

IMPROVED CALCULATION OF THE PROMPT FISSION NEUTRON SPECTRUM FROM THE SPONTANEOUS FISSION OF ^{252}Cf : PRELIMINARY RESULTS

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Abstract: An improved calculation is presented for the prompt fission neutron spectrum $N(E)$ from the spontaneous fission of ^{252}Cf . In this calculation the fission-spectrum model of Madland and Nix is used, but with several improvements leading to a physically more accurate representation of the spectrum. Specifically, the contributions to $N(E)$ from the *entire* fission-fragment mass and charge distributions will be 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. Therefore, values of the energy release in fission, fission-fragment kinetic energy, and compound nucleus cross section for the inverse process will be considered on a point-by-point basis over the fragment yield distributions instead of considering averages of these quantities over the peaks of the distributions. Preliminary results will be presented and compared with a measurement, an earlier calculation, and a recent evaluation of the spectrum.

(Keywords: calculation, prompt fission neutron spectrum, spontaneous fission, ^{252}Cf)

Introduction

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 $^{252}\text{Cf}(sf)$ reaction are presented and compared.

Our previous calculations¹⁻⁵ 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, $\langle E_f \rangle$, and the total average fission-fragment kinetic energy, $\langle E_f^{\text{tot}} \rangle$, 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

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

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 and some tentative conclusions are given.

Refinements in the Model

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

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

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 $\langle E_f \rangle$ 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 $\langle E_f \rangle$ was also performed). In evaluating Eq. (2), experimental masses from the 1986 Audi-Wapstra mid-stream mass evaluation⁶ 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,

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

for all values of A_H observed ($126 \leq A_H \leq 166$). These $E_f^{\text{tot}}(A_H)$ 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 $\langle E_f^{\text{tot}} \rangle$ 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

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

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

In all of our previous work these same equations have been used, but they have been evaluated using $\langle E_f^{\text{tot}} \rangle$ 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 calculations 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

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

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

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

Previously, the average values $\langle E_r \rangle$ and $\langle E_f^{\text{tot}} \rangle$ 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, $\sigma_c = \sigma_c(\epsilon, Z, A)$, (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 Greenlees⁹ 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

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

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

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

and where the width parameter, c , is given by

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

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 ²³⁵U.

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 Boldeman¹² 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

$$N(E) = \sum_A \frac{\bar{\nu}(A)}{\bar{\nu}_{\text{tot}}} Y(A) \sum_Z P(Z) N[E, E_f(A), \sigma_c(Z, A), T_m(Z, A)] \quad (11)$$

where $\bar{\nu}_{\text{tot}} = \sum_A \bar{\nu}(A)Y(A)$ 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 .

Preliminary Results

The first-calculation using the refined model summarized by Eq. (11) is for the spontaneous fission of ²⁵²Cf. 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 $198.061 \text{ MeV} \leq E_r \leq 236.421 \text{ MeV}$,
- (c) 7 experimental values⁸ of the total fragment kinetic energy E_f^{tot} , each accounting for 2 fragment pairs, spanning the range $165.91 \text{ MeV} \leq E_f^{\text{tot}} \leq 195.22 \text{ MeV}$,
- (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 $0.777 \text{ MeV} \leq E_f^L \leq 1.227 \text{ MeV}$, and 7 such pairs for the heavy fragments having values in the range $0.353 \text{ MeV} \leq E_f^H \leq 0.729 \text{ MeV}$,
- (e) 14 calculations of the most probable charge Z_{pL} , one for each pair of isobars, 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 values⁸ 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 values¹² 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. 2-4. For comparison purposes, a calculation of the spectrum reproduced from our earlier work⁴ is shown in Fig. 1. The solid curve here shows the spectrum calculated using Eq. (1), for *two average 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 ($T_M = 1.429 \text{ MeV}$) 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 Tamura¹³ 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 Mannhart¹⁴ 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 $a = A_C/9.40$ 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.

Conclusions

It has been shown that the preliminary calculations using the refined model calculation embodied in Eq. (11) yields *improved agreement* with the experimental spectrum of Poenitz and Tamura¹³ and *unsatisfactory agreement* with the evaluated spectrum of Mannhart.¹⁴ The discrepancy probably arises from two sources. On the one hand, the spectrum of Poenitz and Tamura is one of seven experiments used in the Mannhart evaluation. Therefore, the differences between the various experiments making up the evaluation are likely to be at least as large as the difference between the present calculation and the evaluation. On the other hand, the convergence of the refined model calculation with the number of fragments included must be demonstrated. In addition, the physical effects of (a) center-of-mass anisotropy, and (b) explicit gamma-ray deexcitation should both be taken into account.

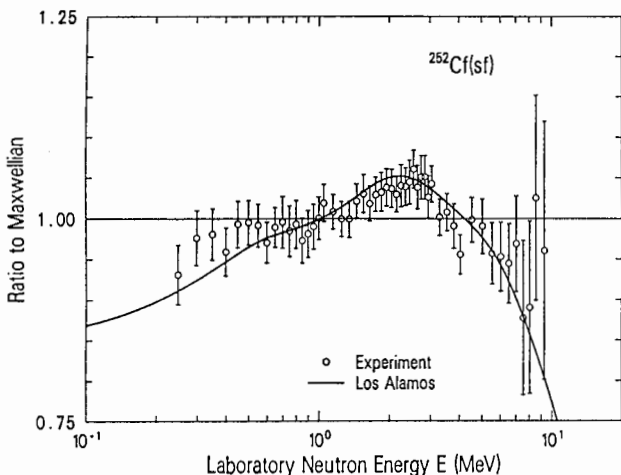


Fig.1. Ratio of the previous least-squares adjusted Los Alamos spectrum and the experimental spectrum of Poenitz and Tamura (1982) to the least-squares adjusted Maxwellian spectrum, for $^{252}\text{Cf}(sf)$.

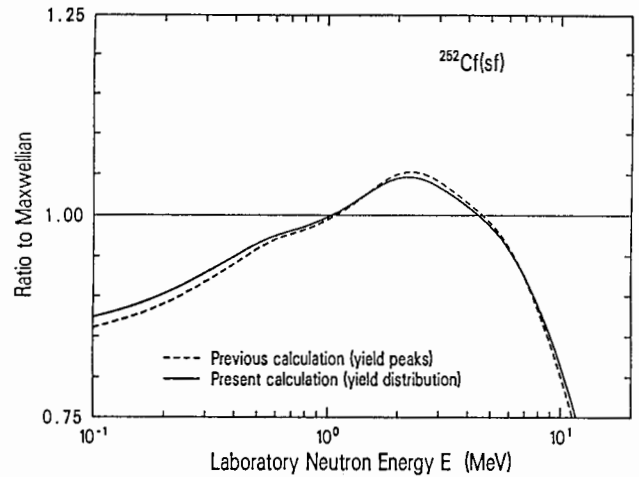


Fig. 2. Ratios of the previous least-squares adjusted Los Alamos spectrum, based on considerations of the *peaks* of the fission-fragment mass and charge distributions, and the present Los Alamos spectrum, based on considerations of the *entire* fission-fragment mass and charge distributions, to a Maxwellian spectrum with $T_M = 1.42$ MeV.

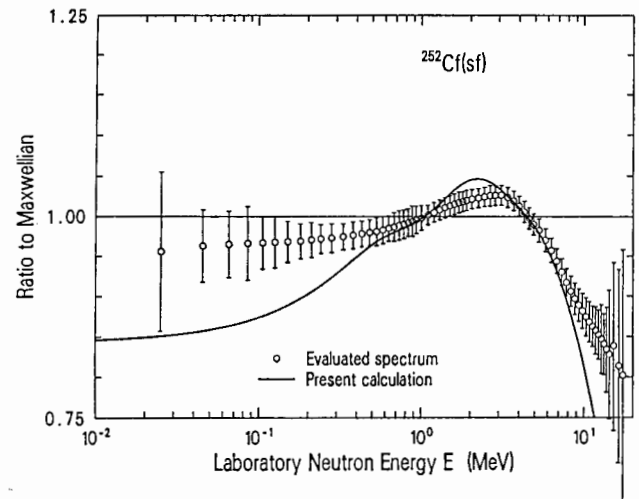


Fig. 3. Ratio of the present Los Alamos spectrum and the evaluated spectrum of Mannhart (1987) to a Maxwellian spectrum with $T_M = 1.42$ MeV, for $^{252}\text{Cf}(sf)$. The nuclear level-density parameter is given by $a = A_C/9.15$ MeV.

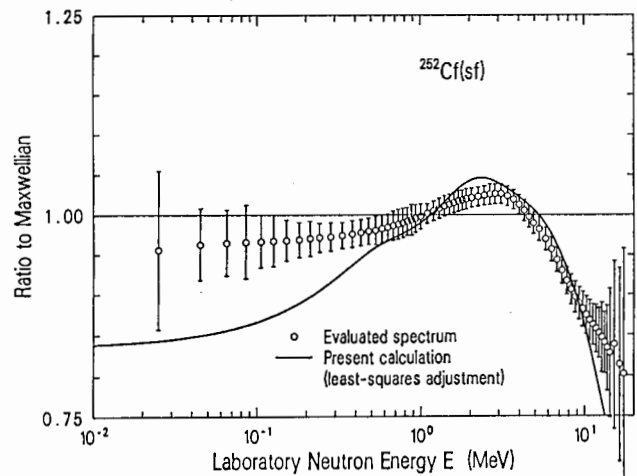


Fig. 4. Ratio of the present least-squares adjusted Los Alamos spectrum and the evaluated spectrum of Mannhart (1987) to a Maxwellian spectrum with $T_M = 1.42$ MeV, for $^{252}\text{Cf}(sf)$. The adjusted nuclear level-density parameter is given by $a = A_C/9.40$ MeV.

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