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# **Nuclear Fission**

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#### **INTRODUCTION**

 $\rm LTHOUGH$  less than a year has passed since the discovery by Hahn and Strassman that the capture of neutrons by uranium nuclei may lead to their disruption to form lighter nuclei, nearly one hundred papers on this subject have already appeared. This number does not include the many older papers written before



the true nature of the process was understood. Many questions about it are still to be answered and extensive work is still in progress. Nevertheless, it now seems that its principal features are clear, both experimentally and theoretically, and that the questions yet to be answered can at least be formulated with reasonable assurance. Because of the large number of papers and the special nature of most of them it seems worth while to review the results obtained to date in a single article. Part I is meant to do this in a general way for readers who are not themselves doing research in this field and do not wish completeness of detail. In Part II some aspects of the subject are considered more closely. It is intended that the references to the literature be sufficiently complete that the article may serve as an analytical reference guide. All papers received in Princeton up to December 6, 1939 have been included.

# PART I

# 1. The apparent discovery of transuranic elements

The first of the experiments to be considered were made in 1934 not long after the discovery of the neutron. Fermi (34Fe2)\* realized that the bombardment of uranium by neutrons might be expected to lead to the production of atoms of atomic number 93, and perhaps to ones of even greater atomic number by successive beta-disintegrations. He and his collaborators had found that nearly all elements could be activated by bombardment with neutrons. The nuclei resulting from the capture of the neutrons were unstable and reverted to stability by the ejection of negative beta-particles. This process would produce nuclei of an atomic number increased by unity over the original one. With thorium they found two beta-activities; the half-life of one was less than 1 min., of the other  $\sim$  15 min. (34Fe4). With uranium they found four halflives of  $10$  sec.,  $40$  sec.,  $13$  min., and  $90$  min. (34Fe4) with some indication of a still longer one (34Fe2, 35Ag1). Since there are but three known isotopes of U the large number of half-lives showed the occurrence of some unusual process. It was reasonable to expect that perhaps one of these half-lives should correspond to an active element of atomic number 93 formed from an active U. Element 93 would be expected to be a chemical homolog of  $_{25}Mn$ ,  $_{43}Ma$ , and  $_{75}Re$ . Accordingly, a salt of Mn was added to an irradiated solution of a salt of U and then

precipitated as  $MnO<sub>2</sub>$ . About 15 percent of the 13-min. and 90-min. activities were brought down in this precipitate, whereas none of the activities of  $_{92}U$ ,  $_{90}UX_1(Th)$ , or  $_{91}UX_2(Pa)$  were to be detected in it. Further experiments (34Fe2, 35Ag1) showed that in the presence of Ba and La neither added  $_{88}MsTh_1(Ra)$ , nor  $_{89}MsTh<sub>2</sub>(Ac)$  were thus precipitated. Since element 86, Rn, would not remain in the solution or be precipitable from it and 87, EkaCs, also would not thus be precipitated it was clear that the activities were not ascribable to isotopes of any of the elements from radon to uranium inclusive (86-92). Whatever the process of activation of the uranium was, it was clearly not mere simple capture of a neutron with reversion to stability by emission of one beta-particle, neither was it an  $(n, p)$  or an  $(n, \alpha)$  process as found with lighter atoms. The only remaining reasonable hypothesis was the one adopted, that the activities in question should be attributed to transuranic elements having atomic numbers of 93 or higher, produced by beta-emission from U nuclei after the capture of neutrons.

This apparent discovery of transuranic elements was naturally of great interest to chemists. Noddack (34No0) criticized Fermi's conclusions on the ground that a great many elements are precipitated with  $MnO<sub>2</sub>$ . Many of them,  $_{84}Po$ included, come down almost quantitatively. She suggested that it might be that the bombarded nuclei split to form elements of lower atomic number, so that certainty of proof of the existence of transuranic elements could be had only by more elaborate tests which would exclude all known elements. If this early suggestion of what has turned out to be the correct explanation was anything more than speculation it is regrettable that the reasons for its being considered plausible were not more fully developed. It seems to have been offered more by way of pointing out a lack of rigor in the argument for the existence of element 93 than as a serious explanation of the observations. However that may be, apparently it was rejected as an improbable alternative wherever considered. It seems to have had no inHuence on the subsequent course of events.

Von Grosse and Agruss (35Gr3), using  $_{91}Pa$ , found that it was precipitated with  $MnO<sub>2</sub>$ , and

<sup>~</sup> The references are listed according to year at the end of the article. The first two numbers of the reference indicate the year (1934), the abbreviation of the author's name and the number identifying the paper are given next.

with rhenium sulphide in the same way as were the apparent transuranic substances. This was contrary to Fermi's result obtained with  $_{91}UX_2$ . They suggested that the  $UX_2$  atoms might have been formed and remained as activated atoms which did not behave in the same way as the ordinary atoms of element 91. They concluded that the 13-min. activity might possibly be attributed to a new isotope of element 91, but suggested trying a precipitate with zirconium phosphate which would surely precipitate Pa without affecting element 93. This experiment was performed by d'Agostino and Segrè  $(35Ag1)$ with results which eliminated Pa as a possibility. In the meantime the discussion had had the result of interesting Hahn and Meitner in the question.

Their first papers (35Ha4) dealt principally with this problem of the possible assignment of the 13-min. and 90-min. activities to isotopes of  $_{91}$ Pa. They added potassium perrhenate and platinum chloride to a solution of an irradiated U compound, and then precipitated with NaOH. This should have removed quantitatively all of elements 90, 91, and 92 but should not have precipitated any elements of atomic numbers greater than 92, if such elements have the expected chemical properties. The filtrate was weakly acidified with HC1, and then the Pt was brought down with  $H_2S$ ; after stronger acidification the Re was also precipitated as a sulphide. The 13-min. and 90-min. substances were found in the platinum precipitate, thus . howing that they are not attributable to isotopes of elements 90, 91., or 92 and that they are somewhat more like Pt than like Re. This precipitation of sulphides with Pt as a carrier became the standard method for isolating the "transuranic" elements.

Finally, almost perfect certainty that the 13 min. and 90-min. substances were not attributable to element number 91 was reached by experiments with irradiated U to which  $_{91}UZ$  had been added. By various precipitations the 13 min. and 90-min. substances were completely separated from the UZ. Consideration of all of the results showed that the two active substances could not be isotopic with any element from  $_{80}Hg$  to  $_{92}U$  inclusive, with the possible exception of 85EkaI.

#### 2. Early experiments with thorium

The half-lives first reported by Fermi et al. (34Fe4) were later revised to 1 min. and 24 min. (35Ag1) and it was shown that the 24-min. activity itself was attributable to an isotope of Th. Experiments with Th were of great interest because of the prospect of producing substances of the hitherto missing  $4n+1$  family of radioactive bodies. The mass numbers of all of the Th series are representable by  $4n$ , *n* being an integer; those of the U series by  $4n+2$ ; those of the Ac series by  $4n+3$ . The capture of a neutron by Th<sup>232</sup> should thus produce Th<sup>233</sup>, a member of the missing  $4n+1$  family. Hahn and Meitner stressed this aspect of the matter (35Ha0). They found activity giving half-lives of 1 min., 11—<sup>12</sup> min. , and 30 min. Only the 30-min. one was enhanced by the use of paraffin to produce slow neutrons. It had the chemical properties of Th and apparently was the same as the substance for which Fermi et al. had found a halflife of 24 min. They believed the 11—12-min. substance to be a daughter of the 1-min. one and suggested that the latter was  $_{88}Ra^{229}$  formed from  $_{90}Th^{232}$  by an  $(n, \alpha)$  process.

Curie, von Halban, and Preiswerk (35Cu1) found activity analyzable to give five half-lives; —<sup>1</sup> min. , 2.<sup>5</sup> min. , <sup>25</sup> min. , 3.<sup>5</sup> hr. In agreement with the others they found the 25-min. substance to be an active Th, and enhanced by the use of paraffin. It is undoubtedly Th<sup>233</sup> formed by the capture of slow neutrons. The 2.5-min. substance seemed to have the chemical properties of  $_{91}Pa$ , and to be descended from the  $25$ -min. Th $^{233}$ . (Meitner, Strassmann and Hahn (38Me2) were later unable to find this 2.5-min. Pa.) The 1-min. substance was precipitable with Ba, and was accordingly ascribed to ssRa. Both the 12-min. and 3.5-hr. substances had the chemical properties of La, and were attributed to Ac. It seemed that the 3.5-hr. Ac, rather than the 12 min. one, was the daughter of the 1-min. Ra. They remarked on the peculiarity of the appearance of Ra formed by an  $(n, \alpha)$  process. This does not occur for other heavy nuclei because of the large potential barrier. They also pointed out that if the  $4n+1$  chain exists in nature the parent of Th $^{233}$  would most probably be U<sup>237</sup>. An attempt to find such Th in U was unsuccessful. If present at all  $U^{237}$  must have an abundance of less than one part in ten thousand.

# 3. Further work on the apparent transuranic elements

During the years 1936 and 1937 Hahn, Meitner, and Strassmann were active in extending the work on the apparent transuranic elements as evidenced by the numerous references to papers by them in those years. A somewhat detailed account of the early work has been given here in the endeavor to make it clear why the hypothesis of the production of transuranic elements seemed so eminently reasonable. It is not desirable to continue the account in the same detailed way but it is worth while to summarize the conclusions reached by Meitner, Hahn, and Strassmann (37Me5). After an extensive series of experiments with different times of irradiation, the use of fast and slow neutrons, and a great variety of chemical tests they concluded that the neutrons produced three diferent active isotopes of U which decayed by successive beta-disintegration according to the following scheme.

(1) 
$$
_{92}U+n \rightarrow_{92}U(10 \text{ sec.}) \rightarrow_{93}EkaRe(2.2 \text{ min.})
$$
  
\n $\rightarrow_{94}Ekao(s(59 \text{ min.}) \rightarrow_{95}Ekalr(66 \text{ hr.})$   
\n $\rightarrow_{96}Ekap(t(2.5 \text{ hr.}) \rightarrow_{97}Ekabu(?) \rightarrow$ 

(2) 
$$
_{92}U+n \rightarrow_{92} U(40 \text{ sec.}) \rightarrow_{93} EkaRe(16 \text{ min.})
$$
  
 $\rightarrow_{94} EkaOs(5.7 \text{ hr.}) \rightarrow_{95} EkaIr(?) \rightarrow$ 

(3)  $_{92}U+n\rightarrow_{92}U(23 \text{ min.})\rightarrow_{93}Ek_2Re(?)\rightarrow$ 

It may be noted that the 13-min. and 90-min. half-lives of the early work have been revised to 16 min. and 59 min. That the 23-min. active body is an isotope of U was proved beyond a doubt by a reaction characteristic of U, its precipitation as uranyl sodium acetate. It was found to be produced by slow neutrons only, in a typical process of resonance capture. Experiments made with absorbers of boron in the usual way showed the energy of the resonance neutrons to be  $25\pm10$  volts (assuming only a single level to be effective in the capture). By the use of Cd it was shown that the thermal neutrons were responsible for only about 5 percent of this activity. Upon using U both as an absorber and detector an absorption coefficient of  $3 \text{ cm}^2/\text{g}$  was found for the resonance neutrons. This large absorption was compatible only with capture by nuclei of the abundant isotope, U<sup>238</sup>. To attribute it to  $U^{235}$  or  $U^{234}$  would require them to have cross sections for capture larger than those theoretically possible  $(\lambda^2/4\pi)$ . The U of a halflife of 23 min. is thus  $U^{239}$ , produced by resonance capture of neutrons, as are many of the active isotopes of the lighter elements.

The assignment of the 10-sec. and 40-sec. activities to U was not made on such a sure basis. The necessary chemical work is extremely difficult with active bodies which disappear so rapidly. Nevertheless, the U precipitate did show evidence of such a short-lived substance. Further, the yields of the two short-lived active substances obtained by varying the time of irradiation indicated that they preceded rather than followed any of the substances of the longer half-lives. The chemical properties of the other substances seemed to fix their atomic numbers as 93 and higher so that the preceding 10-sec. and 40-sec. substances were attributable to U.

All of the active bodies of sequences (1) and (2) were produced both by fast neutrons from  $(Ra+Be)$ , and also by thermal neutrons, but no evidence was found for their production in the resonance absorption. To a first approximation the probabilities for processes (1) and (2) were equal, and the relative quantities of all of the active substances formed were the same whether they had been produced by thermal neutrons or by fast neutrons.

Since the beta-rays emitted by the 16-min. substance were of about the same penetrating power as those of  $UX_2(\sim 3 \text{ Mev})$  it was possible to make a measurement of the yield. From this the cross section of U for the production of the 16-min. substance by fast neutrons was found 16-min. substance by fast neutrons was found<br>to be  $1.6 \times 10^{-25}$  cm<sup>2</sup>, if one assumed U<sup>238</sup> to be the isotope involved. Here again, the assumption of the 139 times less abundant  $U^{235}$  or even rarer  $U^{234}$  would give fantastically large cross sections, not possible theoretically.

The complete discussion of the chemical properties of the active bodies was given in another paper (37Ha5). Detailed comparisons of these with the chemical properties of the supposedly homologous elements are to be found in it. All of these activities attributed to transuranic elements were found in a precipitate of platinum sulphide produced by  $H_2S$  in an acidified solution of an irradiated U compound. One more substance of the group having a half-life of 60 days and of uncertain genetic relationship to the others, was found later (38Ha8). The beta-rays emitted by some of these were studied with a Wilson cloud chamber by Meitner (37Me2).

Although both the chemical and physical results seemed to fit well enough with the scheme of disintegrations given above, it raised many questions. How was one to understand the formation of three isomeric U nuclei of mass 239, two produced by either thermal or fast neutrons, the third by resonance capture? What was the nature of the isomery that it should be inheritable so as to give the isomeric chains of disintegrations? How should the addition of but one neutron to the very stable  $U^{238}$  nucleus produce such great instability that the successive emission of five beta-particles was necessary in order to overcome it?

# 4. The rare-earth-like substance of 3.5-hr. halflife

While Hahn, Meitner, and Strassmann were doing the later work described above Curie and Savitch (37Cu1, 38Cu4) were finding other interesting results with a modified technique. They measured the activity without chemical separation, using an absorbing screen of Cu to remove the beta-rays of  $UX_2$  and any others of energy less than  $\sim$ 2 Mev. The activity so measured showed half-lives of 3.5 hr., 16 min., 2 min, and  $\sim$  40 sec. Similar experiments performed with precipitated sulphides showed the presence of the 16-min. substance but not of the 3.5-hr. one. The nearly equal activities of the two substances were found to be changed in the same proportion by using different mixtures of slow and fast neutrons in their production.

Chemical tests showed the new 3.5-hr. substance to be precipitated with La as a carrier. This suggested that it might be  $_{89}$ Ac, but fractional precipitation of oxalates from a solution containing both La and Ac showed the 3.5-hr. body to be more similar to La. It did not concentrate in the end precipitates as the Ac did. In summarizing this work Curie and Savitch

wrote "Dans l'ensemble, les propriétés de R3.5 hr. sont celles du lanthane, dont il semble jusqu'ici qu'on ne puisse le séparer que par le fractionnement." It is clear that they had come very close to being the discoverers of fission, for we now know that the 3.5-hr. body actually is an isotope of lanthanum. Their qualification concerning separation only by fractionation was based on an experiment in which a salt of La containing some of the  $3.5$ -hr. active substance was partially precipitated by NH<sub>3</sub>. The experiment indicated that the 3.5-hr. stuff tended to concentrate in the portion first precipitated. This result prevented Curie and Savitch from being confronted with perfect chemical identity of La and  $R_{3.5 \text{ hr}}$ . It may be that the presence of the recently discovered active yttrium, also a fission product and also of a half-life of 3.5 hr. (39Li1), was responsible for the observed fractionation.

By measuring the activity produced after different times of irradiation Curie and Savitch showed that the 3.5-hr. body was not descended from any active substance having a half-life of more than 0.5 hr. Assuming that it was another transuranic substance they stressed the difficulty of finding a place for an element having chemical properties like La in the region of the periodic table somewhat beyond U. Thus was added one more puzzle to be solved.

# 5. Apparent beta-active radium from uranium and thorium

Although the experiments of Curie and Savitch did not lead immediately to recognition of the true nature of the 3.5-hr. body, they did nevertheless play an important role in leading to the discovery of fission. Hahn and Strassmann (38Ha14) repeated those experiments and then went on to find that by using both Ba and La as carriers they could precipitate with the Ba active substances having half-lives of 25 min. , 110 min. , and several days. From these grew daughter substances, precipitable with La, having half-lives of  $40$  min.,  $4$  hr., and  $60$  hr., respectively. The former were attributed to isomeric  $_{88}Ra^{231}$  nuclei, and the latter to isomeric  $_{89}Ac^{231}$ nuclei. Here again one apparently had to do with an inheritable threefold isomery, the isomeric nuclei arising by the rapid ejection of two

alpha-particles from the excited  $_{92}U^{239}$  nuclei formed by capture of neutrons. This ejection of two alpha-particles was also very difficult to understand.

The threefold isomery, however, had already appeared in the apparent transuranic elements, and also in work with Th which preceded this latest work with U. Meitner, Strassmann, and Hahn (38Me2) confirmed the earlier work in finding a 26-min. active Th produced by slow neutrons, but found six active substances produced by fast ones. Three of them which were precipitable with Ba were taken to be isomers of Ra<sup>229</sup>. They had half-lives of  $\lt 1$  min., 15 min., and  $\sim$ 4 hr. Apparently they had been formed from Th<sup>232</sup> by an  $(n, \alpha)$  process. Since there is but one reasonably stable isotope of Th, Th<sup>232</sup>, there seemed to be no question as to the necessity of assigning all three active substances to  $Ra^{229}$ , giving another instance of threefold isomery. This isomery was also inheritable, for the three Ra's produced three daughter substances of half-lives of  $\sim$ 18 min.,  $\sim$ 3.5 hr., and 20–30 hr. These were precipitable with La, and were naturally attributed to Ac<sup>229</sup>. A long-lived Ac obtained from Th had already been reported by Rona and Neuninger (36Ro7).

#### 6. Attempts to find alpha-particles

In one of the early papers on Th (35Ha0) it was suggested that the 1-min. activity produced by fast neutrons could be attributed to an active Ra produced in an  $(n, \alpha)$  reaction. Braun, Preiswerk and Scherrer (37Br5) endeavored to detect these alpha-particles by a suitably arranged ionization chamber connected to an amplifier. They reported the finding of alphaparticles of an energy somewhat greater than 9 Mev. It is not clear why they did not find the very large bursts of ionization produced by fission fragments. Experiments of the same sort were later performed by von Droste (38Dr1), using both Th and U. He found no alphaparticles with U and only an uncertain few with Th. He used samples of U and Th covered with thin foils so as to eliminate the natural alphaparticles. Had he followed the simpler but less reasonable procedure of working with uncovered samples he, also, would probably have found the huge pulses from fission.

# 7. Barium from uranium. The discovery of fission

The apparent  $(n, 2\alpha)$  process of production of Ra from U was indeed a peculiar one, especially since it was found to be produced by thermal neutrons as well as by fast ones. In order to be entirely sure of the chemical evidence Hahn and Strassmann (39Ha1) carried out an elaborate series of tests to prove rigorously that the active atoms in question were chemically different from U, Pa, Th, Ac and all of the "transuranic" elements. In the course of these experiments they found evidence for a fourth Ra having a half-life of less than one minute, and revised the other half-lives to get Ra I,  $<$ 1 min.; Ra II, 14 $\pm$ 2 min.; Ra III, 86 $\pm$ 6 min., Ra IV, 250-300 hr. From these developed four active varieties of apparent Ac (La carrier); Ac I,  $\sim$  30 min.; Ac II,  $\sim$  2.5 hr.; Ac III, several days; Ac IV,  $<$  40 hr.

The active "Ra" atoms in question were so similar to Ba chemically that Ra and Ba itself were the only two elements to which they might be ascribed. In order to make sure beyond all doubt as to which of the two was correct special experiments for distinguishing between them were undertaken. Some of the separated "Ra IV" from irradiated U was added to a solution containing Ba and a small amount of ThX or  $MsTh_1$  (isotopes of Ra). After performing fractional precipitations and crystallizations of the kind used for separating Ra from Ba, they found the activity of the "Ra IV" to be uniformly distributed among successive samples, whereas the ThX or  $MsTh<sub>2</sub>$  concentrated in the usual way. Thus, contrary to expectation, it was found that "Ra IV" was an isotope of Ba rather than of Ra. As Hahn and Strassmann expressed it, "Als Chemiker müssten wir eigentlich sagen, bei den neuen Körpern handelt es nicht um Radium, sondern um Barium; denn andere Elements als Radium oder Barium kommen nicht in Frage. "

Having found that "Ra IV" was really Ba, Ba IV, it was reasonable to infer that the other three active bodies were also isotopes of Ba, as implied in Hahn and Strassmann's conclusion quoted above. If so, the daughter "Ac's" should really be isotopes of La. Accordingly, some of the "Ac II" ( $\sim$ 2.5 hr.) was added to a solution containing La and  $MsTh<sub>2</sub>$ , the latter being an

isotope of Ac. The activity of the  $MsTh<sub>2</sub>$  tended to concentrate in oxalate precipitates but not that of the "Ac II." The latter was like La, and not like Ac, as Curie and Savitch had found with their 3.5-hr. substance. Here, in addition to the chemical similarity to La, there was the strong presumption of descent from active Ba, so that there could remain little doubt that "Ac II" was really La rather than Ac.

These results led Hahn and Strassmann to suggest that the transuranic elements might prove to be isotopes of Ma, Ru, etc. The other possibility of Re, Os, etc., was ruled out by the chemical evidence.

These unexpected and startling results, which seemed to be incompatible with the already known properties of nuclei, were offered with much reserve. The authors felt that it was conceivable that some series of unusual accidents might have combined to give misleading results. This paper, however, was the one which led others to perform the experiments of other sorts which were soon to give abundant evidence for the correctness of Hahn and Strassmann's conclusions.

In a second paper (39Ha5) they gave the details of a most beautiful and thorough set of experiments which showed beyond a doubt that both "Ra III" and "Ra IV" were actually isotopes of Ba rather than of Ra. Further, the 15-min. and the  $\sim$  4-hr. "Ra's" obtained from bombarded Th were likewise shown to be isotopes of Ba. Also, they pointed out that these active Ba and La atoms were not all previously unknown ones. The half-life of 86 min. agrees well enough with the known half-life of 85 min. of Ba<sup>139</sup>, for them to be considered the same. La IV with its halflife of  $\langle 40 \text{ hr.}$  is probably to be identified with the known  $La<sup>140</sup>$  of a half-life of 31-46 hr. This fixes Ra IV as  $Ba^{140}$ .

When the U<sup>239</sup> nuclei split to give atoms such as these having masses of  $\sim$ 140 the remaining portions of the nuclei having masses close to 99 must appear elsewhere, presumably as nuclei of lower atomic number. Hahn and Strassmann were able to find an active Sr, and an active Y. When they passed a stream of air through a solution of a U salt it picked up a gas which in turn produced an active solid deposit in a cotton filter. The gas must have been an active noble gas, Kr or Xe. Inactive Sr and Cs were added to a solution made to contain the active substances in the filter. When the Sr and Cs were precipitated both precipitates were active. The gas must have been either active Kr producing Rb and Sr, or active Xe producing Cs and Ba, or a mixture of both gases.

# 8. High energy of the products of fission

The publication of the results of Hahn and Strassmann led to a great outburst of activity elsewhere. It was realized by many experimenters that such a splitting of a heavy U or Th nucleus into two lighter ones would involve the release of an enormous amount of energy, since the sum of the masses of practically all possible pairs of lighter nuclei was less than that of the original one. The pioneers in this work were Frisch  $(39Fr1)$  and Joliot  $(39Jo2)$  who worked by different methods independently and almost simultaneously.\* Several others performed essentially the same experiments independently but somewhat later.

Before discussing the experiments it is desirable to consider the release of energy more closely. Assume, for example, that the U nucleus of a net charge of  $+92$  is to split into two nuclei having charges of  $+54$  and  $+38$ , respectively (Xe and Sr). The heaviest stable atoms of these two elements are Xe<sup>136</sup> and Sr<sup>88</sup>, which have a total mass of 224 instead of the 239 of the splitting U nucleus. There is an excess of 15 neutrons to be divided between the two resulting nuclei. The immediate products af the fission might be something like  $_{54}Xe^{140}$  and  $_{38}Sr^{99}$ . This particular division is the one which, for the assumed charges of the fragments, most nearly preserves in both of them the neutron/proton ratio of the original  $U^{239}$  nucleus. It is not meant to imply, however, that there is any good reason for believing that the splitting will necessarily preserve this ratio. The  $_{54}Xe^{140}$  nucleus would undergo four successive beta-disintegrations,  $Xe \rightarrow Cs \rightarrow Ba \rightarrow La \rightarrow Ce$ , finally becoming stable  $_{58}Ce^{140}$  and the  $_{38}Sr^{99}$  would undergo six,  $Sr\rightarrow Y$ <br> $\rightarrow Zr\rightarrow Nb\rightarrow Mo\rightarrow Ma\rightarrow Ru$ , to become stable

<sup>\*</sup>Frisch's first publication was a letter to Nature dated January 16, 1939 which appeared in the number of February 18. Joliot's first results were reported in the Comptes rendus of January 30, 1939.

 $44Ru^{99}$ . The energy developed in the whole succession of processes will, by Einstein's relation, be  $\Delta E = c^2(M_0 - M_1 - M_2)$  in which  $M_0$ ,  $M_1$ , and  $M_2$  are, respectively, the masses of the original nucleus and the two final nuclei. Introducing the mass numbers,  $A$ 's, and packing fractions,  $f$ 's, one has

$$
\Delta E = K \{ A_0 (1 + f_0) - A_1 (1 + f_1) - A_2 (1 + f_2) \}.
$$

 $K$  is the constant for converting masses on the atomic weight scale into energies in Mev. Since  $A_1 + A_2 = A_0$  this gives  $\Delta E = K(A_0 f_0)$  $-A_1f_1 - A_2f_2$ . From Dempster's work we may take packing fractions of  $+0.00056$ ,  $-0.00030$ and  $-0.00059$  for  $_{92}U^{239}$ ,  $_{58}Ce^{140}$ , and  $_{44}Ru^{99}$ , respectively.  $K$  is 931. For the assumed case we have

$$
\Delta E = 931 \{ 239 \cdot 5.6 - 140(-3.0) -99(-5.9) \} 10^{-4} = 218 \text{ MeV}.
$$

This value, having been obtained with the packing fractions of stable nuclei, gives the sum of the kinetic energy of fission and of the energies of the ten beta-transitions which follow it. More exact calculations, (39Bo9) taking account of the energy evolved in the beta-transitions, give a value of 189 Mev for the particular division here assumed. They show that the available energy is greatest ( $\sim$ 200 Mev) for splitting into nearly equal fragments. For conservation of momentum the energy mill have to divide between the two fragments in the inverse ratio of their masses. It will be 111 Mev for  ${}_{88}Sr^{99}$  and 78 Mev for  ${}_{54}Xe^{140}$ . These figures- must be considered to give upper limiting values for the kinetic energies, since possible excitation of the original fission fragments has been neglected.

Particles of such enormous energy would be expected to produce correspondingly large numbers of ion pairs as they pass through a gas. By putting a thin layer of U in a suitable ionization chamber connected to a linear amplifier and irradiating it with neutrons Frisch and others observed the great bursts of ionization produced by such particles.  $(39Fr1, 39An1, 39Fo1, 39Ro2,$ 39Gri, 39Thi, 39Ma4, 39Je1, 39Dr1). Maximum energies of the order of 100 Mev were found. These pulses of ionization are so extremely large compared with those of single alpha-particles that they are easily recognizable.

Jentschke and Prankl (39Je1) found that the pulses could be divided according to magnitude into two groups at about 60 and 100 Mev. Von Droste (39Dr1) found eight different groups. Later, more elaborate experiments of Booth, Dunning, and Slack (39Bo7), carried out with very thin electrolytically deposited layers of U, showed a continuous distribution of size of pulse with two pronounced maxima. If one assumes these to correspond to two separate superposed groups, the maximum energies in these groups were found to be 100 Mev and 72 Mev, respectively. The wide range of energies and apparently continuous distribution agrees with the chemical evidence in showing that the initial fission can occur in many different ways with a statistical distribution among them, rather than in some single way or small number of alternative ways.

Nearly all who performed experiments of this kind used both Th and U and found such fission bursts with both. They were produced in U by both thermal and fast neutrons but only by fast ones in Th. These results agree with the earlier ones of Meitner, Hahn, and Strassmann as to conditions for the production of Ba from Th and U.

Search has been made for fission with many other elements. Using 91Pa<sup>231</sup>, von Grosse, Booth, and Dunning (39Gr2) found fission produced only by fast neutrons but in amount approximately 3S times that found with Th. No fission has been found with other elements, with the exception of a few very weak effects which lack confirmation. See Section II 4.

Joliot (39Jo2) saw that because of the great amount of available energy the disintegration would occur with explosive violence, sufficient to eject thp fragments from the irradiated sample. He placed a source of neutrons inside a brass cylinder which was coated with a layer of uranium oxide. Surrounding this, at a distance of 3 mm from it, was a cylinder of Bakelite. This soon became coated with active substances on the inside, the activity being like that produced in U. Suitable blank experiments showed that the activity came from the irradiated U. Irradiated Th gave the same result. Surrounding the cylinders mith paraftin increased the yield by a factor of 2, showing that slow neutrons also were effective. The interposition of different thin ab-

sorbing layers between the U and the Bakelite showed that the maximum range of the projected particles was of the order of 3 cm in air.

Similar experiments were suggested by Meitner (39Fri) and carried out in collaboration with Frisch (39Me2). They collected the fission fragments on a water surface and treated the resulting solution chemically after addition of Ba, La, and Pt salts as carriers. Comparison of the total activity of a sample produced by merely drying up part of the solution with one obtained by precipitating the Pt with H2S showed that the sulphide precipitate contained about two-thirds of the total activity. The active substance of the precipitate must always have been present among the "transuranic" elements obtained by this same method of precipitation from irradiated U. Here, however, they must have reached the water by explosive disruption of U nuclei, for mere capture of neutrons and ejection of betaparticles would not give sufficient energy to heavy atoms to eject them from the sample. They were, therefore, of mass less than that of U. This proved that the whole group of interrelated "transuranic" elements must be isotopes of atoms of lower atomic number instead of being truly transuranic.

Bretscher and Cook (39Br2) collected the recoil fragments on glass, dissolved them in aqua regia, added Ba, La, and Pt as carriers, separately precipitated the Ba, the La, and the Pt, and studied the activities of the precipitates. All three precipitates showed some of the already known half-lives. Those appearing in the sulphide precipitate were 16 mm. , 75 min. and 72 hr. which agree well enough with the 16-min., 59-min. , and 66-hr. periods of "transuranic" substances, thus strengthening the already strong evidence that these substances are not truly transuranic.

Both Bretscher and Cook (39Br2) and Meitner (39Me3) performed such experiments with Th and found activity in the precipitate of platinum sulphide. Since transuranic elements could not be produced by any process by neutrons acting on Th, this experiment also cast doubt upon the correctness of ascribing such activity produced in U to transuranic substances. Although Bretscher and Cook, and Meitner, did not agree entirely as to the half-lives of the substances produced

from Th, nevertheless they did agree that the distribution of half-lives and intensities was very diferent from that found in the similar experiment with U. This indicates a different statistical distribution of fission produced in the disintegration of the two atoms which merits more thorough investigation. Such a study can be undertaken with greater prospect of success after the chemical identity of the various active substance has been established.

McMillan (39Mc5) later used a thin sample of U from which nearly all fission products could escape. Any true transuranic atoms would have remained in the U. After irradiation of this sample he found in it only a 25-min. activity of  $U^{239}$  and a strong one with a half-life of 2 days. Segre (39Se7) found that the substance having this latter activity has the chemical properties of a rare earth, and is not a daughter substance coming from the 25-min. U. Its identity has not yet been established.

As discussed above, the fission particles occur in two main groups having diferent energies. Booth, Dunning, and Glasoe (39Bo6), using an ionization chamber, found corresponding ranges of 2.<sup>2</sup> and 1.5 cm in air. Haxel (39Ha13) later found 1.8 and 1.5 cm. Glasoe and Steigman (39G11) used a thin film of Cellophane, of thickness equivalent to 1.4 cm of air, to filter out the particles of the less energetic group. Using 10 min. irradiations they found an active deposit without the filter showing the following halflives;  $\sim$ 9 hr., 80 min., 32 min., 17 min., 10 min., and other shorter unresolved periods. With the filter they got  $\sim$  6 hr., 35 min., 17 min., and 3 min. The 80-min. and 10-min. substances had been removed by the filter. The 80-min. one is perhaps to be identified with the 86-min. Ba III of Hahn and Strassmann. It grows from a 10-min. Cs, as shown by Aten, Heyn, and Bakker (39He1), Apparently some original fission particle of mass 139 gives these active atoms by successive disintegration. Of the activities of particles which penetrate the filter the 17-min. one is known to be attributable to Rb (39He1, 39At1, 39Gr2). It thus appears that the more penetrating fission particles are the lighter ones, as should be expected from the principle of conservation of momentum (assuming nearly the same mean net charge). It should be remarked, however, that there was a half-life of 35 min. also found when the filter was used, and that an active Cs of nearly this half-life is known (39He1, 30 min. ; 39At1, 33 min.; 39Gr2, 30 min.). Also, the 17-min. period of Rb is not distinguishable in these experiments from the 16-min. one of one of the strongly produced "transuranic" substances. The evidence is not unambiguous.

Cloud-chamber pictures showing very clearly the heavy ionization of the fission particles were obtained by Joliot (39Jo3, 39Jo4), and by Corson and Thornton (39Co5). In the picture accompanying this latter paper one can see short tracks diverging from the main heavy one. They are to be attributed to projected C, N, and 0 nuclei. The lack of any corresponding deviation in the main track is an indication of the relatively great mass of the particle producing it.

A more detailed discussion of the kinetic energies and ranges of the fission particles is given in Section II 1.

# 9. Fission Products identified by x-rays and chemical properties

Abelson investigated the radiation emitted by the 66-hr. "transuranic" element (77 hr. according to his measurements) and found x-rays of a hardness which might be expected for the  $L$ x-rays of a transuranic element. Upon hearing of Hahn and Strassmann's results he investigated this radiation more carefully (39Ab4) and found that it actually consisted of  $K$  x-radiation from iodine. The same experiment was performed independently by Feather and Bretscher (39Fe1). Appropriate chemical operations showed that the 2.5-hr. daughter substance which is formed from the 77-hr. one is iodine, and that the 77-hr. active substance is tellurium. This was the first experiment which showed definitely that a supposedly transuranic active element was actually an isotope of an ordinary one.

Numerous workers have carried out experiments like those of Hahn and Strassmann in which an air stream is passed through an irradiated solution of a U or Th compound. The stream picks up an active gas showing several half-lives. It is undoubtedly a mixture of various isotopes of Kr and Xe since active Rb, Sr, Cs and Ba have all been found in the solid deposit produced by the gas. The detailed results of these and other experiments in which fission products are identified chemically are given with references in Section II 2.

So far the following chemical species have been fairly surely identified among the fission products;- Br, Kr, Rb, Sr, Y, Mo, Sb, Te, I, Xe, Cs, Ba, La. It is noteworthy that no elements between  $_{42}$ Mo and  $_{51}Sb$  have yet been found.\* It seems likely that this represents a real absence or relatively low yield of such elements, rather than merely an unfinished stage of comprehensive chemical investigation. Hahn and Strassmann (39Ha5) made special experiments for finding active 47Ag among the "transuranic" elements, and concluded that none of them was Ag. It should be noticed, however, that one could not expect to find either the familiar  $_{47}Ag^{108}$  or  $_{47}Ag^{110}$ active isotopes since the chains of disintegrations of fission products of those masses would terminate with stable  $_{46}Pd^{108}$  and  $_{46}Pd^{110}$ . Ag<sup>111</sup> has a half-life of 7.5 days so that even if present in considerable quantity it would give a relatively feeble activity. The same consideration applies with less force to  $Ag^{112}$  of a half-life of 3.2 hr. Since many of the missing elements from 43Ma to  $_{50}$ Sn would be precipitated in the "transuranic" sulphide group, final information on this point will have to wait upon the identification of all of the elements of the group.

The extreme complexity of the problem is indicated by the results of Abelson and by newer ones of Hahn and Strassmann. Abelson (39A4) reports five active varieties of Sb, seven of Te, four of I. He finds the 77-hr. Te to be descended from a 5-min. Sb, whereas the early experiments of Meitner, Hahn and Strassmann (37Me5) indicated that this was descended from a 59-min. element. Hahn and Strassmann (39Ha12) find that their  $66$ -hr  $(77$ -hr.) substance is a mixture of Te and of Mo of the same half-life. The 59-min. half-life may turn out to be that of an ancestor of the Mo. It should be remembered that when an active substance of short half-life arises in a chain of disintegration between two others of much longer half-lives the short one is difficult to detect and identify unless the proper chemical separation is made. It may develop that intermediate substances of short half-lives are missing from the "transuranic" chains of disintegrations.

<sup>\*</sup>See 6rst paragraph on p. 24.

In the early work there was not perfect agreement between the half-lives of the Ba obtained from Th, with those of the Ba from U. Recently, using more strongly activated samples, Hahn, Strassmann and Flügge (39Ha15) have been able to show that the early results were in error. The half-lives of Ba from Th do agree with those of Ba from U.

As might be expected there are great differences in the magnitude of the activities of the different substances. As mentioned above, Meitner and Frisch find approximately twothirds of the total activity in the sulphide precipitate. Hahn and Strassmann (39Ha14) find the active Br to be only about  $1/10$  of the strength of the active I.

#### 10. Neutrons produced in fission

No matter how the  $U^{239}$  nucleus divides, there is an excess of neutrons over the number contained in normal nuclei of the same charges. As discussed above this excess can be corrected by emission of beta-particles which transforms neutrons into protons. It occurred to several experimenters that the great excess of neutrons might also be overcome by direct neutron emission. Such neutrons were sought for and found by several different methods. Von Halban, Joliot and Kowarski (39Ha7, 39Ha9) measured the distribution of neutrons surrounding a source placed at the center of a large container filled with a solution of uranyl nitrate. Another set of measurements was made in which a solution of ammonium nitrate of the same strength (1.6 normal) was used. The primary neutrons were photo-neutrons produced by the absorption of the  $\gamma$ -rays from Ra (RaC) in Be. Each measured intensity, multiplied by the square of the distance from the source, was plotted against this distance. It had previously been shown that the area under a curve through such points is proportional to the product  $Q_{\tau}$ , where Q is the rate of production of neutrons and  $\tau$  is the mean life of a neutron before capture. The curve obtained with uranyl nitrate might have been expected to give a smaller area than that obtained with ammonium nitrate partly because of a slightly greater total cross section for capture of thermal neutrons, and mostly because of resonance capture of some neutrons before they are slowed

down to become thermal ones. In the experiment the occurrence of this latter process is equivalent to a reduction of the strength of the source. Instead of getting a smaller area under the curve for the uranyl nitrate solution, von Halban, Joliot and Kowarski actually found it to be about 5 percent greater than with ammonium nitrate. This increase showed the production of secondary neutrons in the solution, presumably connected with the fission process. The average number of neutrons produced per fission was calculated to be  $3.5\pm0.7$  (See Section II 5 for more detail). The use of the moderately slow  $Ra\gamma$ -Be neutrons ruled out the possibility of attributing the secondary ones to any  $(n, 2n)$  process.

Somewhat similar experiments were carried out independently by Anderson, Fermi, and Hanstein (39A2). Instead of using a solution of a salt of U they surrounded the source of neutrons with a spherical bulb containing U, and then investigated the distribution of neutrons in a large tank of water surrounding the bulb. They obtained an apparent 6 percent increase of the strength of the source by using the U, corresponding to the product of about 2 neutrons per fission. Since a Rn-Be source of neutrons had been used the possible production of secondary neutrons by an  $(n, 2n)$  process was not entirely excluded. They also used a Ra $\gamma$ -Be source but were unable, with it, to get sufficiently accurate results for useful comparison of the areas under the two curves. These latter experiments did show clearly, however, that a considerable change in the spatial distribution of the neutrons in the water was produced by introducing the U. It indicated an absorption of slow neutrons by the U accompanied by production of new fast ones.

The production of such fast neutrons was demonstrated in an ingenious way by Bode, von Halban, Joliot, and Kowarski (39Do1). They surrounded a Ra $\gamma$ -Be source of neutrons by crystallized uranyl nitrate and placed it at the center of a large flask containing  $CS<sub>2</sub>$ . In this was dissolved 200 mg of P. Any fast neutrons would produce some active  $P^{32}$  by the  $S^{32}(n, p)P^{32}$ process. This reaction is endothermic by 0.9 Mev and requires neutrons of something like 2 Mev for a reasonable yield. The primary neutrons are not sufficiently energetic to produce it. After six days of irradiation the P was isolated by distillation and was found to give an activity of 32 counts per min. A similar sample obtained in a blank run without the uranyl nitrate gave only 5 counts per min. The fast neutrons must have been produced by the U, presumably in connection with fission.

Meanwhile, other workers were investigating these secondary neutrons by observing the ionization attributable to particles projected by them.

Szilard and Zinn (39Sz2) used primary Ra $\gamma$ -Be neutrons and a He-filled ionization chamber with amplifier. By performing experiments with and without Cd screens they were able to measure the number of fast secondary neutrons arising from the absorption of primary thermal ones in U. Their results indicated the production of about two fast neutrons per fission. Von Droste (39Dr1) in his first paper concerning the ionization produced by the fission fragments noted that the apparent total kinetic energy of two fragments was less than the available energy. He suggested that the difference might possibly be attributed to emission of neutrons. An experiment with an ionization chamber filled with He indicated the presence of fast neutrons in the neighborhood of U being irradiated with D-D neutrons. These experiments were extended in collaboration with Reddeman (39Dr2). They investigated the bursts of ionization produced by the He nuclei which had been struck by neutrons of energy greater than 1.5 Mev, when  $U_3O_8$  was bombarded by D-D neutrons. The number of scattered primary neutrons was obtained by similar experiments made with  $PbO<sub>2</sub>$  instead of  $U<sub>3</sub>O<sub>8</sub>$ . The distribution of the fast neutrons present was found to be similar to that of Ra-Be neutrons, and not at all like that of the primary D-D neutrons. Some of them were found to have energies considerably greater than 2.4 Mev, the energy of the primary neutrons. The magnitude of the effect indicated a production of one to two fast neutrons per fission.

Haenny and Rosenberg (39Ha6) used Rn-Be neutrons partially slowed by 4 cm of paraffin and an ionization chamber filled with hexane. It was sensitive only to fast neutrons. Surrounding the source by 8 cm of  $U_3O_8$  increased the number of observed counts by 20 percent. Interposition of paraffin and Cd showed this increase to be attributable to neutrons.

Halban, Joliot and Kowarski (39Ha11) also performed experiments with an ionization chamber filled with oxygen. Projected 0 nuclei of high energy were produced by the neutrons coming from irradiated U. Particles of an energy of  $\sim$ 2.5 Mev were found to be reduced in number by screening the U by Cd. Such fast atoms must have been projected by neutrons of at least 11 Mev energy, which had been liberated from the U by the action of thermal neutrons. This gives a striking indication of the explosive nature of the process. The authors suggest that the neutrons are probably liberated simultaneously with the fission.

In an extension of the above-mentioned experiments with the photo-neutrons (39S32), Zinn and Szilard (39Zi3) studied the distribution in energy of the secondary neutrons by means of the recoils in He. They found that nearly all of the secondary neutrons have energies below 3.5 Mev. Their result is, however, not incompatible with the presence of a few of the much faster ones found by Halban, Joliot, and Kowarski. The number of the secondary neutrons per sec. was determined from observation of the number of projected protons obtained when the ionization chamber was filled with hydrogen. This, divided by the number of fissions per sec., gave a value of 2.3 secondary neutrons per fission. The number of fissions per sec. was obtained from separate experiments made with a different ionization chamber which contained known amounts of U. It was inserted in the position filled by the U sample in the experiments on the secondary neutrons. No estimate is given of the accuracy of the final figure of 2.3 neutrons per fission. The calculation of the number of secondary neutrons is, however, a complicated one. It necessarily involves some simplifying approximations which introduce errors that are difficult to estimate. Also, the final result involves numerous experiments, with a cumulation of experimental error which may be important, although none of the separate experiments involve large errors.

The results of von Droste and Reddeman, of Halban, Joliot, and Kowarski, and of Zinn and Szilard are in substantial agreement, the lastnamed being the most comprehensive.

An intermittent beam of D-D neutrons was used by Gibbs and Thomson (39Gi1) to show that the bulk of these neutrons are emitted within 0.001 sec. of the time of impact of the primary ones. They found only a slight positive effect, indicating the presence of the small number of delayed neutrons which are to be discussed in the next section, Other papers on secondary neutrons are discussed in Section II 5.

# 11. Delayed emission of neutrons

Roberts, Meyer, and Wang (39Ro3) found that neutrons continued to come from irradiated U after cessation of the bombardment. The intensity fell off exponentially with a half-life of  $12.5 \pm 3$  sec. The neutrons were accompanied by a strong, hard  $\gamma$ -ray, decaying with the same period, Roberts, Hafstad, Meyer and Wang (39Ro5) performed numerous blank experiments to eliminate any source of the neutrons other than the U. Upon surrounding the activated U with unactivated U no increase of the number of delayed neutrons was found, thus eliminating the possibility of their being photo-neutrons produced by the  $\gamma$ - rays of the same period.

The delayed neutrons were found to be produced by both thermal and fast ones, but not by those of intermediate energy coming from C bombarded by protons, exactly as with fission itself. The study of particles projected by these neutrons in a cloud chamber indicated that their energy was  $\sim 0.5$  Mev. Several other longer  $\gamma$ -ray periods were found. Delayed neutrons were also produced in Th, the half-life being the same as with U, but the intensity was smaller.

Booth, Dunning, and Slack (39Bo8) also studied the delayed neutrons, using primary neutrons produced in the cyclotron by bombarding Be with protons. In addition to the 10—15-sec. period they found one of 45 sec. The  $\gamma$ -rays showed those two periods and other longer ones. By the use of Cd and B filters they found that the delayed neutrons are produced mostly by slow primary ones, the  $1/v$  law for absorption being approximately followed. The equilibrium number of delayed neutrons per min. was found to be about  $1/60$  of the number of fissions per min.

Most recently (November, 1939) Brostrøm, Koch, and Lauritsen (398r9) have found delayed neutrons with a half-life of 3 sec., and most probably also with a half-life of 0.1—0.3 sec. These half-lives agree reasonably well with ones previously found with penetrating beta-particles (398a5).

#### 12. The instantaneous occurrence of fission

By use of an interrupted beam of neutrons Green and Alvarez (39Gr1) showed that fission must occur within less than 0.003 sec. after the capture of a neutron by a U nucleus.

Feather (39Fe4) showed that this time could be reduced to  $5\times10^{-13}$  sec. by a most ingenious experiment. It depended upon the occurrence of the 6ssion before the loss of forward velocity of the  $U^{239}$  nucleus. When a  $U^{238}$  nucleus captures a fast neutron the resulting  $U^{239}$  nucleus must be projected forward at high velocity (1/239 of that of the neutron) in order that momentum may be conserved. If fission takes place before this  $U^{239}$ nucleus has been brought to rest by collision with other atoms, its velocity will be added to the hssion velocity with respect to the center of mass. The fission fragments sent out in the forward direction will have a greater average velocity and range than those sent in the backward direction. Feather put a layer of U between two equal layers of polythene, but separated from them by Au foils of 7.13 mg/cm'. The plane of the layer of U was set perpendicular to a neutron beam so that the forward moving hssion fragments would be collected on one polythene layer and the backward moving ones on the other. The neutrons came from a Li target bombarded by deuterons. The activity produced on the forward sheet was  $19\pm3$  percent greater than that on the rear one showing that fission must have occurred before loss of forward velocity of the splitting nucleus. A difference of  $10\pm 3$  percent was observed for thinner gold foil (6.74 mg/cm<sup>2</sup>).  $2\times10^{-13}$  sec. is the time calculated for this loss of forward momentum to take place for nuclei which had capmentum to take place for nuclei which had cap-<br>tured the fastest neutrons used  $(5 \times 10^{-13}$  is given as a conservative upper limit). The observed effect of 10-20 percent is somewhat less than a calculated one of 30—40 percent, for reasons not entirely clear.

# 13. Attempts to produce fission by photoexcitation

No fission was observed by Roberts, Meyer, and Hafstad (39Ro2) when their ionization chamber containing a U layer was placed close to a target of Li or of F bombarded by 3 microamp. of 1 Mev protons.

Heyn, Aten, and Bakker (39He1) were unable to find any fission products from U which had been irradiated by the penetrating  $\gamma$ -rays from proton-bombarded Li.

# 14. Excitation functions and nuclear cross sections

Meitner, Hahn, and Strassmann (37Me3, 37Me5) found that the "transuranic" elements were produced by fast Ra-Be neutrons, and also by the slow ones obtained with the use of paraffin. When, with paraffin present, the U was shielded by Cd, the yield of "transuranic" substance was only that which would be expected because of the fast neutrons present. Evidently, of the slow neutrons, only the thermal ones were effective in producing fission. With the thermal neutrons removed by Cd shields there was, nevertheless, a considerable production of 23 min. U by resonance capture of the somewhat faster ones.

When Th was irradiated there was found (35HaO, 35Cu1, 38Ro5) an active Th coming from capture of slow neutrons and several active substances attributed to fast neutrons. None of these latter active substances were produced by thermal or other slow neutrons.

Nearly all of the experimenters who studied the large bursts of ionization attributable to individual fission particles found a paraffin effect for U, but none for Th. Green and Alvarez (39Gr1) and Roberts, Meyer, and Hafstad (39Ro2) used Cd to prove that the thermal neutrons were the effective slow ones. The results were thus consistent with the earlier ones obtained by studying the active substances.

This production by thermal neutrons of active substances which do not result from resonance capture was difficult to understand. With other atoms it is found that when an active isotope is formed by capture of thermal neutrons it is also producible by any resonance capture of somewhat faster ones. This is what would be expected on theoretical grounds by combination of the theoretical expression for the respective capture cross sections with the known distribution in energy of neutrons slowed by paraffin. Because of the difFiculties from the theoretical standpoint in attributing the observed effects of both the thermal and the resonance neutrons to the same nuclei, Bohr (39Bo2) suggested that it is probable that the thermal neutrons are captured by the 139-times-less-abundant (39Ni1)  $U^{235}$  nuclei, and according to the  $1/v$  law. This is discussed further in the next section. The hypothesis has received additional support from the experiments of Anderson et al. (39An1) who find that the number of fission bursts does approximately follow the  $1/v$  law. They used Cd and B filters to vary the distribution in velocity of the neutrons and compared the number of fissions with the number of disintegrations produced in a boron layer in the same ionization chamber. The two numbers were found to vary in nearly the same way. Absorption by boron has already been shown to follow the  $1/v$  law.

Before entering upon the discussion of the various nuclear cross sections for fission and related processes it is perhaps permissible to recall the meaning of a nuclear cross section. It may be defined by  $\sigma$  in the following equation

#### $N = n\nu\sigma$ .

N is the number of occurrences of the kind in question per sq cm of bombarded surface per second. It may refer to the number of neutrons captured, the number scattered, the number of fissions, or whatever is of interest. Here also  $n$ is the number of bombarding particles per sq. cm per sec. (normal incidence assumed) and  $\nu$  is the number of atoms of the kind involved per sq. cm of surface. It is as if each atom presented a target area,  $\sigma$ . If the target is struck the effect in question occurs; if not, nothing happens. It is apparent that  $\sigma$  gives a sort of averaged probability of occurrence of an effect for a random spatial distribution of impinging particles. Of course, this simple consideration applies only to thin layers in which only a very small percentage of the impinging particles produce any effect. With thicker layers proper account must be taken of the exponential falling off of the intensity of the incidental beam.

For thermal neutrons there are cross sections to consider for all of the following processes; scattering, formation of active U, fission, production of secondary neutrons, and production of delayed neutrons.

The cross section for fission produced by thermal neutrons has been measured by Anderson et al. (39An1). The number of fissions per minute with and without a shield of Cd was measured, the difference between them giving the number attributable to thermal neutrons. A thin layer of electrolytically deposited U of known mass, and therefore of known number of atoms per sq. cm, was used. A Rn-Be source of neutrons of known strength produced the fissions, so that all quantities of the above equation except  $\sigma$  were known. tities of the above equation except  $\sigma$  were known<br>A value of  $2\!\times\!10^{-24}$  cm $^2$  was found, no estimat of error being given. An independent repetition of the experiment (39Bo8) gave a value of  $3\times10^{-24}$  cm<sup>2</sup>. In computing these cross sections the total number of U atoms per sq. cm has been used. If, as Bohr suggests, only the  $U^{235}$  nuclei are responsible for this fission by thermal neutrons the above figures must be multiplied by 140 to get the true cross section.

The cross section for production of secondary neutrons is really what is obtained by the experiments with such neutrons. Dividing it by the cross section for 6ssion gives the number of neutrons per fission. Reversing the calculation we get  $4\times10^{-24}$  cm<sup>2</sup> and  $7\times10^{-24}$  cm<sup>2</sup> for the cross section for formation of secondary neutrons from thermal ones from the experiments of Anderson, Fermi, and Hanstein (39An2) and von Halban, Joliot and Kowarski (39Ha9), respectively.

By calibrating their detector of neutrons Booth, Dunning and Slack (39Bo8) found that there is one delayed neutron emitted for every sixty fissions. This gives a cross section for delayed neutrons of  $5 \times 10^{-26}$  cm<sup>2</sup>, if one assumes their value of  $3\times10^{-24}$  for fission. By using Cd and B absorbers the yield of delayed neutrons was found to follow the  $1/v$  law as does the fission produced by slow neutrons.

Since there is a resonance capture of neutrons giving 23-min.  $U^{239}$  it is to be expected that there would be some formation of this substance by capture of thermal neutrons. This was investigated by von Halban, Kowarski and Savitch (39Ha10) who obtained a cross section of (39Ha10) who obtained a cross section of  $(1.3 \pm 0.45) \times 10^{-24}$  cm<sup>2</sup>, and by Anderson and Fermi (39An3) who found  $1.0\times10^{-24}$  and Fermi (39An3) who found 1.0×10<sup>-24</sup> and<br>1.2×10<sup>-24</sup> in two separate experiments. The determination of von Halban et a/. was based on getting a ratio of the cross section for U to the already known one for activating Au. They compared the activities produced in U and Au layers of known thicknesses. Anderson and Fermi compared the U cross section with those of I and Mn in their two experiments. The agreement of all three results is most satisfactory in view of the many difficulties in such work. This cross section indicates that the resonance absorption is to be attributed to several levels rather than to a single one.

So far as is now known, the  $U^{239}$  nucleus formed by capture of a thermal neutron can do only one of two things, either divide by fission, or give out radiation to become 23-min. U<sup>239</sup>. The sum of the two cross sections discussed above,  $(2+1.2) \times 10^{-24} = 3.2 \times 10^{-24}$ , should, therefore, be the cross section for absorption of thermal neutrons. This is somewhat 1ower than a value of trons. This is somewhat lower than a value o<br>5×10<sup>-24</sup> found by Anderson, Fermi, and Han stein (39An2) in their work on secondary neutrons discussed above, and a value of  $5.9 \times 10^{-24}$ found by Michiels, Parry, and Thomson (39Mi1). The discrepancy between these values and the one got from summing the separate cross sections is probably not outside of experimental error.

A much larger value of the cross section for absorption was obtained by a different method by Whittaker, Barton, Bright, and Murphy (39Wh1). They measured the reduction of the intensity of a beam of thermal neutrons by introduction of a known thickness of metallic U between the source and the detector (activated Ag). The initial intensity, and the reduction in intensity, are, respectively, proportional to the number of neutrons impinging on the detector per second without an absorber and the number removed from the beam per sec. Since only the ratio of these two latter quantities is required for determining  $\sigma$ , absolute measurements are not necessary; the ratio is that of the respective responses of the detector. Whittaker et al. found a value of  $(23.1\pm0.5)\times10^{-24}$  cm<sup>2</sup> for the total cross section for removal of thermal neutrons from the beam by combined capture and scattering. With the same sample of U, Dunning independently found a value of  $(23.3 \pm 0.5) \times 10^{-24}$  cm<sup>2</sup>. Goldsmith, Cohen and Dunning (39GoO) reported a smith, Cohen and Dunning (39GoO) reported a<br>value of  $(20\pm2)\times10^{-24}$  cm<sup>2</sup> obtained from

measurements on  $U_3O_8$ , additivity of the cross sections of <sup>U</sup> and 0 being assumed. Reddemann and Bomke (39Re1) reported a value of  $22\pm3$  $\times$ 10<sup>-24</sup> cm<sup>2</sup>. The value of  $(23.2\pm0.5)\times10^{-24}$  cm<sup>2</sup> for the total cross section is well established.

On the basis of further experiments of Khittaker et al. this was divided into a cross section taker *et al*. this was divided into a cross section<br>for absorption of  $(11\pm3)\times10^{-24}$  cm<sup>2</sup>, and of  $(12\pm3)\times10^{-24}$  cm<sup>2</sup> for scattering. The absorber of metallic U was moved close to the detector so that more of the scattered neutrons would be effective. The fraction of the total number of scattered neutrons getting to the detector was determined by auxiliary experiments of the same type made with C and Fe. For these both cross sections are known (no capture by C). With this fraction, and the observed increase of number of neutrons resulting from moving the U close to the detector, the number of scattered neutrons and the above cross section for scattering were obtained. Because of the discrepancy between the resultant large value of the cross section for absorption and the results of the other earlier experiments Reddemann and Bomke (39Re1) undertook similar experiments. As mentioned above, they found nearly the same total cross section, but their scattering experiments indicated a much greater scattering and a total cated a much greater scattering and a total<br>capture cross section of only  $(2\pm 4)\times 10^{-24}$  cm<sup>2</sup>. They state that although their experiments are not extremely accurate they are not reconcilable with the results of Whittaker et al. Further work on this point is needed.

It may be remarked, however, that there is nothing about the experimental data so far obtained which eliminates the possibility of the formation of a shorter-lived isomer of  $U^{239}$  in the capture followed by radiation. Hahn, Meitner, and Strassmann (37Ha5) did find some shortlived activity in the U precipitate which contained the 23-min. U. If such an isomer exists, the cross section for radiative capture will be larger than that determined from the yield of 23-min. U.

Meitner, Hahn, and Strassmann (37Me5) found a mass absorption coefficient of  $3 \text{ cm}^2/\text{g}$  for the resonance neutrons which produce the  $23$ -min. U<sup>239</sup>. This corresponds to an effective 23-min. U $^{239}$ . This corresponds to an effectiv<br>cross section of  $1.2 \times 10^{-21}$  (39Bo9). Halbar Joliot and Kowarski (39Ha10) found that the probability that an originally fast neutron will be so captured while being slowed in a 1.6 normal water solution of uranyl nitrate is  $0.16 \pm 0.025$ .

The diferent measurements on the cross section for fission produced by fast neutrons are not strictly comparable because of the different types of neutron sources used. Using a Ra-Be source Meitner, Hahn, and Strassmann (37Me5) found Meitner, Hahn, and Strassmann (37Me5) found<br>a value of 1.6×10<sup>-25</sup> cm<sup>2</sup> for the production of the 16-min. active substance, and a nearly equal one for the independently produced 59-min. substance, giving a total of  $3.2 \times 10^{-25}$  cm<sup>2</sup>. Since we now know that these activities result from only two out of many sorts of fission, the total cross section for fission by such neutrons must be considerably larger.

Using Rn-Be neutrons Joliot (39jo4) found a cross section for fission of the order of  $1\times10^{-25}$ cm'. This was calculated from the activity of collected recoil fragments. Anderson et al. of collected recoil fragments. Anderson *et al* (39An1) also found a value of  $1 \times 10^{-25}$  cm<sup>2</sup> by direct observation of the fissions produced by Rn-Be neutrons. Thibaud and Moussa (39Th2) found the same value. The Rn-Be neutrons are on the average somewhat more energetic than those from Ra-Be.

For D-D neutrons of an energy of 2.4 Mev Ladenburg et al. (39La1) found cross sections of Ladenburg *et al.* (39La1) found cross sections of<br> $5 \times 10^{-25}$  cm<sup>2</sup> and  $1 \times 10^{-25}$  cm<sup>2</sup> for U and Th, respectively. These are accurate to about 25 percent. The ratio of the cross section for fission of the two elements was constant within 10 percent for variation of the energy of the neutron from 2.1 to 3.1 Mev.

With neutrons from C bombarded by protons, which have a maximum energy of about 0.5 Mev, Roberts et al. (39Ro2) found a slight amount of fission with U and none at all with Th. Von Grosse, Booth and Dunning (39Gr2) find fission in Pa produced by fast neutrons, but not by thermal neutrons, or photo-neutrons from  $Ra\gamma$ -Be.

A process which is of importance with fast neutrons is inelastic scattering. The captured neutrons are re-emitted with assorted energies below the original energy, but with a distribution about a most probable value that corresponds to the "temperature" of the compound nucleus, and is much lower than that of the original neutron. Because of the concurrent emission of secondary neutrons this cannot be directly studied with U.

By scattering experiments Goldstein, Rogozinski, and Walen (39Go1) find that for Pb the total cross section for both elastic and inelastic scattercross section for both elastic and inelastic scatter<br>ing of fast neutrons is 7.4 $\times10^{-24}$  cm², and tha<sup>.</sup> the total cross section for scattering and fission the total cross section for scattering and fissior<br>by U is  $(11.2 \pm 1.5) \times 10^{-24}$  cm<sup>2</sup>. Further, they find that the cross section for inelastic scattering by Pb is  $2\times10^{-24}$  cm<sup>2</sup> divided by  $1-k_i$ , in which  $k_i$  is the average efficiency of production of ions by inelastically scattered neutrons as compared with the original ones. The cross section for inelastic scattering by Pb must, therefore, be inelastic scattering by Pb must, therefore, be<br>greater than  $2\times10^{-24}$  cm<sup>2</sup> and less than  $7.4\times10^{-24}$ cm', the value for the total cross section of both elastic and inelastic scattering. Values of the same order of magnitude would be expected for U.

For the delayed emission Roberts et al. For the delayed emission Roberts *et al*<br>(39Ro5) report a value of  ${\sim}4{\times}10^{-26}$  cm<sup>2</sup>, the activation being produced by Li-D neutrons. This is nearly one-half the cross section for fission by Rn-Be neutrons. Although the two results are not strictly comparable because of the two different sorts of activating neutrons involved, it appears that the ratio, (No. of delayed neutrons)/(No. of fissions), is markedly diferent from what it is with slow neutrons. This apparent great difference between the fission products produced by slow and by fast neutrons must be viewed with some suspicion, since all other properties have been found to be the same.

#### 15. Theory

The discovery of fission by Hahn and Strassmann immediately dispelled the accumulated difficulties concerning the active substances produced from U and Th. It was no longer necessary to assume  $(n, \alpha)$  and  $(n, 2\alpha)$  processes for such heavy elements or the formation of long series of transuranic elements. The active substances of the same chemical nature could be assigned to different isotopes, so that there was no evidence for triple, inheritable isomery. There was no longer any difficulty over finding a place for a rare-earth-like substance among transuranic elements. Obviously, fairly long chains of successive disintegrations were to be expected because of the excess of neutrons.

It did, however, raise new theoretical problems of which the principal one is: how can the fairly moderate activation of the nucleus resulting from

capture of a neutron lead to such a cataclysmic disruption? If one considers the masses of the fragments obtainable by splitting all sorts of nuclei into two parts, it appears that energy could be gained by such splitting of every nucleus of mass over  $\sim$ 110. Why, then, are the heavier nuclei stable? Why do none exist in nature of mass greater than 238? Why does the capture of a neutron lead to the energetically possible splitting of U, Pa and Th while fission is highly improbable for all lighter nuclei?

The first theoretical suggestions were those of Meitner and Frisch (39Me1). They pointed out that, just as a drop of liquid which is set into vibration may split into two drops, so might a nucleus, which in many ways is analogous to a drop of liquid, also split into two smaller nuclei. This becomes more probable for heavy nuclei because of an effective reduction of the surface tension resulting from increasing nuclear charge. Because of the surface tension the spherical shape, which gives the minimum surface for a given volume, is, for a liquid, the one of stable equilibrium. The nuclear forces of short range, which are analogous to the cohesive forces between atoms in a drop of liquid, must tend to produce similar effects of surface tension in nuclei. The electrostatic energy of repulsion of protons, however, tends to produce an opposite effect. For given volume the spherical shape would be the most unstable one, if only the electrostatic forces were important. The actual nudeus will be stable as long as the sum of the surface energy and the electrostatic energy has a minimum for the spherical shape. With increasing size and charge of the nucleus this minimum becomes fiatter and would be expected to disappear altogether for some critical value of Z. Nuclei of greater Z would immediately break apart. Meitner and Frisch estimated that this would occur for value of Z close to 100. Since the U nudeus lies close to this limit of complete instability, it seems reasonable that it should have only slight stability of form and be likely to divide into two nuclei upon receiving a moderate energy of excitation. The name for the process, fission, was suggested by Meitner and Frisch. The stability of form of nuclei has been discussed in several papers by others (39Fa1, 39Fe2, 39Fr2, 39So2, 39We2, 39We3, 39Y1) and also in

the comprehensive paper on the theory of fission by Bohr and Wheeler (398o9).\*

Bohr (39Bo1, 39Bo2), concurring in the hypothesis of Meitner and Frisch as to the nature of fission, discussed its relation to the other more familiar nuclear processes. As with other reactions, one may assume that the initial capture of a neutron produces an excited compound nucleus. Its energy of excitation will be quickly distributed among different modes of vibration and will eventually be dissipated by one of the several processes which are in competition with one another. These are: re-emission of a neutron, fission, and radiation. Their relative probabilities will vary with the amount of energy of excitation. Since the cross sections for the production of fission are of the same order of magnitude as ordinary nuclear cross sections, one may conclude that the required energy of excitation is also of the same order of magnitude. This means that it is so large compared to the zero-point energy of the principal modes of vibration of the nucleus that the quantum aspect is unimportant and the problem may be treated classically. Also, the needed energy is great enough so that one may understand the comparative stability of the 23-min. U<sup>239</sup> nuclei. After radiation of their excess energy, they disintegrate in relatively leisurely manner with a half-life of 23 min. Similarly, one can understand the stability of ordinary heavy nuclei which could release much energy by splitting into two lighter ones.

The probability for radiation would be expected not to change greatly for increasing energy of excitation of the compound nucleus, those for fission and for re-emission of a neutron would be expected to increase rapidly with increasing energy. The experimental results with Th are in good agreement with this theory. The capture of slow neutrons produces excited compound nuclei for which the probability of radiation far outweighs the probability of fission or ejection of a neutron. Only the active Th<sup>233</sup> of a half-life of 26-min. results. With fast neutrons the energy of excitation is high enough so that fission has become a relatively important process.

The same argument would apply to the results

obtained with U were it not for the large yield of fission products given by thermal neutrons.

The probability of fission is negligible for the resonance neutrons of 25 volts or so, and one would expect it to be even less for thermal neutrons. For this reason, Bohr was led to attribute the fission produced by thermal neutrons to one of the lighter, less abundant isotopes of U, probably  $U^{235}$ . The great probability of fission would lead to a shortening of the mean life and corresponding broadening of the levels of the  $U^{236}$ nucleus so that they might overlap and effectively produce a continuum of absorption. In this case the  $1/v$  law for absorption might be expected, as Bohr pointed out. As mentioned above, this was found to be the case experimentally by Anderson, Fermi, and Hanstein (39An2). The correctness of this attribution of the fission by thermal neutrons to  $U^{235}$  is crucial for the theory here outlined. If experimental ingenuity should some day solve the difficult problem of separating the isotopes of U, and it should be found that the abundant  $U^{238}$  isotope is responsible for the fission produced by thermal neutrons, present ideas concerning the properties of nuclear states would have to be modified extensively.

None of the experiments on fission are of a sort to give any direct evidence for the hypothetical re-ejection of the neutrons mentioned above. That it is of importance, nevertheless, is indicated by an experiment of Nishina, Yasaki, and Kimura (38Ni1). They exposed purified Th to the very fast neutrons ( $\sim$ 15 Mev) obtained by bombarding a Li target with 3 Mev deuterons. In a precipitate of Th they found the 26-min. activity of Th $^{233}$  and the 24.5-hr. activity of UY  $(Th<sup>231</sup>)$ . The latter atoms must have been produced by the  $(n, 2n)$  process. The highly excited Th<sup>233</sup> nuclei ejected neutrons to leave excited Th<sup>232</sup> nuclei which, in turn, ejected neutrons. In both nuclei the probability of ejection of a neutron must have been comparable with or greater than those for fission and radiation.

Bohr and Wheeler (398o9) developed the above theoretical considerations in greater detail and more quantitatively. They calculate the energy made available for the various possible fission fragments, the energies of binding of neutrons in the fragments, and the energies of beta-emission from them. Such calculations have

<sup>~</sup> Dr. Seaborg has kindly called my attention to a theoretical paper by B.Ferretti I'39Fe8},It is not available to me. (Added to proof, December 6, 1939.}

also been made by Flugge and von Droste (39Fl1). Bohr and Wheeler discuss the stability of nuclei with respect to deformation. Nuclei which are stable for slight departures from the spherical shape may nevertheless become unstable for small finite distortion. The critical distortion and the energy required for reaching it can be found by interpolation between the calculable limiting cases of small and large influence of nuclear charge. From the resulting curve one can read off the critical energies (in Mev) for fission as follows:  $_{92}U^{235}(5.0)$ ,  $_{92}U^{236}(5.2)$ ,  $_{91}Pa^{232}(5.5)$ ,  $_{92}U^{239}(5.9)$ , and  $_{90}Th^{233}(6.9)$ , the energies of excitation of the compound nuclei resulting from capture of thermal neutrons are:  $_{92}U^{235}(5.4), \quad_{92}U^{236}(6.4), \quad_{91}Pa^{232}(5.4), \quad_{92}U^{239}(5.2),$  $_{90}Th^{233}(5.2).$ \* The threshold energy of neutrons required for producing fission of a given compound nucleus is obtained by subtracting the binding energy from the critical energy for fission. The results for this energy of the bombarding neutrons are as follows for the different bombarded nuclei:  $_{92}U^{238}(0.7)$ ,  $_{92}U^{235}(-1.2)$ ,  $_{92}U^{234}(-0.4)$ ,  $_{91}Pa^{231}(+0.1)$ ,  $_{90}Th^{232}(1.7)$ . The negative values for  $U^{235}$  and  $U^{234}$  mean that these nuclei will undergo fission after capture of thermal neutrons. That the value for  $Th<sup>232</sup>$  is so much higher than that for  $U^{238}$  agrees with the observed smaller cross section of Th for bombardment by 2.4 Mev neutrons. The value for  $Pa^{231}$  agrees with the finding of von Grosse, Booth, and Dunning (39Gr2) that fission is not produced in Pa by thermal neutrons or by photo-neutrons from  $Ra\gamma$ -Be.

The discussion of the statistical mechanics of the excited nucleus leads to the conclusion that for energies of exciting neutrons below, or not far above, the critical one radiation will predominate. With increasing energy the probability of fission will increase rapidly. The cross section for fission will not increase indefinitely, however, because at still higher energies the reemission of neutrons becomes the predominant process.

The detailed comparison of the calculated probabilities for fission for the different U and Th nuclei lead to good agreement with the experimental results. This discussion indicates that it is  $U^{235}$  rather than  $U^{234}$  which is responsible for the fission by thermal neutrons.

Bohr and Wheeler further discuss the emission of secondary neutrons which accompany fission. They give reasons for believing that the delayed ones are to be attributed to evaporation from nuclei which are produced in beta-transformatioris to very highly excited states rather than to the ground state of the product nuclei. Many possible levels of excitation and corresponding energies of the beta-partides are to be expected. The actual beta-radiation will be the result of the superposition of those from the many separate beta-transitions. The results of Barschall, Harris, Kanner, and Turner (39Ba5), who found no quantity of extremely hard betarays comparable with the number of much less energetic ones, give some indication of a greater probability of beta-transition to excited levels. The instantaneous neutrons may arise at the instant of fission or may come very rapidly from the fission fragments because of their rather large energy of excitation resulting from the distorted shape which they are likely to have at the instant of fission.

The statistical distribution in size of the fragments of fission depends on the complicated dynamics of the dividing nudeus. The theory is not developed to the point of giving this distribution, but does indicate that there is a rather wide range of possible fragments even for energies but slightly greater than the critical energy.

The cross sections for fission of U produced by 6 Mev deuterons and protons are estimated to 6 Mev deuterons and protons are estimated t<br>be  $\sim 10^{-29}$  cm<sup>2</sup> and  $\sim 10^{-28}$  cm<sup>2</sup>, respectively Also, the cross section for the photo-fission of U is estimated to be  $\sim 10^{-27}$  cm<sup>2</sup>. This would mean that no observable effect could have been expected in the experiments of Roberts, Meyer, and Hafstad (39Ro2).

As mentioned above, the experimental results indicate that a somewhat asymmetrical splitting of the nucleus is more probable than a symmetrical one. Beck and Havas (39Be2) calculate that the available energy is more in excess of the potential barrier against fission for an asymmetrical division than it is for a symmetrical one. The available energy must be in excess of the potential energy of position of the two

<sup>\*</sup> In the paper of Bohr and Wheeler an erroneous value of 6.4 Mev was given for  $_{91}Pa^{232}$ . See (39Bo12).

fragments with respect to each other as they begin to separate in order that they may do so. This energy, if spherical fragments are assumed, is  $Z_1Z_2e^2/R$ . Beck and Havas calculate these energies for various splittings, using  $R=1.3$  $\times$ 10<sup>-12</sup> cm. They do not discuss the reasons for choice of this particular value of  $R$  or, indeed, for any constant value for all splittings. It may be that this value is chosen as one which is large enough so that the nuclei may surely be separated and exerting only the long range electrostatic forces on each other. The available energy is computed from the masses. The two curves approach each other most closely for  $Z_1 = 37$ ,  $Z_2 = 55$  so that such a splitting would be expected to be the most probable one.

Flügge and von Droste (39F11) presented what is essentially the same idea in somewhat different manner. They gave a plot of the stable nuclei and drew in a curve connecting the points of greatest stability for each Z. This curve has maxima in the neighborhood of  $Z = 35$  and  $Z=55$ . This means that fission fragments of Z's in those neighborhoods would have smaller numbers of excess neutrons and would, therefore, be less highly excited than others. More of the available energy would appear as kinetic energy and there would be greater chance of surmounting the potential barrier. For a fission into  $_{56}Ba$  and  $_{36}Kr$  both fragments have this extra stability so that this fission and others close to it should be the most probable. Further, both Ba and Sr have been found to have especially large values of the (negative) packing fractions. The experimental results seem to be in good agreement with these considerations of Beck and Havas and of Flugge and von Droste.

# 16. Chain reactions. Uranium as a source of nuclear energy

It was realized by many of those who first studied the emission of secondary neutrons (39Ha7, 39Ha6, 39Do1, 39Ha9) connected with fission that there was a possibility of a catastrophic chain reaction. The secondary neutrons might themselves produce still more fissions and neutrons, and so on. The propagation of such a chain would involve the release of terrific amounts of energy in a very short time. Flügge (39F12), in his comprehensive review of this

aspect of fission, estimates that 1 cu. m of  $U_3O_8$ might develop  $27 \times 10^{15}$  m $\cdot$  kg of energy  $(1.0 \times 10^{12})$ kilowatt-hours) in less than 0.01 sec.

In order that such chains may occur it is necessary that the average number of neutrons per fission be enough to compensate for the many which are lost by capture processes which do not result in fission. This condition may be written  $A_f\nu/A_{\text{tot}} > 1$  (39Ha9). Here  $\nu$  is the number of secondary neutrons per fission;  $A_f$  is the cross section for fission multiplied by the concentration of U atoms; and  $A_{\text{tot}}$  is the sum of all such A's for neutron-removing processes.

It is impossible at present to see whether this condition is satisfied for fast neutrons because of uncertainties concerning inelastic scattering. What one needs to know is the cross section for the inelastic scattering which wi11 reduce the energy of the neutron to so low a value that it can no longer produce fission. This will vary greatly with the energy of the neutron being scattered, so that a thorough knowledge of the distribution in energy of secondary neutrons would also be necessary for finding the effective average value.

If slow neutrons are to produce chains it is necessary to introduce some hydrogen-containing substance to slow the secondary ones. Here again we do not know accurately either  $\nu$  or the total cross section for capture with sufhcient accuracy to find whether the condition for a chain reaction is satisfied. Further, the originally fast neutrons will be subject to capture in passing through the region of resonance energy. According to the experiments of von Halban, Kowarski, and Savitch (39Ha10) the chance for such capture is about 1 in 6 in a 1.6 normal solution of uranyl nitrate. Flügge calculates that the critical concentration of  $U_3O_8$  would be 12 kg of  $U_3O_8$  per liter of  $H_2O$  for  $\nu=2$ , and 3.1 kg per liter for  $\nu=3$ . This calculation assumes  $\sigma = 2 \times 10^{-24}$  cm<sup>2</sup> for fission and  $\sigma = 1.3 \times 10^{-24}$  cm<sup>2</sup> for the radiative capture of thermal neutrons by U. The building up of the process of release of energy would be extremely rapid. If, however, the cross section for capture of neutrons is  $5 \times 10^{-24}$  cm<sup>2</sup>, and that for fission is  $2 \times 10^{-24}$ .  $\nu$  would have to exceed 4 for a chain reaction to occur.

The above considerations are valid only for

extremely large masses of  $U_3O_8+H_2O$ . For a sample of reasonable size many neutrons will escape without producing further ones so that the quantity  $A_f\nu / A_{tot}$  will have to be even greater if explosive release of nuclear energy is to occur.

The theory involving this diffusion of neutrons is developed by Flügge and also by Perrin (39Pe2) and by Adler (39Ad2), who calculate minimum radii for the mass of U, assuming a reasonable mean free path for neutrons. They arrive at radii of the order of 100 cm for constants as above, All of these calculations are of doubtful significance because of uncertainties concerning the important cross sections.

Most recently the question has been studied in still another way by Anderson, Fermi, and Szilard (39An4). A powerful source of neutrons was placed at the center of a tank containing a solution of MnSO<sub>4</sub>. About 20 percent of the thermal neutrons were absorbed by the Mn, producing a measurable activity which was proportional to the rate of absorption of neutrons by the solution. Around the source were put tubular cans which could either be left empty or be filled with  $U_3O_8$  of a total mass of 200 kg. The activity of Mn produced in the solution was found to be about 10 percent greater when the cans were filled with the  $U_3O_8$ . An auxiliary experiment showed that about 50 percent of the neutrons from the source were absorbed by the  $U_3O_8$ , so that on the average at least 1.2 secondary neutrons was produced for every thermal one absorbed. This is the ratio of the cross section for the production of secondary neutrons to that for capture. The authors estimate that the ratio should be raised to about 1.5 if account is taken of neutrons absorbed by resonance capture before they have been completely slowed down. A chain reaction would thus be possible in pure U were it not for the necessity of slowing the neutrons to thermal velocities. The addition of the needed hydrogen increases the chance for capture of thermal neutrons. For a chain to be possible it is necessary that more secondary ones be produced than are captured by both U and H. Further, the relative importance of resonance capture would be expected to vary with changing concentration of H. Until more is known about this process, it will be impossible to say whether or not the chain reaction can occur.

There are obvious difhculties connected with the control of the evolution of this large amount of energy. Adler and von Halban (39Ad1) have shown that the development of the chains involving slow neutrons can be controlled by the use of Cd mixed with the U and hydrogencontaining substance. As the energy is released the temperature will rise and the average speed of thermal neutrons will increase. The number of fissions per second will be proportional to  $v\sigma_f$ . Since  $\sigma_f \propto 1/v$  the product will be constant For Cd, however, the cross section is constant so that the number of neutron-removing collisions will increase with rising  $T$  and  $v$ . The fission will become relatively less important until a steady state of development of energy is reached at some temperature which is dependent on the amount of admixed Cd.

For the first time it seems that there is some reasonable possibility of utilizing the enormous nuclear energy of heavy atoms. If later accurate measurements of the cross sections and the number of secondary neutrons show this to be correct the practical difhculties can undoubtedly be overcome in time. The prospects of extremely cheap production of energy are none too bright, however, if the chain can be propagated only by slow neutrons acting on the less abundant isotope,  $U^{235}$ . In a general discussion of this topic, Roberts and Kuper (39Ro9) point out that under those conditions, even at present prices, the cost of the raw material, U, would be about  $\frac{1}{8}$  of that for an equivalent amount of coal.

#### PARr II

#### 1. Energies and ranges of fission particles

The energies of the fission particles have been estimated from the ionization produced as follows:

Frisch (39Fr1), 70 Mev.

- Anderson et al. (39An1), 90 Mev maximum.
- Roberts, Meyer, and Hafstad (39Ro2), 75-150 Mev.
- Green and Alvarez (396r1), 80 Mev.
- Jentschke and Prankl (39Je1), 2 groups at 61 and 98 Mev.

Von Droste (39Dr1), 8 groups at 36, 43, 52, 59, 65, 74, 80, 90 Mev.

- Thibaud and Moussa (39Th2), 10—<sup>65</sup> Mev; (39Th5),  $<$  70 Mev.
- Magnan (39Ma4), 49—98 Mev.

Booth, Dunning, and Slack (398o7}, 2 groups having maximum energies at 100 Mev and 72 Mev; energies at the maxima of the groups,  $\sim 80$  and  $\sim 50$  Mev.

Haxel (39Ha13), 52 and 74 Mev.

Kanner and Barschall (see below).

All of these estimates are based on measurements which give the number of pairs of ions produced by fission fragments. In order to calculate the energy it is necessary to assume a mean energy per pair of ions. This is usually taken to be the same as the value which is known from experiments with alpha-particles. The propriety of using this value is open to question because of the very different nature of the ionizing particles.

Because of the conservation of momentum one would expect a fast light particle to be associated with a heavy slow one. The sum of the energies for the two maxima of Booth, Dunning, and Slack is  $\sim$ 130 Mev. A value of 125 Mev is obtained by adding von Droste's values in pairs, smallest plus the greatest, 36+90, second smallest plus second greatest,  $43+82$ , and so on. From Haxel's values one gets 126 Mev. The total kinetic energy is apparently well below the calculated available kinetic energy. The difference is to be attributed to high excitation of initially distorted fission fragments.

Most recently (December, 1939) new results of this sort have been obtained in Princeton by M. H. Kanner and H. H. Barschall. They have made experiments like those of Booth, Dunning, and Slack except for the use of D-D neutrons instead of thermal neutrons. They obtain a number-energy curve with two fairly sharp peaks, but with these maxima at energies of 65 and 97 Mev. Further, by using a very thin foil of Al upon which U has been sputtered, and by placing it at the center of the ionization chamber, they observe the total energy of the pairs of fission fragments which come from the same original U atoms. The uncorrected curve shows a high peak at 151 Mev. This is corrected for the loss of energy by the fragment which had to penetrate the foil to give 159 Mev. This agrees very well with the sum of the peak energies for the single fragments,  $65+97=162$  Mev. This is the energy of the fragments produced in the most probable splitting. The shapes of the curves give the relative probabilities of the different possible ones. The half-width of the peak for

total energy is about 30 Mev. The highest value observed is about 200 Mev, in good agreement with the calculated value of Bohr and Wheeler (39Bo9). In these experiments a special study of the saturation characteristic of the ionization chamber was made in order to make certain that the full energies were being measured. If we assume that the maxima correspond to the most probable division and that the energies of the two fragments are inversely proportional to their masses we get masses of 96 and 143 (assuming 239 for U). The latter figure corresponds well enough to the masses of the many active atoms from Te to La which have been found in the fission products. It seems likely that the former must then represent the important group of still unidentified "transuranic" active substances, as was suspected by Hahn and Strassmann (39Ha1). In experiments in which recoil fragments are collected, the more energetic lighter fragments would be favored. In their experiments of this latter sort Meitner and Frisch (39Me2) found about  $\frac{2}{3}$  of the total activity to be in elements of the "transuranic" group so we must infer that these arise in highly probable divisions of the U nuclei. The important "transuranic" activities would thus be expected to belong to atoms of atomic numbers somewhat less than 44. (This paragraph added to proof, December, 1939.)

By a calorimetric method, Henderson (39He2) has measured the energy per fission and has found it to be 175 Mev, accurate to about 10 percent. Neutrons, most gamma-rays, and some of the most penetrating beta-rays escaped so that the result would be expected to be smaller than the true average value for the available energy. On the other hand, nearly all beta-rays and all other soft radiations were captured so that this measured energy must be greater than the average kinetic energy of separating fission fragments.

Magnan (39Ma11) found that after covering the layer of  $U_3O_8$  with 1.5 mm of paraffin rather large pulses of ionization of unexplained origin were recorded. The fission particles themselves would have been absorbed in the paraffin and the pulses were too large to be attributed to single protons projected by fast neutrons. They could not have been caused by C nuclei projected by fission fragments because their number increased when the thickness of the layer of  $U<sub>3</sub>O<sub>8</sub>$  was increased beyond one which would absorb all fission fragments.

Values for the ranges of the fission particles have been obtained by measuring the absorption of the fission fragments in foils of known thickness, by varying the pressure in an ionization chamber of known size, and by measuring the tracks in a cloud chamber.

The measurements made by studying absorption in foils are the following: (fission of U)

Joliot (39Jo2, 39Jo4),  $\sim$ 3 cm air equivalent, range >10 $\mu$ in  $UO<sub>2</sub>$ .

Thibaud and Moussa (39Th2),  $\sim$ 5 $\mu$  in Al, (39Th5) <1.8 cm in air.

McMillan (39Ma5), 2.4 cm air equivalent.

From their cloud-chamber pictures Corson and Thornton (39Co5) found a maximum range of  $\sim$ 3 cm.

The results with ionization chambers are:

- Roberts, Meyer, and Wang (39Ro3},1.05 cm for U, 1.<sup>2</sup> cm for Th.
- Anderson et al. (39An1), 1.7 cm.
- Booth, Dunning, and Glasoe (39Bo6), 2 groups;  $2.2 \pm 0.1$ cm, and 1.5 cm.

Haxel (39Ha13), 2 groups;  $1.8 \pm 0.24$  cm,  $1.5 \pm 0.20$  cm.

It appears that the range as found by ionization is definitely less than that determined by penetration of the 6ssion particles. This indicates that the ionization falls off greatly toward the end of the path of the 6ssion fragment instead of increasing as it does with alpha-particles. Beck and Havas (39Be3) have explained this on the basis of a progressively decreasing net charge of the fragment, resulting from the nonradiative capture of electrons from air molecules. Something of the sort is also indicated by an experiment of Haxel (39Ha13) in which he finds that the loss of energy in passing through a thin foil of Al is nearly the same for both the fast and slow groups of fragments. For the same net charge one would expect the ionization and loss of energy to be greater for the slower ones. Haxel calculates mean net charges of 10 and 7 electron units for the two fragments.

# 2. Chemically identified fission products and chains of successive disintegrations

The fission products which have been identified are given below, those of known mass are given

first for each element. Reported half-lives which seem to refer to the same isotope are grouped together. Unless otherwise indicated, all substances are obtained from U. If also obtained from Th or Pa this will be indicated by the chemical symbol. When the same mass number appears for successive elements, the active substances in question are members of a chain of successive disintegrations. Where such chains have been established without the mass of the atoms being known, the corresponding activities in successive elements are all labeled with the same small letter.

 $34Se$ 

None found (39Ha14). Two weak Se activities (398r2) (Te?).

 $35Br$ 82.

230 min. (39 Ha14);  $\sim$ 1.5 hr. (39Th1);  $\sim$ 2.5 hr. (39Br2) Th. (Identification by Langsdorf and Segrè. Both U and Th. Unpublished.) 35 min. (39Ha14);  $\sim$  40 min. (39Do2).  $\sim$ 20 hr. (39Th1); 22 hr. (39Br2).

Hahn and Strassmann (39Ha14) found that the probability of production of active Br is only about  $\frac{1}{10}$  of that for active I. Also see (39Th5) for halogen activities.

agKr

- 83. 113 min. (Langsdorf and Segrè, not yet published Th.
	- An excited state of stable  $Kr^{82}$  decaying with emission of a highly internally converted gammaray.

88. 3 hr. (39La2) Th, (39Ha14).

88. 35 sec. (39G11). Isomery?

88. (39He1); (39At1) Th, (No  $\tau$ ).

 $_{37}Rb$ 

88. 17-18 min. (39Hel), (39At1) Th, (39GI1), (39La2) Th, (39Ha14), (39Gr2) Pa. Also see (39Cu1) and (39Sa3}.

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38Sr
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89. 54 days {39Lii).
a.(>90.) 6 hr (39 Li1).
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(>90.) 7 min. (39Li1).
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 $39Y$ 

a.( $>90$ .) 3.5 hr. (39Li1). Also (39Ha5), no  $\tau$ . 42Mo

99 (or 101}.66 hr. (39Ha12), (39Ha15} Th.

43Ma

47Ag

99 (or 101). Not yet found in fission but known to be the daughter of 66-hr. Mo (Seaborg and Segre, Phys. Rev. 55, 808 (1939)).

None found (39Ha5). (See paragraph added below. )

61Sb {All (39Ab4))

- 127. 80 hr.
- 129. 4.2 hr.
- 132, 134, or 136. 5 min.
- a. &10 min.
- b. &10 min.
- $_{52}Te$ 
	- 127. 10 hr. (39Ab4).
	- 129. 70 min. (39Ab4); 60 min. (39Ha14).
	- 131. 30 hr. and 30 min. (39Ab4) and (39Ha14).
	- 132, 134, or 136. 77 hr, (39Ab4); 66 hr. (39Ha12}, (39Ha14), (39Ha15) Th.
		- a. 43 min. (39Ab4}, (39Ha14).
		- b. 60 min. (39Ab4), (39Ha14).
- 63I
	- 131. 8 days (39Ab4), (39Ha14}.
	- 132, 134, or 136. 2.4 hr. (39Ab4); 2.3 hr. (39Ha12}, (39Ha14), (39Ha15) Th, (39Fe1).
		- a. 54 min. {39Ab4); 45 min. (39Do2} Th, 57 min. (39Ha14).
		- b. 22 hr. (39Ab4); 12 hr. (39Do2) Th, 18.5 hr. (39Ha14).

64Xe

- 139. Few seconds (39Ha8};~0.<sup>5</sup> min. (39Ha1); <sup>5</sup> min. (39611);(39At1) Th.
- 140?  $\sim$ 15 min. (39Ha14). (Uncertainty of mass arises from doubt as to identification of La<sup>140</sup>.)

9 hr. and 5 days (39La2) and (Langsdorf and Segrè, not yet published), both grow from I.

- 55Cs ,<br>139. 10 min. (39He1), 10 min. (39At1) Th; <8 min (39Ha8); 10 min. (39Gl1); 6 min. (39Ha14).
	- 140? 33 min. (39Ha8), (39Ha14); 30 min. {39Hei); 33 min. (39At1) Th; 30 min. (39Gr2) Pa.

1-2 min. (39Hei) (Cs or Rb?). Also unidentified 3-min. activity from gas (39611}.

- 668a
	- 139. 86 min. (39Ha5), (39Ha14), {39Hei), (39At1) Th, (39Ha15) Th, (39611),
	- 140? 300 hr. (39Ha1), (39Ha5), (39Ha15) Th, (39Ha14).
		- a.  $14\pm2$  min. (39Ha1). Not descended from a gas. (39Hei) and (39Ha14), {38Me2)Th.
		- b. &1 min. (39Hai), (38Me2)Th.

67La

- 140? &40 hr. (39Hai), 36 hr. (Hai4), (39Ha5) (398r2}. a. 3.5 hr. (38Cu3), (38Cu4);  $\sim$ 2.5 hr. (39Ha1);
	- (39Cu1) Th, (39Ha14) (39Br2).
	- b.  $<$ 30 min. (39 Hal);  $\sim$ 18 min. (38Me2); 12 min. (35Cui).
	- 80 min. , 14 hr. , 12 days (La carrier} {398r2}.

Since the above table was compiled some new results have been given by Nishina et al. (39Ni3). They exposed purified thorium nitrate to fast neutrons from Li bombarded by deuterons. Chemical separations showed the presence of the following active fractions: (elements not previously found in fission of either Th or U

put in italics)  $Ag$ ,  $Sn$ ,  $Sb$ ,  $Hg$ ,  $Bi$ , an alkali fraction, a halogen fraction, a Mo fraction, a Se+Au fraction, a  $Cu+Cd$  fraction. Neither Pb nor As was found. Only active substances of fairly long half-lives were found, since the chemical separations took 2 to 3 hours. With uranium they found Bi, Hg, Ag,  $Sb+Sn$ , and  $Cu+Cd$ . Also, Chlopin et al. (39Ch3) found with U active bodies which were to be attributed to gases of long half-lives which were not themselves primary fission products. The products which grew from the gases were collected in silica gel, extracted, and separated into Ba, Sr, and La fractions. The half-lives found are as follows:  $La+K$ ,  $\sim$ 30 min.,  $\sim$ 9.5 hr.,  $\sim$ 45 hr.; Sr,  $\sim$ 20 min.,  $\sim$ 7.5 hr., one much longer; Ba,  $\sim$ 10 hr.,  $\sim$ 50 hr. \*

# 3. Unidentified half-lives

There are numerous measured half-lives of active substances produced in fission of which the chemical nature is not yet known. A list of these with references and remarks follows.

0.3–0.9 sec. and  $3-4$  sec. (39Ba5), (39Br9), beta-particles and delayed neutrons. See I 11.

10sec. (34Fe2), (37Me5), perhaps Ma(39Me1), perhaps the same as the 11—15-sec. period for penetrating beta-particles (398a5) and the 12.5 sec. period for delayed neutrons. See I 11.

40 sec. (34Fe2), (37Me5), perhaps Ma(39Me1), perhaps the same as the 45-sec. period for delayed neutron emission. See I 11.

The following half-lives of "transuranic" elements: 2.2 min., 59 min., 16 min., 5.7 hr., 60 days. See I 3. The genetic relationships given for these may possibly omit certain intermediate products of shorter half-lives. Chemical identifications are needed for certainty.

In the sulphide precipitate of fission products from Th Meitner (39Me3) found two strong activities of half-lives of 40 min. and 14.5 hr. In a similar experiment Bretscher and Cook  $(39Br2)$  found 70-min., 11-hr., and 45-day halflives. In the fraction precipitated with Ba there was a 6-hr. activity, and there were 80-min. , 14-hr. , and 12-day ones in the La precipitate. Both Meitner and Bretscher and Cook found some of the ordinary Ba, La, and trans-U half-lives in the similar experiments made with U.

<sup>~</sup> This paragraph added to proof, December 6, 1939.

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As discussed in I 8 McMillan and also Segrè find an active substance remaining in the U as does the 23-min. U<sup>239</sup>. It has a half-live of 2.3 days and the chemical properties of a rare earth.

It is clear from the work of Abelson (39Ab4) that in studying very strong samples of fission products many hitherto undiscovered ones of lesser abundance will be revealed. The above half-lives, however, belong to some of the more probable products, for the most part.

# 4. Fission of elements other than U and. Th

Von Grosse, Booth, and Dunning (39Gr2) studied fission in Pa and found it to be produced by fast neutrons, but not by thermal neutrons or photo-neutrons from  $Ra\gamma - Be$ .

Roberts, Meyer, and Hafstad (39Ro2) found no fission of amount equal to 1/1000 of that for Th with the following elements: Bi, Pb, Tl, Hg, Au, Pt, W, Sn, Ag. They used fast neutrons (up to 13.5 Mev) from a Li target bombarded by 1 Mev deuterons. Magnan (39Ma4), also using such neutrons, found no effect with Bi, Ta, Te, Cd, Ag, Pd, Mo, Zr, and Sr but did get small efFects with Au, W, and Ti. The occurrence of fission in Ti which lies near the minimum of the curve of packing fractions would indeed be surprising.

Others, using weaker sources of neutrons, also report no fission obtained with various ones of the above elements (39Jo2, 39Je1,39Th2, 39My1, 39Gr1, 39Jo4).

#### S. Secondary neutrons

The experiments of von Halban, Joliot and Kowarski (39Ha9), discussed in Section I 10, were based on the measurement of the concentration of neutrons in a large container holding solutions of uranyl nitrate and ammonium nitrate. We may write

$$
S/S' = Q\tau/Q'\tau' = Q\Sigma A'/Q'\Sigma A, \qquad (1)
$$

where S refers to the area under the curve of Ir<sup>2</sup> vs. r, Q to the strength of the source,  $\tau$  to the mean life of a slow neutron,  $\Sigma A$  to the sum of the cross sections for capture of thermal neutrons multiplied by the respective concentrations of the capturing atoms. Let the unprimed quantities refer to the ammonium nitrate solution and the primed ones to the uranyl nitrate solution. Instead of taking an equivalent A for the resonance capture as do von Halban, Joliot, and Kowarski, we may consider the strength of the source to be effectively reduced by this process.

If  $p$  is the chance of capture of a neutron while being slowed, this process will effectively lower the strength of the source to  $Q(1-p)$ . The number of secondary neutrons produced by these will be  $Q(1-p)\nu A_f/\Sigma A$ .  $A_f$  is the A for fission, and  $\nu$  is the average number of secondary neutrons produced per fission. Of these secondary neutrons,  $Q(1-p)^2 \nu A_f / \Sigma A$  will reach thermal velocities. These will in turn generate

$$
Q(1-p)^{3\nu^2} A_f^{2}/(\Sigma A)^2
$$

new ones, and so on. Summation of all of these terms gives the effective strength of the source  $Q'$ ,

$$
Q' = Q(1-p)\frac{1}{1 - (1-p)\nu A_f/\sum A}
$$

Putting this expression for  $O'$  into Eq. (1) and solving for  $\nu$  gives

$$
\nu = \frac{\sum A}{A_f} \left[ \frac{1}{1 - p} - \frac{S}{S'} \cdot \frac{\sum A}{\sum A'} \right].
$$
 (2)

 $\sum A' = \sum A + A_{et} + A_f - \Delta A_H$ .  $A_{et}$  is the A for radiative capture of thermal neutrons and  $\Delta A_H$ is a term which takes account of the difference between the hydrogen contents of the two solutions. By experiment  $S'/S=1.05\pm0.01$ ,  $1-p$  $=0.84\pm0.025$ ,  $\Sigma A = 32\pm3$  (different from von H. , J., and K. because their value was an average which involved  $A_r$ , the equivalent A for resonance capture),  $A_{ct} = 2.1 \pm 0.7$ ,  $A_f = 3.2$ ,  $\Delta A_H$  $=1.2\pm0.1$  (see 39Ha9). With these numerical values one obtains  $\nu=3.5\pm0.7$ .

It is apparent from (2) that the calculated value of  $\nu$  will vary almost inversely as the value of  $A_f$  used. The experiment is really a determination of the  $A$  or  $\sigma$  for production of secondary neutrons.

Michiels, Parry, and Thomson (39Mi1) performed experiments with a source of neutrons at the center of a set of spherical shells which were placed in a tank of water so that only slow neutrons would be measured. The outermost shell contained boron carbide which prevented the escape of thermal neutrons or their penetration inward from the water. An inner space could be filled with uranium oxide. Measurements were made with and without the uranium for fast neutrons and for a mixture of fast and slow neutrons produced by surrounding the source with paraffin. The experimental results are not susceptible of direct interpretation because of the several cross sections for both fast and slow neutrons involved. They seem to indicate that the cross section for production of secondary neutrons by fast primary ones is greater than neutrons by fast primary ones is greater than<br> $4\times10^{-24}$  cm<sup>2</sup>. This value is much greater than that of other experimenters. The interpretation of the results, however, must involve some assumption as to the number and velocity of inelastically scattered neutrons. Hardly enough is known so far concerning that process so that proper account of it may be taken.

Rotblat (39Ro6) measured the increase of the activities of a silver detector when a source of neutrons was surrounded by U, Al, or Cu. The thicknesses of absorbers used were such as to emphasize any effects attributable to inelastic scattering or  $(n, 2n)$  processes with the Al and Cu. In spite of this, the use of U gave a greater apparent increase of the strength of the source. Since the other processes were presumably of less importance than with Al and Cu the increase had to be attributed to the production of secondary neutrons. A value of 6 neutrons per fission was given, assuming a cross section for fission of  $10^{-25}$  cm<sup>2</sup>.

#### 6. Miscellaneous experimental results

a. Differences between distributions of fission products produced in different ways.—In their early experiments Meitner, Hahn, and Strassmann (37Me5) found that practically the same fission products are produced by thermal neutrons and by fast neutrons. It is apparent from the list of active products of fission in Section II 2 that a great many of these are produced from Th as well as from U. Nevertheless, some differences have been reported.

Joliot (39Jo4) found that in the expelled fission fragments the longer half-lives were relatively favored by the use of thermal neutrons.

Meitner (39Me2) and Bretscher and Cook (39Br2) both found an assortment of half-lives

in the sulphide precipitate obtained from Th which was very different from that obtained from U.

Bjerge, Broström, and Koch (39Bj1) collected the fragments expelled from U and Th which had been produced by both slow and by fast neutrons. They found no appreciable differences between any of the decay curves. Frisch (39Fr3) showed, however, that the observed decay curve is what one would calculate as resulting from the summation of a reasonable, random, statistical distribution of half-lives. There would have to be very large differences between the distribution of the activities produced in different ways for them to be detectable in such a curve of decay of the total activity.

b. Energies of beta-particles.—Meitner  $(37Me2)$ studied the beta-rays of some of the active substances of the sulphide precipitate, getting maximum energies in Mev as follows: 16 min. (3.2), 50 min. (0.6), 2.5 hr. (0.7). Curie, Savitch, and Marques de Silva (38Cu5) found a value of 3.2 Mev for 3.5-hr. La in the same way. Curie and Savitch (38Cu4), by the method of coincidences, found an upper limit of 4.8 Mev for the 16-min. activity, and 3.9 Mev for the 3.5-hr. activity. Barschall et al. (39Ba5) found betaparticles of more than 2.9 Mev for the 0.3—0.9 sec., 3—4-sec. and 11—15-sec. periods. No betaparticles of energy  $\sim$ 8–10 Mev have been found during the irradiation (39He1, 39Ke2, 39Ba5).

c. Gamma-rays.—Meitner (37Me2) found  $\gamma$ rays with the 16-min. and 59-min. activities, the latter being much the stronger. Roberts et al. (39Ro3, 39Ro5) found strong  $\gamma$ -ray emission accompanying the production of delayed neutrons with the 12.5-sec. period. They reported the presence of  $\gamma$ -rays of several longer periods. Booth *et al.* (39Bo8) found periods of  $10-15$  sec., 40 sec., and longer. Mouzon, Park, and Richards (39Mo2) studied the  $\gamma$ -rays with a cloud chamber. They found 118 tracks with energies above 2.2 Mev, some as high as 8 Mev. These experiments were continued by Mouzon and Park (39Mo3). They arranged a cloud chamber exposed to gamma-rays coming from irradiated U so that alternate exposures were made during the neutron bombardment by D-D neutrons and after the bombardment. The ones during the bombardment were made 3 sec. after its beginning, the delayed ones were made  $\frac{1}{2}$  sec. after cessation of a bombardment of 11 sec. About four times as many tracks of electrons having energies over 2 Mev were obtained in pictures of the first set, but the distribution in energy was much the same in both sets. Evidently the greater part of the  $\gamma$ -rays either coincide with the fission or follow very soon after it. They may be connected with the 0.3—0.9-sec. period of Barschall et al. (39Ba5).

d. Alpha-particles from fission products.  $-\text{Meit}$ ner, Hahn, and Strassmann (37Me5) reported the presence of weak alpha-particle emission after removal of all U. Its origin is uncertain.

e. Absence of extremely short half-lives.—Kennedy and Seaborg (39Ke2) looked for coincidences between fissions and beta-particles of energy greater than 1 Mev. None was found, and they conclude that at least 90 percent of the fissions are not accompanied by such betaparticles within  $10^{-6}$  sec. Evidently no substance is formed which has such a short half-life.

f. Fission tracks in the photographic emulsion.— Myssowsky and Idanoff (39My1) found very thick tracks in a photographic plate placed near a layer of U. No such tracks were found with Bi, Au, and Pt.

g. The concentration of 23-minute  $U^{239}$ .—Irvine (39Ir1) has developed a method for concentration of  $23$ -min. U<sup>239</sup> based on the fact that the highly excited ions formed will settle down in the tetravalent state which will give a more insoluble hydroxide than do the uranyl compounds. He achieved a concentration by a factor of 10.

h. Attempts to detect element  $93$ .—The 23-min.  $U^{239}$  must decay to form  $93EkaRe^{239}$ . This is presumably unstable. No activity, either of alpha- or beta-emission has been found from this element. Segre (39Se7) worked with a sample of  $U<sub>3</sub>O<sub>8</sub>$  which had been kept close to the Berkeley cyclotron for two years. He could find no alphaactivity in excess of that from a sample which had not been irradiated. When some of the irradiated sample was dissolved and precipitated with Re no activity could be found in the precipitate. Irvine (39Ir1) also was unable to find any such activity. Thus no transuranic element has yet been observed. It may be

that  $93\text{Exa}$ Re<sup>239</sup> is stable. Hulubei and Cauchois (39Hu2) present evidence for the existence of element 93 in minerals.

 $i.$  Detonation of explosives by fission fragments. Feenberg (39Fe6) found that nitrogen iodide could be detonated by fission fragments from U. On the average only about one fission in a thousand was effective. Such experiments have been continued by Fabre, Magnan, and Muraour (39Fa3).

j. Fission produced by deuteron bombardment.  $-G$ ant (39Ga2) found fission produced by deuteron bombardment of U. By the use of a set of foils of properly chosen thickness he could correct the observed activity for activity which could be attributed to the effects of scattered deuterons, and of neutrons. The decay of the activity attributable to fission was like that found when the fission has been produced by neutrons. In these experiments some fission was produced by the neutrons present, but this amounted to only 1/20 of the observed effect.

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#### **REFERENCES**

When several references are given together, the one first given will be that for a late paper which includes the results of the earlier ones.

#### 1934

- Fe2. E. Fermi, Nature 133, 898.
- Fe4. E. Amaldi, O. D'Agostino, F. Rassetti and E. Segrè, Proc. Roy. Soc. A146, 483.
- No0. I. Noddack, Zeits. f. angew. Chimie. 37, 653.

#### 1935

- Ag1. O. D'Agostino and E. Segrè, Gaz. Chim. Ital. 65, 1088.
- Cu<sub>1</sub>. I. Curie, H. von Halban and P. Preiswerk, J. de phys. [7] 6, 361; C.R. 200, 1841; C.R. 200, 2079.
- Gr3. A. von Grosse and M. S. Agruss, J.Am. Chem. Soc. 57, 438.
- HaO. O. Hahn and L. Meitner, Naturwiss. 23, 320.
- Ha4. O. Hahn and L. Meitner, Naturwiss. 23, 37, and 230.
- Ha7. O. Hahn, L. Meitner and F. Strassmann, Naturwiss. 23, 544.
- Sei. E. Segrè, Nuovo Cim. n.s. 12, 232-239. 1936
- Ha3. O. Hahn, L. Meitner and F. Strassmann, Chem. Ber. 69, 905.
- Ha11. O. Hahn, L. Meitner and F. Strassmann, Chem. Ber. A69, 217.
- Me1. L. Meitner and O. Hahn, Naturwiss. 21, 158.
- Me3. L. Meitner, Zurich Vortragswoche 24—42.
- Ro7. E. Rona and E. Neuninger, Naturwiss. 24, 491.

#### 1937

- Br5. A. Braun, P. Preiswerk and P. Scherrer, Nature 140, 682.
- Ha5. O. Hahn, L. Meitner and F. Strassmann, Chem. Ber. 70, 1374,
- Me2. L. Meitner, Ann. d. Physik [3]29, 246.
- Me5. L. Meitner, O. Hahn and F. Strassmann, Zeits. f. Physik 106, 249.
- Cu<sub>1</sub>. I. Curie and P. Savitch, J. de phys.  $[7]$  8, 385.

#### 1938

- Cu3. I. Curie and P. Savitch, C.R. 206, 906 and 1643.
- $Cu<sub>4</sub>$ . I. Curie and P. Savitch, J. de phys. [7] 9, 355.
- $Cu<sub>5</sub>$ . I. Curie, P. Savitch and A. Marques de Silva, J. de phys. [7] 9, 440.
- Drl. G. Von Droste, Zeits. f. Physik 110, 84.
- Ha8. O. Hahn, L. Meitner and F. Strassmann, Naturwiss. 26, 475.
- Ha14. O. Hahn and F. Strassmann, Naturwiss. 26, 755.
- Me2. L. Meitner, F. Strassmann and O. Hahn, Zeits. f. Physik 109, 538.
- Ni1. Y. Nishina, T. Yasaki, K. Kimura and M. Ikawa, Nature 142, 874.
- Ro5. E. Rona, H. Scheichenberger and R. Stangl, Akad. d. miss. Wien Sitz. II A, 147, 209.

#### 1939

- Ab4. P. Abelson, Phys. Rev. 56, 1; 55, 418; 55, 670; 55, 876.
- Adi. F. Adler and H. von Halban, Jr., Nature 143, 793.
- Ad2. M. F. Adler, C.R. 209, 301.
- Ani. H. L. Anderson, E. T. Booth, J. R. Dunning, E. Fermi, G. N. Glasoe and F. G. Slack, Phys. Rev. 55, 511.
- An2. H. L. Anderson, E. Fermi and H. B. Hanstein, Phy. Rev. 55, 797.
- An3. H. L. Anderson and E. Fermi, Phys. Rev. 55, 1106.
- An4. H. L. Anderson, E. Fermi and L. Szilard, Phys. Rev. 56, 284.
- Ati. A. H. W. Aten, Jr., C. J. Bakker and F. A. Heyn, Nature 143, 679.
- Ba5. H. H. Barschall, W. T. Harris, M. H. Kanner and L. A. Turner, Phys. Rev. 55, 989.
- Be2. G. Beck and P. Havas, C.R. 208, 1084.
- Be3. G. Beck and P. Havas, C,R. 208, 1643.
- Bji. T. Bjerge, K. J. Brostrøm and J. Koch, Nature 143, 794.
- Bo1. N. Bohr, Nature 143, 330.
- Bo2. N. Bohr, Phys. Rev. 55, 418.
- Bo6. E.T. Booth, J.R. Dunning and G. N. Glasoe, Phys. Rev. 55, 982.
- Bo7. E.T. Booth, J. R. Dunning and F. G. Slack, Phys. Rev. 55, 981.
- Bo8. E.T. Booth, J. R. Dunning and F. G. Slack, Phys. Rev. SS, 876.
- Bo9. N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426.
- Bo12. N. Bohr and J. A. Wheeler, Phys. Rev. 56, 1065.
- Br2. E. Bretscher and L. G. Cook, Nature 143, 559.
- Br9. K. Brostrgm, J. Koch, and T. Lauritsen, Nature 144, 830.
- Ch3. W. G. Chlopin, M. A. Passwik-Chlopin, and N. F. Wolkov, Nature 144, 595.
- Co5. D. R. Corson and R. L. Thornton, Phys. Rev. 55, 509.
- Cu1. I. Curie and P. Savitch, C.R. 208, 343.
- Do<sub>1</sub>. M. Dode, H. von Halban, Jr., F. Joliot and Lew. Kowarski, C.R. 208, 995.
- Do<sub>2</sub>. R. W. Dodson and R. D. Fowler, Phys. Rev 55, 880.
- Dr1. G. Von Droste, Naturwiss. 27, 198.
- Dr2. G. Von Droste and H. Reddemann, Naturwiss. 27, 371.
- Fai. U. Fano, J. de phys. 10, 229.
- Fa3. P. Fabre, C. Magnan, and H. Muraour, C. R. 209, 436.
- Fei. N. Feather and E. Bretscher, Nature 143, 516.
- Fe2. E. Feenberg, Phys. Rev. 55, 504.
- Fe4. N. Feather, Nature 143, 597.
- Fe6. E. Feenberg, Phys. Rev. 55, 980.
- Fe7. N. Feather, Nature 143, 1027.
- Fe8. B. Ferretti, Ricerca Scient. 10, 332.
- Fli. S, Flugge and G. Von Droste, Zeits, f. physik, Chemic 13, 42, 274.
- F12. S. Flügge, Naturwiss. 27, 402.
- Foi. R. D. Fowler, and R. W. Dodson, Phys. Rev. 55, 417.
- Fr1. O. R. Frisch, Nature 143, 276.
- Fr2. J. Frenkel, Phys. Rev. 55, 981'.
- Fr3. O. R, Frisch, Nature 143, 852.
- Ga2. D. H. T. Gant, Nature 144, 707.
- Gi1. D. F. Gibbs and G. P. Thomson, Nature 144, 202.
- Gli. G. N. Glasoe and J. Steigman, Phys. Rev. 55, 982.
- Go0. H. H. Goldsmith, V. W. Cohen and J. R. Dunning, Phys. Rev. 55, 1124.
- Go1, L. Goldstein, A. Rogozinski and R. J. Walen, Nature 144, 201.
- Gri. G. K. Green and L. W. Alvarez, Phys. Rev. 55, 417.
- Gr2. A. von Grosse, E. T. Booth and J. R. Dunning, Phys. Rev. 56, 382.
- Hai. O. Hahn and F. Strassmann, Naturwiss. 27, 11.
- Ha5. O. Hahn and F. Strassmann, Naturwiss. 27, 89.
- Ha6. C. Haenny and A. Rosenberg, C. R. 208, 898.
- Ha7. H. von Halban, Jr., F. Joliot and L. Kowarski, Nature 143, 470.
- Ha8. O. Hahn and F. Strassmann, Naturwiss. 27, 163.
- Ha9. H. von Halban, Jr., F. Joliot and L. Kowarski, Nature 143, 680.
- Ha10. H. von Halban, Jr., L. Kowarski and P. Savitch, C.R. 208, 1396.
- Ha11. H. von Halban, Jr., F. Joliot and L. Kowarsk Nature 143, 939.
- Hai2. O. Hahn and F. Strassmann, Naturwiss. 27, 451.
- Ha13. O. Haxel, Zeits. f. Physik 112, 681.
- Ha14. 0, Hahn and F. Strassmann, Naturwiss. 27, 529.
- Ha15. O. Hahn, F. Strassmann and S. Flügge, Naturwis 27, 544.
- He1. F. A. Heyn, A. H, W. Aten, Jr. and C. J. Bakker, Nature 143, 516.
- He2. M. C. Henderson, Phys. Rev. 56, 703.
- Hu2. H. Hulubei and Y. Cauchois, C. R. 209, 476.
- Iri. J. W. Irvine, Jr., Phys. Rev. 55, 1105.
- Jei. W. Jentschke and F. Prankl, Naturwiss. 27, 134.
- Jo2. F. Joliot, C.R. 208, 341.
- Jo3. F. Joliot, C.R. 208, 647.
- Jo4. F.Joliot, J. de phys. (7) 10, 159.
- Ke2. J. W. Kennedy and G. T. Seaborg, Phys. Rev. 55, 877.
- Lai. R. Ladenburg, M. H Kanner, H. Barschall and C. C. Van Voorhis, Phys. Rev. 56, 168.
- La2. A. Langsdorf, Jr., Phys. Rev. 56, 205.
- Li1. C. Lieber, Naturwiss. 27, 421.
- Ma4. C. Nagnan, C.R. 208, 742.
- Ma11. C. Magnan, C.R. 208, 1218.
- Mc5. E. McMillan, Phys. Rev. 55, 510.
- Me1. L. Meitner and O. R. Frisch, Nature 143, 239.
- Me2. L. Meitner and O. R. Frisch, Nature 143, 471.
- Me3. L. Meitner, Nature 143, 637.
- Mi1. J. L. Michiels, G. Parry and G. P. Thomson, Nature 143, 760.
- Mo2. J. C. Mouzon, R. D. Park and J. A. Richards, Jr., Phys. Rev. 56, 668.
- Mo3. J. C. Mouzon and R. D. Park, Phys. Rev. 56, 238.
- My1. L. Myssowsky and A. Idanoff, Nature 143, 794.
- Ni1. A. O. Nier, Phys. Rev. 55, 150.
- Ni3. Y, Nishina, T. Yasaki, H, Ezoe, K. Kimura, and M. Ikawa, Nature 144, 547.
- Pe2. F. Perrin, C.R. 208, 1394 and 1573.
- Rei. H. Reddemann and H. Bomke, Naturwiss. 27, 518.
- Ro2. R. B. Roberts, R. C. Meyer and L. R. Hafstad, Phys. Rev. 55, 416.
- RO3. R. B. Roberts, R. C. Meyer and P. Wang, Phys. Rev. 55, 510.
- RO5. R. B. Roberts, L. R. Hafstad, R. C. Meyer and P. Wang, Phys. Rev. 55, 664.
- Ro9. R. B. Roberts and J. B. H. Kuper, J. App. Phys. 10, 612.
- Ro6. J. Rotblat, Nature 143, 852.
- Sa3. P. Savitch, C.R. 208, 646.
- Se7. E. Segre, Phys. Rev. 55, 1104.
- Soi. J. Solomon, C.R. 208, <sup>570</sup> and 896.
- Sz2. L. Szilard and W. H. Zinn, Phys. Rev. 55, 799.
- Thi. J. Thibaud and A. Moussa, C.R. 208, 652.
- Th2. J. Thibaud and A. Moussa, C.R, 208, 744.
- Th5. J. Thibaud and A. Moussa, J. de phys. (7}, 10, 388.
- $We2.$ W. Wefelmeier, Naturwiss. 27, 110.
- We3. C. F. Weizsacker, Naturwiss. 27, 133.
- Wh1. M. D. Whittaker, C. A. Barton, W. C. Bright and E.J. Murphy, Phys. Rev. 55, 793.
- Y1. G. Young, Phys. Rev. 55, 1102.
- Zi3. W. H. Zinn, and L. Szilard, Phys. Rev. 56, 619.