

THE FIRST THREE YEARS OF RADIOACTIVITY THE CONSEQUENCES OF THE DISCOVERIES OF POLONIUM AND RADIUM*

ANDRZEJ KAJETAN WRÓBLEWSKI

Institute of Experimental Physics, Warsaw University
Hoża 69, 00-681 Warszawa, Poland

(Received November 12, 1998)

Natural radioactivity was discovered by Henri Becquerel in February, 1896, but for over two years it remained on the periphery of interest of physicists. It was the research by Maria Skłodowska-Curie and then also by her husband Pierre in 1898 that began a new era in physics.

1. Accidental discovery

It is known that radioactivity of uranium was discovered by accident, as a consequence of the discovery of X-rays by Wilhelm Conrad Röntgen. After publication of Röntgen's paper on December 28, 1895, the whole world became fascinated by extraordinary properties of the new rays. They became the subject of discussion in all circles.

At the meeting of the Academy of Sciences in Paris on January 20, 1895, Henri Poincaré described Röntgen's discovery [1] and proposed a hypothesis that emission of X-rays could be related to phosphorescence, or delayed emission of light by a substance after its exposure to light [2].

Henri Becquerel, who was present at the meeting, decided to check experimentally Poincaré's hypothesis. As he later said in his Nobel lecture [3]: "At the beginning of 1896, on the very day that news reached Paris of Röntgen's experiments and of the extraordinary properties of the rays emitted by the phosphorescent walls of the Crookes tubes, I thought of carrying out research to see whether all phosphorescent material emitted similar rays. The results of the experiment did not justify this idea, but in this research I encountered an unexpected phenomenon."

* Presented at the International Conference "Nuclear Physics Close to the Barrier", Warszawa, Poland, June 30–July 4, 1998.

Becquerel had in his laboratory a sample of double sulphate of uranium and potassium, $\text{K}_2[\text{UO}_2(\text{SO}_4)_2] \cdot (\text{H}_2\text{O}_2)$, well known for its property of phosphorescence. He exposed it to sunlight and then checked that it caused blackening of a photographic plate wrapped in a black paper. On February 24 Becquerel presented this result [4] to the Academy of Sciences, convinced that Poincaré's hypothesis was confirmed.

He, nevertheless, continued experiments. Meanwhile, the weather in Paris changed and the sun was seldom visible. Waiting for weather improvement Becquerel kept the little exposed mineral and the plate in a drawer. After a few days he decided to develop the plate and found, to his surprise, that it was much blackened. He understood that his previous conclusion was incorrect and announced at the next meeting of the Academy of Sciences on February 2 that the uranium mineral emitted unknown penetrating radiation by itself [5]:

"I particularly insist on the following fact, which appears to me exceedingly important and not in accord with the phenomena which one might expect to observe: the same encrusted crystals placed with respect to the photographic plates in the same conditions and acting through the same screens, but protected from the excitation of incident rays and kept in the dark, still produce the same photographic effects. I may relate now how I was led to make this observation: among the preceding experiments some had been ready on Wednesday the 26th and Thursday the 27th of February and as on those days the sun only showed itself intermittently I kept my arrangements all prepared and put back the holders in the dark in the drawer of the case, and left in place the crusts of uranium salt. Since the sun did not show itself again for several days I developed the photographic plates on the 1st of March, expecting to find the images very feeble. The silhouettes appeared on the contrary with great intensity. I at once thought that the action might be able to go on in the dark."

Thus, an erroneous hypothesis and bad weather in Paris led Becquerel to discovery, which later brought him the Nobel prize in physics. The news about the new rays spread rapidly around the world. As reported in "Nature" of March 12, 1896 [6]:

"The curious result has been obtained that although the plate-holder and the uranium salt are not exposed to the light, but kept inside a wooden or cardboard box, the photographic plate shows the same images as when the salt is exposed to light. The author rather tentatively suggests that the uranium salt may continue to emit phosphorescence radiation that is invisible to the eye, but which is capable of traversing paper and aluminium for a time infinitely great compared with the time during which it continues to emit visible light."

2. Becquerel or Niepce de St. Victor?

Let us go back to the circumstances of Becquerel's discovery. As it was mentioned above, there was very bad weather in Paris in the last week of February, 1896. Becquerel's photographic plates wrapped in paper and prepared for exposure to sun rays were kept in his drawer. Nevertheless on March 1 Becquerel decided to rush to his laboratory and develop the unexposed plates. Now, March 1 was a Sunday! What prompted Becquerel to perform such a senseless operation instead of spending time at home with his family ?

It had been known, that in 1867 another Frenchman, Claude Niepce de Saint-Victor, one of the pioneers of photography, noticed that luminescent uranium salts could cause some "fogging" of photographic plates even if there were several sheets of paper between the salts and the plates. He did not follow up this observation but described it in several papers. It was also mentioned in a book [7] published in 1868 by Henri's father, Edmond Becquerel, who was an eminent physicist. At the time when the book was published Henri Becquerel was only 16 years old but he must have read his father's book later. It is therefore quite certain that he had information about Niepce's observation. Nevertheless, Niepce's name was not mentioned in any of Becquerel's papers on uranium radiation. This fact did not escape attention of other scientists. For example, in a 1899 account of "Becquerel rays" by Charles-Édouard Guillaume (who later won the 1920 Nobel Prize for physics) it was said that their penetrating property had been first seen by Niepce "whose work was forgotten" [8] and then seen again by Becquerel, who studied them in more detail.

Becquerel found it necessary to defend himself. In his book [9] published in 1903 he discussed the essence of Niepce's experiments and tried to prove that it was only the chemical effect of uranium salts on the plates which was seen in 1867. However, he said nothing on whether remembrance of Niepce's work could have prompted his uncommon action on March 1, 1896.

From the present perspective there could be no doubt that Henri Becquerel was the true discoverer of radioactivity. While Niepce de Saint-Victor suspected that the "fogging" of photographic plates was somehow related to luminescence, he did not try to check this supposition by further experiments nor seemed to be much surprised by the fact that light could penetrate the paper. Becquerel, who was a first class scientist, performed a series of well-planned experiments in which he systematically studied various properties of the new type of radiation. Although, as we shall see below, some of his results obtained in 1896 were erroneous, there is little reason to diminish his glory. Nevertheless, Niepce de Saint-Victor is still being mentioned in connection with Becquerel's discovery [10].

3. Becquerel's crucial mistake

After the announcement made on March 2, 1896, Becquerel decided to study the newly discovered radiation in more detail. He presented the results of his studies at three meetings of the Academy of Science in March 1896. Firstly, on March 9, he announced that the rays emitted by the double sulphate of uranium and potassium kept in darkness for a few days are capable of discharging an electroscope after passing through a 2 mm thick aluminium plate. He found also that the invisible radiation may be reflected and refracted [11].

Becquerel consequently used photographic plates. He put uranium salt on a wrapped plate and mounted polished steel mirror over one half of the plate. After 55 hours of exposure the blackening of the two halves of the plate was clearly different, which convinced Becquerel that uranium rays underwent reflection by the mirror. In the next experiment he obtained similar result with a spherical mirror. To study the refraction of uranium rays a crown glass prism was used. It covered one end of a bell-shaped glass tube which was filled with uranium nitrate and had a photographic plate at the other end. Again, after a three day exposure Becquerel noticed an effect of refraction similar to that for visible light. In the following contribution, on March 23 he presented more detailed results on the ionizing power of the new rays. On comparing the rate of discharge of a gold leaf electroscope by the radiation from the potassium uranyl sulphate crystal and from a Crookes' tube, the effect of the tube was found to be over one hundred times greater than that of the crystal [12].

On March 30 Becquerel announced [13] that the rays emitted by uranium salts are doubly refracted by a tourmaline, a parallel experiment with a Crookes' tube giving a negative result. This time he used a 0.5 cm thick tourmaline piece with its faces parallel to the optical axis. The plate was then cut into two pieces, which were put on the wrapped plate such that the axes of the two were perpendicular to each other. Another tourmaline plate also cut parallel to the optical axis was laid on top of the first two in such a way that its axis was parallel to the axis of one of the pieces and perpendicular to that of the other. The photographic plate was apparently blackened differently in the parts under the two tourmaline pieces which Becquerel interpreted as due to double refraction and polarisation of the uranium rays.

Becquerel's experiments were carefully planned and performed but we know now that the results obtained by unreliable method of estimating the blackening of photographic plate were incorrect and caused decrease of interest in the new rays. Becquerel was well known and respected physicist so that his results were never put in doubt. The uranium rays appeared to

have “normal” properties, similar to that of ordinary light, hence they were regarded as better known than mysterious X-rays. At the five meetings of the Academy of Sciences in March 1896 there were more than 30 reports on X-rays and in this flood of reports the communications by Becquerel on uranium radiation could not cause great excitement.

Summary on the new rays in the middle of 1896
(unchanged until the spring of 1898)

Property	Röntgen rays	Uranium rays
Penetration through paper and aluminium	Yes	Yes
Penetration through heavier metals	No	No
Action on photographic plates	Yes	Yes
Ionization of air	Yes	Yes
Reflection	No	Yes*
Refraction	No	Yes*
Polarisation	No	Yes*
Nature	?	Very short ether waves

*Erroneous Becquerel’s results of March, 1896

4. A flood of ‘new rays’

There were also other reasons. The new field of invisible penetrating radiation attracted many researchers, some of them casual, who greatly contributed to the confusion by “discovering” great many imagined emissions and emitters. Thus, on January 27, hence before Becquerel’s first note on uranium rays, French physician Gustave Le Bon communicated to the Academy of Sciences his discovery of “black light” (*lumière noire*) [14]. He maintained that an ordinary paraffin lamp emits special radiation, which can penetrate through metallic plates. Le Bon placed a photographic plate under a lead or iron cover and after three hour exposure obtained an image

“which was nearly as vigorous as if no obstacle had been interposed between the light and the plate”. On February 3 Le Bon presented another report on photography with *lumière noire* [15]. This time he carefully eliminated possible influence of heat and light condensed in metal plates. During the same session G.H. Niewenglowski reported that he successfully repeated Le Bon’s experiments in total darkness (that is without any light source!) which seemed to indicate that the images on a photographic plate were due to radiation energy condensed in metals. According to G. Moreau it was possible to obtain an image on a plate closed in a box put near an ordinary induction coil (without the Crookes’ tube). On February 10 Charles Henry reported results of his experiments with zinc sulphide which seemed to make metals transparent to X-rays. On March 9 Troost confirmed that zinc sulphide could produce photographic images as clear as those produced by X-rays from the Crookes’ tube.

According to the reports from the United States at the end of February a certain dr. A. Mau succeeded in obtaining a photograph of a key, covered by a board, by five hours exposure to direct sunlight. Another American, S. Egbert, stated that he demonstrated the action of X-rays through plates of platinum from ordinary sunlight. These false results were corrected after a few months but meanwhile many people tended to “see” everywhere sources of radiation, capable of penetrating covers which stopped ordinary light.

Other researchers “discovered” even more exotic sources of invisible penetrating radiations. It is enough to mention that, for example, professor of electrical engineering at the Alabama Polytechnic Institute, A.F. Mc Kissick, included even sugar and chalk in his list of “active emitters”. Wilhelm Arnold in Erlangen announced that Becquerel rays were emitted by zinc sulphate, fluorite, various mixtures of sulphates and tungstenites, even by the organic compound retene ($C_{18}H_{18}$). Reports presented at the meetings of the Academy of Sciences in Paris confirmed Le Bon’s discovery. On March 16 Ellinger reported on successful repetition of all experiments with *“lumière noire”*. On May 11 Le Bon reported a sensational finding that *“lumière noire”* could be concentrated in metal plates. Such plates exposed for an hour to the light of electric arc were then used to cover photographic plates. Pictures produced in these plates were, according to Le Bon, due to *“lumière noire”* concentrated earlier in a metal.

In this atmosphere of interest in X-rays and numerous reports of many other sources of penetrating radiation there was little interest in the next report by Becquerel [16], who reported on May 23 that also metallic uranium emitted penetrating radiation, four times more intense than that from its salts.

But on July 6 Colson reported to the Academy of Sciences that also zinc with carefully cleaned surface was capable to influence photographic plates

and on July 13 Pellat reported similar finding for steel. Colson attributed the result to metal vapour, whereas Pellat — remembering Becquerel's results for metallic uranium — concluded that it might be a property of metals in general. On August 24 Henry reported that even light emitted by glow-worms could penetrate black paper. It was confirmed soon by Muraoka in Kyoto.

It took quite some time before the erroneous results were corrected. For example, Muraoka withdrew his results on glow-worms only in March 1898, and Le Bon still discussed "*lumière noire*" in 1900 [17].

In the Rede Lecture "The Röntgen Rays" given at Cambridge University on June 10, 1896, John Joseph Thomson said [18]: "Since the discovery of the Röntgen Rays, Becquerel has discovered a new kind of light, which in its properties resembles the Röntgen rays more closely than any kind of light hitherto known ... Becquerel has shown that the radiation from the uranium salts can be polarised, so that it is undoubtedly light: it can also be refracted. It forms a link between the Röntgen rays and ordinary light, it resembles the Röntgen rays in its photographic action, in power of penetrating substances opaque to ordinary light, and in the characteristic electrical effect, while it resembles ordinary light in its capacity for polarisation, in its liability to refraction ...

The radiation from the uranium salts is of special interest from another point of view. Sir George Stokes has shown that in the case of phosphorescence caused by sunlight or the arc lamp, the light emitted by the phosphorescent body is of longer wave-length than the light causing the phosphorescence; in the case, however, of the phosphorescence discovered by Becquerel, the light emitted is of shorter wave-length than the incident light ..."

5. Becquerel leaves the 'uninteresting' field

Meanwhile in August, 1896, Pieter Zeeman in Leyden discovered splitting of spectral lines in the magnetic field. Many physicists, including Becquerel, concentrated their attention on this long awaited connection between magnetism and light. Becquerel read his papers on Zeeman effect and Faraday effect at the meetings of the Academy of Sciences on November 8, 1897, January 17, April 4, July 4, October 31 and December 20, 1898, and on January 16, 1899.

While Becquerel in fact stopped working on seemingly little appealing problem of uranium radiation, he nevertheless published three more short notes on its properties on November 23, 1896 [19], March 1 [20] and April 12, 1897 [21]. In the last note he announced that the activity of the uranium salt did not change measurably during one year of study. But he was almost alone in the study of uranium rays which were generally regarded to be much

less fascinating subject than the X-rays. It is enough to say that in 1896 alone there were over 1000 papers and 50 books and booklets published on X-rays and only about 20 papers on uranium rays [22].

The next Becquerel's paper on radioactivity was read on March 27, 1899, a year after the first paper of Maria Skłodowska-Curie and several months after the discovery of polonium and radium. In the period between May 1896 and the spring of 1898 there was indeed rather little progress in the study of Becquerel rays.

The views of scientists at that period are well characterised in a short passage from the book by Warsaw University professor Wiktor Biernacki [23]. The book was published in Warsaw in the middle of 1898, thus it had been written much earlier, probably at the time when Maria Skłodowska-Curie decided to study uranium rays:

"Becquerel discovered that uranium salts were capable of emitting invisible rays which can penetrate aluminium and black paper impenetrable to light, and diffuse electric charges. Similarly to phosphorescent bodies which after exposure to light may shine up to several hours, so uranium salts send off these invisible rays but for much longer period of time. Some physicists call it hyperphosphorescence. Pure metallic uranium acts even stronger. Many other substances were found, *e.g.* zinc sulphate, calcium sulphate *etc.*, which emitted similar invisible rays. All these bodies are normally phosphorescent, but then, after they stop emitting visible light, still send off these invisible rays of properties similar to those of Röntgen rays. All these invisible rays are called Becquerel rays. They all diffuse electric charges but are absorbed in air even stronger than Röntgen rays. Becquerel also proved that these rays might be reflected, refracted and polarised ...

Glow-worms also emit similar rays, which can pass through aluminium and even thin copper sheets. Again, similar to Becquerel rays, they undergo reflection, refraction, and polarisation.

Each day brings new discoveries in this field. The number of substances capable of emitting rays hitherto unknown is steadily growing. Some of these substances are phosphorescent in visible and invisible light (*i.e.* hyperphosphorescent) and some other sent off only invisible rays of properties similar to those of Röntgen rays.

If we agree to treat X-rays as transverse vibrations of the ether, then the same may be accepted with even greater certainty for Becquerel and similar rays. But since they may be reflected, refracted and polarised, their properties are closer to visible light than to X rays, so that we should treat them as waves of the length between ultraviolet rays and X rays."

Detailed description of X-rays filled few dozen pages in Biernacki's book, while Becquerel rays were treated only shortly, and as we know now, most of the information concerning their properties was incorrect. Other physicists

Becquerel			Others	
			◀	X rays
		24 II	→	
1	Reports	2, 9, 23, 30 III	→	
			→	
8		23 V	→	
9	on			
	uranium			
6		23 XI	→	◀ Zeeman effect
	rays			
		1 III	→	◁ e/m Wiechert
1		12 IV	→	◁ e/m Thompson
8				◁ e/m Kaufmann
9				
7	Reports	8 XI	▶	
		17 I	▶	
	on			◀ 4 II Thorium rays (Schmidt)
1		4 IV	▶	
8	the			◀ 12 IV Thorium rays (MS-C)
		4 VII	▶	
9				◀ 18 VII Polonium
	Zeeman			
8		31 X	▶	
		20 XII	▶	◀ 26 XII Radium
	effect	16 I	▶	◀ Rutherford (α, β rays)
1	Report on uranium	27 III	→	
8	(Erroneous results			
9	of March 1896			
	retracted)			
9				

were of similar opinion. Let us quote for example a few sentences from a review paper by Oscar M. Stewart of Cornell University published in April, 1898 [24]:

“Becquerel rays occupy a unique position, inasmuch as far more is definitely known about them than any of the other ‘new’ ‘rays’. With X-rays nothing has been proven one way or the other about their character, save that if they are ultra-violet rays their wave-length must be extremely small, so small that the refractive index for nearly all bodies is practically unity. With the rays of Becquerel there can be no reasonable doubt that they are short transverse ether waves.”

6. Enter Maria Skłodowska-Curie

It is difficult to say how the history would be shaped if it were not for Maria Skłodowska-Curie who decided at the end of 1897 to study the “uninteresting” subject of uranium radiation. She just completed her study of magnetism of tempered steel; if she continued this applied research her name would probably never be widely known.

Maria’s first independent study of radioactivity [25] — the term she herself proposed — was a real break with the past. Firstly, she applied a precise and sensitive electrometer, much more reliable than the photographic method which gave qualitative, non-repeatable and often erroneous results because of the quality of then manufactured plates. Secondly, she decided to perform a systematic study of all available minerals, rocks and other substances. This at once resulted in a breakthrough, because it was found that the intensity of radiation from various uranium minerals was not proportional to the amount of uranium they contained. This led Maria Skłodowska-Curie to hypothesise on the existence of a new unknown radioactive element. Thanks to her systematic studies she discovered radioactivity of thorium. It was discovered independently by German physicist Gerhardt Schmidt [26]. He used a photographic method similar to that of Becquerel and found that thorium rays can be refracted and reflected (diffused) but not polarised.

Here is an excerpt of Maria Skłodowska-Curie’s paper [25]: “I have studied the conductivity of air under the influence of the rays from uranium, discovered by Mr. Becquerel, and I have sought whether any other bodies than those composed of uranium are able to render air a conductor of electricity. I used for this study a plate condenser; one of the plates was covered with a uniform layer of uranium or another substance pulverised. A difference of potential of 100 Volts was established between the plates. The current, which passed through the condenser, was measured in absolute value by means of an electrometer and a piezo-electric quartz.

I have examined a great number of metals, salts, oxides and minerals ... All the compounds of uranium studied are very active and they are, in general, the more active the more uranium they contain. The compounds of thorium are very active. The oxide of thorium even exceeds metallic thorium in activity. It should be noted that two most active elements, uranium and thorium, are those which have the greatest atomic weight ...

Two ores of uranium, pitchblende (uranium oxide) and chalcocite (phosphate of copper and uranium) are much more active than uranium itself. This fact is very remarkable and leads to the belief that these minerals may contain an element much more active than uranium ... To interpret the spontaneous radiation of uranium and thorium one might imagine that all space is constantly traversed by rays analogous to Röntgen rays but much more penetrating and able to be absorbed only by certain elements of high atomic weight, such as uranium and thorium."

7. Discovery of polonium

"It appeared that the results of my work were so interesting that Pierre Curie put aside his current research and joined me in the effort to extract and study new radioactive substances" — wrote Maria Skłodowska-Curie in the introduction to her doctoral dissertation [27].

Thus, it was the first paper of Maria Skłodowska-Curie published in May 1898, which again concentrated the interest of researchers on Becquerel rays. In July of that year Maria and Pierre Curie announced the discovery of a new radioactive element [28]: "Certain minerals containing uranium and thorium (pitchblende, chalcocite, uranite) are very active from the point of view of emission of Becquerel rays. In earlier work, one of us has shown that their activity is even greater than that of uranium and thorium, and has made the statement that this effect must be due to some other very active substance contained in a very small quantity in these minerals ...

We believe therefore that the substance, which we have recovered from pitchblende contains a metal not yet described, related to bismuth in its analytical properties. If the existence of this new metal is confirmed, we propose to call it polonium, after the native country of one of us."

8. William Crookes remains sceptical

William Crookes, then President of the British Association for the Advancement of Science which had a meeting in Bristol in September 1898, presented an address in which he only briefly mentioned radioactivity. It was clear from his words that he treated radioactivity to be a marginal subject compared with X-rays [29]:

Summary on the new rays in the spring of 1898

Property	Röntgen rays	Uranium rays	Thorium rays
Penetration through paper and aluminium	Yes	Yes	Yes
Penetration through heavier metals	No	No	No
Action on photographic plates	Yes	Yes	Yes
Ionization of air	Yes	Yes	Yes
Reflection	No	Yes*	Yes(?)**
Refraction	No	Yes*	Yes**
Polarisation	No	Yes*	No**
Nature	?	Very short ether waves	?

*Erroneous Becquerel's results of March, 1896

**Schmidt's results of February, 1898

"No other source for Röntgen rays but the Crookes tube has yet been discovered, but rays of kindred sorts are recognised. The Becquerel rays, emitted by uranium and its compounds, have now found their companions in rays — discovered almost simultaneously by Curie and Schmidt — emitted by thorium and its compounds. The thorium rays affect photographic plates through screens of paper or aluminium, and are absorbed by metals and other dense bodies. They ionise the air, making it an electrical conductor; and they can be refracted and probably reflected, at least diffusively. Unlike uranium rays, they are not polarised by transmission through tourmaline, therefore resembling in this respect the Röntgen rays.

Quite recently M. and Mdme. Curie have announced a discovery which, if confirmed, cannot fail to assist the investigation of this obscure branch of physics. They have brought to notice a new constituent of uranium mineral pitchblende, which in a 400-fold degree possesses uranium's mysterious power of emitting a form of energy capable of impressing a photographic plate and of discharging electricity by rendering air a conductor. It also appears that the radiant activity of the new body, to which the discoverers

have given the name of Polonium, needs neither the excitation of light nor the stimulus of electricity; like uranium, it draws its energy from some constantly regenerating and hitherto unsuspected store, exhaustless in amount . . .”

Crookes called polonium a “body” and not an element, although in another part of his address he discussed in detail the discoveries of krypton, neon and metaargon (now called xenon), and also “monium” which he himself discovered and which later proved to be just a mixture of two already known elements. But he gave some attention to the problem of energy source for uranium and thorium rays. He proposed a hypothesis that these heavy elements might absorb energy from the fastest air molecules.

“The reduction of the speed of the quick moving molecules would cool the layer of air to which they belong; but this cooling would rapidly be compensated by radiation and conduction from the surrounding atmosphere; under ordinary circumstances the difference of temperature would scarcely be perceptible, and the uranium would thus appear to perpetually emit rays of energy with no apparent means of restoration.

The total energy of both the translational and internal motions of the molecules locked up in quiescent air at ordinary pressure and temperature is about 140000 foot-pounds in each cubic yard of air. Accordingly the quiet air within a room 12 feet high, 18 feet wide, and 22 feet long contains energy enough to propel a one-horse engine by more than twelve hours. The store drawn upon naturally by uranium and other heavy atoms only awaits the touch of the magic wand of science to enable the twentieth century to cast into the shade the marvels of the nineteenth.”

In the end of 1898 German physicists Julius Elster and Hans Geitel proved experimentally that uranium radiation was the same at normal atmospheric pressure and in a vacuum, and also in a mine at the depth of 853 meters. These results contradicted the hypothesis of Crookes.

9. Ernest Rutherford

The “Philosophical Magazine” for January 1899 carried a paper by Ernest Rutherford “Uranium Radiation and the Electrical Conduction Produced by It” [30]. This thick paper was sent from Cambridge to the editors on September 1, 1898, thus it may be certain that its author began studying radioactivity much before that date, probably at the same time as Maria Skłodowska-Curie.

In the beginning of his paper Rutherford stated that: “The results of Becquerel showed that Röntgen and uranium radiations were very similar in their power of penetrating solid bodies and producing conduction in a gas exposed to them; but there was an essential difference between the two types of radiation. He found that uranium radiation could be refracted and

polarised, while no definite results showing polarisation or refraction have been obtained for Röntgen radiation.”

Rutherford then related his unsuccessful attempts to confirm experimentally that uranium radiation could be refracted and polarised. Since Becquerel was already well known authority, Rutherford, a beginner, concluded modestly:

“All the results that have been obtained point to the conclusion that uranium gives out types of radiation which, as regards their effect on gases, are similar to Röntgen rays and the secondary radiation emitted by metals when Röntgen rays fall upon them. If there is no polarisation or refraction the similarity is complete.”

In his paper Rutherford reported an important finding that uranium radiation contained two components differing in the penetrating power: strongly absorbed alpha radiation and penetrating beta radiation. It convinced Rutherford that uranium radiation is more complicated than it appeared from the study by Becquerel. He thus expressed reservation to whether it was indeed necessary to postulate the existence of new substances:

“It is possible that the apparently very powerful radiation obtained from pitchblende by Curie may be partly due to the very fine state of division of the substance rather than to the presence of a new and powerful radiating substance.”

10. Discovery of radium

Meanwhile Maria and Pierre Curie and Gustave Bémont continued their efforts to extract yet another substance from the pitchblende. The discovery of radium was announced on December 26, 1898 [31] (the report in “Nature” appeared already on January 5, 1899):

“The different reasons which we have enumerated lead us to believe that the new radio-active substance contains a new element to which we propose to give the name of radium... The new radio-active substance certainly contains a very great proportion of barium; in spite of that, the radioactivity is considerable. The radio-activity of radium must therefore be enormous ...”

The discoveries of polonium and radium dispersed earlier doubts concerning the existence of new elements. Now many physicists decided that radioactivity is an exciting field of studies. Becquerel also returned to his research of uranium and on March 27, 1899, presented a new paper to the Academy of Sciences. He stated that the intensity of the uranium radiation, as measured by their photographic action, appeared to be unchanged since May 1896; he also announced that the rays do not appear to be capable of refraction and polarisation, all attempts to repeat two early experiments

Summary on the new rays in the spring of 1899

Property	Röntgen rays	Uranium, thorium polonium, radium rays
Penetration through paper and aluminium	Yes	Yes
Penetration through heavier metals	No	No
Action on photographic plates	Yes	Yes
Ionization of air	Yes	Yes
Reflection	No	No
Refraction	No	No
Polarisation	No	No
Nature	?	?

giving positive results having failed. Thus Becquerel withdrew the results which previously caused that the field was found little interesting.

The following years were full of new discoveries. André Debierne [32] discovered actinium (results presented to the Academy of Sciences on October 16, 1899). Ernest Rutherford made an important impact on the study of radioactivity by the discovery of thorium emanation (1900) and the first theory of radioactive transmutations developed jointly with Frederick Soddy. In 1903 Becquerel and the Curies received the Nobel prize in physics.

The discoveries of polonium and radium have been common achievements of Pierre and Maria Curie. As Eve Curie wrote in the well known biography of her mother [33]:

“We cannot and must not attempt to find out what should be credited to Marie and what to Pierre during these years. It would be exactly what the husband and wife did not want. The personal genius of Pierre Curie is known to us by the original work he had accomplished before this collaborations. His wife’s genius appears to us in the first intuition of discovery, the brilliant start; and it was to reappear to us again, solitary, when Marie Curie the widow unflinchingly carried the weight of a new science and conducted it, through research, step by step, to its harmonious expansion. We therefore

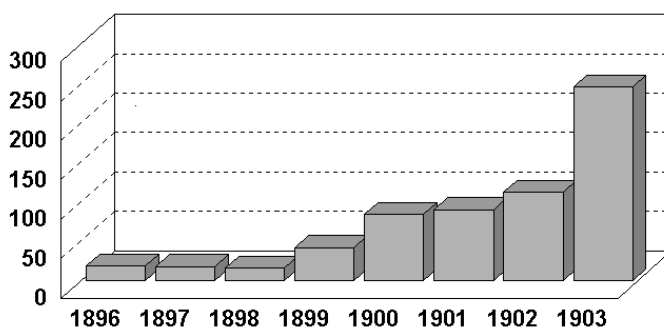
have formal proof that in the fusion of their two efforts, in this superior alliance of man and woman, the exchange was equal.

Let this certainty suffice for our curiosity and admiration. Let us not attempt to separate these creatures full of love, whose handwriting alternates and combines in the working notebooks covered with formulae, these creatures who were to sign nearly all their specific publications together. They were to write “We found” and “We observed”; and when they were constrained by fact to distinguish between their parts, they were to employ this moving locution: Certain minerals containing uranium and thorium (pitchblende, chalcocite, uranite) are very active from the point of view of the emission of Becquerel rays. In a preceding communication, one of us showed that their activity was even greater than that of uranium and thorium, and stated the opinion that this effect was due to some other very active substance contained in small quantity in these minerals.”

It is difficult not to agree with an American historian Lawrence Badash who had this to say on the first years of radioactivity [34]:

“In early 1898, radioactivity was something of a “dead horse” — it was there, but no one knew what to do with it. It took not only the discovery of thorium’s activity, first by Gerhard C. Schmidt and then by Marie Curie, but the subsequent discoveries of polonium and radium by the Curies to produce a sustained renewal of interest. For then it became apparent that this was an atomic phenomenon of great significance.”

Papers on radioactivity



Based on Max Iklé, *Jahrbuch der Radioaktivität und Elektronik*, 1, 413-442 (1904)

REFERENCES

- [1] *Compt. Rend.* **122**, 150 (1896).
- [2] H. Poincaré, Les rayons cathodiques et les rayons Roentgen, *Revue Générale des Sciences* **7**, 52 (1896).
- [3] H. Becquerel, *On Radioactivity, a new Property of Matter, Nobel Lecture, December 11, 1903*, in: *Nobel Lectures. Physics 1901–1921*, Elsevier, New York 1967, p. 52.
- [4] H. Becquerel, Sur les Radiations Émises par Phosphorescence, *Compt. Rend.* **122**, 420 (1896).
- [5] H. Becquerel, *Sur les Radiations Invisibles Émises par les Corps Phosphorescents*, *Compt. Rend.* **122**, 501 (1896). English translation from W.F. Magie, *A Source Book in Physics*, McGraw-Hill, New York 1935.
- [6] *Nature* **53**, 445 (1896).
- [7] E. Becquerel, *La Lumière, ses Causes et ses Effets*. Tome second: *Effets de la Lumière*, Paris 1868, p. 50.
- [8] Ch.F. Guillaoume, Les Rayons de Becquerel et les Corps Nouveaux, *Revue Générale des Sciences* **10**, 890 (1899).
- [9] H. Becquerel, *Recherches sur une Propriété Nouvelle de la Matière*, Paris 1903, pp. 51–52.
- [10] See e.g. Ch. Sutton, *Serendipity or Sound Science?*, *New Scientist*, 27 February 1986, p.30; A. Kohn, *Fortune or Failure*, Basil Blackwell, Oxford 1989, p. 22–24.
- [11] H. Becquerel, Sur Quelques Propriétés Nouvellés des Radiations Invisibles Émises par Divers Corps Phosphorescents, *Compt. Rend.* **122**, 559 (1896).
- [12] H. Becquerel, *Sur les Radiations Invisibles Émises par les Sels d'Uranium*, *Compt. Rend.* **122**, 689 (1896).
- [13] H. Becquerel, Sur les Propriétés Différentes des Radiations Invisibles émises par les Sels d'Uranium, et du Rayonnement de la Paroi Anticathodique d'un Tube de Crookes, *Compt. Rend.* **122**, 762 (1896).
- [14] G. Le Bon, *La Lumière Noire*, *Compt. Rend.* **122**, 188 (1896).
- [15] G. Le Bon, La Photographie a la Lumière Noire, *Compt. Rend.* **122**, 233 (1896).
- [16] H. Becquerel, Émission de Radiations Nouvelles par l'Uranium Métallique, *Compt. Rend.* **122**, 1086 (1896).
- [17] Numerous reports on new kinds of radiation may be found in various periodicals from that period (see e.g. *Nature* **53**, 335, 398, 502 (1896), **54**, 92 (1896)); The reports on French “discoveries” were published in the official *Academy of Sciences Journal Compt. Rend.* **122**, **123**, **124**. Le Bon's late paper on “lumière noire” was published in *Revue Scientifique*, **13**, 449 (1900).
- [18] J.J. Thomson, The Röntgen Rays, *Nature* **54**, 302 (1896).
- [19] H. Becquerel, Sur Diverses Propriétés des Rayons Uraniques, *Compt. Rend.* **123**, 855 (1896).

- [20] H. Becquerel, Recherches sur les Rayons Uraniques, *Compt. Rend.* **124**, 435 (1897).
- [21] H. Becquerel, Sur la Loi de la Decharge dans l'Air de l'Uranium Electrisé, *Compt. Rend.* **124**, 800 (1897).
- [22] O. Glasser, *Wilhelm Conrad Roentgen and the Early History of the Roentgen Rays*, Springfield, 1934.
- [23] W. Biernacki, Nowe Dziedziny Widma, (New Spectral Regions), Warszawa 1898, pp. 79–80.
- [24] O.M. Stewart, A Résumé of the Experiments Dealing with the Properties of Becquerel Rays, *Physical Review*, No. 4, p. 239–251, April 1898.
- [25] M. Skłodowska-Curie, Rayons Émis par le Composés de l'Uranium et du Thorium, *Compt. Rend.* **126**, 110, (1898). English translation from H.M. Davis, *The Chemical Elements, Science Service*, Washington 1952.
- [26] G.C. Schmidt, Über die vom Thorium und den Thorverbindungen ausgehende Strahlung, *Verh. Phys. Ges. Berlin* **17**, 13 (1898); *Ann. der Phys.* **65**, 141 (1898). The results were communicated to the Deutsche Physikalische Gesellschaft in Berlin on February 4, 1898, and his paper sent for publication on March 24; Maria Skłodowska Curie's paper was read at the session of the Academy of Sciences in Paris on April 12.
- [27] M. Skłodowska-Curie, *Recherches sur les Substances Radioactives*, Paris 1903.
- [28] P. Curie, Mme P. Curie, Sur une Substance Nouvelle Radio-Active, Contenne dans la Pechblende, *Compt. Rend.* **127**, 175 (1898). English translation from H.M. Davis, *The Chemical Elements, Science Service*, Washington 1952.
- [29] W. Crookes, Inaugural Address, *Nature* **58**, 438 (1898).
- [30] E. Rutherford, Uranium Radiation and the Electrical Conduction Produced by It, *Philosophical Magazine* ser. 5, 47, 109–163, January 1899.
- [31] P. Curie, Mme P. Curie, G. Bémont, Sur une Nouvelle Substance Fortement Radio-Active, Contenne dans la Pechblende, *Compt. Rend.* **127**, 1215 (1898); English translation from H.M. Davis, *The Chemical Elements, Science Service*, Washington 1952.
- [32] A. Debierne, Sur une Nouvelle Matière Radio-Active, *Compt. Rend.* **129**, 593 (1899); Sur un Nouvel Élément Radio-Actif: l'Actinum, *Compt. Rend.* **130**, 906 (1900).
- [33] Madame Curie, A Biography by Eve Curie, translated by Vincent Sheean, Pocket Books, New York 1946.
- [34] L. Badash, Radioactivity Before the Curies, *Am. J. Phys.* **33**, 128 (1965).