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Upper Limit for the Time between Fission and the Emission of Neutrons*

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The Baker experiment to detect times of delay greater than $\sim 5\times 10^{-9}$ sec for the emission of fission neutrons has been performed by observing the change in fission neutron counting rate when fission fragments are allowed to fly away from a neutron counter as compared to the counting rate when the fragments are kept in the vicinity of the counter. The fission neutrons were detected by observing proton recoils in a thick-paraffin-lined proportional counter. The percent of neutrons delayed by a time of at least 8×10^{-9} sec was found to be (3.6 ± 2.8) percent.

I. INTRODUCTION

A VERY small number (about 0.8 percent) of the neutrons which accompany nuclear fission are delayed by times of the order of seconds.[1] These neutrons are associated with beta-processes in the fission products. It has been assumed that the rest of the neutrons (the "prompt" neutrons) are emitted within the short times characteristic of ordinary nuclear reactions ($<10^{-15}$ sec). Wilson[2] has shown that there is a strong correlation between the direction of a fission fragment and the direction of a fission neutron, indicating that a considerable number of the neutrons are emitted before the fragments are brought to rest. The purpose of the present experiment was to ascertain whether all of the "prompt" neutrons (the ones which are not delayed by at least many milliseconds) are emitted in less than 10^{-8} sec.

The method, originally proposed by C. P. Baker, uses the high speed of the fission fragments to measure the neutron emission time. A foil of U^{235} is wrapped around a counter sensitive only to fast neutrons and the whole immersed in a flux of thermal neutrons. The counter and foil are placed within a chamber that can either be evacuated or filled with a gas of high stopping power. The chamber wall is 3.46 cm away from the foil. Thus when the chamber is evacuated, most of the fission fragments (which have a velocity of $\sim 10^9$ cm/sec) can travel from the foil to the chamber wall in $\sim 5\times 10^{-9}$ sec. On the other hand, when the chamber is filled with the gas, the fragments are brought to rest very close to the counter. If there is a delay in neutron emission, as long as or longer than 5×10^{-9} sec, the neutrons emitted by those fragments which escape from the U^{235} will come from the outer wall of the chamber, far from the counter, when the chamber is evacuated, but from just outside the counter when the chamber is filled with gas. Because the solid angle subtended by the counter is much less at a point on the chamber wall than on the surface of the counter itself, one expects a difference in the neutron counting rates in the two cases if an appreciable number of neutrons are emitted 5×10^{-9} sec or more after fission occurs.

II. APPARATUS

The fast-neutron detector actually used in these experiments was a thick-paraffin-lined proportional counter, illustrated in Fig. 1. The counter cylinder was 17 in. long and 2 in. o.d. with a $\frac{1}{16}$-in. duralumin wall. This large counter was considered desirable because of its high counting rate, since good statistics are imperative in an experiment whose "background" is inevitably greater than 50 percent (because at least half of the fission fragments must remain in the foil). The axial collecting electrode was a Kovar wire 0.005 in. in diameter. The entire counter cylinder was lined with a cerecin-impregnated nickel gauze. The wires of the gauze were scraped free of paraffin on both sides after coating to insure good electrical contact with the counter wall on the one side and to insure an electrical field undistorted by charge accumulation on the other. Cleanliness was important in minimizing the boron contamination since boron gives alpha-particles in a slow-neutron flux which are difficult to distinguish from recoil protons. In addition to boron, any nitrogen present gives an undesirable background owing to protons from the slow-neutron (n,p) reaction. For this reason the counter was filled with spectroscopic argon. With a pressure of 15 cm, the counter was found to operate well at about 820 v.

The counter was placed on the axis of a steel cylinder $4\frac{7}{8}$ in. i.d., $\frac{1}{32}$-in. wall, which could be evacuated or filled

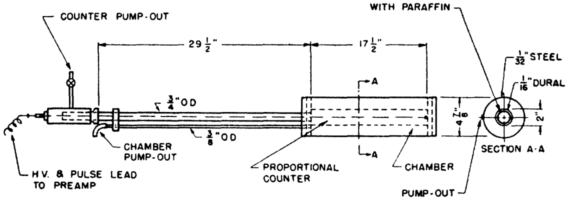

FIG. 1. The recoil counter and gas chamber.

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* This document is based on work performed in 1943 at Los Alamos. A more complete account can be found in the declassified document LADC-252.

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[1] See, for example, de Hoffmann, Feld, and Stein, Phys. Rev. 74, 1330 (1948); Hughes, Dabbs, Cahn, and Hall, Phys. Rev. **73**, 111 (1948).

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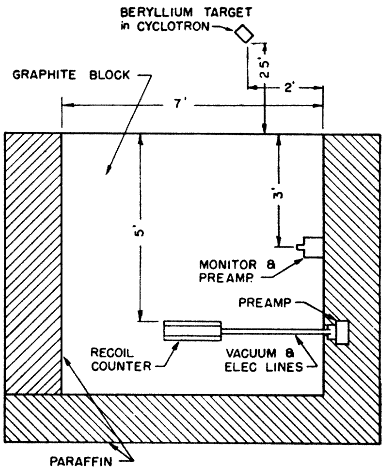

FIG. 2. Plan view of the graphite block, showing location of the counter and monitor.

with commercial propane to a pressure of one atmosphere (see Fig. 1).

To minimize slow-neutron absorbing materials in and near the counter its wire was used both as the collecting and high voltage electrode, and electrical connections were made by a long coaxial lead through the counter filling tube. By using a glass-insulated central wire of sufficient size, spurious electrical effects were avoided. This lead was made long enough so that the pulse preamplifier was far away and had negligible effect on the slow-neutron flux near the counter.

In operation, the counter was placed across the axis of a graphite block 7×5.7 ft on the front face and 7 ft long (see Fig. 2). Fast neutrons from a cyclotron with internal beryllium target were incident on the front face. At a depth of 5 ft, where the counter was located, these neutrons had been slowed down to give a thermal flux $(nv)_{\text{th}}=10^5$ per cm^2 per sec. The ratio of this flux to the flux of neutrons above 100 kev was 25,000. It is important in this experiment to have the number of fast neutrons already present in the block as small as possible compared to the number of fast neutrons produced by fission, which one is attempting to measure. Under the conditions described, background (from all sources) was less than half the total counting rate.

The circuits used in counting consisted of a regulated high voltage supply for the counter, a one-stage preamplifier and cathode follower mounted just outside the graphite block, a stabilized amplifier of the Waddell type, pulse selector, and scale of 32. The pulse selector was a differential discriminator which selected only pulses between two adjustable limits of pulse height. The upper limit was kept as low as seemed consistent with a good counting rate in order to eliminate as many of the boron alpha-particles as possible.

The response of the counter was calibrated by bombardment with monochromatic neutrons from the Li(p,n) source of the Los Alamos electrostatic generator. The purpose of the calibration was to determine whether or not the counter responded selectively to neutrons of certain energies and thus gave preference to some of the fission neutrons while ignoring others. The response was found to be satisfactory for our purposes. The sensitivity had a threshold at about 300 kev, and remained approximately constant on going to higher energies. One Mev was the maximum neutron energy obtained during this calibration. The presence of a plateau beginning at 700 kev means that the counter responds impartially to nearly all the fission neutrons since few have lower energies than 700 kev. The sensitivity of the counter in counts per incident neutron was found from this same calibration to be 9×10^{-5}.

The U^{235} being investigated was a sample of enriched material containing 15.7 percent U^{235} and 84.3 percent U^{238}. It was electroplated onto the outside of a platinum foil 16×6 in. which fitted around the outside of the counter. The average weight of metal deposited (in the form of the fluoride) was about 1 mg of UF_4 per cm^2. Although this deposit was not thin to fission fragments, but rather about one-fourth of their range, it nevertheless permitted most of the fragments to escape with a large part of their initial velocity retained. The disadvantage caused by the inability of all fragments to escape with full velocity was offset by the large number of neutrons produced which gave an improved ratio of fission neutrons counted to background.

For quantitative interpretation of the experiment, it was necessary to know the fraction, E, of fission fragments which escape from the foil. (E would be one-half if the foil were perfectly thin.) The method of determining E and of measuring the uniformity of the foil is described in the Appendix. The value found for the escape fraction was $E=0.435$.

III. PROCEDURE

To compensate for variations in cyclotron output, it was necessary to monitor the thermal-neutron flux incident on the counter. For this purpose a fission ionization chamber employing a semithick foil of unseparated U was placed in the graphite block at a distance of 3 ft from the front face, 3 ft from the bottom, and 1 ft from one side. Any change in intensity and distribution of neutrons incident on the face of the block would therefore be expected to have the same effect on the monitor and on the number of fissions in our apparatus.

The significant quantity for the experiment is therefore the number of recoil counts per unit monitor count. This will be referred to as the counting rate.

In order to learn how sensitive the counting rate is to the point of origin of fission neutrons we observed the counting rate with the U^{235} foil in its normal position, surrounding the counter, then moved the U^{235} foil to the inner surface of the outer cylinder and observed the decrease in counting rate which this caused. These measurements were made with the stopping gas in the chamber. To eliminate any error which might arise from the change in slow-neutron flux with distance from the front face of the graphite block, the foil was placed symmetrically with respect to a plane perpendicular to the axis of the block. It is evident that this measurement, which we have called the "in-out measurement" would correspond approximately to the change in counting rate which would be expected if somehow *all* the fission fragments could escape from the foil, and all the fission neutrons were delayed by at least the time required for the fragments to reach the wall. Of course not more than half the fragments can escape at best, and their arrangement upon striking the wall will be more spread out than it is possible to make the foil, but the result of the experiment expressed in terms of this measurement is, to a first approximation, independent of the *calculation* of the change in counting efficiency as the source of neutrons moves from the surface of the counter to the outer wall of the chamber. This is important because it seems likely that considerations other than the change in solid angle subtended by the counter enter into this efficiency change, making an exact calculation difficult.

Knowing the fraction of fragments that escaped to the walls, we have now a measure of the change in counting rate to be expected from the delayed neutrons when the foil was on the outside of the counter and the stopping gas was removed from the surrounding chamber. The experiment then consisted of repeatedly comparing the counting rate with gas in the chamber (and fragments therefore all confined to the neighborhood of the foil) to the counting rate with the chamber evacuated (and some fraction, less than one-half, of the fragments therefore going to the walls).

If the stopping gas has appreciable absorption for slow neutrons a correction must be made, since the slow-neutron flux on the U^{235} and hence the number of fissions per unit monitor count would then be greater when the gas was removed, and the decrease in counting rate resulting from delayed neutrons might be masked. Propane, having eight hydrogen atoms per molecule, has a small but appreciable probability for slow-neutron capture, and the "gas effect" correction was therefore measured quite carefully and accurately, by covering the U^{235} foil with a foil of Al thick enough to stop the fission fragments, and then comparing the counting rate with and without the gas. A reduction of (0.7 ± 0.2) percent of the total counting rate was observed. About half of this can be attributed to absorption of slow neutrons, according to a simple diffusion calculation, and the rest may reasonably be attributed to slowing of residual fast neutrons.

IV. RESULTS

Table I summarizes the results of this experiment. The first column gives the average for each run of the "in-out" ratio, $I=100\ (\mathrm{C.R.}_{\mathrm{in}}-\mathrm{C.R.}_{\mathrm{out}})/\mathrm{C.R.}_{\mathrm{in}}$. $\mathrm{C.R.}_{\mathrm{in}}$ is the counting rate with the U^{235} foil in its normal position, $\mathrm{C.R.}_{\mathrm{out}}$ the rate with the foil against the outer wall of the chamber. This quantity was measured three times during Run I, twice during Runs II and III, and once during Run IV. It is a criterion of the sensitivity of the experiment and depends, in part, on the background counting rate. Its fluctuations arise from changes in the background, and the improvement between Runs II and III was made by reducing the fast-neutron background. The graphite block was lengthened by adding 12 in. of graphite to its front face; and the hole which contained the recoil counter was changed from a $5\frac{1}{4}$-in. cylinder to an 8×8-in. square channel, in the center of which the counter was supported. The latter change was made to reduce back-scattering into the counter of fast neutrons originating in the U^{235} since such scattering would have a tendency to decrease the change in counting efficiency for delayed neutrons.

The quantity of interest for our experiment is the difference between the counting rate with propane in the outer chamber and the counting rate with outer chamber evacuated. This quantity was converted to percent by multiplication by 100/(C.R. with propane) and designated by P. Each run consisted of about 10 pairs of measurements, a value of P being found for each pair. The average of these values of P for each run is tabulated in the second colum of Table I. The standard deviation of P_{av}, calculated in the usual way from the counting statistics, is given as σ_{calc}. The many values of P for each run permit a determination of the observed value of the standard deviation of P_{av},

$$\sigma_{\mathrm{obs}}=\{\sum_{i=1}^{n}\Delta_i^2/n(n-1)\}^{\frac{1}{2}}.$$

This quantity is given, for each run, in the fourth column of Table I. It will be seen that the internal consistency is satisfactory.

TABLE I. Summary of the measurements on delayed neutrons.

Run no.	I_{av}	P_{av}	σ_{calc}	σ_{obs}
I	28.2%	0.45%	0.46%	0.85%
II	24.8%	−0.16%	0.39%	0.44%
III	35.3%	−0.58%	0.58%	0.62%
IV	35.9%	−0.54%	0.51%	0.38%

Table II gives the results of two runs on the gas absorption effect. The results and errors were obtained in the same way as were those of the delayed neutron

TABLE II. Gas absorption. Foil on counter, covered with 0.001-in. aluminum.

Run no.	Percent difference	σ_{obs}	σ_{calc}
I	0.79%	0.30%	0.28%
II	0.62%	0.32%	0.25%
Average	0.71%	0.22%	

TABLE III. Percentage of delayed neutrons.

Run no.	P, corrected for gas abs.	σ	100f = percent delayed $>8\times10^{-9}$ sec	σ
I	1.15%	0.88%	12.2%	9.3%
II	0.55%	0.49%	6.6%	5.8%
III	0.13%	0.66%	1.1%	5.5%
IV	0.17%	0.55%	1.4%	4.5%
	Average percent delayed 3.6±2.8 percent.			

experiments. The values of P after adding the gas absorption correction and combining errors are given in Table III.

V. DISCUSSION

To a first approximation there is a very simple relation between the in-out measurement, I (defined as the percentage decrease in counting rate when the U^{235} foil is moved from the surface of the counter out to the wall of the chamber, the latter being filled with propane); the fraction E of fission fragments which escape from the foil; the observed counting rate reduction P; and the fraction f of neutrons delayed by at least the time necessary for all the fragments to reach the wall of the chamber. EI is just the counting rate reduction expected if *all* the neutrons were delayed, and f is therefore given by $f=P/EI$. Actually, since the effective length of the counter is about 40 cm, and the average velocity of the fragments about 0.9×10^{9} cm/sec, the time for the fragments which take the longest path to reach the wall is 5×10^{-8} sec, which is unnecessarily long. Also, a correction must be made for the finite range of the fragments in the stopping gas. Some knowledge of the efficiency of the counter as a function of the distance of the neutron source from its axis is therefore necessary.

The details of these rather tedious calculations are given in the original paper[3] and will not be repeated here. The results can be expressed to a good approximation by writing the fraction delayed longer than a time Δt after fission as $f=(P/EI)K(\Delta t)$. For times Δt long enough so that all the fission fragments have come to rest, $K(\Delta t)$ is of course constant, and is equal to 1.3 (it differs from unity mainly because of the finite range of fission fragments in propane, about 1.3 cm). For shorter times, $K(\Delta t)$ involves the spacial distribution of the flying fission fragments.[4]

Figure 3 is a graph of $K(\Delta t)$ against $\log(\Delta t)$. From this graph it is seen that the experiment has very nearly reached its maximum sensitivity, $K=1.3$, at $\Delta t=8\times10^{-9}$ sec. Thus, one can say that $f=1.3\times P/EI$ is the fraction of fission neutrons delayed by at least 8×10^{-9} sec. Table III gives the corrected values of P and corresponding values of f for the four runs. The weighted mean is (3.6±2.8) percent delayed.

This result would include most of the 0.8 percent of fission neutrons which are delayed by long times, although their energies are somewhat lower than those of the normal fission neutrons. There remains (2.8±2.8) percent delayed. The most reasonable interpretation is that this corresponds to zero: within an accuracy of three percent, and excluding the well-known delayed neutrons, none of the neutrons from fission are emitted at times greater than 8×10^{-9} sec after the fission process.

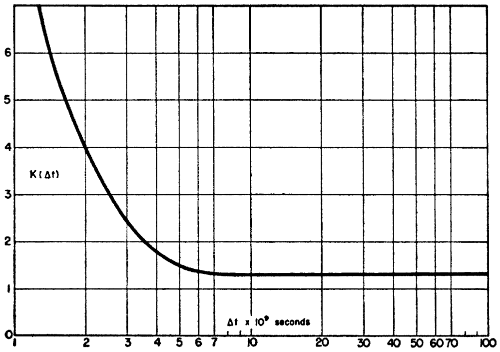

FIG. 3. The function $K(\Delta t)$. $1/K$ is a measure of the sensitivity of the experiment to neutrons emitted at a time Δt after fission.

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APPENDIX

At the conclusion of the experiment, the fraction of fission fragments which escape from the foil was measured by cutting two 2×2-in. samples from representative parts of the foil and comparing the number of fission fragments of energy >6 Mev from them with the number from a thin foil of known weight and the same isotopic constitution. The average E obtained in this way was 0.41. We correct for the fragments of <6 Mev as follows: Let the thickness of the UF_4 deposit be $\beta\leqslant\rho$, where ρ is the range of fragments in the UF_4. Then the fraction of fragments which escape after traversing a thickness less than l is given by the

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[3] Declassified document LADC 252. It is shown there that the probability for a neutron to make a count is approximately proportional to $1/r$, where r is the distance from the neutron source to the counter axis. This law was checked by calculating the value of the in-out ratio, I. The value expected from geometrical considerations was 53 percent. A background measurement (made with the foil removed) showed that 54 percent of the counting ratio with foil in "in" position was due to fission neutrons. One would therefore expect the in-out ratio to be $0.54\times53=28.6$ percent. The measured value at this time was 28 percent.

[4] This calculation included the distribution in velocity of the fission fragments as they emerge from the foil, and therefore involved the range-velocity relation given in the Appendix. It was simplified by assuming that all delayed neutrons were emitted after the same time Δt. An estimate shows that the corresponding mean life for an exponential decay law would be larger than Δt, but by less than a factor of two.

expression

$$1-\beta/2l \text{ when } l \geqslant \beta; \quad l/2\beta \text{ when } l \leqslant \beta. \tag{1}$$

In particular, the total fraction that escape; i.e., the fraction that have traversed any thickness of material up to ρ is

$$1-(\beta/2\rho). \tag{2}$$

We may use the above formulas together with a range-velocity relationship to find both the total fraction of fragments that escape and the thickness of the foil relative to the range; i.e., β/ρ.

A range-velocity relation which agrees with experiment well enough for these purposes is $(v/v_0)=(1-l/\rho)$. From this equation we calculate the value of l/ρ corresponding to 6 Mev, taking an average initial energy of the fragments to be 81 Mev. From Eq. (1) and the measured escape fraction we find $\beta/\rho=0.26$. Inserting this value of β/ρ in Eq. (2) we find that the total fraction of fragments which escape from the foil is $E=0.435$.

The uniformity of the UF_4 deposit was measured by cutting 24 one-inch squares from the foil and alpha-counting them. The standard deviation in thickness of the foil was 18 percent. We have assigned an uncertainty of this magnitude to E in computing the error of our final result.

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Distribution of Slow Neutrons in Free Atmosphere up to 100,000 Feet*†

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Two identical proportional counters filled with boron trifluoride of 96 percent B^{10} have been sent aloft up to an altitude of 102,000 ft at a geomagnetic latitude of 51° 46′ N by means of free balloons. One counter was shielded with 0.030 in. of cadmium, and the other enclosed in tin of the same thickness for the compensation of possible effects caused by "stars" produced in the cadmium shield. The difference in the counts of the two counters is due only to slow neutrons ($E \leqslant 0.4$ ev). The counts and the pressure and temperature were radioed down by an FM telemetering system. Up to about 20 cm of Hg the counts increase exponentially with altitude according to an absorption depth $\lambda=156$ g/cm², in agreement with previous measurements, and roughly with the increase of "stars" in the atmosphere. The counts of both counters as well as their difference show a maximum at high altitude, as expected theoretically. The maximum for the cadmium difference counts appears at about 8.5-cm Hg pressure and drops down sharply to about one-fourth of its maximum value at 1 cm Hg. The counter sensitivity was calibrated against a standard neutron source and the absolute number of slow neutrons absorbed per gram and second in the atmosphere is computed and compared with the number of protons produced at the same altitudes.

I. INTRODUCTION

A CONSIDERABLE number of measurements have been made on the neutron intensity in cosmic radiation by various investigators in this field.[1–9] The essential results thus far show an exponential increase of the neutron intensity as a function of altitude. Since neutrons in the cosmic radiation cannot be considered as primary particles because of their short lifetime, they must be produced in the atmosphere. Thus one can expect that there exists a maximum in neutron intensity distribution as a function of altitude. The position of the maximum has been calculated by Fluegge[10] and recently by Bagge and Fincke[11] on a theoretical basis by considering the absorption, scattering, and diffusion of neutrons in the atmosphere. This maximum is expected to exist at about 10 cm Hg, assuming that the atmospheric neutrons are originally produced in the processes of nuclear disruptions or "stars." However, measurements in the past[7] show a continuous increase in the neutron intensity up to 2 cm Hg. This discrepancy between theory and experiment[7] can probably be attributed to spurious counts obtained at high altitudes because of corona discharge at the high voltage terminals of the proportional counter. As will be described later, a completely pressurized system for the high voltage supply and the counters was employed[12] in the present experiment to eliminate such possible corona effects.

The object of the present experiment is not only to obtain the intensity distribution of the slow neutrons ($E \leqslant 0.4$ ev) in the atmosphere, but also to attempt to measure their absolute intensities at various altitudes.

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