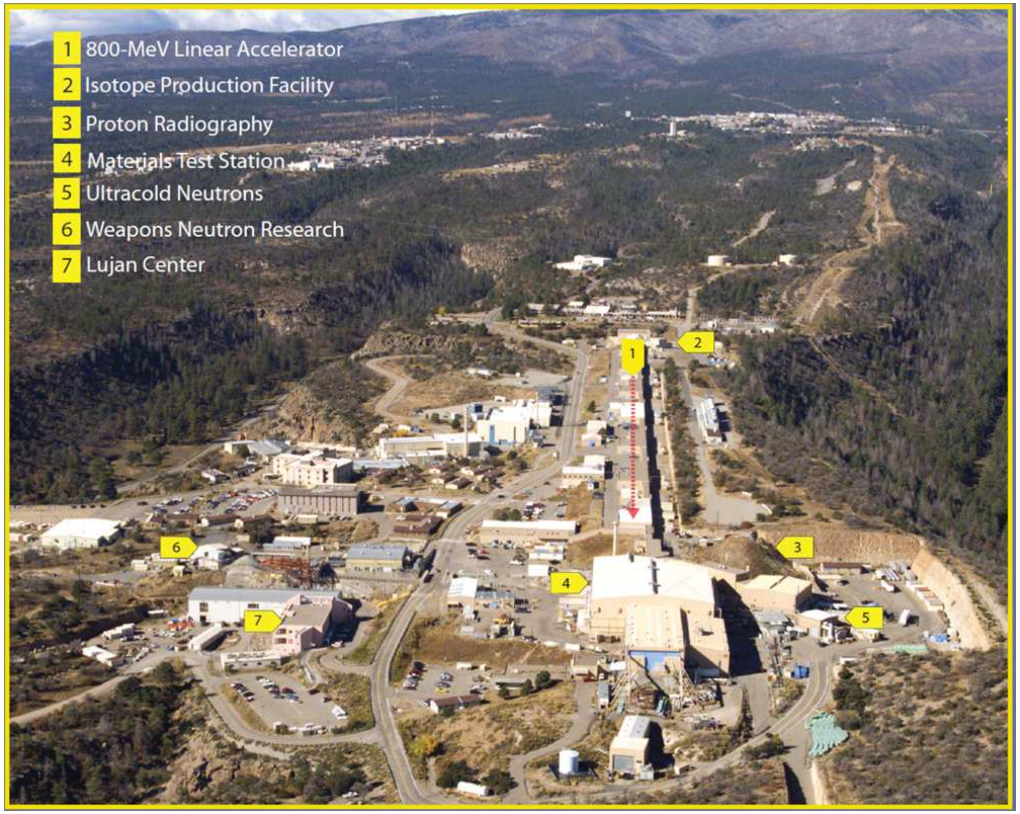
Unfolding with iterative methods support this assumption for simulated response matrices. We are currently evaluating measured data for the purpose of similar unfolding.

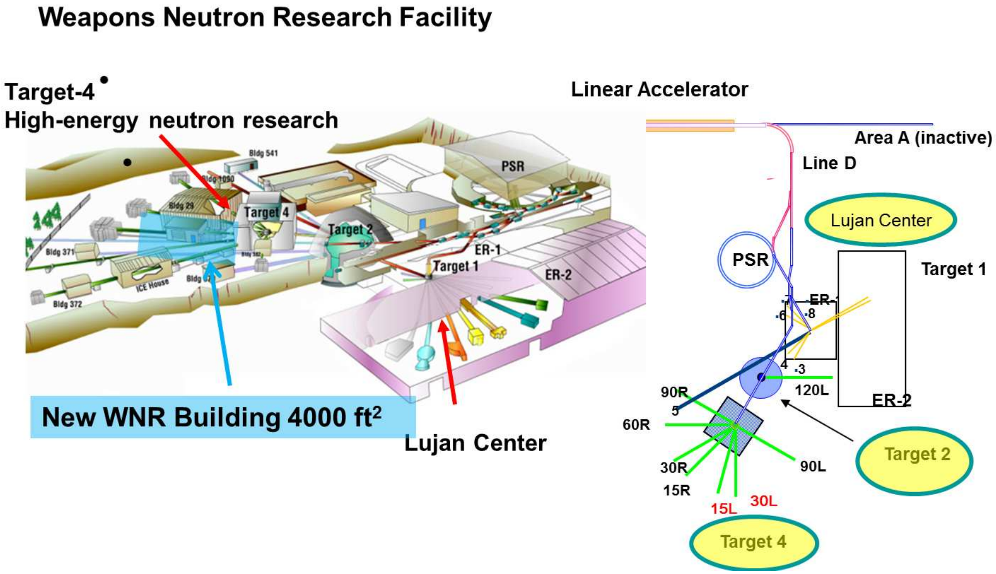
Nuclear Data at LANSCE

Motivation

- ✔ Nuclear data libraries show inconsistencies and a lack of data for relevant induced fission neutron spectra.
- ✔ Main region of interest where data is less reliable is:
  - ✘ Below 1 MeV.
  - ✘ Above 5 MeV.
- ✔ Our liquid scintillation detectors with good pulse shape discrimination (PSD) capabilities can be used in those ranges.
- ✔ Requires a collaborating facility with very specific capabilities and equipment.
- ✔ Collaboration with Dr. Robert Haight of Los Alamos Neutron Science Center (LANSCE), and other universities.

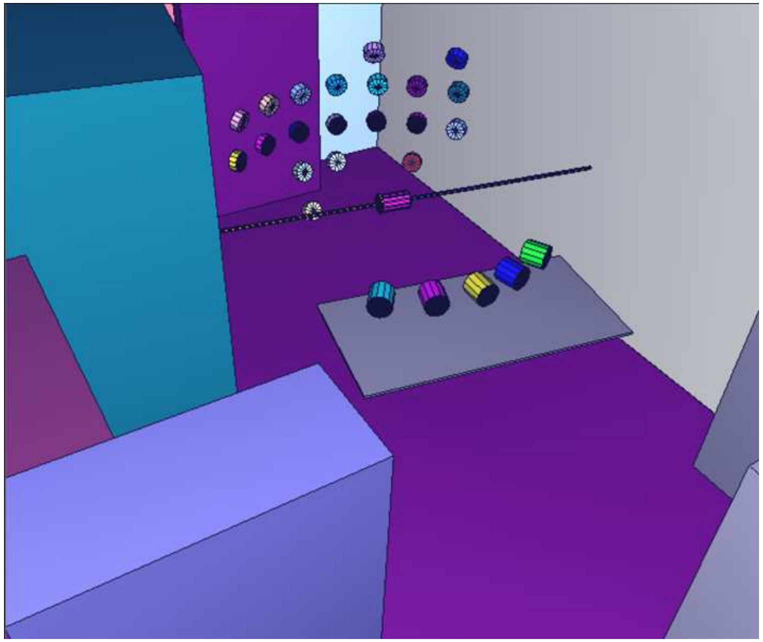
LANSCE: a User Facility





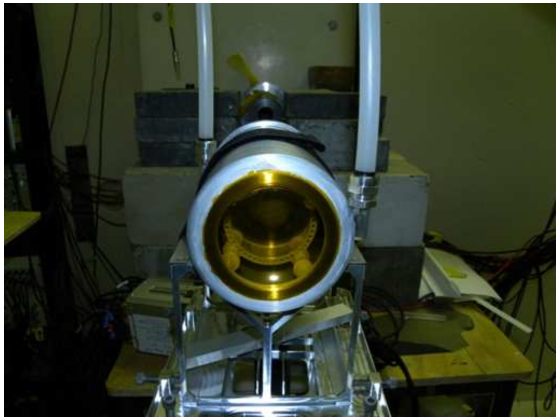
Experimental Setup

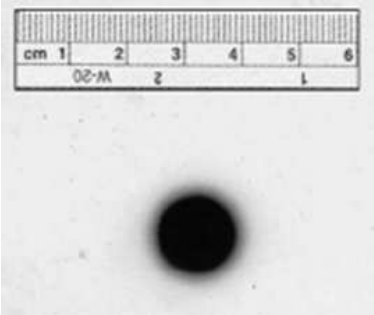
- ✔ WNR 30R beam
- ✔ Flight path 22.657 m
- ✔ White neutron source: neutrons 0.5-600 MeV
- ✔ CAEN V1720, 12-bit, 250-MHz waveform digitizer
- ✔ Five EJ-309 liquid organic scintillation detectors - 80-cm distance between the FC and the detectors
- ✔ Threshold set to ∼50 keVee.
- ✔ ∼1 neutron per second detected (60h total)



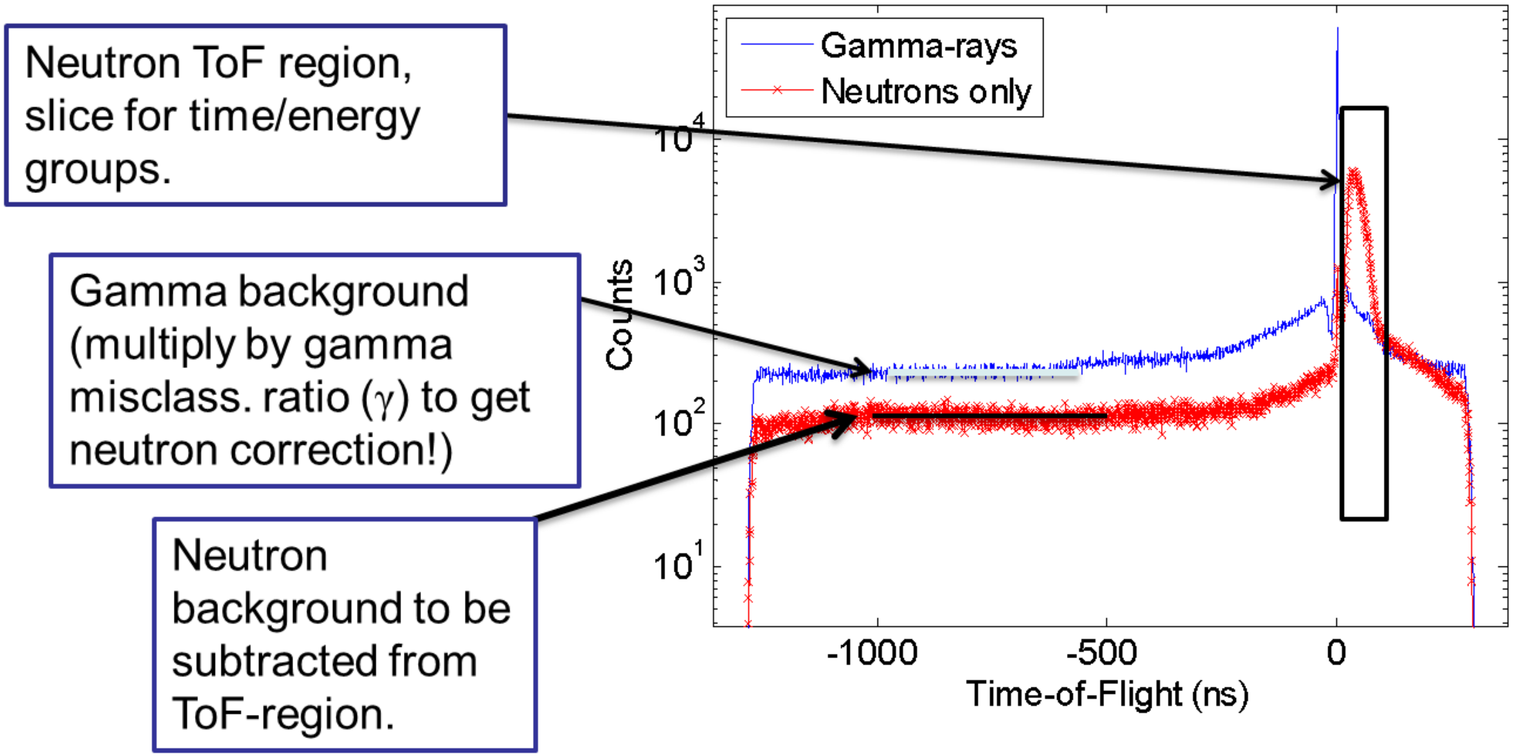
Fission Chamber

- ✔ Parallel plated avalanche chamber (PPAC).
- ✔ 10 plates coated on each side with ^{235}U. 26 triggers per second from alpha decays.
- ✔ Total deposited ^{235}U mass around 112mg.
- ✔ Target are approximately 10cm^2, corresponding beam cross section is on the order of a few square cm.



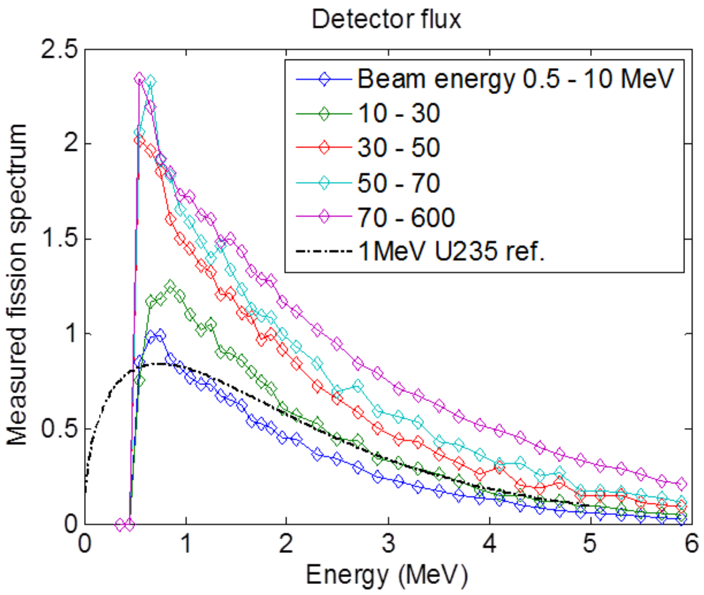


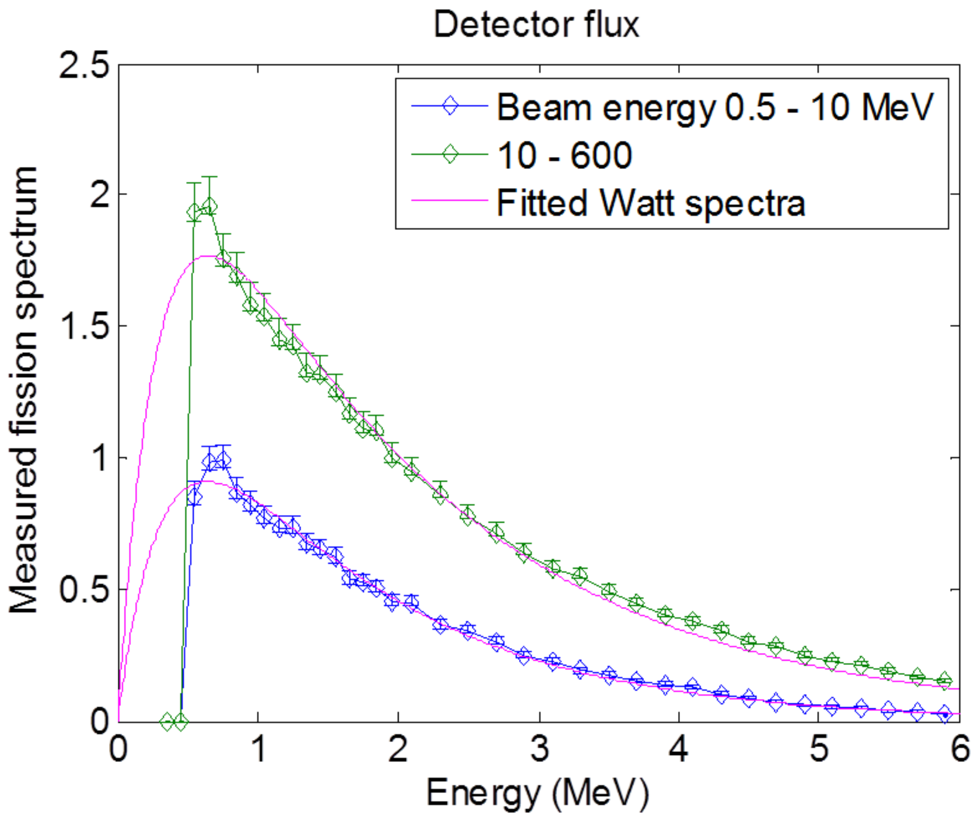
✔ Detected particles as a function of time and particle type (PSD).



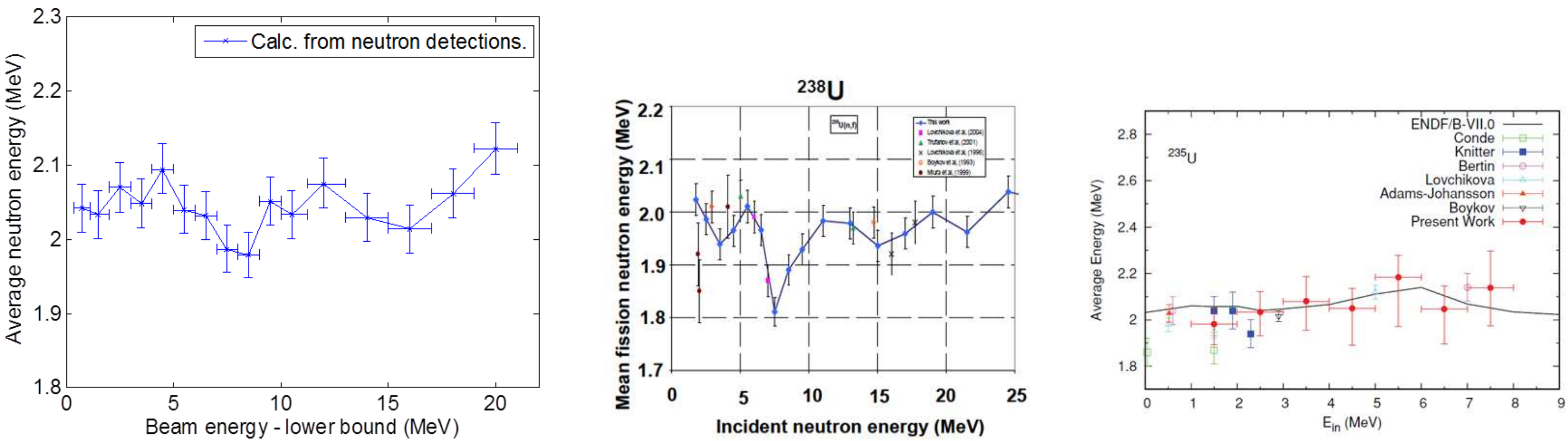
Unfolded Fission Spectrum

- ✔ Dividing data as a function of the incoming beam energy changes the number of emitted fission neutrons and their energy distribution.
- ✔ Absolute fluxes shown.





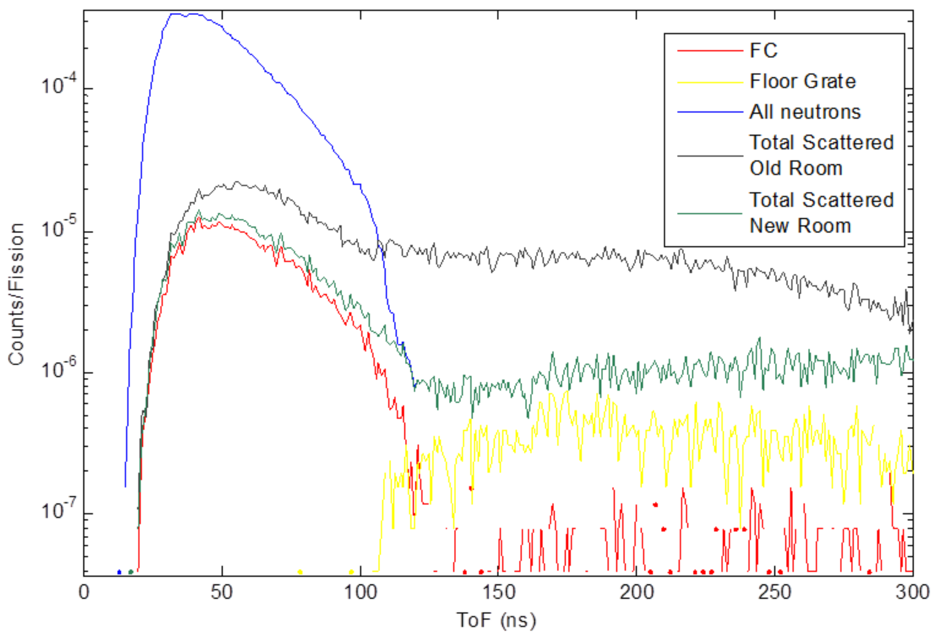
✔ Project results (left); ^{238}U data (center); ^{235}U data (right). Energy calculated from TOF.



✔ (n, n'f) and (n, 2n'f) thresholds at about 7.5 MeV and 15 MeV are observed.

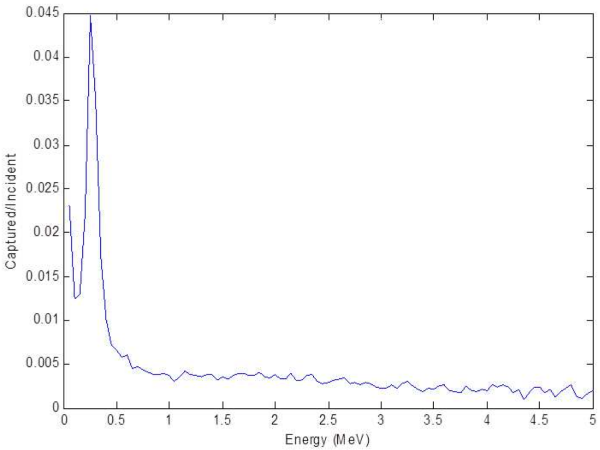
LANSCE Upgrades

- ✔ New facility features a 6-foot deep pit below the fission chamber.
- ✔ This modification results in a significantly lowered amount of neutron scattering in the room.
- ✔ In the lower energy regions (300 keV), a reduction of almost a factor of ten can be seen for the new facility.

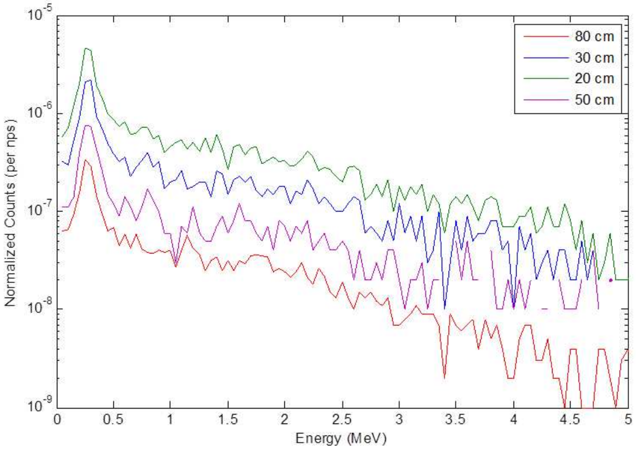


Future measurements

- ✔ New additional detectors:
- ✔ Additional improvement in analysis:
  - ✘ Using compound PSD methods lower energy neutrons can now be detected.
  - ✘ Investigating $\overline{\nu}$ as a function of beam energy.



Efficiency



Count Rate

Conclusions

- ✔ The work has shown the importance of using multiple approaches: models, simulations and experiments all have strengths and weaknesses.
- ✔ Simple physical processes such as neutron scattering gives very complex analyzes and behaviors in for example scintillation detectors.
- ✔ Performing measurements have allowed for a possibility to determine which ideas are applicable, and which are subject to technical limitations.
- ✔ Algorithms and tools of mathematical physics used here, as well as the resulting data, occur not only in the area of safeguards but also other fields such as reactor physics.

Thank You!