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Author(s): P. Talou

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Advanced Modeling of Prompt Fission Neutrons

P. Talou

T-2 Group, Nuclear and Particle Physics, Astrophysics and Cosmology, Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

Abstract. Theoretical and numerical studies of prompt fission neutrons are presented. The main results of the Los Alamos model often used in nuclear data evaluation work are reviewed briefly, and a preliminary assessment of uncertainties associated with the evaluated prompt fission neutron spectrum for n (0.5 MeV)+^{239}Pu is discussed. Advanced modeling of prompt fission neutrons is done by Monte Carlo simulations of the evaporation process of the excited primary fission fragments. The successive emissions of neutrons are followed in the statistical formalism framework, and detailed information, beyond average quantities, can be inferred. This approach is applied to the following reactions: ^{252}Cf (sf), n_{th}+^{239}Pu, n (0.5 MeV)+^{235}U, and ^{236}Pu (sf). A discussion on the merits and present limitations of this approach concludes this presentation.

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INTRODUCTION

Prompt fission neutrons and γ rays are emitted when excited primary fission fragments release their energy to reach a more stable configuration. This happens quickly after the separation of the two fragments and their acceleration due to Coulomb repulsion. Studying prompt neutrons and γ-rays is important not only for deepening our understanding of the nuclear fission process, in particular near the scission point, but also for providing quality data for applied needs (e.g., advanced nuclear reactor concepts).

In this paper, the modeling of prompt neutrons is reviewed through the so-called Los Alamos (LA) model, ubiquitous in nuclear data evaluation works, and through advanced computations that follow the detailed chain of successive neutron emissions. Note that the emission of prompt γ-rays is not treated explicitly in the present work. Because of its importance for current and near-term applications, the quantification of uncertainties associated with Los Alamos model calculations for the ENDF/B-VII library is presented. Both the methodology and preliminary results are discussed.

Results of Monte Carlo simulations are presented for selected reactions: ^{252}Cf (*sf*), n_{th}+^{239}Pu, n (0.5 MeV)+^{235}U, and ^{236}Pu (sf).

THEORETICAL MODELING

The Los Alamos Model

The main results of the Los Alamos or Madland-Nix model [1] are briefly reviewed in this Section. Note that several generalizations of the LA model have been developed over the years for data evaluation work [2, 3], but all of them still use the basic LA equations presented below.

In the Los Alamos model, the neutron energy spectrum $N(E)$, in the laboratory frame, for a fission fragment moving with a kinetic energy per nucleon E_f reads

$$N(E) = \frac{1}{2\sqrt{E_f}T_m^2}\int_{(\sqrt{E}-\sqrt{E_f})^2}^{(\sqrt{E}+\sqrt{E_f})^2} \sigma_c(\varepsilon)\sqrt{\varepsilon}d\varepsilon \times \int_0^{T_m} k(T)Texp(-\varepsilon/T)dT, \qquad (1)$$

where $k(T)$ is the temperature-dependent normalization constant given by

$$k(T)^{-1} = \int_0^{\infty} \sigma_c(\varepsilon)\ \varepsilon\ exp(-\varepsilon/T)d\varepsilon. \qquad (2)$$

$\sigma_c(\varepsilon)$ is the energy-dependent cross section for the inverse process of compound nucleus formation, and T_m is the maximum value of the triangular distribution of temperatures used to describe the excitation energy distribution in the fragments.

Equation (1) was derived under the assumption of prompt neutron emissions occurring from fully accelerated fragments only. Also, the original LA model assumes that the temperatures in the light and heavy fragments are the same.

Considering the most probable fragmentation only, the average neutron energy spectrum in the laboratory frame is given by an average over the spectra for the light and heavy fragments

$$N(E) = \frac{1}{2}\left(N_L(E) + N_H(E)\right), \qquad (3)$$

assuming an equal number of neutrons emitted from the light and heavy fragments.

At higher incident energies, these formulas are generalized to take into account the onset of multiple-chance fissions, i.e. the probability that one or more neutrons are emitted before the residual compound nucleus fissions. The neutron energy spectrum can then be expressed as

$$N(E) = \sum_i P_f^i \times \left(\sum_{j=1}^{i-1} \phi_j(E) + \overline{\nu}_i N_i(E)\right) / \sum_i P_f^i \times (i-1+\overline{\nu}_i), \qquad (4)$$

where $\phi_j(E)$ is the pre-fission evaporation spectrum for the $(j+1)^{th}$-chance fission channel, and P_f^i is the i^{th}-chance fission probability.

The LA model has been very successful in the nuclear data evaluation work due its ability to compute and predict the average prompt neutron spectrum for any fissioning system and incident neutron energies up to 20 MeV, with only few adjustable parameters. In fact, LA model parameters systematics have been developed by A.Tudora *et al.* [4].

Detailed Statistical Evaporation Approach

While the Los Alamos model can be further refined, by allowing different temperatures in the light and heavy fragments for instance, particular questions simply cannot be addressed within its framework. As it averages over all fission fragment masses, charges and kinetic energies, as well as over the sequence of neutrons emission, distributions and correlated data cannot be calculated. The "point-by-point" approach developed by Vladuca and Tudora [3] represents an attempt in that direction, by performing LA model calculations for each fission fragmentation. In this case, one can calculate neutron data for each fission fragment configuration. However, this method still suffers from a few limitations: (i) it cannot assess correlated information on the successive neutron emissions; (ii) it does not treat gamma-rays; (iii) many input parameters are needed.

Monte Carlo Methodology

Going beyond those averages requires to study each step of the evaporation stage specifically. Starting from an initial, pre-neutron emission, fission fragment mass distribution $Y(A,Z,E^*)$ in mass A, charge Z and excitation energy E^*, neutrons are emitted following an evaporation spectrum whose characteristics change at each stage of the process. In the present calculations, γ-rays are only emitted when the residual excitation energy is too low for any additional neutron emission. A more adequate treatment of the neutrons-gamma-rays competition is in progress.

Reliable theoretical predictions for the $Y(A,Z,E^*)$ remain a challenge, but head-ways in this direction are being made and have been reported at this Conference [5, 6]. In our calculations experimental data were used instead.

In the LA model, the light and heavy fragment temperatures are assumed to be equal at scission *and* at the time of neutron emission, i.e., once the fragments are fully accelerated. As shown below, experimental data in low-energy nuclear fission suggest that more neutrons are emitted from the light fragment than from the heavy one, thereby contradicting the equi-temperature assumption. In fact, deformation energies of the fragments at scission, that very quickly transform into intrinsic excitation energies, are likely to be an important factor in this energy sharing. Following Ohsawa [2], we introduce the parameter

$$R_T = \langle T_l \rangle / \langle T_h \rangle , \tag{5}$$

where $\langle T_{l,h} \rangle$ are the average temperatures of the light and heavy fragments respectively. The case $R_T = 1$ corresponds to the LA model assumption. A R_T value greater than one leads to a harder spectrum, more neutrons being emitted from the light fragment. In the following, we treat R_T as a free adjustable parameter, that can be tuned to experimental data.

In the Fermi gas approximation, an excitation energy E^* corresponds to a temperature T through the relation $E^* = aT^2$, where a is the level density parameter of the nucleus. For a given temperature, neutrons are emitted following a Weisskopf-type spectrum

$$\phi_T(\varepsilon) \propto \varepsilon \times exp(-\varepsilon/T) , \tag{6}$$

where T is the temperature in the residual nucleus.

Once the distribution $Y(A,Z,E^*)$ is known for all fragments, Monte Carlo simulations can be initiated using a nuclear temperature given by the Fermi gas value. The corresponding Weisskopf spectrum of Eq. (6) is sampled and a particular outgoing neutron energy ε_n is chosen. A new residual nucleus with $(A-1)$ neutrons is created, with a new temperature corresponding to the residual excitation energy $E^*_{A-1} = E^*_A - B_n(A) - \varepsilon_n$. Emissions continue until the residual energy is too low for the emission of any additional neutron.

LA MODEL UNCERTAINTY QUANTIFICATION

Recently, we performed calculations to quantify uncertainties associated with the evaluated spectra obtained for the ENDF/B-VII.0 library [7]. A Bayesian statistical approach was used to account for uncertainties stemming from both experimental data as well as model parameters uncertainties. In this case, the Los Alamos model was used. A similar approach was used to obtain reaction cross section covariance matrices for ENDF/B-VII.0 in the fast energy range [8], and for prompt fission neutron spectra for the JENDL-3.3 evaluated library [9].

Preliminary results have been obtained for the neutron-induced fission of ^{239}Pu at 0.5 MeV incident neutron energy. Figure 1 shows 100 spectra sampled from the evaluated covariance matrix, and compared with experimental data sets.

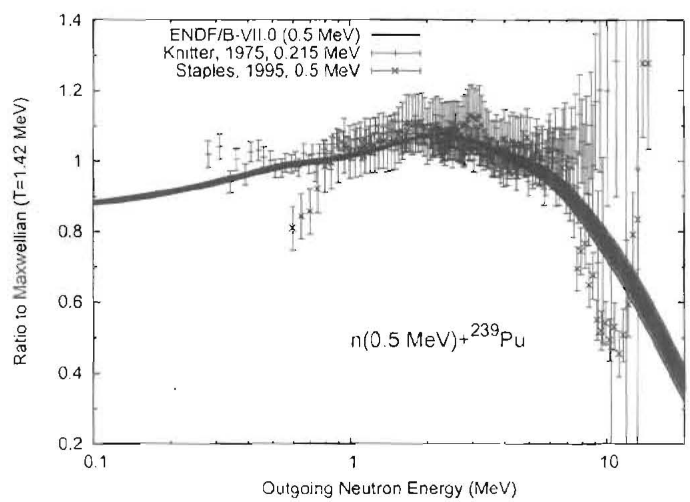

FIGURE 1. Sampling of the evaluated covariance matrix for the prompt fission neutron spectrum of n+^{239}Pu with 0.5 MeV incident neutron energy.

For sufficiently high incident neutron energies, i.e., above 10 MeV, pre-compound neutron emissions become non-negligible and should be taken into account. In the original LA model, such neutrons are calculated in the framework of multiple-chance fission, in a purely statistical evaporation process leading to a spectrum different from a pre-equilibrium spectrum. The LA model can be easily extended to adequately address this physics. The LA model can again be further improved by considering a different number of neutrons emitted from the light vs. heavy fragment. In most observed cases, at low-incident neutron energies, the light fragment emits more neutrons than the heavy fragment does. The spectrum for the light fragment being harder than the one for the heavy fragment will then lead to a harder average spectrum than calculated within the original LA model.

MONTE CARLO APPROACH NUMERICAL RESULTS

We have applied the Monte Carlo approach described above to the spontaneous fission of ^{252}Cf, the low-energy neutron-induced fission of ^{235}U, the thermal energy neutron-induced fission of ^{239}Pu, and the spontaneous fission of ^{236}Pu. Earlier results had been obtained for ^{252}Cf (sf) and thermal-neutron induced fission of ^{235}U, as reported in Ref. [10].

Input Data

For ^{252}Cf (sf), the experimental fission fragment yields Y(A,KE) were taken from Ref. [11]. Hambsch data [12] were also used for the neutron-induced fission of ^{235}U for incident energies ranging from 0.5 to 5.5 MeV, below the threshold of the second-chance fission. For thermal neutron induced fission of ^{239}Pu, the experimental fission fragment yields Y(A,KE) were taken from Ref. [13]. Finally, the fission fragment yields for the spontaneous fission of Pu-236 through Pu-244 were inferred from Ref. [14].

The charge of the fission fragments is obtained from a corrected unchanged charge distribution (UCD). The level density formalism of Gilbert-Cameron-Ignatyuk is used, including the washing-out of shell effects at increasing excitation energies. The level density parameters a are taken from Kawano *et al.* [15]. Finally, nuclear masses are taken from the Audi 2003 compilation [16].

The total excitation energy available at scission can be inferred for a given fragmentation as $TXE = Q_{l,h} - TKE$, where $Q_{l,h}$ is the Q-value for the fission reaction leading to specific light (l) and heavy (h) fragments, and TKE is the total kinetic energy. What is not know however is how much of this total excitation energy goes to the light vs. heavy fragment. In the present work, the R_T temperature ratio parameter, defined in Eq. (5), is treated as a free adjustable parameter.

Results

First, it is interesting to study the results for average quantities such as the prompt neutron multiplicity $\overline{\nu}_p$ and the average outgoing neutron energy $\overline{E}$. A summary of the results is shown in Table 1. The calculated average total number of neutrons $\overline{\nu}$ compares very well with the experimental values in all cases. The results show that the average total $\overline{\nu}$ changes very little when the R_T ratio parameter is changed from 1.0 to 1.2. In fact, the total available excitation energy at scission remains the same in both cases, and only the sharing of this energy between the light and heavy fragments is modified. A value

TABLE 1. Calculated vs. experimental average neutron multiplicities for nuclear reactions considered in the present work.

Reaction		$\overline{\nu}$	$\overline{\nu_l}$	$\overline{\nu_h}$	$\langle E_{lab} \rangle$
$^{252}Cf(sf)$	Calculation R_T=1.0	3.78	1.67	2.11	2.10
	Calculation R_T=1.2	3.79	2.06	1.73	2.03
	Vorobyev, 2004	3.756 ± 0.031	2.051	1.698	
	Boldeman, 1985	3.757	-	-	
n_{th}+^{239}Pu	Calculation R_T=1.0	2.89	1.42	1.47	1.94
	Calculation R_T=1.2	2.89	1.72	1.17	1.97
	Holden, 1988	2.881 ± 0.009	-	-	
n_{th}+^{235}U	Calculation R_T=1.0	2.47	1.11	1.36	1.85
	Calculation R_T=1.2	2.47	1.40	1.07	1.90
	Nishio, 1998	2.47	1.42	1.01	
	Müller, 1984*	2.46	1.44	1.02	
^{236}Pu (*sf*)	Calculation R_T=1.0	2.44	1.05	1.39	1.86
	Calculation R_T=1.2	2.45	1.33	1.12	1.91
	Hicks, 1956	2.30 ± 0.19	-	-	

* E_n=0.5 MeV

R_T greater than unity means that more energy is available in the light fragment, which in turn will emit a larger number of neutrons. Using a value of R_T=1.2, the observed ratios $\overline{\nu_l}/\overline{\nu_h}$ for both ^{252}Cf (sf) and n+^{235}U are well reproduced.

The calculated average energy of the neutron spectrum in the laboratory $\langle E_{lab} \rangle$ is also shown in Table 1. For ^{252}Cf (sf), the calculated values of 2.10 and 2.03 for R_T=1.0 and 1.2 respectively, are close to but a bit lower than the experimental value of about 2.15. In all other cases, the calculated values are also low compared to experimental ones. In fact, the calculated spectra tends to be too soft in most calculated cases.

Obviously, the present Monte Carlo simulations can easily provide much more than average quantities. Figure 2 shows the prompt neutron multiplicity distribution P(ν) in the case of n_{th}+^{239}Pu, compared to the experimental data by Holden [17]. The agreement is very reasonable. In the case of R_T=1.2, the calculated probability of having ν=3 is a bit low compared to the experimental value, compensated by a higher P($\nu = 4$) value.

The choice for the R_T parameter value has a significant influence on the hardness of the calculated spectrum. The reason for this result is easily understood. As R_T gets bigger, more intrinsic excitation energy is available in the light fragment. The light fragment center-of-mass spectrum is harder than the one for the heavy fragment, due to a smaller value of the level density parameter. In addition, the spectrum calculated for the light fragment in the laboratory spectrum gets a stronger kinematic boost than the spectrum for the heavy fragment, simply due to a smaller mass. This result is illustrated in Fig. 3 where the spectra calculated in the case of the 0.5 MeV neutron-induced fission of ^{235}U for R_T=1.0 and 1.4 are shown. It is not argued that 1.4 represents a good value for the R_T parameter, but instead this value is used to illustrate the impact of the parameter on the hardness of the calculated spectrum.

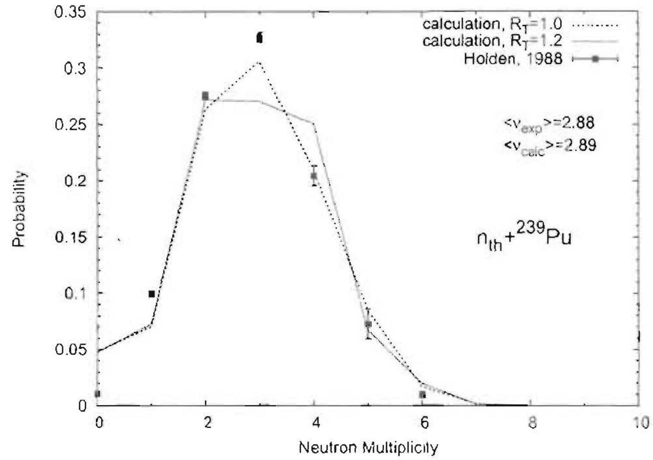

FIGURE 2. Prompt neutron multiplicity distribution $P(\nu)$ for thermal neutron-induced reaction on ^{239}Pu. Experimental data are from Holden [17].

DISCUSSION

The results shown above for the Monte Carlo simulations of the evaporation phase of the excited primary fission fragments are very encouraging. Detailed information on the prompt neutrons can be inferred in this way, such as correlations, distributions, etc., well beyond what can be obtained through the Los Alamos model. However, one of the main reason for the success of the LA model is that the number of adjustable parameters is very small, and that it can be applied to many fissioning systems. In fact, systematics of the LA model parameters have been already been obtained by Tudora [4].

The present Monte Carlo simulations have shown that, given the proper input parameters, in particular the pre-neutron emission fission fragment yields as a function of mass, charge and kinetic energy, they can reproduce the average neutron multiplicity $\overline{\nu}$ very accurately. In addition, they provide a good account for the neutron multiplicity distribution $P(\nu)$. The same cannot be said for the average spectrum $N(E)$ that is still better accounted for by LA model calculations. Additional work remains to be done to understand and solve some of the observed discrepancies. Also, it is very important to assess the sensitivity of the results to the choice of the input data. This work is underway.

Note that the calculations presented here do not include the proper treatment for the prompt γ-ray competition against neutron emission. This has to be done by performing full Hauser-Feshbach calculations, following the evolution of the spin distributions in the primary fragments. The initial spin distributions of the fragments is not known theoretically, and is only scarcely available from measurements. It is however an important endeavor as prompt γ-rays represent an important component of the post-scission processes and have important applications. It is also a quantity that simply cannot be

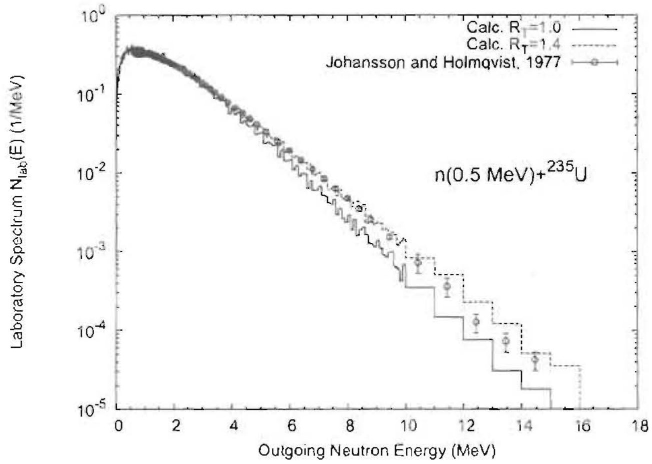

FIGURE 3. Prompt fission neutron spectra calculated in the case of 0.5 MeV neutron-induced fission of ^{235}U, for two values of the R_T parameter, 1.0 and 1.4, respectively. Experimental data are from Johansson and Holmqvist [18].

assessed within the LA model framework.

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