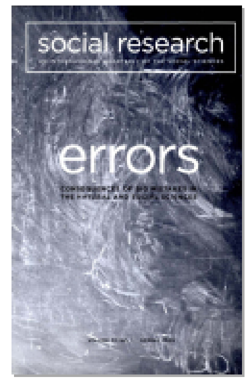
Becquerel's Blunder

Lawrence Badash



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INTRODUCTION

THE DISCOVERY BY WILHELM CONRAD RÖNTGEN OF X-RAYS IN LATE 1895 was the most globally astonishing scientific event prior to the atomic bombing of Hiroshima and Nagasaki in 1945.* Some 50 books and pamphlets and 1,000 papers on X-rays were published in 1896 alone, remarkable testimony to the impact of these penetrating rays.[1]

By contrast, Henri Becquerel, who discovered radioactivity just a few months later, wrote seven papers on the subject in 1896, only two the following year, and then left this seemingly exhausted topic. Others added several papers in this period, but amidst the plethora of various radiations being studied at that time, the radiations from uranium did not seem extraordinary (Badash, 1965a). Especially, uranium rays could not produce the sharp images of bones through the flesh of a living hand that made X-rays so intensely fascinating. Not until Gerhard C. Schmidt and Marie Curie in 1898 independently reported that thorium exhibited the same properties as uranium was interest in radioactivity resurrected, drawing Becquerel and many others back to his discovery.

Over the next century, X-rays were used in increasingly sophisticated medical imaging devices, applied to test the integrity of welds and other structures, told us more about the nature of atoms (the importance of atomic number) through Henry Moseley's examination of their spectra, and led to the revolution in molecular biology through X-ray crystallography. Again by contrast, radioactivity seemingly was left in the dust. Aside from radium's sometimes successful medical application in the treatment of cancer and other diseases, it appeared to have limited import: a pre-World War I use in luminous paint for such showbiz items as poker chips, cocktails, and nightclub dancers' costumes, and its wartime employment in luminous watches and gun sights (Badash, 1978). During the decades before World War II, however, physicists and chemists determined that there was a nucleus to the atom and that nuclear reactions could be induced, and they gained an understanding of the components of the nucleus. The discovery of nuclear fission in late 1938, coupled with the fears of war, led to projects in several countries that ultimately produced both nuclear weapons and nuclear reactors. Then, for nearly half a century, the bomb dominated international relations among the developed countries, in particular the superpowers: the United States and the Soviet Union.

The discoveries by Röntgen and Becquerel thus were both extremely important to society, but that of radioactivity had a far more significant impact on science and the world. It is therefore worthwhile to look more closely at what constitutes discovery and what Becquerel thought he was doing.

BACKGROUND TO THE DISCOVERY

For some time, those who study nature have recognized that the truths of science are written with a lower-case *t*, not a capital *T*. In practice this means that our understanding of Nature is not permanent; it is influenced by the ideas and apparatus at our disposal. More insightful thoughts, new data, and more powerful tools have changed the paradigm.

This suggests that a large percentage of past scientific ideas have been modified, and that many current beliefs will be changed in the future. Scientific error thus is common, if not almost the norm. Why then are we fascinated by scientific mistakes if they are so plentiful? It is not because we wish to gloat with the wisdom of hindsight. Rather, these examples provide us with insights. We learn what were the contemporary beliefs and practices that influenced a new interpretation of Nature, and we see, gratifyingly, that scientific research is a very human enterprise: scientists can fixate possessively on an idea that brought them a measure of fame and ignore other viewpoints that are equally reasonable. We may also speculate whether the mistake hindered the development of science or actually inspired increased activity. For a successful theory should not merely explain existing data but suggest new research to be performed. Success need not be congruent with a later consensus of truth.

The discovery of radioactivity by Antoine Henri Becquerel early in 1896 provides a useful example to explore scientific error. This essay's title, calling his work a *blunder*, is meant to be provocative and alliterative, far more than to convey an accurate depiction of his competence. In fact, Becquerel was a highly talented and innovative physicist, even if his vision was limited. A professor of physics at the National Museum of Natural History in Paris, and at age 43 already a member of the prestigious Académie des Sciences, Becquerel was moved in January 1896 to see if X-rays were emitted from luminescent crystals. X-rays had been discovered in Würzburg, Germany, just a month or two earlier by Röntgen, and were then the most newsworthy topic among scientists.

It is true, of course, that humans were long familiar with the passage of visible light through glass and some liquids, but X-rays were novel not only because they were invisible to the eye, but particularly because they traversed fairly thick pieces of opaque material. Their true nature, whether particle or radiation, remained unclear for several years. But that did not stop feverish investigation. In Paris, for example, the physicians Oudin and Barthélemy, tempted by the diagnostic potential of X-rays, quickly began work with a Crookes tube (a variety of vacuum tube) furnished to them by the physicist Gaston Séguy. Soon, a report of their work and a photograph of the bones of a hand were sent to the Academy of Sciences, where they were communicated at the meeting on January 20, 1896, by the prominent professor of mathematical physics at the Sorbonne, Henri Poincaré.[2]

The speed involved in this report was remarkable, considering that Röntgen's original paper was received by the Physical Medical Society of Würzburg on December 28, 1895, and, although printed and distributed during the early days of the new year, it did not appear translated in other journals for almost a month. For example, *Nature* (London) reprinted the paper on January 23, 1896, the *Electrician* (London) on

January 24, *L'Éclairage Électrique* (Paris) on February 8, and *Science* (New York) on February 14. Yet, news of this discovery traveled so fast and so interested the world that others quickly repeated the experiments and were able to report on them within a few weeks.

Poincaré's attention was drawn less to the rays than to their point of origin, namely the glowing spot where cathode rays struck the wall of the glass tube. "Can we not ask," he wrote, "if all bodies whose fluorescence is sufficiently intense, do not emit, besides luminous rays, the x-rays of Röntgen, *whatever the cause of their fluorescence*?" (Poincaré, 1896: 56) He said the same thing at the weekly meeting of the academy on January 20. Scientists commonly argue by analogy, so this reasonable question inspired several colleagues to investigate (Becquerel, 1903).

For example, Charles Henry and Gaston Niewenglowski, both deeply interested in the related fields of optics and light, published their findings before those of Becquerel. Henry found that phosphorescent zinc sulphide would affect a photographic plate, while Niewenglowski obtained similar results with phosphorescent calcium sulphide. Neither of these studies was in any way connected with radioactivity, and Becquerel, four years later, noted that Henry's work had not been reproducible and Niewenglowski's was still unexplained. The possibility that the black paper used to wrap the photographic plates was not light-tight has been suggested to account for these questionable results (Henry, 1896; Niewenglowski, 1896; Becquerel, 1900, esp. 47-50).

Of the several Parisian physicists who pursued Poincaré's suggestion, Henri Becquerel was best prepared to examine the matter thoroughly. Both his father, Alexandre Edmond Becquerel, and his grandfather, Antoine César Becquerel, had preceded him in the chair of physics at the Museum of Natural History, and both in their day had been distinguished members of the profession. (Henri's son Jean was the fourth in line to hold this chair.) In particular, Henri's grandfather and especially his father had investigated the phenomena of luminescence: fluorescence, the emission of light only while the source is stimulated, and phosphorescence, a glow that persists after the external stimulation ceases. Sunlight, including the ultraviolet part of its spectrum, was the most common stimulus, and numerous minerals were known to glow in a variety of colors and for different periods. The phenomena are so attractive that many natural history and science museums still display such exhibits.

Henri's good fortune was that he inherited his father's laboratory and its fine collection of minerals and was himself familiar with uranium minerals for at least a decade. Science had become more and more specialized throughout the nineteenth century, and Becquerel by chance now found himself in the right position. Almost anyone could produce the novel X-ray, since vacuum tubes, banks of batteries, interrupters, and Ruhmkorff or other coils to convert direct current to high-voltage alternating current, and photographic plates to record the impressions were ubiquitous in physics laboratories. Assortments of crystals that reacted to light, however, were far less likely to be found in these laboratories toward the end of the nineteenth century (Badash, 1965b).

INITIAL INVESTIGATIONS

Becquerel returned to the Muséum, which incorporates the famous botanical gardens and zoo in Paris, and by the next day had begun his famous series of experiments (Becquerel, 1903: 3). He wrapped a photographic plate in light-tight black paper, positioned on the wrapping various minerals that were known to glow when excited by sunlight, and placed these arrangements on the window sill of his second-floor laboratory. Because phosphorescence lasted only a fraction of a second after the stimulus was removed, he planned to bathe his crystals in sunlight for extended periods. After some hours exposure he developed the plates and, *voila!* smudges appeared beneath the spots where certain rocks had sat. Poincaré's hypothesis that luminescent bodies emitted a penetrating radiation (it had to pass through the black paper and affect the photographic emulsion) appeared to be correct (Becquerel, 1896a).

In more detail, Becquerel found several phosphorescent materials to be quite active, but never identified them, other than the uranium salts: the double sulphate of uranium and potassium (potassium uranyl sulphate). This thin transparent crust of crystals, originally prepared some 15 years earlier, had been loaned for photographic experiments to Gabriel Lippmann (who became Marie Curie's doctoral adviser and who won the Nobel Prize in Physics in 1908). As soon as this mineral could be returned to an impatient Becquerel, he tested it (Becquerel, 1903: 8). Recognizing the need for long exposures, he wrapped his Lumière gelatine-bromide photographic plate in two sheets of thick black paper so that a full day in the sun would not fog it; with the uranium salt in place it turned black after several hours. A coin and a sheet of metal cut into a design placed between the plate and the layer of salts left their silhouettes on the plate. To avoid chemical reactions on the emulsion from vapors emitted by the solar-heated crystals, he inserted a thin slip of glass beneath them, with satisfactory results (Becquerel, 1896a).

One week after the February 24, 1896, announcement of his success, Becquerel presented another paper to the academy. He had performed absorption experiments and reported that the rays from potassium uranyl sulphate could pass through sheets of aluminum and copper, although they were somewhat weakened. A piece of copper cut into the shape of a cross left a clear silhouette, but it was apparent that some radiation had passed through the metal.

In an effort to determine the nature of the light required to excite the phosphorescence, Becquerel learned that it could be stimulated by diffuse daylight as well as by the sun. He even tested to see if the necessary phosphorescence could be provoked indirectly by the sun's rays. For this purpose he reflected sunlight by a metal mirror and refracted it through a quartz prism and lens before it struck the salt. The photographic plate was exposed strongly.

However, the choicest piece of information in this second paper, whose importance was clearly recognized by Becquerel, was the observation that *no* immediately prior exposure to light was required for good results. He had prepared a few experimental arrays on February 26 and 27, but since the sun shone only intermittently, he placed the photographic frames in the darkness of a drawer, leaving the uranium crystal layers in position. The sun, uncooperative, remained obscured the next two days. When it presumably shone again, on March 1, Becquerel chose to replace the photographic plates with fresh ones, and to develop one of the old plates. To his amazement, he found silhouettes of great intensity.

Perceptively, Becquerel saw that the next step was to determine if his crystals required *any* stimulation by light to emit the penetrating radiation. Poincaré's hypothesis remained for him a valid guideline, and he joined the majority of physicists toward the end of the century whose belief system regarded all rays as some form of electromagnetic waves (with the exception of cathode rays, which many thought were particles). Working in his darkroom, Becquerel placed directly on the emulsion of a photographic plate a slightly convex layer of uranium salt, *which touched the plate at only a few points*, and a similar layer with a glass slip interposed. This was positioned within two opaque cardboard boxes and then in a drawer. On another plate holder closed by a sheet of aluminum, he placed another uranium salt layer, put that into a cardboard box, and that too into a drawer. When he developed the plates five hours later, he found in all cases the black shape of the layer of crystals, with any differences in intensity accounted for by the aluminum, glass, or the convex shape. Seemingly, visible phosphorescence was not connected to this perplexing radiation (Becquerel, 1896b).

The question that naturally arises is why did Becquerel exchange the old for new plates and develop that historic emulsion? The only comment he made was that he wished to see the weak images. Thus, it is doubtful that he was concerned with the uniformity or stability of his photographic plates. They had simply been exposed to the overcast skies or the diffuse light of the laboratory for a short time, before being placed in the drawer.[3] Certainly, if the plates were exposed somewhat, it would have been reasonable to replace them with fresh ones before resuming such qualitative experiments. In addition, it seems to have been customary at the time to develop all plates before discarding them.[4]

But there is more to this story than the logic of common laboratory procedure just expressed. The first of March was a Sunday, and one wonders why Becquerel was in his darkroom on the weekend. Further, meteorological records show that for every hourly reading between 0400 and 2100, the cloud coverage in Paris on that date was recorded as "ten tenths" (*Annales du Bureau Central Météorologique de France*, 1896). Becquerel, therefore, was not preparing a new experiment and discarding the old one.

Most likely, he felt that, if he could convey little new information at the Monday meeting of the academy, he could at least provide a progress report, describing the expected weak exposures from proportionally weak stimulation of the crystals as something of a control experiment. That Becquerel found the plates as blackened as they would have been had the crystals phosphoresced continuously, and that he recognized the significance of this lack of visible illumination, marks this step in the discovery of radioactivity not simply as a happy accident but the product of genuine scientific genius. It is comforting, however, to note that his genius emerged because he mistakenly believed in a connection between these rays and phosphorescence, and because he felt compelled to speak at the academy meeting. Folly and pride sometimes *are* rewarded (Badash, 1966a; see also Badash, 1996).

Though a major step, this event does not deserve to be called the discovery of radioactivity. The discovery was a process, not an instantaneous occurrence, for even at this point Becquerel had not sufficiently localized the phenomenon. No doubt Becquerel was a skilled and ingenious experimenter. However, in this early research he was not suitably meticulous to exclude extraneous influences and to see that some of his experimental results could bear more than one explanation. Thus, he often concluded that his experiments proved uranium rays to possess a certain physical property, only to have it shown later that the effect was due to another cause. Indeed, his investigations are particularly interesting for their many false trails, unreproducible results, and misinterpreted effects. Yet, his erroneous conclusions inexorably led him to further experiments, which often revealed the true nature of the phenomenon. This uneven progress is perhaps the most striking facet in the story of the discovery of radioactivity. But it must be understood that few scientists are able to avoid false trails to the extent of a Marie Curie or an Ernest Rutherford; Becquerel was typical of scientists, not atypical.

SUSTAINED INVESTIGATIONS

Adhering to the norm of rapid publication, at the March 9, 1896, session of the academy, Becquerel submitted his third consecutive weekly report. Since the phenomenon appeared similar to X-rays, he pursued a line of experimentation analogous to that which identified many of the physical properties of Röntgen rays. This does not imply that Becquerel was unimaginative, but that Röntgen had most thoroughly explored his discovery.

Simply by substituting a layer of uranium salts for the cathode ray tube, he made the separated gold leaves of an electroscope fall, thereby showing that this new radiation could discharge electrified bodies. Becquerel's experimental conditions left no room for doubt, since the instrument was shielded from outside electrical effects by a metal cage and from ultraviolet radiation by yellow glass. His use of an electroscope is somewhat unexpected and indicates a degree of unfamiliarity with electrical measurements; during the first two decades in the history of radioactivity, one could almost tell the physicists from the chemists by whether they used electrometers or electroscopes. Becquerel later used an electrometer, but he seems still to have preferred his simpler device, which was actually satisfactory for his decidedly qualitative results.

Having established this electrical property, Becquerel next investigated the reflection and refraction of uranium rays. When a steel mirror was placed face down on half of a layer of uranium salts, the photographic plate beneath that half was blackened considerably more than the other half, seemingly confirming reflection. Refraction, by phosphorescent powders contained in tubes placed vertically on the plate, is more confusing, because Becquerel apparently used no uranium compounds and could not repeat the experiment. It is enough to say that within a few years both Ernest Rutherford and Marie Curie argued that uranium rays are neither refracted nor polarized. As for reflection, the darkening was caused by the emission of secondary electrons from the mirror, an understanding that came in a few years (Becquerel, 1896c, 1902; Rutherford, 1899; Curie, 1899).

Becquerel tested also for variety and longevity. Several different uranium compounds, whether initially exposed to light or kept entirely in darkness, produced clear images on the photographic plates. In particular, the intensity of emission from a substance kept in darkness for over 160 hours did not decrease appreciably. A final experiment described on March 9 showed that not air molecules but merely distance attenuated uranium rays. Becquerel placed potassium uranyl sulphate salts at 0.0, 0.2, 1.0, and 3.0 millimeters from the emulsions of two photographic plates, both kept in darkness, but one in air and the other in a vacuum. He found no great difference in the images between air and vacuum, but both those at 3 millimeters were much weaker than those of 0-1 millimeter distance. He did not speculate about possible ranges of the rays, no doubt because he still was thinking in terms of electromagnetic radiation and not particles. His earlier absorption experiments through pieces of metal, showing the attenuation of the rays, could have been compared to the diminution of the intensity of light with distance (Becquerel, 1896c).

Becquerel presented no report at the academy's session on March 16, but he was armed with new information the following week. In a relatively rare quantitative experiment, he compared the rate of fall of the gold leaves of an electroscope with, and without, a plate of quartz in the beam. He did this for both a layer of potassium uranyl sulphate and X-rays from a Crookes tube. Since he could not compare the intensity of the two sources, the ratio of absorption for each provided more of an absolute figure. In this case, the ratios for the two types of rays were of the same order of magnitude, their difference in absorption probably, he explained, due to their different wave lengths.

Convinced that the emission of invisible radiations was a phenomenon of phosphorescence, Becquerel felt that he should be able to show excitation by specific, visible radiations. Theory led his experimentation, and expectations led his conclusions—made easier by eyeball qualitative evaluations. By now, the uranium salt had been in a dark box for more than 15 days, still able to expose a photographic plate strongly. A similar salt, just exposed to the sun, produced a silhouette a little darker, he thought. When the salt was stimulated by magnesium light, the difference was, surprisingly, imperceptible. But stimulation by an electric arc and by the discharge of a Leyden jar (a condenser) produced much darker effects. On balance, increased exposure to light of various types seemed to strengthen the uranium rays. Yet, Becquerel was disturbed that he was unable definitively to connect his invisible phosphorescence from uranium salts with visible phosphorescence or fluorescence.

More puzzling was his recognition that uranous sulphate was as active as uranic sulphate. The problem here was that the uranous salt is neither phosphorescent nor fluorescent, in violation of his premise. To pursue this oddity further, Becquerel melted some uranium nitrate, which was known to lose all ability to phosphoresce or fluoresce when in its water of crystallization. He performed this clever experiment in the dark, shielding the tube even from the light of the alcohol lamp used to heat the mineral, and then allowed the solution to recrystallize in darkness. The new crystal should have been energy-free and unable to phosphoresce, and yet it exposed a photographic plate as strongly as before its changes of state (Becquerel, 1896d).

Determined still to keep in the limelight, Becquerel presented his fifth paper concerning uranium rays at the academy's session on March 30. Polarization was proved, he thought, when the photographic image under two tourmaline layers with their axes parallel was darker than the image under the same layers with their axes crossed. Here was a difference between uranium rays and X-rays, for the latter produced equally blackened images. As mentioned earlier, Rutherford could not repeat this experiment; apparently, neither could Becquerel, for he never referred to it again.

Besides reflection, refraction, and polarization, Becquerel sought other differences between uranium rays and X-rays. Absorption experiments showed him that while uranium rays could traverse most bodies, and particularly metals, X-rays could not. Of course, the attenuation was proportional to thickness. Yet, the combined effect of two screens was less than the sum of their individual effects. The logical and correct interpretation, applied by Becquerel, was that the radiation was not homogeneous.

Recalling that he had previously found strong activity in a uranium nitrate crystal that was melted and regrown in darkness, Becquerel now reported continuing activity when the crystal was dissolved. This solution was the second nonphosphorescent substance to emit rays that he had found (the uranous series of salts was the other), and might have helped to plant seeds of doubt concerning the connection with phosphorescence. These seeds, however, fell onto sterile ground (Becquerel, 1896e).

Seven weeks now passed before Becquerel again presented a paper on his discovery, which he modestly called *uranium rays*, while his nationalistic colleagues preferred to term them *Becquerel rays*. (Similarly, Röntgen used the term *X*-rays, the *X* meaning unknown, while others named them after him.) At the session on May 18, Becquerel reported that the radiation from his source kept in darkness had not sensibly decreased over the course of two months, and that recently he had transferred the source to a double lead box kept in his darkroom. This would afford further protection from incident stimulating rays.

More important than this progress report, however, was his announcement that uranium element, not the compound, was the source of the emission. Since all uranium salts gave similar results—whether phosphorescent or not, and whether crystalline, melted, or dissolved—he had been led to the thought that the activity might reside in the element alone. He quickly confirmed this with commercial uranium powder, which he had long had in his laboratory, and then with a piece of the extremely rare uranium metal, which he obtained from Henri Moissan. Both photographic and electroscopic tests showed the metal to be the most active by far.

Acquisition and use of this metal may explain the seven-week delay in Becquerel's reports to the academy. Moissan, famous for his electric furnace, may have required this time to prepare the specimen. There may also have been some question about who should examine the metal's properties, for this was a subject of interest to both men. Since a paper by Moissan, entitled "Préparation et propriétés de l'uranium," follows Becquerel's article in the *Comptes Rendus*, it is possible that the latter was obliged to delay publication until the two reports could appear together.

Since we know the current explanation of the phenomenon that came to be called *radioactivity*, it is all too easy to criticize Becquerel for his stubborn and erroneous single-mindedness. Yet, he tried hard to stay within the paradigm, to explain new phenomena with familiar concepts. In this he was unexceptional. Convinced still that he was dealing with some form of electromagnetic radiation, he concluded that uranium was probably the first example of a metal having a property of the order of an invisible phosphorescence (Becquerel, 1896f).[5]

When was radioactivity discovered? Should we select the first paper in which uranium salts were mentioned? Or the realization that stimulation by sunlight was unnecessary? Or when a nonphosphorescent body was found to be active? Or when only uranium compounds were seen to emit the rays? Or when the action was localized in the metal? Even the last is debatable, for Becquerel still thought of the phenomenon as an invisible phosphorescence. Ultimately, of course, the designation of a discovery date is a matter of definition, and therefore choice. A more satisfactory approach is to see that discovery, as in this instance, is often a process, and need not be limited in time to an event. Moreover, the process can involve testing for many different properties, with some probes into the unknown confirming expectations and others revealing surprises, and with all probes additionally subject to experimental or theoretical error.

This series of a half-dozen papers, extending from February to May 1896, established these important facts among several others: uranium emits rays, the phenomenon is an atomic property of uranium (even if Becquerel did not say so explicitly, we may assume this since he traced the activity to the element), and the metal's activity is about three and a half times stronger than that of the potassium uranyl sulphate. His one remaining paper published in 1896 merely noted that the salts placed in the double lead box on May 3 were still as active in November, and that uranium rays were able to make a gas electrically conducting.

As mentioned earlier, many of Becquerel's experiments were suggested by those used to determine the properties of X-rays, and this latter was no exception. In this case, J. J. Thomson and Ernest Rutherford had shown that X-rays affect a gas, allowing electrified bodies to discharge through it; the phenomenon was named ionization (Thomson and Rutherford, 1896). Becquerel had already learned that the emission from uranium could discharge a body at a distance, and now he passed currents of air and carbon dioxide over the salts and metal and found that the gold leaves of an electroscope fell rapidly. He used both cotton plugs and wrappings of black paper to trap any floating particles, yet obtained positive results. This recognition of the ability of these rays to ionize air was an important contribution (Becquerel, 1896g).

The final two papers in this group of pioneering studies were published in 1897 and were mainly electrical in nature. Becquerel showed that uranium rays would discharge bodies at any voltage and that the charge placed on a piece of uranium was dissipated quickly; he also performed other tests with his electroscope (Becquerel, 1897a, 1897b). True, he was still presenting data on the physical properties of the rays, but the experiments were of the nature of squeezing out every last bit of information and not of developing fresh areas. An impasse thus seems to have been reached. The radiation was an atomic phenomenon, and this was of fundamental significance. Yet, Becquerel apparently was unable to capitalize on this insight and did not even speculate on the mechanism involved (other than allying it to phosphorescence). He had, further, found the rays to be heterogeneous but dropped that trail too. Perhaps, if X-rays had exhibited striking characteristics in a magnetic field, he would have tried such a field on uranium rays and determined some of the components years sooner. Becquerel had run out of ideas.

It is, nonetheless, surprising that he said nothing about attempts to deviate these rays magnetically. Cathode rays were well known to be deflected in a magnetic field and, though unsuccessful, Röntgen had placed such a field on X-rays. Perhaps Becquerel did try the experiment but failed to get a positive result because his source was too weak and uncollimated, and he chose not to mention a negative finding.[6] More likely, he reasoned that the test would after all make sense only if the rays contained charged particles, and he was focused on an electromagnetic explanation.[7] (Uranium rays were magnetically deflected in 1899 by Friedrich Giesel, a chemist at a quinine factory in Braunschweig, Germany, who discovered radium independently and who seems to have produced radioactive materials as something of a hobby. This brought Becquerel back to the subject, and he confirmed magnetic bending.) (Giesel, 1899; Becquerel, 1899. See also Badash, 1966c).

OTHERS ENTER THE FIELD

Silvanus Thompson, professor of physics at the City and Guilds Technical College, Finsbury, London, simultaneously and independently observed the strange action of uranium rays. He was among the many scientists worldwide who, in January 1896, repeated Röntgen's newly announced experiments. Like a number of others, he conceived the idea of placing fluorescent substances in contact with a photographic plate in order to shorten the long exposures then needed to obtain X-ray shadow pictures. The fluorescence stimulated by the X-rays was expected to hasten the action in the emulsion. In practice, this did not succeed well, but Thompson designed a curved cathode, which, by focusing the cathode rays, made the X-ray beam more intense.

A paper by Charles Henry in Paris, mentioned earlier, led Thompson to examine luminous materials not for their enhancement of X-ray images, but for the emission of invisible radiation. He therefore placed a variety of them on a sheet of aluminum under which was a photographic plate. With remarkable similarity to Becquerel's approach, Thompson left the arrangement "for several days upon the sill of a window facing south to receive so much sunlight (several hours as it happened) as penetrates in February into a back street in the heart of London." Upon development, he noted definite photographic action had occurred only beneath the uranium nitrate and uranium ammonium fluoride, and recognized this as significant.

Thompson informed Cambridge mathematical physicist Sir George Gabriel Stokes, a former president of the Royal Society, who was excited by the news, but who told him a week later that he had been anticipated by Becquerel. Having lost priority, Thompson gave no thought to rapid publication, and his paper did not appear until July

1896, when he termed the phenomenon *hyperphosphorescence*. He did not recognize from his own work that uranium rays had nothing to do with luminescence, nor did he localize the properties in the metal (even though he was able to obtain a sample of the metal in London). He was in fact far more interested in X-rays, and when he paid his customary visit to the Société de Physique's Easter meeting in Paris, he notably had no encounter with Becquerel.[8]

The first original research performed after the initial experiments of Becquerel and Thompson was described before the Physical Society of Berlin on October 23, 1896. This in itself is a measure of the indifference with which the discovery was received, for it was more than half a year before others saw fit to investigate the phenomenon. P. Spies then reported that fluorspar, placed between uranium and a piece of bromide paper, caused the paper to be much more darkened than it would ordinarily be. While this effect was perhaps due to some chemical action or secondary emission, it is interesting to see that efforts to concentrate uranium rays proceeded similarly to those for X-rays. The following year, F. Maack put forth the claim that colophony or resin possessed the ability to concentrate these rays (Spies, 1896: 101; Maack, 1897: 366).

The next research contribution came, oddly enough, from the United States. This was not to be expected in the context of nineteenth-century physics, for this country usually lagged far behind the European developments. Yet, A. F. McKissick, professor of electrical engineering at the Alabama Polytechnic Institute (now Auburn University), tested "all such [luminescent] substances that are known as available," hoping to discover other sources of Becquerel rays.

Nor did he stop with known luminescent materials; among a variety of substances, he claimed that granulated sugar was most active. Except for the uranium that he tested, McKissick was completely wrong. The images on his photographic plates were more likely the result of chemical action than penetrating radiation. Still, he is worthy of attention, since he was typical of the casual experimenters who added greatly to the period's confusing array of emitters and emissions (McKissick, 1896: 652; 1897a: 313; 1897b: 17542).

Hanichi Muraoka, who received his training in Germany and was then a professor in the Physical Institute of the Daisan Kotogakko in Kyoto, Japan, also was inspired by the discovery of Becquerel rays in phosphorescent uranium salts. Could not glow worms, which abounded in summertime Japan, also be a source of something similar to X-rays? He found not Becquerel rays, however, but something new. The glow worms produced a photographic impression through cardboard, but not where the cardboard was cut away. Muraoka concluded that cardboard exerted a "suction effect" for his new rays, analogous to the permeability of iron for lines of force. There were others who examined glow worms and fireflies, and also detected types of radiation, but they, fortunately, did not compare them so closely to uranium rays. This line of research was discredited a year later when Muraoka and M. Kasuya published the results of more careful experiments on the next generation of the insects. The photographic effect was due, at least in part, to vapors coming from the glow worms. It had been necessary to keep them moist. Damp brown paper gave the same effect (Muraoka, 1896; Muraoka and Kasuya, 1898).

On March 1 and April 4, 1897, the best executed and most serious investigations of uranium rays in this period of 1896-1897, by scientists other than Becquerel, were described before the Royal Society of Edinburgh. The authors were the illustrious Lord Kelvin, John Carruthers Beattie, and Maryan Smoluchowski de Smolan, the latter two research fellows at the University of Glasgow. Using a quadrant electrometer, they measured the potential difference between an insulated disk of uranium metal (obtained from Moissan) and disks of other metals. They found that each metal acquired a distinct voltage when placed across the air gap from the uranium. Further, any charge given to the metal dissipated quickly and its peculiar voltage was reassumed.

They next performed quantitative absorption experiments with screens of various materials and thicknesses. Then the size of the air gap was varied as well as the pressure and the type of gas. From these last tests came the most significant information: the leakage (a measure of the rate of activity) increased with the applied voltage up to a point, where it effectively leveled off. The discharge of electricity by uranium was not proportional to voltage.[9] Throughout these papers the data are clearly presented. Yet, nowhere are they explained, not even where it might be hoped that the authors would recognize a saturation current. One is left wondering what enables the conduction to occur, for there is no mention of ions, even though J. J. Thomson and Rutherford had almost half a year earlier shown that X-rays produce ions and the current can become saturated. It was, in fact, left to Rutherford to state explicitly that uranium rays produce ions in the gas (Kelvin, Beattie, and Smoluchowski, 1897a, 1897b; Thomson and Rutherford, 1896; Rutherford, 1897).

Subsequently, Beattie alone presented a paper to the Royal Society of Edinburgh in which he tested the electrification of the air surrounding uranium when the containing vessel and the uranium were kept at different potentials. In all cases, the air acquired the same type of charge, positive or negative, as the uranium, and its electrification reached a maximum when the uranium was approximately 10 volts. Beattie attempted no explanation for the decrease above 10 volts, but Kelvin did in an appended note. He suggested that the lines of force around the small piece of uranium have a much greater voltage per centimeter than at the surface of the large container. Above 10 volts the rate of discharge of electricity into the air from the uranium ceases to increase much, while that from the container, of opposite charge, is still increasing. Hence, a maximum is reached, and the electrification must then diminish. It was an ingenious idea, showing that the 73-year-old Kelvin had lost none of his inventiveness. But, in the light of modern understanding of the behavior of electricity in gases, the explanation seems to be that, with increased voltage, the samples of air that Beattie drew off from the container were found lacking in charges because the ions formed by the uranium rays had been too quickly drawn to the electrodes (Beattie, 1897).

In the prestigious Wilde Lecture before the Manchester Literary and Philosophical Society entitled "On the Nature of the Röntgen Rays," Sir George Stokes emphasized the refrangibility and polarizability of uranium rays and, with an apparent love of mechanical models that was so common in British science (for example, Maxwell's demon), he explained the emission process:

> What takes place? My conjecture is that the molecule of uranium has a structure which may be roughly compared to a flexible chain with a small weight at the end of it. Suppose you have vibrations communicated to such a chain at the top; they travel gradually to the bottom, and near the bottom produce a disturbance which deviates more from a simple harmonic motion. So, if a vibration is communicated to what I will call the tail of the molecule of uranium, it may give rise to a disturbance in the ether which is not of a regular periodic character. I conceive, then, that you have vibrations produced in the ether, not of such a permanently regular character as would constitute them vibrations of light, and yet not of so simple a character as in the Röntgen rays—something between. And accordingly, there is enough irregularity to allow the ethereal disturbance to pass through black paper, and enough regularity on the other hand to make possible a certain amount of refraction. You can also obtain evidence of the polarization, and, consequently, of the transverse character of these rays.

His knowledge based only on published accounts of research by others, Stokes had little reason to doubt the electromagnetic character of uranium rays. And, like others, he sought to fit an explanation of their behavior into familiar patterns. Note that the ether was still a valid entity for many physicists at the end of the nineteenth century, and that Stokes' preference for *molecule* instead of *atom* probably reflects continuing ambiguity about what to call fundamental particles.[10]

Throughout 1897 there also appeared other one-paragraph contributions to the study of uranium rays contained in longer papers on related topics. In Paris, Gustave Le Bon found that many bodies, particularly shiny metal surfaces, when struck by light possess the property of discharging the gold leaves of an electroscope. He thereby concluded that the properties of uranium are only a case of a very general law (Le Bon, 1897). In an extract from his Erlangen doctoral dissertation, W. Arnold fed the general confusion by declaring that Becquerel rays are emitted from zinc sulphide, mixtures of various sulphides and of tungstates, fluorspar, and even the organic compound retene (Arnold, 1897). From St. Petersburg, Ivan Ivanovitsch Borgman reported that Becquerel rays and Röntgen rays are capable of producing thermo-luminescence in a mixture of calcium sulphate and manganese sulphate, an effect formerly attributed only to Wiedemann's discharge rays. This paper is interesting in that it shows Borgman exposed the double sulphate of uranium and potassium to ultraviolet light before use. Thus, a year after Becquerel indicated that no external stimulation was necessary for the emission of these rays, that property lay unrecognized as relevant (Borgman, 1897).

But there were also those who appeared to be on the right track. While offering no new properties, they at least confirmed Becquerel's work. Adolf Miethe, the scientific director of the Voigtländer optical works, verified that the rays from uranium salts and the metal pass through aluminum foil (Miethe, 1897a: 63; 1897b: 606). At a secondary school in Wolfenbüttel, Germany, the famous team of Julius Elster and Hans Geitel, who collaborated for almost 40 years, including much later work on radioactivity, also verified the physical properties of the rays and the fact that they persisted when the salts were kept in darkness for months. They showed that zinc, aluminum, and other substances that present photoelectric effects do not emit invisible radiations of sufficient intensity to electrify the surrounding air in the manner of uranium. This enabled them to conclude that the source of the radiant energy was still unknown, and that true photoelectric phenomena cannot be attributed to hyperphosphorescence (a rare instance of the use of Silvanus Thompson's term for uranium radiation) (Elster and Geitel, 1897: 455).

The experiments by the Italian E. Villari also bolstered Becquerel's results, but are more interesting to us historically because of the way they came provokingly close to later, extremely significant, discoveries. Using the mineral uraninite, he tested the ability of a current of air to discharge a gold leaf electroscope. Then he placed the sample in a paper envelope and noticed no electrical action. As was learned later by Rutherford and Ernst Dorn, thorium and radium would have exhibited an effect, since they evolve gaseous emanations (the decay products thoron and radon, respectively), whereas uranium does not. Also interesting is his use of uraninite. This variable mixture, consisting mainly of UO_2 and UO_3, is none other than the ore pitchblende, from which the Curies were to extract the new elements polonium and radium. Had Villari performed quantitative work, he might have discovered that uraninite was far more active than accounted for by its uranium content.[11]

The final experiments to be noted in this 1896-1897 period are those of Charles Thomson Rees Wilson, then the Clerk Maxwell Student in the University of Cambridge. Like Rutherford, he was working in the Cavendish Laboratory under J. J. Thomson, and in the process of perfecting his invaluable cloud chamber. Wilson showed that uranium rays would produce nuclei for the condensation of water upon them. Since the nuclei could be swept away by an electric field, they had to be charged particles, or ions. Whether the uranium compounds were placed within or without the expansion apparatus, they produced nuclei that required the same degree of supersaturation in the chamber as did nuclei produced by X-rays. Prophetically, Wilson wrote that "expansion experiments probably furnish one of the most delicate methods of detecting these rays" (Wilson, 1897, 1898: 128).

TRYING TO UNDERSTAND THE PHENOMENON

Throughout these two years, Becquerel was uncertain just how closely his discovery matched that of Röntgen. Their methods of production were different (sunlight versus high voltage), the exposure times were vastly unequal (often hours for uranium rays), the X-rays penetrated dense material more easily, and X-ray pictures were far sharper. The situation was further clouded by the existence of an overabundance of other radiations, some real and some spurious. As the century ended, scientists were trying to discern the properties and correlate such phenomena as Hertzian waves, cathode rays (including para- and dia-cathodic varieties), canal rays, discharge rays, Finsen rays, radiation from hot bodies and from cleaned metal surfaces, and, as mentioned earlier, light from glow worms, fire flies, and even alleged radiation from granulated sugar.

Although Becquerel's papers were widely abstracted, they were given no special prominence. This subservient role is underscored by the difficulty in locating articles on the subject in the indexes of many scientific periodicals. They are to be found not under the headings of *Becquerel rays*, *radiation* (the name *radioactivity* was coined later, by Marie Curie), or even *uranium*, but often under the term *Röntgen rays*. Becquerel's discovery, thus, must be seen as just another small corner of the radiation physics of his day, and neither he nor others had any reason to be more than mildly impressed by it. There was no suggestion that it was a major phenomenon of nature (Badash, 1965a).

Still, Becquerel laid the foundation of this subject and built much of the edifice, despite a number of wrong turns. Unwilling to abandon his basic theory that luminescent bodies emitted these rays, he opted for the not-unreasonable explanation that he had uncovered a new phenomenon of long-lived luminescence. His minerals had been exposed to sunlight at some time in the past, so he reasoned that they retained their ability to luminesce, even if not visibly, and also their ability to emit penetrating radiations. Even when he traced the activity to nonluminescent uranium compounds he found it difficult to break with the idea of luminescence. Uranium metal posed another challenge, but he embraced it with the novel property of metallic phosphorescence. It is a nice example of a scientific idea not only apparently being verified but leading to new discoveries. Becquerel's tenacity in holding to this idea is not surprising, for it was his greatest claim to fame, and there were as yet no other interpretations of uranium rays.

Becquerel should not be criticized overly for departing from his study of uranium rays when he thought the subject was exhausted.

Some other very competent physicists, such as Lord Kelvin, and Elster and Geitel, did likewise. By the end of 1897, radioactivity was something of a dead horse—it was there, but no one knew what to do with it. Then, Gerhard C. Schmidt in Erlangen and Marie Curie in Paris independently reported in early 1898 that thorium exhibited the same sort of activity as uranium and both considered their high atomic weights to be significant (Schmidt, 1898a, 1898b; Curie, 1898; see also Badash, 1966b). Soon, the Curies located the new elements polonium and radium in uranium ore samples, while Rutherford began his domination of radioactivity and then nuclear physics (P. and M. Curie, 1898; P. and M. Curie and Bémont, 1898; Rutherford, 1899). Still other radioelements began to be found at the heavy end of the periodic table and the experimentation invariably became quantitative. Elster and Geitel in 1899 proposed that radioactivity was an atomic phenomenon—the energy lay within the atom. The Curies would have none of this, opting for an undetectable but pervasive ethereal radiation that excited only the heavy elements, causing them to emit their rays as secondary radiations (Elster and Geitel, 1899; P. and M. Curie, 1900, vol. 3: 79-114).

Through the early years of the twentieth century, Becquerel persisted in his belief of metallic phosphorescence and was criticized harshly by the Curies, after which he no longer communicated their papers to the Academy of Sciences. (Pierre was not yet a member and Marie would never be one, so they needed a sponsor.) The puzzle of radioactivity was resolved in a series of papers in 1902 and 1903 by Rutherford, now at McGill University in Montreal, and a young chemistry colleague named Frederick Soddy. Defying more than a century of modern chemistry from which alchemy had been banished, they argued that radioactive atoms spontaneously and continually transmute from one element into another. These atoms are inherently unstable and contain the energy of their own destruction (Rutherford and Soddy, 1902a, 1902b). Fearful of a hostile reception, they avoided the red-flag term *transmutation*, and called the process *transformation* or *disintegration*. But they need not have feared rejection. Their experimental evidence was overpowering, and their explanation of radioactivity faced little opposition (even if the Curies, like Becquerel uncommonly fond of their own idea, still muttered about an ethereal radiation primary) (Badash, 1966d).

CONCLUSION

In the latter part of the nineteenth century, there was an undercurrent of belief that all the great scientific discoveries had been made. There was but one universe, and Isaac Newton had explained its mechanical stability. In a similar fashion, James Clerk Maxwell had united electricity and magnetism, Charles Darwin had resolved the big question in biology, and the law of conservation of energy wrapped up another major area of science. The towering issues had been settled and those who planned a scientific career could look forward only to determining constants to greater accuracy—the next decimal place (Badash, 1972).

Of course, these naysayers were wrong. The discoveries of long electromagnetic waves by Heinrich Hertz, inert gases in the atmosphere by Lord Rayleigh and William Ramsay, X-rays by Röntgen, radioactivity by Becquerel, the electron (a body smaller than an atom) by J. J. Thomson, the liquefaction of hydrogen (one of the so-called permanent gases) by Karol Stanislaw Olszewski, and the quantum explanation of blackbody radiation by Max Planck made it clear that physical science yet had much to ponder.

Revolutionary as each of these major contributions was, most had roots that extended back to traditional science, and could therefore be accepted with some equanimity. Radioactivity, perhaps having the fewest ties with the past, was the most startling. There never had been any hint of such a remarkable phenomenon in which atoms spontaneously disintegrated with an outpouring of energy, nor was there any explanation in terms of existing knowledge that seemed satisfactory.

Yet, this impressive discovery emerged from a background of false trails, unreproducible results, and misinterpreted effects. This seems more like messy normal science than the logical and cleansed picture of research that is sometimes offered in science textbooks and historical writing. To Becquerel's credit, he not only recognized the odd phenomenon, but sought to fashion an explanation for it. When his incorrectly construed experiments led him to believe that uranium rays were refracted and polarized, it was comforting to assert that they were electromagnetic, located in an unfamiliar part of the spectrum. This was all the more reasonable because of the near-universal blunder that associated X-rays with luminescence—the glowing spot on the vacuum tube. Uranium rays too must be some form of light. Thus, uranium rays poorly navigated an obstacle course: they were not only misinterpreted when found, but their discovery was caused by an incorrect assumption.

Nonetheless, Becquerel's big blunder was not his mistaken experiments or his incorrect theory. His error was that he failed to think through the localization of rays in the element uranium. While he had looked for penetrating rays in a variety of minerals in his very first experiments, and thereby had tested a number of elements, this could hardly be called a systematic survey of the periodic table. A man so well connected that he could obtain a rare sample of metallic uranium from Moissan, and who loaned some of his own uncommon uranium mineral crystals to Lippmann, could surely have borrowed samples of many elements from leading Parisian chemists. But he was not moved to look for other sources of the radiation because he felt that his own novel theory of metallic phosphorescence adequately explained the phenomenon, which surely was located in some remote part of the electromagnetic spectrum—and it was not terribly interesting after all.[12]

Becquerel was capable of the experimental tests called for in the early investigations of the rays he discovered; the necessary craft skills were commonplace. He was also capable of avoiding some of his laboratory missteps, for corrections involved no new techniques. He failed in the intellectual challenge to rethink his theory and search for other active elements. He was not motivated even to look for other metals that might possess long-lived metallic phosphorescence. Nonetheless, by the definition offered as this story began, Becquerel's idea should be termed successful. He introduced order as he went along, methodically incorporating disparate and sometimes unnerving observations into an expanding theory. He explained the existing data and followed the trail to new research questions, even if the next breakthrough was made by Gerhard Schmidt and Marie Curie. Success, as has been argued, need not be congruent with a later consensus of truth.

Was there any consequence of Becquerel's false trails and his unpursued leads? At most, perhaps a year or two were lost. Once the Curies showed that radioactivity was an atomic phenomenon, with quantitative ionization tests as the key to measurement of activity, the subject had a life of its own, distinct from that of X-rays. Moreover, even if the Curies never pursued radioactivity, Ernest Rutherford's examination of the ionization caused by uranium rays would have reopened the field of study. Development of the science of radioactivity thus was not dependent upon the whims of one individual. Further, since major applications of this science were decades in the future, that short delay had little effect on history. The one haunting counterfactual thought is that, if all following research were moved up by two years, nuclear fission might have been discovered in time for Germany to build an atomic bomb during World War II.

When the Nobel Prize was first awarded in 1901, the physics prize was given to Röntgen for his discovery of X-rays. Two years later, Becquerel shared the physics prize with Pierre and Marie Curie, and in 1908 Rutherford won the chemistry prize. With the large cash award, the Nobels were instantly prominent. While the discoverer of X-rays was inevitably a winner, the early prizes for radioactivity suggest that its importance was recognized quickly, once Rutherford and Soddy showed that atoms were unstable.

NOTES

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1. For a translation into English, see Röntgen (1896). See also Glasser (1934, chap. 23).
2. The *Comptes Rendus* for this date contains only a three-line notice of the communication of a photograph by Mm. les Drs Oudin et Barthélemy, with no indication of any text, nor that Poincaré was involved. We

learn these things from Poincaré (1896: 53). We also learn from this article that Poincaré was one of the several scientists to whom Röntgen personally sent X-ray photographs. These were shown at the academy, along with the radiographs by Oudin and Barthélemy.

3. For an affirmation of this view, see the address by Jean Becquerel in *Conférences Prononcées à l'Occasion du Cinquantième Anniversaire de la Découverte de la Radioactivité* (1946): 5.
4. Information in a letter to the author from Yves Le Grand, professor of physics at the Museum of Natural History, Paris, Jan. 17, 1964.
5. For several of Becquerel's papers translated into English, with excellent commentary, see Romer (1964). See also Romer (1960).
6. Successful collimation was not beyond Becquerel's means, however. Rutherford (1899) described his technique of cutting a deep slit through a lead block to obtain a narrow beam from uranium oxide placed underneath.
7. The Zeeman effect, also discovered in 1896, involved a magnetic impression on spectral lines and would not likely have been considered an analogous test.
8. S. P. Thompson (1896: 104). See also Thompson and Thompson (1920). Thompson's letter to Stokes, dated February 28, 1896, is not reproduced in this volume, and curiously contains no mention of uranium compounds. The letter does appear in Larmor (1907, vol. 2: 495).
9. Just eight days later, Becquerel presented a paper to the academy that contained similar findings, and also a formula for the leakage-voltage relationship. See Becquerel (1897b).
10. Stokes (1897: 23-24). The lecture was delivered on July 2, 1897.
11. Villari (1897, 1898). The mineral uraninite, if untreated, would have contained its complement of decay products, including entrained radium emanation, but little would have escaped during the course of his experiment.
12. Interesting studies of error have been produced from sociological and philosophical perspectives. A random sampling includes Cadeddu (2000); Goran (1975); Mulkay and Gilbert (1982); Latour and Woolgar (1986).

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