A Search for Short Period Activities from U^{235} Fission Products*

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A search for gamma-ray activities with period 1–100 msec from thermal neutron fission of U^{235} gave negative results. 0.43±0.03 sec was the shortest observed. The period of B^{12} was found to be 27±3 msec. The cyclotron beam was pulsed to supply 1–10 msec neutron bursts. Coincidence scintillation detectors were employed.

INTRODUCTION

THE shortest period of radioactivity arising from U^{235} fission that has been published is a delayed neutron period of 0.05 sec; the next shortest neutron period is 0.43 sec.[1] It is to be expected that delayed neutrons follow a β- or γ-ray transition, but none such has been reported with half-lives shorter than about 1 sec.[2] Isomeric γ-ray transitions have been observed with periods ranging from 10^{-9} sec to months or years. The very short periods have been observed using electronic time delay techniques.[3] However, there is a conspicuous absence of periods in the millisecond region, although it might be assumed to be due to experimental difficulties. Unpublished studies of fission activities of 3- to 7-millisecond period made at Los Alamos[4] in 1943–1945 are contradictory, and it seems possible that the observations were the result of instrumental limitations. If β-ray activities of such periods exist in the medium weight elements resulting from fission, it would have considerable significance in the interpretation of β-ray theory. This paper reports a search for β-ray and γ-ray activities in the range between 1 and 1000 milliseconds. No periods were found shorter than the known 0.43-sec period corresponding to a delayed neutron activity.

EXPERIMENTAL ARRANGEMENTS

The emergent 11-Mev deuteron beam from the cyclotron was used to provide a pulsed source of neutrons. The beam was passed between electrostatic deflecting plates, through a focusing and analyzing magnet, and through a water tank shielding wall where it could strike a Be target at the end of an evacuated spout. A potential was applied to the deflecting plates which directed the beam off the target, then it was shorted out by an electronic switch for a time interval adjustable between 1 and 10 milliseconds, allowing a pulse of deuterons to strike the target. Neutrons from the Be target were slowed in a paraffin moderator and produced fissions in a U^{235} foil target surrounding a scintillation crystal, which was viewed by two RCA 5819 photomultiplier tubes. Gamma-radiation from the target was attenuated in a Pb cylinder 8 in. long between the target and the crystal. The experimental arrangement is shown in Fig. 1. Pulses from the two tubes were amplified and put through a discriminator coincidence circuit. Coincidence pulses were shaped and applied to the deflection plates of a cathode-ray oscillograph. The trace, to which millisecond time marks were also applied, was photographed by a moving film camera. Data consisted of plots of the number of pulses in each time interval against time.

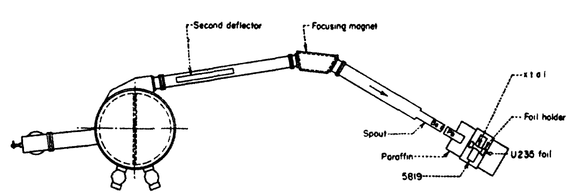

FIG. 1. Arrangement for pulsing and focusing the cyclotron emergent beam and for observing fission activities.

The electronic control circuits for pulsing the cyclotron and operating the camera were activated by a manual switch when the cyclotron beam was tuned to maximum, and performed a sequence of operations as follows:

(a) the camera film drive motor was started and allowed a 0.2-sec run to bring it up to speed;
(b) timing pulses of 1 to 10 millisecond spacing were applied to an electronic switch to turn off and on the electrostatic deflecting field;
(c) a coincident 1 to 10 millisecond square pulse was applied to the fifth dynodes of the photomultipliers to defocus them and prevent circuits from overloading during irradiation;
(d) the first timing pulse started a 100 kilocycle self-excited oscillator with frequency dividers to give 1 and 10 millisecond time marker pulses applied to the cathode-ray tube deflection plates;
(e) the second timing pulse turned off the cyclotron ion source voltage and the cyclotron radio frequency supply voltage;
(f) the camera motor was turned off 100 milliseconds (or more) after the second timing pulse.

Output pulses from the coincidence amplifier were too short (1 μsec) to give photographically observable marks on the film. A pulse-shaping circuit was developed to stretch the pulses to 5 μsec and provide a triangular shape. Amplitude controls for pulse height and timing signals were provided. Films were counted in a projec-

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[1] Hughes, Dabbs, Cahn, and Hall, Phys. Rev. 73, 111 (1948).
[2] Plutonium Project, Revs. Modern Phys. 18, 513 (1946).
[3] S. DeBenedetti and F. K. McGowan, Phys. Rev. 74, 728 (1948).
[4] LAMS-255, LA-253, LA-253A (unpublished).

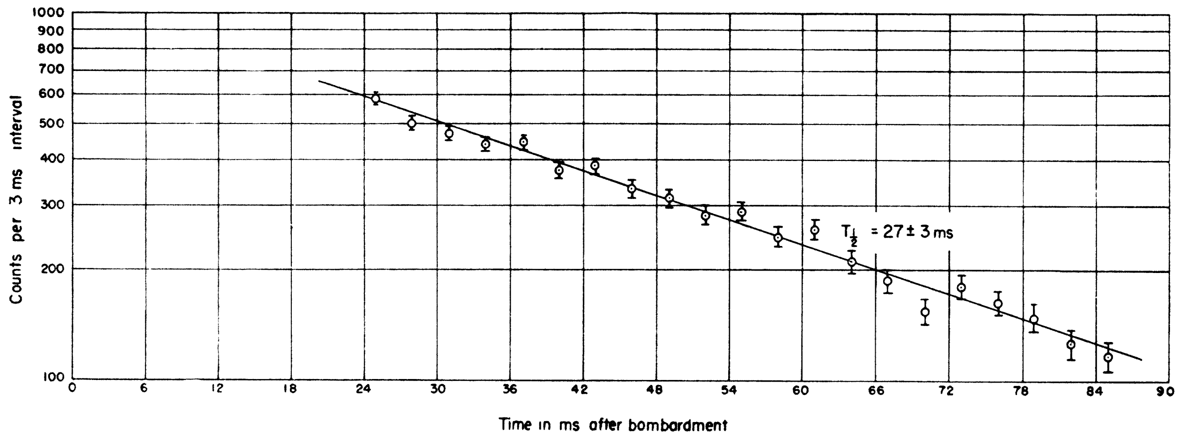

FIG. 2. Decay of B^{12} activity used to check performance of the instruments.

tion microscope. Pulse width, oscilloscope spot size, and photographic resolution limited countable intensities to about 20 per millisecond. Sufficient duplicate runs were made and the counts were added to obtain statistically significant data.

A ballistic galvanometer connected to the insulated Be target measured the total deuteron current pulse during the 1- to 10-millisecond bombardment. The galvanometer was calibrated with standard capacitances. A typical 5-millisecond pulse would record about 5 milli-microcoulombs, indicating an average deuteron beam of 1 microampere on the target.

Amplifiers, discriminator coincidence circuits, scalers, and power supplies were standard laboratory equipment developed by others at Los Alamos. The camera was a General Radio Type 651AE with rebuilt motor drive to give a speed of 100 milliseconds per foot of film. Films used were Eastman Photoflure and Linagraph Pan.

The fission sample was a foil of U^{235}, 0.010 in. thick, 2 in. wide, and 9.5 in. long, bent into a cylindrical shape around a 3-in. diameter crystal holder so as to offer a large solid angle to the crystal for fission-product γ-rays. The 10-mil thickness was chosen because it is approximately one radiation length for 1-Mev γ-rays. Three foils were available, for alternate use in case fission activity was built up in the one in use to give undesirably large background intensities. All three foils could be used simultaneously to obtain maximum intensity of fission activity if required.

The crystal was a cylinder 2 in. in diameter and 1 in. thick, with faces machined to a concave shape to fit the ends of the 5819 photomultiplier tubes. It was cut from a crystalline block of naphthalene having a concentration of 5 percent anthracene. One crystal holder had 0.62 in. of Al between the U^{235} sample and the crystal to absorb β-rays. Another holder was a frame with no absorber to observe both β-rays and γ-rays from the U^{235}. A third crystal holder of Pb 0.62 in. thick and with a Cd foil adjacent to the crystal was used to observe Cd γ-rays from delayed neutrons via the capture process.

The crystal holder and the two photomultiplier tubes were mounted in a light-tight cylinder located in a block of paraffin used as a neutron moderator. A cylindrical mu-metal shield around the cylinder shielded the photomultiplier tubes from the small stray field due to the cyclotron and focus magnets.

The "static" background arising from radioactivity in the uranium was observed by camera runs in which no beam was generated. It was usually small, of the order of 0.1 per millisecond, and could be subtracted directly from runs made with the beam. Background counts with the beam on, arising from causes other than uranium fission, were observed by replacing the U^{235} foil with a Pb foil having the same γ-ray absorption in alternate runs. These were due to activities induced in the surrounding materials and other causes. The "dynamic" background was assumed to be proportional to beam pulse intensity, and so was subtracted in proportion to the intensity as observed by the ballistic galvanometer in the several runs.

Preliminary runs were made to check the operation of the instruments, the sharpness of the pulse and recovery of the amplifiers after an irradiation pulse. In one test the deuteron beam was brought out into air through a thin window, and through a narrow slit onto the moving film of the camera. A direct photograph of the beam pulse was obtained. No tail was found following the pulse beyond the first 0.1 millisecond after termination of bombardment that was more than 0.1 percent the intensity of the main pulse. This proved that the pulsing was sufficiently sharp to carry observations down to one millisecond.

To check the effectiveness of the defocusing pulse on the photomultipliers, runs were made with a strong Co^{60} source placed adjacent to the crystal, pulsing the circuits but with no cyclotron beam. Counting rates were found to be identical in the intervals just before and after the pulse, and no counts were observed during the pulse.

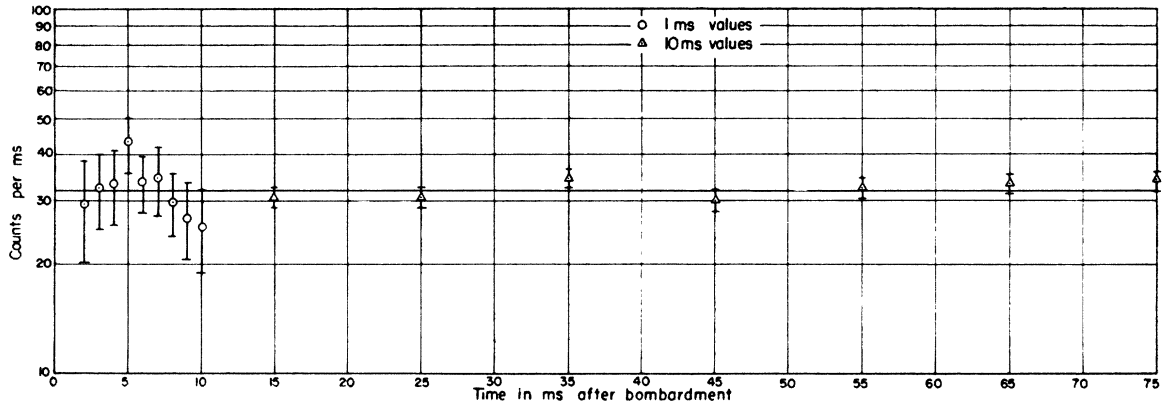

FIG. 3. Total β- and γ-ray activity from U^{235} fission products as a function of time after bombardment showing no millisecond decay periods.

In the preliminary runs a discontinuity in counting rates was observed at about 90 milliseconds, due to the heavy load of the cyclotron radio frequency power supply going off the line. This long time delay was caused by the slow action of several heavy relays and contactors in the circuit. As a consequence, no data are reported in the interval between 75 and 100 milliseconds. The continuous character of the data on both sides of this interval justify the conclusion that no significant information was missed.

As an over-all check on the performance of the system, the decay of B^{12} (produced by $B^{11}(d,p)B^{12}$) was measured, using a 10-millisecond pulse. Absorption between the boron and crystal was sufficiently small to permit the most energetic electrons (end point $\sim$13 Mev) to enter the crystal. Irradiations were made at high and low intensity levels. The high intensity run could not be counted much below 25 milliseconds. Background was negligible. The results are shown in Fig. 2. The observed period of 27 ± 3 milliseconds may be compared with another recent measurement of 27 ± 2 milliseconds.[5] The low intensity run had poorer statistics but could be satisfied by a 27-millisecond period down to the time of termination of bombardment.

RESULTS

No difference was observed, except for intensity, in preliminary runs using 1, 2, 5, and 10 millisecond bombardment pulses. The final runs to accumulate statistically significant data were made with 5 or 10 msec pulses. Runs were also made with 1, 2 or 3 U^{235} foils and found to be identical except for intensity. Three layers were used in the final runs. Five sets of

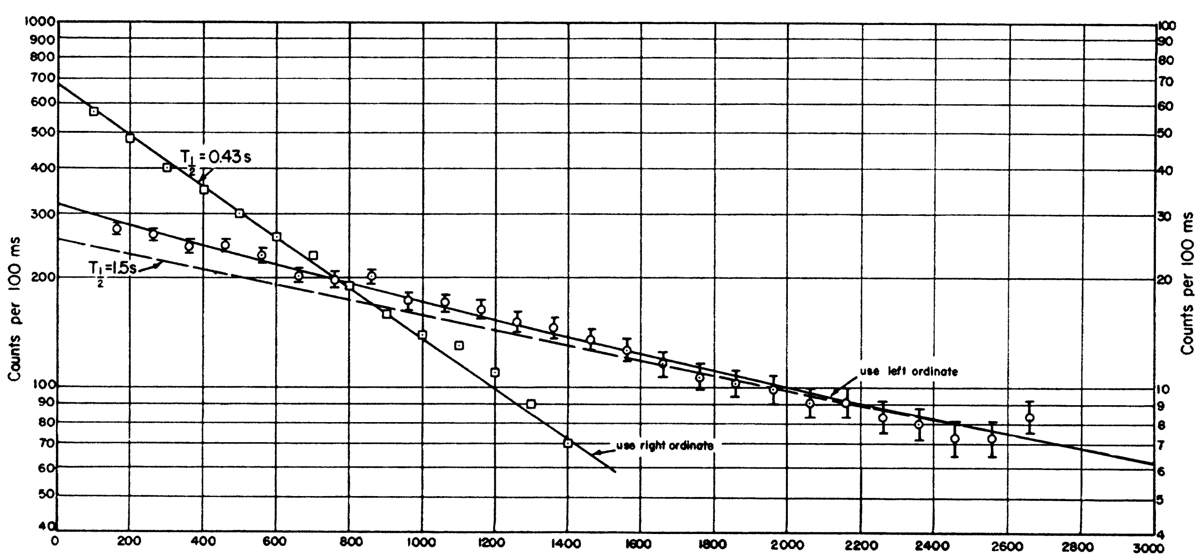

FIG. 4. γ-ray activity for U^{235} fission products as a function of time after bombardment from 100 to 2700 milliseconds. The observed period of about 1 sec can be resolved into 1.52- and 0.43-sec periods if the 1.52-sec period is assumed.

[5] J. V. Jelly and E. B. Paul, Proc. Cambridge Phil. Soc. 44, 133 (1948).

fission measurement runs were made: (1) 5 msec pulse —no absorber—75 msec counting interval; (2) 10 msec pulse—Al absorber—75 msec counting interval; (3) 10 msec pulse—Al absorber—100-2700 msec counting interval; (4) 10 msec pulse—Pb absorber—no Cd—75 msec counting interval; (5) 10 msec pulse—Pb absorber —Cd—75 msec counting interval. Three variations were made in each set except (4) and (5): (a) U in—beam pulsed; (b) U in—no beam; (c) Pb foil in—beam pulsed. To correct the data, the results from (b) which represent chiefly the natural radioactive background of the uranium, were directly subtracted from (a). (c) represents the background arising from substances other than uranium and is assumed proportional to the beam pulse intensity. (a), (b), and (c) were normalized to unit galvanometer deflection and the difference taken. Corrected data from each run in a given set were added together and plotted on a semi-log scale as a function of time.

The results of set (1), which represent the total β-ray and γ-ray intensity, are plotted in Fig. 3. The first point is not plotted, since it represented an interval shorter than 1 msec due to persistence of the defocusing pulse. There was also a strong decay in the first half of the interval in both the U and Pb background runs, not due to fission, which would make large errors in the difference count. The source of this apparent decay period of a few tenths millisecond is not known, but may be associated with the finite slowing down time of neutrons in the moderator. Figure 3 shows clearly that there are no decay periods in the millisecond range, and this is illustrated by passing a horizontal line through the data. It is of course decaying with a period long compared to the interval displayed, which is determined in the longer duration runs. The vertical lines in this and other figures represent statistical errors in the total count for each time interval.

Identical results were obtained with set (2), which measured γ-ray activity only. The lower counting rates made the data less accurate, so the results are not plotted separately.

An estimate of the shortest period occurring in fission comes from the data of set (3), in which the camera was run for the maximum length of film available (100 ft), about 2700 milliseconds. The resulting decay curve shown in Fig. 4 indicates a period of the order of 1 sec. The curve does not extend for a long enough time to analyze it easily for components. However, if it is assumed that the predominant component at 2700 msec is the known 1.52-sec period[1] in the delayed neutron family, and this period is subtracted from the data, a residual activity of 0.43 ± 0.03 sec is revealed as the shortest period in fission activities. This value is in exact agreement, possibly fortuitous, with the shorter delayed neutron period of 0.43 ± 0.05 sec.[1] The analysis showing two periods is also indicated in Fig. 4.

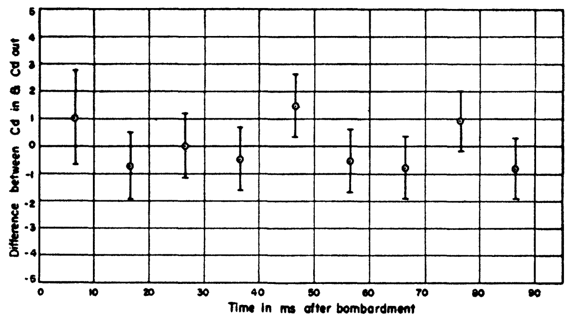

FIG. 5. Difference between runs with Cd in and Cd out plotted against time in an attempt to observe the 50-msec delayed neutron period. Results are negative.

Sets (4) and (5) with and without a Cd radiator surrounding the crystal and with a thick Pb absorber for U β- and γ-rays were made in the hope of accentuating the delayed neutron activities, and to search for a 50-msec period. No measurable difference was observed with and without Cd, the results were consistent with the results from sets (1) and (2). The difference between counts with Cd in and Cd out is plotted in Fig. 5, and shows no evidence for a 50-msec period.

CONCLUSIONS

From the results of this study it seems unlikely that any short period β- or γ-rays (1 to 100 milliseconds) arise from the products of slow neutron fission of U^{235}. Specifically, no periods were observed in the 3- to 7-msec region or in the 50-msec region where previous experiments had indicated the existence of activities. An activity of 5 percent the intensity of the observed 1-sec period could have been detected. The shortest period observed was of the order of 1 sec, which is resolvable into two periods of 1.52 and 0.43 sec if the 1.52-sec period is assumed. No evidence has been obtained for delayed neutron activities, but the β- and γ-ray results throw considerable doubt on the validity of the reported 50-msec neutron period.

These negative results are in agreement with present β-ray theory, which requires excitation energies above the neutron binding energy for medium weight elements in order to have a half-life of less than 0.5 sec. The absence of gamma-ray activity in the millisecond range from isomeric transitions is also significant, since fission products cover such a wide spread of elements.

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