J. Thomson and Ernest Rutherford, 1934. (Courtesy of Juan A. del Regato, M.D.)
PROLOGUE:
The Century of Classical Physics

The nineteenth century was a period of great progress in the physical sciences. By the closing decade of the century both the theoretical and practical aspects of electromagnetism and electrodynamics were firmly established; the existence of atoms and molecules was accepted; the periodic table of the elements provided a guide in the search for the "missing" elements indicated by gaps in the table; the kinetic theory of gases, the laws of electrolysis, thermodynamics, the equivalence of heat and mechanical energy, and the absolute scale of temperature were standard topics in science textbooks; and the wave nature of light was clearly indicated by properties such as polarization, interference, and diffraction.

The triumph of classical physics was such that, by the closing years of the century, many scientists believed that everything about the physical world was already known and that all the physical laws had been discovered. It remained only to fill in the details and to measure known physical properties ever more precisely. By the turn of the century, however, this comfortable illusion had been rudely shattered, and the era of modern science—of quantum physics, relativity, radioactivity, and the nuclear atom—had begun. Although it is difficult to assign a precise date to this changeover, it is safe to say that one of the main instruments of this revolution was the vacuum discharge tube used for studying the passage of electricity in gases at greatly reduced pressure. These studies led directly to the discovery of cathode rays, X rays, and the electron and, indirectly, to the discovery of radioactivity. In fact, perhaps it is not unreasonable to suggest that modern science began on 8 November 1895: the day X rays were discovered.

The study of the conduction of electricity in gases at low pressure was not new; indeed, Michael Faraday had experimented with a discharge tube in 1835 and had described the associated visual phenomena, including a feature still known as the Faraday dark space. It was not until the 1860s, however, that the vacuum tube assumed an important role in the study of electricity and matter. The foremost names in this work were Johann Hittorf, William Crookes, Heinrich Hertz, Philipp Lenard, and J. J. Thomson.

In 1865 Hittorf described two distinct types of discharge-tube spectra for a num-
PIONEERS OF THE VACUUM DISCHARGE TUBE

The four principal scientists whose work on the vacuum discharge tube preceded that of Röntgen were all noted for their other contributions to research. Some of these contributions were as follows:

Johann Hittorf (1824-1914) is best known for his studies in Germany from 1853 through 1859 of variations in the concentrations of electrolytes during electrolysis, which he attributed to differing mobilities of the positive and negative ions. He measured the ratio of mobilities.

Heinrich Hertz (1857-1894) was the first to demonstrate the existence of the electric (radio) waves predicted by James Clerk Maxwell in 1873. Hertz’s principal work was carried out at Karlsruhe, where he was professor of physics from 1885 to 1889. In the last five years of his short life, Hertz was professor of physics at Bonn.

Sir William Crookes (1832-1919), a British chemist, discovered the element thallium in 1861 and made an accurate determination of its (non integral) atomic weight in 1863. He invented the radiometer (for measuring the mechanical effect of radiation) in 1865. Crookes was editor of the Chemical News from 1859 to 1906.

Philipp Lenard (1862-1947) held university appointments in physics in Bonn (1863), Aachen (1895), Heidelberg (1896), Kiel (1898), and again in Heidelberg (1907-1931). Apart from his work on cathode rays, Lenard is noted for his 1903 model of the atom and his work on photoelectricity. He also devised the grid as the control electrode in thermionic valves. Lenard was awarded the Nobel Prize for physics in 1905 for his work on cathode rays.

In 1869 he investigated in detail the changes inside the tube as the gas pressure was decreased, and noted that both the “cathode glimmer” in the gas near the cathode and the phosphorescence in the glass of the tube were subject to shadows when objects were placed near the cathode. This finding proved that the glimmer and the phosphorescence were caused by rays emanating from the cathode. In 1878 and 1879 Crookes amplified these observations and reported on the warming of obstacles by the “molecular rays” proceeding from the cathode and on the deflection of these rays by a powerful magnet.

In 1892 Hertz showed that the cathode rays could penetrate thin metal foils. The work of Crookes and Hertz stimulated Lenard from 1892 to 1894 to examine the cathode rays outside the discharge tube, which he was able to do by sealing a very thin aluminum window into the glass wall of the tube. He found that cathode rays could induce fluorescence and blacken a photographic plate up to a distance of 8 centimeters (cm) in air, and could discharge an electroscope at a distance of 30 cm. There is little doubt that Lenard came close to the discovery of X rays, for which he himself claimed
priority, but he failed to make the crucial investigations that distinguished Röntgen's work.¹

The fifth major investigator of cathode rays, Thomson, carried out his main work in this area after the discovery of X rays in 1895. Thomson's work is discussed later in this chapter.

THE DISCOVERY OF X RAYS

Wilhelm Conrad Röntgen (1845–1923) was born in Lennep, in Germany, but was brought up in Holland. In 1868 he graduated in mechanical engineering from Zürich Polytechnic and a year later received a Ph.D. from the University of Zürich. Junior appointments followed at the universities of Würzburg, Strasburg, and Württemberg and, in 1879, a professorship in physics at the Hessian University of Giessen. In 1888 he returned to Bavaria as professor of physics and director of the newly-established Physical Institute of the University of Würzburg.

In 1894 Röntgen was elected rector of the University of Würzburg but retained his physics posts and duties. The same year he began to study the phenomena of the vacuum discharge tube. Progress was slow because his teaching and administrative duties left little time for research, which he carried out mainly in the late afternoon or evening in his private laboratory in the Physical Institute.

By the autumn of 1895 Röntgen had repeated the investigations of the earlier workers and was ready to begin experiments of his own design. He substituted a pear-shaped Hittorf vacuum tube for the Lenard tube. The Hittorf tube had thicker glass walls and no aluminum window, but Röntgen nevertheless suspected that the cathode rays could penetrate the walls of the tube. To prove this, it was necessary to demonstrate fluorescence in a suitable screen held close to the outside of the tube. When the discharge tube was activated, however, its glass walls became brightly luminescent, and this effect would mask any weak fluorescence of an adjacent screen. His remedy was to cover the tube with a close-fitting mantle of thin black cardboard and to work in a totally dark room.

On the evening of 8 November Röntgen was ready to test the effectiveness of the cardboard enclosure of his tube and of the thick curtains covering the windows of the laboratory. On activating the vacuum tube he noticed in the darkness a shimmer of light coming from a table some distance from the tube. The source of light proved to be the cardboard screen coated with barium platinocyanide, which he intended to use to test for cathode rays immediately outside the tube, but at that moment the screen was well beyond the normal range of these rays in air; in fact, when Röntgen moved the screen it continued to respond up to two meters distance from the vacuum tube. As Röntgen stated in his "Preliminary Communication," "It is easy to convince oneself that the cause of the fluorescence is the discharge apparatus [i.e., the vacuum tube] and nothing else."

In the seven weeks following the discovery Röntgen worked alone, at a fever pitch, to confirm the existence of the mysterious "X" radiation and to investigate its properties. On 28 December 1895 he delivered a handwritten preliminary report of his findings to the secretaries of the Physical-Medical Society of Würzburg. This concise document, titled "Über eine neue Art von Strahlen" ("On a new kind of ray"), comprised seventeen short sections setting out the properties of the newly-discovered rays.²

Röntgen reported that all substances were transparent to X rays in some degree, the transparency depending mainly on the density of the material. The rays induced fluorescence in a number of substances, including calcium compounds, uranium glass, and rock salt, as well as the barium platinocyanide whose glow was responsible for the original discovery. In addition, the rays blackened a photographic plate, although Röntgen was uncertain whether this was a direct effect on the silver salts or an indirect effect of fluorescence. There was no evidence of either refraction or regular reflection of the rays. Finally, the rays were unaffected by even a strong mag-
JOHN COX (1851-1923)

John Cox was born in England in 1851 and educated at the City of London School and Cambridge University, where he was a brilliant student of mathematics and classics. After graduating in 1874 he was elected a fellow of Trinity College, Cambridge, and later became Warden of Cavendish College, Cambridge. (This college no longer exists.)

In 1890 Cox was appointed the first professor of physics at McGill University in Montreal. (Up to that time physics had been taught as part of applied science.) A new physics building, the gift of Sir William Macdonald, was planned, and Cox was asked to visit and study physical laboratories in Europe and the United States. As a result, the Macdonald Physics Building, opened in 1893, was considered to be one of the finest and best-equipped science buildings in the world. It remained in use as a physics teaching and research building until 1977 and is now a library.

The nineteen years Cox spent at McGill as Macdonald Professor of Physics (he retired in 1909) were notable in three ways. First, he expanded and greatly improved the teaching of physics at the university; he himself was an excellent teacher, albeit a poor researcher. Second, he persuaded Macdonald to endow a professorship in experimental physics and made two outstanding appointments to the post: Hugh Callendar, a world authority on the physics of heat, in 1893; and, when Callendar left in 1898, Ernest Rutherford. Throughout Rutherford's nine-year tenure at McGill, Cox supported him in several ways: morally, by encouraging him during the controversial development of the transmutation theory of radioactivity; materially (e.g., by soliciting funds for radium and a liquid-air machine); and practically, by reducing Rutherford's teaching load to a minimum.

Third, Cox is remembered as the first person in Canada to produce (on 7 February 1896) a clinical radiograph that could be used as an adjunct to surgery. Cox followed this by undertaking, with Callendar, a number of measurements on the physical properties of X rays, including their velocity. Unfortunately he did not pursue this work beyond the summer of 1896.

Cox wrote a number of books on science, including Beyond the Atom, an excellent account of the early researches on radioactivity (published by Cambridge University Press in 1913).

magnetic field; this fact differentiated the X rays from the cathode rays.

Röntgen's "Preliminary Communication" ended with a brief discussion of the nature of the new radiation. The concluding words were: "May not the new rays be due to longitudinal vibrations in the ether? I must admit that I have put more and more faith in this idea in the course of my research, and it behooves me therefore to announce my suspicion, although I know well that this explanation requires further collaboration."
The news of Röntgen's discovery spread rapidly round the world via Vienna and London. An English transliteration of the "Preliminary Communication" was published in Nature on 23 January 1896 and in The Electrician (London) on 24 January. The equipment needed to produce the rays was readily available in physical laboratories, and scientists on the Continent and in Britain immediately began to repeat and extend Röntgen's work. At first, however, only summaries reached North America by cable; the full text arrived only toward the end of January. It was published in Electricity (New York) on 5 February 1896, along with a short explanatory article by Michael I. Pupin of Columbia College and reproductions of the two "Röntgen silhouettes" (a hand and a razor in its case) taken by A. A. Campbell-Swinton, a British engineer.

Röntgen himself continued to investigate the X-rays after the publication of his "Preliminary Communication" and wrote two further papers on the "new kind of rays," in March 1896 and March 1897. After 1897 Röntgen lost interest in X-rays, and he made no further contribution to this field. In 1900 he left Würzburg for Munich, where he was appointed professor of physics at the Ludwig-Maximilians University. In 1901 Röntgen was awarded the first Nobel Prize for physics for his discovery of X-rays. He died in 1923.

NORTH AMERICAN PIONEERS

The credit for the first successful X-ray image in North America is a matter of some dispute. Brecher and Brecher list more than fifty physicists, physicians, and others who obtained X-ray photographs in the United States or Canada prior to 1 March 1896. The earliest documented exposures were made on 26 January when Arthur Williams Wright, the professor of experimental physics at Yale, connected a Crookes tube to an induction coil and obtained recognizable X-ray images of several small objects as well as a hand. Other early experimenters in this field were John Trowbridge, Rumford Professor of Science at Harvard; Edwin Brant Frost, professor of astronomy at Dartmouth College; John Cox, professor of physics at McGill; and William Francis Magic, professor of physics at Princeton. Of these Frost merits further mention, since he was the first in North America to take a clinical X-ray photograph, whereas Cox was the first in North America to obtain a radiograph that was later used as an adjunct to a surgical procedure.

Frost and his colleague, C. F. Emerson, a professor of physics, began experimenting with X rays in a physics laboratory at Dartmouth on 1 February 1897. Two days later Frost's brother, a physician, brought a young patient with a broken forearm to the laboratory. A twenty-minute X-ray exposure was made, and, according to the report published in Science on 14 February, "the plate on development showed the fracture of the ulna very distinctly." However, the published report was concerned mainly with the physical investigations made by Emerson and Frost and did not include a reproduction of the clinical radiograph. Incidentally the term radiography, as a substitute for X-ray photography, was suggested in the same issue of Science by Arthur W. Goodspeed, of the University of Pennsylvania.

On 3 February Cox assembled the apparatus to produce X-rays and immediately obtained excellent pictures of the hands of his assistants. On 7 February Robert Kirkpatrick, a surgeon at the Montreal General Hospital, brought to Cox's laboratory a young man who had been shot in the leg during a street brawl. The doctors at the hospital had been unable to locate the bullet by probing, and Kirkpatrick decided to try the new roentgen photography. An exposure of forty-five minutes resulted in a plate showing the bullet lying against the outer edge of the tibia; the bullet was surgically removed the same day with the aid of the radiograph. Unfortunately, despite the long exposure, the plate was significantly underexposed, and the print made from it and subsequently published is unclear (Fig. 1.1). In a later action the patient brought against his assailant, the radiograph was accepted as evidence—the first example in North
true medical physicist in North America, William Duane, was not appointed to a hospital post until 1913, almost at the end of the period under review. Nevertheless, the work discussed in this chapter not only made a significant contribution to twentieth-century science but laid the foundations of modern diagnostic radiology and radiation therapy. Thus the period from 1895 to 1914 effectively established the physical and engineering bases of medical radiology, even though much of the work was not specifically directed to that end.

The Nature of the Cathode Rays

Early workers were hampered in their efforts to understand the production of X rays in a vacuum discharge tube by their lack of knowledge of the nature of the cathode rays that were involved in—or at the least, associated with—the emission of the new penetrating radiation. For almost two years after Röntgen’s discovery the nature of the cathode rays was hotly disputed; as J. J. Thomson stated: “The most diverse opinions are held as to these rays.” In particular, were the cathode rays to be considered material particles or a nonmaterial manifestation of an all-pervasive ether? Both views were held with equal conviction. It was not until late 1897 that Thomson demonstrated beyond dispute the material nature of the cathode rays and showed that they comprise streams of particles much smaller than the lightest atom, hydrogen. These subatomic particles were eventually recognized as a fundamental component of all matter: the electron.

Thomson’s investigation of the cathode rays was the most thorough that had yet been made. He measured their electric charge, their deflection in electrostatic and magnetic fields, the conductivity induced in different gases by the rays, and the rise in temperature of a solid body in which the rays are absorbed. These measurements were combined so as to yield values of the velocity of the particles and the ratio of their mass (m) to charge (e) (the measurements did not yield the absolute

As already indicated, the earliest investigators in this field were all scientists or engineers rather than physicians. This is not surprising, because no commercial X-ray equipment was available, and each experimenter was obliged to assemble his own apparatus. Soon, however, members of the medical profession, especially surgeons, began to assemble the equipment, or to seek the help of friends or colleagues in doing so, and to carry out their own investigations. The potential of the new radiation as a diagnostic tool and as an aid to surgery was obvious from the beginning. In his classic biography of Röntgen, Otto Glasser lists more than a thousand papers, articles, and reports on the production, properties, and applications of X rays published worldwide in the first year after the publication of Röntgen’s discovery.10

Much of the work to be described in this chapter was not specifically directed toward the medical applications of X rays and radioactivity: it was not “medical physics” in the modern sense of the term, and the individual contributors were not “medical physicists.” The first

Fig. 1.1 Radiograph of patient’s leg with bullet embedded. (Cox and Kirkpatrick, Montreal Medical Journal, February 1896)
values of \( m \) and \( e \). Thomson found that the ratio \( m/e \) was consistently of the order of \( 0.5 \times 10^7 \) (the units were not explicitly stated), whereas the corresponding value of the hydrogen ion (as deduced from electrolysis investigations) was \( 10^4 \), about two thousand times larger. Thomson showed that the low value of \( m/e \) for cathode rays could not be attributed to each particle carrying multiple electric charges; only a very low value of \( m \) could explain the results. Thomson concluded that the cathode rays were "primordial atoms," that is, subatomic particles that, in different combinations, form the chemical atoms. Thomson called these subatomic particles "corpuscles" and added: "...if these corpuscles are charged with electricity and projected from the cathode by the electric field, they would behave exactly like the cathode rays."

Fortunately, the name "corpuscle" did not stick. As far back as 1874 a British scientist, G. Johnstone Stoney, had argued that electricity must be atomic in nature, that is, there is a minimum quantity of electricity, the valency-charge, that is indivisible. A similar argument was made by Hermann von Helmholtz in Germany in 1881. In 1894 Stoney reviewed his own and Helmholtz's arguments and proposed the name electron for the fundamental unit of electricity. This name was later adopted not for the unit quantity of electricity but for Thomson's "corpuscle," which carried this charge.

**The X-Ray Tube, the Power Supply, and the Source of the Radiation**

In January and February 1896 the first objective of the investigators who responded to the news of Röntgen's discovery was to prove to themselves, to their scientific and medical colleagues, and to the public at large that the X-radiation was a real phenomenon with unusual properties and enormous potential. Despite the skeptics who argued that Röntgen rays were either nonexistent or simply a variation of known phenomena, this task was soon accomplished, and by March 1896 the pioneers could move to the next stage, in which the output of penetrating radiation would be greatly increased and brought under a measure of control. Clearly, exposures of up to or beyond an hour were inconvenient and needed to be reduced. Furthermore, because X-ray images were a form of shadow photography, the sharpness of the image depended on the size of the source. The first objective was therefore to modify the design of the discharge tube and its source of power, so as to increase the intensity of the X rays emitted while simultaneously reducing the dimensions of the source of the radiation.

Before this could be accomplished, however, an important question needed to be answered: where did the X rays originate? In his "Preliminary Communication," Röntgen stated that the penetrating radiation was clearly produced in the discharge tube, but he did not specify a particular location. There were three possibilities: the cathode, the anode, and the glass walls of the tube. Rowland, Carmichael, and Briggs, of Johns Hopkins University, asserted in March 1896 that the "main source of the rays was a minute point on the anode...in no case did the cathode rays seem to have anything to do with the Röntgen rays." Wilbur M. Sline, of the Armour Institute of Technology in Chicago, reached the opposite conclusion by studying the X-ray shadows cast by a series of concentric brass tubes. In early April he concluded that "the cathode, not the anode, is the source of the X-ray." Progress in this field was rapid, however, and by mid-April 1896 Cory, Le Conte, and Lohman, of the University of California, were able to state that "the source of the Röntgen rays is the solid upon which the cathode rays first strike, giving up either partially or entirely their negative charge." This conclusion was broadly accepted by the scientific community.

This conclusion, however, had been anticipated by workers in other countries, even by Röntgen himself. In paragraph 20 of his second communication on 9 March 1896, Röntgen stated: "According to my experience up to now, platinum is best suited for the pro-
duction of X rays of highest intensity. For several weeks I have used with good success a discharge tube with a concave mirror of aluminum as cathode and a platinum foil as anode, which has been placed in the focus of the cathode and inclined 45 degrees in relation to the axis of the mirror. Again, on 4 April 1896, The Electrical World (New York) reported that the London Electrical Review of 13 March had illustrated a new form of the tube, "designed by one of the professors [Herbert Jackson?] at King's College, London," in which the cathode was a concave disc of aluminum and the anode a flat disc of platinum placed at an angle a short distance beyond the focus. The same report in The Electrical World mentioned a similar tube, attributed to A. A. Campbell-Swinton, described in the London Electrical Engineer, also on 13 March. Editorially The Electrical World considered that "this tube may be considered the greatest advance in X-ray photography since Röntgen's discovery." The so-called "focus tube" soon came into wide, albeit not universal, use. By October 1896 Stine was able to assess the relative merits of the focusing tube, which gave superior definition for scintigraphic purposes, and the glass impact tube, which provided a greater output of penetrating radiation. Incidentally, Stine concluded that "credit for the design [of the focus tube] does not belong to a particular person since it originated simultaneously with a number of investigators, both in this country and in England."20 At first it was thought that the production of X rays and of fluorescence were twin aspects of the same phenomenon. (Indeed, as discussed later in this chapter, it was this belief that prompted Antoine Henri Becquerel to begin his search for naturally occurring sources of X rays, a search that led to his discovery of radioactivity.) The focus tube, with its platinum anode providing the source of X rays, necessitated the modification of this theory. Elihu Thomson was the first to point out that "fluorescence has nothing to do with it [the production of X rays] since substances which are not fluorescent (e.g., platinum) give off the rays on bombardment."21 Modifications of the focus tube soon appeared. The most important (soon to be marketed by the General Electric [GE] Company) was Elihu Thomson's "universal double-focus tube," proposed in The Electrical Engineer on 15 April 1896 (Fig. 1.2).22 It comprised a glass bulb, at the opposite sides of which were mounted concave aluminum discs supported

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Fig. 1.2 Elihu Thomson's "universal double-focus tube." (The Electrical Engineer, 15 April 1896)
on wires sealed through the glass. The foci of these electrodes coincided nearly at the center of the bulb where a V-shaped sheet of platinum, iron, or other metal was located. The two aluminum electrodes acted alternately as the cathode, depending on the polarity of the applied alternating high voltage, while the V-shaped target provided a relatively compact source of X rays. A somewhat similar tube, with two concave cathodes and a single (flat) target was designed at about the same time by Campbell-Swinton in England.23 Although Thomson's design, which he proposed as a "standard tube," was undoubtedly a success, not everyone was convinced. Stine, for example, in the paper previously cited, maintained that there was no need for a standard form of the tube, because the design must vary according to the special need.24

The design and operation of the gas X-ray tube attracted the immediate attention not only of American scientists but also of engineers, technologists, and inventors, including Thomas Edison and Nikola Tesla. As early as March 1896 Edison was able to report the results of intensive investigations on the new radiation. Among Edison's ten conclusions at this stage were: that a very high vacuum in the discharge tube conveyed no advantage; that a tube with thin walls of German glass and length three times the diameter gave the best results; and that the intensity of the roentgen rays could be increased by placing metallic discs, each connected to the respective electrode, opposite the electrodes on the outside of the tube.25

In contrast to Edison, whose early investigations related mainly to the design of the discharge tube and the choice of fluorescent material, Tesla was initially more concerned with the origin, nature, and properties of X rays, including their effects on the human body. Later, however, he developed some new designs for vacuum tubes and posed two questions concerning the location and nature of the "impact body" in which the roentgen rays were produced by collision of particles from the cathode: (1) must the impact body be within the tube? and (2) must the impact body be solid or liquid? Tesla answered both questions in the negative.25

Despite early improvements in the design and efficiency of the X-ray tube, in practice its operation was subject to several serious problems. In particular, neither the quantity nor the quality of the radiation could be properly controlled, although at this stage the distinction between these properties was scarcely appreciated. The basic problem was that the residual air pressure inside the discharge tube—a few micrometers of mercury—changed as the tube was operated. (Some residual air was necessary, because it provided the positive ions that bombarded the cathode and thereby released the cathode rays [electrons] that, in turn, produced the X rays.) With low discharge currents through the tube the vacuum tended to harden (i.e., the gas pressure decreased) until it reached the point at which the tube could no longer function to produce X rays. With high discharge currents the vacuum would change rapidly and erratically in either direction. The remedy for too high gas pressure was to reevacuate the tube, whereas the remedy for low gas pressure (the more usual situation) was to add a side tube, opening into the main discharge tube and containing a chemical which gave off vapor on heating. At first this procedure was carried out as needed by the operator, who applied a Bunsen burner to the side tube. Soon, however, devices were introduced to automate the operation. For example, at a meeting of the Franklin Institute in Philadelphia on 26 April 1897, Lyman Sayen demonstrated a new tube with automatically adjustable vacuum (Fig. 1.3).27 The chemical in the side tube not only gave off vapor on heating but reabsorbed it on cooling, and the heating/cooling was governed by a discharge in an auxiliary tube surrounding the "chemical bulb" and directly connected to the main X-ray tube. In this way, it was claimed, the main vacuum could be adjusted to any desired value and would remain at the chosen level.
THOMAS ALVA EDISON (1847-1931)

Thomas Edison was born in 1847 of Canadian parents in Milan, Ohio. The young Edison received little formal education but at an early age became an avid reader and acquired a strong interest in chemistry and electricity. As a teenager he earned money first by selling newspapers, tobacco, and candy on trains and later by working as a telegraph operator. By the age of twenty he had resolved to become an inventor.

In 1870 Edison sold his stock-ticker inventions for $40,000, a sum that enabled him to set up a private laboratory with fifty employees—a kind of “inventions factory”—first in New York, then in Menlo Park, New Jersey, and finally, in 1887, in West Orange, New Jersey.

Although Edison acquired a considerable knowledge of (nonmathematical) science by private reading, he was essentially a trial-and-error inventor, and in this capacity he excelled, making important contributions to telegraphy, electric lighting, storage batteries, telephony, and, of course, the phonograph (1877). His one important scientific discovery was the “Edison effect” (1883), that is, the emission of electrons from a hot cathode, the basis of the thermionic valve. However, unlike Tesla, he failed to realize that the future of electricity generation and transmission lay with alternating rather than direct current.

When Röntgen’s discovery of X rays became known in January 1896, Edison immediately began experimenting in this field, and in March 1896 published (in Electrical Engineering) the first of several reports on the production and properties of the new radiation. His most important contribution was probably the search for a more sensitive fluorescent material than the barium platinocyanide used by Röntgen in the original discovery. After testing more than eight thousand different compounds, Edison concluded that calcium tungstate was best. Edison’s fluoroscope (a box with a fluorescent screen at one end and a peep hole at the other) aroused enormous interest at the Electrical Exposition in New York in May 1896.

Edison was also one of the first investigators to notice the injurious effects of X rays. Indeed, his assistant, Clarence Dally, suffered severe ulceration and epilation and, in 1904, was probably the first person to die of X-ray injuries.

During World War I Edison was appointed president of a new board set up to advise the United States Navy on the use of science and technology, and this eventually led to the establishment of the Naval Research Laboratory. In 1927 Edison was elected to the National Academy of Sciences; he died four years later. Further information may be found in Matthew Josephson’s Edison: A Biography (New York: McGraw Hill, 1959).
Even though the vacuum in a given tube could be controlled, the problem remained that gas X-ray tubes were inherently variable; no two tubes behaved in exactly the same way. As George Pfahler, a clinical professor of roentgenology in Philadelphia, noted in 1912: "Every experienced Roentgenologist recognizes that the most important part of his apparatus is a good tube." Pfahler pointed out that, during an exposure—even one as short as a half second—movement of the focal point occurred, although the current and vacuum were constant. This movement caused the image to blur; the blurring was more marked in some tubes than others, but in no case was the effect entirely absent. Pfahler described some simple tests, involving a wire mesh, to determine the extent of the movement in each tube and hence to select satisfactory tubes for clinical use. He concluded: "If these tests were made by manufacturers, they would probably learn to make more uniformly good tubes."  

The operation of the gas X-ray tube also depended on the reliability and constancy of the power supply. The required high voltage—many thousands of volts—was produced either by a static generator, such as a Wimshurst machine powered by a DC motor or by a Tesla coil (an air-core transformer with primary and secondary coils tuned to resonate). Many variations of the Tesla coil were developed over the years, both by Tesla himself and by others. For example, in a modified Tesla coil marketed by a Boston company in July 1896, the secondary coil was immersed in insulating oil. An important innovation occurred in 1900 when John Trowbridge, at the Jefferson Physical Laboratory of Harvard University, assembled a battery of twenty thousand storage cells to give over 40,000 volts and a comparatively steady current of 8 to 5 milliamperes through a resistance of 4 million ohms. The significance of this assembly is that, for the first time, it was possible to apply a constant potential across an X-ray tube and to study the relationship between the applied voltage and the characteristics of the resulting X rays. But it was premature to attempt such an investigation in 1900. Indeed, it was not until Trowbridge retired in 1913 that conditions became appropriate for the investigation to be carried out by his successor, William Duane. By 1915 the Coolidge X-ray tube was available, together with X-ray diffraction, which provided a means of measuring the wavelength (and hence the spectrum) of X rays. The outcome was the Duane-Hunt Law, developed between 1915 and 1917, which is discussed in the next chapter. At this point it is sufficient to note that Duane's research would not have been possible without the storage battery assembled in 1900 by Trowbridge.

**The Coolidge Tube**

Despite the many modifications and improvements made to the gas X-ray tube in the decade after Röntgen's discovery, the tube remained, at best, an inconvenient and erratic source of radiation and, at worst, a major obstacle to the progress of radiological imaging. The turning point in this situation came in December 1918, when William
Nikola Tesla (1856-1943)

Nikola Tesla was born in 1856 of Serbian parents in a village in Croatia. He was educated in local schools, in the polytechnical college in Graz (Austria), and in the University of Prague. After working as an engineer in Budapest and Paris, in 1884 he moved to the United States, where he remained for the rest of his life. One of Tesla’s biographers, Kenneth Swezey, noted, “He arrived in New York with a working knowledge of a dozen languages, a book of poetry, four cents, and an introduction to Thomas Edison.”

The Edison introduction provided Tesla with a job, but after a year he left to set up his own business. Thereafter, Tesla was on his own, running his own laboratory or company in several different locations, with or without the collaboration of prominent businessmen such as J. Pierpont Morgan and George Westinghouse.

In his long career as an inventor, engineer, and scientist, Tesla contributed to many branches of science and technology, including electric lighting, robotics, radio transmission, and X rays. Undoubtedly, however, his main work was in the field of electrical engineering: the generation, transmission, and applications of electric power, particularly alternating currents. A crucial decision was made in 1893, when the Niagara Falls Commission (set up to harness the power of the falls to generate electricity) opted for the AC generators patented by Tesla and manufactured by the Westinghouse Corporation.

Like many of his contemporaries, in February 1896 Tesla began investigating the newly discovered phenomenon of Röntgen rays, and he occupies a prominent place in the list of American pioneers. However, his interest was relatively short-lived, and his last contribution to X-ray technology was not specifically developed for this field: the Tesla coil, an air-core transformer with primary and secondary coils tuned to resonate, was widely used as a means of generating the high voltage needed for the production of X rays. Tesla was one of the first investigators of Röntgen rays to realize the harmful effects of the radiation and to propose ways of rendering the apparatus safer. His strongly held belief that the X rays comprised streams of very small material particles proved to be incorrect, but this was the case for most of the early theories of the nature of radiation. A readable biography of Tesla is Margaret Cheney’s Tesla: Man Out of Time (New York: Dorset Press, 1981).
D. Coolidge, a physicist/engineer at the Research Laboratory of the GE Company at Schenectady, New York, published a paper in the Physical Review titled, "A Powerful Röntgen Ray Tube with a Pure Electron Discharge." (A condensed version of the paper, under the same title, appeared in the January 1914 issue of the American Journal of Roentgenology.)

Coolidge did not set out to produce an X-ray tube based on new principles. His initial research was concerned with the design and material of the anode (target) of the conventional X-ray tube. This was usually a platinum disc that tended to melt at the point of impact of the cathode rays and emitted so much gas on heating to redness that the tube had to be re exhausted. Coolidge solved this problem by developing a target of wrought tungsten, which has a very high melting point (≈3400°C centigrade), high density and heat conductivity, and a low vapor pressure at high temperatures. Having solved this problem, Coolidge became interested in the other limitations of the gas tube, eleven of which he listed in the opening of his paper. Apart from the difficulty of maintaining the vacuum at a constant level, these problems included the deposit of metallic copper (from the copper-backed target) on the walls of the tube, the cracking of the glass tube in the vicinity of the cathode, the wandering of the focal spot on the target, the melting of the central portion of the aluminum cathode, and the production of "a very heterogeneous bundle of primary Röntgen rays" when the tube was operated on a periodically intermittent current. Perhaps most important, "no two tubes are exactly alike in their electrical characteristics" and "the penetrating power of the Röntgen rays produced changes with the magnitude of the discharge current." The last point can be restated thus: In a gas X-ray tube it is impossible to control the quality (penetrating power) of the radiation independently of its intensity.

The X-ray tube developed by Coolidge differed from the gas tube by completely eliminating the role of positive ions in releasing electrons (cathode rays) by bombardment of the cathode. This was achieved first by reducing the gas pressure in the tube to the lowest value (a few hundredths of a micrometer of mercury pressure) attainable by the best vacuum technology—far below the level at which the gas tube would cease to operate—and second by employing, in both the anode and the cathode, a material (wrought tungsten) that had a very low vapor pressure and could be made free of occluded gas. This solution, however, necessitated an alternative source of electrons to provide the cathode stream for bombarding the target. Richardson had shown in 1902 and 1908 that the electrons were emitted by a hot filament by the process of thermionic emission, and had derived the relationship between the thermionic current (i) and the absolute temperature (T) of the filament (Richardson's Law). However, Coolidge pointed out that "the values of thermionic currents obtained by different observers had varied between wide limits, so much as to suggest that a Röntgen tube based on this principle might be as unstable in resistance as is the standard tube." Furthermore, the materials hitherto used as filaments—platinum and carbon—were difficult to obtain free of occluded gas and emitted copious vapor at high temperatures.

Fortunately, Coolidge's colleague at the GE Research Laboratory, Irving Langmuir, was studying the phenomenon of thermionic emission at the same time that Coolidge was researching the design of the X-ray tube. Langmuir investigated the effect of the "space charge" set up adjacent to a heated filament in a highly evacuated tube. He showed that as the gas pressure decreased, the thermionic current increased, up to a limiting value corresponding to the rate of emission of electrons by the filament, which depended on its temperature. The X-ray tube ("tube No. 147") described by Coolidge in his 1913 paper is shown in Fig. 1a. The bulb was of German glass about 18 cm in diameter.
William D. Coolidge (1873-1975)

William Coolidge was born in 1873 on a farm in Massachusetts. He attended the local grade and high schools and, in 1891, obtained a scholarship to the Massachusetts Institute of Technology (MIT), where he studied physical chemistry and electrical engineering. After receiving the B.S. degree in 1896, he transferred to the University of Leipzig in Germany, again on a scholarship. Coolidge obtained a Ph.D. in physics from Leipzig in 1899.

On returning to the United States, Coolidge became an instructor/researcher at MIT. His research (with Arthur Noyes) concerned the electrical conductivity of aqueous solutions at high temperatures. He remained at MIT until 1905, when he transferred to the Research Laboratory of the General Electric (GE) Company at Schenectady. The head of the GE Research Laboratory was Willis R. Whitney, whom Coolidge had known since his early days as a student at MIT.

Coolidge's first research at GE related to incandescent lamps. At the time these lamps were made with carbon filaments, but these were so fragile that the use of the lamps was impractical in many situations, such as in moving vehicles. Coolidge developed strong yet ductile tungsten as a replacement for carbon, and this made possible the marketing in 1911 of lamps that were both more efficient and mechanically robust.

Coolidge's work on tungsten-filament lamps led directly to the invention for which he is best known, the Coolidge X-ray tube, although in fact research on vacuum discharge tubes proceeded at the GE laboratory concurrently with that on incandescent lamps. Coolidge's first paper on the new X-ray tube, titled "A Powerful Röntgen Ray Tube with a Pure Electron Discharge," was published in the Physical Review in December 1913, although the first patents for the new tube were not issued until 1916. Despite the obvious advantages of the Coolidge tube over the gas tube, scientists and radiologists greeted the new invention with caution, and it was several years before the replacement of the gas tube was complete.

Coolidge remained at the GE Research Laboratory for the remainder of his career, becoming director in 1932 and, in 1940, vice-president of the company and director of research. During World War II he became a member of the National Academy of Science's Atom Bomb Project. He retired in 1945.

Among the many honors awarded to Coolidge were the Hughes Medal of the (U.K.) Institute of Electrical Engineers and, shortly before his death in 1975, election to the National Inventors Hall of Fame in the United States Patent and Trademark office in Washington, D.C. In 1972 the American Association of Physicists in Medicine established the William D. Coolidge Award in recognition of distinguished contributions to medical physics, and Coolidge was presented with the first award shortly before his one-hundredth birthday.
The filament was a flat, closely-wound spiral of thin tungsten wire surrounded by a tube of molybdenum, which acted as a focusing device. The target, which also served as the anode, comprised a single piece of wrought tungsten, with diameter 1.9 cm facing the cathode, supported by three split rings of molybdenum that fitted in the glass anode arm. Coolidge described the method of exhausting the tube and concluded that the pressure in the finished tube is "certainly not more than a few hundredths of a micron and probably much less than this."

The electrical connections of the original Coolidge tube are shown in Fig. 1.5. The filament current was provided by a small storage battery, and because the high potential was connected to the battery circuit, this circuit was well insulated from the ground. The high potential was supplied by a step-up transformer with oil insulation and a mechanical rectifier. The outside of the tube was cooled by a fan throughout its operation.

The latter half of Coolidge's paper comprised a detailed analysis of the characteristics of the tube. He concluded that "positive ions play no appreciable role" in the operation of the tube and that its main properties were: (1) the tube could be operated from either direct or alternating current; (2) the intensity and penetrating power of the X-rays were under the complete control of the operator and each could be instantly increased or decreased independently of the other; and (3) the tube could be operated continuously for hours, at either high or low discharge [tube] current, without appreciable change in the intensity or penetrating power of the radiation (there was no fluorescence, or local heating, of the glass tube). Coolidge concluded, "The tube permits of the realization of intense homogeneous pri-
Michael Idvorsky Pupin (1858-1935)

Michael Pupin was born in 1858 of Serbian parents in a village in Austria-Hungary. He was educated in local schools and, during vacations, helped at home on the farm. In 1874 his father died, and the young Pupin, then fifteen years old, decided to emigrate to America. For the first five years in his new country Pupin supported himself by manual work, while continuing his education by private study.

In 1879 Pupin was awarded a scholarship to Columbia University, where he studied Greek, mathematics, and physical science; he received the B.A. degree in 1883. After a short period in Cambridge he transferred to Berlin (1885) with a Tyndall Fellowship and received his Ph.D. in 1889 for a dissertation on theoretical physics. He returned to Columbia as an instructor in mathematical physics, and in 1901 he became professor of electromechanics, a post he held until his retirement in 1931.

Pupin’s research covered a wide range of problems in physics and electrical engineering, including vacuum tube discharge phenomena, electromagnetism (he invented the “electric resonator,” a circuit having a variable condenser and inductance), propagation of waves in a vibrating string (this had a practical application in long-distance telephony), and radio transmission.

Pupin was one of the American pioneers in the field of roentgen rays. The translation of Rontgen’s first paper, published in Electricity (New York) on 5 February 1896, was accompanied by a short explanatory article by Pupin, submitted on 1 February. However, the statements by Pupin’s biographers that he produced the first clinical X-ray photograph in the United States, early in January 1896, cannot be substantiated. Pupin himself made no such claim, and his first articles on X rays were illustrated by radiographs taken in England by Campbell-Swinton. The alleged “first,” a radiograph of a man’s hand embedded with lead shot, was published in Scientific American on 21 March 1896, but the date of exposure was not stated. Nevertheless, there is no doubt that Pupin made a significant contribution to the early study of X rays in America, including the observation of X-ray scattering and the first use of an intensifying screen, published in Science on 10 April 1896.

After World War I Pupin gave up research and devoted himself to writing and public speaking. His autobiography, From Immigrant to Inventor (1923), won a Pulitzer Prize in 1924. He was a member of the National Academy of Sciences. Pupin died in 1935.
mary Röntgen rays of any desired penetrating power." In this conclusion he was partially mistaken: even with a constant high potential across the tube, the resulting radiation is not homogeneous, and its penetrating power can be altered by filtration either in a metal filter or in the tissues of a human subject. However, Coolidge was working before the means of measuring X-ray wavelengths—and hence X-ray spectra—became available. The beam appeared to be homogeneous in its radiographic properties, and Coolidge’s conclusion is understandable if incorrect.

THE DETECTION AND RECORDING OF X RAYS

X rays were discovered by their ability to cause fluorescence in a screen coated with barium platinocyanide. To this property of the rays Röntgen soon added their ability to blacken a photographic plate. Both effects were weak, however, and the practical application of the radiation demanded (quite apart from a more intense source) more sensitive photographic emulsions or fluorescent materials. The search for more sensitive emulsions had limited success, except for the innovation of double-coated films, because it was soon realized that photographic sensitivity depended on grain size and was inversely related to image definition. Early work therefore concentrated on the search for better fluorescent screens.

From the beginning it was understood that fluorescence could be used in radiology in two distinct ways: as a means of rendering the X-ray image directly visible to the eye and as an adjunct to the blackening of the photographic emulsion. (It will be recalled that, in his original communication, Röntgen had stated that he did not know whether the blackening of a photographic plate by the X rays was a direct effect or an indirect one caused by fluorescence. It was not realized that the fluorescent material optimal for one effect was not necessarily optimal for the other.)

According to Edison’s biographer, soon after Röntgen’s discovery became known in North America, Michael Pupin of Columbia University appealed to Edison for help in discovering the most effective fluorescent chemicals. Edison and his assistants tested crystals derived from about eight thousand different chemical combinations, and within a few weeks were able to send Pupin a fluoroscope with a calcium tungstate screen, with which Pupin was able to make a clear shadowgraph of a man’s hand filled with shotgun pellets. Edison himself reported that calcium tungstate was six times more sensitive than barium platinocyanide as a fluorescent material. The next best compound was strontium tungstate; several other salts, including rock salt, also fluoresced. Edison constructed a fluoroscope in the form of a box, with a calcium tungstate screen at one end and a peephole at the other, which was exhibited at the Electrical Exposition in New York in May 1896. Thousands of curious visitors to the exposition were able to examine their own hands, held between the X-ray tube and the screen. This was the first display of X rays to the general public in America, possibly in the world.

Pupin made good use of Edison’s discovery of the fluorescence of calcium tungstate. In a paper presented before the New York Academy of Sciences on 6 April 1896, Pupin said that, with a satisfactorily working tube, tungstate of calcium “will show a noticeable fluorescence at a distance of over thirty feet.” By placing the fluorescent screen in contact with the photographic plate, Pupin was able to photograph a hand with an exposure of a few seconds at a distance of 4 inches from the tube, and the whole chest, shoulders, and neck of his assistant on a single plate at a distance of 5 feet and with an exposure of seventy minutes. This was almost certainly the first successful use of an intensifying screen for radiography. Indeed, in another version of his Academy of Sciences lecture, published in Electricity (New York) on 15 April 1896, Pupin specifically claimed priority for the intensifying screen. In an earlier (14 February) article, in which he discussed his own experiments with the new radiation, he wrote, “A fluorescent screen placed in
front of the sensitive plate for the purpose of shortening the time of exposure gave encouraging results. The claims of Edison and Pupin for priority in the use of fluorescent screens were not beyond dispute. As early as 13 March 1896 Edward P. Thompson of New York described, in a British journal, a “kinetoskotoscope” whereby the motion of the bones of the finger, when bent backwards and forwards, could clearly be seen on a fluorescent screen. He wrote “My idea for this use of X rays was conceived and attested on 1 February 1896.”

A modification of the fluoroscope was introduced by J. Mount Bleyer in July 1896. Instead of putting the fluorescent screen in contact with the photographic plate, he placed it at the end of a cone attached to the lens of an ordinary camera (Fig. 1.6). In this way the image on the fluorescent screen could be both seen and photographed, but the technique was not a success and the idea lay dormant for many years. The amount of light reaching the film in a camera is much less than that in the immediate vicinity of the fluorescent screen, and so the exposure required is proportionately longer. Bleyer’s technique was therefore suitable only for static images involving long exposures. Fifteen years later E. W. Caldwell, of New York, reexamined the idea with a view both to producing practical “roentgen cinematography” of moving images and to economizing on film by recording static images of reduced size in a camera. He concluded that it was necessary to develop not only faster films and more efficient screens but camera lenses of much greater relative aperture than those generally available at the time. Commercial production of both intensifying screens and fluoroscopes expanded rapidly, and soon at least eight different makes were available on the United States market. Yet, it was not until 1914 that a systematic study of their characteristics was undertaken. In February 1914 J. S. Shearer of Cornell University published the first of a series of papers in which he examined the properties of various combinations of intensifying screens and photographic plates. This paper was concerned with the relative speeds of screen/plate combinations; later papers dealt with detail, contrast, freedom from grain, lag, and durability.

**THE BASIC PROPERTIES OF X RAYS**

The early experiments with X rays by investigators in North America were essentially general, intended to reveal as much information as possible without focusing on any particular property. It is therefore not surprising that the results were confusing and often conflicting. Soon, however, scientists began to design experiments to elucidate individual properties such as reflection, refraction, and polarization. One of the first such experiments was carried out in March 1896, and modified in August 1896, by O. N. Rood of Columbia University. Rood obtained...
an image of a grid of iron wire by reflection from a platinum foil. He estimated that at 45 degrees the reflected radiation amounted to 1/260th part of the incident X rays and that the radiation reaching the film was a mixture of specularly and diffusely reflected rays.38 Tesla also investigated this problem and concluded that specular reflection did indeed occur; this finding fitted his theory that X rays were streams of particles of minute size.34

Meanwhile, Pupin had reached the opposite conclusion. In the April 1896 paper previously cited, he quoted Riögen himself to the effect that regular reflection does not exist but “bodies behave to the X-rays as muddy media do to light.”55 Pupin believed that the experiments of Rood and Tesla demonstrated diffuse but not specular reflection and that his own experiments proved that roentgen rays were diffusely scattered by all bodies, including gases.

Other early contributors to this topic were Alfred M. Mayer of New York and Elihu Thomson. Mayer supported Rood’s conclusion that specular reflection of X rays did indeed occur, but his experiments added little evidence to this effect.56 Thomson, in contrast, found no evidence of specular reflection. He was one of the first to observe that diffusion of X rays occurred not only from objects placed directly in the path of the beam but also from surrounding objects such as the walls and floor of the room. He noted, too, that bodies differed greatly in diffusing power: Substances such as paraffin wax and wood, which were comparatively transparent to X rays, were also “fairly vigorous” in diffusing them. This property was shared by liquids as well as solids. Furthermore, diffusion was not merely a surface phenomenon but occurred throughout the irradiated body. Thomson pointed out that this fact had an important implication in radiography: The shadow of an opaque object embedded in tissue could never be as dense as the shadow of the same object surrounded by air.57

In 1897 R. W. Wood of the Massachusetts Institute of Technology developed a new type of X-ray tube, consisting of a very small bulb in which an arc was struck between two minute platinum balls in a very high vacuum. This device emitted X rays of intensity, per unit area of radiating surface, up to twenty times greater than the best focus tube, and Wood used the very fine beam in an attempt to detect X-ray diffraction. With the shadow of a fine copper wire, he obtained an effect that he thought was diffraction, but he added, “I am in fact not yet at all sure that we are dealing with true diffraction.”58

Indeed, another fifteen years would elapse before the diffraction of X rays would actually be demonstrated.

Max von Laue was a Privatdozent (Lecturer) at the Institute of Theoretical Physics of Munich University in Germany in May 1912 when, together with his graduate students Friedrich and Knipping, he showed that the parallel layers of atoms in a crystal acted as a diffraction grating for X rays. This effect arose from the fact that the spacing of the layers was of the same order of magnitude as the wavelength of the radiation. In his first demonstration von Laue used a crystal of zinc sulfide, which has a cubic structure.59 Shortly afterwards W. H. Bragg, professor of physics at Leeds University in England, found that the spacing of the spots in the diffraction pattern produced by von Laue conformed to a simple numerical rule, and Bragg’s son, W. L. Bragg, a graduate student at Cambridge, advanced a theory to account for the pattern of spots and to explain his father’s numerical rule.60,61 Bragg Senior acknowledged his son’s theory and suggested that, although the new phenomenon clearly pointed to the wave nature of X rays, other properties of the radiation required a corpuscular theory: “The problem then becomes...not to decide between two theories of X rays, but to find one theory which possesses the capacities of both.”62 That theory is, of course, the quantum theory of radiation, which links the frequency of electromagnetic waves with the energy of the individual photons that constitute the beam.

The discovery of X-ray diffraction had no direct bearing on medical radiology, but it had a profound indirect
effect. Diffraction confirmed that X rays were electromagnetic waves of very short wavelength rather than streams of material particles. Up to 1912 the wave theory was widely believed but remained controversial. Diffraction provided a means of measuring X-ray wavelengths and hence of studying X-ray spectra, and conversely of studying the structure of materials, including in due course biological materials such as proteins and DNA.

The third basic property of electromagnetic radiation—if, indeed, X rays were of this nature, a widely held but unproven assumption before 1912—is polarization. This effect eluded all investigators, from Röntgen on, in the first few years after the discovery of X rays. For example, in March 1896 Alfred Mayer tried to demonstrate the polarization of X rays by passing the radiation through crystal plates of herapathite (iodo-sulfate of quinine), a low-density doubly-refracting medium that gave complete polarization of visible light (i.e., zero transmission through “crossed” crystals). When this configuration had no effect on the transmission of X rays, Mayer concluded that, unlike visible light, roentgen rays were not transverse electromagnetic waves.69

Charles C. Barkla, an assistant lecturer in physics at the University of Liverpool in England reached the opposite conclusion. If X rays were pulsed transverse waves, then electromagnetic theory postulated that the primary radiation should be polarized with respect to the direction of the cathode-ray stream, such that the intensity of X rays was greatest perpendicular to the cathode-ray stream and least in the direction of the stream. Similarly, the secondary radiation scattered by air and light solids should be polarized and show a maximum intensity perpendicular to the direction of the primary beam. In a series of papers published from 1904 to 1906, Barkla showed that both the primary and secondary radiation (produced in low atomic weight materials such as carbon) behaved as predicted.61,65,66,67,68 He concluded that the primary radiation was partially polarized and the secondary radiation fully polarized.

Barkla’s investigations were repeated and extended in 1910 by William Ham, working under R. A. Millikan at the University of Chicago. Ham measured the variation of X-ray intensity around the target of the X-ray tube and the effect of various absorbing materials on this distribution. Like Barkla, Ham found that the maximum intensity occurred in the plane through the target normal to the cathode-ray stream and fell off symmetrically on either side of this plane. The difference in intensity was increased by interposition of sheets of silver and tin but decreased by sheets of lead. The difference in intensity was also greater at lower kilovoltages across the tube. Ham interpreted these results as proof that the primary rays were polarized, as Barkla said, but that (contrary to Barkla’s conclusion) the secondary rays were unpolarized.69 While the general conclusion as to the polarization of primary X rays is accepted today, it is difficult to judge the measurements of Barkla and Ham in quantitative terms. The quality of their beams was not specified and they were unaware of the complexity of the interaction processes between X rays and matter resulting in secondary radiation; nor did they apparently take account of self-absorption in the X-ray target, which modifies the angular distribution of the radiation and gives rise to the so-called “heel” effect. For further discussion of the polarization of X rays, the reader is referred to the article on this topic in the Encyclopedia of X-rays and Gamma Rays.70

**Interchange of X Rays with Air**

Perhaps the most important area of research in the physics of radiology, and certainly the most intensely investigated in the first decade, was the interaction of X rays with air (and, to a lesser extent, gases in general), whereby air becomes a conductor of electricity at ordinary atmospheric pressure. This phenomenon had practical as well as theoretical importance, because the measurement of radiation intensity by means of an
ionization chamber and quadrant electrometer, or alternatively by means of a
gold-leaf electroscope, depended on the
creation of charged atoms (ions) by the
interaction of the radiation and the gas.
It was therefore important to under-
stand the nature of the interaction and
to measure the energy absorbed in the
ionization process. Much of the early
work in this area in North America was
carried out by scientists whose prime
interest was radioactivity rather than X
rays, and this topic is discussed later in
the chapter under the heading of
radioactivity. However, some of the
investigations did involve X rays, partly
because researchers were inherently
interested in this radiation, but also
because it was possible to obtain a much
more intense beam of X rays than rays
from radium or thorium.

One of the earliest workers in this
field was Ernest Rutherford. In January
1896, when Röntgen's discovery of X
rays was announced, Rutherford was a
research student at the Cavendish
Laboratory in Cambridge under J. J.
Thomson, who invited the young gradu-
ate to join him in investigating the con-
ducting power of gases exposed to the
newly discovered radiation. Thomson
and Rutherford published a joint paper
on this problem in November 1896. This
was followed in 1897 and 1898 by three
papers by Rutherford alone, in which he
investigated the relationship between
the ionization current and the radiation
intensity, as well as the rate of recombi-
nation of the ions produced by the radia-
tion. Rutherford's fifth paper on this
topic was published in January 1899, by
which time he had already moved to
Canada, although the work described
was carried out in Cambridge. For details
of these papers, see the Collected Papers
of Lord Rutherford of Nelson.

In 1898 Rutherford, a New Zealander
by birth, was appointed Macdonald
Professor of Experimental Physics at
McGill University. Although most of
Rutherford's work during his nine years
at McGill was specifically concerned with
radioactivity, he also worked with X rays
when the nature of the problem
demanded it or, as previously indicated,
when the experiment required a more
intense beam than could be provided by
a radioactive source. An important
example of this is Rutherford's investiga-
tion, carried out in 1900 and published
in 1901, of the energy required to pro-
duce a single ion in gases exposed to
roentgen rays. In this research Ruther-
ford was assisted by R. K. McIlwraith, a
demonstrator in physics. They first deter-
mined the total energy in the beam by measur-
ing the fraction of the radiation
absorbed in a grid of platinum wound
on a mica frame. The resulting (very
small) rise in temperature of the grid
was deduced from the measured
increase in resistance of the platinum.
Rutherford then measured the ioniza-
tion current produced in air and other
gases by the X rays under saturation
conditions (i.e., no recombination) and the
fraction of the radiation absorbed in a
given volume of the gas. The latter was
measured by means of two identical and
quasi-parallel brass tubes, 118 cm long,
through which a divided X-ray beam
passed before reaching identical ioniza-
tion chambers (Fig. 1.7). With one tube
evacuated and the other filled with a gas
under known pressure, the difference in
the currents measured in the two ioniza-
tion chambers allowed the absorption
coefficient of the gas to be calculated.
These experiments led to a value of W,
the average energy required to produce
an ion (or rather, an ion pair, positive
and negative), at normal atmospheric
pressure and temperature, of $1.90 \times 10^{18}$
ergs, equivalent to 120 electron volts
(eV). This is several times the present
value of 34 eV, but is nevertheless a high-
ly satisfactory result in view of the many
difficulties associated with the investi-
gation, including an unstable and inhomoge-
nous X-ray beam.

While Rutherford turned his atten-
tion to other properties of radioactivity
and ionizing radiation, discussed later in
this chapter, McIlwraith continued to
investigate the phenomena associated
with ionization in gases. In 1902 he
reported that the rate of recombination
of ions in air is proportional to the square
of the number of ions ($n$) present at a
given time, that is $dn/dt = \alpha n^2$, where $\alpha$ is
the coefficient of recombination. McClung found that \( \alpha \) is independent of the air pressure, within the range of 0.125 to 3.0 of the normal atmospheric pressure.  

However, in 1905 L. L. Hendren of Columbia University showed that \( \alpha \) does, in fact, decrease with decreasing pressure, to a value about half that at normal atmospheric pressure when the air pressure falls to 150 millimeters (mm) of mercury and to less than one-third the normal value at 10 mm.  

Hendren suggested that errors in this type of investigation were due mainly to diffusion of ions to the electrodes of the ionization chamber.

The problem of the recombination of ions in air was taken up again in 1909 by Henry Erikson at the University of Minnesota, using essentially the same method as that used by Hendren, except that a radium source was substituted for X-rays. The radium was placed at the center of a copper sphere 3 cm in diameter, which in turn was within a concentric sphere 11 cm in diameter, so that the two spheres constituted an ionization chamber.  

Erikson varied the temperature of the air within the chamber in the range 90° to 400° Absolute (-165° to 136° centigrade) and found that the coefficient of recombination of the ions, \( \alpha \), decreases as the temperature increases, by a factor of about 18 in the range of temperature indicated. In 1913 S. J. Plimpton of Yale University found that the recombination relationship \( \frac{dn}{dt} = \alpha n^2 \) is valid only if sufficient time has elapsed to enable an effectively uniform distribution of ions to be established in the gas. The coefficient \( \alpha \) has an initial maximum value and becomes a constant only when a uniform distribution in ions is reached.

The recombination of ions was an essential aspect of the interaction of X-rays with air, but it was by no means the only aspect worthy of study. A different type of investigation was undertaken by John Zelemy, a physicist at the University of Minnesota. Zelemy's interest in this subject began in 1898 when he was a graduate student at the Cavendish Laboratory under J. J. Thomson. Zelemy showed that when a metal surface is exposed to air that has been ionized by X-rays, the metal acquires a negative charge while the air itself becomes positively charged. This effect could be increased by blowing the charged gas away from the metal surface. Zelemy explained this result by postulating that the negative gaseous ions are smaller than the positive ions and therefore move more quickly in an electric field.  

Subsequently an Italian physicist, Emilio Villari, proposed a different hypothesis: When air that has been exposed to X-rays flows past a metal surface, the friction produces a separation of positively and negatively charged particles. After returning to America in 1903, Zelemy took up the problem again.
He devised an experiment in which air was exposed to X rays in a vessel and then blown into a metal tube in such a way that the length and shape of the tube, the velocity of the air, and other experimental conditions could be varied. The results confirmed Zeleny’s previous hypothesis concerning the unequal velocities of diffusion of the positive and negative ions in the gas, but did not uphold Villari’s frictional theory.80

Another property of ionized air was investigated in 1904 by Frederick Reynolds in the College of the City of New York. Reynolds’s main interest was the viscosity of air; he devised an experiment to determine whether the viscosity coefficient was altered when the air was exposed to X rays, but his careful and sensitive measurements showed no significant change.81

A somewhat different approach was taken by A. S. Eve in Montreal, also in 1904. Eve was a colleague, lifelong friend, and subsequent official biographer of Rutherford. Eve was primarily concerned with the nature of the gamma rays emitted by radium and other radioactive sources. Were gamma rays essentially highly penetrating X rays? Evidence to this effect was accumulating, but the ionizing properties of the two radiations in various gases and vapors were quite different. For gamma rays the ionizing currents in different gases, relative to that in air, are approximately proportional to the densities of the gases. For diagnostic X rays, however, no such proportionality is found; for example, the ionization current in methyl iodide is seventy-two times that in air, although the relative density is only 5.0. Rutherford suspected that the discrepancy was due to the softness of the X rays employed in the test, because the result varied considerably with the type of X-ray tube employed. He therefore suggested that the measurements be repeated with the penetrating X rays produced in a “hard” tube. Eve found that, with this radiation, after filtration through 1.7 mm of lead, the discrepancy was much reduced, although not completely eliminated. Thus the relative ionization current for methyl iodide decreased to 13.5 and that for carbon tetrachloride decreased from 45 (earlier value) to 49, for a relative density of 5.8.82

In evaluating these early results it must be borne in mind that at the time (1904) there was no way of expressing the quality of an X-ray or gamma-ray beam and no possibility of measuring their spectra. Furthermore, the mechanisms of interaction between the radiation and air or other absorbing material, such as photoelectric absorption and the Compton process, were totally unknown. Rutherford and Eve were right in suggesting the essential similarity of X and gamma rays, but they did not realize the magnitude of the discrepancy in wavelength (today we would say photon energy) between the gamma rays and the X rays then available.

Eve continued his ionization studies at McGill for many years after Rutherford’s departure in 1907. (It is perhaps worth noting that, although they were close colleagues, Rutherford and Eve never published a joint paper.) In 1910 Eve studied the effect of dust, smoke, and mist on the ionization of air produced by the gamma rays of radium. He found that the number of ions detected can vary by as much as 50 percent, depending on the amount of these contaminants present in the atmosphere. Negative ions rapidly combine with particles of dust and smoke; in this way small ions are replaced by large ions, which move and recombine more slowly. The overall effect is to reduce the conductivity.83

Two other ionization studies by Eve, both published in 1912, require mention. In the first investigation, Eve, with a graduate student, F. H. Day, measured the absorption coefficient of X rays in air for radiation described as “soft,” “moderate,” and “hard.” The absorbing column of air was either “short,” (4 to 10 meters) or “long” (40 to 60 meters). Corrections were applied for the effect of scattered radiation reaching the measuring electroscope from the air, roof, floor, and walls of the laboratory, and a monitoring electroscope in a fixed position allowed corrections to be made for fluctuations in the X-ray out
For the moderate X-ray tube, the coefficients of absorption were about 0.0004 and 0.0003 for the short and long ranges, respectively (the units were not stated). The corresponding values for the hard X-rays were about 0.0003 and 0.0001 respectively. Thus not only does the absorption of X-rays in air depend on the beam quality, but the beam was shown to harden (i.e., increase in penetrating power) on its passage through air.85

The second study by Eve, published shortly after the paper just described, concerned the role of the walls of a metal container in determining the level of ionization in an electroscope housed inside the vessel. Identical cylindrical vessels were constructed of copper, zinc, iron, and aluminum about 0.5 mm thick. With soft X-rays the ionization measured by the electroscope increased by a factor of more than three as the wall changed from copper to aluminum. With hard X-rays the corresponding increase was about 50 percent. However, with gamma rays from the radium source the corresponding change was a decrease of about 15 percent. Eve explained these results by postulating that the metal wall absorbed both the primary and the secondary X-rays and thereby excluded them from producing ions within the vessel. This effect decreased as the wall material changed from copper to aluminum of the same thickness. However, with gamma rays the measured ionization was due to high-velocity electrons expelled from the walls by the radiation, and this effect was greater for copper than aluminum.86

An investigation of great importance, not merely for the physics of radiology but for science as a whole, was that of R. A. Millikan, professor of physics in the University of Chicago. Between 1909 and 1913 Millikan carried out historical experiments that resulted in the first precise value of the charge on a single ion. The method was to isolate a single droplet of a liquid, charge it with a single ion, and measure the speed of its fall in a gravitational field and then in a combined gravitational and vertical electrical field. In his first experiments Millikan used droplets of water or alcohol, but these were subject to errors due to evaporation and other causes, and in his later investigations he substituted minute droplets of oil.87 The apparatus used by Millikan in 1911 is shown in Fig. 1.8.88 An atomizer (A) enabled a cloud of fine droplets of oil to be blown by dust-free air into a dust-free chamber (C). One or more of the droplets were allowed to fall through a pinhole (p) into the space between the plate (M, N) of a horizontal air condenser, and the pinhole was then closed by means of an electromagnetically operated shutter. The movement of the droplet between the plates was observed by means of a telescope and a narrow beam of light illuminating the drop. The electric field between the plates—about 5,000 volts in magnitude—pulled the droplet upwards against the force of gravity. The time required for the droplet to fall under gravity through a known distance (determined by crosshairs of the observing telescope) was measured, with and without the counteracting electric field. Millikan's 1913 apparatus was a modified version of that shown in Fig. 1.8, but the principle was unchanged.89
Millikan’s 1913 measurements resulted in a value of the elementary electric charge of $4.774 \times 10^{-10}$ electrostatic units (esu), equivalent to $1.592 \times 10^{-10}$ coulombs. The present accepted value is $1.602 \times 10^{-19}$ coulombs. This work also enabled a number of other basic constants to be derived, including Avogadro’s number (the number of molecules in a gram molecule of any substance) = $6.026 \times 10^{23}$ (modern value $6.022 \times 10^{23}$) and the mass of the hydrogen atom = $1.70 \times 10^{-24}$ grams (modern value $1.67 \times 10^{-24}$ grams).

An important offshoot of Millikan’s oil-drop experiments was his 1911 investigation of valency in gaseous ionization. Although the negative ions were considered to constitute a single elementary electric charge, there was some evidence, albeit inconclusive, that positive ions produced by X rays were bivalent (i.e., the act of ionization was thought to detach two elementary charges from the neutral molecule). Millikan investigated this problem by means of the capture on oil drops of the products of ionization by X rays. He concluded that his experiments gave “direct, unmistakable evidence” that the act of ionization of air molecules by hard and soft X rays, as well as by beta and gamma rays, always resulted in the detachment from a neutral molecule of a single elementary electrical charge.\textsuperscript{90}

**THE INTERACTION OF X RAYS WITH SOLIDS AND LIQUIDS**

The interaction of X rays with solid and liquid materials is a topic of great interest and importance. It is the key to understanding both the formation of radiological images and the biological effects of radiation. Furthermore, the design of protective barriers depends on a knowledge of the attenuation of radiation in different materials and the emission of secondary radiation. It is not surprising, therefore, that in the period under review in this chapter, a considerable effort was directed to this area of investigation, both in North America and in Europe.

Much of the early work on attenuation involved gamma rays rather than X rays, because gamma-ray sources were considerably more penetrating than even so-called hard X rays, and their penetration could be studied for greater thicknesses of material. Where appropriate, therefore, this section includes investigations involving gamma rays as well as X rays.

One of the earliest systematic studies of X-ray absorption in liquids was the 1902 investigation by McClung and McIntosh, students of Rutherford at McGill University. They used an ionization chamber and quadrant electrometer as the measuring system. A narrow X-ray beam (5 x 0.6 cm) was passed through a rectangular box holding the layer of liquid under investigation, 2.6 cm thick, and solutions of chlorides and sulfates of various densities were measured. McClung and McIntosh found that the percentage absorption increased with the density of the solution, but with an approximately logarithmic, rather than a linear, relationship.\textsuperscript{91}

In 1906 Henry Bunstead of the Sloan Physical Laboratory at Yale found that when X rays were equally absorbed in sheets of lead and zinc, about twice the amount of heat was generated in the lead as in the zinc.\textsuperscript{92} However, when Bunstead repeated the experiment in 1908, he concluded that his previous result was erroneous, because there was a greater loss of heat by the zinc plate. Bunstead now concluded that for equal absorption there was equal generation of heat, within the experimental uncertainty of 5 to 10 percent.\textsuperscript{93}

In 1907 John Mead Adams, working in the Jefferson Physical Laboratory at Harvard, investigated the absorptive properties of aluminum, copper, silver, and tin, each by itself and in pairs.\textsuperscript{94} His detector was a platinum disc, 3.8 mm in diameter and 0.014 mm thick, connected to a copper-constantan thermocouple that in turn was incorporated in a loop of copper wire carrying a small mirror (Fig. 19a). The loop was suspended between the poles of a thermally insulated permanent magnet (NS), and a thin aluminum window (A) in the housing allowed a narrow X-ray beam to reach the platinum
disc (Fig. 1.9b). The intensity of the X rays was measured by
the deflection of the mirror, which, in turn, depended on
the rise in temperature of the disc. The total energy in the
beam at the position of the disc, 0.003 gram-calories per
second, was calculated from the measured rise in tempera-
ture and found to be in line with values deduced by other
investigators by different methods.

Adams found that the sur-
face of the metal sheet had no
influence on its absorption of
the X rays. The absorption
curves in copper and alu-
minum were the same, within
experimental error, whether measured
with single-piece or laminated plates
(Fig. 1.10). By investigating metals in
pairs he found that the effect of one
metal was to harden the beam for
transmission through the other. But
there appeared to be an exception;
Transmission through aluminum appar-
ently softened the beam for subsequent
transmission through silver. Adams con-
cluded that X rays are heterogeneous
and that the absorption in metals "is
more or less selective." 93

Adams followed up the previous
publication with another short paper,
published only a month later. 94 For this
investigation he constructed an X-ray
tube with the target in the form of a
platinum disc bent into an arc of radius
5 cm. The cathode stream was deflected
by a strong magnetic field, and the
deflection of the individual particles
depended on their velocity. The focus
was thus spread out on the target into a
spectrum, and the X-ray beam spread
into a band when the target was viewed
on a fluorescent screen through a pin-
hole. The separation of the X rays into a
spectrum was confirmed by putting a
sheet of copper lengthwise over one-half
of the band. The existence of selective
absorption in the case of aluminum and
silver was shown by photographing half
of the spectrum through a sheet of one

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Fig. 1.9a Adams's thermojunction-type detector used for measuring the absorptive properties of metals.

Fig. 1.9b Detector suspended between the poles of a thermally insulated permanent magnet. (Phil. Mag., 1907)

Fig. 1.10 Adams's absorption curves of copper and aluminium, measured with single sheets of varying thickness (dots) and laminated places (crosses). (Phil. Mag., 1907)
metal and the other half through the other metal, their thicknesses being such as to equalize absorption of the hardest rays. The rays in the other part of the spectrum were transmitted more by the silver than by the aluminum sheet. Adams concluded that selective absorption by metals was proved but does not follow the same law in all metals. However, he had no explanation for the effect.

It is difficult to offer a precise explanation of Adams's results, especially in as much as he specified neither the kilovoltage applied to the X-ray tube nor the beam quality in terms acceptable today. From the absorption curves in Figure 1.10 we may deduce a value of about 27 kiloelectron volts (keV) for the photon energy of an equivalent homogeneous beam of X rays, corresponding to a heterogeneous beam generated at 55 to 60 kilovolts-peak (kVp). It is highly probable that Adams had observed "K-edge" absorption and the production of secondary characteristic radiation, which occurs in silver at 23.5 keV. These phenomena were investigated by Barkla at King's College, London, between 1909 and 1911. In a series of papers relating to the absorption of X rays in metals and the resulting secondary radiations, Barkla showed that, in addition to scattered radiation similar to the primary beam, "homogeneous fluorescent" radiation was produced, characteristic of the bombarded atom.97,98,99,100

A follow-up investigation was made by S. J. Allen at the University of Cincinnati in 1912 and 1913. In the first of two papers, Allen used a gold-leaf electroscope to measure the absorption of radium gamma rays in a large number of metals as well as in liquids and aqueous solutions.101 He divided the measured coefficient of absorption (A) by the density (d) of the absorbing substance and obtained the stopping power per unit mass of material. A/d was found to be a function of the atomic weight (A) of the absorber, increasing very slowly with atomic weight until A = 100, then more rapidly. Allen confirmed that the radiation was hardened by passage through any material, the degree of hardening being a function of atomic weight. He also noted that the absorption of gamma rays by liquids is an additive function of the atomic weights of the constituent atoms, independent of their chemical grouping.

In a follow-up paper Allen and Lorentz compared the absorption of X rays and soft gamma rays (as distinct from the hard gammas used in their previous work).102 He had hoped to close the gap between the transmission of X and gamma rays but was unable to do so, the value of λ/d for hard X rays being four times greater than that of the softest gammas. Allen observed that the absorption of X rays increased in general with the atomic weight of the absorber, but like Adams he found that some substances, notably silver, showed anomalous results. He concluded that the anomalous behavior had not yet been completely explained, but "it is quite probably that the fluorescent secondary X radiation discovered by Barkla must have much to do with it."

Finally, in the period under review, Harold Wilson, of the Rice Institute in Houston, Texas, published a short paper in 1914 in which he showed that the observed distribution of scattered radiation around an absorbing layer of metal could be explained by assuming approximately equal contributions of electrons distributed at random and of electrons distributed regularly in crystal arrays and subject to diffraction according to von Laue. He concluded as follows:

The only property of Röntgen rays which is not easily explicable on this theory seems to be that of causing the emission of high-velocity electrons. The most reasonable way of explaining the emission of these electrons seems to be that proposed by Planck, according to which matter absorbs radiant energy continuously but only emits it in definite amounts proportional to the wave length of the radiation.103

**THE MEASUREMENT OF X RAYS**

In the preceding sections of this chapter many investigations have been described in which a property of X rays
such as their ability to penetrate different materials—was measured by means of some other property of the radiation. The "other" property was initially the blackening of a photographic plate, but it was soon realized that this method was slow and inaccurate. Apart from the variability of the sensitivity of photographic plates and of development techniques, the opacity of the plate was not readily related to the "quantity" or "intensity" of the radiation, however these terms might be defined. The ionization chamber soon replaced the photographic plate as the technique of choice for investigating the properties of X rays, although the photographic plate (or, alternatively, the fluorescent screen) remained supreme in the clinic, where the main concern was to compare the imaging capabilities of different X-ray tubes. As stated earlier, the physical basis of the ionization chamber was investigated at the Cavendish Laboratory in 1896 by Thomson and Rutherford and in 1897 and 1898 by Rutherford alone. Rutherford's first paper as Macdonald Professor of Physics at McGill University (although the work described was carried out at Cambridge), titled "Uranium Radiation and the Electrical Conduction Produced by It," was published in January 1899. Thus the measurement of ionization in air, as a means of quantifying both X rays and the radiations emitted by radioactive substances, was well established by the turn of the century and remained the principal measurement technique throughout the period under review.

Usually the change in potential across a parallel-plate or cylindrical ionization vessel was measured by means of a quadrant electrometer, one pair of quadrants being connected to the charged plate of the ion chamber and the other pair to earth. The electrometer was essentially a voltmeter of very high resistance, and the rate of deflection of the needle suspended between the quadrants, usually described as the "rate of leak," was a measure of the intensity of the radiation incident on the ion chamber. Nevertheless the quadrant electrometer had certain limitations for accurate quantitative work. The rate of movement of the needle was not always uniform but was subject to sudden variations arising from static charges in the neighborhood or other artifacts. Furthermore, the relevant quantity was a rate (i.e., the change in needle deflection in a given time), not the instantaneous deflection. This rate was difficult to measure both for strong ionizations (which produced a very rapid change in deflection) and for rapidly decaying radioactivity (in which the activity changed significantly during the period of measurement).

In 1905 Howard Bronson, an American research student working under Rutherford at McGill, devised a solution to this problem: He connected a pair of metal plates in parallel with the charging quadrants of the electrometer. The lower plate was connected to earth and held a strong radioactive material of long half life. This converted the air between the plates into a high resistance, with a discharge current opposing the ionization current in the measuring chamber. When the two currents were equal, the electrometer needle took up a position of rest proportional to the intensity of ionization in the measuring chamber at that instant.

A modification of the Bronson device was described in 1907 by S. J. Allen, an instructor in physics at the University of Cincinnati. The principle of Allen's method is illustrated in Fig. 1.11. The radioactive material on the lower plate of the balancing chamber (S) was a thin uniform layer of uranium oxide, which was constant in activity and gave off no emanation. The ionization current in S could be controlled by a sliding cover (g), which effectively shielded the radioactive layer. Balance was obtained by sliding the cover until a steady reading of the electrometer needle was obtained; this served merely as an indicator of the balance, the relevant reading being that on the scale attached to the sliding cover.

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The ionization chamber with quadrant electrometer, with or without a balancing current as just described, was an excellent means of investigating the properties of X rays and radioactive materials in relative terms but did not measure the absolute intensity or quantity of radiation in standard physical units. For that purpose a defined and reproducible unit of radiation quantity was required or, alternatively, a standard radiation source against which all other sources could be calibrated. Such a standard was clearly unattainable in the case of X rays, although a standard radioactive (radium) source was feasible. In fact, it was prepared by Marie Curie in 1913, following a decision of the International Radium Standards Committee, which had been set up in 1910 at the first International Congress of Radiology (ICR) and Electricity in Brussels. For X rays, however, there was no alternative to a defined unit of radiation quantity.

The credit for first defining a realizable unit of radiation quantity is usually given to Paul-Ulrich Villard, a French physicist who, in 1908, proposed that “The unit of X-ray quantity is that which liberates, by ionization, one electrostatic unit per cubic centimeter of air under normal conditions of temperature and pressure.” It was, in fact, twenty years later that a modified version of Villard’s definition was adopted by the second ICR as the unit of radiation quantity, the roentgen. (The roentgen was subsequently redefined as a unit of “dose” and then of “exposure,” before being discarded altogether; see, for example, the historical review given by Harold O. Wyckoff in 1980.)

The basic idea of the roentgen unit had apparently occurred to others even before Villard’s proposal. For example, in 1907 C. L. Leonard of Philadelphia, in the discussion following a lecture on “The Standardization of Radiations,” made the following suggestion: “Electrical units are definitely known and determinable.... Why should not a unit be adopted that can be expressed in these known and commonly employed terms: A unit of radiation will then be the unit quantity of electricity passing across a unit gap, in unit time, under the influence of radiations at unit distance under standard conditions of barometer and temperature.” Leonard did not realize that neither the unit time nor the unit distance factor was needed in such a definition.

A completely different approach to the problem of X-ray measurement was taken by a Dr. Peabody in a lecture published in the American Quarterly of Roentgenology in 1910. (The author’s initials and affiliation were not stated.) He analyzed the factors determining the exposure needed for any radiographic examination and produced a formula, based on a large number of measurements, involving the distance between the X-ray target and the photographic plate, the thickness of the tissue traversed (deducting the
thickness of cavities but not differentiating bone and muscular tissue), the tube current, and the reading of a Wehniert penetrometer. However, in the discussion following the lecture, Dr. E. W. Caldwell, of New York City, commented that:

...the very excellent penetrometer which he described is not a safe nor convenient instrument to use...in making rapid exposures the change in penetration in a tube in one second may be enormous...I do not believe the chief difficulty we have in making plates is in estimating the length of exposure...What bothers us most is to get the right kind of ray for a sufficient length of time and...the suppression of undesirable rays, which are always present in a more or less degree.

At the time of this discussion the Coolidge X-ray tube was still over three years in the future.

THE DISCOVERY OF RADIOACTIVITY

The news of Röntgen's discovery of X rays was disseminated early in January 1896. One of the first scientists to consider the consequences of the new phenomenon was Antoine Henri Becquerel, a professor of physics at the Conservatoire National des Arts et Métiers (National Academy of Arts and Crafts) and the Museum of Natural History in Paris. Becquerel's interest was aroused by the fact that visible fluorescence and penetrating X rays were apparently generated simultaneously by the action of cathode rays on the glass walls of the discharge tube. Could it be that the same mechanism was producing both the visible and the invisible radiation? If so, were X rays produced also in luminescent crystals, that is, crystals that emitted visible light on absorption of the ultraviolet radiation in sunlight? In late January 1896 Becquerel began a systematic search for crystals that emitted penetrating radiation.

On 24 February 1896 Becquerel reported to the Academy of Sciences in Paris that fluorescent crystals of a uranium salt, potassium uranyl sulfate, exposed for several hours in sunlight, had blackened a photographic plate wrapped in opaque paper. This result appeared to confirm Becquerel's theory that the penetrating radiation was a fluorescent phenomenon, activated by the ultraviolet rays in sunlight. On 26 February he prepared the items needed to repeat the experiment, i.e., plaques of fluorescent crystal in contact with wrapped photographic plates, with a layer of aluminum partially interposed between the uranium salt and the plate. Unfortunately, on that day the weather was dull and, in the absence of bright sunshine, Becquerel put the plaques away in a drawer. The following days were also cloudy, and the plaques remained in the drawer in total darkness. On 1 March Becquerel took out the plaques and decided to develop the photographic plates, expecting to find, at best, a very weak blackening. To his surprise, an intense image of the uranyl plaque appeared. On 2 March he reported this finding to the Academy and stated his intention to investigate the phenomenon systematically with the hope of identifying the "exciting radiation" responsible for both the instantaneous emission of visible light (fluorescence) and the long-lived emission of an invisible but penetrating radiation. In the following weeks Becquerel published three more reports in which he showed that the properties of the penetrating radiation were similar to those of Röntgen's recently-discovered X radiation, including its ability to render air conducting and to discharge an electroscope. This phase culminated in a short paper, read to the Academy on 18 May 1896, announcing that the penetrating radiation was emitted not only by the salts of uranium—the presence of the element uranium was essential—but even by uranium metal itself, which had no fluorescent property. Thus the assumed link between fluorescence and the emission of the unknown radiation was finally broken, although Becquerel continued to believe that the underlying cause of the phenome-
non was energy absorbed from an unidentified exterior source.

It is interesting to note that X rays and radioactivity were discovered by chance, and it has been suggested that neither Röntgen nor Becquerel deserves the credit usually given to them for these discoveries. As Lawrence Badash has forcibly argued, however, only the "prepared mind" recognizes the significance of an unexpected observation or event and follows it through to a logical conclusion.¹⁰⁷

**Pioneers of Radioactivity: The Curies**

The five principal pioneers of radioactivity were Becquerel, Pierre and Marie Curie, Rutherford, and Frederick Soddy. Becquerel is remembered chiefly for his discovery of the phenomenon, as first discussed. He continued to investigate radioactivity for five years after his initial discovery and made further significant contributions, although his work was limited by his unwillingness to theorize on the nature of the phenomena involved. Rutherford and Soddy, in contrast, did not hesitate to advance theories to explain their experimental results and to suggest new approaches, including their joint theory of radioactive transformation (1902) and Rutherford's nuclear model of the atom (1911). The work of Rutherford and Soddy is discussed later in this chapter; here we are concerned with the contributions of the remaining two pioneers, Pierre Curie and Marie Skłodowska Curie.

The most important scientific work of the Curies was carried out between 1897 and 1903. In April 1898 Marie Curie reported to the Academy of Sciences that she had examined a large number of metals, salts, oxides, and minerals in order to determine whether any substances, other than the potassium uranyl sulfate used by Becquerel, emitted radiation capable of rendering air conductive.¹¹⁸ For this purpose she used a parallel-plate ionization chamber, with a thin layer of substance under test on the lower plate. The distinguishing feature of this study was the use of the piezoelectric quartz electrometer, invented earlier by Pierre Curie and his brother Jacques, to provide a quantitative measure of the activity of each sample. Marie Curie found that only compounds of uranium and thorium, besides the metals themselves, showed any significant activity, and she noted that both elements were of high atomic weight. (However, her discovery of the radioactivity of thorium had been narrowly preceded by Gerhard Schmidt in Erlangen.)¹¹⁹ Although the activities of the uranium-containing samples varied, each activity was roughly proportional to the amount of uranium element in the compounds and independent of the physical state of the sample: The activity was clearly an atomic rather than a molecular phenomenon. Furthermore, measurements of absorption by layers of various materials showed that the same radiation was emitted by all the compounds of uranium. The radiation emitted by thorium, however, was both more copious and more penetrating than that of uranium. Most important of all, the uranium minerals pitchblende and chalcolite were both more active than uranium metal itself. Marie Curie concluded that "these minerals may contain an element much more active than uranium." The search for this element—or elements—was the central theme of the Curies' work during the next four years.

On 18 July 1898, three months after her first report, Marie Curie and her husband reported to the Academy of Sciences that repeated chemical separations on a sample of pitchblende had yielded a "radio-active" substance—this was the first time the term was used in a publication—whose emissive power, as measured by the technique already described, was four hundred times that of uranium metal.¹²⁰ The new substance appeared to be an element with chemical properties similar to bismuth—it was, in fact, difficult to separate from the bismuth in the mineral—and "if the existence of this new metal is confirmed, we propose to call it polonium, after the name of the country of origin of one of us."

The Curies continued their efforts to separate polonium from pitchblende and on 26 December 1898 published a
further report with G. Bémont in which they noted the discovery of a second radioactive element even more powerful than polonium. The new substance, which they called radium, was chemically similar to barium (but barium was inactive), and it was difficult to separate radium from barium by standard chemical procedures. The existence of a new element was indicated by the appearance, in the spectrum of the sample, of a strong line unknown in any previous spectra. The radioactivity of the sample was nine hundred times that of uranium metal. Furthermore, samples of both radium and polonium gave strong photographic impressions with exposures of thirty seconds, whereas several hours were required with uranium and thorium.

During the next three years the Curies worked tirelessly on the difficult task of isolating pure polonium and radium from the ton of uranium mining residues donated by the Austrian government; the story is too well known to need repeating here. By November 1899 they had prepared samples of the two metals with activities five thousand to fifty thousand times that of uranium, and in a second paper published that month Marie Curie reported her measurements of the atomic weight of the metal in radium-containing samples of barium chloride. She showed that the atomic weight increased with the measured radioactivity of the sample, reaching 146 for a sample of activity seventy-five hundred times that of uranium, compared with 137.5 for pure (nonradioactive) barium. At this stage she could conclude only that the atomic weight of radium was greater than that of barium; it was not until July 1902 that she was able to make a definitive measurement, using the 100 milligrams (mg) of pure radium chloride obtained by repeated fractionation of the barium radoflue at her disposal. The result was 225±1, and radium was finally confirmed as a bivalent element.

The active samples of radium-containing uranium minerals prepared in 1899 enabled the Curies to demonstrate, in November of that year, the phenomenon of induced radioactivity: Substances irradiated by polonium and radium became themselves radioactive; the degree of activity increased with the duration of exposure, up to a limit, and gradually decreased after the removal of the exciting radiation.

In 1901 Pierre Curie and his colleague, André Debierne, published three papers on radioactivity induced by radium salts; they showed that this activity could be transmitted even to distilled water when the water and a solution of the radium salt, each in a separate open jar, were placed side by side in a closed vessel. The radioactivity was said to be communicated through the medium of the air in the vessel. The radioactive water lost its activity over a period of several days if kept in a closed vessel but much more rapidly in an open vessel. Similarly, the activity of a solution of radium salt decreased if left in an open vessel, especially if the fluid surface was large, but regained its original activity in about twelve days if the deactivated solution was placed in a sealed vessel. Curie suggested that the "radioactive energy" accumulated in radium was dissipated in two different ways: by the emission of radiation and by "conduction" to a nearby body through a gas or liquid.

Concurrent with the Curies' investigation in Paris of induced radioactivity were studies of the same phenomena by Ernest Rutherford and his group in Montreal. Rutherford demonstrated radioactivity induced (he used the term excited) by thorium, not directly by the primary radiation but indirectly via a vapor-like substance, associated with the thorium, which he called emanation. By mid-1901 Rutherford had not only demonstrated the gaseous nature of emanation but had investigated the emanation of radium as well as thorium and estimated its atomic weight. This work is described in more detail in the next section. By June 1901 Rutherford had published five papers on emanation and excited radioactivity, including a summary in Nature, but the papers by the Curies in this period...
contained no mention of Rutherford's work.\textsuperscript{126} They suggested that induced radioactivity was a phosphorescent phenomenon analogous to the production of secondary radiation by X rays.

Between 1900 and 1902, in addition to the papers already cited, the Curies published several other reports relating to the properties of radium rays, which were "deviable" and "non-deviable" in a magnetic field, and showed that the deviable rays were more penetrating and were negatively charged.\textsuperscript{129,130} They did not, however, adopt the terminology suggested by Rutherford in January 1899 (i.e., alpha and beta rays for the non-deviable and deviable rays, respectively). In January 1902, in a rare theoretical paper, Pierre and Marie Curie suggested that the most plausible explanation of the phenomenon of radioactivity was the continuous absorption and transformation, by radioactive bodies, of an unknown source of energy pervading the universe.\textsuperscript{131}

This phase of the Curies' work ended in June 1903, when Marie Curie presented her doctoral thesis, "Recherches sur les substances radioactives" ("Investigations on Radioactive Substances") to the Faculty of Sciences of the University of Paris. By this time she had isolated 100 mg of pure radium. Thereafter her research was concerned mainly with the properties of the new element and its applications, especially in medicine.

**Rutherford in Montreal: Emanation and Excited Radioactivity**

In our discussion on the measurement of X rays we mentioned that Ernest Rutherford's interest in radioactivity began in 1898 when, as a graduate student in Cambridge, he investigated the ionization of air induced by uranium and its compounds. Previously, his main focus had been the mechanism of ionization, but this project marked a shift of interest to the use of ionization as a tool for the study of radioactivity. The uranium investigation was published in January 1899, by which time Rutherford had moved to McGill.\textsuperscript{132} He spread uranium metal or compounds in powdered form on a horizontal zinc plate that formed the lower electrode of a parallel-plate ionization chamber. The resulting ionization current was measured by a quadrant electrometer connected to the upper electrode. Rutherford placed successive layers of thin aluminum foil over the uranium compound and noted the radiation intensity (i.e., the "rate of leak" of the electrometer) for each additional layer. He found that the intensity diminished according to the ordinary (exponential) law for the first three layers, but that after the fourth thickness the intensity fell only slightly when another eight layers of aluminum were added. Rutherford concluded that the uranium radiation is of two distinct types, a readily absorbed component and a more penetrating component, which he termed the alpha and beta rays respectively. The third, even more penetrating, component of the radiation was discovered by Villard in 1900 and later termed gamma rays.\textsuperscript{133,134}

The investigation in which Rutherford identified the alpha and beta components of uranium radiation also included a study of the radiation emitted by thorium compounds.\textsuperscript{135} At this time—late 1898 and early 1899—radium sources, even of low purity, were unavailable (the very existence of radium was unknown before the end of 1898), but high-purity thorium compounds were readily available.\textsuperscript{136} Rutherford noted that thorium radiation was more penetrating than uranium radiation and that thorium nitrate was not a steady source of radiation because "the rate of leak varied very capriciously, being sometimes five times as great as at others." This casual observation proved to be of great importance: He found that the "capricious" behavior of thorium nitrate was even more marked with the oxide, and was caused by slight air drafts around the radioactive material in the measuring chamber. In a paper published in January 1900 Rutherford explained the effect by postulating the emission by thorium, in addition to radiation, of large numbers of "radioactive particles" that could penetrate consider-
able thicknesses of materials such as paper.\textsuperscript{137} The nature of these particles was unknown, and in order to avoid the use of terms such as gas or vapor, Rutherford coined the term \textit{emanation}. Emanation (thoron and radon) was destined to play a central role in elucidating the nature of radioactivity.

In an ingenious experiment (Fig. 1.12) Rutherford separated the emanation from the thorium compound and showed that its radioactivity diminished rapidly, with a half life of about a minute, while the activity of new emanation from the "de-emanated" thorium built up at an equal but inverse rate (Fig. 1.13). Furthermore, the positive ions produced in a gas by the emanation were able to induce radioactivity in all substances on which they fell. In February 1900 Rutherford termed this effect \textit{excited radioactivity}.\textsuperscript{138} It was clearly the same phenomenon as \textit{induced radioactivity} observed by the Curies, but the suggested mechanisms were quite different.

Toward the end of 1900 Rutherford obtained a sample of reasonably pure radium from Germany and was able to undertake studies of the emanation produced by this newly discovered element whose radioactivity was much greater than that of thorium. He showed that when a radium source was heated below red heat, its emanating power increased over ten thousand times, but this power was largely destroyed at higher tempera-

![Graph](image-url)
Fig. 1.14 Rutherford and Brook's apparatus for estimating the atomic weight of radium emanation by measuring its rate of diffusion from ionization chamber A into chamber B, after removal of the dividing slide S, 1901.
(Trans. Roy. Soc. Canada, 1901)

Fig. 1.15 Photograph of apparatus in Fig. 1.14. (Courtesy of Rutherford Museum, McGill University)

The emanation, obtained by passing a slow current of air over heated radium, preserved its radioactivity and exciting power for weeks when kept in a closed metal container. The longer life of radium emanation, as compared with that of thorium, was clearly an advantage in studying its properties, such as its molecular (or atomic) weight. This was the subject of the next investigation, carried out with the help of a young graduate student, Harriet Brooks.

The amount of emanation available was far too small to enable its molecular weight to be measured by standard chemical techniques. Instead, Rutherford and Brooks used an indirect method, based on the fact that the rate of diffusion of gas in air was known to depend on its molecular weight. They passed air containing radium emanation into one half of a long cylindrical ionization chamber, divided into two airtight halves by means of a metal slide (Figs. 1.14 and 1.15). The ionization current was measured with an electrometer; when the current became steady, the input of emanation was stopped, the chamber sealed, and the dividing slide removed. The ionization in the other half chamber, as measured by a second electrometer, depended on the rate of diffusion of the emanation and hence on its molecular weight. The result obtained, between 40 and 100, was actually far too low—the experiment was subject to considerable error—but was sufficient to convince Rutherford that emanation was not simply radium in vapor form. As we have seen, Marie Curie had already shown the atomic weight of radium was at least 140, although her final value of 225 was still a year ahead. As it was, the result of the diffusion experiment was a powerful piece of evidence that radium, known to be an element, was producing a substance of different atomic weight and apparently different properties. This evidence supported the transmutation theory, which was beginning to take shape in Rutherford's mind.

In the subsequent history of radioactivity, emanation and excited radioactivity
continued to play an important role, especially in elucidating the sequence of changes that were later designated as the uranium-radium, thorium, and actinium radioactive series. Rutherford's group at McGill made important contributions to this field, both before and after Rutherford's own departure for Manchester in 1907. Other major North American contributions were made by William Duane of Harvard University and E. M. Wellisch of Yale.

In 1904 Harriet Brooks made a detailed study of the decay of excited radioactivity from thorium, radium, and actinium. This work began in Montreal and continued at the Cavendish Laboratory, although still under Rutherford's direction. She showed that in measuring the buildup and decay curves of excited radioactivity, it was essential to avoid the deposition, on the electrode whose induced activity was to be determined, of dust already rendered radioactive by the emanation. The following year, Duane, working at that time in the laboratory of the Curie's in Paris, made a quantitative investigation of the ionization arising from radium emanation. He concluded that the decay of a single molecule of emanation results in 14,500 ions of either sign in air.

From 1904 to 1908 Eve, working in Rutherford's laboratory at McGill, demonstrated that radium emanation is a normal constituent of atmospheric air, the average amount in 1 cubic meter of air in Montreal being that which would be in equilibrium with 60 x 10⁻¹² gram of radium. The actual amount at any given time varies according to weather conditions, with a ratio to maximum of minimum of about 7:1. At about the same time, G. Rümeli, a German physicist working temporarily in Rutherford's laboratory, made an accurate measurement of the half life of radium emanation; his result, 3.75 days, was slightly less than the values measured by previous workers, which ranged from 3.77 to 3.99 days. A further study of emanation, also in the McGill physics laboratory, was made by Howard Bronson, who concluded that the emanations of both thorium and actinium emitted multiple alpha particles on disintegration, four in the case of thorium, two in the case of actinium. However, he rejected the explanation, which later proved to be correct, of the presence of several short-lived decay products.

Finally, in the period under review, the work of E. M. Wellisch of Yale, between 1912 and 1914, must be mentioned. Wellisch studied the distribution of the active deposit of radium in an electric field in order to determine the mechanism of the production of excited radioactivity on the electrodes in an electric field or on exposed surfaces in the absence of such a field. He showed that a large fraction of the active deposit particles are positively charged, the remainder being neutral; the fraction is 80 to 90 percent, depending on the nature of the gas in which the emanation is mixed. In part of this work Wellisch collaborated with Bronson, who by 1912 had moved from McGill to Dalhousie College in Halifax, Nova Scotia, as professor of physics.

Rutherford and Soddy: The Transformation Theory

We now return to Rutherford at McGill University in the opening years of the twentieth century. He had shown that radium and thorium both gave rise to a gaseous emanation which, in turn, resulted in the deposition of radioactive particles on solid surfaces. Furthermore, the radiations emitted by the original substance (radium or thorium) and by the emanations and deposits had different penetrating powers, and their radioactivity decayed at different rates. In addition, the atomic weight of the radium emanation was significantly lower than that of radium itself. Further progress now depended on physically separating the original radioactive substance from both the emanation and the excited radioactivity and on proving that each component was chemically distinct. This task required the collaboration of a skilled chemist.

Fortunately one presented himself at the right time: He was Frederick Soddy, a
young demonstrator in chemistry at McGill. The Rutherford-Soddy collaboration lasted only eighteen months (October 1901 to March 1903) but was one of the most fruitful in the history of science. Soddy carried out the chemical procedures in the chemistry building, while Rutherford, and to some extent also Soddy, made the physical measurements in the physics building. Their procedure was to use standard chemical techniques, such as heat, solvents, precipitation, and filtration, to separate the elements in a given compound or mixture and to identify the presence and nature of a radioactive element by measuring the characteristics of the radiation emitted (i.e., penetrating power and the rate of change with time). The actual masses of radioactive elements were, of course, far too small to be measured on a balance or even spectroscopically.

Between January 1902 and May 1905 Rutherford and Soddy published nine joint papers—about 160 printed pages—including one on "The Cause and Nature of Radioactivity," which appeared in two parts in September and November 1902. For a detailed account of the collaboration, including the development and maturation of the transformation theory as new facts came to light, the reader is referred to The Self-Splitting Atom by Thaddeus Trenn. Further information may be found in the biography of Soddy by Muriel Howorth.

The important stages in the development of the transformation theory—the term transmutation was avoided because it was associated with medieval alchemy—were as follows:

1. The separation of a new radioactive substance, thorium X, from thorium and demonstration of its role as the "active constituent" of thorium;

2. Proof that the emanations behave as chemically inert gases, with fixed condensation points, and are produced by radium and thorium at a constant rate under all conditions;

3. Proof that radioactivity is due to the continuous and spontaneous production of new kinds of matter;

4. Indication (not yet proof) that helium is produced from radioactive substances; and

5. Evidence that the energy associated with radioactive matter is very large compared with ordinary chemical energy.

At first Rutherford and Soddy assumed that the emission of radiation and the transformation of the atom were separate processes, because it appeared that the only material particles emitted were the beta rays, with very small mass compared even with the lightest atom, hydrogen. The alpha rays were assumed to be nonmaterial, similar to soft X rays, because attempts to deviate the rays in a magnetic or electric field had been unsuccessful. However, by mid-1902 evidence was accumulating that alpha rays were actually heavy charged particles and that the emission of radiation and the transformation of the emitting atom occurred simultaneously. Indeed, the alpha particle proved to be central to the understanding both of radioactivity and the nature of the atom.

**The Alpha Particle**

The evidence that the alpha rays were heavy charged particles was persuasive but indirect, and before 1902 no one had demonstrated deviation of these rays in a magnetic or electric field, as would be expected for charged particles. Such a demonstration required a very strong field, a strong source of alpha rays, and a very sensitive detector. Toward the end of 1902 Rutherford was able to fulfill all three conditions, using a powerful electromagnet, a strong source of radium, and a gold-leaf electroscope, which was more sensitive than a quadrant electrometer. Using the apparatus shown in Fig. 1.16a, he showed that the reading of the electroscope was reduced to 11 percent of its initial value when a magnetic field was applied across the parallel brass plates, because the rays were deviated into the plates instead of passing into the electroscope. Furthermore, by half covering the exit slits between the plates (Fig. 1.16b) he proved that the particles were positively charged. Calculations based on this experiment indicated that
Frederick Soddy (1877-1956)

Frederick Soddy was born in 1877 in Eastbourne, England, the son of a London merchant. In 1895 he was awarded a science scholarship to Oxford, where he obtained a B.A. in 1898. He remained at Oxford for two more years as a research student in chemistry. In the spring of 1900 Soddy applied for the vacant professorship in chemistry at the University of Toronto. Without waiting even for an acknowledgment of his application, he set sail for Canada, only to find on his arrival in Toronto that the post had been filled. Returning to England via Montreal he visited McGill University, where he was offered, and accepted, a post as a demonstrator in chemistry.

Soddy's first encounter with Rutherford occurred in March 1901, when Soddy defended the classical concept of the atom in a famous debate at the McGill Physical Society on "The existence of particles smaller than the atom." Despite this, he worked with Rutherford in research that led in 1902 and 1903 to the transformation theory of radioactivity. The Rutherford-Soddy collaboration lasted only eighteen months but resulted in nine joint papers on various aspects of radioactivity.

In March 1903 Soddy returned to England, where he joined Sir William Ramsey at University College, London. In July of the same year Ramsey and Soddy spectroscopically confirmed the continuous production of helium by radium predicted a year earlier by Rutherford and Soddy.

In 1904 Soddy moved to the University of Glasgow, where he studied the properties of different members of the three natural radioactive series and, as early as 1910, became convinced that it was possible for elements of different atomic weight to have identical chemical properties, although he did not introduce the term isotopes until 1913. Meanwhile, in 1911, Soddy proposed the "displacement rule" for radioactive transformations involving the expulsion of an alpha particle.

In 1914 Soddy was appointed professor of chemistry at Aberdeen, and in 1919 he received the chair of chemistry at Oxford, where he remained until 1936, when he retired following the untimely death of his wife. During the latter years of his career, from 1919 onward, Soddy devoted much of his time to the impact of science on society, and he is acknowledged as one of the pioneers of this field.

Apart from the Nobel Prize in chemistry, which he was awarded in 1921 for his work on the origin and nature of isotopes, Soddy received many honors, including the Fellowship of the Royal Society and foreign membership in the Swedish, Italian, and Russian academies of science. He died in 1956.
the ratio of charge to mass of the particles was about half that of hydrogen, and it therefore appeared that alpha rays were rapidly moving particles of about twice the mass of hydrogen.

The experiment just described was an important step, but by no means the decisive one, in determining the identity of the alpha particle. In particular, what was the relationship, if any, between the alpha particle and the noble gas helium? In the summer of 1903 Ramsey and Soddy (who had left McGill for a post in University College, London, in March 1903) reported the presence of helium in gases obtained from radium bromide and the production of helium by radium emanation.105,106 Rutherford was now convinced of the identity of the alpha particle with the helium atom, but there was still no firm proof. A major (but not the final) step was taken in 1906, when Rutherford carried out the experiment illustrated in Fig. 1.17.107 The alpha particles from a line source passed through a thin mica window into an evacuated cylinder and hence between two parallel brass plates about 0.2 mm apart. An image of the exit slit between the plates was registered on a small photographic plate higher up in the cylinder. When a deflecting voltage was applied across the brass plate, and the polarity reversed halfway through the exposure, the image split into two, the separation depending on the applied voltage, on the geometry of the system, and, of
It was two years before Rutherford followed up this observation. In 1908 he suggested to Hans Geiger, a young German scientist in Manchester on a fellowship, that he carry out a full investigation of the scattering of alpha particles in foils of different metals. In part of this investigation Geiger was joined by a graduate student, Ernest Marsden. In the famous “gold-leaf experiment,” Geiger and Marsden found that a small percentage of the alphas were scattered by a thin gold foil through 90° or more, a phenomenon described as “diffuse reflection.”

Rutherford’s explanation of alpha scattering—the nuclear model of the atom—followed in 1911, although it was not until two years later that the model, as modified by Niels Bohr on the basis of Planck’s quantum theory, became widely accepted. Rutherford himself followed in 1914 with a further paper on the nuclear atom. The Rutherford-Bohr atom remains today the essential basis of the chemical atom, although of course the detailed structure of the nucleus has since been explored in much greater detail.

The Energy of Radioactive Transformations

The early 1900 and 1901 work of Rutherford and McClung in measuring the energy of roentgen and Becquerel rays, together with the energy required to produce an ion-pair in air, was discussed earlier in this chapter. In the same paper Rutherford and McClung used their value of the energy per ion-pair to deduce the energy radiated per second by 1 gram of uranium oxide. Assuming that radium was at least one-hundred thousand times more active than uranium, they concluded that radium emitted energy at an apparently constant rate (the slow decay of radium had not yet been detected) of at least 8,000 calories per gram per year. This value was actually far too low.

In 1903, soon after Soddy’s departure from McGill, Rutherford returned to this problem. Pierre Curie and André Laborde had just published a new value for the heat output of 1 gram of radium: about 100 calories per hour. 

40

1895–1914
In his new investigation Rutherford was assisted by Howard Barnes, an assistant professor of physics who specialized in precision calorimetry. They used a differential air calorimeter (Fig. 1.18) in which the expansion of air resulting from the heat emitted by a radium source in a glass flask (immersed in a constant-temperature water bath) could be compared with the heat generated in a wire of known resistance carrying a known electric current. The experiment was arranged so that the components of the heat emission could be measured separately by introducing into the flask either radium bromide, from which the emanation had been expelled, or pure emanation.  

The results indicated a reasonably good correlation between the rate of heat emission and the emission of alpha rays, assuming that the energy contributed by the beta and gamma rays was only a small fraction of that associated with the alphas. However, in response to criticism, Rutherford and Barnes later repeated the experiment in a modified form, with lead cylinders surrounding the radioactive source so as to absorb the beta and gamma rays (Fig. 1.19). The results confirmed their original assumption that most of the energy of radioactive transformations was carried off by the alpha rays. These experiments indicated that if it were possible to obtain 1 cubic cm of emanation (the actual amounts available were minute) the heat liberated per hour would lie between $1.25 \times 10^5$ and $1.25 \times 10^6$ gram-calories, an amount "probably
Bertram Borden Boltwood (1870-1927)

Bertram Boltwood was born in 1870 in Amherst, Massachusetts. His parents were of British and Dutch descent. His father died in 1872, and the young boy was brought up by his mother in a village in New York.

After graduating in chemistry at Yale in 1892, Boltwood spent two years at the University of Munich, where he learned analytical techniques, with special reference to the rare earths. He returned to Yale in 1894 as a laboratory assistant/graduate student in analytical chemistry, obtained his Ph.D. in 1897, and remained at Yale as an instructor in analytical and physical chemistry until 1900, when he left to set up a private consulting laboratory.

Boltwood's partner in the consulting enterprise was Joseph Pratt, a mining engineer who was also a Yale graduate. Pratt sent Boltwood samples of ores, mainly from the Carolinas, which the latter analyzed in the New Haven laboratory. Many of the ores contained rare earths, uranium, and thorium. It was natural, therefore, for Boltwood to become interested in radioactivity following Becquerel's discovery in 1896, although he did not begin his own research in the field until 1904.

Boltwood's first investigation in the field of radioactivity concerned the relationship between uranium and radium. In 1903 Rutherford and Soddy put forward the transformation theory of radioactivity and suggested that radium might be a transformation product of uranium. Using the radioactivity of radium emanation (radon) as a measure of the amount of radium present in a sample of uranium mineral, Boltwood was able to show that the amounts of radium and uranium in different minerals were always in the same proportion. However, he was unable to demonstrate the growth of radium in a purified sample of uranium, even over a period as long as a year. Clearly, there was an intermediate product, and Boltwood set out to isolate this substance. By 1907 he had identified the immediate parent of radium as "ionium," later shown to be an isotope of thorium.

Another of Boltwood's achievements in radioactivity was the demonstration that the amount of lead in minerals of a given geological age is proportional to their uranium content and increases with geological age. He also investigated the rate of production of helium by radium (1910) and made the first accurate measurement of the half life of radium (1913-1914).

Boltwood returned to Yale in 1906 as assistant professor of physics and in 1910 became professor of radiochemistry; he played an active part in building new chemistry laboratories at Yale. He collaborated with Rutherford for many years by correspondence and in person in Manchester in 1909 and 1910. The Rutherford-Boltwood correspondence was published in 1969 by Badash, L., ed., Rutherford and Boltwood. Letters on Radioactivity, Yale University Press.

In 1922 Boltwood suffered a nervous breakdown from which he never fully recovered, and he committed suicide in August 1927. In an obituary published in Nature on 14 January 1928, Rutherford wrote that "[Boltwood's] premature death will be mourned by a wide circle of friends, who held him in high esteem for his personal qualities as well as his outstanding scientific achievements."
sufficient to raise to a red heat, if not melt down, the glass containing it [i.e., the 1 cubic centimeter of emanation]." Rutherford and Barnes concluded: "There is thus no doubt that matter under special conditions is capable of emitting an amount of energy enormous compared with that released in the most intense chemical reactions."\textsuperscript{172}

The work of Rutherford and Barnes just described, together with that of their European contemporaries, had some unexpected repercussions. At the time the age of the earth was highly controversial. On the basis of the known temperature gradient in the earth's crust, and on the assumption that the earth had originally been a molten sphere, Lord Kelvin had calculated that the age of the earth was, at most, one-hundred million years, a duration totally insufficient to account for geological and biological evolution. But, it was known that everything—earth, rocks, water, air—contained one or two parts per million of radioactive material. Rutherford showed that the heat supplied by this tiny amount of radioactivity, if uniformly distributed in the form of uranium, with a half life of hundreds of millions of years, was sufficient to extend the origin of the earth back several billion years. Rutherford argued this case in a popular article in \textit{Harper's Magazine}\.\textsuperscript{173}

\textbf{THE RADIOACTIVE SERIES}

As mentioned earlier, Soddy left McGill for London in 1905, but Rutherford remained in Montreal until 1907. These four years were a period of important consolidation: The nature of radioactivity was further explored with reference to the identity of the alpha rays, the energy associated with radioactive transformation, and the radioactive series. The first two of those topics have already been discussed; the third—the elucidation of the uranium-radium series—was in many ways the most difficult problem of all, because its solution depended on an analysis of the relationship between uranium and radium, and between radium and lead in naturally occurring minerals, along with detailed studies of the radiations emitted at each stage of the transformation process. The alpha particle was an essential link in the identification of the stages in the series, especially the mass of the particle. As we have seen, it was not until 1900 that the identities of the alpha particle and the helium atom were finally proven.\textsuperscript{174}

In May 1904, in the Bakerian lecture given at the Royal Society in London, Rutherford reviewed the evidence for four radioactive series, those of uranium, radium, thorium, and actinium, and derived the mathematical theory of successive changes.\textsuperscript{175} At this stage the link between uranium and radium was unproven, although Rutherford discussed the evidence that uranium was the parent of radium and that radium could also be derived from thorium and actinium. (The concept of isotopes had not yet been developed.) It was not until 1908 that Rutherford was able to define, with some certainty, the link between uranium and radium.\textsuperscript{176}

By September 1905 Rutherford had extended the radium series, via emanation, from radium A, B, C, and D (now considered the primary constituent of radio-lead) to radium E and, finally, the active constituent in radio-thelrium and polonium.\textsuperscript{177} He suggested, but could not prove, that the transformation production of radium F was lead. A similar suggestion had been made by Bertram Boltwood, on the basis of a study of twenty-two minerals containing uranium.\textsuperscript{178} J. C. McLennan, a professor of physics at the University of Toronto, also contributed to this study by measuring the radioactivity of lead and other metals.\textsuperscript{179}

\textbf{OTHER ASPECTS OF RADIOACTIVITY}

As we have indicated, the most important studies in radioactivity in the period under review related to the nature of the radioactive process and of the radiations emitted during that process. But many other studies of different aspects of radioactivity were made, both in Rutherford's group in Montreal and elsewhere in North America. Rutherford's colleague (and subsequent biographer) at McGill,
Arthur Eve, made an extensive study of the properties of the beta and gamma rays of radium.\textsuperscript{180,181,182,183,184} Other members of the physics department at McGill who made significant contributions to the study of radioactivity after Rutherford's departure were Louis King and J. A. Gray.\textsuperscript{185,186}

CONCLUSION

The period from 1895 to 1914 was the most important in the history of radiological physics. This statement may seem an exaggeration for a time that lacked most of the modalities taken for granted today, including image intensifiers, digital radiography, computed tomography, magnetic resonance imaging, ultrasound, artificial radionuclides, and high-energy accelerators. Even the terms “radiological physics” and “medical physics” were unknown; physicists and engineers investigated the phenomena of X rays and radioactivity, and developed new techniques for generating and measuring penetrating radiation, independent of their medical colleagues.

In spite of these deficiencies, the importance of this period of radiological physics cannot be overstated. At the beginning of 1895 neither X rays nor radioactivity was known or even envisaged, and both physics and chemistry were mired in the self-satisfied classical era of immutable and indivisible atoms. By the end of 1914 X rays had become an indispensable diagnostic tool in medicine, and their basic properties—including absorption, scattering, polarization, and diffraction—were known, although the mechanisms and laws of these properties remained to be discovered. Radioactivity had been intensively investigated, several new radioactive elements (including radium) had been discovered, the nature and laws of radioactive decay had been elucidated, and a start had been made in investigating the biological effects of radiation. This period also saw the beginning of radiation therapy (especially using radium), of radiation protection, and—with the first tentative definitions of a unit of radiation quantity—of radiation dosimetry.

Perhaps most important, modern science took firm root in this period. Apart from the discoveries of X rays and radioactivity, such advances as Planck's quantum theory, Einstein's theory of relativity and the equivalence of matter and energy, Thomson's discovery of the electron, Bragg's explanation of X-ray diffraction, and the Rutherford-Bohr nuclear atom all date from this period. Strictly speaking, none of these advances was in the field of radiological physics, yet in a broader sense all of them were in this field, because they provided the foundation on which the modern edifices of radiation diagnosis, therapy, and investigation were built.

Although this volume is intended to highlight the contributions to radiological physics by scientists and others who worked in North America, any account of the beginnings of radiological science must discuss the work of Röntgen and von Laue in Germany, Becquerel and the Curies in France, and Thomson, Barkla, and the Braggs in Britain. What does this leave for North America? In basic radiation physics, excluding radioactivity, very little, apart from Millikan's measurements of the electronic charge from 1910 to 1913; the work of Duane, Hunt, and Compton was still to come.

In applied physics and technology, however, the American contribution in the years prior to World War I was important both to the development of the X-ray tube and its power supply and to the means of detecting and recording the radiation by photographic plates and fluorescent screens. The important names here were Edison, Tesla, Elihu Thomson, Pupin, and above all, Coolidge. Indeed, the invention of the hot-cathode X-ray tube was crucial to the further progress of both radiation physics and practical radiology: Here, for the first time, was a means of independently setting and controlling the quantum and quality of the radiation emitted by the tube. Coolidge described his invention in December 1913, toward the end of the period reviewed in this chapter, but it was years before the new source finally displaced the gas X-ray tube.
As for the North American contribution to the development of radioactivity in the early years, the outstanding group was that of Rutherford at McGill University in Montreal, rivaled only by the Curies in Paris. But the contributions of the two centers were different, with the Curies working mainly on the chemical aspects of radioactivity, such as the separation of radium and other radioactive elements, while Rutherford and his group, especially Soddy, investigated the basic nature and laws of radioactivity. Rutherford’s achievements in Montreal included the exponential law of radioactive decay, the transmutation theory of radioactivity, the relationship between radium emanation (radon) and excited radioactivity, the nature of the alpha particle, the energy involved in a radioactive transformation, and the elucidation of the uranium-radium radioactive series. The foundations of radiological science and practice laid down in the first twenty years after the discovery of X rays were solid and have withstood the test of time. The next twenty years, to be described in the following chapter, were a period of both consolidation and further advance.

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Strictly speaking, we should refer to phosphorescence rather than fluorescence. The difference is that phosphorescent light persists, sometimes for hours, after the exciting radiation has ceased, whereas fluorescent light is emitted only while the exciting radiation is present. In the case of potassium uranyl sulfate, however, the phosphorescence remains only for a fraction of a second after the sunlight is removed, and for practical purposes we are dealing with fluorescence.


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