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Prompt neutron emission in the neutron-induced fission of ^{239}Pu and the spontaneous fission of ^{252}Cf: an unfolding story

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Prompt neutron emission is one of the major observables in fission and critical to a sustained chain reaction. The prompt neutron emission spectrum and its precision are essential for adequately modeling the chain reaction for any given application. Within this context, a joint program, called Chi-Nu, was initiated by LANL [1] with the objective to improve the precision of experimental measurements of prompt neutron emission for neutron-induced fission of ^{235}U and ^{239}Pu as well as the spontaneous fission of ^{252}Cf. The measurements were carried out at the Weapons Neutron Research Facility at Los Alamos Neutron Science Center using two scintillator arrays for neutron detection and a PPAC (parallel-plate avalanche counter) for fission-fragment detection.

Two neutron detection arrays were developed at LANL [2,3], one for neutron energy below 2.5 MeV with 22 Li-glass scintillators and another one for neutron energy above 700 keV with 54 organic liquid scintillators. A PPAC of multiple target foils with minimal mass construction was developed at LLNL [4] to detect the fission fragments. A total of 10 individual PPAC's was accommodated in one enclosure, each holding a mass up to 10 mg of ^{235}U or ^{239}Pu, which was electrodeposited on both sides of a thin titanium foil of 3 μm thickness [5]. A PPAC with two individual foils were installed for ^{252}Cf with a strength of ~ 2μCi each. This uniquely designed PPAC has a time resolution of ~ 1 ns and the derived timing has no fission-fragment induced time-walk and can be utilized as the time zero for fission occurrence. The time difference between PPAC signal and the beam pulse is a measure of the incident neutron energy for a given flight path. The time difference between PPAC signal and neutron detector signal is a measure of the fission neutron energy. This so-called double time-of-flight technique is used to determine the prompt fission neutron spectrum (PFNS) as a function of incident neutron energy, which is known as the χ matrix.

The measured PFNS requires corrections to detector responses that include the detection efficiency and the multiple-scattering effect from the experimental environment, to obtain the physical spectrum. The detector responses were simulated using the "MCNP-PoliMi", a modified version of MCNP®X, with a Maxwellian input spectrum [6,7,8]. LANL adopted the ratio-of-ratios method to correct the data documented in Ref. [9]. It is done with one iteration after recalculating the simulated detector response using the first corrected PFNS as input to substitute the initial Maxwellian spectrum.

We propose to correct the measured PFNS directly using the unfolding technique with the same detector response matrix for the ratio-of-ratios method, to obtain the physical spectrum. In this report, we limit the discussion to four unfold packages available in the public domain [10] applied to the PFNS data obtained from the array of 54 liquid scintillators for the spontaneous fission of ^{252}Cf and the neutron-induced fission of ^{239}Pu. The preliminary results are presented below.

Bin-by-bin: This unfolding technique is the simplest of those presented here and, in general, is not suitable for use where strong off-diagonal elements exist in the response matrix. In other words, if the response matrix does more than perform a simple efficiency correction and causes a shifting of the data. Since this is a common feature of especially lower-energy neutron spectra, this method should not be used in isolation. That being said, in the energy range over which the liquid scintillators perform well (1-10 MeV) the bin-by-bin unfolding method performed very similarly to the other methods. This is indicative of the predominantly diagonal nature of the detector response in this energy region [11] (see Fig. 1).

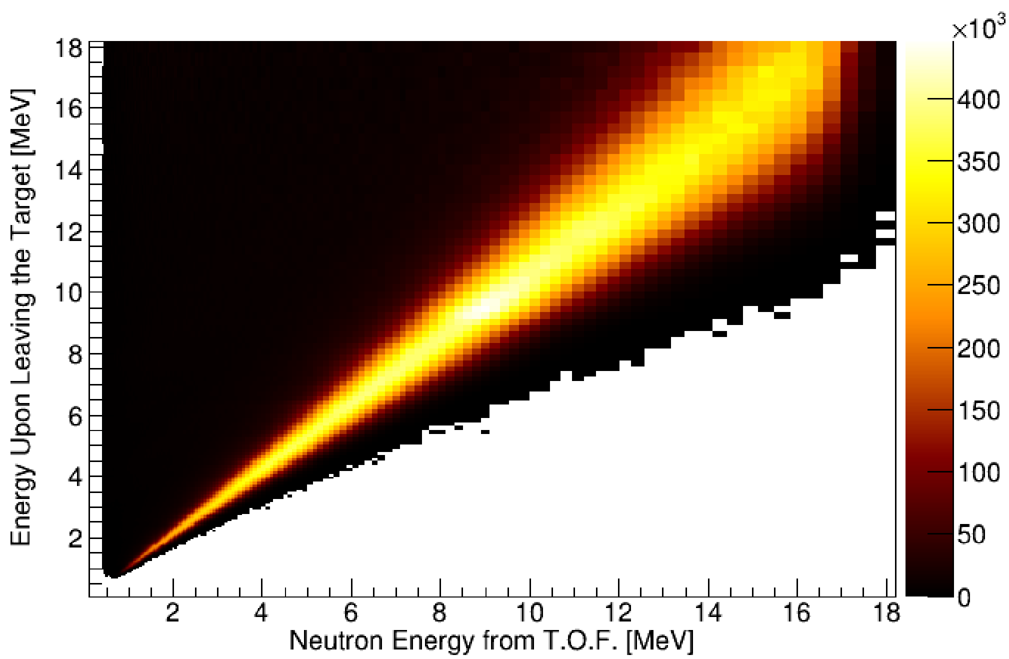

Figure 1: Response matrix for liquid scintillator detectors in the ^{239}Pu(n,f) experiment.

Singular value decomposition (SVD) [11]: This method uses a regularization parameter to correct for induced noise to the final spectrum. The choice of regularization parameter is a particular limitation of the method. One solution to this is to perform simulations using known distributions and choose a parameter such as to best reproduce the data.

Iterative Bayesian [12]: This method uses an iterative technique to overcome a limitation in the traditional Bayesian methods, namely the requirement for a prior. Instead, using this method, one can use a flat initial prior which is iteratively changed. The number of iterations required to achieve convergence is typically low (5-10). Iterations beyond that point are likely to induce noise – albeit while marginally improving the absolute accuracy of the result. For large numbers of iterations therefore, sensitivity to genuine high-frequency events in the data can be difficult to distinguish from those induced by the unfolding.

Maximum-likelihood expectation maximization:

The maximum-likelihood expectation maximization (MLEM) method [13] is widely used to reconstruct data, for example in positron emission tomography to determine the probability distribution of the source position. The method is based on the maximization of the log-likelihood function of the data, defined as:

$$\ln P_r = \sum_{i=1}^{I} \left[-\sum_{j=1}^{J} r_{ij} x_j + y_i \ln \sum_{j=1}^{J} r_{ij} x_j - \ln(y_i!) \right].$$

Where r_{ij} is an element of the response matrix, y_i is the experimentally measured bin content and x_j corresponds to the emitted value. One can construct an algorithm in order to find non-negative values for $\frac{d \ln P_r}{d x_j}$, allowing the calculation of x_j values:

$$x_j^{(k+1)} = \frac{x_j^{(k)}}{\sum_{i=1}^{I} r_{ij}} \sum_{i=1}^{I} r_{ij} \frac{y_i}{\sum_{l=1}^{J} r_{il} x_l^{(k)}}, j = 1, \dots, J$$

where, $x_j^{(k)}$ is the *k*th estimate. This unfolding technique begins with only a response matrix and experimental data, requiring no further prior assumption and increases the likelihood of the unfolded neutron spectrum with every iteration. The convergence of the method is, however, relatively slow and the propagation of uncertainties requires repeating the unfolding method many tens of times, further slowing the process. For thirty bins of experimental data however, fifty iterations with error propagation takes considerably less than a minute. A further issue with the MLEM method is that, while the total likelihood of the unfolded spectrum increases with every iteration, the method does introduce the noise level of the spectrum. The MLEM method behaves similarly to the iterative Bayesian, requiring more iterations but taking a similar time to achieve similar convergence.

An unfolding graphical user interface

The unfolding process is highly repetitive, with a number of different unfolding methods, each with a number of options, such as the number of iterations. Every unfolding method uses common elements however, in particular: an input spectrum (experimental data) and a response matrix. This combination of features makes the unfolding process well suited to a graphical user interface (GUI), allowing a user to select an unfolding mechanism and perform unfolding with minimal input. To this end an unfolding GUI has been created within the ROOT framework, allowing the user a common, easy-to-use format for performing unfolding, shown in Fig. 2.

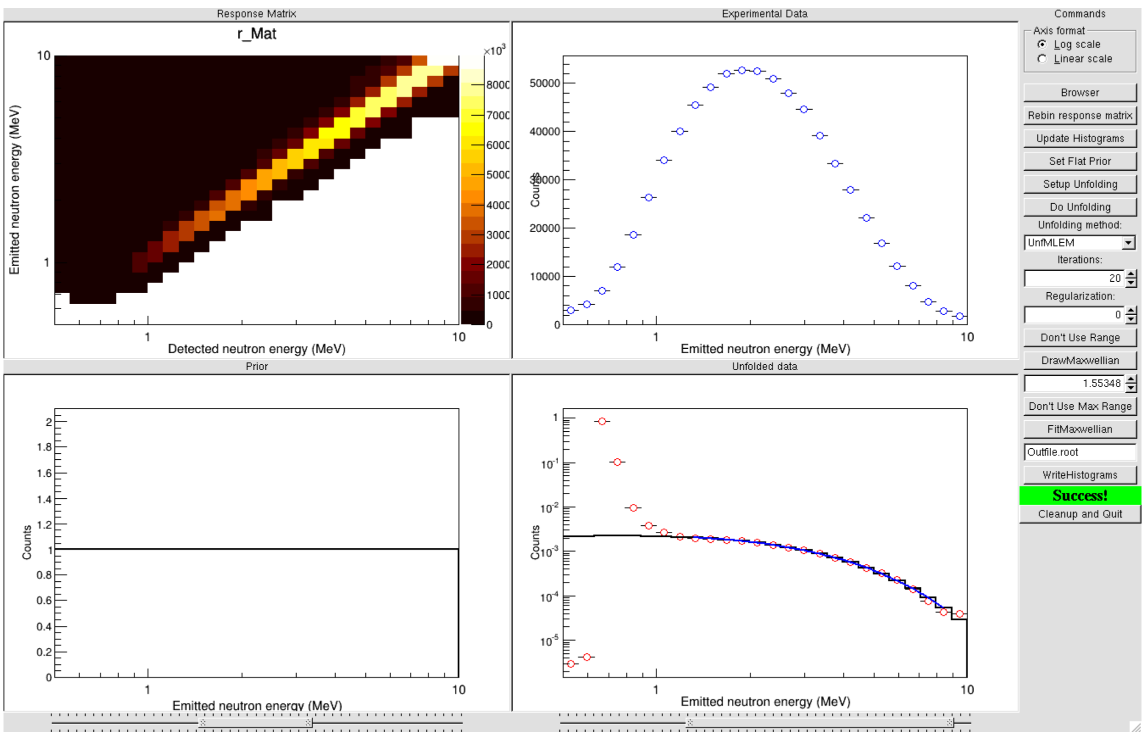

Figure 2. The unfolding GUI developed. Clockwise from top left: Response matrix, experimental data, unfolded spectrum and prior (assuming a Bayesian unfolding method is used). See text for further details. The data in this figure were from ^{239}Pu(n,f) using liquid scintillator detectors and with incident neutron energy of 15-17.5 MeV, unfolded using the maximum likelihood expectation maximization method.

The interface takes experimental data and a user-provided response matrix (selected using the common ROOT TBrowser package). It then rebins the response matrix appropriately and can be used to unfold the data using any of the available unfolding methods: Iterative Bayesian, singular-value decomposition, bin-by-bin and maximum-likelihood expectation maximization. The first three options are part of the RooUnfold package while the latter was developed as part of this work and is discussed elsewhere in this report. In addition to unfolding the experimental data, the GUI also allows for simple functionalities such as saving the spectra to a file and fitting them with a Maxwellian curve, either with temperature as a free variable or with a fixed temperature. The range over which this fit is performed can be easily varied by the user.

Summary: Common to all non-trivial (i.e. excluding bin-by-bin) unfolding methods is a strong sensitivity to the correct propagation of experimental energy thresholds, either physical (trigger/resolution thresholds) or analysis based (locations of particle ID cuts, for example). In Figures 3 and 4 for ^{239}Pu(n,f) and ^{252}Cf(SF), respectively, all three methods exhibit a significant excess compared to a Maxwellian below 1.3 MeV. However, all three methods show excellent agreement between 1.3 MeV and 10 MeV, giving good confidence in the results in that region. The exact origin of the behavior below 1.3 MeV is still being investigated.

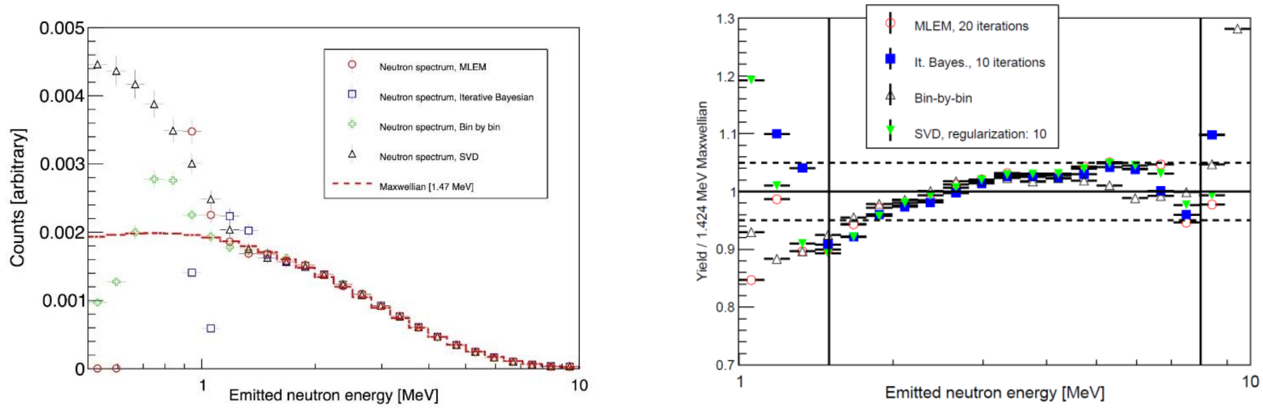

Figure 3. Comparison of unfolding methods for ^{239}Pu(n,f) liquid scintillator data with incident neutron energies of 3-4 MeV. MLEM performed with 40 iterations, Iterative Bayesian with 10 iterations and SVD with a regularization parameter of 10. These values are approximately optimal for convergence. A 1.47 MeV Maxwellian is shown for comparison and corresponds to the approximate best fit for the MLEM data presented here between 3-10 MeV but should be considered preliminary. (R) The unfolded data from various unfolding methods were normalized to 1.424 MeV Maxwellian, the current adopted value. The dashed lines indicate a deviation of $\pm$5%.

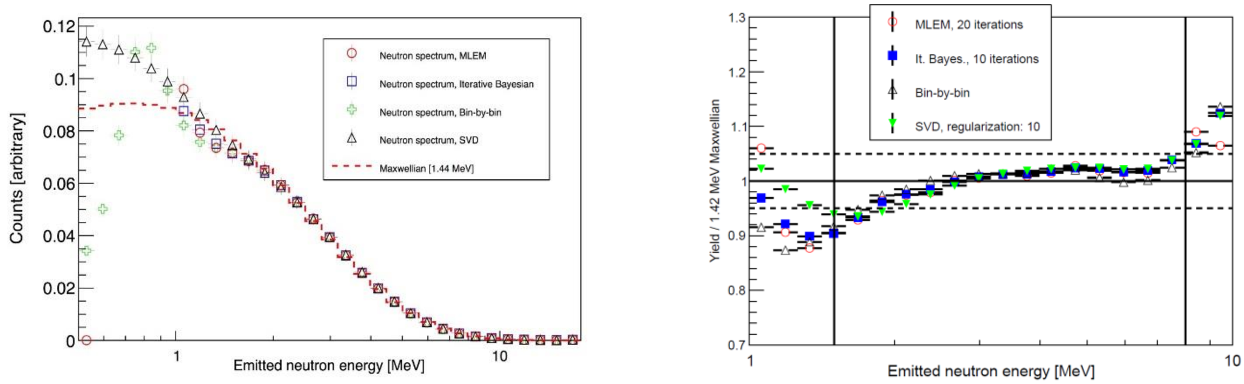

Figure 4. Comparison of unfolding methods for ^{252}Cf spontaneous fission neutron spectrum. (L) MLEM performed with 40 iterations, Iterative Bayesian with 10 iterations and SVD with a regularization parameter of 10. These values are approximately optimal for convergence. A 1.44 MeV Maxwellian is shown for comparison and corresponds to the approximate best fit for the MLEM data presented here between 3-10 MeV but should be considered preliminary. (R) The unfolded data from various unfolding methods were normalized to 1.42 MeV Maxwellian, the current adopted value. The dashed lines indicate a deviation of $\pm$5%.

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References:

[1] R.C. Haight, C.Y. Wu, H.Y. Lee *et al*., Nucl. Data Sheets 123, 130 (2015)

[2] R.C. Haight, H.Y. Lee, T.N. Taddeucci *et al*., Journal of Instrumentation 7, C03028 (2012)

[3] H.Y. Lee, T.N. Taddeucci, R.C. Haight *et al*., Nucl. Instrum. Methods Phys. Res. A 703, 213 (2013)

[4] C.Y. Wu, R.A. Henderson, R.C. Haight *et al*., Nucl. Instrum. Methods Phys. Res. A 794, 76 (2015)

[5] R.A. Henderson, J.M. Gostic, J.T. Burke, S.F. Fisher, and C.Y. Wu, Nucl. Instrum. Methods Phys. Rese. A 655, 66 (2011)

[6] S.A. Pozzi, E. Padovani, and M. Marseguerra, Nucl. Instrum. Methods Phys. Res. A 513, 550 (2003)

[7] D.B. Pelowitz, J.W. Durkee, J.S. Elson *et al*., Los Alamos National Laboratory Report No LA-UR-11-02295 (2011)

[8] T.N. Taddeucci, R.C. Haight, H.Y. Lee *et al*., Nucl. Data Sheets 123, 135 (2015)

[9] M. Devlin, J.A. Gomez, K.J. Kelly *et al.*, Nucl. Data Sheets 148, 322 (2018)

[10] http://hepunx.rl.ac.uk/~adye/software/unfold/RooUnfold.html

[11] K.J. Kelly, J.M. O'Donnell, J.A. Gomez *et al.*, Nucl. Instrum. Methods Phys. Res. A 866, 182 (2017)

[12] A. Hocker and V. Kartvelishvili, Nucl. Instrum. Methods Phys. Res. A 372, 469 (1996)

[13] G.D. Agostini, Nucl. Instrum. Methods Phys. Res. A 362, 487 (1995)

[14] B.Pehlivanovic, Radiation Measurements 49, 109 (2013)