# Story of Fission

## Unlocking Power of the Nucleus

*Amit Roy*

**The discovery of nuclear fission is the culmination of sustained efforts involving many scientists led by Hahn and Meitner to understand the production of artificial radioactivity induced by neutrons bombarding uranium. The large energy release in this process was almost immediately used for both military and civilian purposes. It also started the large-scale funding of scientific research by governments across the world. Nuclear energy is one of the clean sources of energy and contributes very little to global warming.**

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The discovery of fission of uranium in 1939 changed forever the way society at large supported scientific research. Till that time, individual researchers or small groups would pursue their subjects of interest with whatever resources they could muster either from government or private individuals. With fission promising the prospect of energy release of unprecedented amounts, governments all across the globe started funding research in a big way leading to big organised research by large teams.

How did it all begin? It all started with the chance discovery of radioactivity in 1896 by Henri Becquerel, which paved the way for the study of matter at the atomic scale for the first time. J J Thomson showed that atoms were no longer indivisible with his discovery of the electron in 1897. The radiations emitted by radioactive elements were identified in the works of Ernest Rutherford and Frederick Soddy at Montreal and Pierre Curie in Paris. They found that these radiations carried unprecedented amounts of energy. Work on radioactivity was carried on fruitfully by Pierre and Marie Curie, who discovered the elements radium and polonium. Rutherford took the next major step in the progress towards understanding the heart of the matter. Rutherford, from the experiments performed with his colleagues Hans

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Geiger and Ernest Marsden, discovered the nucleus in 1911. Investigating further, Rutherford and Blackett discovered transmutation of elements in 1919 when they observed proton tracks on bombarding nitrogen with alpha particles in the reaction $^{14}N + \alpha \Rightarrow p + {}^{17}O$.

The stage was set for rapid progress towards exploration of the nuclear world with the discovery of neutron by Rutherford's student, James Chadwick in 1932.

A flurry of activities started when Irene Curie and Frederick Joliot discovered artificial radioactivity by bombarding aluminium with alpha particles producing radioactive phosphorous in their laboratory at Paris in 1934. In his laboratory at Rome, Enrico Fermi with his band of students used the recently discovered neutron to bombard almost every available element, producing artificial radioactivity in many elements. The most significant discovery was that the artificial radioactivity induced by slow neutrons was very much larger than that by fast neutrons. These experiments have been described in detail in the article on Fermi in *Resonance* [1].

Fermi had found that in all the heavy elements, n-bombardment resulted in one of the following scenarios: (i) capture of the neutron leading to a heavier isotope of the element or (ii) the next element with higher proton number following beta decay, or (iii) the emission of a proton or alpha particle. These processes generated radioactive products usually with only one or two different half-lives. But, in case of n-bombardment of uranium, activities[1] with a large number of different decay half-lives were measured. Since all of these products emitted beta rays and none of these had chemical properties similar to that of U or nearby elements, Fermi and collaborators attributed these activities to the production of new transuranic elements with Z = 93 and 94. Aristid von Grosse, working in Chicago, repeated the experiment and thought that one of the elements with half-life of 13 minutes produced by n-bombardment of U behaved like protactinium (Pa).

[1] The radioactive nuclei produced in a nuclear reaction such as neutron irradiation are termed 'activities'.

Ida Noddack (a German chemist who, with her husband Walter, had discovered the element rhenium) made a criticism of Fermi's conclusion and suggested that "when heavy nuclei are bombarded by neutrons, it is conceivable that the nucleus breaks up into several large fragments...". However, she did not suggest any mechanism for this type of a break up. She wrote to both Fermi and Hahn but her suggestion was considered too preposterous and unlikely and was ignored. She also did not pursue this line of thought any further.

Otto Hahn and Lise Meitner had discovered the element Pa in Berlin. Aristid von Grosse had worked with Hahn earlier in Berlin on Pa and had differences with Hahn before leaving for US. Hahn and Meitner decided to further cross-check the identification of Pa in n-bombardment of U. They had little trouble in showing that von Grosse was wrong by using the method of co-precipitation (*Box* 1). The task of identification of all the activities was not at all easy since the chemistry had to be done with very tiny amounts of the unknown activities in the presence of much more intense U activity. They asked Fritz Strassmann to join them in their quest. They followed traditional analytical methods of chemical separation and precipitation whenever possible. But in all their analyses, they concentrated only on the identity of the 'transuranes', i.e., elements they thought to be heavier than uranium. To do this, they dissolved the products of n-bombardment of U and separated out the 'transuranes' from the solution for follow-up. They never looked at the leftover solute for any activity since it contained the dominant natural activity from U. Meitner used cloud chamber

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**Box 1. Co-precipitation**

This is a procedure where a compound of an element is added to precipitate out all elements in the same group out of a solution containing a mix of many elements. Hahn and Meitner used this technique to separate the activities corresponding to different group elements. Expecting the 'transuranes' to be similar to Re and Pt, they added potassium perrhenate and platinum chloride to the solution containing all the products of n-bombardment of U, and then precipitated out the elements with Z = 90, 91 and 92 with sodium hydroxide. The higher Z elements were expected to remain in solution that could be co-precipitated with Re and Pt by passing hydrogen sulphide through the solution to form corresponding sulphides. Similarly, Pa was ruled out by adding the known element Pa to the solution and precipitating it out completely. No activity corresponding to Pa was found.

1. $_{92}U + n \rightarrow (_{92}U + n)$ [β, 10 sec] $\rightarrow$ $_{93}$EkaRe [β, 2.2 min] $\rightarrow$ $_{94}$EkaOs [β, 59 min] $\rightarrow$ $_{95}$EkaIr [β, 66 hr] $\rightarrow$ $_{96}$EkaPt [β, 2.5 hr] $\rightarrow$ $_{97}$EkaAu?

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

3. $_{92}U + n \rightarrow (_{92}U + n)$ [β, 23 min] $\rightarrow$ $_{93}$EkaRe?

**Figure 1.** The term 'eka' was used first by Mendeleev for naming elements he found similar to others in chemical properties and put them in the same group in the periodic table. For example, Germanium (Ge) was termed Eka Silicon (Si). Origin from Sanskrit, *ekam* = same/similar.

photographs to establish that the 'transuranes' emitted only beta particles. They identified nine radioactive species based on their decay half-lives and proposed three chains of radioactive elements resulting from the n-bombardment of U in their publication in *Zeitschrift fuer Physik* and *Chemische Berichte* in 1937 [2]. These are shown below as they appeared in their publication. Out of these, only process 3 identifying the 23 min activity to U was on firm ground (*Figure* 1).

The results were unusual on two counts: first, there were three parallel chains and secondly, the length of the first two chains exceeded hitherto all known beta decay chains.

The curious results on radioactivity induced in U by neutrons also attracted the attention of Irene Curie and her collaborator Pavel Savitch in Paris. Instead of chemically separating the 'transuranes', they measured the activities present in the entire solution. They used a pair of ion chambers with absorbers of different thicknesses to eliminate the effect of natural activity of U. They found the known 40-second, 2-minute, and 16-minute activities, and in addition, a new strong 3.5-hour activity with the highest energy beta rays. They selected this activity by covering their samples with thick sheets of brass and measuring only the radiation penetrating the brass sheet. Since the 3.5-hour activity remained in the filtrate after the transuranes were precipitated out and this activity could be separated from uranium, it was, therefore, neither a 'transurane' nor uranium. First they postulated [3] it to be an isotope of thorium (Th) from its chemical behaviour in their publication. The Berlin group repeated their measurements but could not find any trace of Th; after they informed Curie about

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their negative result, Curie and Savitch retracted their Th result. After more tests, Curie and Savitch found the activity to behave more like actinium (Ac). This was not considered very likely by others as it would require emission of an alpha particle on n-bombardment and that was considered highly unlikely due to the Coulomb barrier.

Around this time, Meitner asked her student Gottfried von Droste to look for long-range alpha particles from U during n-bombardment using an ionization chamber. Droste used an aluminium foil to stop the alpha particles naturally emitted by U and did not observe any long-range alpha particles, which had to be present if any radium or actinium isotopes were produced by n-bombardment. If only Droste had not used the foils to stop the natural alpha particles, the fission fragments would not be stopped and the ionization chamber would have recorded the fragments.

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Curie and Savitch persisted in studying the 3.5 hr activity and in May 1938 found that this activity could be precipitated with a lanthanum (La) carrier and could be separated from Ac completely. However, they did not firmly identify the activity with La and in their publication with the French Academy of Sciences, wrote, "It appears, therefore, that this substance can only be a transuranic element, with properties very different from those of the other known transuranic elements."

In the meanwhile, Lise Meitner lost her job at the Kaiser Wilhelm Institute due to the policies of the Nazi regime and left Germany in July, 1938 for Holland without a visa and then to Sweden. Strassmann and Hahn continued with the experiments at Berlin and Hahn continued to inform Meitner about their results through his letters.

Strassmann thought that the 3.5-hour activity could be an isotope of radium (Ra) and decided to separate out this activity with their refined chemical analysis that resulted in much lower contaminations. First they removed the transuranic elements from the solution and found the activities to resemble radium isomers and their actinium decay products. Hahn reasoned in their publication

**Box 2. Fractional Crystallization**

This is a method of purifying substances based on differences in solubility. Marie Curie had used this process for isolating radium from the ore containing barium by converting radium and barium to their chloride and bromide salts. In this process, the temperature of the solution containing a mixture of two or more substances in solution is reduced resulting in crystallization; the least soluble substance would crystallize first. The proportion of components of the mixture in the precipitate will depend on their solubility products. If the solubility products are very similar, the crystallization process has to be repeated a number of times for a complete separation.

in the November issue of *Naturwissenschaften* [4] that the formation of radium was the result of emission of two alpha particles successively, represented as

$$^{238}\mathrm{U} + {}^{1}\mathrm{n} \rightarrow \alpha + {}^{235}\mathrm{Th} \rightarrow {}^{231}\mathrm{Ra} + \alpha.$$

Among the products, they listed three radium and three actinium isomers. Hahn and Strassmann had found, as had Curie and Savitch before them, that the process leading to radium was significantly enhanced when slow neutrons were used. The emission of alpha particles with a high probability was hard to understand, especially for slow neutrons since the alpha particles would face the Coulomb barrier, and this was criticized by Bohr and Meitner when Hahn visited Copenhagen in November 1938.

Hahn and Strassmann decided to reinvestigate the evidence for radium. From the chemical reactions, these could only be either radium or barium. They tried to separate artificial "radium" from the inactive barium ballast material by fractional crystallization (*Box* 2). Despite the most varied techniques, Hahn and Strassmann failed to separate the "radium" from the barium. Starting with a solution of the presumed barium–radium mixture, Hahn and Strassmann added bromide to the solution in four steps; with each step, a fraction of the barium (and radium) would precipitate as crystals of barium bromide. Because radium was known to co-precipitate preferentially with barium bromide, i.e., the proportion of radium that precipitated was larger than the proportion of radium in solution, the first barium bromide fraction was expected to be richer in radium than those that followed. To their surprise, Hahn and Strassmann measured no difference at all: the "radium" activities were evenly distributed among the successive barium bromide fractions. The sequence of the reactions they followed is schematically given in *Figure* 1. They identified the

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**"RaI"? [β, <1 min] → AcI [β, <30 min] → Th?**
**"RaII"? [β, <14+2 min] → AcII [β, ~2.5 h] → Th?**
**"RaIII"? [β, 86+6 min] → AcIII [β, ~several days] → Th?**
**"RaIV"? [β, 250-300 h] → AcIV [β, <40 h] → Th?**

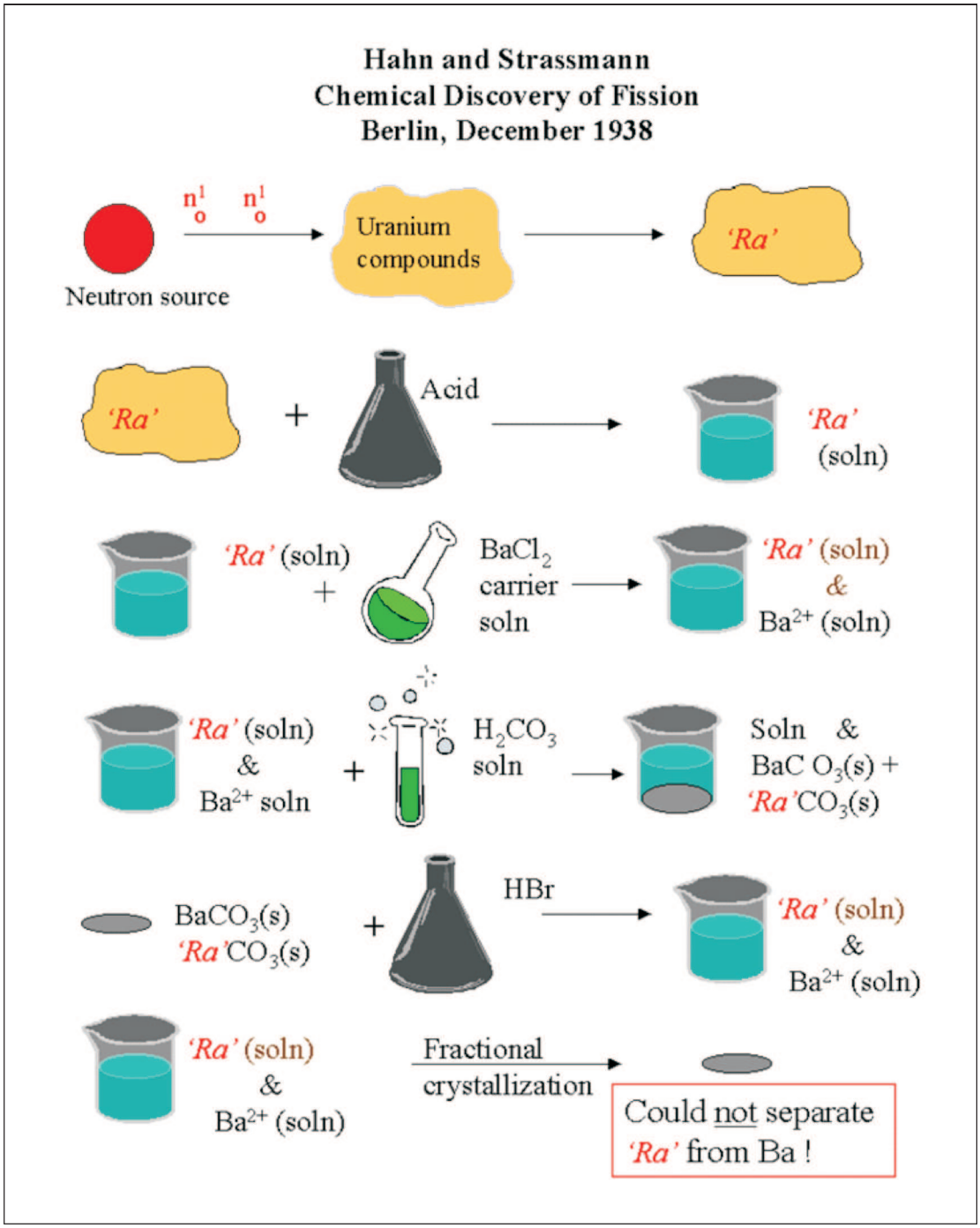


**Figure 1.** Schematic of the sequence of experiments leading to the discovery of fission by Hahn and Strassmann. (From http://chemwiki.ucdavis.edu/)

following sequence of decays:

Thinking that something might have gone wrong with their procedure, they ran control experiments using known radium isotopes. They carried out two separate fractionations adding natural radium isotopes, mesothorium 1 and 2; $MsTh_1$($^{228}$Th) and $MsTh_2$($^{228}$Ac) as tracers to the long-lived activity they called Ra IV. On using $MsTh_1$, they found that with barium bromide, the concentration of $MsTh_1$ was increased whereas that of Ra IV was not, and the activity remained at the same level for fractions with equivalent barium content. Ra IV was behaving as barium itself.

Following this, Hahn wrote [5] to Lise Meitner on 19 December, 1938.

> "...But we are coming steadily closer to the frightful conclusion: our Ra isotopes do not act like Ra but like Ba. ... All other elements, transuranes, U, Th, Ac, Pa, Pb, Bi, Po are out of the question. I have agreed with Strassmann that for now we shall tell only *you. Perhaps you can come up with some sort of fantastic explanation. We know ourselves that it can't actually burst apart into Ba. Now* we want to test whether the Ac-isotopes derived from the "Ra" behave not like Ac but La."

Similarly, for the activity labelled Ac II, they found on adding pure actinium isotope $MsTh_2$ ($^{228}$Ac) and precipitating with lanthanum oxalate, the $MsTh_2$ was concentrated whereas AC II was not, leading to the inexorable conclusion that it was lanthanum itself. They submitted their findings to *Naturwissenschaften* on 22 December and it was published on 6 January 1939 [6], with the very hesitant conclusion given below.

> "We come to the conclusion that our 'radium isotopes' have the properties of barium. As chemists we should actually state that the new products are not radium, but rather barium itself.... As chemists we really ought to revise the decay scheme given above and insert the symbols Ba, La, Ce, in place of Ra, Ac, Th. However, as nuclear chemists, working

very close to the field of physics, we cannot bring ourselves yet to take such a drastic step which goes against all previous experience in nuclear physics. There could perhaps be a series of unusual coincidences which has given us false indications." (Translated by H G Graetzer) [7].

Otto Robert Frisch went to visit his aunt, Meitner in Sweden during Xmas vacation. Meitner had meanwhile received Hahn's letter with the irrefutable evidence for production of barium from uranium on neutron irradiation. While taking a stroll in the snow, the idea of splitting of a nucleus came to Meitner and Frisch on the basis of Bohr's 'liquid drop model'[2]. According to this model such a drop might, on getting excited, execute collective vibrations leading to elongation, and divide itself into two smaller drops. They worked out the energy that would be released in such a process following two methods. One was to consider the change in the binding energy in this process from the mass defects and the other was to calculate the electric repulsion energy between the fragments from their atomic numbers. The two calculations matched and gave approximately an energy release of 200 MeV.

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Frisch returned to Copenhagen and found Bohr getting ready to leave for America. He describes what transpired between him and Bohr in a letter to his aunt [5],

"Dear Tanterl, I was able to speak with Bohr only today [3 January] about the splitting of uranium. The conversation lasted only five minutes as Bohr agreed with us immediately about everything. He just couldn't imagine why he hadn't thought of this before, as it is such a direct consequence of the current concept of nuclear structure. He agreed with us completely that this splitting of a heavy nucleus into two big pieces is practically a classical phenomenon, which does not occur at all below certain energy, but goes readily above it...."

Frisch got busy, following a suggestion of George Placzek, to measure the fragments released in the splitting of U directly. In Frisch's own words [8],

[2] Niels Bohr and Fritz Kalcker postulated the liquid drop model to explain the collective behaviour of a nucleus. A nucleus exhibits many collective features similar to those of a liquid drop, e.g., vibrations, rotations, evaporation of smaller particles on heating.

> "I rigged up a pulse amplifier for the special purpose, and I also built a small ionization chamber; but the whole thing only took me about two days, and then I worked most of the night through to do the measurements because the counting rates were very low. But by three in the morning I had the evidence of the big pulses."

Following a caution from Meitner about the presence of large background from natural alpha decay of U, which her assistant Droste had suppressed by using aluminium foil (thereby stopping the fragments as well), Frisch electronically biased his counters against detecting the natural alphas.

Frisch had used a U-lined ionization chamber and a Ra$\alpha$ + Be source of neutrons, for measuring the fragment pulses. Following a caution from Meitner about the presence of large background from natural alpha decay of U, which her assistant Droste had suppressed by using aluminium foil (thereby stopping the fragments as well), Frisch electronically biased his counters against detecting the natural alphas. He set a threshold of $> 5 \times 10^5$ ion pairs so that it could count the relatively rarer fragments from the splitting of U. This is because the binary fission fragments carry an energy of roughly 100 MeV each and will produce a large number of ion pairs. (The energy required for creation of an ion pair is typically 30 eV and hence, complete stopping of one of the 100 MeV fission fragments should yield approximately $3 \times 10^6$ ion pairs. The natural alphas emitted by U having typically 5 MeV energy when fully stopped, will produce only $10^5$ ion pairs). He observed about 15 such large pulses per minute in his set-up. Presence of paraffin around the neutron source doubled the rate of the large fragments. Substituting U by Th also resulted in large pulses, except that surrounding the neutron source with paraffin reduced the count rate in the case of Th.

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Frisch communicated two papers to *Nature* and these were published in consecutive issues of *Nature*. The first one [9] was the paper by L Meitner and O R Frisch, in which the term 'fission' occurs for the first time. Frisch had asked an American biologist, William A Arnold working in von Hevsey's laboratory about the biological name for cell division. The second paper was by Frisch alone on 'Physical Evidence for the Division of Heavy Nuclei under n-bombardment' [10].

Bohr worked on the idea of splitting of U during the six-day journey by sea and within this short span of time he had the sketch of a theory. After reaching USA he worked with John Wheeler on the theory of fission that was published in *Physical Review* in 1939 [11]. In this paper, Bohr and Wheeler also pointed out that slow neutrons cause fission in $^{235}U$ and fast neutrons in $^{238}U$. Immediately experiments were conducted successfully to verify the predictions of this theory by Alfred O. Nier, E.T. Booth, J.R. Dunning and A.V. Grosse at Columbia University in 1940 [12]. It led to a flurry of activity in all leading laboratories of the world including Bose Institute, Kolkata [13].

Subsequent work on the demonstration of chain reaction in a nuclear reactor carried out under the leadership of Fermi and the development of the atomic bomb have been described in detail in a previous article on Fermi in *Resonance* [1].

Otto Hahn received the Nobel Prize for Chemistry in 1944 "for his discovery of fission of heavy nuclei". It was announced only after the end of World War II in 1945. Many including this author feel that both Lise Meitner and Fritz Strassmann should have shared the award.

Electricity was generated for the first time from a nuclear reactor at Idaho, USA on 20 December 1951. Today more than 10% of the world's electricity needs are met by nuclear reactors employing nuclear fission as the source of heat energy. In India we have nearly 20 reactors generating about 3% of the electricity. Discovery of nuclear fission is a prime example of fundamental research leading to a phenomenon that has given rise to technology of lasting value to society at large. Even after 75 years of its discovery, this field of research continues to be an enigma, a process whose finer details are still being investigated. The happenings in this exciting field of research were highlighted in the recent conference on 75 years of nuclear fission [14].

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