BECQUEREL’S EXPERIMENTAL MISTAKES

Roberto de Andrade Martins

Abstract: In 1896, Henri Becquerel detected a penetrating radiation emitted by some uranium salts and met a phenomenon that nowadays we call “radioactivity”. Becquerel’s study of uranium radiation was not casual or blind. It was guided by his acceptance of Poincaré’s conjecture concerning a possible relation between X rays and luminescence. What Becquerel expected to find was the emission of a penetrating electromagnetic radiation (something similar to ultraviolet rays) emitted by a special phenomenon of fluorescence or phosphorescence that violated Stokes’ law. Guided by his preconceptions, Becquerel described experiments that seemed to support the view that uranium radiation had the usual properties of known electromagnetic waves: reflection, refraction and polarization. He also described an increase in the emission of radiation when uranium compounds were stimulated by sunlight. Those and several other aspects of Becquerel’s experimental work must nowadays be interpreted as experimental mistakes. Becquerel’s mistakes were gradually corrected by other researchers. As the study of radioactivity developed, Becquerel reinterpreted his own early work, hiding his mistakes or ascribing to himself their correction. The aim of this article is to discuss one particular episode of experimentation – Becquerel’s study of the phenomenon we call radioactivity – and the methodological problems aroused by his mistakes.

Keywords: radioactivity; experimental errors; history of physics; Becquerel, Henri

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1. INTRODUCTION

The study of experimental research has recently deserved more attention from historians of science (Holmes, 1992). The aim of this article is to discuss one particular episode of experimentation – Becquerel's study of the phenomenon we call radioactivity – and the methodological problems aroused by his mistakes.

In 1896, Henri Becquerel detected a penetrating radiation emitted by some uranium salts and came accross a phenomenon that nowadays we call "radioactivity". Becquerel's study of uranium radiation was not casual or blind. It was guided by his acceptance of Poincaré's conjecture[1], together with his previous knowledge and expectations concerning the properties of uranium compounds (Martins, 1997). What Becquerel expected to find was the emission of penetrating electromagnetic radiation (something similar to ultraviolet rays) emitted by a special kind of fluorescence or phosphorescence that violated Stokes' law. That was, indeed, what he thought he had found. Guided by his preconceptions, Becquerel ascribed to uranium radiation the usual properties of known electromagnetic waves: reflection, refraction and polarization. As he thought the phenomenon to be a kind of phosphorescence, he also expected to observe an increase in the emission of radiation when uranium compounds were stimulated by sunlight – and he verified this increase.

Those and several other aspects of Becquerel's experimental work must nowadays be interpreted as full of experimental mistakes. There is nothing new in the observation that scientists sometimes are misled by their theoretical expectations – but it is remarkeable how far Becquerel was led by his preconceptions. He was even led to support the claims for existence of N-rays, because they seemed able to explain some of his own experimental anomalies.

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[1] See the first paper in this volume: MARTINS, Roberto de Andrade. A pool of radiations: Becquerel and Poincaré's conjecture.

Becquerel's mistakes were gradually corrected by other researchers. As the study of radioactivity developed, Becquerel reinterpreted his own early work, hiding his mistakes or ascribing to himself their correction. He was successful, and in a few years his errors were forgotten – and he was accorded the Nobel Prize.

Of course, Becquerel could not succeed in his personal endeavour without the support of colleagues and the French Academy of Sciences. This paper will not try to disclose the sociological aspects of the episode. It will only analyse the evidence relating to Becquerel's mistakes and his strategy of occultation of his own failure.

2. HENRI BECQUEREL'S EARLY WORK

Becquerel began his search for penetrating radiations emitted by luminescent bodies in January 1896. On the 24th February, he presented to the French Academy of Sciences his first positive results: he succeeded to detect a penetrating radiation (similar to X-rays) emitted by crystals of double sulphate of uranyl and potassium (Becquerel, 1896a). In this and in the next communication (Becquerel, 1896b), Henri Becquerel did not discuss the nature of the penetrating radiation. He only described that it was able to pass through black paper and thin glass plates, and to affect photographic plates. At this time, he believed that those radiations "[...] could be invisible radiations emitted by phosphorescence with a persistence infinitely larger than the persistence of luminous radiations emitted by those bodies." (Becquerel, 1896b)

Nowadays, a modern physicist will find nothing strange or unexpected in those two earlier communications. From Becquerel's third "radioactivity" paper onwards, however, he reported several phenomena that seem to us completely anomalous, in the following sense: they conflict with our physical knowledge and it is doubtful that anyone nowadays could reproduce Becquerel's observations. Becquerel and coetaneous scientists, however, found nothing strange in those phenomena. Indeed, they completely accorded with Becquerel's expectations.

In his third "radioactivity" paper (9th March 1896), Henri Becquerel began the study of the properties of the radiation emitted by the uranium phosphorescent compound he was using (Becquerel, 1896c)[2]. He was guided by his expectation that the radiation was an invisible light, and by Röntgen's investigation of X-rays. One of the known properties of X-rays and ultraviolet light was their ability of discharging an electroscope. Becquerel observed that the uranium rays were also able to discherge an electroscope. Röntgen had tried to observe reflection and refraction of X-rays, with negative results. Becquerel tried similar experiments with uranium radiation – and he apparently succeeded.

3. REFLECTION OF URANIUM RADIATION

One of Becquerel's experiments seemed to show clear evidence of regular reflection of the radiation emitted by the uranium salt. He cut a concave mirror in a small tin block. This mirror was polished and produced visual images. There was, however, a small defect of the metal, and at this point the mirror could not be polished. A thin crystal of uranium and potassium sulphate was attached to the focal plate of the mirror. Below this device, Becquerel placed a photographic plate, separed from the crystal by paper.

Becquerel reported that when the photographic plate was developed, he observed a triangular black figure, corresponding to the crystal flake. It was surrounded by a dark circle, and in this circle there was a spot corresponding to the defect of the mirror. A hypothetical reconstruction of this experiment is shown in Fig. 1.

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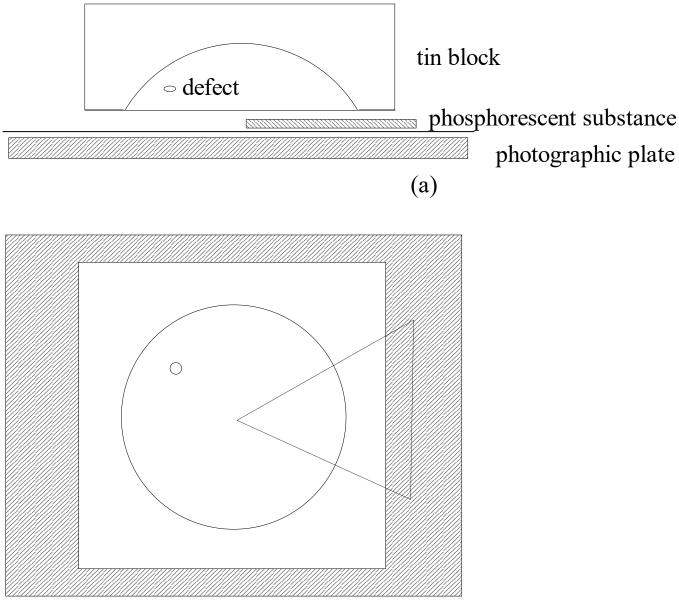
[2] At this time, Becquerel did not use the phrase "uranium rays" and there was no evidence that the radiation he was studying was peculiar to uranium compounds.



(a)

(b)

Fig. 1. A hypothetical reconstruction of Becquerel's experiment on the reflection of uranium rays.

Becquerel concluded:

> This halo, with a quite sharp border, is therefore due to radiations that, after being reflected on the mirror, were sent to the plate in nearly parallel directions. (Becquerel, 1896c, p. 561)

This seemed a conclusive evidence of regular reflection (not diffusion or scattering) of radiation. According to our present-day knowledge, uranium radiation does not suffer reflection in polished metal. Besides that, Becquerel's argument is wrong. Only if the source of radiation were very small and if it were placed at the focus of an spherical mirror, the radiation could be reflected as parallel rays. However, in the case of an extended, large source (as was the case in Becquerel's experiment), even if there were specular reflection, rays emitted from different points of the substance would have different directions after reflection and there could never arise any spot corresponding to the defect of the mirror. It is impossible to understand what happened in this experiment.

4. REFRACTION OF URANIUM RADIATION

In the same paper presented on 9th March 1896, Henri Becquerel described evidence for the existence of refraction of the penetrating radiation emitted by phosphorescent compounds. Becquerel first tried to detect refraction of uranium radiation using a prism, and stated that those experiments "gave signs of refraction, but the signs were too weak to be presented today. Moreover, it will be seen from results that will be described below, that some images clearly reveal the fact of refraction and total reflection in glass" (Becquerel, 1896c, p. 561).

The positive evidence referred to by Becquerel was obtained in the study of uranium nitrate. This substance strongly absorbs moisture from the air and its crystals must therefore be protected from the atmosphere. Henri Becquerel put the uranium nitrate in a glass tube, closed with a thin glass plate (0.2 mm thick) sealed with paraffin (Fig. 2). The crystal sample was several milimeters high.

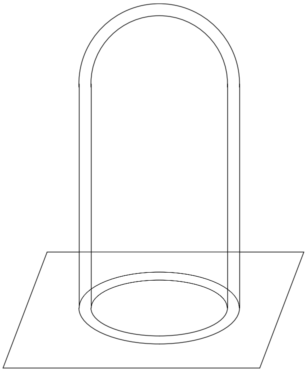


Fig. 2. A hypothetical reconstruction of the glass tube used by Becquerel in his experiment with uranium nitrate.

This device was put (the glass plate downwards) over a photographic plate wraped in black paper. After two days, the photographic plate was developed and showed a black spot corresponding to the base of the uranium nitrate crystal. This spot was surrounded by a "slightly dark" band, limited by the border of the glass tube. Becquerel concluded:

> This band is due to the action of the radiations obliquely emitted by the vertical faces of the [cylinder of powdered] crystal which is several milimetres thick; the radiations stoped by this tube were refracted and totally reflected inside it, as light rays inside a liquid vein. The action is stronger at the places that are in contact with the uranium nitrate crystal. (Becquerel, 1896c, p. 563)

Becquerel *expected* uranium radiation to be refracted and reflected, because he thought it was some kind of penetrating ultraviolet light. What he observed confirmed his expectations. However, we know that uranium radiation is not refracted or reflected by glass. This anomalous effect cannot be explained according to our present knowledge.

Besides that, in a later paper, Becquerel described that he obtained deflection of the radiation of uranium nitrate using a crown glass prism (Becquerel, 1896d, p. 693). Becquerel never published the photographic evidence of those experiments.

5. EMISSION OF PENETRATING RADIATION BY CALCIUM SULPHIDE

In his first two "radioactivity" papers, Henri Becquerel had studied the radiation emitted by double sulphate of uranyl and potassium. In his third (9th March 1896) paper of this series (Becquerel, 1896c), he described for the first time some attempts to detect penetrating radiations emitted by other substances. In a first series of experiments, he tried double sulphates of uranyl and sodium, of uranyl and potassium, of uranyl and ammonium, uranium nitrate, and zinc sulphide.

Those substances were put over thin glass plates over the same photographic plate, without exposition to any strong light. All uranium salts produced similar photographic effects through black paper. The zinc sulphide sample produced no effect. Before Becquerel's experiments, both Charles Henry (1896) and Louis Joseph Troost (1896) had observed strong effects with zinc sulphide, but in their experiments there was stimulation by sunlight or magnesium light.

In another series of experiments, Becquerel tried another set of substances: orange calcium sulphide,[3] green strontium sulphide, blende (zinc sulphide), blue calcium sulphide and greenish blue calcium sulphide. Some of those substances were altered by the air, and therefore he was led to enclose them in glass containers similar to those used for uranium nitrate. Between those tubes and the photographic plate (wrapped in black paper) there was an aluminium plate, 2 mm thick. This set of samples was exposed to ambient light (not directly to sunlight) and left over the photographic plate from 4 p. m. on the 7th March to 9:30 a. m. on the 9th March 1896 – that is, 41½ hours[4]. Only two of those substances produced observable effects on the photographic plate: the blue and greenish blue calcium sulphides. The effects produced were very strong – even stronger than those obtained with uranium compounds:

> [...] the two blue and greenish blue luminous calcium sulphides gave very energetic actions, the most intense that I have yet obtained in those experiments. The fact relative to the blue calcium sulphide accords with the observation of Mr.

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[3] The colours described here refer to the light emitted by those substances in darkness, after excitation by light. The colour of light emitted by phosphorescent bodies usually depend on the presence of impurities.

[4] In different publications, Becquerel provides different numbers, varying from 43 to 48 hours. The exact times of the experiment are only found in the caption accompanying the image published in 1903.

> Niewenglowski [that the radiation passes] through black paper. (Becquerel, 1896c, p. 563)

Besides confirming Gaston Niewenglowski's earlier observation (Niewenglowski, 1896)[5], this experiment was relevant for another reason: it provided evidence for reflection and refraction of the penetrating radiation:

> The images I have obtained with the two calcium sulphides through aluminium are worth pointing out as offering very important peculiarities. The quantity of phosphorescent powder contained in the tubes formed a column several milimetres high over the plane glass slide basis, and about one centimetre for the blue sulphide. The radiation of the lateral surface produced large black spots, exceedingly strong, in the middle of which it was possible to distinguish a clearer image of the section of the glass tube, and especially the very neat edge of the glass plate. Those edges, black inside and surrounded by an absolutely white line, show that the oblique radiations have penetrated the glass plate and have been refracted and completely reflected there at the separation surface between glass and air. Both calcium sulphide tubes presented the same appearance, in different degrees, and the radiations have even attained the nearby tube containing strontium sulphide and produced the appearance, with the same character, of part of this tube and of the slide that supported it. If the phenomena of refraction and reflection had not been evidenced before by other experiments, it would be made manifest by this single test. (Becquerel, 1896c, pp. 564-565)

Hence, this was the strongest evidence provided by Becquerel for reflection and refraction of the invisible radiation. Notice that he made no distinction here between uranium radiation and the radiation presumably emitted by calcium

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[5] See MARTINS, Roberto de Andrade. A pool of radiations: Becquerel and Poincaré's conjecture, published in this volume.

sulphide. This is a strong evidence that, at this time, he still accepted Poincaré's conjecture as true and thought his experiments were equivalent to those of Gaston Niewenglowski, Charles Henry and others.

Most of Becquerel's work used the photographic method of detection of radiation. It is likely that Becquerel presented at the meetings of the Academy of Sciences the negatives he obtained, but they were not published in the *Comptes Rendus*. Several years later, after the appearance of the works of Marie Curie, Ernst Rutherford and others, when Becquerel's preliminary work grew in importance, he published his early photographic evidence concerning calcium sulphide (Becquerel, 1900, p. 49, fig. 1; Becquerel, 1902, plate 2, fig. 4; Becquerel, 1903a, plate II, fig. 5). There is, however, an older copy of Becquerel's photograph, sent by himself to Lord Kelvin in 1897, that deserve notice.

In the beginning of 1897, Lord Kelvin began a series of researches on uranium radiation. He was specially surprised with the electrical properties of the uranium radiation, as he told Stokes:

> Two days ago I received from Moissan a specimen of Uranium, and have seen with my own eyes its effectiveness in discharging an electrified conductor which is more like magic than anything I have ever seen or heard of in Science.[6]

Kelvin sent a copy of his first paper (Kelvin, Beattie & de Smolan, 1897) to Henri Becquerel, who replied to him on the 3rd August 1897. Becquerel enclosed with his letter two copies of the photographs of his experiment[7]:

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[6] Letter from Lord Kelvin to Stokes, 25th February 1897. Original. manuscript kept at the Cambridge University Library, CUL Add 7656.K328. Other manuscripts consulted at this library will hitherto be identified as CUL.

[7] Letter from Henri Becquerel to Lord Kelvin, 3rd August 1897, CUL Add 7342.B52.

> I send you two facsimile prints of the negatives I have obtained last year, at the very beginning of my researches.
>
> One is the print of an aluminium medal traversed by the rays emitted by double sulphate of uranium and potassium.
>
> The other was obtained with a powdered phosphorescent calcium sulphide that later showed itself inert. The phosphorescent powder was enclosed in a small glass tube that rested upon a glass slide sealed with paraffin, and separed from the photographic plate by an aluminium sheet 2 milimetres thick. The print shows the refraction and the total inflexion [of the radiation] at the edges of the glass slide and the paraffin.

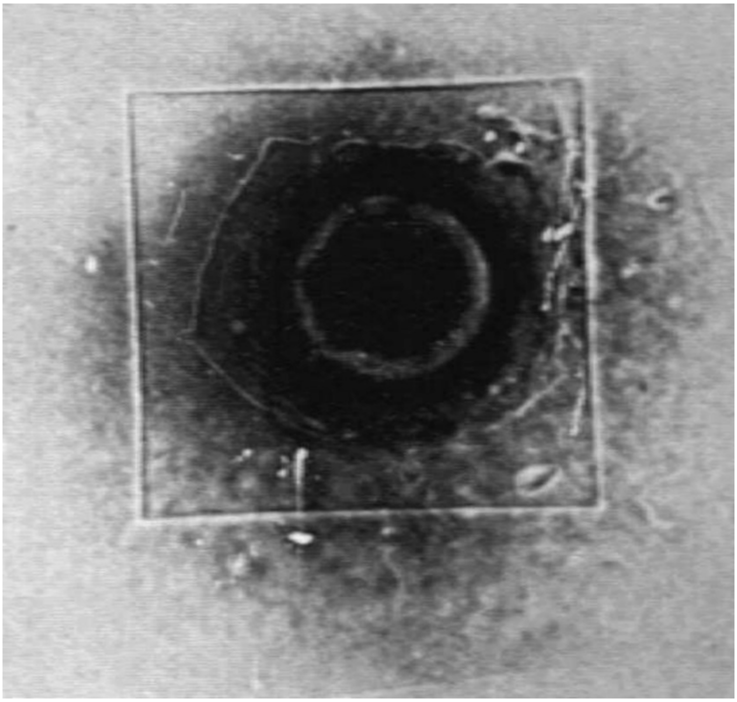


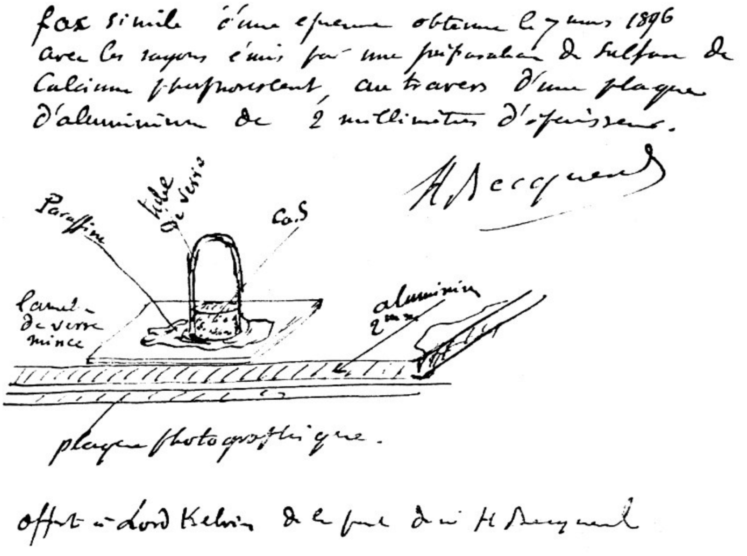
Fig. 3. Detail of the second photograph sent by Becquerel to Lord Kelvin in 1897 (CUL Add 7342.B52).

Part of the second photograph is reproduced in Fig. 3. Notice that at this time Becquerel still accepted as valid and relevant his observations of the calcium sulphide samples and the evidence for the refraction of radiation. For this reason, the second photograph is the most relevant for the present discussion.

At the back of this photograph, Becquerel sketched the experimental setup and wrote:

> Facsimile of a print obtained the 7th March 1897 with the rays emitted by a preparation of phosphorescent calcium sulphide, through an aluminium sheet 2 milimetres thick.
>
> [signed] H Becquerel

Then follows the sketch of the experiment, and, at the bottom of the print: “Offered to Lord Kelvin by Mr. H. Becquerel”.



Fac simile d'une épreuve obtenue le 7 mars 1896 avec les rayons émis par une préparation de sulfure de Calcium phosphorescent, au travers d'une plaque d'aluminium de 2 millimètres d'épaisseur.

H Becquerel

offert à Lord Kelvin de la part de M H Becquerel

Fig. 4. Becquerel’s sketch of the experiment with calcium sulphide (CUL Add 7342.B52).

Both photographs sent by Becquerel to Kelvin measured 9x12 cm. This is the exact size of Becquerel’s original Lumière plates (Becquerel, 1903a, p. 42). Of course, they have been reproduced by a contact method. The experimental setup is also shown in a photograph published by Becquerel (Becquerel, 1902, plate 2, fig. 3; Becquerel, 1903a, plate II, fig. 7). The photograph shows indeed the details described by Becquerel and, at this time, was compatible with his interpretation. Becquerel seemed particularly proud of this evidence, and he provided copies of the refraction photograph to other researchers (Bouty, 1896, p. 615).

In later works, after the dismissal of refraction of uranium radiation by other researchers, Becquerel ascribed his mistake to this single calcium sulphide experiment. In his 1902 account, Becquerel describes this experiment and comments: "Those facts and some other had led me to think that the new radiation could be a transversal motion of the ether analogous to light; the absence of refraction and a large number of different experiments made me give up this hypothesis". In his 1903 book, Becquerel also attributes his mistakes to this anomalous experiment: "Unfortunately, the assimilation [of the calcium sulphide effects] with the effects produced by uranium rays and the appearance of reflection and refraction effects have led me at that time to attribute to the uranium rays properties analogous to those of light – properties that they do not have" (Becquerel, 1903b, p. 51).

However, as shown above, Becquerel had described a similar effect he observed in an experiment using uranium nitrate. The calcium sulphide experiment was not the only evidence he presented for refraction of the penetrating radiations.

The emission of penetrating radiation by calcium sulphide observed by Becquerel cannot be explained by our physical knowledge. In a later communication, Becquerel reported that the samples of calcium sulphide that had formerly given strong effects in previous experiments were now inactive. He tried to stimulate those samples by light, heat and cold, with no results (Becquerel, 1896d). A similar phenomenon had occurred with the samples of zinc sulphide used by Troost: recently prepared blende produced penetrating radiation when excited by magnesium light, but the activity gradually decreased and after some time the phosphorescent substance was unable to react to illumination (Troost, 1896).

After his samples of calcium sulphide died out, we might expect that Becquerel would perceived that the phenomenon he was studying was peculiar to uranium compounds. However, even several months after this time, he still mentioned his calcium sulphide observations as related to his uranium experiments.

6. PERSISTENCE AND STIMULATION OF EMISSION OF THE INVISIBLE RADIATIONS

There was a conflict between Becquerel's expectations and his observations concerning the persistence of the invisible radiations emitted by uranium salts. He observed that the substances he was using emitted the penetrating rays for a long time, when kept in the dark. Nowadays, we believe that this is one of the main characteristics of radioactivity: it is a spontaneous emission of radiation, that cannot be increased or decreased by common physical stimuli (light, heat, etc.). In the case of uranium, the emission decreases very slowly in time – a decrease that cannot be detected in a few years.

In his third "radioactivity" paper (Becquerel, 1896c), Henri Becquerel described the long persistence of the invisible radiations emitted by the phosphorescent crystals of uranium compounds, that he had kept in darkness for 160 hours. During this time, there was no perceivable decrease of the penetrating radiation. However, this observation did not led him to the conclusion that this was a new phenomenon:

> Perhaps this fact should be compared to the indefinite conservation of absorbed energy in some bodies, that emit it when one heats them, a fact to which I have already called the attention in a work on the phosphorecence by heat. (Becquerel, 1896c, pp. 562-563)

Henri Becquerel was still being guided by his knowledge of luminescence phenomena. The phenomenon he recalled here had been well studied by his father. When a phosphorescent substance is exposed to light and brought to a dark room, it will shine during some time, but its luminosity will decrease and after a longer or shorter time it will seem to have lost all its phosphorescence. However, there are several phosphorescent substances that can shine again after becoming dark, if they are heated, a phenomenon that had been studied by his father Edmond Becquerel:

> Once the phosphor is exposed to light and placed in the darkness, the change acquired under the influence of radiation will remain for some time, even when the sulphide does not sensibly shine any more at room temperature, and an increase of temperature can afterwards give rise to a light emission. (Edmond Becquerel, 1848)

Five years before his "radioactivity" researches, Henri Becquerel had also studied those phenomena:

> From the moment when they are submitted to the exciting action of light onwards, phosphorescent bodies kept at a constant temperature emit light that ceases being perceptible after a shorter or longer time, varying from a fraction of a second to several days, and then the body extinguishes itself. If we then rise the temperature and keept it constant again, the body becomes luminous, then extinguishes itself again; [...] so, for a given temperature, there is, on one side, a faster or slower lost of energy by light radiation, and, on the other side, an amount of energy that remains in the body in a latent state, to be emitted at a higher temperature. This latent portion of light stored in the body seems to stay therein in a permanent way, if that body is kept at a temperature equal or smaller to the regarded temperature. (Becquerel, 1891, pp. 561-562)

In the same paper, Becquerel remarked:

> There is a fact worth calling our attention, the indefinite conservation in bodies of an amount of energy that they absorb and that they emit when they are heated. By what mechanism is this energy kept without any sensible lost? Is there a particular state of matter comparable to that of magnetized bodies? Is the lost of energy continually compensated? Those are questions that we cannot answer now and that may perhaps be elucidated by future studies. (Becquerel, 1891, p. 563)

There is one sentence in the above citation that deserves carefull analysis: "Is the lost of energy continually compensated?". According to Lommel's theory, the vibrations of the particles of solid luminescent bodies are submitted to resistive forces (see Martins, 1997). There should therefore be a continuous energy lost, and if those bodies keep for a very long time (or indefinitely) their ability of emitting light when heated, this loss had to be compensated. It is very difficult to imagine any process of this kind, but the above citation shows that in 1891 Henri Becquerel considered the possibility of luminescence phenomena with continuous loss of energy and continuous compensation of this loss.

7. OTHER ANOMALOUS PROPERTIES OF BECQUEREL'S RAYS

Further experiments made by Becquerel provided new anomalous phenomena: (a) the intensity of the radiation emitted by uranium salts increased when they were stimulated by light; (b) the radiation exhibit polarization effects. Of course, according to present physical knowledge those effects could not exist, but Becquerel reported them an they strengthened the belief that the phenomenon was a kind of insivisible phosphorescence and that the emitted radiation was invisible electromagnetic radiation (similar to ultraviolet rays).

7.1 *Excitation of emission of radiation by light*

When Henri Becquerel reported the emission of radiation by his phosphorescent samples kept in darkness, he concluded that it could be due to some kind of invisible, long lived phosphorescence. In his third "radioactivity" paper, he reported that the effect was still observed when the samples were kept in darkness for 7 days. In his communication presented on the 23rd March 1896, Becquerel presented new evidence:

> If the phenomenon of emission of invisible radiations that we study is a phosphorescence phenomenon, it should be possible to exhibit its excitation by given radiations. That research becomes very difficult because of the prodigious persistence of the emission when those bodies are kept in darkness, protected from all luminous radiations and from invisible radiations of known nature. After more than 15 days, uranium salts still emit radiations almost as intense as on the first day. Placing on the same photographic plate, with black paper, a flake kept for a long time in darkness and another that had just been exposed to daylight, the impression of the silhouette of the second is a little bit stronger than the first. Magnesium light, in the same conditions, produces only an imperceptible effect. If the flakes of double sulphate of uranyl and potassium are lively illuminated by an electric arc, or by the bright sparks of the discharge of a Leyden bottle, the impressions are noticeably darker. Therefore the phenomenon seems indeed an invisible phosphorescence phenomenon, but it does not seem intimately related to the visible phosphorescence and fluorescence. (Becquerel, 1896d, p. 691)

Becquerel was not the only one who reported this effect. Silvanus Thompson also stated that stimulation by light increased the emission of penetrating rays by uranium nitrate (Thompson, 1896a, p. 713). In the case of metallic uranium, Thompson stated that "the hyperphosphorescence of uranium in the metallic state is about equal in darkness and when exposed to light" (*ibid.*).

Becquerel's study of increased emission required the visual comparison between two black spots produced by different phosphorescent flakes upon photographic plates. It is very difficult to arrive at any definite conclusion if the spots are similar, and he could be misled by his theoretical expectation. However, there was an independent, objective method that could be tried: the measurement of the effect of the radiation upon the discharge of an electroscope.

Henri Becquerel also used this second method:

> The electroscope allowed me also to display the weak difference between the emission of a flake of uranium salt kept in darkness for eleven days, and the emission of the same flake vigorously iluminated by magnesium. In the first case, the speed of fall of the [electroscope] leaves was 20.69 [seconds of arc per second] and after luminous excitation it became 23.08. (Becquerel, 1896d, p. 691)

There was a second series of measurements. On the 28th March 1896, he measured the speed of discharge of an electroscope due to the action of a flake of double sulphate of uranyl and potassium. At 1:45 p.m., shortly after the uranium salt had been exposed to light, the speed was 38.18" /s; at 6:20 p.m., the speed was 33.60" /s. On the next day, at 5:40 p.m., the speed was 33.00"/s. He concluded:

> The numbers cited above show that, a short time after being exposed to light, the action of the flake of the uranium salt was a little bit stronger. In five hours there happened a slight weakening, and afterwards the action remained sensibly constant up to the next day. (Becquerel, 1896^{e}, p. 765)

The two series of measurements are in qualitative agreement. In both cases, a decrease of the intensity of radiation larger than 10% was observed when the uranium salt was kept in darkness. There seemed to exist strong evidence for accepting the increase of radiation intensity under stimulation by light and to interpret the phenomenon as a kind of phosphorescence. In later papers, Becquerel still held the same opinion (Becquerel, 1896f).

7.2 *Polarization of Becquerel's rays*

An important property of transversal waves is the possibility of polarizing them. After the discovery of X-rays, several researchers have tried to polarize them by reflection and using tourmaline crystals. If X-rays could be polarized, this would be a great step in the search for their nature. Although some papers reported positive results (Galitzine & Karnojitsky, 1896), most observers had failed to detect polarization of X-rays (Thomson, 1896).

Henri Becquerel believed that the radiation he was studying was similar to light. He had already "proved" that it could be refracted and reflected. It was natural to check whether it could be polarized. On the 30th March 1896, he reported positive evidence for the polarization of uranium radiation (Becquerel, 1896e).

A photographic plate was wrapped in black paper. Over the paper Becquerel placed two pieces of a thin tourmaline sheet (0.50 mm), oriented in perpendicular directions. Over them he put a single tourmaline layer (0.88 mm thick), with its axis parallel to that of oe of the small tourmalines and perpendicular to the other. In this condition, light passed through the parallel tourmalines and is stopped by the crossed tourmalines. A flake of double sulphate of uranyl and potassium was placed over this device.

> After 60 hours of exposition, the photographic plate was developed; it clearly showed the silhouette of the tourmalines, and the action through the parallel tourmalines was considerably stronger than through the crosses tourmalines. [...]
>
> This experiment therefore shows at the same time, for the invisible rays emitted by uranium salts, the double refraction, the polarization of both rays and their different absorption through the tourmaline. (Becquerel, 1896e, p. 763)

Becquerel repeated the same experiment using X-rays instead of uranium radiation, and observed no polarization effect.

In this case, as in some others, Becquerel had very scarce experimental evidence: his polarization experiment was *a single test* (perhaps repeated once, several days later). One of the most crucial pieces of evidence for the interpretation of the nature of uranium rays was the difference between two diffuse dark spots in a photographic plate.

In his 1903 book, Henri Becquerel published for the first time his photographic evidence for polarization of uranium rays (Becquerel, 1903b, plate II, fig. 6). It is very difficult to recognize the effect described by Becquerel (Fig. 5).

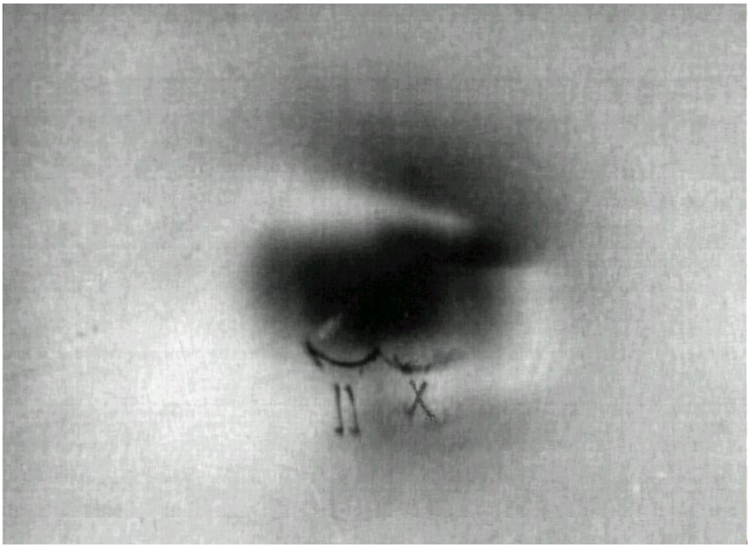


Fig. 5. Photograph of Becquerel's experiment to show the polarization of uranium rays. The region identified by II corresponds to parallel tourmaline plates, and the regions identified by X corresponds to crossed plates..

7.3 *Conclusions drawn from Becquerel's experiments*

At this time, Becquerel's experiments seemed to clearly prove that the radiation emitted by uranium compounds was a kind of electromagnetic transversal wave. Silvanus Thompson discussed the nature of the uranium radiations, and remarked:

> The extraordinary property exhibited by the uranium compounds of emitting a persistent invisible radiation that will pass through aluminium and produce photographic action would suggest that these rays are identical with Röntgen's, were it not that Becquerel's success in reflecting, refracting, and polarizing them proves that they are more akin to ultraviolet light. (Thompson, 1896b, p. 107)

8. RECTIFICATION OF BECQUEREL'S MISTAKES

Before 1898, Becquerel's work was not submitted to systematic duplication or criticism. It was simply reviewed and accepted as a contribution that did not strongly contrast with other known phenomena and therefore called for no deeper thoughts. The presentation speech of Becquerel's Nobel Prize completely misrepresents the historical situation, when refers to the reception of Becquerel's work, *before the work of Pierre and Marie Curie*, in the following words:

> It goes without saying that a discovery such as this was bound to excite the liveliest interest in the scientific world and give birth to a whole host of new investigations with the aim of making a thorough study of the nature of the Becquerel rays and determining their origin. (Törnebladh, 1967, p. 48)

Before 1898, only two aspects of Becquerel's work had been criticized: the polarization of uranium rays and the excitation of this radiation by light. Gustave le Bon, whose work on "black light" had been strongly criticized by Henri Becquerel, was the first to deny the polarization of uranium radiation, in May 1897:

> I suppose that the qualification "pretended" applied to black light means simply that my experiments do not always succeed. They do indeed not invariably succeed mainly because of the difficulty in preparing plates that are sensible

> to those new radiations. But if the qualification "pretended" should be applied to all experiments that do not always succeed, couldn't we apply it also to uranium rays? Would Mr. Becquerel like to state, for instance, that his experiments on the polarization of uranium rays – the polarization that if demonstrated would have a fundamental importance concerning the nature of those rays – can be repeated at will? Will he achieve repeating them safely? I have some reason to doubt it. (Le Bon, 1897, p. 691)

Notice that in his comment Le Bon did not clearly state that uranium rays cannot be polarized. He presented a *doubt*, not a clear *denial*. He also didn't state that he had repeated Becquerel's experiments. In January 1899, however, he clearly stated that he repeated the experiments and that uranium radiation cannot be polarized:

> Metallic radiations, including uranium radiations, have never shown any trace of polarization, by any of several methods I have used, even after an exposure of three months. It is certain, therefore, that those radiations [...] that cannot be a kind of light, are a new form of energy, as I have formerly said. (Le Bon, 1899, pp. 108-109)

Of course, the non-observation of polarization does not *prove* that something is not an electromagnetic radiation: recall that no polarization effect of X-rays had been observed. However, the denial of Becquerel's supposed prove of polarization of uranium rays destroyed one of the strongest arguments for their interpretation as high frequency ultraviolet rays.

In his first paper on the radiation emitted by thorium, Schmidt believed that he had found evidence for refraction, but found no sign of polarization by tourmalines (Schmidt, 1898).

In the beginning of 1899, Ernest Rutherford reproduced Becquerel's polarization experiment and could not perceive "the slightest difference in the intensity" of radiation passing through parallel or crossed tourmalines (Rutherford, 1899, p. 112). In the same paper, Rutherford described experiments to test refraction of uranium rays. He used prims of glass, aluminium and paraffin. The prisms were crossed by uranium radiation emerging from a slit cut in a thick lead plate. He observed no deflection of the radiation.

Johann Philipp Elster and Hans Friedrich Geitel found, in 1897, that the emission of uranium radiation could not be increased by excitation by sunlight (Elster & Geitel, 1897). The intensity was found to be constant (not "slightly decreasing", as Henri Becquerel described it) over several months. In this paper, Elster and Geitel stressed that the radiation from uranium can be distinguished from the effects produced by other substances (aluminium, zinc, phosphorescent paind and fluorspar) because these do not impart electrical conductivity to the air. Notwithstanding the title of their paper, they concluded that the name "hyperphosphorescence" cannot be applied to the observed phenomenon. This seems the first time that the concept of an invisible phosphorescence of uranium was criticized.

In April 1898, thorium was discovered to emit radiations similar to those of uranium (Badash, 1966). This led to an increased interest in the phenomenon. In this same year, polonium and radium were also found. In 1898, Marie Curie also rejected the name "hyperphosphorescence" and proposed the name "radioactivity":

> Uranium rays have frequently been called *Becquerel rays*. This name can be generalized and applied not only to uranium rays but also to the rays of thorium and to all similar radiations.
>
> I will call *radioactives* the substances that emit Becquerel rays. The name *hyperphosphorescence* that had been proposed for the phenomenon seems to me to convey a wrong idea about its nature. (Curie, 1899, p. 50)

The Curies rejected the old "invisible phosphorescence" concept, but proposed an explanation of radioactivity related to invisible fluorescence. Indeed, for several years they claimed that there existed an unknown, invisible, very penetrating cosmic radiation (similar to extremely hard X-rays), that could be transformed by radioactive bodies into less penetrating, detectable rays, by a process similar to fluorescence.[8]

The Curies concentrated their attention on the substances emitting radiation and not on the radiations. In 1899, Rutherford identified two kinds of radiation (called by him α and β) using as criterion the absorption of radiation by thin aluminium foils (Rutherford, 1899). A few months later, Fritz Giesel (1899) showed that β radiation could be deflected by a magnet and therefore could not be an electromagnetic radiation (see Malley, 1971). After a few years, a completely new view emerged: radioactive bodies emitted three kinds of radiation, two of them (α and β) deviable by magnetic fields (and, therefore, carrying electrical charges), and the third (γ), non deviable, similar to X-rays. The nature of the radiation emitted by uranium and other radioactive bodies was completely different from what Becquerel had believed and "proved" by his experiments.

The central core of our present theory of radioactivity was built by Rutherford and Soddy in 1902-3. They presented strong evidence for the gradual transformation of radioactive elements, the existence of radioactive series and spontaneous release of internal energy (Rutherford & Soddy, 1902a; 1902b; 1903; cf. Malley, 1979; TRENN, 1975).

9. BECQUEREL'S STRATEGY

In 1899, Henri Becquerel acknowledged for the first time some of his early mistakes, but tried to convey the impression that he had corrected them himself (Becquerel, 1899). From this time onwards, he devoted much of his energy to establish himself as the successful discoverer of radioactivity.

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[8] See MARTINS, Roberto de Andrade. The guiding hypothesis of the Curies' radioactivity research: secondary X-rays and the Sagnac connection, in this volume.

It is remarkable that, at one point of his 1903 book, which presented the state of the art of radioactivity up to that time, Becquerel stated that his only aim was to describe his own researches: "To describe the beautiful work of Mr. and Mrs. Curie is outside the scope of this memoir, that in principle contains only my personal researches" (Becquerel, 1903b, p. 105). Maybe this meant that the researches of other people, described in his book, were accessory to his own work.

Henri Becquerel used a systematic strategy: he turned his old mistakes into as so many successes; he described as his own the discoveries of others; he distorted the whole history of radioactivity and tried to show that he was the central protagonist. Let us show some instances of this strategy.

9.1 *Spontaneity of radiation*

Before 1898, Becquerel had never described the emission of uranium radiation as "spontaneous". Afterwards, when this was seen to be one of the fundamental aspects of radioactivity, Becquerel reinterpreted his work:

> Among the properties that I have pointed out at the beginning of my researches as characteristic of this radiation that was unknown, there are three fundamental ones that have been afterwards verified by all observers; they are: the spontaneity of radiation, its constancy and the property of imparting electrical conductivity to gases. (Becquerel, 1899, p. 771)

After describing his first "radioactivity" paper, Becquerel stated:

> Under those conditions, the phenomenon could be attributed to a transformation of solar energy, of the same kind as phosphorescence, but I soon recognized that emission was independent of any excitation of known nature – luminous, electric or thermic.

> We were therefore in face of a spontaneous phenomenon of a new kind. Here I show you the first print which revealed the spontaneity of the radiation emitted by the uranium salt. (Becquerel, 1903c, p. 2)

and at this point, Becquerel mentioned the first photograph taken in darkness, described in his second "radioactivity" paper. At other places, Becquerel explicitly stated that he recognized at this time the spontaneity of uranium radiation:

> This observation establishes the fundamental new fact of an emission of penetrating rays without apparent exciting cause. (Becquerel, 1903a, p. 13)

> [...] some days later, from 27th February to 1st March, I recognized that the emission was produced spontaneously, even when the uranium salt was kept protected from luminous excitation [...]. On the 2nd March 1896, I reported to the Academy of Sciences the conditions under which I have been led to observe the spontaneity of the radiation, the new fact from which follow all later studies. (Becquerel, 1900, p. 48)

9.2 *Constancy (in time) of emission*

Up to 1898, Becquerel described that the emission of radiation by uranium salts decreased with time, after stimulation by light. Afterwards, the story was changed.

According to Becquerel, after noticing that the uranium salt emitted radiation in darkness, he already supposed that the intensity was constant:

> As the uranium salts used had been prepared a long time ago, it was to be supposed that the intensity of the phenomenon was independent of time, and hence that emission should appear constant. All later experiments showed that the activity of uranium presented no appreciable decrease with time.
>
> [...] The photographic method was primarily a qualitative one while the electrical method gave numerical data, and the

> early measurements revealed the constancy of the radiation with time. (Becquerel, 1903c, p. 2)

In 1899, Becquerel still accepted that the intenstity of uranium radiation exhibited a decrease with time:

> It seems that there is a slight decrease of intensity during the first months and afterwards the intensity seems unchanged. (Becquerel, 1899, p. 772)

In this same paper, Becquerel stated that uranium radiation cannot be stimulated by physical influences, but did not acknowledge that Schmidt corrected him:

> [...] it was impossible to produce any noticeable change of the intensity of this emission by physical influences. (Becquerel, 1899, p. 777)

At other places, Becquerel stated that his early experiments showed that the intensity was constant:

> From the begining of those studies I have checked whether one could observe a progressive weakening of the radiated energy by subtracting those bodies to all known external excitation. A first series of experiments, pursued during two months, has initially showed that this energy did not decrease in an appreciable way. (Becquerel, 1900, p. 52; cf. Becquerel, 1903a, pp. 14-15)

At some places Becquerel referred to experiments that had shown an increase of the radiation when uranium salts were excited by light, but he did not state that himself had reported those effects:

> None of the attempts to exhibit an excitation by ultraviolet, infrared or light rays produced a [positive] result; the same was the case when uranium salts were excited by X-rays. However, in several experiments, after exposing the double

> sulphate of uranyl and potassium flakes to the action of sparks and electrical arc, a slight temporary increase of emission was observed, but this very weak effect seems another phenomenon superposed upon the constant and continuous emission by uranium. (Becquerel, 1900, p. 53)

Finally, in his 1903 book Becquerel stated that, using the electroscopic method, he had been able, as early as 14th March 1896, to prove that the intensity of radiation was not increased when the uranium salt was excited by magnesium light:

> In some cases, the photographic impression produced by samples of a salt exposed to light or strongly iluminated by electric sparks seemed stronger thatn the impression produced by the same bodies carefully kept away from any excitation. [...] But it seems that hose facts are accidental, because electrical measurements and experiments made in order to analyse the active rays have not allowed us to detect any action of this kind. This is, for instance, one of the earlier measurements made to detect this effect. (Becquerel, 1903a, p. 21)

Below this paragraph, Becquerel presented this table:

Double sulphate of uranyl and potassium in the electroscope, 1 cm. below the gold leaves (14 March 1896)

URANIUM SALT PROTECTED FROM EXCITATION			URANIUM SALT EXCITED BY MAGNESIUM LIGHT		
t	α	dα/dt	t	α	dα/dt
4h38m	15°.8		5h12m	19°.6	
5h07m	5°.8	0.34	5h39m	10°.0	0.35

The table seems, indeed, to exhibit a constancy of rate of discharge of the electroscope. Why, then, Becquerel did not publish this result in the early 1896, establishing the lack of excitation of uranium radiation by light? Well, it seems that he *did* refer to this experiment, on his communication to the

Academy of Sciences of 23rd March 1896 – but the data and conclusion were different. In that paper, Becquerel stated:

> The electroscope also allowed me to exhibit the slight difference between the emission of an uranium salt flake kept for eleven days in darkness, and the emission of the same flake strongly illuminated by magnesium. In the first case, the speed of fall of the [gold] leaves was 20.69, and after luminous excitation it became 23.08. (Becquerel, 1896d, p. 690)

If we transform those speeds from seconds of ar per second of time, to degrees per minute, we obtain respectively the values 0.34 and 0.38. The first value agrees with the above table (before excitation), but the second value is different.

It could have happened that Becquerel made several series of measurements, and that the series published in 1903 is not the same referred to in his 1896 communication. Notice, however, that the speed without excitation, computed from the data of the table, is not exactly 0.54, but 10/29 degrees per minute, equivalent to 20.69 seconds of arc per second – exactly the value published in the 1896 paper. This coincidence suggests that the communication of 23rd March was indeed reporting the same experiment described in 1903, but either in 1896 or in 1903 Becquerel made-up one of the measured speeds.

9.3 *Reflection and refraction of uranium radiation*

In 1899, Becquerel acknowledged that other researchers had shown that uranium radiation cannot be polarized, reflected or refracted, but his description implies that he had also, independently, arrived to that conclusion:

> Of the other properties that I have mentioned, polarization, reflection and refraction have not been verified by several observers that have repeated those experiments. The observations that I have made for three years have also

> disconfirmed my early conclusions and have shown that the phenomena were more complex. (Becquerel, 1899, p. 772)

In some of his accounts, Becquerel provided no hint about someone else having corrected his work:

> [...] I had been led to attribute to uranium radiation the properties of light, while all later experiments have demonstrated that this radiation cannot be reflected and refracted like light rays. (Becquerel, 1903c, p. 3; cf. Becquerel, 1902, p. 86)

Becquerel referred to the reflection experiments with metal mirrors:

> However, those experiments, and other that I do not cite here, do not allow us to conclude that there is regular reflection. I have repeated many times, with variations, one of my earlier experiments, in which a fragment of active substance was placed below a small concave tin mirror that produced nice optical images and that was adjusted to produce the image of the substance on the [photographic] plate; in this way I have obtained the print of no image and, in most cases, the mirror surface seemed the source of a new radiation, producing a stronger impression of the borders of the mirror than of the central regions that were farther from the plate. (Becquerel, 1899, p. 773; cf. Becquerel, 1903a, p. 26)

After this description, which is completely at variance with his early publications, Becquerel referred to Schmidt's work:

> The experiments of Mr. Schmidt with thorium have also led him to admit a phenomenon of diffuse reflection. (Becquerel, 1899, p. 773)

Hence, according to Becquerel, Schmidt dit not correct his mistake – he just confirmed his findings.

At other places, Becquerel described the concave mirror experiments without acknowledging the authorship of the mistakes:

> The effect, initially attributed to reflection, is due to the emission of secondary rays produced at the mirror by uranium radiation. Those rays produce diffuse prints; images like those of light rays are not obtained. (Becquerel, 1900, p. 54)

The same omission of the author of the mistake is used at several other places:

> It was observed that a flake covered by a steel mirror produces a stronger impression under the mirror than another non-covered flake. This phenomenon, initially attributed to reflection, is a secondary phenomenon to which we will return at another chapter. (Becquerel, 1903a, p. 16)

While describing his early acceptance of refraction of the uranium radiation, Becquerel usually only described his experiments with calcium sulphide and did not recall his similar experiments with uranium nitrate.

At some places, Becquerel acknowledged that Rutherford was the first to find out that uranium radiation is not refracted by glass and aluminium:

> Later experiments made by Mr. Rutherford and that I could repeat have shown that the uranium radiation was not deflected by glass, paraffin wax or aluminium prisms [...] (Becquerel, 1900, p. 54)

However, sometimes Becquerel attributed to himself the discovery that uranium radiation suffers no refraction:

> The explanation given above could only be accepted if it was verified that it is possible to deflect the studied radiation with a prism of a transparent substance. Now, experiment

> shows that the radiation passes without sensible deviation through glass and aluminium prims.
>
> These are some of the arrangements that allowed me to find out this fact: [...] (Becquerel, 1899, p. 775)

Becquerel did not state here that his prism experiments had apparently exhibited refraction, and that it was Rutherford who refuted his conclusions. He only stated that:

> This experiment is analogous to one of Mr. Rutherford's experiments, that gave him the same negatif result. (Becquerel, 1899, p. 775; cf. Becquerel, 1903a, pp. 110-113)

In his 1903 book, Becquerel also implied that Rutherford had just repeated his experiments:

> In 1899, nearly three years after my first publications, Mr. Rutherford methodically retook, one by one, most of the acpects that I had pointed out, and measured them. (Becquerel, 1903a, p. 93)

> Mr. Rutherford, after applying the photographic method to verify the absence of refraction and polarization of uranium rays, used only the electrical method. (Becquerel, 1903a, pp. 94-95)

Of course, Becquerel should have written: "after applying the photographic method to refute the refraction and polarization of uranium rays" ...

9.4 *Polarization*

At several places, Becquerel suggested to his readers that he had disproved the polarization of uranium rays before other researchers:

> One first photographic print, that I have shown to the Academy in March 1896 had presented a difference of absorption through tourmaline plates, according as they were

> crossed or parallel. A second trial obtained a few weeks later had given a similar result, but all other later experiments, whether with uranium or with radium, have been negative. Other observers have also arrived to this same result. (Becquerel, 1899, p. 772)

> In two successive tests, the impression through crossed tourmalines was smaller than through parallel tourmalines, but later experiments have not given the same results, whether with the same tourmalines or with other systems. The same negative conclusions were observed by Mr. Rutherford and Mr. Le Bon. (Becquerel, 1900, p. 54)

At other places, Becquerel omitted the works of other researchers:

> A second trial made with the same tourmalines gave a result similar to the first [...], but the effect is doubtless due to an accident, because all tests made afterwards, either with the same tourmalines or with a large number of different ones, gave negative results and both transmitted beams were equally strong. (Becquerel, 1903a, p. 25)

9.5 *Becquerel's criticism of Rutherford's work*

In the first chapter of his book, where the fundamental properties of uranium radiation are presented, Henri Becquerel describes all evidence as if he alone had arrived to all the correct conclusions: the radiation is spontaneous, is not increased by excitation by physical agents, the rays cannot be reflected, refractes or polarized, etc. No credit is given to Rutherford, Schmidt, Le Bon and others. He also conveys the view that he was the main protagonist of radioactivity research after the discovery of emision of radiation by thorium. Still worst: Henri Becquerel criticizes and detracts the works of several researchers – particularly that of Rutherford[9]. For instance:

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[9] It would be impossible to list all evidence of systematic depreciation of other researchers in Becquerel's book. As an example, see his

> Mr. Rutherford [...] concluded, as I had previously recognized, that the radiation [of uranium] is heterogeneous. Mr. Rutherford interprets his experiments considering two parts in the radiation: one, α, stronger, but easily absorbable; the other, β, much weaker, but very penetrating; he also supposes that β radiation is almost homogeneous.
>
> We are going to see below that the phenomenon is not that simple [...]. (Becquerel, 1903a, p. 95)

Of course, this remark was unjust. Becquerel had previously ascertained that uranium radiation was not homogenous *in the same sense that the X-rays emitted by a Crookes tube are not homogeneous* (Becquerel, 1896e, p. 765). Rutherford's conclusion about the existence of two different kinds of radiations (α and β) emitted by radium was a completely different thing. Besides that, the treacherous comment that "the phenomenon is not that simple" is clearly intended to discredit Rutherford's work.

At many places, Becquerel stated that Rutherford's experiments were a later reproduction of his own work: "I should however recall that this work [Rutherford's] was made two years after the measurements I had published on the same subject" (Becquerel, 1903a, p. 100). At other places, he also called the attention of the reader to limitations of Rutherford's 1899 research: "In those experiments and the conclusions drawn therefrom, the heterogeneity of the emitted radiation beam has not been taken into account" (Becquerel, 1903a, p. 97).

While describing Rutherford's discovery of the magnetic deviation of rays, Becquerel was also critical:

> Mr. Rutherford employed with a high competency, as we see, a relatively rough electrical method to exhibit a highly delicate phenomenon.

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description of the works of Giesel, Meyer and Schweidler on the magnetic deviation of radiation (Becquerel, 1903a, pp. 126-34).

> However, the experimental disposition rises a serious objection. [...]
>
> The conclusions of Mr. Rutherford could therefore be put in doubt, if they were not confirmed by other experiments. (Becquerel, 1903a, pp. 187-188)

Rutherford read Becquerel's book and was not very happy with what he found there. He wrote letters to several of his friends, asking their oppinion about Becquerel's book. Soddy wrote to him:

> I have just got Becquerel's book, but beyond the impression he gives that he, Becquerel, is a very ignorant person, have not yet formed any opinion of it.I don't think it is of any account.[10]

Oliver Lodge also replied to Rutherford:

> I have indeed not read Becquerel's book. I have hardly glanced at it.
>
> I know that Frenchmen have a great tendency to write so as practically to claim everything for France. It seems to be the fashion there. [...]
>
> I think that in this country your reputation is safe.[11]

In the long run, Rutherford's work became widely recognized, but in 1903, after the publication of Becquerel's book and his nomination for the physics Nobel Prize, Rutherford might have felt that his researches could be misinterpreted or forgotten. It was probably in order to present his own version of the story – the one generally accepted nowadays – that he immediatly began to write his famous book *Radioactivity* (Rutherford, 1904).

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[10] Letter from Frederick Soddy to Ernest Rutherford, 12th December 1903. CUL Add 7653.S117.

[11] Letter from Oliver Lodge to Ernest Rutherford, 11th December 1903. CUL Add 7653.L109.

This episode shows that scientists should never be trusted as faithful historians of their own work and that even a famous researcher may have been the actor of a comedy of errors.

10. HENRI BECQUEREL AND N-RAYS

In 1903, René Prosper Blondot (1849-1930) “discovered” a new kind of radiation while studying X-rays. He called them “N-rays”, as a homage to his home city, Nancy (Blondlot, 1903a, 1903b). The story of this episode has been described by several authors (Nye, 1980; Lagemann, 1977; Klotz, 1980; Rosmorduc, 1972; Martins, 2007).

Blondot claimed the observation of a penetrating radiation that could not be detected by some other physicists. The method of detection usually involved the visual observation of delicate changes of intensity of electric sparks and phosphorescent screens. Such method is strongly affected by subjective factors, such as suggestion and expectations. In 1904, the lack of reproductibility of Blondot’s results led to strong attacks by the scientific community and by 1905 the vast majority of researchers believed that the N-rays did not exist.

Most of the work on N-rays was developed at Nancy. However, some researchers from other places visited Blondot’s laboratory and afterwards carried out investigations on those radiations. One of them was Jean Becquerel (Nye, 1980, pp. 141, 152), the only son of Henri Becquerel – the fourth generation of the Becquerel scientific dynasty.

Shortly after the “death” of N-rays, Jean Becquerel’s work was strongly criticized:

> Mr. Jean Becquerel was compeled to go to Nancy to succeed, but afterwards he threw himself with passion in a series of researchers that rendered N-rays as suspect for physicists as those of Mr. Charpentier for physiologists. [...] And this rendered still less admissible the results of Mr. Charpentier, and those of Mr. Jean Becquerel, who successfully chloroformed metals, and noticed their

> anaesthesia by the diminution of emission. (Piéron, 1907, pp. 148-149)

Some recent authors have supposed that Jean Becquerel was a "reputed scholar" (Rosmorduc, 1972, p. 20) and that he had his own research laboratory at this time (Klotz, 1980, p. 131). As a matter of fact, in 1903 Jean Becquerel was a 25 years old engineer who had just become his father's assistant at the Paris Museum of Natural History[12]. He had never published a single article before his papers on N-rays. Jean Becquerel was not an experienced scientist working on his own laboratory – he was certainly working at his father's laboratory, under Henri Becquerel's supervision. Jean Becquerel's communications on N-rays to the Paris Academy of Sciences were not read by himself – they were presented by his father. Although the explicit authorship of the N-rays papers did not include the name of Henri Becquerel, he completely assumed their content.[13] Indeed, in December 1904, when most of the scientific community had strong doubts concerning the reality of N-rays, Henri Becquerel presented this opinion on the subject:

> Mr. Becquerel declares that his opinion on the question is well known, from the notes of Mr. Jean Becquerel that he communicated to the Institute, and that at this moment he, as well as his son, has nothing to change therein. (Becquerel, 1904, p. 718)

There is further evidence of Henri Becquerel's support of N-rays. In 1904, he was a member of the Lecomte Prize commitee of the Academy of Sciences. He prepared a positive report on Blondot's work, including N-rays, and defended that the Prize

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[12] The four physicists of the Becquerel family have successively occupied the same physics chair at the Museum of Natural History.

[13] It is likely that Henri Becquerel preferred to present those papers in the name of Jean Becquerel to give his young son the opportunity of recognition by the scientific community.

should be given to Blondot (Piéron, 1907, p. 153)[14]. Besides that, in his course of 1904-1905, Henri Becquerel lectured on X-rays, uranium rays and N-rays (Nye, 1980, p. 1545; Piéron, 1907, p. 161).

Blondot was smart enough to use Becquerel's support to strenghten his claims:

> In Paris, Mr. Jean Becquerel has published in the *Comptes Rendus* many notes on N and N_1 rays that were presented to the Academy of Sciences by his father, Mr. H. Becquerel, the eminent physicist. Is it possible that Mr. Jean Becquerel, with the concordance of his father, risked to endanger one of the most illustrious names of science, publishing observations that would leave the least doubt? (Blondlot, 1904, p. 621)

Why did Henri Becquerel accept as decisive proof the elusive visual observation of flickering sparks and phosphorescent screens? And why did he get so deeply involved with N-rays?

The first question is easly to answer: this was just a repetition of Henri Becquerel's mistakes in his early work on uranium radiation, when he accepted as decisive proof of delicate effects an irregular spot on a photographic plate. George Stradling commented on the effect of expectancy on N-rays research:

> A perusal of the literature on the N rays leads to the thought that the investigators often had the satisfaction of finding what they expected to find. (Stradling, 1907, p. 186)

Once Henri Becquerel was convinced that a phenomenon should exist, any dim effect was sufficient proof for him. In the case of uranium radiation, we have already discussed the

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[14] Henri Becquerel's initial report was replaced by another one written by Henri Poincaré, who circumvented the delicate situation by proposing that the prize should be given to Blondot because of all his scientific contributions.

theoretical expectations that guided his work. What about the N-rays? This leads us to the second question.

Let us examine Blondot's first paper "On a new kind of light" (Blondlot, 1903a), presented at the Paris Academy of Sciences meeting on 23rd March 1903. Blondot studied the radiation emitted by an X-ray focus tube, using as detector a small spark jumping between the points of two wires connected to a high voltage source (usually the same induction coil used to produce the X-rays). With this detector, Blondot claimed to detect polarization, reflection, refraction and diffusion of the radiation and therefore he concluded that they could not be X-rays – they should be a new kind of rays.

> From all that precedes it results that the rays that I have thus studied are not those of Röntgen, since the latter do not suffer refraction or reflection. Indeed, the small spark reveals a new kind of radiations emitted by the focus tube: those radiations pass through aluminium, black paper, wood, etc.; they are plane-polarized when they are emitted, they are susceptible to circular and elliptical polarization, they are refracted, reflected, diffused, but produce no fluorescence, nor photographic action. (Blondlot, 1903a, p. 737)

Let us recall that Becquerel had claimed the detection of reflection, refraction and polarization of uranium radiations. After the retification of those claims by other researchers, Becquerel wrote:

> One should therefore conclude that the most active part of the uranium radiation in those experiments suffers no reflection, no refraction and cannot be polarized as light. [...] But it has not been proved that this radiation is not accompanied by radiations identical with those of light.
>
> Future experiments will probably bring us the explanation of the contradictory phenomena that have just been pointed out. (Becquerel, 1900, pp. 54-55)

In his first communication on N-rays, Blondot remarked that his new radiation could provide the explanation of Becquerel's anomalous results:

> It is interesting to approximate the foregoing to the opinion issued by Mr. Henri Becquerel that, in some of his experiments, "appearances identical with those produced by refraction and total reflection of light could have been produced by luminous rays that had crossed aluminium". (Blondlot, 1903a, p. 738)[15]

Of course, Henri Becquerel did not assume that common (visible) light can pass through aluminium, but conjectured that radiations similar to light (polarizable, reflectable and refractable) could have produced the anomalous effects he described in his early experiments. Now, in 1903, Blondot was offering him a radiation suitable for the explanation of those anomalies.

There is one particular paper by Jean Becquerel that reminds us of Henri Becquerel's old ideas on the spectrum of uranium compounds. Blondot had measured the wavelengths of N-rays; Jean Becquerel searched – and found – a periodicity in this spectrum, as Henri Becquerel had done for uranium compounds.

> It seems possible to look for the origin of N and N_1 rays in the molecular motions that are produced in all bodies in state of deformation or molecular transformation. [...]
>
> Those considerations have led me to examine whether the wavelenghts measured by Mr. Blondot in the beam emitted by a Nerst lamp would present simple relations between them, as the wavelenghts of the motions produced by vibrating bodies. (Jean Becquerel, 1904, p. 1332)

The remarkable similarity between this work and Henri Becquerel's work on uranium spectral bands (Becquerel, 1885;

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[15] Blondot referred to Becquerel (1901).

see Martins, 1997) suggests that the same theoretical considerations were behind both studies.

All this suggests that Henri Becquerel's first stimulous for the study of N-rays was the expectation that those radiations could provide a suitable explanation for his anomalous results. As in the case of uranium radiation, his preconception intereferred with his observations and led him to describe non-existent phenomena.

Jean Becquerel published 11 communications between May and August 1904. After the "death" of N-rays, Jean Becquerel did not acknowledge his mistakes (Piéron, 1907, p. 154). During one year he published nothing and then turned, under the direction of his father, to "normal" researches.

11. ANALYSIS OF BECQUEREL'S MISTAKES

Becquerel was the author of a variety of experimental error. Most of them cannot be classified among the usual kinds of laboratory mistakes[16].

In order to discuss Becquerel's mistakes, it will be necessary to depend on the use of current scientific knowledge. There is nowadays some resistance against the use of anachronic knowledge within history of science, but there are also good arguments for their use, in this case: it allows reconstruction of the object of investigation (see Pickstone, 1995). It could happen, in principle, that Becquerel was never wrong, that all his experiments were correctly done and successful, but due to nationalistic preconceptions scientists of other countries (England, Germany, etc.) criticized him and were able to convince the scientific community that Becquerel was wrong. In that case, it would be ridiculous to try to understand what led Becquerel to make mistakes (there would be no mistakes to be explain); instead, it would be relevant to understand why he accepted the claims of his critics and changed his ideas about

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[16] Giora Hon (1989) has recently proposed a classification that does not include some of the crude errors commited by Becquerel.

uranium radiation. If, on the other hand, Becquerel commited a series of mistakes, it is relevant to understand the source of those mistakes, and discuss the existence of methodological rules that could help to avoid such errors.

11.1 *Reflection*

Consider, first, Becquerel's observation of specular reflection of uranium radiation. According to the accepted laws of geometrical optics, *no kind of radiation* could produce the effect described by Becquerel, even if it did suffer regular reflection at the surface of the metal mirror. There are a few possibilities of interpreting what happened:

a) Becquerel did make that experiment, and
  a1) he obtained a photograph exactly as he described it; or
  a2) he obtained a photograph with an irregular fogged circle where he thought there was a spot corresponding to the defect of the mirror; or
  a3) he obtained a photograph that showed no spot corresponding to the defect of the mirror, and misreported his observation.
b) Becquerel never made that experiment and described an imaginary effect.

In my opinion, the first case (a1) is unacceptable because it would conflict with our current knowledge. Becquerel never published the photograph of this experiment. If it existed, if it was as described by Becquerel, and if it were shown to me, I would be as surprised as if someone had shown to me a *perpetuum mobile*.

The second case (a2) is possible – it is the most benevolent interpretation of Becquerel's mistake. In that case, Becquerel was deceived by his theoretical expectation, and saw what he expected to see in a photograph where nothing or anything could be seen (as in a Rorschach spot). However, if Becquerel did believe that his photograph showed a spot corresponding to the defect of the mirror, why didn't he publish the photograph, as he did in other cases? It could have happened that, at first, Becquerel was misled by his photograph (a2), and afterwards, convinced that the effect did not exist, noticed that the photograph was not sufficiently sharp to support any claim, and chose not to publish it. In that case, he concealed his former mistake, but did not conterfeit his early report.

Alternatives (a3) and (b) would correspond to fraud.

11.2 *Refraction*

As described above, Becquerel found evidence for the refraction of his penetrating radiation in three different experiments: two with uranium nitrate (sealed tube and prism experiments) and one with calcium sulphide. The photographs corresponding to the uranium nitrate experiments have never been published. The photograph of the calcium sulphide experiment was published several times.

In the case of each uranium nitrate experiment, it is possible to distinguish several possibilities, as above:

a) Becquerel did make that experiment, and
  a1) he obtained a photograph exactly as he described it; or
  a2) he obtained a photograph with irregular fogged spots where he thought there was positive evidence for the effect he described; or
  a3) he obtained a photograph that showed no evidence for the effect he expected, and misreported his observation.
b) Becquerel never made that experiment and described an imaginary effect.

As in the case of reflection, case (a1) is unlikely because it would conflict with our current knowledge, and case (a2) is possible but problematic, because Becquerel never published those photographs.

In the experiment with calcium sulphide, however, the case is completely different. We know that Becquerel made the experiment and obtained a photograph exactly as he described it. Therefore, this experiment deserves a special analysis.

11.3 *Calcium sulphide radiation*

It is necessary to discuss, first, how could Becquerel observe the emission of penetrating radiation by calcium sulphide, given that we know that calcium sulphide emits no penetrating radiation. Then, it is necessary to discuss the evidence for reflection and refraction of radiation in that experiment.

There are several possible interpretations of Becquerel's observation of a penetrating radiation emitted by calcium sulphide:

a) Contrary to our current belief, calcium sulphide does indeed emit a penetrating radiation in some unknown circumstances.
b) Calcium sulphide emits no penetrating radiation, and Becquerel observed the radiation emitted by another substance:
  b1) there was an impurity mixed with calcium sulphide; or
  b2) Becquerel took a radioactive (uranium) sample for calcium sulphide.
c) The photographic plate was not adequately protected against light emitted from calcium sulphide; what Becquerel observed was light, not a penetrating radiation.

Interpretation (a) is unlikely, but at that time it could be supported by Niewenglowsky's experiments that had also detected penetrating radiation emitted by calcium sulphide. The impurity interpretation (b1) is unlikely because the effect observed by Becquerel was very strong (even stronger than those obtained with pure uranium compounds) and because it disappeared after some time. We know, of course, that there are some short-lived strongly radioactive substances, but they were not available in Becquerel's laboratory. Interpretation (b2) is very unlikely, given that Becquerel was well acquainted with luminescent substances, and calcium sulphide has a long-lived, strong phosphorescence of peculiar colour.

Interpretation (c) – a rude negligence – is the most likely interpretation, because the radiation emitted by calcium sulphide exhibited reflection and refraction. There is a difficulty, however: Becquerel described that in those experiments the photographic plate was wrapped in black paper and there was an aluminium plate, 2 mm thick, between the samples and the plate. Even if the paper was not opaque enough, light would never pass through that metal plate. Light could only affect the photographic plate *if the aluminium plate was missing*.

Could that be the case? It seems unbelievable that Becquerel could be so negligent. Such an error could perhaps be ascribed to Becquerel's laboratory assistant, called Louis Matou.

Is there any independent evidence that the aluminium plate was missing? Yes, there is. Contrary to his uranium photographs, that exhibited diffuse boundaries, Becquerel's calcium sulphide photograph is remarkably sharp. It shows details that could never be produced by any radiation, if the aluminium plate were between sample and photographic plate. Indeed: a point source of radiation can project a well defined shadow of an object at a large distance; however, an extended source of radiation (the phosphorescent sample inside the glass tube) will produce a poorly defined shadow. In the circumstances of Becquerel's experiment, if there were a distance of 2 mm between the thin glass plate that was used to close the tube and the photographic plate, the border of the glass plate could not be as sharp as it is. Scientifically speaking, the photograph published by Becquerel cannot have been produced under the circumstances he described.

Even if the metal plate was missing, it would be difficult for light emitted by calcium sulphide to pass through black paper. Hence, it is likely that the paper wrapping was also missing.

This interpretation of Becquerel's experiment elucidates other misterious aspects: Why was the radiation of calcium sulphide reflected and refracted by glass? Because the observed radiation was simply light. Why didn't the same sample of calcium sulphide emit penetrating radiation in later experiments? Because in later experiments Becquerel was careful enough to wrap the plate and/or to use an aluminium plate.

Only this interpretation (rude negligence) can fit our current physical knowledge and the effects described and registered by Becquerel. Notice that, in that case, Becquerel never suspected that he comitted such an error, since he was proud of his experiment, he exhibited and published his photograph, and he never concealed that evidence.

11.4 *Stimulation of emission of radiation by light*

As described above, Becquerel provided two classes of evidence for the excitation of uranium radiation by light. One kind was the comparison between the spots produced by two samples upon a photographic plate. The other one was the measurement of the speed of discharge of an electroscope.

In the case of the first method, only a very strong difference could be unabiguously detected. Indeed: the samples used by Becquerel were not exactly equal, but only roughly similar – and any observed small difference in radiation could be ascribed to a difference in the samples themselves. Besides that, visual comparison between two dark spots in a photographic plate is highly subjective, except if one is much darker than the other. Becquerel reported that when a flake of the uranium salt was illuminated by an electric arc or by discharge of a Leyden bottle, "the impressions are noticeably darker" (*les impressions sont notablement plus noires*) (Becquerel, 1896d, p. 691). "Noticeable", of course, can be interpreted either as striking or as merely observable.

According to our current physical knowledge, Becquerel could not have observed any strong increase in radiation emission, because uranium radiation is not excited by light. The electric arc could *heat* the uranium salt flake, and that could increase the photographic effect below the illuminated sample – but discharge of a Leyden bottle would not produce the same effect. It seems likely that the spots were very similar to one another, but Becquerel saw one of them darker than the other because he expected the effect to occur. The photographic evidence was never published by Becquerel. This case is comparable to Becquerel's mistaken experiments on reflection and refraction of uranium radiation.

On the other hand, Becquerel's electroscopic experiment showed a new kind of error. In this case, it is possible to detect Becquerel's error using information published seven years later. The table containing the 28th March 1896 measurements was published in Becquerel's book (Becquerel, 1903b, p. 20) and it allows us to recognize several problems: (a) lack of precision of measuments; (b) lack of reproductibility; (c) only a single series of measurements was made. Let us briefly describe those problems.

Becquerel measured time in hours and minutes, and measured angles in degrees and tenths of degree. Given the size of the electroscope leaves, it is likely that his instrument only allowed estimation of tenths of degree by visual interpolation[17]. Parallax and mistaken interpolation could easily introduce random errors of the order of half degree.

The inicial rate of discharge of the electroscope was computed from *a single pair of measurements*: at 1:44 hours the angle between the electroscope leafs was 10.3 degrees, and at 1:54 hours the angle was 4.5 degrees. Therefore, there was a motion corresponding to 5.8 degrees in 10 minutes, or 0.58 degrees/minute. Transforming to seconds of arc per second of time, one obtains 34.8"/s, with an error that could amount to about 10%.

In his paper of 1896, as described above, Becquerel presented a table that showed that at 1:45 p.m. the speed was 38.18"/s. Besides the computational error, Becquerel's figures published in 1896 seemed to imply a very large precision.

The second measurement (without any screen), between 6:12 and 6:25 p.m., gave a discharge speed of 0.54 degrees/minute or 32.4"/s (again, the number published in 1896 is wrong). And

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[17] If the electroscope leaves had a length of 6 cm, a displacement of one millimetre would correspond to one degree. It is unlikely that the scale of the electroscope had divisions smaller than that.

finally, the next day, Becquerel measured four different speeds of discharge of the electrocope: 0.43, 0.50, 0.51 and 0.56 degrees/minute, with a mean speed of 0.53 degrees/minute or 31.8"/s. The difference between the first and second (or third) measurement is smaller than 10% and does not seem significative.

There is, however, a deeper problem with Becquerel's measurements. In the series of measurements of 29th March, as just shown above, the measured speeds varied between 0.43 and 0.56 degrees/minute. Why was there so large a variation? The reason is very simple: the speed of the electroscope leaves was not constant, but depended on the initial angle. It can be easily perceived from Becquerel's tables that the angular speed was smaller for larger initial angles. Becquerel, however, had wrongly stated that "when we follow the progressive approach of the electroscope golden leaves during discharge, it is recognized that, for apertures that do not exceed 30°, the angular changes are very sensibly proportional to time [...]" (Becquerel, 1896d, pp. 689-690).

Of course, in order to admit *quantitive comparison* between measurements, his experiments should always have been done between the same initial and final angles. *This was never the case*. In the first measurement described above, the angle varied from 10.3 to 4.5 degrees; in the second, from 13.8 to 6.8 degrees; and in the third, from 30.0 to 0.5 degrees. The third measurement cannot, of course, be compared with the first or second. If we select from the third measurement the data taken between 5:36 and 5:59 o'clock, when the angle varied from 13.5 to 0.5 degrees, the rate of discharge was 0.56 degrees/minute – that is, a value that is *between* the results of the first and second measurements.

What conclusion could be drawn? Of course, that no decrease of radiation intensity was detected. Becquerel drew, however, the opposite conclusion. It seems that Becquerel was not skilful in quantitative research. He overlooked elementary rules about measurement and interpretation of data.

11.5 *Polarization of uranium radiation*

Becquerel's evidence for polarization of uranium radiation was, in a sense, similar to that for reflection and refraction: he compared two spots on a photographic plate, in a single trial, and concluded that one of them was "considerably stronger" than the other. However, in this case, Becquerel published the corresponding photograph and the occurrence of fraud is excluded. The photograph cannot be as easily interpreted as Becquerel claimed: indeed, it exhibits a diffuse dark spot where it is very difficult to see any difference between the intensity of radiation transmited through parallel or crossed tourmalines. Becquerel was certainly misled by his theoretical expectations, as in the case of the photographic evidence for the increase of radiation by light stimulation.

11.6 *Summary*

In all cases, Becquerel was strongly influenced by his theoretical preconceptions. His mistakes, however, belong to different kinds.

a) Calcium sulphide: he (or his assistant) committed a rude mistake – the experiment was not performed as planned and described. Once this error was done and overlooked, the photographic evidence did support Becquerel's interpretation.

b) Electroscopic measurement of stimulation of radiation emission by light: Becquerel was unable to follow some well known rules about measurement and manipulation of quantitative data.

c) Photographic evidence for polarization and stimulation of radiation by light: the photographic evidence was inconclusive, but Becquerel arrived nevertheless to definite conclusions. The first case cannot be classified as fraud, as Becquerel published the corresponding photograph and claimed that it supported his interpretation.

d) Photographic evidence for reflection and refraction of uranium radiation: either the photographic evidence was inconclusive, as above, or Becquerel frauded the experiment.

12. COULD BECQUEREL'S ERRORS HAVE BEEN AVOIDED?

In a few years, Becquerel's mistakes were corrected by the scientific community. However, why should we trust the authors that reported that Becquerel was wrong, instead of Becquerel? Can we interpret the episode as a blind conflict between people with different preconceptions – each one seeing in his/her experiment what he/she expected to observe?

It seems impossible, in this episode, to adopt a relativist interpretation, since Becquerel was soon convinced that his earlier results were wrong. In some cases, he presented his early evidence and tried to convince the scientific community that he had been unavoidably misled by objective data, but he never claimed that the refutation of his claims was problematic or wrong.

Were Becquerel's mistakes unavoidable? Could a better experimental methodology lead Becquerel to correct results? Maybe. Had Becquerel been as careful and critical as later tradition said he was[18], he would *deserve* the name of "discoverer of radioactivity".

The possibility of mistakes in empirical investigation had been discussed since the beginning of the scientific revolution, by Francis Bacon and other authors. In the late 19th century, we find several works calling again the attention of scientists to the need for special care in experimental research. William Stanley Jevons, for instance, stresseed the role of preconceptions:[19]

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[18] According to Oliver Lodge, "Henri Becquerel set himself carefully and critically to examine the kind of penetrating radiation which fluorescent substances exposed to light might possibly be found to emit [...]" (Lodge, 1912).

[19] The first edition of William Stanley Jevons' book *The principles of science* was published in 1874. I present citations of this book, not of more recent books on philosophy or methodology of science, to avoid anachronism.

> Every observation must in a certain sense be true, for the observing and recording of an event is in itself an event. But before we proceed to deal with the supposed meaning of the record, and draw inferences concerning the course of nature, we must take care to ascertain that the character and feelings of the observer are not to a great extent the phenomenon recorded. The mind of man, as Fancis Bacon said, is like an uneven mirror, and does not reflect the events of nature without distortion. [...]
>
> It is difficult to find persons who can with perfect fairness register facts for and against their own peculiar views. Among uncultivated observers the tendency to remark favourable and forget unfavourable events is so great, that no reliance can be placed upon their supposed observations. (Jevons, 1958, p. 402)

Accordingly, Jevons recommended:

> Thus the successful investigator must combine diverse qualities: he must have clear notions of the result he expects and confidence in the truth of his theories, and yet he must have that candour and flexibility of mind which enable him to accept unfavourable results and abandon mistaken views. (Jevons, 1958, p. 404)

Of course, it is easier to talk about the correct attitude than to show how it can be attained. There is, however, a simple methodological rule: to repeat and to vary experiments.

> Even when we are not aware by previous experience of the probable presence of a special disturbing agent, we ought not to assume the absence of unsuspected interference. If an experiment is of really high importance, so that any considerable branch of science rests upon it, we ought to try it again and again, in as varied conditions as possible. We should intentionally disturb the apparatus in various ways, so as if possible to hit by accident upon any weak point. (Jevons, 1958, p. 431)

Suppose Becquerel repeated and varied his experiments: he would probably find contradictory results. If he were not too stubborn, it is likely that he would have been able to correct his former mistakes. Or was he too obstinate?

In several cases (such as the polarization experiment), Becquerel made a single experiment and jumped to conclusions. In other cases, he did repeat and vary his experiments – but that did not lead him to correct his former view. Such was the case of his refraction and light excitation experiments. He also repeated his calcium sulphide experiments, noticed a contradictory result, but only concluded that the sample had lost its capacity of emitting penetrating radiation. It seems that Henri Becquerel was, indeed, too stubborn to correct his own views. His theoretical confidence in his interpretation of the radiation emitted by uranium compounds as an invisible phosphorescence was so strong that he acted exactly as Jevons described the "uncultivated observers" would do. No methodological rule will lead to safe results in the hands of the wrong scientist.

Becquerel's mistakes were avoidable and a better experimental methodology could have lead to correct results – in the hands of another scientist. Becquerel himself, however, did not have the adequate experimental training and correct attitude.

We must acknowledge that there are highly qualified observers, such as Newton and Faraday (Jevons, 1958, chapter xxvi, pp. 574-593) and unreliable observers (such as Becquerel and Blondot). One of the signs of a good experimenter is the capacity of finding and accepting evidence opposite to his own preferred ideas. Of course, even the best experimenter will commit mistakes. For that reason, it is unavoidable that the construction of science should be a collective enterprise.

13. FINAL COMMENTS

Henri Becquerel's experimental research on the phenomenon we now call "radioactivity" was full of serious mistakes. He ascribed to uranium radiation several properties – such as reflection, refraction, polarization, and increase by light stimulation – that were corrected by other researchers. In the study of uranium radiation – as in the case of N-rays – Henri Becquerel was a careless and perhaps unfair observer, misguided by his preconceptions. Later, however, he was socially successful in reinterpreting his early work and convincing the scientific community that his research was seldom mistaken, and that he had himself corrected his earlier mistakes.

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BIBLIOGRAPHIC REFERENCES

BADASH, Lawrence. The discovery of thorium's radioactivity. *Journal of Chemical Education* **43**: 219-220, 1966.

BECQUEREL, Edmond. Note sur la phosphorescence produite par insolation. *Annales de Chimie et de Physique* [3] **22**: 244-255, 1848.

BECQUEREL, Henri. Relations entre l'absorption de la lumière et l'émission de la phosphorescence dans les composés d'uranium. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **101**: 1252-1256, 1885.

BECQUEREL, Henri. Sur les différentes manifestations de la phosphorescence des minéraux sous l'influence de la lumière ou de la chaleur. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **112**: 557-563, 1891.

BECQUEREL, Henri. Sur les radiations émises par phosphorescence. *Comptes Rendus Hebdomadaires des*

Séances de l'Académie des Sciences de Paris **122**: 420-421, 1896 (a).

BECQUEREL, Henri. Sur les radiations invisibles émises par les corps phosphorescents. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 501-503, 1896 (b).

BECQUEREL, Henri. Sur quelquer propriétés nouvelles des radiations invisibles émises par divers corps phosphorescents. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 559-564, 1896 (c).

BECQUEREL, Henri. Sur les radiations invisibles émises par les sels d'uranium. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 689-694, 1896 (d).

BECQUEREL, Henri. 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. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 762-767, 1896 (e).

BECQUEREL, Henri. Émission de radiations nouvelles par l'uranium métallique. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 1086-1088, 1896 (f).

BECQUEREL, Henri. Note sur quelques propriétés du rayonnement de l'uranium et des corps radio-actifs. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **128**: 771-777, 1899.

BECQUEREL, Henri. Sur le rayonnement de l'uranium et sur diverses propriétés physiques du rayonnement des corps radio-actifs. Vol. 3, pp. 47-78, in: GUILLAUME, Charles-Édouard; POINCARÉ, Lucien (eds.). *Rapports Présentés au Congrès International de Physique réuni a Paris en 1900.* 3 vols. Paris: Gauthier-Villars, 1900.

BECQUEREL, Henri. Sur la radioactivité secondaire. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **132**: 734-739, 1901.

BECQUEREL, Henri. Sur la radio-activité de la matière. *Notice of the Proceedings of the Meetings of the Members of the Royal Institution of Great Britain* **17**: 85-94, 1902.

BECQUEREL, Henri. Recherches sur une propriété nouvelle de la matière – activité radiante spontanée ou radioactivité de la matière. *Mémoires de l'Académie des Sciences de l'Institut de France* **46**: 1-360, 1903 (a).

BECQUEREL, Henri. *Recherches sur une propriété nouvelle de la matière: activité radiante spontanée ou radioactivité de la matière*. Paris: Firmin-Didot, 1903 (b).

BECQUEREL, Henri. Sur une propriété nouvelle de la matière, la radio-activité. *Les Prix Nobel* **3**: 1-15, 1903 (c).

BECQUEREL, Henri. Enquête. Les rayons N existent-ils? Déclaration de M. H. Becquerel, membre de l'Institut, professeur de physique au Muséum et à l'École Polytechnique. *Revue Scientifique* [5] **2**: 718, 1904.

BECQUEREL, Jean. Sur l'émission simultanée des rayons N et N_1. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **138**: 1332-5, 1904.

BLONDOT, René. Sur une nouvelle espèce de lumière. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **136**: 735-738, 1903 (a).

BLONDOT, René. Sur de nouvelles sources de radiations susceptibles de traverser les métaux, le bois, etc., e sur de nouvelles actions produites par ces radiations. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **136**: 1227-1229, 1903 (b).

BLONDOT, René. Enquête. Les rayons N existent-ils? Déclaration de M. Blondot, Professeur à l'Université de Nancy. *Revue Scientifique* [5] **2**: 620-621, 1904.

BOUTY, E. Les rayons de Röntgen et la photographie à travers les corps opaques. *Revue Scientifique* [4] **5**: 610-617, 1896.

CURIE, Marie Sklodowska. Les rayons de Becquerel et le polonium. *Révue Génerale des Sciences* **10**: 41-50, 1899.

ELSTER, Johann Philipp Ludwig Julius & GEITEL, Hans Friedrich Karl. Versuche über Hyperphosphorescenz. *Jahresbericht des Vereins für Naturwissenschaft zu Braunschweig* **10**: 149-153, 1897.

GALITZINE, B. & KARNOJITSKY, A. de. Recherches concernant les propriétés des rayons X. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 717-718, 1896.

GIESEL, Fritz. Über die Ablenkbarkeit der Becquerelstrahlen im magnetischen Felde. *Annalen der Physik und Chemie* [2] **69**: 834-836, 1899.

HENRY, Charles. Augmentation du rendement photographique des rayons Roentgen par le sulfure de zinc phosphorescent. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 312-4, 1896.

HOLMES, Frederic L. Do we understand historically how experimental knowledge is acquired? *History of Science* **30**: 119-136, 1992.

HON, Giora. Towards a typology of experimental errors: an epistemological view. *Studies in History and Philosophy of Science* **20**: 469-504, 1989.

JEVONS, William Stanley. *The principles of science: a treatise on logic and scientific method*. New York: Dover, 1958.

KELVIN, Lord; BEATTIE, J. C.; DE SMOLAN, M. S. Experiments on the electrical phenomena produced in gases by Röntgen rays, by ultraviolet light, and by uranium. *Proceedings of the Royal Society of Edinburgh* **21**: 393-428, 1897.

KLOTZ, Irving M. The N-ray affair. *Scientific American* **242** (5): 122-129, May 1980;

LAGEMANN, Robert T. New light on old rays: N rays. *American Journal of Physics* **45**: 281-284, 1977.

LE BON, Gustave. La lumière noire et les propriétés de certaines radiations du spectre. *Revue Scientifique* [4] **7**: 689-691, 1897.

LE BON, Gustave. La luminescence invisible. *Revue Scientifique* [4] **11**: 106-109, 1899.

LODGE, Oliver. The discovery of radioactivity, and its influence on the course of physical science (Becquerel memorial lecture). *Journal of the Chemical Society* **101**: 2005-2032, 1912.

MALLEY, Marjorie. The discovery of atomic transmutation: scientific styles and philosophies in France and Britain. *Isis* **70**: 213-223, 1979.

MALLEY, Marjorie. The discovery of the beta particle. *American Journal of Physics* **39**: 1454-1460, 1971.

MARTINS, Roberto de Andrade. Becquerel and the choice of uranium compounds. *Archive for History of Exact Sciences* **51** (1): 67-81, 1997.

MARTINS, Roberto de Andrade. *Os "raios N" de René Blondlot: uma anomalia na história da física*. Rio de Janeiro: Booklink; São Paulo, FAPESP; Campinas, GHTC, 2007.

NIEWENGLOWSKI, Gaston Henri. Sur la proprieté qu'ont les radiations émises par les corps phosphorescents, de traverser certains corps opaques à la lumière solaire, et sur les expériences de M. G. Le Bon, sur la lumière noire. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 385-386, 1896.

NYE, Mary Jo. N-rays: an episode in the history and psychology of science. *Historical Studies in the Physical Sciences* **11**: 125-156, 1980.

PICKSTONE, John V. Past and present knowledges in the practice of the history of science. *History of Science* **33**: 203-224, 1995.

PIÉRON, Henri. Grandeur et décadence des rayons N. Histoire d'une croyance. *L'Année Psychologique* **13**: 143-169, 1907.

ROSMORDUC, Jean. Une erreur scientifique au début du siècle: "les rayons N". *Revue d'Histoire des Sciences et de leurs Applications* **25**: 13-25, 1972.

RUTHERFORD, Ernest. Uranium radiation and the electrical conduction produced by it. *London, Edinburgh and Dublin Philosophical Magazine and Journal of Science* [5] **47**: 109-163, 1899.

RUTHERFORD, Ernest. *Radioactivity*. Cambridge: Cambridge University Press, 1904.

RUTHERFORD, Ernest & SODDY, Frederick. The radioactivity of thorium compounds. I. An investigation of the radioactive emanation. II. The cause and nature of radioactivity. *Journal of the Chemical Society, Transactions* **81**: 321-350, 837-860, 1902 (a).

RUTHERFORD, Ernest & SODDY, Frederick. The cause and nature of radioactivity. *Philosophical Magazine (series 6)* **4**: 370-396, 569-585, 1902 (b).

RUTHERFORD, Ernest & SODDY, Frederick. Radioactive change. *Philosophical Magazine (series 6)* **5**: 561-576, 576-591, 1903.

SCHMIDT, Gerhard C. Ueber die von den Thorverbindungen und einigen anderen Substanzen ausgehende Strahlung. *Annalen der Physik und Chemie* [2] **65**: 141-151, 1898.

STRADLING, George Flowers. A resumé of the literature of the N rays, the N_1 rays, the physiological rays and the heavy emission, with a bibliography. *Journal of the Franklin Institute* **164**: 57-74, 113-130, 177-199, 1907.

THOMPSON, Silvanus P. On hyperphosphorescence. *Report of the 66th Meeting of the British Association for the Advancement of Science* [**66**]: 713, 1896 (a).

THOMPSON, Silvanus P. On hyperphosphorescence. *The London, Edinburgh and Dublin Philosophical Magazine and Journal of Science* [5] **42**: 103-107, 1896 (b).

THOMSON, J. J. The Röntgen rays. *Nature* **53**: 581-583, 1896.

TÖRNEBLADH, H. R. Physics 1903. Presentation speech by Dr. H. R. Törnebladh, President of the Royal Swedish

Academy of Sciences. Pp. 47-51, in: *Nobel lectures including presentation speeches and laureates' biographies. Physics. 1901-1921.* Amsterdam: Elsevier, 1967.

TRENN, Thaddeus. J. *The self-splitting atom: the history of the Rutherford-Soddy collaboration.* London: Taylor and Francis, 1975.

TROOST, Louis Joseph. Observation à l'occasion de la communication de M. H. Becquerel. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences de Paris* **122**: 694, 1896.

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