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NUMBER OF NEUTRONS EMITTED BY U^{235} *FISSION FRAGMENTS*

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The number ν of neutrons emitted by the fragments of U^{235} fission induced by thermal neutrons are measured with an ionization chamber and a cadmium neutron detector. The data obtained contradict the hypothesis of a universal dependence of ν on the fragment mass. Calculations show that the fragment excitation energy increases by ~ 20 MeV in the vicinity of symmetrical fission.

INTRODUCTION

It has been reliably established recently that the total kinetic energy of fragments decreases sharply in the region of symmetrical fission[1,2]. The magnitude of this effect cannot yet be regarded as firmly determined, but at any rate it amounts to tens of MeV.

As is well known, the sum of the fragment excitation energy and the fragment kinetic energy (the total energy release) can be calculated for each fragment mass ratio from a semi-empirical formula. The energy release calculated in this manner does not experience strong changes in the symmetrical-fission region. Consequently the question of reviewing our notions concerning the energy balance arose immediately after the publication of [1,2]. The only known method of determining the fragment excitation energy is to determine the number of neutrons (and gamma quanta) emitted by the fragments. Experiments of this type were undertaken earlier[3-5], but in view of certain difficulties no information existed concerning the excitation energy of the Pu^{240} and $U^{234,236}$ fragments in the mass-ratio region in question. Very valuable information obtained by Whetstone[6] concerned the Cf^{252} spontaneous fission electrons, and especially the data obtained by Bowman et al.[7], did not solve the problem, since no noticeable singularities were observed in the behavior of the total kinetic energy of the fragments of this isotope near the symmetrical fission.

We have measured in [8,9] the total number of neutrons emitted by both fragments in the case of the fission of U^{233}, U^{235}, and Pu^{239} by thermal neutrons. A noticeable increase in the neutron number ν was observed in the region of symmetrical fission, and the minimum of the neutron number coincided in position with the maximum of the total kinetic energy. However, one could not conclude from these investigations with any degree of certainty that a quantitative agreement exists between the increase in the excitation energy and the decrease in the kinetic energy, in view of the insufficient resolution of the fragment-mass analysis.

In the present investigation we succeeded in improving the mass analysis resolution. We measured with the aid of the new apparatus the number of neutrons emitted by individual U^{235} fission fragments.

EXPERIMENT

The work was done with a neutron beam ($\sim 5 \times 10^6$ cm^2/sec) from the thermal column of the IRT-1000 reactor. The arrangement of the apparatus is shown schematically in Fig. 1. The following is a brief description of the units of the apparatus and of its operation as a whole.

1. Neutron detector. The detector was a $40 \times 40 \times 20$ cm parallelepiped filled with a liquid organic scintillator. The scintillating substance was a solution of 2.5-diphenyloxazole (PPO) in dioxane with a concentration of 5 g/liter, to which was added an almost-saturated aqueous solution of cadmium nitrate in an amount making the lifetime of the thermal neutrons 12 microseconds. To increase the light yield, 30 g/liter of naphthalene was added to the solution. The scintillations produced by the gamma quanta due to neutron capture in cadmium were registered by six FÉU-24 photomultipliers, the outputs of which were connected together.

2. Ionization chamber. The construction of the chamber was governed by two main requirements: collimation of the fragments and attainment of high resolution.

A layer of uranium of thickness $\sim 20\ \mu g/cm^2$ deposited by electric spraying of the solution on a gilded ($\sim 15\ \mu g/cm^2$ of gold) collodion film

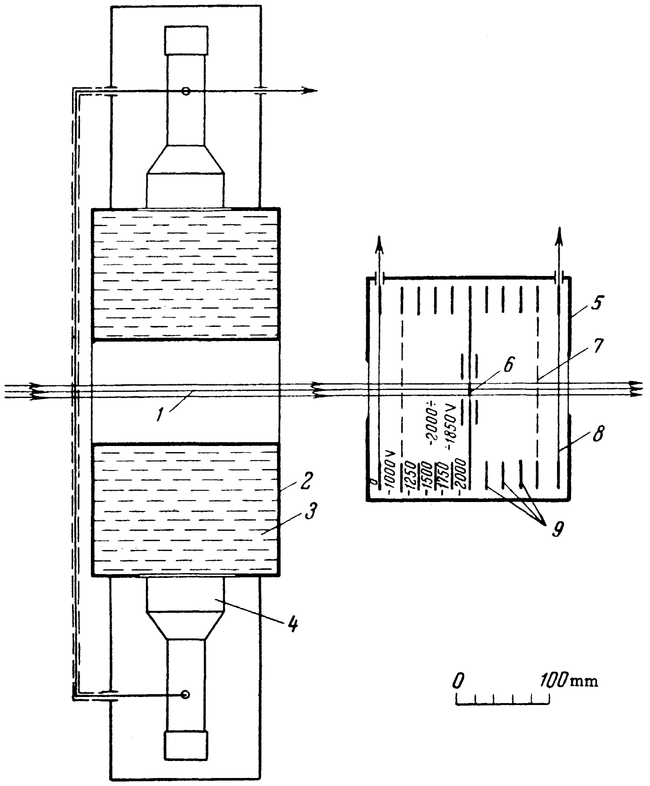

FIG. 1. Diagram of the apparatus: 1–neutron beam, 2–housing of neutron detector, 3–scintillator, 4–FÉU-24 photomultiplier, 5–housing of ionization chamber, 6–uranium layer, 7–grid, 8–collecting electrode, 9–rings to adjust the potential on the boundary.

~ 4 μg/cm^2 thick was placed on a high-voltage electrode. The layer diameter was 13—14 mm. The distance to the grids was chosen to be 80 mm. This made it possible to reduce the pressure in the chamber (argon + 3% CO_2) to 220 mm Hg. To produce a uniform field in the gap between the grid and the high-voltage electrode, three rings were placed in each half of the chamber, and suitable potentials applied to them.

The fragment collimation was with the aid of two diaphragms 50 mm in diameter, located 7 mm off both sides of the high-voltage electrode. The fragment in the working volume of the chamber left noticeable ionization in the gap between the layer and the diaphragm. In order to draw out the ionization products to the working volume, the field in the gap was set to be about half as weak as in the main part of the chamber. This resulted in the necessary bending of the equipotential surfaces and did not allow the electrons to collect on the diaphragm. It was established experimentally that a change of 30% (accurate to 1%) in the gap field does not change the amplitude of the pulse produced by the fragment. As to the cases when one or both fragments came to rest on the diaphragm, the resultant pulses were either too small to be registered by the apparatus, or else the ratio was larger than 3.5:1 and far outside the range of the analyzed events.

3. Operation of the installation as a whole. The pulses from the ionization chamber were amplified and fed to a coincidence circuit, the signal from which controlled the operation of the entire apparatus. This pulse gated for 10 microseconds, with a delay of 1.2 sec, the input to the scaler, which memorized the information fed from the neutron detector. The resolution of the scaler was 0.6 μsec. In the case when the amplitude of the pulses from the ionization chamber exceeded a level corresponding to 35 MeV, the input of the analyzer serving for amplitude division was gated, and a pulse for reading the information recorded in the neutron-count channel was generated. A change in the threshold by ± 10 MeV did not change the fragment yield curve.

Information (amplitude ratio and neutron number) concerning each event was registered in a multichannel recorder. The ratio range from 2.2 to 1/2.2 was covered by 60 analyzer channels. The cases in which a light fragment was emitted in the detector direction were registered by the first 30 channels of the analyzer, and the cases when a heavy fragment traveled to the detector was registered by the remaining 30 channels. The channel width averaged over the mass range corresponded to 1.4 mass units.

Approximately five events per second were recorded under the operating conditions. Altogether 5.0×10^6 fissions were recorded during the course of measurements. In the mean, 8.3×10^{-2} neutrons and 7.0×10^{-2} background counts were registered for each event. The background measurement was made by repeated triggering of the neutron counting circuit (200 μsec after each fission).

CORRECTIONS

1. Corrections to mass distribution. A correction for the ionization defect had to be introduced into the amplitude ratio of the pulses produced in the chamber. It was assumed that the magnitude of the defect is linearly dependent on the mass M of the fragment: $\Delta E(M) = (4 + 0.019\,M)$ MeV[3]. The coefficients were chosen here to satisfy the most probable defects, 5.7 and 6.7 MeV for the light and heavy fragments respectively. The correction so calculated increased with increasing fission asymmetry, reaching ~ 5 mass units in the 2.2:1 ratio region. Inasmuch as the actual depend-

ence of the ionization defect on the mass is unknown, it is possible that the error accompanying the introduction of the corrections to the mass scale ranges in size from zero at symmetrical fission to apparently 1—1.5 mass units in the case of strongly asymmetrical fission.

In addition to the correction just discussed, a correction was made also for the neutron emission. As can be readily shown (in the case of isotropic evaporation of neutrons), the ratio of the fragment energies is equal to $(M_h/M_l)(1 - \nu_l/M_l)/(1 - \nu_h/M_h)$, where M—masses of the fragments prior to the emission of the neutrons, and ν—number of neutrons emitted by a fragment of given mass. In this correction we used the values of ν obtained in the present work.

2. Corrections to the neutron registration efficiency. As is well known, the possibility of measuring the number of neutrons emitted by a single fragment is based on the fact that the angular distribution of the neutrons is strongly elongated in the direction of flight of the fragment in the laboratory coordinate frame. The degree of elongation in the case of isotropic neutron evaporation relative to the fragment is determined by the velocity of the fragment and by the spectrum of the neutrons in the coordinate system in which the fragment is at rest. Therefore the probability that the neutrons will enter the detector is an implicit function of the fragment mass. The angular distribution of the neutrons is discussed in detail in a recent paper by Terrel[5], where an analytic expression is given for the integral of the angular distribution. The integrand depends only on two parameters: the kinetic energy per fragment nucleon, and the average energy of the evaporated neutrons. In our calculations we used Terrel's formula. The calculations were made with allowance for the following circumstances and assumptions.

a) The values of the kinetic energy used for the fragments of each mass were taken from [1], with the energy scatter disregarded.

b) The average neutron energy q in the fragment system was obtained on the basis of the evaporation model[5]: $q = 0.635(2\nu + 1)^{1/2}$.

c) It was assumed that the neutrons from the fragment approaching the detector are registered in a cone subtending polar angles from 0 to 30°, while that from the outgoing fragment—from 150 to 180°, the scatter in the fragment emission directions being disregarded.

d) It was assumed that the registration efficiency for neutrons entering the detector does not depend on the neutron energy.

3) The absolute values of ν were obtained by normalization to the known average number of neutrons per fission event, $\bar{\nu} = 2.44$.

It is difficult to estimate beforehand the errors due to the approximate character of the calculations. To determine the possible values of errors we obtained the total value of $\nu_l + \nu_h$ for the light and corresponding heavy fragments, and compared the data with the result of our paper[8], in which $\nu_h + \nu_l$ was determined directly in a near-4π geometry. A comparison has shown that in the mass interval from 88 to 149 (the region covered by the measurements in [8]) the error in question can amount to 5—7%.

RESULTS AND DISCUSSIONS

The measurement results are shown in Fig. 2. The abscissas show the fragment mass prior to neutron emission. The ratio of the yield at the maximum to the yield for symmetrical fission is 370 : 1.

The following remarks can be made concerning the results.

1) On the average the light fragments emit 30% more neutrons than the heavy ones: $\bar{\nu}_l/\bar{\nu}_h = 1.30$. The statistical error of this ratio is negligibly small. The uncertainties in the calculations of the efficiency make this result accurate to 5—7%.

2) In the mass region 128—131, the $\nu(M)$ curve has a deep minimum: such fragments emit on the average 0.25—0.3 neutrons. The position of the minimum is shifted somewhat relative to the masses 131—133 corresponding to the closed shells Z = 50 and N = 82.

3) A sharp increase in $\nu(M)$ takes place in the narrow mass range 105—110. Approximately 3.5 neutrons are emitted at the maximum where M = 110.

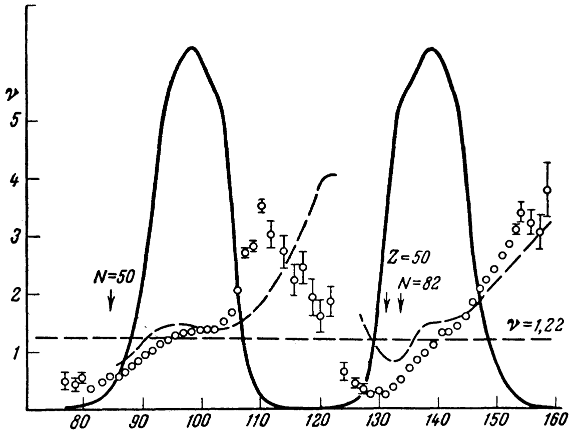

FIG. 2. Dependence of the number ν of neutrons emitted from the fragment on the fragment mass for U^{236}; solid curve—mass distribution of fragments; dashed curve—dependence ν (M) for Cf^{252} [7].

4) The number of emitted neutrons varies approximately linearly in the symmetrical fission region (masses 111—125).

5) In the region M = 97—103 $\nu(M)$ remains practically constant. The growth of $\nu(M)$ is slowed down also in the case of heavy fragments at M = 140—142.

6) Near the closed N = 50 shell, a change takes place in the slope of the $\nu(M)$ curve for light fragments with $M \leq 82$ and the complementary fragments with $M \geq 154$. This apparently confirms Terrel's hypothesis[5] regarding the role of the "magic" number 50 in the fission. It must be noted, however, that the measurement accuracy in this mass region is no longer high.

7) The dashed curve in Fig. 2 shows the variation of $\nu(M)$ obtained by Bowman et al[7] for the spontaneous fission of Cf^{252}. In the mass ranges 84—104 and 124—150, the curves for U^{236} and Cf^{252} are qualitatively of the same character and are close quantitatively almost everywhere. The main difference between the two curves is that their maxima lie at different mass values: at 110 in the case of U^{236} and at 120—122 in the case of Cf^{252}, so that the hypothesis that a universal $\nu(M)$ exists for all fissioning nuclei, in the form given by Terrel[5], must be discarded. The data show that in both cases the masses of the fragments that emit the maximum and minimum number of neutrons are quite close to the complementary ones. At low excitation energy the minimum for nuclei with atomic weight from 230 to ~256 lies apparently in the mass region 130, while the maximum shifts in position from mass ~105 to mass 126.

The experimental values of $\nu(M)$ were used to calculate the energies carried away by the neutrons from the fragments

$$E_x(M) = \nu(M)[B(M) + q(M)],$$

where B(M)—binding energy and q(M)—average kinetic energy of the evaporating neutron. The values of B(M) were taken from Milton's tables[10], and q(M) was obtained from the simple evaporation model without shell effects: $q(M) = 0.65\,(2\nu + 1)^{1/2}$. It was shown by direct measurements in[7] that this relation is well satisfied everywhere except the mass region 125—140, that is, near the closed shells. However, the difference is quite small from the point of view of interest to us here.

The results of the calculations are shown in Figs. 3a and b. In Fig. 3c is shown the total excitation energy carried away by the neutrons. As can be seen from the figure, the difference between the maximum and minimum excitation energies ranges from 18 to 22 MeV.

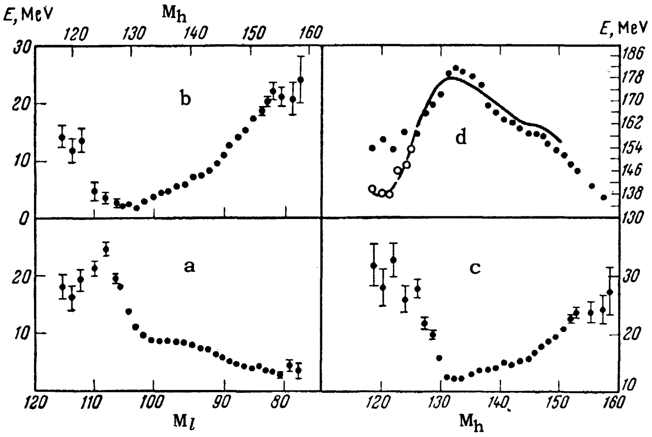

FIG. 3. Energies carried away from the fragments by the neutrons, and the total kinetic energy of the fragments: a—energy carried away from the light fragment, b—energy carried away from the heavy fragment, c—total energy carried away by the neutrons from both fragments, d—total kinetic energy of the fragments, calculated from the semiempirical formula and the results of the present work. Solid and dashed lines—results of [1].

Before we speak of the energy balance, we must discuss the reliability of the results obtained for symmetrical fission. Here (masses 114—122) a little more than 60% of the events are genuine, and the rest have entered this region from other mass regions owing to apparatus effects. The total value of $\nu_l + \nu_h$ was found to be ~4.5. In our earlier work we obtained for $\nu_l + \nu_h$ a value 3.6, and the number of genuine events was only 33%, in which connection we have advanced the hypothesis that actually 6.3 neutrons are emitted in symmetrical fission of U^{236}[8,9]. However, the increase in the number of genuine events, attained in the present work, shows that, within the limits of experimental error, the total number of neutrons and the calculated excitation energy in the mass region 112—124 does not depend on the mass. The appearance of such a plateau shows that the results obtained here are already quite close to the true ones. Apparently a further improvement in the experimental conditions will lead essentially only to some broadening of the plateau.

The black dots in Fig. 3d show the difference between the total energy release and the fragment excitation energy, which should appear in the form of fragment kinetic energy. In determining the correction to the energy carried away by the neutrons from the fragments, we added the energy of the gamma quanta and took account of the fact that the latter does not depend on the mass ratio and is equal to 8 MeV.

The total energy release was determined in accordance with the cited work by Milton[10]. As can be seen from the figure, the decrease in the energy thus obtained, occurring in the region of symmetrical fission, amounts to 24—28 MeV. The same figure shows the results of direct measurements of the kinetic energy by the fragment time of flight method[1], according to which the drop amounts to 38—40 MeV, that is, disagreed with our results by 10—16 MeV. The authors of [1] note that in their measurements the number of genuine events in the mass region 112—124 amounts to only one-third, and are therefore very cautious with respect to the numerical values obtained for symmetrical fission. It must be noted that the inclusion of events from other mass regions into the region of symmetrical fission is accompanied by a decrease in the kinetic energy. A particularly strong decrease occurs in time-of-flight measurements. Gibson et al.[2], who used the ionization method, obtained an energy drop of ~ 31 MeV, which is close to our value (in the figure from which this number is taken, the total kinetic energy is given as a function of the masses of the light and heavy fragments, and the curves in the region of the strongly asymmetrical fission are for some reason not identical).

The ranges of the fragments of symmetrical fission were measured by Niday[11] and by Alexander et al.[12]. The necessary range-energy ratio was obtained by extrapolating the data from the region of the most probable fission, where the measurement of the energy by the time-of-flight method is quite reliable. According to [12], the value of the drop amounts to 24—27 MeV, which agrees with our result.

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