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The discovery of radioactivity

La découverte de la radioactivité

Pierre Radvanyi [a], Jacques Villain [b]

[a] *Institut de physique nucléaire d'Orsay, 15, rue Georges-Clemenceau, 91406 Orsay cedex, France*
[b] *Institut Laue-Langevin, 71, avenue des Martyrs, CS 20156, 38042, Grenoble cedex 9, France*

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ABSTRACT

The radioactivity of uranium was discovered in 1896 by Henri Becquerel who, starting from a wrong idea, progressively realized what he was observing, regularly informing the French Academy of Sciences of the progress he was doing. In the next years, it was found that thorium was radioactive too, and two new radioactive elements, polonium and radium, were discovered by Pierre and Marie Curie, while a third one, actinium, was identified by André Debierne. The study of the penetrating power and of the effect of electric and magnetic fields allowed scientists to demonstrate the complexity of nuclear radiation with its three components α, β, γ. The *Comptes rendus de l'Académie des sciences* allow the reader to see how difficult it was to understand the nature of radioactivity, which was essentially elucidated by Ernest Rutherford and Frederick Soddy.

RÉSUMÉ

La radioactivité de l'uranium a été découverte en 1896 par Henri Becquerel qui, partant d'une idée fausse, a progressivement réalisé ce qu'il était en train d'observer, informant régulièrement l'Académie des sciences des progrès qu'il faisait. Au cours des années qui ont suivi, il fut découvert que le thorium était également radioactif, et deux nouveaux éléments radioactifs, le polonium et le radium, furent mis en évidence par by Pierre et Marie Curie, tandis qu'un troisième, l'actinium, était identifié par André Debierne. L'étude du pouvoir de pénétration et de l'effet des champs électriques et magnétiques permit aux scientifiques de démontrer la complexité de la radiation nucléaire, avec ses trois composantes α, β et γ. Les *Comptes rendus de l'Académie des sciences* permettent au lecteur de réaliser combien il fut difficile de comprendre la nature de la radioactivité, qui a été essentiellement élucidée par Ernest Rutherford et Frederick Soddy.

E-mail address: villain@ill.fr (J. Villain).

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1. Henri Becquerel's finding: uranium spontaneously emits radiation

Five years before the end of the nineteenth century, nobody suspected that matter can emit radiation, except if it is heated, or submitted to a high voltage. The best understood radiation was light, well described by Maxwell's theory, although not yet quantized. The nature of cathode rays, discovered in 1869 by the German physicist Johann Hittorf, was still debated. And the radiation discovered in 1895 by Röntgen was particularly mysterious, and for that reason was called X-rays by its discoverer.

During the following years, a new phenomenon was discovered: radioactivity. The seeds of the discovery were sown by the person of Henri Poincaré and the subject of X rays. The mathematician was interested in physics, and he could read German. He had received a copy of Röntgen's original paper, and on 20 January 1896 he gave a talk on X-rays at the "Académie des sciences" in Paris. He had an idea on X-ray emission: he suggested that it might be a result of the fluorescence of the glass of the Crookes tube in which cathode rays were produced at low pressure, and that this phenomenon might be a general effect of fluorescence. Among the academicians of the audience, there was in fact a specialist of fluorescence: Henri Becquerel, Professor at the "Muséum d'histoire naturelle". He was very interested and, when he returned to the Muséum, he decided to perform experiments on a fluorescent material he possessed, and which turned out to be potassium-uranyl sulfate.

Luminescence (phosphorescence or fluorescence) is the property of certain materials to absorb light and reemit a light at a different frequency, after a time that may reach several hours, but is much shorter in the case of potassium-uranyl sulfate. The most common light source at that time was the Sun, and this detail will turn out to be important.

During the following weeks and months, Becquerel progressively realized that (*i*) the initial idea was wrong and (*ii*) starting from this wrong idea, he was making a great discovery. He periodically kept his colleagues of the "Académie des sciences" aware of his findings, and we can follow the development of his reasoning in the *Comptes rendus* [1].

In the middle of February Becquerel placed his uranium salt on a photographic plate, wrapped in a very thick black paper, and exposed the package to sunlight during a few hours. "After developing the plate, one sees the outline of the phosphorescent substance in black", he wrote, and then concluded that "the phosphorescent substance emits radiations able to traverse a paper which is opaque for light" [1a].

This statement was carefully formulated and correct, but it is likely that in February 1896 most of Becquerel's colleagues believed that it was a general property. One of them wrote: "Fluorescent bodies emit radiations which have the same properties as X rays, as suggested by Mr. Poincaré" [2]. He used the plural, bodies, and again the plural, properties, while Becquerel was careful enough to specify which property had been observed, and to speak of "the substance", the single one which he had studied. The next task was to check whether the radiation was really that discovered by Röntgen. For this purpose, Becquerel investigated the absorption of the rays emitted by his potassium uranyl sulfate, and found they can cross not only a thick sheet of black paper, but also an aluminum plate or a thin copper foil, as he mentioned on 2 March [1b]. However, he also reported an unexpected fact, related to the intermittence of the light source he was using: "Some of these experiments had been prepared on 26 and 27 February, and, since the Sun shined only intermittently on those days, I had kept the experiments that I had already prepared in a drawer, leaving the uranium salt [on the photographic plate]. As the Sun did not show up during the next days either, I developed the photographic plates on 1 March, expecting to find very weak images. Instead, the outlines appeared very intense. I concluded that the action had probably continued in the dark ...."

Becquerel began to doubt that the radiation he was discovering was a general property of luminescent materials. On 9 March, he announced [1c] that he investigated the possibility of radiation from hexagonal blende (α ZnS), which is highly phosphorescent, and nothing happened.[1] However, the title of his note, as those of the two preceding ones, still contains the word "phosphorescent". Two weeks later, it had disappeared, and the new note [1d] is devoted to *radiations emitted by uranium salts*. In May, Becquerel announced that metallic uranium radiates even more intensely than its salts. It is, he wrote, "the first example of a metal showing a phenomenon analogous to an invisible phosphorescence" [1e]. Thus, he had not yet abandoned the idea that he was observing something resembling phosphorescence. However, in November, the uranium samples, although they had been kept in the dark since 3 March, were still emitting a radiation, which he now called "uranic rays" [1g]. "The duration of the emission of these uranic rays is far beyond ordinary phosphorescence phenomena, and one cannot yet understand where uranium takes the energy it emits with such a long persistence."

Becquerel then tried to determine whether these radiations were really X-rays. On 9 March, he observed [1c] that the radiation from uranium could discharge a gold leaf electroscope, as do X-rays. The effect was studied more quantitatively in the next two weeks, with various materials inserted to slow down and absorb the radiation [1d]. In November, he concluded that "the discharge of electric bodies by gases which have been exposed to uranic rays [...] establishes a new relation between X rays and uranic rays which, with respect to reflection and refraction, appear to be quite different phenomena."

In April of 1897, Becquerel put an end to his study of "uranic rays". The newly discovered Zeeman effect looked more promising. Two newcomers, however, were attracted by uranic rays: Pierre and Marie Curie. Becquerel has always adhered

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[1] Becquerel's paper [1c] is followed by a paper of Academician Troost "on the use of artificial hexagonal blende to replace Crookes tubes" for X-ray production! He claimed to have obtained "results which confirm the hypothesis of our colleague Henri Poincaré" [...] and "allow us to substitute a simple instrument, easy to handle and of an infinite lifetime to Crookes tubes [...] which easily break." Thus, on the same day, in the same journal, two academicians reported conflicting experimental results! This should not be possible now.

to the photographic method of measurement, sometimes to the gold foil electroscope, never to the "electric method", which would have allowed more quantitative measurements. Pierre and Marie Curie, as well as Ernest Rutherford will use the electric method.

2. Not only uranium spontaneously emits radiation!

Pierre Curie, 38, was already a well-known physicist. For his works on piezoelectricity, together with his brother Jacques Curie, he had obtained a prize of the Académie des Sciences. He had then made a systematic study of the magnetic susceptibility of a number of materials as a function of temperature, and the well-known Curie law testifies to his successful research. In 1895 he had married Maria Skłodowska, a Polish student, 8 years younger than him. At this time, female students were not accepted in the universities in Poland, which was a part of the Russian empire. Pierre had obtained his doctorate and Marie her "licence", and, after the birth of their first daughter, Irène, she was looking for a subject for a doctorate. Uranic rays might be a good idea, suggested Pierre. He provided his wife with instruments that allowed precise measurements of the radiation: "I used", wrote Marie Curie [3] "a capacitor (*now called ionization chamber*) made of two plates; one of the plates was covered with a uniform layer of uranium or any other substance in fine powder [...] The voltage between the two plates was 100 volts. The current through the capacitor was measured by an electrometer and a piezoelectric quartz." The quadrant electrometer she used was an invention of Lord Kelvin, modified by Pierre Curie. The properties of piezoelectric quartz had been discovered by Jacques and Pierre Curie.

"I studied the electrical conductivity of air under the effect of the rays of uranium", wrote Marie Curie [3], "and I sought other materials than uranium compounds which could provide air with an electrical conductivity [...] I examined a large number of metals, salts, oxides and minerals." One of the problems was to obtain the samples, but the young woman was able to find help from older colleagues. The result was positive: not only uranium emits radiation, thorium is active too! "It is remarkable", writes Marie, "that the two most active elements, uranium and thorium, are those that have the highest atomic weight." It is now known that indeed heavy nuclei are unstable because short-range nuclear forces are not able to compensate the Coulomb repulsion between protons.

At the same time, in Germany, Gerhard Carl Schmidt had discovered that thorium emits radiation [4]. Marie Curie's three-page note to the *Comptes rendus* was published on 12 April and Schmidt's eleven-page article on 23 April, but had been sent on 24 March.

Marie Curie's star was only beginning to shine. She had noticed something important: two uranium ores, pitchblende (uranium oxide) and chalcolite (copper and uranium phosphate) were much more active than uranium. "This fact is very remarkable and suggests that these ores may contain an element much more active than uranium." Her guess was correct. Pierre Curie ceased his research on magnetism and joined his wife in the study of radioactivity. They decided to use a new method of chemical analysis: it was based on the emission of radiations. "Our chemical research has been guided by the control of the radiative activity of the products separated at each step of the operation." The element looked for was concentrated in parts, which became more and more radioactive as separation progressed. On 18 July 1898, they jointly announced the discovery of a new element [5], which they proposed to call *polonium*. For the first time, the word *radio-active* appears in a scientific publication, while the former "uranic rays" are now called "Becquerel rays", since uranium is not the only element emitting them.

On 19 December Pierre and Marie Curie announced the discovery of another, very radioactive element in pitchblende [6]. The help of a chemist, Gustave Bémont, and of a spectroscopist, Eugène Demarçay, made this discovery undisputable. The new element was chemically analogous to barium, and therefore quite difficult to separate, but the presence of an unknown spectroscopic line testified to its presence. Its discoverers called it *radium*.

Pierre and Marie Curie did not exclude finding new radioactive elements in pitchblende, but they asked a coworker to look for them. Indeed, André Debierne discovered a new radioactive element, as he reported in a note of 16 October 1899 [8]. This element was later called *actinium*.

2.1. The importance of radium

The half-life of radium is 1600 years, which is very much shorter than the half-life of uranium ($4.5 \cdot 10^9$ years), and therefore, for a same weight, its radiation is much more intense. For the study of radioactivity, radium was much more convenient than the very weakly radioactive uranium. The rays emitted by radium became a fantastic tool to explore the microscopic structure of matter. Medical applications appeared at the end of 1901. However, radium is also very rare and very expensive. Indeed there is 0.15 g of radium in 1 ton of pitchblende.

While uranium was the first radioactive element to be discovered, radium was much more popular, as it was a spontaneously luminous material that emitted an incredible quantity of radiation. The popularity of radium is exemplified by a novel by Maurice Leblanc, *The Island of Thirty Coffins*, published in 1919 where a central role is played by a stone "shivering with radium, from where goes steadily a bombardment of invigorating and miraculous atoms."

The research that led to the discovery of radium in 1898 had been performed in very difficult conditions, in premises that were not adequate, without funding. Pierre Curie succeeded in getting uranium ore from Bohemia, at that time pertaining to Austria. Two years later, Pierre and Marie Curie had become famous throughout the world and the situation had improved very much. "The preparation of radium has been very expensive. We thank the Académie des sciences [...]" wrote Marie

Curie a little later [7]. The help of the Austrian government, which gave one ton of pitchblende, was also acknowledged, as well as the help of the chairman of the Austrian Academy of Sciences.

The collaboration between Pierre and Marie Curie was exemplary in many respects. They complemented each other. Pierre was dreamy and imaginative, ready to undertake various difficult projects at the same time or successively. Marie was full of energy and tenacity when pursuing the goal she had in mind.

2.2. The nature of the radiations emitted by radioactive atoms

What are Becquerel rays? The answer came from various countries. Research on radioactivity was becoming international. From New Zealand, a bright, young physicist had landed in England: Ernest Rutherford. He carefully investigated the absorption of uranium radiation by an increasing aluminum thickness and he concluded that "[t]hese experiments show that the uranium radiation is complex, and that there are present at least two distinct types of radiation, one that is very readily absorbed, which will be termed for convenience the α radiation, and the other of a more penetrative character, which will be termed the β radiation" [9].

The readily absorbed radiation is still now called α radiation. The more penetrative one was complex, as it appeared in the next years. A natural idea was to investigate the effect of a magnetic or an electric field, which were known to deviate cathode rays. Actually, a part of the rays emitted by radium were found to be easily deviated by a magnetic field, as was first demonstrated by Friedrich Oskar Giesel [10] in Germany, then checked by Stefan Meyer and Egon von Schweidler [11] in Austria and by Henri Becquerel [12]. In 1900, it was known that "radium's radiation contains two groups: rays which are deviated by a magnetic field and rays which are not deviated by a magnetic field" [13]. The "deviable" rays, now called β^- rays, were found to be identical with cathode rays, the nature of which had been clarified by Jean Perrin and Joseph John Thomson in the last decade of the nineteenth century: they were electrons. Thus, if classified according to their "deviability", there were two categories of radiations, and if classified according to their penetrative power, there were also two categories. It was realized a little later that there are three. The problem was that (i) the magnetic fields available at that time did not appreciably deviate α rays, and (ii) gamma rays, which have a high penetrating power, were not easy to detect.

However, they were observed in 1900 by Paul Villard [14]. He reported experiments in which he compared the penetration in glass of magnetic field deviable rays and non-deviable rays. The glass (actually photographic plates) had been wrapped in a thick black paper that eliminated α rays. From his experiment, Villard concluded that "the non-deviable part of radium's radiation contains very penetrative rays." This was the act of birth of gamma rays, that Villard called "X rays emitted by radium"; the word "gamma rays" was coined in 1903 by Rutherford. Becquerel, at the beginning, was skeptical[2] [15], but recognized that "if Mr. Villard's observations are right, the reason of the disagreement might be the existence of less intense and very penetrating rays" (in fact less ionizing rays).

It took more than six years to elucidate the nature of α rays [16], the range of which in air at normal pressure is quite short. Ernest Rutherford showed in 1902 [17] that they can be deflected by a very strong magnetic field and that they carry a positive charge. In 1903, the experiments of Pierre Curie and Albert Laborde [18] indicated that the α particles are strongly ionizing. Ernest Rutherford and Frederick Soddy concluded that each succession of decays of radioactive atoms should finally lead to the formation of stable atoms [19,20]. Experiments performed in London by William Ramsay (the discoverer of rare gases) and Frederick Soddy [21] showed conclusively that helium appeared in radium and emanation (radon) sources. Finally, the measurement of the ratio e/m of the α particles indicated that these are completely ionized helium atoms.

2.3. Where does the energy come from? The nature of radioactivity

One learns now at school that, about 14 billion years ago, the Universe was very hot and allowed for the formation of many stable or unstable elements, among which uranium and thorium, which are both unstable, but very weakly unstable. The period of ^{238}U is 4.5 billion years and the period of ^{232}Th is 14 billion years, so that there is still some uranium and a lot of thorium left. There is also radium and radon because these are constantly generated by uranium and thorium (see Fig. 1, Table 1 and Fig. 2).

Rutherford and Soddy came rather quickly (in 1902) to a picture of this type, but Pierre and Marie Curie were hesitating between two hypotheses: "either a rather large quantity of energy was stored a long time ago [in radioactive materials], or there are energy sources in space, that these materials are able to use" [22]. In January 1902, a Note by Pierre and Marie Curie [23] shows that they were on a wrong trail. They made indeed the hypothesis that "each atom of a radioactive material works as a constant energy source." They added however: "Experiments of several years show that for uranium, thorium, radium [. . .] the radioactivity [. . .] does not change in time." This was a mistake: they should rather have given an upper limit to the rate of change. From the values of the periods we know now, it is readily seen that this rate of change

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[2] « Si le chlorure de radium qui a servi à ces expériences émettait avec une intensité comparable à celle du rayonnement étudié des rayons non déviables très pénétrants, l'existence de ces rayons n'aurait pu échapper aux expériences de M. et M^{me} Curie ou aux miennes, et, si les observations de M. Villard sont exactes, il faudrait chercher la cause du désaccord, soit dans la nature du produit actif qu'il a employé, soit dans l'existence de rayons moins intenses [* en réalité moins ionisants] et très pénétrants, comme ceux de l'uranium, dont l'effet n'apparaîtrait qu'après une longue pose. »

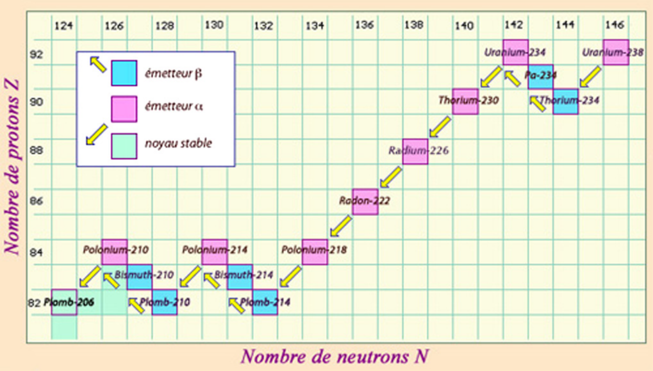

Fig. 1. The radioactive series of uranium-238, as we know it today. Atoms have nuclei (Rutherford, 1911) that are composed of protons and neutrons (Chadwick, 1932). (http://www.laradioactivite.com/site/pages/desintegrationencascade.htm).

Table 1
The periods of uranium 238 and its daughters (http://www.laradioactivite.com/site/pages/desintegrationencascade.htm).

	Period	Unit	Emitter
Uranium-238	4.468	billion years	alpha
Thorium-234	2.410	days	beta^{-}
Protactinium-234	6.70	hours	beta^{-}
Uranium-234	24.5500	years	alpha
Thorium-230	75,380	years	alpha
Radium-226	1600	years	alpha
Radon-222	3.8235	days	alpha
Polonium-218	3.10	minutes	alpha
Lead-214	26.8	minutes	beta^{-}
Bismuth-214	19.9	minutes	beta^{-}
Polonium-214	164.3	microseconds	alpha
Lead-210	22.3	years	beta
Bismuth-210	5.015	days	beta
Polonium-210	138.376	days	alpha
Lead-206	Stable		

is not directly measurable for uranium and thorium. The period of radium is much shorter (1600 years), but still too long to produce an appreciable decrease in activity in a few weeks. The law of radioactive decay could only be observed directly for an element with a much shorter lifetime, i.e. polonium (138 days) or radon (3.8 days); such elements, generated by the decay of uranium or thorium, must be separated chemically. Becquerel [24] suspected some kind of transformation by chemistry: he observed: “after certain [chemical] treatments, certain uranium compounds became less active.” The chemical treatment included the separation of precipitates that “could become appreciably more radioactive than uranium”. After a few months, uranium compounds had again become as active as they were before the chemical treatment. However, Becquerel could not explain these observations (they indicated the return to radioactive equilibrium).

In 1902, working together in Montreal on the radioactive daughter atoms of thorium, Ernest Rutherford and Frederick Soddy showed experimentally that radioactivity is the spontaneous transformation of one element into another through the emission of radiation [19,20]. A succession of such transformations forms a radioactive series.

While Becquerel’s observations were only qualitative, Rutherford and Soddy [19,20] derived quantitative laws from their experiments, however not performed on uranium, but on thorium. They established that radioactive decay is exponential, following the law:

$$N = N_0\, e^{-\lambda t}$$

N being the number of radioactive atoms present at time t and N_0 being this number at $t = 0$; λ is the probability for any particular atom to disintegrate per unit time; the period or half-life T for a specific species of radioactive atoms is: $T = \ln 2/\lambda = 0.693/\lambda$.

If the radioactive atoms resulting from such a decay are also radioactive, they will in turn decay with their own period. In this way, we have whole series of successive decays. Even though Rutherford and Soddy did not know yet the whole sequence of atomic decays, the exponential behaviors observed by them suggested that radioactivity represents an irreversible transformation of matter. Thus, the first of the two possibilities considered by Pierre Curie was correct: a huge quantity of energy was stored a long time ago in radioactive atoms. The second hypothesis, that there are energy sources in space, a sort of ether, which these atoms can use, is not valid.

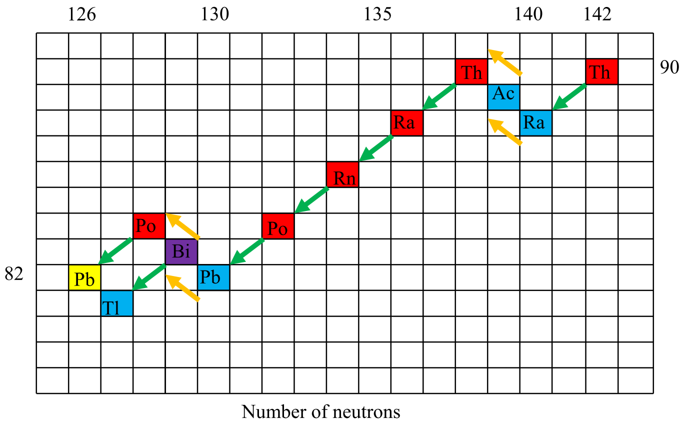

Fig. 2. The radioactive series of thorium, as we know it today. The vertical direction shows the number of protons. Green arrows indicate alpha decay, yellow arrows indicate beta decay. The elements in red are alpha emitters, those in blue are beta emitters; the one in purple means both, that in yellow is stable. Rutherford and Soddy identified the first part of this cascade in 1902–1903.

One should remember that, in 1903, scientists did not know neither the existence of isotopes (Frederick Soddy, 1911), nor the existence of a nucleus inside the atom (Ernest Rutherford, 1911), radioactivity being a nuclear property. The presence of electrons moving inside the atom had been demonstrated by P. Zeeman and H.A. Lorentz (1897).

It will be the merit of Pierre and Marie's daughter, Irène, and their son-in-law, Frédéric Joliot, to discover that one can create radioactive species (radioactive isotopes) of all chemical elements [25].

2.4. The Comptes rendus de l'Académie des sciences: a unique case

The *Comptes rendus de l'Académie des sciences* allow one to follow the progress of Becquerel's discovery from week to week, or at least from month to month during the year 1996 [1]. He informed the Academy about his latest discoveries, but also his plans and his doubts. In 1900 again, the quarrel between Becquerel and Villard [14,15] yields a vivid testimony of the difficulty to reach the truth in physics. Becquerel's colleagues acted in a similar way. They were not particularly eager to publish well organized articles, in contrast with Rutherford and Soddy. Pierre and Marie Curie published their discoveries in the *Comptes rendus*, but, not being members of the Academy, had fewer pages at their disposal. However, an exception was Henri Poincaré, who published many notes in the *Comptes rendus de l'Académie des sciences*, but also books and detailed articles.

Appendix A. Supplementary material

A French version of this article can be found on-line at https://doi.org/10.1016/j.crhy.2017.10.008.

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