The Average Number of Neutrons per Fission Event in the Photodisintegration of Uranium and Thorium

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The average number of neutrons ν from a single fission event has been measured in the photodisintegration of uranium and thorium with a mean nuclear excitation energy of 12 mev. For uranium, the value of ν is 6.2 ± 0.5, for thorium 14.2 ± 1.2. The measured quantities allowed evaluation of the relative fission probability in photodisintegration of uranium and thorium nuclei.

IN the laboratory of V. I. Veksler during 1951 - 1952, the average number of photoneutrons ν from a single fission event was measured in the photodisintegration of uranium and thorium by bremsstrahlung with maximum energy $E_{max} = 18.6$ mev (average energy of nuclear excitation ~ 12 mev). The work was done with a synchroton which gave 150 γ - pulses per second. The width of a single pulse was ~ 20 μ sec. To determine the values of ν, the absolute emission of photoneutrons and fission fragments was measured, using the same number of nuclei and the same radiation dosage.

The intensity of γ radiation was measured with a thin walled ionization chamber placed between the target and the sample (Fig. 1). The chamber was connected to an integrating circuit which acted as a dosimeter.

MEASUREMENT OF THE ABSOLUTE NUMBER OF NEUTRONS

Neutrons emitted from the sample during γ irradiation were slowed in the surrounding paraffin, and registered in the ionization chamber (KN - 14) filled with BF_3. Figure 1 shows the positions of

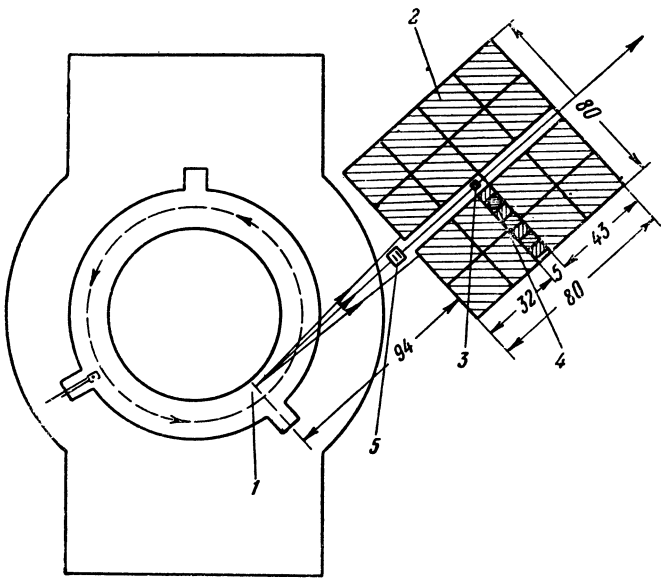

FIG. 1. Physical arrangement (dimension in cm) : *1* – internal synchroton target; *2* — paraffin block 70 cm high; *3* – sample; *4* – BF_3 ionization chamber; *5* – thin walled chamber with plane parallel electrodes for measurement of γ radiation dose (electrodes made of 50μ aluminum foil; windows in the shielding cover made of of 50 μ aluminum foil).

the paraffin block, sample, and ionization chamber. The paraffin block is 80 × 80cm, with a height of 70 cm. Samples in the form of 4cm diameter disks were placed in the center of the block. Incident γ– rays paseed through a round, 5 cm diameter, opening in the paraffin. The KN-14 chamber could be moved along a channel perpendicular to the γ - ray axis in order to measure the spatial distribution of neutron density.

Strong ionization occurred in the chamber during the passage of the γ - rays. Therefore,neutrons were registerd in the time interval between γ pulses, which followed each other every 6667 μsec. The final tube of the ionization chamber amplifier was gated after each γ pulse, passing neutron pulses into counting circuits, and closed again before the next γ pulse. By varying the beginning of the neutron counting time referred to the γ pulse, the dependence of the neutron count on time was measured. The neutron lifetime determined in this way was

$\tau_0 = 183 \pm 3\,\mu$ sec.

Counting of neutrons began after a delay of 20 μ sec, and ended after 2500 μ sec. Practically all neutrons were absorbed by the paraffin during this time interval. The correction for neutrons missed in the first 20 μsec after the γ pulse was 12%. Uncertainty in the delay time did not exceed 5 μsec, and the error in neutron count in a single series of measurements did not exceed $\sim$ 3%.

Calibration of the absolute emission of neutrons from irradiation of uranium and thorium samples was done with a Ra +Be source. The intensity of this source, measured by several methods, was $(4.69 \pm 0.27) \times 10^5$ neutrons per second.

Figures 2 and 3 show the spatial distribution of neutrons $\rho(r)r^2$ for the Ra + Be source,uranium and thorium, superimposed at the point r=10 cm, where the ionization chamber was located during the measurements. The points for $r > 35$ are obtained by exponential extrapolation of the curve ends. The area under the curve $\rho(r)r^2$ for the source is $k = 1.27 \pm 0.05$ times larger than the area under the uranium curve, and $k = 1.34 \pm 0.05$ times larger than the area under the thorium curve.

Due to the large neutron background accompanying the γ - ray bundle, it was necessary to work with large samples. Sample weight for uranium was 284 gm, and for thorium, 120 gm. Neutron background was respectively $\sim$ 10% and 20%.

In order to maintain the same neutron measuring conditions in the presence of the samples, which were strongly neutron absorbing, and in the absence of the samples during the background measurements, samples were placed in a cadmium box (wall thickness 0.5 mm). An identical empty box was placed at the sample position during background measurements. During calibration, the Ra +Be source was also placed inside such a box.

During passage through such thick samples as are necessary in this experiment, the γ - ray intensity decreases considerably, and the original energy distribution changes. The true neutron emission was obtained by extrapolating specific neutron emissions for various sample thicknesses to zero sample thickness. Both samples were made of four flat aluminum containers with an inner diameter of 4 cm, and top and bottom thickness 0.5 mm. For the uranium sample each container was filled with 71 gm of the powdered metal; for thorium four such containers were filled with pressed shavings (30 gm in each container). Neutron emission was measured when one, two, three and four containers were placed inside the cadmium box. For better extrapolation, neutron emission was also measured from sample thicknesses of one half that of a container. Fig. 4 shows the specific neutron emission for various sample thicknesses of the two materials. The specific emission obtained from the entire sample (4 containers) has been taken as unity. Specific neutron emission, extrapolated to zero sample thickness, is $1/\varkappa$, where $\varkappa = 0.59 \pm 0.02$ for uranium, and $\varkappa = 0.80 \pm 0.02$ for thorium.

Measurements of the number of neutrons from Ra +Be source in the presence of the samples showed that resonance absorption in the sample of neutrons passing through the cadmium, and fast neutron induced fission both effect the measured neutron emission by not more than one percent.

MEASUREMENT OF THE ABSOLUTE NUMBER OF PHOTOFISSION EVENTS

The absolute number of fission events was measured by observing fission fragments in an ionization chamber placed in the paraffin block instead of the sample.

To record fission fragments during the passage of γ quanta, when large ionization was produced, a differential chamber was used. The chamber had three electrodes: a central collecting, and two outside electrodes to which voltages of opposite polarities were applied. A thin layer of oxide (U_3O_8) or (ThO_2) with a diameter of 4 cm was deposited on the negative chamber electrode. Ionization impulses arising simultaneously in the identical halves of the chamber from γ quanta arrive at the amplifier grid with opposite sign, and cancel. It is thus possible to observe pulses from fission fragments. Pulses due to incomplete cancellation were several times smaller than those

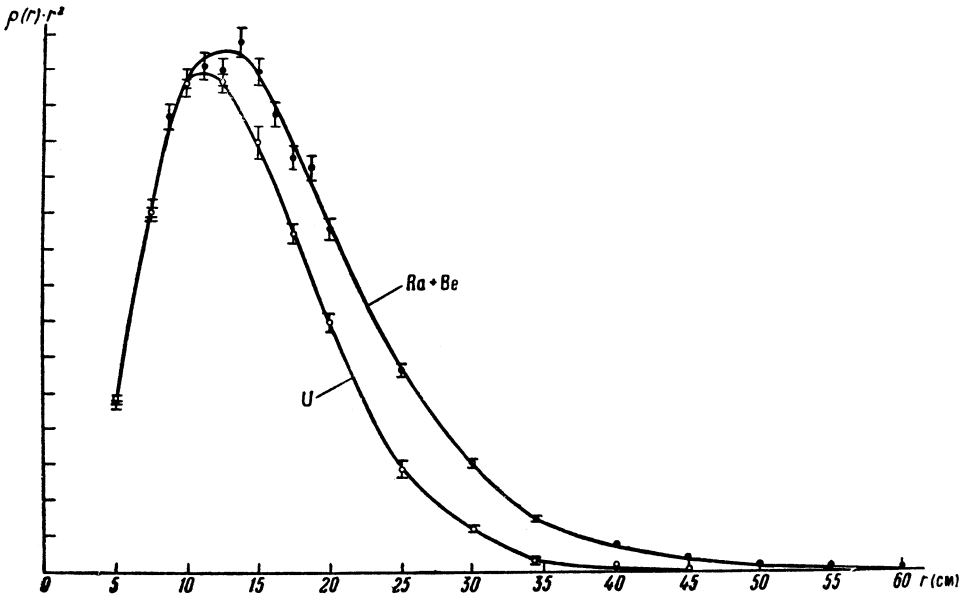

FIG. 2. Spatial distribution of neutrons from uranium.

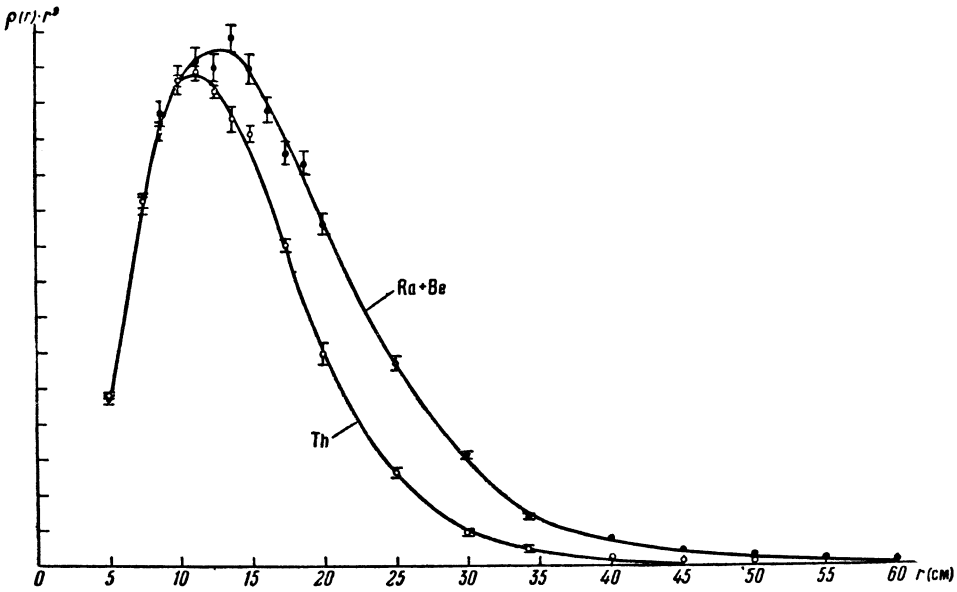

FIG. 3. Spatial distribution of neutrons from thorium.

due to α-particles emitted by the uranium and thorium layers. These pulses were cut off during the fission fragment count.

The chamber wae filled with chemically pure argon at a pressure of 1.5 atmospheres, and utilized the electron part of the pulse, which insured low noise and an absence of microphonic effects. The electrodes had 7 cm diameter in order that fragments emitted from the edge of the layer would not leave the confines of the chamber and be missed.

An electronic circuit counted pulses which were > 2.5 times the maximum α-particle pulse.

The oxide layers used in the measurements were prepared by baking water solutions of uranium nitrate and thorium in nitric acid. The deposition was on aluminum foil in the course of several hours at 500°.

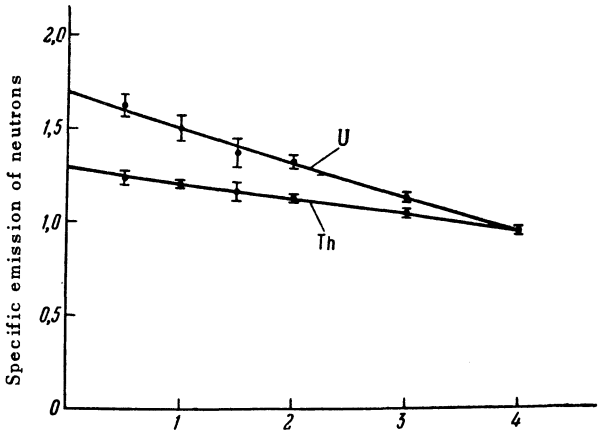

FIG. 4. Relative emission of photoneutrons during irradiation of various thickness targets. The abscissa is marked in thicknesses of 1, 2, 3, 4 containers. The thickness of each container with uranium was 5.65 gm/cm^2, and with thorium, 2.38 gm/cm^2.

The uranium measurements were carried out with a single layer whose weight was determined by the number of α-particles emitted. For the layer used, $w = 1.78 \pm 0.02$ mg, which corresponded to a thickness of 0.17 mg/cm^2 of the oxide. Thorium measurements were made with two layers: $w_1 = 1.38 \pm 0.04$ mg; $w_2 = 2.36 \pm 0.07$ mg. The weight of the thorium layers was determined from the amount of solution of known concentration which was deposited on the surface used in the measurements. The solution concentration was found by weighing the ThO_2 produced after slow evaporation and subsequent baking of a known volume of the solution. A correction to the fission fragment count was made for each layer. Since the areal density of a layer was of the order of several tenths mg/cm^2, the correction to the fission fragment count was not over several percent.

During γ irradiation of the layers, various fission events could have been caused by background neutrons or photoneutrons emitted from the ionization chamber walls. Measurement of fission fragments during the bombardment of the layers by neutrons from the Ra + Be source showed that the neutron induced fission events did not exceed 0.2% of the total number of registered fissions.

RESULTS

The average number of neutrons ν emitted per photofission event was obtained: for uranium 6.2 ± 0.5; for thorium 14.2 ± 1.2. The value of ν for thorium is the average of two values $\nu_1 = 14.1 \pm 1.1$ and $\nu_2 = 14.4 \pm 1.3$ which were obtained for the two thorium layers.

The measured values of ν allow evaluation of the photofission probability in uranium and thorium. During irradiation of these nuclei with γ-rays of $E_{max} = 18.6$ mev, four reactions involving photoneutrons are energetically possible:

$$(\gamma, f),\ (\gamma, n),\ (\gamma, nf) \quad \text{and} \quad (\gamma, 2n).$$

The following Table shows the various reaction thresholds[1]. The probability for photoproton emission for nuclei of $Z > 80$ is a few tenths of a percent. The probability for a (γ, γ') reaction should not exceed several percent[2].

Table

Reaction*	U^{238} (MeV)	Th^{232} (MeV)
(γ, f)	5.08 ± 0.15	$5,40 \pm 0,22$
(γ, n)	5.97 ± 0.10	$6.35 \pm 0,04$
$(\gamma, 2n)$	$11,4 \pm 0.2$	$11.85 \pm 0,2$
(γ, nf)	11.6 ± 0.3	12.15 ± 0.3

*Experimentally determined thresholds for (γ, n) and (γ, f) are given[1].

Thresholds of (γ, nf) and $(\gamma, 2n)$ are the sum of the experimentally determined first neutron binding energy, and the calculated second neutron binding energy and fission threshhold of U^{237} and Th^{231}.

[1] J. R. Huizenga, L. B. Magnusson, P. R. Fields, M. H. Studier and R. B. Duffield, Phys. Rev. **82**, 561 (1951); L. B. Magnusson, J. R. Huizenga, P. R. Fields, M. H. Studier, Phys. Rev. **84**, 166(1951)

[2] A. G. W. Cameron and L. Katz, Phys. Rev. **84**, 608 (1951)

Thus at the present excitation energies, photofission and photoneutron emission are essentially the only competing processes of uranium and thorium photodisintegration.

The fraction of nuclei α which fission as a result of (γ, f) and (γ, nf) reactions is $(\bar{\sigma}_{\gamma, f} + \bar{\sigma}_{\gamma, nf}) / \bar{\sigma}_{\gamma}$; the fraction of nuclei β which decay by the emission of one or two neutrons is $(\bar{\sigma}_{\gamma, n} + \bar{\sigma}_{\gamma, 2n}) / \bar{\sigma}_{\gamma}$ where $\bar{\sigma}_{\gamma}$ is the average absorption cross section for the effective quanta; this is equal to the sum of the average cross sections of all competing reactions $(\bar{\sigma}_{\gamma, n} + \bar{\sigma}_{\gamma, f} + \bar{\sigma}_{\gamma, nf} + \bar{\sigma}_{\gamma, 2n})$.

The measured quantity $\nu = \nu_0 + \bar{n}\,(\beta/\alpha)$, where ν_0 is the average number of neutrons emitted from the nucleus undergoing fission and $\bar{n}$ the average number of neutrons emitted in the photodisintegration of a nucleus through the reactions (γ, n) and $(\gamma, 2n)$.

In agreement with the γ quantum energy dependence of the photofission cross section obtained by I. V. Chuvilo, the average excitation energy of fissioning uranium and thorium nuclei irradiated by bremsstrahlung of $E_{max} = 18.6$ mev is ~ 12 mev. With such excitation the average number of neutrons accompanying fission is around three. From the known reaction yields of $(\gamma, 2n)$ for bismuth and tantalum[3], and also from calculations based on statistical theory, $\bar{n} \approx 1.2 - 1.4$. Substituting these values for ν_0 and $\bar{n}$ into the expression for ν, we obtain:

$$\left(\frac{\beta}{\alpha}\right)_U = \left(\frac{1-\alpha}{\alpha}\right)_U = \frac{3.2}{\bar{n}} \approx 2.7 \div 2.3,$$

$$\alpha_U \approx 0.27 \div 0.30;$$

$$\left(\frac{\beta}{\alpha}\right)_{Th} = \left(\frac{1-\alpha}{\alpha}\right)_{Th} = \frac{11.2}{\bar{n}} \approx 9.3 \div 8.0.$$

$$\alpha_{Th} \approx 0.10 \div 0.11.$$

The values obtained for α_U and α_{Th} give an upper limit to the average fission probability of an excited U^{238} or Th^{232} nucleus, since α includes fission events resulting from the (γ, nf) reaction, where the nucleus remaining after emission of the first neutron undergoes fission.

The above values show that during the absorption of 12 mev γ quanta, the fission probability of uranium is $\sim 1/4$ to $1/5$ and the fission probability of thorium is $\sim 1/10$. These agree with similar evaluations made by other authors[4].

The average absorption cross sections for γ quanta by uranium and thorium nuclei do not differ by more than a few percent[5], and therefore the ratio α_U / α_{Th} must be equal to the ratio of the photofission cross sections of these elements. The value obtained for the ratio $\alpha_U / \alpha_{Th} = 2.7$ is in good agreement with results given in other work[6].

There is only one measurement of ν in the literature, where a similar method was used to measure ν for uranium for $E_{max} = 23$ mev[7]. The value obtained, $\nu = 10.5 \pm 2$, contradicts all available data for the average photofission probability of uranium. No reports of measurements of ν for thorium have appeared in the literature.

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