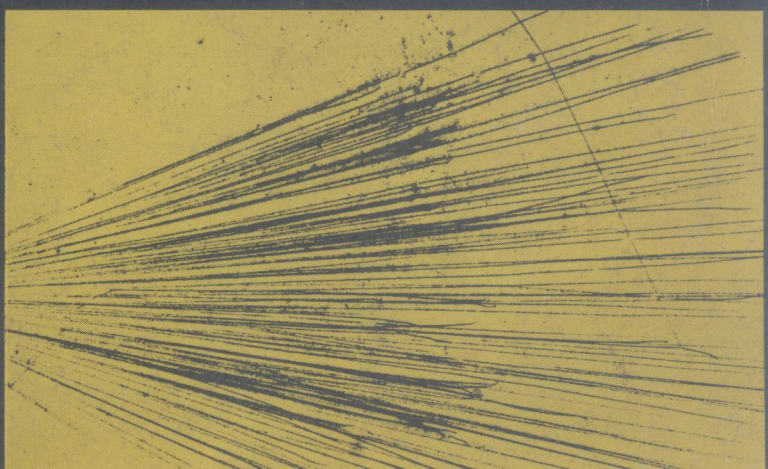
VAN NOSTRAND MOMENTUM BOOK #10

COMMISSION ON COLLEGE PHYSICS

Radioactivity and Its Measurement

WILFRID B. MANN AND S. B. GARFINKEL



WILFRID B. MANN
Chief, Radioactivity Section
National Bureau of Standards

and

S. B. GARFINKEL
Physicist, Radioactivity Section
National Bureau of Standards
University of Florida

RADIOACTIVITY AND ITS MEASUREMENT

Published for
The Commission on College Physics

D. VAN NOSTRAND COMPANY, INC.
Princeton, New Jersey
Toronto *London* *New York*

D. VAN NOSTRAND COMPANY, INC.
120 Alexander St., Princeton, New Jersey
(*Principal Office*)
24 West 40 Street, New York 18, New York

D. VAN NOSTRAND COMPANY, LTD.
358, Kensington High Street, London, W.14, England

D. VAN NOSTRAND COMPANY (Canada), LTD.
25 Hollinger Road, Toronto 16, Canada

Published simultaneously in Canada by
D. VAN NOSTRAND COMPANY (Canada), LTD.

PRINTED IN THE UNITED STATES OF AMERICA

Preface

This book, which has been written by the authors in a personal capacity, attempts to chronicle the discovery of radioactivity and the historical researches of the Curies, Rutherford, and other pioneers in its early development. It also deals with the interactions of α and β particles and of x and γ rays with matter and the consequent methods of detection of such radiations based upon the transfers of energy resulting from such interactions with matter. The energetics of the different modes of nuclear disintegration are also considered in relation to the Einstein mass-energy relationship as applied to radioactive transformations.

Based on these premises, the last three chapters of this book describe the nucleonic instrumentation that has been developed as a result of the observed interactions with matter that have been discussed in the earlier chapters. Various kinds of counters and detectors are described as well as methods, such as that of coincidence counting, of using these detectors to make direct and accurate measurements of radioactivity.

For references to recent methods of measuring radioactivity we would commend to our readers the National Bureau of Standards Handbook 80, entitled *A Manual of Radioactivity Procedures*.

Finally we wish to express our appreciation and thanks to several friends and colleagues, namely U. Fano for reading and commenting on Chapter 5; L. A. Currie and W. J. Youden for discussions on some parts of Chapter 8; J. W. Motz for discussions on bremsstrahlung; and H. A. Olsen for discussions on the energetics of radioactive decay.

We would also like to thank L. A. Currie, U. Fano and R. W. Hayward for reading all or parts of two chapters on atomic and nuclear structure which, because of lack of space, have been eliminated but the influence of which, like the grin of the Cheshire cat, may still appear unexpectedly in what remains of this monograph! We are also greatly indebted to Professor P. M. S. Blackett for kindly sending us the cloud-chamber photograph, showing the disintegra-

tion of a nitrogen nucleus by an alpha particle, which is reproduced on the cover of this book, and to him and to the Council of the Royal Society for permission to publish it.

W. B. MANN
S. B. GARFINKEL

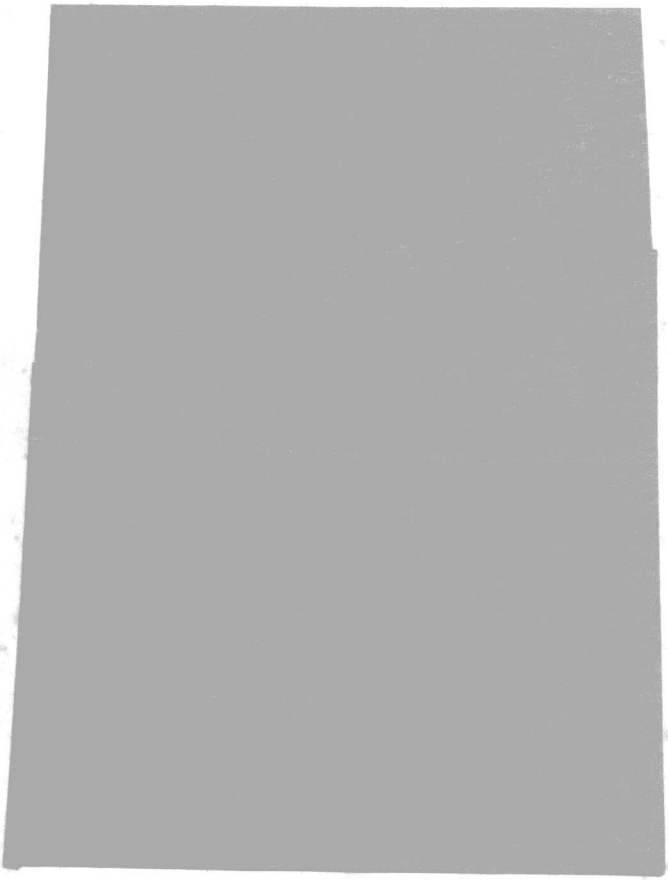


Table of Contents

TO

David, Janet, Kristine, Peter, and Phillip
in the hope that it may be of interest to them
and to their generations

1 *The Discovery of Radioactivity and Early Experiments Into Its Nature*

Radioactivity, the name coined by Marie Curie to describe the outward manifestations of atomic transformation, is the subject of this book.

Three discoveries in the closing years of the nineteenth century were not only to have a profound effect on scientific progress and thought in the first half of the twentieth century, but were also to create a new pattern for international political power and even, possibly, for life itself. These discoveries were those of x rays by Röntgen in 1895, of radioactivity by Becquerel in 1896, and the discovery, or postulation, of the corpuscular nature of the cathode rays by J. J. Thomson in 1897. In 1900 Planck also put forward the fundamental concept of the quantum in order to meet the failure of classical theory to describe the spectral distribution of black-body radiation; the so-called "ultraviolet catastrophe." Thus in the brief space of five exciting years were laid some very important foundations for the nuclear era upon whose threshold the world now stands!

The discovery of radioactivity was a direct consequence of that of the x rays or Röntgen rays. These had been produced in a discharge tube by the action of the cathode rays impinging upon the glass walls of the tube itself. A strong luminescence of the glass thus accompanied the production of the x rays, and there was considerable speculation whether such luminescence, or fluorescence, might of necessity be associated with the production of x rays. This possibility thus led to an intensive search for x-ray emission by materials rendered fluorescent or phosphorescent by visible light.

In the course of this search Antoine Henri Becquerel experimented with a double sulfate of uranium and potassium, a substance whose phosphorescence had been investigated previously by him and his father, Alexandre Edmond Becquerel. In the session of February 24, 1896, of the Académie des Sciences of Paris, Becquerel presented a short note announcing that he had carried out experiments using a crystal of this double sulfate of uranium and potassium, in the form of a thin transparent "crust." In these experiments he took a photographic plate that was wrapped in black paper of such thickness that a day's exposure to the sun did not fog the plate. He then placed the fluorescent salt on the outside of the black paper and exposed the whole package to sunlight for several hours. On development, the silhouette of the fluorescent crystal appeared in black upon the photographic plate. Further, on interposing a piece of money or a metal plate pierced with a design, its outline also appeared on the plate. Becquerel also interposed a thin sheet of glass between the fluorescent salt and the paper wrapping of the plate to exclude the possibility of chemical action from the vapors that might result from heating in the solar radiation. (In retrospect we now know that the effect observed by Becquerel must have been caused chiefly by the β rays, as the less penetrating α particles could not have passed through the black paper in which the plate was wrapped.)

At the session of the Académie one week later, on Monday, March 2, 1896, Becquerel reported a most significant development. On the preceding Wednesday and Thursday he had prepared experiments similar to those which he had described at the beginning of that week. But, and so typical of that time of the year, the sun only appeared intermittently, and so he had returned the wrapped photographic plates to the darkness of a drawer, leaving in place the small crystalline layers of uranium salt. As the sun failed to appear on the following days, he developed the plates on March 1, expecting to find quite weak images. "The silhouettes appeared, to the contrary, with great intensity." Becquerel had discovered that the effect on the photographic plate was independent of the phenomenon of fluorescence which, he pointed out, could scarcely be perceived after one-hundredth of a second. In this communication he also referred to the "*radiations actives*," thereby anticipating the term "radioactiv-

ity" used later by Mme. Curie. Subsequently, on May 18, 1896, Becquerel reported that all compounds of uranium that he had examined, whether fluorescent or not, and uranium itself, exhibited this phenomenon. He also showed that metallic uranium was more active than its compounds, when disposed over roughly the same surface area, and that the radiations were weakened by interposing thin foils of aluminum, copper, and other absorbers between the source of radiation and the photographic plate, as witness the earlier experiments giving the silhouettes of money and other objects. Becquerel also demonstrated that these radiations, like the x rays, possessed the property of discharging electrified bodies; it is this property that was and is the basis of most methods for detecting radiation from radioactive substances. Becquerel also thought that he had been able to demonstrate the reflection, refraction, and polarization through tourmaline of these radiations, but these results could not be repeated by other physicists at that time. It is possible that some of Becquerel's results, particularly that of the reflection experiment, could have been accounted for by the back-scattering of radiation. Finally, Becquerel kept a piece of uranium for several years in darkness at the end of which time its action on a photographic plate was essentially unaltered. He thus demonstrated that these radiations from uranium occurred independently of the phenomenon of luminescence.

Apart from the discovery of radioactivity itself, one of the most important contributions made by Becquerel was to show that the radiations from uranium would cause an electrified body in air to be discharged, and he used an elementary form of electroscope, designed by Dragomir Hurmuzesen, to make semiquantitative measurements of the radioactive intensity. This method is considerably faster and more convenient than that of observing the photographic effect.

Two years after the discovery of radioactivity, Marie Sklodowska Curie turned her attention to the possibility of there being other radioactive elements. She therefore examined a large number of materials for radioactivity, using for this purpose the electric method of measurement in conjunction with the phenomenon of piezoelectricity which had been discovered by the brothers Jacques and Pierre Curie, the latter of whom she had married in 1895. Her

apparatus consisted essentially of a parallel-plate condenser (*AB* in Fig. 1-1) on the lower plate of which the material under test was placed in the form of a layer of powder, the thickness of this layer being from 0.25 to 6.0 mm. The diameter of each of the plates was

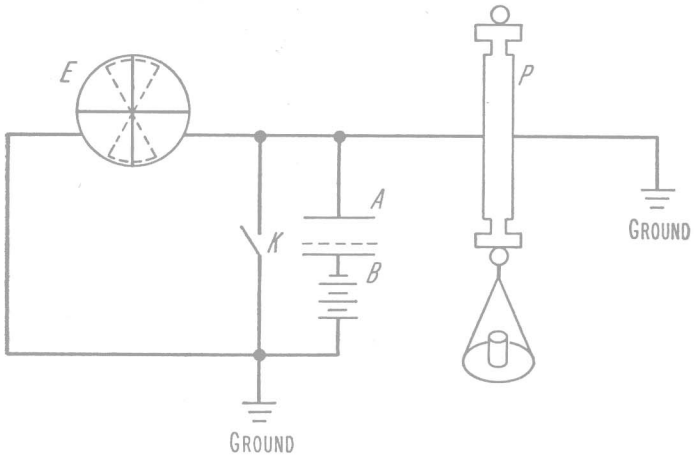


FIG. 1-1 Schematic diagrams of apparatus used by Pierre and Marie Curie to test the radioactivity of materials. The material to be tested was placed on the lower plate *B* of the parallel-plate condenser *AB*. The deflection of the electrometer *E* was maintained at zero by loading the quartz piezoelectric crystal *P* at a suitable rate with weights. *K* is a grounding key which is opened at zero time.

8.0 cm, and they were usually 3 cm apart. The lower plate *B* of this condenser was connected to one pole of a source of potential, the other pole of which was connected to ground. The other plate, *A*, of the condenser was connected to one pair of plates of a quadrant electrometer and also to one electrode of a piezoelectric crystal, of quartz, the other electrode of which was grounded. Under the influence of the radiations from radioactive materials the air between the plates *A* and *B* became ionized and conducting, and a current, of the order 10^{-11} ampere, flowed between the plates. One pair of quadrants of the electrometer *E* thus acquired an electric potential which would, in turn, cause the electrometer needle to be deflected. On now applying weights to the piezoelectric crystal a compensating

potential is developed which neutralizes that due to the current across the condenser, and the electrometer deflection can be reduced to zero. In this condition the mass added in a given time gives a measure of the current flowing across the condenser as a result of the air being rendered conducting by the action of radiation. At the beginning of any measurement, plate A and the pair of quadrants to which it is connected are grounded by means of the key K. On increasing the voltage on plate B the current reaches a limiting value, called the *saturation current*, which is a rough relative measure of the radioactive intensity of the layer of material on plate B.

Using this electrical method, Marie Curie examined a great many chemical compounds and also a large number of rocks and minerals to see whether any other elements exhibited the phenomenon of radioactivity found in uranium. As a result she found that thorium was the only element which was radioactive to the same degree as uranium, a result which was obtained independently by G. C. Schmidt and published a few weeks earlier by him. Marie Curie also showed that the radioactivity of compounds containing uranium and thorium was in proportion to the amounts of these elements present. As it was also found to be independent of any change in physical state or chemical decomposition, she concluded that radioactivity was an atomic phenomenon.

Mme. Curie also found, however, that some minerals, notably pitchblende, chalcolite, autunite, and carnotite, were more radioactive than uranium itself. Thus measurements with these materials gave the following currents, in 10^{-11} ampere, across the condenser AB: uranium, 2.3; pitchblende from various localities, 1.6 to 8.3; chalcolite, 5.2; autunite, 2.7; carnotite, 6.2.

The question then arose: If radioactivity were an atomic phenomenon, how could minerals containing uranium, and perhaps also some traces of thorium, be more radioactive than pure uranium itself? To test this point Mme. Curie prepared crystals of artificial chalcolite, which is a double phosphate of copper and uranium, also known as tobernite. This artificial chalcolite exhibited a normal radioactivity given by its composition, namely some two-and-a-half times less than that of uranium. She concluded therefore that these minerals must contain traces of some radioactive element many times more active than uranium itself.

Pierre and Marie Curie therefore set themselves the task of isolating this hypothetical element which had no known property except its radioactivity. Their procedures could therefore be based only on the methods of chemistry supplemented by observation of the radioactivity, and their investigation was, in a sense, the first application of the techniques of radiochemistry. Thus they observed the radioactivity of a compound which was then subjected to chemical decomposition, and the radioactivities of all the products were determined. In this way the radioactive substances could be followed through the various stages of the chemical process.

Eventually two new radioactive elements were discovered, the one chemically similar to bismuth and separating with it, and the other chemically similar to, and separating out with, barium. The former they named polonium (in honor of Poland, the country of Mme. Curie's birth) and the latter, which they discovered in collaboration with G. Bemont, radium. In their paper in 1898 describing the discovery of radium, they comment that both polonium and radium when placed near a luminescent salt, render it fluorescent. They go on to comment that this weak source of light which functions without a source of energy (as they then thought) is in contradiction to the Carnot Principle.

It was now important to be sure that these activities that separated with bismuth and radium should be identified as being due to new elements. This proved difficult in the case of polonium, but by repeated fractional crystallization, radium chloride, which is less soluble than barium chloride, was eventually concentrated to a point where E. Demarçay was able to identify spectroscopically first one and then more new emission lines due to the new element. The first new line identified was that of greatest intensity at 3814.8 A in the ultraviolet.

Finally, in collaboration with A. Debierne and with the help of a gift of one ton of residues from Joachimstal pitchblende by the Austrian Government, Mme. Curie was able to separate about 8 kg of barium and radium chloride having an activity of some sixty times that of metallic uranium. Finally, working with only about 0.1 g of radium chloride which had been found by Demarçay spectroscopically to have only the slightest trace of barium, Mme. Curie determined the atomic weight of radium and found it to be 225.

The presently accepted value of this quantity is 226.05, as determined by O. Hönigschmid in 1934. In 1903 the Nobel Prize in Physics was awarded to Pierre and Marie Curie and Henri Becquerel for their work. In 1911 the Nobel Prize in Chemistry was also awarded to Marie Curie for isolating radium. She died in 1934 as a result of prolonged exposure to the radiations from the element which she had successfully isolated.

Eventually many more naturally occurring radioactive elements were discovered and their relationships to each other largely established by the work of Ernest Rutherford and Frederick Soddy. To Becquerel and the Curies belong the credit of the discovery of radioactivity and the elements polonium and radium, but in the next four decades the field was dominated by Rutherford and his colleagues. By 1904 some 20 radioactive elements had been discovered, mainly by the work of Rutherford in a series of brilliant and incisive researches, often in collaboration with Soddy. It was also a happy accident in this connection, as will be explained later, that P. Curie discovered an "emanation" from radium and Rutherford and Soddy an "emanation" from thorium and that the latter investigators were able to conclude that these emanations were rare gases, namely, radon and thoron (recognized later as an isotope of radon). It was also found that there exist three "families" of radioactive elements.* The parent of the third family, actinium, was discovered by Debierne in 1899 and independently by F. Giesel in 1901. Rutherford showed, by investigating their penetrating power, that there were two types of radiation emitted by uranium. These were the α rays, which could easily be stopped by a sheet of paper or a few centimeters path in air (the "range" in air), and the β rays, which were far more penetrating and could pass through several millimeters of aluminum. In 1900 a third, much more penetrating type of radiation was found, by P. Villard, to be emitted by radium, and the rays comprising this type were named γ rays.

As early as 1899 F. Giesel, S. Meyer and E. von Schweidler, and Becquerel showed that the β rays could be deflected in a magnetic field in the same sense as the cathode rays. By a deflection through 180° onto a photographic plate (P in Fig. 1-2), Becquerel also

* These are described in more detail in Chapter 2 and illustrated in Figs. 2-3, 2-4, and 2-5.

showed that the spectrum of the β rays varies continuously in energy, and he also demonstrated that those which impinge upon the plate nearest to the source, S, are most readily absorbed. This he showed by placing different absorbers of paper, glass, and metals directly on

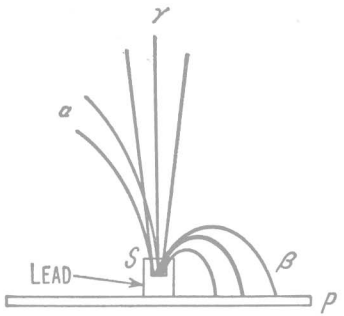


FIG. 1-2 Schematic of the paths of α, β, and γ radiation under the influence of a magnetic field at right angles to the plane of the drawing. The radioactive source *S* is contained in a lead cup. By allowing the β rays to fall on the photographic plate *P*, Becquerel showed that their energies varied continuously from zero, for any given substance, to a maximum.

the photographic plate so that different amounts of the continuous spectrum were suppressed. Becquerel and E. Dorn showed in 1900 that the β rays could also be deflected, in the same manner as cathode rays, by an electric field. In the same year Pierre and Marie Curie confirmed, by collecting them in a Faraday cylinder, that the β rays are negatively charged, while Becquerel, by observing their deflections in magnetic and electric fields, showed that β rays, having a velocity of some 1.6×10^{10} cm per second, have an approximate ratio of charge to mass ($e/m = 10^7$ emu per gram) of the same order of magnitude as that for the cathode rays. Also in 1900, W. Kaufmann showed that for β rays having velocities between 2.36×10^{10} and 2.83×10^{10} cm per second, the ratio of charge to mass varies from 1.31×10^7 to 0.63×10^7 emu per gram, respectively, thereby also indirectly demonstrating the relativistic dependence of mass on velocity. As the corresponding values for cathode rays, determined by J. J. Thomson in 1897, were 3×10^9 cm per second and $0.7 \times$

10^7 emu per gram, it was readily concluded that the β rays were of the same nature as the cathode rays, which were negatively charged and, according to Thomson, corpuscular in form.

The identity of the α and γ rays eluded elucidation for a great many more years. Initially, they were both believed to be unaffected by an electromagnetic field, but in 1902 Rutherford showed that the α rays were indeed deflected in powerful electric or magnetic fields. For a given field their deflection was much less than that for the β rays and in an opposite direction, indicating that the α rays consist of positively charged particles. Also in 1902, by measuring their deflection in electric and magnetic fields, Rutherford concluded that the α rays from radium were traveling with a velocity of about 2.5×10^9 cm per second and that they had a charge-to-mass ratio of about 6000 emu per gram. The method used by Rutherford to make these measurements is illustrated in Fig. 1-3. In 1903, T. des Coudres, using the less sensitive photographic method of detection and radium bromide as a source, obtained values of velocity equal to 1.65×10^9 cm per second and e/m equal to 6400 emu per gram for the α rays from radium. Unlike the β rays, which were found to have continuously varying velocities (Fig. 1-2), the α particles from any given radioactive element were found to have discrete velocities and, hence, to be homogeneous in energy.

Helium had been discovered on earth in 1895 by W. Ramsay, its spectrum having first been observed in the solar spectrum by J. N. Lockyer. In 1903 Ramsay and Soddy showed conclusively that helium is produced from radium. Previously, Rutherford and Soddy had pointed out that one could speculate on whether the invariable presence of helium in minerals containing uranium and radium might be connected with their radioactivity. In the experiments of Ramsay and Soddy radium bromide in solution was used to supply radium emanation to a vacuum system containing a discharge tube to excite the helium spectrum, if helium were present. Initially, no helium spectrum could be observed after condensing the emanation in liquid air, but on repeating the experiment after the equipment had stood for four days the characteristic lines of the helium spectrum could be observed. The accounts of this work are fascinating, especially in describing how the liquid air trap shone brilliantly in the dark and how, on removing the liquid air, a brilliant phenome-

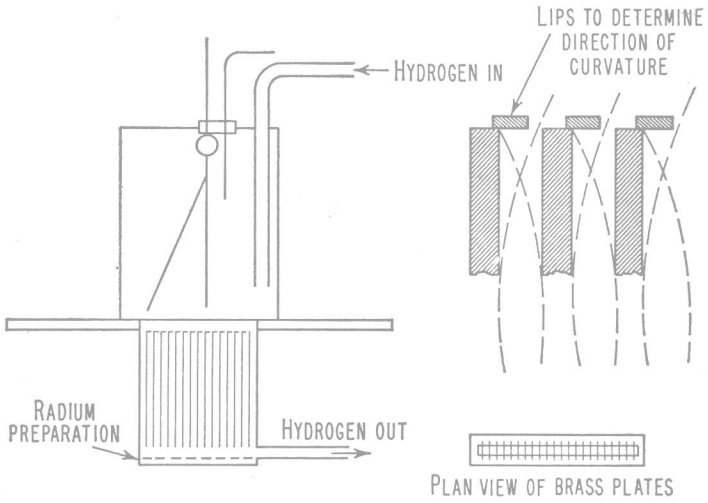


FIG. 1-3 Apparatus used by Rutherford for the magnetic and electrostatic deflection of the α rays from radium. Twenty to twenty-five parallel brass plates insulated from each other by ebonite were disposed below a gold-leaf electroscope in the manner shown. By applying a magnetic field normal to the plane of this page or an electric field between the plates, the α rays could be made to travel in arcs of circles. By increasing the field strengths until the α rays just failed to enter the electroscope, the respective limiting radii of curvature could be determined, and hence mv/e and mv^2/e could be calculated. The direction of curvature was determined by using plates with lips as illustrated. Radium emanation was flushed from the apparatus by a stream of hydrogen.

non was observed as the emanation expanded through the equipment—slowly through the capillaries, delayed by the phosphorus pentoxide trap, and rushing through the wider glass tubes. A charming reference is also made to the vacation which intervened and the precautions which were taken to prevent the apparatus' bursting while gases accumulated for 60 days!

In 1906, in a paper written from Berkeley, California, Rutherford describes further and more precise measurements of the velocity and ratio of charge to mass of the α rays from radium C, obtained by exposing a wire to radium emanation, and from radium F and also