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SOME FEATURES OF THE SPONTANEOUS FISSION OF U^{238}

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The average number of neutrons emitted per event of spontaneous U^{238} fission, $\overline{\nu} = 2.1 \pm 0.1$, and the quantity $\Delta = (\overline{\nu^2} - \overline{\nu})/\overline{\nu}^2 = 0.95 \pm 0.05$, which characterizes the neutron distribution, were measured by a double-coincidence technique. These values, as well as the results of previous studies of neutron emission from spontaneous U^{238} fission do not agree with the semi-empirical laws valid for most investigated nuclei. The number of neutrons emitted was determined to be 64.5 ± 2.0 per gram-hour. The decay constant and spontaneous fission half-life computed from the data obtained in the present investigation are 31 ± 1.5 fissions per gram-hour and $(6.5 \pm 0.3) \times 10^{15}$ years, respectively.

INTRODUCTION

THOROUGH investigations of prompt neutrons from fission have clarified many details of the process of neutron emission by excited fragments. A number of experiments[1,2] and calculations[3] have shown that the average number $\overline{\nu}$ of neutrons emitted per fission event increases approximately linearly with the excitation energy E_X of the fissioning nucleus. A comparison of the data on $\overline{\nu}$ for the spontaneous fission[2] of Pu^{240} and Pu^{242} and the neutron-induced fission of Pu^{239} and Pu^{241} shows that the linear dependence of $\overline{\nu}$ on E_X can evidently be extended into the sub-threshold region of excitation energies as far as the spontaneous fission of an unexcited compound nucleus.

The excitation energies E_X of fragments from neutron-induced fission and from spontaneous fission differ by the amount $E_{inc} + E_{comp}$ where E_{inc} is the kinetic energy of the incident neutron and E_{comp} is its binding energy in the compound nucleus. It is assumed that the average kinetic energy of fragments is independent of the excitation energy of the fissioning nucleus.[3] Therefore the linear dependence of $\overline{\nu}$ on E_X can be applied to experimental data on neutron-induced fission to obtain $\overline{\nu}$ for the spontaneous fission of a number of nuclides for which the direct measurement of this quantity is practically impossible.

In references 4 and 1 an attempt was made to calculate the dependence of $\overline{\nu}$ on the number of nucleons A and charge Z of a spontaneously fissioning nucleus. The results of these calculations are in satisfactory agreement with both the experimental values of $\overline{\nu}$ for spontaneous fission and with the values obtained by extrapolating data on induced fission. An exception is provided by the values of $\overline{\nu}$ for the spontaneous fission of U^{238}, 2.26 ± 0.16 and 2.4 ± 0.2, which do not agree with the calculation and appreciably exceed the extrapolated values of $\overline{\nu}$ for neighboring uranium isotopes. Extrapolation of the experimental values of $\overline{\nu}$ for photofission of U^{238} yields a result which is considerably lower than the two values given above.[5]

In addition to this discrepancy an anomaly in the distribution of the number of neutrons is observed in the spontaneous fission of U^{238}. The probability $P(\nu)$ of emitting a given number ν of neutrons in one fission event is represented satisfactorily for the great majority of the investigated nuclei by the binomial law[6]

$$P(\nu) = \frac{\nu_m!}{\nu!\,(\nu_m - \nu)!}\left(\frac{\overline{\nu}}{\nu_m}\right)^{\nu}\left(1 - \frac{\overline{\nu}}{\nu_m}\right)^{\nu_m - \nu}, \tag{1}$$

where ν_m is the maximum possible number of emitted neutrons. From (1) we obtain the relation

$$\Delta = (\overline{\nu^2} - \overline{\nu}) / \overline{\nu}^2 = 1 - 1/\nu_m. \tag{2}$$

An experimental study of the distribution $P(\nu)$[6] determined that Δ for both spontaneous and neutron-induced fission of nuclides from U^{233} to Cf^{252} is given approximately by the semi-empirical expression

$$\Delta = 0{,}714 \pm 0{,}035\,\overline{\nu} \tag{3}$$

and thus depends only slightly on $\overline{\nu}$; its values for the aforementioned nuclides lie in the interval $0.79 - 0.85$.

Geiger and Rose[7] have measured the ratios of the first three moments of the distribution $P(\nu)$ for spontaneous fission of U^{238} ($\overline{\nu^2}/\overline{\nu} = 3.26$, $\overline{\nu^3}/\overline{\nu} = 12.73$), the relation between which is better represented by the Poisson distribution

$$P(\nu) = \bar{\nu}^{\nu} e^{-\bar{\nu}} / \nu!, \qquad (4)$$

than by the binomial law (1). From these results it follows that for spontaneous fission of U^{238} we have $\Delta = 1$, whereas from (3) for $\bar{\nu} = 2.3$ we obtain $\Delta = 0.79$.

Since the experimental values of $\bar{\nu}$ and Δ for spontaneous fission of U^{238} were obtained by indirect methods[7,8] and do not obey the same relations as for a number of other nuclides, the direct experimental measurement of these quantities was of definite interest.

MEASUREMENT OF $\bar{\nu}$

$\bar{\nu}$ was determined by relative measurements of the number of coincidences between detectors of prompt neutrons and of spontaneous fissions of U^{238} and Pu^{240}. Two multilayer ionization chambers in parallel (Fig. 1) were used to detect spontaneous fissions of U^{238}. 12 g of uranium depleted of U^{235} were deposited in layers 2 mg/cm^2 thick on both sides of aluminum foils. An identical chamber contained 2.5 mg of Pu^{240} (92% Pu^{240} and 8% Pu^{239}) on platinum foil. The chamber containing U^{238} was filled with argon to a pressure of 5 atm, while the chamber containing plutonium was filled with a mixture, 90% Ar + 10% CO_2, to 35 mm Hg. The lower pressure of this mixture considerably improved the discrimination between fission fragments and piled-up α particles. The operating point on the characteristic of the fission chambers was selected by extrapolating the counting rate for α particles to 0.1% of the spontaneous fission intensity.

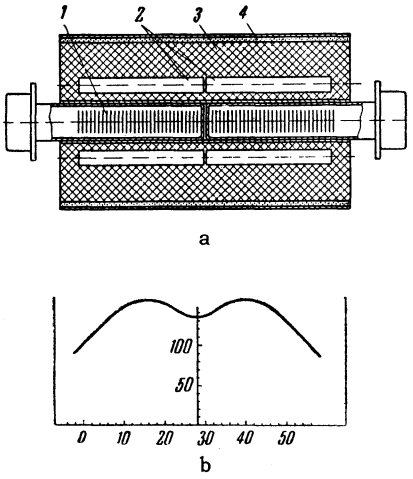

FIG. 1. a) Diagram of apparatus for measurement of $\bar{\nu}$ (U^{238}). 1 – layers of U^{238}, 2 – $B^{10}F_3$ counters, 3 – paraffin, 4 – shielding layer; b) efficiency of neutron registration along the detector: The vertical axis represents the number of counts per unit time, while the horizontal axis represents the distance from the edge of the detector in cm.

The fission chambers were surrounded by 24 proportional $B^{10}F_3$ counters in paraffin, as shown in Fig. 1, which were connected in parallel. The efficiency of registering prompt fission neutrons was ~ 4%. The electronic equipment registered pulses from the chambers, counters and coincidence circuits, which had a resolving time of ~ 6×10^{-4} sec. Accidental coincidences during measurements with Pu^{240} amounted to less than 0.2%, and ~ 0.01% with U^{238}, and were therefore not taken into account.

The total number of registered coincidences was ~ 2400 for U^{238} and ~ 12,000 for Pu^{240}; as a result of these measurements a correction was introduced for nonuniform efficiency along the neutron detector. Three experimental runs were in satisfactory agreement and yielded the ratio $\bar{\nu}(U^{238})/\bar{\nu}(Pu^{240}) = 0.92 \pm 0.03$. Using $\bar{\nu}(Pu^{240}) = 2.26 \pm 0.05$,[2] we obtain $\bar{\nu}(U^{238}) = 2.1 \pm 0.1$.

MEASUREMENT OF Δ

The measurement of Δ was based on double coincidences between neutrons from a single fission event. These methods have been described in detail by Geiger and Rose.[7] Samples of uranium and plutonium were placed inside a paraffin block together with two six-counter groups of $B^{10}F_3$ counters connected for coincidences (Fig. 2). Eight disks, each consisting of 200 g of depleted uranium, were spaced uniformly along the length of the neutron detector. Calibration was performed by means of a thin spherical plutonium sphere containing about 2% Pu^{240}. With a shell thickness of 1.5 mm the multiplication constant was 1.05. For Pu^{240} Δ is known to have the value 0.807 ± 0.008.[6]

The pulse counting rate of each channel was

$$N_i = \bar{\nu} F \varepsilon_i, \qquad i = 1, 2. \qquad (5)$$

The double coincidence counting rate was

$$N_c = (\overline{\nu^2} - \bar{\nu}) F \varepsilon_1 \varepsilon_2 \eta, \qquad (6)$$

where F is the number of spontaneous fissions

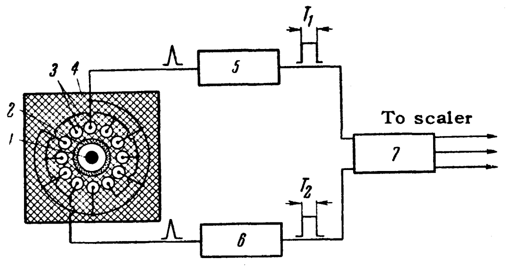

FIG. 2. Diagram of apparatus for measurement of Δ: 1 – U^{238}, 2 – shielding layer of B_4C, 3 – $B^{10}F_3$ counters, 4 – paraffin, 5 and 6 – amplifiers, 7 – coincidence circuit.

per unit time in the sample: ϵ_1, ϵ_2 are the efficiencies of the counting channels; η is the double-coincidence selection factor which takes into account the finite resolving time of the coincidence circuit.

For U^{238} and Pu^{240} (5) and (6) with the notation $\delta = (\overline{\nu^2} - \overline{\nu})/\overline{\nu}$ give

$$\frac{\delta_U}{\delta_{Pu}} = \frac{N_c(U)}{N_c(Pu)} \frac{(N_1+N_2)_{Pu}}{(N_1+N_2)_U} \frac{\varepsilon_{Pu}}{\varepsilon_U} \frac{\overline{\varepsilon_U^2}}{\overline{\varepsilon}_U^2}. \tag{7}$$

Our experimental ratio was $\delta_U/\delta_{Pu} = 1.085 \pm 0.02$.

Using the known values of $\overline{\nu}$ and Δ for Pu^{240} and the measured value of $\overline{\nu}$ for U^{238}, we obtain

$$\Delta_U = 0.95 \pm 0.05.$$

This result includes a correction for the multiplication of spontaneous fission neutrons in the samples and geometric factors ϵ_{Pu}/ϵ_U and $\overline{\epsilon_U^2}/\overline{\epsilon}_U^2$ accounting for the difference in the arrangements of the uranium and plutonium samples.

MEASUREMENT OF THE SPONTANEOUS FISSION HALF-LIFE

In addition to giving the values of $\overline{\nu}_U$ and Δ_U, (5) and (6) can be used to determine the number of neutrons $Q = \overline{\nu}F/M$ emitted in unit time by a gram of uranium. Q was measured in three ways.

(a) Comparison of the channel counts for the uranium and plutonium samples gives

$$\frac{(N_1+N_2)_U}{(N_1+N_2)_{Pu}} = \frac{(\bar{\nu}F)_U}{(\bar{\nu}F)_{Pu}} \frac{\varepsilon_U}{\varepsilon_{Pu}}. \tag{8}$$

The determination of $(\overline{\nu}F)_U$ from this equation required knowledge of F_{Pu}, which can be calculated if the concentration of Pu^{240} in the spherical shell is known. This concentration was measured by comparing the emitted neutron intensity from the sphere and from a standard plutonium disk of known concentration and weight. From (8) we then obtained Q = 60.3 ± 3.6 neutrons/g-hr.

b) Using the value of F_{Pu} measured above, it is easy to determine $(\overline{\nu}F)_U$ and therefore Q from the relation

$$\frac{(N_c/N_1N_2)_U}{(N_c/N_1N_2)_{Pu}} = \frac{(F\bar{\nu})_{Pu}}{(F\bar{\nu})_U} \frac{\overline{\varepsilon_U^2}}{\bar{\varepsilon}_U^2} \frac{\delta_U}{\delta_{Pu}}. \tag{9}$$

In this way we obtain Q = 67.5 ± 3.7 neutrons/g-hr.

c) Q can be determined by measuring the selection factor η, independently of F_{Pu}, using

$$\left(\frac{N_c}{N_1N_2}\right)_U = \frac{\eta}{(F\bar{\nu})_U} \frac{\overline{\varepsilon_U^2}}{\bar{\varepsilon}_U^2} \delta_U. \tag{10}$$

The selection factor was determined by studying the dependence of the number of true coincidences on the resolving time of the coincidence circuit. With τ as the mean life of slowed-down neutrons in the detector the time distribution of registered neutrons obeys the law $\tau^{-1}e^{-t/\tau}$. The probability of registering the coincidence of two neutrons can be obtained from the relation

$$\eta = \int_0^\infty e^{-t/\tau}\frac{dt}{\tau}\int_t^{t+T_1} e^{-t'/\tau}\frac{dt'}{\tau} + \int_0^\infty e^{-t/\tau}\frac{dt}{\tau}\int_t^{t+T_2} e^{-t'/\tau}\frac{dt'}{\tau}$$
$$= 1 - (e^{-T_1/\tau} + e^{-T_2/\tau})/2, \tag{11}$$

where T_1 and T_2 are the durations of pulses reaching the coincidence circuit from the first and second channels, respectively.

In the present experiment we have

$$\eta \approx 1 - e^{-T/\tau}, \tag{12}$$

since the pulse durations in both channels were selected to be approximately equal: $T_1 \approx T_2 = T$. T also becomes the resolving time of the coincidence circuit. The mean neutron lifetime τ was determined by bringing the experimental relation between the number of coincidences and the resolving time T into best agreement with (11).

From all of the foregoing data we obtained

$$\tau = 1.44 \cdot 10^{-4} \text{ sec}, \quad \eta = 0.82 \pm 0.02$$

$$\text{for } T = 2.38 \cdot 10^{-4} \text{ sec}.$$

A calculation by means of (10) gives Q = 65.0 ± 2.3 neutrons/g-hr. The number of neutrons emitted by a gram of uranium per hour, which was obtained by averaging the values of Q determined by the methods described above, is

$$Q = (64.5 \pm 2) \text{ neutrons/g-hr}.$$

Hence the decay constant $\lambda = Q/\overline{\nu}$ and the spontaneous fission half-life $T_{1/2}$ become

$$\lambda = (31 \pm 1.5) \text{ fissions/g-hr},$$

$$T_{1/2} = (6.5 \pm 0.3) \cdot 10^{15} \text{ years}$$

DISCUSSION OF RESULTS

Our measured result $\Delta = 0.95 \pm 0.05$ agrees with the measurements of Geiger and Rose. However, we cannot conclude on the basis of these experiments that the number of neutrons emitted per spontaneous fission of U^{238} obeys a Poisson distribution. Neither a binomial distribution nor a Poisson distribution for $P(\nu)$ can apparently be justified theoretically, since the probability for the emission of a given number ν of neutrons is determined by the distribution of the excitation energy. Nevertheless, the Poisson law is a worse

TABLE I

Authors	Q neutrons/g-hr	Authors	Q neutrons/g-hr
Fermi[11]	54		
Scharff-Goldhaber and Klaiber[12]	63	Hanson[6]	57.5±3.6
		Rotblat[6]	53 ±13
Pose[13]	55.5	Littler[8]	59.5±3.3
		Present authors	64,5±2.0

approximation than the binomial law to the real distribution P (ν), since with the former $\nu_m = \infty$ and $\Delta \equiv 1$, independently of the excitation energy of the fissioning nucleus, and this conflicts with experimental results.

The binomial distribution is also insufficiently accurate since, for example, Eqs. (2) and (3) now give $\nu_m = 1/(0.286 - 0.035)$, which is smaller than the observed maximum number of neutrons per fission, at least for the spontaneous fission of Cm^{244} and Cf^{252}.[6,9] The real distribution P (ν) is evidently somewhere between the two distributions mentioned.

The deviation of the experimental value of Δ from (3) clearly indicates that the width of the excitation energy distribution in the spontaneous fission of U^{238} is greater than for other nuclides with the same mean excitation energy. Leachman calculated $\overline{\nu^2}/\nu = 2.70$, which is closer to our value (2.98 ± 0.05) than to the result obtained by Geiger and Rose (3.26 ± 0.16).

Our result, $\bar{\nu} = 2.08 \pm 0.08$, for the spontaneous fission of U^{238} agrees within experimental error with the value obtained by Geiger and Rose (2.26 ± 0.16) but is smaller than Littler's result (2.4 ± 0.2). The data indicate, as previously, that $\bar{\nu}$ for the spontaneous fission of U^{238} is ~ 0.4 above the results calculated in references 1 and 4. In these papers it was assumed that the kinetic energy of fragments depends linearly on $Z^2/A^{1/3}$, which is only slightly different for close isotopes, so that the computed kinetic energy of fragments is almost identical for neighboring uranium isotopes. However, Kovrigin and Petrzhak have shown in a recent paper[10] that the mean kinetic energy of fragments from the spontaneous fission of U^{238} is 3 or 4 Mev lower than in the fission of U^{235} induced by thermal neutrons. This indicates that the actual excitation energy of fragments in the spontaneous fission of U^{238} is 3 or 4 Mev higher than that calculated in references 1 and 4; $\bar{\nu}$ is thus increased by 0.4 or 0.5.

It is interesting to compare $\bar{\nu}$ for the spontaneous fission and photofission of U^{238}. Reference 5 gives $\bar{\nu} = 1.6 \pm 0.5$ for U^{238} fission induced by γ rays with about 5 Mev. Assuming that with rising excitation energy $\bar{\nu}$ increases linearly and $d\bar{\nu}/dE = 0.13$ Mev^{-1},[1] we would expect $\bar{\nu} \approx 2.8$ in photofission induced by 5-Mev γ rays.

The features of U^{238} fission discussed in the present paper show the inadequacy of semi-empirical laws which ignore the characteristics of individual nuclides.

Tables I and II compare the values of Q and λ obtained in the present work and by other investigators.

TABLE II

Authors	λ, fissions/g-hr
Segre[11]	24.8±0.9
Whitehouse and Galbraith[14]	24,2±1.5
Flerov et al.[15]	19.8±2.9
Present authors	31.0±1.5

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