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IMPROVED CALCULATION OF THE PROMPT FISSION NEUTRON SPECTRUM FROM THE SPONTANEOUS FISSION OF 252Cf: 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 ²⁵²Cf. 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, 252Cf)

introduction

The prompt fission neutron spectrum N(E) from the spontaneous fission of ²⁵²Cf 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 ²⁵²Cf(sf) reaction are 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, , and the total average fission-fragment kinetic energy, <E' >, 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

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

where E is the laboratory neutron energy, E_f^L and E_f^H are the average kinetic energies per pucleon, or the light and heavy fragments, respectively, and σ_c^L and σ_c^L 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_t for each binary fission considered is given by

$$E_{r} = M(Z_{c}, A_{c}) - M_{L}(Z_{L}, A_{L}) - M_{H}(Z_{H}, A_{H})$$
, (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 are used where they exist and otherwise the calculated masses of Möller and Niv 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 \leq A_H \leq 166). These E_{ℓ}^{D} (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. Recal! 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_{ℓ}^{D} by Schmitt *et al.*, represented by Eq. (3), replaces the average value of the total fission-fragment kinetic energy $\leq E_{\ell}^{D}$ used in Refs. 1-5

tission 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 $E_f^{(0)}$ by Schmitt et al., represented by Eq. (3), replaces the average value of the total fission-fragment kinetic energy $\langle E_f^{(0)} \rangle$ used in Refs. 1-5.

The values of $E_f^{(0)}$ 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, $E_f^{(0)}$ and $E_f^{(0)}$, of the light and heavy fragments. These are obtained by use of momentum conservation, as before, and are given by

$$\mathbf{E}_{\mathbf{f}}^{\mathbf{L}} = (\mathbf{A}_{\mathbf{H}}/\mathbf{A}_{\mathbf{l}}) (\mathbf{E}_{\mathbf{f}}^{\mathbf{tot}}/\mathbf{A}_{\mathbf{c}}), \text{ and}$$
 (4)

$$E_t^H = (A_L/A_H) (E_t^{tot}/A_c)$$
. (5)

In all of our previous work these same equations have been used, but they have been evaluated using $\langle E_f^{10l} \rangle$ instead of

E^{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 fissionfragment 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

$$T_{\rm m} = [(E_{\rm r} - E_{\rm f}^{\rm tot})/a]^{1/2},$$
 (6)

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

$$\mathbf{a} = \mathbf{A}_{\mathbf{C}}/(\mathbf{const}) \ . \tag{7}$$

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 oc for the inverse process is computed for the two fragments occurring in each binary fission considered. Thus, $\sigma_c = \sigma_c(\epsilon, Z, A)$, $(Z_L \text{ or } Z_H,$ AL or AH), where e 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, (AL or AH), the charge distribution in Z, (Z_L or Z_H), approximates a Gaussian distribution

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

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

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

and where the width parameter, c, is given by

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

where σ is the average charge dispersion. A value of $\sigma = 0.40$ ± 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), (AL or AH), and (b) the average number of prompt neutrons emitted for each fragment mass V(A), (AL or AH). 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 2 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

$$N(E) = \sum_{A} \frac{\nabla(A)}{\nabla_{tot}} Y(A) \sum_{Z} P(Z) N[E, E_{p}(A), \sigma_{c}(Z, A), T_{m}(Z, A)]$$
(11)

where $\overline{v}_{tot} = \sum_{i} \overline{v}(A)Y(A)$ is the total average prompt neutron multiplicity and the sums occurr g are over ZL and ZH as well as over AL and AH.

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 $88 \le A \le 164$, 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 propable charge Z₀.

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 of the total fragment kinetic energy $E_f^{(0)}$, each accounting for 2 fragment pairs, spanning the range 165.91 MeV $\leq E_f^{(0)} \leq 195.22$
- (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 $\leq E_f \leq 1.227$ MeV, and 7 such pairs for the heavy fragments having values in the range 0.353 MeV $\leq Z_f^{11} \leq 0.729$ MeV,
- (e) 14 calculations of the most probable charge Z_p one for each pair of iosbars, yielding 7 values of Z_p for the light fragments and 7 values of Z_p for the heavy iragments,
- (f) 7 experimental values of the fragment mass yield Y(A), each accounting for 2 fragment pairs, spanning the range $0.17\% \le Y(A) \le 5.55\%$, and
- (g) 14 experimental values 12 of the average neutron multiplicity as a function of fragment mass $\nabla(A)$, one for each pair of isobars, spanning the range 0.71 ≤

The preliminary results obtained using Eq. (11) with 28 fission fragments to explicitly represent the total fission-fing ment 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 averge fragments from the yield peaks, with a nuclear level-density parameter $a = A_c/(9.15 \text{ MeV})$ obtained in a leastsquares 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 .4 MeV and above approximately 8.8 MeV, and is decreased

in the region between approximately 1.4 MeV and so MeV. A comparison of Figs. 1 and 2 clearly shows that these effects are in eactly the right direction to give even better agreement with the experiment of Poenitz and Tamura 13 than was obtained in the previous calculation. 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, $a = A_c/(9.15 \text{ MeV})$, identical to that used in Fig. 1, and also that the reference Maxwellian of Fig. 2 is calculated with $T_M = 1.42 \text{ MeV}$.

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 $a = A_c/(9.40 \text{ MeV})$, 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.

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.

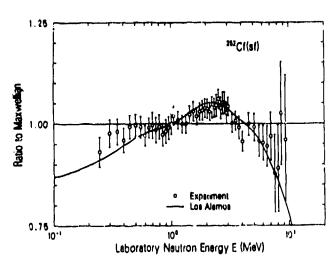


Fig.1. Ratio of the previous least-squares adjusted Los Alamos spectrum and the experimental spectrum of Poenitz and Tamura (1982) to the least-squares adjusted Maxwellian spectrum, for ²⁵²Cf(sf).

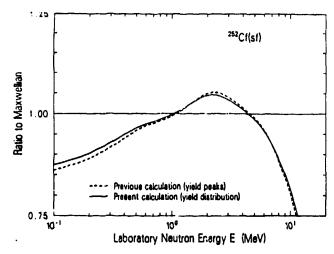


Fig. 2. Ratios of the previous least-squares adjusted Los Alamos spectrum based on considerations of the peaks of the fission-fragment mass and charge distributions, and the present Los Alamos spectrum, based on considerations of the entire fission-fragment mass and charge distributions, to a Maxwellian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

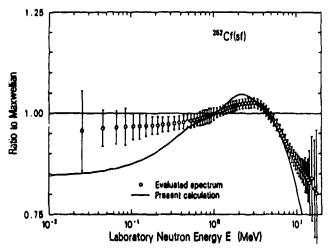


Fig. 3. Ratio of the present Los Alamos spectrum and the evaluated spectrum of Mannhari (1987) to a Maxwellian spectrum with $T_M = 1.42$ MeV for 232 Cf(sf). The nuclear level-density parameter is given by $a = A_c/(9.15 \text{ MeV})$.

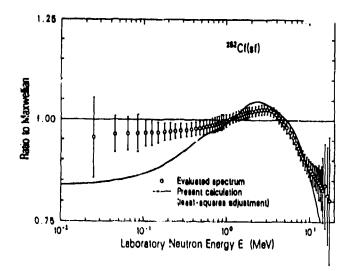


Fig. 4. Ratio of the present least-squares adjusted Los Alamos spectrum and the evaluated spectrum of Mannhart (1987) to a Maxwellian spectrum with $T_M = 1.42$ MeV, for 252 Cf(sf). The adjusted nuclear level-density parameter is given by $a = A_0/(9.40 \text{ MeV})$.

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