On radioactivity, a new property of matter

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The subject which I propose to speak to you about has become, in only a few years, so vast that in order to deal with it in a single lecture I should have to confine myself to listing the main facts following the chronological order of their discovery. But as M. and Mme. Curie are to describe to you their fine work on radium, I will simply summarize the subject and give some account of my own research.

At the beginning of 1896, on the very day that news reached Paris of the experiments of Röntgen and of the extraordinary properties of the rays emitted by the phosphorescent walls of Crookes’ tubes, I thought of carrying out research to see whether all phosphorescent material emitted similar rays. The results of the experiment did not justify this idea, but in this research I encountered an unexpected phenomenon.

Of all the phosphorescent materials, uranium salts seemed particularly suitable for the investigations, because of the exceptional structure indicated by the harmonic series of the bands making up their absorption and phosphorescence spectra. Thus I placed sheets of double sulphate of uranium and potassium on photographic plates enveloped in black paper or protected by a sheet of aluminium and exposed them to light for several hours. On developing the plates, I found that the uranium salt had emitted rays which reproduced the silhouettes of the crystalline sheets through the black paper and various screens of metal or thin glass laid on the plates.

Under these conditions the phenomenon could be ascribed to a transformation of solar energy, like phosphorescence, but I soon recognized that the emission was independent of any familiar source of excitation, such as light, electricity or heat.

We were thus faced with a spontaneous phenomenon of a new order. Figure 1 shows the first print, which revealed the spontaneity of the radiation emitted by the uranium salt. The rays passed through both the black paper which enveloped the plate, and a thin sheet of copper in the shape of a cross. Figure 2 shows the radiograph of an aluminium medal; the non-uniform absorption of the radiation by the different thicknesses of metal reveals the...
effigy. As the uranium salts used had been prepared a very long time before-hand, it was to be supposed that the intensity of the phenomenon was independent of time, and hence that the emission was constant. All the later experiments have shown that the activity of uranium does not diminish appreciably with time.

In obtaining these first results I noticed that the radiation of uranium discharged electrically-charged materials located some distance away, and this phenomenon provided a second method of studying the new rays. The pho-
The two methods showed that all uranium salts, whatever their origin, emitted radiation of the same type, that this property was an atomic property connected with the element uranium, and that metallic uranium was about three and a half times as active as the salt used in the first experiments.

A sphere of charged uranium, which discharges spontaneously in the air under the influence of its own radiation, retains its charge in an absolute vacuum. The exchanges of electrical charges that take place between charged bodies under the influence of the new rays, are the result of a special conductivity imparted to the surrounding gases, a conductivity that persists for several moments after the radiation has ceased to act.

These fundamental properties of the radiation emitted by uranium were verified later by numerous investigators; of these I will only mention Rutherford, who established that the conducting properties of the gases through which the radiation of uranium passes, are completely equal to the ionization caused by other factors.

During the course of these first experiments, the observation of a number of phenomena that were hitherto unexplained led me aside from the path to which my later experiments were to bring me back. Various samples of phosphorescent calcium sulphide resting on small plates of glass and covered with a small bell-jar of glass had been laid on a photographic plate protected by a sheet of aluminium 2 mm thick, as shown in Fig. 3. The print developed after 48 hours (Fig. 4) revealed silhouettes of the plates of glass, reproduced with the details which would have been produced by refraction and total
reflection of the light rays. Although at the same time Troost and Nieweglowski observed the emission of penetrating rays by phosphorescent materials, the above experiment could not be reproduced; the cause of the momentaneous appearance and the disappearance of the activity of these products is unknown. But since, in the experiments with uranium, the silhouettes of all the screens put in between are edged with white lines outlining projected shadows, and similar to those of the print which we have just mentioned, I was led to attribute the properties of light to the radiation from uranium, while all later experiments have shown that this radiation cannot be reflected or refracted like light rays.

In 1898 Schmidt and Mme. Curie observed almost simultaneously that thorium has properties similar to those of uranium; these properties were studied by Owens and by Rutherford. Mme. Curie, after measuring, by means of the ionization of air, the radioactivity of a large number of minerals containing uranium or thorium, noted the remarkable fact that some minerals were more active than metallic uranium. After having verified the fact that the activity accompanied the uranium molecule in its various combinations, M. and Mme. Curie concluded from this that there must be present in the minerals mentioned a substance more active than uranium, and they set about isolating it. They treated one of the most active of these minerals, Joachimsthal pitchblende, and first separated from it active bismuth which they supposed contained a new substance, polonium, and then soon afterwards they obtained extremely active barium, associated with a new element, radium. These products were obtained by fractional crystallizations in conjunction with electrometer readings. The activity of the products in-
creased with their richness in new elements. The activity of pure radium is about one million times greater than that of uranium.

Giesel has succeeded in obtaining very active preparations, and Debierne has studied a product intimately associated with thorium, which he has called actinium.

Of these different preparations, radium alone has the characteristics which we associate with simple substances; it has an emission spectrum formed of lines not belonging to any other known element, and the molecular weights of radium salts increase with their radium content.

The intensive radiation of radium excites the phosphorescence of different materials and restores the ability of crystals to phosphoresce under heat, when they have lost this as a result of a previous rise in temperature; moreover, radium salts become luminous spontaneously under the influence of their own radiation.

The first samples of polonium and radium that M. and Mme. Curie were good enough to lend me, revealed a striking difference in the nature of the radiation emitted by each of these substances. They were put into small paper cylinders closed at the bottom by very thin flakes of mica or aluminium, and the cylinders were placed on a photographic plate. The print in Fig. 5 shows that the radium radiation has easily passed through the various envelopes while the polonium radiation has not penetrated the wall of the paper cylinder; thus the polonium rays are only slightly penetrating.

Towards the end of 1899, first Giesel and then Meyer and von Schweidler observed that the radiation of active preparations is deflected by a magnetic
field. At the same time, unaware of these experiments, I noted the same in the case of radium radiation, after first having noted that in a non-uniform field the radiation is concentrated on the poles, as are cathode rays in the experiment of Birkeland. Figures 6 and 7 show some pictures which were obtained in the following way:

![Fig. 6.](image)

![Fig. 7.](image)

Almost at once I realised that the rays emitted by polonium were not deflected under the above experimental conditions and that there are two kinds of rays, one highly deflected by a magnetic field, and the other apparently not deflected. When studying the emission from radium, M. and Mme. Curie observed the simultaneous presence there of the two types of rays and saw that the rays from polonium, in common with those of the same type emitted by radium, were increasingly absorbable according as they had passed through a greater thickness of absorbent material: the in-
verse occurs in the case of the heterogeneous beams of the other rays. The photograph in Fig. 8 shows the two types of rays known nowadays as \( \alpha \)-rays for those emitted by polonium, and \( \beta \)-rays for the magnetically deflectable part of the radiation from radium. I further found that thorium emits the two types of rays and that, even in a vacuum, uranium emits only \( p \)-rays (Fig. 9) not excluding the existence of much less active, non-deflectable rays.

There is, in fact, a third type of rays, \( \gamma \)-rays, not deflected by a magnetic field, attention to which was first drawn by an experiment conducted by Villard and which seem analogous to X-rays.

The action of a magnetic field enables the various components of the radiation from radioactive substances to be separated and analysed.

\( \beta \)-Rays behave like cathode rays; it may be assumed, as has been for the latter, that they are made up of masses \( m \) carrying at velocity \( v \) negative charges \( e \). In a uniform magnetic field of intensity \( H \) the trajectories normal to the field must be circular paths, the radius \( R \) of which is given by the relation

\[
RH = \frac{mv}{e}.
\]

For an original direction making an angle \( \alpha \) with the lines of force, the trajectories are helices which wind on cylinders of radius \( R \sin \alpha \). I have verified the various geometrical consequences of this similarity.

The beam of \( p \)-rays is made up of an infinity of rays with trajectories having different radii of curvature; the magnetic field disperses them as a prism disperses the light rays of various colours. A photographic plate without black paper wrapping can be placed in, and parallel to, a uniform, horizontal magnetic field, for instance, then on the plate a small lead dish containing a few grains of radiferous barium forming a source of very small diameter. The radiation is directed at the plate and produces an image on just one side. If small strips of different substances - paper, aluminium or various metals - have been placed on this side it will be found that in the diffuse image which represents a kind of spectrum there are rays with varying penetration which, under each screen, give images whose limits are different and which constitute absorption spectra. It will be seen in the following that the image is mainly caused by the secondary radiation which originates at the face where the incident rays emerge. Figure 10 is an example of the photo-
graphs obtained; the screens are: a strip of black paper, a strip of aluminium 0.1 mm thick, and a platinum foil 0.03 mm thick. To secure what might be termed a pure spectrum, i.e. such that each point on the plate is struck by a single beam, all of whose trajectories have the same curvature, the radiation from the point source must be made to pass through a narrow aperture. The result is the same as the foregoing (Fig. 11). The photograph shows, moreover, an image produced by the secondary rays emitted by the inside face of a lead semi-cylinder which covered the source and in which was pierced the small aperture in question. Finally, when the radiation is gathered through a second small aperture, the result is a single ray.

Each single ray may be defined by the value of the product RH corresponding to it. In the above tests, the RH values of the active rays ranged from 600 to about 2,500.

A more complete analysis of the radiation can be made by applying the following general method:

The active material is placed at the bottom of a deep, narrow groove in a small block of lead to produce a very thin vertical beam issuing from a linear source a few millimetres long. This system is placed in, and parallel to, the uniform field of a magnet. A photographic plate is then arranged above the
source, at right angles to the field and intercepting the deflected beam. The very oblique rays reaching the plate produce thereon an image which differs very little from the actual trajectory of a skimming ray emitted normally to the field. On the photographic plate are placed a number of screens pierced by small slits normal to the plate; these then allow either portions of pure spectra or single rays to pass. To eliminate the light emitted by the source, the latter is covered with a thin aluminium foil. In practice the screens are fixed by means of an adhesive to a glass plate which serves to press them against the photographic plate. Figures 12, 13, 14 and 15 show the arrangement of the screens and some of the photographs obtained by this method. For two of these photographs an aluminium foil 0.1 mm thick, concentric with the pierced screen, was mounted beyond the pure spectra. It can be seen that the least deflectable rays pass through this foil as if it were not there; other more deflectable rays produce, on emerging, secondary rays which are photographically more active than the incident rays, and lastly the most deflectable rays do not pass through the aluminium foil and produce at the entry face secondary rays which give an intense image. The general image within the contours of the screens is also attributable to secondary radiation. The magnetic field had an intensity of 859 c.g.s. units. In these tests the α-rays were blocked but the γ-rays give straight line images revealing a discontinuity between these rays and the less deflectable β-rays; for these latter the product RH is about 10⁴. In general the not very deflectable β-rays are very penetrating and the rays which are highly deflected are also very readily absorbed. These tests illustrate the advantages of this method for analysing the effects produced for each single ray.

When applied to secondary radiation this method has shown that the rays were deflected by a magnetic field in the same direction as the cathode rays.

While I was conducting these experiments, M. and Mme. Curie demonstrated that the β-rays from radium actually carry negative electrical charges; the bodies which receive the radiation become negatively charged while the source itself becomes positively charged. For this double phenomenon to be observed, all the conductors and the source itself must be completely surrounded by insulating materials, e.g. paraffin, or be placed in a vacuum.

On the other hand I have shown that the β-rays from radium were deflected by an electrostatic field. When F is the intensity of this field, the trajectory of a single ray, as characterized by the quantities m, e and v defined above, is a parabola with parameter \((m/e) \left(\frac{v^2}{F}\right)\), and the size of this parameter combined with that of the radius of curvature of the same ray’s trajectory in
a known magnetic field enables m/e and v to be determined. The experiment performed with the apparatus in Fig. 16 provided the image shown in Fig.17 which reveals the electrical deflection by the shadow projected by a vertical screen normal to the field. By combining for a single beam the crossed electrical and magnetic deflections, Kaufmann has made much more precise measurements than the ones that can be deduced from the earlier experiments. His measurements showed him that the ratio e/m was a function of the velocity v which, for the least deflectable β-rays, tends towards the speed of light. Interpretation of this fact in terms of Max.Abraham's concepts suggests that the mass of the electrons is at least in part, if not entirely, the outcome of electromagnetic reactions - a result which prompts fresh ideas about the nature of the inertia of matter.

Apart from the β-rays, identical with cathode rays, α-rays make up an important part of the radiation from active substances. I mentioned earlier how I had observed them for the first time with polonium, and how their apparent non-deflectability and the peculiarities of their low penetrability...
had caused them to be classified apart. By means of a very delicate electrical experiment, Rutherford discovered that the $\alpha$-rays could be deflected very slightly by a very strong magnetic field, and that the deflection was in the opposite direction to that of $\beta$-rays. They hence behave as if carrying positive electrical charges and appear identical with Goldstein's "Kanalstrahlen".

Using the above arrangement I recorded photographically the trajectory described in a magnetic field by $\alpha$-rays from radium and the rays from polonium which are identical. Figure 18 shows a photograph (enlarged) obtained in a magnetic field of 10,000 c.g.s.units. The two concurrent paths each correspond to a direction of the magnetic field which has been reversed in the middle of the exposure. They do not present any trace of dispersion, which allows us to regard the active beam as homogeneous. It is noted, moreover, that the radius of curvature calculated for the different points of the trajectory continues to increase as the path in the air increases. This phenomenon can be attributed to the fact that the positive charges, with a velocity which is ten times less than that of the $\beta$-rays and a real or supposed mass a thousand times larger, attract the neutral molecules of the air, or are discharged progressively in the ionized air.

As the $\alpha$-rays are very absorbable, they appear in consequence to constitute the most active part of the radiation when this is measured by the ionization of the air in the neighbourhood of the source. These rays are also the most active in exciting the phosphorescence of zinc blende, and of diamond, whereas barium platinocyanide becomes equally luminous under the influence of the $\alpha$- and $\beta$-rays and the phosphorescence of the double sulphate of uranium and potassium is particularly excited by the $\beta$-rays. The curious effect of Sir W. Crookes' spinthariscope should be attributed to the
cc-rays, and this effect seems due to the cleavages accompanied by flashes identical with those produced when various crystals are fractured.

The third kind of rays, $\gamma$-rays, are characterized by their great penetrability and their non-deflectability in a magnetic field. In the attached illustration (Fig. 19) made by the method described earlier, the $\alpha$-rays have been stopped near their source, the $\beta$-rays are deflected by the magnetic field, whilst the $\gamma$-rays and the light emitted by the radium salt form a rectilinear beam which falls upon a quartz prism. The luminous rays are deflected, whereas the $\gamma$-rays leave a trace which can be followed without deflection not only past the prism but even through the prism itself.

The great penetrability of the $\gamma$-rays and also of the less deflectable $\beta$-rays means that neither the ionization of the air nor the photographic plate can give an exact idea of their intensity, since they pass through the gases and the silver salts without being absorbed.

If fairly thick metal plates are placed in their path, these rays are transformed and, either on the entry face or on the exit face, give rise to secondary rays which are more absorbable, so much so that the effect observed immediately behind these screens is more intense than if this screen did not
exist. This transformation reminds one of the effect produced by placing a fluorescent screen in the path of a beam of invisible radiations.

A photographic plate receiving the radiation of radium filtered by lead 1 cm thick is affected more under a strip of lead 1 mm thick than in the regions not covered by this screen. Figure 20 shows the effect produced by the radiation leaving through the walls of a lead parallelepiped after passing through 5 to 12 mm of metal.

Fig. 20.

These secondary phenomena can account in part for the appearance of cast shadows on the edges of all the more or less transparent screens placed on the photographic plates.

Uranium and polonium emit penetrating rays which appear to be identical with $\gamma$-rays.

Radiation of radioactive substances produces various chemical actions; it acts upon the substances used in photography, and the $\alpha$- and $\beta$-rays are the most active in this respect; it colours glass violet or brown, and the alkaline salts are coloured yellow, violet, blue or green. Under its action paraffin, celluloid and paper turn yellow; white phosphorus is transformed into red phosphorus. This transformation has been noted with $\beta$-rays, but it is probable that $\alpha$-rays are equally active. Ozone is produced in the air around active bodies. Not only gases but also liquid dielectrics (petroleum, liquid air) and insulating solids like paraffin are ionized when they have been penetrated by radium radiation, and they preserve their conductive properties for a few
moments after the radiation has ceased to act. Giesel has observed that an aqueous solution of radium bromide continuously liberates oxygen and hydrogen at the rate of approximately 10 cm$^3$ per gram per day. According to Ramsay and Soddy these gases would also contain helium.

M. Curie has discovered moreover that radium salts continually emit heat; pure radium would emit approximately 80 calories per gram per hour. Curie and Dewar observed that in plunging a tube containing radium into liquid hydrogen there was a continuous release of hydrogen gas; with 0.7 g of radium bromide 73 cm$^3$ of hydrogen were obtained per minute.

Various physiological effects have been observed with radium rays; they excite phosphorescence in the interior of the eye; when an active product is brought near to the temple, a sensation of light is perceived. They act upon the epidermis and profoundly disorganize the skin, as do X-rays. The effect is produced without any sensation being felt at first and it only develops after several weeks; it then produces more or less deep lesions which can take several months to heal and which leave scars. At present an effort is being made to utilize this action in the treatment of lupus and cancers.

Radium rays have an active effect on the nerve centres and can then cause paralysis and death; they seem to act with particular intensity on living tissues in the process of evolution.

Up to now we have only spoken of the radiation which is transmitted through glass, mica, opaque bodies and metals. In the emission of radioactive bodies there is another phenomenon of a different nature which appears to be intimately connected with radioactivity, if it is not the primordial phenomenon. Thorium and radium emit energy in a particular form; the resultant activity is propagated in the form of an active vapour which has been called emanation and which is arrested by any covering, however thin, which is impermeable to gases.

This emanation seems to settle upon all bodies in order to make them radioactive, but the activity disappears when the latter are no longer under the influence of the active source, even when they are in a closed chamber. These facts were discovered simultaneously at the end of 1899 by Rutherford for thorium, and by M. and Mme. Curie for radium. Rutherford when studying the activity of thorium saw that besides the ordinary radiation there was an effect produced by an emanation comparable with an active vapour. This latter is deposited on all bodies, principally on those which are negatively charged, and it makes them momentarily active.
At the same time M. and Mme. Curie discovered that under the influence of radium the bodies become temporarily active; this induced activity persists for some time after the bodies have been removed from the radium; in the open air it diminishes by about one half in half an hour.

The phenomenon is produced with regularity and with great intensity in a closed space; the induced activity is then the same on all the bodies; it is independent of the nature of the gas and of the pressure within the chamber, but the activation is no longer produced if a vacuum is constantly maintained by removing the gases which are liberated as soon as they are produced. Solutions of radium salts produce activation with greater intensity than the solids.

The water of crystallization extracted from the active salts, or the water separated from an active solution by a semi-permeable celluloid wall, become strongly radioactive. The bodies activated by the radium produce the same effects as radium; the glass walls activated by the emanation emit a penetrating radiation which passes through them and makes them luminous, whilst the activating solution can only emit feeble radiation.

The activating property is diffused gradually through the gases inside a closed chamber, through capillary tubes or imperceptible fissures; the bodies become more active as the volume of gas at their surface is greater.

A gas which has been kept near to radium and has acquired the property of making solid bodies radioactive is itself radioactive, but it only emits rays with very little penetrating power which will not pass through a glass wall. When it is removed from the radium it continues to emit rays and to cause radioactivity. Its activity from this double point of view diminishes by half during each successive period of four days and finally becomes extinct. This period of four days is a time constant characteristic of radium emanation.

Air charged with emanation produces phosphorescence of various substances (glass, zinc blende, etc.) and this phenomenon allows various striking experiments to be conducted with relation to the propagation of emanation.

Radium emanation behaves like a gas from many points of view. M. and Mme. Curie have shown that it divides like a gas between two intercommunicating gas chambers and that it is diffused in the air in accordance with the diffusion law for gases; its diffusion coefficient in air appears to be close to that of carbonic acid.

Rutherford and Soddy have discovered that the emanation condenses at the temperature of liquid air. All experiments lead one to regard the emanation as a material gas. However, the hypothesis of the existence of such a gas
is based solely on the radioactive manifestations, and contrary to what happens with ordinary matter, the emanation disappears spontaneously in a sealed tube which encloses it.

Ramsay and Soddy have recently found that by enclosing the emanation from radium in a sealed tube and studying the emanation spectrum, the spectrum of helium could be seen progressively appearing, which was not observable at first. The writers explain these facts by a transformation that would be half complete in four days, and fully complete in 28. Without resorting to the hypothesis of the transformation of matter, while one is still waiting for a more complete demonstration of such an important fact, it would be possible to explain the facts by agreeing that the helium already exists in the gases of the emanation in the sealed tube, but that the special state of the emanation prevents the lines of the helium spectrum from appearing. The latter would gradually appear as the emanation is transformed; and it was observed that the activity of the emanation was reduced by half by the end of four days.

Further, it is relevant to quote a striking method of activation which leads one to make certain reservations about the conclusions to be drawn regarding the presence of new elements in radioactive preparations. Every inactive substance which is put into solutions of uranium, radium or thorium, and then precipitated out, becomes radioactive, but slowly loses its radioactivity. This fact was first noted by Curie and Giesel; the latter activated bismuth in this way.

With uranium, a trace of barium precipitated as the sulphate becomes noticeably more active than uranium; when activated in this way barium, like uranium, gives off only $\beta$-rays. After precipitation, the uranium salt, which is collected as a solid, is less active than before. This reduction in activity can be carried further by repeating the process, but the products gradually regain spontaneously their former activity.

The temporary reduction in activity after dissolution is a general phenomenon. Radium salts, when they are taken out of solution again are less active than before they were dissolved; their activity then increases over several months before reaching a maximum. They give off heat following this increase in activity.

According to Debierne, barium activated by actinium can be separated from inactive barium; it fractionates as radiiferous barium chloride, the least active portion being least soluble in water and hydrochloric acid. Debierne obtained by this method a product which was one thousand times more ac-
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activated than uranium. Activated barium behaves like a false radium, but differs from true radium in lacking the characteristic spectrum, and in losing its activity with time.

The spontaneous recovery of radioactivity by substances in which it has been reduced may be explained in terms of the substances themselves trapping their emanation, either on the active molecules, or on the inert molecules associated with them.

Following on this research, various workers have found more examples of radioactivity, sometimes as trace elements in metals, sometimes in natural phenomena.

Elster and Geitel discovered that atmospheric air shows to a slight extent the same properties as activated gases. By stretching long negatively-charged wires in the air they were able to recover traces of activated substances on them. Gases trapped in enclosed spaces, and air extracted from the ground or near waterfalls, show these properties, as do gases extracted from certain types of water.

To sum up, the radioactive substances whose nature is now well established are: uranium, thorium, radium, and polonium; actinium can be added, although very little information has been published about this last product. Reservations must be made about various other products obtained by Giesel, and about a preparation of active bismuth or active tellurium obtained electrolytically by Markwald.

Uranium gives off β- and γ-rays; it does not give off an emanation in air, but the activation which it produces in solution can be explained as the effect of an emanation.

Thorium and radium give off α-, β- and γ-rays, and an activating emanation in gases.

Polonium does not give off β-rays. It gives off α- and γ-rays, but loses its activity with time.

Actinium is said to possess a remarkable activation power.

Besides uranium and thorium, only radium has characteristics which enable it to be considered as an element with properties related to, but distinct from, those of barium. However, it is worth noting that this substance is never found, even as a trace element, in ordinary barium minerals, and that it is only met with in uranium minerals, where it is found with barium. This fact may well have a significance which will become clear to us later.

Radioactive substances, especially radium, give off energy in all the known
forms: heat, light, chemical reactions, electrical charges, \( \gamma \)-radiation. They seem to maintain the same state indefinitely, and the source from which they derive the energy they give off escapes us.

Among the hypotheses which suggest themselves to fill the gaps left by current experiments, one of the most likely lies in supposing that the emission of energy is the result of a slow modification of the atoms of the radioactive substances. Such a modification, which the methods at our disposal are unable to bring about, could certainly release energy in sufficiently large quantities to produce the observed effects, without the changes in matter being large enough to be detectable by our methods of investigation.

In this scheme, there would still be scope to wonder whether the transformation of the atom comprises a slow, spontaneous evolution, or whether it is the result of the absorption of external radiation beyond the range of our senses. If such a radiation were to exist, one could still picture the radioactive substances transforming it without themselves being altered. So far no experiment has confirmed or invalidated these hypotheses.