

Probabilities of Prompt-Neutron Emission from Spontaneous Fission*

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The neutron number distributions from the spontaneous fission of seven isotopes have been measured by use of a cadmium-loaded liquid scintillation tank. The experimental distributions can be roughly approximated by binomial distributions.

The average number of neutrons per spontaneous fission have been found to be 2.30 ± 0.19 for Pu^{236} , 2.33 ± 0.08 for Pu^{238} , 2.18 ± 0.09 for Pu^{242} , 2.65 ± 0.09 for Cm^{242} , 2.84 ± 0.09 for Cm^{244} , and 3.82 ± 0.12 for Cf^{252} , all based on 2.257 ± 0.046 for Pu^{240} .

INTRODUCTION

THE multiplicities of the neutrons from fission—i.e., the fraction of fissions producing 0, 1, 2, . . . , neutrons—are of interest both in theoretical considerations of the fission process and in reactor design. Earlier studies have been limited by the low neutron-detection efficiency of the apparatus to measurements of the first and second moments of the multiplicity distributions.¹⁻¹¹ The cadmium-loaded liquid scintillation tank¹² with its high neutron-detection efficiency permits an accurate determination of the neutron emission probabilities, recent measurements of which have been made by groups at Berkeley and Los Alamos.^{13,14} A theoretical study of these multiplicity distributions has been made by Leachman.¹⁴

APPARATUS

The nuclide to be investigated was mounted as a very thin sample upon a platinum foil, which served as

the cathode in a 3-inch-diameter parallel-plate fission chamber. The fission chamber was positioned at the center of the scintillator tank by a 3-inch-diameter well along the axis of the tank. The neutron detector is nearly identical to that built at Los Alamos.^{12,15} The tank is 30 inches long and 30 inches in diameter, with $\frac{1}{4}$ -inch steel walls. The inside surfaces have been sprayed with molten aluminum as a reflecting and protective coating. Although the reflectivity of aluminum is less than that of some other types of coatings, it was sufficient for our purposes and has shown no tendency to deteriorate over long periods of time.

The scintillator consists of toluene, cadmium propionate, *p*-terphenyl, methanol, and alpha-NPO, and is mixed as described by Reines *et al.*¹² The cadmium-to-hydrogen atom ratio used is 0.0019, resulting in a mean capture time for the neutrons, after thermalization, of approximately 10 microseconds (Fig. 1).

The curved surface of the tank has ninety $\frac{1}{4}$ -inch glass windows of $2\frac{1}{8}$ in. diameter, sealed with Neoprene O-rings. A Dumont 6292 photomultiplier tube, mounted in a soft steel collar, is placed against the outside of each window, with the cathode end immersed in mineral oil to give good optical contact. The photomultiplier tubes are wired in parallel in two banks of 45 tubes each, both banks observing all portions of the scintillator. A copper shield is placed around the entire photomultiplier tube assembly, increasing the tank diameter to 48 inches. The entire apparatus is surrounded by 2 in. of lead and 0.5 in. of boric acid powder, the boric acid powder external to the lead. In order to keep the background low and constant, the apparatus was gated off during all cyclotron and linear accelerator beam pulses.

The external wiring diagram is shown in Fig. 2. The phototube linear amplifiers were designed by William Goldsworthy and have a low-noise, wide-band 950-ohm triode input, matching the 950-ohm 1-microsecond delay line. The fission chamber amplifier and pre-amplifier are of standard design. The pulses from the fission chamber and the photomultiplier tubes are

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¹ de Hoffmann, Serber, and Feynman, Los Alamos Laboratory Report LA-101, 1944 (unpublished); F. de Hoffmann *et al.*, Los Alamos Laboratory Report LA-183 (unpublished); F. de Hoffmann and R. Serber, Los Alamos Laboratory Report LAMS-336 (unpublished); F. de Hoffmann, Los Alamos Laboratory Report LA-471, 1945 (unpublished).

² J. Rotblat, George Holt Physics Laboratory, Liverpool, Report Br-241, 1943 (unpublished).

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⁶ Barclay, Galbraith, and Whitehouse, *Proc. Phys. Soc. (London)* **A65**, 73 (1952).

⁷ F. Barclay and W. Whitehouse, *Proc. Phys. Soc. (London)* **A65**, 73 (1952).

⁸ Crane, Higgins, and Thompson, *Phys. Rev.* **97**, 242 (1955), and *Phys. Rev.* **97**, 1727 (1955).

⁹ Higgins, Crane, and Gunn, *Phys. Rev.* **99**, 183 (1955).

¹⁰ D. Hughes and J. Harvey, Brookhaven National Laboratory Report BNL-325, 1955 (unpublished).

¹¹ K. Geiger and D. Rose, *Can. J. Phys.* **32**, 498 (1955).

¹² Reines, Cowan, Harrison, and Carter, *Rev. Sci. Instr.* **25**, 1061 (1954).

¹³ Hicks, Ise, and Pyle, *Phys. Rev.* **97**, 564 (1955), and *Phys. Rev.* **98**, 1521 (1955).

¹⁴ International Conference on the Peaceful Uses of Atomic Energy, A/Conf. 8/P/592 (1955), compiled by R. B. Leachman. See also R. B. Leachman, accompanying paper [*Phys. Rev.* **101**, 1005 (1956)].

¹⁵ The use of a loaded scintillator tank as an efficient neutron detector at this laboratory was conceived by Dr. Walter E. Crandall for use on another project. In the early stages of its development, information was received about the Los Alamos tank and its design was utilized.

displayed on the sweep of a Tektronix Model 517 oscilloscope, the sweep speed being exponential, i.e.,

$$x = x_0(1 - e^{-t/\tau}),$$

where x is the distance that the sweep has progressed in time t . The sweep duration is set at 30 microseconds and the mean time τ is set at 10 microseconds. A Dumont Model 321 oscillographic record camera was used to record the data. The data were read after projection with a Recordak film projector.

EXPERIMENTAL PROCEDURE

The spontaneous fission event occurring in the fission chamber is accompanied by the release of prompt

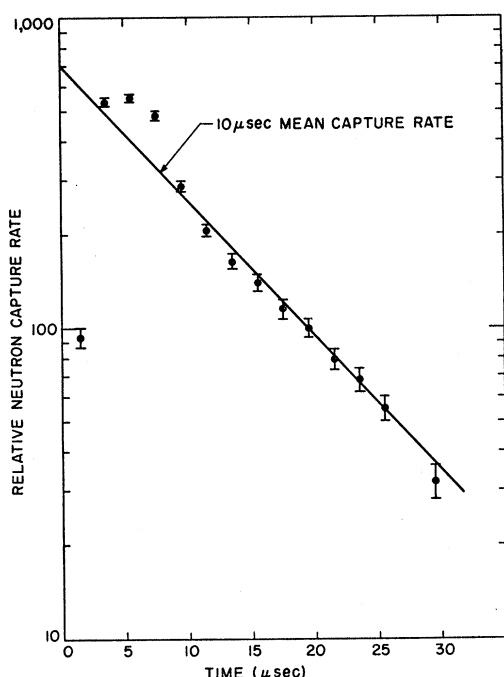


FIG. 1. Experimental capture rates of fission neutrons in the liquid scintillator tank.

neutrons and γ rays. The neutrons transmit practically all of their energies to recoil protons in a time much shorter than a microsecond, although not all the neutrons are completely thermalized until about four microseconds after the event. These recoil protons and any of the converted γ rays from the fission appear as one prompt pulse from the liquid scintillator tank. The thermalized neutrons then are captured exponentially in time by the cadmium-113 (or the hydrogen) in the solution. The Cd^{113} radiative capture immediately gives a cascade decay with a total energy of 9.2 Mev,¹⁶ some fraction of which is converted in the tank and gives a pulse indicating the neutron capture.

¹⁶ G. A. Bartholomew and B. B. Kinsey, Phys. Rev. **90**, 355 (1953); Can. J. Phys. **31**, 1051 (1953); H. T. Motz, Phys. Rev. **90**, 355 (1953).

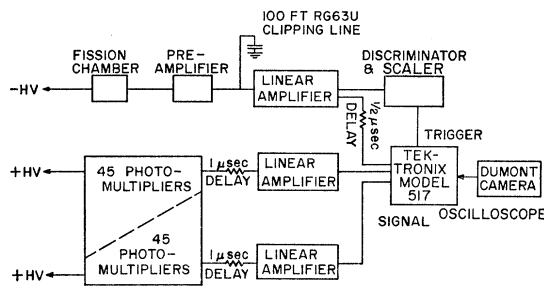


FIG. 2. External wiring diagram.

A fission-chamber discriminator plateau was obtained for each isotope except Pu^{236} , and the discriminator bias was set so that all fissions and no alpha pile-up pulses were observed. In all but 0.3 percent of the events the fission pulse was accompanied by a prompt tank pulse. With Pu^{236} , although the sample was so thin that a good discriminator plateau could in principle have been obtained, the activity was so low as to render this experimentally impractical. In this case the discriminator bias was set so low that some alpha pile-up pulses were observed, but these were readily distinguishable from the fission pulses by the lack of prompt tank pulse.

When a fission occurs, the oscilloscope is triggered and the fission pulse is displayed at the beginning of the sweep. The tank pulses are delayed one microsecond and also displayed on the sweep. Both the fission pulse and the prompt tank pulse were required before any trace was counted as a fission event. This eliminated any chance of mistaking pile-up alpha pulses for fission pulses (Fig. 3).

Californium-252 was run periodically as a calibration for every isotope investigated; this procedure reveals immediately any changes in the over-all efficiency of the apparatus. Experience has shown that after several months the cadmium salt begins to come out of solution, and since appreciable periods of time elapsed between the availability dates of the isotopes, this method of obtaining the relative tank efficiency was the most satisfactory one. In the recent runs the tank efficiency was actually a few percent lower than in the earlier runs. Background pulses with the chamber fission in place were also recorded several times during each run by random triggering of the oscilloscope, using a rate generator.

The absolute tank efficiency for detection of fission neutrons is based on \bar{p} (the mean number of prompt neutrons emitted per fission) for Pu^{240} obtained by



FIG. 3. Sweep triggered by fission-chamber pulse. Left to right: fission pulse, prompt tank pulse from fission γ rays and recoil protons, and four neutron-capture pulses.

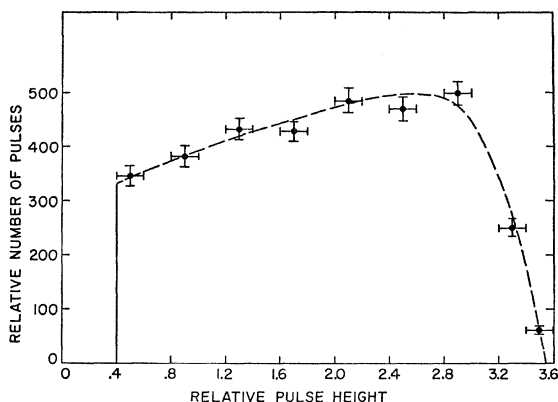


FIG. 4. Differential pulse-height distribution of the neutron-capture pulses on fission traces. Pulse heights less than 0.4 were not read because of rapidly increasing background in this region.

Diven *et al.*,¹⁷ this value having been measured relative to $\bar{\nu}$ for the fission of U^{235} by thermal neutrons.¹⁰ The absolute tank efficiency at the time the Pu^{240} was run was 76.9 ± 2.2 percent.

There are various reasons for the tank efficiency to be less than 100 percent. Some of the neutrons escape from the tank without being captured. Some are captured but give a capture gamma-ray pulse too small to be observed. Because the background counting rate increases rapidly as smaller pulses are accepted, a lower pulse-height limit was set, and this caused the loss of all neutron pulses smaller than this lower limit. The differential pulse-height distribution of the pulses on fission trace is given in Fig. 4, which also shows the lower acceptance limit. Since the oscilloscope sweep length was only 30 microseconds, approximately 7 percent of the neutrons captured in the tank were captured after the sweep had ended.

Since a possibility exists that the number of neutrons per fission event is correlated with the neutron kinetic energy, the neutron-capture efficiency of the scintillator tank as a function of the neutron energy must be considered. Monte Carlo calculations performed at the

University of California Livermore Laboratory show that the efficiency is about 99 percent for 1-Mev neutrons, 95 percent for 3-Mev neutrons, 89 percent for 5-Mev neutrons, and 84 percent for 7-Mev neutrons, when the neutrons are emitted isotropically at the center of the tank. If it is assumed that the energy spectra of the neutrons from all the isotopes approximate that from the fission of U^{235} by thermal neutrons, an over-all efficiency for capturing a fission neutron in the scintillator is calculated to be about 95 percent. Theoretical considerations by Leachman¹⁴ indicate that the above assumption is quite accurate. In addition, preliminary results of an experiment being performed at the Nobel Institute¹⁸ indicate that the peak of the energy spectrum of the neutrons from the spontaneous fission of Cf^{252} is only slightly higher than the peak energy for U^{235} fission by thermal neutrons.

If all the high-energy neutrons originate from fissions giving only a single neutron, some bias would be expected in the data. Hammel and Kephart,¹⁴ however, working with a 16-in. diameter tank, have obtained emission probabilities of neutrons from Pu^{240} fission that are in good agreement with those obtained in this work, and since their capture efficiencies for high-energy neutrons are considerably less than for the larger tank, it can be concluded that no very marked relation exists between neutron energy and number of neutrons per fission.

We have considered the possibility that some of the pulses on a fission sweep were due to the decay of fission products with half-lives of a few microseconds. To check on this possibility, we repeated the Cm^{244} run using a scintillator solution with no cadmium, and time and pulse-height distributions were measured for the tank pulses. The time distribution was consistent with an exponential capture rate for neutrons in toluene. The pulse-height distribution showed the 2.2-Mev gamma ray from hydrogen capture, with a resolution of approximately 20 percent. We conclude from this that the effect of secondary fission products is negligible.

TABLE I. Observed neutron multiplicity distributions from spontaneous fission and background pulses per sweep.

	Pu^{238}	Pu^{240}	Pu^{242}	Cm^{242}	Cm^{244}	Cf^{252}	
F_0''	42	642	368	471	569	999	
F_1''	85	1532	1067	920	2495	4259	
F_2''	78	1450	1086	767	3584	5814	
F_3''	31	620	553	282	2312	3750	
F_4''	7	179	174	44	723	1174	
F_5''	1	25	20	10	159	188	
F_6''	0	0	1	0	13	16	
F_7''	0	0	0	0	2	0	
F_8''	0	0	0	0	0	8	
Total	244	4448	3269	2494	9857	16200	
Back-ground counts per sweep	0.0106	0.0108	0.0108	0.0109	0.0109	0.0081	0.0150

¹⁷ B. Diven and J. Terrell (private communication). See also Diven, Martin, Taschek, and Terrell, preceding paper [Phys. Rev. **101**, 1012 (1956)].

ANALYSIS OF DATA

The observed numbers of fissions giving 0, 1, 2, \dots , pulses per sweep are given in Table I. Two corrections are made before the true multiplicity distribution is calculated.

(a) "Resolving Time"

Because of the exponential sweep the neutron-capture pulses are uniformly distributed distancewise along the sweep trace, so it is better to use the "resolving distance" ΔL . The probability that two pulses fall within a cell ΔL long is

$$2\Delta L \int_0^L \frac{1}{L^2} dx = 2\Delta L/L.$$

¹⁸ H. Slätis (private communication, reporting work of E. Hjalmar, M. Punnis, H. Slätis, and S. Thompson).

TABLE II. Calculated probabilities of emitting ν neutrons per spontaneous fission, $P(\nu)$, and the average number of neutrons per spontaneous fission, $\bar{\nu}$, based on $\bar{\nu}=2.257\pm 0.046$ for Pu^{240} .

$P(\nu)$	Pu^{236}	Pu^{238}	Pu^{240}	Pu^{242}	Cm^{242}	Cm^{244}	Cf^{252}
P_0	0.062 ± 0.035	0.044 ± 0.009	0.041 ± 0.009	0.063 ± 0.013	0.011 ± 0.005	0.001 ± 0.004	0.001 ± 0.001
P_1	0.156 ± 0.090	0.175 ± 0.026	0.219 ± 0.021	0.192 ± 0.034	0.126 ± 0.018	0.099 ± 0.017	0.021 ± 0.007
P_2	0.38 ± 0.13	0.384 ± 0.026	0.351 ± 0.021	0.351 ± 0.041	0.323 ± 0.018	0.281 ± 0.022	0.111 ± 0.019
P_3	0.28 ± 0.12	0.237 ± 0.027	0.241 ± 0.020	0.324 ± 0.047	0.347 ± 0.020	0.365 ± 0.018	0.271 ± 0.019
P_4	0.096 ± 0.086	0.124 ± 0.021	0.127 ± 0.018	0.033 ± 0.026	0.139 ± 0.013	0.198 ± 0.020	0.326 ± 0.018
P_5	0.033 ± 0.036	0.036 ± 0.009	0.020 ± 0.006	0.036 ± 0.013	0.050 ± 0.009	0.049 ± 0.009	0.178 ± 0.016
P_6			0.001 ± 0.002		0.004 ± 0.002	0.007 ± 0.002	0.077 ± 0.013
P_7					0.001 ± 0.001		0.013 ± 0.004
P_8							0.003 ± 0.001
$\bar{\nu}$	2.30 ± 0.19	2.33 ± 0.08	2.257 ± 0.046	2.18 ± 0.09	2.65 ± 0.09	2.84 ± 0.09	3.82 ± 0.12

* See reference 17.

If n pulses are observed on a trace, it is possible that there were n pulses, all of them resolved, or that there were $n+1$ pulses, two of which are not resolved. Higher order corrections have been neglected. If $F''(n)$ is the number of fissions observed with n pulses per fission, and $F'(n)$ the corresponding number for $\Delta L=0$, then

$$F''(n) = F'(n) \left[1 - \frac{n!}{(n-2)!} \frac{2\Delta L}{L} \right] + F'_{n+1} \frac{(n+1)!}{(n-1)!} \frac{2\Delta L}{L}$$

The $F'(n)$ were then calculated, with $2\Delta L/L=0.0030\pm 0.0015$, and with the value for ΔL estimated from the appearance of the sweeps. The separation of two pulses was measured for a number of cases, at all positions along the sweep, where two pulses could just be identified as separate pulses.

(b) Background

The background count rate γ was about 0.01 pulse per sweep. The corrected number of fissions is calculated from

$$F(n) = [(F'(n) - \gamma F(n))] / (1 - \gamma)$$

The number of background sweeps with two or more pulses was completely negligible. The background rates for the various isotopes are given in Table I.

If $F(n)$ is the corrected multiplicity distribution, the true distribution $P(\nu)$ is obtained from

$$P(\nu) = \sum_{n=\nu}^{n=n_{\max}} F(n) \frac{n!}{\nu!(n-\nu)!} \epsilon^{-n} (\epsilon-1)^{n-\nu}$$

The errors on the calculated $P(\nu)$ have five sources: (a) counting statistics, (b) uncertainty in the efficiency because of the uncertainty of $\bar{\nu}$ for Pu^{240} , (c) background fluctuations (which are negligible), (d) uncertainty in the value of ΔL used in the resolution corrections, (e) reading errors. [There are small differences in the data obtained when one person reads the film twice, and similar fluctuations among different readers. All film has been read at least twice. The uncertainty is small compared to (a), (b), and (d).]

These calculations were made on an IBM Type 650 calculator at Livermore. The calculated values of $P(\nu)$ and their standard errors are given in Table II.

DISCUSSION

The multiplicity distributions of Pu^{240} , Cm^{242} , and Cf^{252} are shown in Fig. 5. The distributions of the Pu^{236} , Pu^{238} , Pu^{242} , and Cm^{244} are similar to those shown in the figure.

Points obtained from the binomial distribution,

$$\frac{\nu_{\max}!}{\nu!(\nu_{\max}-\nu)!} \left(\frac{\bar{\nu}}{\nu_{\max}} \right)^\nu \left(1 - \frac{\bar{\nu}}{\nu_{\max}} \right)^{\nu_{\max}-\nu}$$

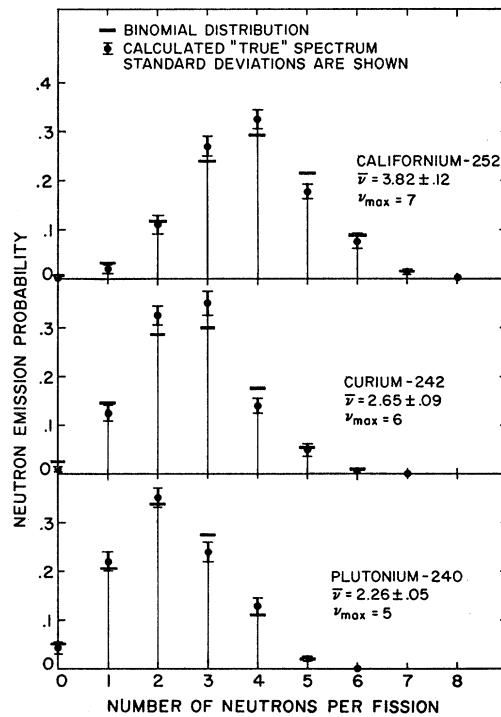


FIG. 5. The probabilities for prompt-neutron emission from the spontaneous fission of plutonium-240, curium-242, and californium-252. Binomial distributions roughly approximating the emission probabilities are also shown.

also are shown in Fig. 5. It can be seen that this distribution gives a fair approximation to the experimental points, but using the χ^2 test we have been unable to find any binomial distributions that fit the experimental results with more than a negligible probability.

The theoretical emission probabilities calculated by Leachman¹⁴ are in good agreement with the results given in Table II.

It should be pointed out that the discrepancy as reported earlier¹³ between our preliminary value for $\bar{\nu}$ of Cf²⁵² based on Pu²⁴⁰ and that of Crane, Higgins, and Thompson⁸ has been partially resolved. The Pu²⁴⁰ source used in our preliminary work was rather thick, and about one-quarter of the fissions were lost by foil absorption. The error in the early value might be explained by assuming that the lower-energy fissions, which would be more easily lost by foil absorption, have a higher internal excitation and give off more neutrons than the high-energy fissions. Preliminary work indicates that this is true. We are comparing the average number of neutrons and neutron multiplicities with the energy mode of fission using a "back-to-back" fission chamber. The remaining discrepancy may be due to the different methods of absolute calibration.

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Relativistic Radiative Transitions*

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We have calculated the oscillator strength for electric dipole transitions with retardation between the $1s$ state and higher discrete states for a single Dirac electron in a Coulomb field. Numerical values for atomic number 82 show that the retarded relativistic oscillator strength for each shell is about 0.8 of the nonrelativistic value. We give numerical values for the relative intensity of the different K x-ray lines for lead, neglecting screening: the $K\alpha_1/K\alpha_2$ doublet intensity ratio is 1.93 as compared to the NR value of 2.0. We introduce an average "oscillator density" for transitions for discrete states, and extrapolate to the series limit to find the photoelectric cross section at threshold. Our

numerical value for lead of 740 barns is 23% larger than the value given by Hulme *et al.* We also calculate the summed oscillator strength, using a cutoff at an arbitrary high energy to give convergence. The numerical results for lead are insensitive to the numerical value of the cutoff used: using 10 Mev, we find a summed oscillator strength of 0.85, with an increase of 0.01 for each factor of two increase in the cutoff energy. Our value of the summed oscillator strength is less than that calculated nonrelativistically by Thomas, Reiche, and Kuhn; or relativistically by Gell-Mann, Goldberger, and Thirring.

I. INTRODUCTION

THE oscillator strengths for electric dipole ($E1$) radiative transitions between discrete nonrelativistic (NR) states in a Coulomb field are given in closed form and tabulated numerically by Bethe.¹ The

same approximation gives the Stobbe formula^{1,2} for the atomic photoeffect. The much more difficult relativistic calculations of the photoeffect have been performed numerically by Hulme *et al.*³ and approximate analytic forms given by Hall,⁴ and Sauter.⁵

Jacobsohn's calculations⁶ for discrete-discrete transi-

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