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Radioactivity

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IN the year 1896, which fell near the beginning of the great transformation of modern physics, Henri Becquerel heard that Roentgen had discovered strange rays proceeding from an electric discharge-tube while the discharge was passing and the glass walls of the tube were phosphorescing. Suspecting that the new rays were connected with the phosphorescence, Becquerel tested samples of some of the substances which naturally phosphoresce. It happened that one which he tested was a compound of uranium. He wrapped the sample in paper to shut in the light of its phosphorescence, and set it beside a photographic plate; for the rays of Roentgen had disclosed themselves by acting on such plates. Becquerel had made a happy guess; for the compound affected the plate. Yet his original idea was altogether wrong; for the effect had nothing to do with the phosphorescence of the compound, it was due to the uranium itself and faithfully reappeared when other and non-phosphorescent compounds were used instead, and even when a piece of the pure metal was set beside the plate. It was an instance of a fallacious idea having guided a keen observer to a great discovery—not the first in the history of physics, and assuredly not the last.

Thereupon Pierre and Marie Curie, having verified that the effect of any quantity of any compound of pure uranium is strictly proportional to the amount of uranium in it, noticed that the effect of certain natural rocks and minerals containing uranium was much greater than that which their content of the metal should produce. Suspecting that there was some constituent of the rocks having the same property as the uranium but in a degree much greater, they set about the task of getting the uranium and the inert matter out of the way and isolating the more potent substance. It was a long task; to speak of "winnowing" the pile of rock would be to use a comically feeble metaphor, and as for the proverbial needle in a haystack, it could have been extracted with incomparably less trouble than the few hundredths of a gramme of the active substance which were latent in the ton of raw material. Eventually the Curies did liberate it, or rather them, for there were several active substances; and one of them was named radium, and their strange property was called radioactivity. This was the first of the words containing the magic syllables *radio*, syllables which are one of the special symbols of our epoch; were the

literature of these times to disappear all but a few scraps, posterity could date them by the appearance of that word, as Latin manuscripts are dated through containing some word or some trick of style that came into use at a definite moment of history. A word must bear almost magical connotations, to enter so thoroughly into popular usage; and the phenomena of radioactivity endowed it with these in abundance, with suggestions of rays piercing all matter, and inexhaustible stores of energy, and transmutation of elements, and influences having power even over life and death. Wonderful to relate, the suggestions for once were justified by the truth.

From 1898 onward there was a tremendous rush of investigators into the new field, and in a few years there were explorers of almost every conceivable aspect of radioactivity—chemists ascertaining the chemical properties of the radioactive elements. physicists observing their physical properties, and a great host of students investigating the numerous and striking effects of the rays. The subject presently became so wide that books on radioactivity written before and during the War resemble treatises on the contemporary physics of their dates of publication; for the new rays seemed to be able to invade all the provinces of physics as easily as they could penetrate matter in all its forms.

Eventually, however, it became clear that many of the topics classed at first with radioactivity should be removed into other fields of science. The radioactive elements all have their places in the Periodic Table, and their chemical properties are what should be expected from elements thus placed; peculiar as radium is in its one famous feature, there is nothing abnormal about its chemical reactions, and they may justly be relegated to the handbooks of chemistry and to the manuals written for those who wish to prepare or purify the element. The same thing is true of the physical properties of radium; nothing in its optical spectrum suggests that it is other than an ordinary member of the second column of the Periodic Table, nothing in its X-ray spectrum intimates that it is more than just the 88th member of the Procession of the Elements. None of these needs to be taken into account in the study of radioactivity.

The various effects of the rays which the radioelements emit are likewise quite irrelevant. At the beginning it was natural and proper for every writer to describe all that was known of the actions of the alpha-rays, the beta-rays and the gamma-rays, after having said that these are the three kinds of rays which radioactive substances emit. Indeed it was essential, for at first there was no way of defining the rays, much less of ascertaining their real nature, except by considering *en bloc*

everything that was known about their actions. This condition prevails no longer. It is established that alpha-rays are atoms of helium each bearing a charge $+2e$; that beta-rays are electrons, that gamma-rays are composed of electromagnetic radiation. Information about the first two belongs to the vast body of doctrine concerning the properties of fast-flying electrified particles; information about the last belongs to the science of the properties of radiation. I do not mean to imply that the information is redundant. One can produce in the laboratory fast-flying electrons, but none so fast as the fastest beta-rays; swift positively-charged atoms, but none nearly so swift as the alpha-particles; electromagnetic waves of many wavelengths, but none nearly so short as the shortest to be found among gamma-rays. The knowledge acquired from studying the properties of the rays is exceedingly important, and if the radioelements had not been discovered, most of it would not been acquired so early, and some of it would still be unattainable; but it is not knowledge of radioactivity.

What then is knowledge of radioactivity? So far as now appears, we know all that can be known about the radioactivity of a radioelement if we know what are the speeds of the alpha-particles emitted from it, if any; what are the speeds of the electrons emitted from it, if any; what are the wavelengths of the electromagnetic waves which it emits, if any; how many of each kind of particle (for we may speak of the waves as particles also, meaning by "particle" a quantum) are emitted from a given number of atoms in a given time; and what element or elements result from these processes. Apparently, if we could know all of these things for a particular radioelement, we should know everything which determines its peculiar actions upon the outside world. This unfortunately is not the same thing as being able to solve the problems of predicting all of these actions or understanding them; but these problems are now transferred out of the field of radioactivity into the field of the science of fast-flying charged particles and short-wave radiation. Let us leave them there, and restrict the field of radioactivity to the speeds of the particles and the frequencies of the waves which issue from each radioelement, and the rates at which they come forth, and the condition of the atoms they leave behind.[1]

[1] The specific statements made in this article are derived chiefly from three recent synopses of the data of radioactivity: the National Research Council bulletin *Radioactivity*, by A. F. Kovarik and L. W. McKeehan; the *Manual of Radioactivity*, by G. v. Hevesy and F. Paneth; and the relevant articles by St. Meyer, L. Meitner, W. Bothe and O. Hahn in volume 22 of the new Geiger-Scheel *Handbuch der Physik*. As these are all well supplied with bibliographies (and so likewise, I presume, is the new edition of Meyer and von Schweidler's *Radioaktivität*) I have omitted references to individual papers except a few published since 1923. At several places I venture to refer under the name " Introduction " to my *Introduction to Contemporary Physics* for topics not falling within the field of radioactivity as here defined.

During the composition of this article I have had the advantage of frequent consultation with my colleague Dr. L. W. McKeehan.

Already in expressing these restrictions, certain principles of radioactivity have been implicitly assumed; it is necessary to state them explicitly.

In the first place, I have spoken of the radioactivity of the elements alone; this is permissible, because radioactivity is definitely a property of individual elements. This does not mean merely that radioactivity is a property of a limited number of elements in certain states and a limited number of compounds of these and other elements, as seems to be true of ferromagnetism. It means that wherever there is a particular radioactive element, free or compounded, gaseous or liquid or solid, the characteristic rays of that element are emitted in a degree proportional to the amount of the element and not affected in the least by its condition or its state of combination. A given amount of radium emits the same kinds of rays at the same rate whether it is a piece of pure metal, or is combined with chlorine in radium chloride, or with sulphur and oxygen in radium sulphate. A given amount of radon emits rays of the same sort at the same rate whether it is gaseous as at normal temperatures, or frozen by submerging its enclosing tube in liquid air. Samples of some of the radioelements have passed through combination after combination in the chemical laboratory, being released from one compound only to enter into another; their activity was meanwhile being measured by the most delicate available tests, but it was never found to be affected in any perceptible degree. There is no other property of an element, excepting mass, of which this can be said without reservation.[2]

The indifference of radioactivity to the state of combination of the elements which display it extends also to all their other circumstances. In modern laboratories it is feasible to subject pieces of matter to very powerful, severe and violent agencies; heat enough to melt any element, cold enough to freeze any substance, electric fieldstrength high enough to tear electrons out, high magnetic fields, intense illumination, bombardment by multitudes of fast moving charged particles— and all of these have been tried to some extent, some to the utmost humanly possible extent, upon radioactive elements; but in every instance the radioactivity has remained constant without detectable variation, inaccessible and immune to all the powers within human control or under human observation.[3]

[2] It can be said of the higher-frequency emission-lines and absorption-edges of the X-ray spectra of the elements, but not unreservedly; for since the lower-frequency lines and edges of an element do vary slightly but perceptibly when its state of combination is altered, there is a strong presumption that the higher-frequency spectra will likewise be found to vary as soon as the accuracy of the measurements is increased say five- or tenfold.

[3] Influences of sunlight upon radioactivity are reported now and then in the *Comptes Rendus;* but it seems exceedingly unlikely that something immune to every other known agency should be susceptible to this particular one.

Sooner or later, in expounding almost any topic in physics, one arrives at a place where the introduction of an atom-model greatly simplifies what remains to be said. In the present article, this is the place.

Physicists commonly employ an atom-model in which a certain number of electrons are arranged around a nucleus bearing a charge equal in magnitude and opposite in sign to the sum of their charges. For any particular element the number of electrons assigned to its atom-model is equal to its atomic number, which can be obtained from any modern chart of the Periodic Table. In such a chart the elements are arranged in the order of their atomic numbers from 1 to 92, composing what I shall call the *procession of the elements*—a procession from which only two are now missing. In dealing with an element of high atomic number—all of the radioactive elements are of this character, ranging in atomic number from 81 upwards [4]—the electrons are assigned to various locations, some being close to the nucleus and others intermediate and others at the periphery of the atom-model. In fitting the various regions and divisions of the atom-model to the various properties of the element which it represents, the outermost electrons are assigned to the task of accounting for those properties which vary exceedingly with the state of chemical combination and with the other circumstances of the element; for being at the surface of the atom they should be most exposed to outer influences. The inner electrons, being partly shielded, are used to account for such properties as the X-ray frequencies, which depend so little upon the circumstances of the element that their variations are scarcely perceptible or not at all. The nucleus is the best shielded of all, and it receives for its quota the two properties which within the accuracy of experiment are immune from change—radioactivity and mass.

There are additional reasons for assigning mass and radioactivity to the nucleus. As for the mass: since the sum of the masses of the electrons constituting an atom-model never attains 1/1800 of the known mass of the atom, the balance which the nucleus must take is practically the whole of it. Again, there are experiments which show that a single chemical element may have several kinds of atoms differing in mass and yet quite alike in chemical properties, in their line-spectra, in their X-ray spectra; since these similarities require that the same nucleus-charge and the same number and arrangement of electrons be imposed upon all these atoms, the outstanding difference in their masses must be ascribed to their nuclei.[5] Again, there are slight

[4] Except potassium and rubidium (compare footnote 13).

[5] *Introduction*, pp. 29–39, 65–66.

differences between the band-spectra of compounds involving such atoms, which are well explained by attributing to the several atoms identical nucleus-charges and electron-systems, but nucleus-masses standing to one another in the same ratios as the observed masses of the atoms do.[6]   But I must not give all the evidence for the nuclear atom-model, or this article will be swamped.

Among the reasons for ascribing radioactivity to the nucleus, the primary one has already been introduced—radioactivity, like mass, is unalterable; and another has already been stated, though without mentioning its relevance to this question.   Certain radioactive elements emit charged atoms of helium; and since outside of the nucleus nothing except electrons is provided in the atom-model, these charged atoms must be supposed to proceed out of the nuclei.   This argument could not be used upon the radioelements which emit electrons; but even for these there are reasons for suspecting that some of the electrons which issue from them do not come out of the family of electrons surrounding the nucleus, but from some other place.   For instance, it is possible and usual to pry electrons out of various locations in the circumnuclear family; but when this is done, the resulting "ionized" atoms promptly take in one electron or as many more as they have lost, and revert to their original state and nature.   This does not happen with the radioactive atoms which emit beta-rays; the departure of the electron effects an irreversible change, the atom is altered for good and all.   It does not however acquire a permanent positive charge; it takes on an electron and makes good its loss of charge. This is best explained by supposing that the original atom lost an electron originally located in the nucleus, and added one to the circumnuclear family, keeping its net charge equal to zero but undergoing a rearrangement of its charges.

By accepting the idea that certain of the charged particles emerging from a radioactive element issue from the nuclei of its atoms, it is possible to express and explain very simply a celebrated law of radioactivity which was discovered by Fajans and Soddy in the early days of the nuclear atom-model and helped greatly to establish it.

When an atom of a radioelement of atomic number Z emits an alpha-particle with its charge $+ 2e$, its nuclear charge diminishes by that amount.   It becomes an atom with nuclear charge $(Z - 2)e$ and Z electrons.   The diminished nuclear charge cannot hold the entire electron-family; two of its members depart, and the atom becomes an atom of nuclear charge $(Z - 2)e$ and $(Z - 2)$ circumnuclear or orbital electrons.   The radioelement changes into an element two steps farther down in the procession of the elements.

[6] *Introduction*, p. 400.

This is the first half of the displacement-law of Fajans and Soddy. It signifies that *the emission of α-particles by a radioelement is the sign of a transmutation of that element into the next but one of those preceding it in the procession of the elements.* If the properties of this latter element are known already, the law can be tested with all the accuracy desired. Polonium stands two places after lead in the procession; it emits alpha-particles; its atoms should turn into atoms possessing all the chemical qualities of lead, and they do. If a radio-element which lies two steps ahead of an element not previously known is discovered to emit alpha-particles, we are still not without information as to the qualities of the element into which it should transmute itself. For if we know the column of the Periodic Table in which the original element lies, we know also the column in which the element two steps ahead of it should lie; and the chemists know what features are common to all the known elements of that column and presumptively extend also to the unknown one. Radium lies in the second column of the Periodic Table; it emits alpha-particles; it should be transmuted into an element lying in the "zero" column. That element was not known until after radium was discovered; but it was known that the other elements of the zero column are inert gases, and consequently that the one into which radium transmutes itself should be an inert gas. This is verified; and as a general rule it is verified that when a radioelement emits alpha-particles the substance left behind possesses the particular chemical features of the elements belonging to that column of the Periodic Table to which the element two steps preceding the original one belongs. From this fact of experience it is only a short step to the first part of the Fajans-Soddy displacement-law—and a step which is put quite beyond criticism by the relations presently to be cited which connect the atomic weights of the radioelements.

The second half of the law relates to the other radioelements, those which eject electrons from their nuclei. When an atom of atomic number Z emits an electron from its nucleus, the nuclear charge increases to $(Z + 1)e$, which is sufficient to hold another electron beyond the Z electrons of the original family. The atom does pick up another electron which enters into the circumnuclear set (*not* into the nucleus); and it becomes an atom of nuclear charge $(Z + 1)e$ and $(Z + 1)$ orbital electrons. *The radioelement changes over into another which is one step farther up the procession of the elements.* This is the second half of the displacement-law of Fajans and Soddy.

The evidence for this second part is extensive; but on the whole it is not so imposing as the evidence for the first part. Largely this is

due to the difference between the two types of emission. Alpha-particle emission is violent and unique; positively-charged particles moving with a speed like theirs are not produced in any other way known to man. Beta-particle emission is considerably less violent, and there are so many known processes for producing fast-flying electrons that one must always keep in mind the possibility that some of the electrons proceeding from radioelements may be due to one or another of these; in fact, many certainly are. Perhaps the best way to state the evidence is this: every radioelement which does not emit alpha-particles transmutes itself into an element lying one step farther up the procession,[7] and all but one (actinium) of these elements is known to emit electrons, all of which agrees with the assumption that in each of these transmutations one electron is extruded from each participating nucleus. Stated thus, it may not sound very convincing; but if the second part of the Fajans-Soddy law were not true, we should hardly have failed thus far to find something definitely inconsistent with it.

Were gamma-rays without an accompanying beta-particle or alpha-particle to be emitted from a nucleus we could scarcely call the result a transmutation, since it would not affect the nuclear charge nor the electron-family of the atom. There is no reason for denying that this might happen; but I am not aware that it is known ever to happen, except in cases of nuclei which have just previously undergone a transmutation—cases which we shall eventually examine.

If now each radioelement is passing over into another element, one step before it or two steps behind it in the procession according as it emits beta-rays or alpha-rays—then it must be possible to draw up *genealogies* of radioelements, series of elements of which each member is transmuted out of the foregoing and transmutes itself into the following one. All of the known radioelements fall into one or another of several such series. To represent all these relations, and one more, it is convenient and suitable to draw a graph in which the atomic numbers of the elements are laid off horizontally, and their atomic weights are laid off vertically. Each element is represented by a point upon this graph; when the element transmutes itself it moves to another point, two units to the left if an alpha-particle is emitted and one to the right if the change is a beta-ray change. Now the emission of an alpha-particle involves the departure of four units of mass from the nucleus which it leaves; the loss of an electron however involves a loss

[7] More precisely, into an element having the chemical features distinguishing the column of the Periodic Table containing the element lying one step farther up the procession than the original one.

of only 1/1850 of a unit of mass, which is quite inappreciable.[8]   Hence in a transmutation of the former sort, the point representing the element in the graph moves four units downward as well as two to the left; in one of the latter sort, the point simply slides horizontally to the right.   The meaning of Fig. 1 will now be clear.
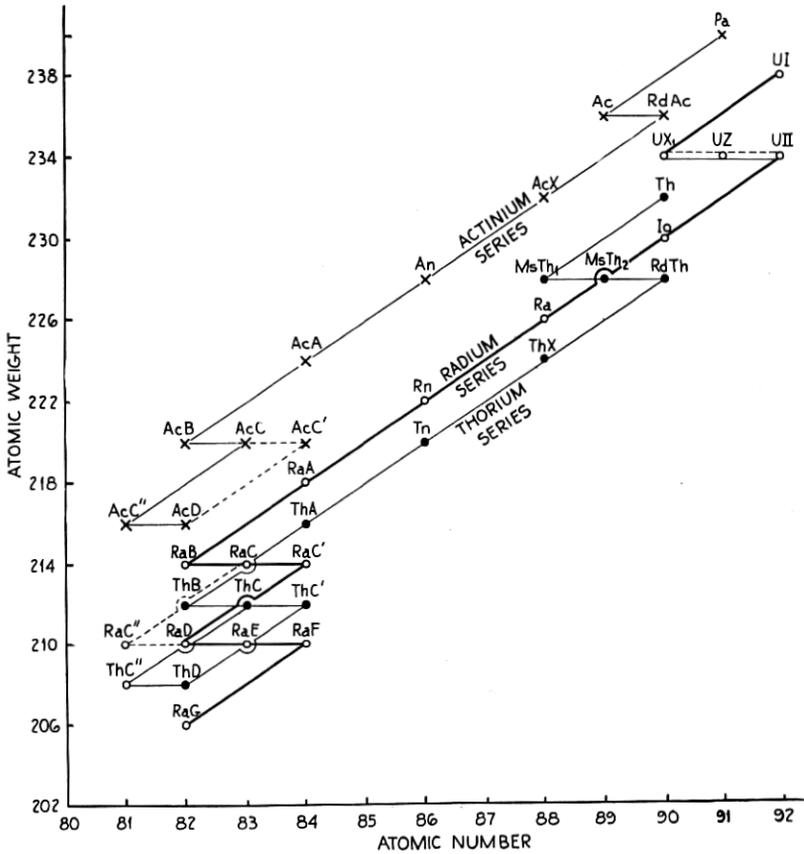

Fig. 1.   Genealogies of the radioelements

(The actinium series is plotted some distance above the others for legibility, but should almost certainly lie lower.)

The lines in Fig. 1 which represent the family trees of the radioelements descend in zigzags, which signifies that the "decline and fall"

[8] These masses are given in terms of the unit of mass in which atomic weights are measured, of which 16 constitute the mass of an oxygen atom.   Were the mass of an electron appreciable in these measurements, we should have to allow for the electrons added to or lost from the circumnuclear family to balance the change in the nucleus-charge.   But then we should also have to make decisions about the mass to be assigned to the energy carried away by the particles and the waves.

of a radioactive atom does not proceed continually downward to lower and ever lower atomic numbers, but is interrupted by occasional partial recoveries. Whenever there are three consecutive transmutations of which two involve the emission of beta-rays and one that of alpha-rays, the element in which the third ends has the same atomic number as the element from which the first originates. In each of the three lines of descent made visible in Fig. 1 there are instances of this; the one including radium for instance touches three times at atomic number 82, the one commencing with thorium twice. In the sense in which I have thus far used the word "element," the element 84 recurs three times in one series and twice in the other. Here is an ambiguity which the time has come to dispel.

The ambiguity in the use of the term *element* is a question of words, but not wholly a linguistic, much less a trivial one; it is such a question as arises when a field of knowledge is expanded and enriched to such an extent that its old vocabulary ceases to be adequate. This particular question arose after the discovery that certain substances differing in radioactivity are very much alike in their chemical properties—another of the facts which the atom-model is especially adapted to explain.

Consider, for an example, the three elements radium B and radium D and radium G, which lie upon the same line of descent, the "radium series." The first transmutes itself into the second, and the second transmutes itself into the third, each in a three-stage process involving the departures of two electrons and an alpha-particle (with the order of their exits we are not now concerned) from the nucleus. The mass of the third is four units less than that of the second and eight units less than that of the first; in radioactivity also they differ. But all three are alike in nuclear charge, and hence in the size and presumably in the arrangement of their circumnuclear families of electrons; and hence the presumption arises, that in their physical and chemical properties apart from radioactivity and mass they should be quite alike.

This presumption about the chemical properties is confirmed by the fact that RaB and RaD and RaG cannot be separated from one another by chemical means after they are once mixed. In general, whenever two of these "radioelements" coinciding in atomic number are subjected to any of the very considerable variety of agents in the chemists' armory, they respond in so nearly, if not exactly, the same way that there is no method known for taking one and leaving the other. Crystallization out of a mixture of salts of two such elements merely produces crystals containing the two salts in the same proportion as the liquid; sublimation merely produces a deposit containing the elements

in their original ratio; electrolysis does not favor one above the other, nor does osmosis, and if a mixture of the two elements or of salts of theirs is presented to an absorbent or an adsorbent or a solvent, it accepts them in the same proportions as they are presented, while any element willing to react with either reacts in precisely the same degree with the other.[9] Similarity such as this goes far beyond the interresemblances of the alkali metals, for instance, or even those of the rare-earth elements, difficult as the task of separating these latter from one another sometimes proves; it is not similarity merely, it amounts to identity.

As for the presumption that radioelements sharing the same atomic number should be alike in what are loosely termed the "physical properties," it is more difficult to test.   In fact, there seems to be no instance of two such elements, each radioactive and each obtainable quite unmixed with the other in quantities large enough for such experiments.   The three elements sharing atomic number 86 are all gaseous at ordinary temperatures, but they are too scanty and two of the three are much too fugitive for making accurate comparative measurements of such qualities as viscosity or elasticity or ionizing potential.   The elements sharing atomic number 82 are, as I shall presently bring out, mostly stable, and upon them it is possible to test the expected coincidence in optical line-spectra and X-ray spectra, which is verified except for certain very minute (but unexplained!) differences in the wavelengths of certain lines.   The band-spectra of these elements (more precisely, of their compounds) display slight differences which are beautifully explained by the contemporary theory of band-spectra, involving as it does a participation of the nucleus with its mass in the production of the bands.[10]   Mixtures of two of the elements sharing atomic number 90 (thorium and ionium) display precisely the same optical spectrum as pure thorium.[11]   In addition to these observations, a great many have been made upon the physical

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[9] There is a huge literature of the attempts to separate elements of identical atomic number and to discriminate between their chemical properties, for a review and bibliography of which I refer again to von Hevesy and Paneth (*l.c. supra*, chapter XII).

[10] E. S. Bieler, *Nature*, **115**, p. 980 (1925).

[11] This is vividly illustrated by a passage in the classical treatise upon radioactivity which Rutherford wrote in 1912.   Boltwood had isolated from uranium ores a sample of thorium oxide which emitted, along with the alpha-particles from the thorium, a considerable number coming from ionium.   Russell and Rossi produced its arc spectrum and "the spectrum of thorium was obtained, but not a single line was observed that could be attributed to ionium.   On the assumption that ionium has a life of 100,000 years, the preparation should have contained 10% of ionium.   Since probably the presence of 1% of ionium would have been detected spectroscopically, it would appear that the ionium was present in small amount, indicating that the life of ionium must be much less than 100,000 years."   As a matter of fact there was probably more than 10% of ionium in the mixture; but its spectrum lines were identical with those of thorium.

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properties of other non-radioactive elements which share particular atomic numbers and are mixed together in varying proportions; and they establish that such "elements" are indistinguishable except in such properties as are influenced to a measurable extent by the mass of the nucleus.

These facts make it necessary to redefine the word *element*, which in its long journey through the centuries from Lucretius has modified its meaning time and time again to keep pace with the gradual refinement of scientific thought, though all the while it kept its spelling intact. These are the alternatives: *either* to confer the status of a separate "element" upon each substance (apart, of course, from the compounds!) possessing a distinctive mass and radioactivity of its own, so that there may be several distinct elements sharing a given set of chemical properties— *or* to link the term "element" with a characteristic set of chemical and physical properties, with a specific atomic number and position in the Periodic Table, so that a given element may be an ensemble of several different kinds of matter differing in radioactivity or mass or both.   Reasons of science require that one or the other of these alternatives be chosen, but the actual choice is determined by reasons of language and expediency.   These reasons—I will not pause to develop them—favor the second alternative.   Inconvenient though it may be to refer to RaB and RaD and RaG and ThB and several other radioactive substances as the same element, the inconveniences entailed by the other policy would in the end be immensely greater.   One element to each atomic number, one place in the Periodic Table to each element— this is the choice which the prior usage and the associations of the word *element* recommend; and some other name must be selected to distinguish the substances which share a common atomic number but differ in mass or radioactivity or both.

Such a name is Soddy's word *isotope*, constructed out of Greek words to signify "in the same place."   Radium B and RaD and all the other substances which appear in the column labelled "82" in Fig. 1 are isotopes of the element 82; radon and thoron and actinon are isotopes of the element 86.   In these eleven places of the Periodic Table extending from 81 to 92, the individual isotopes enjoy names of their own, the elements are best known by their numbers.   The names *thallium, lead, bismuth* and *uranium* are, it is true, generally attached to the elements 81, 82, 83 and 92; but the first three of these names are used by some people to mean the elements in question and by others to designate only those of their isotopes which are not radioactive, and there is danger of confusion.[12]   Elsewhere in the Periodic Table, where

[12] The names *polonium, radium, actinium, thorium* and *protactinium* signify par-

all the isotopes of each element are stable, the elements have individual names and the isotopes are designated only by their masses. The elements 81, 82 and 83 have some isotopes which are radioactive and others which are not; thus the word "radioelement" is misleading, and should be replaced by "radioactive isotope." Consistency indeed requires that one speak of the successive members of a family of radioactive substances not as consecutive elements, but as consecutive isotopes of diverse elements. At this point however consistency almost ceases to be a jewel. I can find no satisfactory compromise, and will hereafter refer to the various radioactive materials simply as "substances"—so bringing to an end this long analysis of words, which is justified only in so far as it may have concentrated the reader's attention upon the facts underlying them.

We return to Fig. 1.

The radioactive substances are grouped into three main lines of descent or sequences, commonly called *series*. Each of these throws off one or two branches, which however cannot be followed far; these I will discuss further on, pausing here only to mention that one of the three main sequences, the *actinium series*, is believed by many to branch in this manner out of the *uranium-radium series*. This however is not certainly established, and it is suitable to regard these two and the *thorium series* as independent sequences, which between them comprise all the known radioactive isotopes among the elements.[13]

Uranium and thorium, the first elements of the series to which they have given their names, are even yet after all the aeons of the earth's existence to be found in abundance among its rocks. This practically proves that uranium, at least, disintegrates with exceeding slowness; for all the other known elements are lighter than it is, and consequently there is none of them out of which the steadily-dwindling supply of uranium might be replenished by transmutation. We shall presently learn methods of estimating the duration of uranium, by which it is shown to be truly colossal.

The atomic weights of uranium and thorium are known, and amount to 238.18 and 232.12 respectively. From these it should be possible

ticular isotopes of the elements 84, 88, 89, 90 and 91 respectively, but are sometimes used as names for these elements—another dangerous source of misunderstanding. The name *niton* was formerly used for the isotope *radon* of element 86, and might well be used for this element now that the isotopes are individually named.

[13] Apart from the elements potassium and rubidium, which will continually demand to be mentioned as exceptions unless they are disposed of once for all at this point. Let it be stated, then, that these elements emit electrons, so feebly however that they are much less active than even uranium, which ranks among the least radioactive of all the known radioactive substances; and that no one has identified the substances into which they are transmuted, though presumably those are isotopes of calcium and strontium respectively. Cf. an account of the radioactivity of these elements by A. Holmes and R. W. Lawson: *Phil. Mag.* (7) **2**, pp. 1218–1233 (1926).

to deduce the atomic weights of all the other members of the two sequences; thus, a radium atom is what is left behind after a uranium atom has ejected three alpha-particles (mass, 4 apiece) and two electrons (mass negligible) and its atomic weight should therefore be 226.18. Here we meet a troublous fact. The value of the atomic weight of radium, as measured by no less an expert than the celebrated Hönigschmid, is 225.97 with an uncertainty believed not to exceed three units in the last place. This might be explained by supposing that the *element* uranium as found in nature is a mixture of several isotopes in relatively large proportions, only one of which is the parent of the uranium-radium series, while the others may be stable or perchance the ancestors of the other series; indeed it is hard to think of any other adequate explanation.[14]

All three of the sequences terminate in isotopes of the element 82, commonly known (but remember the caution on page 110!) as lead. It is a curious fact that the most rare and precious of all substances should die away by self-transmutation into the one which serves as the symbol for everything which is commonplace, dull and cheap. The atomic weights of the terminating isotopes of the radium and thorium sequences may be guessed in the same manner as that of radium from that of uranium. Starting from radium and from thorium respectively and noting that an atom of radium is destined to eject five alpha-particles and an atom of thorium six during the transformations whereby they turn into atoms of RaG and ThD respectively, we calculate the values 206.0 and 208.1 for the atomic weights of these two isotopes of element 82. Now nearly every sample of lead that has ever served for an atomic-weight determination has yielded a value near 207.2. Yet, when the lead-content of certain minerals rich in uranium and its posterity and deficient in thorium was extracted and investigated, the atomic weights of these samples were found to lie extremely near to 206—some of the values recorded are 206.046, 206.048 and 206.08. On the other hand, samples of lead extracted from various minerals rich in thorium and poor in uranium displayed abnormally high atomic weights, values attaining in some instances to 207.9. These are data much more dramatic than the customary outcome of the tedious process of determining an atomic weight; one wonders vainly what chemists would have felt, if they had been published before

[14] What is commonly called "uranium" contains not only the ancestor of the uranium-radium series, but also one of its descendants, which however is not present in sufficient amount to affect the atomic weight. This is the reason for inserting the words "in relatively large proportions" in the above sentence. The fact that the atomic weight of uranium is not integral might be taken to suggest that it is a mixture of integral-weight isotopes. Aston's latest experiments on stable elements of non-integral atomic weight show, however, that the premise does not necessarily lead to the conclusion.

radioactivity was discovered. They disclose the only known instance of distinct stable isotopes of an element being found separately from one another in nature. Whether "ordinary lead" of atomic weight 207.2 is a mixture of these two isotopes, or contains still others, is as yet an unsolved question.[15]

The three series resemble one another not only in the nature of their terminal substances, but in other regards as well. The substance in the radium series known as ionium, the member of the actinium series called radioactinium, the member of the thorium series named radio-thorium, are isotopes all three of the element 90; and these three substances evolve through the same succession of transformations, alpha-ray emissions and beta-ray emissions following after one another in the same order. The *n*th descendants of these three substances, for each value of *n* from 1 to 6, are isotopic with one another—a statement which will probably be made clearer by Fig. 1 than by these words. This parallelism, which from the grandchildren of Io and RdAc and RdTh onward is reflected in the names of the substances, includes also the "branchings" which occur in each sequence at the substance labelled C—radium C and actinium C and thorium C. It is limited in its range, for the earlier parts of the three sequences are by no means alike, while the radium sequence continues onward for three stages longer than the two others. Something within the radium atom impels it to continue evolving even after it has twice taken and left the atomic number which it is destined eventually to take and keep, although the atoms which were once actinium or thorium are contented to stop at the atomic number 82 when for the second time they reach it.

The phenomenon of *branching*, which I have twice casually mentioned, is worthy of a few paragraphs. It signifies that a certain proportion of the atoms of such a substance as (for instance) thorium C transmute themselves in one fashion, the remainder in another. Sixty-five per cent of the atoms of ThC extant at any moment are destined to emit beta-rays and become atoms of a substance ThC' lying one step further up the procession of the elements; the other thirty-five per cent eventually emit alpha-particles and become atoms of ThC" placed two steps further down the procession. Such a "dual transmutation" occurs also at RaC and at AcC—an instance of the parallelism just mentioned, which however does not extend to the relative frequency of the two modes of transformation; 9996 out of ten thousand atoms of RaC, but only three out of a thousand atoms of

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[15] Not however a definitely insoluble question, since the Thomson-Aston method of resolving mixtures of isotopes (*Introduction*, pp. 14–29) and measuring their individual masses should be applicable to lead—that is to say, certain difficulties have thus far prevented it from being applied to the very heavy elements, but these difficulties may not prove insuperable.

AcC, transmute themselves by ejecting electrons. As the disintegration of a sample of any of these substances proceeds, the relative proportions of the atoms disintegrating in the two ways remain unchanging. This makes it seem inadvisable to describe ThC (for instance) as a mixture of two distinct substances; rather it appears that the atoms may be all alike, but the destiny of each particular atom is a matter of "chance," with the chances favoring one type of disintegration over the other by nearly two to one. This is not the only circumstance in radioactivity which suggests the operations of "chance."

The substances labelled C′ and C″, which result from the dual disintegration of any of the three substances labelled C, differ in atomic weight and in atomic number, and in radioactivity as well; for the C′ substances which were born out of beta-ray transformations emit alpha-rays, while the C″ substances which resulted from alpha-ray transmutations send forth beta-rays. Consequently their immediate descendants, the two grandchildren of each C-substance, are isotopes with one another—and isotopes which should be alike not only in atomic number but in atomic weight as well. Is there any respect in which they differ? We cannot tell. Both of the grandchildren of ThC are apparently non-radioactive and stable; probably they are one and the same isotope of lead.   Both grandchildren of AcC likewise seem to be stable.   The predominant grandchild of RaC is the radioactive substance RaD; but in this case the number of atoms of RaC electing the less popular path of disintegration is so exceedingly small that we can neither discern any distinctive radiation to be ascribed to a substance isotopic with RaD but distinct from it, nor yet conclude from our failure that no such substance exists. Concerning the fourth of the known branchings, which occurs at UX_1, the state of affairs is the same as with RaC; we can neither detect more than one kind of grandchild, nor be sure that there is only one.   In this case, by the way, both modes of transmutation of the parent element involve the emission of beta-rays.

Although among the four substances which are known to disintegrate in two alternative ways there is thus none for which both of the two lines of posterity can be traced through more than two generations, it is believed by many that there must be a fifth such substance in the uranium series, from which the actinium series goes off as a branch while the main proportion of the atoms continue evolving down the radium sequence. The reason for this idea is that in the ores of uranium the members of the actinium sequence are as a rule to be found about three per cent as abundantly as the members of the radium sequence. This fact could be deduced by assuming that

uranium II suffers a dual alpha-ray disintegration, about 97 per cent of the atoms transmuting themselves into ionium and the other 3 per cent into the mysterious substance uranium Y which is always found mixed with uranium, and which is known to emit beta-rays and hence to pass over into an isotope of element 91 which may well be protactinium, the first known member of the actinium series. On following out these presumptive transformations in Fig. 1 the reader will see that they would lead to the actually-observed result; but that is not quite the same thing as proving that the observed result is attained in just that way. The branching may occur elsewhere in the posterity of uranium; or the observed constancy of the ratio of actinium to radium in the rocks may mean that actinium and its family all descend from a separate isotope of element 92, not concerned in the production of radium. Much light would be shed upon this question if someone would only determine the atomic weight of even one member of the actinium sequence—an achievement which would settle at once those of all the others, and is most eagerly awaited.

Having dealt with the filiation of the radioactive substances, having specified the substance from which each is born and the substance to which it gives birth, and the sort of particle which is emitted in each process of transmutation, it remains to specify the rates at which the transmutations occur, and the speeds of the particles which are emitted, and the wavelengths of the quanta of radiation which sometimes come out also, and how many there are of these. The fundamental assumptions of the theory of radioactivity, which the experiments have sustained, require that in a transmutation only one alpha-particle or one beta-ray be emitted from the nucleus of one self-transmuting atom; but there is no such limitation upon the radiation-quanta, nor upon the electrons incidentally ejected from the circumnuclear family.

The rate of transmutation of every radioactive substance, so far as we know, is governed by the famous exponential law which signifies that *equal fractions perish in equal times*—that if one were to take a sample of the substance and determine the quantities extant at two instants an hour apart, and also those existing at two other instants an hour apart, and at any number of pairs of instants separated by intervals of one hour, then the mutual ratios of the two measured values of all those pairs would be the same. Half of any sample of thorium C transmutes itself in one hour; half the remainder in the next hour; half the remainder in the next hour, and so forth *ad infinitum* (or, to speak more carefully, up to the limit of the observations).

This law is described by the following formula relating the quantity Q of the substance existing at any time t, and the quantity Q_0 existing

at any other time t_0 (provided that no replenishment of the supply is taking place!):

$$Q = Q_0 \exp\left(\frac{t_0 - t}{\tau}\right). \tag{1}$$

Furthermore the rate dQ/dt at which the substance is being transmuted at any instant is related to the amount Q existing at that instant as follows:

$$dQ/dt = -\frac{Q}{\tau} = -\frac{Q_0}{\tau} \exp\left(\frac{t_0 - t}{\tau}\right). \tag{2}$$

These formulæ contain only a single constant characteristic of the substance. Nothing simpler could be desired. A phenomenon that
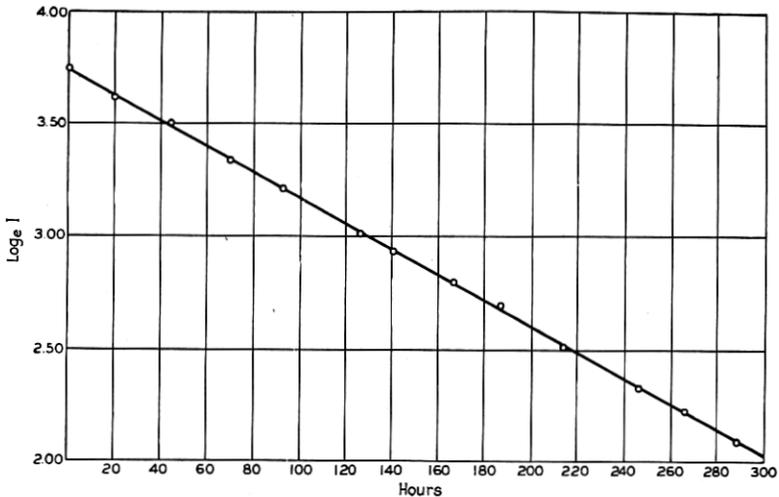

Fig. 2.   Decay-curve of radium E (R. F. Curtiss)

(Being plotted on logarithmic paper, the graph of the exponential law is a straight line.)

can be described by a formula involving only one constant which has to be varied to distinguish one case from another is a rare gift of nature.

While the equations (1) and (2) are naturally valid whatever the unit in which we choose to measure Q, it is desirable as a rule (and necessary, in comparing the radioactivity of different substances) to express Q either in gramme-molecules, or in actual numbers of atoms. In some places I shall use N as a symbol for Q measured in the latter manner.

The exponential law is a law of chance. It may be expressed by saying that the chance of an atom disintegrating within a given time-

interval is precisely the same, whichever atom one chooses to consider and whenever the instant at which one chooses to let the given time-interval begin.   I will quote a passage from Poincaré [16], taking only the liberty of writing 'nucleus' where he wrote 'atom.'   "If we reflect on the form of the exponential law, we see that it is a statistical law; we recognize the imprint of chance.   In this case of radioactivity, the influence of chance is not due to haphazard encounters between atoms or other haphazard external agencies.   The causes of the transmutation, I mean the immediate cause as well as the underlying one (*la cause occasionnelle aussi bien que la cause profonde*) are to be found in the interior of the atom [*read*, in the nucleus]; for otherwise, external circumstances would affect the value of the coefficient in the exponent. . . . The chance which governs these transmutations is therefore internal; that is to say, the nucleus of the radioactive substance is a world, and a world subject to chance.   But, take note! to say 'chance' is the same as to say 'large numbers'—a world built of a small number of parts will obey laws which are more or less complicated, but not statistical.   Hence the nucleus must be a complicated world. . . ." I shall make no further allusion to theories of radioactivity.[17]

The constant τ may be interpreted as the time-interval during which the fraction $\dfrac{e-1}{e} Q$ (or approximately $0.632Q$) of any initially-present quantity Q of the substance would undergo its change.   It is greater by the factor $1/log_e2$ (or approximately 1.44) than the *half-period* of the substance, the interval (designated by T) during which one half of the initially extant atoms are transmuted.   It is also the average duration of the life of a single atom.   All of these statements may be proved without difficulty from the formula (1).   From the similarity between (1) and (2) it follows that the rate at which transmutations occur in an unreplenished sample of a radioactive substance, and the rate at which rays shoot out of such a sample, and the intensity of all the effects which the rays produce, vary exponentially with time; and the constant T which is the half-period for the extant quantity of the substance is likewise the half-period for all of these.   The constant τ likewise has the same meaning for them all, and so does its reciprocal

[16] *Dernières Pensées*, pp. 204–205; he credits Debierne with the idea.

[17] Further and very valuable evidence that the transmutations of individual atoms are governed by the "laws of chance" operating within their own nuclei is furnished by the variations or fluctuations (*Schwankungen*) of the numbers of alpha-particles emitted from a sample of any radioactive substance in consecutive equal time-intervals very short compared with the half-period of the substance (*v.i.*).   These are precisely analogous to the fluctuations in thermionic emission known by the name of "Schroteffekt" (*Introduction*, p. 10).   Consult an article by K. W. F. Kohlrausch in *Ergebnisse der exakten Naturwissenschaften*, 5 (1926).

λ, which is called the *disintegration constant*, and is often specified instead of τ or T.

The values of the half-period T for the various radioactive substances are collated in the accompanying Table, which contains also the names of the substances, their usual symbols (those used in Fig.1), the symbols embodying their atomic numbers proposed by Kovarik and McKeehan, and the types of particle which they emit from their nuclei.

SYMBOLS, NAMES AND HALF–PERIODS OF THE RADIOACTIVE SUBSTANCES

The first column of this Table contains the usual symbols for the substances; the third, their usual names; the fourth, their half-periods as collated by A. F. Kovarik and L. W. McKeehan (*l. c. supra*); the fifth, the nature of the particles which they emit at transmutation. In the second column, the symbols proposed by Kovarik and McKeehan are given; each is composed of the atomic number of the substance, of a symbol denoting the series to which it belongs, and sometimes of a second numeral which, when the substance is an isotope of one or more others in the same series, denotes whether it is the first, second or third of these isotopes reached in the course of the transmutations. In cases of branching, the less common of the two resulting substances is italicized. The annotation *est.* signifies that the half-periods in question are estimated by extrapolating the Geiger-Nuttall relation (*v.i.*). The abbreviations s, m, d, a stand for second, minute, day, year.

URANIUM-RADIUM SERIES:

UI	92UI	Uranium I.............	$4.6 \cdot 10^9 a$	α
UX$_1$	90UI	Uranium X$_1$............	24.5d	β to UX$_2$, β to UZ
UX$_2$	91U	Uranium X$_2$............	1.138m	β
UZ	*91Ua*	*Uranium Z............*	*6.69h*	β
UII	92UII	Uranium II............	$1.2 \cdot 10^8 a$ (*est.*)	α
Io	90Ra	Ionium...............	$7.43 \cdot 10^4 a$	α
Ra	88Ra	Radium..............	$1.69 \cdot 10^3 a$	α
Rn	86Ra	Radon, radium emana-tion...............	3.810d	α
RaA	84RaI	Radium A............	3.0m	α
RaB	82RaI	Radium B............	26.8m	β
RaC	83RaI	Radium C	19.5m	β to RaC', α to RaC''
RaC'	84RaII	Radium C'............	$10^{-6}s$	α
RaC''	*81Ra*	*Radium C''...........*	*1.32m*	β
RaD	82RaII	Radium D............	16a	β
RaE	83RaII	Radium E............	4.85d	β
RaF	84RaIII	Radium F, polonium 136.3d		α
RaG	82RaIII	Radium G, radium lead apparently stable		

THORIUM SERIES:

Th	90ThI	Thorium..............	$1.3 \cdot 10^{10} a$	α
MsTh1	88ThI	Mesothorium 1........	6.7a	β
MsTh2	89Th	Mesothorium 2........	6.20h	β
RdTh	90ThII	Radiothorium.........	1.90a	α
ThX	88ThII	Thorium X...........	3.64d	α

Tn	86Th	Thoron, thorium emana-tion..............54.5s	α
ThA	84ThI	Thorium A............ 0.145s	α
ThB	82ThI	Thorium B.............10.6h	β
ThC	83ThI	Thorium C............60.6m	β to ThC', α to ThC''
ThC'	84ThII	Thorium C'...........$10^{-11}s$ (est.)	α
ThC''	81Th	Thorium C''........... 3.20m	β
ThD	82ThII	Thorium D, thorium lead apparently stable	

ACTINIUM SERIES:

Pa	91Ac	protactinium........... $1.6 \cdot 10^4 a$	α
Ac	89Ac	actinium..............20a	β
RdAc	90Ac	radioactinium.........18.9d	α
AcX	88Ac	actinium X...........11.2d	α
An	86Ac	actinon, actinium emana-tion................ 3.92s	α
AcA	84AcI	actinium A............ 2.00s	α
AcB	82AcI	actinium B............36.1m	β
AcC	83Ac	actinium C............ 2.16m	α to AcC'', β to AcC'
AcC'	84AcII	actinium C'............ .009s	α
AcC''	81Ac	actinium C''........... 4.71m	β
AcD	82AcII	actinium D, actinium lead apparently stable	

UY, K, Rb not assigned to series. They emit beta-rays, and their half-periods are given respectively as 24.6h (St. Meyer, *l. c.* footnote 1), $1.5 \cdot 10^{+12} a$ and $10^{11} a$ (Holmes and Lawson, *l. c.* footnote 13).

To measure a disintegration-constant seems an easy task, since one has only to choose the most convenient effect of the rays of the substance in question, and measure it at sufficiently many times to establish a sufficiently long arc of its decay-curve. Yet there is, I suppose, no other problem of which the general solution involves as many of the typical difficulties of research in this field; partly because some of the half-periods to be measured are so exceedingly short and some so tremendously long, largely because no radioactive substance ever exists by itself. Some can be separated completely from their ancestors, but none can ever be totally isolated from its posterity, especially since its rate of producing its posterity is the very thing which is being measured. Its own gradually-declining rays are mixed with the gradually-augmenting rays of its descendants, and while the specific effects of the former can indeed in some cases be distinguished from those of the latter, this is often difficult and sometimes impracticable. Frequently the observer is required to deduce the half-periods of individual substances from observations upon a continually-changing mixture; and most of the mathematical formulæ used in the study of radioactivity are developed out of equations (1) and (2) for interpreting such observations, or inversely for predicting the evolution

of a mixture of substances of which the initial composition is taken for granted. There is no better way of conveying a notion of the methods by which radioactivity was and is studied than to describe how some of the known half-periods were actually ascertained.

The simplest of all the cases are those in which a substance which can easily be separated from its ancestors transmutes itself into one which either is not radioactive at all, or else decays so slowly that the rays which it emits are not strong enough to interfere with the observations on the rays of its parent. The penultimate substances of the various series are candidates for this class, but the only one among them which is abundant enough and lasts long enough to be easily isolated from its ancestors is radium F, otherwise known as polonium. This therefore is the classical instance of a substance of which the decay-curve is determined directly from observations on rays of its own. Another is radium E, of which the half-period is so short (about 5 days) and the half-period of its daughter-substance so long (more than four months) that its decay-curve can be traced practically as if it changed into a stable element (Fig. 2).

Almost as simple are certain cases in which a radioactive substance is isolated both from its ancestors and from its posterity, and then the growth of its immediate descendant is measured. This method is available when the parent-substance is much longer-lived than its child, so that the rate at which atoms of the latter come into being is practically constant throughout the period of observation. Let B represent this rate; let Q represent the quantity of the daughter-substance extant at any time t, the time being measured from the instant when the isolation of the parent-substance is perfected, so that $Q = 0$ at $t = 0$; let λ stand for the disintegration-constant of the daughter-substance, so that the rate at which its atoms are disappearing through transmutation is equal to λQ. The net rate of growth of the daughter-substance is therefore

$$dQ/dt = -\lambda Q + B \qquad (3)$$

from which we obtain by integration

$$Q = \frac{B}{\lambda}(1 - e^{-\lambda t}), \qquad (4)$$

so that the quantity of the daughter-substance, and the intensity of its rays vary as exponential functions of time with the disintegration-constant standing in the exponent. This function, it is true, rises from zero to a positive final limiting-value instead of falling to zero from a positive initial value, as the decay-curve would; but the value

of λ is determined from it quite easily, and as a matter of fact the decay-curve itself can be obtained merely by plotting as function of time the difference between Q and the limiting-value ($= B/\lambda$) which Q approaches as t increases indefinitely. Determining a half-period from a rate of growth is therefore mathematically the same process as determining it from a rate of decay. This is one of the ways in which the half-period of uranium X_1 is measured; and the standard method for determining that of radium is based partly upon it, as we shall presently see.

Eventually the grandchild and the remoter posterity of the parent-substance must make their presence known. This is not always a disadvantage. Letting λ_1 and Q_1 stand for the disintegration-constant and the extant quantity of the daughter-substance, λ_2 and Q_2 for those of the granddaughter, we have as basis for the theory these equations:

$$dQ_1/dt = B - \lambda_1 Q_1, \qquad dQ_2/dt = \lambda_1 Q_1 - \lambda_2 Q_2, \qquad (5)$$

integrating which, and supposing that at $t = 0$ the parent-substance has just been isolated so that the building-up of the two descendants from zero is just commencing, we obtain for Q_1 the expression (4) with λ_1 in the place of λ, and for Q_2 the function

$$Q_2 = B\left[\frac{1}{\lambda_2} + \frac{1}{\lambda_1 - \lambda_2} e^{-\lambda_1 t} - \frac{\lambda_1}{\lambda_2(\lambda_1 - \lambda_2)} e^{-\lambda_2 t}\right], \qquad (6)$$

which to second approximation is equivalent to

$$Q_2 = \frac{1}{2} B\lambda_1 t^2. \qquad (7)$$

The amount of the grandchild therefore should increase at first as the square of the time elapsed, whereas the amount of the child increases proportionally to the time. There are instances, in the history of the study of radioactivity, of a substance being regarded as the child of another until measurements were made upon its rate of growth in an isolated sample of its putative parent, whereupon through its conformity to (7) it was proved to be the grandchild and not the child. The question whether radium comes directly out of uranium II, or out of an intermediate substance, was settled in this fashion; and by observing a sample of uranium II at intervals over a period of almost twenty years, and measuring the radium which was being developed within it, Soddy was able through equation (7) to calculate the half-period of this intermediate substance (ionium).

The method used in deriving the equations (4) and (7) can always be

applied to any number of consecutive radioactive substances; there are always just equations enough to determine all the constants and describe completely the future history of any mixture of the members of a single family line, provided that their relative proportions in the mixture are specified for some particular moment. Even with only three substances the behavior of the mixture may be extraordinarily complicated; but there are simpler cases which are instructive.

If for instance one sets aside a substance with a much longer half-period than any of its posterity possesses, the extant quantity of each and every one of the descendants will first increase and then begin to decrease, and eventually diminish along the same exponential curve as the long-lived ancestor itself—not because the half-periods of the descendants are actually changed, but because of the partial balancing between the decay and the replenishment of each. Thus the half-period of the long-lived ancestor may be determined by plotting against time the total intensity of all its rays and all the rays of its descendants, or that of any particularly convenient kind of ray emitted by any member of the family. The most carefully measured and accurately known of all disintegration-constants, that of radon, is usually determined in this way; its half-period amounts to four days, those of its three next descendants radium A and radium B and radium C to only a few minutes each, so that after isolating a sample of radon and waiting a few hours one can set up any device for measuring the gamma-rays of radium C, plot their decay-curve, and from it determine a value of λ which is not that of radium C, but that of radon.

If in such a case as the foregoing the long-lived ancestor is so very long-lived that no appreciable decrease in its rate of transmutation can be detected over a period of years, then eventually the quantities and the radiations of all of its descendants assume values which likewise do not change appreciably for years; "radioactive equilibrium" is attained. In a unit of time, equal numbers of atoms are transmuted out of each substance into the substance following, into each substance out of the one preceding. Representing by N_n the number of atoms of the nth member of the series (counting the very long-lived ancestor as the first) extant in the mixture in radioactive equilibrium, by λ_n its disintegration-constant, and remembering that $\lambda_n N_n$ is the rate at which its atoms perish by transmutation, we have the chain of equations:

$$- dN_1/dt = \lambda_1 N_1 = \lambda_2 N_2 = \lambda_3 N_3 = \lambda_4 N_4 = \ldots \tag{8}$$

from which, if we know the relative quantities of any two members in

a mixture in equilibrium, and the half-period of either, we can determine the half-period of the other.[18]

This method could be applied to estimate the half-period of radium, which is so long that in the years since it was first isolated no sample has yet become perceptibly feebler in emitting its rays, while the half-periods of its descendants are all much shorter, and that of its child is only 3.82 days and is rather accurately known. However, the volume of radon gas in equilibrium with one gramme of radium (about the largest quantity of radium which has ever been gathered together in one place) is at normal temperature and pressure only about .0006 cc, and the measurement of so small a quantity of gas is inevitably so inexact that this method cannot compete even with the not-very-accurate alternative methods which we shall presently meet. However, its results do not disagree with theirs.

The most fascinating application of this method is made upon the rocks of the earth, which have presumably been existing so long that there has been ample time for the longest-lived member of the uranium-radium series to attain equilibrium with all of its descendants. As it happens, the longest-lived member of this series is the first, uranium I. Probably this is no mere accident; if uranium is the descendant of less lasting ancestors, they would all be gone by now. However that may be, it is a fair presumption that at least the older rocks of the earth have been formed and buried long enough for the uranium in them to have attained to equilibrium with its descendants. The ratio of the concentration of uranium to the concentration of any member of its posterity, radium for example, should then be equal to the reciprocal of the ratio of their half-periods. Great numbers of samples of rock from all over the world were analyzed by Rutherford and his pupils, and in the laboratories of France and Germany; and for a large proportion among them the ratios of the radium content to the uranium content were found to lie close to one another, and to a mean value which Rutherford assigns as $3.40 \cdot 10^{-7}$. Accepting this as the equilibrium-ratio, and 1690 years as the half-period of radium, we obtain for the half-period of uranium the truly colossal figure of 4.4 billions of years! This value is substantiated, as we shall presently see, by an altogether different method.[19]

[18] In some of the older rocks of the earth, uranium and its descendants have attained mutual equilibrium, and the value of λN for uranium in such a rock is equal to the rate at which the inert end-product (RaG) of the series is accumulating, so that by measuring the amount of RaG already accumulated and the amount of uranium still remaining one can estimate the age of the rock. Consult O. Hahn, *Handbuch der Physik*, **22**, pp. 289–306.

[19] This is a fortunate circumstance, as it gives greater confidence in rejecting the data obtained with samples of rock which yield values of the radium-to-uranium ratio differing considerably from $3.4 \cdot 10^{-7}$. In some cases these deviations may be

One of the two best methods for determining the half-period of radium is a combination of this last-named method with one of those which I described earlier. Let us suppose that a sample of ionium, equal to the amount which would be in equilibrium with one gramme of uranium and $3.40 \cdot 10^{-7}$ grammes of radium, is purified of its original radium-content and set aside for occasional observations of the rate of growth of fresh radium in it. Representing by N_1 the number of ionium atoms in the sample (which diminishes in so small a proportion that we may consider it constant), by N_2 the number of radium atoms extant at time t after the new supply begins to grow, by λ_1 and λ_2 the disintegration-constants of these two substances; translating equation (4) into this notation, and remembering that the rate of transmutation of the parent substance which was there called B is now (measured in atoms transmuted per second) equal to $\lambda_1 N_1$, we have

$$N_2 = \frac{\lambda_1 N_1}{\lambda_2} (1 - e^{-\lambda_2 t}). \tag{9}$$

Represent by N_{20} the number of atoms of radium which would be in radioactive equilibrium with the sample of ionium, that is to say, the number of atoms in $3.40 \cdot 10^{-7}$ grammes of radium; by equation (8) we have

$$\lambda_1 N_1 = \lambda_2 N_{20}, \tag{10}$$

so that equation (9) may be transformed into one containing no constants except the known one N_{20} and the object λ_2 of the investigation. The gain is still greater; developing the exponential function in (9) as a power-series in t and retaining only the first term, we have

$$N_2 = N_{20}(1 - e^{-\lambda_2 t}) = N_{20}\lambda_2 t + \text{terms of higher order.} \tag{11}$$

This means that we need to trace the growth-curve of radium out of ionium only so far as is necessary to determine its initial slope, the initial rate at which the radium increases before its own transmutation begins to tell. This as it happens is all that there has yet been time to trace, so that this combination of the two methods is the only way yet available of interpreting the growth-curves.[20] After a sample of ionium has been kept for a century or two, it may be possible to trace a long enough arc of the curve to determine by the first method. After

ascribed to the comparative youth of the rocks, in others to the selective action of flowing water and other geological agents in removing some and leaving others of the members of the radioactive family.

[20] This method, it will be perceived, is essentially a measurement of one and hence of all of the terms $\lambda_n N_n$ which are equated in equation (9); the rate of growth of radium out of ionium being ascertained, it is possible to calculate the value of λ_n for any substance in the radium series for which N_n, the quantity in equilibrium with the preassigned quantity of ionium, can be measured.

our descendants have solved the other problems of physics, they may be able to entertain themselves by keeping records of the behavior of long-lived radioactive substances, and so determining half-periods with an accuracy improving from millennium to millennium.

Another and the most picturesque of all the ways of determining a disintegration-constant consists in counting the atoms which in a measured quantity of the substance disintegrate in each second. It sounds almost unbelievable that this should be feasible; but it is really practicable to count the alpha-particles which proceed from a radioactive substance, for they make individual visible scintillations upon a fluorescent screen placed across their paths. If this device is inconvenient, one can measure the total charge which the alpha-particles carry into a chamber arranged to receive them, and divide it by the specific charge borne by each, which is very accurately known. The particles and consequently the transmuted atoms having been counted, it is necessary to weigh the substance which is emitting them; and this requirement is less easy to fulfil, being fulfillable in fact only for three substances—radium, and the long-lived ancestors thorium and uranium. Dividing the mass of the weighed sample by the mass of an atom, and dividing the quotient into the number of alpha-particles emitted per second, we obtain the value of λ_1. This of course does not prove that the transmutation is actually proceeding according to the exponential law; that is proved only for certain substances of which the half-periods amount to a few months, days or hours. Nevertheless we assume it, and multiply the reciprocal of λ_1 so measured by $log_e 2$, and call the product the half-period. The values thus obtained are close to 1700 years for radium, agreeing well with the results of the method just above described; 4.7 billions of years for uranium I, agreeing with the result derived from the relative proportions of uranium and radium in the rocks; and 22 billions of years for thorium.

There are yet other ways of estimating half-periods, some of them very ingenious. Extremely short-lived substances require special methods. Thoron, a gas with the half-period of fifty-four seconds, is blown with a measured velocity through a tube along which various electrodes are placed for measuring its activity as it flows past them. Actinium A, of which the half-period is only .002 second, is projected upon the rim of a rapidly revolving wheel, and whirled past various instruments which measure its activity at successive points of its transit through space and time. The projection is due to a very simple but none the less striking natural phenomenon; when an alpha-particle is fired out of an atom of its parent-substance actinon, the residual particle—the atom of actinium A—rebounds or recoils like the

6

gun which fires a shell.   The speed of this "recoil atom" is calculable, standing as it does to the speed of the ejected particle in the inverse ratio of their masses; and it has been utilized for measuring an excessively short half-period, that of RaC', which amounts to only 10^{-6} second; a tube was oriented so that some of the recoil atoms flew along it, and their activity at various points of their flight was measured as in the case of thoron.[21]

Many of the half-periods, finally, are determined by analyzing the curves which represent the variation in time of the rays from continually-changing mixtures of growing and decaying substances: curves which presumably can be represented as sums of three, four or even more terms like the exponential terms in equation (6), not however independently known—that is to say, their coefficients and their exponents must be determined by inspecting the activity-curve itself and trying to build one like it.   This operation sometimes requires a great deal of skill and discernment and intuition.   It seems little short of marvelous that all the radioactive substances of the known series should have been recognized and their half-periods measured.   That they have all been recognized there can be little doubt; for let us consider what it would imply if another substance lay undetected between (let us say) radon and radium A.   There would have to be not one such substance but three, one of them emitting alpha-rays and the two others beta-rays—for otherwise the displacement-law of Fajans and Soddy would be broken.   But if there were an undetected alpha-ray-emitting substance between radon and radium A, the atomic weight of the latter would be eight units below that of the former, instead of only four as we now suppose; and this difference of four units would follow step by step all the way down the radium series, ending in a to-be-expected value of 202 instead of 206; which would vitiate the excellent agreement between the latter figure and the observed atomic weight of the samples of element 82 contained in the uranium ores.   The same argument can be used in the thorium series; in the actinium family the basis for the argument is lacking, but the parallelism between this and the other two families conduces to the same belief.   It is all but certain, therefore, that the explorers of radioactivity have done their work so thoroughly that no substance yet remains unknown in the direct genealogical line from uranium I to radium G, nor in that from thorium to thorium D, nor between radioactinium and actinium D.

[21] This experiment was first performed by J. C. Jacobsen, and later by A. W. Barton, whose paper (*Phil. Mag.* (7) 2, pp. 1275–1282; 1926) should be consulted for details.   It is a very delicate one, especially as the atoms recoil because they have emitted not alpha-particles but electrons, which are comparatively light and are emitted with various speeds.

We turn to the rays themselves.

The alpha-rays are particles of mass $6.60 \cdot 10^{-25}$ gramme and positive charge $2e$ or $9.55 \cdot 10^{-10}$ electrostatic unit. The particles emitted from different radioactive substances differ, so far as we know, only in their initial speeds. The range of variation is astonishingly small; the slowest known alpha-particles issue from their sources (atoms of uranium I) with a speed of $1.423 \cdot 10^9$ cm/sec, the fastest [22] emerge from atoms of thorium C' with a speed of $2.069 \cdot 10^9$ cm/sec. The differences in speed between different alpha-particles emerging from a substance are imperceptibly small.

As a rule the speed of the alpha-rays from a substance is neither measured nor quoted directly; one measures by preference their *range in air*, a thing which can be defined because alpha-particles ionize air (and other substances) more readily the more slowly they are moving, until their speeds drop below 10^8 cm/sec and they suddenly cease to ionize altogether. Consequently, if alpha-rays shoot out from a bit of radioactive substance into environing matter, the concentration of the ions which they produce increases steadily and rapidly from the emitting substance outwards, up to a distance where it attains a sharp maximum and then suddenly falls to zero.[23] This distance is the range in the material in question; it is greater the faster the alpha-rays, varying as the cube of their initial speed. It is a property of the alpha-rays and not of the substance which emits them, and I should not have introduced it here but for a certain relation between ranges and half-periods, and as a pretext for showing some pictures of pleochroic haloes.

These haloes occur in certain ancient minerals, chiefly mica; they are systems of concentric spheres of discoloration, of which the pictures represent cross-sections. No one could imagine what they were when they were first discovered; but the explanation is simple and beautiful. Particles of uranium in some cases, of thorium in others, bubbles of radon in yet others, were caught ages ago and held in the points which were to become the centres of the haloes; the spheres of discoloration are the regions of maximum intensity of ionization, where the alpha-rays emitted from the central source were slowed down to their speed of optimum ionizing-power and were on the verge of

[22] Among the particles issuing from samples of thorium C and producing scintillations on fluorescent screens, very occasional ones (one in ten thousand, or fewer) have a much greater range than the rest, or than the characteristic particles of other substances. A few corpuscles of abnormally long range issue from samples of radium C. It is a controversial question whether these particles come from nuclei disintegrating in a rare and abnormal manner, or from nuclei struck and broken by alpha-particles ejected from other atoms, as sometimes happens. Even the published data are not all in accord, and it is unsafe to make further statements.

[23] *Introduction*, pp. 200–204.

ceasing from ionization altogether.   The radius of the outer boundary of every such sphere is the range, in mica, of the kind of alpha-rays which caused it.   All of the alpha-ray-emitting descendants of the initially-imprisoned substance form their individual spheres; in the cross-sections of the best haloes one can discern nearly all of the rings due to uranium I and its seven alpha-ray-emitting descendants on the direct line to radium G, or those of the seven members of the thorium
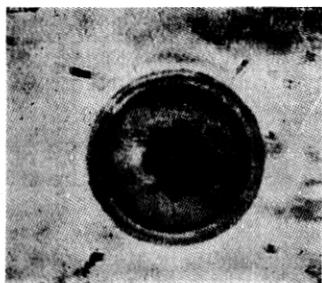
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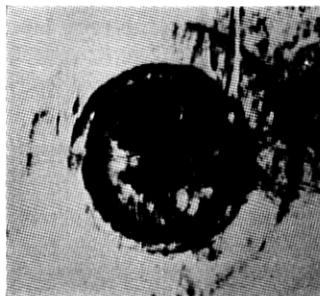
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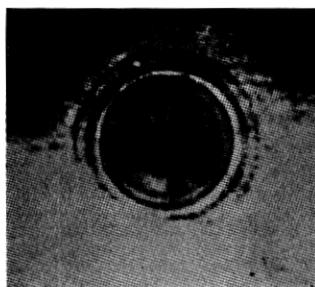
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Fig. 3.   Pleochroic haloes (B. Gudden, *ZS. f. Physik*)

a. Rings of UI and UII (innermost, merged into a single broad ring), Io, and Ra.
b. Rings of RaF (innermost), Rn, RaA and RaC'.
c. Rings of various substances of the uranium-radium series.   Magnifications 665, 500, 480 respectively.

series which disintegrate in this way.   There are no extra rings in these haloes, which strengthens the presumption that no radioactive substances in either series lie undetected.   But there are also haloes of which the rings have not the proper radii to be identified with any known radiating substance.   Are these possibly evidence for the prehistoric existence of others belonging to other series, all of which were too short-lived to survive into the days of scientific research, but disappeared with the dinosaur and the pterodactyl?

There is an interesting and important relation between the initial speeds of alpha-rays and the half-periods of the substances which emit them. One varies as an exceedingly high (negative) power of the other, so that when speed is plotted versus half-period upon logarithmic plotting-paper the resulting curve is a straight line; or, rather, three parallel straight lines, one for each of the three series. This remains true if we plot any power of the speed (for instance, the third) against
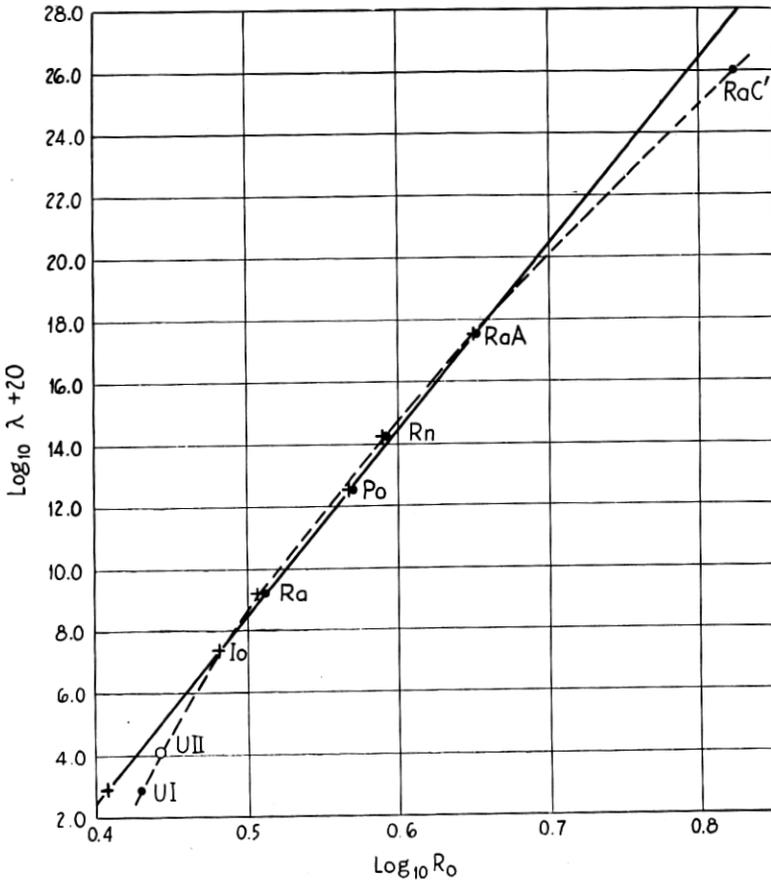

Fig. 4. The Geiger-Nuttall relation

Data for the uranium-radium series; the values for alpha-particle range denoted by dots are taken from pleochroic haloes, those marked by crosses from experimental data. The value of half-period for UII is not known, but is placed by interpolation upon the smooth curve. The straight line represents the best attainable approximation by a linear relation between logarithms of half-period and range; the smooth curve is that drawn by B. Gudden, from whom the data are taken.

any power of the half-period (for instance, the power −1), so that the logarithm of the range of the emitted particles varies linearly with the

logarithm of the disintegration-constant of the emitting substance. This is the way in which this, the "Geiger-Nuttall" relation, is usually expressed:

$$\log \lambda = A + B \log R. \tag{12}$$

The constant B is given (by Hevesy and Paneth) as 53.9 for all three series of radioactive substances, which signifies that the disintegration-constant varies as the fifty-fourth power of the range of the ejected particles! I do not know of any other relation between physical variables in which so high a power occurs; radioactivity, like astronomy, is the home of colossal numbers. The constant A varies from one series to another; it is given as -37.7 for the radium series.

The Geiger-Nuttall relation, like most simple formulae, is a mere approximation. For the radium series its degree of accuracy is illustrated by Fig. 4; the curve drawn through the various points is not quite a straight line. In the actinium series there is a jolt; the point for actinium X lies quite away from the place it should occupy on the straight line drawn to fit closest to the points for the other members, and in fact the half-period of AcX is shorter than that of RdAc, though its alpha-particles are slower. A straight line can be drawn to pass near the points for the remaining members of this series, and another to pass near the points for the descendants of thorium, about as successfully as the line in Fig. 4 fits the points for the radium family. Extending the line drawn for the thorium family to the value of the range for the fastest of all alpha-particles,[24] those of thorium C', one obtains by extrapolation for the half-period of this substance the fantastically small value 10^{-11} second. There is no discernible prospect of verifying this by direct measurement, and in quoting it one should remember the risks of extrapolation.

An alpha-particle is a helium nucleus; when it acquires two electrons, the combination is a helium atom. Helium therefore is a daughter-substance of every radioactive substance which transmutes itself by emitting alpha-rays.

Passing over from alpha-rays to beta-rays, we take at once a great step backward from the clear to the obscure.

The great trouble arises from the fact that beta-rays are electrons, and electrons exist both in the atom-nuclei and in the electron-systems which surround them, or at least they come out of both localities. Whereas the emergence of an alpha-particle from a substance is a clear sign of the transmutation of one atom of that substance, the emergence of a beta-particle need not mean anything of the sort; it may

[24] Reservation being made for the particles mentioned in footnote 22.

simply mean that an alpha-particle, or a gamma-ray quantum, or a different beta-particle coming out of one atom-nucleus operated on its way out the expulsion of that electron from the outer electron-family of that atom or some other. To take one instance only: radium C and radioactinium both emit beta-rays and alpha-rays together, but in the former case there is as we have seen a dual transmutation, in the latter the beta-rays appear to be electrons torn out of the electron-shells surrounding the atom-nuclei as the alpha-particles pass by on their way out. Electrons have the same charge and the same mass, whatever their origin; although it is essential to distinguish how they originate in all these cases of beta-ray-emitting substances, there is no way to make the distinction except by performing experiments on distribution-in-speed of the beta-rays and invoking various theories, not always of the highest order of reliability, to interpret the results. This is the reason why, as Meitner says, the beta-rays actually emitted from self-transmuting nuclei "are the least clarified point in the entire problem of the radioactive transformations."
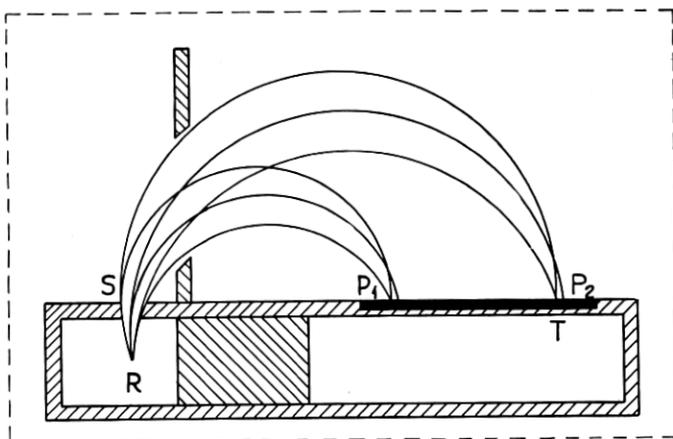

Fig. 5. Apparatus for photographing beta-ray spectra

(After C. D. Ellis and H. W. B. Skinner. Source at R, magnetic field and photographic plate perpendicular to plane of paper, which the plate intersects along P_1P_2.)

In attacking the beta-rays the first thing to do, and indeed the only thing which can be done by experiment, is to determine their distribution-in-speed—the function which gives the relative number of electrons issuing from the substance with speeds comprised between any preassigned limits. The process consists in isolating, by a proper system of narrow perforations and slits, a narrow beam or pencil of electrons, and applying to this pencil a magnetic field which bends the paths

of the electrons (see Fig. 5). The slower the electron, the more its trajectory is curved; if the beam comprises particles of more than a single speed, it is spread into a fan, and a photographic plate placed across the path of the fan records the "magnetic spectrum" of the electron-beam. If the beam comprises several groups of electrons, each with its own sharply marked and definite speed, each group falls upon a distinct part of the plate; if the slit limiting the beam is long and narrow, the groups form long and narrow discolored bands upon the plate, and these bands or "lines" constitute an electronic line-spectrum. The appearance of lines in a magnetic spectrum is taken as practically convincing evidence that the electrons in question issue from the circumnuclear electron-families of the atoms, not from their nuclei.

For this view there is direct evidence of a very convincing character: namely, that beta-ray spectra containing the same lines can be elicited from ordinary stable elements not undergoing transmutation, by the simple process of playing gamma-rays upon them from the radioactive substance in question. Take a sample of the substance, and envelop
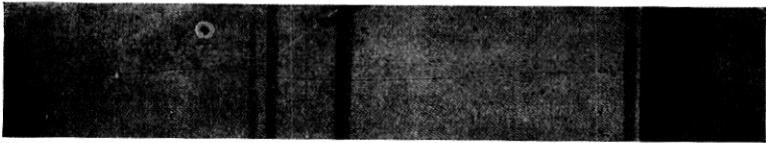

Fig. 6.   Part of the beta ray spectrum of radium B

(After Ellis and Skinner, *Proc. Roy. Soc.* Range 0.037 to 0.054 millions of equivalent volts.)

it in a metal sheath thick enough to stop all of the electrons or alpha-particles issuing from it. Some of the gamma-rays will pass through the sheath, for generally some of them (not necessarily all) are more penetrating than any other radiations which the substance can emit. Let these fall upon another piece of metal nearby; apply a magnetic field to the electrons expelled from this metal, or indeed to the electrons which the gamma-rays expel through the outer surface of the sheath enclosing their source; photograph the resulting spectrum. If the atomic number of the irradiated metal does not depart too far from that of the radioactive source—if for instance the irradiated metal is uranium or lead or platinum or tungsten—the spectrum of the electrons expelled from it will resemble the natural beta-ray spectrum of the source, closely enough so that strong lines of the one spectrum can obviously be identified with corresponding strong lines of the other. Corresponding lines in the two spectra may or may not coincide with

one another; that depends on the kind of metal irradiated; a prominent line in the spectrum of (for instance) radium B will be composed of electrons having energy somewhat greater than that of the electrons forming the corresponding line in the spectrum elicited from uranium, somewhat less than that for the corresponding line from platinum. But if the irradiated metal be isotopic with the substance into which the radioactive source is being transmuted, corresponding lines will be found to coincide with one another. One obtains a beta-ray spectrum having many lines in common with that of radium B, by allowing the gamma-rays to play upon and expel electrons from a piece of a metal isotopic with radium C—that is to say, bismuth.[25]

Whether one uses an atom-model or not, these facts suggest that some at least of the electrons emerging from a radioactive substance are hurled out by some sort of a secondary process operated upon the already-transmuted atoms by the accompanying gamma-rays, working in the same manner as they work upon atoms exposed to them outside. This suggestion becomes much more precise when the atom-model is invoked; for the contemporary model is designed to give a vivid explanation of the lines in the electronic spectra elicited by X-rays and gamma-rays playing upon the atoms of the stable metals.

Every such line is composed of electrons extracted from a particular group, in the circumnuclear electron-family of the atom, by radiation of a particular frequency. Think of the most tightly-bound electrons of all, the so-called K-electrons, to be imagined as lying or revolving closer than any of the others to the nucleus. Merely to extract one electron of this set, a definite amount of energy W_K must be imparted to the atom. Conceive a beam of radiation of frequency ν pouring over a multitude of similar atoms; to each it communicates either no energy at all, or else a definite amount of energy equal to $h\nu = 6.57\cdot10^{-27}\nu$. If this "quantum" unit of energy exceeds W_K, and if the radiation extracts a K-electron from an atom, the liberated electron will fly away with a kinetic energy equal to the excess of the imparted energy $h\nu$ over the extraction-energy or "binding-energy" W_K.

(13) $$\text{Kinetic Energy} = T = h\nu - W_K.$$

This equation determines the initial speed of the departing K-electrons.[26]

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[25] *Introduction*, pp. 184–192; to this I refer also for reproductions of some very beautiful photographs of beta-ray spectra taken by J. Danysz and M. de Broglie.

[26] If the speed v of the electrons is inferior to $3\cdot10^9$ cm/sec, it is permissible to set for T the familiar expression $\frac{1}{2}mv^2$, putting for m the "rest-mass" $m_0 = 9\cdot10^{-28}$g of the electron. Otherwise it is necessary to take account of the dependence of the mass of the electron upon its speed, preferably by using the formula derived from the

If there is but one frequency in the inflowing radiation, the spectrum of the emitted electrons will contain one line composed of what were formerly K-electrons. It will contain others, composed of electrons which originally belonged to other and less firmly-bound sets within the atoms. We distinguish, in order of decreasing binding-energy, K and L_I and L_{II} and L_{III} and M_I and M_{II} and M_{III} and M_{IV} and M_V and still further classes of electrons. The electron-spectrum due to radiation of a single frequency attacking atoms of a single kind comprises a line for each of these classes (apart from those, if any, for which the binding-energy exceeds the quantum-energy $h\nu$ so that the radiation cannot detach them) and the speed of the electrons composing each line is determined by an equation like (13), with the appropriate extraction-energy W_{LI} or W_{LII} or whichever it may be inserted in place of W_K. If there is more than one frequency in the incident radiation, each produces its own system of lines. These statements are proved, and the binding-energies are determined for all the classes of electrons and most of the kinds of metallic atoms, by irradiating metals with X-rays of which the frequencies are known, for they can be separately measured.[27] To ascertain the binding-energy of, let us say, the L_{II} electrons of platinum, one has only to look into the standard tables.

Now we have seen already that the physical and chemical properties of each radioactive substance, so far as they are known, are almost exactly like those of its stable isotope (if it has one); and with this rule the resemblance between the beta-ray spectrum of a radioactive substance and the electronic spectrum which its gamma-rays elicit from its stable isotope most admirably conforms. When a line in the former spectrum obviously corresponds to a line in the latter, both presumably are composed of electrons extracted from the same level by the same radiation. The same gamma-rays are working upon atoms isotopic with one another, and therefore endowed with electron-

Theory of Relativity, to wit:

$$T = m_0c^2 \left[\frac{1}{\sqrt{1 - v^2/c^2}} - 1 \right] = h\nu - W_K$$

X-rays generated by artificial means never have frequencies so high that the electrons which they expel move rapidly enough for the simple substitution $T = \frac{1}{2}mv^2$ to be inadequate; but the frequencies of some of the gamma-rays are so great that the electrons which they extract even from the K-layers of massive atoms depart with speeds much exceeding $3 \cdot 10^9$ cm/sec. J. Thibaud has made direct measurements of a certain gamma-ray frequency and of the speed of the electrons which it ejects from a certain group of known extraction-energy, which are compatible with one another and with equation (13) if the relativity-formula for T is used, but decidedly incompatible if T be set equal to $\frac{1}{2}m_0v^2$ or to the once well-known expression derived by Abraham (J. Thibaud, *L'effet photoélectrique composé;* Paris (Masson) 1926).

[27] *Introduction*, pp. 192–195, 273–282.

families classified into identical classes with identical binding-energies. There is an evident difference; the atoms in the latter case are ionized by radiation poured upon them from without, in the latter by processes which occur within their own nuclei. (Whether in the latter case a wave-train does actually leave a nucleus, and enjoy a real existence during the brief time before it reaches the circumnuclear electron which it is destined to eject, is a question to which it is not easy to give a
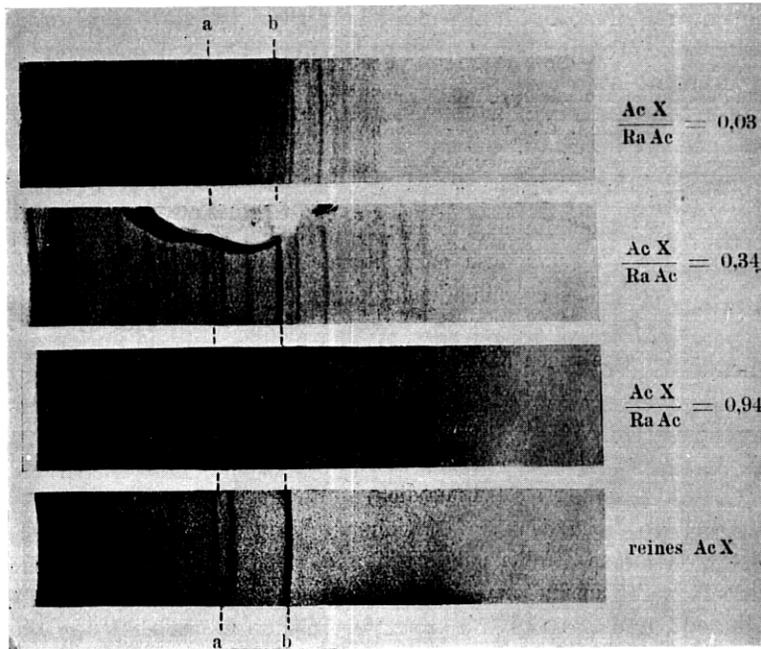

Fig. 7.   Beta ray spectra of radioactinium and actinium X

(After O. Hahn and L. Meitner, *ZS. f. Physik.*   The three upper pictures repre-sent portions of the beta-ray spectrum photographed respectively a few hours, 6 days, and 20 days after the preparation of a pure sample of RdAc, in which the daughter-substance AcX was steadily growing; the lowest, the corresponding portion of the spectrum of a sample of AcX with its descendants.   The lines which diminish in intensity from top to bottom are those of RdAc, those which increase belong to AcX and its descendants (note especially the lines marked *a* and *b*).)

sensible answer!)   But the difference does not affect the energies of the ejected electrons; only their number, for, as seems natural enough, the beta-rays expelled from atoms of which the nuclei are emitting gamma-rays are much more abundant than those which an equal amount of gamma-radiation extracts from atoms on which it falls from without.   Corresponding electron-groups have the same energy.

This is why we know in some cases, and suppose in the others, that when the electrons issuing from a radioactive substance constitute the lines of a line-spectrum they are not themselves coming from the nuclei; they are merely the signs of gamma-rays coming from the nuclei.

This discovery disposes of one potential objection to the displacement-law of Fajans and Soddy. Radioactinium (for instance) is a substance which emits alpha-rays and passes over into a substance two steps farther down the procession of the elements, as the displacement-law requires; but it also emits beta-rays, and since no alternative product one step farther up the procession has been discovered, the displacement-law would be gravely threatened if it were necessary to suppose that these come from the nuclei. There is no such necessity; and since the beta-rays display a line-spectrum, it is intrinsically all the more likely that they come from the circumnuclear electron-shells.

En revanche the character of the thus-far-analyzed beta-ray line-spectra makes it all the more difficult to understand what becomes of the electrons which must truly be emitted from the nuclei, in the transmutations in which the daughter-substance is displaced one step up the procession from its parent. When a substance is undergoing a transmutation of the other kind, the alpha-particles which its atoms emit all have very nearly the same speed. One would certainly expect that the electrons emitted from the nuclei of all atoms of radium B at their instants of transmutation emerge with the same speed. If so, they should compose a sharp line in the beta-ray spectrum of radium B. Now there are certainly some lines in this particularly rich spectrum which have not yet been definitely and exactly explained by the theory which I described before; but it appears that none of them is very prominent, and most of the experts refuse to admit that any one of them is composed of electrons coming forth direct and unretarded from the nucleus. There are other substances which display beta-ray spectra comprising but a few lines, one of which some physicists believe to contain the nuclear electrons.

If the nuclear electrons are not to be assigned to the lines, there remains but one alternative; they must be identified with the electrons composing the continuous beta-ray spectrum which underlies the lines and intervenes between them. The best way to study this spectrum is to dispense with the photographic plate, and set a Faraday-chamber to receive the electrons, with its aperture somewhere in the plane which the plate formerly occupied; if then the magnetic field is continuously varied, the spectrum slides across the aperture, and at each particular value of the fieldstrength the electrons of a particular limited speed-range pass into the chamber and are counted (more precisely, the total charge which they bear is measured, which comes to the same thing).

Curves obtained in this way are copied in Fig. 8. The peaks are the traces of lines (not so many as a photograph would show, for the method in this respect is not so delicate) rising up not from the zero-level but from a smooth sweeping curve, carried (hypothetically) in
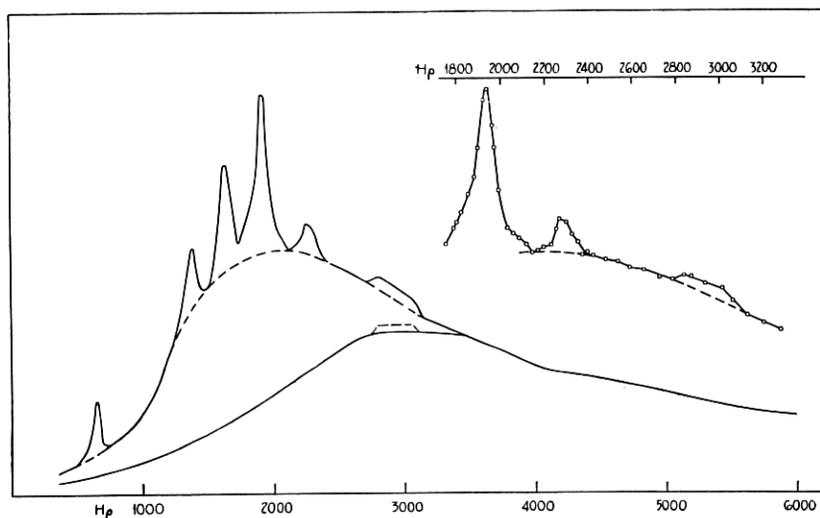

Fig. 8. Beta-ray spectra measured with a Faraday chamber
(Lower curve for RaC, upper curves for RaB + RaC. After R. W. Gurney.)

dashes across the base of the peaks. This is the distribution-curve of the electrons forming the continuous spectrum; integrating it, one obtains the total number of these electrons.[28] This number has been measured for radium B and radium C by Gurney; it amounts to somewhat more than one electron per self-transmuting atom.[29] A much smaller number, had such a one been found, would have rendered untenable the notion that all the nuclear electrons go into the continuous spectrum; the result proves that there is no such obstacle, not at least in these cases. The beta-ray spectrum of radium E consists of a single diffuse band; there are no lines. Emeléus counted the emitted electrons and found a value equivalent to 1.1 electrons per self-transmuting atom.[30] Perhaps then it is a quality of the nuclear

[28] This statement is not exact. The curves may be transformed into true distribution-curves, resembling them but not identically like them, by processes involving allowances for the geometry of the apparatus. The area under these true curves must then be found by integration, and gives the total charge borne by the electrons, the quotient of which by e is the desired number of electrons.

[29] R. W. Gurney, *Proc. Roy. Soc.*, **A 109**, pp. 540–561 (1925).

[30] K. G. Emeléus, *Proc. Camb. Phil. Soc.*, **22**, pp. 400–404 (1924). It is frequently pointed out that RaE emits no perceptible gamma-rays, a fact which makes it seem additionally probable that all the electrons which it emits come from the nuclei. This does not prove anything, as it is conceivable that gamma-rays are emitted which extract electrons from the electron-layers with such efficiency that no appreciable fraction of them escapes unconverted.

electrons that they issue from their atoms with widely and irregularly scattered speeds. If this is true, the presumption is that they escape from the nuclei with equal speeds, the differences resulting from experiences of theirs during the transit through the circumnuclear electron-shells. But it must not be forgotten that a continuous electronic spectrum appears together with the lines, when the gamma-rays from a radioactive substance fall upon one of its stable isotopes; and some allowance must certainly be made for this.

Refreshingly in contrast with the status of this perplexing question is the condition of another, for years the subject of a fervid controversy. Are the gamma-rays from a self-transmuting atom emitted before or after the transmutation occurs? There is only one way of settling this question, and perhaps the question itself ought to be so phrased as to bring this way into prominence. Granting the theory of beta-ray line-spectra expounded in these pages, and granting that certain lines in a certain spectrum have been recognized as being composed of electrons expelled by gamma-rays of one and the same frequency from various K, L, M classes in the circumnuclear electron-family, do the energy-values of these lines show that the electrons come from atoms as yet untransmuted, or from atoms which have already undergone their transmutation—from the atoms of the parent, or those of the daughter-substance? There is no forceful *a priori* reason for expecting either of these alternatives rather than the other; the question must be put to experiment.

If one knew with all desirable accuracy the frequency of the gamma-ray responsible for a particular set of lines, and the class of electrons contributing each line—if one knew for instance that a certain line is composed of K-electrons extracted by gamma-rays of a known frequency ν, one would measure the speed of these electrons, calculate their kinetic energy, subtract it from $h\nu$, identify the difference with the binding-energy W_K according to equation (11), and consult the standard tables to locate the element possessing that value of the extraction-energy for its K-electrons. But there are few gamma-rays of which the frequencies are independently known, and for these the values are not very accurate; so that this method is not generally available.

If however two lines are composed, the one of K-electrons and the other of L_I electrons ejected by gamma-rays of the same though unknown frequency, then the difference between the values of kinetic energy for the electrons of the two lines is equal to the difference between W_K and W_{L_I}; and as this difference varies from element to element, one can consult the tables to locate the element for which the

difference between the K and the L_{I} extraction-energies agrees with the measured value. This is the usual method.

There is still another way, which may be explained by describing an experiment performed by C. D. Ellis and W. D. Wooster.[31] They enclosed a sample of radium B mixed with radium C in a rather thick-walled platinum tube, and deposited a thin layer of the same mixture upon the outer surface of the tube. The thin layer contributed the beta-ray spectrum of radium B and radium C. The beta-rays from the substances inside the tube were stopped by its walls, but the gamma-rays went through and expelled electrons from the platinum, which mingled with those from the covering film; so that upon the photographic plate there appeared side by side the spectrum-lines composed of electrons extracted from atoms of radium B and radium C by their own gamma-rays, and the spectrum-lines composed of electrons extracted from atoms of platinum by gamma-rays of the identical frequencies. Side by side there appeared, for instance, the lines due to K-electrons extracted by the same radiation from radium B and from platinum. The electrons from the radioactive substance had less energy than those from the platinum, for more had been spent in extracting them; the difference between the values of kinetic energy of the electrons was equal to the difference between the values of the K binding-energy for the atoms, with sign reversed; the K binding-energy for platinum is known, that of the other atom is calculated at once.[32]

The six or eight investigations, performed by these methods upon diverse substances by various physicists during the past two years, have all come to concordant results. The atoms from which the electrons of the beta-ray line-spectra are detached are the atoms of the daughter-substances; the gamma-rays are emitted, or at least they act (and it would be a daring person who would say that they exist for a while before they act!) after the transmutation occurs. The controversy

[31] *Proc. Camb. Phil. Soc.*, **23**, pp. 844–848 (1925). There are several important articles in this (November, 1925) number of the *Proceedings* which deal with the problem of the emission of gamma-rays, secondary X-rays, and electrons emitted from the nucleus or ejected from the circumnuclear family by these rays.

[32] All the methods require the observer to guess which lines are composed of electrons from the K-class, which of electrons from the L_{I} class, and so forth; and this is the major difficulty of the problem, for there is nothing intrinsically distinctive about the lines. In many cases, especially when there are several gamma-ray frequencies and a multitude of beta-ray lines, it is necessary to proceed by trial and error, assigning a line first to one class of electrons and then to another, and finally adopting the systematization which leaves the smallest number of lines unexplained or at odds. Sometimes only one out of the three L classes yields a perceptible number of electrons; there is a rule, which if general is very valuable, that when the product of h into the frequency of the gamma-ray exceeds the extraction-energies of all the L classes very greatly, then the L_{I} class is the only one out of which electrons enough are extracted to make a noticeable line.

is settled, and the triumphant side is that of which Meitner was the protagonist. Evidently we must conceive that the electron departing from a nucleus leaves it in a very unstable state, from which it speedily passes over into a comparatively though not absolutely stable state by one or a series of transitions, of which the gamma-rays are the manifestations.

We have still the gamma-rays to consider. Throughout this article I have taken it for granted that the gamma-rays are electromagnetic waves of definite frequencies. The evidence that they are electromagnetic waves has been known so long that it need not be rehearsed. The classical way of determining the frequency of such a wave is to measure its wave-length. With ordinary light-waves this is effected by dispersing them with a prism or diffracting them with a ruled grating. It used to be thought that these methods do not avail with X-rays, because of the shortness of their waves; natural crystal gratings, in which close-ordered files of atoms play the rôle of the rulings in artificial gratings, are used to diffract these. Applying the crystals to gamma-rays, one meets the same difficulty as the discoverers of X-ray met when they applied prisms and ruled gratings; the waves are mostly too short to be diffracted appreciably by natural crystals. The gamma-rays are spread out into a spectrum, and sometimes lines are discernible in the spectrum (Fig.9); but the line of shortest wave-length thus far measured (so far as I know) is at 0.052 Angstrom units or 52 X-units, and there are certainly many others at much shorter wave-lengths which the crystal spectroscope does not diffract far enough outward to be located. Recently people have renewed the attempt to measure wave-lengths of X-rays by the methods appropriate to visible light, and have attained values of astonishing accuracy; perhaps it is not too much to hope that a comparable advance in technique will bring the shortest gamma-rays into the scope of crystal gratings.
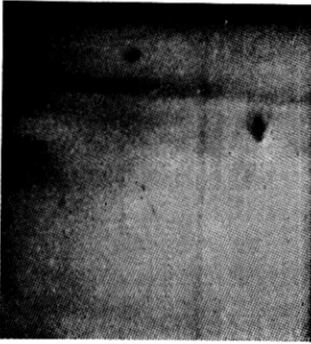

Fig. 9. Gamma-ray spectrum of radiothorium and thorium B

(Wavelengths of the lines 52X, 145X, 168X from right to left. After J. Thibaud, *l. c.* footnote 26.)

The usual method for estimating the frequencies of gamma-rays consists in guessing the class to which the electrons forming a beta-ray line originally belonged; taking its binding-energy from the tables; measuring the kinetic energy of the electrons forming the line; adding

it to the binding-energy, and dividing the quotient by h. This is in a sense the reverse of the usual process of ascertaining which is the element from which the beta-ray line-spectrum proceeds; and as a matter of fact the two have frequently been carried out as parts of one single investigation. The line-spectrum is photographed and its lines are measured, and then the student works over the data until he succeeds in setting up a hypothetical gamma-ray spectrum in which not too small a fraction of the beta-ray lines are explained by the action of not too great a number of gamma-ray frequencies upon the K and L and M classes of electrons, and reversely there is no too obtrusive case of a beta-ray line being predicted from his hypothetical gamma-spectrum and failing to appear.

For illustration I will quote some actual results. Meitner and Hahn located forty-nine lines in the beta-ray spectrum of radioactinium; among these, thirty-seven could be attributed to the action of one or another of twelve gamma-ray frequencies upon one or another of nine classes of electrons. In the spectrum of actinium X they observed twenty-nine lines and explained fourteen of them by postulating seven frequencies. Happily there are much more perspicuous cases. The spectrum of radium D consists of only a few lines—five, according to L. F. Curtiss,[33] whose measurements show that four may be supposed to consist of electrons ejected from the L_I, L_{II}, M_I, and N_I shells by a single gamma-radiation of wave-length 0.26A, while the energy of the electrons forming the fifth line is not perceptibly different from the quantum-energy $h\nu$ of the rays themselves. These last electrons may have been extracted from the outer layers of the atoms, where the binding-energy is so small that it makes but an insignificant deduction from the energy transferred to the electron. Another instance is that of radium itself, of which the three lines composing the beta-ray spectrum may be ascribed to a single gamma-ray of wave-length 0.066A expelling electrons from the K group, the L_I and the M_I group. Such cases as these are so simple that the theory in general and the wave-lengths calculated for the gamma-rays in particular are almost beyond all question.

Certain of the gamma-ray frequencies thus determined, and some which are directly measured with the crystal spectroscope, are found to agree with characteristic X-ray frequencies of the atoms whence they come. This is true of the solitary gamma-ray which is necessary and sufficient to explain the beta-ray spectrum of UX_1, and of two of the rays postulated by Meitner to account for the spectrum of RdAc

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[33] L. F. Curtiss: *Phys. Rev.* (2), **27**, pp. 257–265 (1926). A previous investigation by L. Meitner (*ZS. f. Physik*, **11**, pp. 35–54; 1922) had led to substantially the same conclusion regarding the gamma-ray spectrum.

and two of those for AcX. This is precisely what was to be expected; for in the contemporary atom-model, the characteristic X-rays of an atom are conceived to arise from its circumnuclear electron-family, and to arise after and because an electron has been evicted from the family —a cause which the primary gamma-rays, or the alpha-rays or the electrons coming out of the nuclei, can themselves supply. The electrons expelled by these "secondary" gamma-rays or X-rays (the latter term is now preferred, in all cases where the identification can be surely made) are ejected as the *fourth* stage of a complicated process: first, the primary quantum or particle departs from the nucleus, then a tightly-bound electron is ejected from the electron-family, then a rearrangement of the remaining electrons brings about the emission of an X-ray, which in turn expels the loosely-bound electron. (It seems unlikely, as I intimated before, that the four stages are really separate; probably the passage from the initial state to the final takes place in a single operation, in a flash; but one does not see how to conceive that single operation without resolving it into four.) Since even the primary gamma-rays are emitted after the transmutation, the secondary X-rays *a fortiori* must come from the atoms of the daughter-substance; and this they do.[34]

The gamma-ray spectra thus far mapped out consist of from one to fourteen frequencies, not counting the secondary X-rays; the palm, in this respect, is awarded to radium C. The highest frequency thus far recorded is $5.4 \cdot 10^{20}$, corresponding to a wave-length of 5.57X (5.57 $\times 10^{-11}$ cm) and a quantum-energy amounting to $3.54 \cdot 10^{-6}$ erg or 2.22 millions of equivalent volts; it has twenty times the frequency of the highest X-ray known, and twenty times as great an energy in each quantum as is required to tear the most tightly-bound electron from the family of the most massive atom. It emerges from the nuclei of atoms which have just transmuted themselves out of radium C into radium C′. The fastest electrons forming a definitely-known line in a beta-ray line-spectrum occur in that of thorium C″; their speed amounts to 0.986 of that of light, their energy to almost $2.5 \cdot 10^6$ equivalent volts; but there are still faster ones in the continuous spectrum of radium C, which extends at least as far as to 0.998 of the speed of light. The energy of the alpha-particles of the various substances which emit them

[34] The strongest single piece of evidence is the measurement upon two radiations of radium B, performed with the crystal spectroscope by Rutherford and Wooster (*Proc. Camb. Phil. Soc.*, **23**, pp. 834–837; 1925) who found that the difference between the angles at which they were diffracted from the crystals agreed closely with that to be expected for two prominent X-ray lines of the *L* series of the daughter element (atomic number 83) and disagreed unmistakably with that to be expected for the parent element. This invalidated a contrary result obtained in 1914, which long had stood as an obstacle in the way of the conclusion that gamma-rays are emitted after the transmutation.

ranges from somewhat over four to somewhat under nine millions of equivalent volts. The greatest amount of energy which men have yet succeeded in loading upon a single charged particle or crowding into a single quantum of radiation lies well below the first million of equivalent volts; it still lay well below the first hundred thousand, ten years after radium was discovered. The step from the tens of thousands to the millions is a great one; this supplement voluntarily offered by Nature, transcending immensely the greatest amounts of energy which men can concentrate into a compact parcel, is chiefly responsible for the advances in the understanding of energy and matter which radio-activity made possible.

The advances have indeed been great. Consider what ensued from the discovery of the alpha-rays alone. With alpha-particles Ruther-ford explored the interiors of atoms, and the results of his explorations led him to the nuclear atom-model. The particles themselves he proved to be atom-nuclei of a certain element, and they established the amounts of electric charge which must be assigned to the atoms of that element and all the others. The nuclear atom-model in turn supplied Niels Bohr with the substructure of his theory; and Bohr's theory, together with the phenomena which it inspired men to seek and find, forms the half of contemporary physics. In the edifice of modern physical theory, the alpha-particle is the cornerstone. Had Nature not dispersed the radioactive substances through the rocks of the earth, had there not been one or two of them long-lived enough to survive and maintain a supply of their descendants until man arrived and became scientific—or if the faint outward signs of the radioactivity latent in the rocks had been overlooked, or having once been noticed had been left unstudied—in any of these cases, centuries more might have passed before a proper foundation was located for the edifice. That is the prime reason for honoring those who detected radioactivity, and then did not rest until they had brought it fully into the light. Theirs is an illustrious history, and one not without pathos; for some of those who had worked with the greatest zeal found themselves in later years the prey of a terrible and inexorable disease; like Prometheus in the myth, they were consumed for having brought benefits to the human race. Even yet the benefits which they gave have not been fully ex-ploited; marvelous things may still be discovered, in the process of understanding the actions of the rays on living matter. But that will be another story, and a long one.