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Subatomic Determinism and Causal Models of Radioactive Decay, 1903–1923

Helge Kragh

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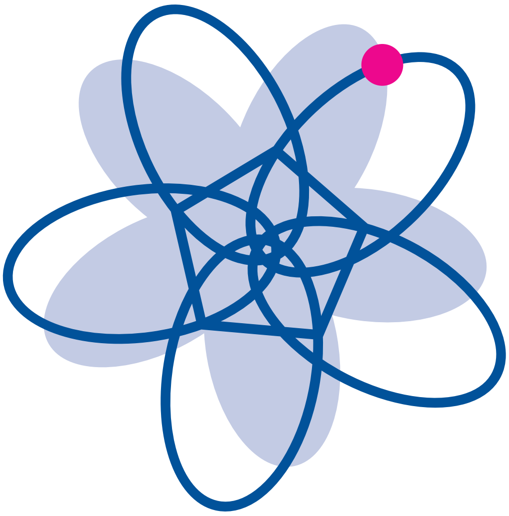

Centre for Science Studies, University of Aarhus, Denmark
Research group: History and philosophy of science

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Subatomic Determinism and Causal Models of Radioactive Decay, 1903-1923[*]

HELGE KRAGH[**]

Introduction

Science is essentially problem-solving – "the art of the soluble," to use the phrase of Nobel laureate Peter Medawar.[1] When faced with reliable experiments that cannot be explained according to accepted theory, scientists typically modify the theory or develop new theoretical tools that may or may not constitute a break with the existing theory. Within the new theoretical framework the problem or anomaly disappears, that is, it is turned into a non-problem. However, this is not the only kind of problem-shift processes met in the sciences. In some cases it is realized that a certain problem either cannot be solved in principle or is a pseudoproblem, that is, it is meaningless within the existing theoretical framework; in both cases, it will be dismissed, if generally for different reasons. Such problem-shifts are typically connected with deep changes in the paradigmatic background.

A good example is the density of the luminiferous ether, which was

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[*] This is a slightly revised version of a manuscript intended for a book that never appeared. A version of it was presented under a different title ("Probabilistic physics and the enigma of radioactivity") at the joint DHS-DLMPS Commission Conference on "Probabilité et Réalité Physique" at the Palais des Académies in Brussels on 19 November 2004.

[**] Department of Science Studies, University of Aarhus, Denmark. E-mail: helge.kragh@ivs.au.dk.

considered a meaningful and interesting problem in the late nineteenth century. Many leading physicists, including William Thomson, Francis Fitzgerald and Oliver Lodge, calculated the density of the ether and related their calculations to experiments. However, the issue was redefined as a pseudoproblem after relativity had made the ether superfluous. The history of science offers several other examples of a similar kind.

There is a third way in which a problem may be brought to disappear, namely, if it is decided that the problem is no longer worth dealing with or if it is judged to be so complicated that it cannot be solved. This is a pragmatic strategy that should be distinguished from the ones just mentioned. Problems within the third category are not turned into non-problems because they have no solution, but because they cannot be solved in practice or because the lack of solution is of no significance for progress in the field of science to which it belongs.

The kind of pragmatic problem-shifts mentioned above is quite common in the sciences and is nicely exemplified be the early history of radioactivity. How can radioactivity be explained? And what qualifies as an explanation? About 1906 almost all physicists believed that the phenomenon was of intra-atomic origin and could be given a causal explanation if only the structure and dynamics of the atom was sufficiently well understood. Unfortunately, such an understanding was lacking. For this reason there was no way in which scientists could explain why some elements are radioactive and some are not, or when a particular atom of radium would decay. However, this was seen as a practical difficulty, not a difficulty that could not be solved in principle. It is a fact of history that for more than a decade a minority of physicists attempted to understand radioactivity on a deterministic and microphysical basis.[2] We know that this work was futile and that radioactivity can only be explained quantum-mechanically, which implies that no definite cause can be ascribed to why an atom decays at a particular time. Radioactivity is fundamentally indeterministic. Yet, although the statistical nature of radioactivity was recognized at an early date, this was not taken to imply that the phenomenon defied the law of causality. The recognition of radioactivity as being truly acausal and indeterministic was a by-product of the more general indeterminism associated with quantum mechanics after 1925.

Radioactivity as intra-atomic rearrangements

The early work in radioactivity can be divided roughly into three, partly overlapping phases or research topics. The earliest one was purely phenomenological, an attempt to determine the basic *properties* of radioactive substances. Another, slightly later phase was preoccupied with determining the *nature* of the rays emanating from uranium and some other metals. At the same time, there was an interest in the *origin* of the rays or, even more ambitiously, attempts to find the *mechanism* responsible for radioactivity. On the whole, this theoretical or speculative approach attracted much less interest than the two experimentally oriented approaches. Moreover, whereas the study of the properties and nature of radioactivity was highly successful, no real progress was made in the attempts to understand the cause of radioactivity. Nonetheless, it was a question that was considered significant and a natural part of the study of radioactivity. Walter Kaufmann, the eminent German electron physicist, discussed in 1901 "the source of energy as well as the whole mechanism of this phenomenon [radioactivity]" and concluded that the physicists were confronted with "a complete puzzle."[3] Two years later the first part of the puzzle had been largely understood, in the sense that most physicist agreed that the energy came from the atom itself rather than from some external agent. But the second part of the puzzle remained.

In a review article of early 1904 in his recently founded *Jahrbuch der Radioactivität und Elektronik*, Johannes Stark discussed the state of affairs in radioactivity with special emphasis on the question of the origin of the rays. He supported the still controversial transformation theory suggested two years earlier by Ernest Rutherford and Frederick Soddy, according to which radioactivity was a result of subatomic changes of a physico-chemical nature. However, although Stark recognized the Rutherford-Soddy theory as a great step forward, he also pointed out that it had nothing to say about the ultimate cause of radioactive change, that is, it lacked a microphysical explanation of radioactivity. There was one enigma left, the German physicist wrote: "This shadow is the question of the cause of the chemical atom's instability. Why do chemical atoms decay spontaneously, while they are permanent and immune to intensive thermal disturbances from the outside?"[4]

Stark realized that the answer might lie in the internal structure of the atom, and he suggested that the new atomic model proposed by J. J. Thomson might be able to throw light on the question. In his address of 1901, Kaufmann, too, refererred to the model, which he in a general sense characterized as the view that "all material atoms consist of conglomerates of electrons … whose different groupings would form the chemical elements, [so that] the old alchemists' dream of the transformation of the elements would be brought a good deal nearer realisation."[5] In fact, Thomson was greatly interested in radioactivity, which was an integrated part of his electron-atomic research program starting in 1897.[6] As early as 1898, in a discussion of X rays (which initially were not sharply distinguished from Becquerel's rays), Thomson suggested tentatively a mechanism for the rays emanating from uranium. Might it not be, he suggested "that in the case of a complicated structure like the uranium atom regrouping of the constituents of the atom may give rise to electrical effects similar to those which occur in ionization and might possibly be the origin of the uranium radiation?"[7]

Thomson's model of atomic structure, first vaguely suggested in 1897 and developed into a detailed atomic theory in 1903-04, assumed that a large number of electrons ("corpuscles" in Thomson's terminology) moved in concentric circular orbits in a positively charged but massless fluid of atomic dimensions.[8] The criterion for the acceptable electron configurations was that the electrons were in stable equilibrium positions. In part by calculations and in part by model experiments, Thomson deduced the number of electrons in the various rotating rings, assuming for simplicity that his atom was two-dimensional. However, the precise electron configurations were unimportant in his explanation of radioactivity. Thomson originally believed that the entire mass of an atom was made up of electrons, implying that there must be about a quarter of a million electrons in a uranium atom. Although experiments made in 1906 forced him to drastically reduce the number of electrons – and that by a factor of about one thousand – he kept the basic features of the atomic model until about 1910.

Thomson's view concerning the origin of radioactivity changed somewhat over the years, but not significantly. His favored view, developed from 1903 to 1907, was the radiation-drain hypothesis, which may be briefly sketched as follows. Because of the electromagnetic radiation emitted by the accelerating electrons, their angular velocity would gradually decrease and eventually reach a certain critical value. As a result, the configuration would become unstable and cause the electrons to rearrange into a structure with a larger kinetic energy. In Thomson’s words:

> There will be what is equivalent to an explosion of the corpuscles, the corpuscles will move far away from their original positions, their potential energy will decrease, while their kinetic energy will increase. The kinetic energy gained in this way might be sufficient to carry the system [of electrons] out of the atom, and we should have, as in the case of radium, a part of the atom shot off. In consequence of the very slow dissipation of energy by radiation the life of the atom would be very long.[9]

Thomson’s idea was not the only suggestion of the mechanism behind atomic earthquakes. Oliver Lodge, James Jeans, Jean Perrin, and Hantaro Nagaoka made their own suggestions about the origin of radioactivity, different from Thomson’s but sharing with it the basic idea of radioactivity being the manifestation of intra-atomic instabilities. These and other ideas were shortlived speculations, and it was only Thomson’s radiation-drain mechanism that enjoyed a more general support. For example, Norman Campbell claimed in 1907 that “[Thomson’s] view is now generally accepted.”[10] This might be an exaggeration, but if so it was not a gross one. Rutherford found the mechanism “probable” and discussed it in detail in his 1904 textbook on radioactivity entitled *Radio-Activity*. Moreover, in his 1906 Silliman Lectures Rutherford argued confidently that radioactivity would be explained in purely intra-atomic and causal terms by some future development of atomic theory. “We are almost forced to the conclusion that the α particle was originally in rapid motion within the atom and for some reason suddenly escaped from the atomic system with the velocity it originally possessed in its orbit," he wrote. Rutherford did not claim to know the reason for the escape, but he did not doubt that there was one: "A promising beginning has already ben made," he wrote, referring to Thomson's theory, "and there is every hope that still further advances will soon be made in the elucidation of the mystery of atomic structure."[11]

Soddy largely shared the views of Rutherford, although he cautiously avoided supporting Thomson's radiation-drain hypothesis. In his *Radio-Activity* – published the same year as Rutherford's book with the same title – Soddy stressed that the decay law was phenomenological and independent of particular ideas of atomic constitution. It would be true even if atoms were structureless Daltonian particles. The decay law, Soddy wrote, "greatly limits the field of speculation on the one hand, and, on the other, it raises new problems of its own which any satisfactory theory will have to account for." These qualities he found sadly missing in ideas concerning the origin of radioactivity, a problem scientists could speculate about but of which there was no theory: "It is not until we enquire as to the ultimate cause of radio-activity, and seek a knowledge of the forces at work which bring about the observed disintregration, that we enter a region to which the term *hypothesis* in the ordinary sense of a probable explanation would apply."[12]

The decay law and indeterminism

For reasons of convenience I shall refer to models explaining radioactivity microphysically and deterministically, such as Thomson's, as dynamical models. The Rutherford-Soddy decay law, well established by 1905, might seem to pose grave problems to such models. According to this law, the number of atoms that decay in a certain period of time depends only on the decay constant λ. If N_0 is the original number of atoms, the fraction of atoms that have decayed after a period t is given by

$$N(t) = N_0 \exp(-\lambda t)$$

Since λ depends only on the kind of the element (or isotope), this implies that the probability of decay is wholly independent of the age of the atom. But then, according to the dynamical hypothesis, all atoms formed at the same time should have the same lifetime, contrary to observation. As Lord Kelvin put it, in a letter to Thomson of 1906: "What would be the difference between radium atoms in a piece of radium bromide, of the performance of those 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?"[13]

Although not easily reconcilable with the dynamical hypothesis, the problem of the non-ageing atoms could be avoided by introducing suitable statistical assumptions, such as did Paul Langevin in 1904 and Thomson in 1909. Nor did Egon von Schweidler's important work on radioactive fluctuations, which included a purely statistical derivation of the decay law, cause the dynamical models to be discredited. Schweidler's thoroughly probabilistic approach led to a phenomenological understanding, but not to an *explanation* of radioactivity on a more fundamental, microphysical level. It was an approach that differed completely from Thomson's model-oriented approach and in principle made dynamical explanations superfluous.

However, probabilistic approaches à la Schweidler and dynamical-deterministic approaches à la Thomson are not necessarily irreconcilable. More to the point, contemporary physicists did not interpret the statistical nature of radioactivity as an indication of the inadequacy of dynamical models of lack of determinism in radioactive processes. Nor does this seem to have been implied by Schweidler, who wrote that "the atoms of an active substance are unstable systems with a 'mean life' determined by their structure,"[14] a view that Thomson would not have contradicted. That the decay law is not necessarily connected with lack of causality is furthermore illustrated if we consider that the law is not a consequence of radioactivity being an inherent property of the atom. As Rutherford pointed out in 1906, the decay law does not contradict the rival hypothesis (at the time considered unlikely) that radioactive instability is triggered by some external agent that then acts as the cause.[15]

Soddy's view of the matter further illustrates how the relationship between the complex atom, determinism, and the decay law was perceived in the first decade of the twentieth centure. The essence of the decay law was the constancy of λ, this "most fundamentally remarkable feature of radio-active systems."[16] According to Soddy, it precluded the idea that radioactivity occurred as the result of the electrons being configurated recurrently in certain unstable structures. For this would lead to a maximum life of any atom and to a definite number of atoms, rather than a definite fraction, decaying per unit period. All the same, Soddy accepted Thomson's idea that the components of the atoms must be in violent motion and that this was somehow the cause of radioactive decay:

> [The view] that rapidly recurring motions within the atom, giving rise to orientations exhibiting individual differences, but reverting to a

> general average in extremely short intervals of time after separation, can, at least, be entertained. ... The action appears to be due to "chance" – i.e., the orientation assumed at one instant has no determining influence on the orientation about to be assumed at the next instant. The conclusion is thus arrived at that the internal movements of the atomns must be highly irregular and cannot follow a definite sequence if the law of radio-active change is to hold good. The unstable position appears to be rather the result of a chance collocation of the parts than be due to the operation of any simple law.

Soddy seems here to argue for indeterminism, but I think this is not the way the quotation should be understood. That his reference to chance phenomena does not make him a probabilist in any strong sense is supported by the example he chose to illustrate a typical chance event, namely, the motion of molecules in a gas. Because of the enormous number of molecules it was impossible to determine the precise motion of individual gas molecules; but in principle it could be done, both in the case of a gas and in the case of radioactive atoms. This is how I think Soddy should be understood when he wrote: "The causes at work appear to be so complex that the results can only at present be described as 'chance' or 'accidental' happenings, in the sense of being impossible to predict." Had Soddy believed in strict indeterminism in the sense of the later quantum mechanics there would be no point in including the words "at present."

Radioactivity and statistical behavior

By the mid-1910s the Thomson model had been largely replaced by the nuclear atom, an important change in the history of atomic theory but not of great consequence with regard to possible explanations of radioactivity. The change merely meant that the atomic instabilities causing radioactive decay should no longer be sought in atomic electrons but in the motions of the nuclear particles. Dynamical explanations could be, and in a number of cases were, argued. Contrary to later physicists, the contemporaries of Thomson and Rutherford did not associate statistical behavior with lack of causality, and they did not consider probabilistic theory as an alternative to causal microphysical explanation. An early and rather typical view was that of Johannes van der Waals, who in an address of 1903 reflected on what he called the statistical view of nature.[17] According to the Dutch physicist, Boltzmann's statistical theory of heat and gases was perfectly compatible with deterministic behavior on the level of individual atoms and molecules. Physicists thought about radioactivity in a roughly similar way.

The general attitude with respect to the microphysical origin of radioactivity – a problem not demanding much attention after 1910 – was that it was a legitimate question to ask and one that could in principle be answered. It was considered somewhat peripheral compared with other problems of radioactivity, but not beyond solution and not a matter of great conceptual importance. The reason for the relaxed attitude was, I think, the general understanding of statistical phenomena that prevailed among most physicists. Consider the position of Max Planck, as he expressed it in a 1914 address on the relationship between dynamical and statistical laws in physics. "[It] appears ... at present hopeless even to guess at dynamical laws," he said, referring to the peculiarities of radioactive decay. Radioactivity could be accounted for statistically, but not dynamically. "How is this possible?" Planck asked. "How can physical laws be derived by considering phenomena the cause of which has, provisionally, to be left complety unexplained?"[18]

Planck, it should be noted, did not declare dynamical models of radioactivity hopeless in principle, but only "at present," and he considered the lack of a causal explanation to be provisional. The Polish physicist Marian von Smoluchowski, an expert in statistical phenomena, was more inclined than Planck to consider radioactivity a phenomenon beyond mechanical explanation. Yet he did not conclude from Schweidler's derivation of the decay law that radioactivity cannot be causally explained. Smoluchowski had no problem in constructing an *ad hoc* dynamical model of the radium atom that led to the decay law. Although he did not believe his model had anything to do with the real structure of the radium atom, he considered it instructive because it exemplified that "the decay ... can very well be produced by precisely defined, lawlike causes."[19]

Also the eminent mathematician Emile Borel referred to radioactivity in order to illustrate the difference between statistics in a strict mathematical sence and the statistical explanations used in physics. As he noted in 1920, the experimentally confirmed invariance of the decay constant of a certain substance may seem to be a proof of radioactivity being a purely indeterministic phenomenon on the atomic level, although ruled by "global determinism" on the macroscopic level. But, Borel pointed out, experiments are unable to decide between the indeterminism hypothesis and its alternative, namely, that the global determinism is the result of a huge number of deterministic events in which the lifetime of every single atom is precisely determined by the atom's constitution.[20] As far as Borel was concerned there was no need to conclude that radioactivity violated the determinism of natural laws. The kind of hidden determinism discussed as a possibility by Smoluchowski and Borel was illustrated by the mechanisms suggested qualitatively by Thomson and Langevin in the first decade of the century. In his report to the 1904 St. Louis Congres of Science and Arts, Langevin came up with the following suggestion:

> Perhaps the reorganization of the atomic structure might result from its accidental passage through a particularly unstable configuration, the probability that a like configuration should be reproduced being independent, in the mean, of the previous history of the atom, and the mean life of the latter would be short in proportion as this probability is great.[21]

In the 1910s, this line of argument was developed quantitatively by Frederick Lindemann, André Debierne, and a few others. Lindemann and Debierne shared with other physicists the belief that radioactivity was deterministic in principle, the probabilistic feature being the result of some averaging over a large number of processes, particles or states. For example, Debierne suggested that the central region of the atom consisted of “sub-atoms”, each of which included a large number of unspecified “elements.” The agitation of the elements would then create disorder among the sub-atoms “like the thermal agitation of molecules in a liquid creates the disorder of Brownian motion among the particles in a suspension.”[22] The analogy with Brownian motion, the characteristic example of deterministic random processes, was a favourite among the few physicists occupied with the origin of radioactive change. Although anachronistic, it may not be inappropriate to point out that the models of radioactivity in the Langevin-Debierne-Lindemann tradition bear a certain similarity to the much later quantum-mechanical models operating with hidden parameters as an alternative to the standard-indeterministic interpretation à la Copenhagen.[23]

In an important paper of 1918, Einstein formulated a quantum theory of radiation and remarked that "the statistical law which we assumed corresponds to that of a radioactive reaction."[24] On the assumption that Einstein took radioactivity to be a fundamentally statistical phenomenon, the remark has sometimes been interpreted as a recognition of the equally fundamentally statistical nature of radiation processes, a precursor of quantum indeterminism. However, as has been pointed out by historians of science, this is a misconception. Einstein's comment concerned a mathematical analogy and neither he nor other physicists at the time considered radioactivity to be a paradigmatic example of acausal processes such as later physicists would do.

Post-1915 explanation attempts

From about 1910 explanations of radioactivity in terms of dynamical models came to be seen as increasingly unimportant among mainstream atomic physicists. This was not because such explanations were considered suspect in principle, but rather because it was recognized that the insufficient knowledge of the constitution of the atom made it almost impossible to construct reliable models. In addition, there was no real need for such models, for the phenomenological and statistical theory provided the necessary connection between theory and experiment. All the same, the dream of explaining radioactivity never vanished completely from the scene of pre-quantum-mechanical physics. Between 1915 and 1925 a dozen physicists or so were occupied with the problem. Some of these explanations, such as those proposed by Hans Wolff, Georg Kirsch and William Harkins, were semi-quantitative and included elaborate and imaginative models of the atomic nucleus.[25] However, to give the flavour of this class of theories it suffices to look at a qualitative and very simple proposal of 1923.

At Niels Bohr's institute in Copenhagen, the young Norwegian physicist (and later prominent astrophysicist) Svein Rosseland suggested in 1923 a possible explanation of radioactivity based on the Bohr-Sommerfeld atomic model. His suggestion was inspired by an idea of Robert Pease, a physical chemist at Princeton University. Pease speculated that with the large number of electrons rotating with different periods about the nuclei of the radioactive element, "there will evidently come some times periodically when numbers of electrons in excess of the average will all be exerting attractive forces on the nucleus in the same direction." The result of such an atomic tidal effect might conceivably be "that a positively charged constituent of the nucleus might be drawn out of its normal equilibrium position and ... be sent on its path as an α-particle."[26] Rosseland criticized Pease's idea for not taking quantum theory into account, but his own model did not differ substantially from that of Pease. According to Rosseland, "it does not appear excluded that the presence of radioactivity among the heaviest known elements as well as the apparent absence of elements of higher atomic numbers may be connected with some sort of interaction between the nuclear and the external electrons."[27]

The remark should be seen in relation to the attempts at the time to find an explanation of why there exists only a limited number of chemical elements. Why is uranium with its 92 electrons the heaviest element found in nature? Several physicists tried to calculate the maximum atomic number and relate it to the cause of radioactivity, an interest shared by Bohr.[28] Although

Rosseland hastened to add that "our knowledge of nuclear structure is probably far too scanty to permit any definite conclusions," he clearly believed that the problem could be solved and did not associate it with any irreducible indeterminism. Since papers from the young scientists at the Copenhagen institute always had to be accepted by its director we may assume that Bohr agreed with Rosseland's conclusion. It is remarkable that the qualitative suggestions of Pease and Rosseland were in essentially the same spirit as the explanation of radioactive decay that Thomson pioneered almost twenty years earlier.

Concluding Remarks

The study of the history of causal mechanisms of radioactivity invites a couple of more general remarks. From a historiographical point of view it is obvious that it is anachronistic, hence unhistorical, to interpret the early history of radioactivity in accordance with the knowledge of a later generation, namely that radioactive decay is an acausal process. That this kind of anachronism can be found in textbooks in physics is understandable and even exusable. It is less exusable that they can be found in some historical works as well.[29]

As I have argued, in full agreement with J. van Brakel's valuable study of 1985, radioactivity's statistical nature was not interpreted as a failure of the principle of causality until after the emergence of quantum mechanics. When George Gamow proposed the first quantum-mechanical explanation of alpha radioactivity, including a derivation of the empirical Geiger-Nuttall law, he did not comment on the relationship between the in principle statistical quantum mechanics and the phenomenologically statistical radioactivity. For such a comment we must look to Ronald Gurney and Edward Condon, who derived the theory of alpha radioactivity almost simultaneously with Gamow. In 1929, referring to earlier attempts to make sense of the statistical nature of radioactivity, they wrote:

> This has been very puzzling so long as we have accepted a dynamics by which the behaviour of particles is definitely fixed by the conditions. We have had to consider the disintregration as due to the extraordinary conjunction of scores of independent events in the orbital motions of nuclear particles. Now, however, we throw the whole responsibility on to the laws of quantum mechanics, recognizing that the behaviour of particles everywhere is equally governed by probability.[30]

That is, from the point of view of quantum mechanics radioactivity is a statistical phenomenon simply because it is governed by the inherently statistical laws of quantum mechanics. The decay of a particular radium atom cannot be precisely predicted, not because it is a radioactive phenomenon but because it is a quantum phenomenon. As indicated by Gurney and Condon, the quantum physicists of the 1920s had to make statistical assumptions in order to reproduce the phenomenological laws of radioactivity, apparently in the same way as Thomson, Langevin and others twenty years earlier. But the difference in methodology is crucial: Quantum mechanics makes use of one general assumption, namely, that the theory applies to the atomic nucleus; the physicists in the Thomson tradion had to make very special, *ad hoc* assumptions that did not apply to domains outside the atom.

The importance of radioactivity as a key instance of statistical quantum mechanics is further exemplified by Einstein, who in 1949 used the decay of radioactive atoms to illustrate his dissatisfaction with quantum mechanics, according to which "the ψ–function does not imply any assertion concerning the time instant of the disintegration of the radioactive atom." Einstein considered this a grave deficiency and argued that it implied that the quantum mechanical description must be incomplete. He was convinced that a complete description of a single atomic system was possible and realized that "for such complete description there is no room for the conceptual world of statistical quantum theory."[31]

NOTES

[1] Peter B. Medawar, *The Art of the Soluble* (London: Methuen, 1967).

[2] The present paper builds on and develops ideas in H. Kragh, "The origin of radioactivity: from solvable problem to unsolved non-problem", *Archive for History of Exact Sciences* 50 (1997): 311-358. The subject is not virgin land to the historian of science and for this reason I deal with it in a more general perspective. For earlier treatments, see Abraham Pais, "Radioactivity's two early puzzles", *Reviews of Modern Physics* 49 (1997): 925-38; Edoardo Amaldi, "Radioactivity, a pragmatic pillar of probabilistic conceptions" in G. Toraldo di Francia, ed., *Problems in the Foundations of Physics* (Amsterdam: North-Holland, 1979), pp. 1-28. J. van Brakel, "The possible influence of the discovery of radio-active decay on the concept of physical probability", *Archive for History of Exact Sciences* 31 (1985): 369-85; Anne C. van Helden, "Modellen voor radioactief verval (1911-1928)", *Tijdschrift Geschiedenis der Geneeskunde, Naturwetenschapen, Wiskunde en Techniek* 11 (1988): 1-11; and M. G. Ianniello and F. Sebastiani, "La legge esponenziale del decadimento radioattivo", *Physics* 29 (1992): 711-808.

[3] W. Kaufmann, "Die Entwicklung der Elektronenbegriffs", *Physikalische Zeitschrift* 3 (1901): 9-15, on p. 14. Lecture delivered before the 73rd meeting of German

Scientists and Physicians in Hamburg. An English translation appeared as "The development of the electron idea", *The Electrician* 48 (1901), 95-97.

[4] J. Stark, "Vorgeschlagene Erklärungen der Radioactivität", *Jahrbuch der Radioactivität und Elektronik* 1 (1904): 70-82, on p. 80.

[5] Kaufmann, "Die Entwicklung der Elektronenbegriffs" (note 3), p. 15.

[6] For details, see Steve B. Sinclair, "J . J. Thomson and radioactivity", *Ambix* 35 (1988): 91-104, 113-126.

[7] J. J. Thomson, "On the diffuse reflection of Röntgen's rays", *Procedings of the Cambridge Philosophical Society* 9 (1898): 393-97, on p. 397.

[8] J. J. Thomson, *Electricity and Matter* (London: Constable & Co., 1904. For historical analysis, see John L. Heibron, "J. J. Thomson and the Bohr atom", *Physics Today* 30 (1977): 23-33 and H. Kragh, "J. J. Thomson, the electron, and atomic architechture", *The Physics Teacher* 35 (1997): 328-32.

[9] J. J. Thomson, "On the structure of the atom", *Philosophical Magazine* 7 (1904): 237-65, on p. 265. See also Thomson's letter to Rutherford of February 18, 1904, as cited in Per F. Dahl, *Flash of the Cathode Rays: A History of J. J. Thomson's Electron* (Bristol: Institute of Physics Publishing, 1997), p. 325.

[10] N. R. Campbell, *Modern Electrical Theory* (Cambridge: Cambridge University Press, 1904, p. 283.

[11] E. Rutherford, *Radio-Activity* (Cambridge: Cambridge University Press, 1904), pp. 316-22, 338-42. Rutherford, *Radioactive Transformations* (London: Constable & Co., 1906), p. 42 and p. 265.

[12] F. Soddy, *Radio-Activity: An Elementary Treatise from the Standpoint of the Disintegration Theory* (London: The Electrician, 1904), p. 55.

[13] Kelvin to Thomson, 12. November 1906, as reproduced in Lord Rayleigh, *The Life of Sir J. J. Thomson* (Cambridge: Cambridge University Press, 1942), p. 141. A similar point was made in Rutherford, *Radioactive Transformations* (ref. 11), p. 267.

[14] E. R. Schweidler, "Über Schwankungen der radioaktiven Umwandlung", *Comptes Rendus du Premier Congrès International pour l'Etude de la Radiologie et de l'Jonisation* (Brussel, 1905), p. 1.

[15] Rutherford, *Radioactive Transformations* (ref. 11), p. 267.

[16] Soddy, *Radio-Activity* (ref. 12), p. 179, which is also the source of the following

quotations.

[17] J. D. van der Waals, "Die statistische Naturanschauung", *Physikalische Zeitschrift* 4 (1902-03): 508-14.

[18] M. Planck, "Dynamische und statistische Gesetzmässigkeiten", in Planck, *Vorträge und Erinnerungen* (Darmstadt: Wissenschaftliche Buchgesellschaft, 1983), pp. 81-94, on p. 83.

[19] M. von Smoluchowski, "Über den Begriff des Zufalls und den Ursprung der Wahrscheinlichkeitsgesetze in der Physik", *Die Naturwissenschaften* 6 (1918): 253-263.

[20] E. Borel, "Radioactivité, probabilité, déterminisme", *Revue du Mois* 21 (1920): 33-40. That any statistical phenomenon can in principle be explained by a suitably chosen dynamical model is shown in Robert B. Lindsay and Henry Margenau, *Foundations of Physics* (New York: Dover, 1975), pp. 190-92.

[21] P. Langevin, "The relations of physics of electrons to other branches of science", in Katherine R. Sopka and Albert E. Moyer, eds., *Physics for a New Century: Papers Presented at the 1904 St. Louis Congress* (New York: American Institute of Physics, 1986), pp. 195-232, on p. 223.

[22] A. Debierne, "Considerations sur la mécanisme des transformations radioactives et la constitution des atomes", *Annales de Physique* 4 (1915): 323-45, on p. 338. Lindemann's theory is analyzed in Amaldi, "Radioactivity" and van Helden, "Modellen".

[23] See, for example, the contributions of David Bohm and Jean-Pierre Vigier in Stephan Körner, ed., *Observation and Interpretation in the Philosophy of Physics* (New York: Dover, 1957).

[24] A. Einstein, "Zur Quantentheorie der Strahlung", *Physikalische Zeitschrift* 18 (1918): 121-130, on p. 129. The relationship between radioactivity and Einstein's theory is discussed in van Brakel, "The possible influence" (ref. 2) and Amaldi, "Radioactivity" (ref. 2).

[25] Roger H. Stuewer, "The nuclear electron hypothesis", in William R. Shea, ed., *Otto Hahn and the Rise of Nuclear Physics* (Dordrecht: Reidel, 1983), pp. 19-67.

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[28] H. Kragh and Bruno Carazza, “A historical note on the maximum atomic number of chemical elements”, *Annales de Fondations Louis de Broglie* 20 (1995): 207-215.

[29] E.g., Max von Laue, *Geschichte der Physik* (Frankfurt am Main: Ullstein, 1950), pp. 122-24, Abraham Pais, *Inward Bound: Of Matter and Forces in the Physical World* (Oxford: Clarendon Press, 1986), p. 121, and Philip Stehle, *Order, Chaos, Order: The Transition from Classical to Quantum Physics* (New York: Oxford University Press, 1994), p. 209. According to Stephen Brush, in the first two decades of the twentieth century the theory of radioactive decay led scientists to think that “the determinism or causality characteristic of the Newtonian world-machine ... was slipping away.” S. G. Brush, *Statistical Physics and the Atomic Theory of Matter* (Princeton: Princeton University Press, 1983), p. 127. However, although there was indeed a trend against determinism and causality in the period, it seems not to have been nourished by the science of radioactivity.

[30] R. W. Gurney and E. U. Condon, “Quantum mechanics and radioactive disintegration”, *Physical Review* 33 (1929): 127-40, on p. 134.

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Centre for Science Studies
University of Aarhus
Ny Munkegade 120, building 1520
DK-8000 Aarhus C
DENMARK
`www.css.au.dk`

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Department of Science Studies
AARHUS UNIVERSITET