December 5, 1941

Preliminary Report on the Capture of Neutrons by Uranium in the Energy Region of Photo Neutrons from Radium-Beryllium Sources

By

John Marshall, Jr., and Leo Szilard

The capture of neutrons by uranium which leads to the formation of uranium 239 plays an important role from the point of view of the chain reaction in a system in which a lattice of uranium spheres is embedded in graphite. If the cross-section for this process were known as a function of the energy, then this knowledge could be utilized in determining the most favorable arrangement in this system. While one may assume that this cross-section falls off inversely proportionally to the square root of the energy of the neutron in the region between a few hundred volts and a few ten thousands of volts, the absolute value of the cross-section is not known for any value of the energy. We have for this reason attempted to determine this cross-section for photo neutrons from a radium-beryllium source. A preliminary rough measurement gives a cross-section of about

As a source of neutrons we used a beryllium block 8 cm. high having a diameter of 8 cm. and an inside bore of 3 cm. diameter. A small box containing a uranium salt, from which the natural β-active components were removed before irradiation, was placed inside the bore of the beryllium block and two grams of radium were placed next to it inside the bore. While this uranium sample was being irradiated, another identical uranium sample was

placed in a flat container below a Geiger counter and the growth of its activity was observed. After irradiation (43 minutes) the non-irradiated sample was replaced by the irradiated sample and the activity of the latter was followed. The difference in initial activity between the two samples (taken at the time when the irradiation was stopped) amounted to 0.93 expressed in units of the activity which corresponds to one hour's growth of activity of the two samples (which is due to the growth of uranium X in the sample). This value has to be multiplied by 4/3 in order to correct for infinite time of irradiation. We may write for the capture cross-section σ of uranium leading to the formation of uranium 239

$$(1) \qquad \sigma = .93 \times \frac{4}{3} \frac{\beta}{N\alpha} \frac{1}{85 \times 2.08 \times 10^{17}}$$

In this formula N is the number of neutrons emitted by the photo source, α is the geometrical factor so defined that αN gives the number of neutrons going per cm.^{2} and second through the uranium sample. β is a factor which correlates the penetrating power of the β rays of uranium 239 to the β rays of uranium X; if these two β rays were equally penetrating, the factor β would have the value 1. In reality the β rays of uranium 239 are softer, and the value of β is roughly estimated to be about 3 for the case of our particular geometry. This value is a rough estimate which will be checked by later measurements. The above given formula would hold if the total initial activity of 0.93 growth units would be due to the 24-minute period of uranium 239. In reality a fraction of about .18 of this activity is due to fission, and a corresponding correction will be applied to the cross-section given by equation (1).

In order to determine the fraction of the initial activity which is due to fission, and in order to determine the value of the factor a, the following experiments have been carried out.

(a) A box filled with 3 grams of the above-mentioned uranium salt was irradiated by a fast neutron source (one gram of radium mixed with beryllium) at 7 cm. distance for 48 minutes, and the initial activity was observed by switching over from a non-irradiated sample to the irradiated sample at the end of the irradiation. This initial activity was observed to be 1.1 growth units on a 3 gram sample placed below the counter in the same geometry as used before.

(b) The number of fissions produced in an ionization chamber coated with a thick uranium oxide layer was compared for the photo neutron source and the radium-alpha-beryllium source. We obtained 5 fissions for the photo neutron source at a distance of 11 cm. between the center of the source and the center of the spherical ionization chamber. We obtained 184 fissions per minute for the radium-beryllium mixture at a distance of 15 cm. from the center of the spherical ionization chamber.

(c) We compared the activity obtained in iodine with photo neutrons for two positions, one being the same position in which the uranium was irradiated by photo neutrons in the first mentioned experiment, and the other being 10 cm. away from the center of the photo neutron source. We found the ratio of the activities to be 13.6.

From the accessory experiments a, b and c we may conclude that about 0.16 of the initial activity is due to fission, and 0.93-0.16 = 0.77 is due to the 24-minute period.

From the accessory experiment c we may conclude that the factor a has the value of $a = 15.6 \times \frac{1}{4\pi 10^2}$. The number of neutrons emitted by the photo source per second (N) was estimated sometime ago by Anderson and Fermi who compared this photo neutron source with a fast neutron source (radium mixed with beryllium) to be about 5.8×10^6. This value is now being re-measured by B. T. Feld under the supervision of E. Fermi. With the above value and the application of the above-mentioned correction of the initial activity observed after photo neutron irradiation for fission activity, we find a cross-section for the formation of uranium 239 of about

$$\sigma = 6.15 \times 10^{-25} \text{ cm}^2$$

Since this value appeared to be exceptionally high if compared with the capture cross-sections of elements other than uranium which were found by Halban and Griffiths, on the other hand, for photo neutrons from radiothorium-deuterium and radium-beryllium, respectively. We have, therefore, also tried to determine the capture cross-section for iodine for which Griffiths has reported a value of questionable value. Again we find a very high value, about ten times as large as the value quoted by Griffiths, and in this particular case we have compared the activity of a thin sheet of iodine irradiated by photo neutrons in a fairly well defined geometry and compared this activity with the activity of uranium x and radium E standards. It should be noted that our measurement of

the uranium cross-section is not finished and, therefore, more accurate determinations are being made at present.

Should be
March 8, 1941
not 1940
(K.W. 1/17/68)

F52 (2)

420 West 116th Street
New York City
March 8, 1940

Mr. Morton Adams
Pennie, Davis, Marvin and Edmonds
165 Broadway
New York, New York

Dear Mr. Adams;

I telephoned your secretary on Friday to say that you need not file an amendment in reference to my pending patent application and that my application may be abandoned. Today I have changed my mind on the subject after having had time to study the reference cited against my application

Since the amendment has to be filed before March 11th it seemed best to write up and send off something today. I have, therefore, written and mailed a letter of which I enclose a copy.

There is perhaps a slight risk that the Examiner will be dissatisfied and that my application will be held abandoned, but this is a risk which I am willing to take.

In the circumstances it does not seem necessary for you to do aything in connection with the amendment due March 11th. If you care to send me a bill for the consultation which we had on Thursday in connection with this amendment I should be pleased to settle it at the end of the month.

Please note that it is my intention eventually to abandon this patent application and I only want to keep it alive for a while in connection with the possibility of filing a continuation in part.

Very truly yours,

L. Sz

(Leo Szillard)

December 5, 1941

Dr. Gregory Breit
National Bureau of Standards
Washington, D. C.

Dear Breit:

Many thanks for your letter of December 2. I am sorry to take your time with this correspondence. Do you think it would be possible to make the statement which you propose to attach to the report more precise, for instance by saying the following:

> "Report A-55 is, with the exception of the summary and page 21, a copy of a paper submitted for publication in the PHYSICAL REVIEW on February 16, 1940. The paper has been withheld from publication at the request of the author in the interests of national defense."

If you should find it convenient to do so, you might attach to this statement a copy of the original summary and a copy of the original page 21, but I personally have no opinion as to whether this is desirable or not.

Many thanks again.

Sincerely yours,

LS:H

Leo Szilard

Background of The Paper "Divergent Chain Reaction in Systems Composed of Uranium and Carbon" of February 14, 1940.

In May, 1939, Anderson, Fermi, and I carried out an experiment[12] in which neutrons were slowed down by water and reacted with uranium. This experiment showed that more neutrons are emitted than absorbed by uranium under the particular conditions in which the experiment was performed.

Our system contained 17 H atoms per U atom and the thermal neutron absorption of hydrogen rules out the possibility of a chain reaction at such concentrations. When we started this experiment we had hoped, however, that we might perhaps be able to draw from it the conclusions that a chain reaction could be maintained at a lower hydrogen-uranium ratio.

That the possibility of a chain reaction can not be proved in such a way had been first realized by G. Placzek. He pointed out that with increasing uranium concentrations the fraction of neutrons captured by uranium at resonanc increases so rapidly that a chain reaction might be very well impossible at any hydrogen-uranium ratio. Placzek put forward the suggestion of using helium in place of hydrogen for slowing down the neutrons. He emphasized that the absence of a thermal neutron absorption in helium would make it possible to use very high helium uranium ratios with the result that a high fraction of the neutrons would be slowed down to thermal energies without being captured by uranium at resonance.

In the experiment of Anderson, Fermi, and Szilard[12] tubes filled with uranium oxide were immersed in a water tank. Adjacent tubes were separated by a few centimeters of water in order to keep these distances within the range of thermal neutrons in this medium. For the interpretation of this experiment it was necessary to calculate the fraction of the neutrons absorbed at resonance in these uranium-oxide filled tubes. Fermi carried out such a calculation and noticed that the arrangement which we had used was more favorable with respect to resonance absorption than a homogeneous mixture of uranium oxide and water. Assuming a sharp absorption line in uranium it is indeed quite easy to see a posteriori that most of the resonance absorption will take place in a thin outside layer of the uranium oxide, whereas the inside of the cylinder, which will not appreciably contribute to the resonance neutron absorption, will still materially contribute to the useful thermal neutron absorption. Thus the use of uranium oxide layers of finite thickness has an advantage over the use of infinitely thin uranium oxide layers which would be equivalent to a homogeneous mixture of uranium oxide and water. Fermi calculated in detail the balance of

neutron absorption and emission for systems built up from alternating layers of uranium oxide and water. These calculations of Fermi showed that there was a much better chance for obtaining a chain reaction in such an inhomogeneous system of water and uranium oxide than in a homogeneous system of these two substances. For the purposes of such calculations it might be sufficient to consider only the lowest resonance absorption line of uranium but the width and intensity of this line enter into these calculations as major determining factors, and these quantities were known only within rather wide limits of experimental error. For this reason it was difficult to say whether or not a chain reaction can be maintained in the heterogeneous uranium oxide water systems which were considered by Fermi. (June, 1939)

Meanwhile it occurred to me that from the point of view of practical applications it would be very much better to use carbon in place of hydrogen for slowing down the neutrons. Led by this point of view, I became interested in this possibility in spite of the fact that carbon, on account of its larger atomic weight and smaller scattering cross-section, is very much less efficient in slowing down neutrons than hydrogen. (June, 1939)

The rate at which a nuclear chain reaction can be maintained will obviously be determined by the rate at which the heat which is developed in the reaction can be dissipated. Since carbon can withstand high temperatures it is much more efficient in this respect than water or paraffin. Led by these considerations, I made inquiries about carbon and found that it can be obtained in the form of graphite bricks, very pure, and at a moderate price. A structure composed of alternate layers of graphite bricks and uranium oxide appeared to be the simplest from a purely practical point of view and I attempted to form an estimate of the chances of a chain reaction in such a structure. As soon as this problem was seriously considered it became evident that these chances were appreciable even if one assumed that the uranium layers would absorb all the neutrons which reach them while the energy of the neutron is between 10 and 100 volts. (July, 1939)

In this respect, a system in which the neutrons are slowed down by carbon is very different from a system in which the neutrons are slowed down by water. Of the two, carbon is very much superior and this is mainly due to the fact that the range of thermal neutrons in carbon, is considerably larger than the range of the resonance neutrons of uranium in carbon, whereas the ranges of these two categories of neutrons are about equal in water.

Further simple considerations showed that the lattice of uranium spheres

embedded in graphite appeared even more favorable from the point of view of a chain reaction than the system of plane uranium layers which was initially considered. The efficiency of such a system was calculated and it was found that if small spheres of uranium metal were embedded in graphite there would be quite a good chance of obtaining a chain reaction in an experiment performed on a large but entirely practicable scale. There appeared to be an appreciable chance for success even if uranium oxide had to be used in place of uranium metal In the circumstances, the conclusion was reached that it would be better to perform an experiment on a large scale rather than to wait for measurements to be performed for the purpose of determining all the nuclear constants involved. (July 9, 1939)

Note for Memorandum:

Tentative steps were thereupon taken in this direction and among others, E. P. Wigner, and E. Teller were informed of these considerations. They shared the opinion that no time must be lost in following up this line of development and in the discussion that followed, the opinion crystallized that an attempt ought to be made to enlist the support of the Government rather than that of private industry. Dr. Wigner, in particular, urged very strongly that the Government of the United States be advised of certain possible consequences of this as well as some other lines of work connected with uranium. With this in mind, we approached Professor Albert Einstein and Dr. Alexander Sachs, and after a number of consultations, Dr. Einstein wrote a letter to the President of the United States recommending that a person or committee be appointed to act as a permanent liason between the Government and the physicists who are working on uranium. (August 2, 1939)

Einstein's letter and a memorandum which I was asked to write were submitted by Dr. Sachs to the President and Dr. Lyman J. Briggs was appointed as chairman of a Government committee. Dr. Sachs, Dr. Wigner, Dr. Teller and I were given an opportunity to explain to this committee why we believed that the work which is being done on uranium deserved the attention and the support of the Government. (October 21, 1939)

On this occasion a plea was made for the Government's support either financial or moral both for the work on uranium in general and for the work along the lines indicated in the paper in particular. It was stated that a lattice of uranium metal spheres embedded in graphite appeared to offer the greatest chance for immediate success; that about 100 tons of graphite and 10 to 20 tons of uranium would have to be used in a large scale experiment in order to produce a divergent chain reaction. A recommendation was made that steps be taken to prepare for the performance of such a large scale experiment and that methods of producing uranium metal from uranium oxide be explored. It was emphasized that before starting a large scale experiement the capture cross-section of carbon would be measured and that a few tons of graphite were required for this purpose. A memorandum summarizing these statements and recommendations was submitted to Dr. Briggs (October, 1939)

The value of the capture cross-section of carbon for thermal neutrons was not known at that time; only an upper limit for this magnitude of 0.01×10^{-24} cm^2 was published by Frisch, v. Halban, and Koch. To measure this value appeared, therefore, to be an urgent task, indeed. The usual methods for measuring small absorption cross-sections did not seem to be adequate for this purpose and so a method was devised which called for the study of the spatial distribution of the thermal neutron density in a large mass of graphite. The thermal neutron

density in graphite obeys a diffusion equation which contains the mean free path for scattering and the ratio of the capture and scattering cross-sections. Since the thermal neutrons are produced in the graphite through the slowing down of the fast neutrons emitted from the neutron source, this diffusion equation is not homogeneous. By introducing screens which are black for thermal neutrons one can obtain, however, experimental values which obey the homogeneous diffusion equation. The first experiment of this type which was originally planned (July 5, 1939) is described in the paper.

Due to various circumstances, experimentation along this line was halted between July, 1939, and March, 1940. While no information on the capture cross-section of carbon was as yet forth-coming, a increasingly optimistic view on the chances of a chain reaction in a uranium - graphite system appeared to be justified when it was realized that the neutron absorption in carbon could be materially reduced by allowing the bulk of the graphite to heat up to high temperatures. Thus having finally reached the conclusion that we may expect a divergent chain reaction in such a system, the paper was submitted in February, 1941, to the Physical Review for publication. After its acceptance, and after consultations with a number of my colleagues and Dr. Lyman J. Briggs, its publication was deferred at my request.

Aug 1941 shortened version. Attached to 4-page "Background" (K.W. 11/5/68) SBS 19

Conditions for a chain reaction

If q denotes the fraction of fast neutrons emitted by uranium which are sloved down to the thermal region and are absorbed as thermal neutrons by uranium and if μ denotes the number of fast neutrons produced on the average by uranium for one thermal neutron absorbed by uranium then obviously

(49) $$\mu q > 1$$

is the condition for the possibility of a chain reaction. If this condition is fulfilled then a divergent chain reaction can be maintained in a sufficiently large system from which only a small fraction of the neutrons emitted by the uranium within can escape across the boundary of the system without being absorbed within.

In order to be on the conservative side we shall consider as a sufficient condition for a chain reaction

(50) $$\mu q \cdot 0.9 > 1$$

From this we find using equation No. 27 for ε

(51) $$\varepsilon > 11.3$$

as a sufficient condition.

In order to see now whether a chain reaction is possible we have to calculate from our formulae the numerical value of ε. We shall do that in the following under the assumption that the energy liberated in the chain reaction will maintain the carbon at a temperature of about 900C and in order to be on the conservative side we shall assume that the temperature of the uranium spheres in which most of the energy is liberated is, in spite of efficient cooling, about the same.

Since we have at room temperature $\sigma_c < 0.01$ we shall have at 900 C. a capture cross-section of carbon half of this value. The scattering cross-section of uranium for thermal neutrons we take to be $\sigma_{sc}(u) \sim 9$. Finally, at room temperature we take $\frac{\sigma_a(u)}{\sigma_{sc}(u)} = \frac{1}{2}$ and correspondingly we take at 900 C. $\frac{\sigma_a(u)}{\sigma_{sc}(u)} = \frac{1}{4}$. For a density of graphite of 1.7 and a density of uranium of 15 we then obtain from No. 14 for $R = \rho_{cm}$

$$\varepsilon = 14$$

This being larger than the value required by No. 51 we conclude that in the circumstances we can expect a divergent chain reaction to take place in the system which we have investigated.

In reality the capture cross-section of carbon is perhaps much smaller than

the upper limit which has so far been established and consequently there is hope that conditions will be much more favorable for a chain reaction than would seem from the values so far quoted.

The amount of carbon and uranium required to reach the point of divergence at which nuclear transmutation will proceed at a rate limited only by the necessity of avoiding over-heating is essentially determined by the value of

In the following we shall calculate how the value of this expression depends on the value of the carbon capture cross-section at room temperature.

We shall take the density of graphite to 1.7; the density of uranium metal to be 15 and choose R = 8 cm. We then obtain for a capture cross-section of carbon at room temperature of 0.005 the following set of values at 900° C:

$A = 64\ cm$; $\frac{A^2}{B^2} = 90$; $\gamma = 0.666$; $\varepsilon = 27$; $g_m = 0.68$

and for $\mu = 2$ $(\mu g\, 0.9 - 1) = 0.32$

Critical Dimensions

For a large sphere of graphite which contains a large number of small spheres of uranium the critical value, l, for the radius of the graphite sphere for which the chain reaction becomes divergent may be calculated for various distributions of uranium within the graphite sphere. The treatment of this question may as well be postponed until the value of the carbon capture cross-section is known. In the meantime, a very rough approximation may give an idea of the order of magnitudes which are involved. In graphite of 1.7 density the meandistance $\tilde{\tau}$ to which a fast neutron emitted by uranium diffuses away from its point of origin until it becomes a thermal neutron and reacts with uranium or carbon is about 50cm

For $(\mu g - 1) = \frac{1}{\delta}$ we find for the critical radius $l \sim 250\ cm$.

$$l \sim \sqrt{\frac{3\tilde{\tau}^2}{\mu g - 1}} \quad \text{or} \quad l \sim \sqrt{\frac{7500}{\mu g - 1}}$$

The amount of uranium required can be calculated from equation No. 33a

$$\frac{4\pi}{3} R^3 / V = \frac{1 - g_m}{6} \frac{R^2}{B^2} \frac{1}{1 + R/B}$$

It may be kept down by choosing a smaller value for $\tilde{\tau}$ than the value corresponding to the

Note for Memorandum

The approximation used here corresponds to a treatment of the problem which was first given in a patent application that was filed in England in 1934, and was assigned to the British Admiralty. Appendix No. is a copy of pages of the corresponding American patent application which was filed in March, 1935. The mean distance $\tilde{\tau}$ in this paper has to be identified with the mean free path "a" in the appendix. The differential equation quoted in the appendix leads, if written in the notation of the present paper, to a critical radius of

$$l = \pi \tilde{\tau} \sqrt{\frac{1}{3(\mu g - 1)}}$$

It may be kept down by choosing a smaller value for R than the value corresponding to the maximum value of Σ. For R = 5 cm. and $q_\mu = 0.6$ 33a gives $\frac{4\pi}{3} R^3/v = 0.022$ corresponding to 20 tons of uranium for 100 tons of graphite.

MEASUREMENTS

In order to determine the critical dimensions and the most favorable distribution of the small uranium spheres within a large graphite sphere it is essential to have an accurate value for μ and for the capture cross-section of carbon for thermal neutrons.

In order to have an accurate value for μ it will be necessary to have a direct measurement of p for small hydrogen concentrations preferably for n = 3.

The upper limit which has been reported for the carbon cross-section by Halban, Frisch, and Koch is already so low that it would be difficult to improve upon it unless a method is used which is specifically designed to measure extremely small capture cross-sections. Such a method will be described in the following:

Let us consider a sphere of carbon and a neutron source in the center of the sphere. The thermal neutron density inside the carbon will then obey equation No. 3 only in this case Q is a function of r of which we must not assume $\frac{dQ}{dr} = 0$. Let the carbon sphere be immersed in a water tank or surrounded by paraffin wax. The thermal neutron density will then have a certain value at the surface of the sphere and inside the sphere it will be some function of r, $\rho_1(r)$. If in a second experiment the surface of the sphere in the water is covered by cadmium the thermal neutron density at the surface of the sphere is then reduced to practically zero and inside the sphere it will be another function of r, $\rho_2(r)$. The difference $\rho(r) = \rho_1 - \rho_2$ obeys the homogeneous equation

$$D \frac{d^2(r\rho)}{dr^2} - \Sigma r \rho = 0$$

which has the solution

$$\rho(r) = C \frac{e^{r/A} - e^{-r/A}}{r}$$

We can thus find A by determining the value of ρ for two values of r, for instance: r = 0 and r = r. It is

$$\frac{\rho(r)}{\rho(0)} = \frac{e^{r/A} - e^{-r/A}}{2\, r/A}$$

Or for small values of r/A

$$\frac{\rho(r)}{\rho(0)} \cong 1 + \frac{1}{6}\left(\frac{r}{A}\right)^2$$

Using a sphere of graphite of fifty to seventy centimeters of radius it should be possible to measure the range A with sufficient accuracy. There is a limit to using very large spheres which arises out of the fact that for a very large sphere most of the neutrons emitted in the center of the sphere will be slowed down to the thermal region within the carbon and the thermal neutron density near the surface of the carbon may become very low. If that happens, then the difference $\rho = \rho_1 - \rho_2$ will become small and will therefore set a limit to the accuracy of the measurement.

Stabilizing the chain reaction

Soon after the discovery of an abundant neutron emission from uranium the question of stabilizing such a reaction was a subject of discussion (14,15) but the situation as we see it appears to be rather different.

If a chain reaction could be maintained in a homogeneous mixture of water and uranium or carbon and uranium it would have a certain natural stability in the sense that with rising temperature there would be a decrease in the neutron production. The reason for this is the fact that the absorption of both uranium and hydrogen obey the 1/v law in the thermal region and thus at higher temperatures the range of thermal neutrons in the mixture is larger. Correspondingly, at higher temperatures a larger fraction of the thermal neutrons will escape across the boundary of the mixture without having reacted with the uranium in the mixture. This natural stability could even be enhanced by having bodies of strong thermal neutron absorbers inserted in the mixture. Fairly thin sheets of such absorbers as boron, for instance, are practically "black" for thermal neutrons and any strong thermal neutron absorber would stabilize equally well.

A system, on the other hand, in which uranium bodies which are almost "black" for thermal neutrons are embedded in carbon, like the system which we have considered in great detail in the present paper, has no such stability. This is due to the fact that with rising temperature the capture cross-section of the carbon decreases whereas the absorption by the uranium spheres remains almost unchanged. Accordingly, at higher temperatures, a larger fraction of the thermal neutrons is absorbed by uranium and a smaller fraction is absorbed by carbon and this leads to an increase in q and thermal instability.

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E-1885 February 17, 1941

Dr. Leo Szilard,
Kings Crown Hotel,
420 West 116th Street,
New York, N.Y.

Re: Apparatus for Nuclear
Transmutation, Serial No. 263,017
Filed March 20, 1939

Dear Dr. Szilard:

This is to remind you that some action must be taken before March 11th in the above entitled application or it will be held abandoned.

Very truly yours,

RMadams

Backpocket 9

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420 West 116th Street
New York City
March 8, 1941

Hon. Commissioner of Patents
Div.56 Room 4725
Washington, D. C.

Sir:

In response to your communication of September 11th addressed to Pennie, Davis, Marvin and Edmonds, 165 Broadway, New York City, referring to my application Serial No. 263,017, filed March 20, 1939, I beg to state the following:

The patent of Fermi et al. which you cite does not appear to be pertinent since it is not possible to maintain a chain reaction in beryllium. Moreover, the priority date of Fermi el al. is October 26, 1934, whereas the conception of the chain reaction is contained in my patent application of 1935 which has a priority of English patent applications which were filed before October, 1934

The first two lines of the specification may be amended to say that my application is "in part a continuation" of my earlier application, if such an amendment is acceptable to the examiner.

I submit that my patent application describes the method which makes it possible for persons skilled in the art to determine the proper thickness in various layers. I further submit that your reference to Egloff 464 O.G. 3 does not apply since no chain reaction can be set up by following the directions given in the specification of Fermi et al.

Very truly yours,

(Leo Szilard)

U. S. DEPARTMENT OF COMMERCE

NATIONAL BUREAU OF STANDARDS

WASHINGTON

ADDRESS REPLY TO
NATIONAL BUREAU OF STANDARDS

IN YOUR REPLY
REFER TO FILE

GB:KSV December 2, 1941. D

Dr. L. Szilard,
Department of Physics,
Columbia University,
New York, N. Y.

Dear Szilard:

In your letter of December 1, you suggest that I date your report A-55. According to the Physical Review records, your paper has been submitted on February 16, 1940, which corresponds to your date of February 14, 1940, if account is taken of the time it takes for a manuscript to reach Minneapolis from New York. Report A-55 does not appear to be identical with the manuscript submitted to the Physical Review. The summary in A-55 is longer and speaks of 30 tons of uranium instead of 10 tons in the Phys. Rev. paper. Also p.21 of A-55 does not correspond to the analogous page in the Phys. Rev. manuscript.

In view of these differences I wonder whether it would be satisfactory to you if I were to send out the following statement to be attached to the report:

> "Report A-55 is substantially the same and in most parts identical with a paper submitted for publication in the Physical Review on February 16, 1940, and which was generously withheld from publication at the request of the author in the interests of national defense."

Sincerely yours,

Gregory Breit

Gregory Breit.

This document contains information affecting the national defense of the United States within the meaning of the Espionage Act, U.S.C. 50: 31 and ff. Its transmission or the revelation of its contents in any manner to an unauthorized person is prohibited by law.

November 14, 1941

PRELIMINARY REPORT ON FISSION CAUSED BY FISSION NEUTRONS

By

John Marshall, Jr., and Leo Szilard

Columbia University

It appeared to be ~~important~~ of interest to determine whether fission can be caused in uranium by fission neutrons and if so to learn something about the cross-section of this process.

This process may play ~~an important~~ a role in a chain reaction in a system in which spheres of uranium are imbedded in a large mass of graphite. In such a system thermal neutrons diffuse from the graphite into the uranium sphere and lead to the emission of fast fission neutrons. The fast neutrons thus produced will cause a certain number of fission processes in the same uranium sphere from which they originate and thus produce secondary fast fission neutrons. Some of these secondary neutrons will again cause fission in the uranium sphere from which they originate and lead to tertiary fission neutrons, etc. This process might increase by perhaps ~~as much as~~ a factor of $\beta = 1.25$ the number of fast neutrons emitted by uranium per thermal neutron absorbed by uranium. The magnitude of the factor β depends primarily upon certain nuclear properties of uranium and in the first approximation on the product

$\sigma_f(\nu-1)$

where σ_f is the cross-section of uranium for fast fission neutrons and ν is the number of neutrons emitted per fission process. The magnitude of the factor β also depends on the cross-section of uranium for inelastic collisions which slow down fast fission neutrons below the fission threshold of U238. This slowing-down phenomenon of uranium has been studied by Szilard and Zinn and will form the subject of another report.

Secondarily, the magnitude of the factor β depends on the size and density of the uranium sphere and also on the presence in the uranium sphere of oxygen or other elements which slow down the neutrons by means of elastic collisions. It may very well be that, due to the process which is the subject of this report, there may be a ten per cent increase in the number of fast neutrons emitted by a uranium sphere per thermal neutron absorbed by the uranium sphere if we change over from uranium oxide at a density of about 4 to 6 gm./cc. to uranium metal of density 18 to 20 gm./cc. The experiment which we performed has the purpose of measuring the quantity

$$\sigma_f \nu$$

The principle of the experiment is illustrated in the enclosed diagram which shows the experimental arrangement. In this diagram Be is a beryllium block which serves as a source of photo neutrons and which is placed in the axis of a cylindrical paraffin block. Ra represents about two grams of radium which are placed in the center of the beryllium block. A cadmium diaphragm A leaves a circular opening free through which thermal neutrons can emerge from the paraffin and can enter in the

absence of the cadmium screen B (at H). ~~These thermal neutrons can enter~~ the cylindrical box U which contains uranium in the amount of about 25 grams per square cm. A spherical ionization chamber D which is coated with a thick layer of uranium and has a uranium-coated surface of about 800 cm. square is used to record fission which takes place in the uranium coating of this chamber. This fission chamber is protected from the action of thermal neutrons by thick ~~layers~~ walls of boron carbide, one of the walls and E is a boron carbide screen which can be removed if it is desired to admit thermal neutrons to the fission chamber.

The basic experiment which we performed is the following: With the boron screen E and the cadmium screen placed at H ~~in position~~, the fission chamber registers a background of about .5 fissions per minute. Thermal neutrons which come from the circular opening C in the center of the cadmium diaphragm A are prevented by the cadmium screen at H from reaching the uranium during this "control" experiment. If the cadmium screen is now brought from the position H into the position K, thermal neutrons are admitted to the uranium, will cause fission in the uranium and will lead to the emission of fast fission neutrons. A considerable fraction of these neutrons passes through the uranium filled box U ~~whole thickness of the uranium layer~~ and through the boron screen E and will cause fission in the ~~fission~~ ionization chamber D. By changing the position of the cadmium screen from position H to position K, we obtained a fission count which was more than double of the background count, the difference corresponding to about .7 counts per minute. A preliminary estimate of the quantities involved

leads to the conclusion that the observed effect corresponds to a value of about

$$\sigma_f \nu \cong 1.3 \times 10^{-24} \text{ cm}^2$$

Taking for ν the value of 2.6 reported by Zinn and Szilard,* we obtain

$$\sigma_f = 0.5 \times 10^{-24}; \quad \sigma_f(\nu - 1) = 0.8 \times 10^{-24}$$

It has to be emphasied that this is a preliminary result, and that ~~the experiments will be repeated, and that~~ an attempt will be made to determine the value more accurately than it was possible to do in the first rough experiment. In the following we describe two methods which were used in interpreting the observed effect. The following designations will be used:

F_f is the fission count which we obtain from the fast neutrons when we remove the cadmium screen B at N

F_{th} is the fission count which we obtain in the chamber (in the absence of the uranium, the cadmium screen at B or at K and the boron screen E) due to the action of thermal neutrons which emerge from the window in the center of cadmium diaphragm A, and which can be cut off by placing the cadmium screen in the position B.

N is the number of thermal neutrons which emerge from the circular window in the center of the cadmium diaphragm A.

α is the fraction of these thermal neutrons which are absorbed by the uranium in the box U.

$1 - \varepsilon$ is the fraction of the fast neutrons which are emitted by uranium and which are prevented from reaching the ionization chamber by scattering either in uranium in the box U by ~~or by~~ ~~B or E~~ in the boron screen E.

* Zinn and Szilard, Phys. Rev. 1939

$N b/2\pi$ is the number of thermal neutrons emitted (per unit solid angle) from the paraffin wall in the circular opening G in the center of the cadmium diaphragm A in the forward direction (towards the chamber D) per unit solid angle.

U is the number of uranium atoms which are exposed to the thermal neutrons.

ρ is the thermal neutron density in the uranium.

r is the distance of a volume element of uranium in the Box U from a surface element of the uranium layer in the spherical ionization chamber.

σ_{th} (fission) is the fission cross-section of uranium for thermal neutrons.

σ_{th} is the total absorption cross-section of uranium for thermal neutrons.

ρ_0 is a thermal neutron density at the location of the uranium surface of the spherical ionization chamber in the absence of the uranium, Box U, the cadmium screens H B and K C and the boron screen E.

The main quantities which we measure are F_{th} and F_f. One of the methods employed makes use of the fact that we may write:

$$F_{th} = \frac{N b}{2\pi \bar{r}_{th}} \sigma_{th}(\text{fission}) n^{th}$$

$$F_f = \frac{N}{4\pi \bar{r}_f^2} \alpha \varepsilon \frac{\sigma^{th}(\text{fission})}{\sigma_{th}} (\sigma \rho V) n^f$$

or

$$\sigma \rho V = \frac{F_f}{F_{th}} \sigma_{th} \frac{2b}{\alpha \varepsilon} \frac{n^{th}}{n^f} \frac{\bar{r}_f^2}{\bar{r}_{th}^2} \qquad (1)$$

In these formulae $N_{th}(U)$ is the number of uranium atoms which is effective in producing an impulse in the chamber if excited by thermal neutrons, and $N_f(U)$ is the number of uranium atoms which is effective in producing an impulse in the chamber if excited by fast fission neutrons. We have actually assumed that these two numbers are equal, i.e., that the range of the fission particles is about the same when the fission is due to thermal neutrons as it is when the fission is due to fast fission neutrons.

~~From these two equations,~~ we obtain:

Our experiments gave for F_{th} a value of 43 counts per minute (cadmium difference), and for F_f we obtained from 25 fifteen-minute readings with the cadmium screen at the position H and K, ~~respectively~~, a value of

$F_f = 0.77$ count/min

above a background count of .6 per minute.

α, the fraction of thermal neutrons which are absorbed by the uranium in the box, was estimated to be about 0.5 The uranium box contains about 27.7 gms./sq.cm. of uranium carbide or about 25.2 gms./sq.cm. of uranium. Neglecting scattering of the thermal neutrons, of the thermal neutrons passing through the uranium box parallel to the axis of the arrangement, a fraction of .31 would be absorbed, and a fraction of .687 would be transmitted by the uranium box. By measuring the thermal neutron density with a vanadium indicator in front and right behind the uranium box, we find that the thermal neutron density drops by a factor of 8, ~~in our arrangement.~~ Part of this drop is due to the scattering of thermal neutrons

and part of it is due to oblique passage

in the uranium box, and a very rough estimate leads us to believe that actually about .5 of the neutrons entering the uranium box are absorbed.

For b we take the value of

$$b = 2.53$$

which corresponds to ~~distribution of~~ an angular distribution of

$$\cos\varphi + \sqrt{3} \times \cos^2\varphi$$

~~for the spatial~~ angular ~~distribution~~ of thermal neutrons leaving the paraffin window G inside the cadmium diaphragm A.

For ε we took the value of .75 by estimating the effect of the scattering in the boron carbide screen E containing 3 gms./sq.cm. of boron carbide and the effect of scattering of the uranium carbide in the uranium box.

For $\frac{\overline{r^2}}{r\mu}$ we took the value of $\left(\frac{13}{14}\right)^2 = 0.87$

For σ^{μ} we took the value of 5.9×10^{-24} sq.cm.

With these values we obtained from (1)

$$\sigma_f V = 1.24$$

The second method employed makes use of the fact that we may write

(2) $$\sigma_f V = 4\pi \rho_0 \overline{\left(\frac{r^2}{\rho}\right)} \frac{1}{u} \frac{1}{\varepsilon} \frac{F^f}{F^{\mu}} \frac{n^{\mu}}{n^t}$$

We estimated ρ inside the uranium, and ρ_0, the thermal neutron density in the space occupied by the ionization chamber D by observing the activity of a vanadium foil in front

of the window G on both sides of the uranium box and also in the absence of the uranium box, the cadmium screens at H or K and the boron screen E with a vanadium foil taking the place of the ionization chamber D. Assuming an exponential fall of the thermal neutron density inside the uranium box, we then find for $4\pi \rho_0 \left(\frac{r^2}{\rho^2}\right) = (4\pi r_1^2)\frac{\rho_0}{\rho} = 250$

For the number of uranium atoms in front of the window G we take the number of U = 4.63×10^{24} corresponding to 1830 gms. of uranium. For ε we again take the value of 0.75

With these values we obtain from (2)

(2)a) $\eta f V = 1.28$