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TITLE "Recent Improvements in the Calculation of Prompt Fission Neutron Spectra: Preliminary Results"

AUTHOR(S) David G. Madland, T-2
Raphael J. LaBauve, T-2
J. Rayford Nix, T-9

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Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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RECENT IMPROVEMENTS IN THE CALCULATION OF PROMPT FISSION NEUTRON SPECTRA: PRELIMINARY RESULTS

David G. Madland, Raphael J. LaBauve, and J. Rayford Nix

Theoretical Division, Los Alamos National Laboratory
Los Alamos, New Mexico 87545

ABSTRACT

We consider three topics in the refinement and improvement of our original calculations of prompt fission neutron spectra. These are an improved calculation of the prompt fission neutron spectrum N(E) from the spontaneous fission of ^{252}Cf, a complete calculation of the prompt fission neutron spectrum matrix $N(E,E_n)$ from the neutron-induced fission of ^{235}U, at incident neutron energies ranging from 0 to 15 MeV, and an assessment of the scission neutron component of the prompt fission neutron spectrum. Preliminary results will be presented and compared with experimental measurements and an evaluation. A suggestion is made for new integral cross-section measurements.

I. INTRODUCTION

In this paper we report on initial efforts to refine and improve our original theoretical description of prompt fission neutron spectra and average prompt neutron multiplicities.[1] Although the refinements and improvements performed to date affect both the spectra and multiplicities, our work so far has been on the spectra alone. We consider three topics in the refinement of our original calculations of prompt fission neutron spectra. These are (a) an improved calculation of the *standard* prompt fission neutron spectrum N(E) from the spontaneous fission of ^{252}Cf, (b) a complete example of the incident neutron energy dependence of the prompt fission neutron spectrum N(E) for the neutron-induced fission of ^{235}U, resulting in the prompt fission neutron spectrum matrix $N(E,E_n)$, and (c) an assessment of the scission neutron question arising in prompt fission neutron spectra.

On the first topic, (a), the contributions to N(E) from the *entire* fission-fragment mass and charge distributions are calculated instead of calculating on the basis of a *seven-point approximation* to the peaks of these distributions as has been done in the past. Preliminary results are presented and compared with a measurement, an earlier calculation, and a recent evaluation of the spectrum, as well as recent integral cross-section measurements in this field. On the second topic, (b), we use the exact energy-dependent approach from our original work and calculate the entire fission spectrum matrix $N(E,E_n)$, for incident neutron energies in the range 0 MeV $\leq E_n \leq$ 15

MeV. At the higher incident neutron energies, we use the multiple-chance fission probabilities determined in our original calculation. Results are presented and compared with recent integral cross-section measurements in the thermal field, other comparisons with experiments at specific incident-neutron energies having been performed earlier. Due to the effort that would be required to experimentally verify a theoretical calculation of the complete fission spectrum matrix $N(E,E_n)$, we instead suggest that a number of crucial integral cross-section measurements be performed. On the third topic, (c), we discuss the experimental evidence for scission neutrons and the most likely physical mechanism for their production. However, at this time we do not calculate the scission-neutron component of N(E) due to lack of *conclusive* evidence for its existence.

In Sec. II we consider the fission spectrum N(E) for the ^{252}Cf(sf) *standard reaction* and in Sec. III we consider the fission spectrum matrix $N(E,E_n)$ for the neutron-induced fission of ^{235}U. We discuss scission neutrons in Sec. IV and our conclusions in Sec. V.

II. IMPROVED CALCULATION OF THE PROMPT FISSION NEUTRON SPECTRUM FROM THE SPONTANEOUS FISSION OF ^{252}Cf.

The prompt fission neutron spectrum N(E) from the spontaneous fission of ^{252}Cf is important due to its use as a *standard* neutron field. In addition, because of extensive experimental studies on this spectrum, it is used as a test case in the development of theoretical models of prompt fission neutron spectra for spontaneous as well as neutron-induced fission. In this paper, a measurement, an earlier calculation, an evaluation, and preliminary results from an improved calculation of N(E) for the ^{252}Cf(sf) reaction are presented and compared. In addition, measured and calculated integral cross sections for the ^{252}Cf(sf) spectrum are also presented and compared.

Our previous calculations[1-5] of the prompt fission neutron spectrum have utilized input parameters based upon *average* values of the fission-fragment mass, charge, and kinetic energy distributions. In particular, values of the average energy release in fission, $\langle E_r \rangle$, and the total average fission-fragment kinetic energy, $\langle E_f^{tot} \rangle$, have been used instead of the specific values occurring from all possible binary mass and charge divisions in fission. Likewise, the calculations of the inverse process to neutron emission, compound nucleus formation, have been restricted to two nuclei: the average central light fragment and the average central heavy fragment. Finally, it was noted that in the vicinity of the average fragments, the average numbers of neutrons emitted from the light and heavy fragments are approximately equal. The spectrum N(E) has therefore been given by the *average* of the spectra calculated from the light and heavy fragments, namely

$$N(E) = \frac{1}{2}\left[N(E,E_f^L,\sigma_c^L) + N(E,E_f^H,\sigma_c^H)\right], \qquad (1)$$

where E is the laboratory neutron energy, E_f^L and E_f^H are the average kinetic energies per nucleon of the light and heavy fragments, respectively, and σ_c^L and σ_c^H are the cross sections for the inverse process in the average light and heavy fragments, respectively.

In the present work, the use of input parameters based upon *average* values of the fission-fragment mass, charge, and kinetic energy distributions is replaced by *direct use, on a point-by-point basis, of the distributions themselves*. Following a description of the refinements to our original calculations, in the next section, preliminary results are presented and discussed.

II. A. REFINEMENTS IN THE MODEL

The energy release E_r for *each binary fission considered* is given by

$$E_r = M(Z_c,A_c) - M_L(Z_L,A_L) - M_H(Z_H,A_H) \ , \qquad (2)$$

where M is a mass excess expressed in MeV and c, L, and H refer to compound fissioning nucleus, light fission fragment, and heavy fission fragment, respectively. Use of Eq. (2) over the fission-fragment mass and charge distributions *replaces* the average value $\langle E_r \rangle$ obtained using the seven-point approximation given in Ref. 1 and used in Refs. 1-5 (note that in Ref. 2, an exact calculation of $\langle E_r \rangle$ was also performed). In evaluating Eq. (2), experimental masses from the 1986 Audi-Wapstra mid-stream mass evaluation[6] are used where they exist and otherwise the calculated masses of Möller and Nix.[7]

The total fission-fragment kinetic energy E_f^{tot} for *each binary fission considered* is taken from the experimental results of Schmitt *et al.*,[8] in which E_f^{tot} is given as a function of heavy fragment mass,

$$E_f^{tot} = E_f^{tot}(A_H) \ , \qquad (3)$$

for all values of A_H observed ($126 \le A_H \le 166$). These $E_f^{tot}(A_H)$ values are themselves averages due to the fission-fragment distributions in charge $P(Z_L)$ and $P(Z_H)$, for fixed values of A_L and A_H, respectively. Recall that the binary fission assumption demands that the sets (A_L,A_H,A_c) and (Z_L,Z_H,Z_c) simultaneously satisfy complementarity. Use of the measurements of E_f^{tot} by Schmitt *et al.*,[8] represented by Eq. (3), *replaces* the average value of the total fission-fragment kinetic energy $\langle E_f^{tot} \rangle$ used in Refs. 1-5.

The values of E_f^{tot} are used in two ways in the calculation of N(E). The first way is in the calculation of the average kinetic energies per nucleon, E_f^L and E_f^H, of the light and heavy fragments. These are obtained by use of momentum conservation, as before, and are given by

$$E_f^L = (A_H/A_L)(E_f^{tot}/A_c \text{, and} \qquad (4)$$

$$E_f^H = (A_L/A_H)(E_f^{tot}/A_c). \qquad (5)$$

In all of our previous work these same equations have been used, but they have been evaluated using $\langle E_f^{tot} \rangle$ instead of E_f^{tot}, the average central light fragment instead of A_L, and the average central heavy fragment instead of A_H.

The values of E_f^{tot} are also used, together with the values of the energy release in fission E_r, to calculate the maximum temperatures T_m of the temperature distributions P(T) representing the corresponding distributions of fission-fragment excitation energy. In the present calculation this is done for *each binary fission considered*, whereas in our previous calculations *one average value* of T_m was used. For spontaneous fission, T_m is now given by

$$T_m = [(E_r - E_f^{tot})/a]^{1/2} , \qquad (6)$$

where E_r and E_f^{tot} are given by Eqs. (2) and (3), respectively, and a is the Fermi gas level density parameter

$$a = A_c/(\text{const}) . \qquad (7)$$

Previously, the average values $\langle E_r \rangle$ and $\langle E_f^{tot} \rangle$ were used in evaluating Eq. (6).

The compound nucleus cross section σ_c for the inverse process is computed for the two fragments occurring in *each binary fission considered.* Thus, $\sigma_c = \sigma_c(\varepsilon,Z,A)$, ($Z_L$ or Z_H, A_L or A_H), where ε is the center-of-mass neutron energy. The optical-model potential of Becchetti and Greenlees[9] is used on a 100-point grid extending to 40 MeV, as in our earlier work for the *average light and heavy fragments.*

Given the above refinements to calculate the prompt fission neutron spectrum for each pair of complementary points on the fission-fragment mass and charge distributions, it remains to combine the results from all contributing pairs. For a given fragment mass number A, (A_L or A_H), the charge distribution in Z, (Z_L or Z_H), approximates a Gaussian distribution

$$P(Z) = (1/\sqrt{c\pi})\, \exp[-(Z - Z_p)^2/c] \ , \qquad (8)$$

where the most probable charge Z_p, (Z_p^L or Z_p^H), is obtained using a corrected unchanged charge distribution (UCD) assumption due to Unik *et al.*,[10]

$$(Z_p^L - \tfrac{1}{2})/A_L = (Z_c/A_c) = (Z_p^H + \tfrac{1}{2})/A_H \ , \qquad (9)$$

and where the width parameter, c, is given by

$$c = 2(\sigma^2 + \tfrac{1}{12}) \ , \qquad (10)$$

where σ is the average charge dispersion. A value of $\sigma = 0.40 \pm 0.05$ is used, which was determined in the experiments of Reisdorf *et al.*[11] for the pre-neutron emission charge distribution in the thermal-neutron-induced fission of ^{235}U.

Given the charge distribution P(Z) for each fragment mass number A, the contributions from all fragment masses are summed. This is accomplished by use of weighting factors comprised of (a) the fragment mass yields Y(A), (A_L or A_H), and (b) the average number of prompt neutrons emitted for each fragment mass $\bar{\nu}(A)$, (A_L or A_H). In the present work, the pre-neutron emission experimental fragment-yields of Schmitt *et al.*[8] are used and the average prompt neutron multiplicities measured as a function of fragment mass by Walsh and Boldeman[12] are also used.

Using Eqs. (2)-(10), the expression for the prompt fission neutron spectrum N(E) in the preliminary refined model is given by

$$N(E) = \sum_A \frac{\bar{\nu}(A)}{\bar{\nu}_{tot}}\, Y(A) \sum_Z P(Z)\, N\left[E, E_F(A), \sigma_c(Z,A), T_m(Z,A)\right] \qquad (11)$$

where $\bar{\nu}_{tot} = \sum_A \bar{\nu}(A) Y(A)$ is the total average prompt neutron multiplicity and the sums occurring are over Z_L and Z_H as well as over A_L and A_H.

II. B. PRELIMINARY RESULTS

The first-calculation using the refined model summarized by Eq. (11) is for the spontaneous fission of ^{252}Cf. In this calculation, the fission-fragment mass and charge distributions are represented by 28 fragments:

(a) 14 approximately equispaced fragment masses in the range $88 \leq A \leq 164$, with a spacing of about 6 in mass number, and

(b) 2 isobars per fragment mass, with values of Z that are the nearest integer values above and below the most probable charge Z_p.

The contributions to the prompt neutron spectrum from *each binary fission considered* therefore include:

(a) 28 optical-model calculations of the compound nucleus formation cross section $\sigma_c(Z,A)$ for the inverse process, using Ref. 9,

(b) 14 calculations of the energy release in fission E_r, one for each fragment pair, with values spanning the range 198.061 MeV $\leq E_r \leq$ 236.421 MeV,

(c) 7 experimental values[8] of the total fragment kinetic energy E_f^{tot}, each accounting for 2 fragment pairs, spanning the range 165.91 MeV $\leq E_f^{tot} \leq$ 195.22 MeV,

(d) 14 calculations of the average kinetic energy per nucleon one for each pair of isobars, with 7 such pairs for the light fragments having values in the range 0.777 MeV $\leq E_f^L \leq$ 1.227 MeV, and 7 such pairs for the heavy fragments having values in the range 0.353 MeV $\leq E_f^H \leq$ 0.729 MeV,

(e) 14 calculations of the most probable charge Z_p, one for each pair of iosbars, yielding 7 values of Z_p^L for the light fragments and 7 values of Z_p^H for the heavy fragments,

(f) 7 experimental values[8] of the fragment mass yield Y(A), each accounting for 2 fragment pairs, spanning the range 0.17% $\leq$ Y(A) $\leq$ 5.55%, and

(g) 14 experimental values[12] of the average neutron multiplicity as a function of fragment mass $\bar{\nu}(A)$, one for each pair of isobars, spanning the range $0.71 \leq \bar{\nu}(A) \leq 3.89$.

The preliminary results obtained using Eq. (11) with *28 fission fragments* to explicitly represent the total fission-fragment mass and charge distributions are illustrated in Figs. 2-7. For comparison purposes, a calculation of the spectrum reproduced from our earlier work[4] is shown in Fig. 1. The solid curve here shows the spectrum calculated using Eq. (1), for *two averge fragments* from the yield peaks, with a nuclear level-density parameter $a = A_c/(9.15\ \text{MeV})$ obtained in a least-squares adjustment to the experimental spectrum of Poenitz and Tamura.[13] Ratios to the least-squares adjusted Maxwellian spectrum (T_M = 1.429 MeV) were used as the basis for comparison.

In Fig. 2 we show our earlier calculation again, as the dashed curve, together with the present calculation using Eq. (11), as the solid curve. The effects of the refined model calculation compared with the previous model calculation are that the spectrum is increased in the regions below approximately 1.4 MeV and above approximately 8.8 MeV, and is decreased in the region between approximately 1.4 MeV and 8.8 MeV. A comparison of Figs. 1 and 2 clearly shows that these effects are in *exactly the right direction* to give even better agreement with the experiment of Poenitz and Tamura[13] than was obtained in the previous calculation.[4] However, it is equally clear that the refined calculation does not yet exactly reproduce the experiment. Namely, an even larger increase would be possible in the low and high energy regions of the calculated spectrum. Note that the spectra shown in Fig. 2 are both calculated with a level-density parameter, $a = A_c/(9.15$ MeV), identical to that used in Fig. 1, and also that the reference Maxwellian of Fig. 2 is calculated with T_M = 1 42 MeV.

The present calculation shown in Fig. 2 is compared with a recent evaluation of the spectrum by Mannhart[14] in Fig. 3. The "data" shown are from the "group averages" spectrum obtained by Mannhart. Again, a reference Maxwellian with T_M = 1.42 MeV has been used. The agreement between the present calculation and the evaluated spectrum is not nearly as good as in the case of the experimental spectrum of Poenitz and Tamura.[13] A least-squares adjustment to the level-density parameter was then performed resulting in the value $a = A_c/(9.40\ \text{MeV})$, which improved the χ^2 approximately by a factor of two. The comparison of this spectrum with the evaluation of Mannhart is shown in Fig. 4 using the same reference Maxwellian spectrum. Although the agreement with the evaluated spectrum is improved, it is again not nearly as good as in the case of the experimental spectrum of Poenitz and Tamura and the unadjusted present calculation.

Comparisons of integral cross sections calculated using these spectra and experimental values are shown in Figs. 5-7. Recall that the integral cross section, $\langle\sigma_i\rangle$, represents the net effect of the pointwise cross section $\sigma_i(E)$ in the presence of the neutron field N(E), and is given by

$$\langle\sigma_i\rangle = \frac{\int_{E_1}^{E_2} \sigma_i(E)N(E)dE}{\int_{E_1}^{E_2} N(E)dE}, \tag{12}$$

where E is the neutron energy and E_1 and E_2 are the energy limits of the field. A specific reaction with a known cross section and a threshold, at $E = E_{th}$, serves as a means by which integral comparisons can be made of different neutron fields for energies $E \geq E_{th}$. For the present calculations, we use ENDF/B-V pointwise cross sections[15] in all cases except for four high threshold reactions (E_{th} > 12 meV) where we use recent evaluations by Young[16] and measurements by Bayhurst *et al.*[17] and Mannhart and Vonach.[18] We compare our calculated values obtained using Eq. (12) with the experimental integral cross sections measured by Kobayshi *et al.*[19] and Mannhart.[20-22]

The ratios of the calculated integral cross sections using the present spectrum, shown as the solid curve in Figs. 2 and 3, to the experimental integral cross sections are plotted in Fig. 5 as a function of the threshold energy of the reaction (defined here as the energy at which the integral of the pointwise cross section reaches 0.01% of its total value.) The figure shows that the present spectrum is compatible with the experimental integral cross sections for emitted neutron energies below about 9.5 MeV, but is too soft for higher emitted neutron energies. Similarly, the ratios of the calculated integral cross sections using the present least-squares adjusted spectrum, shown as the solid curve in Fig. 4, to the experimental integral cross sections are plotted in Fig. 6. This figure shows that the present least-squares adjusted spectrum, shown as the solid curve in Fig. 4, to the experimental integral cross sections are plotted in Fig. 6. This figure shows that the present least-squares adjusted spectrum is also too soft, but only for emitted neutron energies in excess of about 11.5 MeV, compared to 9.5 MeV before. Moreover, the departure from experiment for higher threshold energies is clearly less than before. Thus, the present least-squares adjusted spectrum is compatible with the experimental integral cross sections for emitted neutron energies up to about 11.5 MeV, but is somewhat soft for higher energies. Therefore, further improvement is needed.

Finally, the rations of the calculated integral cross sections using Mannhart's spline fit[14] to his evaluated spectrum, shown as the points in Figs. 3 and 4, and the experimental integral cross sections are plo d in Fig. 7 (since the spline fit extends only to 20 MeV, integral cross sections cannot be calculated for reaction thresholds above about 12 MeV.) This figure shows that the spline fit to the evaluated spectrum is compatible with the experimental integral cross sections for emitted neutron energies up to about 12.5 MeV. However, there is clearly a trend in the ratios indicating that the spline fit is increasingly too hard for energies in the range of about 7 to 12 MeV. Thus, the evaluated spectrum may also require some revision in this energy range. Clearly, further work must be done on the prompt fission neutron spectrum for the spontaneous fission of ^{252}Cf, especially if this spectrum is to be used as a *standard* spectrum.

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FIGURE CAPTIONS

Fig. 1. Ratio of the previous least-squares adjusted Los Alamos spectrum and the experimental spectrum of Poenitz and Tamura (1982) to the least-squares adjusted Maxwellian spectrum, for ^{252}Cf(sf).

Fig. 2. Ratio of the previous least-squares adjusted Los Alamos spectrum, based on considerations of the *peaks* of the fission-fragment mass and charge distributions, and the present Los Alamos spectrum, based on considerations of the *entire* fission-fragment mass and charge distributions, to a Maxwellian spectrum with T_m = 1.42 MeV. The nuclear level-density parameter in both calculations is given by $a = A_c/(9.15$ MeV).

Fig. 3. Ratio of the present Los Alamos spectrum and the evaluated spectrum of Mannhart (1987) to a Maxwellian spectrum with T_m = 1.42 MeV, for ^{252}Cf(sf). The nuclear level-density parameter is given by $a = A_c/(9.15$ MeV).

Fig. 4. Ratio of the present least-squares adjusted Los Alamos spectrum and the evaluated spectrum of Mannhart (1987) to a Maxwellian spectrum with T_m = 1.42 MeV, for ^{252}Cf(sf). The adjusted nuclear level-density parameter is given by $a = A_c/(9.40$ MeV).

Fig. 5. Ratio of calculated to experimental integral cross sections for the neutron field from the spontaneous fission of ^{252}Cf, as a function of the threshold energy for the reaction. The calculated values are obtained using the present spectrum from Eq. (11) in Eq. (12) together with ENDF/B-V pointwise cross sections, except for four high threshold reactions extending beyond 20 MeV (see text.)

Fig. 6. Ratio of calculated to experimental integral cross sections for the neutron field from the spontaneous fission of ^{252}Cf, as a function of the threshold energy for the reaction. The calculated values are obtained using the present least-squares adjusted spectrum from Eq. (11) in Eq. (12) together with ENDF/B-V pointwise cross sections, except for four high threshold reactions extending beyond 20 MeV (see text.)

Fig. 7. Ratio of calculated to experimental integral cross sections for the neutron field from the spontaneous fission of ^{252}Cf, as a function of the threshold energy for the reaction. The calculated values are obtained using Mannhart's spline fit to his evaluated spectrum (1987.)