Hauser-Feshbach calculation of the ^{252}Cf spontaneous-fission neutron spectrum*

J. C. Browne and F. S. Dietrich
Lawrence Livermore Laboratory, University of California, Livermore, California 94550
(Received 1 July 1974)

The neutron spectrum from ^{252}Cf spontaneous fission has been calculated using the Hauser-Feshbach formalism. Details of the excitation energy and spin distributions have been included for 40 fission fragments chosen in a manner to average over the mass distribution and over pairing-energy effects. No arbitrary parameters were introduced into the calculation to produce a fit to the data. The results of the calculation are in good agreement with experimental data.

[RADIOACTIVITY, FISSION ^{252}Cf(sf); calculated neutron spectrum; Hauser-Feshbach calculation.]

I. INTRODUCTION

The neutron energy spectrum from ^{252}Cf spontaneous fission has been measured by many experimenters; the most recent measurements are those of Meadows[1] and of Green, Mitchell, and Steen.[2] Various theoretical calculations have been made to describe the shape of the measured spectrum with the most complete descriptions given perhaps by Terrell,[3] Lang,[4] and Kluge.[5] In all three cases the authors use a center-of-mass energy distribution for the neutrons predicted by Weisskopf's evaporation model.[6] In recent years much detailed information concerning ^{252}Cf spontaneous fission has become available so that it is possible to consider the decay of individual fission fragments in determining the neutron spectrum. We have therefore performed a statistical calculation of the ^{252}Cf fission neutron spectrum in the Hauser-Feshbach formalism[7] using experimental information to determine initial fragment excitation energies and spin distributions, and kinetic energy and mass distributions. No arbitrary parameters were introduced into the calculation to produce a fit to the data.

II. CALCULATION

For each excited fragment (Z,A), the c.m. partial neutron energy spectrum $N(E_n)$ was calculated assuming competition between only neutron and γ-ray emission using the following expression:

$$N(E_n)dE_n = \sum_J \int dE\, P(E,J)\frac{1}{D(E,J)} \times \sum_{J'} G_n(E\to E',J\to J')dE_n \ , \qquad (1)$$

where $P(E,J)$ is the initial excitation energy and spin distribution and

$$D(E,J) = \sum_{J'} \int dE' G_n(E\to E',J\to J') + \sum_{J''} \int dE'' G_\gamma(E\to E'',J\to J'') \ , \qquad (2)$$

where

$$G_n(E\to E',J\to J') = \sum_{l,s} T_l(E\to E',J\to J')\rho(E',J',Z,A-1) \qquad (3)$$

and

$$G_\gamma(E\to E'',J\to J'') = T_\gamma(E\to E'',J\to J'')\rho(E'',J'',Z,A) \ . \qquad (4)$$

In the above expressions, T_l and T_γ are the neutron and γ-ray transmission coefficients and the ρ are the relevant level densities. The excitation energy after neutron emission is $E'=E-S_n-E_n$ where S_n is the neutron separation energy of the fragment Z,A, and E_n is the c.m. kinetic energy carried off by the neutron. The excitation energy after γ-ray emission is $E''=E-E_\gamma$. The form taken for the initial excitation energy-spin distribution was

$$P(E,J) \propto (2J+1)\exp\left\{-\frac{[E-E_0(Z,A)]^2}{2(\sigma_{E(Z,A)})^2} - \frac{J(J+1)}{(B_{Z,A,E})^2}\right\} .$$

For a given pair of fission fragments, the total available energy for fission was calculated from the Garvey-Kelson mass tables.[8] The mean *total* excitation energy, E_{0T}, was found by subtracting the mean total kinetic energy of the fragment pair[9] from the total available energy. The division of excitation energy between the light and heavy fragment was computed using the neutron number distribution $\bar{\nu}(A)$ reported by Nifenecker *et al.*[10] in the following manner:

$$E_{0L} = \bar{\nu}_L \bar{E}_{nL} / (\bar{\nu}_H \bar{E}_{nH} + \bar{\nu}_L \bar{E}_{nL}) E_{0T} ,$$

where E_{0L} is the average excitation energy of the light fragment, $\bar{\nu}_L(\bar{\nu}_H)$ is the average number of prompt neutrons emitted by the light (heavy) fragment, and $\bar{E}_{nL}(\bar{E}_{nH})$ is the average excitation energy carried off by a neutron emitted from the light (heavy) fragment. The quantities $\bar{\nu}_L$, $\bar{\nu}_H$, $\bar{E}_{nL}$, and $\bar{E}_{nH}$ were taken from Ref. 10. We took the variance in the total excitation energy of a given fragment pair to be 86 MeV^2, which is a reasonable average for all pairs except close to symmetric fission.[10] The variances $(\sigma_{E(Z,A)})^2$ in the excitation energy of the individual fragments were taken to be equal for the light and heavy fragment, as was shown to be a reasonable approximation in Ref. 10. Gavron and Fraenkel[11] and Signarbieux *et al.*[12] have shown that there is little correlation between the excitation energies of the two fragments so that the $(\sigma_{E(Z,A)})^2$ were taken to be 43 MeV^2. The spin parameter $B_{Z,A,E}$ was calculated from

$$B_{Z,A,E} = \bar{B} + [E - E_0(Z,A)]/(8\text{ MeV})$$

with $\bar{B}=6$ for $A<130$ and $\bar{B}=7.2$ for $A>130$ which is consistent with angular momentum distribution studies by Wilhelmy *et al.*[13] The calculation was not sensitive to the details of the form of the spin distribution.

The neutron transmission coefficients T_l in Eq. (3) were calculated using the Wilmore-Hodgson potential[14] for $A=110$ (140) for the light (heavy) fragments. The sensitivity of the calculation to the form of the transmission coefficient is discussed below. The γ rays were assumed to be electric dipole ($E1$) and the γ-transmission coefficients T_γ were used in a form which incorporated the dipole sum rule. The T_γ are discussed in detail in a previous paper[15] in which we describe a measurement and a calculation of high-energy γ rays (> 10 MeV) from ^{252}Cf spontaneous fission.

The level densities $\rho(Z,A,E,J)$ were calculated using the semiempirical method of Gilbert and Cameron.[16] The sensitivity of the calculation to this choice of level-density parametrization is discussed below.

In using Eq. (1) to calculate the neutron spectrum, the angular distribution of neutrons in the c.m. system was assumed to be isotropic. A typical calculation of the c.m. spectrum for the mass pair ($Z, A=42$, 108, and 56, 144) is shown in Fig. 1. It can be seen that the usual c.m. spectrum of the Weisskopf evaporation form $\phi(\epsilon) \propto \epsilon \exp(-\epsilon/T)$ cannot describe the whole c.m. neutron energy (ϵ) range for a single temperature T. The low-energy neutrons deviate from the straight line ($T=0.76$) in exactly the same manner as was found in the neutron measurements of Bowman *et al.*[17] The c.m. neutron spectra were transformed to the laboratory using nonrelativistic kinematic expressions assuming isotropic emission. To perform a complete calculation for all the pairs of fragments that are typically involved in the mass and charge distributions from fission would involve prohibitively long computer times. However it is possible to perform a numerical integration over the mass distribution taking advantage of the fact that the two-hump mass distribution from ^{252}Cf spontaneous fission can be reasonably approximated by two Gaussian distributions. The integral to be evaluated for one of the Gaussians is

$$\frac{1}{\sqrt{\pi}} \int_0^\infty \exp(-\chi^2) f(E_n, \chi) d\chi \approx \sum_{i=1}^{k} w_i f(\chi_i) , \quad (5)$$

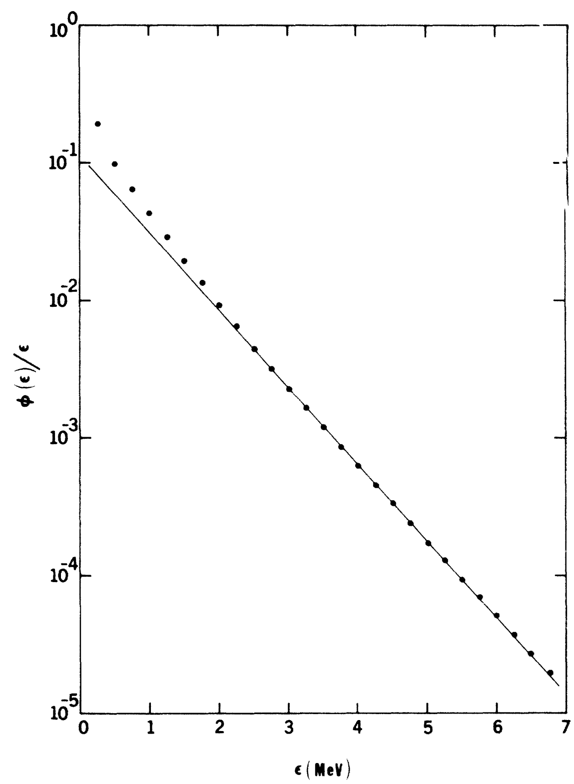

FIG. 1. A typical center-of-mass neutron spectrum $\phi(\epsilon)$ (●) calculated for a light ($Z, A=42$, 108) and a heavy fragment ($Z, A=56$, 144). The results were essentially indistinguishable so the average for the two fragments is shown. The straight line represents a c.m. evaporation spectrum $\phi(\epsilon) \sim \epsilon \exp(-\epsilon/T)$ for a temperature $T=0.76$ MeV. The calculation deviates from this single temperature form similarly to the neutron data of Bowman *et al.* (Ref. 17).

where $\chi = (A - \bar{A})/\sigma\sqrt{2}$ and $f(E_n, \chi)$ is the partial neutron spectrum for a particular fragment. The variance σ of the Gaussian was taken to be 6.5 mass units from the work of Schmitt, Kiker, and Williams.[18] The right-hand side of Eq. (5) is the polynomial approximation to the integral. The χ_i are the zeros of the Hermite polynomials and the weights w_i are calculable from these polynomials[19]; k is the number of Hermite polynomials used to approximate the integral. We therefore chose our fission fragment pairs for the calculation so that the χ_i corresponded to the Hermite polynomial zeros. Calculations were performed for the following mass pairs (light, heavy): (108, 144), (102, 150), (114, 138), (95, 157), and (122, 130). For a given A, the most probable Z was chosen from the charge distribution studies of Reisdorf *et al.*[20] To average over pairing energy effects, we calculated for each mass split listed above an even-even, even-odd, odd-even, and odd-odd case. In total, the calculation was performed for 20 mass pairs or 40 fission fragments.

For each excited fragment Z, A considered, the neutron spectrum was calculated, and then the calculation repeated for $Z, A-1$ with a new mean excitation energy $E_0(Z, A-1) = E_0(Z, A) - S_n(A) - \bar{E}_n$ where $\bar{E}_n$ was the calculated mean c.m. kinetic energy carried off by the emitted neutron. The neutron separation energies $S_n(A)$ were taken from Ref. 8. Third, fourth, and fifth stages were added to this calculation as necessary until the final excitation energy distribution was sufficiently low in energy to prevent further neutron emission.

III. RESULTS AND DISCUSSION

The results of this calculation of the ^{252}Cf fission neutron spectrum along with experimental results extracted from the papers by Meadows[1] and by Green *et al.*[2] are shown in Fig. 2. The calculation resulted in a value for the average total number of neutrons emitted ($\bar{\nu}$) equal to 3.64 which differs from the accepted value of $\bar{\nu} = 3.73$ by 2.5%. In Fig. 2, the calculated $N(E)$ have been divided by 3.64 so that the integral of $N(E)$ is equal to one. This allows a direct comparison with Meadows's results which are presented in the same manner in Ref. 1. The data of Green *et al.* have been arbitrarily normalized to Meadows's data at $E_n = 2$ MeV where the best agreement between the two experiments is expected.

The calculation shows good agreement with the Green data with the calculation being 25% higher than the data at 2 MeV and 70% lower at 10 MeV. The agreement with the Meadows data is not as good since these data imply more high-energy neutrons than the Green data. Both authors pro-

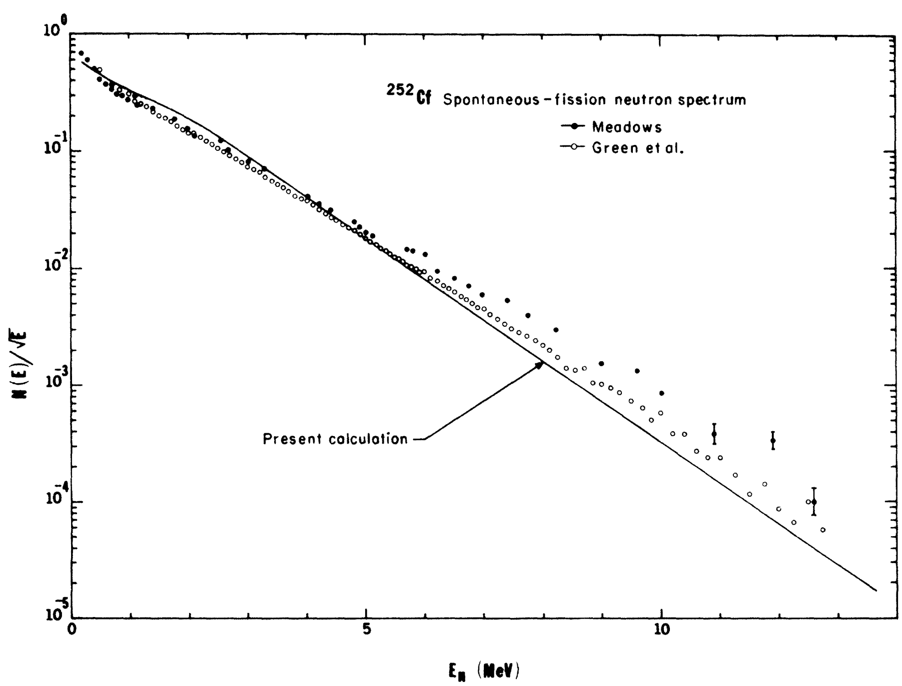

FIG. 2. Experimental and calculated neutron spectra (laboratory energy) for ^{252}Cf spontaneous fission. The solid line represents the results obtained by dividing the calculated $N(E)$ by the calculated value of $\bar{\nu} = 3.64$ so that the integral of $N(E)$ is equal to one as is the case for Meadows's data. The Green *et al.* data are normalized to Meadows's data at 2 MeV.

duced fits to their data assuming Maxwellian distributions of the form $N(E) \sim \sqrt{E}\exp(-E/T)$ with the Meadows data implying $T = 1.57$ MeV, while the Green data yielded $T = 1.40$ MeV. A new measurement of the ^{252}Cf fission neutron spectrum has recently been performed by Auchampaugh[21] and co-workers with results in excellent agreement with the Green data and their derived temperature. A similar Maxwellian fitted to our data yields $T = 1.25$ MeV for $E_n > 2$ MeV.

The greatest sensitivity in our calculation is associated with the level densities which are not known over a wide energy range, especially for fission fragments which are far from the line of stability. An alternate approach to the Gilbert and Cameron method is a thermodynamic calculation described by Huizenga and Moretto[22] which uses a set of shell-model levels and includes a residual interaction in the form of a pairing energy. This procedure was used for two cases ($Z, A = 46$, 116 and 55, 142) to compare with the Gilbert-Cameron level densities. Approximating the results of the two methods by a constant temperature form $\rho \propto \exp(-E/T)$, we find that there is a 10% difference in T between the two methods of calculating level densities. This temperature difference has the effect of raising our calculated values for the neutron spectrum at 10 MeV by approximately 60% and lowering them at 2 MeV by 5%. This improves the agreement between the calculation and the data considerably.

To determine whether the spin-cutoff parameter σ^2 used in the level-density calculation strongly affects the calculated neutron spectrum, we repeated a test case ($Z, A = 42$, 108) for an unreasonably low cutoff of $\sigma^2 = 2$ and for an infinite cutoff. The small cutoff lowers the calculation by approximately 20% at 2 and 10 MeV relative to a value of $\sigma^2 = 36$ which is a typical value for this test case. The value of $\sigma^2 = \infty$ lowered the calculation by 1% at 2 MeV and raised it by 8% at 10 MeV. For the results shown in Fig. 2, σ^2 was calculated for each fragment according to the Gilbert-Cameron procedure. The $\sigma^2 = \infty$ case shows that the calculation is not strongly affected by a reasonable choice of the spin-cutoff parameter.

To check the sensitivity of the calculation to the form of the neutron transmission coefficients we repeated the calculation for several nuclei using neutron transmission coefficients for the Moldauer potential[23] and for a "black" nucleus. The Moldauer T_l resulted in lowering the calculation by 1% at 2 MeV and raising it by 8% at 10 MeV. Similarly, the black nucleus values lower the calculation by 4% at 2 MeV and raise it by 11% at 10 MeV. These results show that the calculation is not extremely sensitive to the form of the neutron transmission coefficient although both the Moldauer and black nucleus values improve the agreement with the experimental data. The calculated neutron spectrum is also not very sensitive to the details of the γ-ray emission spectrum.

Another assumption which would affect the shape of the spectrum is the isotropy of neutron emission in the c.m. system. Bowman *et al.*[17] concluded from their data on neutron emission from ^{252}Cf fission that no more than 90% of the neutrons could have an isotropic distribution in the c.m. system. However, they also concluded that a c.m. angular distribution of the form $1 + a_2P_2(\cos\theta)$ was not able to explain the additional 10% but that it was necessary to assume that these neutrons were emitted isotropically in the laboratory system. Such a possibility is outside the framework of our calculation. However, it should be noted that angular anisotropies will probably not account for most of the differences between the data and the calculation shown in Fig. 2.

In summary, we were able to obtain good agreement with the experimental neutron spectra from ^{252}Cf spontaneous fission using a statistical calculation of the neutron emission from the excited fission fragments employing the Hauser-Feshbach formalism. The calculation was most sensitive to the level densities used, with most of the difference between the calculation and the experimental data probably attributable to this source. This agreement with the data was obtained without introducing arbitrary normalizations or free parameters to fit the data.

---

*Work performed under the auspices of the U.S. Atomic Energy Commission.

[1]J. W. Meadows, Phys. Rev. 157, 1076 (1967).
[2]L. Green, J. A. Mitchell, and N. M. Steen, Nucl. Sci. Eng. 50, 257 (1973).
[3]J. Terrell, Phys. Rev. 113, 527 (1959).
[4]D. W. Lang, Nucl. Phys. 53, 113 (1964).
[5]G. Kluge, Phys. Lett. 37B, 217 (1971).
[6]V. F. Weisskopf, Phys. Rev. 52, 295 (1937).
[7]E. Vogt, in *Advances in Nuclear Physics*, edited by M. Baranger and E. Vogt (Plenum, New York, 1968), Vol. I, p. 270.
[8]G. T. Garvey, W. J. Gerace, R. L. Jaffe, I. Talmi, and I. Kelson, Rev. Mod. Phys. 41, S1 (1969).
[9]S. L. Whetstone, Phys. Rev. 131, 1232 (1963).
[10]H. Nifenecker, C. Signarbieux, R. Babinet, and J. Poitou, in *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1974), Vol. II, p. 117.
[11]A. Gavron and Z. Fraenkel, Phys. Rev. Lett. 27, 1148

(1973).
[12]C. Signarbieux, R. Babinet, H. Nifenecker, and J. Poitou, in *Physics and Chemistry of Fission* (see Ref. 10), Vol. II, p. 179.
[13]J. B. Wilhelmy, E. Cheifetz, R. C. Jared, S. G. Thompson, H. R. Bowman, and J. O. Rasmussen, Phys. Rev. C 5, 2041 (1972).
[14]P. E. Hodgson, Ann. Rev. Nucl. Sci. 17, 1 (1967).
[15]F. S. Dietrich, J. C. Browne, W. J. O'Connell, and M. J. Kay, Phys. Rev. C 10, 795 (1974).
[16]A. Gilbert and A. G. W. Cameron, Can. J. Phys. 43, 1446 (1965).
[17]H. R. Bowman, S. G. Thompson, J. C. D. Milton, and W. J. Swiatecki, Phys. Rev. 126, 2120 (1962).
[18]H. W. Schmitt, W. E. Kiker, and C. W. Williams, Phys. Rev. 137, B837 (1965).
[19]*Handbook of Mathematical Functions*, edited by M. Abramowitz and I. Stegun (U.S. Dept. of Commerce, National Bureau of Standards, 1964), p. 890.
[20]W. Reisdorf, J. P. Unik, H. C. Griffin, and L. E. Glendenin, Nucl. Phys. A177, 377 (1971).
[21]G. F. Auchampaugh, private communication.
[22]J. R. Huizenga and L. G. Moretto, Annu. Rev. Nucl. Sci. 22, 47 (1972).
[23]P. A. Moldauer, Nucl. Phys. 47, 65 (1963).