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Title: Monte Carlo Hauser-Feshbach Calculations of Prompt Fission Neutrons and Gamma Rays: Application to Thermal Neutron-Induced Fission Reactions on U-235 and Pu-239

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Monte Carlo Hauser-Feshbach Calculations of Prompt Fission Neutrons and Gamma Rays: Application to Thermal Neutron-Induced Fission Reactions on ^{235}U and ^{239}Pu

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Abstract

We have developed a new code `CGMF` , implementing a Monte Carlo version of the Hauser-Feshbach statistical theory of nuclear reactions and applied to the de-excitation of fission fragments. A parallel version of the code has been developed as well, and was used to produced the results presented in this report. Prompt fission neutron and gamma-ray characteristics have been computed for the thermal neutron-induced fission reactions on ^{235}U and ^{239}Pu. We discuss the status of these calculations and how the results compare to available experimental data.

1 Hauser-Feshbach Theory to Predict Prompt Fission Particle Emissions

Modern evaluated nuclear data libraries contain information on prompt fission neutrons, and possibly gamma rays, only through average quantities, e.g., the average prompt fission neutron spectrum (PFNS) and average prompt fission neutron multiplicity (PFNM). Moreover, those quantities are often obtained through simplified models or through experimental data alone.

The Madland-Nix or Los Alamos (LA) model [1] has been widely used to evaluate PFNS for a wide range of actinides, and for incident neutron energies from thermal up to 20 or 30 MeV. Several improvements have been implemented over the years [2, 3, 4], leading to better fits to measured PFNS. However, the LA model is limited in its scope to the prediction of average PFNS and PFNM, and does not say anything about prompt gamma rays.

The statistical Hauser-Feshbach theory [5] is very well suited to describe the evaporation of primary fission fragments, which can be considered as compound nuclei shortly after scission. They are formed in an excited nuclear configuration, and will tend to reach a ground-state or isomeric state by emitting neutrons and gamma rays.

A Monte Carlo implementation of the Hauser-Feshbach theory allows the prediction of distributions and correlations, difficult to extract from traditional deterministic Hauser-Feshbach codes, such as GNASH or TALYS. Quantities such as P(ν), $\langle\nu\rangle$(A,TKE), n-n correlations, etc, have been simply ignored in evaluated data files, as they are difficult to measure and could not be predicted from previous theoretical tools.

1.1 Monte Carlo Hauser-Feshbach Code: `CGMF`

Recently, we have developed a new code, `CGM` [6] that implements a Monte Carlo version of the Hauser-Feshbach equations. While `CGM` was primarily intended as a complemented to more traditional Hauser-Feshbach deterministic codes, no focus was placed on fission. Combining `CGM` with another T-2 code, `FFD` [7], which has been used to compute the de-excitation of fission fragments in a Weisskopf approximation, the `CGMF` code extends `CGM` capabilities by including two entirely new C++ classes to treat the question of fission fragment evaporation.

Because Monte Carlo sampling is more time-consuming than traditional HF codes, we have developed a parallel version of `CGMF` , using MPI calls. It has been tested successfully on our T-2 cluster composed of 4 nodes of 32 processors. This code has been used to obtain all the results discussed in this report.

*** This represented a FY12 Level 3 milestone (M3FT-12LA0210025).*

1.2 Monte Carlo Hauser-Feshbach Input Parameters

While the LA model requires only a handful of input parameters, Monte Carlo Hauser-Feshbach calculations require a vast array of input data. Some of them are indeed model input parameters, which can be varied to best fit experimental data, while others are simply input data that cannot be tempered with as easily, e.g., nuclear structure or fission fragment yields.

Performing `CGMF` calculations require the knowledge of the initial conditions of the fission fragments, right after scission. In particular, the yields of primary fission fragments, i.e., prior to prompt neutron emission, have to be known for each mass and charge of the fragments, and for each fragment, the initial excitation energy distribution as well as spin and parity.

Such data simply cannot be predicted by theory at this time, and one has to use experimental data or/and systematics commonly derived from known experimental data. This is the case for the yields $Y(A, Z, TKE)$ that can only be reconstructed from data.

The initial excitation energy, spin and parity distribution $P(U, J, \pi)$ is known neither from experiment nor theory. For a given pair of fission fragments, if the total kinetic energy TKE is known, then one can infer the total excitation energy available in the fission reaction. However, the exact partitioning of this total energy among the two fragments remains unknown. We have introduced a parameter to describe this partitioning.

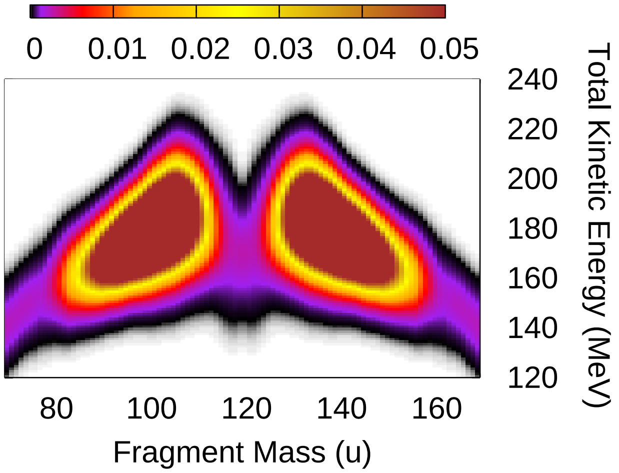

Figure 1: Primary fission fragment yields as a function of mass and total kinetic energy formed in the thermal neutron-induced fission reaction of ^{239}Pu. The units of the color scale are in percent. See Ref. [7] for details on how this distribution was reconstructed from experimental data.

2 Numerical Results

Results similar to the ones presented below are discussed in a paper recently submitted for publication in Physical Review C [8].

2.1 Pu-239 (n_{th}, f) Reaction

The fission fragment yields $Y(A, Z, TKE)$ used in the present work were obtained following the approach described in [7]. The primary fission fragment yields as a function of mass and total kinetic are shown in Fig. 1.

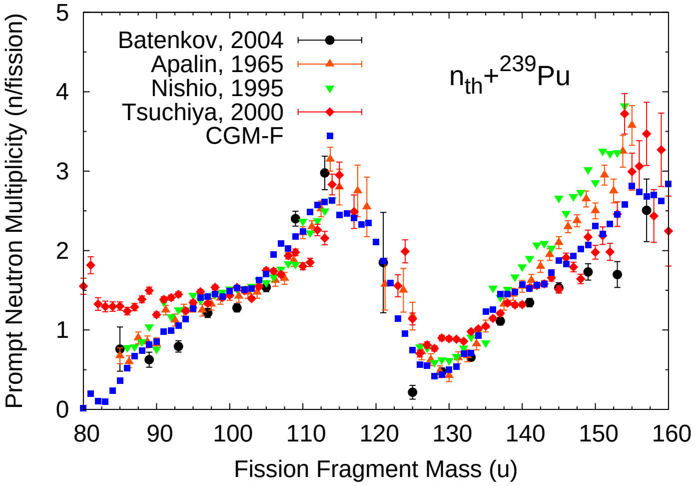

Figure 2: The Monte Carlo Hauser-Feshbach-calculated (`CGMF` code) average neutron multiplicity as a function of the fragment mass is compared to available experimental data.

The results of the `CGMF` calculations are illustrated in Figs. 2−4 for the prompt neutrons, and Figs. 5−6 for the gamma rays.

The average neutron multiplicity as a function of the fragment mass is fairly well reproduced by the `CGMF` calculations. However, the calculated average neutron multiplicity is too high (about 5-10%) and the multiplicity distribution $P(\nu)$ fails to reproduce experimental data, especially for $\nu = 2$ (see Fig. 3). The average prompt fission neutron spectrum also deviates significantly from the ENDF/B-VII.0 evaluation (see Fig. 4) and we are investigating different hypotheses to explain this result. Note that results obtained with a simpler Weisskopf spectrum led to a much better agreement with experimental data and evaluations [7].

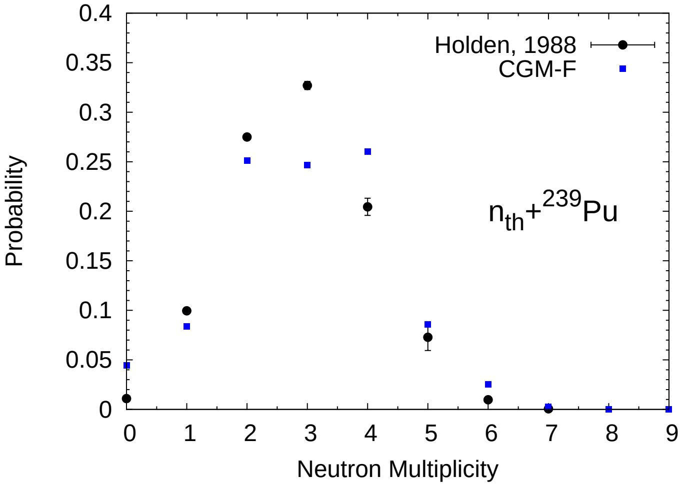

Figure 3: The calculated prompt fission neutron multiplicity distribution $P(\nu)$ for the thermal neutron-induced fission reaction on ^{239}Pu is compared to the evaluation by Holden and Zucker [9]. The results of `CGMF` calculations are not as good as the ones obtained in the simpler Weisskopf approach [7]. We are still investigating this deficiency for the production of prompt neutrons.

While the calculated results for the prompt neutrons are not as satisfactory as one would hope for, the results for the prompt fission gamma rays should not be impacted as much, since they are mostly emitted at a later stage of the evaporation process, except for average quantities such as average gamma-ray multiplicity $\langle N_\gamma \rangle$ and average total gamma-ray energy $\langle E_\gamma \rangle$. The average prompt fission gamma ray spectrum calculated for the first time in a Monte Carlo Hauser-Feshbach formalism is shown in Fig. 5 and compared to the ENDF/B-VII.0 evaluation based entirely on the experimental data by Verbinski *et al.* [10]. While the figure is in log-log, the agreement between calculations and experiment is nevertheless reasonable, in particular given the target accuracy of current nuclear applications. In fact, very little has been done so far to evaluate prompt fission gamma rays beyond what is known from very scarce experiments and for very few important isotopes.

Finally, the calculated prompt fission gamma-ray multiplicity distribution $P(N_\gamma)$ is plotted in Fig. 6. It follows quite well the negative binomial model proposed by Valentine [11] to describe experimental data by Brun-

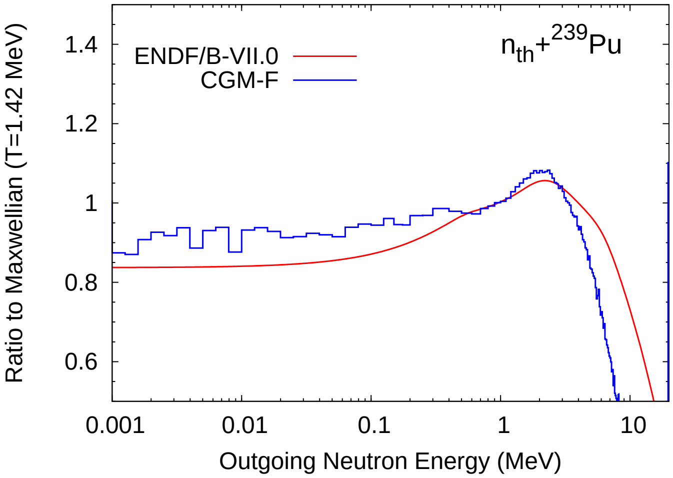

Figure 4: The prompt fission neutron spectrum in the laboratory frame is shown as a ratio to a Maxwellian at temperature T=1.42 MeV, and compared to the current ENDF/B-VII.0 evaluation, based on Los Alamos model calculations tuned to fit available experimental data. As for the prompt neutron multiplicity distribution (Fig. 3), the evaluated spectrum is not well reproduced by `CGMF` calculations.

son [12].

2.2 U-235 (n_{th}, f) Reaction

Similar results were obtained for the thermal neutron-induced fission on ^{235}U. In this case, the initial fission fragment distribution in mass and total kinetic energy $Y(A, TKE)$ were taken directly from experimental data by Romano *et al.* [13].

The evaluated prompt fission neutron multiplicity distribution for this reaction is much better reproduced by the `CGMF` calculations than for $n_{th}+^{239}$Pu, as seen in Fig. 7. The average neutron multiplicity remains about 5% too high though. The prompt fission gamma-ray spectrum is shown in Fig. 8 and is compared to experimental data by Peelle and Maienschein [14]. The agreement is very good almost throughout the entire outgoing energy range. Only at the lowest energies do the `CGMF` calculations over-estimate the observed gamma-ray spectrum.

Another test for the validity of the calculations is to compute the average gamma-ray energy as a function of the fission fragment mass, $\langle E_\gamma \rangle (A)$. The

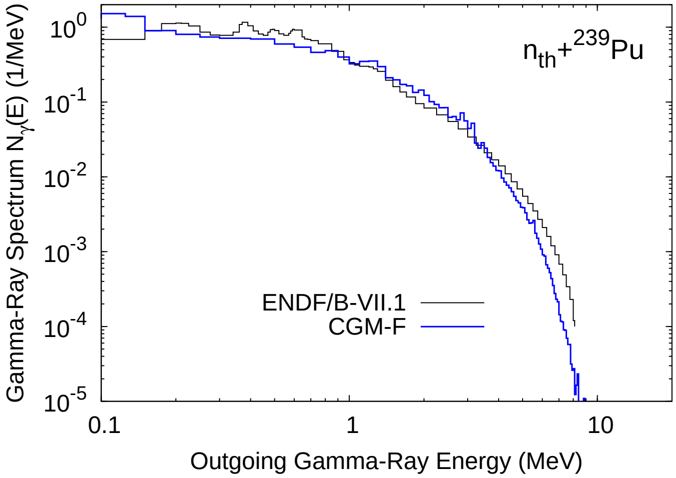

Figure 5: The `CGMF` -calculated prompt fission gamma spectrum is compared to the ENDF/B-VII.0 evaluation, based entirely on the experimental data by Verbinski *et al.* [10]. The agreement between calculation and experiment is reasonable for the target accuracy of current nuclear applications.

results are shown in Fig. 9. The evolution of $\langle E_\gamma \rangle$ as a function of the fragment mass clearly follows our expectations: near closed-shell fragments, the average gamma-ray energy increases, reflecting a higher temperature or lower level density in those fragments.

3 Monte Carlo Hauser-Feshbach Calculations and DANCE Experimental Data

Results of Monte Carlo Hauser-Feshbach calculations for the prompt fission gamma rays have been provided to experimentalists at the Los Alamos Neutron Science Center, working with the DANCE 4π-calorimeter. While the prompt fission gamma-ray spectrum cannot be measured directly in DANCE, a gamma-ray energy spectrum and multiplicity, convoluted with the complicated DANCE detector response, is measured. Therefore, a possible comparison of calculation vs. experiment is to perform a forward propagation of model-calculated prompt gamma rays, perform a transport simulation of the DANCE detector response, and compare the final product with the measured data. This work was done by M. Jandel *et al.* for n_{th}+^{235}U [15] and by J. Ullmann *et al.* for n_{th}+^{239}Pu [16].

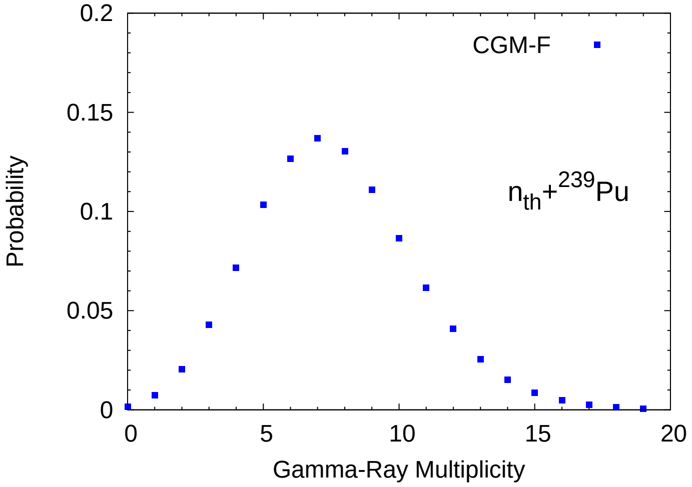

Figure 6: The prompt fission gamma-ray multiplicity distribution $P(N_\gamma)$ as calculated in the Monte Carlo Hauser-Feshbach formalism. It follows nicely the Negative Binomial model proposed by Valentine [11].

In Fig. 10, the results for the prompt fission gamma-ray multiplicity (left), energy (center), and total gamma-ray energy (right) are shown for the ^{235}U case. Different `CGMF` calculations using different α_I parameter values correspond to different initial spin distributions. A value of 1.0 mean that the rigid-body moment of inertia is used for all fragments to infer the spin distribution. A higher value for α_I translates into a higher average initial spin in the fragments. The agreement between experiment and calculations is reasonable, but the calculated spectra are always softer than the experimental values, most noticeably above 3 MeV.

Figure 11 shows the primary prompt fission gamma-ray spectrum calculated with `CGMF` (red), compared to a simpler parameterization of the gamma-ray cascade (blue points) that lead to an excellent agreement between data and propagated calculations through a `GEANT4` simulation. The agreement between the red curve and blue points is remarkable, and deviate significantly from the existing evaluation in the ENDF/B-VII.0 library.

4 Conclusion

We have developed a new code, `CGMF` , implementing the Hauser-Feshbach statistical theory of nuclear reactions for the de-excitation of fission frag-

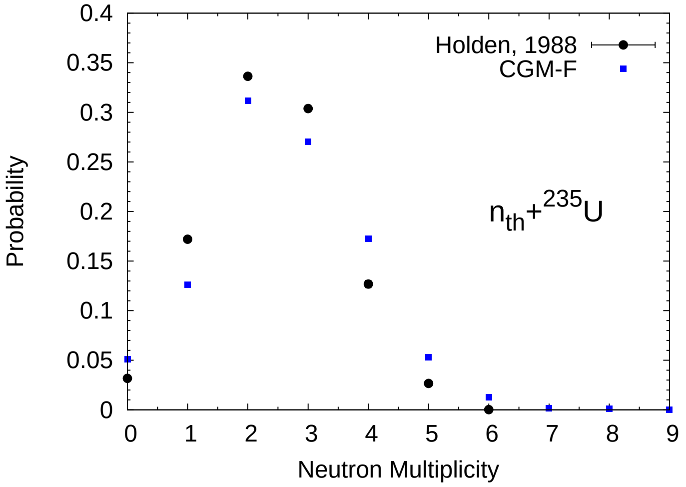

Figure 7: The calculated prompt fission neutron multiplicity distribution $P(\nu)$ for the thermal neutron-induced fission reaction on ^{235}U is compared to the evaluation by Holden and Zucker [9]. The agreement is reasonable, although the calculated average neutron multiplicity is slightly higher than the evaluated value.

ments, using the Monte Carlo sampling technique. Furthermore, this code has been parallelized and is currently used on a small cluster of 32x4 processors.

We have used `CGMF` to produce all results shown in this report, for thermal neutron-induced fission on ^{235}U and ^{239}Pu. The results include, but are not limited to: average prompt fission neutron spectrum $\langle\chi\rangle$ and multiplicity $\overline{\nu}$, average neutron multiplicity as a function of fragment mass $\overline{\nu}(A)$, prompt fission neutron multiplicity distribution P(ν), average prompt fission gamma spectrum $\langle\chi_\gamma\rangle$ and multiplicity $\langle N_\gamma\rangle$, average gamma-ray multiplicity $\langle N_\gamma\rangle$ and energy $\langle\epsilon_\gamma\rangle$ as a function of the fission fragment mass, prompt fission gamma-ray multiplicity distribution P(N_γ).

The agreement between `CGMF` calculations and experimental data varies: for both n$_{th}$+^{239}Pu and n$_{th}$+^{235}U, the calculated average neutron spectra are much softer than the evaluated and experimental results, and the calculated average neutron multiplicity is too low. It is interesting to note that our previous results [7] obtained with the simplified Weisskopf-type evaporation mechanism led to much better results for the prompt neutrons. The present results for ^{235}U are better than the ones obtained for ^{239}Pu though.

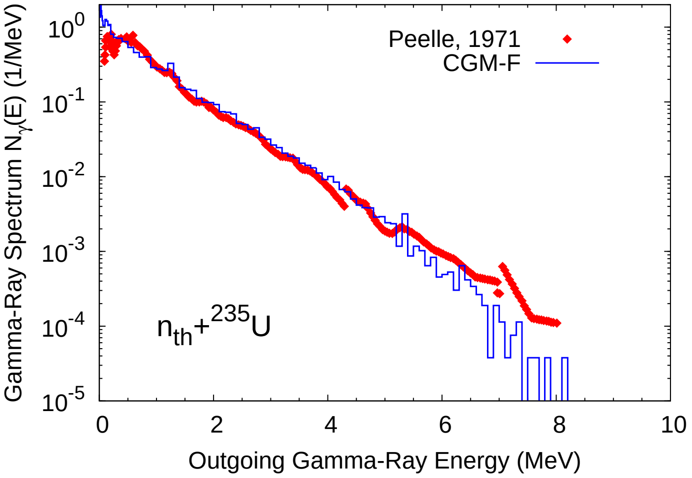

Figure 8: The `CGMF` -calculated average prompt fission gamma ray spectrum for the thermal neutron-induced fission reaction on ^{235}U is compared to experimental data by Peelle and Maienschein [14]. The agreement is very good almost throughout the entire range of outgoing energies, except below 500 keV where the calculated spectrum is too high.

For prompt fission gamma rays, however, `CGMF` calculations are in very reasonable agreement with experimental data, at least within the target accuracies required for current applications, and which are much less stringent than for neutrons at this point. For both fission reactions, the evaluated prompt fission gamma-ray multiplicity distributions P(N_γ) are very well reproduced in our calculations. The PFGS is also quite reasonable, especially given the large uncertainties in the existing experimental data.

Several important questions remain in order to produce a very robust and predictive capability. In particular, the energy sorting mechanisms at play near scission, and which determine the initial energy distribution in each fragment, remain poorly known. In addition, Monte Carlo Hauser-Feshbach calculations require the initial knowledge of the spin and parity distribution in the fragments. Little is known about these quantities, and only limited and indirect experimental data are available for comparison. We are currently working on the calculation of isomeric ratios in fission products, for which limited data exist. Such calculations lead to a better understanding of the initial conditions in the primary fission fragments.

*** This report represents the completion of our Level3 Milestone M3FT-*

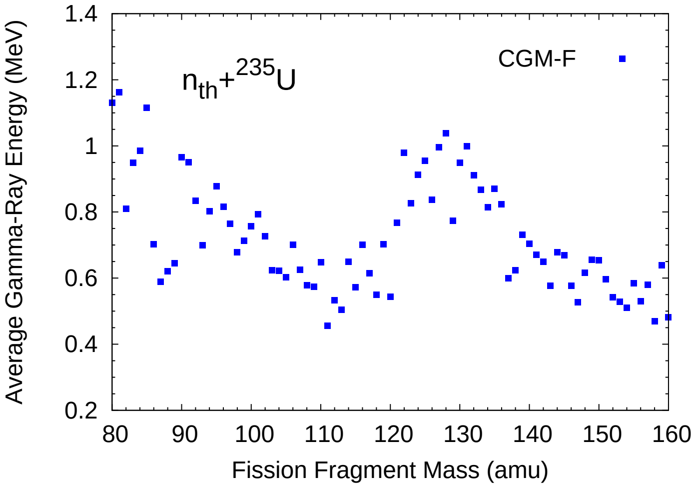

Figure 9: The average prompt fission gamma-ray energy is plotted as a function of the fission fragment mass. As expected, the average energy increases near closed-shell fragments for which the level density is lower.

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Acknowledgements

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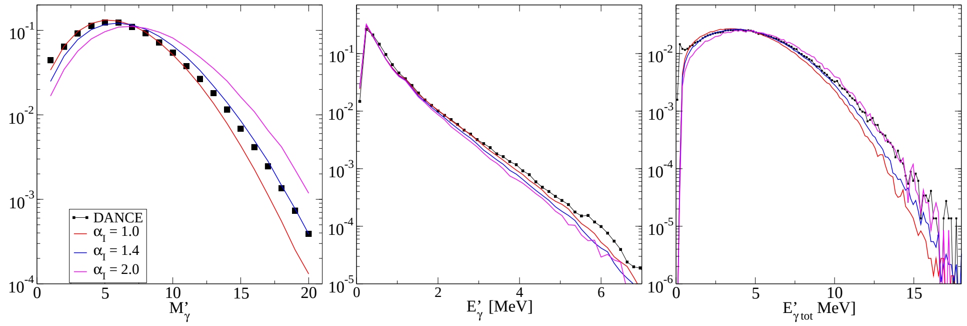

Figure 10: Comparison of DANCE experimental data on prompt fission gamma-ray multiplicity (left), energy (center) and total gamma-ray energy (right) with `CGMF` results convoluted with the DANCE detector response through a `GEANT4` simulation. The different α_I values correspond to different spin distribution assumptions. A higher α_I value means higher average initial spin in the fragments. (Courtesy of M. Jandel, LANL)

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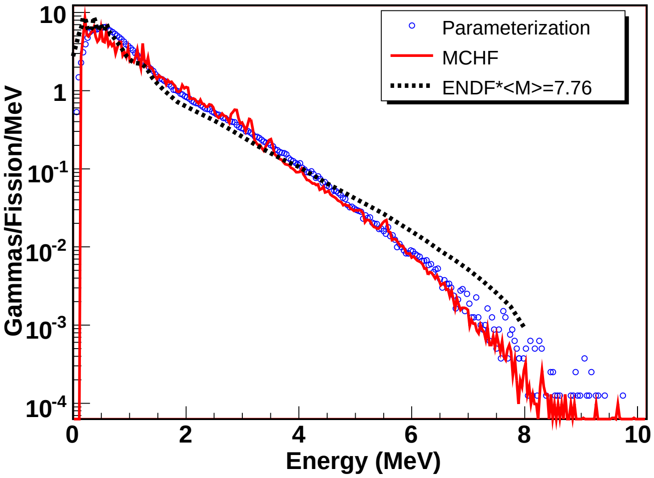

Figure 11: The primary prompt fission gamma-ray spectrum calculated in the Monte Carlo Hauser-Feshbach formalism (red line) is compared to a simplified parameterization (blue points) that lead to an excellent agreement between `GEANT4`-propagated results and the DANCE experimental data. Also, the ENDF/B-VII.0 evaluated spectrum is shown for comparison (black dotted line). (Courtesy of J. Ullmann, LANL)