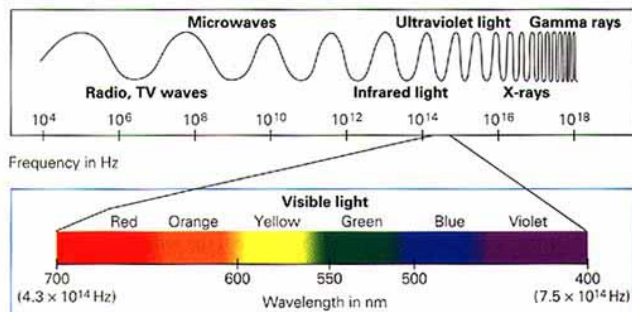# The History of the Discovery of Radiation and Radioactivity

Introduction:

Radiation can be defined as the propagation of energy through matter or space. It can be in the form of electromagnetic waves or energetic particles.

Ionizing radiation has the ability to knock an electron from an atom, i.e. to ionize. Examples of ionizing radiation include:

- alpha particles
- beta particles
- neutrons
- gamma rays
- x-rays

Non-ionizing radiation does not have enough energy to ionize atoms in the material it interacts with. Examples of non-ionizing radiation include:

- microwaves
- visible light
- radio waves
- TV waves
- Ultraviolet radiation (except for the very shortest wavelengths)

The earth has been radioactive ever since its formation into a solid mass over 4½ billion years ago. However, we have only known about radiation and radioactivity for just over one hundred years. [One hundred and four years and 10 months to be exact.]

You are probably familiar with the radiation warning sign, the "trefoil". See http://www.orau.com/ptp/articlesstories/radwarnsymbstory.htm for a history of the symbol.
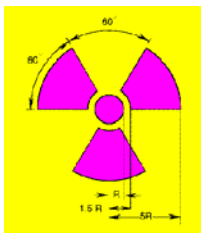
A Brief History of the Discovery of Radiation and Radioactivity
The History of the Discovery of Radiation and Radioactivity
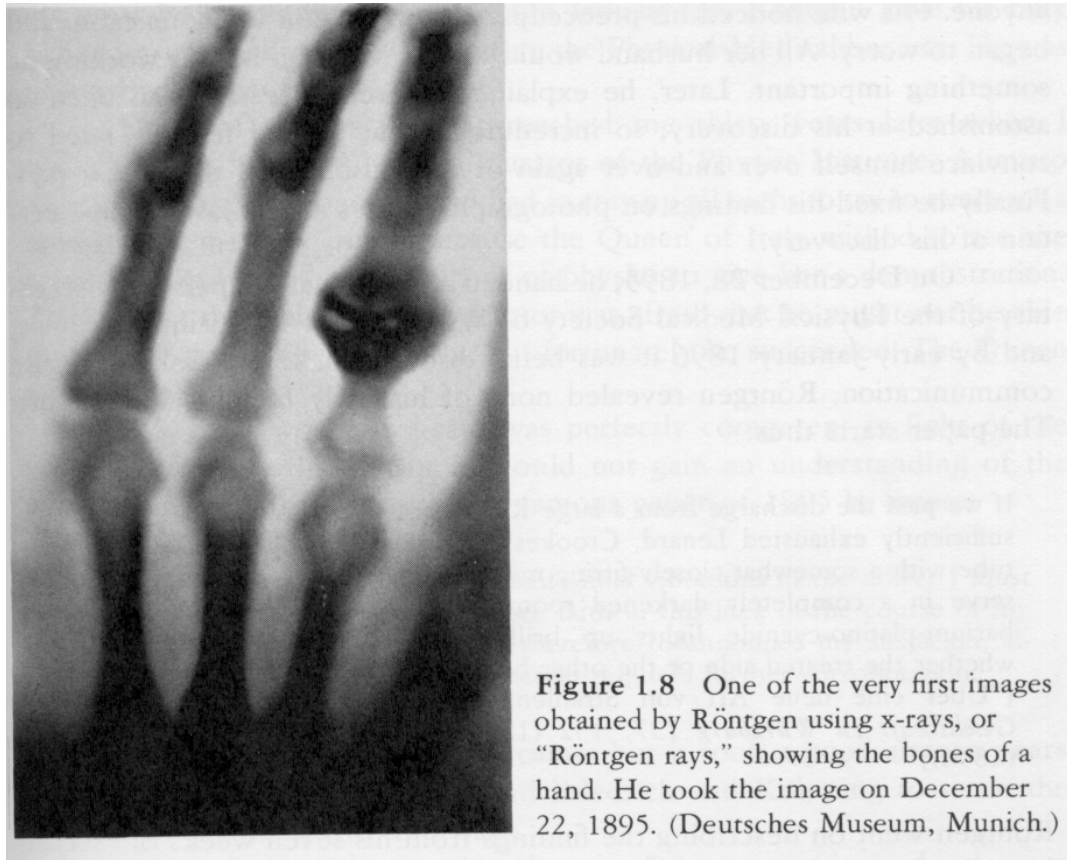
1895: The Discovery of the X-ray

Radiation was discovered by Wilhelm Conrad Roentgen on November 8, 1895. http://www.orcbs.msu.edu/radiation/radhistory/wilhelmrontgen.html Roentgen, like many other physicists at that time, had been experimenting in his laboratory with the discharge of electricity in "vacuum tubes". The glass tubes were evacuated (and made airtight using Bank of England sealing wax) and had metal plates sealed at the ends. The metal plates could be connected to a battery or an induction coil. This flow of electricity was necessary in order for the tube to glow. The glow emerged from the negative plate (the cathode) and disappeared into the positive plate (the anode). If a circular anode was sealed into the middle of the tube, the glow (the cathode rays) could be projected through the circle and into the other end of the tube. If the beam of cathode rays were energetic enough to hit the glass, the glass would glow (fluoresce).
The glass tubes were given various names, e.g. Crookes tubes (named after William Crookes) or Hittorf tubes (Johann Hittorf) based on the individual able to design a tube better able to generate or hold vacuum. Roentgen's work was carried out using a Hittorf tube. [See www.chem.uiuc.edu/demos/cathode.html for a demo of an early cathode ray tube in operation.]
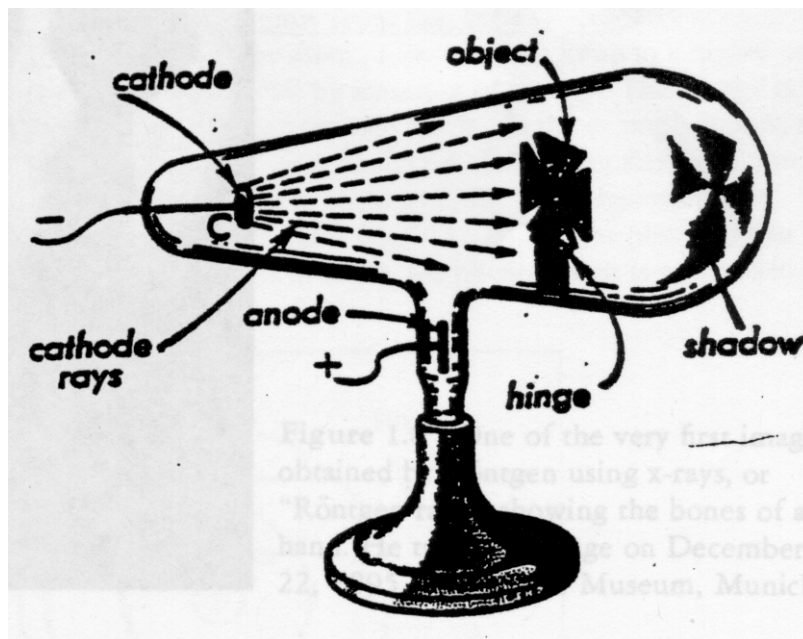
It had already been demonstrated that the emission coming from the cathode, the "cathode rays", were not very penetrating. On the evening of November 8, 1895, Roentgen had covered the tube completely with black cardboard. The laboratory was completely dark. Several feet away from the tube was a piece of paper, used as a screen, covered with barium-platinum cyanide. In the darkened room, Roentgen noticed the screen fluorescing, emitting light. Something must have hit the screen if it reacted by

emitting light. However, since the Hittorf tube was covered in cardboard, no cathode rays or light could have come from Hittorf tube.

Very surprised, Roentgen began investigating this strange occurrence. He turned the paper screen so that the side without barium platinum-cyanide faced the tube – still the screen fluoresced. He moved the screen farther from the tube and still the screen fluoresced. Then he placed several objects between the tube and the screen but all appeared to be transparent. When he put his hand in front of the tube he saw his bones on the screen.

Figure 1.8 One of the very first images obtained by Röntgen using x-rays, or "Röntgen rays," showing the bones of a hand. He took the image on December 22, 1895. (Deutsches Museum, Munich.)

Roentgen experimented with these "new rays", alone in his laboratory, for many weeks. He found that objects were transparent to these rays in different degrees. Photographic plates were sensitive to x-rays. He could not detect any appreciable reflection or refraction of the rays, nor could he deflect them with a magnetic field. The rays originated in the area of the discharge tube where the cathode rays impinge on the wall of the glass tube.

On December 28, 1895, Roentgen delivered a preliminary paper to the secretary of the Physical-Medical Society of Würzburg. It was printed immediately and by early January of 1896 it was being distributed. The paper caused a great stir (as can be imagined) and may have been ignored as unbelievable had he not also included x-ray photographs of hands (taking 3-10 minutes to acquire). This evidence could not be easily dismissed.

The response to this new discovery was tremendous. Roentgen was invited to lecture all over the world, however he declined all invitations (except one – he made a demonstration to the Kaiser on January 13, 1896) in order to continue his investigations.

In the year 1896 there were over 1,000 papers written on the subject of x-rays. Roentgen himself wrote only two more papers on x-rays, in 1896 and 1897. He then went back to working on his former interests but received the first Nobel Prize for physics in 1901.

What Happens in the Crookes/Hittorf Tube?

1. Phosphorescence/ Fluorescence: The glow from the tube
Electrons exist in discrete orbits in the atom. Electrons can jump to a higher orbit only by absorption of energy or to a lower orbit by emission of energy. The energy is equal to the difference in energy level between the two orbits. In the example below, an electron in the outer shell (E_i) drops into the inner shell (E_f). In making this transition, a photon is given off. The energy of the photon is equal to E_i-E_f. If the emission of a photon (following energy absorption) is very quick (eg 10^{-8} - 10^{-9} s), the phenomenon is called "fluorescence". If the emission is slow to occur, the phenomenon is referred to as "phosphorescence".



A jumping electron emits a photon
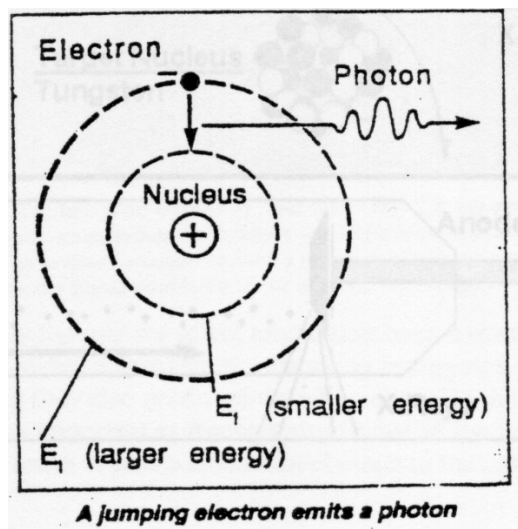
Since the energy levels of each atom are unique to that atom, the photons emitted during fluorescence are very distinctive and act as 'fingerprints' for that particular element. This fact is often used in the identification and quantification of elements in a sample. Because the fluorescent (or phosphorescent) photons are unique to the atom, these photons are sometimes referred to as "characteristic radiation".

2. Bremsstrahlung (X-rays): The Mysterious Rays:

What Roentgen discovered was the phenomenon we now know as "bremsstrahlung". He himself didn't know what the rays were – they were new, penetrating and mysterious. For lack of a better name he called them "x-rays".

The cathode rays generated in the Hittorf tube, the electrons, struck the glass wall of the tube. When electrons interact with matter they typically do one of two things. They (1) dislodge orbital electrons in the material (via elastic collisions); these electrons make an ionization track themselves in the material; eventually the energy is given to the material as heat. Orbital vacancies are filled from electrons dropping in from higher shells and characteristic radiation (fluorescence) may be given off.

Or, they (2) are decelerated in the field of the nuclei in the glass tube. Remember that the electrons are negatively charged and will be attracted to the positively-charged nuclei. The trajectory of the electron will change, bending toward the nucleus and slowing down. Classical theory tells us that when any charged particle undergoes a change in acceleration, electromagnetic radiation (an x-ray) is emitted. This phenomenon is called bremsstrahlung, which is German for 'braking radiation', and is the phenomenon observed by roentgen. [This process, which is the basis for all x-ray tubes in use today, will be detail with in more detail later in the course.]
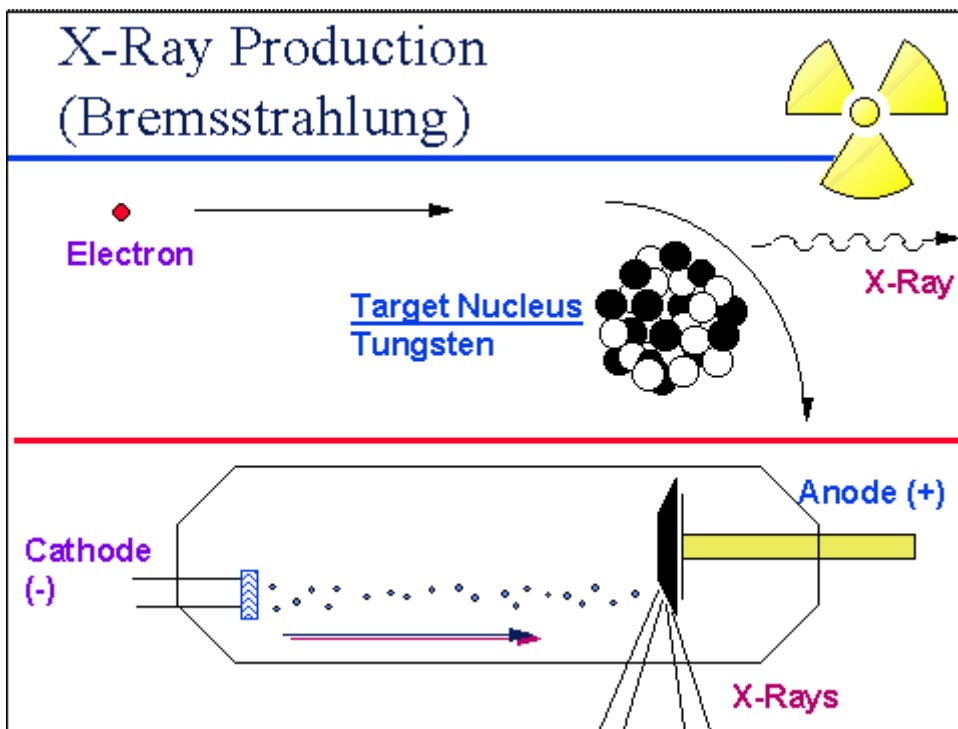
1896: The Discovery of Radioactivity

The discovery of radioactivity followed the discovery of x-rays by only two-and-a-half months. Henri Becquerel, a third generation expert on fluorescence and phosphorescence, heard a report (and saw radiographs) of Roentgen's work at the weekly meeting of the Académie des Sciences on January 20, 1896.
[http://www.orcbs.msu.edu/radiation/radhistory/antoinebecquerel.html] Upon learning that the x-rays emerged from the fluorescing glass of the Hittorf tube, he immediately conjectured that other fluorescing materials might also emit x-rays. He worked for many days, testing a number of phosphorescent and fluorescent substances without success, until he tried a uranium salt, uranyl potassium sulfate.

Sealing a photographic plate in black paper, he sprinkled a layer of the uranium salt onto the paper and put the whole thing in the sun for several hours. When he later developed the plate, he saw the silhouette of the phosphorescent substance in black on the negative. He then assumed (mistakenly) that the sunlight *caused* the emission of the penetrating rays (and that x-rays came automatically from fluorescing substances).

The next part of the story is famous. Becquerel tried repeating the experiment on February 26 and again on February 27, however Paris was cloudy. He put the covered plate, with uranyl potassium sulfate on top, away in a drawer. Then, on March 1 he went ahead and developed the plate, expecting to see very faint images. Instead, the images appeared very intense. Thus, the uranium salt emitted rays capable of penetrating black paper, whether or not it had been previously exposed to sunlight.
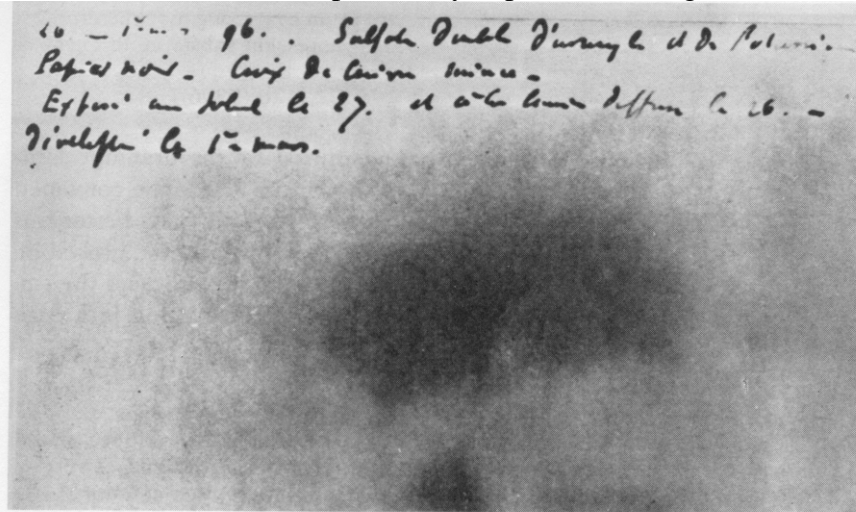

Figure 2.2 The first plate marked by "Becquerel rays." It was placed under some uranyl potassium sulfate on February 26, 1896; after Becquerel developed it on March 1, he realized uranium emitted a radiation of unknown nature that did not depend on the phosphorescence of the uranium salt, as had been believed. The discovery of "Becquerel rays" was published in the *Comptes-rendus de l'Académie des Sciences de Paris* [*122*, 501 (1896)]. (CEA.)

Becquerel initially thought the effect might have been a result of long-lived invisible phosphorescence, but disproved this theory by testing non-phosphorescent uranium salts and finding they also produced the silhouettes. At the time, Becquerel's discovery did not seem as important as Roentgen's. (After all the images were very fuzzy and there was not much uranium around, in contrast to the comparatively high

resolution images made with "Roentgen Rays" using a Crooks or Hittorf tube already in the hands of many, many investigators.)

Becquerel found that the "Becquerel Rays" ionized gases. Thus it was possible to measure the activity in a sample simply by measuring the ionization that it produced. At the time this could be done using a crude gold-leaf electroscope. Becquerel shared the 1903 Nobel Prize in physics and is known as the discoverer of radioactivity.

Becquerel continued to investigate the rays emitted by uranium. However, he restricted himself to uranium since, first, it was the material he had most experience with and, second, he felt it to be improbable that another substance would be a more powerful emitter since the rays were first discovered from uranium. Thus it was not Becquerel but the Curies who investigated other elements and, by discovering first polonium and then radium, were able to make very powerful sources which helped pave the way toward a full understanding of the process of radioactive decay.

1897: Elucidating the Nature of Cathode Rays

In 1897, the third director of the prestigious Cavendish Laboratory at Cambridge University, J.J. Thomson, carried out a number of experiments with cathode rays. [See http://www.orcbs.msu.edu/radiation/radhistory/josephthomson.html for a brief history of J.J. Thompson, and http://webster.aip.org/history/electron/ for an interesting description of his experiments.] By virtue of the fact that he was able to establish a good vacuum in the cathode tube, Thomson was able to show that the cathode rays could be deflected in an electric field. [He had connected two metal plates placed inside the tube to a battery. He showed that a narrow cathode ray beam, made using a pair of separated slits, created a fluorescent patch on the end of the tube in a different place, depending on whether the battery was connected or not.] Although it was known that cathode rays could be deflected in a magnetic field, no one had yet observed their deflection in an electric field. The reason was that J.J. Thomson was able to obtain a better vacuum in his tube. Unless there is a good vacuum, no electric field can be established. [A poor vacuum is a good conductor.]
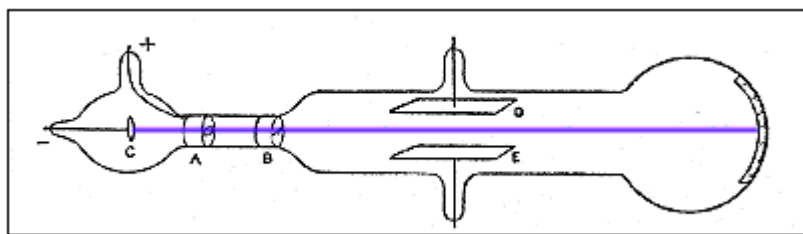
Schematic drawing of Thomson's apparatus in the second experiment. Rays from the cathode (C) pass through a slit in the anode (A) and through a slit in a grounded metal plug (B). An electrical voltage is established between aluminum plates (D and E), and a scale pasted on the outside of the end of the tube measures the deflection of the rays.

In 1897 Thomson described his experiments carried out to determine the ratio of charge to mass of the particles making up the cathode rays. Thomson is credited with the determination that cathode rays ("electrons") are particulates. He also found that the

corpuscles that constituted the cathode rays were the same no matter what the composition of the cathode or the gas in the tube. This, and the fact that the electrons were lighter than hydrogen, the lightest known kind of matter, implied that the atom was not indivisible.

J.J. Thomson won the Nobel Prize in physics for demonstrating the particulate nature of the electron. Interestingly, in 1937, his son George Thomson won the Nobel Prize in physics for demonstrating the wave properties of the electron.

1897: The Discoveries of Marie Sklodowska Curie and Pierre Curie

In September of 1897, just after the birth of her first daughter, Marie Curie, having already passed what we would call her doctoral qualifying exam, sought her husband's advice regarding the subject of her Ph.D. thesis. Pierre suggested she investigate the new phenomenon discovered by Becquerel.
[http://www.orcbs.msu.edu/radiation/radhistory/pierremariecurie.html]

She began by repeating Becquerel's experiments using better electrometers (containing a piezoelectric quartz – Pierre and his brother Jacques had discussed piezoelectricity). She confirmed that the extent of ionization (in air) created by a uranium sample and thus the intensity of its radiation emission, was proportional to the amount of uranium in the compound and independent of its chemical state. She then decided to examine all the elements then known and found that only thorium emitted rays similar to uranium (a fact discovered earlier by G.C. Schmidt). Marie Curie then proposed the word "radioactivity" to describe the phenomenon.

Her next step was to investigate natural ores rather than limit herself to simple compounds of uranium and thorium. To her surprise, the radioactivity of some of the ores were three to four times greater than could be accounted for on the basis of just their uranium and thorium content. Her conclusion was correct. The samples of ore must contain something even more radioactive than u or Th.

She began her search for the new substance which, by a clear experiment, she had determined was some type of impurity in the uranium ore. [That is, when she fabricated the mineral herself out of pure substances, the mineral was no more radioactive than what was expected based on the uranium content.] To find the new substance she had to take the natural ore, dissolve it, separate it into its components and then measure the radioactivity of each component using her electrometer.

The task was immense and she recruited help from her husband in the process of grinding sample after sample of pitchblende ore. They eventually concentrated the highly radioactive part of the sample in a small residue. They decided they had found a new element since, although their sample was far from pure, it was far more radioactive than either U or Th. They called it polonium (after Poland, Marie's native country). They also found that the substance disappeared spontaneously reducing itself to one half in a characteristic time (called the half-life).

During their chemical analysis they had found polonium in the group of sulphides, insoluble in acid solutions. But then they also found radioactivity in the barium group (barium, strontium and calcium). When they finally succeeded in separating the radioactivity from barium they found a new substance which they called

radium. Obtaining a hundred kilograms of residues from the pitchblende mines of Joachimstal, Czechoslovakia (practically the only active center of uranium mining at the time) from which the uranium had been already removed. Marie and Pierre ground it all, by hand, dissolved it in various solvents, heating and stirring it in a laboratory with no fume hoods. After two years they had enough radium in their sample to detect a change in the atomic weight of the substance. [Barium has an atomic weight of 137 but their samples showed an atomic weight of 174. We now know that pure radium has an atomic weight of 226. It took Marie 12 more years to produce a pure sample.]

By 1902 Marie had lost about 20 pounds as a result of the heavy labor involved in the extraction of radium from pitchblende ores. But, she had enough radium to enable her to measure its atomic weight. She found it to be 225, almost the correct value.

In 1903, the Curies shared the Nobel Prize in Physics with Henri Becquerel. In 1911 Marie Curie received a second Nobel Prize, this time in chemistry, for her purification of Ra metal. She is the first person ever to have won the prize twice.

Pierre died in a carriage accident in 1906 but Marie lived to the age of 67. She had been ill a long time and ultimately died of aplastic anemia. [This is a condition that can be caused by overexposure to radiation] If you look at the periodic table you'll notice that radium and calcium are both in Group I. They thus share similar chemistry. Calcium, as you know, is a bone-seeker too. Aplastic anemia is a disease of the blood caused by defective function of the blood-forming organs (i.e. the bone marrow). Although there is no way to know for sure that Marie Curie died of a radiation-induced aplastic anemia, it is, of course, quite likely. After her death, it was found that her laboratory notebooks were strongly contaminated. Even her cookbooks, at home, were radioactive fifty years after her death.

1899/1900 Alpha, Beta and Gamma

Ernest Rutherford, http://www.orcbs.msu.edu/radiation/radhistory/ernestrutherford.html, a physicist from New Zealand, was working under J.J. Thompson at the Cavendish Laboratory in 1898-1899. Through a long series of experiments he realized that there were two kinds of radiation emitted from Uranium. He called them *alpha* and *beta*, the first two letters in the Greek alphabet. In a few years it was concluded that beta rays were cathode rays, that is, electrons. The precise nature of alpha particles remained a mystery although both Rutherford and the Curies suspected they were particles, atoms electrically charged and projected at high speed. They also knew they could be stopped by extremely thin shields (e.g. paper) and were deflected to only a small extent in a magnetic field.

In 1900, P. Villard in France had also found an additional penetrating radiation from Uranium. This radiation was not alpha nor beta, but more penetrating than the beta particles; it was given the name *gamma*. [It was the effects of gamma radiation that Becquerel had observed when he developed his photographic plates. The gamma rays are penetrating enough to both make it out of the solid uranium salts, and through the paper.]

The figure below, taken from the Ph.D. thesis of Marie Curie, shows the behavior of the α, β, and γ rays in a magnetic field. Over the next few years many experiments were carried out to determine the exact nature and behavior of these rays.
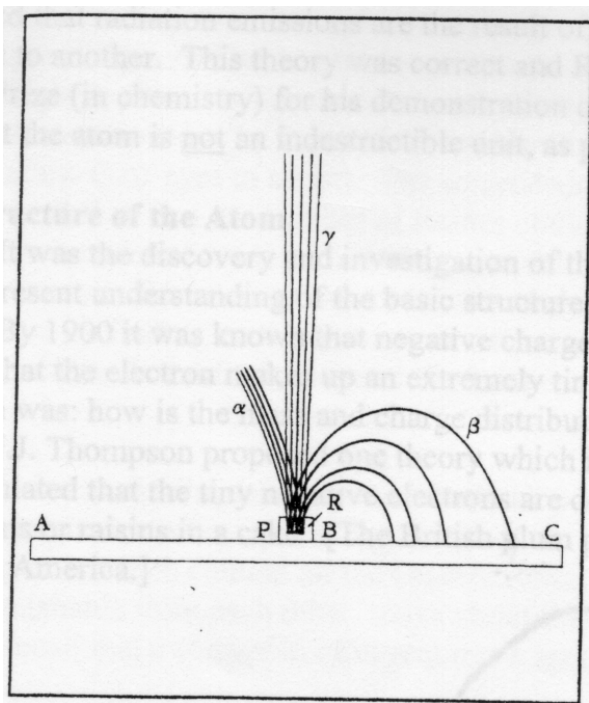
Figure 3.3 The three kinds of rays: α, β, and γ. They are distinguished by their trajectories in a magnetic field at right angles to the direction of motion. The α rays (helium nuclei) are positively charged; β rays (electrons) are much lighter and negatively charged; γ rays, analogous to x-rays, are quanta of electromagnetic radiation (photons). As they are neutral, they are not deflected by the magnetic field. Rutherford was responsible for the nomenclature. [From Marie Curie, *Thesis* (Paris: Gauthier-Villars, 1904).]

What is this "radioactivity"?

The first use of the word "radioactivity" appeared in an 1899 publication by the Curies. But the exact nature of radioactivity stumped everyone. The emissions did not seem to be affected by temperature or state. That is, the penetrating Becquerel rays were observed whether the Uranium was in a solid lump or in a heated solution. The emissions could change the color of glass, they could burn skin, and they could create heat. Eventually it was noted that the emissions were not restricted to Uranium, Radium or Polonium. They were found, to some extent in rain, soil, and air. To make it more confusing, some elements showed the same rate of emission (i.e. "activity") while others appeared to decrease according to a specific half-life. [We now know that all radioactive elements decay with a specific half-life. Those with extremely long half-lives only appeared to be decaying at a constant rate.]

Data to consider were supplied by both physicists and chemists. Crookes discovered in 1900 that if iron hydroxide was precipitated in a uranyl salt solution, all the radioactivity went into the precipitate, and the uranium remained inactive. However, after a few days the precipitate lost its activity, and the uranium reacquired it. Similar phenomena occurred in other radioactive substances on different time scales – sometimes hours, sometimes minutes.

There were many theories put forward to explain the phenomenon of radiation. Becquerel at first proposed that radioactivity was a form of delayed phosphorescence. This was proved incorrect since some things that did not exhibit phosphorescence were shown to be radioactive. Crookes, the inventor of the Crookes tube, thought that energy is extracted from fast moving air molecules. This was disproved by conducting experiments in a vacuum. Marie Curie proposed that all space is constantly traversed by X-rays which are only absorbed by heavy elements. This theory was tested (and disproved) in a mineshaft. Rutherford and Soddy (a chemist at McGill with Rutherford)

proposed that radiation emissions are the result of the spontaneous transmutation of one element to another. This theory was correct and Rutherford eventually won the 1908 Nobel Prize (in chemistry) for his demonstration of this. However, this theory basically said that the atom is <u>not</u> an indestructible unit, as previously believed!
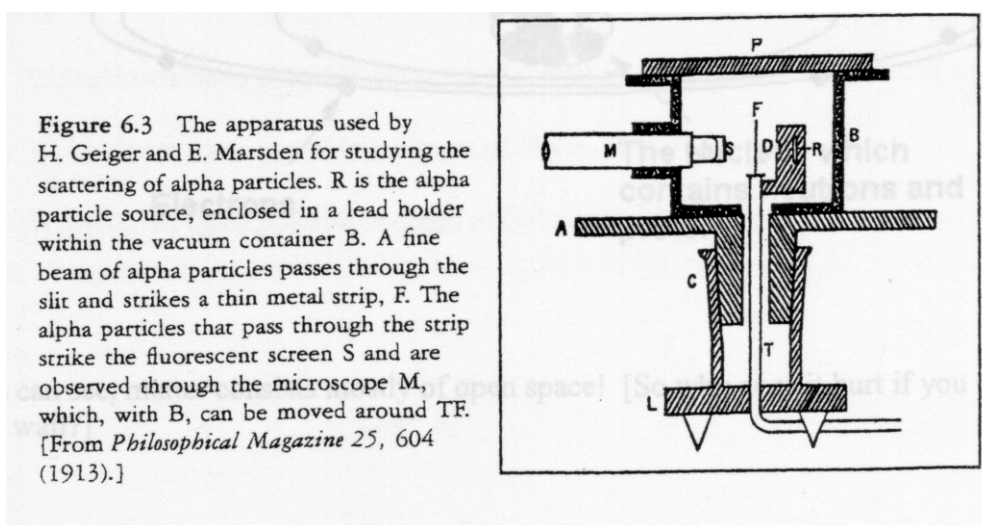
The Structure of the Atom
It was the discovery and investigation of the phenomenon of radioactivity that led to our present understanding of the basic structure of the atom.
By 1900 it was known that negative charge is carried by the electron. It was also known that the electron makes up an extremely tiny fraction of the mass of the atom. The question was: how is the mass and charge distributed in an atom?
J.J. Thompson proposed one theory which is called the "plum pudding model". He postulated that the tiny negative electrons are dotted about the very massive nucleus like plums or raisins in a cake. [The British plum pudding would be referred to as a cake in North America.]
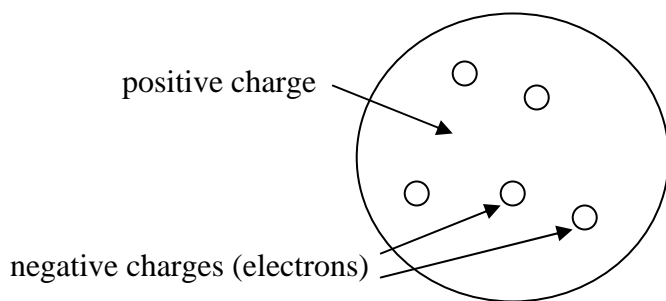
Rutherford, however, carried out a famous scattering experiment which prove that the atom was <u>not</u> structured like a plum pudding. The experiment involved a radiation source) (e.g. ^{214}Po), a thin foil of material, a zinc sulphide screen, a microscope and many hours of sitting in the dark. The apparatus and set-up is shown below.

Figure 6.3 The apparatus used by H. Geiger and E. Marsden for studying the scattering of alpha particles. R is the alpha particle source, enclosed in a lead holder within the vacuum container B. A fine beam of alpha particles passes through the slit and strikes a thin metal strip, F. The alpha particles that pass through the strip strike the fluorescent screen S and are observed through the microscope M, which, with B, can be moved around TF. [From *Philosophical Magazine* 25, 604 (1913).]

The idea was to allow alpha particles to strike the thin foil and then to look at their angle of scatter. The screen, coupled with the microscope, is the detector. A tiny flash of light appeared when an alpha particle hit the screen; this flash was observed through the microscope by Rutherford and his students (following hours of sitting in the dark to allow their eyes to adapt). The screen/microscope combination could move around the foil so that the number of flashes could be logged as a function of angle.

The results of the experiment were as follows. Most alpha particles went straight through the foil. Some scattered at various angles. But only one in 8000 scatter backwards! [a finding certainly inconsistent with the plum pudding model!] On the basis of these results, Rutherford proposed the "planetary model" of the nucleus.

The Atom

In Rutherford's planetary model, all the positive charge and almost all of the mass are concentrated in a tiny spot in the center of the atom. Surrounding the nucleus are the light electrons which contain all the negative charge. The electrons, which are indistinguishable from each other, move about throughout the atomic volume in specific orbits. Recall that a change in orbit requires a specific change in energy.
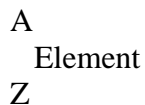
Atomic radius: 10^{-8} cm

Nuclear radius: ~5 x 10^{-13} cm
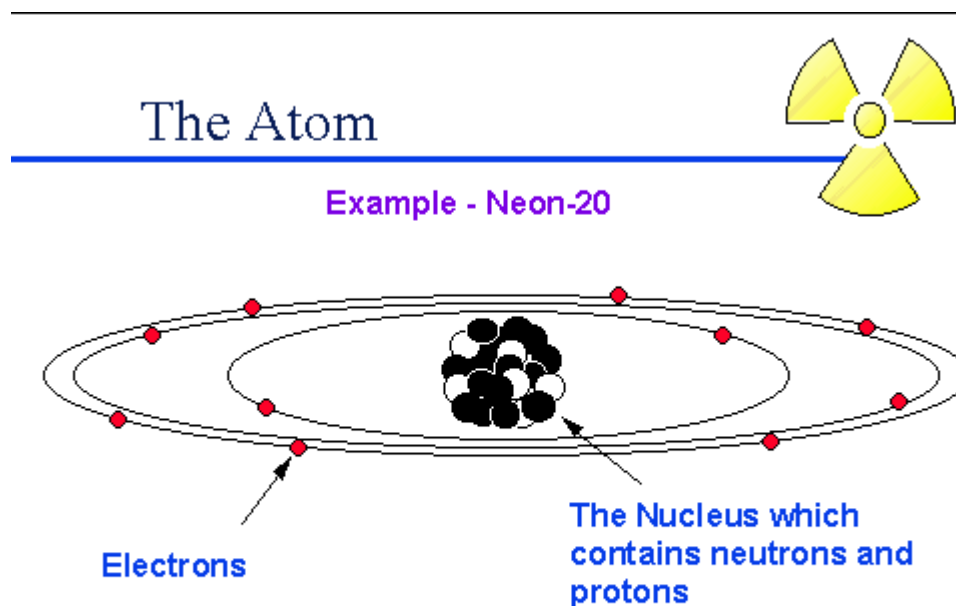
As you can see, matter consists mostly of open space! [So why does it hurt if you punch a brick wall?]
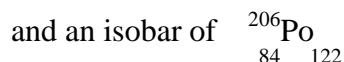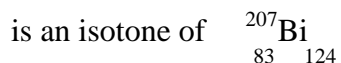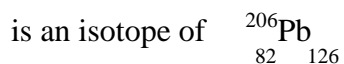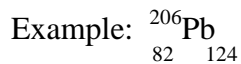
The Nucleus

Nuclei are referred to using the following nomenclature:

$$^{A}_{Z}\text{Element}$$

Z is the atomic number. It characterizes the element. It is also the number of protons. Since protons carry all the positive charge in a nucleus, **Z** also is the number of electrons in a neutral atom. Note that it is the electrons, the electronic configuration that determines the chemical behavior of an element.

N is the number of neutrons [The neutron was discovered by Chadwick in 1932]. **A** is called the "mass number" and A is equal to the sum of **N** and **Z**. Collectively, neutrons and protons are called "nucleons".

A species of nucleus of a given **Z** and **A** (or **N**) is called a <u>nuclide</u>. Nuclides of an element (i.e. same **Z**) with different **A** (or **N**) are called <u>isotopes</u>. [Meaning "same place" i.e. on the periodic table.] Nuclides having the same **N** are called <u>isotones</u>, and nuclides having the same **A** are called <u>isobars</u>. [A handy way to keep these straight is to note that isoto<u>p</u>e includes the letter "p" (same <u>p</u>roton number), isoto<u>n</u>e the letter "n" (same <u>n</u>eutron number), and isob<u>a</u>r the letter "a" (same **A**). \

Example: $^{206}_{82}\text{Pb}_{124}$

is an isotope of $^{206}_{82}\text{Pb}_{126}$

is an isotone of $^{207}_{83}\text{Bi}_{124}$

and an isobar of $^{206}_{84}\text{Po}_{122}$

Of course, lead-206 has many other isotopes, isotones and isobars.

Using a Periodic Table of Elements, (eg. http://pearl1.lanl.gov/periodic/)try the following:

(i)   Write down three possible isotopes of Calcium-44 (^{44}Ca).

(ii)   Write down three possible isotones of Neon-21 (^{21}Ne).

(iii)   Write down three possible isobars of Selenium-78 (^{78}Se).