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The Discovery of Artificial Radioactivity

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We reconstruct Frédéric Joliot and Irène Curie's discovery of artificial radioactivity in January 1934 based in part on documents preserved in the Joliot-Curie Archives in Paris, France. We argue that their discovery followed from the convergence of two parallel lines of research, on the neutron and on the positron, that were focused on a well-defined experimental problem, the nuclear transmutation of aluminum and other light elements. We suggest that a key role was played by a suggestion that Francis Perrin made at the seventh Solvay Conference at the end of October 1933, that the alpha-particle bombardment of aluminum produces an intermediate unstable isotope of phosphorus, which then decays by positron emission. We also suggest that a further idea that Perrin published in December 1933, and the pioneering theory of beta decay that Enrico Fermi also first published in December 1933, established a new theoretical framework that stimulated Joliot to resume the researches that he and Curie had interrupted after the Solvay Conference, now for the first time using a Geiger-Müller counter to detect the positrons emitted when he bombarded aluminum with polonium alpha particles.

Key words: Frédérick Joliot; Irène Curie; Francis Perrin; Enrico Fermi; Carl D. Anderson; Patrick M.S. Blackett; Niels Bohr; James Chadwick; Paul A.M. Dirac; Lise Meitner; Giuseppe Occhialini; Wolfgang Pauli; Ernest Rutherford; seventh Solvay Conference; Institut

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du Radium; Geiger-Müller counter; beta decay; artificial radioactivity; history of nuclear physics.

Introduction

Jean Perrin communicated Irène Curie and Frédéric Joliot's report on their discovery of artificial radioactivity to the Paris *Academie des Sciences* on January 15, 1934,[1] which gained worldwide attention when it was reported in the February 10 issue of *Nature*.[2] *Science News* emphasized that, "Never before [had] radioactivity been created by an external cause,"[3] and Ernest Rutherford declared:

> [It] is remarkable that the life of the unstable atom produced is as long as it is. We do not know whether the atoms so far made artificially radioactive are typical or whether other unstable atoms which may be produced will have a longer or shorter life. The discovery of the Joliots shows how little we really know about radioactivity.[4]

That became clear in March 1934 when Enrico Fermi in Rome demonstrated that artificial radioactivity was also produced when neutrons bombard elements of high atomic number.[5]

While Joliot and Curie's discovery of artificial radioactivity has been discussed earlier by historians and physicists, most do not go into detail regarding its timing,[6] or do not offer an explanation for its timing.[7] Joliot first observed artificial radioactivity on the afternoon of January 11, 1934,[8] but in retrospect it seems that it could have been discovered soon after the seventh Solvay Conference at the end of October 1933,[9] since Francis Perrin pointed out in the discussion following Joliot and Curie's report that when α (alpha) particles bombard aluminum a neutron and an unstable isotope of phosphorus could be produced that then decays by positron emission. Joliot and Curie, however, did not take up Perrin's suggestion; instead, after the Solvay Conference they interrupted their researches for two months before resuming them at the end of December 1933.

To gain insight into Joliot and Curie's puzzling interruption of their researches, we examined the notebooks, manuscripts, and other materials preserved in the the Joliot-Curie Archives in the *Institut du Radium* in Paris. We first set their work into historical context by discussing the discoveries of the neutron and positron in 1932.

Joliot and Curie and the Discovery of the Neutron

In 1930 Walther Bothe and Herbert Becker in Berlin bombarded beryllium (Be) with polonium α particles (${}_2He^4$) and reported that a highly penetrating gamma radiation was produced.[10] This was the first in a series of experiments that led James Chadwick to conclude, in February 1932, that not gamma rays, but neutrons and an isotope of carbon were being produced according to the reaction ${}_4Be^9 + {}_2He^4 \rightarrow {}_6C^{12} + {}_0n^1$.[11]

Joliot and Curie (figure 1) recognized that they had missed a major discovery, but they did not allow that to impede their researches. They were well positioned to respond to Chadwick's discovery. Curie, an expert radiochemist, had received her doctorate in 1925, a few months after Joliot had entered the *Institut du Radium* as an assistant to her mother Marie. They married in 1926, and Joliot received his doctorate in 1930 with a thesis on the electrochemistry of the radioelements that displayed his deep understanding of physics and chemistry. Lew Kowarski, who came to know them well, characterized Curie as an "exquisite technician" who "worked very beautifully, very thoroughly," and had "a profound understanding of what she was doing," while Joliot had a "more brilliant, more soaring imagination." Thus, although having very different personalities, "they complemented each other marvelously, and they knew it."[12]

Curie and Joliot soon reported that when beryllium is bombarded with polonium α particles both neutrons and gamma rays are produced,[13] and in further experiments they showed that the energy of the latter was on the order of 4 MeV (million electron volts).[14] In

June 1932 they concluded that both neutrons and gamma rays "are emitted simultaneously" according to the reaction ${}_4\mathrm{Be}^9 + {}_2\mathrm{He}^4 \rightarrow {}_6\mathrm{C}^{12} + {}_0\mathrm{n}^1 + h\nu$.[15] Then, over the next seven months, they set out to prove definitely that the products of the reaction consist "at least partially" of neutrons. To this end, they used an ionization chamber filled with methane (CH_4) connected to a Hoffmann electrometer. Methane had the advantage of reducing the effect of gamma rays and clearly revealing the presence of neutrons by the recoil protons they expelled from it. Jean Perrin communicated their results to the *Académie des Sciences* at a meeting on February 6, 1933.[16] They found that when aluminum (${}_{13}\mathrm{Al}^{27}$) was bombarded with polonium α particles "a major part" of the products consisted of neutrons, and that the minimum α-particle energy required to produce them was "close to" 5 MeV. They carried out similar experiments with fluorine (${}_9\mathrm{F}^{19}$) and concluded:

> [It] is known that Al^{27} and F^{19} nuclei are able to emit protons under the action of polonium α rays. *These nuclei can therefore undergo two different modes of transmutation, one with the emission of protons, and one with the emission of neutrons (unless a proton and a neutron are jointly emitted).* Such a case has not yet been observed....[17]

Fourteen days later, on February 20, Jean Perrin communicated a note by Pierre Auger and Gabriel Monod Herzen to the *Académie des Sciences* that confirmed Joliot and Curie's results on aluminum.[18]

Joliot and Curie and the Discovery of the Positron

Two of us have recently discussed Joliot and Curie's reasearches on the positron.[19] We recall here that they began in April 1932, four months before Carl D. Anderson's discovery,[20] when they bombarded beryllium with polonium α particles and sent the neutrons that were produced through paraffin and other hydrogenous substances, detecting the recoil protons in their ionization chamber and photographing their tracks after entering Joliot's cloud chamber when immersed in a 1500-gauss magnetic field.[21] They also photographed the tracks of electrons that were expelled from a lead filter, one of which, they found, was "curved in an opposite sense to the others."[22] They realized they had photographed the track of a positron only after learning of its discovery, either from Patrick M.S. Blackett and Giuseppe P.S. Occhialini's paper in the *Proceedings of the Royal Society*,[23] where they identified it with Paul A. M. Dirac's anti-electron, or from Chadwick, Blackett, and Occhialini's note in the April 1, 1933, issue of *Nature*.[24] They then realized that they again had missed a major discovery.

Maurice de Broglie communicated Curie and Joliot's first paper on the positron to the *Académie des Sciences* on April 10, 1933.[25] In it they reported that they had bombarded beryllium with polonium α particles, had sent the emitted gamma rays through a lead foil and the positrons that were then produced into Joliot's cloud chamber when immersed in a magnetic field, finding that the positron tracks emerged from the point at which the gamma rays struck the lead foil. Seven weeks later, on May 22, 1933, they found that the gamma rays emitted from thorium active deposit ThC″ ($_{81}\mathrm{Tl}^{208}$) also produced electron-positron pairs.[26] They concluded that "when a high energy gamma photon encounters a heavy nucleus it is transformed into two electrons of opposite sign."[27] They called them, on Marie Curie's suggestion, "materialization electrons."[28]

On June 19, 1933, Jean Perrin communicated Curie and Joliot's report on further far-reaching experiments to the *Académie des Sciences*.[29] They had used a 20-millicurie polonium α-particle source, had placed an aluminum foil in front of the opening to Joliot's cloud chamber when immersed in a 400-gauss magnetic field, and were surprised to observe that the emitted positrons (figure 2) were not accompanied by electrons of comparable energy. This meant that the positrons had not been created by the materialization of electron-positron pairs. The positrons emitted from boron exhibited the same behavior. Curie and Joliot called them “transmutation” positrons to distinguish them from their earlier “materialization” positrons.

Less than one month later, on July 10, 1933, Curie and Joliot submitted a paper for publication that contained the first ever photograph of the materialization of an electron-positron pair, but they also emphasized their discovery of transmutation positrons:

> The transmutations hitherto known, caused by α particles, fast protons or neutrons, occur with the emission of protons, neutrons or α particles; [until now] the emission of nuclear electrons [positrons] has never been observed in these phenomena.[30]

The Intersection of Research on the Neutron and Positron

Joliot and Curie’s discovery of transmutation positrons allowed them to settle an earlier problematic result concerning neutrons. Thus, in February 1933 they had found that when polonium α particles bombard aluminum and fluorine they emit neutrons, which a few months later they found was also true for sodium.[31] This meant that both a neutron and an unknown isotope should be produced in the reaction.[32] For example, in the case of aluminium, an unknown isotope of phosphorus ($_{13}P^{30}$) should be produced according to the reaction $_{13}Al^{27} + {}_{2}He^{4} \rightarrow {}_{13}P^{30} + {}_{0}n^{1}$. Similarly, in the cases of fluorine ($_{9}F^{19}$) and sodium ($_{11}Na^{23}$), the unknown isotopes $_{11}Na^{22}$ and $_{13}Al^{26}$ should be produced. Returning to the case of aluminium, they wrote that “*unless a proton and a neutron are emitted at the same time*,”[33] in which case the silicon isotope $_{14}S^{i29}$ would be produced according to an unlikely reaction whose “energetic balance [would be] strongly negative,” the only possibility was that the unknown phosphorus isotope $_{15}P^{30}$ “spontaneously changes into” the stable silicon isotope $_{14}Si^{30}$ “by capture of an extra-nuclear electron,”[34] according to the reaction $_{13}Al^{27} + {}_{2}He^{4} + {}_{-1}e^{0} \rightarrow {}_{14}Si^{30} + {}_{0}n^{1}$.

By June 1933, however, with their discovery that positrons as well as neutrons are emitted by aluminium, the possibility existed, Joliot and Curie suggested, that when α particles bombard aluminium both neutrons and positrons are emitted, and the stable silicon isotope ${}_{14}\mathrm{Si}^{30}$ is then produced according to the transmutation reaction ${}_{13}\mathrm{Al}^{27} + {}_{2}\mathrm{He}^{4} \rightarrow {}_{14}\mathrm{Si}^{30} + {}_{0}\mathrm{n}^{1} + {}_{+1}e$.[35] It was known since the early 1920s, however, that under α-particle bombardment aluminium emits protons (${}_{1}\mathrm{H}^{1}$), producing the stable silicon isotope ${}_{14}\mathrm{Si}^{30}$ according to the reaction ${}_{13}\mathrm{Al}^{27} + {}_{2}\mathrm{He}^{4} \rightarrow {}_{14}\mathrm{Si}^{30} + {}_{1}\mathrm{H}^{1}$. Now, since only one aluminium isotope was known to exist, this in turn meant that under α-particle bombardment *either* a neutron and a positron, *or* a proton, could be produced along with the stable silicon isotope ${}_{14}\mathrm{Si}^{30}$. They therefore asserted that "*sometimes a neutron and a positive electron are emitted instead of a proton*."[36] This implied "the hypothesis that the proton is composed of a neutron and a positive electron,"[37] which was in direct conflict with Rutherford's and Chadwick's belief that the neutron is composed of a proton and an electron—a conflict that led the two teams to calculate very different values for the mass of the neutron.[38]

The Seventh Solvay Conference

The seventh Solvay Conference, which was held at the Free University of Brussels on the subject, "Structure and Properties of Atomic Nuclei," from October 22-29, 1933,"[39] occupies a special place in the history of nuclear physics.[40] Eleven countries were represented by forty-one participants.[41] In addition to Joliot and Curie, the invited speakers were John D. Cockcroft, Chadwick, Dirac, George Gamow, and Werner Heisenberg (figure 3).

Joliot and Curie divided their report, "Penetrating Radiation of Atoms under the Action of α Rays," into two parts, on the neutron and on the positron, the former of which, as the original manuscript reveals, was written by Curie and the latter by Joliot. In the cases of the α-particle bombardment of boron and aluminum, they reiterated their contention that *either* a neutron and a positron, *or* a proton, is produced in the reaction.

During the discussion, Lise Meitner pointed out that in her own cloud-chamber experiments in Berlin on the α-particle bombardment of aluminum and fluorine, although she had found unequivocal evidence for the emission of positrons, she had failed to see any recoil proton tracks arising from the emission of neutrons, even though four times as many positrons were emitted from aluminum as from fluorine. She concluded that "this does not support the idea that ... the emission of a neutron takes place at the same time as that of a positive electron."[42] She was convinced that, unlike neutrons, positrons did not originate in the nucleus as was suggested by Joliot and Curie's idea of "transmutation positrons." The positrons from aluminum, as Blackett suggested during the discussion,[43] could be created as electron-positron pairs following the "internal conversion" (materialization) of gamma rays emitted by the nucleus.

In his reply, Joliot emphasized that above a certain threshold energy only positrons are emitted from aluminum, in contrast to beryllium where mostly electrons are emitted,[44] as he had shown experimentally.[45] This supported his and Curie's view that the positrons from aluminum are genuine transmutation positrons, while those from beryllium were probably produced by the "internal materialization" of gamma rays, as Blackett had suggested.[46] Moreover, if the positrons from aluminum were created by materialization rather than transmutation, then the emission of neutrons from aluminum would not produce an unknown unstable isotope of phosphorus.

Immediately following Meitner's intervention, Francis Perrin (figure 4) offered a new hypothesis on the origin of the positrons. He observed that, according to Joliot, the emitted positrons display a continuous energy spectrum, similar to that of the beta particles emitted in natural radioactive decay. Thus, if Joliot were correct in claiming that neutrons and positrons were emitted simultaneously, then the total energy of emission should be shared between them. That, however, could not be the case for the neutrons and positrons emitted from aluminum, because the observed energy of the neutrons was less than 1 MeV, and that of the positrons was about 2.6 MeV, so the energy of the former could never be as large as that of the latter. Therefore, Perrin argued:

> It also seems reasonable to assume that the mechanism proposed by Mr. Joliot is decomposed into two successive emissions, [first] of a neutron and [then of] a positive electron, with the intermediate formation of an unstable nucleus ($_{15}P^{30}$ in the case of aluminum); this nucleus shows, in short, a radioactivity by positive electrons, and it is not surprising that one finds in this case a continuous spectrum as for the β rays in the natural radioactivities.[47]

Perrin concluded:

> One is therefore led to suppose that the phenomena of radioactivity with the emission of positive electrons and perhaps the phenomena of materialization of electron pairs involve mechanisms analogous to those observed in the natural β radioactivities.[48]

We have found the original transcript of Perrin's intervention (typed on onionskin) in the Joliot-Curie Archives (figure 5), which Paul Langevin, as President of the Scientific Committee of the seventh Solvay Conference, probably sent to Joliot and Curie along with the transcripts of the other interventions after their report. Joliot and Curie received Perrin's transcript on February 1, 1934, and returned it on February 8. Perrin had made a number of handwritten changes, deletions, and clarifications in pencil, which were taken completely into account in the published proceedings. His key addition probably was that of the chemical symbol of the intermediate unstable isotope of phosphorus, $_{15}P^{30}$, which should be produced when α particles bombard aluminum.

Wolfgang Pauli and Niels Bohr responded immediately, but very differently, to Perrin's suggestion regarding the analogy of the emission of positrons to that of electrons in natural β decay. Pauli declared:

> The conclusion, that there is a strong analogy between the emission of positrons in artificial disintegration and the spontaneous emission of β rays seems to me not very certain. In the first case, it may well be that the particles are always produced at the periphery or outside the nucleus itself, while β rays seem to emerge from the nucleus.[49]

Bohr declared instead:

> It is a problem of the highest importance to know if in the process in which aluminum is bombarded with α particles, energy is conserved or not. *Without doubt*, the observation that positrons do not all have the same speed, is not *by itself* an argument against energy conservation, *as we do not yet know how the emission of positrons is produced*. If indeed, as assumed by Mr. Joliot, positrons come from the interior of the nucleus, the circumstances will be very similar to those of β rays.[50]

Another subject of crucial significance that came up in the discussion following Joliot and Curie's report was whether the proton was composed of a neutron and a positron, as Joliot and Curie believed, or whether the neutron was composed of a proton and an electron, as Rutherford and Chadwick believed. Each hypothesis entailed radically different values for the mass of the neutron and grave difficulties regarding the spin and statistics of nuclei. These problems remained unresolved until Chadwick and Maurice Goldhaber carried out an experiment at the Cavendish Laboratory on the photodisintegration of the deuteron in the summer of 1934, which proved that the neutron is a new elementary particle.[51]

Aftermath

Less than three weeks after the Solvay Conference, Lise Meitner (figure 6), in a letter to Irène Curie on November 18, 1933, withdrew her criticism of Joliot and Curie's observation that neutrons and positrons are emitted simultaneously when aluminum is bombarded with polonium α particles:

> Dear Madame Joliot,
>
> After my return [to Berlin] I looked again at your measurements of Al and F, and I came to the conclusion that the absence of protons [recoiling from neutrons] from aluminum in the gas can be interpreted very well by statistical errors.... So I made some hundreds of measurements with a stronger preparation of polonium and I found in the gas, in the case of aluminum, about six protons per one hundred measurements, and in fluorine two protons per hundred measurements. Thus, one of the observations in the discussion is not correct. I have written to Brussels and I have asked them to allow me to add a small note to the remarks I made during the debate.... I have reached the conclusion that your interesting views on the process of disintegration of aluminum are correct and that positive electrons really do come out of the aluminum nucleus.[52]

Meitner's note duly appeared in the conference proceedings.[53] Curie, on her part, responded to Meitner immediately, on November 22:

> Dear Miss,
>
> We, Mr. Joliot and I, have read your letter with interest and thank you very much for letting us know about these new results. Would you have the kindness to tell us what is the maximum energy of the Po+Al neutrons, on the basis of the recoil H rays [protons] that you have observed? It is interesting to know if this energy is in agreement with what we have regarded as probable following our experiments.[54]

Thus, by the end of November 1933, Curie and Joliot knew that Meitner supported their observation that positrons are emitted in a nuclear-transmutation process.

A decade and a half later, in notes that Joliot made for a course he taught at the Sorbonne in the academic year 1948-1949, exclaimed: “2 months of tranquility thanks to Meitner!! [“*2 moins de tranquillité à cause de Meitner!!*].”[55] Thus, Joliot and Curie decided not to pursue their experiments on the simultaneous emission of positrons and neutrons from aluminum; instead, as seen in materials preserved in the Joliot-Curie Archives, after the Solvay Conference they worked independently on different researches.[56]

Prior to the conference, after October 19, 1933, Curie had worked mostly on the maintenance of their polonium α-particle sources. Then, after the conference, on five days between November 18-27 she used Joliot’s cloud chamber to investigate the “positive electrons emitted from Al by alpha particles slowed down or not” (figure 7), and on seven days between December 2-13 she used two different ionization chambers, a “small” and a “large” one filled with butane at various pressures, to study the “limit of excitation of Al neutrons.” Her experiments thus did not touch on the central problem, the link between the emission of neutrons and positrons when aluminum was bombarded with polonium α particles.

Joliot, on his part, resumed experiments on the properties of the “materialization positrons” by employing an “intense source of positrons” produced by “80 millicuries of Po deposited by volatilization on Al and covered with Al.” Then, between the middle of December 1933 and early January 1934, he studied positron-electron annihilation, finding results in complete agreement with Dirac’s theory, which he published in the *Comptes rendus*.[57] In these experiments, Joliot for the first time used a Geiger-Müller counter whose circuit diagram he sketched (figure 8), but which was actually built by Wolfgang Gentner in early January 1933 after his arrival in the *Institut du Radium* as the first German postdoctoral researcher.[58]

The Discovery of Artificial Radioactivity

Jean Perrin communicated Curie and Joliot's paper, "A New Type of Radioactivity,"[59] to the *Académie des Sciences* on January 15, 1934, announcing their discovery of artificial radioactivity.

Joliot had irradiated an aluminum foil for about ten minutes with a 60-millicurie source of polonium α particles and had detected the emitted positrons with his new Geiger-Müller counter, thus replacing his cloud chamber, an apparatus for photographing an event at a given instant of time, with one for recording the evolution of an event over time. He discovered, much to his surprise, that when he placed the aluminum foil in front of the window of his Geiger-Müller counter and removed the polonium α-particle source, the emission of positrons did not stop but decreased exponentially with a half-life of 3 minutes, 15 seconds. When he reduced the energy of the incident α particles by inserting a thin lead absorber in their path, the number of emitted positrons decreased, but their half-life did not. He found an identical effect when irradiating boron (half-life 14 minutes) and magnesium (half-life 2 minutes, 30 seconds), but not when irradiating a number of other light elements (hydrogen, lithium, carbon, beryllium, nitrogen, oxygen, fluorine, sodium, calcium, nickel, silver), which showed that the new phenomenon could not be ascribed to any "contamination" of the polonium α-particle source. Rather, "for some of these elements, the phenomenon does not occur," whereas "for others the half-life is perhaps too short" to be measured.

Since a **Geiger-Müller counter cannot distinguish between a positive and a negative** ionizing particle, to confirm that the emitted particles were positrons, Joliot carried out further experiments on aluminium and boron "by the Wilson [cloud-chamber] method and by the Thibaud trochoid method,” [60] which proved that “the irradiation … is made of positive electrons,” and that in all likelihood “it is the same for the magnesium irradiation.” Therefore, “it has been possible for the first time to create a radioactivity by an external cause in a certain number of atomic nuclei that exists for a measurable time in the absence of the exciting cause.”[61]

To explain the emission of positrons, Curie and Joliot abandoned their earlier hypothesis that when a light element like aluminum is bombarded with *α* particles it simultaneously emits a neutron and a positron through a single nuclear transmutation. Instead, a neutron and an unstable nucleus are first produced and the latter then decays by positron emission, in the case of aluminium according to the two reactions:

$$ {}_{13}\mathrm{Al}^{27} + {}_{2}\mathrm{He}^{4} \rightarrow {}_{15}\mathrm{P}^{30} + {}_{0}\mathrm{n}^{1} $$

$$ {}_{15}\mathrm{P}^{30} \rightarrow {}_{14}\mathrm{Si}^{30} + {}_{+1}e^{0}. $$

For boron and magnesium, the analogous reactions are:

$$ {}_{5}\mathrm{B}^{10} + {}_{2}\mathrm{He}^{4} \rightarrow {}_{7}\mathrm{N}^{13} + {}_{0}\mathrm{n}^{1} $$

$$ {}_{7}\mathrm{N}^{13} \rightarrow {}_{6}\mathrm{C}^{13} + {}_{+1}e^{0}; $$

$$ {}_{12}\mathrm{Mg}^{24} + {}_{2}\mathrm{He}^{4} \rightarrow {}_{14}\mathrm{Si}^{27} + {}_{0}\mathrm{n}^{1} $$

$$ {}_{14}\mathrm{Si}^{27} \rightarrow {}_{13}\mathrm{Al}^{27} + {}_{+1}e^{0}. $$

Curie and Joliot stressed that the unstable isotopes ${}_{15}\mathrm{P}^{30}$, ${}_{7}\mathrm{N}^{13}$, and ${}_{14}\mathrm{Si}^{27}$ “can exist only for very short times, which is why they are not observed in Nature.”[62]

Theoretical Considerations: Perrin and Fermi

We have seen that at the seventh Solvay Conference at the end of October 1933 Francis Perrin proposed that positrons are emitted in Joliot and Curie’s experiments just as they themselves now proposed. We suggest that Joliot and Curie finally embraced Perrin’s interpretation because in December 1933 they learned that Perrin had published an important paper on Wolfgang Pauli's neutrino hypothesis, and that Enrico Fermi had published his pioneering theory of beta decay.

Jean Perrin communicated his son Francis's paper, "On the possibility of the emission of neutral particles of zero intrinsic mass in β radioactivities,"[63] to the *Académie des Sciences* on December 18, 1933. Note the plural, "β radioactivities," which underlined Perrin's contention, which he had expressed at the Solvay conference, that there are two kinds beta decay, the well-known one in which an electron is emitted, and a new one in which a positron is emitted. Perrin went on to conclude:

> [If] the neutrino has zero intrinsic mass we must also think that it does not pre-exist in atomic nuclei and that, as for the photon, it is created at the time of emission. Finally, it seems that we should assign to it spin ½ so that we have conservation of spin in β radioactivities and, more generally, in any transformation of neutrons into protons (or *vice versa*) with absorption and emission of electrons and neutrinos.[64]

Two of us have recently discussed Fermi's theory of beta decay,[65] which Fermi first published in an abridged version in late December 1933 in *La Ricerca Scientifica*,[66] the bulletin of the Italian National Research Council, and then in expanded versions in January 1934 in *Il Nuovo Cimento* and the *Zeitschrift für Physik*,[67] the latter giving as its date of receipt January 15, 1934. Fermi did not cite Perrin's paper in his first paper (he and Perrin probably submitted their papers for publication at about the same time), but he did in his second and third papers.

Perrin's new ideas and Fermi pioneering theory constituted a new theoretical framework for understanding beta decay and the emission of positrons in it. It seems that Joliot and Curie may not have pursued Perrin's suggestion of the formation of an unstable isotope of phosphorous after the Solvay Conference because there then was no experimental support for it. Now, however, a new theoretical framework emerged with Perrin's new idea that a proton could be transformed into a neutron, and *vice versa*, and with Fermi's theory of beta decay, in which Fermi developed this same idea in detail and mathematically. We believe that Joliot and Curie learned about these new theoretical developments at the end of 1933 or in early 1934, which stimulated Joliot to resume his and Curie's earlier experiments, ending their "two months of tranquility."

Joliot and Curie, after all, were personal friends of Perrin and kept abreast of his new ideas and publications. They also were fully aware of Pauli's neutrino hypothesis, which Pauli presented for the first time for publication at the Solvay Conference,[68] with Fermi in the audience. Then, two months, later Fermi's theory of beta decay was rapidly disseminated throughout the community of physicists. For example, Pauli, at the Swiss Federal Institute of Technology (*Eidgenössische Technische Hochschule*) in Zurich, learned about Fermi's theory through Felix Bloch, who was then with Fermi in Rome on a Rochefeller Foundation fellowship, and who wrote to Gregor Wentzel, Pauli's colleague at the University of Zurich, on December 24, 1933, reporting that:

> Fermi [figure 9] has made a beautiful theory of β-decay emission, introducing the neutrino, which so simply reproduces the empirical facts that I believe in it strongly. The mass of the neutrino should be essentially zero, or in any case much smaller than that of the electron.[69]

It thus seems likely that Joliot and Curie also learned about Fermi's pioneering theory of beta decay in late 1933 or early 1934.

Joliot and Curie's Laboratory Notebook

Joliot first observed artificial radioactivity on the afternoon of Thursday, January 11, 1934.[70] Since, however, he and his wife Irène had a dinner engagement that evening, he asked Wolfgang Gentner to check whether his Geiger-Müller counter was functioning properly.

The following morning Joliot found a note from Gentner saying that it was,[71] so Joliot immediately began making entries into a new laboratory notebook, which today is preserved in the Joliot-Curie Archives. Its title, in Joliot's handwriting, is "*Radioactivité artificielle – 1re Experiences. Wilson statistique energie.*" It is 146 pages long. With the exception of pages 42-44, which are in Curie's handwriting, all are in Joliot's handwriting. Page 1, dated January 12, 1934, through page 62, dated March 22, 1934, deal with measurements of artificial radioactivity induced by alpha particles; page 63, dated April 17, 1934, through page 138, dated December 26, 1934, deal with artificial radioactivity induced by neutrons.

As seen in figure 10 (upper left-hand corner), Joliot first measured the background radiation, his Geiger-Müller counter recording 27 counts (4847–4820) in 3 minutes, or 9 counts per minute, after which he placed an aluminum foil (thickness 5/100 millimeter) ½ millimeter above the source, recording 48 counts (4895–4847) in 5 minutes, or 9.6 counts per minute. He then placed the aluminum foil closer to his Geiger-Müller counter and recorded 85 counts (4980–4895), which, assuming these also were due to background radiation, means that it took him around 9 minutes to reposition the aluminum foil. He therefore spent about 17 minutes taking background-radiation measurements before beginning his actual measurements at count 4980, recording 16 counts, one every ¼ minute for a total of 4 minutes, until count 5601. Note that in the first ¼-minute interval he recorded 51 counts (5031–4980) or 204 counts per minute, in other words, more than 20 times that of the background radiation at 9.6 counts per minute. At the end of this run, the number dropped to 124 counts per minute, still around 13 times higher than the background radiation. He then took 6 measurements, one every ½ minute for a total of 3 minutes (from count 5601 to count 5829) and 5 measurements, one every 1 minute for a total of 5 minutes (from count 5880 to count 6002), by which time the number dropped to 17 counts per minute. On the following page (not shown), he recorded 3 more 2-minute measurements (to count 6111), the last interval being at 13 counts per minute. He then took another background-radiation measurement, recording 75 counts in 8 minutes, or 9.4 counts per minute, which was consistent with his initial background-radiation measurement.

Joliot next irradiated a nickel foil (thickness 5/100 millimeter), recording 88 counts (6363–6275), which he reported as 9 counts per minute and as a "negative" result. This allowed him, as he and Curie reported in their paper of January 15,[72] to exclude the possibility that his earlier positive results for aluminum were due to some "contamination" of his polonium α-particle source.

Joliot next inserted a lead absorber (thickness 1 millimeter, which corresponds to a thickness in air of 1.5 meters for positrons) between the aluminum foil (thickness 5/100 millimeter) and his Geiger-Müller counter, recording 17 counts (6481–6464) in 1 minute. Over the next couple of minutes, between counts 6481 to 6538, he reduced the thickness of the lead absorber to ½ millimeter and then took 8 1-minute measurements (between counts 6538 and 6673), finding 31, 18, 18, 17, 10, 14, 17, and 11 counts per minute, which proved that the emitted positrons decayed exponentially even after passing through a lead absorber. In all, Joliot's experiments on aluminum took around an hour and a half, after which he carried out similar experiments on boron with a positive result, and on lithium (negative) , magnesium (positive), calcium fluoride (negative), and sodium (negative). He recorded his last experiments on January 12, 1934, the day after he first observed artificial radioactivity, on page 11 of his notebook.

Immediate Reception

The Editor of *Nature*, who was especially impressed by the long 14-minute half-life of the positrons emitted when polonium α particles bombarded boron, wrote to Joliot on January 31, 1934:

> The Editor of Nature presents his compliments to Monsieur F. Joliot and in confirmation of a telegram just dispatched to monsieur Joliot begs to say that he will be very glad indeed to publish in NATURE an account of Monsieur Joliot's experiments upon the atomic bombardment of boron with the result that a new source of radioactivity is produced. If Monsieur Joliot has already published a paper upon the subject, perhaps he could send a copy of that to the Editor for publication in NATURE, either in the original French language or translated into English. In any event, the Editor is sure that scientific readers all over the world would welcome any account which Monsieur Joliot may be able to send the Editor on his recent work and the results obtained.[73]

The Editor acknowledged receipt of Curie and Joliot's paper around February 6, 1934, and published it in the February 10 issue of *Nature* under the title, "Artificial Production of a New Kind of Radio-Element."[74] In it they presented the results of their experiments and signaled, as Perrin had suggested at the Solvay Conference, that the "positrons of aluminium seem to form a continuous spectrum similar to the β-ray spectrum," so as "in the case of the continuous spectrum of β-rays, it will be perhaps necessary to admit the simultaneous emission of a neutrino (or of an antineutrino of Louis de Broglie) in order to satisfy the principle of the conservation of energy and of the conservation of the spin in the transmutation."[75]

Joliot and Curie continued by reporting on their recent measurements, as they already had in a letter to Ernest Rutherford on February 2 and in a note to the *Académie des Sciences* on February 5,[76] that despite the miniscule amount of various radioactive elements that were produced (on the order of 10^{-16} gram),[77] and despite their short life-times, they had managed to "identify the chemical nature of the atoms formed," that is, nitrogen and phosphorus in the case of boron and aluminum, which "provide the first chemical proof of these transmutations and of the capture of α particles by these nuclear transformations."[78]

Joliot and Curie's discovery suggested that new radioactive elements could be produced in other ways. Rutherford, for example, in a letter of January 29, 1934, congratulated them on their "fine piece of work which I am sure will ultimately prove of much importance," adding by hand that "we shall try to see whether similar effects appear in proton-diplon [proton-deuteron] bombardament."[79] Joliot and Curie replied on February 2, endorsing Rutherford's suggestion: "It's actually very important to try to bring these types of radioactivities by bombarding the matter with other projectiles beyond alpha particles."[80] And in their paper in *Nature*, they reiterated: "These elements and similar ones may possibly be formed in different nuclear reactions with other bombarding particles: protons, deutrons [deuterons], neutrons."[81]

Positive results were soon found for protons at the Cavendish Laboratory in Cambridge, England, and for deuterons at the University of California in Berkeley and at the California Institute of Technology in Pasadena.[82] Neutron-induced artificial radioactivity was more difficult to achieve, because neutrons could be produced only at very low intensities. Chadwick, for example, noted that the "greatest effect is given by beryllium, where the yield is probably about 30 neutrons for every million α-particles of polonium which fall on a thick layer of beryllium."[83] Fermi, however, stimulated by his theory of beta decay, made a bet in March 1934 that it would be possible to induce artificial radioactivity by neutron bombardment provided that special attention was paid to the geometry of the experimental apparatus. As we now know, he soon won his bet.

Conclusions

On January 2, 1935, The Nobel Prize for Chemistry Committee invited Rutherford to nominate candidates for 1935, and in response he proposed "Monsieur and Madame Curie-

Joliot jointly for a Nobel Prize in Chemistry." After referring to "the great interest and importance" of their discovery, he observed:

> It is not as if attempts had not been made before to produce artificial radio-elements by bombardment with α-particles. I, and I believe also Madame Curie, had tried such experiments in the old days using electroscopes, but without success. It required a fortunate combination of strong α-ray sources with the use of the sensitive β-ray counter to make success possible. [84]

We have shown that both the use of an intense polonium α-particle source of about 60 millicuries--that is, of the same order of magnitude as those used in the experiments that led to the discoveries of the neutron and positron--and the introduction of the Geiger-Müller counter played crucial roles in the discovery of artificial radioactivity. These technical factors, however, were not all that were required. The choice of aluminum as the sample to be irradiated with α particles was also crucial. It was first necessary to show that aluminum emits neutrons, and then that positrons are also emitted. Only then was it possible to focus on the origin of these particles and to suggest, as Perrin did at the seventh Solvay Conference, that an intermediate unstable isotope of phosphorus could be formed, leading to a new type of positron beta decay. We have argued, however, that the transition from Perrin's suggestion to Joliot's experiment also required the emergence of a new theoretical framework that gave credence to Perrin's ideas. Only then was the groundwork prepared, both experimental and theoretical, for Joliot's fortunate combination of an intense α-particle source and a sensitive β-particle detector, which led him and Curie to their discovery of artificial radioactivity.

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