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DEUTERON INDUCED FISSION OF URANIUM AND THORIUM

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1. Introduction.

The first account of experiments with the object of producing nuclear fission with artificially accelerated charged particles was published by GANT [1]. Using deuteron beams of 8 MeV from the cyclotron in the Cavendish Laboratory in Cambridge, GANT obtained detectable fission effects in uranium, but he did not attempt a quantitative determination of the cross-section for the process. In view of the desirability of a closer study of the problem and especially of testing the theoretical considerations developed by BOHR and WHEELER [2], further experiments were carried out by the writers by means of the cyclotron constructed in the Institute of Theoretical Physics at Copenhagen and recently described by one of us [3]. In these experiments, of which a preliminary account has already been published [4], deuteron beams up to 9,5 MeV were used and cross-sections for fission in uranium and thorium for varying deuteron energies were measured. In particular, it was found that the value of the relative fission cross-section for thorium and uranium at 9 MeV deuteron energy was about 0,7, in close agreement with the value obtained independently by KRISHNAN and BANKS [5] in the Cavendish Laboratory. In the present paper, a fuller account of the method used in our experiments and of the results obtained is given together with a closer comparison with the theoretical expectations.

2. Experimental method.

The irradiation of the thorium and uranium samples was carried out within the vacuum tank of the cyclotron since the deflecting field was not sufficient to bring the deuteron beam outside the tank wall. The targets were inserted and removed from the cyclotron by means of a vacuum lock which, besides avoiding air penetration into the tank, had the advantage that the targets could be brought close to the deflecting electrode, where the cross-section of the beam is smallest. This part of the arrangement was described in the earlier paper [3].

In most of the experiments with uranium, the target was a thin layer of metallic uranium supported on a sheet of aluminium. The target was prepared by evaporation of metallic uranium in the form of powder from a tungsten ribbon in vacuum. The thickness of the uranium layer was determined by weighing one of the targets; the amount of uranium in the other targets was then determined relatively to this one by counting the β-particles emitted by the samples. The thickness of the uranium layers used was about 0,5 mg per cm^2. For the experiments with thorium, a quantity of thorium in the form of powder was available. Still, attempts to obtain a layer of metallic thorium by evaporation were unsuccessful because, what was soon found, the powder consisted mainly of thorium oxide. The thorium targets finally used in the experiments were in all cases "thick" targets obtained by compressing thorium oxide.

The target holder is shown in Fig. 1. The targets are fastened by screws in the positions a_1, a_2, a_3 and a_4 on the four sides of a brass block supported by a brass tube.

The latter is insulated by a pyrex tube from an outer brass tube which slides through a rubber gaskett (not shown in the figure) so that the target holder can be moved in and out and rotated. The outer brass tube supports a cylindrical brass case with four openings, c_1, c_2, c_3 and c_4, facing the four sides of the brass block; the energy of

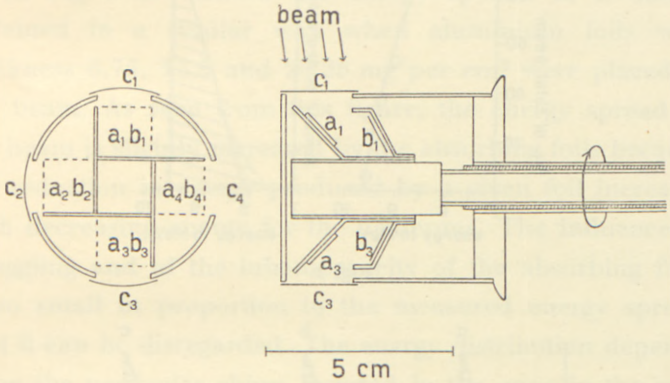

Fig. 1.

the deuterons hitting the target can be varied by covering the openings in the case with aluminium foils. The space inside the brass case is divided into four partitions by copper sheets. Facing the targets, aluminium foils of thickness 6,5 mg per cm^2 used for collecting the fission products could be placed in the positions b_1, b_2, b_3 and b_4. The β-ray activity of the fission products was measured by a counter connected to a scale-of-eight amplifier. The stopping power of the window of the counter corresponded to 4 cm of air, the solid angle occupied by the window, as seen from the radioactive source measured, was about $^1/_{10} \cdot 4\pi$.

The deuterons in the beams used had energies up to 9,5 MeV. The total beam current was 0,1—0,2 microamp.,

which was sufficient for the experiments; with larger currents the cooling of the targets would also have caused difficulties with the present arrangement. The energy distribution of the deuteron beam was determined by

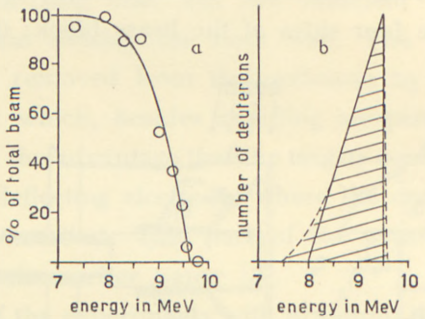

Fig. 2.

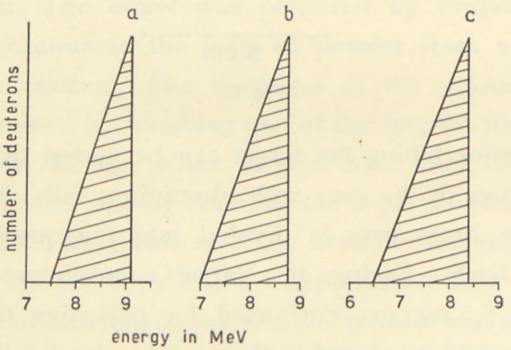

Fig. 3.

placing aluminium foils in the beam and measuring the transmitted current; the connection between range and energy was obtained by use of the range curves given by LIVINGSTON and BETHE [6]. The ordinates of the curve *a* in Fig. 2 indicate, for one of the deuteron beams used, the part of the beam current with energy greater than the values plotted as abscissae. The corresponding energy

distribution *b*, shown in Fig. 2, is obtained by differentiation of the absorption curve *a*. The distribution thus obtained is given by the dotted line. For the evaluation of the results, however, the curve shown by the full line was used. It will appear from the later discussion of the results that the difference between the two curves is insignificant.

In Fig. 3 are shown the energy spectra a, b and c obtained in a similar way when aluminium foils with thickness 6,75, 13,5 and 20,25 mg per cm^2 were placed in the beam. As seen from this figure, the energy spread of the beam is slightly increased by the absorbing foils because the reduction in energy produced by a given foil increases with decreasing energy for the deuterons. The influence of straggling and of the inhomogeneity of the absorbing foils is so small in proportion to the measured energy spread that it can be disregarded. The energy distribution depends upon the particular shims inserted in the gaps in the pole pieces of the cyclotron magnet (see [3] p. 13) and, each time when a change had been made in the arrangement of shims, a new determination of the energy distribution had, therefore, to be made.

3. Yield of fission as a function of deuteron energy.

The procedure used in the determination of the yield of fission products as a function of the deuteron energy was to irradiate two targets of uranium or thorium with deuterons with different energies but with the same beam current and to measure the ratio of the amounts of fission products collected. For a definite length of the time of irradiation, the decay curves obtained with different deuteron energies run very nearly parallel. In relative determ-

inations, it was thus sufficient to measure the activity of the fission products at some definite time after the end of the irradiation. In the actual experiments, this time was 90 minutes.

During the irradiation, which lasted for 40 minutes, the target holder was kept in constant rotation in order to eliminate the influence of the fluctuations in the beam current. The rate of rotation was about 1 revolution in 5 seconds, which was sufficient to ensure that each of the four sides of the target holder received the same number of deuterons. The two uranium targets used were "thin" targets made by evaporation of uranium on aluminium and measured in the way mentioned above. It was found that the uranium amount on the targets, which we shall refer to as uranium I and uranium II, were in the ratio I:II = 1:1,08. They were placed in the two positions in the target holder denoted by a_1 and a_2 in Fig. 1. In the position a_3 was placed an aluminium sheet without uranium and, facing all three targets, were placed the collecting foils b_1, b_2 and b_3. The activity of b_3 gave a control on the activity of the collecting foils produced by neutrons and by scattered deuterons; this activity was found to have a period of about 10 minutes due to the formation of 27 Mg (^{27}Al (n, p) ^{27}Mg), but it was not strong and, 90 minutes after the end of the irradiation, it had completely disappeared. The activity of b_1 and b_2 at that time was then a direct measurement of the number of fission particles collected. In the position a_4 of the target holder was placed a copper sheet which was used as a current integrator to obtain a relative measurement of the total number of deuterons for comparison with other experiments; the amount of ^{64}Cu (period 13 hours) produced during the

irradiation is proportional to the total number of deuterons hitting the target.

Table I gives the data from one of the experiments.

Table I.

Target	Thickness of absorbing foil	Activity of collecting foil (90 minutes after the irradiation)
1. Uranium I	13,5 mg/cm²	14,0 counts per min.
2. — II	20,25 —	6,4 — — —
3. Aluminium	6,75 —	0 — — —
4. Copper	0 —	no collecting foil; activity of the copper target itself in relative measure: 620

At first we consider the case of the target 2 which was hit by deuterons with the upper limit 8,4 MeV (Fig. 3 c); here the conditions are simple, because almost the whole fission effect can be ascribed to deuterons with energy near the upper limit. In fact, in other experiments with deuteron energies below 8 MeV a fission effect was found, which was small compared with that found with 8,4 MeV deuterons. This shows, firstly, that the effect due to fission by neutrons is small, and, secondly, that the effect found with the target 2 can be ascribed to deuterons in the energy interval 8,0 to 8,4 MeV. The fission effect for deuterons in this energy interval is overestimated, since part of the effect is due to deuterons with smaller energy; but the error is without influence on the results obtained for higher deuteron energies. From the energy distribution curve (Fig. 3 c) it is found that 38 % of the beam lies in the region between 8,0 and 8,4 MeV; the activity observed

was 6,4; now, if the whole beam had been lying in this region, then the effect would have been 6,4:0,38 = 17.

We then consider the case of the target 1, which was hit by deuterons with the upper limit 8,75 MeV (Fig. 3 b). The effect 14,0 is at first corrected to 14,0·1,08 = 15,1 due to the difference in the thicknesses of the targets. In the same way as before, we can neglect the effect due to deuterons with energies smaller than 8,0 MeV and ascribe the whole effect to deuterons in the two regions 8,0—8,4 MeV and 8,4—8,75 MeV; from the energy distribution curve (Fig. 3 b) it is found that 31 % and 36 % of the beam lie in these two regions, respectively. The deuterons in the region 8,0—8,4 MeV must give the effect 17·0,31 = 5,3 and the deuterons in the other region 8,4—8,75 must then give the rest of the effect 15,1 — 5,3 = 9,8; if now the whole beam had been lying in the last region, the effect would have been 9,8:0,36 = 27.

The results of this experiment can be summarized as follows:

Mean deuteron energy	8,2 MeV	8,58 MeV
Relative fission yield.....	17	27

In another experiment, the thicknesses of the foils c_1 and c_2 were 0 and 6,75 mg per cm², corresponding to the energy distribution curves in Fig. 2 b and Fig. 3 a, respectively. Before comparing with the experiment above we must apply a correction to the activities found in this experiment, because the activity of the copper target was now only 550, measured in the same units as in the former experiment. As before, we divide the effect into parts due to deuterons in smaller energy regions. The effect of the deuterons in the regions 8,0—8,4 MeV and 8,4—8,75 MeV may be cal-

culated from the figures given above and the energy distribution curves. In quite the same manner as just described, we get further figures corresponding to the regions 8,75—9,1 MeV and 9,1—9,5 MeV.

Table II gives the results of these two experiments.

Table II.

Mean deuteron energy	Relative fission yield of uranium
8,2 MeV	17
8,58 —	27
8,92 —	48
9,3 —	81

The stopping power of the targets was about 5 mm of air so that the energy loss of the deuterons in passing through the targets was negligible. The results are, therefore, an approximation to those which would be obtained by bombarding an infinitely thin target with a homogeneous beam.

In the experiments with thorium, thick targets of ThO_2 were used, but the method was the same. The results are given in Table III.

Table III.

Mean deuteron energy	Relative fission yield of thorium
7,6 MeV	7
8,12 —	12
8,55 —	52
3,92 —	85
9,3 —	175

It may be stressed that the figures in Tables II and III which refer to "thin" and "thick" targets, respectively, give no information about the ratio between the fission yields in uranium and thorium under similar conditions. Since, however, the fission products are only escaping from a thin upper layer of the thick targets and the deuterons loose only a small part of their energy in passing through this layer, the results are in relative measure also for thorium approximately the same which would be obtained by using a thin target and a homogeneous beam.

4. Comparison between uranium and thorium.

In some of the experiments with thorium, thick targets of U_3O_8 were placed on the target holder together with the targets of ThO_2; in this way a comparison between the yield of fission from uranium and thorium was obtained. In Fig. 4 are shown the decay curves of the collecting foils from such an experiment; the data of this experiment are given in Table IV.

Table IV.

Target	Max. deuteron energy	Corresp. curve in fig. 4
1. ThO_2	9,5 MeV	1
2. U_3O_8	9,5 —	2
3. ThO_2	8,75 —	3
4. Copper	9,5 —	4

Curve 1 gives the activity of the collecting foil facing the target 1 (ThO_2), corrected for the neutron effect given by curve 4. The vertical line in the figure shows the time at which the relative activities of the fission products were

measured; this was chosen so that the correction due to neutrons could be omitted. The mean value of the yield from thorium relative to that from uranium of two such experiments, which were in very good agreement, was

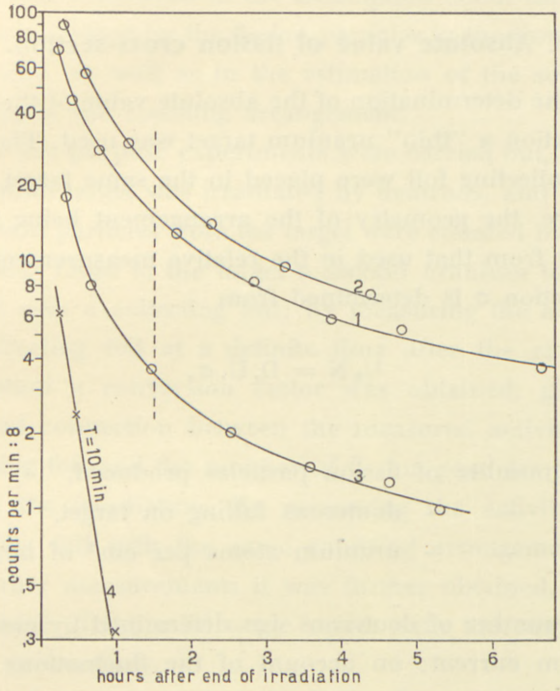

Fig. 4.

75 %. In estimating the ratio for the fission cross-section in the two elements it should be taken into account that the stopping power in the uranium layer for the fission fragments escaping from it will be somewhat larger in proportion to the amount of heavy element present. In fact, due to the greater relative number of oxygen atoms in U_3O_8 than in ThO_2, the stopping power per atom of heavy element for the fission fragments must be expected

to be about 10 % greater in the uranium than in the thorium target. We may conclude, therefore, the fission cross-section of thorium for 9 MeV deuterons to be about 70 % of that of uranium.

5. Absolute value of fission cross-section.

For the determination of the absolute value of the fission cross-section a "thin" uranium target was used. The target and a collecting foil were placed in the same target holder as before, the geometry of the arrangement being slightly different from that used in the relative measurements. The cross-section σ is determined from

$$^1/_2 N = D.U.\sigma, \qquad (1)$$

where

N = number of fission particles produced,
D = — - deuterons falling on target,
U = — - uranium atoms per cm^2 of target.

The number of deuterons was determined by measuring the beam current; on account of the fluctuations in the beam it was somewhat difficult to fix a mean value of the current. The uncertainty in the determination of the number of deuterons was estimated to be about 25 %.

A preliminary determination of N was obtained by assuming that the number of β-particles emitted from the fission products within some 10 hours after the end of the irradiation was 3 per atom. From a determination of the number of β-particles emitted from the collecting foil in this time interval and from the geometry of the arrangement, N could thus be determined.

In this way we got for the deuteron energy 9 MeV the value $\sigma = 5.10^{-27}$ cm^2 mentioned in our first note (4). In continuing the work we have, however, attempted to obtain a more reliable value for the cross-section by avoiding the uncertainties involved in the assumption about the number of β-rays emitted by the fission particles in the time interval concerned, as well as in the estimation of the solid angle entering in the counting arrangement.

For this purpose experiments were carried out, in which a uranium target was irradiated by neutrons, and in which the fission particles from the target were counted by a linear amplifier. Close to the target a second uranium target was placed with a collecting foil; by measuring the activity of the collecting foil at a definite time after the end of the irradiation a conversion factor was obtained, giving the required connection between the measured activity of the collecting foil and the number of fission particles produced during the irradiation. By measuring the activity of the collecting foil with the same counting arrangement as in the earlier measurements it was further obtained, that the knowledge of the solid angle underneath the counter was made unnecessary.

The apparatus used in the experiments with neutrons is shown in Fig. 5. In a hemispherical ionization chamber with radius 2 cm a target T_1 consisting of a brass disc with a thin layer of uranium hydroxide ($(NH_4)_2U_2O_7$) or of metallic uranium was placed at the centre; the brass disc was connected to the grid of the first valve of a linear amplifier. The hemisphere forming the other electrode of the ionization chamber was kept at a potential of 500 volts. The ionization chamber was surrounded by a cylindrical screen to shield it against high frequency disturbances.

Inside the screen the second uranium target T_2 was placed with a collecting foil, which in this case was a thin lead foil (48 mg/cm^2) placed in direct contact with the target. As a consequence of the geometry of the apparatus the number of fission particles received by the collecting foil

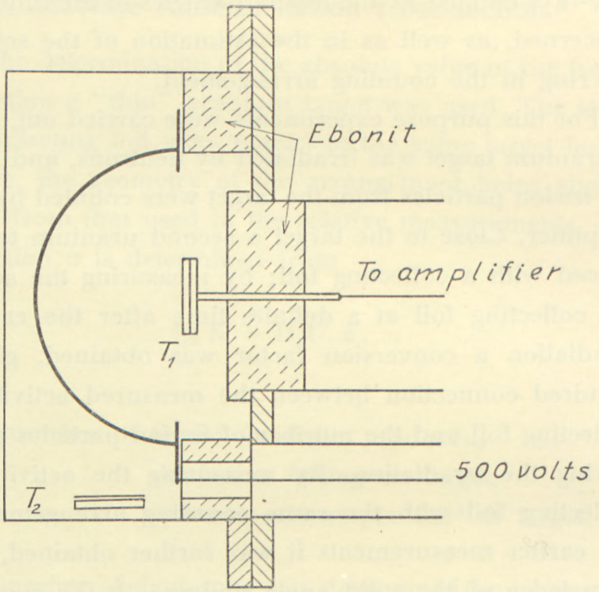

Fig. 5.

will be the same as the number of particles counted by the amplifier, when referred to the same amount of uranium in the two targets.

The ionization chamber was placed at a distance of about 30 cm from the neutron source, a beryllium target bombarded by deuterons in the cyclotron. The time of irradiation was 40 minutes as in the earlier experiments. In the present experiments, where the activity of the collecting foil was much smaller than in the experiments with deuterons, the activity of the collecting foil was measured

20 minutes after the end of the irradiation. In a particular experiment the following data were obtained: the amounts of uranium were 0,92 and 0,20 mg on T_2 and T_1 respectively. During the irradiation period of 40 minutes 2270 fission particles were counted by the amplifier; the activity of the collecting foil, when measured 20 minutes after the end of the irradiation, was 1,3 in the same unit as used previously. This gives for the ratio (number of fission particles) : (activity of collecting foil) $f = \frac{2270}{1,3} \cdot \frac{0,92}{0,20} = 8000$. In four experiments with different targets the values obtained for f were f = 8000, 5400, 8600, and 5300 with a mean value of f = 7000.

We now return to the determination of the fission cross-section for deuteron impact. In one of our experiments the beam current was 0,045 microamp. during 40 minutes and for D we get thus the value $D = 6,8 \cdot 10^{14}$. Further, U was $1,8 \cdot 10^{18}$ atoms per cm^2, and the activity of the collecting foil (corrected for effect of neutrons and scattered deuterons), when measured 20 minutes after the end of the irradiation, was found to be 170. Thus, $170 \cdot 7000$ fission particles were collected on the foil, and the solid angle of the collecting foil as seen from the target being $\frac{1}{50} \cdot 4\pi$, the total number of fission particles produced was $N = 170 \cdot 7000 \cdot 50 = 6,0 \cdot 10^7$. From these figures and the equation (1) we get therefore $\sigma = 2,4 \cdot 10^{-26}$ cm^2 as a mean value of the cross-section over the deuteron energy interval concerned.

In the experiments the deuteron energy spectrum was that shown in Fig. 2 b. From the energy distribution curve and from the variation of σ with deuteron energy determined above, the cross section $\sigma_{9,0}$ for the deuteron energy 9,0 MeV

can be determined by a simple numerical calculation. The result is

$$\sigma_{9,0} = 2{,}2 \cdot 10^{-26}\ \text{cm}^2 \qquad (2)$$

Besides this experiment we have performed two experiments with lower deuteron energies corresponding to the energy spectra in Figs. 3 a and 3 b. These measurements, treated in the same way as above, gave values for $\sigma_{9,0}$ of about $3 \cdot 10^{-26}$ cm^2. On account of the much larger and more uncertain corrections due to the influence of the lower part of the energy spectrum these values are, however, less reliable than the value (2). From a consideration of the probable errors in the various steps in the calculations we estimate the uncertainty of this value to be about 50 %.

6. Comparison of the results with the theory.

The theory of nuclear fission is based on the assumption that fission in competition with other nuclear transformations takes place in a highly excited intermediate compound nucleus. In neutron induced fission, this compound nucleus is just formed by the temporary capture of the neutron by the original nucleus. In deuteron induced fission, however, two types of processes of forming a compound nucleus must be taken into account. In the one process, the intermediate state is formed by the fusion of the whole deuteron with the original nucleus while, in the other process, the deuteron breaks up during the impact with the result that the proton escapes and the neutron is captured in the nucleus. In the former process, the excitation of the compound system will for heavy nuclei be much higher than the neutron binding energy as well as the

critical fission energy. In the latter process, however, the excitation will, on the average, be lower than the neutron binding energy and, therefore, for almost all nuclei be insufficient to produce fission. As pointed out by BOHR and WHEELER [2], the intermediate capture of the whole deuteron by the original nucleus will, thus, be necessary in order to obtain a considerable fission probability by deuteron impact.

For the high excitation concerned, the probability of fission of the compound nucleus will be almost independent of its energy and the fission output will, therefore, vary with varying deuteron energy in the same way as the cross-section σ_0 for the penetration of the deuteron into the original nucleus. This cross-section will be approximately given by the formula (see [2], p. 449)

$$\sigma_0 = \pi R^2 e^{-P}. \tag{3}$$

R is the nuclear radius, and P is given by

$$P = \frac{4 Z\varepsilon^2}{\hbar \cdot v} \left(\text{are cos } x^{1/2} - x^{1/2} (1-x)^{1/2}\right), \; x = \frac{ER}{Z\varepsilon^2} \tag{4}$$

where v and E denote the velocity and energy of the deuteron, respectively, and Z is the atomic number of the nucleus concerned.

The value of R to be introduced into (3) and (4) is rather uncertain; but, choosing, in agreement with the value given in [2] for uranium

$$R = 1{,}45 \sqrt[3]{A} \cdot 10^{-13} \text{ cm} \tag{5}$$

where A is the atomic weight, we obtain for thorium (Z = 90, A = 232) and for the abundant uranium isotope (Z = 92,

A = 238) the two curves in Fig. 6 which, at any rate as regards the order of magnitude, agree with the fission cross-section obtained for both elements by deuterons of 9 MeV. For a closer comparison of the theory with the

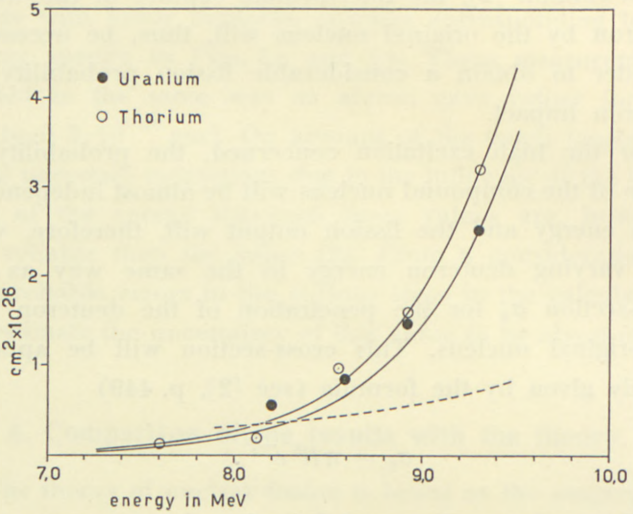

Fig. 6. The points plotted are the experimental values for the fission cross-section multiplied by a suitable factor (see text).

experiments, we have by the full and open circles indicated the experimental values from Tables II and III, respectively, the scales being in each case chosen in such a way that the point for the highest energy falls on the corresponding curve. We see that the variation of the fission output with energy for both elements approximately corresponds to formula (3). For thorium, the agreement is almost perfect; for uranium, however, the deviation from the curve is for smaller energies rather outside the limits of experimental error.

A more sensitive test of the theory can, however, be obtained from the consideration of the ratio between the

fission outputs in thorium and uranium which, for 9 MeV deuteron energy, was found to be 0,7 with an estimated uncertainty of few per cent. As regards the theoretical expectation for this ratio, it was pointed out by Bohr and Wheeler [2] that the excitation of the compound nucleus, formed by the fusion of the deuteron with the heavy nucleus, is not only sufficient for fission to occur in competition with neutron escape, but there will even be a great probability that fission may take place after the escape of a neutron from the compound nucleus. Such successive transformations have been discussed more closely by Bohr [7] who estimated the probability of fission of the compound nucleus in both steps for uranium to be nearly 1 and for thorium to be about 0,8.

In our preliminary note [4], the experimental ratio between the outputs in thorium and uranium was regarded as a confirmation of the value estimated from the theory. From the closer discussion of the experiments it appeared, however, that the difference of the cross-section for the formation of the compound system cannot be neglected in such a comparison. In fact, for 9 MeV deuteron energy, this cross-section must, as seen from the curves in Fig. 6, be expected to be about 25 % greater for thorium than for uranium. The theoretically estimated ratio between the fission outputs in thorium and uranium should, therefore, instead of 0,8 be $1{,}25 \cdot 0{,}8 = 1{,}0$, while the value found experimentally was, as mentioned above, only 0,7.

As pointed out to us by Professor Bohr, this rather great discrepancy between theory and experiment seems to indicate that a part of the fission effect in uranium, in contrast to thorium, has to be ascribed to a process in which the deuteron breaks up during the impact and only the

neutron is captured by the nucleus. In fact, if such processes, in spite of the low average excitation of the compound nucleus, contribute materially to the fission, we should expect a larger effect in uranium than in thorium. Thus, as known from experiments on neutron induced fission, the critical fission energy is, in thorium, almost 2 MeV higher than the neutron binding energy in the compound nucleus while, for the abundant uranium isotope (238), this difference is smaller than 1 MeV. Moreover, in the energy region concerned, where the cross-section of the process is less than 1 $^0/_0$ of the geometrical nuclear cross-section, it is also not excluded that the presence of the lighter rare uranium isotope (235), for which the critical fission energy is still lower, may contribute sensibly to the fission effects observed.

Indications for the correctness of such views are also obtained from a consideration of the fission effects in thorium and uranium for lower energies. Thus, while the variation of the output in thorium agrees closely with the theoretical curve in Fig. 6, the values for uranium show, as already mentioned, for energies about 8 MeV rather large deviations from the curve, just as would be expected if a part of the effect in uranium was due to processes more probable for lower energies. In Fig. 6 is, by the dotted line, roughly indicated the contribution of such processes sufficient to ensure an agreement with the theoretical expectations regarding the variation with energy of the fission effect in uranium for processes initiated by deuteron capture as well as the ratio between these effects in uranium and thorium.

The experimental material so far obtained is, however, not sufficient to settle the last questions. For this purpose,

experiments over a wider energy region will be necessary. It is planned to take up such experiments as soon as the reconstruction of the cyclotron with the object of obtaining higher energies and currents is completed.

The present investigation has been carried out at the Institute of Theoretical Physics, Copenhagen, and we wish to thank Professor NIELS BOHR for his continual interest in this work and for many helpful discussions on the subject. Our thanks are also due Mr. S. HØFFER JENSEN for valuable assistance in the experiments.

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