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A Brief History of the *Delayed* Discovery of Nuclear Fission

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Abstract

This year marks the Fiftieth Anniversary of the discovery of Nuclear Fission. In the early 1930's, the neutron was discovered, followed by the discovery of artificial radioactivity and then the use of the neutron to produce artificial radioactivity. The first experiments resulting in the fission of uranium took place in 1934. A paper which speculated on fission as an explanation was almost immediately published, yet no one took it seriously not even the author herself. Why did it take an additional five years before anyone realized what had occurred? This is an abnormally long time in a period when discoveries, particularly in nuclear physics, seemed to be almost a daily occurrence.

The events which led up to the discovery are recounted, with an attempt made to put them into their historical perspective. The role played by Mendeleev's Periodic Table, the role of the natural radioactive decay chain of uranium, the discovery of protactinium, the apparent discovery of masurium (technetium) and a speculation on the reason why Iréne Curie may have missed the discovery of nuclear fission will all be discussed.

Proloque

In reviewing historical developments, it is difficult to ignore one's own knowledge of events subsequent to the historical period being studied. In this case – the discovery of nuclear fission – one is aware of the present form of the latest version of the Periodic Table and the Chart of the Nuclides. We now have available separated isotopes, sophisticated experimental instrumentation off the shelf, methods of radiochemical separation of elements and of mass separation of the isotopes of any chemical element, extensive computer databases giving the properties of targets, projectiles and reactions as well as the latest versions of all the theories of nuclear matter. This makes it all the more difficult to appreciate the problems that scientists might have encountered in a prior time period. To understand why five years elapsed after the experimental realization of nuclear fission before scientists discovered what had happened, it is necessary to go back in time and try to describe the atmosphere that the scientists were dealing with in those years.

Early Background

Let us begin by going back in time, not 50 years but 120 years. Between 1869 and 1871, the Russian chemist Dmitri Mendeleev proposed his form of the Periodic Table of the Chemical Elements. Although others had preceded him by decades in presenting and developing portions of such a Table, only Mendeleev is now associated with it because he used the periodic properties of the elements as a function of their atomic weight values to revise measured values and to predict the properties of undiscovered elements[1]. Over the next fifteen years, all three of his predicted elements were discovered, *i.e.*, gallium (Mendeleev's eka–aluminum), scandium (eka–boron), and germanium (eka–silicon), where Mendeleev used the prefix 'eka', 'dvi' and 'tri' (*Sanskrit: one, two, three*) to indicate an element with similar properties in the next, second, or third removed period (next higher Z elements in the same group) of the Table.

When Lecoq de Boisbaudran discovered gallium in 1879[2], he measured a value of 4.7 for the density. However, Mendeleev insisted that this element was his eka–aluminum and that de Boisbaudran's value was incorrect and should be closer to his prediction of 5.9. To answer Mendeleev, de Boisbaudran obtained 5.956 for the density on remeasurement. Scientists were extremely impressed by the fact that Mendeleev knew more about a new element from theory than the discoverer of that element did from his experiment. Scientists began to rely on the Periodic Table as an aid in the discovery of new elements.

Toward the close of the Nineteenth Century, a prevailing view was that 'all processes of science had been discovered and the only thing left to do was to extend the values of physical parameters to the next decimal place'.

The Twentieth Century

By the start of the Twentieth Century, the discovery of radioactivity, x-rays, and radioactive transformations heralded the beginning of a new age. In 1913, the English physicist Henry G. J. Moseley compared the energy of the x-ray spectral lines of various elements against their atomic weight and obtained an approximate straight line[3]. To avoid breaks in this graph, it was necessary to place argon-potassium, cobalt-nickel, and tellurium-iodine in the order demanded by chemical properties rather than increasing atomic weight. According to his curve of x-ray lines each element has a constant value, its atomic number (nuclear charge = Z), which increases by a constant amount from element to element. A major activity for scientists seemed to be the attempt to fill in the missing places in Mendeleev's Table using Moseley's law to determine the atomic number. One problem was the rare earth elements which did not seem to fit in, so they were usually omitted. Danish physicist Niels Bohr[4] proposed the electronic orbital structure of the atom indicating the filling of the inner electron shells, which helped to clarify the position of these elements.

In 1913, Lise Meitner and Otto Hahn, a German physicist and chemist respectively, decided to search various ores to locate the immediate precursor of actinium. Hahn had previously discovered radio-thorium, ^{228}Th, as well as meso-thorium, ^{228}Ra, and he had developed expertise in separating radium from other radioactive substances. He futilely attempted to separate meso-thorium and radium without success. It would prove useful later. Actinium was a very scarce element with an unknown atomic weight and an uncertain chemistry. It headed a natural decay chain, which was distinct from the uranium-radium series but was found in uranium bearing minerals in amounts proportional to uranium. Protactinium was discovered in 1918 after a painstaking search, which Meitner continued on her own between 1916 and 1918 while Hahn was in the Army heading the German chemical warfare effort under Fritz Haber. The name *Lisottonium* was suggested for this element after the discoverers, but they declined[5].

In 1922, Bohr applied his theory and the periodic table to help discover an element. In 1911, the French chemist George Urbain claimed the discovery of an element with Z = 72, 'celtium', having properties similar to those of the rare earth elements[6]. Bohr knew his theory required element 72 to have properties similar to those of zirconium and not of the rare earths. He recommended to scientists working at his Institute in Copenhagen that they search for the missing element in zirconium ores. They found hafnium[7]; it was later shown that 'celtium' had resulted from a mixture of other rare earth elements.

In 1925, Berlin chemists Ida Tacke and Walter Noddack claimed[8] the discovery of eka-manganese and dvi-manganese, which they called masurium (Z=43) and rhenium (Z=75). They observed Moseley's x-ray lines for these elements in various ores selected on the basis of possible overlap with the hypothetical properties of the missing elements. Rhenium was later confirmed by others, but masurium never was. In 1937, Emilio Segre and co-workers discovered technetium (Z=43) in molybdenum samples[9] that had been supplied by the Berkeley California

physicist, Ernest Lawrence, who had bombarded the samples with deuterons (heavy hydrogen nuclei) at his cyclotron. Since it is now known that there are no stable isotopes of element 43, the Tacke–Noddack claim to have observed x–rays from element 43 in naturally occurring ore is no longer accepted; Segre is now accepted as the discoverer. Pieter van Assche[10] has recently claimed that the discrediting of the masurium discovery caused scientists to ignore a 1934 paper by Ida Tacke Noddack, which as we shall see, could have explained Fermi's first fission measurement in 1934. Van Assche went on to defend the original paper on masurium by observing that the ore in which the x–rays were detected had a significant amount of uranium and the spontaneous fission process in ^{238}U could explain the existence of masurium as a fission product in the ore. Whether or not Noddack's claim is considered valid, Segre only discovered technetium in 1937. In any case, the non acceptance of the masurium claim could not have affected judgments about Noddack's fission proposal paper in 1934, three years earlier.

The Fateful and Event–full Decade

As the 1930's began, a burst of activity marked the beginning of the era of nuclear physics. At that time, the nucleus was thought to be made up of both protons and electrons. The existence of electrons in the nucleus was introduced to explain the β^- (negative–electron) decay of natural radio–isotopes. In 1919, New Zealand physicist Ernest Rutherford, who was first to split an atomic nucleus, was appointed to head the Cavendish Laboratory at Cambridge University, after the retirement of the discoverer of the electron, J.J. Thomson. In a 1920 Bakerian lecture[11], Rutherford said that there must exist in nature a particle with the same mass as a hydrogen atom, but with a zero electrical charge, to explain the phenomenon of radioactivity. French physicist Frédéric Joliot, later indicated that he had never bothered to read Rutherford's remarks (much to his regret) since he wrongly assumed that a public lecture would contain the usual display of oratory but no new ideas[12]. In addition, theoretical work was never highly regarded in the Paris laboratory where Joliot worked. His mother–in–law, the double Nobel prize winner Marie Curie, once responded to a theoretical physicist's recommendation that a particular experiment be performed with a comment that they might even perform the experiment in spite of that suggestion[13].

Rutherford and his chief assistant, James Chadwick, spent ten years trying to find the neutron without success. In 1932, Joliot and his chemist wife, Irène Curie, reported that α particles from a polonium source bombarding a beryllium target had produced radiation that knocked protons out of hydrogen atoms. The Joliots thought that they had observed a γ–ray Compton effect[14], even though there were theoretical problems with that assumption of γ–rays. Theory was not highly regarded at the Paris lab as was just mentioned. Chadwick did not believe that this radiation was γ–rays. He proposed that the observed radiation was the elusive neutron that Joliot had not previously heard about. He proceeded to verify this assumption[15] and is now credited as the discoverer of the neutron.

Having missed this discovery, Joliot and Joliot–Curie were not to miss their next opportunity. Two years later in 1934, they bombarded aluminum with α particles from polonium and the emission of β^+ (positrons – positively charged electrons) did not cease immediately when the polonium source was removed from the target, but continued to be emitted with an exponential decay characteristic of natural radionuclides[16]. Ernest Lawrence had noted the same phenomenon using his cyclotron as the source of charged particles. He found that his counters misbehaved after the cyclotron was shut off. However, he 'corrected' the malfunction by arranging to automatically shut off the counters as soon as the cyclotron was turned off[17].

In those days, Geiger counters were very erratic. Joliot thought – "either my counter is not working or I have made a fundamental discovery". He was just leaving for a dinner party and asked a colleague to check out the counter and leave him a note. The Joliots arrived at the laboratory in the morning to find a message stating that the counter was working perfectly[18]. *Artificial radioactivity had been discovered.* The Joliots determined that this only occurred with light element targets. The Coulomb barrier would prevent the polonium α particles from approaching the heavy target nucleus closely enough to cause a nuclear reaction. As soon as Joliot published, everyone rushed to duplicate his work. Instead of trying to duplicate this work, in a matter of weeks the University of Rome physicist Enrico Fermi put the neutron and the idea of artificial radioactivity together and produced nuclear fission.

Chemical Techniques and the Periodic Table

At this point in the story, let us pause to discuss chemical techniques that will be critical to understanding much of what follows. It's already been mentioned earlier that the Periodic Table was useful for indicating that elements in the same group would have similar chemical properties. They would all combine with the same elements, have similar solubility properties and in general would behave in a similar manner. This was useful when radioactivity was discovered. Chemical identification of a radioactive nuclide usually required precipitation of a compound of that nuclide from a solution. Following a nuclear reaction, the irradiated sample is put into solution and a very small amount of a stable element, the *carrier*, is added. If the carrier is isotopic to the radioactivity or belongs to the same group in the Periodic Table, this radioactivity will precipitate out of solution with the carrier when a sufficient amount of carrier is added to saturate the solution. The presence of the radioactivity in the precipitate is easily recognized by its characteristic half–life and radiation.

Another chemical technique is the tracer or indicator method, where a small amount of known radioactive material is added to a mixture and the result is precipitated. If the radioactivity is found to be precipitated, then it's known that the mixture is an element isotopic to the radioactive tracer.

A further point to be discussed is the form of the Periodic Table in those days. Although the rare earth elements were recognized as belonging in a separate location in the Table, the concept of actinide elements had not yet been identified. This would have to await their discovery and a study of their chemical properties. Thorium, protactinium, and uranium were considered to be the analogues of hafnium, tantalum, and tungsten, respectively[19]. Thus, elements 93 and 94 were considered to be analogues of rhenium and osmium. This affected some of the thinking in the complicated analysis of the experiments to follow.

Fermi's Neutron-Induced Reactions

While most scientists knew that sources of neutrons would be very much weaker than α sources, Fermi did his experiments realizing that proton, deuteron and α particles would all suffer Coulomb repulsion in a heavy nucleus and would never cause a nuclear reaction. Thus neutrally charged neutrons, although weak, would still be more effective than the α. He proposed to irradiate all elements, even the heavy ones, with neutrons to study artificial radioactivity. In a series of papers in 1934, Fermi and his Rome group[20] reported the irradiation of all available elements up to uranium with neutrons, and the production of radiations with characteristic half-lives. Most of the radioisotopes produced, decayed by emitting β^- particles (negatively charged electrons) to become atoms of the next (higher Z) chemical element. Whereas most target elements produced one radiation and half-life, Fermi obtained four radiations with the half-lives 10 and 40 seconds and 13 and 90 minutes from uranium. Reasoning by analogy from the behavior of medium weight elements, Fermi's group thought they had produced radioisotopes by neutron capture. Since β^- s were emitted, they concluded the isotopes were of elements beyond uranium in the Periodic Table. By various chemical techniques, they eliminated all of the possible candidate elements between lead and thorium and concluded that they had *created* element 93 and possibly 94, which did not exist in nature[21]. The Rome group would call them ausenium, and hesperium, respectively[22]. In addition, Fermi used paraffin to slow down the neutron's velocity and discovered that slow neutrons would produce increased radioactivity in the target element[23]. Thus, the likelihood of forming radioactivity was not proportional to neutron energy (velocity), as one might initially have expected.

One member of Fermi's five man group, Edoardo Amaldi[24], noted that the group wrote short letters in Italian to La Ricerca Scientifica, received what we now call preprints, and distributed these to forty prominent and active nuclear physicists all over the world in order to communicate these results rapidly. Fermi, who up to that time was probably best known for his theoretical work explaining β^- decay, received a reply from Rutherford thanking him for the preprint and congratulating him on his "escape" from theoretical physics. Thus, we can see that many prominent experimentalists of that day did not view theoreticians in a very favorable light.

Reports immediately appeared from Aristid von Grosse[25], a German-born chemist from the University of Chicago and from Ida Tacke Noddack[26]. Von Grosse had done early work on protactinium with Hahn soon after its discovery and he mistakenly thought that one of the activities behaved like that element. Von Grosse did not use irradiated uranium but a strongly acid solution of uranil nitrate, which Fermi's group had used. A small amount of protactinium was added. Von Grosse precipitated manganese dioxide along with protactinium. The experiment failed when it was repeated with rhenium, which was considered a lower period member of the same group as element 93 in the Periodic Table of that day. Von Grosse concluded that protactinium, and not element 93, was the activity produced.

Because of her work on its discovery, Lise Meitner considered protactinium to be "her" element. By 1934, she had not worked with Hahn for over a decade. She convinced him that they should take up this protactinium problem[27]. Their interest in the uranium problem would continue long after von Grosse was proven wrong about protactinium.

Von Grosse also published[28,29] additional articles questioning the claim of elements 93 and 94. He suggested that they might belong to a second group of rare earth elements in the Niels Bohr sense. Apparently no nuclear physicist or radiochemist actively involved read these latter articles until after fission was discovered. Even Niels Bohr, whose theory had explained the first rare earth series, never gave this issue any thought. Thus, the actinide concept would be delayed for a decade.

Noddack's article contained the concept of nuclear fission, although no one took it seriously. She stated that it was conceivable that in the bombardment of heavy elements with neutrons the nuclei might break into larger pieces, which are isotopes of known elements, but not neighbors of those irradiated. Noddack's proposal was viewed as speculation, pointing out a lack of rigor on Fermi's part in not eliminating all other possibile elements before he claimed the discovery of transuranium elements. It was never taken seriously. Noddack never attempted an experiment to follow up on her own proposal, and no one else ever did either. Her proposal was considered wholly incompatible with the known laws of physics. The known fact that the Coulomb barrier prevented charged particle emission in heavy elements for the α particle and the proton would argue against an even heavier charged particle emission of a fission fragment. Keep in mind that Bohr's liquid drop model of the nucleus would not be published for a few more years and no one up to that time had considered the possibility of the collective motion of the nucleons.

By 1936, the Rome group itself fissioned as three of the members moved on to positions at other locations in Italy and the USA. Only Fermi and Amaldi remained and their interest turned to the problem of the selective energy

absorption of neutrons. They left this uranium problem in the hands of groups more experienced in the area of radiochemistry, such as Meitner, Hahn and the Joliot–Curies. Fermi eventually left for the USA in 1938, after he was awarded the Nobel prize for his neutron work, because his wife was Jewish and he was concerned about the effect of racial laws.

Paris and Berlin Get Involved

After dismissing Noddack's paper as not worth pursuing, Meitner and Hahn[30] repeated von Grosse's 1934 protactinium work, and concluded that the elements from 92 down to 80 were excluded, so the radioactivities seen probably were from elements beyond uranium, Z = 92. The 10 and 40 second activities were too short to work with but they stated that the two half–lives of 13 and 90 minutes reported by Fermi could be separated and that the 90 minute activity was from a mixture of two elements.

The Paris group headed by Irene Joliot–Curie did not initiate work with uranium but with thorium[31]. There were three naturally occurring radioactive decay chains known at the time, i.e., 4n, 4n+2, and 4n+3. They reasoned that neutron irradiation of thorium might produce the non–existent 4n+1 decay chain. When Meitner and Hahn began to measure a number of different chains of radio–active half–lives in the uranium irradiation[32], the Paris group did not accept Hahn's results. Even Meitner thought it disturbing to find a long chain of successive β^- disintegrations[27], which were unknown in other naturally occurring heavy element decay. A upper limit of two β^- decays were usually followed by an α particle decay.

Joliot–Curie decided to follow a single radioisotope rather than Hahn's mixture. A major problem in this type of radio–chemical work was the existence of the natural radioactive background of the uranium decay chain. The Berlin group had avoided this problem by purifying the uranium before the experiment. In contrast to the Berlin group, Joliot–Curie and the Yugoslavian chemist Pavel Savić wrapped the irradiated uranium target in copper foil to measure the activity, while suppressing the natural radio–active background of the uranium decay chain.

The Paris procedure removed the β^- radiation of UX_2 (^{234}Pa) and any other β^- radiation with an energy of 2 MeV or less. With the lower energy radiations removed, the French group found a 3.5 hour half–life that was missed by Meitner and Hahn. At first, Curie and Savić reported that the 3.5 hour radiation behaved like thorium[33], but later discovered that they could separate it from thorium. Chemical tests revealed that it precipitated with a lanthanum carrier and a check of the Periodic Table suggested that it must be actinium from the same group[34]. It is interesting to note that the 3.5 hour activity did not completely follow lanthanum. If it had, this might have led Curie and Savić to question why it could not be separated from the lanthanum. From our present vantage point, it might well have been a mixture of 3.9 hour ^{141}La

with a β^- endpoint of 2.43 MeV, with 3.54 hour ^{92}Y with a β^- endpoint of 3.6 MeV. Curie made the comment in 1938 that she sometimes thought that she had all the chemical elements in her bombarded uranium.[27]

These publications by Curie irritated Hahn, who told Frédéric Joliot[35] in 1938, "the work of your wife is so wrong but as she is a lady, I don't want to ridicule and publish what I think of her work". Hahn thought that since all of his work was now in trouble because of Curie, he would have to take time off to "straighten her out". Hahn called this 3.5 hour half-life element 'curiosum'[36].

Effect of the Political Situation in Germany

After Meitner and Hahn's initial experiments, they asked a young chemist, Fritz Strassman to join in the work. They worked together until 1938. With the advent of Adolph Hitler's rise to political power in 1933, Nazi Germany passed a series of racial laws which circumscribed the rights of people of Jewish heritage. Although Lise Meitner was Catholic, she had 50% Jewish blood. However, she was protected because she was not German, but Austrian. When Germany annexed Austria in the 'Anschluss' in 1938, Meitner came under the German racial laws and would be forced to leave Berlin. The German Chancellor, Adolph Hitler created the Brown Shirt militia (storm troopers), whose members often wore their uniforms in the workplace and informed on any anti-government attitudes or comments by their fellow workers. There were a number of these Brown Shirts working in Hahn's institute at that time. Concern about Meitner's situation caused Hahn considerable mental anguish at the time that he was trying to unravel the mystery of fission.

When the Berlin group repeated Curie's experiments[37], they used both barium and lanthanum as carriers of their 'radium' activity and of its 'actinium' daughter and found three of the radioactive half-lives were precipitated with the barium carrier. From the Periodic Table, this implies that the activities belonged to radium, which must be formed via two α decays from ^{239}U. This led to a search for these α particles. Meitner asked one of the Brown Shirt physicists in the laboratory named Gottfried von Droste to search for these α particles with an oscilloscope and air chamber but he was not successful in finding the two α particles[38]. Von Droste complained that since Hahn the chemist knew he had radium, there must be alphas, and Meitner called von Droste a lousy physicist because he couldn't find them. The high voltage air chamber took in the α particles and they were displayed on the screen of the oscilloscope. Occasionally, the beam would soar off the screen and disappear and they had to wait for it to return. Von Droste attributed it to electrostatic effects, which was anything that didn't turn out right[39]. He solved the problem by using thin aluminum foil which stopped the pulses. If he had turned down the magnification, he could have seen the fission fragment pulses. Other labs had a similar problem. At the Cavendish Laboratory, for example, Chadwick and Maurice Goldhaber used a foil to eliminate the α particles from the uranium natural decay chain and in addition eliminated any fission fragments as well[40].

In July, following the 1938 Anschluss, Meitner was smuggled out of Germany to Holland and finally to Sweden, where Manne Siegbahn offered her a position in his Institue in Stockholm. Hahn and Strassman continued their experiment, trying to separate their 'radium activity' from the barium carrier. They added ^{228}Ra ($MsTh_1$) to their solution. Hahn had unsuccessfully tried to separate $MsTh_1$ from radium at the beginning of his career in radiochemistry as mentioned above. When barium bromide is precipitated fractionwide, radium is strongly enriched in the first fractions and depleted in the last fractions. However, instead of the 'radium activity' going with the $MsTh_1$, after successive the fractional precipitations and crystalizations, it remained uniformally distributed. They had *barium* and not *radium*. They concluded their famous paper with – "on the basis of the briefly reported experiments, we should as chemists replace the symbols Ra, Ac, Th with Ba, La, Ce. As 'nuclear chemists' who are close to physics in certain ways, we cannot yet bring ourselves to take this jump which contradicts all of the experiences of nuclear physics to date. Perhaps it is still possible that a number of strange chance incidents have distorted our results[41]." Hahn later indicated that his reluctance to take a stronger position was not due to any doubts about the results but as a chemist he hestitated to announce a revolutionary discovery in physics.[42]

Events now moved very quickly. Hahn wrote to Meitner, who replied: "I find it very difficult to assume such a degree of bursting, but we've had so many surprises in nuclear physics that one can't very well say it's impossible." Meitner's nephew, the Austrian physicist Otto Frisch, visited her in Stockholm while on Christmas leave from Niels Bohr's Institute in Copenhagen. Using Bohr's liquid drop model, they were finally able to understand what had occurred[43].

Epiloque

The series of events following the publication of the Hahn–Strassman paper make another very interesting story, but since it is subsequent to the discovery of fission, it will have to wait for another day and another paper.

It is interesting to note at this time, when there is a major concern about the number of woman participating in the sciences, that three of the principal players in this drama were women, i.e.; Meitner, Joliot–Curie, and Noddack.

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