JANUARY 1, 1941 PHYSICAL REVIEW VOLUME 59

Photo-Fission of Uranium and Thorium

R. O. Haxby,*† W. E. Shoupp,* W. E. Stephens,*‡ and W. H. Wells
Westinghouse Research Laboratories, East Pittsburgh, Pennsylvania

(Received August 30, 1940)

Fission of uranium and thorium has been observed to be produced by irradiation with γ-rays. The cross section for this photo-fission produced by the γ-ray from fluorine bombarded with protons has been measured and found to be:

$$\sigma_{\mathrm{U}} = 3.5 \pm 1.0 \times 10^{-27}\ \mathrm{cm}^2,$$
$$\sigma_{\mathrm{Th}} = 1.7 \pm 0.5 \times 10^{-27}\ \mathrm{cm}^2.$$

Introduction

Soon after neutron-induced fission of uranium and thorium was discovered it was pointed out that sufficient excitation of the heavier nuclei by γ-rays might also cause fission.[1, 2] A search was made in several laboratories for fission caused by γ-rays, but no effect was observed.[3, 4] The failure to observe fission of this type was thought to be caused by insufficient γ-ray intensities, as calculated from the yields of the F(p, γ) and Li(p, γ) reactions given by Livingston and Bethe.[5] However, we looked for and discovered photo-fission. This was made possible by the fact that the yield of γ-rays from F(p, γ) is actually much greater than quoted[5] and increases rapidly with proton energy. A preliminary report[6] has been published, and this paper gives a full account of our experiments.

Apparatus

The arrangement of apparatus used to observe photo-fission is shown in Fig. 1. High energy protons from the Westinghouse pressure electrostatic generator were magnetically analyzed and directed on to a CaF_2 target in a Faraday cage. This proton current was measured by a current integrator and on a microammeter connected to ground through a 45-volt battery; the Faraday cage being made negative. Gamma-rays produced in the fluorite crystal target irradiated a 12-cm^2 piece of uranium metal, placed on the high voltage plate of the ionization chamber. The ionization pulse of the fission recoils was amplified and observed on an oscilloscope. The pulses greater than twice the α-particle background were observed visually, and counted with a hand counter. The ionization chamber filled with air at atmospheric pressure was 1 cm deep, and 2000 volts were applied to the collector plate. The intense γ-ray background made it necessary to ground the grid of the first tube of the amplifier through a resistance of 20 megohms, so that the background would not change the grid bias, and consequently the gain of the amplifier. With thick uranium samples in the ionization chamber, the α-particle intensity was high enough to give α-coincidences, and hence only kicks greater than twice the highest observed coincidence kick were counted as fissions. It was determined that the

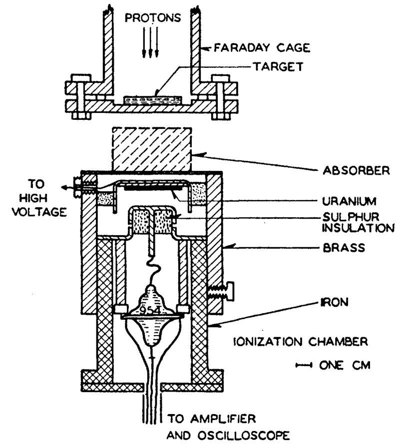

Fig. 1. Arrangement for detecting photo-fission.

---

* Westinghouse Research Fellow.

† Now with the Sperry Gyroscope Company.

‡ Now at Stanford University.

[1] E. Feenberg, Phys. Rev. **55**, 504 (1939).

[2] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

[3] Roberts, Meyer and Hafstad, Phys. Rev. **55**, 417 (1939).

[4] Heyn, Aten and Bakker, Nature **143**, 516 (1939).

[5] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 316 (1937).

[6] Haxby, Shoupp, Stephens and Wells, Phys. Rev. **58**, 92 (1940).

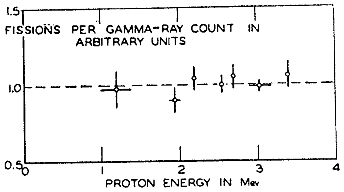

FIG. 2. Fissions per γ-ray count *vs.* proton energy.

presence of intense γ-radiation did not change the distribution of α-kicks. With thin layers of uranium, practically all the fission pulses were of a maximum height determined by saturation of the linear amplifier.

The γ-ray intensity was measured with a Geiger counter shielded with one inch of lead, except for a window over which was placed only $\frac{1}{8}$ inch of lead in the direction of the target. The counting was done with either a scale-of-four and Cenco recorder or a counting rate meter. The counter tube was placed at a distance of 3 to 8 feet from the target to prevent the counter from saturating. To correct for that fraction of the counts due to radiation scattered by the lead shield, a 4-cm plug of lead was inserted in the window and the difference with and without this plug was taken to be proportional to the intensity of γ-rays coming from the target.

EXPERIMENTS

To establish that the fission recoils observed were due to γ-rays and not to neutrons, we have found that: (1) Too few neutrons were given off from the target to cause the observed fissions; (2) the fission rate was proportional to the γ-ray intensity, the latter being varied by changing the proton energy or the proton current, and (3) the absorption of the fission-producing radiation was similar to that for high energy γ-rays and not like that for neutrons.

A few neutrons were observed to come from the CaF_2 target when it was bombarded with protons. These neutrons were detected by means of a BF_3 ionization chamber coupled to a linear amplifier, and the voltage excitation curve was measured. The threshold for neutron production of these neutrons was approximately $E_p=1.8$ Mev, and the yield was of the order of 1/800 that previously observed from Li (p, n).[7] The (p, n) reaction responsible for this background was not identified, but is reasonably ascribed to either Ca (p, n), whose excitation curve it resembles, or to lithium contamination. The intensity of neutrons from the bombardment of CaF_2 is approximately half that from calcium metal, and has, approximately, the same threshold. However, no fissions were observed when uranium was exposed to irradiation from calcium metal bombarded with protons, all other conditions being identical with the experiment in which CaF_2 was bombarded and fission observed. This proves definitely that the phenomenon we attribute to photo-fission is not caused by neutrons from the calcium (p, n) reaction. Furthermore, fissions have been observed at proton energies as low as 1 Mev, at which energy no neutrons more than the natural background were observed. As a further check, the expected number of fissions were observed when the target was changed to AlF_3, but no fissions occurred when a target of aluminum metal was used.

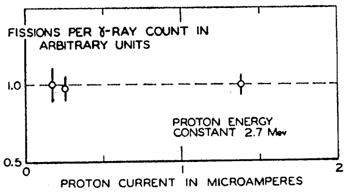

FIG. 3. Fissions per γ-ray count *vs.* proton current.

Although the γ-ray intensity, and hence the fission rate, is very low at lower proton energies, we were able to observe fissions down to proton energies of 1 Mev, and to measure the fission rate per unit of γ-ray intensity. A curve showing fissions per γ-quantum in arbitrary units as a function of proton energy from 1 to 3.4 Mev is given in Fig. 2. The γ-ray yield changes by about a factor of 70 in going from 1 Mev to 3.4 Mev. Part of this increase was compensated by increasing the proton current at lower proton energies. The points in Fig. 2 are averages of several runs, and the vertical lines through the points indicate the statistical uncertainty. The number of fissions per unit of γ-ray intensity was measured as the proton current was changed. These results are given in Fig. 3.

The absorption of the fission-producing radiation from the target was measured by interposing various absorbers between the target and the

---

[7] Haxby, Shoupp, Stephens and Wells, Phys. Rev. 57, 348A (1940).

uranium. The geometry used is shown in Fig. 1. The Geiger counter and shield used to measure the γ-ray intensity were at right angles to the proton beam, and were 228 cm away. The γ-ray intensity readings registered by the Geiger counter, were not corrected for radiation scattered by the lead shield as the γ-ray intensity remained practically constant. The presence of the various absorbers placed between the target and the fission counter caused no change in γ-ray intensity. The number of fissions per unit of γ-ray intensity, in this case per Geiger count, is plotted in Fig. 4 for different absorbers and different thicknesses of lead absorber. These readings were taken with a proton beam current of 0.5 μa of 3.4-Mev protons. The absorption coefficients taken from the curves of Fig. 4 are tabulated in the second column of Table I. In the third column the absorption coefficients calculated for 6-Mev γ-rays are tabulated. The agreement between columns two and three is quite satisfactory. In contrast, similar absorption measurements with the neutrons from Li (p, n) to cause fissions, were made. The absorption coefficients determined in the same way for lead, iron, and paraffin, were 0.23±0.05, 0.13±0.05 and 0.13±0.05 cm^{-1}, respectively. The vast difference in absorption shows conclusively that the observed fissions are caused by γ-rays.

It seems unlikely that a secondary reaction such as the photo-disintegration of the plate on which the uranium was mounted should give enough neutrons to cause an appreciable number of the observed fissions. We observed no change in fission rate when the backing plate was doubled in thickness, or when it was changed from brass to aluminum. Furthermore, the experiment which measured the neutrons produced when CaF_2 was bombarded with protons should likewise have measured neutrons produced by photo-disintegration or brass or aluminum since the BF_3 counter contained large quantities of both substances. The fact that approximately half as many neutrons were observed from CaF_2 as from Ca indicates that the fluorine γ-rays do not cause appreciable photo-disintegration in aluminum or brass. Therefore, the fission observed cannot be caused by photo-neutrons from these substances.

TABLE I. *Absorption coefficient of fission-producing radiation.*

	ABSORPTION COEFFICIENTS (CM^{-1})	
ABSORBER	OBS.	CALC. FOR 6 MEV γ-RAYS
Paraffin	0.01±0.07	0.04
Aluminum	0.14±0.07	0.07
Iron	0.28±0.07	0.26
Lead	0.53±0.08	0.50

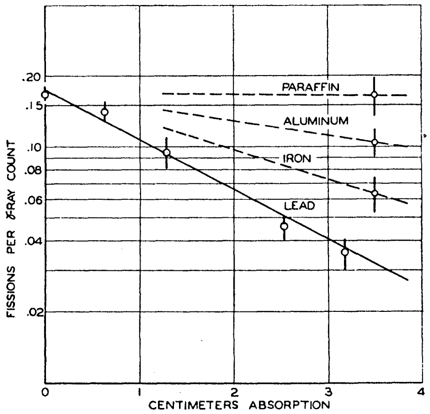

FIG. 4. Fissions per γ-ray count *vs.* thickness of γ-ray absorber.

When uranium was removed from the ionization chamber there were no observed fission kicks. Likewise, with the proton beam hitting a quartz plate instead of the fluorite target, no fissions were ever observed. Occasional pulses induced from switch transients, sparking in the high voltage, or spray voltage were observed, but since the counting was visual, these could easily be distinguished from fission kicks by their wave form and were, consequently, not counted.

CROSS SECTION FOR PHOTO-FISSION

The cross section for the photo-fission process was measured by using a uranium film thin enough so that every fission process could be observed in the ionization chamber. Since each fission produces a *pair* of recoil atoms having roughly the same mass, one of the particles will be in a direction to be recorded by the ionization, while the other will be absorbed in the backing plate on which the uranium is deposited. The evaporated films we used contained approximately 1 mg/cm^2, which is roughly $\frac{1}{4}$ cm air equivalent in stopping power.[8]

---

[8] We wish to thank Dr. E. D. Wilson of these laboratories for preparing the films for us.

If such a film contains N atoms of uranium and if it is traversed by a γ-ray beam of δ quanta/cm^2-sec., and if σ is the cross section for the fission process in cm^2, then the number of fissions per second, f, is

$$f = \sigma N \delta. \tag{1}$$

To find N for the thin films of uranium, we measured the weight of the film and also the number of α-particles emitted from it. The first film was gray, and was assumed to be U_3O_8, while the second was black, assumed to be UO_2. The weight of the first film was 4.06 mg, giving $N = 0.87 \times 10^{19}$. The uranium used was separated chemically about 10 years ago; consequently it does not contain an appreciable amount of ionium, and the U II should be in approximate equilibrium. Actino-uranium should be present in the normal ratio. Using Nier's disintegration constants[9] we calculate a total α-emission per second per atom of uranium to be 9.76×10^{-18}. The observed number of α-particles was assumed to be half the total number, although actually, because of the geometry, it will be somewhat less. For the first film the observed number of α-particles per second was 40, giving $N = 0.82 \times 10^{19}$. This is in good agreement with the estimate made from the weight of the film. The second, black, film (UO_2) weighed 5.75 mg, giving 1.27×10^{19} atoms from the weight. It emitted 57 α-particles per second, giving 1.17×10^{19} atoms from the activity.

We were unable to get such a good check in the analogous procedure for a thin evaporated film of thorium. Therefore, the cross section for thorium was obtained by comparing the fission yields from thick targets of uranium and thorium. In each case the same exposed area of freshly sandpapered metal was used. The relative yield gives:

$$\sigma_{\mathrm{U}}/\sigma_{\mathrm{Th}} = 2.0 \pm 0.1. \tag{2}$$

This is the average of several runs at different proton energies. The uncertainty arises mainly from the statistical fluctuations involved. The thorium used was spectroscopically free of uranium, and the uranium had less than a few percent of thorium contamination.

Next we consider the factor δ in Eq. (1). Let Γ be the total number of quanta/sec. from the target. Then $\delta = \Gamma/4\pi r^2$, where r is the distance from the uranium to the γ-ray source. We have

$$\Gamma = pqy, \tag{3}$$

where p is the number of protons per second striking the fluorite crystal, y is the yield in quanta per proton at 1-Mev proton energy, and q is the ratio of the yield at the energy used to that at 1 Mev.

With a Geiger counter cylinder 1×5 cm located 82 cm from the fluorite target, we observed 7.4 γ-counts/sec. when the proton current was 1.4 μa (8.7×10^{12} proton/sec.) at 1 Mev. Assuming the counter efficiency to be 2 percent[10] this gives roughly 7.3×10^{-7} γ-quanta/proton at 1 Mev. This yield has been independently and more carefully measured by Lauritsen, Fowler, and Streib to be 7.6×10^{-7} γ-quanta/proton at 1 Mev.[11] The agreement between our rough Geiger counter measurements and this more accurate electroscope measurement is reassuring. However, the number of quanta per proton we obtain *is about 1000 times greater* than the value given by Livingston and Bethe.[5] That value is presumably derived from Hafstad's[12] comparison of the lithium and fluorine γ-ray intensities, together with a rough guess for the Li (p, γ) yield. Hence, the value for Li (p, γ) must also be much too low. In the calculations which follow, we will use $y = 7.6 \times 10^{-7}$ quanta/proton.

We have taken many excitation curves to determine the value of q in Eq. (3). Some runs were taken with protons, mass 1; others were taken by comparing the yield from mass 1 at 3 Mev with mass 3 at 3 Mev (effectively 1 Mev per proton). The averages of several runs are given in Table II. These are probably not accurate to better than 10 percent. The value $q = 10.5$ at 2.0 Mev checks with that obtained at Wisconsin[13] where they obtained $q = 11$.

TABLE II. *Yield of quanta/proton as a function of proton energy*

E_p (Mev)	1.5	2.0	2.9	3.0	3.2	3.4
q	3.4	10.5	52	55	63	70

The notation F (p, γ) is a convenient but in-

---

[9] A. O. Nier, Phys. Rev. **55**, 152 (1939).

[10] J. V. Dunworth, Rev. Sci. Inst. **11**, 167 (1940).

[11] We wish to thank Dr. Fowler for informing us of this result before publication.

[12] Hafstad, Heydenburg and Tuve, Phys. Rev. **50**, 504 (1936).

[13] Herb, Kerst and McKibben, Phys. Rev. **51**, 691 (1937).

exact designation for the γ-rays from fluorine bombarded with protons, since it has been shown[14] that most of the high energy rays are emitted from an excited state of O^{16} derived from $(Ne^{20})^*$ by emission of an α-particle. The energy of the γ-rays has been measured to be 6.3 Mev[14] and the character of the γ-rays does not seem to change when the proton energy is varied from 0.33 to 1.36 Mev. Since we observed fission at 1-Mev proton energy it seems probable that the 6.3-Mev quanta cause the fissions. Also since the fissions/(γ-quantum) remain constant with increase of proton energy from 1 Mev to 3.4 Mev, it seems probable that there is no marked change in the γ-ray spectrum over this range. For the cross-section calculation we assume as equally effective all the quanta recorded on the counter (through $\frac{1}{8}$ inch of lead). This assumption neglects the possibility that the γ-rays consist of several close lines or have weak high energy components.

Fifteen runs were taken in the determination of the fission rate per proton current. Two uranium films were used, the proton energy and current were varied as well as the distance of the uranium to the target. In a sample run, 41 fissions were observed in 295 seconds from a uranium film containing 1.22×10^{19} atoms placed so that $1/4\pi r^2$ was 0.0134 cm^{-2} when the CaF_2 target was bombarded with 0.91 μa of 3.0-Mev protons. The mean value of these fifteen determinations of the uranium cross section is $\sigma=3.47\times 10^{-27}$ cm^2. The distribution of the individual values gives a root-mean-square deviation from the mean of 0.81×10^{-27}, most of which arises from the fluctuation statistics caused by the finite number of fissions in each run. Other sources of error are such that this value of the cross section may well have a probable error of 30 percent. Coupled with the result already cited for the ratio of σ_U to σ_{Th}, we get $\sigma_{Th}=1.73\times 10^{-27}$ cm^2 also with a probable error of about 30 percent.

We have also observed fission in both uranium and thorium produced by the 17-Mev γ-rays from lithium bombarded with protons. The effect is quite weak, because of the smaller number of quanta available, but the cross sections seem to be at least of the same order of magnitude as with the fluorine γ-rays, and are being studied further. We have also observed neutrons emitted when thorium is irradiated with fluorine γ-rays. These are probably released in the fission process, and are being investigated further. We have observed no fission in bismuth, thallium, or mercury, with either lithium or fluorine γ-rays. However, the intensity of lithium γ-rays used was relatively small.

DISCUSSION

Bohr and Wheeler[2] predicted a cross section for photo-fission of 10^{-27} and 10^{-28} cm^2 for uranium and thorium, respectively, on the basis of Bothe and Gentner's[15] photo-disintegration cross section of 5×10^{-26} cm^2 and estimates of the partial widths of nuclear levels caused by fission and by neutron emission founded partly on theoretical considerations, partly on the observational material available at that time.

Additional experiments performed in the meantime make it possible to give in the present instance a somewhat more detailed treatment of the balance of energy which, according to Weisskopf[16] and Bohr and Wheeler[2] determines the partial widths of nuclear levels.

In particular, in view of our recent measurements of the neutron fission thresholds[17] it seems that Bohr and Wheeler's curve of the critical energy of fission should have its ordinate scale lowered by about $\frac{1}{2}$ Mev. The critical energy for fission E_f is the sum of the neutron binding energy E_n and the neutron energy necessary to produce fission, Q_n. Since Bohr and Wheeler have calculated E_n to be 5.1 Mev for U^{239} and 5.2 Mev for Th^{233} and our measurements suggest Q_n is 0.3 and 1.1 Mev,[17] then E_f should be 5.4 and 6.3 Mev, respectively. These values are just 0.5 Mev less than those estimated by Bohr and Wheeler.[18] This revision of their E_f curve gives for U^{238} and Th^{232} (the nuclei concerned in photo-fission) critical energies of fission of 5.2 and 6.1 Mev. The neutron binding energies of these same nuclei

---

[14] W. A. Fowler and C. C. Lauritsen, Phys. Rev. **56**, 840 (1939).

[15] W. Bothe and W. Gentner, Zeits. f. Physik **106**, 236 (1937).

[16] V. F. Weisskopf and D. H. Ewing, Phys. Rev. **57**, 472 (1940).

[17] Haxby, Shoupp, Stephens and Wells, Phys. Rev. **57**, 1088A; **58**, 199A (1940).

[18] This revision makes slow neutron fission of Pa to be expected. The cross section may, however, be small enough to have been missed at Columbia.

calculated as in Bohr and Wheeler's paper are 6.1 (U^{238}) and 6.2 Mev (Th^{232}).[19] Excitation of U^{238} by a 6.3-Mev γ-ray, then, raises the nuclear energy to 1.1 Mev above the critical fission energy but only 0.2 Mev above the energy necessary to emit a neutron. Hence the Γ_f as read from Bohr and Wheeler's Fig. 5 is certainly larger than Γ_n and Γ_r. For Th^{232}, on the other hand, the 6.3-Mev γ-excitation raises the nucleus to an energy 0.2 Mev above the critical fission energy and 0.1 Mev above the energy necessary to emit a neutron. Since these energies are not accurately known, and since Γ_n, especially, depends very critically on the excess energy in this region, all we may say is that Γ_f, Γ_n, and Γ_r may be of the same order of magnitude.

The cross section for the ejection of neutrons from various heavy nuclei by γ-rays of 17 Mev energy has been measured by Bothe and Gentner[15] to be 5×10^{-26} cm². In these experiments the nuclei are raised to excited states of such energy that neutron emission is overwhelmingly more probable than radiative de-excitation. This will no longer necessarily be the case when the excitation is produced by the fluorine γ-rays and when, as in our experiments, fission provides an additional means of disposing of excited nuclei. The cross section will be reduced below the figure of Bothe and Gentner by the fraction $\Gamma_f/(\Gamma_f+\Gamma_n+\Gamma_r)$ on this account. It will be further reduced because the probability of radiative excitation of the normal nucleus varies (in the region of continuous level distribution with which we are concerned) approximately as the cube of the quantum energy.[20] Thus we have as the cross section for the radiative production of fission

$$\sigma\sim\Gamma_f/(\Gamma_f+\Gamma_n+\Gamma_r)5\times 10^{-26}(6.3/17)^3$$
$$=\Gamma_f/(\Gamma_f+\Gamma_n+\Gamma_r)2.5\times 10^{-27}\text{ cm}^2.$$

As we have seen, Γ_f for U^{238} is larger than Γ_n and Γ_r, so that $\Gamma_f/(\Gamma_f+\Gamma_n+\Gamma_r)$ is approximately one. This gives $\sigma_U=2.5\times 10^{-27}$ cm² in agreement with the observed value 3.5×10^{-27} cm². For Th^{232}, on the other hand, since Γ_f, Γ_n and Γ_r may all be of the same order of magnitude, we can say only that the fraction, $\Gamma_f/(\Gamma_f+\Gamma_n+\Gamma_r)$ is between 0 and 1, giving $\sigma_{Th}=0$ to 2.5×10^{-27} cm². Whether competition from neutron emission, or radiation, or both, is the cause of the lower cross section for thorium will have to be decided by more accurate knowledge of the energies involved and the partial widths, or by more experimental information on the neutrons emitted. As Dr. Wheeler suggests, there seems to be more chance that neutron emission is the competing mechanism since there is more likelihood that the neutron width will exceed rather than fall below the radiation width.

We wish to thank Dr. F. W. Stallman for assistance with some of the measurements, and Dr. E. U. Condon for his help and encouragement throughout all these experiments.

---

[19] We are indebted to Dr. Wheeler for these calculations and for his emphasis that these values are accurate to 0.2 Mev because of the use of accurately known natural disintegration energies to calculate the packing fractions. We also wish to thank him for his help in revising this section.

[20] We wish to thank Dr. Weisskopf for suggesting these considerations.