Radioactivity's two early puzzles

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Since the discovery of radioactivity predates the birth of quantum mechanics by nearly thirty years, it was inevitable that the most fundamental aspect of radioactive processes, its spontaneity, was also the most baffling one to physicists during the first quarter of the twentieth century. The ensuing struggles in that period with regard to the origins of the radioactive energy release, and to the significance of the life time concept are discussed.

CONTENTS

I. PROLOGUE

In the course of a recent discussion with an experimentalist colleague about a subtle weak-interaction experiment he planned to perform, the question came up as to how long this effort would take. Such actuarial questions are inevitable these days, not only because of the high technical and logistic demands of modern experimentation, especially in high-energy physics, but also because an experimental proposal must first be scrutinized by a committee of peers and, even when accepted, there remain the dictates of beam availability for its execution. As we went on to reflect on these factors which influence the pace of discovery, one of his younger colleagues who was present wondered out loud about the days of yore when physics was done by small groups, if not by individuals, when the space needed for an experiment was still table-top-sized, and when the time needed could often be counted in weeks if not less. This led one of us to remind him that it took more than thirty years from the discovery of β radioactivity to the postulation of the neutrino. He was astonished and asked: what did people do in between? It is the purpose of this paper to give part of the answer to this question. The main theme of the present paper will be stated more precisely in Section III.

I am sure that many of us particle physicists have often wondered about the factors which determined the pace of past discovery in our field, which is still in such a rapid state of flux. Through the years I have often inquired of senior colleagues about their reminiscences in this regard. Such is the speed of development of twentieth-century physics in general, and of particle physics in particular, that memories of past events tend to blur even in the clearest of minds. It has led me to a growing conviction that it will serve a purpose to record systematically the history of particle physics from its inception, the days of Roentgen, Becquerel, J. J. Thomson, the Curies, and Rutherford. The present paper is a small fragment of a fuller projected account. It is respectfully dedicated to my questioner whom I just mentioned and to all others who occasionally wonder about the roots of this discipline.

Particle physics has already produced many surprises. There are good reasons to expect that there is much more to come. Obviously these reasons have nothing to do with the history of the subject, if only because in this kind of history, wonderfully exciting and totally useless as it is, nothing repeats itself beyond two things: first, plus ça change, plus ça change and, secondly, the ever recurring necessity to forget much of the past. For this last reason, one historical extrapolation can safely be made. There will be a time in the twenty-first century when some older particle physicists will sit together and pontificate about events past to them, present and future to us. A younger colleague will listen and ask in astonishment: what did people do in between?

II. SOME INTRODUCTORY CHRONOLOGY

The question raised in Sec. I can be stated more specifically as follows: What did people do between March 1, 1896 and December 4, 1930? As a prelude to a precise statement of the scope of this paper let us first consider a brief chronology of some events directly relevant to this subject.

(1) "Le 1er Mars 1896, Henri Becquerel a découvert la radioactivité."[1] Thus begin the recollections of Jean Becquerel (1878–1953) of the observation, made in Paris by his father Antoine Henri Becquerel (1852-1908) on a cloudy Sunday, that $K_2UO_2(SO_4)_2 \cdot 2H_2O$ emits radiation of a kind never detected before (H. Becquerel, 1896). Henri Becquerel did not name his new phenomenon radioactivity—that term was first coined by the Curies and G. Bémont in 1898. (It entered the English literature for the first time in the November 16, 1898 issue of *Nature*.) Becquerel was unaware that this was the first observa-

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[1] On the first of March 1896, H. B. discovered radioactivity (J. Becquerel, 1924).

tion ever of a nuclear process because even the existence of the nucleus was not as yet known. Nor could he realize at once that the bulk of the radiation he had observed consisted of electrons—because neither was the existence of the electron yet known. It was clear to him, however, that he had made a startling discovery. His further experiments performed during that year demonstrated that the new radiation was an intrinsic property of uranium. He called the new phenomenon uranic rays.

(2). Next follows the discovery of the electron. In a series of papers, the first of which was dated Oct. 31, 1896, Pieter Zeeman (1865–1943) reported on the effect since named after him (Zeeman, 1896a). With a helping hand from Hendrik Antoon Lorentz (1865–1928) he could interpret his effect as being due to the motion of an "ion" within the atom, with an $e/m \sim 10^3$ times the corresponding quantity for ionized hydrogen. This result is found in his second paper (Zeeman, 1896b),[2] Nov. 28, 1896, in which, curiously, no surprise is expressed at this large value of e/m for the "ion". There is only the laconic comment: "Natuurlijk kan deze uitkomst van uit de theorie slechts als een eerste benadering worden beschouwd."[3]

Six weeks later we find the first statement ever to occur in print of the existence of a subatomic particle. It was made by Johann Emil Wiechert (1861–1928), on Jan. 7, 1897, in a lecture with demonstrations before the Physical Economical Society of Königsberg in East Prussia. In a discussion of his recent experiments on cathode rays, he noted that these "showed that we are not dealing with the atoms known from chemistry, because the mass of the moving particles turned out to be 2000–4000 times smaller than the one of hydrogen atoms, the lightest of the known chemical atoms" (Wiechert, 1897).[4]

The second, independent announcement also came in a lecture, this one by Joseph John Thomson (1856–1940), on April 30, 1897 before the Royal Institution in London. While Wiechert's method could only set upper and lower bounds on the e/m for cathode rays, Thomson had found a way to determine this quantity precisely. In the lecture he reported his preliminary results which led him to the "somewhat startling" conclusion of "a state of matter more finely subdivided than the atom" (reprinted in Thomson, 1970). Both Wiechert's and Thomson's conclusions rested on the assumption that each cathode-ray particle carries one fundamental unit of charge (the latter quantity being determined from electrolysis). Work on the charge determination started right away, and the correctness of the assumption was soon confirmed.

The combined outcome of the experiments by Zeeman and by Thomson [who was aware (Thomson, 1970) of Zeeman's result on e/m] was rapidly realized to be: electrons are universal atomic constituents. Their motions are responsible for atomic spectra. But this view did not at once receive wide acceptance. Referring to his Royal Institution lecture, Thomson has recalled that "I was ... told long afterwards by a distinguished physicist who had been present at my lecture that he thought I had been 'pulling their legs'" (Thomson, 1936).

Thus at the end of 1897 there were some who knew of radioactivity and of the existence of at least one subatomic particle. But no one knew as yet that radioactive rays *consist* of subatomic particles.

(3) In 1898, Marie Curie (1867–1934) announced, in the second paper she ever published (S. Curie, 1898) that the emission of uranic rays is an atomic property, that is, the intensity of the rays is proportional to the amount of uranium present. In the same paper she reported the discovery that thorium compounds exhibit activities similar to those of uranium, a result which, unbeknownst to her, had been obtained somewhat earlier by Gerhard Carl Nathaniel Schmidt (1865–1949) in Erlangen (Schmidt, 1898). (In that same year, she and Pierre Curie also discovered polonium and, again in 1898, the two of them together with G. Bémont discovered radium. Incidentally, Marie Curie received her Ph.D. in 1903 with the mention "très honorable".)

(4) Also in that same year, 1898, Ernest Rutherford (1871–1937) found that the "Becquerel rays" (as they were now called) emitted by uranium are "complex, and that there are at present at least two distinct types of radiation—one that is very readily absorbed, which will be termed for convenience the α radiation, and the other of a more penetrative character, which will be called the β radiation" (Rutherford, 1899).

[Investigations beginning in 1899 soon established that β rays are electrons (as defined from the cathode rays). It took ten years before it was incontrovertibly established that α rays consist of doubly ionized helium atoms.[5] γ rays were first observed in 1900 (Villard, 1900). It took fourteen years before it was established beyond doubt that these are high-frequency electromagnetic radiations (Rutherford and Andrade, 1914).]

These few data may suffice to set the stage for the developments to be described in the subsequent sections. Let us also recall that the discovery of the nucleus by Rutherford dates from 1911 (Rutherford, 1911a,b), that the first one to state that β radioactivity is a nuclear process was Niels Bohr (1885–1962), in 1913, (Bohr, 1913), and that the first one to observe the continuous nature of the primary β spectrum was James Chadwick (1891–1974), in 1914, (Chadwick 1914). To conclude this short chronology, I shall note a few events related to the other date: December 4, 1930, mentioned at the beginning of this section.

Exactly half a century ago, a paper by Charles Drummond Ellis (1895–) and William Alfred Wooster

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[2] Both papers were published in English as a single article in early 1897 (Zeeman, 1897).

[3] Of course this result from the theory is only to be considered as a first approximation.

[4] The original reads: "Sie ergab, dass wir es nicht mit den von der Chemie her bekannten Atome zu thun haben, denn die Masse der bewegten Teilchen zeigte sich 2000–4000 mal kleiner als die der Wasserstofatome, also der leichtesten der bekannten chemischen Atome." Also Walter Kaufmann (1871–1947) had found an $e/m \sim 10^7$ for these rays. However, he concluded that the assumption of a corpuscular structure of cathode rays did not give a satisfactory explanation of his observations (Kaufmann, 1897).

[5] For an account of the first decade of α radioactivity see Rutherford's Clark University lecture (Rutherford, 1912) given in 1909. At that time it was not yet known that the doubly charged helium atom is in fact the helium nucleus!

(1903–) appeared which marked the end of the beginning in the developments of β decay. In 1927 they reported (Ellis and Wooster, 1927) on calorimetric measurements in which they showed that the average energy of disintegration in the decay of RaE equals the mean energy of the continuous spectrum rather than its upper limit. It was quite a difficult experiment for its time. Early in 1977, Professor Wooster wrote to me about those days. He recalled that the galvanometer they used in their 1927 experiment "was so sensitive to external changes in the magnetic field that we had to work between 12 midnight and 3 a.m. for a fortnight. Even so when the policeman walked by in the street, the nails of his boots disturbed the galvanometer ..." (Wooster, 1977).

In any event, there remained quite a few who doubted that the Ellis–Wooster result was definitive, among them Wolfgang Pauli (1900–1958). A year and a half later, on February 18, 1929, Pauli wrote as follows to Oskar Klein: "...I myself am rather sure (Heisenberg is not so certain) that γ rays must be the cause of the continuous spectrum...".[6] Only after Lise Meitner (1878–1968) and Wilhelm Orthmann confirmed the Ellis–Wooster result and also showed that RaE (Bi_{83}^{210}) does not produce any γ rays to speak of [their paper was submitted on Dec. 18, 1929 (Meitner and Orthmann, 1930)] did Pauli come to the realization that a crisis was at hand. This, in turn, led him to postulate the neutrino, in a letter to colleagues dated Dec. 4, 1930 (collected in Pauli, 1964).

This abbreviated chronology raises many further questions. To mention but a few: Why did it take 16 years between the discovery of β radioactivity and the first observation of the continuous character of the β spectrum? Why did it take two years from the discovery of the nucleus to the realization that β decay is a nuclear process? Why did it take from 1914 to 1927 before the first serious evidence appeared for an "unusual" origin of the continuous β spectrum? Again, but now on a finer scale of time, the same old question arises: what did people do in between?

I intend to return elsewhere to these and other questions, but shall not answer them in this paper. However, there is a design in raising them. I should like to warn the reader: the major problems in radioactivity which physicists had to cope with in the first quarter of the twentieth century represent a highly complex pattern. What will be related in the rest of this paper concerns some major facets thereof. The full story of that period is still more intricate, however.

III. THE MAIN THEME

I now turn to the main topic of this paper, the discussion of two puzzles which baffled physicists during the early decades immediately following the first observations of radioactive phenomena.

The first one was: What is the source of the energy that continues to be released by radioactive materials? Already in the year of discovery, Becquerel himself had been quite surprised at the persistence of the energy produced by the "uranic rays." From 1898 on, physicists began to pose such questions as: Could it be that energy is not conserved in these processes? Could there be something amiss with the second law of thermodynamics in radioactive transformation? Does the source of energy reside outside the atom or inside?

The second puzzle was: What is the significance of the characteristic half life for such transformations? (The first determination of a lifetime for radioactive decay dates from the year 1900.) If in a given radioactive transformation all parent atoms are identical, and if the same is true for all daughter products, then why does one radioactive parent atom live longer than another, and what decides when a specific parent atom disintegrates?

It should be stressed that these problems did not hold center stage throughout the period under discussion, a period so rich in other developments. Rather, the puzzles to be discussed in this paper were principally the concern of a fairly modest-sized but élite club of experimental radioactivists. In those days, theoretical physicists did not play any role of consequence in the development of this subject, both because they were not particularly needed for its descriptive aspects and because the deeper questions were too difficult for their time. It is true that distinguished theorists (especially those belonging to an older generation) would on occasion express views on these issues from which we gain revealing insights into the climate of thought of the times. But these comments were not to be of lasting significance—with one most notable exception: the contribution by Einstein.

In the second of his 1905 papers on relativity Einstein stated that "If a body gives off the energy L in the form of radiation, its mass diminishes by L/c^2.... The mass of a body is a measure of its energy .... It is not impossible that with bodies whose energy content is variable to a high degree (e.g., with radium salts) the theory may be successfully put to the test" (Einstein, 1905).

With the help of Einstein's discovery of the mass–energy equivalence, some of the questions related to the origins of the radioactive energy release could have been answered, at least in principle (see further Sec. IV below). However, as a matter of historical fact this did not come to pass in the period under discussion. There appear to be three reasons for this. (1) The precepts of relativity were assimilated rather slowly. (2) The level of accuracy of mass measurements was not adequate during this period. Thus in his review of relativity theory, Pauli (1921) notes that "*perhaps* the theorem of the equivalence of mass and energy can be checked *at some future date* by observations on the stability of nuclei" (my italics).[7] (3) The lifetime question had to remain entirely unresolved until the advent of quantum mechanics, when it became possible for the first time

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[6] "Ich selbst bin ziemlich sicher (Heisenberg nicht so unbedingt) dasz γ-Strahlen die Ursache des kontinuirlichen Spektrums sein müssen" (Pauli, 1929). I am indebted to Mrs. Franca Pauli for permission to quote from the W. Pauli correspondence.

[7] The validity of the energy–mass–velocity relation required by special relativity was well verified for the electron by about 1915.

to understand the *mechanisms* of radioactive decay. Prior to the understanding of these mechanisms it was inevitable that the origin of radioactive energy had to remain hazy as well, even though, after the fact, much can be explained about the energy release by the simple application of conservation laws, independently from quantum mechanical arguments.

Yet, well before the proofs were there, a correct consensus began to emerge with regard to the energy puzzle. If around 1910 those who had labored and thought seriously about this question had been polled, there is no doubt that a majority would have expressed the belief that energy is conserved, and that the energy source resides in the atomic interior. Had they further been asked about the explanation of the lifetime puzzle, however, then the wisest would have readily admitted that this was a question beyond their horizon.

In any event, these questions, first stated around the turn of the century, remained unresolved until the summer of 1928, when (see Sec. VII) it was found that "... it has hitherto been necessary to postulate some special arbitrary 'instability' of the nucleus ... but ... disintegration is a natural consequence of the laws of quantum mechanics, without any special hypothesis ..." (Gurney and Condon, 1928).

That was good enough for α but not for β radioactivity. In fact, in just about that same year, 1928, new paradoxes emerged regarding the energy loss in β decay. And so it has continued. The developments which started in the 1890's have posed challenge after challenge until this day. Now, as then, our endeavors are based on a jumble consisting of some fine but incomplete dynamics, some good but incomplete ordering principles, some consensus with no basis in facts—the whole of which is presently called particle physics.

IV. THE FIRST ENERGY CRISIS

Between 1898 and the early 1930's it happened three times that the discoveries of new natural phenomena were so unsettling as to make prominent physicists waver in their faith in the universal validity of the law of conservation of energy. The first of these crises, referred to in Sec. III, concerned radioactivity. Thirty years later it was radioactivity again (more specifically, β decay) which caused temporary doubt in some quarters about energy conservation. In between, agonizing attempts to reconcile quantum effects with classical reasoning led likewise, and again briefly, to suggestions that energy conservation might not hold strictly. It is the first of these three instances which shall concern us here. As a prelude to this subject, let us look briefly at the status of the conservation law toward the end of the nineteenth century.

In 1775, the Paris Academy of Sciences (still the Académie Royale, at that time) formally announced a significant decision (Académie Royale, 1775): "The Academy has resolved, this year, to examine no longer any solutions to problems on the following subjects: the duplication of the cube, the trisection of the angle, the quadrature of the circle, or any machine claiming to be a perpetuum mobile." The resolution was expatiated upon in a motivation with many a curious turn of phrase; for us, this simple categorical statement is of interest: "The construction of a perpetual motion machine is absolutely impossible." Evidently the illustrious Academicians grew tired of finding the inevitable flaws in papers submitted on this subject.

We now call the machine excommunicated by the Academy a perpetuum mobile of the first kind. The growing insight that such a device, which spontaneously creates energy, cannot be made was one of the main contributing factors to the formulation, more than fifty years later, of the universal energy principle, a major achievement of nineteenth century physics. Insofar as purely mechanical systems are concerned, the law of conservation of energy has much older roots (see Hiebert, 1962; Elkana, 1974). Several of science's most illustrious names are associated with these early developments in mechanics. But the principle in its broader sense emerged only when the need arose to express quantitatively the convertibility of diverse forms of energy (mechanical, electrical, magnetic, chemical, physiological, etc.) into each other. The period of discovery of the macroscopic energy law (the first law of thermodynamics) in its generality, that is, applied to any form or several forms of energy, is approximately 1830–1850.

No single year can be associated with this discovery because it was made not by any one person but by many, working most often without initial awareness of each other's activities. A list of pioneers on the subject (Kuhn, 1962) contains no less than twelve names: Sadi Carnot, Colding, Faraday, Grove, Helmholtz, Hirn, Holtzmann, Joule, Liebig, Mayer, Mohr, Séguin. Four of these (Carnot, Hirn, Holtzmann, Séguin) became involved because of their interest in the effectiveness of steam engines, while two others (Helmholtz, Mayer) were initially intrigued by physiological questions. Given this large a number of dramatis personae, priority disputes were inevitable: "most intense battles took place about the priority of [these] ideas, during which execrable personal accusations and repugnant national chauvinism came into the open" (Mach, 1896).[8] These controversies will not be discussed here. The interested reader can find several detailed accounts elsewhere (Mach, 1896; Planck, 1887; Kuhn, 1962).

The curious case of Sadi Carnot (1796–1832) should be mentioned, however. He is of course justly famous as the discoverer of the second law of thermodynamics (for reversible systems). In actual fact he also discovered the first law. In the early 1820's he stated in his diaries that wherever there is destruction of mechanical work (puissance motrice) there is generation of heat (production de chaleur) and concluded: "one can therefore pose the general thesis that mechanical work is an invariable quantity in nature, that properly speaking it is never produced nor destroyed" (Mach, 1896). In addition he gave an estimate (somewhat low, but not at all so bad) of the mechanical equivalent of heat. But he never published! Long after his death, in 1878, this material was handed over to the French Academy by his surviving

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[8] Earlier, Mach had devoted a separate essay to the history of the energy conservation law.

younger brother (H. Carnot, 1878). As Max Planck put it: "He [S.C.] has unquestionably the merit of having given the first evaluation of the mechanical equivalent of heat" (Kuhn, 1962, p. 16). As Ernst Mach put it: "Since for practical reasons one cannot name the law [of the equivalence of heat and mechanical work] for all the people who took part in its discovery and its justification, it is advisable to associate [the law] with the names of those who in both respects must be accorded the priority of publication" (Elkana, 1974, p. 241). For this reason Mach speaks of the Mayer–Joule principle for the case that only mechanical work and heat are considered, and of the energy conservation law when *all* forms of energy are included. It is understandable that the first law is often referred to as *le principe de Carnot* in the French literature. Since the same appellation is also used for the second law, the reader of such papers is advised to find out from the context what the issue is.

Mach also observed (Elkana, 1974, p. 241) that the strongest emphasis on the universality of the conservation of energy stems from Robert Mayer (1814–1878) and Hermann Helmholtz (1821–1894). Already the title of Helmholtz' important essay on the subject[9]: "Über die Erhalhung der Kraft: eine physikalische Abhandlung" ("On the conservation of force: a physical memoir"), is of considerable interest. What is here called force is what we now call energy. Current terminology in this respect is itself of nineteenth-century origin. The first one to use the term "energy" in its modern technical meaning was Thomas Young (1773–1829): "The term energy may be applied with great propriety to the product of the mass or weight of the body, into the square of the number expressing its velocity ..." (Young, 1807). This quantity Mv^2 is the *vis viva* of Leibnitz. A factor 1/2 is still lacking before we arrive at our familiar kinetic energy. This factor seems to have been supplied first by Gaspard Gustave de Coriolis (1792–1843) (Coriolis, 1829).

Nor should one fail to notice Helmholtz' emphasis on his subject as a treatise in *physics*. As he strongly urges, we are not dealing here with an axiomatic statement or a philosophical tenet, nor with a tautology (all such views were expressed at one time or another) but with a physical hypothesis which needs verification in each instance.[10] The key to doing this is to find the equivalent of each energy form (via direct or indirect processes) in terms of mechanical work. Moreover, such an equivalence has an unambiguous meaning only after it is realized that the change in energy of a system from an initial to a final state is independent of the way in which the transition between these states takes place.

These, briefly, are the lines along which the conservation of energy came to be clearly understood as a physical principle of universal validity, as the nineteenth century drew to a close. In two respects there have been fundamental developments of the subject in the twentieth century. First, a unification has taken place of the two basic laws: conservation of energy and conservation of matter. Unlike the former law, the latter one, a product of the eighteenth century, is associated with the name of one single scientist: Antoine-Laurent Lavoisier (1743–1794), a man to whom a grateful nation expressed its debt by putting him under the guillotine, an event of which Laplace has said: "It took them only an instant to cut off that head, and a hundred years may not produce another one like it." Secondly, the energy law appears in thermodynamic context as a macroscopic principle and on a different footing than the conservation of momentum and angular momentum. The modern association between conservation laws and invariance principles emphasizes the microscopic foundations of all three laws, treats them very much on a common level, and frees the conditions for their validity to a larger extent than before from dynamical details.

I shall return elsewhere to both these subjects. For the present purposes it is enough to conclude this brief survey with a comment by Max Planck, found on the opening page of his 1887 prize essay (Planck, 1887): "... if today a quite new natural phenomenon were to be discovered, one would be able to obtain at once from [the energy conservation principle] a law for this new effect, while otherwise there does not exist any other axiom which could be extended with the same confidence to all processes in nature."

To the best of our present knowledge, Planck was right. Yet in years to come the paradoxes posed by several new discoveries were initially so grave as to cause a temporary lack of confidence in the energy principle. Let us now turn to the first of these events.[11]

Becquerel's surprise at the persistence with which the uranic rays kept pouring out energy was already mentioned in Sec. III. In 1910, Marie Curie reminisced as follows about those early days: "The constancy of the uranic radiation caused profound astonishment to those physicists who were the first to be interested in the discovery of H. Becquerel. This constancy appears in fact to be surprising; the radiation does not seem to vary spontaneously with time ..." (M. Curie, 1910). In order to appreciate this statement fully, three facts should be borne in mind. (1) The radiation emitted by uranium when unseparated from its daughter products does in-

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[9] Both this essay and the first paper by Robert Mayer on the subject share the distinction of having been rejected for publication by Poggendorf's Annalen, the later Annalen der Physik. Helmholtz' essay is most easily accessible in Kahl (1971).

[10] "The principle is presented and should be understood as a hypothesis within physics totally divorced from philosophical considerations" (in Kahl, 1971).

[11] It may be noted that in 1882 Helmholtz expressed uncertainty about the applicability of the thermodynamic principles to "the fine structures of the organic living tissues" (Helmholtz, 1883). Likewise Louis-Georges Gouy (1854–1926), one of the pioneers in refined experiments on Brownian motion, wondered in 1888 whether "le principe de Carnot... serait seulement exact pour les mécanismes grossiers... et cesserait d'être applicable... [pour] des dimensions comparable à 1 micron," ("... whether the principle of C. would be exact only for large scale mechanisms... and would cease to be applicable... [for] dimensions of the order of one micron.") (Gouy, 1888). However, these comments are in the nature of asides and were not raised as central issues. For what follows, it may be of interest to observe that Marie Curie was aware of these remarks by Helmholtz and by Gouy.

deed represent to a very high degree a steady state of affairs. (2) It took two years from Becquerel's initial discovery until the first parent–daughter separation was effected. (3) It took another two years until it was firmly established that radioactivity does diminish with time.[12]

Speculation on the origin of radioactive energy started with Marie Curie's very first paper on radioactivity [the one in which she announced her discovery of the activity of thorium (1898)]. There, cautiously, she suggests the possibility that the energy might be due to an outside source: "One might imagine that all of space is constantly traversed by rays similar to Roentgen rays only much more penetrating and being able to be absorbed only by certain elements with large atomic weight, such as uranium and thorium" (S. Curie, 1898). Also Becquerel made an analogy with an externally induced process, phosphorescence: "It would not be contrary to what we know about phosphorescence to suppose that these [U and Th] substances have a relatively considerable energy reserve, which they can emit for years, as radiation, without noticeable weakening" (H. Becquerel, 1899). However, he also stated that this analogy had its limitations. Phosphorescent phenomena exhibit a finite lifetime (as Becquerel and his father well knew), and they can be affected by external agents. Neither of these properties seemed to apply to radioactivity: "...however, it has not been possible to induce any appreciable variation in the intensity of this emission" (H. Becquerel, 1899).

In that same year, 1898, Marie Curie discovered polonium, for which the liberated energy per unit weight of separated material was even larger than for uranium and thorium. Thus the question of the origin of this energy became an even more burning one and she returned to it, listing a number of possible answers (M. Curie, 1899). Here we find the first mention that one might have to face a contradiction with the conservation of energy. Furthermore she emphasized that the assumption of an external source would be nothing but an evasion of energy nonconservation—unless the nature of the external source were determined: "Any exception to Carnot's principle [first law!] can be evaded by the intervention of an unknown energy which comes to us from space. To adopt such an explanation or to put in doubt the generality of the Carnot principle are in fact two points of view which to us amount to one and the same as long as the nature of the energy here invoked stays entirely 'dans le domaine de l'arbitraire'." She also pointed out that the interior of the atom could be the energy source: "The radiation [may be] an emission of matter accompanied by a loss of weight of the radioactive substances" (M. Curie, 1899).

Not only the first but also the second law of thermodynamics was sometimes questioned as a result of this energy puzzle. For example, in his 1898 inaugural address as President of the British Association, the brilliant and erratic Sir William Crookes speculated, somewhere in between dissertations on food shortages and psychical research, whether one can "mentally modify Maxwell's demons" in such a way that radioactive substances release energy drawn from the air surrounding the active material (Crookes, 1898; also 1899).

These various speculations set in motion a set of experiments designed to locate a possible outside source of radioactive energy. In an attempt to see whether the sun could be the cause, the Curies looked for diurnal variations in the activity of uranium. They found no effect (M. Curie, 1910, Vol. 1, p. 129). Among others who addressed the same question, particular mention should be made of the team of Elster and Geitel.

Julius Elster (1854–1920) and Hans Geitel (1855–1923) had been high school friends. They both became teachers at the Gymnasium[13] in Wolfenbüttel near Braunschweig. When Elster married and had a house built, Geitel moved in with the young couple and together the two friends built a laboratory in the new home. Here they started their research (often financed from their own pockets) which were to make them internationally renowned.[14] They experimented on photoelectric effects, on spectroscopy, on the conduction of electricity through gases, and especially on atmospheric electricity. These last experiments led to their classic work on the radioactivity of the atmosphere, research about which Rutherford spoke with great respect. Simultaneously with Crookes they discovered the scintillations of zinc sulfide screens by α rays (Elster and Geitel, 1903).

In later years the two men loved to relate (Pohl, 1924) their experiences in Berlin, where the Prussian Minister of Education tried to convince them to accept a joint offer as university professors at a first-rate institute. They listened modestly but did not react. The minister believed that "die kleinen Oberlehrer der Provinz" were probably too awed and suggested that they take a few hours to think it over. They did so, came back and said no thank you. They had decided that the transition to the academic world would inhibit their independent research. They were grateful for the honor but preferred to stay in Wolfenbüttel.

The two were inseparable. I cannot resist mentioning an anecdote related by D'Andrade (1964). "In their time there was a man who much resembled Geitel in appearance. A stranger meeting him said 'Good morning, Herr Elster...', to which he replied, '...Firstly I am not Elster but Geitel, and secondly I am not Geitel'." Almost their complete oeuvre consists of joint publications. "We shall doubtless search in vain for a similar instance of private scientific partnership throughout a lifelong friendship. Each ascribed to the other the credit for a discovery published jointly" (Lawson, 1924).

Their work on the origins of radioactive processes is contained in two papers. In the first one (Elster and Geitel, 1898) they begin with the observation that if Crookes were right and the radioactive energy is taken from the surrounding air (Crookes, 1898, 1899), then the activity should decrease when the source is placed in a

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[12] See further Sec. VII of this paper.

[13] It is not evocative enough to translate Gymnasium simply as high school. Let us say it is an academic high school preparatory to going to university.

[14] There were others as well who did their most creative work while teaching in a high school, Weierstrass for example.

vacuum. They find no such effect. Next they turn to the conjecture of the Curies that the energy may be supplied by an X-ray-like radiation which is all pervasive in the atmosphere and reasoned that, if this were so, there should be a decrease in activity if the source were placed deep underground. So they requested and obtained permission to do an experiment in the Clausthal mines in the Harz mountains, under 300 meters of rock. They found no effect. They admit that perhaps the rock layer may not be all that good an absorber. Nevertheless they conclude, as early as 1898, that "the hypothesis of the excitation of Becquerel rays by radiation preexisting in space appears improbable to the highest degree (im höchsten Grade unwahrscheinlich)." In their second paper (Elster and Geitel, 1899) they report on attempts to increase the radioactive emissions by exposing a source to cathode rays; or to sunlight. They find no effect and conclude "man wird vielmehr aus dem Atome des betreffenden Elementes selber die Lichtquellen ableiten müssen."[15]

It is important to stress at this point that the fascinating puzzles discussed in this chapter were never any hindrance to progress in their days. If anything, the contrary is true. The field of radioactivity was young at the time when these questions arose, the tasks were enormous. While these problems were given much thought by the Curies, that never inhibited them from continuing their superb research. They were a stimulus to men like Elster and Geitel, as we have just seen. Others chose to state them as unresolved issues and then to move on to other pursuits. Such was largely the attitude of the English school. Rutherford, for example, simply noted in his 1899 memoir that "the cause and origin of the radiation continuously emitted by uranium and its salts still remain a mystery" (Rutherford, 1899). J. J. Thomson always took the attitude that the atom itself was the energy source—"...the [radioactive] changes we are considering are changes in the configuration of the atom ..." (Thomson, 1903).

I referred earlier to the article by Marie Curie in which she listed possible options for the explanation of the energy release. It was written before but published after the discovery of radium by her, Pierre Curie, and Bêmont. This last development once again brought the issue to the fore. The radium radiation was even more intense than for polonium! The question of nonconservation of energy came up once again: "On réalise ainsi une source de lumière, à vrai dire très faible, mais qui fonctionne sans source d'énergie. Il y a là une contradiction tout au moins apparente avec le principe de Carnot" (P. and S. Curie and G. Bêmont, 1898).[16]

Nonconservation of energy was never a widely held explanation of these effects. In 1902 the Curies again gave a list of possible interpretations, on which this possibility no longer appears (P. and M. Curie, 1902). Yet in that same year, a visitor to England recalled that he "had been dining seated between Lord Kelvin and Professor Becquerel, ... Lord Kelvin had turned to him and said that the discovery of Becquerel radiations had placed the first question mark against the principle of conservation of energy which had been placed against it since the principle was enunciated" (Hammer, 1903).

It should also be stressed that such options as nonconservation of energy or external sources were not proposed lightheartedly. The idea that the atom itself is the source was not so easily swallowed at that time, since it meant giving up the concept of an atom as an immutable entity. By 1900 the debate over the reality of atoms was well past its peak; but at that time the question was not universally regarded as settled. The Curies were proponents of the existence of real atoms, as their writings make abundantly clear. But to accept the atom itself as the source of the energy could only mean one thing to them: transmutation. And they could not simply accept this since to them at that time it seemed in conflict with the principles of chemistry as then known—which indeed it was. In 1900 Marie Curie summed up the dilemma in the following way (M. Curie, 1900):

"Uranium exhibits no appreciable change of state, no visible chemical transformation, it remains, or so it seems, identical with itself, the source of energy which it emits remains undetectable—and therein lies the profound interest of the phenomenon. There is perhaps a disagreement with the fundamental laws of science which until now have been considered as general ... The materialistic theory of radioactivity[17] is very attractive. It does explain the phenomena of radioactivity. However, if we adopt this theory, we have to decide to admit that radioactive matter is not in an ordinary chemical state; according to it, the atoms do not constitute a stable state, since particles smaller than the atom are emitted. The atoms, *indivisible from the chemical point of view* (my italics), are here divisible, and the subatoms are in motion.... The materialist theory of radioactivity leads us...quite far. If we refuse to admit its consequences, our embarassment will not lessen. If radioactive matter does not modify itself, then we find ourselves again in the presence of the question: from where comes the radioactive energy? And if the source of energy cannot be found we are in conflict with Carnot's principle, a principle fundamental to thermodynamics.... We are then forced to admit that Carnot's principle [the second law!] is not absolutely general [and]...that the radioactive substances are able to transform heat from the ambiant environment into work. This hypothesis undermines the accepted ideas in physics as seriously as the hypothesis of the transformation of the elements does in chemistry, and one sees that the question cannot easily be resolved." (Cette hypothèse porte une atteinte aussi grave aux idées admises en physique que l'hypothèse de la transformation des éléments aux principes de la chimie, et on voit que la question n'est pas facile à résoudre.)

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[15] Rather, one will have to derive the source of light (sic) from the atom itself of the element concerned.

[16] Thus one realizes a source of light (sic), quite weak to be sure, but which functions without a source of energy. There is here a contradiction, or so it seems, with the principle of Carnot" [the first law!].

[17] According to which radioactive atoms expel subatomic particles.

The transformation theory of Rutherford and Soddy, proposed in 1902, provided the break with the past which was clearly needed in order to answer Marie Curie's question. In this "great theory of radioactivity which these young men sprung on the learned, timid, rather unbelieving, and, as yet, unquantized world of physics of 1902 and 1903" (Russell, 1951), they unabashedly put forward the idea that some atomic species are subject to spontaneous transmutation (Rutherford and Soddy, 1902). Forty years later, a witness to the events characterized the mood of the times as follows (Robinson, 1943): "It must be difficult if not impossible for the young physicist or chemist of the present day to realize how extremely bold it was and how unacceptable to the atomists of the time... this is a point which must be stressed, for the younger generation is more likely to be familiar with the ordered simplicity of the radioactive series as we know them than with the chaotic state which preceded the transformation theory."[18]

The main tenet of the transformation theory[19] is: radioactive bodies contain unstable atoms of which a fixed fraction decay per unit time. The rest of the decayed atom is a new radio element which decays again, and so forth, till finally a stable element is reached.

As Rutherford himself emphasized some time later (see below), there is no explicit reference in this theory as to the energy mechanism. Nevertheless, the successes of the transformation theory led Rutherford to express the following opinion: "This [transformation] theory is found to account in a satisfactory way for all the known facts of radioactivity and a mass of disconnected facts into one homogeneous whole. On this view, the continuous emission of energy from the active bodies is derived from the internal energy inherent in the atom, and does not in any way contradict the law of conservation of energy" (Rutherford, 1904).

And so the energy debate might have quieted down were it not that, in March 1903, new fuel was added to it by the discovery that radioactive energy release surpassed *in magnitude* anything that had been known until then from chemical reactions. In that year Pierre Curie and Laborde (1903) measured the amount of energy released within a Bunsen's ice calorimeter by a known quantity of radium. They found that 1 g of radium can heat ~4/3 g of water from the melting point to the boiling point in 1 hr. These results caused a tremendous stir. The authors themselves referred once again to a possible outside energy source: "This release of heat can also be explained by supposing that radium utilises an exterior energy of unknown nature." In a discussion of the new discovery, Kelvin spoke of THE mystery of radium (his capitals) and continued: "It seems to me, therefore, absolutely certain that if emission of heat can go on month after month... energy must be supplied from without... I venture to suggest that somehow etherial waves may supply energy to radium..." (Kelvin, 1904). In a lecture on "The present crisis of mathematical physics" given in 1904, Poincaré also brought up the energy conservation question: "... These principles on which we have built everything, are they about to crumble away in turn?... When I speak thus, you no doubt think of radium, that grand revolutionist of the present time.... At least, the principle of the conservation of energy still remained with us, and this seemed more solid. Shall I recall to you how it was in its turn thrown into discredit?... This [activity of radium] was itself a strain on the principles.... But these quantities of [radioactive] energy were too slight to be measured; at least that was the belief, and we were not much troubled. The scene changed when Curie bethought himself to put radium in a calorimeter; it was then seen that the quantity of heat created incessantly was very notable..." (Poincaré, 1913).

Section V of this paper will relate how these developments became general public knowledge. From the physics point of view, these results became even more remarkable when it was found from additional experiments that ~75% of this effect was due to a daughter product of radium, the radium emanation (radon, Rn_{86}^{222}), although the amount of emanation present was actually extremely small. In fact the energy released by radon (Rutherford and Barnes, 1904; Ramsey and Soddy, 1904) is more than a million times greater than the heat evolved by the same volume of hydrogen and oxygen when they explode to form water. In 1905 Soddy wrote of these discoveries: "It is probably the most far reaching and revolutionary fact that has yet transpired in the study of radioactive substances. This enormous evolution of energy which accompanies the production of helium from the radium emanation establishes beyond question the new and fundamental character of radioactive change" (reprinted in Soddy, 1975).

Soddy also pointed out that the magnitude of the energy production made it ever more difficult to imagine it to be due to an external source: "It has been suggested... that all space is traversed by undiscovered radiations to which ordinary matter is completely transparent, but to which radioactive substances are opaque. On this view, the energy traversing a cubic centimeter of space must be at least 60.000 calories per hour [in order to explain the heating effects due to radium]. The total quantity in the universe must therefore be so great that the hypothesis involves far greater difficulties than the effects it is designed to explain" (Soddy, 1975).

Still the external source idea would not quite die.

In 1906, Sagnac raised a new possibility (Sagnac, 1906). He asked: could gravitational energy be the external source? Might it be that the Newtonian attraction is universal for nonradioactive bodies while yet the Newtonian constant could have a "valeur spéciale" for radium? This led him to do a torsion balance experiment in which he compared the oscillations of equal weights of barium

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[18] The events surrounding the enunciation of this theory have been described in more detail by Badash (1966).

[19] Why did Rutherford and Soddy not use the term "transmutation" but rather the more neutral one, "transformation"? The following exchange took place while they were at work on the separation of thorium *X* (Howorth, 1958). Soddy: "Rutherford this is transmutation...". Rutherford: "For Mike's sake, Soddy, don't call it transmutation. They'll have our heads off as alchemists."

and radium. He found no effect.[20]

In 1911, Rutherford again referred to the energy issue but expressed himself more cautiously than he had done earlier (1904). He observed (1911d) that the transformation theory leaves open the question of the inside versus the outside source, since all results of the transformation theory remain true for either hypothesis.

As late as 1919, Jean Perrin came forth with a new "ultra X-ray" mechanism for explaining radioactivity as an externally induced effect (Perrin, 1919, 1920). This time the new radiation was supposed to come not from outer space but "from under our feet, from the fiery center of the planet." The scheme is discussed in detail on its astronomical and cosmological implications.

The matter was still being discussed in the year in which quantum mechanics was born (Briner, 1925). This is not to say that the external source idea was in any way a serious issue at that late time. It does bring home the fact, however, that only in the quantum-mechanical era could the definitive proof of the existence of internal mechanisms for energy generation be given, which settled the question for the ages.

The negative outcome of these various searches for an external energy source was a positive contributing factor to a major insight which emerged early in the twentieth century: physical and chemical actions do not affect radioactive phenomena. In 1903, Rutherford and Soddy elevated this to a new principle, the "conservation of radioactivity": "Radioactivity, according to our present knowledge, must be regarded as a process which lies wholly outside the sphere of known controllable forces, and cannot be created, altered or destroyed" (Rutherford and Soddy, 1903).

There is a vast body of experiments which bear on this question. Temperature independence was established by many. Pierre Curie went to London, to repeat with Dewar his radium heat production experiment, at liquid air temperatures. Marie Curie later went to Leyden to do the same at liquid hydrogen temperatures, with Kamerlingh Onnes. Rutherford stuck 4 mg of radium bromide inside a steel-enclosed cordite bomb, exploded the device, and concluded that there was no change in radioactivity at temperatures ~2500 °C. Independence of pressure, of concentration, of the presence of strong magnetic fields, of irradiations of various kinds were established. Detailed discussions of these results are beyond the compass of this paper. For more information the interested reader is referred to older textbooks.[21]

Yet, strictly speaking, there is no such thing as the conservation of radioactivity. This became clear after the Second World War. I shall come back to this in Sec. VII of this paper. Even so, the "conservation of radioactivity" served an excellent purpose in its time. In particular it helped Bohr to locate the atomic nucleus as the seat of all radioactive phenomena, as will be discussed elsewhere. As this "principle" so well illustrates, it is often better at an early stage to know the truth than to know the whole truth.

Nowhere in this section has there been mention of relativity theory. Some reasons for this absence have already been given in Sec. III. To conclude this section, I should like to make a further comment on this point.

As far as I know, Einstein's work did not lead to theoretical studies on the role of relativity in nuclear phenomena until the year 1913, when the question was raised whether small deviations from Prout's rule of integer mass multiples could be associated with an equivalent mass of the interaction energy between nuclear constituents.[22] Also the related (and still open) question why some but not all nuclei are radioactive came under scrutiny. As a good example of such early work, a paper by Swinne (1913) entitled, "On an application of the principle of relativity in radiochemistry" should be mentioned. It contains a relativistic treatment (in the sense of kinematics) of the α-radioactive process, based on the tacit assumption that this process is a *decay* of the type $A \to B + \alpha$—that is, there is no external source,[23] no doubt in accord with the general consensus of that time. However, it would have been just as easy to apply the same ideas to the "external source model" according to which α radioactivity should be a *reaction* of the type $X + A \to B + \alpha$. It is obvious that a simple discussion of this reaction would yield criteria for the need of the source X. Yet (to my knowledge) such a reasoning was not proposed at any time.

V. INTERLUDE: ATOMIC ENERGY

Speculations on the possible good and the possible evil of the atom go back to the founding fathers of radioactivity.

Here is Becquerel, in an early lecture (quoted by Crookes, 1910): "Today the [radioactive] phenomena are of transcendent interest, but in them almost infinitesimal amounts of energy are utilized. Whether ultimately science will have so far advanced as to permit of the practical utilization of the abundant store of energy locked up in every atom of matter is a problem which only the future can answer. Remember, at the dawn of electricity this was looked on as a mere toy, suitable only to amuse children by attracting bits of paper with a stick of rubbed sealing wax."

Here is Pierre Curie, also in a lecture (reprinted in P. Curie, 1967): "... it can even be thought that Radium could become very dangerous in criminal hands, and here the question can be raised whether mankind benefits from the secrets of Nature..."

And here is Soddy in an early popular book (1912): "If we pause but for a moment to reflect what energy means

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[20] A related negative result was obtained by Thomson (1905), who determined the swinging time of a pendulum to which a bob of RaBr was attached. His work was differently motivated, however. Much later, Rutherford and Compton (1919) also obtained a negative result when looking for the influence of gravitation on radioactivity.

[21] See, for example, Meyer and Schweidler (1927).

[22] The existence of radioactive isotopes was known by then, and J. J. Thomson had achieved the first isotope separation in neon in that same year.

[23] The author states that the masses are not known precisely enough to draw any firm conclusions on the questions just mentioned.

for the present, we may gain some faint notion as to what the question of transmutation may mean for the future to a fuelless world, once more dependent on a hand-to-mouth method of subsistence. It may still be centuries before this occurs, but neither the application of the discoveries of science nor even their achievement is to be compared with the struggle in winning them."

It is also noteworthy that, even in these early stages, questions concerning radioactive energy, and their implications, seized the imagination of the general public. This was due mainly to reports which began to circulate in the press, soon after the discovery by Curie and Laborde (1903), about a mysterious new energy stored in radium. For example, in a preview of the forthcoming 1904 International Electrical Congress in St Louis,[24] the *St. Louis Post Despatch* of October 4, 1903 carried an item about the unusual properties of radium. The article contains sweeping statements to the effect that radioactivity could cause a holocaust.[25]

In that same year, 1903, the term "atomic energy" entered the language for the first time, in a most appropriate sense. It was first used by Rutherford and Soddy (1903) *not* just for the energy released by a radioactive element, but much more generally for the energy locked up in *any* atom: "All these considerations point to the conclusion that the energy latent in the atom must be enormous compared with that rendered free in ordinary chemical change. Now the radio elements differ in no way from the other elements in their chemical and physical behavior. On the one hand they resemble chemically their inactive prototypes in the periodic table very closely, and on the other they possess no common chemical characteristic which could be associated with their radioactivity. Hence there is no reason to assume that this enormous store of energy is possessed by the radio elements alone. It seems probable that *atomic energy* (my italics) in general is of a similar high order of magnitude, although the absence of change prevents its existence being manifested." This, truly, is the physics of the twentieth century.

The fact that today "atomic energy" is an expression firmly anchored in our everyday language has nothing to do, however, with the above marvelous lines. Rather, the present common usage of the term derives in the first instance from a report released by the President of the United States on the evening of Saturday, August 11, 1945. The title of this report came eventually to be "Atomic Energy for Military Purposes. The Official Report on the Development of the Atomic Bomb under the Auspices of the United States Government, 1940–45." It is now generally known as the Smyth Report.

Here is how I became aware of the fact that "atomic energy" is an expression reinvented, as it were, in 1945. After I finished a draft of the present paper, sometime in November, 1976, I went to see my friend Henry DeWolf Smyth. We had the following conversation, the essence of which I report here with his permission.

A. P. When you were writing your report, were you aware that the term atomic energy dates back to the beginning of this century?

H. D. S. No, I was not.

A. P. I have a second question. In the days of Rutherford and Soddy, the nucleus was not yet discovered. Therefore, the expression "atomic energy" was, so to speak, the only natural one they *could* use. Now I have been puzzled, ever since your report came out: why in fact did you not speak of "nuclear energy," "nuclear bomb," etc.? Having just finished writing the paper of which I told you, this seems a good time for me to ask you about this.

H. D. S. Your question comes at a very opportune moment, since I have just published an article on the history of the Smyth Report. You will be glad to know that in my original draft I did use the word "nuclear" instead of "atomic." After the writing was done there followed a period of consultation with (Major General Leslie R.) Groves. In turn, Groves must have discussed my draft with his advisers (James B.) Conant and (Richard C.) Tolman, and possibly with others as well. In a subsequent discussion, we decided that the word "nuclear" was either totally unfamiliar to the public or primarily had a biological flavor, whereas "atomic" has a definite association with chemistry and physics. Since it became clear that the report was aimed at a wider audience than nuclear physicists, we decided that "atomic" was less likely to frighten off readers than "nuclear." So I accepted the change after a somewhat painful suppression of my purist principles.

With these words he gave me a copy of his article (Smyth, 1976). I gratefully accepted this gift from a man my respect for whom I have already expressed elsewhere (Pais, 1969).

VI. WHY A HALF LIFE?

The first determination of a half life for a radioactive decay was made by Rutherford (1900). In a study of the properties of thorium emanation (Rn_{86}^{220}) he found that the intensity of the radiations given out by his sample fell off with time in a geometric progression. Thus he was the first to note that if $N(t)$ is the number of active atoms at time t, then the decrease of N with t is well described[26] by

$$dN/dt = -\lambda N, \quad \text{or } N(t) = N(0)e^{-\lambda t}. \tag{1}$$

He called λ the radioactive constant, and "it has been shown that $e^{-\lambda t} = 1/2$ when $t = 60$ sec," a quite respectable half life determination (the modern value is about 55 sec). It is of course no accident that this first discovery

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[24] This meeting was attended by J. J. Thomson and by Rutherford.

[25] The article was headed: "Priceless mysterious radium will be exhibited in St. Louis. A gram of the most wonderful and mysterious metal to be shown in St. Louis in 1904." The text contained these lines: "Its power will be inconceivable. By means of the metal all the arsenals in the world would be destroyed. It could make war impossible by exhausting all the accumulated explosives in the world. . . . It is even possible that an instrument might be invented which at the touch of a key would blow up the whole earth and bring about the end of the world" (quoted in Jauncey, 1946).

[26] In the absence of any replenishing mechanism.

concerned an element of medium-short life. Much longer half lives (such as the one for radium, ~1600 yr) were also well established within the next few years, with the help of the theory of radioactive equilibrium between parent and daughter substances.[27] Equation (1) and its generalization to sequential decays was the first of two contributions by Rutherford to theoretical physics, an activity which he did not always hold in the highest esteem (his second contribution was his theoretical discovery of the central nucleus from the results of scattering experiments).

Today, even though we may not always be able to compute λ theoretically for any given radioactive decay, the meaning of λ is certainly quite clear. However different the respective mechanisms for α, β and γ decay are, in each case λ is a quantum mechanical transition probability per unit time. Thus radioactivity represents one instance among very many of a situation in which physicists of earlier days were unwittingly dealing with quantum effects.

At the turn of the century there already existed a body of knowledge on unstable systems of atomic dimensions. For example, much work had been done at that time on luminescent phenomena. This had made the lifetime concept familiar. It is true that insuperable problems arose for those who attempted to find mechanisms for these and similar processes; consider for instance Boltzmann's struggles with molecular dissociation.[28] However, if these various unstable systems were not amenable to theoretical treatment, they did not appear to pose any manifest paradoxes, principally since the causes for instability could at least be identified on a phenomenological level. It is in this last respect that radioactivity created problems unique for their time. It seemed (in fact, it was true) that radioactive decays were contrary to the classical concepts of cause and effect. During the first two decades of this century, physicists had no reason to suspect that these paradoxes were not by any means typical for radioactivity only.

Jeans has given a graphic description of the situation: "Interesting but difficult questions arise when we discuss which atoms will disintegrate first, and which will live longest without disintegration. [Suppose that] 500 million atoms are due to disintegrate in the next second. What, we may inquire, determines which particular atoms will fill the quota?.... it seemed to remove causality from a large part of our picture of the physical world. If we are told the position and the speed of motion of every one [of a set of radium atoms], we might expect that Laplace's super-mathematician would be able to predict the future of every atom. And so he would if their motion conformed to the classical mechanics. But the new laws merely tell him that one of his atoms is destined to disintegrate today, another tomorrow, and so on. No amount of calculation will tell him which atoms will do this..." (Jeans, 1943).

Nevertheless, there were those who, early on, began to think of dynamical models which would incorporate radioactivity. One of these early model builders was J. J. Thomson. Some details of his work on models will be discussed elsewhere. Here it suffices to state that he attempted to describe radioactivity in terms of classical mechanical pictures. Thus we can well understand the objections which Lord Kelvin wrote to Thomson: "What would be the difference, between radium atoms in a piece of radium bromide, of the atoms which are nearly ripe for explosion, and those which have the prospect of several thousand years of stable diminishing motions before explosion?" (Rayleigh, 1969). Rutherford also saw this weak point of the classical model: "...all atoms formed at the same time should last for a definite interval. This, however, is contrary to the observed law of transformation, in which the atoms have a life embracing all values from zero to infinity" (Rutherford, 1911d).

In despair, one might of course reply to the question: how is it possible that one species of identical atoms is made up out of particles some of which live longer than others? by saying that "the different atoms of a radioactive substance are not in every respect identical" (Rayleigh, 1969, p. 142), a possibility mentioned by Thomson in an address in 1909. However he never came back to this.

Those who wisely left aside the question: "why does a radioactive atom change?" and focused on the more modest problem: "how does it change?" were able to make some further progress on a more descriptive level, however. They focused on the essential content of Eq. (1), which is probabilistic: the probability that a given unstable atom decays is the same for all atoms (in a sample of a given species) and is independent of its age, but does depend on the specific element under consideration. "If the destroying angel selected out of all those alive in the world a fixed proportion to die every minute, independently of their age,...and chose purely at random...then our expectation of life would be that of the radioactive atoms" (Soddy, 1920). With the help of this probability Ansatz, it is of course possible to refine Eq. (1) (which can hold strictly only in the limit of very large N, since it ignores the discreteness of N) for the case of finite samples, to predict the average number of events in any finite time interval (for given λ) and to study the fluctuations of that number (the "Schweidler fluctuations"[29]) as well as fluctuations for other variables such as heat production, ionization, spatial distributions, etc. (see Meyer and Schweidler, 1927; Fürth, 1920).

Beyond that, there was not much more that could be done. The lifetime paradox simply did not lend itself to the statement of new hypotheses subject to test. The problem was so difficult that it was hard even to get a wrong idea about it.

In a review of alternatives by Debierne in his thesis (1914) an old acquaintance briefly returns: the exterior source of radioactive decay. He notes that exponential

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[27] See, for example, Rutherford, Chadwick, and Ellis, 1930, *Radiations from Radioactive Substances* (Cambridge University, Cambridge, England).

[28] See the English translation: L. Boltzmann, 1964, *Lectures on Gas Theory* (University of California, Berkeley), Part 2, Chap. 6.

[29] After Egon von Schweidler who was the first to draw attention to such fluctuation phenomena (1905).

decays occur in several chemical processes, such as monomolecular irreversible reactions (dissociations etc.). He notes that in such instances thermal disorder plays a role. This leads him to ask whether the radioactive decay processes could be due to some exterior action which, however, cannot respond to temperature variations. He concludes that such a mechanism is hard to conceive. Pursuing the thermal disorder analogy, he speculates that each unstable atom contains an extremely complex system in which high-velocity particles create a state of "internal disorder." Several others likewise tried to associate the decay properties with fluctuations in highly complex internal motions (Lindemann, 1915; Wolff, 1920).

Considerations of this kind were the subject of an address by Marie Curie to the second Solvay Conference in 1913. In the subsequent discussion, Rutherford expressed his interest in the ideas of Debierne and summarized his own view as follows: "The law of radioactive transformation, which is universal for all radioactive substances, seems only to be explicable as a consequence of accidental disturbances ("troubles fortuits") in the nucleus, in conformity with probability laws. But, in the present state of knowledge, it does not seem possible to form a clear idea as to the very constitution of the atomic nucleus, nor of the causes which lead to its disintegration" (Rutherford, 1921).

And so these problems remained unresolved until several years after the birth of quantum mechanics.

VII. POSTSCRIPTA: MODERN TIMES

(1) 1928: *α decay explained.* It was realized by George Gamow (1928) in Goettingen, in August, 1928, and independently by Ronald W. Gurney and Edward U. Condon in Princeton, one month later (Gurney and Condon, 1928, 1929), that α decay results as a consequence of quantum-mechanical tunneling through a potential barrier. Moreover, all authors had a further significant advance to report: the first explanation of the Geiger-Nuttall relation, known phenomenologically since 1912 (Geiger and Nuttall, 1912), which establishes a connection between the lifetime of an α-emitter and the range of the produced α particles.

In the letter to *Nature* by Gurney and Condon we hear for the last time the echoes of a confusing past: "It has hitherto been necessary to postulate some special arbitrary 'instability' of the nucleus; but in the following note it is pointed out that disintegration is a natural consequence of the laws of quantum mechanics without any special hypothesis.... Much has been written about the explosive violence with which the α particle is hurled from its place in the nucleus. But from the process pictured above, one would rather say that the α particle slips away almost unnoticed."

(2) *Nonconservation of radioactivity.* If the principle of conservation of radioactivity (mentioned toward the end of Sec. IV) were strictly valid, then it would follow that the decay constant λ is independent of all chemical and physical changes, for any radioactive process.

The first suggestions that this cannot be universally true were published independently by Segrè (1947) and by Daudel (1947). The process they chose to discuss, K-capture in Be,[7] was not known to the founding fathers, to be sure. They noted that the chemical environment should affect the electron capture rate, especially in light nuclei, since chemical changes imply changes in the electron density at the position of the nucleus. During the next decade, effects ~0.1% were indeed established experimentally.[30] In 1951 it was noted that similar considerations also apply to decays involving internal conversion, and an effect ~0.3% was observed by comparing different chemical embeddings for a cleverly chosen technetium 99 isomer (Bainbridge *et al.*, 1951, 1953); in 1965, a niobium-90 isomer turned out to be even better, yielding an effect one order of magnitude larger (Cooper *et al.*, 1965).

These and other manifestations of "nonconservation of radioactivity" have become a lively subject of research in recent times.[31] This is of course also due to the discovery of a quite different influence of the environment on radioactive decay: The Mössbauer effect, where a nuclear decay, even though it originates in a single nucleus, is properly described only by treating the decay as a collective quantum-mechanical property of the entire crystal in which that nucleus resides (see Frauenfelder, 1962).

(3) *The exponential law of radioactive decay.* The question how well Eq. (1) describes the temporal behavior of radioactive substances is quite an old one. Its early version was: is λ a constant independent of time? Through the years this was verified experimentally, for long periods of time by Rutherford (1911c), and later for "very young" sources of radium emanation (Poole, 1914), eventually down to 10^{-5} sec after their creation (Joliot, 1930). A more recent study of ^{56}Mn showed no deviations of the exponential law during the first 34 half lives (Winter, 1962).

Quantum-mechanical arguments show nevertheless that Eq. (1) is not mathematically rigorous. Deviations occur[32] for times both very small and very large compared with λ^{-1}. Asymptotically for large times the exponential behavior turns into a power behavior. Experimental situations in which such deviations play a noticeable role have not been found to date.

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[30] See R. Bouchez *et al.* (1956) for references to earlier experimental literature.

[31] See the review by G. T. Emery (1972).

[32] See, for example, Hellund, 1953; Höhler, 1958; Lévy, 1959; Matthews and Salam, 1959; Petzold, 1959a,b; Schwinger, 1960.

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