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SHORT PERIOD γ-RAYS FROM U^{235} FISSION PRODUCTS

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PHYSICS - FISSION

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Abstract

A search for γ-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.

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SHORT PERIOD γ-RAYS FROM U^{235} FISSION PRODUCTS

Introduction

The purpose of the experiment is to study the γ and β radio-activities from fission in the time interval 1 to 75 milliseconds. This region has been investigated previously but results are contradictory. The pertinent earlier results can be briefly summarized as follows:

1) CC-1128 "Decay of Gross Fission Product γ-Ray Activity and Energy after Short Bombardments" - Katcoff et al: Decay curves for γ's after cyclotron bombardments of 5 sec, 10 sec, 1 min, 5 min. Results were extrapolated to 0.1 sec; the shortest half-life observed was $\simeq$ 1.0 sec.

2) LAMS-255 "F-Division Progress Report, May 1945": Slug of U^{235} * shot through water boiler pile was measured at times > .040 sec.

3) LA-253 "Short Period γ's from Fission of 25" - Moon and de Hoffman: Used "dragon" critical assembly to observe γ's at times from 0.001 to 10 sec; observed a period 3-4 milliseconds at the extreme lower limit of time resolution.

4) LA-253A "Short Period γ's from Fission of 25 and 49" - Halpern

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* L.D.P. King informs us that their data yield half-lives of 0.44 sec and 2.7 sec with indication of additional activity $\ll$ 0.44 sec.

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and Moon: A comparison of LA-253 with LAMS-255 and some new data from the water boiler shows discrepancies. The results of LA-253 were probably due to (n, γ) processes caused by neutrons released from the critical assembly after it had supposedly become sub-critical.

5) LA-252 "Delayed Neutrons from 25 after Short Irradiations" - de Hoffman: "Dragon" shots showed five delayed neutron periods > 0.5 sec and one short period of 6.3 milliseconds.

6) Phys. Rev. 73, 111 (1948) "Delayed Neutrons from Fission of U^{235}" - Hughes et al: "Chopper" used with pile showed five delayed periods > 0.4 sec and one short period of 50 milliseconds.

7) CC-3032 "Summary of Rate of Decay of Fission Products" - Wigner and Way: Statistical analysis of number and average energy of β- and γ-rays with theoretical arguments showing improbability of periods shorter than 0.5 sec from fission products.

8) LA-170 "Fission γ-Rays" - Deutsch and Rotblat: A search for metastable states with delayed coincidence techniques was negative; periods longer than 100 microseconds could have been detected; upper limit not estimated.

The results listed above are mutually contradictory. The 3-4 msec γ-ray period is almost certainly the result of an error in interpretation. The 6.3 and 50 msec delayed neutron periods are in both cases found near the limit of time resolution of the apparatus and may be due to instrumental effects. Delayed neutrons should be

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preceded by β-rays of the same period, although not necessarily by γ-rays. Periods less than 0.5 sec cannot be explained by present β-ray theories. Isomeric γ-rays with periods as long as msec are possible but extremely unlikely. Furthermore, experiments able to detect even shorter lived isomeric states in fission fragments have given negative results. A necessary conclusion is that the evidence is insufficient to prove the existence of decay periods in the msec range.

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Experimental Arrangement

The present equipment was planned to utilize short pulses of beam from the cyclotron to produce fission, and scintillation detectors to observe fission activities in the time interval 1 to 75 msec after bombardment. The emergent 11 Mev deuteron beam from the cyclotron passed between horizontal plates of an electrostatic deflector system, was focused and analyzed by a magnet, passed through the water-tank shielding wall and struck an insulated Be target at the end of an evacuated spout. Gamma radiation from the target was attenuated in a cylinder of lead 8 in. long between the Be and the crystal. 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 as illustrated in Fig. 1. The crystal was one in. thick and two in. in diameter. It was machined from a crystalline block of naphthalene containing 5% anthracene. Pulses from the two tubes were amplified and put through a discriminator coincidence circuit; only large pulses coincident in time were counted. Output pulses were applied to the deflection plates of a cathode-ray tube, and the trace was photographed by a moving-film camera. Time marks applied to the film allowed the pulses in each msec to be counted. Data consisted of plots of the number of pulses in each time interval versus time.

The electronic control circuits for pulsing the cyclotron

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and operating the camera were designed by C. Sewell and constructed in the Electronics shop. A timing circuit was arranged to be triggered manually or operated automatically on a 100 sec cycle. On each triggering impulse the camera film drive motor was started and allowed a 0.2 sec run to bring it up to speed. At this time a pair of timing pulses of 1 to 10 msec spacing were initiated. These pulses turned off and on the electrostatic deflecting field during this interval, allowing the deuteron beam to pass through the focusing magnet and strike the Be target. While the Be was being irradiated, a rectangular defocussing pulse was applied to the fifth dynodes of the photomultipliers and kept them defocussed until 200 μsec after bombardment. The second of these timing pulses also turned off the cyclotron ion source voltage (estimated as $< 50\,\mu$sec) and the cyclotron radio frequency supply voltage (delayed by contactor for about 90 msec). Finally the camera motor was turned off 100 msec or more after the second timing pulse.

Precisely at the beginning of the deflector pulse a 100 KC self-excited oscillator started. The self-excited oscillator was standardized by observing beats against a 100 KC crystal oscillator in a cathode-ray tube. One and 10 msec timing marks were generated by dividing the signal from the gated oscillator. These marks accurately terminated the deflector pulse at an integral number of msec.

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 γ-ray pulses to 5

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μsec and provide a triangular form. Amplitude controls for pulse height and timing signals were provided.

A ballistic galvanometer connected to the insulated Be target read the total deuteron current pulse during the 1 to 10 msec bombardment. Calibration with standard capacitances gave a value of 1.864 cm deflection/milli-micro-coulomb.

Voltage supply for the 5819 photomultipliers came from a thermostatically regulated battery box. Voltages were set to give equal tube amplifications of 10^5. Two channel amplifiers with constant amplification of 2×10^5 were supplied by B. Watt (Dwg. No. 4Y26230), as were also the discriminator coincidence circuit (Dwg. No. 4Y26229) and three scalers (Dwg. No. 4Y26140), with their power supplies. The scope had a Model 200 power supply (Dwg. No. 535A). The camera was a General Radio (type No. 651AE) with re-built motor drive to give the desired speed. The speed was sufficient to give about 100 msec/ft of film. Films used were Eastman Photoflure and Linagraph Pan.

Preliminary observation of counting rates with a test source of Co^{60} showed a drop in counting efficiency when the cyclotron magnet and focus magnet were turned on. This was interpreted as due to a small magnetic field at the location of the 5819 photomultipliers, and was measured to be about 3 gauss. A mu-metal shield to surround the tubes was installed and was found to give adequate shielding.

The fission sample was a foil of U^{235}, 10 mils thick, 2 in.

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wide and 9.5 in. long, bent into a cylindrical shape around a 3 in. dia. crystal holder in order to offer a large solid angle to the fission product γ-rays reaching the crystal. The thickness of 10 mils was chosen because it is about one radiation length for 1 Mev γ-rays. Three foils* were available, for alternate use in case fission activity was built up to give undesirably large background intensities. All three foils could be used to obtain maximum intensity of fission activity if required. One sample holder was of solid Al, 0.62 in. thick, to absorb β-rays; another holder was a frame with no absorber to observe β-rays plus γ-rays from the U^{235}. A third crystal holder of Pb was planned for use with a Cd foil around the crystal to observe γ-rays from delayed neutrons via the capture process.

Background arising from causes other than the uranium were measured by replacing the uranium with a lead foil of about the same γ-ray absorption.

Biases were always set to count Co^{60} γ-rays.

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* Sample numbers 8123, 8130, 8128.

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Results

Since activities had been reported in the 1 to 10 msec region, we desired to prove unambiguously that the beam was indeed sharply pulsed because a small tail could lead to spurious activity in this region. The Be target was replaced by an Al window in order to bring the beam out into the air. The lens of the General Radio camera was replaced by a narrow slit whence the camera was mounted to allow the deuterons to pass through the slit and impinge on the moving film. The camera was connected to the timing circuits described earlier and direct photography of the beam pulse was performed. No tail was found. If any did exist, beyond the first tenth msec after termination of bombardment it was at least a thousand times less intense than the main pulse.

The defocusing pulse in the 5819's prevented activities generated during the Be irradiation from paralyzing subsequent electronic circuits in the counting system. In order to demonstrate that pulsing did not affect sensitivity immediately after the beam pulse, a strong Co^{60} source was placed adjacent to the crystal. The circuits were then pulsed in the usual manner with no cyclotron beam. The counting rate on the film prior to the pulse was also observed immediately after the pulse.

As an overall check on the performance of the system, the decay of B^{12} produced by the bombardment of a thick B target* with

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* Sintered boron supplied by Wm. Wellborn.

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deuterons was measured using a 10 msec pulse. The absorption between target and crystal was sufficiently low to permit γ-rays, if any, and the more energetic β-rays (end-point 13.4 Mev)[9] to strike the

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9 W. Hornyak and T. Lauritsen, Phys. Rev. 77, 160 (1950).

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crystal. Irradiations at low and high levels were performed. The high level irradiation films were counted from 25 to 85 msec after bombardment and added together to produce the curve shown in Fig. 2. The films could not be counted before 25 msec because of overlapping of the pulses. Background as determined by a run with no beam was insignificant. The half-life, 27 ± 3 msec, deduced from the curve may be compared with another recent measurement of 27 ± 2 msec.[10] The low-

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10 J. V. Jelly and E. B. Paul, Proc. Cam. Phil. Soc. 44, 133 (1948).

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level run had very poor statistics but was satisfied by 27 msec half-life up to the termination of bombardment.

Four types of fission measurements were performed using three layers of uranium foil as a lamina. (1) 5 msec bombardment - no Al absorber - 75 msec counting interval. (2) 10 msec bombardment-with and without Cd - 75 msec counting interval. (3) 10 msec bombardment - thick Al absorber - 100 to 2700 msec counting interval. (4) 10 msec bombardment - thick Al absorber - 75 msec counting interval. At about 90 msec the heavy RF load of the cyclotron went off the line

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and caused a small discontinuity in amplifier gain, thus prohibiting measurements in this region. Three types of measurements were made in each set except (2). (A) Uranium in - beam pulsed. (B) Uranium in - no beam. (C) Pb in - beam pulsed. (B) was the natural radioactive background of the uranium and was directly subtracted from (A). (C) represents the background arising from substances outside uranium. It was assumed proportional to the beam pulse (galvanometer deflection). (A) -(B) and (C) were normalized to 1 cm galvanometer deflection and the difference taken. In the case of run (1) counting was done in one msec intervals in the range 1 to 10 msec and in 10 msec intervals thereafter. The first msec interval could not be counted because the defocussing pulse did not open it for 0.2 msec after bombardment and also due to a strong decay in the interval which appeared in both uranium and Pb background runs. Fig. 3 illustrates the results of run (1) when both β- and γ-rays were counted. The errors shown in this and other figures represent statistical errors of counting. No 3 to 4 msec or 50 msec periods are obvious. The activity is practically constant over the interval and we illustrate this by passing a horizontal line through the data (it is of course decaying with a period long compared to the interval displayed). In the hope of accentuating the detection of neutrons, run (2) was made over the interval 1 to 75 msec. The addition of Cd next to the crystal caused no measurable difference in the counting rate within the limits of error. Fig. 4 illustrates this

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result.

To estimate the shortest period occuring, we allowed the camera to run for the maximum length of film (100 ft); about 2700 msec. The resulting decay curves shown in Fig. 5 indicate a decay of the order of 1 sec found in earlier work. The curve does not extend for a long enough time to easily analyze it for components. However, if it is assumed that the predominant component at 2700 msec is of 1.5 sec half-life, the earlier part of the curve yields another activity of 0.43 ± 0.03 sec. We have estimated this error on the basis of extreme fits of the primary curve to the experimental points. The value of 1.5 sec was suggested by the known value of 1.52 ± 0.05[6] half-life in the delayed neutron family. Our value of 0.43 ± 0.03 may be compared with the shortest half-life of the delayed neutron which is well-known, namely 0.43 ± 0.05.[6] The agreement would appear fortuitous. An analysis of the first 75 msec of the long run was made. The statistics were poor since only a few films were shot. However, the results were consistent with the conclusions of run (1). From this work it seems unlikely that short-period γ-rays (2 to 75 msec) arise from the fragments generated by thermal neutron fission of 25 with energy above 1 Mev. Since the instrument was adjusted to count Co^{60} γ-rays, it is not known how far below one Mev the instrument was responsive. On the basis of a crude analysis of the long decay curve the shortest period observed is 0.43 sec. Had more time been available, the experiment

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would have been done with electronic storage of counts rather than film. Such a system would have considerably enhanced the precision and rate of accumulating data.

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Acknowledgements

We are indebted to B. Watt for advice and some equipment. H. Fishbine, W. Johnstone and C. Sewell designed and produced most of the electronic equipment. The cyclotron crew gave generous cooperation. A. Armstrong and her assistants gave invaluable aid in counting films.

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Captions for Figures

Fig. 1 - Schematic arrangement of apparatus.

Fig. 2 - Decay of B^{12}.

Fig. 3 - Decay of $\gamma + \beta$ activities arising from slow neutron fission of "25".

Fig. 4 - Cd difference plot to emphasize neutron counting.

Fig. 5 - Decay of fission products from 100-2700 msec with Al absorber in.

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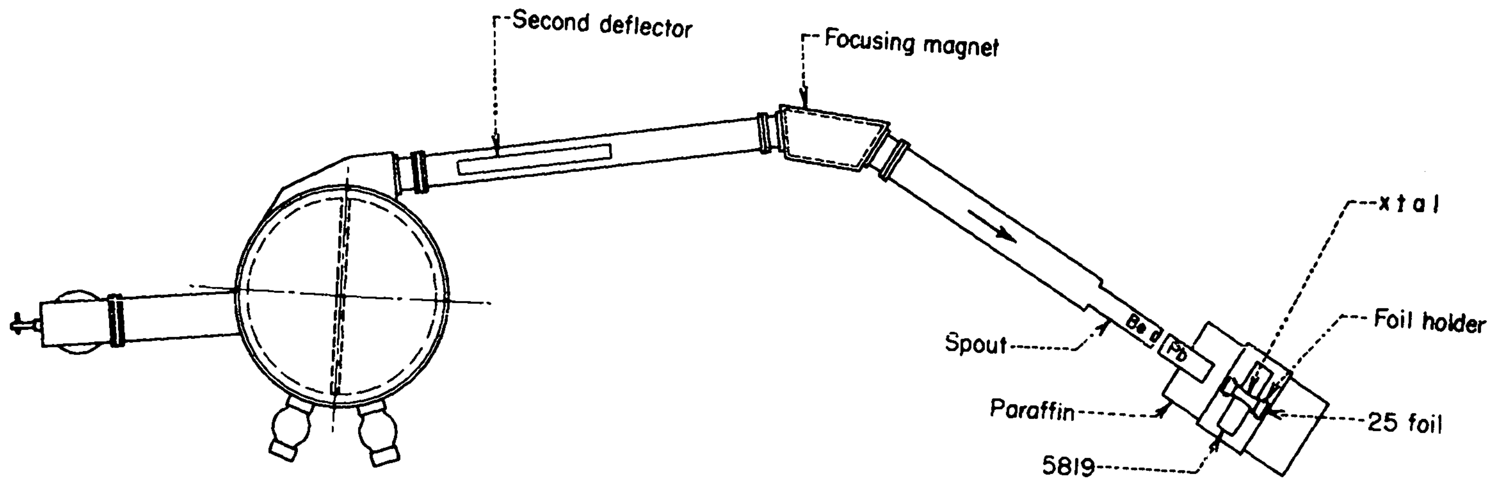

Fig. 1

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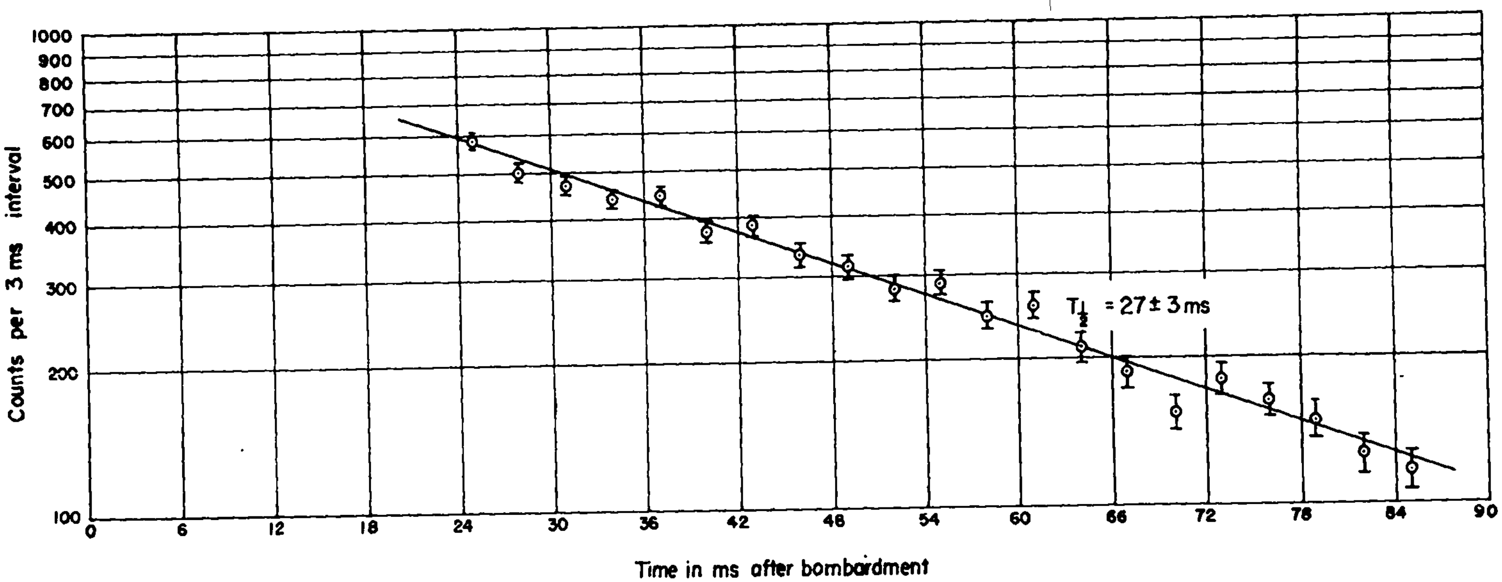

Fig. 2

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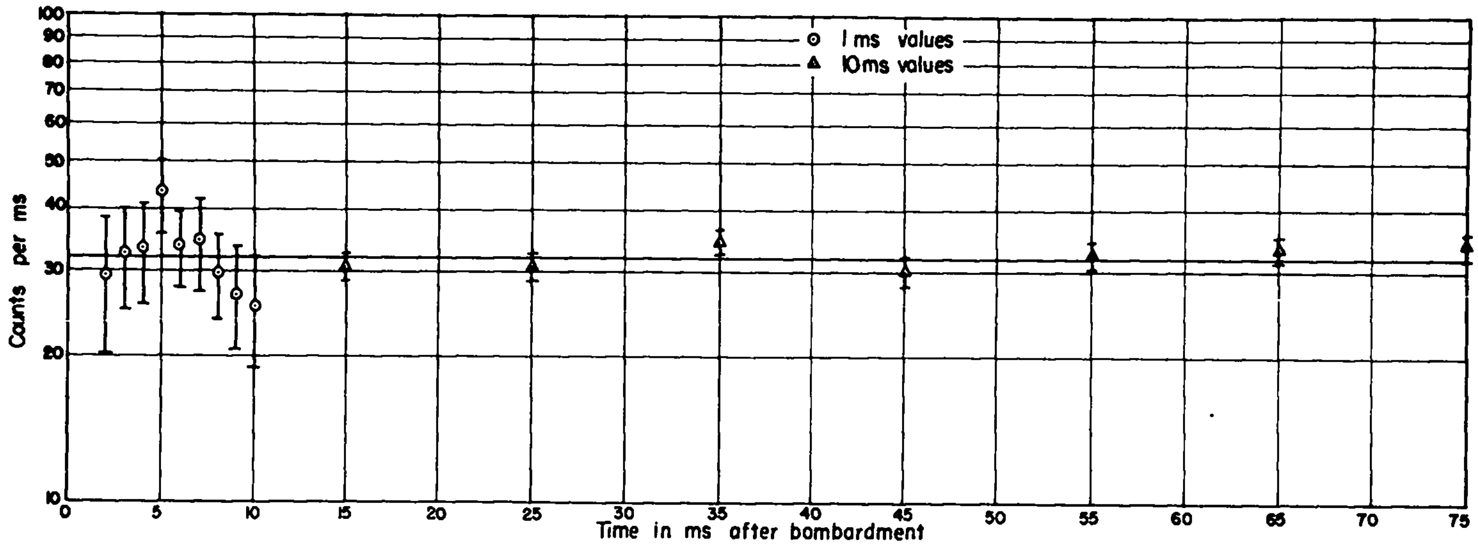

Fig. 3

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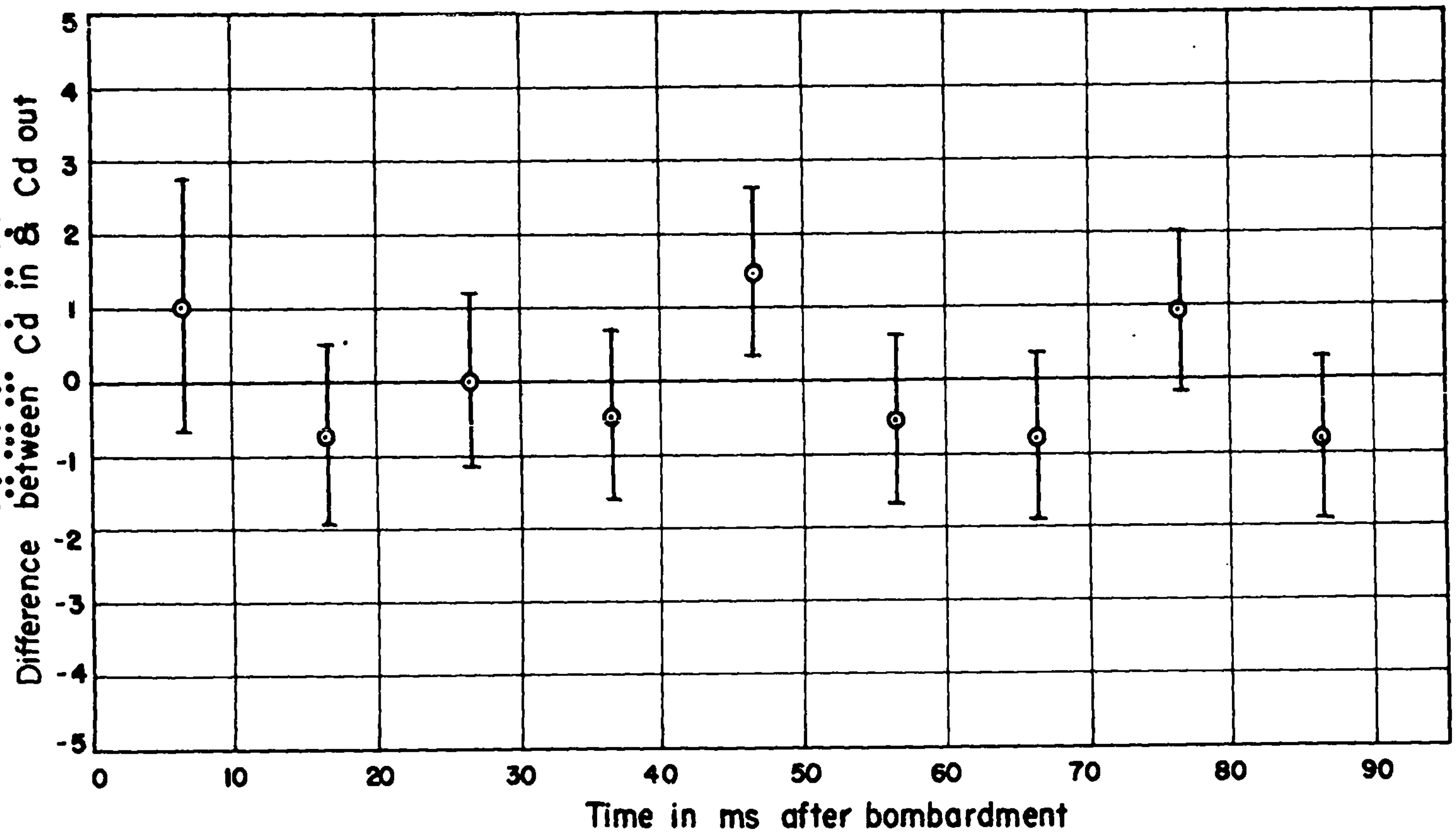
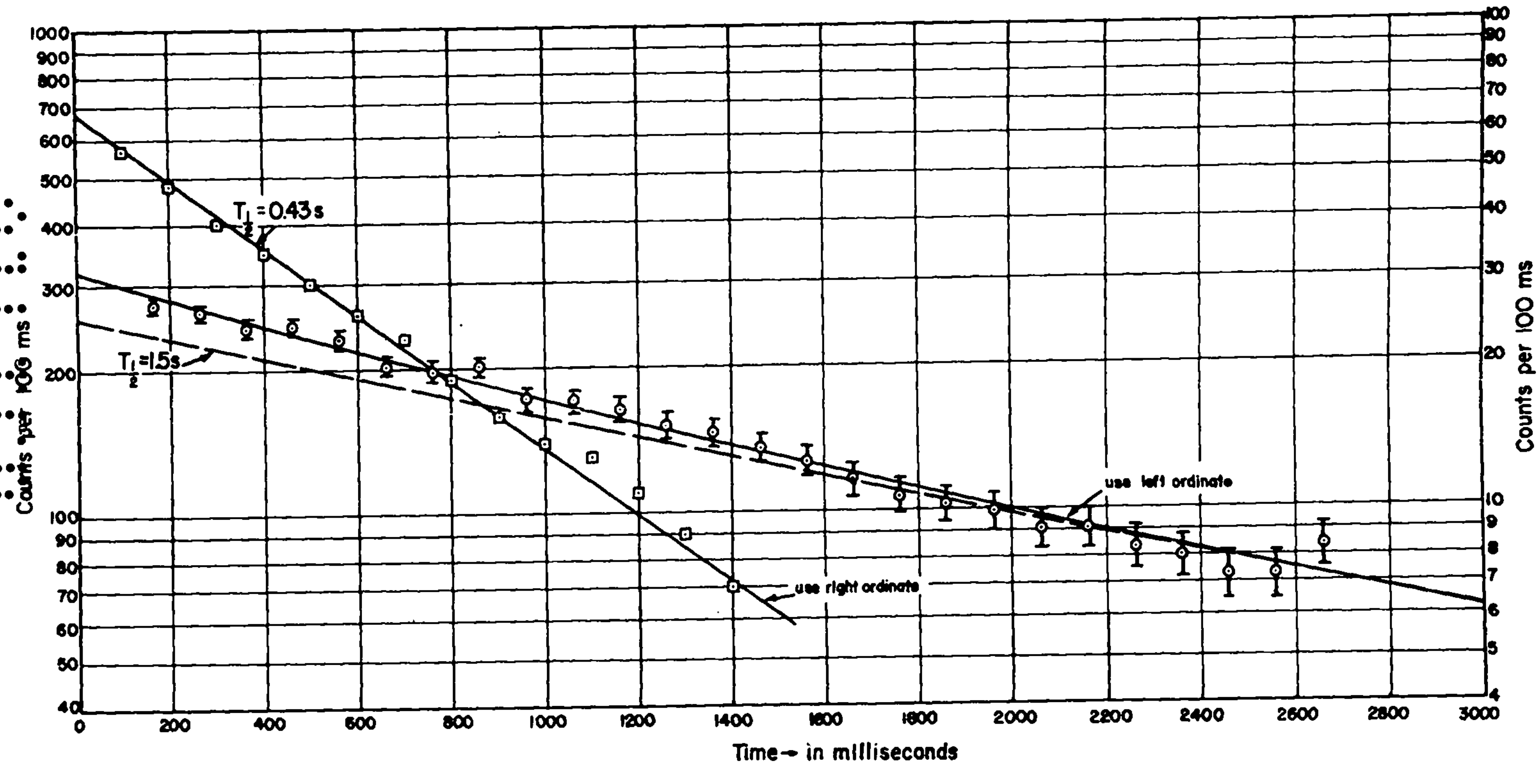


Difference between Cd in & Cd out
5
4
3
2
1
0
-1
-2
-3
-4
-5
0
10
20
30
40
50
60
70
80
90
Time in ms after bombardment

Fig. 4


$T_{\frac{1}{2}}$ = 0.43 s
$T_{\frac{1}{2}}$ = 1.5 s
use left ordinate
use right ordinate
Counts per 100 ms
Counts per 100 ms
Time → in milliseconds
0
200
400
600
800
1000
1200
1400
1600
1800
2000
2200
2400
2600
2800
3000
1000
900
800
700
600
500
400
300
200
100
90
80
70
60
50
40
100
90
80
70
60
50
40
30
20
10
9
8
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6
5
4

Fig. 5

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