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The discovery of radioactivity

La découverte de la radioactivité

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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.

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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

[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.

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