RADIO-ACTIVITY:

AN ELEMENTARY TREATISE,

From the Standpoint of the Disintegration Theory.

Brown

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PREFACE.

IN this book I have attempted to give a connected account of the remarkable series of investigations which have followed M. Becquerel's discovery, in 1896, of a new property of the element uranium. The discovery of this new property of self-radiance, or "radio-activity," has proved to be the beginning of a new science, in the development of which physics and chemistry have worked together in harmony. The pioneer in the chemical development of the subject was Mme. Curie, who, by the discovery of radium, extended our knowledge of the new property out of the region of the infinitely small effects in which it had its beginning, and demonstrated it on a scale that could neither be explained nor explained away. On the physical side, the brilliant and elaborate researches of Prof. Rutherford, at first mainly with thorium—an element which, like uranium, is so feebly active that it had been studied for a century before its radio-activity was discovered—paved the way for a complete and general theory of the cause and nature of the new property. According to this theory the elements exhibiting radio-activity are in the process of evolution into lighter and more stable forms, and the radiations spontaneously emitted are due to the incessant flight, radially from the substance, of a swarm of light fragments of the original atoms, expelled in the course of their explosive disintegration. This theory has recently received a direct experimental confirmation by the discovery of the continuous production of the element helium from radium. In these advances physics and chemistry have borne equal shares, and in the close communion between the two sciences throughout the investigations the secret of the rapidity and definiteness of the progress is to be found. Radio-activity has passed from the position of a descriptive to that of an independent philosophical science, based upon principles, only the germ of which is to be found in physics and chemistry as they were understood *before* its coming.

It has been recognised that there is a vast and hitherto almost unsuspected store of energy bound up in, and in some way associated with, the unit of elementary matter, represented by the atom of Dalton. It is possible to arrive at this result by two independent processes of reasoning, the purely physical and the purely chemical. The paths are different but the conclusions are identical. Since the relations between energy and matter constitute the ultimate groundwork of every philosophical science, the influence of these generalisations on allied branches of knowledge is a matter of extreme interest at the present time.

It would seem that they must effect, sooner or later, little short of a revolution in astronomy and cosmology. They will certainly be eagerly received, for it is only fair to add that they have been long awaited, by the biologist and geologist. By the extension of the conception of evolution to the inanimate world, which the study of radio-activity has justified, not only has that conception achieved universality, but the difficulties which hitherto have retarded its logical development in the biological sciences have been cleared away.

Most sciences offer some evidence of the extent of time over which the laws of Nature may be regarded as having been in continuous operation without external interference. It has been a reproach in the past that the conclusions arrived at were mutually inconsistent, and its final removal marks a not unimportant step in the history of science.

The object of the book has been to give to students and those interested in the subject generally a connected account of the main arguments and chief experimental data by which the results have been achieved. This task would hardly have been possible to me but for the training I have received in the subject from Prof. Rutherford. I have had the great advantage of having witnessed the gradual development in the laboratory of many of his most intricate and difficult researches, and I have endeavoured to pass on to the reader something of the clearness of aim directing these researches as it was interpreted and demonstrated to me. No attempt at completeness has been made, for the great wealth of detail in which the subject abounds, while it is the opportunity of the investigator, is probably also the chief cause of difficulty experienced by the student. It is hoped that while this book may serve as an introduction, more complete treatises, or the original communications, will be studied in conjunction with it.

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CHAPTER I.

RADIATION PHENOMENA.

Advances of the Last Decade.—The Undulatory and Corpuscular Types of Radiation.—The Work of Crookes and Lenard on Cathode Rays.—X-Rays.—The Focus Tube.—The "Density Law" of Absorption.—Deviation of Cathode Ray by Magnetic and Electric Forces.—Becquerel's Discovery of the Radio-activity of Uranium.—Methods of detecting the New Types of Radiation.—General Account of the α, β and γ-Rays.—Resemblance between the Effects of the Undulatory and Corpuscular Forms of Radiation.

The last decade, 1894–1904, will probably always be considered a remarkable one in the history of scientific progress on account of the advances made in connection with the phenomena of radiation. Not only has there been a great extension of knowledge with regard to those types of radiation, allied to light, which enter into every-day experience, and which have been the object of enquiry for centuries, but, in addition, entirely new kinds of rays have been discovered, and to account for them new conceptions have arisen, fresh fields of research have been opened up, and problems, before deemed insoluble, have been brought within the range of direct experimental attack. The previously existing foundations upon which the vast fabric of modern science has been successfully raised are being exchanged, without injury or alteration to the structure, for others one step deeper and more fundamental. The work of transition has been proceeding quietly and simultaneously from many sides, and in this respect the last decade is inseparably connected with those proceeding; but until quite lately few, except those actually engaged in the work, realised the magnitude of the results being obtained or the real consequences of the conclusions being arrived at. Most recently, however, the advent of radium, and the prominence given to the almost daily discoveries that followed, have drawn universal attention to the newly explored regions. The chemist's atom is no longer the unit of the sub-division of matter, and the internal structure of the atom is now the object of experimental study. The particular development which forms the subject of the present book arose out of a discovery in 1896 by M. Henri Becquerel, that certain kinds of matter have the property of emitting a new and peculiar type of radiation continuously and spontaneously, and this class of bodies has been termed "radio-active." This discovery was, however, directly connected with previous discoveries of Crookes, Lenard and Röntgen of other new kinds of radiation, and it will be convenient to devote this opening chapter to a brief review of the present position that has been reached in these allied subjects, treating each advance in the order of its discovery, and tracing its historical connection with those preceding. This review can at best be but superficial, but some idea of the modern conceptions of the nature of radiations generally is essential to the correct appreciation of the discoveries in the most recent field of radio-activity.

The term *radiation* is properly applied to indicate an influence transmitted *radially* from its source to its surroundings, and capable of traversing vacuous space, without occupying in its transmission a period of time great enough to be sensible under ordinary circumstances. It was used in this sense to express the transmission of light from distant objects before any definite ideas as to the mechanism of propagation had been advanced. Some of the most fundamental ideas in science owe their inception to the necessity of accounting for radiation phenomena. Newton was the first to recognise the difficulty underlying the problem, for to him, as to present-day philosophers, action at a distance through space of one body on another is inconceivable unless some connection exists between the two bodies. He put forward the only explanation possible in his day to account for the radiation of light, and his corpuscular theory, as it is called, although long since disproved so far as light is concerned, has a remarkable bearing on the discoveries which form the subject-matter of the present book. Light, to Newton, was propagated by the agency of minute material particles or corpuscles, which are emitted from the radiating object and travel in straight lines from their source in all directions outward through space, at a speed which is practically infinite. This expressed an otherwise inconceivable process in the terms of matter and the motion of matter. Science outgrew, as it advanced, these two Newtonian conceptions. It came to be realised that something else existed besides matter which was capable of motion. The idea of a universal ether filling all space and constituting a medium for the communication of motion over interstellar distances was naturally only admitted after it had been conclusively proved that the older ideas were insufficient. The crucial test, which showed that the emission or corpuscular theory was untenable, arose out of the properties of light radiation, and from that time the rival view, known as the undulatory theory, which postulated the existence of a luminiferous or light-carrying ether, has held the field. According to this view, light is propagated as a succession of waves in the ether, much in the same way as the waves of the sea are propagated. In the latter, the moving particles of water have an oscillatory motion in a vertical line, whereas the wave travels forward horizontally. In the transmission of light the oscillatory movement of the ether is also transverse to the direction of motion. The velocity of propagation is constant for all kinds of radiation which result from the transverse vibration of the ether, but the wave-length, and, correspondingly, the frequency or number of vibrations per second, vary over a very wide range. As the former decreases and the latter increases we pass, without any sudden break of continuity, from the invisible heat-radiations occupying the infra-red region of the spectrum through the region of visible light from red to violet to the extreme ultra-violet waves, so easily absorbed even by air that their investigation at last reaches the limit of experimental possibility.

During the last century the undulatory theory received an enormous extension at the hands, primarily, of Clerk-Maxwell, who put forward the electromagnetic theory of light which is based on the discovery of Faraday, of the phenomenon of electromagnetic induction. The original undulatory theory postulated only one attribute—namely, motion—to the ether. Physicists occupied themselves with investigating the laws which the motions of the ether obey. One school, the most brilliant of whom was Lord Kelvin, for a long time attempted to explain the motion on ordinary mechanical conceptions, in which the ether was given certain material attributes, such as elasticity and incompressibility, and the forces acting on it were assumed to be ordinary mechanical stresses and strains. To the newer school the discovery of Faraday of electromagnetic induction supplied the key to the nature of the strains and stresses in the ether which produce light waves. Clerk-Maxwell developed the view that a ray of light was due to waves of electromagnetic induction set up in the ether by the transverse oscillatory movement of an electric charge, and this view was confirmed by the proof that the velocity of propagation of electromagnetic induction through space is the same as that of light. But it was not until after 1888 that the view was generally accepted. In this year Hertz produced electromagnetic waves from the oscillatory discharge of the Leyden jar, and showed that these waves, although of wave-length in some cases several feet long, and hundreds of millions of times longer than the longest wave-length of visible light, yet travel at the same velocity as light waves and are subject to the same laws of reflection, rerfaction and polarisation. The length of wave is regulated simply by the form and dimensions of the apparatus employed. To produce waves of the length of visible light we should have to use single *atoms* or molecules of matter. So the general position has been reached that light waves are caused by the rapid vibration or oscillation of electric charges within the atomic or molecular structure, the oscillation being continuously maintained at a certain definite period corresponding to each wave-length emitted. For the application of this idea to the problems of the spectroscope, and the effect of the magnetic field on the lines of a spectrum, works on spectroscopy must be consulted.

Thus, in 1895, at the beginning of the decade with which we are more nearly concerned, all the known radiations, from the extreme waves of the ultra-violet region of the spectrum to the long Hertzian waves now employed in wireless telegraphy, were satisfactorily accounted for as undulations or vibrations in the luminiferous ether, essentially the same in nature, and differing only in their wave-length and frequency. At the present day, ten years later, this explanation holds good without modification so far as these types of radiation are concerned, but there has been a great extension in our knowledge of radiations. It has been recognised that the undulatory variety is by no means the only example of radiation existing in Nature. The rays we shall be most concerned with are of a totally different character, and constitute the realisation of the conception by which Newton, in his corpuscular theory of radiation, sought to explain the character of light. Thus the term *radiation* to-day applies with equal propriety to two fundamentally distinct phenomena: (1) The older-known class of ether vibrations; (2) a new class recognised within the last decade to be caused by the radiant expulsion of corpuscles or minute particles of matter, projected through space at exceedingly high velocity. The first example of this latter class was correctly recognised by Sir William Crookes 30 years ago, but his explanation was not then accepted. Crookes found that when the electric discharge is passed through a nearly vacuous space, as the degree of exhaustion is increased the character of the discharge alters, the so-called "dark space" around the cathode fills the whole tube, and "rays" proceed from the cathode normally to its surface which travel in straight lines through the tube and cause strong phosphorescence where their passage is arrested by the glass walls opposite the cathode. These "cathode rays" have very remarkable properties. If any obstacle is placed in their path it is heated and may be brought to incandescence if the discharge is sufficiently powerful. Sharply defined shadows of the obstacle are projected on the opposite walls of the tube, the glass not phosphorescing where it is protected by the obstacle from the impact of the rays. Small lightly-poised vanes of mica placed in the path of the rays are driven round with great speed. But the most remarkable property of the rays is that they are deviated by a magnet, and, if the latter is sufficiently strong, can be made to assume circular or helical paths around the lines of force. Crookes gave as the explanation of the phenomenon that matter existed in high vacua in a fourth state, which he named *ultra gaseous* or *radiant* matter. He supposed the effects to be produced by the flight of charged particles or atoms repelled from the cathode, and attaining in the electric field very high velocity and kinetic energy. This explanation has now been substantially adopted, but only within the last decade. We shall see (Chapter III.) that we are forced to regard the particles as smaller than any known atoms, and that they would be more appropriately known as atoms of negative electricity. Without, however, anticipating this conclusion, it may safely be stated that the cathode ray constituted the first known type of a corpuscular radiation caused by the emission of small material particles or, at least, of particles which possess the ordinary attributes of matter. In one sense it is not a radiation, for it is propagated normally to the surface of the cathode, so that with a flat cathode the rays are propagated as a parallel beam. The phenomenon could only strictly be described as a radiation when a spherical cathode is employed. This is due to the radiation being directed along the lines of the electric force which produces the motion of the charged radiant particle. New types were soon to be discovered to which this restriction does not apply, and which in every sense conform to the original Newtonian conception of light.

But many successive discoveries had first to be made, and the historical order in which this result has been achieved is a very interesting chapter in the progress of science. The chapter opens in 1895, at the beginning of the present decade, and this date may be well said to commence an era of new and remarkably rapid advance in the physical sciences. In this year Röntgen discovered the X-rays. For over 20 years Crookes tubes had been in general use for demonstration purposes, but it was reserved for Röntgen to discover, and then by an accident, that they emit a new and remarkable kind of radiation which, unlike the cathode rays, are capable of passing through the walls of the tube and so penetrating the external space. The rays are invisible, but, like the cathode rays *inside* the tube, their presence is manifested by their power to cause strong fluorescence when they impinge upon certain substances, of which the platino-cyanide of barium is the most commonly used. In this way they are virtually made visible to the eye, and they were so discovered. Another property they possess is the power of affecting sensitised photographic plates in the same way as light, and a third very remarkable property is their power of making the air, or other gases, through which they pass, and which, under ordinary circumstances, are practically perfect insulators, capable of conveying limited quantities of both positive and negative electricity. This process is known as *ionisation*, and the rays are said to *ionise* the gases—*i.e.*, to make them for the time being partial conductors of electricity.

These three methods furnish the means whereby the invisible X-rays of Röntgen may be investigated. An explanation of the manner of their production from a Crookes tube was soon forthcoming. Their source was traced to the obstacles bombarded by the cathode rays. If the latter had an uninterrupted passage through the tube, the X-rays resulted at the glass against which the cathode rays impinged. But it was soon found that far more powerful effects could be obtained by inserting into the path of the cathode rays an obstacle in the form of a plate of one of the heaviest metals—platinum, iridium, osmium or uranium. Platinum, on account of its other valuable qualities, is most frequently employed. Moreover, if the cathode is made concave, the rays, being expelled normally to its surface, converge to a "focus." At this focus the platinum plate, or "anti-cathode" as it has come to be called, is placed somewhat obliquely to the path of the rays. This is the disposition in the ordinary "focus-tube" which was first designed by Prof. Herbert Jackson, of King's College. The effect is represented in Fig. 1. AB is the concave cathode, and the dotted lines represent the cathode rays converging to the focus F, where they strike the anti-cathode. The X-rays radiate away from the plane surface of the anti-cathode in all directions, and cause the glass of the tube to fluoresce strongly and uniformly over a hemispherical area, CDE, bounded by the plane of the anti-cathode. A satisfactory theoretical explanation, on the electromagnetic theory, of the nature of the X-rays was soon put forward. When the charge carried by the cathode-ray particle is suddenly accelerated (negatively), and its velocity reduced to zero almost instantaneously by its collision with the dense material of the anti-cathode, a pulse of an electromagnetic nature radiates from the latter, and this pulse constitutes the X-ray.

Thus the X-rays, like light, are ether waves, and the difference seems to be that in the former the disturbances are of the nature of sudden pulses very rapidly dying away, whereas in light there is a regular succession of undulations of the same kind. This, together with their probably extremely short wave-length, would account for the fact that the X-rays have not been reflected or refracted or polarised, although in their nature they so nearly resemble light rays.

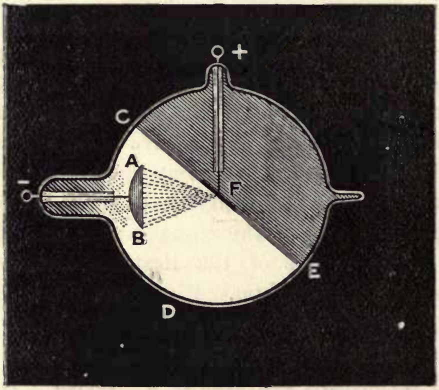


FIG. 1.

The X-rays are of a far more penetrating character than the cathode rays, and easily penetrate the glass walls of the tube. The higher the degree of exhaustion of the tube the greater the penetrating power of the X-rays produced. A tube in which the exhaustion has not been carried to the extreme limit gives out an easily absorbed type of X-ray, and is known as a "soft" tube. In a "hard" tube, on the other hand, the vacuum is so good that a very great difference of potential between the electrodes is necessary to force the discharge through; the cathode rays in consequence attain a very high velocity, and the X-rays they produce on impact with the anti-cathode are of a high penetrating power.

The difference of penetrating power between the X-rays and the cathode rays is one of degree only, and not of kind. The cathode rays share to a limited extent the property of the X-rays of penetrating matter which is completely opaque to ordinary light. The ordinary ideas connected with the terms "transparency" and "opacity" are derived solely from the behaviour of light in every-day experience, and a type of radiation to which these ideas does not apply seems strange and abnormal. In reality the behaviour of light is more remarkable than that of the X-rays and the cathode rays. To these new kinds of radiation all matter is, as a first approximation, equally opaque or equally transparent if equal *weights* are compared. For equal *thickness* the absorption is roughly proportional to the density, and independent of the nature of the matter traversed. Thus Lenard found that, if windows of thin aluminium foil are inserted in the walls of a Crookes tube in the path of the cathode rays, the latter are able to penetrate the foil, and their absorption by different kinds of matter could be investigated outside the tube. He found that bodies as different in their nature as air and gold absorbed the rays very approximately in proportion to their density.

Compare with this simple result the behaviour of light. Certain bodies, like glass, most liquids and gases, rock salt, quartz, calcite and other dense crystals, hardly absorb light at all, while other substances, especially the metals, are almost completely opaque even in the thinnest layers. There has been found to be a connection between the transparency of matter and its electrical properties. With but few exceptions the transparent bodies are the best insulators. The whole range of frequencies should be taken into account, and not merely the visible part of the spectrum. Thus ebonite, according to Prof. Silvanus P. Thompson, is very transparent to the infra-red region of the spectrum, while Prof. Wood has recently described a dye, nitroso di-methyl aniline, which is opaque to the visible rays, but transparent to the extreme ultra-violet rays. When a body exists in two forms, one of which is transparent or translucent and the other is opaque—for example, selenium, the double iodide of mercury and copper, &c.—the transparent form is always the best insulator. These facts serve to indicate that there probably exists a connection between the electrical properties of matter and its transmission of light and are here introduced for the sake of contrast. To the new types of radiation it does not seem to make much difference whether the matter traversed is gaseous, liquid or solid, insulating or conducting, " transparent " or " opaque." The absorption is regulated, at least mainly, by the mass of matter traversed and not by its nature. Thus, although the cathode rays are considered to consist of radiant particles and the X-rays of electromagnetic pulses, the two types exhibit a surprisingly close resemblance in their properties. Both ionise gases, affect the photographic plate, and excite fluorescence, and both are independent of the more or less arbitrary considerations which apply to the absorption of light by matter. They are, however, distinguished, first, by the fact already mentioned, that the X-rays are propagated radially with uniform intensity in all directions from their source, and secondly by the action of a magnetic field.

The path of the cathode rays is deviated by a magnet, whereas the path of the X-rays is not known to suffer deviation. In this way it is easy to show that the cathode rays are the cause and the anti-cathode the source of the X-rays, for, if the former are deviated by a magnet so as to no longer impinge on the latter, the production of X-rays to a large extent ceases.

In 1896, with the awakening that followed the researches of Lenard and Röntgen to the existence of new types of radiations of a character utterly different from those of light, came the discovery of the property of radio-activity. At first it was thought that the fluorescence of the glass in a Crookes tube under the impact of the cathode rays was the cause, rather than, as we now know, an accidental accompaniment of the X-radiation. M. Poincaré suggested that the production of X-rays might be an effect general to fluorescence which had previously been overlooked, in the same way as it had been overlooked during the 30 years in which the Crookes tube had been in use.

M. Becquerel, acting on this idea, examined some fluorescent compounds of uranium. His method was to place the bare salt above a photographic plate, which was carefully wrapped up in opaque material, and so protected completely from the direct action of light, and to expose the salt to direct sunlight, so as to cause it to fluoresce. He found that his plate was affected in these circumstances, even when a layer of copper foil or aluminium was interposed between the substance and the plate. But M. Becquerel also soon found that the exposure to sunlight was unnecessary, and the same effect was obtained in absolute darkness, even when compounds of uranium were employed which, since their preparation, had never been exposed to light. Moreover, he found he obtained the same result whatever compound of uranium was employed, whether it was fluorescent or not, and he soon satisfied himself that he was investigating an entirely new property *of the element uranium* which was completely unconnected with the property of fluorescence (see *Comptes Rendus*, CXXII., 1896, pp. 420, 501, 559, 609, 762 and 1,086). This property is now generally known by the special term "Radio-activity," and its investigation has already lead to some remarkable conclusions of a very far-reaching character. The characteristics of the radiation from uranium are very similar to those of the X-rays. The penetrability of the rays for ordinary matter is, as a first approximation, a function of the density of the matter and not of its nature. The rays ionise the air, or other gases through which they pass, and make them for the time being limited conductors of electricity. This can be easily shown by a gold-leaf electroscope, which is one of the oldest of electrical instruments, and has suddenly assumed a new importance as a means of detecting and measuring the ionisation of the air, and, therefore, the intensity of the new kinds of radiation. It is strange to reflect that it was not for the lack of means of detection that the property of radio-activity remained for so long unknown.

Fig. 2 represents a simple form of gold-leaf electroscope, charged and uncharged. It consists ordinarily of two narrow gold leaves fastened to either side of a strip of metal, which is supported inside of a glass vessel coated internally with tinfoil, by means of a rod passing through a stopper of some good insulator, such as paraffin, shellac or ebonite. To charge it a piece of rubbed sealing-wax, or ebonite, is brought in contact with the upper end of the rod, and the leaves diverge owing to the repulsion of similar electrical charges. If the insulating stopper is in proper condition, the leaves once charged should remain so for hours, or even days. If a small quantity of uranium compound is put inside the vessel the leaves collapse within a few minutes. By making the instrument small the effect may be sufficiently rapid to be followed with the eye. The sensitiveness of the instrument, and the rate of collapse under given conditions, depends on its capacity, and this is regulated mainly by the area of the leaves and their support which receive the charge.

It may be pointed out that the electroscope would retain its charge indefinitely *in vacuo*, even in the presence of uranium, or when acted upon by the X-rays—*i.e.*, the gas present plays a direct part in the transport of the electricity. The uranium rays fail to excite fluorescence (for example, in barium platinocyanide), but this almost certainly is because of their *excessively feeble character* compared with the X-rays from an ordinary tube. Rutherford has also conclusively shown (*Phil. Mag.*, 1899, V., 47, p. 111, that the uranium rays are like X-rays, non-polarisable and non-refrangible.

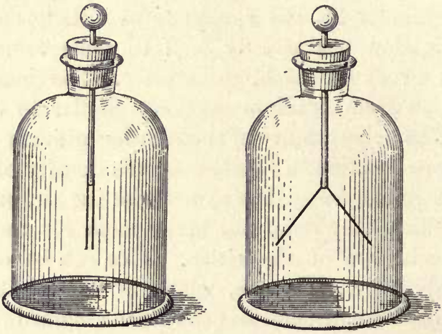


FIG. 2.—Two Gold-leaf Electroscopes, Uncharged and Charged.

It will be convenient at the present stage to anticipate the results dealt with in Chapter V. to the extent of giving a general account of the nature of the rays from uranium and the other radio-active substances. These radiations have been analysed into three distinct types, which are differentiated in the first instance by their power of penetrating matter. The three types have been termed respectively α, β and γ-rays. The former are so feebly penetrating that they are completely stopped by a single sheet of note paper or by a few centimetres of air. The β-rays resemble ordinary X-rays in penetrating power, and pass with ease through thin metal foil, glass, &c., but would be nearly all stopped by a single coin. The γ-rays are by far the most penetrating kind of ray known, and pass through a pile of 12 coins without being completely absorbed. In each case it has been found that the simple law is approximately followed, the absorption being mainly proportional to the density and independent of the nature of the matter. Of the three types the α-rays are the most and the γ-rays the least important. The γ-rays from a kilogramme of uranium, for example, can only just be experimentally detected by the most sensitive test known. We shall see that Becquerel has proved the β-rays to be identical in type with the cathode rays of the Crookes tube. The difference is that the β-rays are travelling with a far higher speed than the cathode rays ever attain, and in consequence have a much greater power of penetrating matter. The proof rests upon the fact that the β-rays are easily deviated by a magnet in the same direction as the cathode rays. The γ-ray is not at all deviated by a magnet, and it is probable that the γ-ray stands in a similar relation to the β-ray as the X-ray stands to the cathode ray, being produced when the charged particle which constitutes the latter is suddenly accelerated at the moment of its expulsion. The β-rays are most active in impressing the photographic plate. They represent, however, but a very small fraction of the total radiation if the ionisation of the air is used as the basis of detection. The α-rays are mainly operative in producing this effect. These have been shown by Rutherford to be very slightly deviated in a powerful magnetic field in the opposite direction to the cathode or β-ray. We shall see that, like the latter, they are caused by the radial expulsion of particles, which, however, carry a positive instead of a negative charge. Moreover, the radiant particle of the α-ray is by no means small, but is a body of about the mass of a hydrogen atom, and it is expelled from the radio-active substance with the colossal speed of 20,000 miles a second. In the α and β types of rays we have exactly the realisation of Newton's corpuscular theory of light. At first they were mistaken for a type of extreme ultra-violet light radiation, and their wave-lengths were actually estimated. These experiments are now known to be erroneous. But it is curious that the first wave-form of radiation known was initially considered to be due to the emission of corpuscles, and that the first type of the latter class of radiation was conversely mistaken for a peculiar kind of wave vibration.

In some respects these recent discoveries may be said to have provided an additional indication of the scientific intuition and foresight of Newton. To the triumphant supporters of the electromagnetic theory of light the corpuscular theory must have appeared too crude to merit consideration. To-day we know that its conception anticipated by three centuries the march of scientific progress.

This chapter was designed to give at the outset a review of the generally accepted theories of the nature of the many kinds of radiation, old and new, which come within the range of modern physics. Two classes of radiation have been recognised, and yet the various types of both classes seem to be distinguished from one another by a gradual rather than a sudden break of properties. There is no single criterion which serves to differentiate an ethereal disturbance from a corpuscular radiation, except, perhaps, the action of a magnetic field. Even this test would fail in the easily-conceived case of an *uncharged* radiant particle.

The power of ionising a gas, which is a common characteristic of the newly discovered radiations, has recently been shown to be possessed by ultra-violet light of *extremely* short wave-length (below 2,000 Ångstrom units). The explanation of the transparency of insulators to electromagnetic undulations of the character of light would seem to necessitate that the X-rays and the γ-rays should be more easily absorbed by conductors than by insulators. Yet this has not been observed. Indeed, from their properties, these two types seem to be more nearly allied to the cathode rays and the α and β-rays from the radio-active substances than to light. These considerations may serve to show that the present division adopted is somewhat unsatisfactory, and that probably in the future it will be seen that a closer connection exists between the corpuscular and undulatory forms of radiation than our present views of the relations between electricity and matter reveal.

CHAPTER II.

THE RADIO-ACTIVE ELEMENTS—URANIUM, THORIUM, RADIUM, POLONIUM AND ACTINIUM.

Uranium and Thorium, the only examples of Radio-activity among the known Elements.—Abnormal Radio-activity of Pitchblende.—Discovery of Radium and Polonium.—Radio-activity an Atomic Property of Matter.—Radium.—Source.—Method of Extraction.—Atomic Weight.—Spectrum.—Radio-active Properties contrasted with ordinary Material Properties.—Polonium.—Work of Marckwald.—Actinium.—Work of Giesel.—The Five Radio-elements and their Distinguishing Characteristics.—Source of the Energy of Radio-activity.—The Two Alternatives.

Soon after M. Becquerel's discovery of the radio-activity of the element uranium, Mme. Curie (Thesis presented to the Faculté des Sciences de Paris, *Chem. News*, 1903, p. 85, *et seq.*) made a careful examination of practically all the known elements, and found that one only—viz., thorium—possessed the new property. Schmidt simultaneously discovered the radio-activity of thorium. With regard to the other elements, Mme. Curie states that if they possess radio-activity it can only be to an extent at least 100 times more feeble than that possessed by uranium and thorium.

The compounds of these two elements, when examined for their power of ionising a gas and discharging an electroscope, are found to possess a very similar degree of radio-activity. To the photographic plate, however, thorium is several times less active than uranium. This result we now know is to be explained by the fact that the α-radiation from the two bodies is of similar intensity, but the β-radiation of thorium compounds is much feebler than in the case of the uranium compounds.

Although none of the known elements in the pure state possessed appreciable radio-activity, Mme. Curie observed that certain ores of uranium and thorium, notably the pitchblendes, were several times as radio-active as the element uranium or any of its compounds. The following is a list of the minerals examined and their activities :—

	$\times 10^{-11}$ amperes.
Pitchblende (Johanngeorgenstadt)	8·3
Do. (Joachimsthal)	7·0
Do. (Pzibran)	6·5
Do. (Cornwall)	1·6
Clèveite	1·4
Chalcolite	5·2
Autunite	2·7
Thorite	1·4
Crangeite	2·0
Monazite	0·5
Æschynite	0·7
Fergusonite	0·4
Samarskite	1·1
Niobite	0·3
Carnotite	6·2

The method of measurement will be considered later, but the activity is expressed by the current in amperes which was enabled to pass through the air, under the action of the rays, from a layer of the powdered substance of area 64 sq. cm.

The following table gives the radio-activity, on the same scale, of uranium and its pure compounds :—

	$\times 10^{-11}$ amperes.
Metallic uranium (containing carbon)	2·3
Black oxide, U_2O_5	2·6
Green oxide, U_3O_8	1·8
Uranic hydrate, $UO_3 . 2H_2O$	0·6
Sodium uranate, $Na_2U_2O_7$	1·2
Potassium uranate, $K_2U_2O_7$	1·2
Ammonium uranate, $(NH_4)_2U_2O_7$	1·3
Uranium sulphate, $UO_2SO_4, 3H_2O$	0·7
Potassium uranyl sulphate, $UO_2SO_4, K_2SO_4, 2H_2O$	0·7
Uranium nitrate, $UO_2(NO_3)_2 . 6H_2O$	0·7
Phosphate of copper and uranium	0·9
Uranyl sulphide, UO_2S	1·2

It will be seen from the above table that the radio-activity depends mainly on the percentage of uranium present in the compound, although the density and state of division of the compound, by altering the proportion of the rays absorbed in the substance itself, exerts an influence on the observed activity. But the Johanngeorgenstadt pitchblende, which probably contains about 70 per cent. of uranium, is nearly four-times as active as the most active uranium compound. Mme. Curie next tried whether an artificial mineral, prepared from pure materials, would show any higher activity than the materials of which it is compounded. She chose for the purpose chalcolite, which is a phosphate of copper and uranium, and she found the artificial preparation possessed an activity corresponding to its composition—viz., 0·9, compared with 5·2, the activity of the natural mineral.

Mme. Curie came to the conclusion, as the result of these experiments, that *radio-activity is an atomic property* which is independent of the physical or chemical state of the active element, but is proportional only to the quantity of active element present, when compounds of similar density and states of division are compared. On this view, the high activity of the natural minerals examined could only be explained by supposing them to contain a *new* element (since none of the known elements except uranium and thorium are radio-active) many times more active than uranium, which had hitherto escaped detection by the analyst.

M. and Mme. Curie proceeded to the decomposition of pitchblende, and its separation by chemical analysis into its constituent elements. Each preparation was tested for radio-activity. If the high activity of pitchblende is due to the presence of highly active new elements, it is to be expected that the radio-activity in certain of the fractions will increase at the expense of the activity of the others.

The expected result was obtained, and it was found that the bismuth and the barium separated from pitchblende were both strongly radio-active. These active preparations were first termed "radio-active bismuth" and "radio-active barium" respectively, but afterwards, when it became clear that the activity could only be caused by the presence of new elements, the names "polonium" and "radium" respectively were given to the latter.

Radium.—It will be convenient to consider the case of radium first, in the discovery of which M. Bemont was associated with M. and Mme. Curie. Radium is the only new radio-element that has so far been isolated in the form of pure compounds, or which has been found to give a new spectrum. It was discovered that the barium separated from pitchblende possessed marked activity, and, if the active barium chloride was fractionally crystallised, the activity tended to concentrate in the least soluble fraction. The active barium preparations were, as soon as possible, submitted to M. Demarçay for spectroscopic examination, in order that the hypothesis of the existence of a new element might be confirmed. In the first specimens a new line in the ultraviolet ($\lambda = 381{\cdot}47 \mu\mu$) of considerable intensity was detected, and, as preparations were obtained of higher and higher activity, this line became stronger and new lines appeared. As the fractionation of large quantities of material was proceeded with small quantities of intensly active products were prepared, in which the new spectrum predominated and only the three strongest barium lines were visible. Finally a product was obtained, in the spectrum of which the presence of barium lines was scarcely detectable.

The quantities of pure radium compounds so obtained are excessively small. A few tenths of a gramme only of radium chloride can be extracted from a ton of pitchblende, and this is in the ratio of one part to several millions of the original mineral. On the other hand, the radio-activity of the pure compound is correspondingly increased, and the tiny quantity extracted from a ton of ore retains in concentrated form the greater part of the radio-activity of the original mineral. Weight for weight, the radium compounds are at least a million times more active than the compounds of uranium and thorium. This numerical comparison hardly conveys a true impression of the relative activities. A clearer idea is given by a concrete example. If a radium compound produced a given effect, for example, on a photographic plate, in one second, a similar weight of uranium compound would take several *weeks* to produce the same effect. This is illustrated by Figs. 3 and 4. The first illustrates a radiograph of an aluminium medal taken with uranium rays in which probably several grammes of a uranium compound were employed, and the exposure given was 14 days. The second illustrates a negative obtained by merely writing slowly on a photographic plate with a tube containing a few thousandths of a gramme of a pure radium preparation.

The use made by M. and Mme. Curie of the property of radio-activity as a means of detecting and separating new

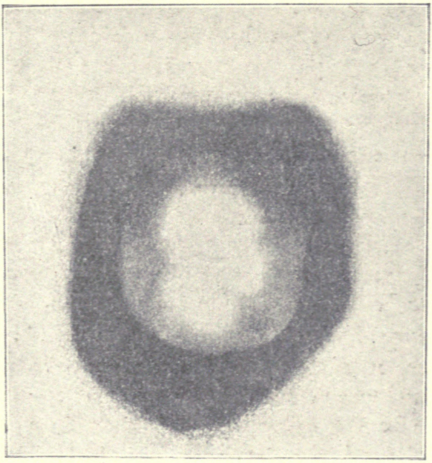


FIG. 3.—Radiograph of an Aluminium Medal taken by M. Becquerel with the Uranium Rays. Exposure: A fortnight. (From "Rapports du Congrès International de Physique, 1900," Tome III., p. 51.)

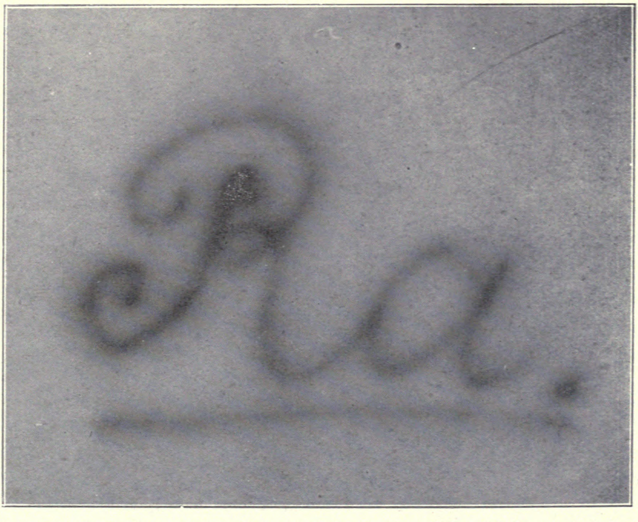


FIG. 4.—Negative produced by Writing with a Glass Tube containing a few milligrammes of Pure Radium Bromide on a Photographic Plate wrapped in Black Paper.

elements is strictly analogous to the use of the spectroscope made by Bunsen and Kirchhoff for the same purpose, in their discovery forty years ago of two new elements, rubidium and caesium. Thus 44,200kg. of Dürkheim water were worked up by these investigators and 7·272 grammes of caesium chloride obtained therefrom, the sole guide to the existence and, in the early stages of the separation, to the analytical behaviour of this new element being the two bright blue lines in the flame spectrum of caesium.

Mme. Curie states that the strongest line of radium cannot be observed with specimens less radio-active than fifty times that of uranium. Since radio-activity $\frac{1}{150000}$th of this can be directly detected (compare Strutt, *Phil. Mag.*, 1903, VI., 5, p. 683), it follows that for radium the radio-active test is 150,000 times more sensitive than the spectroscopic. Nevertheless, Demarçay classes radium among the elements which possess the most sensitive spectrum reaction. In pitchblende itself the quantity of radium is probably about one-twentieth of the minimum quantity detectable by the spectroscope. Recently it has been demonstrated that radium is very widely distributed in nature. The test used is not the direct radiation, but depends on the fact that radium produces an *emanation*, or radio-active gas, whose presence can be detected in almost inconceivably minute quantities. The presence of the radium emanation, and, by deduction, the existence of radium, has been detected in the soil of many localities, in tap water and in various springs, also (compare J. J. Thomson, Cambridge Phil. Soc. *Proceedings*, February 15th, 1904) in the commonest materials, such as flour, the sand of the sea shore, &c. In fact, it is hardly too much to expect that, as the methods of investigation are improved, and sufficiently large quantities of material are worked with, radium will appear as a universal constituent. This is mainly due to the fact that the means of detecting it are almost infinitely more sensitive than those possible for any of the older known elements. Conversely, the property of radio-activity has supplied the chemist with so powerful a new weapon in the search for excessively rare elements possessing the property that a great extension of our knowledge in this direction is to be anticipated in the near future.

The principle underlying this method of research, and which is also at the root of the important theoretical advances that have been so quickly made in the subject, depends upon the fact that radio-activity is an atomic property. When the principle was first enunciated by Mme. Curie, it connoted that the activity of any radio-element was not influenced by its state of chemical combination or by the conditions under which it was tested. Thus any mineral containing uranium or thorium is radio-active without reference to the other inactive elements it contains, or to the conditions of its past history. We now know that, if proper precautions are taken, the radio-activity of a substance furnishes a good quantitative measure of the amount of the radio-element it contains. The radio-activity of matter is a property which can neither be increased nor diminished by any artificial means.

Matter, strictly speaking, possesses only one other "atomic property," and that is *mass*, which, like radio-activity, is an additive property of the atoms, each constituent atom contributing its quota without possibility of its being changed by any agency we know of. This is shown by the fact that mass, or, as it is usually measured, *weight*, is the only property that can be used universally as the basis of chemical analysis. Of other properties spectrum reactions represent probably the nearest approach to an atomic property, for, within certain limits of temperature, the spectrum of an element is the same whatever the compound employed. But the spectrum reaction is not an additive property, and the intensity of a spectrum is by no means a sure indication of the quantity of element present, for many spectra are "masked" by comparatively small quantities of impurities. Radio-activity is, of course, distinguished from the property of mass or weight in being a very special property of a certain few types of atoms, and not a universal property of all.

In spite of the discovery of many new facts, which at first sight seemed to contradict the view that radio-activity is an atomic property, there is no doubt of its truth at the present time. It has not been found possible to influence radio-activity by any known agency, any more than it has been found possible to alter weight, and the more free from disturbing causes (such as absorption) the measurements are, the more accurately this result holds good. Hence radio-activity is fitted in every respect as a property upon which a method of quantitative chemical analysis could be based. We shall see later that radio-activity furnishes a means of qualitative analysis also. There are certain peculiarities which enable us instantly to recognise thorium, or radium, from uranium (even were all these elements mixed) by the character of their radio-activity, without performing a single chemical or spectroscopic test with the substance. Conversely, by an examination of a radio-active mineral, one can be sure, before a single chemical test is employed, whether it contains any new and hitherto unrecognised active elements, or whether its activity is due to the known radio-elements. The finest and best methods of chemical analysis do not surpass in ease of operation and certainty the methods of "radio-active analysis," whereas the latter tests are applicable for quantities millions of times less than the former.

Source of Radium.—So far radium has only been found in notable amount in minerals containing uranium, and very few of these appear to contain a sufficient quantity to repay extraction. Not all pitchblendes are equally rich in the element; for example, the Cornish pitchblende, although it contains a high percentage of uranium, possesses such a low activity (1·6 compared to 8·3 for the Johanngeorgenstadt ore) that it can only contain little, if any, of the element. Practically the ore from Joachimsthal provides the whole of the available supply. A method for the accurate estimation of radium in ores will be given later, after its properties have been more closely studied. The quantity obtainable even from the best ores is excessively small. Mme. Curie states that from 2 tons of residues from the Joachimsthal ore she obtained a few hundredths of a gramme of the pure chloride and a few tenths of a gramme of considerably less active material. Giesel, who has probably prepared the largest quantities of concentrated radium compounds, obtains about 0·25 gramme of pure radium bromide from 1 ton of the residues of the pitchblende after the commercially valuable uranium has been extracted.

Method of Extraction.—The following method of treatment of the residues from the Joachimsthal ores has been worked out by Debierne and is taken from Mme. Curie's Thesis :—To extract the uranium, the ore is roasted with sodium carbonate, lixiviated with warm water and then with dilute sulphuric acid, when the uranium passes into solution. The insoluble part consists of the sulphates of lead and calcium, alumina, silica and iron oxide, together with greater or less quantities of nearly all the metals. These residues possess an activity four and a-half times that of uranium, and constitute the raw material used for the extraction of radium. The insoluble sulphates are converted into carbonates by boiling with a concentrated solution of carbonate of soda, and the soluble sodium sulphate produced is removed by repeated washing. The residue is treated with hydrochloric acid, which dissolves most of it, including the polonium and actinium, but the radium remains undissolved, as unconverted sulphate. It is washed with water, again boiled with concentrated sodium carbonate (which completes the transformation of the sulphates into carbonates), again thoroughly washed, and then treated with dilute hydrochloric acid free from sulphuric acid. Polonium and actinium are still present in the solution, from which the radium and barium are removed by precipitating with sulphuric acid. From 1 ton of residues 10kg. to 20kg. of crude sulphates are thus obtained of activity about 60 times that of uranium, and these contain calcium, lead, iron and a trace of actinium. The sulphates are transformed into chlorides as before, and the solution treated with sulphuretted hydrogen, filtered, oxidised with chlorine and precipitated with ammonia. The activity of the precipitated hydrates and oxides is due to actinium. The filtrate is precipitated with sodium carbonate, and the precipitate washed and converted into chlorides, evaporated to dryness, and the chlorides washed with concentrated hydrochloric acid, which removes calcium. The purified chlorides of barium and radium thus obtained possess an activity of about 60. Eight kilos are obtained from 1 ton of residues. At this stage the material leaves the factory, and is now fractionated in the laboratory.

Mme. Curie's system was based upon the fact that if the mixed chlorides are dissolved in water, raised to the boiling point, and allowed to cool, the small part that crystallises out is five times more active than the part left in solution. By successive repetitions of this process the mixture is divided into two parts—the one consisting of the great bulk of the material, which may be obtained only one-tenth as active as uranium, the other consisting of continuously diminishing quantities of preparations with correspondingly increasing activity. As the fractionation advances it is convenient to dissolve in water containing gradually increasing quantities of hydrochloric acid. The salts are less soluble in the latter than in water, and larger volumes of solution are obtained. When the proportion of radium increases to a certain point, the crystals of mixed barium and radium chloride become coloured after a few hours, the colour disappearing when the crystals are re-dissolved. A maximum coloration is reached when the radium present is of a certain concentration, and then again diminishes as the preparation becomes more concentrated. This serves as a check on the progress of the fractionation.

Giesel (*Berichte der Deutschen Chemischen Gesellschaft*, 1902, p. 3,609) has improved upon the method by using the bromide instead of the chloride for fractionation. He states that eight crystallisations suffice to remove the easier soluble barium bromide, leaving the radium bromide approximately pure. But it is probable that to approach the purity ultimately attained by Mme. Curie with her method very many more fractionations would be required. Such a degree of purity is, however, only required in certain special cases, and Giesel's method furnishes preparations with sensibly the maximum obtainable radioactivity. Giesel discovered that salts of radium impart to the colourless flame of a bunsen burner a fine carmine red colour, which serves as a useful control over the progress of fractionation. In this respect radium shows its relationship to the other alkaline-earths, which all yield fine flame colorations.

Atomic Weight.—The atomic weight of radium has been determined by Mme. Curie with the preparations obtained during the progress of the fractionations. The method employed was to precipitate the chlorine in a known weight of the chloride with silver nitrate, and to weigh the silver chloride obtained. Besides being accurate and applicable to very small quantities of material, the method possesses the advantage of retaining the radium in the soluble form, as nitrate, after the performance of the operation.

The atomic weight of preparations of activity 230 and 600 times as active as uranium was indistinguishable from that of barium—viz., 137. With a preparation of activity 3,500 the number 140 was obtained, showing a small but distinct difference. Another sample of activity 7,500 gave the number 145·8, while two samples of very high activity, each of the order of one million times that of uranium, gave 173·8 and 225 respectively. In the latter the barium could only just be detected spectroscopically. A mean of three determinations of the purest sample prepared also gave the number 225.

It may be noted that the radio-activity of pure radium salts is generally taken to be of the order of a million times that of uranium, but the exact value is unknown. This is because it is experimentally impossible to directly compare together activities which are so widely different. An absolute measure in electrical units of the radio-activity of pure radium salts has not yet been published.

The value 225 for its atomic weight places radium in the position of the third heaviest element known, the two heavier being the other radio-elements—thorium 232 and uranium 238. It is surely not merely a coincidence that the three elements distinguished by their radio-activity should possess the three heaviest atomic weights. Except in this respect they are chemically very dissimilar, radium being divalent like barium, thorium tetravalent like tin, and uranium hexavalent like tungsten. On the other hand, these elements, which they most resemble, show no trace of radio-activity.

Spectrum.—The following table shows the measurements of Demarçay of the spark spectrum of radium between the limits $\lambda = 500{\cdot}0$ and $\lambda = 350{\cdot}0 \mu\mu$. The intensity of the strongest line is put at 16 :—

	λ	Intensity.		λ	Intensity.
Blue	482·63	10	Violet	460·03	3
	472·69	5		453·35	9
	469·98	3		443·61	8
	469·21	7	Ultra-violet	434·06	12
	468·30	14		381·47	16
	464·19	4		364·96	12

In addition there are two well-marked misty bands in the spectrum, and in this and other characteristics the spectrum is completely similar to those of the other alkaline earth metals.

Runge and Precht (*Phil. Mag.*, 1903, VI., 5, p. 476) have examined the spark spectrum of radium in a magnetic field, and have found that the strongest lines are exactly analogous in their behaviour to the strongest lines of barium, and to the corresponding lines of the related elements magnesium, calcium and strontium. Like the latter, the radium lines may be grouped into three series, a pair of lines in each series. In the second series of each element there occurs, besides the two lines, a third, which they term a "satellite." In calculating the distance apart of the lines of each series, the oscillation-frequency which is the reciprocal of the wave-length is employed. Runge and Precht find that the distance apart between the two lines of the first and third series, and of one of the lines and the "satellite" in the second series, is the *same* for each element and different for different elements. The following table shows the corresponding lines in the spectra of magnesium, calcium, strontium, barium and radium, expressed in Ångstrom units ($\mu = 10$ Ångstrom units) :—

—	Mg.	Ca.	Sr.	Ba.	Ra.
Primary series..........	2,803	3,969	4,216	4,934	4,682
	2,796	3,934	4,078	4,554	3,815
1st secondary series	—	3,181	3,475	4,166	4,436
	2,798	3,179	3,465	4,131	4,341
	2,791	3,159	3,381	3,892	3,650
2nd secondary series	2,937	3,737	4,306	4,900	5,814
	2,929	3,706	4,162	4,525	4,533

The following is a table of the distances apart of the three pairs in the case of radium measured on the scale of frequency :—

—	λ	$10^8/\lambda$	Difference.
Primary series	4,682·35 3,814·59	21,356·8 26,215·1	4,858·3
1st secondary series	4,436·45 3,649·77	22,540·5 27,399·0	4,858·5
2nd secondary series	5,813·9 4,533·33	17,200·2 22,058·8	4,858·6

Now, in a series of elements of the same family in the periodic table, the distances apart of the lines increases with the atomic weight of the element, and from the values in the case of Mg, Ca, Sr and Ba Runge and Precht have extrapolated for the case of radium, and obtained for the atomic weight of radium, the number 257·8. This is considerably above the experimental number found by Mme. Curie, and the discrepancy has not been explained. This matter needs further investigation, but for the present we must accept the experimental value, 225, especially as it is that required by the periodic law.

The position is a very interesting one. Both the chemical and spectroscopic evidence point most strongly to the conclusion that radium is a heavy representative of the alkaline-earth family of element. If old tables of the periodic law are consulted there are two vacancies possible beyond barium, the then heaviest known member of the family. One corresponds to atomic weight about 172, the other to 225. The former is too small to agree with either determination, and need not be considered, whereas the latter is identical with Mme. Curie's number. If the value 257·8 indicated by the spectroscopic evidence were correct, radium would appear in the periodic table allied to the mercury-cadmium family of elements and not to the alkaline-earths.

The line 4,826·14 in the radium spark spectrum is stated by Runge and Precht to be the one most prominent in the flame spectrum, and is analogous to the strongest flame lines Ba 5,535, Sr 4,607, Ca 4,426, for all these lines are resolved into triplets by a magnetic field, the lines in each case being equidistant from one another on the scale of frequency. We thus see that the spectrum of radium, when subjected to the strictest investigation by the modern methods of spectroscopy, is the normal spectrum to be expected for an alkaline-earth element heavier than barium. Radium shows in its spectrum no indication of any special peculiarities which might be connected with its many unique radio-active properties, and this is the more surprising when it is considered that the methods of spectroscopy have thrown a great deal of light on the internal mechanism of the chemical atom.

In marked contrast to its strictly normal character as a chemical element occupying a definite and predicted place in the periodic table, radium possesses in addition a totally different set of properties, which make it in many ways the most extraordinary substance known. These will be generally referred to as its radio-active properties, in contradistinction to the other set, which will be termed the ordinary material properties. There is thus a double nature to the radio-elements, according as their ordinary material properties or their radioactive properties are considered. We shall see later that it is probable that the same set of atoms is not concerned in the two cases, the ordinary material properties being caused by what may be called the normal atoms, which are in overwhelming numerical majority, while the radio-active properties are caused by a very minute and constant fraction of the total in a very critical and unusual state.

It will be convenient to pass in review in a preliminary manner some of the most important of the radio-active properties of radium. The radiations, even from a few milligrammes, are of an extraordinarily powerful character. We have seen that they can be divided into three classes, in the order of their penetrating power. The α or non-penetrating type are stopped by less than an inch of air. They instantly discharge an electroscope or an electrified silk tassel, and cause zinc sulphide to phosphoresce very brilliantly. The scintillations of Sir W. Crookes' "spinthariscope" are brought about by the impact of the α-rays on a zinc sulphide screen. Owing to their enormous kinetic energy and the ease with which they are stopped by material obstacles, the α-rays are the main cause of the heat evolution from radium. By far the greater part of the α radiations are absorbed in the solid compounds of radium, and their energy is converted into heat. In this way radium liberates enough heat every hour to raise its own weight of water from the freezing to the boiling point. They continuously maintain themselves a few degrees above the temperature of their surroundings. The α-rays bring about chemical decompositions, the most remarkable of which is the decomposition of water into its constituent elements. A gramme of radium in aqueous solution liberates 10 cubic cm. of hydrogen and oxygen per day continuously.

The β-rays of radium pass with ease through plates of copper and other metals of the thickness of a visiting card, and will instantly affect a photographic plate or produce brilliant fluorescence on a crystal of barium platino-cyanide on the other side. As they carry away a negative charge, the radium, under favourable circumstances, is continuously kept charged with positive electricity. The γ-rays of radium will affect a photographic plate and produce visible fluorescence on a screen of platino-cyanide through metal screens over an inch thick, or through a pile of 12 coins. In addition, there occurs simultaneously a continuous production of a radio-active emanation, or gas, the absolute amount of which is so small that it can scarcely be directly perceived. Yet this invisible quantity of gas emits enough energy to give astonishing indications of its existence. The quantity of emanation obtainable from a few milligrammes of radium salt will keep a phosphorescent mineral like willemite continuously luminous for several days or even weeks, to a sufficient extent to be visible even in a brightly lighted room.

A fuller consideration of this property and of others connected with it must be reserved till later (Chapter VII., *et seq.*). At this stage the point must be emphasised that, in spite of the properties above mentioned, the radio-activity of radium appears in no fundamental point different from that of uranium and thorium. It is mainly a matter of the phenomena in the former case being exhibited to an extraordinarily greater degree than in the latter cases.

Polonium.—Besides radium there is strong evidence that there exists in pitchblende two other new radio-active bodies, polonium and actinium, and, although neither of these has yet been obtained in sufficient quantity to give a spectrum reaction or any other evidence of their presence, except their radio-activity, the peculiar nature of the latter in each case leaves no doubt that each is a specific new form of matter, whose ultimate separation may be confidently expected. In the fractionation of pitchblende by M. and Mme. Curie, the polonium appeared with the bismuth in the second group of metals precipitated from acid solution by hydrogen sulphide. It resembles bismuth in analytical properties, but it can be concentrated by special methods even from the bismuth, which shows that it is chemically distinct. Mme. Curie describes three methods: (1) By subliming the active sulphide in vacuo, the active part, being the more volatile, tends to deposit on the cooler parts of the tube. (2) By fractional precipitation of the solution of the nitrates with water it is found that the basic nitrate precipitated is more active than the fraction which remains dissolved. (3) By precipitation of the chlorides in strong hydrochloric acid solution with hydrogen sulphide the fraction precipitated as sulphide is again the more active.

So far Mme. Curie has not succeeded in obtaining her preparations free from bismuth. Its radio-activity is peculiar in two respects: (1) It comprises only α or non-penetrating rays, the penetrating β-rays being completely absent. In this respect it is sharply distinguished from the other radio-active substances, uranium, thorium, radium and actinium. (2) Its radio-activity is not permanent, but decays slowly with the lapse of time, falling to half its original value at the expiration of about one year. The discussion of this important point, which throws a great light on the nature of polonium, and why it has so far been impossible to separate it as a distinct element, must be reserved until the numerous other cases of temporary activity which are now known have been considered.

Recently Marckwald (*Berichte der Deutschen Chemischen Gesellschaft*, 1902, p. 2,285) has shown that polonium may be easily separated from the bismuth extracted from pitchblende residues by immersing a polished plate of bismuth in the solution. The whole of the active substance is deposited in highly concentrated form on the plate, while the solution is left inactive. The deposit, which is metallic, can be easily detached, and weighs but a few milligrammes, which, however, when dissolved still shows the presence of bismuth. In a later communication (*ibid.*, p. 4,239) the same investigator has shown that the metallic deposit more nearly resembles tellurium than bismuth in its chemical nature, and he has proposed the name *radio-tellurium* for his preparation, believing it to be distinct from Mme. Curie's polonium. He finds that it is precipitated out of its solutions by stannous chloride as a black precipitate, and recommends this method as giving far purer preparations than that first employed. The activity of these is far higher than the earlier products, but the weight obtained is correspondingly smaller. In a still later communication (*ibid.*, 1903, p. 2,662) Marckwald gives some very interesting details of the actual amount of the new substance present in pitchblende. 2,000kg. of the latter yielded 6kg. of bismuth oxychloride, which gave 1·5 grammes of the tellurium-like precipitate with stannous chloride. He now finds that this is almost entirely ordinary inactive tellurium, which he separates from the active product by precipitating the chloride with hydrazin hydrochloride in not too acid solution. The active substance remains in the filtrate, and after precipitating with stannous chloride was found to weigh 4 milligrammes ! Even so, the author does not consider it proved to be a homogeneous substance. He states that from $\frac{1}{100}$th milligramme of this preparation he has obtained sufficient light, by means of the phosphorescence produced by the rays in a preparation of zinc sulphide, to be plainly visible to an audience of several hundred people.

There seems to be no justification for supposing that the active constituent of Marckwald's preparation is different from that of Mme. Curie. Both preparations are identical in their radio-active properties and are obtained from the same mineral. In this book it will be assumed that "radio-tellurium" is identical with polonium. For a full discussion of this point compare *Nature*, March 17, 1904, p. 461.

On the view that polonium is a new element, this work of Marckwald establishes a record in the achievements of chemical analysis. Before it, the separation of xenon from the atmosphere, one part of the former in between 10 and 100 millions of the latter, by Sir William Ramsay and Dr. Travers, was probably the best example of the possibilities of modern methods in the detection and separation of minute quantities of new elements as they exist in Nature. But in Marckwald's separations the proportion of the new substance in pitchblende is certainly less than this, and may be estimated at less than one five hundred millionth part of the whole. We shall see as we proceed that the minute quantity of polonium in pitchblende is intimately connected with its relatively short-lived activity after separation.

Actinium.—Little has been published about this substance, and the only investigator who has examined it is Debierne, who discovered it in the ammonium hydrate group in the analysis of pitchblende (*Comptes Rendus*, 1899, 129, p. 593; 1900, 130, p. 906; 1903, 136, pp. 446 and 767). The element most nearly allied to actinium in its analytical behaviour was first stated to be titanium and subsequently thorium. In fact, the preparations were shown by a spectroscopic examination by M. Demarçay to consist chiefly of thorium. Like radium, it gives penetrating rays deviable in a magnetic field, and also a characteristic emanation capable of imparting temporary activity to objects in the neighbourhood, but distinguished from the emanations of radium and thorium by a more rapid rate of decay of its activity. On these latterly discovered facts its chief claim to be considered a new radio-active element rests.

Giesel (*Ber.*, 1902, p. 3,608; or *Chemical News*, 1903, p. 97) has also described a substance which appears to be identical with Debierne's actinium. He calls his body "The Emanation Substance from Pitchblende," because it is characterised by great power of giving a radio-active emanation. Both Debierne and Giesel have suggested that the new body is the cause of the radio-activity of thorium, but this seems impossible, on account of the totally distinct character of its emanation. It resembles thorium and the rare earths in its analytical reactions, being precipitated from its solutions by hydrogen peroxide and by oxalic acid. But Giesel has obtained it free from thorium and nearly free from cerium and didymium, and finds the main bulk of the substance consists of lanthanum. Lanthanum is, however, inactive, and stands to actinium in just the same relation as barium to radium, or bismuth to polonium, as the analytically similar element. Giesel's preparation was about one-thousandth as active as pure radium salts. The close resemblance between certain very curious and unique properties of the emanation from both Debierne's actinium and Giesel's emanation-substance (these properties being discovered independently by the two observers for their respective preparations) show that the source of the radio-activity is almost certainly the same substance in the two cases. These special properties of the actinium emanation will be considered later, after the other emanations have been studied more in detail.

The Five Radio-elements.—Five radio-active substances, possessing characteristic radio-activity are thus known—uranium, thorium, radium, polonium and actinium. The activity of the last two probably only persists over a short term of years, while that of the first three is sensibly permanent at least for many centuries. These bodies present many peculiarities among themselves in the kind of radio-activity they exhibit (compare Rutherford and Miss Brooks, *Phil. Mag.*, VI. 4, p. 1, 1902), so that it is impossible that the radio-activity of any two or more of them can be caused by the presence, possibly in minute quantity, of the same substance.

These peculiarities can be grouped mainly under two heads. The first is the character of the rays and their penetration-power. Three types of rays, known as the α, β and γ rays, arranged in the ascending order of their power to penetrate matter, are recognised, and, although the rays of any one kind differ slightly among themselves in penetrating power for different radio-active substances, this is always of the same order for the same type of ray and of a completely different order for different types of rays.

Substance.	α-rays.	β-rays.	γ-rays.	Radio-active emanation.	Imparts activity to surrounding objects.
Uranium	Yes.	Yes.	Yes.	No.	No.
Thorium	Yes.	Yes.	Yes.	Yes.	Yes.
Radium	Yes.	Yes.	Yes.	Yes.	Yes.
Polonium ..	Yes.	No.	No.	No.	No.
Actinium	Yes.	Yes.	?	Yes.	Yes.

The second distinctive feature which serves to differentiate radio-active substances is their power to produce, besides rays, radio-active gases, not necessarily in visible quantities, but possessing *sufficient radio-activity* to be easily detectable. This property was discovered for thorium by Prof. Rutherford, who gave the name "Emanation" to the radio-active gas produced from thorium. Uranium and polonium do not, while thorium, radium and actinium do, possess this property. The respective emanations of the last three are distinguished from one another by the time their activity lasts, the activity of the radium emanation lasting several weeks, that of the thorium emanation only a few minutes, and that of the actinium emanation a few seconds. With the power of a radio-active element to produce a radio-active emanation is bound up its power to impart radio-activity to objects in the neighbourhood. Rutherford discovered this property for the thorium emanation, and designated it the *excited* *activity*. M. and Mme. Curie discovered it simultaneously for the element radium (the emanation of radium was not then known), and called it the *induced activity*. As both terms are misleading in light of present knowledge, the term "Imparted radio-activity" will generally be applied in this book to this phenomenon. In the preceding table a list is given of the radio-active substances and the characteristics of their radio-active properties with respect to the features just enumerated.

Although radium is the only one of the three new radio-elements that has been obtained in quantity sufficient for its ordinary material properties to be investigated, the strongest evidence in favour of considering it a new element is the unique and specific character of its radio-active properties. The emanation of radium in the rate of decay of its activity and its power of imparting a peculiar sort of temporary activity to bodies in contact with it is a far finer test for the presence of radium than the spectrum of the latter element. On this account it is certain that the activity of the other four radio-elements is not, for example, due to the presence of radium. Conversely, it appears likely that certain supposed new radio-elements, as, for example, the so-called radio-lead, consisted merely of inactive matter admixed with a trace of radium. The radio-active gas that has been described from metallic mercury, from soils and water, &c., has the characteristics of the radium emanation, and owes its origin, in all probability, to the same cause. On the other hand, a new type of emanation, for example, indicates the presence of a new radio-element, and many such will, no doubt, be discovered by this test in the future.

Source of Radio-active Energy.—The fundamental problem presented by the property of radio-activity is the same for all the radio-elements, whether they exhibit it to a feeble degree, like uranium and thorium, or to an intense degree, like radium. In the latter case the magnitude of the effects render the nature of the problem more obvious, but the difficulty in the way of an explanation is no greater on that account. To cause substances to fluoresce, to ionise a gas, or even to fog a photographic plate requires energy. In the case of the production of cathode-rays and X-rays from a Crookes tube, the source of the energy is, of course, to be found in the electrical forces employed. But in the case of the radio-elements the source of the energy is not apparent. The emission of energy from radium, for example, is at once spontaneous and persistent. If in one hour sufficient heat is given out by this element to raise its own weight of water from the freezing to the boiling point, it is easy to see that the quantities of energy liberated in a year, or in a thousand years, must make an enormous total. In only two days radium gives out more energy, weight for weight, than the most powerful explosive known liberates during its explosion. If the energy liberated in a thousand years could be released instantaneously, a single milligramme of radium would equal in its effect *a ton* of any known explosive. There are only two general explanations of the source of this energy possible. *Either* radium must possess the power of responding to some hitherto unknown and unsuspected source of *external* energy in such a way as to convert the latter into forms which come within our powers of recognition, *or*, the energy must be derived from some hitherto untapped *internal* store bound up and latent in the structure of the atom. The arguments at the present time are overwhelmingly in favour of the latter view. Both views necessitate very substantial additions to our accepted ideas. But whereas the extension required by the latter view of the existence of forces within the atom—large compared with ordinary chemical or molecular forces—is in accordance with recent advances that have been made in our knowledge of atomic structure, the extension required by the first view of unknown forms of energy in external space is wholly new, and much more sweeping in its consequences. For, to explain the energy emitted in one place by radium, *all* space, it would seem, must, equally with the place where the phenomenon is manifested, contain a similar store of potential energy, and the total amount of existing energy in the universe postulated by the latter view is far greater than in the former case. Moreover, it only explains the source of the energy, and leaves unanswered how it is that it is manifested in those particular ways—the emission of rays, the production of emanations, &c.—which comprise all the complex manifestations of radio-activity. On this view also, an element once radio-active should continue so indefinitely without change of activity or loss of substance. On the alternative view the opposite holds true. The element must be undergoing change, and in the exhaustion of its large, but still not infinitely large, store of energy the element must itself change into other forms which possess a smaller energy-content. It is the main object of the present book to show that the radio-elements are undergoing such changes, and that these changes are new, in that they involve the disappearance of the atom of the element and its transformation into other atoms. In this way it will be shown that not only is the energy-emission explained, but also that all the complex and varied phenomena exhibited by the radio-elements are simple and necessary consequences of the one fundamental conception.

CHAPTER III.

THE ELECTRICAL PROPERTIES OF GASES.

The Ions of Gaseous Conduction.—Distinction between the Ions and the Radio-active Emanations.—The Saturation Current.—Equation of Current flowing through a Gas.—Ionic Velocities.—Coefficients of Diffusion.—Determination of the Charge carried by an Ion.—The "Atomic Charge."—C. T. R. Wilson's Condensation Experiments.—Determination of the Number of Molecules in a Cubic Centimetre of Hydrogen.—Radiant Ions.—Their power of Ionising Gases.—Striæ.—Determination of the Velocity and of the Ratio of the Charge to the Mass of the Radiant Ion.—Direct Determination of Velocity.—The Negative Ions produced by Metals under the action of Ultra-violet Light.—The Mass of the Negative Ion or Corpuscle.—Positive-Rays.—Electrical Inertia or Mass.—Variation of Electrical Mass with the Velocity of the Corpuscle up to the Speeds of Light.—The Electronic Constitution of Matter.

The measurement of radio-activity in an accurate and quantitative manner depends upon the property possessed by the rays emitted of ionising the gas through which they pass—*i.e.*, of rendering it capable of carrying limited quantities of both positive and negative electricity. This was shown for the uranium rays very early by Becquerel. The use of this property as a means of quantitative measurement dates, however, from the work of Rutherford (*Phil. Mag.*, 1899, V., 50, p. 109), who showed that the ionisation produced by the uranium radiation is of the same character as that produced by the X-rays, and equally with the latter can be simply and directly explained by the theory put forward by Prof. J. J. Thomson that the conduction is brought about by the transport of negatively and positively charged carriers to the positive and negative electrodes respectively. The function of the rays, both the X-rays and the rays from radio-active substances, is to *produce* these ions throughout the volume of the gas. The energy of the rays absorbed by the gas is utilised in the production of ions, so that the number produced in unit time is a measure of the intensity of the radiation absorbed. The current flowing through a gas under certain special conditions will be shown later to be a measure of the number of ions produced per second. Hence it becomes possible to measure radio-activity by electrical means. The actual methods employed will be dealt with in the next chapter. It is first necessary to obtain an idea of the nature of the ions and their behaviour under given conditions.

It will be convenient to consider first the ions of normal gaseous conduction which are produced in a gas whenever it is acted on by certain types of radiation of which the X-rays, the cathode or Lenard rays, and Becquerel rays are the most important, but which we now know are produced also by ultra-violet light of excessively short wave-length. Ordinary air screened from extraneous influences is in all probability an absolute non-conductor of electricity. Experimentally, it always exhibits an excessively minute conductivity, which is, however, caused by ions, and generally spoken of as the natural or, less correctly, the spontaneous ionisation of the air. But since this continuously decreases as greater precautions are taken to screen it from the action of infinitesimal quantities of radio-active matter, it is a fair inference to regard the air as a perfect insulator when in its normal condition. If it is got into a conducting state by the action of X-rays, for example, it is found that its conductivity persists for a short time after the rays are cut off, so that the conducting air can be blown away from the neighbourhood of the X-ray tube, and will discharge an electrified body, as, for example, an electroscope, at some distance out of the direct line of fire of the rays. Fig. 5 represents an arrangement for showing this. The air under the influence of the X-rays is drawn by a pump from B through wide tubes into the electroscope A and discharges it. If, however, the air is passed through a filter, like cotton or glass wool, or bubbled through liquids, or exposed in its passage to the action of an electric field, it ceases to conduct. Thus if (Fig. 5) an electric field is made to operate on the gas in its passage by connecting the outside of the metal tube C with one pole of a battery, the other pole being connected with a central wire insulated and stretched along the tube, the air passing into the electroscope no longer discharges it. Something is removed out of the air by these means which is different from the main bulk of the air, and this something consists of the charged ions which carry the electricity. Since the gas as a whole shows no charge, it follows that an equal number of positive and negative ions exist in the gas, for it has been shown that the charge carried by the negative ion is equal to that carried by the positive.

It is necessary here to call attention to the distinction between the ions and the radio-active emanations produced by radium and thorium. The latter also possess the power of being blown through tubes and discharging an electroscope at some distance from the radio-active substance. The emanations, unlike the ions, pass unchanged through filters and liquids which destroy the ions, and they survive the action of the electric field. The reason why the electroscope is discharged in the latter case is that the emanation is an *ionising* agency, and continually produces *fresh* ions out of the gas after its passage through the plug. The emanation passes through the latter like an ordinary gas, and immediately produces fresh ions out of the gas on the further side.

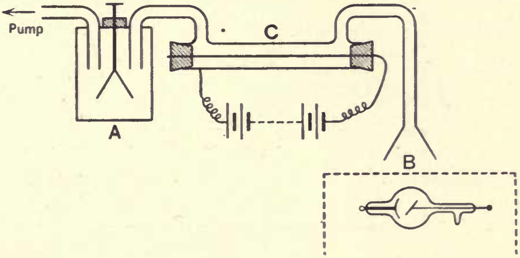


Fig. 5.

The most characteristic feature of the conductivity of an ionised gas, by which it is easily distinguished from all other types of electrical conduction, is the existence of a maximum or "saturation" current, which limits the quantity of electricity the gas is able to convey. In electrolytic and metallic conduction the current obeys Ohm's law, and increases proportionally to the voltage between the electrodes. This is true also of ionised gases under very small voltages. As the latter is increased, the current soon begins to increase less rapidly than the voltage, until, finally, a stage is reached at which a further very great increase of voltage scarcely increases the current that passes. This is the *saturation current*, and it serves experimentally as a very valuable criterion of a true ionisation current. Fig. 6 shows the variation of the ionisation current with voltage.

In the electrical measurements of radio-activity, it is always necessary to make sure either (1) that the current measured is the saturation current, or (2) less commonly, and with very active substances, that the current is proportional to the E.M.F., in order that the results should possess a definite meaning, and be comparable among themselves. The ions move to the electrodes through the gas under the influence of the electric field. They obey in this respect the ordinary law of a body moving through a viscous medium, and their velocity over the ordinary experimental range may be taken as being directly proportional to the force acting on them—*i.e.*, to the strength of the electric field. The number arriving at the electrodes, and, therefore, the strength of the current flowing, will depend (1) on the total number present in the gas and (2) on their velocity. With a definite rate of reproduction of ions by the ionising agency the current should, if there were no disturbing factor to be considered, be independent of the strength of the electric field—*i.e.*, of the voltage. The product

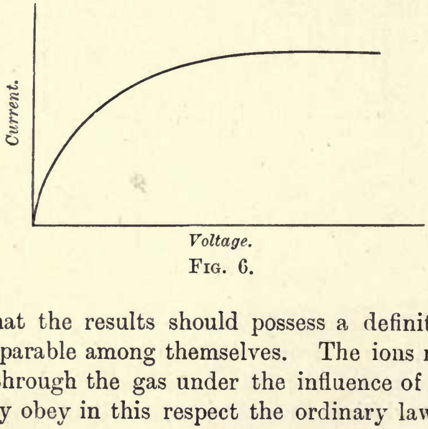


FIG. 6.

of the number present and their velocity would be constant, for as the latter decreases the number accumulating in the gas under the steady rate of reproduction will correspondingly increase. The disturbing factor is that oppositely charged ions attract each other and tend to recombine to form the neutral molecule. With high voltages acting this influence will be reduced to a minimum and the *saturation current* will be obtained. This represents the condition in which the ions in the gas are removed by passage to the electrodes, as fast as they are produced by the ionising agency, and before any appreciable proportion recombine. When this condition is substantially realised the current will be independent of further increase of voltage. Let q positive and q negative ions be produced per second, and let e be the charge carried by the individual ion. If i is the current passing through the gas, i represents the value of the charge received by each electrode per second. i/e, therefore, is the number of ions which give up their charge to either electrode per second, so that i/e positive and i/e negative ions are withdrawn from the gas per second. The number withdrawn by the saturation current represents the number produced. Therefore, $i/e=q$ and $i=qe$ when current is the maximum. If, as is usually assumed to be the case, the ions are produced uniformly throughout the volume of the gas, and q_0 represents the number of ions produced in each cubic centimetre per second, and if A is the area of the electrodes and l the distance between them, $q=q_0\mathrm{A}l$, and the saturation current through the gas $\mathrm{I}=q_0\mathrm{A}le$.

With currents less than the saturation current, some of the ions are destroyed in other ways than by passage to the electrodes. Some will re-combine, one positive and one negative ion reproducing the neutral molecule, while others will give up their charge by diffusion to the walls of the containing vessel and so cease to exist as such. The number that re-combine will be proportional to the number of collisions between ions of opposite sign and, therefore to n_1n_2, where n_1 and n_2 represent the number of positive and negative ions respectively present in 1 cubic cm. of gas. Since, in the cases generally considered, $n_1=n_2$, the number that re-combine will be proportional to n^2, and may be expressed by an^2, where a is a quantity independent of n. The number that give up their charge to the walls of the containing vessel is proportional to n and not to n^2, and may be expressed by κn, where κ is a coefficient depending on the size and shape of the vessel.

If i is made to represent the current flowing through unit area of the gas, i/le represents the number of ions of each kind withdrawn by the current for unit volume of gas. So that, when the gas is in a steady state, the number of ions withdrawn being equal to the number produced,

$$q_0 = an^2 + \kappa n + \frac{i}{le}.$$

If the number of ions present in the gas has not reached a steady state,

$$\frac{dn}{dt} = q_0 - an^2 - \kappa n - \frac{i}{le}.$$

This is the simplest general equation representing the current flowing through a gas ionised by Becquerel or X-rays. The meaning of the saturation current is, thus, the current that is able to flow through the gas when the ions are withdrawn as quickly as produced by the action of a strong E.M.F. before any appreciable number have time to be dissipated by recombination or by diffusion to the walls of the containing vessel.

With no current flowing, $q_0 = an^2 + \kappa n$. Except for narrow tubes, the number lost by diffusion to the walls is small compared to the number lost by recombination, and, consequently, it is often unnecessary to take the quantity κn into account. With regard to the quantity an^2, Langevin has recently shown ("Recherches sur les gaz ionisés," University of Paris, 1902) that recombination is brought about by the mutual attraction of the positive and negative ions, for if it were not for this attraction the rate of recombination would be much smaller than it actually is. For further details Prof. J. J. Thomson's "Conduction of Electricity through Gases," p. 545, should be consulted. The simple statement of the ionisation theory here given, and the account that follows, is taken from that work.

The rate at which the ions move in an electric field was first measured by Rutherford (*Phil. Mag.*, 1897, V., 46, p. 422), and was found to be comparatively slow. Zeleny subsequently showed that the negative ions move appreciably faster than the positive ions, and the ratio of the two velocities varies very much with different gases and with their state of dryness. Water vapour tends to equalise the velocities by diminishing that of the negative ion. The following table of ionic velocities, taken from Prof. Thomson's book, expresses the results obtained by Zeleny (*Phil. Trans.*, A. 195, p. 193, 1900). The ionisation was produced by X-rays:—

Ionic Velocities.

Gas.	Velocities in cm. per sec. under a potential gradient of 1 volt per cm.		Ratio of velocities of negative and positive ions.	Temperature. Degrees Centigrade.
	Positive ions.	Negative ions.		
Air dry........	1·36	1·87	1·375	13·5
Air moist......	1·37	1·51	1·10	14·0
Oxygen dry....	1·36	1·80	1·32	17·0
Oxygen moist..	1·29	1·52	1·18	16·0
CO_2 dry	0·76	0·81	1·07	17·5
CO_2 moist	0·82	0·75	0·915	17·0
Hydrogen dry..	6·70	7·95	1·19	20·0
Hydrogen moist	5·30	5·60	1·05	20·0

Townsend has also measured the coefficients of diffusion of the ions produced by various agencies, and his results are given in the following tables. The X-rays were the ionising agent used:—

Coefficients of Diffusion in Dry Gases.

Gas.	D for + ions.	D for − ions.	Mean value of D.	Ratio of D for + and − ions.
Air	0·028	0·043	0·0347	1·54
Oxygen	0·025	0·0396	0·0323	1·58
Carbon dioxide	0·023	0·026	0·0245	1·13
Hydrogen	0·123	0·190	0·156	1·54

Coefficients of Diffusion in Moist Gases.

Gas.	D for + ions.	D for − ions.	Mean value of D.	Ratio of D for + and − ions.
Air	0·032	0·035	0·0335	1·09
Oxygen	0·0288	0·0358	0·0323	1·24
Carbon dioxide	0·0245	0·0255	0·025	1·04
Hydrogen	0·128	0·142	0·135	1·11

These coefficients of diffusion are remarkably slow compared with the values obtained for the coefficients of diffusion of the uncharged gas molecules. For example, the value for the positive ion in air (0·028) is less than the value of a heavy vapour like ether or isobutylamine diffusing into carbon dioxide (0·055 and 0·03 respectively).

But the ions show the same coefficient of diffusion and the same velocity under unit electric force whatever the agency employed to produce them. The negative ion diffuses about half again as fast as the positive in dry gases like oxygen or hydrogen, but this difference is much diminished by moisture. The diffusion coefficient D is connected simply with the velocity u of the ions under unit electric force, by the equation

$$\frac{u}{D} = \frac{Ne}{\Pi},$$

where N is the number of molecules in 1 cubic cm. of a gas, e the charge on the ion and Π the pressure of the gas in dynes per square centimetre. It is thus possible to determine Ne, and, since N is known from the kinetic theory of gases within certain limits, to obtain an estimate of e, the charge carried by the individual ion. But the quantity NE, where E is the charge carried by the hydrogen ion in electrolysis, is accurately known, for 2NE (since one molecule of hydrogen contains two atoms) represents the number of electrostatic units of electricity which must pass through acidulated water to liberate 1 cubic cm. of hydrogen as gas. Now $NE = 1{\cdot}22 \times 10^{10}$, whereas the mean of the values of Ne obtained by the aid of the above equation is $1{\cdot}24 \times 10^{10}$. Hence it may be inferred that the charge carried by the positive or negative ion in gases is independent of the nature of the gas, of the ionising agency used to produce it, and is equal to the charge carried by the hydrogen atom in electrolysis. This charge will, in future, be referred to as the "atomic charge."

One of the most beautiful and striking developments of the ionisation theory arises out of a discovery by C. T. R. Wilson that, if dust-free air, free from ions and saturated with water vapour, is suddenly expanded, so as to chill it and cause supersaturation, no deposition of moisture occurs provided the expansion is not too great. If, however, positive or negative ions are present, these serve as nuclei for the condensation, and a cloud is produced, provided the expansion is above a certain limiting value. If the ratio of the volumes, after and before expansion, is below 1·25, no deposition of moisture occurs. For the ratio 1·25 up to 1·3 only the negative ions serve as nuclei, while for ratios above 1·3 both positive and negative ions are caught and carried down by the cloud. The drops are so small that the cloud takes a measurable time to settle down under the action of gravity, so that the rate of fall can be used to determine the size of the water drops. If the quantity of water present in the cloud is known, the number of drops can be determined, and, therefore, the number of ions which served as the nuclei for the formation of these drops can actually be counted. The total charge carried by the ions can be simply determined by electrical means, and thus a direct measure can be obtained of the charge carried by the ion. Prof. Thomson has accomplished this result for the ions produced by X-rays, by the radium rays, and for the negative ions which are emitted from metals under the influence of ultra-violet light, and he has proved that the value of e is the same for all ions, positive or negative, and is equal to $3{\cdot}4 \times 10^{-10}$ electrostatic units $((\text{gr.})^{\frac{1}{2}}(\text{cm.})^{\frac{3}{2}}(\text{sec.})^{-1})$. From $NE = 1{\cdot}22 \times 10^{10}$, and the identity of E and e, N can be directly calculated for the first time in an accurate manner. The value thus obtained for the number of molecules in 1 cubic cm. of hydrogen gas at normal temperature and pressure is $3{\cdot}9 \times 10^{19}$, a value which is in good agreement with the estimates based on the kinetic theory of gases. For fuller particulars and for the experimental methods employed to obtain these results the reader is again referred to Prof. Thomson's book. (Compare also Sir Oliver Lodge: "On Electrons," *The Electrician*, Vols. L. and LI.)

No direct measurements of the masses of the ions produced from gases have yet been accomplished. From considerations connected with the slow rate of diffusion and of movement in an electric field it seems probable that they consist of aggregates of several molecules of the gas, perhaps held together by the attraction of the field due to the charge carried by the ion. This cluster of molecules seems to be smaller for the negative ion than for the positive in dry gases, whereas when moisture or vapours like alcohol or ether are present they tend to condense on the negative ion more than on the positive, and so diminish its freedom of movement.

Prof. Thomson's ionisation theory of gaseous conduction possesses, however, another quite independent and more fundamental bearing upon radio-activity.

The terms "ion" and "ionisation" appear in so many different connections in science that they are apt to be somewhat confusing. We have, for example, the ionisation of gases, the ionic theory of electrolytic conduction, and the ionic dissociation theory of solution. The fundamental meaning of the word *ion* should be carefully kept in view. The term in all these cases is used strictly in the original sense of Faraday—to express a moving particle carrying an electric charge. Thus Prof. Thomson speaks in his book of the positive and negative ions expelled by radium and the other radio-elements, alluding to the α and *β-rays.* The α and β-rays consist of moving particles carrying an electric charge, and, strictly speaking therefore, are ions.

The α-rays are positive ions and the β-rays negative ions. There is, however, this distinction between the two classes.—The ions produced in gases by the passage of the X-rays and similar agents from radio-active substances are characterised solely by their charge. Their movement in an electric field is the result of the charge carried, and the charge is the only means we possess of detecting and investigating them. With the projected charged particles shot out by radium, the kinetic energy of the moving body and not its charge is its main experimental characteristic. In consequence, this class of ions has a great many properties not possessed by the ions of the first class, and it might be convenient to designate it by the name of "radiant ion." The means of investigation are quite distinct for the two classes. Yet it is possible to start with an ion of the first class and impart to it so much energy that it passes into the second class and resembles in properties the radiant ions of radium and uranium. The problems of radio-activity are thus intimately bound up with the electrical properties of gases.

We have seen that under ordinary circumstance the ions of gaseous conduction travel slowly in an electric field, as if hampered by a cluster of molecules attached to it. A remarkable change, however, comes over the property of the ion when the pressure of the gas is sufficiently reduced. Its movement then appears to be no longer hampered by the condensation of an aggregate of molecules around it, and it travels free. Under the action of a sufficiently powerful electric force it then acquires enormous velocity, and the increase in its kinetic energy above a certain critical value causes a change to come over its properties; that is, the ion passes from the first class, where its charge is its characteristic, to the second class, where its energy begins to dominate its experimental properties. It is true in the case just considered the energy is the result of the action of an electric force on the charge carried, but in the case of radio-active substances projected charged particles are shot out spontaneously which do not owe their velocity or energy to the action of any external force upon the charge they carry. The velocity of these particles is far higher than any that have been obtained by the action of the electric field.

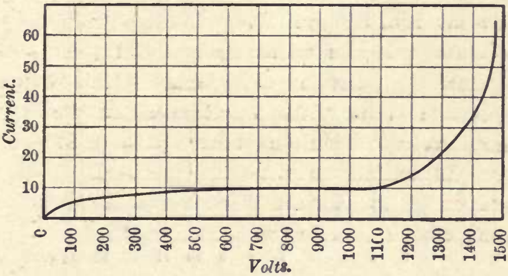
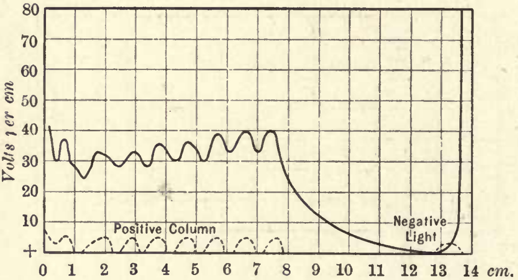


FIG. 7.

The most characteristic property of the radiant ions above their critical velocity is their power to ionise the gas through which they pass. This has been particularly studied by Townsend (*Phil. Mag.*, 1901, VI., 1, pp. 198 and 630). If the voltage is increased far beyond that required to produce the saturation current in a gas at low pressure, it is found that above a certain point the current again commences to increase, and does so with great rapidity as the voltage increases, until finally the point is reached at which a spark or luminous discharge passes through the gas. This is shown in Fig. 7, which would represent the relation between current and voltage for plates 10cm. apart at about 3mm. pressure. A potential gradient of 40 volts per centimetre distance between the electrodes is sufficient for the ions to acquire sufficient velocity at a pressure of 1mm to start ionising the gas. It can be shown that at first it is only the negative ion which acquires this power, and the number of ions produced, although largely increased, still remains finite. For the negative ions all travel in the one direction, and of the ions they produce by collision with the gas molecules, it is only the negative ions which acquire sufficient velocity to ionise fresh molecules. Hence, as soon as these ions reach the electrode, the action is at an end. But when the positive ion, which at these low pressures is far more massive than the negative ion, reaches the critical velocity and begins to produce fresh ions, the ions travelling in *both* directions produce ions in their path. This action must go on cumulatively, for it no longer ceases when the ions reach the electrode, as the ions of opposite sign are always travelling anew back to the other electrode and ionising the gas in their path. A little consideration will show that the number of ions increases rapidly to an almost infinite extent, provided the electrical force driving the ions is maintained unaltered in value. At this point a spark passes through the gas. Under some circumstances, however, the sudden increase of the number of the ions and their electrical convection tends to so diminish the force of the effective electric field that this is lowered below the point at which ions are produced by collision. It is probable that the striated discharge in vacuum tubes is produced by this action, as the distribution of electric force through the tube shows a rapid alternation in intensity corresponding with the position of the striæ. This is shown



Hydrogen Pressure 2·25 mm. Current 0·586 m.a.

FIG. 8.

in Fig. 8, which represents the electric force per centimetre on the vertical axis for different parts of a tube showing striæ plotted on the horizontal axis (compare H. A. Wilson, *Phil. Mag.*, 1900, V., 49, p. 505). The discharge thus consists of a succession of periods in which the electric force alternately rises above and sinks below the critical value necessary for the ions present to produce other ions out of the gas. The well-known experiment of Hittorf, who showed that a discharge refused to pass between electrodes 1mm. apart in a gas at low pressure, but preferred a circuitous route 375cm. long (Fig. 9), is to be explained by similar considerations. In the space of 1mm. the ions moving under the electric field cannot acquire the critical velocity required to ionise the gas.

An independent way of regarding the phenomena of discharge at low pressure is arrived at from the consideration that the viscosity of the gas is much diminished. In consequence, the ions do not obey the law followed at atmospheric pressure that their velocity is proportional to the force acting. The force *accelerates* the unhampered ion, and its velocity is, therefore, greater, the greater the distance traversed under the action of a constant force

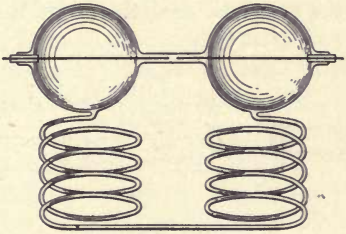


FIG. 9.

The most instructive property of a charged particle moving with great velocity is its power of being deviated out of a straight line by the action of electric and of magnetic forces, since, by a study of these deviations, we can arrive at the determination of the velocity of the particle and of the ratio e/m of the charge of the particle to its mass. These considerations are of great importance in radio-activity, for by aid of them knowledge has been obtained of the mass and velocity of the projected particles from radio-active substances.

If the charged particle is projected with uniform velocity at right angles to the lines of magnetic force, no electrical forces acting, its path is changed to that of a circle whose radius of curvature, ρ, is given by

$$\rho=\frac{mv}{He},$$

where m is the mass of the ion, e its charge, v its velocity, and H the magnetic force. This enables us to determine the value of e/mv. If an electrostatic force is made to act simultaneously on the moving ion, so that the lines of electric force are at right angles to the lines of magnetic force, and at right angles also to the direction of motion of the ion, the electric force can be made either to neutralise or assist the deviation produced by the magnetic force according as the sign of the P.D. is in the one direction or the other. If it is adjusted so as to oppose the magnetic deviation, a point can be reached, by the suitable variation of one of the forces, the other remaining constant, at which the projected particle is not deviated. At this point it can be shown that the force Fe, where F is the electric force and e the charge on the ion, is balanced by the force Hev. Fe = Hev. Therefore, $v = F/H$, and the velocity of the ion can be determined. When v is known the ratio e/m can also be determined from the ratio e/mv.

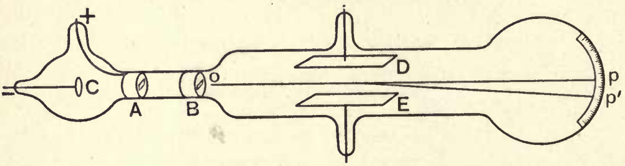


FIG. 10.

In this way Prof. Thomson, in 1897, succeeded in determining the value of e/m and of v for the negatively charged particle which constitutes the cathode ray of the Crookes tube. The apparatus used is shown in Fig. 10. Cathode rays starting from C are projected through the diaphragms AB, which consist of fine horizontal slits in metal discs; they then pass between the two horizontal plates ED, maintained at a constant P.D., and fall upon the phosphorescent screen at the end of the tube. The vacuum in the tube must be of the highest, or otherwise the ionised gas, being a conductor, screens the rays from the action of the electrostatic field E D. The value e/mv was determined by measuring the deviation of the spot of light on the screen pp' under the action of a known magnetic field. An electric field of known intensity is then applied at E D, so as to bring back the spot of light to its original position, and the value of v and of e/m thus found. Prof. Thomson obtained $v = 2{\cdot}8 \times 10^9$ cm. per second and $e/m = 7{\cdot}7 \times 10^6$ as the mean result of his determinations. The velocity of light is about 3×10^{10} cm. per second, so that the cathode rays consist of streams of charged particles travelling with one-tenth of the velocity of light. From the direction of the deviation it follows that the particles must be negatively charged. The value e/m for the hydrogen ion in electrolysis is only 10^4, so that the value for the cathode ray particle is 770 times greater than for the hydrogen ion. This value was found to be independent of the nature of the gas in the tube, although a wide range of gases, represented by the extremes of hydrogen and methyl iodide, was experimented on, and it has been proved to be the same whatever the nature of the metal of which the cathode is made. Lenard investigated the values of v and e/m for the cathode rays from an exhausted tube that escaped through a window of very thin aluminium foil, and found that while e/m was in good agreement with Thomson's value the value of v was much higher—viz., 7×10^9, indicating that the aluminium foil had sorted out the more rapidly moving particles, but that the value e/m was independent of the velocity.

The theoretical reasoning on which these determinations are based has been confirmed in the most brilliant manner by the direct determination of v by Wiechert (*Wied. Ann.* 1899, p. 739). The principle of the method was to send a Leyden-jar discharge through two circuits at different positions along the tube through which the cathode rays passed. The Leyden-jar discharge consists of very rapidly alternating currents whose frequency can be accurately determined. If the distance between the two circuits is so arranged that the cathode ray takes the time of a complete oscillation to traverse it, the deflection produced by the first circuit will be augmented by the second circuit. If, however, the distance is so arranged that by the time the cathode ray leaves the first circuit to the time it reaches the second circuit only half a period of oscillation has elapsed, then the deflections produced will neutralise one another, and the path of the ray will appear straight. The value of v found by Wiechert in this way was, for one experiment, 5×10^9. From the magnetic deviation of the same rays the value mv/e was determined, and combining this with the direct experimental value of v the value $e/m =$ from $1{\cdot}5$ to $1{\cdot}0 \times 10^7$ was obtained. Considering the difficulty of the experiments, this value is in good agreement with the others, and supports the general argument in the strongest possible manner.

The connection between the negative ion produced by X-rays, &c., and the negative radiant ion which constitutes the cathode particle is shown by the following experiments:—If ordinary ultra-violet light (not of the extreme wave-lengths before mentioned, which are able to ionise the air like X-rays and to produce both + and – ions) is allowed to fall on a polished surface of zinc, the latter expels negative ions only into the gas. If the zinc is negatively charged it rapidly loses its charge in ultra-violet light, but remains charged indefinitely when its electrification is positive. If the experiment is repeated in air at low pressure, the negative ions produced by the zinc acquire in an electric field the characteristics of the cathode-ray particle, and are shot out normally to its surface. The deviation produced by magnetic and electric forces can be measured as in the case of the cathode rays. The values of v and of e/m for these ions have been determined by Prof. Thomson (*Phil. Mag.*, 1899, p. 547) and by Lenard (*Drudes Annalen*, 1900, p. 359). The former found for e/m the value $7{\cdot}3 \times 10^6$, which is in very good agreement with the value for the cathode-ray particle—viz., $7{\cdot}7 \times 10^6$. These negative ions have been very carefully studied. Produced in a gas at atmospheric pressure, they have been shown to be identical with the negative ions produced by other agencies (as, for example, the X-rays) in their coefficient of diffusion and their velocity under unit potential gradient and, therefore, in the value of e, the charge carried.

Moreover, a direct determination of the value of e has been accomplished by Prof. Thomson by the condensation method, and found to be the same as that carried by the hydrogen ion in electrolysis. Yet this same ion, when the air is removed and it travels free from entanglement, then resembles the cathode ray in all its properties, and is found to possess a value of e/m 700 times greater than that possessed by the hydrogen ion in electrolysis. If the charge on the ion is assumed to be invariably the "atomic charge" possessed in gases at ordinary pressure, the conclusion is reached that the negative ion, when it travels free, possesses a mass $\frac{1}{700}$th of the mass of the hydrogen atom. Since the value of e/m agrees for all negative ions for which it has been determined, it follows that this ion possesses an invariable mass about $\frac{1}{700}$th of that possessed by the lightest atom known, and that negative electrification in high vacua, when but little ordinary matter is present, consists of an assemblage of discrete charged "corpuscles" of far smaller mass than anything before observed.

The presence of positive ions in the electric discharge in vacuo is manifested in a type of radiation discovered by Goldstein, called by him "Canalstrahlen" (Canal Rays), but which may be termed the positive rays. They are deflected by electric and magnetic fields in the opposite sense to the deflection produced in the path of the cathode rays, but only with very much greater difficulty. Wein showed the value of e/m for these rays to be 3×10^2, while $v = 3{\cdot}6 \times 10^7$cm. per second. The subject has since been investigated by J. J. Thomson for the positive ions emitted from heated wires in vacuo, and he found a similar value, $e/m = 4 \times 10^2$. Thus this value is only about $\frac{1}{30000}$th part of that shown by the negative ion, and is of the same order as in the case of electrolysis for heavy ions like those of oxygen or the metals. Since the value e/m of the positive ion has never been observed to be greater than that observed for the hydrogen ion in electrolysis, it follows that the positive ion, unlike the negative, is never dissociated from the atom which carries it. On this view Prof. Thomson has developed the "corpuscular theory of electricity" which supposes that the negative charges are discrete particles or *corpuscles* (called *electrons* by other investigators), and that the absence of one of these corpuscles from the atom of matter produces the positive ion.

It has been shown mathematically by Thomson and Heaviside that an electric charge concentrated on a sphere of sufficiently minute radius would possess inertia by virtue of the electromagnetic field of force it creates in the surrounding ether. That is to say, it will tend to resist change of motion, and in general will behave as if possessing a certain mass. Electricity will, therefore, under these conditions, resemble matter in its most fundamental property. One of the consequences deduced from this view is that the speed of the ion cannot be increased beyond a certain limited value, represented by the velocity of light, because at this value the inertia, and therefore the mass, of the particle would be infinite. For values of v below one-half the velocity of light the inertia due to the moving charge, or as it is convenient to call it, the *electrical mass*, is approximately constant, but above that speed the mass increases rapidly. Sir Oliver Lodge (*Nature*, June 11, 1903, p. 129) has given some calculations of the ratio of the electrical mass m_0 for slow motions to the mass m when the velocity of light is approached. At half the speed of light $m = 1{\cdot}12m_0$; at three-quarters $m = 1{\cdot}37m_0$; at nine-tenths $m = 1{\cdot}8m_0$; when the speed is 99 per cent. of that of light $m = 3{\cdot}28m_0$, for 99·9 per cent. $m = 5m_0$, whereas between this last value and that of light the mass increases to infinity. It is to be remarked how nearly the velocity of light must be approached before the action becomes pronounced. One of the most remarkable facts of radio-activity has been the discovery that radium projects negative corpuscles, or β-rays, with speeds varying within the limits between which an increase of mass is to be theoretically expected, *if this mass is electrical in origin.* This increase has been looked for and found by Kaufmann and his work will be considered when the rays from radium are studied.

It will be seen that the tendency of these developments has been to replace the attributes of matter by those of electricity, and to look upon the atom as a very complex system made up of much smaller units of electricity, or corpuscles. If one excludes the minute and unexplained effect of gravitation, which is only so prominent to us on account of the nearness of the overwhelming mass of the earth, the main attribute of matter is inertia. If the corpuscles possess inertia equal to one-thousandth of the mass of the hydrogen atom, it follows that an assembleage of a thousand corpuscles might represent in inertia and other properties the hydrogen atom as we know it. This point of view would seem to account for the behaviour of matter towards X-rays, cathode rays, and the rays from radio-active substances. For if the corpuscle rather than the chemical atom were incidental in causing the absorption, the latter would depend on the number of the corpuscles in the path of the ray, and would be independent of the nature of the complexes—chemical atoms or molecules—into which these corpuscles were grouped. This view of the constitution of matter is, of course, far from established, and mainly doubtful points require further elucidation. These cannot be here considered, but the nature of positive as opposed to negative electrification may be mentioned as one of the most fundamental. The above is to be regarded as merely a superficial summary of some of the more important physical aspects of the work on the conduction of electricity through gases, and the nature of electrons or corpuscles. The mathematical analysis upon which much of the reasoning depends is beyond the scope of the present work, and the original authorities must be consulted.

It may at once be pointed out that the theory of atomic disintegration, to which, in the succeeding chapters, the study of radio-activity will lead, is independent of the electrical or electronic view of atomic constitution. It postulates no view of atomic structure beyond the original conception of Dalton; greatly limits the field of speculation on the one hand, and, on the other, it raises new problems of its own which any satisfactory theory will have to account for. The term "disintegration" is indeed little more than a convenient and short means of expressing certain experimental facts. It is not until we enquire as to the ultimate cause of radio-activity, and seek a knowledge of the forces at work which bring about the observed disintegration, that we enter a region to which the term *hypothesis* in the ordinary sense of a probable explanation would apply.

CHAPTER IV.

METHODS OF MEASURING RADIO-ACTIVITY.

Electrical, Photographic and Fluorescence Methods of Measurement.—Different Fluorescers behave Differently to the Three Types of Rays.—Electrical Method of Measurement.—Apparatus of Rutherford and of Curie.—The Use of the Gold-leaf Electroscope.—Distinction between Ionisation Currents and Leaks due to Defective Insulation.

The means of investigating the rays from radio-active substances are similar to those employed for the X-rays, and may be divided into three classes: the electrical, photographic and fluorescence methods respectively. The first only, the electrical method, is suitable for accurate quantitative investigations of radio-activity, but the photographic and fluorescence methods have frequently been employed for rough comparisons and for experiments in which accurate measurements of the intensity of the rays are not required. Thus much of the work on the magnetic and electric deviation of the β-rays by Becquerel has been done by causing the latter to trace their paths on a photographic plate. It must be pointed out that the relative strengths of the different types of rays are often very different with the three methods. Thus the electrical method depends on the absorption of the rays by the air, the energy of the rays going to produce ionisation, and with this method the easily absorbed or α-rays give by far the greater effect. These same rays, on the other hand, give relatively very little effect on the photographic plate. Thus the α rays of uranium do not perceptibly affect a photographic plate, although they contribute over 99 per cent. of the total electrical effect when examined under ordinary circumstances (Soddy, *Jour.* Chem. Soc., 1902, 81, p. 860). This is probably due to the fact that the rays are unable to penetrate the gelatin film sufficiently to affect appreciably the sensitive silver salt.

Many fluorescent substances behave differently under the action of the α and β-rays. Thus, zinc sulphide (Sidot's hexagonal blende) is extremely sensitive to the action of the α-rays and much less to the action of the β-rays, while with barium platinocyanide and willemite (zinc silicate) the contrary holds true. The diamond glows by the action of the α-rays.

Other fluorescers are kunzite, a transparent gem-like variety of spodumene (lithium aluminium silicate) which fluoresces a rose yellow; sparteite, a form of calcite containing manganese, which shows a remarkable deep orange-red fluorescence; and scheelite, a native calcium tungstate, which gives a fine deep blue colour. The first two hardly seem to respond to the α-rays of radium, being excited solely by the β-rays. The platinocyanides furnish at once the most beautiful and intense fluorescent effects with the β and γ-rays of radium. Being transparent, the whole body of the matter adds to the light produced by a penetrating type of radiation. But it is rather a curious fact that magnesium platinocyanide, which is one of the best fluorescers under the X-rays, does not respond to the radium rays. Of the other platinocyanides, barium and calcium fluoresce green, sodium lemon-yellow and lithium red, the same as for the X-rays. If the salts are in the form of large crystals most beautiful effects can be obtained, even with only a few milligrammes of the pure radium compounds.

The electrical methods alone demand a more detailed consideration. It has been shown in previous articles that, to obtain definite results, the conditions must be such that the "saturation current" is measured, as this current is an expression of the number of ions produced per second, and the latter is proportional to the intensity of the radiations absorbed by the air. Rutherford, in the research in which he showed that the ionisation from uranium was of the same character as that produced by X-rays (*Phil. Mag.*, 1899, V., 47, p. 109), showed also that the ionisation was proportional to the pressure of the gas, while the absorption of the rays was also proportional to the pressure. For different gases, although for thin layers the ionisation increases as the density of the gas, for thicker layers the total ionisation when all the rays are absorbed reaches a maximum, and then is approximately the same for all gases. A very careful series of measurements by Strutt (*Phil. Trans.*, A. 196, p. 507, 1901) has led to the conclusion that, with the exception of hydrogen, all gases are ionised by the various rays from radio-active substances to an extent sensibly proportional to the density and independent of the nature of the gas. With hydrogen the ionisation is greater than it should be if proportional to the density, and this divergence is greater for the α than for the β-rays. We may conclude, as a first approximation, that the energy required to produce an ion is the same for the ions of all gases except hydrogen.

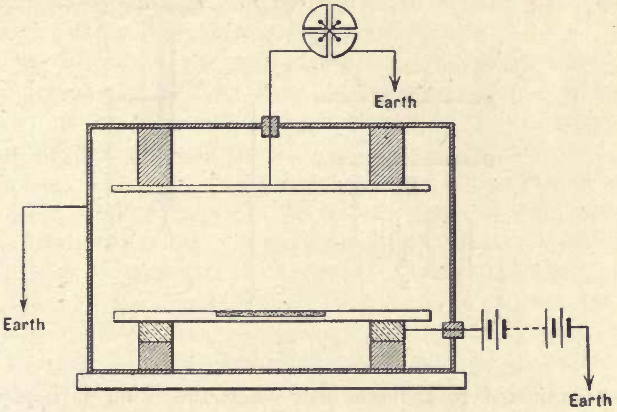


FIG. 11.

Fig. 11 represents an apparatus constantly employed by Rutherford for the electrical measurement of radio-activity. The two plates are each of about 36 sq. cm. area, and a known weight of the radio-active substance in fine powder is sifted uniformly over the lower plate, which is insulated and connected with one pole of a battery of 300 volts, the other pole being earthed. The upper plate is about 5cm. from the lower, and is connected with one pair of quadrants of a Kelvin electrometer of the White pattern, the other pair being connected to earth. This upper plate is most carefully insulated, and the whole apparatus is placed in a metal box, provided with a door at the side through which the lower plate can be inserted or removed. The saturation current through the air, between the upper and lower plates, furnishes a measure of the radio-activity. To use the apparatus, both pairs of quadrants are at first connected to earth; the earth connection of the pair connected to the upper plate is then broken, and the *rate* of deflection of the electrometer needle from zero is measured by means of a stop-watch by noting the time taken for it to pass over a given number of divisions of the scale. The deflection of the electrometer needle is a measure of the difference of potential between the two pairs of quadrants, and, since the capacity of the system is constant, of the charge communicated to the one pair when the other is kept connected to earth. The rate at which this charge is communicated is a measure of the current through the gas. 0·5 gramme of uranium or thorium oxide in this apparatus produces a current of approximately 10^{-11} amperes. The measurements are usually comparative, the radio-activity of some standard, such as a known weight of uranium oxide spread over a given area, being compared with the activity of the substance to be measured. In this way the sensitiveness of the electrometer, which varies from day to day according to the extent to which the needle is charged, is eliminated. M. and Mme. Curie use a similar apparatus, which is represented in Fig. 12, in which the charge communicated to the one pair of quadrants by the current through the air under the action of the

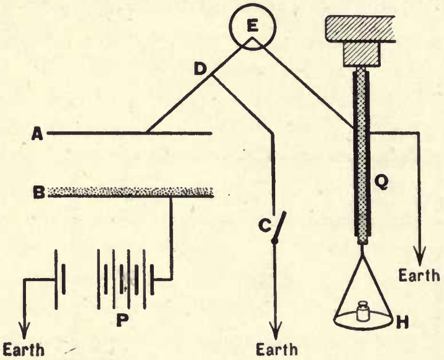


Fig. 12.

radiations being measured, is balanced by the communication of a similar charge of opposite sign, so that the electrometer needle shows no deflection. This is effected by means of a quartz electric balance which consists of a quartz lamina, which can be subjected to a known tension by means of weights, the two sides of the lamina receiving opposite charges of electricity, the amount of which is known for any known weight. The application of the weight is so regulated by the hand that at each instant the charges of opposite sign communicated to the electrometer are equal, and the time from the commencement to the completion of the application of the tension furnishes a measure in absolute units of the intensity of the current flowing through the air under the action of the radio-active substance. In this way the operation is performed by a null method, and is independent of the sensitiveness of the electrometer.

For delicate work an electrometer designed by Dolezalek and constructed by Herr Bartels of Göttingen has been much used. It is very small, and the needle is of silvered paper suspended by a quartz thread. If the latter is made very fine the sensitiveness of the instrument is very high, and may be as much as 20,000 divisions of the scale for one volt P.D. between the quadrants. Great precautions against external disturbances due to accidental electrification of the surroundings must be taken in working with these very sensitive instruments.

The simplest means of measuring radio-activity electrically is by means of a gold-leaf electroscope, and this instrument is very useful in certain cases, although its application is somewhat limited. The results obtained with it need very careful scrutiny if accidental errors are to be avoided. It can, however, be arranged to detect far feebler ionisation than the most sensitive electrometer, with the exception, perhaps, of the Dolezalek, and has proved invaluable in many important researches. Fig. 13 represents an electroscope suitable for the testing of radio-active substances. It is modelled after one first employed by C. T. R. Wilson. It consists of a tin can with a removable lid, E, at the bottom for the insertion of the substance to be tested. A paraffined rubber cork, H, is pierced in the centre by the metal wire G, which carries at its end a rod of fused quartz, A. A thin brass strip, B, to which a single gold-leaf, C, is attached, is fastened to the lower end of the quartz rod. The charge is communicated through the rod D, which can be turned by the ebonite handle F so as to make contact with B. When the electroscope is charged this connection is broken, and the rods C and G and the outside of the metal vessel are connected to earth. The electroscope then represents a perfectly isolated charged system, completely surrounded by metal and free from all external disturbances. Unless the electroscope is required to be made airtight, it is convenient to insert a short paraffined glass tube into the cork to serve as a bearing in which the charging-rod turns. If the latter is pierced directly through the cork the rod is apt to stick annoyingly to the rubber. The rate of collapse of the gold-leaf is observed by means of a reading microscope provided with a micrometer scale, through windows in the case of the instrument. For the measurement of radioactivity the substances to be compared can either be placed inside the electroscope by spreading known weights over suitable metal discs of similar area, or, in the case of more active preparations, can be placed at a fixed distance outside the electroscope so that the rays have to penetrate the metal walls. If the deflections are only taken between certain limits, it is found that they are very approximately proportional to the potential of the leaves. If the dimensions of the instrument are not too large and the radio-activity is not too great, the

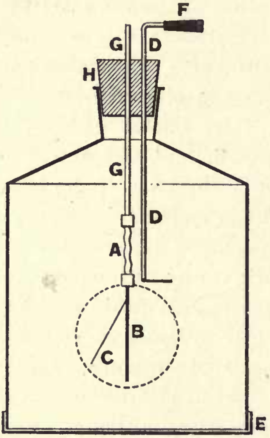


FIG. 13.

saturation current through the gas is attained and the rate of collapse is uniform between these limits. But all of these considerations must be continually borne in mind in working with an electroscope: The potential required to make the leaf diverge to a convenient extent is usually about 200 to 400 volts. For accurate work the uniform rate of collapse of the leaf over the part of the scale employed must be frequently verified with some standard quantity of uranium oxide. A far more serious source of error is the possibility of the insulation of the leaves deteriorating during a series of measurements, and so simulating the effect being looked for. For this reason the natural leak of the instrument when no active substance is present must be frequently redetermined. In working with an electrometer an ionisation leak is at once distinguished from a conduction leak, due to defective insulation, by increasing the voltage. The former, being the saturation current, is not appreciably increased, whereas the latter is proportional to the voltage.

The α-rays are so easily absorbed that the radiation does not much increase beyond a certain thickness of the layer of the active substance. To make the measurements of different substances as comparative as possible, the smallest possible weight spread over a large area should be employed. But even then the activities of different compounds of different densities and states of division can only be approximately compared. On the other hand, the *change* of the activity of any given preparation with time can be very accurately studied by leaving it undisturbed on its original plate throughout the course of the measurements.

For the measurement of more active preparations than uranium and thorium the sensitiveness of the measuring instruments must be decreased by connecting them in parallel with suitable capacities. But it must be remembered that the voltage required to produce the saturation current increases as the current increases. For work with preparations of uranium a greater voltage than 300 is seldom required, and this can be obtained by means of a battery of Planté cells charged in parallel and discharged in series. But for work with radium much higher voltages are necessary to produce the saturation current, unless the intensity of the rays is cut down by absorption in suitable screens.

CHAPTER V.

THE α, β AND γ-RAYS.

General Consideration of the α, β and γ-Rays.—γ-Rays.—Relation between γ and β-Rays.—Explanation of High Penetrating Power of γ-Ray on the view that it is an X-Ray Pulse.—β-Rays—Photographic Action—Magnetic and Electrostatic Deviation—Value of e/m and v.—Charge carried by the β-Ray.—Self-electrification of Radium.—Kaufmann's Work on β-Rays Approaching the Velocity of Light.—α-Rays.—Curves of Penetrating Powers of Various Types of α-Rays.—Diminution of Penetrating Power with Distance Traversed.—Magnetic and Electrostatic Deviation.—Value of e/m and v.—Charge carried by α-Rays.—The Spinthariscope.—Confirmation of the Corpuscular Theory of Electricity.

It has been stated that the rays from the radio-active substances, with the exception of polonium, are complex, and three types, the α, β and γ-rays, have been distinguished. The earliest work on this subject was done by Rutherford (*Phil. Mag*, 1899, V., 47, p. 109), who showed the complexity of the rays from uranium by measuring the ionisation from a bare layer of the compound, and comparing it with that produced when the layer is covered with successive layers of metal foil of uniform thickness. The apparatus employed was that described in the last chapter (Fig. 11). Fig. 14 represents the curve obtained by plotting the intensity of the rays transmitted on the vertical, and the number of layers of aluminium foil of thickness 0·00012cm. on the horizontal axis. It will be seen that the radiation is quickly diminished to a few per cent. of the original value and after 15 layers of foil have been traversed remains constant at a minimum value. This shows that the radiation is complex and consists of an easily-absorbed kind, which was called the α radiation, and a much more penetrating kind, which was called the β radiation. The easily-absorbed or α-rays contribute over 95 per cent. of the total ionisation, and are completely stopped by 15 to 20 layers of the foil. The more penetrating or β radiation is able to pass through 50 layers with hardly any absorption. The latter was found to be rather more than 100 times more penetrating in character than the former, the α-rays being reduced to half value by passage through aluminium foil 0·0003cm. thick, the β-rays by passage through 0·05cm. of aluminium. In the case of the thorium radiation the α-rays are able to pass through ·0005cm. of aluminium before being reduced to half-value. The β-rays from thorium constitute a far smaller proportion of the total radiation than in the case of uranium. The ratio of the β to the α-rays for the latter is four times greater than for thorium (compare Rutherford and Grier, *Phil Mag.*, 1902, VI., 4, p. 326). The existence of these two types of rays has since been observed for radium, and, in addition, a more penetrating type than either, known as the γ-rays was first noticed by Villard for

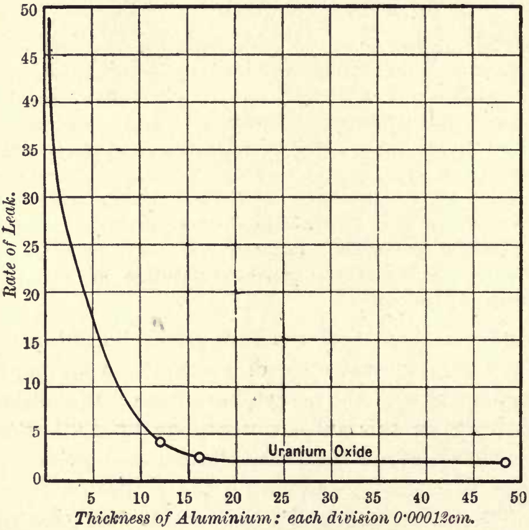


Fig. 14.

this element (*Comptes Rendus*, 1900, 130, p. 1,010). γ-rays have also been detected by Rutherford in the radiations of thorium and uranium, but large quantities of these substances must be used and the finest and most sensitive electrical methods employed in order to detect them. The γ-rays are, roughly speaking, about 100 times more penetrating than the β-rays, being cut down to half value by 6cm. or 7cm. of glass or aluminium.

In addition to their varying powers of penetrating matter, the three types of rays are characterised by different behaviour under the influence of the magnetic and of the electrostatic field. The β-rays are easily deflected into circular paths, and in this respect, and in the direction of the deflection, they resemble the cathode rays. The α-rays, at first thought to be undeflected, have now been shown by Rutherford (*Phil. Mag.*, VI., 5, p. 177, 1903) to be slightly deviated by intense magnetic and electrostatic fields, and the deviation is in the opposite sense to that of the β or cathode ray. The γ-rays remain quite undeflected. These facts have been conveniently represented by Mme. Curie by a diagram (Fig. 15) in which the rays are represented to be issuing from radium placed in a deep cavity in a block of lead under the action of an intense magnetic field at right angles to the direction of the beam and the plane of the paper. The thin pencil of undeflected rays represents the relatively feeble γ-rays. The intense pencil slightly deviated the α-rays, and the circular paths of

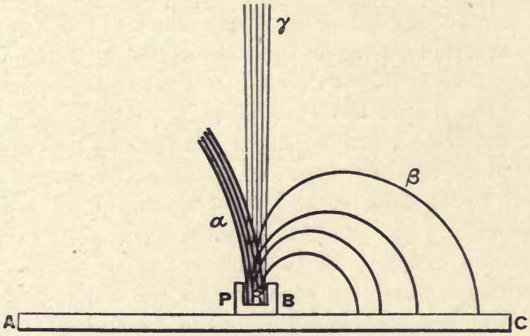


FIG. 15.

varying radii the β-rays, deflected in the opposite direction to the α. It must be noted that the scale of the deviation of the α-rays, compared with that of the β, is enormously exaggerated in the diagram for the sake of clearness.

γ-Rays.—It will be convenient to consider the three types of rays in detail in the inverse order of their importance, and to commence with the least known and, except in the case of powerful radium compounds, least obvious type—the γ-rays. They are characterised by extraordinarily great penetrating power, being able to pass through 7cm. of lead, 19cm. of iron and 150cm. of water before they are reduced in intensity to 1 per cent. of the original (Rutherford, *Nature*, 1902, 66, p. 318). No deviation of these rays has been detected in the most powerful magnetic fields it is possible to obtain, and in this respect they are more nearly allied to the X-rays than to the α and β-rays; but in their absorption by different kinds of matter they exhibit an almost complete parallelism to the β-rays. The simple law enunciated in the first chapter—and which for shortness will be called the Density Law—that the absorption of the new types of radiation by matter is proportional to its density, and independent of its nature, is followed strictly by none of the known types. The exceptions are, however, different for the different types. Thus, for all gases, with the exception of hydrogen, the absorption of the rays in the gas, as measured by the amount of ionisation produced, is in good agreement with the density law for all three types of rays. In the case of hydrogen, the ionisation is about twice as great for the β and γ-rays and about three times as great for the α rays as the density would lead us to expect. But for ordinary X-rays the ionisation in the case of heavy gases and vapours—as, for example, chloroform, methyl iodide and carbon tetrachloride—is from 7 to 15 times greater than the density law requires (compare Strutt, *Proc.* Roy. Soc., 1903, 72, p. 208).

At first sight this seems to point to the γ-rays being more nearly allied to the β-rays than to the X-rays. But it must be remembered that the β-rays themselves differ from the ordinary cathode rays of the Crookes tube in possessing a far higher velocity, and it is to be expected that the X-ray from radium produced from the β-ray at the moment of its sudden expulsion will also differ from the X-rays of the Crookes tube. It has recently been found (A. S. Eve, *Nature*, March 10, 1904) that if the absorption of only the most penetrating X-rays produced from a "hard" tube in dense gases is measured, the deviations from the density law noticed for the easily-absorbed X rays to a large extent disappear. That is, very highly penetrating X rays are similar to the γ-rays of radium in their absorption by dense gases. Rutherford has pointed out (*loc. cit.*) that it is to be expected that a narrow electromagnetic pulse—*i.e.*, a penetrating type of X-ray—will be generated at the sudden expulsion, with extreme velocity, of the β-corpuscle from radium. On the other hand, on account of the great penetrating power of the β-corpuscle, it is impossible to stop it suddenly, even by the densest matter, and the X-rays produced by it on impact with matter will be broad pulses of non-penetrating character, which experimentally would be difficult to detect. There is thus very strong evidence for supposing that the γ-rays of radium are to the β-rays as the X-rays of a Crookes tube are to the cathode rays, with the difference that the γ-rays accompany the expulsion of the β-ray, whereas the X-rays accompany the stoppage of the cathode rays. This point of view is borne out by the fact observed by Rutherford that the γ-rays of radium are, under all circumstances, proportional to the β-rays. There is always a complete parallelism in the intensity of the β and γ-rays; and throughout the various processes of decay and recovery, which will be considered later, the ratio between these types remains unchanged.

It may be pointed out here that these new facts strongly confirm the view that will be developed later as to the nature of radio-activity. X-rays should result from cathode rays whenever the corpuscles constituting the latter are accelerated either positively or negatively, and are the more penetrating the greater the acceleration. Sir George Stokes pointed out that X-rays should accompany the production of the cathode rays as well as their stoppage. But in the Crookes tube the cathode ray corpuscle acquires its velocity gradually as it moves under the influence of the electric field. Its acceleration is, therefore, relatively small, and the pulse which results has feeble penetrating power and is incapable of detection. When the corpuscle strikes the anti-cathode the resulting acceleration is very great, and a pulse of high penetrating power is emitted, giving rise to the penetrating X-rays which escape from the tube. In the radio-active substances the converse holds true. The β-corpuscle is suddenly expelled as the result of the explosion of the atom, and the value of the acceleration is probably far higher than that of the cathode ray corpuscles at the anti-cathode. In consequence, the γ-rays are far more penetrating than the X-rays. But the acceleration suffered by the β corpuscles when they encounter obstacles is relatively small because they are very penetrating and difficult to stop. Hence, the X-rays resulting from the stoppage of the β-rays will be of low penetrating power, and will not be capable of detection, for the γ-rays can only be detected by virtue of their extreme penetrating power and of the possibility of sorting them out from all the other types. Their effects are insignificant compared with that of the other two types.

β-rays.—On account of their relatively intense photographic action, and of their power of penetrating opaque screens of considerable thickness, the β radiation was at first the most studied. In much of the earlier photographic work, the term radiation must be interpreted to mean β radiation, and the work considered to refer only to this type. The existence of rays easily deviable in the magnetic field in the radiations from radium was discovered simultaneously by Giesel, Meyer and von Schweidler, and Becquerel. The latter subsequently (*Comptes Rendus*, 1900, 130, p. 1,584) showed the same to be true of the radiations from uranium. Rutherford and Grier (*Phil. Mag.*, 1902, VI., 4, p. 315) proved that thorium also gave out deviable rays, although the proportion of β-rays in the radiations of the latter is much less than in that of uranium. This explains the fact that, although the radio-activities of thorium and uranium are very similar when tested by the electrical method, the former is much less active than the latter to a photographic plate. In all cases it is the photographically active, penetrating β-ray which is deviated by a magnetic field. Curie (*Comptes Rendus*, 1900, 130, p. 73) using the apparatus shown in Fig. 16 showed that the easily-absorbed type of radiation from radium is not appreciably affected by a magnetic field, whereas the penetrating radiation was completely deviated. A is the radium preparation, BB′ are blocks of lead of varying heights, PP′ plates connected to the electrometer and battery respectively between which the ionisation due to the rays is measured. A powerful electromagnet (not shown in the figure) is arranged so as to deviate the β-rays into the lead blocks BB′. One pole of the magnet would be placed beneath and the other above the plane of the paper to secure this effect. If the distance AD is greater than 7cm., all the rays are deviated by a magnetic field, and are absorbed by the lead blocks. For smaller distances only a fraction is deviated. This proves that the non-deviable or α radiation of radium is completely absorbed in 7cm. of air. Under the conditions of experiment the effect of the γ-rays was too small to be detected. Becquerel investigated the magnetic deviation of the β-ray of radium by placing the substance in a metal capsule upon the sensitive film of a photographic plate wrapped in paper and placed in an exhausted space. Under these circumstances the rays formed a pencil proceeding normally away from the plate, and by applying a magnetic field they could be curved into circles cutting the plate at some distance from the point of origin. The disposition of the experiment is the same as in the

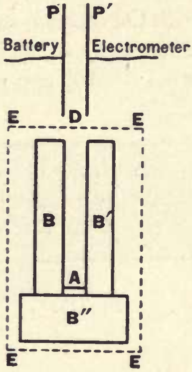


Fig. 16.

diagram, Fig. 15, where AC is the photographic plate. Fig. 17 represents a negative so obtained by first applying the field in the one direction and then in the other. The central black spot represents the position of the radio-active substance; the curved areas of darkening on either side the points where the rays, travelling upwards initially, are curved round by the field and cut the plane of the paper. Becquerel (*Comptes Rendus*, 1900, 130, p. 809) showed that the β radiation of radium is complex, and different rays suffer different deviations in a uniform magnetic field. From the equation $H\rho = mv/e$ (compare p. 50), Becquerel found values for mv/e varying from 350 (for rays capable of penetrating 0·01mm. aluminium) to 2,600 (for rays penetrating 0·13mm. of lead). Becquerel identified the β-rays with the cathode rays by proving that they were deviated in the same sense as the latter by an electrostatic field. He deduced $v = 1{\cdot}6 \times 10^{10}$ cm. per sec. for the average velocity of the rays, and this is several times greater than the value found by Prof. Thomson for the velocity of the cathode ray. This fact explains the much greater penetrating power of the β-rays. The great variation in penetrating power of the β-rays of radium is due simply to the wide range of velocities possessed by different rays. The value of e/m was found to be the same as for the cathode ray—viz., 10^7, and the β-rays must, therefore, be regarded as negatively charged particles of mass one-thousandth of the hydrogen atom moving at speeds comparable with that of light. This conclusion has been confirmed by the direct determination of the negative charge carried by the β ray by M. and Mme. Curie (*Comptes Rendus*, 1900, 130, p. 647), using the apparatus shown in Fig. 18. The plate MM on which the β-rays from the radium R impinge is completely

FIG. 17.

enclosed in paraffin and connected with an electrometer. In these circumstances any charge communicated to the plate will be retained, whereas if the plate were exposed bare to the strongly ionised air in the neighbourhood of radium it would tend to be discharged as fast as the charge carried by the rays were communicated to it. It is found that a negative charge is conveyed to the plate MM from the radium R *through the screen of thin metal, pp, connected to earth.* The charge is extremely feeble, and can only be determined for very active radium preparations. It is to be noted that it is, probably, only about the $\frac{1}{1000000}$th of the charge carried by the ions produced by the complete absorption of the rays in air. This fact cannot be too strongly emphasised. The ions shot out by radium are of the second class, and are detected by their kinetic energy. The part played by the air or gas in the measurement of the ionisation produced by radio-activity is a secondary feature, which serves as a convenient means of measurement. In vacuo, the ionisation produced by a radio-active substance would be *nil*, but the radio-activity would be the same as in air. In hydrogen the ionisation would usually be feebler than in air or carbon-dioxide, but the activity of the substance is not affected by the atmosphere surrounding it. In the case of the phenomenon of imparted activity already mentioned we measure the *ions* produced by the *rays* produced by the matter causing the imparted radio-activity, which is produced by the emanation which is produced by radium. The number of the steps involved in the production of the phenomena observed is one of the initial difficulties of the subject.

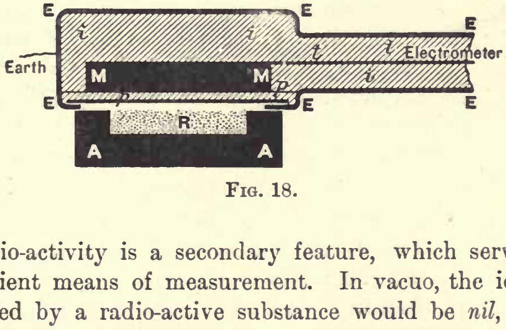


FIG. 18.

The existence of the negative charge carried away from radium by the β-rays has recently been shown as a lecture experiment by Strutt (*Phil. Mag.*, 1903, VI., 6, p. 588), with only a small quantity of radium, by the apparatus represented in Fig. 19. A tube, *a*, containing radium, is supported in a perfectly exhausted vessel by a quartz rod, *b*. To the lower end of the tube an electroscope, *cc*, is attached, and the surface of the tube *a* is made conducting with phosphoric acid. The positive charge left behind after the expulsion of the β-rays is communicated to the leaves, which diverge until they touch the sides of the vessel connected to earth. They then collapse and again commence to diverge, this cycle of operations being repeated indefinitely. In an apparatus constructed by the Author, about 2 milligrammes of pure radium bromide was used, and the cycle of operations occupied about four minutes.

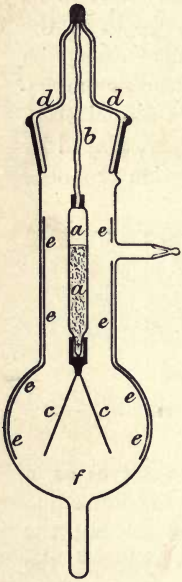


Fig. 19.

It is to be noticed that the charge thus measured is an *induced* positive charge on the outside of the glass repelled to the leaves by the accumulation of excess of positive electricity within. The latter accumulates continuously, and so increases until the strain becomes too great and the glass of the tube is punctured, as in the case of an over-charged Leyden jar. Several cases are on record in which sealed tubes of active radium preparations have shattered with the accompaniment of a bright spark and explosion after having been kept sealed up for some months.

Kaufmann (*Nachrichten* der K. Gesells. der Wiss. zu Göttingen, 1901, No. 2) has measured both v and e/m for the more penetrating kinds of β-rays emitted by radium, and obtained for v values between 2·36 and $2·83 \times 10^{10}$—*i e.*, between 80 and 95 per cent. of the velocity of light, Now, it has already been pointed out (p. 54) that if the mass of the corpuscle is electrical in origin and due to the inertia of the moving charge, it should, theoretically, tend to increase as the velocity of light is approached, and become infinite when that value is attained. Now Kaufmann found that the ratio e/m decreased from $1·31 \times 10^7$ when $v = 2·36 \times 10^{10}$ to $0·63 \times 10^7$ when $v = 2·83 \times 10^{10}$, and these results are in good agreement with the theory that the mass of the corpuscle or electron—*i.e.*, the negative atomic charge, which constitutes the cathode ray and the β-rays from radio-active substances—is, at least mainly, electrical in origin.

α-rays.—These are given out by all radio-active substances, with the possible exception of uranium X (p. 85), and, although the least striking, are much the most important of the three types, representing, in each case, as Rutherford has shown, by far the greater part of the total energy radiated. In measurements by the electrical method, unless special precautions are adopted, the effect of the β and γ-rays is negligible compared with that of the α-rays.

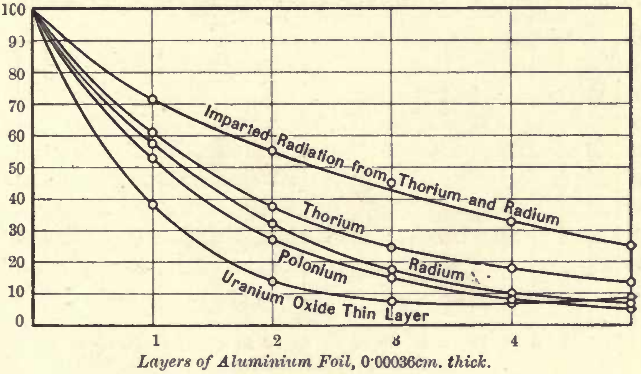


FIG. 20.

Throughout the book measurements of radio-activity must be taken to mean measurements of the intensity of the α-rays, unless otherwise stated. Their power of penetration is always very small, but varies considerably for the different radio-active substances. The diagrams (Figs. 20 and 21) were given by Rutherford and Miss Brooks (*Phil. Mag.*, 1902, VI. 4, p. 1), and show the penetrating power of the α-rays of uranium, polonium, radium, thorium, together with that of the imparted radio-activities produced by the radium and thorium emanation. In each case the vertical axis represents the rays transmitted, while the horizontal axis represents the thickness of matter penetrated, this being, in Fig. 20, made up of successive layers of aluminium foil of thickness 0·00036cm., and in Fig. 21 of the thickness of air in millimetres. It will be seen that the order is the same for the different rays in the case of aluminium and air, those from uranium being the least and from the imparted activities the most penetrating. The density law, that the absorption of the rays by various substances is proportional to the density of the latter, holds fairly generally for the α-rays. Tin furnishes an exception, its opacity being about the same as for aluminium, although it is three times as dense as the latter. Tin is also an exception in its power of absorbing the β-rays, but in the opposite direction, being three times more absorbing than its density would indicate. It thus absorbs β-rays nine times more than the α-rays compared with other metals. Lead also absorbs the β-rays abnormally, being twice as opaque as other metals in proportion to its density. In this respect the γ-rays resemble the β-rays completely.

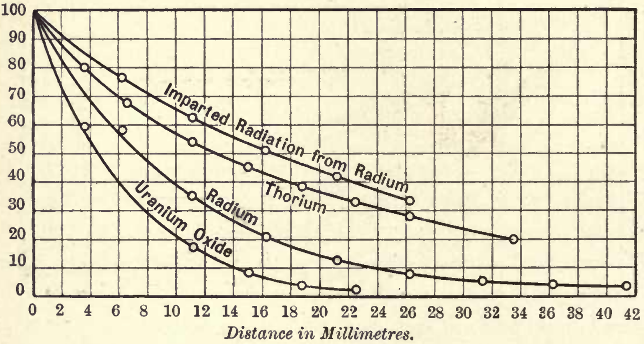


Fig. 21.

A very characteristic property of the α rays is that they are absorbed the *more* readily the greater the thickness of matter traversed (compare Mme. Curie, "Thesis," Chap. III.; Rutherford, *Phil. Mag.*, 1903, VI., 5, p. 114). The apparatus employed by Mme. Curie for the investigation of the α-rays of polonium is shown in Fig. 22. The object of the experiment is to measure the ionisation current due to the rays between the plates PP, P′P′ from a polonium preparation at A when the distance AT was varied. The hole T in the plate P′P′ is covered with the thinnest possible sheet of aluminium foil (not shown) to prevent the ions formed in CCCC finding their way into the space PPP′P′. When AT is over 4cm. no current passes, indicating that the rays are all absorbed in this thickness of air. The appearance of the rays between the plates is manifested somewhat suddenly as the distance AT is reduced. If successive layers of foil are placed over the polonium, the absorption caused by the second layer is greater than that produced by the first.

These considerations and the existence of this peculiarity for α-rays generally led Rutherford (*Phil. Mag.*, 1903, VI., 5, p. 177) to regard the α-rays as consisting of projectiles, and to examine them carefully in the most intense magnetic fields under conditions where the slightest deviation could be detected. His apparatus is represented in Fig. 23. The radium rays pass upwards through a series of very narrow slits placed between the poles of a very powerful electromagnet and emerge inside the gold-leaf electroscope placed above. A current of hydrogen is kept flowing through the electroscope and the slits in a downward direction, and this serves to prevent the diffusion of any emanation from the radium into the electroscope.* It

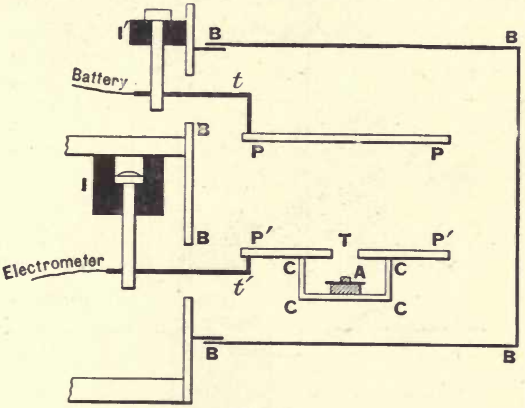


Fig. 22.

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* The aluminium foil placed above the top of the slits serves the same purpose as in Mme. Curie's apparatus just described. It is excessively thin and, being porous, offers no obstruction to the passage of the hydrogen.

also greatly reduces the absorption of the α-rays in passing through the slits, and so increases the effect to be measured in the electroscope. The effect due to the β and γ-rays is, on the other hand, very much diminished by using hydrogen, owing to the ionisation in the latter gas being much less for a penetrating radiation than it is in air. The ionisation due to the α-rays is not reduced because there is a sufficient thickness of gas to ensure their *complete* absorption inside the electroscope. The plan of the experiment was to measure the rate of collapse of the leaves when the radium salt was bare and when covered

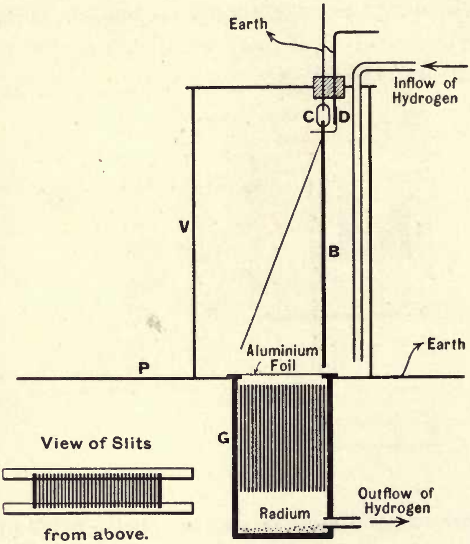

FIG. 23.

with a sheet of mica sufficient to absorb the α-rays, both with and without the magnetic field. Under proper conditions the rate of collapse is the same with the salt bare and the magnetic field on as with the salt covered and the field off. But in the latter condition the residual magnetism was always sufficiently strong to eliminate the β-rays, so that these, under no circumstances, enter the electroscope. In this way Rutherford showed that the α-rays were completely deviated by passage through slits 4·5cm. long and 0·55cm. wide placed in a uniform magnetic field of 8,400 C.G.S. units. Deviation was also obtained by an electrostatic field; and the direction of deviation in each case is opposite to that suffered by the cathode ray or the β-ray of radioactive substances. The direction of deviation was determined by using the device shown in Fig. 24, in which the openings of the slits are half covered from the one side by metal plates, so that a greater deflection is necessary in the one direction than in the other if no rays are to enter the electroscope. From a combination of his results for both the electrostatic and electro-magnetic deviation, Rutherford has deduced that the velocity of the charged particle $v = 2{\cdot}5 \times 10^9$cm. a second, while the ratio of the charge to the mass $e/m = 6 \times 10^3$. If e is assumed to be the "atomic charge," and equal to the charge carried by the hydrogen ion in electrolysis, the mass of the projected particle is thus about 1·6 times that of the hydrogen atom. The value of $H\rho$ in these experiments was 390,000, and this result may be expressed differently by the statement that the α-rays would be coiled into circles of 39cm. radius under conditions which would coil the cathode rays into circles of 0·01cm.

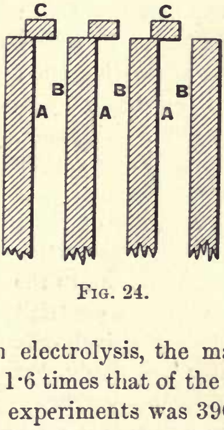

FIG. 24.

The magnetic deviability of the α-ray of radium has since been confirmed by a photographic method by Becquerel (*Comptes Rendus*, 1903, 136, pp. 199, 431 and 1,517), and he also shows that the α-ray of polonium is deviated to a similar extent. Des Coudres also (*Phys. Zeitschr.*, 1903, p. 483), working by a photographic method in vacuo, has confirmed both the electrostatic and electromagnetic deviation, and obtained a value of e/m in close agreement with that given by Rutherford.

Since the α-rays of all radio-active substances are very similar in type, there is strong evidence for considering them all as consisting of positive-charged particles of mass about twice that of the hydrogen atom travelling with a speed one-tenth of that of light. Their large size compared with that of the β-ray accounts for their small penetrating power, and their great kinetic energy for their very great ionising action, while their projection character well accords with the property already mentioned of being more easily stopped the greater the thickness of matter penetrated.

This view of the discrete nature of the α-radiation has recently been confirmed by an experiment of Sir William Crookes' (*Chem. News*, 1903, 87, p. 241), which was discovered simultaneously in Germany by Elster and Geitel (*Chem. News*, 1903, 88, p. 37), that the phosphorescence of a zinc sulphide screen under the action of the α-rays consists, when viewed through a lens, of a number of momentary flashing points of light or scintillations, presenting the appearance of an incessant bombardment of the screen by a rain of projectiles from the radium, each impact being marked by a separate flash of light. An instrument, known as the "Spinthariscope," has been designed to show this effect.

We thus see that the rays of radium have furnished most striking and unexpected evidence of the correctness of the views adopted by Prof. J. J. Thomson, as the result of his investigations into the relations between electricity and matter. The α-ray has been shown to be a positively-charged particle of atomic dimensions, the β-ray a negatively-charged particle of corpuscular dimensions, in accordance with the corpuscular theory of electricity, which requires that positive electrification does not exist apart from matter, whereas negative electrification is composed of discrete charges of far smaller mass than the lightest atom of matter. In addition the β-ray of radium has afforded a welcome means of putting to experimental test one of the most fundamental consequences which follow from the view that an electric charge possesses inertia and, therefore, mass.

CHAPTER VI.

URANIUM X AND THORIUM X.

Radio-activity an Atomic Property.—The First Idea Untenable.—Crookes' Uranium X.—Decay of Activity of Uranium X.—Hypothesis of Radio-active Induction.—Uranium X gives only β-Rays.—α Radiation a Specific Property of Uranium.—Thorium X.—A Specific Type of Matter.—Greater Part of α-Rays, all the β-Rays, and the Emanating Power due to Thorium X.—Decay and Recovery of Activity of Thorium X and Thorium.—Continuous Reproduction of Thorium X.—Radio-active Change.—Radio-active Equilibrium.—Radio-active Constant λ.—Independence of Radio-active Change towards all known Agencies.—Curves of Decay and Recovery of Activity of Uranium X and Uranium.—Homogeneity of the Elements Thorium and Uranium.

It has been shown in the preceding chapters that the phenomenon of radio-activity consists essentially in the incessant and spontaneous expulsion of positively and negatively-charged particles from the three heaviest chemical elements that are recognised—viz., radium, thorium and uranium. The velocity of both the positively and negatively-charged particles shot out spontaneously from the radio-elements is greater than is ever impressed by external agencies upon the positive and negative ions of the electric discharge, even when the latter are caused to move in electric fields of very great intensity. In consequence, the kinetic energy associated with the α and β particles is greater than in the case of the ions artificially produced, and is, in fact, considered with reference to their mass, of a far higher order of magnitude than is associated with matter in any other form.

We have now to consider the radio-active matter from which these particles are projected, and from which their energy must in some way be derived. It must be first pointed out that matter of this kind, from which energy is being continuously dissipated in a new and easily detected form, is naturally far more easy to investigate and trace than non-radio-active matter. New experimental methods of great delicacy are available which are capable of detecting the presence of such matter in quantity so minute that no other indication of its presence exists. The first fruits of the researches in this direction were, of course, Mme. Curie's discoveries of radium and polonium, which have already been discussed. It will be recalled that these discoveries resulted from the view that radio-activity is an atomic property of the element, in that it is exhibited by each radio-element in amount sensibly proportional to the quantity present, and independent of its state of chemical combination, of physical conditions, and of previous history. On this view, a substance like pitchblende, more radio-active than any known element, owes its radio-activity to the presence of new elements of proportionally great radio-activity, and not to any cause increasing the activity of the known radio-elements contained therein beyond the normal value. In spite of the successful separation of these predicted new elements, a series of researches, which have now to be considered, have shown that the view that radio-activity is an atomic property of the element in question cannot be accepted in its original form. Mme. Curie, in one of her earlier papers, describes radio-activity as a property exhibited by *each* atom of the radio-active element, which acts as a continual source of energy. This view is now known to be untenable, for we shall see that the normal radio-activity of a single element, like thorium, is due to several different types of matter radiating simultaneously, and the thorium contributes but a small part of the total radio-activity.

The discrepancy has been reconciled by Rutherford and Soddy in their disintegration theory of radio-activity, and as this view accounts in a simple manner for all the new facts at present known with regard to the radio-elements, and greatly assists the presentation of the subject as a connected whole, it is used as the basis of arrangement in the remaining chapters. It must be borne in mind that the order in which the researches will be considered is frequently the inverse of the historical order.

Uranium, of the three radio-elements, furnishes the simplest example and will be considered first. Sir William Crookes

(*Proc.* Roy. Soc., 1900, 66, p. 409), showed that it was possible by various processes to free uranium completely from rays which affect a photographic plate, and to concentrate the whole of the photographic activity in a minute fraction of the total matter which contained no uranium. He gave several methods, of which two may be considered. If crystallised uranium nitrate is dissolved in ether, the solution separates into two layers: (1) an ethereal solution containing the bulk of the dissolved uranium salt; (2) an aqueous solution consisting of the water of crystallisation of the original salt, in which a relatively small amount of uranium is dissolved. He found that the uranium salt obtained from (1) was completely inactive to the photographic plate, while that obtained from (2) possessed all the activity of the original uranium salt in concentrated form. Again, if a solution of a uranium salt be precipitated by ammonium carbonate solution, and a sufficiency of the latter is added in excess of that necessary to cause precipitation, the precipitated uranium carbonate again dissolves, and on filtering the solution a very minute precipitate of insoluble matter is obtained. This consists mainly of iron and aluminium present in the uranium as impurities, and by the manner of its preparation is chemically free from uranium. Yet this minute precipitate possessed all the photographic activity of the original uranium, and the latter obtained from the solution was completely inactive. In this way Crookes obtained preparations many hundred times more active to the photographic plate than the uranium salt itself. Thinking that possibly he had separated a new radio-element of intense activity, which caused the radio-activity of uranium, Sir William Crookes proposed the name "Uranium X" for the new substance.

It must be noticed that the actual amount of uranium X obtained is probably quite incapable of detection except by its radio-activity. The bulk of the substance is iron, aluminium, &c., present in the uranium as minute impurities which afford the necessary solid nuclei for the separation of the uranium X by the relatively gross process of filtration.* No new lines

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* The Author has noticed that this separation may fail if the uranium preparation is too pure, until a trace of ferric salt or similar impurity is added to act as nuclei in the filtration.

were detected by Crookes in the spectrum, and everything points to the conclusion that the actual amount of matter causing the radio-activity of uranium X is infinitesimal.

Becquerel (*Comptes Rendus*, 1900, 131, p. 137), independently of Crookes, obtained evidence of the enfeeblement of the radio-activity of uranium by chemical treatment. Acting on a suggestion of Debierne, he dissolved barium salts in solutions of uranium, and precipitated the barium with sulphuric acid. He found that the precipitated barium sulphate tended to "drag down" the activity of the uranium, leaving it more or less inactive. He so obtained specimens of barium sulphate more radio-active, weight for weight, than the salts of uranium used in their preparation.

The constancy of the radio-activity of uranium, independent of its source, and previous history, which had been shown by Mme. Curie, and was, indeed, the fundamental starting point of her subsequent discoveries, caused Becquerel to consider as improbable Crookes' hypothesis that the radio-activity of uranium was due to a small intensely active impurity (uranium X). He concluded (*Comptes Rendus*, 1901, 133, p. 977) that, if uranium, like radium, possessed the power of making admixed inactive bodies temporarily radio-active by association with them, the phenomena could be explained. This supposed communication of radio-activity to an inactive molecule had been termed "radio-active induction." Thus M. and Mme. Curie found that objects in the neighbourhood of radium became temporarily radio-active, and called this phenomenon "the induced activity." The theory of Rutherford, which we shall deal with more fully in the sequel, that this radio-activity was *imparted* to the inactive matter as a deposit of active matter from the radio-active emanation of radium, had not then been accepted by the Continental investigators. This effect of radio-active induction, Becquerel argued, would be produced equally on the active and inactive matter present, so that by induction on itself the activity of the mixture should increase spontaneously at first. In a similar way the activity of solid radium salts was known to increase from the moment of their preparation. If barium were rendered active by induction when it was mixed with uranium, by precipitating the barium it would be possible to remove some of the activity, and, then, after removal, the induced activity of the barium should gradually decay, in a manner similar to the induced activity of radium, while that of the uranium should gradually increase. Becquerel, therefore, examined his preparations 18 months afterwards and found that the activity of the barium sulphate had completely disappeared, while the uranium had fully recovered its normal activity. Subsequently it was shown (Soddy, *Journal* Chem. Soc., 1902, 81, p. 860) that the processes of Crookes and Becquerel resulted only in the separation of the β-radiation of uranium, and that the relatively far more intense α-radiation of uranium was completely unaffected. Thus, when the activity of the uncovered salt is examined by the electrical method, it is found to be sensibly the same after chemical treatment as it was before, although the same preparation has been rendered completely inactive to the photographic plate. On the other hand, the uranium X obtained from it by Crookes' ammonium carbonate process was but feebly active by the electrical, but intensely active by the photographic, method. But a very thin sheet of aluminium (0·005cm. thick) placed over the uranium completely absorbed the radiation, whereas the rays from uranium X are not appreciably absorbed by this thickness of metal. The view that the β-radiation of ordinary uranium is caused by uranium X, a non-uranium type of matter, which can be chemically separated, whereas the α-radiation is a specific property of the uranium and is not affected by this separation, was confirmed by Rutherford and Grier (*Phil. Mag.*, 1902., VI., 4, p. 315), who examined the radiations from the two preparations in a magnetic field. They found the radiation from the uranium X to consist entirely of rays easily deviated, while that from the uranium wholly consisted of non-deviable rays. The chemical separation had analysed the radiation into its two components (α and β) without affecting the nature or intensity of the rays in any way. Uranium is the simplest example of the three radio-elements, for its two main types of radiation result from two different kinds of matter, the one constituting practically the entire mass of the substance, the other present in proportion so minute that its radio-activity is at present the sole evidence we have of its existence. The recovery of the β-radiation of uranium with time, and the decay of the

β-radiation from the uranium X with time, thus appear to be phenomena of momentous import.

Simultaneously with the above researches Rutherford and Soddy (*Trans.* Chem. Soc., 1902, 81, pp. 321 and 837) had obtained results for the case of thorium similar in character to those obtained by Becquerel for uranium. If thorium nitrate in aqueous solution is precipitated by ammonia,* it is found that a large part of the radio-activity of the thorium remains in the solution, so that, by evaporating the filtrate to dryness and igniting to remove ammonium salts, minute residues are left possessing radio-activity relatively intense compared to that of the original thorium. The thorium hydroxide, which is precipitated completely by this process, on the other hand, is found to have lost the greater part of its radio-activity, and by suitable repetitions may be obtained only one-quarter as active, weight for weight, as the original salt. Now this behaviour is by no means due to the *precipitation* of thorium from its solution. If reagents other than ammonia are employed—for example, sodium or ammonium carbonate, ammonium oxalate and alkaline sodium phosphate—the thorium is completely precipitated in the normally active condition, and the residues obtained from the several filtrates by evaporation and ignition are in each case quite inactive.

Rutherford and Soddy, therefore, concluded that the major part of the radio-activity of thorium is due to the presence of a non-thorium type of matter, not precipitated by ammonia, and they gave the name "Thorium X" to this body, in accordance with the nomenclature adopted by Sir W. Crookes in the case of uranium. Ammonia is the only reagent at present known which will separate thorium X from thorium. Besides being responsible for the greater part of the radio-activity of thorium, the power of thorium preparations of giving a radio-active emanation and of imparting radio-activity to surrounding objects is wholly due to the presence of thorium X. For the power of a thorium solution to give the emanation is retained unaltered by the *solution* after

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* The ordinary chemical equation for this reaction is—

$$Th(NO_3)_4 + 4(NH_4)OH = Th(OH)_4 + 4(NH_4)NO_3.$$

Thorium Nitrate (soluble).	Ammonium Hydrate (soluble).	Thorium Hydroxide (insoluble).	Ammonium Nitrate (soluble).

the thorium has been precipitated as hydroxide, and the latter when re-dissolved, possesses at first, no emanating power at all. As in the case of uranium X, the actual quantity of matter causing these radio-active manifestations is, probably, infinitesimal. Thorium X has no definite analytical reactions which would lead to its separation from the impurities always present, but is dragged down by any precipitate formed in its solution to a greater or less extent by "adsorption." Barium sulphate, pre-eminently, appears to possess the general power of removing in this way the infinitesimal traces of radio-active matter (ThX, UrX, &c.) present in solutions of the radio-elements. In the language of the theory of radio-active induction this result would be expressed by saying that barium is easily induced into activity by contact with radio-active matter. The character of the activity of barium, so "induced," is, however, different according as it is precipitated in solutions of uranium or thorium, and in each case is distinctive of the original element. The character of induced activity is quite independent of the nature of the matter "induced," whether barium sulphate or any other precipitate which shows the same property.

If the preparations of thorium and thorium X so obtained are allowed to remain untouched, it is found that the thorium gradually regains the activity that it has lost until it is just as active as at first, while the thorium X loses its activity gradually, until it becomes at length completely inactive. These processes occur rapidly compared with the similar case of uranium and uranium X. In three weeks or a month the thorium X preparation has lost practically all its radio-activity, while the thorium in the same interval has completely regained its lost radio-activity. The same holds true equally of the emanating power of the preparations. The thorium X at first possesses all the emanating power of the original thorium solution and loses it completely in one month, whereas the precipitated thorium hydroxide, if re-dissolved in nitric acid and kept in solution, at first possesses no emanating power at all, but gradually recovers it, until, at the end of a month, it again possesses its original power.

When a thorium preparation is precipitated in the above manner several times successively at short intervals, its radio-activity is reduced to about one-fourth of the normal, and this *non-separable radio-activity*, which is, as far as we know, a specific property of the element itself, consists only of α-rays. The β-radiation is wholly retained by the thorium X. These results may be conveniently represented in tabular form.

Ordinary Uranium by addition of ammonium carbonate in excess to the solution		*Ordinary Thorium* by addition of ammonia in excess to the solution	
(Insoluble.) *Uranium X* giving all the β-rays, no α-rays.	(Soluble.) *Uranium* giving all the α-rays, no β-rays.	(Soluble.) *Thorium X* giving 75% α-rays, all the β-rays, all the emanating power.	(Insoluble.) *Thorium* giving 25% α-rays, no β-rays, no emanating power.

Thus, of the three main phenomena which go to make up the normal radio-activity of thorium compounds (1) α-radiation, (2) β-radiation, (3) production of a radio-active emanation, only 25 per cent. of the α-radiation can be considered to be specifically due to the thorium atoms, the remaining α-radiation, together with the β-radiation and the emanating power, being caused by the presence of a non-thorium type of matter—thorium X—present in infinitesimal quantity. Whereas, however, the thorium X, on keeping, rapidly loses its radio-activity, the thorium from which it was separated regains its lost radio-activity, until at the end of a month it again possesses its normal or maximum value. In the same time the thorium X becomes completely inactive. If, now, the thorium that has completely recovered its radio-activity is again precipitated with ammonia, *a new amount of thorium X* is obtained from the filtrate of the same radio-activity as that originally extracted, and the thorium is again freed from the major part of its activity. This cycle of operations can be repeated indefinitely. If, however, the precipitations are carried out without lapse of time, the radio-activity of the thorium is not further reduced below a certain minimum value, and when this is reached no more thorium X is obtained and the residues from the filtrates are inactive. By waiting for any period less than one month between successive precipitations a new quantity of thorium X is obtained, in amount (as judged by its activity, which is the only criterion we have of its presence) the greater the longer the time waited.

If it is admitted that thorium X is a specific kind of matter, the only explanation of this behaviour is that it is being *continuously produced* by thorium. This is equivalent to saying that thorium is *changing* into thorium X, unless we suppose that thorium has the power of *creating* thorium X, which is hardly conceivable in the present state of science. The view that thorium X is a specific kind of matter therefore necessitates the belief that an element is capable of undergoing a slow spontaneous change, and it will be convenient to designate this by the special name of *radio-active change.* As this is the point of departure from which springs the whole theory of atomic disintegration as applied to explain radio-activity, it is necessary to bear in mind the precise character of the fundamental proposition. The alternative is to suppose that thorium X is not a specific kind of matter, but is ordinary inactive matter of some kind unknown, in which strong radio-activity has been "induced" by association with the thorium, and which loses again this property when it is removed from the thorium. The evidence in favour of the first and opposed to the second view is drawn not only from the behaviour of thorium X and uranium X, but from the behaviour of at least six other similar types of temporarily radio-active matter. The most conclusive evidence is drawn from the study of the radio-active emanations of thorium and radium, which have still to be considered. On the other hand, the second view finds to-day no support, and no attempt has been made, since the processes of radio-activity have become better understood, to reconcile the idea of radio-active "induction" with the overwhelming evidence that can now be brought against it. The effects to which the term "induction" was first applied are now admitted to be capable of a direct explanation, and the process, in the sense of the communication of radio-activity to an inactive molecule, has never been shown to take place. Even if it did, it would merely constitute an additional fact to be explained without itself aiding in the explanation of any of those already known.

Once, however, the possibility of an element undergoing change is admitted, the varied phenomena of radio-activity can be explained from a consistent point of view without further difficulty.

A quantitative study of the rate of recovery of the activity of thorium and the rate of decay of the activity of thorium X (Rutherford and Soddy, *Phil. Mag*, 1902, VI., 4, p. 378) revealed a simple connection between the two processes. Fig. 25 represents the curves obtained in which the radio-activity is plotted on the vertical and the time in days on the horizontal axis. Curve I. represents the decay of activity of thorium X, the initial activity being represented as 100, and curve II. the recovery of activity of thorium to a constant maximum value, represented also as 100. For the first day the effects are abnormal, the activity of the thorium X increasing slightly before commencing to diminish, while that of the thorium at first slightly diminishes and then proceeds to increase in a regular manner. These initial irregularities will be examined later. For the

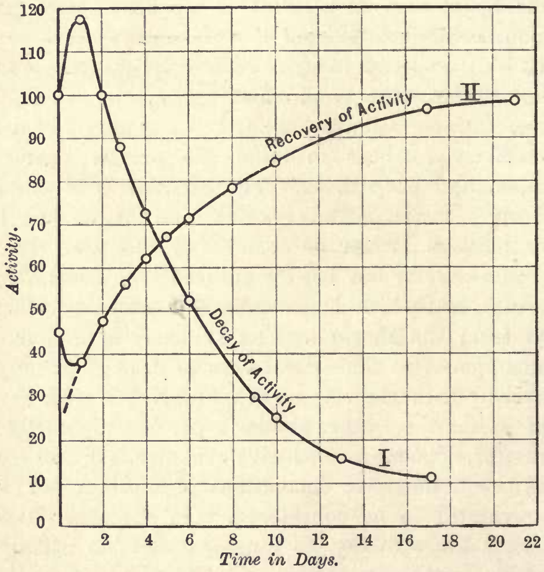

Fig. 25.

present they may be neglected, and the subsequent regular changes be considered, without involving any serious error. It will be noticed that the recovery curve, if produced backwards, cuts the vertical axis at about 25 per cent., and this can be shown to be due to the thorium possessing a constant non-separable radio-activity specific to the element itself. In Fig. 26 the relation between the two curves is more clearly brought out. Curve I. represents the percentage proportion of the activity recovered by the thorium after the second day, the activity recovered from the beginning to the end being taken as 100. Curve II. represents the activity of thorium X on the same scale during the same period.

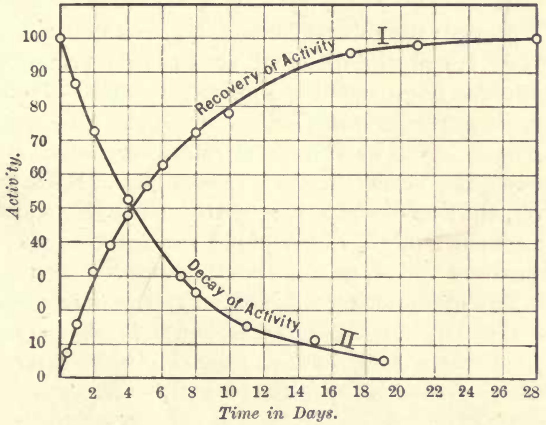

FIG. 26.

It will be seen that the proportion of the radio-activity recovered by the thorium during any interval throughout the whole period is equal to the proportion of the radio-activity lost by the thorium X during the same interval. If I_0 represents the original activity of the thorium X, and I_t the activity after any time t, I'_t the activity recovered by the thorium after time t, and I'_∞ the maximum activity recovered by the thorium when the constant value is attained,

$$\frac{I'_t}{I'_\infty}=\frac{I_0-I_t}{I_0}=1-\frac{I_t}{I_0} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

The activity of the thorium X decays very approximately in a geometrical progression with the time, falling to half-value in four days. Thus, in eight days it is one-quarter of the original, in 12 days one-eighth, and so on. This is expressed by the equation

$$\frac{I_t}{I_0} = e^{-\lambda t}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

where λ is a constant and e the base of natural logarithms. Since $\frac{I_t}{I_0} = \frac{1}{2}$ when $t = 4$ days $= 345{,}600$ seconds, $\lambda = 2 \times 10^{-6}$, when t is expressed in seconds.

From equations (1) and (2) we can write at once

$$\frac{I'_t}{I'_\infty} = 1 - e^{-\lambda t}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

which represents the recovery of activity by the thorium with time. The law of the decay of activity of thorium X in a G.P. with the time (equation 2) is a general law for temporarily radio-active substances. Many such substances are known, decaying at characteristic rates, varying from a few seconds to a few years, but always according to the same law. Moreover, the rate of decay has been found to be independent of the most powerful chemical and physical agencies. Extremes of temperature and drastic chemical treatment do not at all affect it. In consequence, λ (equation 2) serves as a perfectly definite constant for the identification and characterisation of any of these new bodies, none of which have been obtained in sufficient quantity for the ordinary methods of investigation. λ may be called the *radio-active constant*, and we shall see later that it possesses a profound physical meaning.

The chemical separation of thorium X from thorium, therefore, cannot have any effect on the activity of the former in causing it to decay. The activity of thorium X must always be decaying at the same rate, whether it is present in the thorium compound producing it or whether it is separated from it. The apparent constancy of activity of the radio-elements results from the continuous production of fresh matter possessing continuously diminishing radio-activity. When the increase in radio-activity through the production of new active matter balances the decrease through the decay of the activity of that already formed, the activity remains constant. This point may be termed the *radio-active equilibrium.* The mystery of the constancy and permanence of the emission of rays from radio-active substances, which from the first riveted attention on the new property, is, therefore, one step nearer elucidation. The rays in question are always dying down and are always being renewed. The constancy is apparent rather than real, and is due to the balance or equilibrium between opposed processes.

The next consideration that follows from equations (2) and (3) is that a uniform quantity of thorium X, as measured by its radio activity, is produced in each unit of time. The constancy of the activity after radio-active equilibrium is attained shows that the production of new matter must proceed at a constant rate. This holds true, not only for the condition of radio-active equilibrium, but generally. Thus, equation (3), which was obtained as the experimental relation holding for the recovery of activity of thorium, is that theoretically required for the rise of activity of a system in which (1) the rate of supply of fresh radio-active matter proceeds at a uniform rate ; (2) the radio-activity of the matter supplied decays with time, according to equation (2) (compare Rutherford, *Phil. Mag.*, 1900, V., 49, p. 179).

The form of the recovery curve is not altered by the conditions under which the change occurs. It was found that different parts of the same specimen of thorium, which had been freed from thorium X, kept under widely different conditions of temperature, state of chemical combination, &c., recovered their radio-activity at the same rate. This absolute independence, so far as we know at present, of the rates of recovery and decay, towards all the agencies at our disposal, is one of the most striking features of radio-active change. In this respect the latter is sharply differentiated from all the other kinds of material change, whether physical or chemical. The process which proceeds spontaneously in Nature, and gives rise to the phenomenon of radio-activity, is entirely beyond the range of ordinary molecular forces. This of itself furnishes strong presumptive evidence in favour of the view that radio-active change involves an alteration in the internal structure of the chemical atom and its transmutation into other atoms. For, since molecular forces are quite incapable of transmuting one elementary form of matter into others, it is not to be expected that such forces would produce any effect on the course of a natural process of transmutation which was proceeding spontaneously. For a similar reason it is extremely improbable that any such effect as radio-active induction, or the rendering of an inactive substance radio-active, exists. If it did it would constitute a case of artificial transmutation, which, at least, requires further proof before being accepted.

On the new views, the source of the energy dissipated by radio-active substances no longer presents any fundamental difficulty. The explanation is similar in kind and differs only in degree from the well-known cases of slow spontaneous chemical changes in which energy is liberated, as, for example, in the ordinary processes of living organisms. In each case matter is "consumed," that is, transformed into new kinds which possess less energy. Direct experimental data, to be considered in the sequel, show that the energy of radio-active change is of the order of a million times greater than is ever manifested in ordinary chemical change. It is in the enormous stores of energy liberated during radio-active change that the disintegration theory derives its most unanswerable argument. A fuller consideration of this aspect of the question must wait until the case of radium has been considered. It is probable that the order of the energy liberated is, *for equal weights of matter changing*, similar in all the radio-elements. But in the case of radium the change proceeds a million times faster than in the case of uranium or thorium, and this accounts for the surprising nature of the properties of the first-named element.

An examination of the recovery and decay of the radio-activity of uranium and uranium X (Rutherford and Soddy, *Phil. Mag.*, 1903, VI., 5, p. 422) showed that the considerations just developed for thorium apply equally well for the case of uranium. Nearly six months is required, however, before the radio-active equilibrium is attained in this case. Since the uranium X gives all the β or penetrating rays of uranium, the course of the production of uranium X can be followed by measuring the penetrating radiation from uranium after the uranium X has been separated chemically. Fig. 27 shows the curves of recovery and decay of the penetrating radiation of uranium and uranium X with time. When separated from the uranium producing it, the activity of uranium X decays in a geometrical progression with the time, as shown in Fig. 27, and falls to half-value in about 22 days. The value of λ (equation 2) for this case is, therefore, about $3{\cdot}6 \times 10^{-7}$, when t is expressed in seconds. When free from uranium X, uranium gives practically no penetrating rays. The gradual recovery of the β-radiation by the uranium is shown by the second curve of Fig. 27. The proportionate loss of activity of uranium X is for any time approximately equal to the proportionate recovery of the β activity of the uranium, and equation (3), with the value of λ just given, applies to the recovery of the activity of uranium equally to that of thorium.

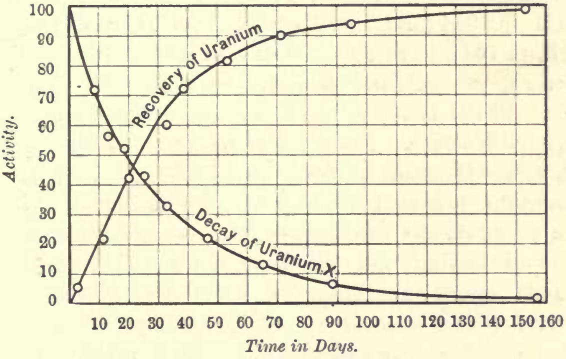

FIG. 27.

Special interest attaches to the case of uranium on account of its simplicity. The radiation can be analysed into its two constituents and the uranium shown to be responsible for the whole of the one type, the uranium X for the whole of the other. Then, again, uranium, unlike thorium and radium, does not produce radio-active emanations or impart radio-activity to its surroundings. These secondary effects cause the initial irregularities in the curves of decay and recovery for thorium which are absent in the case of uranium. On the other hand, the non-separable activity, which in the case of thorium is a relatively small fraction of the whole, in the case of uranium comprises the whole of the α-radiation, and therefore the greater part of the total energy radiated. The existence of this non-separable radio-activity is general for all three radio-elements, and does not appear to be merely due to the insufficiency of our chemical methods in analysing the sources of the radio-activity. It is possible that further chemical investigation will show that what is here termed non-separable activity is in reality due *in part* to new types of matter not yet separated from the radio-elements. But it is unlikely that the latter will ever be obtained by any chemical process entirely free from radio-activity, and then recover it with the lapse of time.* In fact, the non-separable activity has a very important and fundamental bearing on the final view we adopt of the cause and nature of radio-activity.

Two observers (Brauner, *Trans.* Chem. Soc., 1898, Vol. LXXIII., p. 951; Baskerville, *Journ.* Am. Chem. Soc., 1901, XXIII., p. 761), have obtained evidence of a new element of heavier atomic weight, associated with thorium in its compounds, and the latter believes that pure thorium compounds are not radio-active. Recently, Hoffman and Zerban (*Berichte der Deutschen Chemischen Gesellschaft*, 1903, p. 3,093) state that the thorium prepared from gadolinite is not radio-active. Before these results can be accepted, more details as to the method of testing and the time since preparation of the specimens employed must be made known. For both Becquerel (*Comptes Rendus*, 1902, 134, p. 208) and Sir William Crookes (*Proc.* Roy. Soc., 1900, Vol. LXVI., p. 409) considered that they had separated the radio-activity from uranium, although we know now that this result was only partially true (compare *Trans.* Chem. Soc., 1902, Vol. LXXXI., p. 860). The non-activity of thorium from gadolinite has already been called into question (George F. Barker, *Am. Joun. Science*, 1903 [4], 16, p. 161), and reaffirmed

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* It is, of course, possible that what we call thorium and uranium respectively may prove to be mixtures of more than one element, only one of which is radio-active. In this case the proportion of the respective active constituents must be the same in all preparations of these elements that have been examined, for they all possess similar activity. Moreover, on separating the active constituent the uranium and thorium will then *not* recover their lost activity.

(Zerban, *Ber.*, 1903, p. 3,911). Even if it is ultimately established it does not interfere with the above reasoning, for when once the active constituent is separated from thorium the latter will *not* recover its activity with time. The considerations deduced with regard to the non-separable activity would then apply without modification to the active constituent, and whether this is thorium or so like thorium that it has not yet been separated from it, is a matter of little theoretical importance in the present connection.

CHAPTER VII.

THE RADIO-ACTIVE EMANATION OF THORIUM.

The Variability of the Radio-activity of Thorium.—Effect of Air-currents.—Uranium and Thorium Contrasted.—The Radio-active Emanation of Thorium.—Emanating Power Proportional to Weight, Radiating Power to Surface.—The Radiation of the Emanation consist of only α-Rays.—Rate of Decay of Activity.—The Emanation Analogous to the Argon Family of Elements.—Decay of Activity Unaffected by Temperature, &c.—Emanating Power Persists in Absence of the Atmosphere.—Emanation Condensed by Liquid Air.—Produced by Radio-active Change of Thorium X.—Imparted Radio-activity.—Due to Matter Deposited from the Emanation.—Rate of Decay.—Imparted Activity Concentrated by an Electric Field.—Produced by Radio-active Change of Emanation.

A detailed investigation of the radiation from thorium (Owens, *Phil. Mag.*, 1899, V., 48, p. 360) brought to light a curious difference between the radio-activity of this element and that of thorium. It was found that the ionisation current through the air in a closed space under the influence of the rays from the various thorium compounds increased with time up to a maximum value. If air was then drawn continuously through the apparatus by means of a pump, the current immediately decreased to a minimum value. This effect was more marked for a thick layer of thorium compound than for a thin layer. In the former case the maximum current was three times as great as the minimum. Rutherford (*Phil. Mag.*, 1900, V., 49, p. 1) showed that this effect was due to thorium compounds possessing the property of continuously emitting into the air particles of some kind which possessed temporary radio-activity. That is, the air in the neighbourhood of a thorium compound possessed the power of giving out radiations on its own account, and this power persisted in the air for some time after the thorium compound was withdrawn. Rutherford named the radio-active substance communicated to the air in this way the "thorium emanation," and although it is now known to be a distinct type of matter, like thorium X or uranium X, the original name has been retained. He found that the rays of thorium are completely absorbed by covering the salt with a single thickness of foolscap paper, whereas the emanation readily finds its way through 20 layers, so that the effect of the emanation alone can be well studied by enclosing the thorium salt in paper. Under these circumstances the ionisation current observed when the package is placed between the plates of the testing apparatus is reduced to a very small fraction of its original value when the blast of air is directed between the plates. The effect of β-rays of thorium in experiments by the electrical method is exceedingly small and would hardly be detected except by special arrangements. The thinnest sheet of mica or glass is completely impervious to the passage of the emanation. In this and all other respects in which it has been examined the emanation behaves like a gas distributed in infinitesimal amount through the atmosphere which carries it, each particle emitting rays on its own account and acting as a centre of ionisation in the atmosphere.

The "emanating power," or ability to give the radio-active emanation, is proportional to the quantity of the thorium compound employed, whereas the radiating power is proportional to the surface exposed. Thus, in very thin layers the ionisation is caused chiefly by the radiation, and as the layer is increased in thickness the additional ionisation due to the emanation is increased. Hence, in the former case a blast of air has little effect on the value of the radio-activity. Thin layers of thorium preparations are in this respect little different from those of uranium. With thicker layers the difference is, however, very marked, and the value of the radio-activity is greatly affected by slight currents of air. All these phenomena are quite independent of the nature of the atmosphere employed.

The properties of the radio-active emanation itself will be first considered, without reference to the manner in which it comes to be produced by the thorium. The experiments quoted show that thorium transmits to the air an ionising agent with the properties of a gas, but the question remains open as to whether the ionisation is produced by *rays* of the kind which constitute radio-activity. This is a difficult point to establish experimentally in the case of the thorium emanation, and was accomplished by Rutherford in the following manner. A vessel was constructed through which the thorium emanation could be led by a current of air. In one side of the vessel was a window of the thinnest mica sheet, which, although gas-tight and impervious to the passage of the emanation, was thin enough to allow a sufficient fraction of α-radiation of ordinary penetrating power to pass through unabsorbed. The effects of the imparted activity (*q.v.*) were eliminated by keeping the vessel positively charged with respect to an internal electrode placed out of the direct line of fire to the mica window. In this way any imparted activity, produced by the emanation, was concentrated on the negative electrode, the radiations from which were unable to penetrate to the outside of the vessel. Under these circumstances the outside air in the line of fire from the mica window was found to be ionised by rays which could only come from the gaseous emanation inside the vessel. Rutherford proved with this arrangement that the rays from the thorium emanation consist entirely of the easily-absorbed and difficultly-deviable α-rays, and no detectable amount of β-rays are present. This experiment, difficult for the thorium emanation, has been shown in the simplest manner for the millionfold more powerful radium emanation (*q.v.*). The emanation particles do not appear to move appreciably under the action of an electric field, and Rutherford concluded that they are uncharged. Our present knowledge shows that this result proves, rather, that if the emanation particle is charged it can give up its charge without losing either of its essential characteristics—viz., its volatility or radio-activity. The point will be again referred to when the actinium emanation is dealt with.

Rate of Decay of the Thorium Emanation.—If the emanation is blown away from the thorium by a blast of air into a metal cylinder (which can be closed air-tight) provided with a central insulated wire, the duration of its radio-activity can be investigated by measuring the ionisation current between the central wire and the cylinder, after the air stream is stopped and the cylinder closed. In this way Rutherford showed that the radio-activity of the emanation, after separation from the thorium compound, decays in a geometrical progression with the time, and falls to half-value in about one minute. This is expressed by the equation $I_t/I_0 = e^{-\lambda t}$, in which the radio-active constant λ has a value of 1/87, or $1{\cdot}16 \times 10^{-2}$. In 10 minutes after the removal of the emanation from the thorium the radio-activity falls to one-thousandth of its original value, a quantity generally too small to be detected. In 20 minutes the activity is one-thousandth of the value at 10 minutes or one-millionth of the initial.

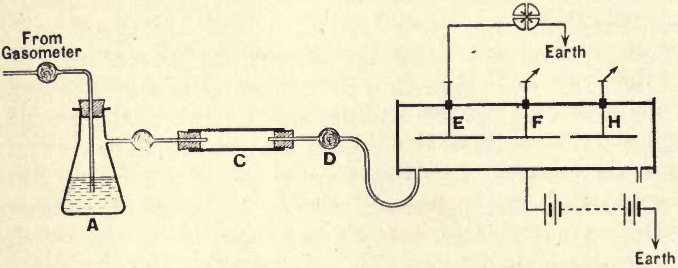

FIG. 28.

A convenient apparatus for the study of the thorium emanation is shown in Fig. 28. A known weight of the thorium compound to be tested is placed in C, and a steady current of air from a gas bag passed over it. The cotton wool in D removes from the air the ions caused by the action of the direct radiation. The emanation passes through the plug and causes the ionisation in the cylinder. With a uranium compound in C no current would be observed with this apparatus, for all the ions present in the gas would be removed on passing through D. With 10 grammes of ordinary thorium oxide in C, and a fairly rapid stream of air, a current between 10^{-10} and 10^{-11} ampere passes between the cylinder and the electrodes. The central electrode is divided into three separate parts, E, F, H, for the purpose of determining the rate of decay of the emanation. If the velocity of the gas stream is known, the ionisation currents respectively measured by connecting the three electrodes successively with the electrometer will afford a measure of the decay of activity suffered by the emanation in the time taken for the gas stream to pass from E to F and from F to H. If the gas stream is kept constant, the rates of decay of the activity of the emanations from different compounds of thorium, and from the preparations of thorium X, can be compared together. It is found that the rate of decay, and therefore the radio-active constant λ, is the same in all cases. In ordinary work when the rate of decay is not being investigated the three electrodes E, F, H, are usually connected together.

The thorium emanation was the first type of matter possessing temporary radio-activity to be recognised, and its nature had been studied pretty completely before thorium X was discovered. In this work (Rutherford and Soddy, *Phil. Mag.*, 1902, VI., 4, p. 569) the conclusion was drawn that it was a specific type of radio-active matter of a gaseous character in infinitesimal quantity. The latter presents no drawback to its accurate investigation, for its radio-activity affords adequate and convenient methods for its experimental study. Various experiments were performed on the action of temperature and chemical reagents on the emanation. The general result showed that it was completely unaffected by any of the means employed. It is not altered by passage through a tube of platinum raised to the temperature of bright white heat. It may be bubbled unchanged through any acid or reagent. It is not absorbed by passing through red-hot lead chromate, magnesium powder, zinc dust, &c., and in this respect it shows the same chemical inertness as the members of the argon family of gases. The latter, and the emanations of thorium and radium, are the only known gases which resist absorption by these reagents. The apparatus employed in these experiments was that already described (Fig. 28), the tubes containing the reagents used being placed in the gas stream between D and the testing cylinder. A steady stream of gas was maintained and the ionisation current measured with the tubes hot and cold. Since the value of the ionisation current was not altered by this treatment, it not only proves that the emanation is not absorbed, but also that the rate of decay of the activity of the emanation is the same throughout, and is not affected either by temperature or by chemical reagents.

Some experiments were performed by the Author with a view to determine if the presence of the air were necessary for the production of the emanation by thorium. An apparatus for producing hydrogen and oxygen by the electrolysis of dilute sulphuric acid was constructed, and the gas passed over a thorium compound. All the joints were of sealed glass, so that there was no possibility of air leaking into the apparatus. Over a thousand litres of hydrogen were passed over the thorium compound, without opening the apparatus to the air, in an uninterrupted stream lasting for three months. At the end of this period the amount of emanation carried over by the gas stream was no less than at the beginning. Hence the emanation is not, for example, one of the inert constituents of the atmosphere rendered radio-active by contact with thorium, and there is no escape from the conclusion that the emanation is a specific type of matter of a new kind derived from the element thorium. This conclusion was confirmed by an investigation of the behaviour of the emanations of thorium and radium at the low temperatures attainable by the use of liquid air (Rutherford and Soddy, *Phil. Mag.*, 1903, VI., 5, p. 561). It was found that the emanations of both thorium and radium were completely condensed if passed through a tube cooled with liquid air. The gas stream used to convey it passes on completely freed from the emanation. If the tube is allowed to warm up, the condensed emanation again volatilises unchanged (compare Chapter X.).

Now, the experiments already recorded on thorium X show that the emanation is not produced by thorium directly, for when freed from thorium X the thorium possesses no emanating power at all at first, but gradually recovers it. The thorium X, on the other hand, possesses the full emanating power of the original substance from which it was separated, and gradually loses its power as the thorium recovers it.

Without, at the present stage, considering more fully the nature of these phenomena, a simple explanation may be developed on the lines laid down in the discussion of thorium X.

It is clear that, just as thorium X must be considered as the product of the radio-active change of thorium, so the emanation in turn must be regarded as the product of a second change of a similar kind suffered by thorium X. Thorium is changing into thorium X, and the latter, in turn, is changing into the emanation. A chemical separation is necessary to effect the removal of thorium X from thorium, because both types of matter are non-volatile. But to remove the emanation no such operation is needed, because in this case the matter is gaseous in character, and separates itself from the matter producing it by ordinary gaseous diffusion.

Imparted Radio-activity.—The most remarkable property of the emanation is its power to impart temporary radio-activity of a specific kind to solid bodies with which it comes into contact (Rutherford, *Phil. Mag.*, 1900, V., 49, p. 161).

If a thorium compound is kept in a closed vessel for several hours, so that the emanation is retained and prevented from diffusing away, it is found that the interior of the vessel, after removal of the thorium, is itself strongly radio-active. The character of the rays from this imparted radio-activity is distinct from that of the thorium itself, being more penetrating (compare Fig. 20). The decay of this imparted radio-activity follows the usual law, decreasing in a G. P with the time, and reaches half-value after 11 hours. λ, the radio-active constant for this case, is, therefore, $1{\cdot}7 \times 10^{-5}$. All objects become radio active in this manner independent of their nature, and the imparted radio-activity is of the same character. Moreover, if the surface of the body made active is scrubbed with sand paper, the activity is to a large extent removed and transferred to the sandpaper. Rutherford found that certain acids were capable of dissolving off the matter causing the radio-activity from a platinum wire rendered active by the thorium emanation. Hydrochloric acid and sulphuric acid especially possessed this power, whereas nitric acid and alkalies showed very little effect. The activity of the matter removed is in no case destroyed. Thus, if the hydrochloric acid used be evaporated in a platinum dish, the activity removed from the wire is left behind on the dish after the evaporation of the acid. By evaporating aliquot portions of the acid at definite intervals of time, Rutherford proved that the activity of the matter causing the phenomenon decayed at the same rate when dissolved in the acid as it does when kept on the wire on which it was originally deposited. As in the other cases, the matter causing these phenomena is in otherwise undetectable quantity. The film produced is quite invisible and unweighable.

An interesting feature experimentally distinguishing the type of active matter causing the imparted radio-activity of thorium is its behaviour in an electric field. If a central electrode is kept negatively charged with respect to the vessel, the imparted activity deposited inside the vessel from the emanation is confined solely to the negative electrode. The matter causing the imparted activity at the moment of its production carries a positive charge and travels in a field to the negative electrode. In this way it is possible to concentrate the whole of the imparted activity on a fine wire, so that the latter is, weight for weight, many hundred times more active than the original thorium compound. Under these circumstances the slightest gain in weight of the wire could be observed, but none has been detected. The action of the field does not in any way affect the total amount of radio-activity imparted by the emanation, but merely alters the distribution. For a fuller consideration of this phenomenon, see Rutherford, *Phil. Mag.*, 1903, VI., 5, p. 95.

The interpretation of the foregoing facts follows without difficulty on the explanation adopted in the other cases. The deposition of a film of matter by the emanation on solids, the imparted matter again possessing a specific kind of temporary radio-activity, obviously points to a third radioactive change, the gaseous emanation changing in turn into a non-volatile type of matter which settles down out of the gas upon whatever provides it a resting-place. Being positively charged at the moment of production, it will move in a field and be directed to the negative electrode exclusively. The possession of this charge, no doubt, assists its deposition, even when no field is acting, by causing it to be attracted to uncharged objects.

Analysis of the radio-activity of the element thorium has thus shown that it is made up of at least four parts, due to four specifically distinct types of matter—the parent thorium and three new types continuously being produced from it by successive change :

Thorium
↓
Thorium X
↓
Emanation
↓
Matter causing the
imparted radio-activity.

It must be admitted that a series of changes of this kind would be extremely remarkable if they were considered to be due to ordinary molecular or chemical change. The absolute dissimilarity of the successive products in chemical and physical nature, the matter in question passing from a non-volatile state to the gaseous condition and again back into the non-volatile form, is, however, what is to be expected of a sub-atomic transformation. If radio-active change consisted in the successive degradations of a heavy atom into lighter ones, unless it happened that all the products were members of the same family of elements in the periodic table, it is to be expected that the successive elements resulting would exhibit the widest variation in their nature. The fact that some families of elements increase in volatility with increasing atomic mass (for example, the series lithium-caesium, magnesium-mercury), while others (oxygen-tellurium, fluorine-iodine) follow the opposite course, together with the existence of very light elements which are extremely non-volatile—for example, carbon and silicon—shows that nothing can be predicted of the physical nature of the products resulting from the break up of a heavy atom unless they chance to belong to the same family in the periodic table.

CHAPTER VIII.

THE THEORY OF ATOMIC DISINTEGRATION.

Radiations remain Unaltered in Character during Decay and Recovery.—Relation between Radiation and Continuous Production of New Matter.—Both Rays and New Matter Result from Parent Element by the Same Change.—Radio-activity Measures the Number of Atoms Changing.—Non-separable Activity.—Rate of Decay a Measure of Rate of Change.—λN Atoms Change per Second when N are Present.—Mono-molecular Types of Change.—Disintegration.—Conservation of Radio-activity.—Ultimate Products of Radio-active Change.—Prediction with regard to Helium.—Cause of the Initial Irregularities in the Curves of Decay and Recovery of Thorium X and Thorium.—De-emanation.—Due to Alteration of Rate of Escape of Emanation.—Rate of Production Constant as Theory requires.—Disturbing Effects of Subsequent Changes.—General Résumé of the Theory of Atomic Disintegration.—Latent Energy of Atomic Structure.—Explosive Character of the Individual Disintegrations.—Metabolons.—Infinitesimal Quantity of the Transition-forms.—Unstable Elements.—Criteria of Identification—Average Life.—Reason of the Stability of the Elements.

In preceding chapters the products of radio-active change have been studied by their radio-activity, which has been used as the means by which infinitesimal quantities of matter have been brought within range of experimental investigation. No enquiry has, so far, been pursued as to the nature of radio-activity except that it has been shown to be for the most part maintained by the continuous production of fresh active matter. It remains to frame a consistent physical explanation of the nature of radio-activity itself (Rutherford and Soddy, *Phil. Mag.*, 1903, VI., 5, p. 576). The discussion in Chapter V. of the nature of the α and β radiations assists us materially in this task. Evidence was then brought forward to show that the α-rays are caused by the radial expulsion from the active substance of positively-charged particles of atomic dimensions (Rutherford), while the β-rays are high-velocity cathode rays, or negatively-charged particles of sub-atomic dimensions (J. J. Thomson and Becquerel). Now, in the changes suffered by radio-activity during the processes of decay and recovery that have been considered the character of the radiation is not sensibly affected. Hence we must regard these changes in radio-activity as being caused by alterations in the number of particles projected as rays in unit time. This consideration is important, for it precludes the view that the decay of the activity of thorium X, for example, is analogous to the decay of the radiations from a hot body cooling. The character of the particles projected remains unaltered, and the weakening of the activity is caused by a diminution in the number emitted in unit time. Similarly, the only distinction between the radio-activity of a powerfully active element like radium and a feebly active element like thorium is that a greater number of radiant particles are expelled per unit-weight in unit time. Considered from this point of view the emanation of thorium, thorium X, &c., are far more powerfully active substances than radium. We have seen that, in the case of thorium, thorium X, the emanation, and the matter causing the imparted activity, each type of matter not only expels rays, but produces the next new type of matter also. So radio-activity will be completely defined as the simultaneous occurrence of two processes: (1) The expulsion of charged particles with enormous velocity; (2) the production of new types of matter in minute amount, which can be investigated when they also are radio-active. What is the precise connection between these two processes? Thorium X furnishes a convenient example, for (1) the expulsion of rays can be determined by measuring its radio-activity, (2) the production of new matter, by measuring its emanating power. It was found (Rutherford and Soddy, *Phil. Mag.*, 1902, VI., 4, p. 579) that, when one part of thorium X is kept in solution and its *emanating power* tested from day to day, it decayed with time according to the same law and at the same rate as the *radiations* from another part of the same sample that had been evaporated to dryness in a platinum dish. This points to the conclusion that the two processes

(1) and (2) are causally connected. Another example, giving the same result, is to be found in the first paper of Rutherford (*Phil. Mag.*, 1900, V., 49, p. 170) on the imparted activity of thorium. Here we have (1) the radiation from the radio-active emanation, (2) the production of the new matter which causes the imparted activity. A steady current of air was blown over a thorium compound into the tube shown in Fig. 29 in the direction of the arrows for several hours continuously. At the commencement the ionisation current due to the rays from the emanation was measured by connecting the four electrodes A, B, C, D separately with the electrometer, the exterior of the tube being connected with the + pole of a battery giving 300 volts, the other pole of which was earthed. The decay of activity suffered by the emanation in the time required for its passage through the tube caused the current to the electrode A to be greater than to B, and the ratio of the currents due to the emanation at the four electrodes was determined.

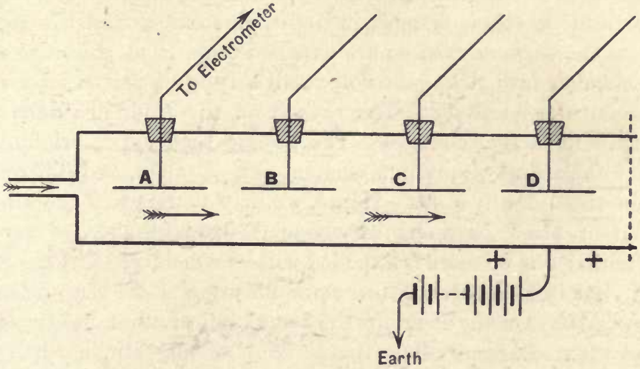

FIG. 29.

With the above arrangement the whole of the imparted activity produced by the thorium emanation is deposited on the electrodes, which are negatively charged with respect to the tube. After the emanation had been kept passing for several hours the electrodes were removed from the apparatus and the imparted radio-activity on each determined. It was found that the four electrodes exhibited substantially the same ratio in their imparted activities as in the determination of the current due to the emanation. That is to say, as the radio-activity of the emanation decays with time, its power to impart radio-activity to its surroundings correspondingly diminishes. The number of particles projected as rays from the emanation during its change into the new matter causing the imparted activity is proportional to the amount of the latter (as measured by its radio-activity) produced.

It follows, therefore, that both the projected particles, or rays, and the new matter are products of the parent type of matter by the *same* change. The rays constituting the radio-activity of thorium are expelled at the moment the atom of thorium changes into thorium X, the rays constituting the radio-activity of thorium X are expelled at the moment the thorium X atom changes into the emanation, and the rays from the emanation atom are expelled from it at the moment it changes into the again non-volatile type of matter causing the imparted activity. The rays from the latter are derived from still further changes. The atomic theory, put briefly, is that the atoms of any one element are all alike, and different from those of any other. Hence, we may assume that the same kind of atom changing expels some fixed number of rays. Whether one or more is expelled will be considered in Chapter XI., but it is immaterial to the present argument. The number must always be the same for the same kind of atom undergoing the same change. Thus the activity of any single type of radio-active matter, in the sense of the number of particles expelled from it in unit time, furnishes a measure of the number of atoms undergoing change.

Several important and definite conclusions follow from this point of view, which have in many cases been found capable of direct experimental test. In the first place, since the production of thorium X from thorium proceeds at a constant rate in any given mass of thorium compound without reference to the physical and chemical conditions (see p. 93), it follows that a definite fraction of the total number of thorium atoms are changing in each unit of time into thorium X, and this change is accompanied by the projection into space of a definite number of α particles. Hence, to whatever chemical process thorium is subjected, it should prove impossible to entirely free it from radio-activity—that is to say, there must exist a certain constant non-separable radio-activity. Now this has been shown to be true, so far as our experimental knowledge goes, not only of thorium, but also of uranium and radium, and in each case it is important to note that the non-separable activity consists entirely of α-rays. The next deduction from the position that the expulsion of the rays accompanies the change of the atom is that radio-active matter must be continuously diminishing in quantity. A radio-active atom is a changing atom. For feeble examples, like uranium and thorium, it is not necessary that the change should be great enough to be appreciable. It is probable that in these cases the quantity in existence remains practically constant over very long epochs of time. But if this is true, it follows equally that the actual quantity of uranium X and thorium X produced from them must also be excessively small. In order that this insignificant amount of matter should be able to give sufficient rays to be appreciated it must be changing much faster than the original element, and its quantity in consequence must rapidly diminish. Hence its radio-activity decays, and the rate of decay, together with the intensity of the radio-activity of unit quantity, depend upon whether it is changing slowly or rapidly.

The radio-activity of an intensely active substance must quickly decay. On the general assumption that the order of the total quantity of energy liberated in the form of rays may be expected to be similar for each type of radio-active atom, the argument may be inverted. The more rapidly the activity decays the more intensely radio-active (considered with reference to unit-weight) must the type of matter be. Marckwald's work on polonium (Chapter II.) is an example of this principle. We have seen that the activities of uranium X, thorium X, the emanation of thorium, and the matter imparted by the latter to other objects, all decay in a G.P. with the time at characteristic rates, represented by the equation (p. 92)

$$\frac{I_t}{I_0}=e^{-\lambda t},$$

in which λ is a numerical constant different for each type of matter. The expression $\frac{I_t}{I_0}$ represents the ratio of the number of rays expelled in unit time for any time t to the number expelled initially, and may be replaced by $\frac{n}{n_0}$, where n_t represents the number of atoms of the substance changing in unit time, for time t, and n_0 the number initially, so that

$$\frac{n}{n_0}=e^{-\lambda t}.$$

To find the rate of change of the original system, let N_0 = the total number of atoms originally present and N_t the number left unchanged at time t,

$$N_t=\int_t^\infty n_t \,.\, dt=\frac{n_0}{\lambda}e^{-\lambda t},$$

$$N_0=N_t \text{ when } t=0,$$

$$N_0=\frac{n_0}{\lambda},$$

$$\frac{N_t}{N_0}=e^{-\lambda t}.$$

Differentiating, $$\frac{dN}{dt}=-\lambda N.$$

The result is thus a very simple one. On the view that the number of rays expelled is a measure of the number of atoms of the system undergoing change, the geometrical decrease of the radio-activity is due to the geometrical decrease of the number of atoms of the changing system. The rate of change of the atoms undergoing radio-active change is always proportional to the amount remaining unchanged. If N atoms of any type of radio-active matter are present, λN change per second, where λ is the radio-active constant. From the values of λ already given it follows that about $\frac{1}{87}$th of the thorium emanation, $\frac{1}{500000}$th of thorium X, and $\frac{1}{3000000}$th of uranium X undergoes change per second.

Now, the law of radio-active change just developed is already well known to us as the law followed by a particular kind of chemical reaction—viz., that known as mono-molecular. In reactions which are of the type of the decomposition of the single molecule into simpler parts the quantity of the substance undergoing change under constant conditions decreases in a geometrical progression with the time, owing to the amount changing in unit-time being a constant fraction of the amount present. Both are simple consequences of the law of probabilities, the number of changes occurring under constant conditions increasing with the number of changing systems. But the case is completely different when the reaction is not of the type of a decomposition but of a combination. For here, in order for the change to occur, the combining molecules or atoms must first approach within each other's spheres of influence. The rate of change, in consequence, depends upon the concentration, and this diminishes with the progress of the change. The rate of change proceeds in these cases according to some power of the number of changing systems present. Hence, radio-active change cannot be of the nature of a combination together, or, as a chemist would say, the polymerisation, of the atoms of the active element, but must be due to their decomposition or disintegration. The term *changing atom* can now be logically replaced by the more definite conception expressed by the use of the term *disintegrating atom*, with considerable advantage to the clearness of the mental picture conveyed.

The Conservation of Radio-activity.—The simple law of radio-active change—that in N atoms of any type of radio-active matter λN disintegrate per second, where λ is a constant characteristic for the type of matter considered and unalterable by any known agency—leads to some general deductions of a far-reaching character. Radio-activity is derived either (1) from the slow disintegration of some "permanent" radio-element—*i.e.*, an element changing excessively slowly, so that the amount does not perceptibly alter from year to year; or (2) the more rapid disintegration of transition-forms of matter which are maintained in existence because they are continually being reproduced by (1). The radio-activity due to (1) must be constant over ordinary periods of time. With regard to (2), a little consideration shows that the quantity of each transition-form in existence is also constant, and must bear some fixed relation to the quantity of the parent element, for it is that quantity at which so much has accumulated that the quantity breaking up per second exactly balances the constant quantity produced per second. Therefore, each transition-form contributes a fixed proportion of the radio-activity, so that the sum total of radio-activity is a constant. All that is effected by the chemical separations that have been discussed is to remove one or more of the transition forms. But the sum of the activities is still the same as before separation. Hence, if the radio-activity of a preparation increases with lapse of time, this of itself is evidence that during its preparation a transition-form has been separated from it, and if looked for it will be found that the activity of the transition-form steadily decays concomitantly with the steady rise of activity of the preparation. This is the principle of the conservation of radio-activity, and holds true under all conditions that have yet been investigated. If it were otherwise, the value of λ would suffer alteration, and this is contrary to experience, so far as our present experimental knowledge is concerned. After very long periods, probably comparable with the age of the earth, the radio-activity of any given quantity of one of the parent radio-elements, uranium or thorium for example, must diminish, it is true, unless there is some upward process of evolution not yet recognised which is re-forming the heavy elements out of the lighter ones, and so maintaining their quantity. But the principle of conservation applies rigidly only when some fixed amount of radio-element is considered, and this is spontaneously growing less. The proportionate change in these cases is so small that it may be neglected for ordinary periods of time.

Ultimate Products.—The question arises, since all radio-active matter must be changing, What are the final products resulting from the disintegration of the radio-elements when they at length assume a stable—*i.e.*, a non-radio-active—form? Obviously no evidence can be obtained in this direction by direct experiment, at least for the feebly-active elements uranium and thorium, except by operating with very large quantities over long periods. This has, fortunately, been done for us by Nature in the minerals containing these elements, for we must suppose, having no evidence to the contrary, that the changes have been proceeding continuously over geological epochs in these elements as they occur in the crust of the earth, and that, therefore, the ultimate products of the changes tend to accumulate in the radio-active minerals in quantities sufficient to be detected by ordinary chemical means. Under favourable circumstances they should appear in the minerals as invariable companions of the radio-elements. An examination of the minerals containing uranium and thorium, many of which, as we now know, contain radium also, shows that there are usually present a very large number of elements in larger or smaller quantities. The frequent presence of one—viz., helium—is, however, very remarkable. When Sir William Ramsay discovered this element for the first time in terrestrial sources, he drew attention to the curious fact that it only occurs in those minerals which contain uranium or thorium—*i.e.*, as we know now, are radio-active. On this account the case of helium is very suggestive. For the element belongs to the family of inert gases which are incapable of entering into chemical combination, and, once separated from the mineral, by heat or solution, cannot be made to re-combine with it again. If, however, helium were liberated by radio-active change, through the mass of the substance, it is possible that it might be mechanically prevented from escaping. Rutherford and Soddy discovered (*Phil. Mag.*, 1903, VI., 5, p. 453) that this is the case with the gaseous emanations of thorium and radium which, under certain circumstances, remain stored up in the compound producing them, and do not escape. They suggested that helium might be a disintegration product of one of the radio-elements. Thanks to the great advances made of recent years in gas manipulation by Sir William Ramsay and his colleagues in their work on the rare gases of the atmosphere, which necessitated the handling of vanishingly small quantities of gases, this prediction has been verified experimentally in the case of radium (p. 157).

We are now in a position to study somewhat more exactly than was before possible the nature of the radio-active changes occurring in thorium. Owing to its complex nature, the parent element producing in turn several products, each of which is radio-active, the simple laws arrived at by considering each change by itself, without reference to the effect of the subsequent changes, are not often strictly applicable without modification. Thus, the experimental curves obtained (Fig. 30, reproduced from Fig. 25) in the determinations of the rate of decay of the activity of thorium X, and the rate of recovery of the activity of thorium, exhibit for the first two days divergencies from the normal course afterwards followed. These are at once capable of explanation if the other products of the change besides thorium X —viz., the emanation and the matter causing the imparted activity—are taken into account. Before the separation of thorium X, the activity of a thorium compound is due to the several activities of (1) thorium, (2) thorium X, (3) emanation, (4) matter causing imparted activity. By the chemical operation (2) is separated, leaving (1), (3), (4).

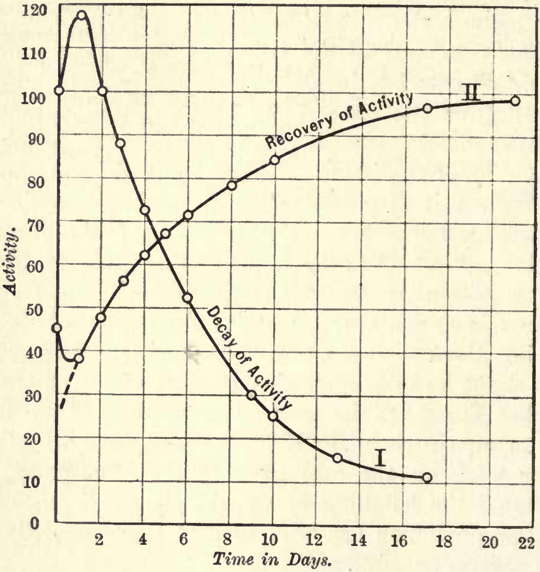

FIG. 30.

The emanation (3), being very short-lived, loses its activity within a few minutes after the separation, and need not be practically considered. But (4), the matter causing imparted activity, requires 11 hours for the activity to fall to half value. Hence the recovery of activity of the thorium, due to the regeneration of thorium X, will be offset by the simultaneous decay of the activity of the matter causing the imparted activity. Thus the activity of the thorium compound does not commence to recover at once, but at first diminishes owing to the decay of the imparted activity present. The truth of this conclusion was tested by separating thorium X a great many times at short intervals from the thorium, so as to give the imparted activity produced by the thorium X, originally present before the first separation, time to completely decay. Under these circumstances, it is found that the recovery curve commences to rise immediately in the normal manner from a 25 per cent. minimum, and the initial irregularity is completely absent. The same considerations hold good for the decay curve of thorium X. At first the imparted activity produced in the further change of thorium X causes the activity of the latter to increase, until the point is reached in about two days at which the amount of imparted activity produced balances its decay with time. Then the augmentation of the activity stops, and the steady decay of the activity of thorium X, to half-value in four days, proceeds after the second day according to the simple geometrical progression.

De-emanation.—Another point which will repay discussion before leaving the element thorium, is the great variation in the emanating powers of different compounds, and of the same compound under different conditions. For the law that λN atoms change when N atoms are present necessitates equally, with a definite and unalterable radio-activity, a correspondingly definite rate of production of the emanation. Now, although the radio-activity of thorium compounds is proportional to the quantity present, and is not affected by chemical or physical means, it is found that the emanating power of a compound often bears little relation to the amount of thorium contained in it. Certain solid compounds, like the oxide, but more particularly the hydroxide and carbonate, give off a large amount of emanation, while others, like the nitrate, oxalate, &c., give off very little indeed. It was found however, that, when dissolved in aqueous solutions, all compounds of thorium are equally effective in this respect, and the emanating power may be taken to be at its maximum in solution. Rutherford noticed, in his original observations of the emanating power of thorium, that if the oxide is intensely heated the emanating power is permanently reduced to a few per cent. of its original value, and preparations so treated are frequently referred to as "de-emanated." But if the de-emanated oxide is subjected to a series of chemical processes, and again converted into a soluble compound, it is found that the normal maximum emanating power is possessed by the solution, showing that the phenomenon of de-emanation is to be associated with a particular physical state. The emanating power of a given thorium preparation is much affected by temperature, being very small at temperatures of about −100°C., and being increased by rising temperature up to the point—about a red-heat—at which the process of de-emanation sets in The power of a thorium compound to impart activity to its surroundings varies correspondingly with its emanating power.

All these variations are satisfactorily accounted for by the view that the *rate of production* of the emanation is, as theory requires, constant and proportional to the quantity of thorium (or more strictly of thorium X) present, but that the *rate of escape* of the gaseous emanation from the compound producing it is much affected by temperature, solution, and the chemical and physical state of the compound. De-emanation, for example, is brought about by an alteration in the physical state of the oxide, which retards the escape into the surrounding atmosphere of the emanation, which is formed uniformly through the mass of the compound. Owing to the rapid decay of the activity of the thorium emanation, a relatively slight decrease in its rate of escape will greatly affect the value of the observed emanating power. In consequence, the radio-activity of the emanation appears as a part of that of the solid, and the imparted activity it produces is retained in the compound instead of being dissipated among surrounding bodies. Hence the *radio-activity* of a highly emanating compound like thorium hydroxide should be increased by strong ignition, and the increase should reach a maximum about two days after the process, when the imparted activity produced by the imprisoned emanation obtains its equilibrium-value. This was found to be the case. The activity of a sample of thorium hydroxide rises slowly during the first two days after ignition to a maximum about 120 per cent. of the former value. The converse experiment is to take a non-emanating compound and suddenly convert it into a highly-emanating one. This was accomplished by keeping the highly-emanating hydroxide in liquid air for some days, so that the emanation is condensed and does not escape, and its radio-activity and the imparted activity it produces is added on to the activity of the compound. On removing it from the liquid air and allowing it to warm up the activity slowly decays about 20 per cent. during the first few days, owing to the emanation now being free to escape from the system and to carry away the imparted activity to surrounding objects.

No simple general treatment is here possible of the disturbing effects of subsequent changes. The following cases may be qualitatively recognised. If a body, A, changes into B, and B changes into C more rapidly than A into B, the decay curve of A will show a preliminary increase in activity, and then a decrease to zero at the rate of change of A into B. If, however, B changes into C at a slower rate than A into B, the decay curve will exhibit a normal course down to a certain point, and, if this point is calculated as zero, the rate of change may be considered without error as the true rate of change of A into B. From this point the activity decays to zero at a rate slower than before—viz., that of B into C. These cases are complicated experimentally when one of the stages consists of a gaseous type of matter, like the emanation, but each case can be worked out for itself from first principles. Further very instructive illustrations will be considered in the case of radium. It has been assumed in the above cases that α-rays are expelled at each change.

In the above consideration of the nature of radio-active change, for the sake of simplicity and to avoid confusion, the case of thorium has been taken, this being the element for which the theory was first worked out. But it applies equally to every case, so far as is at present known, and the treatment of the remaining cases is much simplified by the consistent application of the theory.

It will be convenient to conclude the chapter by a summary in general terms of the nature of the processes that the study of the radio-elements has revealed.

Instead of regarding each atom as a constant source of energy, in the form of rays, the disintegration theory regards the property as due to a fixed proportion of the total number of atoms which are undergoing disintegration. During the instant of their disruption they fling away fragments of themselves into space in the form of radiant particles. Radio-activity, being thus a property of a *fixed proportion* of the active matter in each instant, behaves in many ways as an atomic property contributed equally by all the atoms. The vast majority are for the time being ordinary inactive atoms with a specific and characteristic material nature which is not affected by, or does not exert any influence on, the character of the additional set of properties contributed by the fraction disintegrating. The dual character of the properties of the radio-elements is thus simply explained.

The disruption of the individual atom is a sudden and explosive process, but differs from that of ordinary explosion in the fact that the explosion of an atom exerts no influence on the rate of explosion of its neighbours. In an explosive substance—as, for example, fulminate of mercury or acetylene—the disturbance occasioned by the disintegration of a single molecule is the *cause of the explosion* of the surrounding molecules, so that the whole mass detonates in a very short interval of time. In the case of the disintegrating atoms the cause of the disintegration is at present unknown. It proceeds at a definite rate, a fixed fraction of the total atoms disintegrating in the unit of time, without hindrance or acceleration by any agency known. Radio-activity and the processes that give rise to it appear to lie totally outside the sphere of known molecular forces. No change in the chemical and physical properties of the atom appears to occur as it approaches the termination of its existence as a separate entity. Suddenly, and without previous indication, it flies to pieces by some internal cataclysm, the cause of which we can only conjecture. The vast store of energy bound up with its internal structure, and which make it a stable system to all the relatively insignificant forms of energy manifested in an ordinary chemical and physical change, are revealed when the structure flies to pieces. The internal energy of the chemical atom becomes for the first time knowable when it disintegrates. Because the atom has never in any other process been observed to change, and because it does change in exhibiting radio-activity, we have the explanation of the fact that the energy associated with matter in the rays of the radio-elements is of a higher order of magnitude than has ever been dealt with in science before. Radio-activity thus draws its supply of energy from a previously untapped reservoir—the latent energy associated with the atomic structure.

Atomic disintegration would have been a difficult process to establish by direct experiment as the cause of radio-activity had the phenomenon not been somewhat complex. The complexity of radio-activity, especially in the case of the element thorium, made it possible that so far-reaching a conception as the one put forward should be subjected to rigorous experimental verification. For if the disintegrating atom passed from its initial to its final state in one change, as in the analogous molecular cases of explosion, it would have been difficult to have obtained much knowledge of the process. Fortunately, this is not the case. The thorium atom only passes to its final state after at least five separate and successive disintegrations, each of which, considered by itself, is suddenly and explosively consummated, so far as the individual atom is concerned, by the radial expulsion of fragments in the form of α particles. In consequence, there exist a certain number of short-lived transition-forms of matter intermediate between the initial and the final atoms which result from the process. The term "Metabolon" has been proposed by Rutherford and Soddy to denote unstable atoms of this character, and the name indicates *change*, the essential feature which characterises them, and the reason why they come within the range of our methods of investigation. A metabolon is an atom with a limited life. While it exists it is a normal atom, possessing the ordinary attributes of matter. At the moment of its disruptive change it exhibits the property of radio-activity.

Situated as we are, the observers for a short time only of a process of evolution which has been going on for indefinite ages, our knowledge of the unstable atoms is necessarily limited by certain definite considerations. In the first place, since the process is not affected by any known agency, we must assume that it has proceeded continuously during past ages at the same rate, for any given type of matter, as it is proceeding at present. Hence it follows that the initial process of disintegration must be excessively slow, in order that some of the matter disintegrating should survive to the present day. The initial type of matter will be termed the parent-element. For any ordinary quantity of parent-element, considered over any ordinary period of time, the actual quantity of the transition-forms of matter resulting from the disintegration must be excessively minute. The quantity of the transition-form produced, moreover, cannot accumulate beyond a certain limiting or equilibrium value, which is attained when the amount changing per second equals the amount produced per second, which is the condition of radio-active equilibrium. Since the rate of production is slow, and the rate of change of the transition-form is rapid, the equilibrium quantity must always be practically infinitesimal, and quite below that needed for detection by ordinary methods. But the rapid rate of change of the transition-form, and the expulsion of α particles which accompanies the change, afford the means whereby these transition-forms can readily be detected and studied, although only present in so small quantity that no other evidence of their existence is manifested.

Radio-activity, as the manifestation of atomic disintegration, has thus introduced us to a whole series of new unstable elements which at present find no place in the periodic table, and of which, except in the single case of radium, we posses but little chemical knowledge. After the initial stage is passed the evolution proceeds rapidly, and passes from stage to stage so quickly that almost all we know at present of the intermediate forms is derived from the energy-phenomena which mark their appearance and destruction. Yet these phenomena—viz., the expulsion of rays—are so characteristic and so fitted for exact study that our knowledge is by no means necessarily scanty, although it is of a kind not usually associated with ordinary matter. This appears at once when the means of identifying and distinguishing the different types of metabolons from one another are compared with those employed in the case of the stable atoms. For the latter, atomic weight, spectrum reaction, and general chemical and physical nature are the distinctive criteria employed. In the case of the metabolons, physical and chemical nature can sometimes be fairly completely studied. But the most valuable criterion, which is universally applicable, is the rate of change as defined by the radio-active constant λ, the proportion of the total changing per second.

A relation perhaps more generally useful than the radio-active constant is its reciprocal $1/\lambda$, which has a very interesting physical significance. It represents the *average life* of the metabolon in seconds, and affords a more concrete mental picture than the rate of change. Thus, for the thorium emanation the average life of the metabolon is 87 seconds, for the radium emanation 5·3 days (see Appendix, Chapter IX.) The average life of a metabolon may be compared with the atomic weight in the case of a stable atom as a constant well suited for its experimental identification. It may be pointed out that the actual life of the different atoms of the same unstable element has all values between zero and infinity. Some break up during the first second of existence, and, since only a fraction of the total changes per second, the quantity is, theoretically, never reduced to zero, and some persist indefinitely. This constitutes the first difference in properties between the individual atoms of the same element that has ever been discovered. It may be likened to the individual differences of velocity that exist between the molecules of a gas at constant temperature, according to the kinetic theory. This is important, as it suggests the question whether all atomic properties are not really average properties, the individual atoms continually passing with great rapidity through phases varying widely among themselves in chemical and physical nature.

Of all possible groupings of matter, the atoms of the periodic law probably represent only a selected number—viz., the forms with longest life—which exist to-day because they have survived a long process of evolution in which those physically unfit have disappeared. The transition-forms already spoken of represent, on the other hand, the elementary forms of matter physically unfit to survive, but which are brought within our powers of knowledge because they constitute the temporary halting places through which matter is passing in a scheme of slow continuous evolution from the heavier to the lighter forms. The original radio-active elements may be described as the connecting links between the two classes, partaking of the properties of each. They are changing, but so excessively slowly that some still survive. This gives us an insight into the reason of the stability of the elements. Matter has passed to its present position of apparent immutability by a long process of natural selection. The elements known to the chemist are stable *because* they exist and have survived. On the other hand, it is now possible to examine some excessively unstable forms of matter. Radio-activity might be defined as the science of the ephemeral elements.

CHAPTER IX.

THE RADIO-ACTIVE PROPERTIES OF RADIUM.

Intensity and Permanence of the Activity of Radium.—Chemical Actions of the Radium Rays.—Physiological Actions.—The Emanation of Radium.—The Imparted Activity of Radium.—Rate of Decay of the Emanation.—β-Radiations Produced in the Last Stages of the Disintegrations.—Summary of Radio-active Changes of Radium.—Evidence of the Complexity of the Changes giving rise to the Imparted Activity.—"Induced Activity of Radium with Slow Rate of Dissipation."—The Actinium Emanation.—Charge Carried by the Emanations.—Methods of Radio-active Analysis of Minerals.

APPENDIX.—Table I., Course of Disintegration of Uranium, Thorium, Radium, Actinium.—Table II., Radio-active Constants and the Values of the Average Life of the Unstable Elements.

The main interest attaching to radium is on account of the intensity and permanence of its radio-activity. We have seen that, according to the disintegration theory, the product of the intensity of the activity into the length of time it persists may be expected to be similar in all cases. The more intensely active any type of matter is the more rapidly will it tend to exhaustion, and therefore the intensely-active or rapidly-changing forms can only exist at the present time if they are being continuously reproduced in the slower change of a more permanent element. Radium occupies a very interesting position. Its rate of change is fast enough to cause its radio-activity to be very remarkable. The energy emission is so great that it is manifested in ways other than the direct radiation. The preparations continuously generate heat and produce marked chemical actions. On the other hand, the change is so slow that the substance does not measurably exhaust itself in a short term of years, and appears at first sight to be not changing at all. In brief, the position of radium is intermediate between that of a slow-changing parent element like uranium or thorium and a rapidly-changing transition-form like uranium X or thorium X. Its rate of change is just slow enough not to be obvious in a short period and yet sufficiently rapid for the change to be detectable by direct observation, as, for example, by the production of helium, if methods of the utmost delicacy are employed and a moderate lapse of time allowed. Or, to consider another aspect of the question, the rate of change is so rapid that only an infinitesimal quantity can accumulate in any mineral in which it is being produced, and yet is sufficiently slow that when, by the expenditure of infinite patience, as in Mme. Curie's research, tons of mineral are extracted, a sufficient weight can be accumulated to come within the range of ordinary chemical and spectroscopic methods of recognition. For we shall see as we proceed that it is as certain, in the present state of knowledge, that radium is being continuously reproduced in minerals containing it as that the emanation is being produced by radium, or helium by the emanation. The element has thus been the means of opening out an entirely fresh field of research, in which the requirements of the theory of atomic disintegration have been subjected to direct verification by ordinary physical and chemical methods.

Chemical Actions of the Radium Rays.—The radiations from radium are so much more powerful than those of uranium and thorium that they produce many effects not strong enough to be appreciable in the latter cases. They are able, for example, to bring about many chemical changes. The most important actions noticed are the conversion of oxygen into ozone (Demarçay), the conversion of yellow phosphorus into the red variety (Becquerel), the decomposition of water into hydrogen and oxygen (Giesel), and the colouration of glass violet or brown (Curie). In the conversion of oxygen to ozone and the decomposition of water a large absorption of energy occurs, and it was from evidence of this nature that the large amount of energy continuously emitted by radium was first recognised. A gramme of pure radium bromide in solution in water will evolve about 10 cubic cm. of mixed hydrogen and oxygen per day, which corresponds to the production of 20 gramme-calories of heat per day. This evolution goes on steadily month after month at constant rate. An unexplained point about this reaction is that there is always a slight excess of hydrogen over the composition required to re-form water. This has been variously accounted for, but at first seemed to be due to the oxygen being used up in effecting oxidations. In recent experiments, however, in which the possibility of this effect has been avoided, the excess of hydrogen still appeared. Giesel has stated that solutions of radium bromide generate bromine as well as hydrogen and oxygen, and this, if correct, would possibly account for the excess of hydrogen. But in the experiments mentioned no trace of bromine was detected, and the genesis of the excess hydrogen is still unelucidated.

There seems some doubt also about the actual production of ozone from oxygen. Certainly, in many cases it is not formed even where it might reasonably be expected. But there is no doubt that oxygen under the action of the rays from radium, or from the emanation, is extremely active and can effect many oxidations at ordinary temperatures which normally it is quite unable to do. Mercury is converted into the yellow oxide and carbon dioxide is produced if carbonaceous matter is present when the emanation is stored in an atmosphere of oxygen (Ramsay and Soddy, *Proc.* Roy. Soc., 1903, 72, p. 204). One may suppose that the oxygen molecule is dissociated by the rays, and the single atoms of oxygen effect these oxidations in a manner analogous to "nascent oxygen." If an atom and a molecule combined, ozone, O_3, would be produced, but this certainly does not always occur. The production of ozone, if real, may, therefore, be considered as merely a special effect of the general case—the formation of atomic oxygen. The effect of the rays on the photographic plate is, of course, to be classed under the chemical reactions of the rays. Various fluorescent substances, as barium platinocyanide and zinc-blende, are changed by prolonged action of the rays, and then become more or less insensitive. Willemite is probably one of the most lasting fluorescers we possess, and does not deteriorate appreciably even after long exposure.

Physiological Effects of the Radium Rays.—Powerful preparations of radium, if allowed to act at close quarters on the skin for a sufficiently prolonged period, produce ulcerated wounds which are very difficult to heal. The effect has not yet been exactly studied, but it would seem that the β-rays are mainly operative in producing this effect as ordinarily observed. On the other hand, the α-rays would probably be found to be even more harmful than the β-rays if precautions were taken against their being absorbed before reaching the skin. This is a condition not easy to observe, although it is possible that if the radium were enclosed in a celluloid capsule some of the emanation would find its way through and produce its α-rays in direct contact with the skin (see footnote below).

The most interesting physiological effect of the rays of radium is on the retina of the eye. If, after the eye has been rested in absolute darkness for some minutes, a quantity of a radium compound is brought suddenly near to the upper part of the forehead, a diffuse light seems to fill the back of the head even if the eyelids are closed and the radium is cased in lead or steel. This effect was discovered by Giesel, who explains it as being due to the fluorescence of the centre of the eye. Blind people with the retina uninjured experience the effect, but if the retina is diseased the effect is not obtained. The test may prove of some use in obtaining information as to the cause of blindness.

Radium Emanation.—Radium resembles thorium very closely in the character of its radio-active disintegration products. It gives a gaseous emanation possessing an almost completely similar physical and chemical nature to the thorium emanation, resisting absorption by all known reagents,* condensing and becoming non-volatile at low temperatures, and exhibiting to an even more marked degree than the thorium emanation the property of being retained by the dry solid compounds producing it, and of being liberated therefrom by heat and solution. It is distinguished by a much slower rate of change. Dorn (*Abk. der Naturforsch. Ges. für Halle,* 1900) first recognised the existence of the radium emanation by testing a sample of radio-active barium prepared from pitchblende in a manner similar to that already described by Rutherford for

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* M. and Mme. Curie have described some experiments which, when interpreted, seem to indicate that celluloid has the power of absorbing the emanation and retaining it. It would be interesting if further experiments could be carried out on this point.

thorium, and pointed out that its activity lasted a much longer time than in the case of the thorium emanation. It can still be detected several weeks after it has been separated from the radium producing it.

Owing to the comparatively slow rate of disappearance, the radium emanation, when it does not succeed in escaping, accumulates in the compound producing it to a much more marked extent than in the case of thorium. It has been shown that the amount of any transition-form capable of accumulating reaches an equilibrium value when the amount produced per second equals the amount disintegrating per second. By definition λ is the fraction of the total amount disintegrating per second. If N_0, represents the equilibrium quantity of emanation, and q_0 the quantity produced per second, $q_0 = \lambda N_0$, or $N_0/q_0 = 1/\lambda$. λ for the thorium emanation is 1/87, and for the radium emanation 1/463,000, or 6,000 times smaller. Hence, if compounds of radium and thorium are taken and kept under conditions in which the radio-active emanation does not escape (most conveniently in the form of solutions in stoppered vessels) for a sufficient length of time for the equilibrium point to be reached in each case, the maximum amount of radium emanation accumulating in the bottle will be 463,000 times the amount produced per second, while in the case of the thorium emanation the maximum amount will be only 87 times the amount produced per second. These relations have been quantitatively verified by actual experiments (Rutherford and Soddy, *Phil. Mag.*, 1903, VI., 5., p. 450).

Nearly all dry solid radium compounds give out very little emanation until they are warmed or dissolved, when there occurs a sudden evolution of the stored-up emanation several thousand times greater than that ordinarily given by the cold dry salt. If this is swept away by a current of gas or extracted by a mercury pump and stored, it will be found that the gas is spontaneously luminous in the dark and remains so for several weeks, the luminosity continuously diminishing till finally it completely disappears. Speaking more strictly, the luminosity is not due to the gas itself but to the fluorescence of the walls of the containing vessel. Once the emanation is extracted in this manner from any given sample of radium, no appreciable further quantity can be then obtained. As the activity of the gas extracted diminishes with lapse of time, a fresh crop of emanation is manufactured by the radium, the total radio-activity due to emanation being at all times constant. The explanation is the same as in the case of thorium and thorium X, and the rate of decay of the activity of the radium emanation is similar to that of thorium X, being reduced to half the initial value after the lapse of about 3·7 days. The radium emanation, although otherwise similar in general properties to the thorium emanation, escapes into the air with even less facility from solid compounds. In both cases, so far as we know, the emanation escapes instantaneously from the compounds in aqueous solution. The emanation is being continuously produced by radium at a constant rate, in exactly the same manner as thorium X is produced from thorium. It depends entirely on the physical conditions whether the emanation escapes or is accumulated. The amount *produced* is, however, always constant and independent of the conditions.

The Imparted Activity of Radium.—M. and Mme. Curie (*Comptes Rendus*, 1899, 129, p. 823) discovered that radium has the power of conferring radio-activity on its surroundings, and this phenomenon was later studied by Curie and Debeirne (*Comptes Rendus*, 1901, 132, p. 768; 1902, 133, pp. 276 and 931). For a long time these investigators held the view that the radio-activity was "induced" in the inactive matter by the *radiations* of the radium. But it has now been admitted that the "induced" or imparted activity of radium is caused by the emanation in a precisely analogous manner to that of the allied phenomena in the case of thorium. The imparted radio-activity from radium decays more rapidly than that from thorium, falling to half value in 28 minutes, but possesses the same property as the latter of travelling in the electric field and being concentrated entirely on the negative electrode. The matter causing the radium-imparted activity possesses interesting features, in that its radio-activity is derived from three separate and successive disintegrations. In a first consideration only the last of the three changes need be considered, as this is the one usually most important. The rate of decay given above for the imparted activity of radium—viz., to half-value in 28 minutes ($\lambda = 4{\cdot}1 \times 10^{-4}$)—applies to the last disintegration. The stored-up emanation and the imparted activity it produces in the mass of the dry solid compounds of radium cause the greater part of the radio-activity. When they are removed, by keeping the radium in solution in the open air for some hours, the activity of the radium compound after evaporation to dryness is very much enfeebled, but slowly recovers as the emanation and imparted activity is regenerated in the compound. A non-separable radio-activity of the radium is retained after solution, or any other treatment, and constitutes about 25 per cent. of the total activity. As in the case of thorium and uranium, it comprises only α-radiations.

This enfeeblement of the activity of radium preparations through damp and the corresponding increase of activity of dry, solid salts from the moment of their preparation from their solution was known many years before the changes were connected with the escape and re-accumulation respectively of the radio-active emanation (Giesel, *Wied. Ann.*, 1899, VIA., p. 91; M. and Mme. Curie, *Comptes Rendus*, 1902, 134, p. 85).

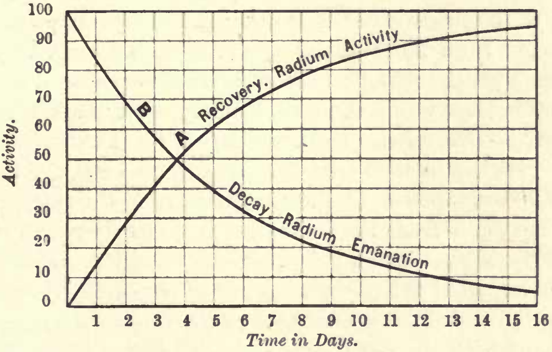


FIG. 31.

Fig. 31 shows the curves obtained (Rutherford and Soddy, *Phil. Mag.*, 1903, VI., 5, p. 455) for the recovery of the activity of radium compounds and the decay of the activity of the emanation. They are in every respect analogous to those given for the case of thorium and thorium X (see Fig. 26). In curve A the activity regained, between the minimum and maximum activities, is taken as 100. Curve B was obtained by storing the emanation mixed with air in a gasholder over mercury, and removing from day to day an aliquot portion, which was introduced into a testing vessel provided with an internal electrode, so that the ionisation current through the gas due to the emanation could be determined. The current is measured immediately after the introduction of the emanation, for the activity rises rapidly at first owing to the production of the matter causing the imparted activity from the emanation. It will be seen that the activity of the emanation falls to half value in about 3·7 days, and the value of λ is about $2{\cdot}2 \times 10^{-6}$. M. Curie (*Comptes Rendus*, 1902, 135, p. 857) found that the penetrating rays from a sealed glass tube containing the radium emanation decayed to half value in four days, and this constitutes a separate determination of the decay of the emanation. For although the rays in question do not come from the emanation, which we know gives no β-rays, but from the imparted activity, it can be shown by the disintegration theory that the rate of decay after the first few hours must be that of the radio-activity of the emanation in the tube. For we have seen in the case of thorium (p. 111) that the radiating power of the matter causing the imparted activity deposited from the emanation is proportional to the radio-activity of the emanation producing it, and as the latter decays the imparted activity diminishes correspondingly. By subjecting the sealed tubes during the experiments to temperatures varying between 450°C. and -180°C. it was shown that the rate of decay, and therefore the radio-active constant λ, is completely unaffected by change of temperature.

The disturbing effects of the later products of the disintegration, as, *e.g.*, the production of the imparted activity, upon the course of the earlier ones, which has already been referred to for thorium, is much more marked in the case of radium. Curve A (Fig. 29) was obtained by working with radium that had been kept in solution some hours before evaporating to dryness. If, however, the dry salt containing its maximum charge of occluded or stored-up emanation is dissolved in water and *immediately* evaporated to dryness, the emanation completely escapes, but the imparted activity produced by it in the compound remains with the radium. The activity, instead of being reduced by this treatment to the 25 per cent. constant minimum, is only reduced to about 60 per cent. of the maximum value. But a very rapid decay to nearly the 25 per cent. value occurs in the course of the first three or four hours after the evaporation. This is due to the imparted activity produced in the compound by the stored-up emanation rapidly decaying when the latter is removed. After a few hours the activity again commences to increase, and the subsequent progress of the recovery is the same as in curve A. The converse of this phenomenon is to be found in the rapid increase of activity for the first few hours when the emanation is introduced into a fresh vessel. If the central electrode is made negative with respect to the vessel, the imparted activity is concentrated solely upon it, as in the case of that from thorium. After the introduction of the emanation the activity steadily rises, very rapidly indeed for the first few minutes, and then slowly for three or four hours until it attains a maximum nearly twice as great as at first. If at any time the central electrode is removed, the imparted activity upon it will be found to account for the increase of the value of the activity above the original value. Or, the emanation may be suddenly blown out at any stage, when the current through the gas is reduced to the proportion of the total activity due to the imparted matter on the negative electrode.

β-rays.—Radium furnishes valuable evidence of the stage of the disintegration at which the β-ray is produced. It will be recalled that in the case of uranium the α-radiation is produced in the first change of uranium into uranium X and the β-radiation in the subsequent disintegration of uranium X. The cases of thorium and radium are less simple. With regard to the former a complete separation of the matter causing the imparted activity from thorium X has not yet been effected. Now, the imparted activity gives β as well as α-rays, and it is possible that the β-radiation of thorium X is due entirely to this source. The emanation (compare p. 101) is known to give only α-rays, and the same is true of the non-separable activity of thorium. The lower the emanating power of a thorium compound—that is, the more the emanation is retained by the compound to produce the imparted activity within it—

the greater the proporton of β-radiation from the compound. Although it has not been definitely proved, the facts are in agreement with the possibility that the β-radiation of thorium occurs only in the last stage of the disintegration, as in the case of uranium.

In the case of radium the evidence is more definite. If the radium emanation is introduced into a vessel the walls of which are thick enough to completely absorb the α-rays and thin enough to allow the β-rays to pass through, it is found that at first there is no external radiation from the vessel—*i.e.*, there are no β-rays produced (Curie, *Comptes Rendus*, 1902, 135, p. 857; Rutherford and Soddy, *Phil. Mag.*, 1903, VI., 5, p. 456). In a short time after the introduction of the emanation, an external or β-radiation is produced from the vessel, and this rapidly and regularly increases, attaining a practical maximum in three or four hours. If the emanation is then swept out of the vessel by a blast of air, the value of the external radiation from the vessel is not suddenly affected, but commences to decay regularly, falling to half value in 30 minutes, and decreasing practically to zero in three or four hours. Thus the radium emanation gives only α-rays. The β-rays are produced after it has changed into the matter causing the imparted activity. Since the decay of the β-radiation of the latter is regular, falling to half value in 30 minutes, it follows that the β-rays are produced in the later changes of this matter, for it will be shown later that the first change suffered by the matter causing the imparted activity is very rapid, and the time-constant characteristic of the change of the β-radiation—to half value in 30 minutes—is that of the last change that can be experimentally traced. Hence, in both uranium and radium, and probably also in thorium, the β-rays only appear at the last stage of the disintegration. Now the γ-rays accompany the β-rays, and are always produced with them (Rutherford). Hence we must regard the α-ray as being the most characteristic feature of radio-activity, and the β and γ-rays as less important and, in point of time, secondary. Polonium, for example, gives only α-rays, showing that the β and γ-rays are not essential to the phenomenon. Atomic disintegration, in the majority of instances we know of, when the separate stages are considered each by itself, proceeds with the expulsion of α-rays alone. The important issue raised as to how the equivalence of electric charges is maintained, if positively charged particles are being continually expelled alone, will be considered in the last chapter.

The foregoing results may be briefly summarised. The radio-activity of a compound of radium that has been kept for some time in the dry solid state—a sufficient time to attain its maximum activity—is made up as follows:—(1) A non-separable activity, comprising 25 per cent. of the total α-radiation; (2) a part due to the occluded emanation, comprising about 40 per cent. of the total α-radiation; (3) the imparted activity, comprising about 35 per cent. of the total α-radiation and all the β and γ-radiation. If the compound is dissolved in water and immediately evaporated to dryness, or if heated and cooled, the activity of the radium is reduced and is now due to (1) and (3), comprising about 60 per cent. of the α-rays and all the β and γ-rays. On keeping for five hours its activity decays further and then comprises only (1), viz., 25 per cent. of the α rays and no β or γ-rays. The emanation given off on heat or solution at first comprises 40 per cent. of the total α-radiation and gives no β or γ-rays. If kept for five hours in a closed vessel it then comprises (3) as well as (2), and represents 75 per cent. of the α and all the β and γ-radiation. If now the emanation is blown out of the old vessel into a new vessel, the latter will again show the 40 per cent. α-radiation due to the emanation alone, whereas the old vessel will retain 35 per cent. of the α-radiation and all the β and γ-radiation. A complete separation of the constituents (1), (2), (3) has thus been simply effected. In five hours the activity of the old vessel almost completely disappears, whereas the new vessel in which the emanation has been stored again shows an increased α and all the β and γ-radiation. Now, consider the radium preparation and the stored emanation over a further period of one month. During this interval the radium gradually regains its radiating power, and the vessel in which the emanation is stored correspondingly loses its radiating power. In these changes the α, β and γ rays now grow and decay together. At the end of a month the radium is again in its initial condition of maximum activity, and the activity of the separated products is practically nil. The whole of this complicated series of changes, every step of which is predicted by the disintegration theory, is followed in the minutest detail when the simple experiment of heating a radium compound is investigated in the laboratory. The above example furnishes a good instance of the principle of the conservation of radio-activity.

Evidence of the Complexity of the Changes Causing the Imparted Activity of Radium and Thorium.—The curve of decay of the radium-imparted activity is very irregular (Rutherford and Miss Brooks, *Phil. Mag.*, 1902, VI., 4, p. 18). It consists of (1) a very rapid decrease to possibly 20 per cent. of the original value in the course of a few minutes; (2) a period of a few minutes of slow change, which is, under some circumstances, a slight increase, and, under others, a slight decrease; (3) a final geometrical decrease to zero, the activity falling to half value in 30 minutes. Now, under certain circumstances, an irregular curve can be obtained also for the decay of the thorium-imparted activity (Rutherford, *Phys. Zeit.*, 1902.) If a wire is exposed for a *short* time only to the emanation of thorium, it is found on removal that the imparted activity increases at first instead of decaying, and may rise in the course of the first few hours to three or four times its initial value, from that point decaying at the normal rate to half value in 11 hours. So also in the case of radium, the initial rapid decrease of the imparted activity is the more marked the shorter the time of exposure to the action of the emanation. These effects would be produced in each case if the disintegration causing the activity of the imparted matter, instead of being single, consisted of more than one successive change whose combined effects give rise to the radio-activity.

It is necessary to assume two changes in the case of the thorium-imparted activity, the radio-active constant of the first type being in the neighbourhood of 2×10^{-4}, so that the change is half complete in about an hour. For the last type the value of λ is $1{\cdot}7 \times 10^{-5}$, as already mentioned.

For the case of radium it is necessary to suppose that three types are successively produced. The rate of change of the first is very rapid, being comparable with that of the thorium emanation, and this accounts for the initial rapid increase of activity when the emanation is transferred to a new vessel, as well as for the initial rapid decay of the imparted activity after the emanation is removed.

Most recently the decay of the imparted activity of radium has been subjected to more detailed examination by Rutherford (*Phil. Mag.*, 1904, VI., 7, p. 216) and Curie and Danne (*Comptes Rendus*, 1904, 138, p. 683), and both of these independent investigations have led to identical conclusions. The course of the decay is explained on the disintegration theory if three successive disintegrations occur in the imparted activity matter after its production from the emanation. The respective radio-active constants are approximately as follows: For the first type, $\lambda = 4 \times 10^{-3}$; for the second, $\lambda = 5{\cdot}4 \times 10^{-4}$; and for the third, $\lambda = 4{\cdot}1 \times 10^{-4}$. The first change is half completed in three minutes, and is accompanied by α-radiations alone; the second is half completed in 21 minutes, and *is not accompanied by any radiation*; the third and last change is half completed in 28 minutes, and gives rise to α, β and γ-rays (Rutherford). It would seem also, from a cursory examination, that the first type in the case of thorium changes into the second without the expulsion of rays, although this case has not yet been accurately studied from this point of view.

Thus, even in these most complex cases, the simple law holds that a constant fraction of each type changes in the unit of time, and the complexity is due solely to the number of successive types produced. In addition, we are introduced for the first time to a sub-atomic change, unaccompanied by radio-activity, which is detected because it is intermediate between two other changes which are. This should serve to emphasise how dependent we are for our knowledge of natural phenomena on the means of experimental investigation at our disposal. Sub-atomic change has been established in the case of the radio-elements on account of the accompanying expulsion of radiant particles. The example just considered shows that this is not necessarily an essential accompaniment of the process in all cases. Hence the absence of radio-activity in the cases of the inactive elements does not imply that these necessarily are not undergoing change, but rather that no means are at present available of detecting such changes unless they happen to be accompanied by the expulsion of radiant particles. There seems little doubt that our knowledge of the scheme of material evolution is at present only in its infancy, and that when fully elucidated it will be found to embrace all the elements within its scope. To arrive at this result it is not necessary to suppose that all the elements are really radio-active, and that those we call inactive are in reality so feebly active as to be beyond the range of the present methods. It seems more reasonable to regard radio-activity as a somewhat special and unusual accompaniment of sub-atomic change. Such changes without such accompaniment may be occurring even in the commonest materials at rates relatively rapid when expressed in cosmical time-units, and still have remained wholly unsuspected.

The "Induced Activity of Radium with Slow Rate of Dissipation."—The statements made in the foregoing account of the imparted activity of radium, that this decays to zero in the course of five or six hours, are not strictly accurate. Mme. Curie describes in her Thesis a phenomenon, under the above title, which, under ordinary conditions, is only observed with very active specimens of radium. If the imparted activity is left to itself it decays regularly after the first half-hour in a geometrical progression with the time ($\lambda = 4 \cdot 1 \times 10^{-4}$) until it attains a value only $\frac{1}{20000}$th of the initial, and then remains sensibly constant over many months. Hence, beyond the last type of matter causing the imparted activity, there is a further very slowly-changing transition-form produced. It has been suggested (Soddy, Wilde Lecture, 1904, Manchester Literary and Philosophical Society) that this type is Mme. Curie's polonium, and the evidence in favour of this view will be considered in the last chapter.

The Actinium Emanation.—The rate of decay of this emanation and of the imparted activity it produces has recently been determined by Debierne (*Comptes Rendus*, 1904, 138, p. 411). The activity of the emanation decays in a geometrical progression with the time, falling to half-value in 3·9 seconds, and the imparted activity produced from it also decays regularly, falling to half-value in 40 minutes. The value of λ in the first case is, therefore, 0·17 and in the second 3×10^{-4}. The results obtained indicate, however, that an intermediate product exists between these two types, which appears to be gaseous and, therefore, of the nature of an emanation, which changes even more rapidly than the first emanation, but apparently does not expel rays. The evidence is as follows:—When the experiment described on p. 111 (Fig. 29) is repeated for the actinium emanation, the amount of activity imparted to the first electrode is less than to the succeeding, and the results indicate that when first produced the emanation imparts no activity to its surroundings, but this power rapidly increases to a maximum and then decays at the same rate as the activity of the emanation. The decay of the activity of the emanation itself shows no initial irregularity. More detailed investigations must be awaited before any definite conclusions can be drawn.

Do the Emanations carry an Electric Charge?—Rutherford showed that the thorium emanation is not affected by an electric field, and concluded that it was not charged (p. 101). He has also shown that the radium emanation survives prolonged action of the electric field without being removed from the atmosphere in which it is present. Recently McClelland (*Phil. Mag.*, May, 1904, p. 355) has shown that no charge is communicated to a vessel by admitting the radium emanation through a plug of glass wool, and concludes that it is unchanged. All these results, however, merely show that the emanation particle, if charged when produced, loses its charge like an ion and, as is to be expected, presents the same essential properties of radio-activity and volatility whether charged or uncharged. Since, on the average, the radium emanation atom exists 5·3 days before disintegrating, it would be remarkable if it persisted charged so long under ordinary conditions of storage. There are theoretical reasons for expecting the atom to be charged at the moment of its production, since it is derived from a neutral radium atom by the expulsion of a positive ion.

The case of the actinium emanation, for which the average period of existence is only 5·7 seconds, therefore presents features of great interest. The expected difference exists. When produced, the atom of the actinium emanation appears to carry a positive charge, which endows it with certain peculiarities which are not exhibited by the radium and thorium emanations. Debierne (*Comptes Rendus*, 1903, 136, pp 446 and 767) showed that the actinium emanation moves to the negative electrode in an electric field, and Giesel

(*Ber der D. Chem. Gesel.*, 1903, 36, p. 343) showed that the emanation from his "emanation substance," which we have seen is probably identical with Debierne's actinium, shows the same property. The experiments of Geisel may be described. He placed the active substance in a metal cylinder, open at one end, 5cm. to 10cm. from a zinc-blende screen negatively charged by means of an influence machine. Under these circumstances the particles of emanation are attracted to the screen and travel to it in straight lines, so that a phosphorescent image of the opening of the cylinder is formed on the screen. From this image "secondary emanation" streams away, lighting up the screen surrounding. A blast of air now does not affect the phosphorescent image, but only the "secondary emanation." Thus the emanation in an electric field travels to the negative electrode in straight lines like a "ray," and Geisel proposes the term "E-ray" for this phenomenon. Having given up its charge, it is no longer controlled by the electric field, but can be blown away, so that it seems probable that what Geisel calls "secondary emanation" consists of the emanation that has given up its charge, but has not yet disintegrated. The theoretical bearing of this result will be considered in Chapter XII.

Methods of Experimentally Recognising Radium, Thorium and Actinium, and the Analysis of Radio-active Minerals.—The three distinctive emanations and the three equally-distinctive types of imparted activity they produce furnish sufficiently characteristic analytical tests for the presence or absence of the elements radium, thorium and actinium in the radio-active minerals. To test for radium, these should be brought into the state of solution and kept in closed bottles for a few days. The gas above the solution should then be drawn off and stored for 10 minutes. In this time the thorium and actinium emanations will have completely disappeared, and if a sample of the gas blown into an electroscope discharges it the presence of the radium emanation may be inferred. The conclusion may be confirmed in two ways. In the first place, if the amount is adjusted to give a moderate rate of leak in the electroscope, it will be found that the leak will increase notably—usually 30 or 40 per cent.—in the first few minutes after the gas is introduced. If then the emanation is blown out and the rate of leak again tested it will be found to rapidly decrease to a very small value in the first few minutes. A larger quantity of the emanation may then be introduced and allowed to remain some 30 minutes before being blown out. The activity remaining will again rapidly diminish, but will now be quite noticeable after half an hour. If after this period it is tested from time to time, it will be found to decay to half-value after the lapse of 30 minutes, to one-quarter in an hour, and so on. The second test for the radium emanation is to store the gas in an air-tight gas-holder, and to remove equal portions at intervals of four days. Its power of discharging the electroscope will persist noticeably for many weeks, diminishing to one-half during each interval of four days, or correspondingly for longer or shorter intervals. If it fulfils these tests, the presence of radium in the mineral may be considered proved and its quantity deduced by comparative tests, using a known sample of Joachimsthal pitchblende as the standard.

To test for thorium, the gas swept out of the bottle containing the mineral is rejected, and the emanation, allowed to re-accumulate for five minutes, is removed to a gas-holder and stored for one minute. In this time the actinium emanation completely disappears, and if, when a portion is blown into an electroscope, it discharges it the presence of the thorium emanation may be inferred. This may be proved as follows: After introduction of the gas the discharging power will rapidly decay to half-value in one minute, to quarter-value in two minutes, and so on, and in five minutes will have *completely* disappeared. No appreciable amount of thorium-imparted activity is produced under these circumstances. To confirm, a continuous stream of air may be passed through the solution into a metal vessel containing an insulated negative electrode, the vessel itself being connected with the positive pole of the battery. The voltage is not important, but may be from 50 to 300 volts, according to the size of the vessel. The rate of decay of the imparted activity on the negative electrode may then be determined, commencing after the lapse of five or six hours from the time it is removed from the vessel. In this time any radium or actinium-imparted activity will have completely decayed, whereas the thorium-imparted activity decays to half the initial value in 11 hours, and should still be noticeable after two days.

To test for the presence of actinium, the air is blown out from the solution as before, and then a rapid current is blown directly into the electroscope for some two or three minutes. While the air-stream is passing the electroscope should leak rapidly. The rate of decay of the actinium emanation is so rapid that it cannot be well measured by this rough means, but its presence may be confirmed by continuing the air stream for some time, when there should be left in the electroscope an imparted activity very similar to that of radium in its rate of decay, but slightly slower. If care is taken to completely blow out all the accumulated radium emanation from the solution before the air-stream is sent into the electroscope, the possibility of the radium-imparted activity is excluded, and the presence of actinium may be inferred.

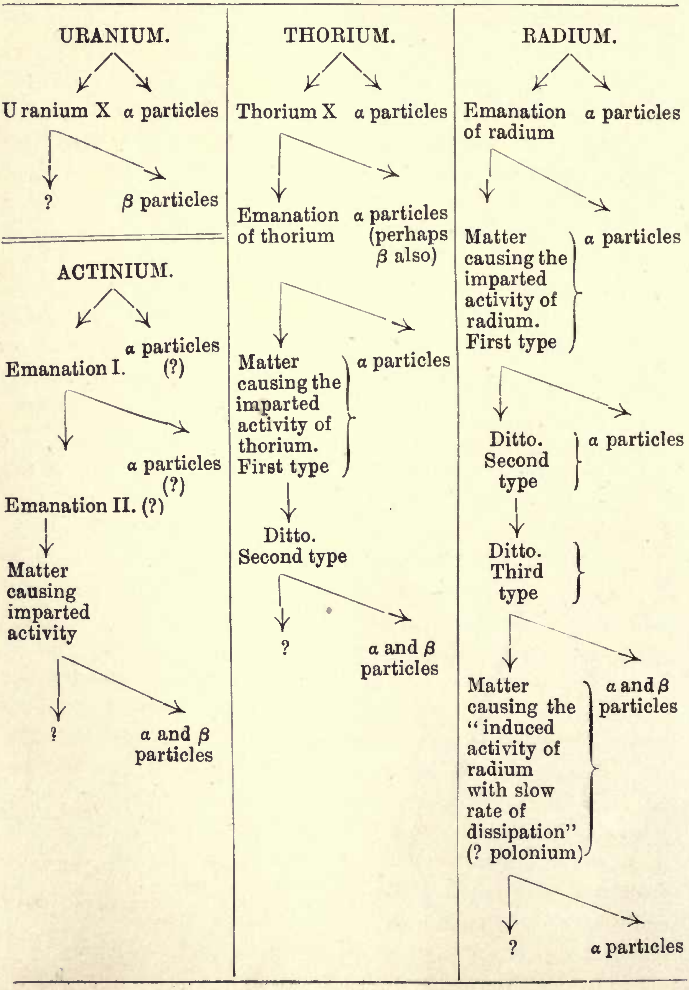
If in these tests an emanation is obtained which differs essentially from those above described, the existence of a new radio-element may be inferred. The characteristics of its radio-activity may be worked out and the element sufficiently defined before a single chemical separation of the mineral is effected.

The principles underlying these methods of radio-active analysis will be readily understood from previous chapters. The most important is that the radio-activity measured is the product of the radio-active constant λ and the number of atoms present of the type of matter in question. If λ is relatively small, as in the case of the radium emanation, λN is small when N is considerable. Hence, although the number of atoms of radium emanation present after the lapse of five minutes' accumulation may be as great as the number of thorium emanation atoms, only the latter exhibits a detectable radio-activity. The more slowly a transition-form loses its activity the longer must it be allowed to re-accumulate in order for it to be detected by its activity. Similarly, only the first type of the radium-imparted activity matter is experimentally observed when the emanation is allowed to remain in the electroscope only a minute or so, and this, after the latter is blown out, decays practically to zero in five or six minutes. If left in half an hour or more, the emanation produces sufficient of the slower-changing third type to be quite apparent after the rapidly-changing first type has decayed. These examples might be multiplied indefinitely. Extended to the radio-active minerals themselves over a period of years or centuries, they throw light on the fundamental problem, with which this chapter opened, of the present existence of so powerful a radio-active element as radium.

APPENDIX.

Two tables are appended which summarise the present knowledge of the course of atomic disintegration. In the first, the complete series of disintegrations in the case of uranium,

Table I.



thorium, radium and actinium have been tabulated. In the second, the unstable elements have been arranged in the order of their instability together with their radio-active constants, λ, or the fraction changing per second, and the period of their average lives, as given by $1/\lambda$. The latter is the time required for the quantity to be reduced to $1/e$ (0·368) of the initial, which is 1·45 times longer than that required for the quantity to be reduced to half. The values of λ for the last three examples are deduced in Chapter XI.

Table II.

Element.	λ	$1/\lambda$
Actinium emanation	$1\cdot7\times10^{-1}$	5·7 seconds
Thorium emanation	$1\cdot16\times10^{-2}$	87 ,,
Radium-imparted activity I....	4×10^{-3}	4 minutes
Ditto ditto II....	$5\cdot4\times10^{-4}$	30 ,,
Ditto ditto III....	$4\cdot1\times10^{-4}$	41 ,,
Actinium imparted activity ...	3×10^{-4}	58 ,,
Thorium-imparted activity I....	?	?
Ditto ditto II....	$1\cdot7\times10^{-5}$	16 hrs. 30 min.
Radium emanation	$2\cdot2\times10^{-6}$	5 days 8 hours
Thorium X	2×10^{-6}	5 days 19 hours
Uranium X	$3\ 6\times10^{-7}$	32 days
Polonium ... ... about	2×10^{-8}	about 16 months
Actinium	?	?
Radium	3×10^{-11}	1,150 years
Uranium ... } about	3×10^{-17}	about 10^9 years
Thorium ... }		

CHAPTER X.

THE MATERIAL PROPERTIES OF THE RADIUM EMANATION AND ITS TRANSMUTATION INTO HELIUM.

Material Properties of Radio-active Matter.—Condensation of the Emanations by Liquid Air.—Differences of Behaviour between the Thorium and Radium Emanations.—Volatility of Matter causing Imparted Activity.—Diffusion Coefficient of Radium Emanation.—Production of Helium from Radium.—Production from the Emanation.—Phosphorescent Spectrum of Radium Compounds.—Volume occupied by Radium Emanation.—Diminution on Keeping.—Possibility that the a-Particle is a Helium Atom.—Law of the Conservation of Mass need not apply to Sub-atomic Changes.

Radio-activity is the property of but a few atoms only of the total mass in any given instant. A certain fraction, represented by λ, contributes the activity in any given second, while the vast preponderance, $1 - \lambda$, is, strictly speaking, not radio-active at all. It is, for the time being, ordinary matter which must possess a normal chemical and physical nature in addition to and independent of the radio-active properties contributed by the fraction undergoing disintegration. Each transition-form is a new element, although an unstable one, and some information about its ordinary material properties may be obtained. An example may make this clear. The radium emanation passes through tubes containing various reagents which would completely absorb all the known gases except the members of the argon family. Like the latter, it may be sparked over potash in an atmosphere of oxygen or kept in contact with red-hot magnesium and calcium for many hours without being appreciably absorbed. The conclusion is indicated that the emanation is a chemically inert element of the argon family. In this and similar work the chemical nature of the matter is the real object of study, the radio-activity merely furnishing the means of detection and measurement. The special interest about this work is the smallness of the quantity of material employed. Consider, for example, the case of the phenomenon exhibited by the emanations of being condensed at low temperatures. If the volatility or vapour pressure of an ordinary gas were investigated by direct methods, the limit of detection would be reached with pressures of $\frac{1}{100}$mm. of mercury. In the case of the emanations we can study accurately the effect of changing temperature on the volatility of quantities a million million times less, and the results, therefore, are of great general interest from a purely physical point of view, apart altogether from the special means by which the result is obtained.

Condensation of the Radio-active Emanations.—This property, in the case of the radium emanation, may be made the subject of a striking and beautiful lecture experiment. If the emanation, obtained by heating or dissolving a few milligrammes of pure radium bromide, or by storing a solution of the latter in a closed bottle for a week since the emanation was last removed, is swept by a slow stream of dry air into a glass spiral tube immersed in liquid air, practically the whole of the emanation condenses at the point where the spiral enters the liquid air, causing the glass to fluoresce brilliantly. The apparatus usually employed by the Author is shown in Fig. 32. Some hours before the lecture the emanation from the radium is swept into A and the taps closed. A is a glass tube in which has been sealed a piece of willemite. It has been mentioned that the β-rays of radium are mainly operative in causing this mineral to fluoresce, so that the luminosity of the mineral increases for some hours after the emanation is introduced into the tube, and after five or six hours is mainly due to the imparted activity, and not to the emanation. Kunzite would show this effect even better, but is less brilliantly luminous. For showing the condensation of the emanation the tube is attached to the glass spiral B immersed in a vessel containing liquid air C. To the other end of the spiral a long glass tube D is attached. In D is placed a long narrow strip of mica painted with phosphorescent zinc sulphide. The easiest way to coat the mica is to cover it with a hot film of gelatine solution, and sieve on the zinc sulphide after the gelatine has partly set. With this apparatus, provided that an extremely slow air-stream is used, the whole of the emanation is condensed at a point in the spiral. The tube A may then be removed and the air-stream sent into B direct. On removing C the stage at which the emanation volatilises may be clearly seen by the passage of the glowing gas along the spiral into D. Then the gradual ascent of the emanation as a zone of intense luminosity may be watched in D as the air-stream is continued. The lower parts of the screen in D, the whole spiral, and also the willemite in A, continue to glow, owing to the imparted activity, after the emanation has passed on, but this glow completely disappears in the course of a few hours. To illustrate the properties of the emanation a long narrow tube may be inserted between A and B before the condensation

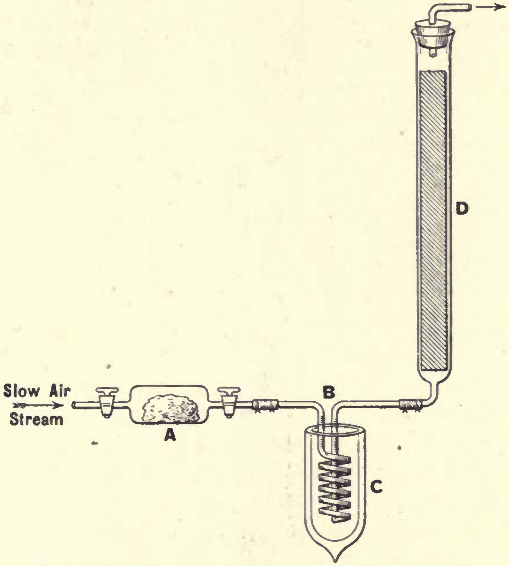


FIG. 32.—Lecture Experiment for Showing the Condensation of the Radium Emanation by Liquid Air.

is carried out. This tube might be made to pass round the lecture table, and if small diamonds are introduced at different points the arrival of the emanation at the various points could be rendered visible.

The apparatus used in the detailed investigation of the condensation of the emanations (Rutherford and Soddy, *Phil. Mag.*, 1903, VI., 5, p. 561) is shown in Fig. 33. The emanation was carried by means of a steady stream of some suitable gas through the doubly-wound copper spiral, which was immersed in a bath of liquid, kept well stirred, at the temperature required. The liquid employed was ethylene, which boils at – 103°C. and freezes at – 169°C. The outside tube was cooled with liquid air, and ethylene gas passed in through A until the level of the liquefied ethylene was well above the top of the spiral. The emanation was passed through the spiral after the bath had been cooled by means of liquid air slightly below

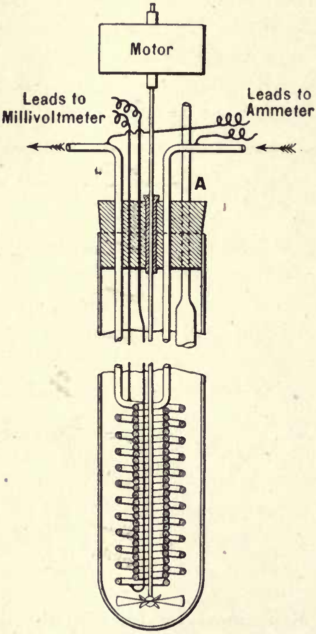


FIG. 33.

the temperature of condensation. The apparatus was wrapped round with felt and allowed to warm up slowly, and the temperature was noted at which the emanation volatilised and was swept out of the spiral by the gas stream.

The general arrangement of the apparatus for the case of the radium emanation is shown in Fig. 34. The emanation was stored in the gas holder B, and small quantities could be sent as required into the gas stream kept passing continually into the spiral from A. The temperature of the bath was known by the electrical resistance of the copper spiral, and this was determined by keeping a constant current of about an ampere flowing through it, and measuring the difference of potential between two points at the top and bottom of the spiral by means of a milli-voltmeter. In Fig. 35 the resistance of the copper spiral is plotted on the vertical axis, and the temperatures in degrees Centigrade on the horizontal axis. The fixed points chosen in the calibration of the spiral were the boiling and freezing points of water, the boiling and freezing points of ethylene, and the boiling point of liquid air of known composition. The temperatures of the condensation of the thorium emanation (–120°C.) and of the radium emanation (–150°C.) were found by interpolation. With the radium emanation the point of volatilisation and condensation is very sharp. In one experiment at –154°C. no emanation

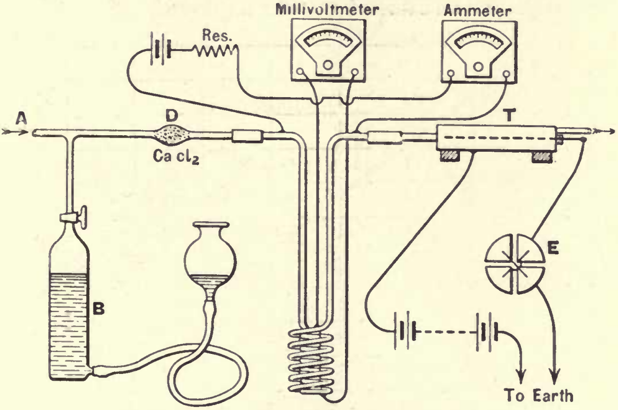


FIG. 34.

was observed in the gas stream, although less than $\frac{1}{10000}$th part could have been detected, while at −150°C. the greater part had volatilised. The decay of the activity of the thorium emanation is so rapid that the emanating compound has to be retained continuously in the gas stream. It is then found that some emanation escapes condensation when the temperature is as low as −150°C., but the greater part is condensed at much higher temperatures. The point given above, −120°C., represents the temperature at which condensation commences to take place. In the case of the radium emanation, no great difference is observed between the temperature of initial volatilisation and complete condensation, provided sufficiently slow streams of gas are employed.

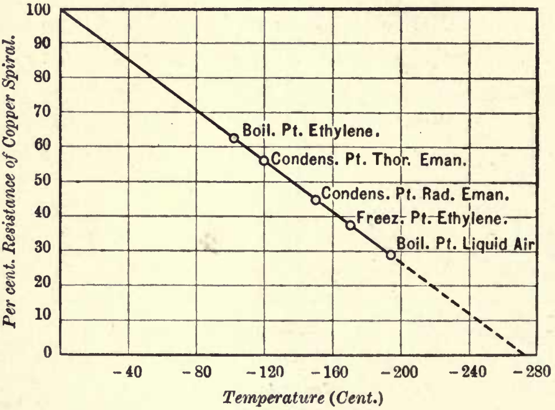


Fig. 35.

This difference between the two emanations was further investigated by means of the apparatus shown in Fig. 36, which is arranged for the case of the thorium emanation. The gasholder B and T-piece of Fig. 34 replaces the thorium oxide tube A for experiments with the radium emanation. Instead of the emanation being carried by a steady current of gas it is transferred to the spiral, and thence to the testing vessel, by means of the mercury pump P. This was effected by raising or lowering the mercury in the pump and by opening and closing the various taps as required. In this way the experiments could be performed in an atmosphere of any gas at any pressure below atmospheric, and the emanation allowed to remain in the condensing spiral any definite time before being removed. It was found that the temperature of condensation in the case of the radium emanation was substantially the same however the experiment was performed. But for the thorium emanation the proportion of the total condensing at any temperature varied (1) with the nature of the gas used, a greater proportion condensing in an atmosphere of hydrogen than of oxygen; (2) with the time allowed, a greater proportion condensing the longer the emanation was allowed to remain in the spiral. The proportion escaping condensation was always greater when a current of air was kept passing than when the experiments were performed by means of the pump in a stationary atmosphere. The temperature of initial condensation, – 120°C., was, however, always substantially the same whatever the conditions.

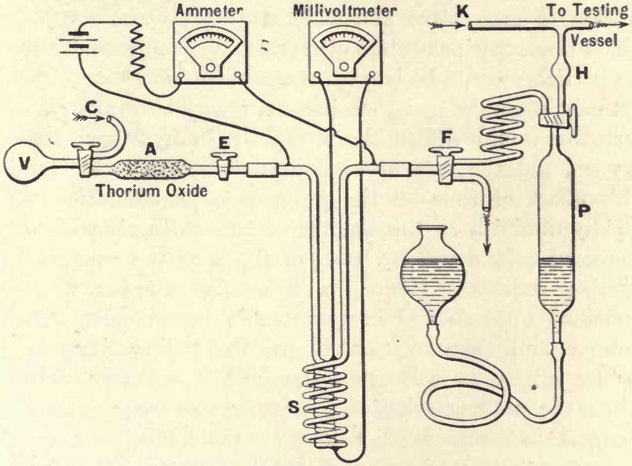


FIG. 36.

The difference between the behaviour of the thorium and the radium emanation is capable of a simple explanation on the disintegration theory. Consider quantities of the two emanations possessing similar radio-activity—*i.e.*, giving a similar number of α-rays per second. The radio-activity is proportional to the number of atoms disintegrating per second—*i.e.*, to λN, where N is the total number present and λ the radio-active constant. Since λ for the radium emanation is 6,000 times smaller than for the thorium emanation, it follows that, when λN is equal for the two cases, the number of atoms of emanation present must be 6,000 times greater in the case of the radium emanation than in the case of the thorium emanation. Condensation can only take place by the approach of two or more of the condensing molecules into each other's sphere of influence, and it is to be expected, when the quantities present are extremely small, that the amount condensing will depend on the concentration of the particles, the time allowed for the process to take place, and the nature of the atmosphere in which the experiments are performed. The concentration of the particles would be less when the emanation was carried by a steady stream of gas than in a stationary atmosphere. The particles would diffuse more rapidly in hydrogen than in oxygen, and the rate of condensation would be accelerated. This effect of time on the progress of condensation would rapidly diminish as the number of condensing particles were increased. It would probably diminish as the square of the number present. Hence, if a similar number of atoms remained uncondensed in comparable experiments with the radium and thorium emanations, the radio-activity of the former might be well below the limit of ordinary detection when the radio-activity of the latter was large. To obtain comparable results with the two emanations the measuring instruments should be made 6,000 times more sensitive in the case of that of radium than for that of thorium. The ionisation current in the experiments on the thorium emanation was probably about 10^{-10} amperes. An ionisation current of 10^{-14} amperes is quite within the experimental range, and it is, therefore, quite possible to subject this explanation to direct experimental test. This has not yet been done.

From considerations to be deduced in the next chapter, it is probable that the thorium emanation obtainable from 10 grammes of thorium oxide if confined in 1 cubic cm. volume would only exert a pressure of between 10^{-16} and 10^{-17} atmosphere, so that the emanation obtainable from a million tons of thorium oxide, if concentrated in this volume, would only just produce a pressure that could be directly measured. It is, therefore, matter for remark that the emanation can be made to condense completely at any temperature, and the question remains, perhaps, somewhat doubtful as to whether the phenomenon is really one of simple gaseous condensation at all. It has been suggested that the gas used to carry the emanation condenses on the walls of the spiral by the action of capillary forces as a surface film, and carries with it the particles of emanation by attraction, possibly of a chemical nature. Such a surface condensation would presumably only occur in a gas below its critical temperature, and should, therefore, be absent in perfectly pure hydrogen or helium. It would be interesting to repeat the experiments in a stream of hydrogen of the highest possible purity. It is possible that condensation in these circumstances might not occur.

In this connection may be mentioned the work of Miss Gates (*Physical Review*, 1903, 16, p. 300), who found that the matter causing the imparted activity of thorium and radium could be volatilised at temperatures above a red heat and be deposited on neighbouring cold surfaces. This indicates that phenomena similar to the condensation and volatilisation of the emanations take place with the imparted activity matter at high temperatures.

Another property of the emanation of radium which has been investigated is its density, as shown by its coefficient of diffusion into air. The first experiments were performed by Rutherford and Miss Brooks. They determined the rate at which the emanation confined in one half of a long cylinder diffused into the other half when a partition between the two halves was withdrawn. They found a value for the coefficient of diffusion of about 0·08, which is comparable with that of ethyl ether, and indicates a density in the neighbourhood of 80 (H=1). If the molecule of the emanation is assumed to be monatomic, like argon, the atomic weight is, therefore, in the neighbourhood of 160.

The great difficulty in the experiments is to eliminate the effect of the imparted activity produced from the emanation from that of the latter by itself, and the rate of diffusion tends in consequence to appear lower than the true value. Owing to the very rapid initial changes in the radio-activity of the matter causing the imparted activity it is difficult to correct accurately for this error. The value given above, 0·08, is 20 per cent. greater than the uncorrected value. A determination of the coefficient has also been made by Curie and Danne (*Comptes Rendus*, 1903, 136, p. 1,314), who obtained the value 0·1, which indicates a density nearer to that of alcohol (46) than of ether. Most recently Bumstead and Wheeler (*Am. Journ. of Science*, February, 1904, p. 97) obtained a value indicating a density of 180, so that the evidence is conflicting, probably for the reason already given. The latter have definitely proved that the radio-active gas obtained from the tap-water at New Haven, U.S.A., and from the soil in the same neighbourhood, is the radium emanation, for the rate of decay of its activity and its rate of diffusion are similar to the latter. The existence of the radio-active gas in tap-water was first shown by J. J. Thomson in the Cambridge water, and experiments by H. S. Allen on the rate of decay of its activity indicated that it was the same as the radium emanation. The existence of this emanation in the soil was first noticed by Elster and Geitel.

The Production of Helium from Radium.—(Ramsay and Soddy, *Proc.* Roy. Soc., Vol. LXXII., 1903, p. 204.) The intense radio-activity of pure radium compounds, and the relatively rapid rate, therefore, at which it must be supposed to be disintergrating, made it appear likely that if helium were one of the ultimate products formed, as has already been suggested (p. 117), it might be possible to detect it by direct spectroscopic methods if a reasonable time were allowed for it to accumulate. The apparatus used is shown in Fig. 37. In two separate experiments 20 and 30 mgs. of pure radium bromide, which had been kept for some months in the dry solid state, were employed. From the general analogy of helium to the radium emanation, both in chemical nature and the phenomenon they exhibit of being occluded and of being liberated by heat and solution, it was assumed that the helium if produced would not escape from the radium compound until it was heated or dissolved. The radium bromide was introduced into the bulb A which was then sealed at the dotted line M to the bulb B, carrying two taps. This was then completely exhausted, the lower tap closed, and water, boiled free from air, allowed to flow into B. The seal at the dotted line N was then made. C is a tube in which is a thin copper spiral wire which can be heated by an electric current. D is a phosphorous pentoxide tube to absorb moisture. E is a capillary U tube which is cooled in liquid air during the experiment. The emanation and any CO_2 present are condensed here and prevented from entering the spectrum tube.

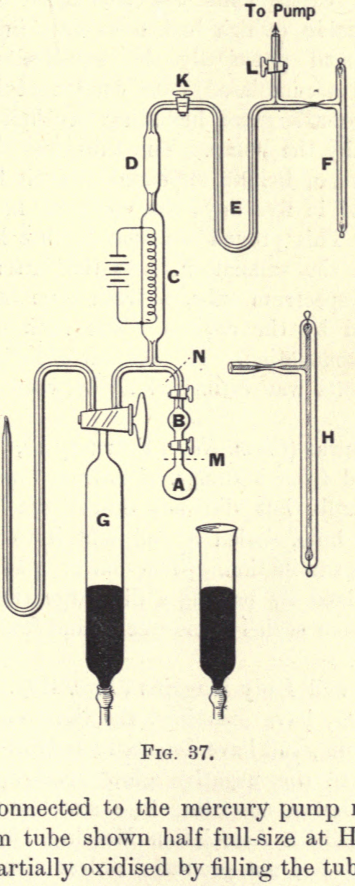


FIG. 37.

The tap L is connected to the mercury pump not shown. F is the spectrum tube shown half full-size at H. The copper spiral is first partially oxidised by filling the tube with oxygen from the burette G and keeping the spiral at dull redness by a current. The whole apparatus is thoroughly exhausted and all taps closed. Water is now admitted from B into A, the radium bromide dissolves and gives up its occluded gases, which are admitted into C. The red-hot spiral absorbs the hydrogen and oxygen, and the water produced is absorbed in D. Mercury is now allowed to flow up from G, and the gases forced into the spectrum tube through the cooled U tube. The spectrum on examination proved to be practically the complete spectrum of helium.

In the next experiments the emanation from 50 mgs. of radium bromide (which had been kept in solution in a closed vessel until practically the equilibrium amount of emanation had accumulated) was condensed in a spectrum tube, and all volatile gases, including any helium, if present, were removed by the pump. The tube was then sealed off. The yellow line of helium appeared after it had been kept three days, and in five days the complete helium spectrum was obtained. This proved conclusively that helium is being produced from the emanation. As the latter changes into helium in the spectrum tube, a fresh crop of emanation is being produced by the radium. The radium changes into helium *via* the emanation. The amount of helium obtained in these experiments was estimated at between 10^{-4} and 10^{-5} cubic cm.

Curie and Dewar (*Chem. News*, February 19, 1904) examined the gas evolved from 400 mgs. of radium bromide on fusion in a quartz bulb, but did not detect the helium. They exhausted the bulb, sealed it and sent it to Deslandres for spectroscopic examination. The latter photographed the spectrum obtained by passing a discharge through the bulb after it had been sealed some weeks, and found it to be that of pure helium.

Sir William and Lady Huggins (*Proc.* Roy. Soc., 1903, 72, pp. 196 and 409) have examined the light emitted by compounds of radium, and have detected bands in the ultra-violet corresponding to the negative glow spectrum of nitrogen. Crookes and Dewar, according to a statement made by the latter at the British Association Meeting, 1903, have shown that these bands are not produced unless nitrogen is present in the atmosphere to which the radium is exposed. In vacuo, and also in pure helium, the spectrum of the luminosity is continuous and no bands are present.

Volume of the Radium Emanation.—Continuing the research on the production of helium from radium, Ramsay and Soddy have made a determination of the volume occupied by the emanation produced from a known quantity of radium in a known time. This is even less than the volume of the helium produced, for two reasons. First, the emanation is much denser, and, secondly, the quantity does not continuously accumulate, but attains an equilibrium value when the rate of production equals the rate of disintegration. Since (p. 131) $q_0=\lambda N_0$, the equilibrium quantity N_0 is the quantity produced per second, q_0, divided by the radio-active constant. Or, since $1/\lambda$ represents the average life of the atom, the equilibrium quantity is the amount produced in a period equal to the average life, which for the radium emanation is 5·3 days.

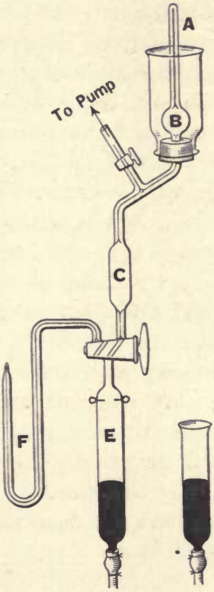


Fig. 38.

The apparatus employed is shown in Fig. 38. The hydrogen, oxygen and emanation that have accumulated a known time in a previously exhausted bulb containing the radium solution are introduced into E and exploded. A small excess of hydrogen always remains. In E a lump of solid caustic potash floats on the surface of the mercury and absorbs carbon dioxide. C is a phosphorus pentoxide tube, B is a small bulb that can be cooled externally by liquid air, and A is a piece of fine thermometer tubing sealed at the upper end. The whole is carefully exhausted, the pump connection closed, and the hydrogen and emanation admitted into the cooled bulb C. The hydrogen is then pumped out completely and mercury allowed to flow up from E until it cuts off the pump-connection. B now contains only the condensed emanation freed from all other gases. The liquid air is removed, the emanation allowed to volatilise, and mercury allowed to fill the bulb and compress the emanation into A. At atmospheric pressure the volume occupied was found to be excessively small, and it was measured under considerably reduced pressure. The equilibrium quantity of emanation from 60 mgs. of radium bromide was found to occupy between 0·03 and 0·04 cubic mm. at 0°C. and 760mm. The emanation produced in 5·3 days by 1 gramme of radium (element) therefore occupies a volume of about 1·3 cubic mm.

The behaviour of the tiny bubble of gas was investigated from day to day. In one experiment the volume steadily diminished with time, roughly at the same rate and according to the same law as the activity decays. In about three weeks the volume had shrunk practically to zero, being less than 1 per cent. of that initially occupied.

The following table shows the decrease of volume measured in this experiment:—

Volume.	Cubic mm.
After 1st day	0·027
„ 3rd day	0 011
„ 6th day	0·0063
„ 9th day	0·0041
Final volume	0·0004

This uniform decrease of volume is, however, not always observed. In some experiments the volume increased, but whether this is due to surface films of gas on the glass or other disturbing cause or is a real effect has not been settled.

Rutherford has suggested that the α particle is an atom of helium. The mass of the particle, according to his preliminary determinations, is 1·6 times that of the hydrogen atom, whereas the mass of the helium atom in terms of hydrogen is 4. It seems likely that this discrepancy may be accounted for, and that the α particle will ultimately be shown to be a helium atom. It seems certain that the α particles of all the radio-active bodies are likely to be of the same mass, and that the comparatively narrow limits in which they differ in penetrating power, &c., result from their possessing different velocities. As Strutt has pointed out in a recent Paper (*Proc.* Roy. Soc. 1904, 73, p. 191), helium is very abundant in minerals like monazite, which contain large quantities of thorium, but very little radium. This hypothesis would, therefore, lead to the belief that helium is a common product in the disintegration of all the radio-elements.

The diminution of the volume of the emanation to zero is well explained on this view, for, if the helium is produced from the emanation in the form of projected α particles, it is to be expected that the latter will bury themselves in the glass, owing to their enormous velocity. The experiment was, therefore, performed of "washing" out the thermometer tube with oxygen after the emanation had spent itself, re-exhausting and heating the glass. On passing a discharge through the tube, by means of external tin-foil caps attached to the ends, the helium spectrum was clearly seen, but the tube punctured before it could be confirmed. The experiments are difficult, and the results, in so far as they bear on the question as to whether the α particle is an atom of helium, must be considered only provisional at the present stage.

It may be well to define the limits of our present knowledge with regard to the disintegration of the radium atom. At each stage we recognise without difficulty (1) expelled radiant particles, by means of their kinetic energy; (2) unstable transition-forms, on account of their radio-activity. Independently, helium has been recognised as a product, on account of its excessively sensitive spectrum reaction, and it is possible this may ultimately be identified with (1). It is important to note that we should not be able to detect, even if it were being simultaneously produced, any stable, and therefore (presumably) known, element not expelled in the form of radiant particles, unless it were as easily detected as the element helium. The interesting experiments of Sir William and Lady Huggins, in which the nitrogen spectrum was detected in the radium glow, may ultimately prove to have a bearing on this question.

A point of great theoretical importance is whether the mass of the products of the disintegrating atom equals the original mass, and this cannot be attacked on the assumption that the recognised products are the sole products. On the other hand, it can be directly tested experimentally by looking for a spontaneous change of weight in a quantity of radium kept under conditions in which the products of disintegration cannot escape. From the recent views, dealt with at the end of Chapter III., on the nature of inertia and its possible dependence on the internal energy of the atom, it is indicated that the law of the conservation of mass will not apply to the case of atomic transformation. Considerations such as these serve to show how useless it is to attempt to find numerical relations between the atomic weights of the elements of the Periodic Table. It is notorious that all such efforts have been fruitless, but it is only recently that the reasons for the failure have been indicated.

CHAPTER XI.

THE ENERGY OF RADIO-ACTIVE CHANGE.

Energy Evolved by Uranium—by Radium—by the Radium Emanation.—Energy Evolved on Disintegration a million-fold greater than during Chemical Changes.—Average Life of the Radium Atom.—Theoretical Estimate.—Experimental Determination.—Internal Energy of Radium Atom.—Probability that all Atoms possess great Internal Atomic Energy.

The first measurements in this connection are those of Rutherford and McClung (*Trans.* Roy. Soc., 1901, 196, p. 55), who deduced from a determination of the energy required to produce an ion in air, and from the number of ions produced by uranium, that 1 gramme of uranium oxide must radiate every year energy equivalent to 0·03 calorie as a minimum estimate. This amount would raise a weight of water equal to that of the uranium compound 1°C. in 30 years. If the radioactivity of radium in terms of uranium is taken to be 10^6, the heat evolved by this element would raise its own weight of water 1 deg. in 15 minutes. This has proved to be much below the truth.

The energy emitted by radium compounds has recently been experimentally determined by Curie and Laborde (*Comptes Rendus*, 1903, 136, p. 673). Two methods were employed. In one a known quantity of the radium compound was placed in a Bunsen ice calorimeter, and the heat evolved in a definite time measured by determining the quantity of ice melted in the usual way. In the second the radium was placed in a metal block, and the temperature of the block above that of the surrounding air-bath determined. The radium was then replaced by a coil of wire that could be electrically heated, and the current measured which it was necessary to pass through the coil to maintain it at the same temperature above the surroundings as was maintained by the radium.

The quantity of heat supplied in unit time could be determined from the electrical measurements, and this was equal to that supplied by the radium. Curie and Laborde give, as the result of their measurements, that 1 gramme of radium evolves about 100 gramme-calories of heat per hour, or sufficient to raise its own weight of water 1 deg. in 36 seconds. In about 40 hours sufficient energy is evolved to decompose its own weight of water completely into its constituents, hydrogen and oxygen. Now, in this reaction the change of hydrogen and oxygen into water, or vice versa, more energy is involved, weight for weight of matter employed, than in any other chemical reaction known. Hence, in two days the radio-active change of radium evolves more energy than is ever evolved by the same quantity of matter in ordinary chemical change. The change of radium, however, in two days is quite inappreciable by ordinary measurement.

On the disintegration theory, it is to be expected that the heat-emission from a radio-active substance will be proportional to the α radiation from it. For the latter is a measure of the amount disintegrating. This conclusion has been directly proved by Rutherford and Barnes (*Phil. Mag.*, 1904, VI., 7, p. 202). They measured the heat effect from a quantity of solid radium bromide which had been kept for some weeks in the solid state. They then removed the emanation by heating it, and condensed the emanation by liquid air in a glass tube, which was then sealed. They found that the heat evolution from the radium was much reduced by this treatment, and decreased further for the first few hours, as the imparted activity in the radium compound decayed, until a minimum of about 25 per cent. of the original was attained. From this point the evolution of heat commenced to slowly increase. The emanation sealed up in the glass tube gave, on the other hand, a large heat evolution, so that the sum total of the heat given out by the radium and the emanation together was at any time equal to that given out by the radium originally. In a few hours after separation the heat effect of the emanation, and of the activity imparted by it to the walls of the tube, equalled 75 per cent. of that originally given by the radium. The heat effect from this point slowly decreased at the same rate as the activity of the emanation decayed. Therefore, the heat effect is at all times proportional to the α-radiation, and follows the same changes as the latter when the emanation is removed. The actual quantity of emanation obtainable from radium is almost infinitesimally small, being for any available quantity of radium compound a practically invisible bubble of gas. It was deduced on p. 162 that the volume of emanation present in 1 gramme of radium, when equilibrium is attained, is about 1·3 cubic mm. It, together with the imparted activity produced by it, evolves 75 calories per hour, and the total evolution of heat is given by multiplying the emission per hour by the average life $1/\lambda$, expressed in hours, and is, therefore, about 10^4 calories. 1 cubic cm. would, therefore, evolve 7×10^6 calories during its complete life. This is a purely experimental result, and is independent of all hypothesis. The energy liberated on explosion by 1 cubic cm. of hydrogen and oxygen, in the proportion required to form water, is about 2 calories. The energy of the disintegration of the radium emanation is thus several million times greater than the energy of explosion of an equal volume of hydrogen and oxygen. This, it would seem, is an unanswerable argument in favour of the view that the energy emitted by radium comes from the internal energy of the atom when it disintegrates, and is not derived from a hypothetical external source, which radium alone has power to respond to.

The argument may be pushed one stage further. It is probable that by far the greater part of the energy liberated in disintegration appears as the kinetic energy of the α-particle expelled. For, if one atom of radium disintegrates into one α-particle and one atom of emanation, by a well-known mechanical principle, after disintegration the momentum of the α-ray particle will be equal and opposite to that of the emanation particle. The velocities will, therefore, be inversely proportional to the masses. From evidence of the rate of diffusion of the emanation, which has been discussed (p. 157), it is probable that the atom of emanation is at least 50 times heavier than that of the α-particle, and the velocity of the latter will, therefore, be 50 times greater. The kinetic energy is, however, proportional to the square of the velocity, so that the kinetic energy of the α-particle must be at least 50 times greater than that of the rest of the atom. If more than one α-particle is produced by the disintegration, the probable result would be that an even less proportion of the energy is received by the heavier particle, for the expulsion of two α particles in opposite directions would neutralise each other's effect, leaving the heavy nucleus at rest. Hence, on the disintegration theory, all but a few per cent. of the heat effect should be due to the bombardment of the matter of the radium and the walls of the containing vessel by the α particles expelled.

The energy emitted by one gramme of radium per hour is 100 calories $= 4{\cdot}2 \times 10^9$ ergs. This equals the kinetic energy, $\frac{1}{2}mv^2$, of the α-particles expelled, where m is the mass of the α particles expelled per hour and v their velocity. Since, from the measurements of Rutherford (p. 79), $v = 2{\cdot}5 \times 10^9$ cm. per second, we can find m.—$4{\cdot}2 \times 10^9 = \frac{1}{2}m(2{\cdot}5 \times 10^9)^2$. The mass of the α particles expelled from a gramme of radium per hour is, therefore, $1{\cdot}34 \times 10^{-9}$ gramme, and per second is $3{\cdot}73 \times 10^{-13}$.

Average Life of the Radium Atom.—To find the mass of radium breaking up per second it is necessary to know how many α particles are expelled from each atom. Now, in the case of radium there is known to be at least four stages in the disintegration, each of which occurs with the expulsion of at least one α particle. Hence four is the minimum possible number of α particles produced from each atom in this case. The mass of the α particle is 1·6 ($H = 1$), and at least four, representing a total mass of 6·4, are expelled from each radium atom of mass 225. Hence the mass of the radium breaking up per gramme per second is given by $(225 \div 6{\cdot}4) \times 3{\cdot}73 \times 10^{-13} = 1{\cdot}3 \times 10^{-11}$ gramme. This is a maximum estimate of the proportion of the radium breaking up per second, which by definition is λ, the radio-active constant of radium. The average life is $1/\lambda$, and is, therefore, about 2,450 years, as a minimum estimate.

A completely independent determination, which is free from any important assumption, follows from the volume of the emanation. The assumptions made are: (1) That the molecule of the emanation is monatomic, like argon, which seems likely, since it shows no power of chemical combination; (2) That only one atom of emanation results from one atom of radium, which follows from the high density, as shown by the co-efficient of diffusion, and from (1). If the density is taken as 80 (H = 1), the atomic weight must be 160, and not more than one atom of emanation could be produced from one atom of radium (225). These assumptions, even if untrue, can hardly affect the *order* of the result obtained.

One gramme of hydrogen occupies the volume of 11·2 litres, and, if its molecule were monatomic, would occupy 22·4 litres. One gramme of radium, if it could be obtained in the form of a monatomic gas, would, therefore, occupy a volume of $22{\cdot}4 \div 225 = 0{\cdot}1$ litre $= 10^5$ cubic mm. One gramme of radium produces 1·3 cubic mm. of emanation in 5·3 days, and, by Avogadro's law, therefore, the ratio of the number of atoms of emanation to the number of atoms of radium is 1·3 : 100,000, which gives the proportion of the radium undergoing disintegration in 5·3 days. The value of λ is, therefore, about $2{\cdot}8 \times 10^{-11}$, and the average life of the atom is 1,150 years. Although this value is not likely to be very exact, and must be regarded as a preliminary estimate, it is probable that, at least, it approximates to the truth. This is in fair agreement with the theoretical estimate obtained indirectly above, and the result indicates that probably the assumption that only one α-particle is expelled at each disintegration is correct. For, if more than one were expelled, the theoretical estimate would be still further removed from the experimental value. The same would hold if the emanation were considered to possess a polyatomic molecule. On the other hand, if the single atom of radium were assumed to produce two atoms of the emanation, the value found for the average life of the radium atom must be doubled, and this would agree almost exactly with the theoretical estimate. But the data are not sufficiently accurate to render this deduction legitimate, for, from the nature of the experiment, the volume found for the emanation was the *maximum* volume, and any trace of other gas present as impurity would tend to make the value greater than it should be. If the nature of the data are considered, the agreement between the two values must be considered to be as close as could reasonably be expected.

Internal Energy of the Radium Atom.—Since $2{\cdot}8 \times 10^{-11}$ is the fraction of the radium disintegrating per second, and 1 gramme of radium evolves 100 calories per hour, it follows that 100 calories result from the disintegration of $3{,}600 \times 2{\cdot}8 \times 10^{-11}$

grammes of radium. The total energy evolved in the complete disintegration of 1 gramme of radium is, therefore, about 10^9 calories. The energy produced in the formation of 1 gramme of water from its elements is approximately 4×10^3. We thus see that, in the atomic disintegration of radium, about 250,000 times as much energy is produced as is produced in any known chemical change. The question naturally arises whether this energy is only possessed by radium, or is not common to heavy elements in general. The normal character of radium as a chemical element, and its close resemblance to the other elements of the same family which are not radio-active—viz., barium, strontium, &c.—points to the conclusion that the internal atomic energy of the inactive heavy elements is probably of the same order of magnitude as in radium, but the absence of change in these cases prevents our obtaining any information concerning it. In the case of uranium and thorium, the internal energy must be regarded as of a similar order of magnitude to that of radium, and their relatively feeble radio-activity is to be ascribed solely to their slow rate of disintegration. These considerations force us to the conclusion that there is associated with the internal structure of the atom an enormous store of energy which, in the majority of cases, remains latent and unknowable. For the heaviest elements the property of spontaneous atomic disintegration reveals its existence and enables its amount to be calculated.

CHAPTER XII.

ANTICIPATIONS.

Five Main Lines of Enquiry at Present Indicated—(1) The Maintenance of Radium and Polonium in the Radio-active Minerals.—The View that these Elements are Transition-forms.—Evidence with regard to Polonium.—The Present Stage of the Enquiry with regard to Radium.—(2) The Nature of the Atom.—The Meaning of the Law of Radio-active Change.—The requirements of Chemistry.—Differences between Individual Atoms of the same Element.—Deduction that the Component Parts of the Atoms must be in Violent, Irregular Motion.—(3) The Law of the Equivalence of Electric Charges.—Its Applicability to Sub-atomic Change.—Evidence of the Simultaneous Production of two Positive Charges.—The Problem of Chemical Valency.—(4) The Age of the Earth.—The Controversy between Physics and Biology.—Lord Kelvin's Estimate.—Consideration of the three Arguments in light of Recent Knowledge.—A Great Extension of the Older Estimates allowable.—Estimate of Maximum Age of the Earth based on Radio-active Considerations.—10^{10}-year Limit.—(5) The possibility of the Reconstruction of the Heavier Elements.—The Difficulty of the Source of Available Energy.—The Applicability of the Second Law of Thermodynamics to Sub-atomic Changes.—Clerk-Maxwell's "Sorting Demon."—The Two Alternative Views.—The possibility of Cyclic Evolution in Cosmical Processes.

The theory of atomic disintegration, at first put forward on radio-active evidence alone, has received such a full measure of direct experimental confirmation that it will be well to devote this final chapter to the consideration of some features of the theory which are clearly indicated at the present time, but which have not yet received experimental verification. The most important of these have to do (1) with the means by which the quantity of radium (and polonium) is maintained in the earth, (2) with the nature of the chemical atom and closely bound up with it, (3) the question as to whether the equivalence of electric charges is maintained during atomic disintegration, (4) the error in the older physical estimates of the possible age of the earth, and (5) the possibility of a continuous cycle of cosmical evolution to which the second law of thermo-dynamics does not apply.

The determination of the average life of the radium atom (p. 169) shows clearly that in the course of only a few thousand years the quantity of radium existing in a mineral would be reduced to practically nothing unless a concomitant process of reproduction were taking place within the mineral. In the case of polonium the same would occur in the course of only a few years. The readiest explanation is to suppose that the radium and polonium atoms are themselves really metabolons (in the sense of having been themselves produced by sub-atomic change), but with a somewhat more extended period of life than in the other cases. From this point of view radium and polonium are merely slow-changing transition-forms in the disintegration of a heavier and much more slowly changing parent radio-element, as, for example, uranium or thorium.

Consider the case of a disintegration series in which there is a parent element, A, disintegrating at an excessively slow rate through successive more rapidly changing transition-forms, B, C, D, E, the respective rates of change being λ_A, λ_B, &c. If N_A, N_B, &c., represent the number of atoms of each type present when the condition of radio-active equilibrium is attained, the total radio-activity is the sum of $\lambda_A N_A$, $\lambda_B N_B$, &c. A little consideration will show that $\lambda_A N_A = \lambda_B N_B = \lambda_C N_C$, &c., since at equilibrium the amount of each type changing equals the amount produced. In this reasoning it is assumed that one atom of A produces one atom of B, &c. Hence the amount of each transition-form accumulating is inversely proportional to its rate of change or directly proportional to its average life. If the parent element A exists in a natural mineral the quantities of B, C, D, &c., present in the mineral will be proportional to their average lives.

Suppose now that B, C, D are rapidily changing types, and that E is a very slowly changing type. It follows that when the mineral is separated into its various constituents by chemical analysis, E will not appear as a transition-form like

B, C, D, but as a new radio-element whose activity is sensibly permanent. Whether E is detected by its ordinary chemical reactions depends simply on its quantity, and this we have seen is the greater the longer its average life. It thus appears that the existence of a slow-changing transition-form at the end of a series would probably be overlooked in work with only small quantities of material, for both its radio-activity and actual quantity will be relatively small. But when tons of a mineral containing the parent element are worked up these forms may be separated, and will appear as new powerfully radio-elements possessing sensibly permanent activity.

The application of these considerations leads to the view that polonium and radium may both be transition-forms in the disintegration series of uranium. The rate of change of uranium, judging by the radio-activity of unit weight compared to that of radium, is about one million times less than the rate of change of radium. Hence in a mineral in radio-active equilibrium the quantity of uranium should be about one million times the quantity of radium. The rate of change of polonium is about one thousand times that of radium, and the quantity of polonium should be about one-thousandth of the quantity of radium, or 10^{-9} of the quantity of uranium. The work of Mme. Curie, Giesel and Marckwald on the proportion of radium and polonium in pitchblende (Chapter II.) supports the view that radium and polonium are produced from uranium.

Consider, first, the case of polonium, and what would occur if it were being produced as a transition-form in a disintegration series. Owing to its comparatively slow rate of change, the activity of the polonium for short periods of production would be very feeble compared with the activity of the earlier-produced and more rapidly changing transition-forms in the same series. But if the process of production were extended for some years, the activity of the polonium would increase to an equilibrium value comparable with that of the other types. This suggests that possibly Mme. Curie's discovery of the "induced activity of radium with slow rate of dissipation" (p. 140) is caused by polonium, and that this type results as the product of the change of the matter causing the ordinary imparted activity of radium. Consider some definite quantity of the latter changing into polonium. After the change is complete the relative activities of the polonium and the matter producing it may be expected to be of the same order as the relative rates of change of the two types. Since for polonium $\lambda = 2 \times 10^{-8}$, and for the last stage of the imparted activity matter $\lambda = 4 \times 10^{-4}$, the activity of the polonium should be about 5×10^{-5}, or $\frac{1}{20000}$th of that of the imparted activity matter producing it. Now Mme. Curie found that the residual activity left after the ordinary imparted activity of radium had completely decayed was about $\frac{1}{20000}$th of the original activity. The agreement is certainly striking, but it must be borne in mind that sufficient data are not available as to how the above experimental value was obtained to make it clear whether these considerations hold. The ratio of the two activities must be determined for only a short period of production of the imparted activity. If a negatively-charged wire were left in a vessel containing the emanation for several weeks until the latter had completely changed, the residual activity left on the wire will represent the result of the change of the total emanation, and the ratio of the initial activity of the *emanation* to that of the residual activity will then be the ratio of the rate of change of the emanation (not the imparted activity) to the rate of change of polonium, and would be far higher than $\frac{1}{20000}$th. If, on the other hand, the wire had been exposed to the emanation for only an hour or two, then Mme. Curie's result is to be expected.

The view that polonium is a disintegration product of radium is at first sight strongly confirmed by some experiments of Giesel (*Ber. d. Deutsch. Chem. Ges.*, 1903, p. 2,368). Giesel repeated the method applied by Marckwald for the separation of polonium from pitchblende, and kept pieces of bismuth, palladium and platinum respectively in solutions of powerful radium preparations. He obtained a sensibly permanent activity, consisting only of α rays, from the metals after they had been freed from every trace of radium and were left to themselves for a few days. He further found that the radio-active deposit was formed on a wire held in the air above the solution, which is in accordance with the above conclusion that polonium is produced from the matter causing the induced activity, which itself is produced from the emanation, which would diffuse into the air above a radium solution.

Now the characteristic of the polonium radiation is that it consists only of α-rays. In some experiments performed by the Author on the character of the radiation from the residual activity left by the radium emanation, it was found that both β and α-rays were present, although the former were not very obvious and the ratio of the two types was much less than in the case of uranium. It is possible that these β-rays result from intermediate products, and that they will be found to have decayed when the radiations are re-examined after a sufficient interval. Or, possibly, we have here a case of the simultaneous production of two different types of active matter in the same disintegration. But on this account the question as to whether polonium is the last type produced in the disintegration of radium still remains open. A conclusive proof could only be obtained by comparing the rate of decay of the activity of the two types, and this must of necessity take several years.

With regard to the case of radium, the Author has recently found that the quantity of radium produced by uranium in the first year of accumulation is not $\frac{1}{100000}$th part of the quantity that would be expected to result, assuming a direct change of uranium into radium. The method was to completely free 1kg. of uranium nitrate from radium by precipitating barium in its solution by means of sulphuric acid. It was found that the barium sulphate drags down even the last traces of radium present. The test for the latter was the emanation, which was allowed to accumulate about four days in a closed vessel before being sent into an electroscope. As uranium does not give an emanation, the presence of radium could thus be very simply detected. After the lapse of about a year the quantity of radium present, using the same test, was found to be less than 10^{-11} gramme. If the average life of uranium atom is assumed to be 10^9 years, in 1kg. of the nitrate about 5×10^{-7} gramme would change per year, and if this had turned into radium it would have been 100,000 times as much as could have been detected under the conditions of measurement. Hence, the source of radium in minerals still remains a completely open question. It is possible that if an intermediate form (other than uranium X, which changes too quickly to affect the result) exists between uranium and radium—as, for example, actinium—with a very slow rate of change, that it would account for the above result. The same explanation might also account for the known fact that some minerals containing uranium appear to contain but little radium, for if the matter passed through a gaseous stage (or an emanation) before the radium was produced, the amount of the latter present in a mineral would depend upon whether the emanation was able to escape or was accumulated by the mineral. But it seems more probable from the available evidence that uranium is not the parent element of radium, and the question remains at present unsettled.

The Nature of the Atom.—It has been pointed out that the law followed by radio-active change, that a constant proportion of the total number of atoms changes in unit time, is that followed by a monomolecular chemical reaction. It is difficult to find a true case of monomolecular reaction analogous to radio-active change. Many so-called monomolecular reactions are polymolecular, in which one molecule is present in overwhelming preponderance, as in the historic case for which the law was discovered by Wilhelmy—the hydrolysis of cane sugar in aqueous solution.

The true monomolecular reactions which have been studied accurately are nearly all endothermic, *i.e.*, proceed with absorption of heat, instead of with evolution as in radio-active change. An example is the dissociation of the diatomic iodine molecule into single atoms. The course of a monomolecular reaction proceeding with heat evolution cannot be accurately studied, because the heat produced itself increases the rate of change, which often proceeds to explosion, as in the case of the decomposition of any high explosive. Chemistry, therefore, does not offer much assistance in the inquiry as to the physical meaning of the law of radio-active change. Since external conditions do not affect the rate of change, and the change proceeds with evolution of energy, the question naturally arises as to the controlling influence which regulates the speed of the reaction and ensures that only a certain fraction of the total number of systems shall change in unit time. In short, why do not all atoms of a radio-active substance break up together, since all ultimately break up, and in so doing evolve vast quantities of energy ? Obviously there must exist individual differences between the atoms, and, since temperature and other influences are without effect, these differences cannot be merely due to external differences, such as varying speed of translation, &c. They must be sought for *within* the atom structure.

This is an absolutely fundamental conclusion in its bearing on chemistry. The atomic hypothesis of Dalton, although frequently associated vaguely with other quite alien considerations as to the stability of atoms, differed in one essential particular from earlier atomistic notions, prevalent as far back as recorded history. The atoms of one type of matter, or element, according to the Daltonian theory, are *all alike*, and different in essential and recognisable properties from the atoms of any other type. Until the discovery of radioactivity this held true without exception, although in the case of the rare-earth elements the differences are so slight—the different types seeming to merge almost continuously into one another—that the validity of the law might be questioned. In the case of the radio-active elements, although for ordinary properties they may be considered to be among the best defined and most sharply distinguished of the Daltonian atoms, their additional property of disintegration reveals an undoubted difference between the individual atoms. An example will serve to emphasise the point. It was shown on p. 125 that the average life of each typical unstable atom is a constant well suited for its experimental identification. Yet, if a quantity of a homogeneous radio-active element is considered from *any* instant, some of the atoms break up in the first second, and their life is therefore something less than a second, whereas, even after infinite time, some atoms will, theoretically, survive. The actual life of an atom of radium has all values between zero and infinity. This precludes the idea that the atom structure is similar to an isolated solar system in which the parts go through cycles of motion unperturbed by external agencies. If disintegration occurred at a certain orientation of the planets, then, although in a collection of a large number of such systems all possible orientations might be imagined to exist at the same instant, and some would therefore disintegrate immediately, the maximum life of any system could not exceed a certain value, represented in an elementary case by one complete cycle. The proportion of the total changing would increase with lapse of time, for a definite number rather than a definite fraction would come into the unstable position every second. The average life of the systems would depend on the extent of their previous history, whereas the most fundamentally remarkable feature of radio-active systems is that the average life is a constant, independent of the length of time the systems have survived. Hence, the idea of such solar systems is excluded. It is, also, quite untenable on purely chemical grounds. For, if cycles of motion existed—the orientation of the atom passing through a defined course and returning again to its initial position after a finite time—chemical differences should exist between the individual atoms, and it should be easy by chemical analysis to separate the homogeneous elements into groups of similar phase, the difference of the groups disappearing slowly with lapse of time. Possibly such an idea, or a development, might apply to the case of the rare-earth elements already alluded to.

The question remains, Is it possible to frame a first view of atomic structure which shall be consistent with the law of radio-active change and the well-known demands of chemistry? It is simpler to consider the latter first. We have seen that no slow-recurring cycle of internal motion can be entertained. Nevertheless, individual differences must be admitted. There would seem no escape from the conclusion that the internal parts of an atom must be in extremely *rapid* motion, so that, even although chemical differences exist between atoms of different phase, and these are capable of separation by suitable processes, the reversion of the opposed phases to the general average must take place so quickly that no difference of properties between the two parts after separation is detectable Hence the conclusion that the internal structure of an atom is in violent motion, which is, of course, one of the first principles of the electronic view of the constitution of matter, can be deduced from quite independent considerations.

The bearing of the idea on chemical theory cannot be discussed here. It will be seen at once that chemical properties must be regarded as average properties, the individual atoms varying continuously in nature between certain limits, which need not necessarily be narrow. Whether this will assist the elucidation of any of the problems of chemistry remains for the future.

The position is reached that rapidly recurring motions within the atom, giving rise to orientations exhibiting individual differences, but reverting to a general average in extremely short intervals of time after separation, can, at least, be entertained. The problem remains as to how the slow periods of radio-active changes are brought about. The mutual action of the systems on one another would seem to be excluded, otherwise radio-active change should vary with concentration. Hence the case may be considered as between the single atom and its action on the universal ether. It would appear, from the nature of the case, that the cause of the disintegration cannot be due to any regular or continuous action between the matter and the ether, which produces a permanent effect on the internal nature of the atom. If, for example, one supposes, with Sir Oliver Lodge, that the reaction between the revolving parts of the atom and the ether gives rise to an electromagnetic wave-radiation, which diminishes the total internal energy to a point below which disintegration is possible, it is difficult to see how the average life of the systems surviving at each instant is constant and independent of the extent of their previous existence. It seems certain that no permanent alteration of a gradual character can occur in the structure of an atom with lapse of time. An atom that has just failed at one instant to disintegrate may in the next instant be as far removed as any from the unstable condition. In other words, the action appears to be due to "chance"—*i.e.*, the orientation assumed at one instant has no determining influence on the orientation about to be assumed at the next instant. The conclusion is thus arrived at that the internal movements of the atom must be highly irregular and cannot follow a definite sequence if the law of radio-active change is to hold good. The unstable position appears to be rather the result of a chance collocation of the parts than to be due to the operation of any simple law. An analogy might be drawn from the kinetic theory of gases, in which certain of the molecules are regarded as possessing momentarily much higher and others much lower temperature than the average, and the acting causes are so complex that, although the proportion of the whole at any temperature may possibly be calculated when the total number of systems is exceedingly great, the individual history of any one molecule is quite indefinite. In a radio-active substance a definite fraction of the total assumes a peculiar orientation and disintegrates in each second, but the life of any single atom is quite indefinite. The causes at work appear to be so complex that the results can only at present be described as "chance" or "accidental" happenings, in the sense of being impossible to predict. The conclusion is reached that the structure of the atom must be in excessively rapid and *irregular motion.* This is a step further than purely electrical considerations at present indicate.

Atomic disintegration is of such a completely novel character, and is so far removed from all ordinary atomic and molecular phenomena, that none of the fundamental laws which regulate the latter necessarily apply, by analogy, to the former. It was pointed out on p. 164 that it is not to be expected that the law of the conservation of mass will hold true for radio-active phenomena. The work of Kaufmann may be taken as an experimental proof of the increase of apparent mass of the electron when its speed approaches that of light. Since during disintegration electrons are expelled at speeds very near that of light, which, after expulsion, experience resistance and suffer diminution of velocity, the total mass must be less after disintegration than before. On this view atomic mass must be regarded as a function of the internal energy, and the dissipation of the latter in radio-activity occurs at the expense, to some extent at least, of the mass of the system.

The Law of the Equivalence of Electric Charges.—Of other generalities which have found universal acceptance in the domain of molecular physics, the fundamental law of electricity, that in an originally electrically neutral system positive and negative electricity always result together in equal quantities, remains to be considered with reference to radio-activity. It must be understood that the evidence at present available is little more than sufficient to justify the question being raised. Although apparently pointing in one direction, so many possibilities arise, which have not yet been critically examined, that the bearing of the evidence must be accepted with reserve. The main phenomenon of radioactivity has been shown to consist in the successive expulsion of positively charged particles from an originally neutral atom. The deduction, therefore, is that the atomic residue after each expulsion should be left negatively charged. In the cases which have been most completely examined it is found to be left *positively* charged, so that it would appear that there is the simultaneous production of two positive charges during many cases of atomic disintegration. The best established cases are those, worked out by Rutherford, of the change of the emanations of thorium and radium into the matter causing the imparted activity. It was pointed out (p. 141) that the emanation atom, whether charged or not at the moment of formation, rapidly becomes uncharged, and at the moment of disintegration must be considered neutral. In the case of radium the positive charge carried by the α particle expelled has been proved by Rutherford by the direction of the deviation of the particle in a magnetic field. All the α particles from radium are deviated as though positively charged, and since about 40 per cent of the total α radiation is, under ordinary circumstances, derived from the emanation, the α radiation of the emanation must carry a positive charge. The matter produced from the atom after the expulsion of the positive ray is also positively charged, and moves in an electric field to the negative electrode. The same holds true for the thorium emanation and imparted activity. In other cases there is evidence that the residual system is left positively charged at the moment of the expulsion of the ray.

Prof. J. J. Thomson* recently performed the following experiment:—

A quantity of radium was completely enclosed in a block of lead to which two gold leaves were externally attached. The system was suspended by a quartz fibre in a highly exhausted space. It was found that if the lead was of sufficient thickness to completely absorb the β radiation the leaves did not charge. With smaller thickness of lead the leaves diverged with a positive charge as in Strutt's apparatus (Fig. 19). This would seem to prove that the positive and negative electricity produced during the disintegration are equal and neutralise one another.

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* Private communication.

It must be pointed out that no direct evidence has yet been obtained of the positive charge carried by the α particle. Wien, in some recent experiments, sought to perform a similar experiment to that of M. and Mme. Curie for the β-rays of radium (p. 73) for the α-rays, but failed to obtain a positive result.

The reason why these considerations are here introduced is to draw attention to the bearing of the evidence on the problem of chemical valency. Assume, for the sake of example, that the α particle is an atom of helium. The disruption of the divalent radium atom produces two non-valent atoms—helium and the emanation. This is such a novel change that the question may reasonably be asked whether it is right to assume that the electrical neutrality of the system is preserved under these conditions. We have at present no certain knowledge of the meaning either of electrical neutrality or valency. Is it necessarily to be assumed that the older ideas of the nature of the electric charge will apply without modification to the profound changes within the atom that occur in radio-active processes ?

The Age of the Earth.—The discovery of the enormous store of available energy latent in certain atomic structures, and the probable existence of this energy—although not yet shown to be available—in all atoms, which was discussed in the last chapter, have an intimate connection with the problems of cosmical evolution, and the requirements of biology and geology. Throughout the latter part of the last century a controversy, as to the possible age of the earth as a planet fitted for habitation, existed between two schools, represented by the physicists on the one side and the biologists on the other. Some of the arguments advanced by the former make strange reading at the present time. The case for the physicists was presented by the late Prof. Tait in some "Lectures on Recent Advances in Physical Science,"* and the following extracts may be quoted :—

"Thus we can say at once to the geologists, that, granting this premiss—that physical laws have remained as they are now and *that we know of all the physical laws that have been operating during that time*—we cannot give more scope for their speculations than about 10 or (say at most) 15 million

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* Macmillan & Co., 1895.

years. But I dare say many of you are acquainted with the speculation of Lyell and others, especially of Darwin, who tell us that for even a comparatively brief portion of recent geological history three hundred millions of years will not suffice! We say—so much the worse for geology as at present understood by its chief authorities, for, as you will presently see, physical considerations from various independent points of view render it utterly impossible that more than 10 or 15 million years can be granted." The sentence here italicised needs no further comment, in light of present-day knowledge, to decide the question in favour of the geologists. The demands of the latter, at first considered extravagant, are well within the range of even the present limited knowledge of the energy associated with matter.

Lord Kelvin arrived at the 10 million year estimate of the past age of the earth as a habitable world from three independent considerations. The first was based on the internal heat of the earth and the rise in temperature beneath the surface. From the temperature gradient—about 1°C. for every 100ft. of descent—the loss of heat from the centre outward and the temperature of the surface at any time previous could be calculated. This leads to the conclusion that not more than about 10 million years ago the surface of the earth must have been still molten. This argument rests on the assumption that the earth was a self-cooling planet, whereas we now know that it is self-heating also. If only a small fraction of the matter of the earth possessed the property of heat-evolution to only the same extent as the element uranium (which probably evolves about 1 calorie per gramme per year and lasts for thousands of millions of years), it will readily be seen that the rise of temperature from the surface of the earth inwards might have been attained by supposing it to have originally been a cold body spontaneously self-heating, rather than a hot body spontaneously self-cooling.

The second argument rested on what is known as tidal retardation. The earth rotates faster than the moon revolves. The moveable parts of the earth's surface—the oceans—are attracted towards the moon and tend to rotate slower than the rest of the earth. The consequences are twofold. In attracting the water the moon is itself attracted to an equal and opposite extent, and its period of revolution—the lunar month—is decreasing. The period of the earth's rotation—the terrestrial day—is being correspondingly increased by the friction with which it opposes the tidal movements of the oceans. At some distant date the earth will rotate in the same period as the moon revolves, and the lunar month and terrestrial day will be identical. The rate of increase of the earth's period of rotation being known, its value for any past time can be calculated. The problem was to find the time at which the earth was rotating at such a rate that it took the particular shape it possesses at present. It was assumed that the polar flattening now existing was that prevalent at the time the crust of the earth solidified, and that after this event this shape has been retained unaltered. The result went to show that the diurnal period must have been almost what it is now to have produced the existing flattening, and hence it was argued that the earth must have been fluid less than 10 million years ago. The recent work of Prof. F. G. Adams, the geologist, on the flow of crystalline rocks under pressure, would seem to indicate that the earth could not be regarded as a rigid body even if it were solid to the centre. It would seem more reasonable to suppose that the earth's shape would conform to its existing period of rotation, like a viscous fluid, exhibiting, possibly, a certain amount of lag.

Even more important is the possibility that the energy of planetary rotation is being *maintained*, possibly by the radiant pressure of light.* This most recently discovered factor in the processes of cosmical evolution has not yet been reckoned with. Like sub-atomic change, it is one of those infinitely small agencies which, acting over infinite time, may profoundly modify the conditions of the problem to be solved.

The third argument was based upon the length of past time that the sun could have maintained its radiation. Prof. Tait, in the lectures referred to, said, "Take (in mass equal to the sun's mass) the most energetic chemicals known to us and the proper proportion for giving the greatest amount of heat

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* For some mechanical experiments designed on this idea, see R. W. O. Kestel, "Radiant Energy, a Working Power in the Mechanism of the Universe" (Port Adelaide, 1898). No mathematical analysis of the effects of light pressure on planetary motion seems as yet to have been undertaken. The problem is of extreme interest.

by actual chemical combination, and, so far as we yet know their properties, we cannot see the means of supplying the sun's present waste for even 5,000 years. . . . It is quite obvious that the heat of the sun cannot possibly be supplied by any chemical process of which we have the slightest conception. . . . This question is totally unanswerable, unless there be chemical agencies at work in the sun of a far more powerful order than anything we meet with on the earth's surface." Failing any such agency Tait proceeded to fall back on gravitational energy as the source of the sun's heat. Adopting the nebular hypothesis of Laplace, that the various planets and suns are formed by the falling together of small masses originally attracted to one another from great distances, it was pointed out that the heat evolved on the impact of such masses must be of a far higher order than the same masses could produce by chemical combination, and that there was no difficulty in accounting for very long periods of solar radiation on this hypothesis. "But, on the very highest computation that can be permitted, it cannot have supplied the earth [during the past], even at the present rate, for more than about 15 or 20 million years."

The almost prophetic qualification with regard to "chemical agencies of a far more powerful order than anything we meet with" will be appreciated at the present time. It remains to be seen whether the meteoric or any similar more recent hypothesis, such as that of shrinkage, is not rendered unnecessary, or at least of subsidiary importance, by the existence of the available energy of atomic structures undergoing degradation. Undoubtedly radio-activity is but one manifestation of such energy. If all matter, on the average, were disintegrating at the same rate as uranium without exhibiting radioactivity the effects would have escaped observation in the laboratory but would still be great enough to be the ultimate controlling factor of cosmical evolution. It is to be hoped that the whole question will be gone into exhaustively in its various bearings by those able to speak with authority in their respective sciences.

With regard to radio-activity, an independent limit of the past age of the earth is set by our present ignorance of any concomitant process of atomic reconstruction. If it be assumed that no such process has been going on, at least for the last thousand or ten thousand million years, the past age of the radio-active minerals is fixed simply by the period of the average life of the elements uranium and thorium. We have seen that it is probable that all the other cases of radio-active elements will ultimately come to be regarded as products of more slow-changing elements, such as uranium and thorium. From this point of view the minerals containing a large proportion of uranium must have been formed within the period of the average life of the atom of this element. The value given for the latter—10^9 years—is somewhat uncertain, and a margin as between 10^8 and 10^9 years should be allowed. We thus see that the pitchblendes and uraninites must have been formed within, say, the last thousand million years, and possibly within the last hundred million years. With regard to the age of the earth, some further information is obtained by finding the proportion of uranium remaining at various intervals, assuming 10^8 years as the period of average life. In 10^8 years the quantity would be reduced to $\frac{1}{e}$, in 2×10^8 years to $\frac{1}{e^2}$ and so on; so that in 10^9 years the total quantity would be reduced to 0·004 per cent. of the original. In 10^{10} years it would be reduced to 4×10^{-56}. So that, even if the whole earth were originally uranium, and no reproduction has taken place, it cannot have survived more than 10^9 or 10^{10} years.

The Possibility of the Reconstruction of Matter.—The question, therefore, naturally arises as to whether this really sets the limits of the earth's history—or, indeed, of that of any body in which uranium is present; or whether it is not more philosophical to suppose that even the parent radio-elements are being reconstructed. In this supposition we are met at once with what appears to be an almost insurmountable difficulty. We know, from the energy emitted by a disintegrating atom, the absorption that must take place if that atom is to be reconstructed out of its constituents, and this is so great that it is difficult to imagine how it can be furnished from its surroundings unless a considerable area is taxed for the supply. At first sight it seems that the atomic theory, which bears out and is borne out so strikingly by atomic disintegration, opposes a barrier to any conception of atomic up-building. A gradual and continuous accretion of atomic mass, in which the energy was supplied in very small steps as available from the surroundings, seems the only process readily conceivable. But the atomic theory appears to demand equally with a *per saltum* degradation a *per saltum* accretion. This difficulty is, however, not real. We have only to account for the step-by-step increase in the complexity of atoms *as known to us*. It is not necessary to assume that it is impossible for intermediate forms (representing a practically continuous increase of atomic mass from the lightest to the heaviest known atom) to be altogether incapable of existence. All that is required is that the rate of accretion of mass should be more rapid between the points of stability as represented by the atoms of the Periodic Table. This would ensure that the intermediate forms in the up-building process, like the transition—forms in that of disintegration, would never accumulate in sufficient quantity to have been yet detected by direct methods.

The only question, therefore, is as to the source of the necessary energy. We have seen that, with the advent of the possibility of atomic changes, the applicability of the long-accepted generalisations of science—the conservation of mass, the equivalence of electric charges, &c.—to the new conditions are legitimate objects of inquiry. The principle that is now called into question is the second law of thermo-dynamics. This undoubtedly, with the others, must be put upon its trial, and its claim to universal application be tested anew. Fortunately, owing to the genius of Clerk-Maxwell, the limitations of this law have long been clearly recognised. From the kinetic theory of gases it follows that in a gas of uniform temperature the individual molecules must be moving at varying velocities, and therefore possess different kinetic energies. The temperature of the gas represents the average temperature, as measured by the kinetic energy, of the molecules. Clerk-Maxwell imagined some supernatural agency to sort the molecules into two parts, the one possessing kinetic energy higher than, and the other lower than, the average. Such a process can be imagined to be effected without the performance of work, and the gas can therefore be obtained in two portions at different temperatures. The second law of thermodynamics states that such a process as this is impossible without the performance of work. One part of a body of uniform temperature cannot grow hotter at the expense of the heat of the remainder unless work is performed upon it. Yet, according to the kinetic theory of gases, a gas in which all the molecules were originally at the same temperature, would, after a short time, consist of molecules at different temperatures, the *average* temperature only remaining unchanged.

The question therefore arises, Can the second law of thermodynamics be applied to sub-atomic change ? Or, has the law, like that of the immutability of the elements, only a limited range ? All processes of evolution at present revealed proceed in the one direction, the energy being "degraded" into heat of uniform temperature, which is no longer available for any useful purpose. A limit is thus fixed when all change must cease unless there is an upward process going on in Nature, whereby the energy passes back again into available forms. This is the real question underlying the possibility of a reconstruction of the elements with the absorption of energy. The latter is so enormous that it is difficult to imagine any other source than heat of uniform temperature, to be sufficient for the purpose.

The position reached is at least definite. If the doctrine of the transformation of energy proceeding spontaneously in only one direction is universally true, the limits of the possible age of the earth would seem to be fixed at a maximum of 10^9 or 10^{10} years. The end of evolution is definitely fixed as occurring when all the available energy shall have run its course to exhaustion. Correspondingly, a sudden beginning of the universe—the time when present laws began to operate—is also fixed. It is necessary to suppose that the universe, as a thing in being, had its origin in some initial creative act, in which a certain amount of energy was conferred upon it sufficient to keep it in being for some period of years. It is possible, it is true, to avoid the end indefinitely, since the rate of change will diminish as the end is approached, and theoretically the end will require an infinite time to be attained. But the difficulty connected with the beginning cannot be so avoided.

The alternative view, which is only beginning to appear even possible, is that the second law with regard to the availability of energy does not universally apply, and that, in the infinitely varied processes of Nature a cyclic scheme of evolution is possible. The heavy elements, on one side of the cycle, may yield a continuous supply of available energy, while the lighter elements are continuously growing, by the gradual accretion of masses, possibly of electronic dimensions, and at the same time storing up the "waste" energy produced in the opposite process. The essential law of the universe may be that the total quantity of energy is constant, and that what we call low-grade and high-grade energy is the expression of the limited means at our disposal for utilising it. If we lived in a sub-atomic instead of a molecular world possibly the significance of the terms might be reversed. The universe would then appear as a conservative system, limited with reference neither to the future nor the past, and demanding neither an initial creative act to start it nor a final state of exhaustion as its necessary termination. This point of view is, in every way, such a much more complete and satisfactory one that it is perhaps necessary again to emphasise that it is only at present a mere speculation, or perhaps one of those coming events which cast their shadows before! The really essential advance that the results dealt with in this book lead to, is that the limitations with respect to the past and future history of the universe have been enormously extended. Exactly how much time can be given to the biologist in which to work out his processes is a matter which will require some considerable discussion to adjudge. The unconscious application of generalisations which have only a limited scope to the universal problems of evolution has served in the past to restrain the legitimate aspirations of science. Out of the revolution, which the new knowledge must effect in every branch of philosophy, one thought, long nascent, at length begins to take definite shape. Each new advance increases the period of time over which the laws of Nature can be regarded as having been in continuous operation without external interference.

The limitations, with respect to a beginning and an end, which one era imposes, disappear in the next. So that it is not unreasonable to anticipate that ultimately these laws will be recognised to operate, not only universally with regard to space, which has long been admitted, but also consistently with reference to time.

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[Figures in *Italics* denote main references.]