

The Quantum Mechanics of Alpha Decay

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The purpose of this experiment is to explore the relation between the mean lives of alpha-active nuclides and the energies of the alpha particles they emit. You will study portions of the sequences of radioactive transformations whereby uranium is transmuted into lead. Various procedures will be used to measure the alpha-particle energies and lifetimes of five alpha-active nuclides with mean lives ranging from days to microseconds, as well as the mean lives of two beta-active nuclides.

PREPARATORY QUESTIONS

1. With the help of Figure 7 and the associated reference, make charts of the decay chains that start with the naturally occurring isotopes of uranium, U , and terminating with a stable isotopes of lead. For each nuclide place a box in a coordinate system with atomic number and element name on the X axis and mass number (total number of nucleons) on the Y axis. In each box write the half life, decay mode(s) and energies, and branching probabilities.
2. Create a log-log plot of the expected relation between the mean lives of alpha-active nuclides and the energies of the alpha particles they emit.
3. Write and solve the equations that describe the growth and decay of alpha radioactivity in a sample of an isotope that decays by alpha-particle emission into another alpha-emitting isotope. Specifically, call $A(t)$ and $B(t)$ the numbers of atoms of the two isotopes, and τ_A , τ_B their mean lives, respectively. Assume the initial conditions $A_{t=0} = A_0$ and $B_{t=0} = B_0$. Plot A and B against time for $A = 1$, $B = 0$ and two special cases: $\tau_A \gg \tau_B$, and $\tau_A \ll \tau_B$.
4. Why are there no natural nuclides with $Z > 83$ and $A = 4n + 1$?
5. Consider two counters, #1 and #2, each producing random, uncorrelated pulses at average rates of r_1 and r_2 , respectively, where $r_1 \gg r_2$. What is the rate at which a pulse from #1 is succeeded by a pulse from #2 within the time interval from t to $t + dt$? What is the mean value of such time intervals? (In a measurement of the mean life of a nuclide by the method of delayed coincidences you will have to take care that your data are not swamped by events produced by random and uncorrelated events in the two detectors.)
6. Explain how a silicon barrier solid state detector works. (see Reference[7]).

INTRODUCTION

The first Nobel Prize in physics was awarded in 1901 to Wilhelm Röntgen for his discovery on November 8, 1895 that a penetrating radiation (X rays) is emitted by the fluorescing glass of a cathode-ray tube under bombardment by cathode rays inside the tube. (Frederick Smith at Oxford missed that discovery: When he earlier noticed that films left near a cathode ray tube were darkened, he told his assistant to move the films away.) Antoine Becquerel heard Röntgen's report on January 20, 1896 at the Académie des Sciences. He immediately set about to investigate whether other fluorescent materials emitted penetrating radiations. After ten days without success he read about the fluorescent properties of uranium salts. On January 30 he sprinkled some uranyl potassium sulfate on top of a photographic plate wrapped in light-tight paper, exposed the salt to sunlight for a few hours, developed the plate, and found it darkened. He prepared to repeat the experiment on February 26 and 27, but the weather was cloudy, so he put the plate and salt away in a drawer to wait for a sunny day. On March 1 he developed the plate, "expecting to find the images very feeble. On the contrary, the silhouettes appeared with great intensity. I thought at once that the action might be able to go on in the dark." Thus Becquerel discovered the phenomena that was later dubbed "radioactivity" by Marie Curie.

Marie Sklodowska came from Warsaw to Paris to study at the Sorbonne in 1891. A few years later she began her studies of radioactivity in collaboration with her husband, Pierre Curie. By 1898 she had discovered the radioactivity of thorium and had isolated from residues of uranium refinement a new element, radium, and showed that its activity per gram was a million times greater than that of uranium.

Ernest Rutherford, a young New Zealander working at the Cavendish Laboratory of Cambridge University, showed that the radiation emitted by uranium is of two types distinguished by their penetrating power. In a publication, completed in 1899 after he had moved to McGill University in Montreal, Rutherford called the easily absorbed radiation "alpha rays" and the more penetrating radiation "beta rays". Becquerel discovered that the beta rays are deflected by electric and magnetic fields

like charged particles with the same value of e/m as the recently discovered electrons. Then Rutherford demonstrated that alpha particles are doubly charged ions of helium, whose spectrum had been discovered first in the sun only a few years previously, and then in the gases emitted by uranium ores. Shortly thereafter Paul Villard in France identified a third type of radiation he called “gamma”, which turned out to be high energy electromagnetic radiation.

In 1900 Rutherford and Soddy found that thorium emits a short-lived radioactive gaseous element (an isotope of radon (Rn^{228} called thoron) that is chemically inert, like argon. From measurements of its radioactivity they drew the epochal conclusion that thoron decays into other radioactive elements — the discovery of the transmutation of elements. A year later Marie Curie discovered a similar gaseous element Rn^{226} emitted by radium. In a series of papers on radioactivity Rutherford and Soddy unraveled most of the complex relations between the decay products of uranium and thorium. They discovered the exponential law of radioactive decay

$$N(t) = N_0 e^{-t/\tau} \quad (1)$$

in which the mean life τ of the substance is a fundamental characteristic that is unaffected by heat, chemical combination, or any other condition that affects the electronic structure of the atom (except the absence of K electrons in the case of beta decay by electron capture). Also in 1903 Soddy published a calculation of the energy released by the decay of radium and wrote that it is “at least twenty-thousand times, and may be a million times, as great as the energy of any molecular change.” Four years later Albert Einstein, in his discovery of the theory of relativity, deduced the equivalence of mass and energy expressed in his equation $E = mc^2$ and suggested that the energy released in radioactivity was converted mass.

Rutherford dominated experimental nuclear physics for the next thirty years. His greatest discovery was the nucleus itself which he perceived in the results of the alpha-particle scattering experiments he initiated at Manchester University in 1910. A fascinating account of this era has been written by Richard Rhodes [1986] in *The Making of the Atomic Bomb* from which the above history is taken.

RADIOACTIVITY

Most of the natural elements with atomic numbers from 1 (hydrogen) to 82 (lead) are stable. The few exceptions include the isotope of potassium, K^{40} , which constitutes 0.01% of natural potassium and suffers beta decay with a mean life of 1.9 billion years. All the isotopes of the elements with $Z > 82$, with the possible exception of Bi^{209} , are unstable and decay with half lives ranging from microseconds to billions of years. So where, when, and

how did the natural radioactive elements arise? Since the 1950’s, and particularly from the work of William Fowler of Cal Tech and his collaborators, it has become clear that all the elements with $Z > 26$ (iron) are built up suddenly from lighter elements by absorption of neutrons in the neutron-rich region of a supernova during the first ~ 1 sec of the explosion, the so-called rapid process (r-process). Supernovas occur in our galaxy at a rate of about 2 or 3 per century. One of these probably triggered the formation of the solar system and enriched its raw material with freshly synthesized heavy elements of which all with $Z > 82$ have been decaying ever since.

Light nuclei with equal numbers of protons and neutrons (e.g., He^4 , C^{12} , O^{16}) are stable. Heavier nuclei ($Z > 10$), to be stable, need a higher proportion of neutrons to provide sufficient nuclear-force binding to overcome the Coulomb-force repulsion of the protons (e.g., K^{41} , where $A = 2Z + 3$). Thus a nucleus with too few neutrons will convert a proton to a neutron by emission of a positron and an electron neutrino. On the other hand, a free neutron is slightly heavier than the proton and decays into a proton, electron, and anti-neutrino with a mean life of ~ 12 minutes. Therefore, it can be energetically favorable for a nucleus with too many neutrons to convert a neutron to a proton by emission of an electron and anti-neutrino. Thus nuclides with too few or too many neutrons transmute by electron emission toward the stable region along the bottom of the valley. A 3-D plot of neutron number (y-axis) versus proton number (x-axis) versus potential energy per nucleon (z-axis) for all the isotopes of the elements shows a steep potential energy “valley of stability” with a positive curvature in the x-y plane. The three kinds of radioactive transmutation are:

1. Alpha decay (ejection of a helium nucleus consisting of two neutrons and two protons) is the most common decay mode for elements above lead and near the bottom of the stability valley.
2. Beta decay (emission of a positive or negative electron or capture of a K-shell electron together with emission of a neutrino or anti-neutrino) generally occurs in nuclides on the valley sides and serves to correct unstable proportions of neutrons and protons.
3. Spontaneous fission (division into two nuclei) occurs above uranium and is a mode of rapid decay of the artificial elements above $Z = 100$. Theoretical speculation about possible high- Z islands of stability have stimulated numerous, and so far unsuccessful, experiments to produce elements with $Z=114$ or other so-called *magic numbers*.

A chain of radioactive decays, starting with a heavy nuclide near the bottom of the stability valley (e.g., U^{238})

and proceeding by alpha decays, produces nuclides with increasing deficiencies of neutrons and corresponding positions higher up the valley wall. The deficiency is corrected at various steps in the chain by emission of a negative electron, i.e., beta decay. The three decay chains found in nature are illustrated in an appendix. The fourth doesn't exist in nature because it has no long-lived parent nuclide that could have survived in significant quantity since the creation of earth's elements. Alpha decay occurs whenever permitted by energy conservation, unless preempted by beta decay. Consider the decay process



The total mass-energy ($mc^2 + \text{kinetic energy}$) must be conserved. Thus, in the spontaneous alpha-decay of a radium nucleus we have,

$$\text{Ra}^{226} = \text{Rn}^{222*} + \text{He}^4 + Q, \quad (3)$$

where $Q=4.869$ MeV is the total kinetic energy of the product nuclei if they are in their ground states. (Precise values of isotopic masses are listed in the CRC nuclide table). The energy Q is shared in accordance with the conservation of momentum between the mutually repelled He^4 and Rn^{222} nuclei. However, the latter, being a complex system of many constituent particles, may be left in an excited state from which it later decays by gamma emission. Thus the kinetic energy of the remnant particles (alpha particle and Rn^{222} nucleus) just after the decay event depends on the energy of the excited state of the Rn^{222} nucleus. High resolution spectrometry of the alpha particles from Ra^{226} reveals a "fine structure" [4.782 (94.6%), 4.599 (5.4%), 4.340 (0.0051%), 4.194 (7×10^{-4})], each energy corresponding to a particular excited state of the daughter nuclide Rn^{222} .

Generally, a nuclide, created in an excited state by alpha or beta decay of its parent nuclide, decays to its ground state by gamma-ray emission. In some cases it may undergo alpha decay before it has had time to settle to its ground state, thereby giving rise to a more energetic alpha particle, which is another source of fine structure in alpha spectra.

THE CORRELATION BETWEEN ENERGY AND HALF LIFE IN ALPHA DECAY

The principal aim of this experiment is to explore the correlation between the half lives of radioactive nuclides and the energies of the alpha particles they emit – the shorter the half life the higher the energy. The most abundant isotope of uranium, U^{238} , with a half life of 4.5 billion years, emits alpha particles with an energy of

4.2 MeV; Po^{212} with a half life of $0.304 \mu\text{s}$ emits alpha particles with an energy of 8.785 MeV – in this case a ratio of $\sim 4 \times 10^{23}$ in half life corresponds to a factor of only ~ 2.1 in energy! The quantitative expression of this correlation, discovered early in the investigation of radioactivity, is called the Geiger–Nuttall law.

Then, in 1927, Rutherford observed that the scattering of 8.57 MeV alpha particles (from $\text{ThC}'_{84}\text{Po}^{212}$) by a thin uranium foil is exactly what is expected for elastic scattering by a perfect $1/r$ Coulomb potential down to the closest distance of approach for such particles, namely $\sim 3 \times 10^{-12}$ cm. Thus the Coulomb barrier around a uranium nucleus must be at least as high as 8.57 MeV, and any alpha particle going over the top to get out must be pushed away by Coulomb repulsion resulting in a final kinetic energy of at least that amount. And yet the energy of alpha particles emitted by uranium is only 4.2 MeV!

The explanation of these remarkable facts was an early triumph of the quantum mechanics discovered independently and in different forms by Erwin Schrödinger and Werner Heisenberg in 1926. Two years later, George Gamow, a postdoc from Russia studying with Niels Bohr at the Institute for Theoretical Physics in Copenhagen, derived the equation for quantum mechanical tunneling of alpha particles *through* the Coulomb barrier of the nucleus. Two young Americans studying in Europe, Edward Condon and Ronald Gurney, discovered the same explanation independently. Their publications on the topic are reproduced in an appendix to this guide located in the Junior Lab File Cabinet.

As mentioned earlier, radioactive decay is described by Equation 1 where τ is the **mean life**. The **half life**, $\tau_{1/2}$, is the solution of the equation

$$e^{(-\tau_{1/2}/\tau_A)} = \frac{1}{2}, \quad (4)$$

which is

$$\tau_{1/2} = \tau_A \ln 2 = 0.693\tau_A. \quad (5)$$

THE QUANTUM MECHANICS OF ALPHA-PARTICLE EMISSION

Nuclei consist of protons and neutrons held together by the strong, short range nuclear force which exceeds the Coulomb repulsion between the protons at distances less than of the order of 1 fermi (10^{-13} cm). The potential function (a plot of the potential energy per unit charge of a positive test particle against position) for a spherical high- Z nucleus is sketched in Figure 1. Inside the nuclear radius the potential can be approximated by a square well potential. Outside, the barrier is accurately represented by a potential proportional to $1/r$. The wave

function of an alpha particle, initially localized inside the well, can be represented as a traveling wave that is partially reflected and partially transmitted at the barrier. The transmission coefficient, very small, is the probability that the alpha particle will penetrate the barrier. As it bounces back and forth inside the well the particle has multiple chances of penetrating the barrier and appearing on the outside. If it does, then the potential energy which the alpha particle had inside the nucleus is converted to kinetic energy as it slides down the outer slope of the barrier.

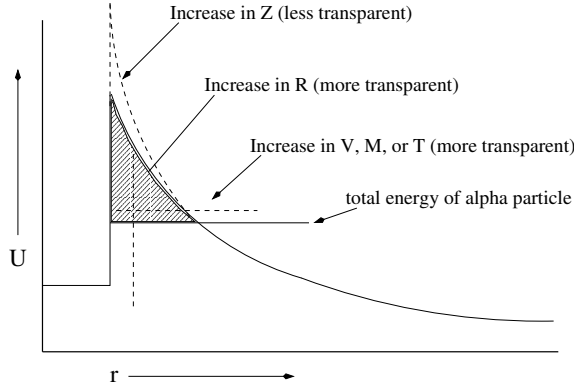


FIG. 1: Plot based on Figure 5.7 from Evans [1955] illustrating the nuclear potential barrier, and the effects of changes in the nuclear parameters on the transparency of the barrier.

EVOLUTION OF RADIOACTIVE MIXTURES: THE BATEMAN EQUATIONS

The source of radioactive nuclides in the present experiment is an ancient ($\sim 10^9$ years old) sample of the mineral uraninite (mainly UO_2 and U_2O_8), borrowed from the Harvard mineral collection to which it was donated by a young rock hound who pried it out of a deposit between layers of mica in the granite shield of Maine.

The sample is evolving in time as the various components of the decay chains are born and decay. The rates of growth and decay of radioactive isotopes in such mixed samples is governed by a set of first-order linear differential equations. Analysis of the variations of the counting rates of the various activities in light of these equations and their solutions can yield measures of the mean lives and relative abundances of the various nuclides.

A full exposition of the differential equations that describe the growth and decay of activities, the so-called Bateman equations, is given by Evans [1955], pp 470–510. Here we consider only the two simple cases that will be of greatest use in planning and analyzing your measurements. The fundamental law of radioactive decay is that at any instant the change, dA , in the number of atoms

of any given radioactive nuclide in a time interval, dt , is proportional to the number, A , present at that particular instant times dt , i.e.,

$$dA/dt = -A/\tau_A \quad (6)$$

where τ_A is a constant characteristic of the nuclide. Integration of this equation leads immediately to the expression, previously cited, for the number at any time t , namely

$$A(t) = A_0 e^{(-t/\tau_A)}, \quad (7)$$

where A_0 is the number present at $t = 0$. Now consider the relations between the activities (decays/second) of two radioactive isotopes, A and B , such that B arises from the decay of A . Let $A(t)$ and $B(t)$ represent the numbers of atoms of the two isotopes present at any given time, A_0 and B_0 the numbers present at $t = 0$, and τ_A and τ_B their mean lives, respectively. The rates of decay are A/τ_A and B/τ_B . As before, equation 6 describes the change of A . The rate of change of B is the algebraic sum of the rates of build-up and decay of B ,

$$dB/dt = A/\tau_A - B/\tau_B. \quad (8)$$

Equations 6 and 8 are simultaneous, first-order, differential equations that must be solved subject to the initial conditions. The general solution to equation 6 is equation 7. The general solution of equation 8 is given by the sum of the solution of the homogeneous equation, which is identical in structure to equation 6 and therefore has the solution $B_h = f_h e^{(-t/\tau_B)}$ where f_h is any constant, and a particular solution. To find the particular solution, we use the homogeneous solution as a bootstrap and set $B_p = f_p(t) e^{(-t/\tau_B)}$ (this is the method of variation of parameters). Substituting B_p for B and the solution for A in equation 8 and rearranging, we obtain for f_p the differential equation

$$df_p/dt = (A_0/\tau_A) e^{-t(1/\tau_A - 1/\tau_B)}. \quad (9)$$

Integration of equation 9 and imposition of the initial condition $B(t = 0) = B_h(t = 0) + B_p(t = 0) = B_0$ yields the solution (for $\tau_B \neq \tau_A$)

$$B(t) = B_0 e^{(-t/\tau_B)} + A_0 \frac{\tau_B}{\tau_A - \tau_B} [e^{(-t/\tau_A)} - e^{(-t/\tau_B)}]. \quad (10)$$

One form of equation 10 is of special importance to the interpretation of data from this experiment. Suppose $B_0 = 0$, $\tau_A \gg \tau_B$ and $t \ll \tau_A$. Then the rate of decay of B is

EXPERIMENTS

$$r_B(t) = B(t)/\tau_B \approx [A_0/\tau_A][1 - e^{(-t/\tau_B)}] \quad (11)$$

Under these circumstances the decay rate of B rises from zero and approaches asymptotically the decay rate of A .

These simple analyses provide us with at least two means of determining a nuclide's lifetime:

1. One can derive an estimate of τ_B as the negative slope of a plot of $\ln[r_B(t = \infty) - r_B(t)]$ against t . The diffusion of radon out of uraninite rocks occurs at a nearly steady rate and can be considered the equivalent of production by radioactive decay of a very long-lived precursor.
2. A second way to determine τ_B is to turn off the supply of B after its decay rate has reached a respectable value. Then the B rate will simply decline according to

$$r_B = (B_0/\tau_B)e^{(-t/\tau_B)}. \quad (12)$$

When a nuclide is third or later in a decay chain, the equation describing its decay rate is more complex, and interpretation of the variation of an observed decay rate in terms of its mean life becomes more difficult or impractical. If the mean lives of the secondary nuclides in the chain are short compared to τ_A , and if one waits for a time long compared to any of those mean lives, then a steady state is approached in which *all* of the decay rates are the same and equal to A_0/τ_A .

Numerical solutions to the Bateman Equations

In spite of the complexity of the analytical solution, a set of differential equations for a multi-nuclide decay chain is readily solved by numerical integration with results that can be displayed as plots of the various decay rates against time. A LabVIEW code which calculates numerical solutions of the Bateman equations is available from <http://jls.mit.edu/shared/software>. You supply the data on initial quantities of isotopes, their decay modes, and their mean lives. The program will plot the alpha-decay rates of the individual isotopes and the total alpha-decay rate.

A second option, using Matlab, is described in Appendix A and which performs the same calculations using matrices. Incidentally, it was Rutherford's observation that the heat generated by the decay of a fresh sample of thorium emanation (Rn^{220}) initially increased with time (the total rate of release of alpha-decay energy increased as the quantity of daughter nuclides increased) that led him to the conclusion that he had discovered the transmutation of elements.

The uraninite sample, consisting of a few small black rocks, is contained in the can which will be assigned to you for the duration of your experiment, along with an expensive and extremely delicate silicon barrier detector that will serve as the sensor of your alpha-spectrometer (take great care not to let anything touch the surface of the detector). The rocks leak radon gas which can be trapped in the can by sealing it with the rubber-stopper in which the detector is mounted.

In this experiment samples of such generically related nuclides are created on the surface of the silicon detector. Each alpha-decay process involved in a decay chain reveals itself as a distinct peak in the energy spectrum, and its rate can be measured by appropriate manipulation of the recording sequence. Thus all the necessary data for an exploration of the energy-mean life relation in alpha decay can be obtained with this one "simple" device. One can imagine that Lord Rutherford, in 1905, would have given his eyeteeth for such a detector to replace the ion chambers, cloud chambers, calorimeters, and spinthariscopes he used in his discoveries.

When a radon atom suffers alpha decay, the daughter polonium atom recoils with sufficient velocity to have several of its outer electrons stripped in collisions with air molecules. An electric field, established by application of high voltage (~ 425 VDC) between the can and the detector, draws the polonium ion to the surface of the detector where it sticks. Its subsequent decay and the decays of its daughter nuclides yield alpha and beta particles. If an alpha particle from an atom stuck on the detector surface is emitted in a direction into the detector, then nearly all of its energy will be dissipated in the sensitive volume of the silicon where the number of electrons raised to the conduction band is almost exactly proportional to the dissipated energy. The conduction electrons are swept out of the silicon by the field established by the bias voltage (~ 45 VDC) applied to the detector. The resulting charge pulse is detected by the preamplifier as a voltage pulse which is amplified and analyzed with a multichannel analyzer (MCA) card located in the Dell computer. This latter device accumulates a record of the distribution in height of the voltage pulses which is a spectrum of the energies of the detected particles.

You will be assigned a silicon barrier detector and a can containing a sample of uraninite rocks for your exclusive use during this experiment. The mixture of decaying nuclides stuck on the surface of the detector depends on the history of its exposure to the decay products of radon that emanates from the uraninite rocks, and on the compositions of the particular uraninite rocks. Therefore it is essential that you control the exposures for your own purposes during the two-weeks of the experiment. You will find a lot of things going on in the can and on the

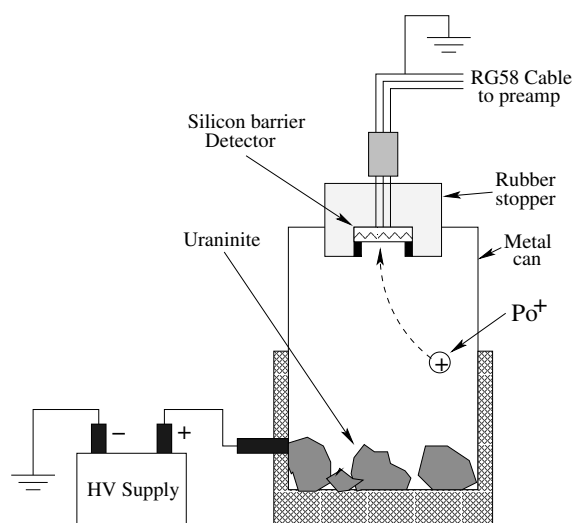


FIG. 2: Schematic diagram of the metal can containing uraninite, sealed by the rubber stopper carrying the silicon barrier detector, and attached to the high voltage supply. A polonium ion is shown as it is about to be drawn to the detector by the electric field.

surface of the detector that are not explicitly mentioned in this lab guide. To figure out exactly what is going on you will have to keep a meticulous timed and dated record of your manipulations and observations.

CAUTION

Precursor nuclides of all the activities studied in this experiment are isotopes of the noble gas, radon, which diffuse at steady rates out of the rocks in the can. The nuclides drawn to the surface of the detector by the imposed electric field are the isotopes of polonium (215, 216, and 218) which are the decay products of those radon isotopes. Therefore, to initiate a useful amount of the action that follows deposition of one of those polonium isotopes on the detector surface there must be an adequate activity of its parent radon isotope in the gas *in the can* when you apply the high voltage, and the voltage must be left on for a time of the order of the longest-lived nuclide in the subsequent chain leading to the isotope under investigation. For example, if you want to study the rapid decay of Po^{212} , you must accumulate its 10.64-hour precursor Pb^{212} in the on the surface of the detector for a day or more with the high voltage on.

To study the decay of Po^{218} you will need to keep the can sealed for a couple of days to accumulate the long lived precursor Rn^{222} gas that leaks from the rocks. its activity will build toward its equilibrium value in proportion to the function $1 - e^{(-t/\tau)}$ where τ is its mean life of about 3 days. **If you open the can, the radon will escape, and you will have to wait for a time of the order of τ for the activity to build up to a useful**

value again. When you have enough of the gas in the can, you can measure the meanlife of its daughter nuclide Po^{218} by turning on and off the high voltage applied to the can. In the case of the 219 and 220 radon isotopes those times are of the order of minutes and seconds. So **THINK** before you open your can.

A technique particularly well suited for determining the mean life of very short-lived species is to use the decay event that gives birth to a particular nuclide as a start signal for a time-to-amplitude converter (TAC), and the decay event of the nuclide as the stop signal. The distribution of time intervals between the start and stop signals is the decay curve of the nuclide. This method will be used to measure the mean lives of Po^{214} and Po^{212} . The sensitive layer of the silicon detector is too thin to yield a usable start pulse from the lightly-ionizing electrons emitted in the beta decay of the precursor bismuth nuclei. Therefore it is necessary to employ a separate detector, a plastic scintillation detector with a thin window, to detect the beta-decay electrons that signal the birth of the short-lived polonium nuclei.

Historical Note: This experiment has also been done using a commercial Ra^{226} source placed in a vacuum jar in which the pressure can be controlled to adjust the thickness of air between source and detector. The uraninite rocks provide a richer alpha-spectrum as they contain daughters from ALL the naturally occurring radioactive chains.

Suggested Experiment Schedule: Overview

- Day 1.** Seal a can containing uraninite rocks with a rubber stopper in which a certified “good” alpha particle detector is embedded. Apply a voltage ($\sim 450\text{V}$) between the can and lab ground (e.g., one of the cable connectors) so as to draw to the surface of the detector *positive* ions of decay products from radon isotopes leaking out of the rocks. **Make sure the ground terminal of the floating 450V power supply is connected to earth ground using an alligator clip to the side of the NIM bin!** Record the energy spectra of the alpha particles. With the help of your charts from the preparatory questions, identify the features in the energy spectra and explain the shapes of the lines. Prepare a sealed sample of radon in liquid scintillator and initiate a measurement of the relatively long mean life of Rn^{222} .
- Day 2.** Determine the mean lives of various alpha and beta activities by measuring changes in counting rates measured with the silicon detector. Measure the alpha decay activity in the liquid scintillator.
- Day 3.** Measure the very short mean life of Po^{212} by the delayed coincidence method using the auxiliary

plastic scintillation counter to detect the precursor beta decays of bismuth. Measure the alpha decay activity in the liquid scintillator.

Day 4. Refine whatever parts of the experiment need additional attention. Measure the remaining alpha decay activity in the liquid scintillator.

DETAILED EXPERIMENTAL NOTES

Day 1: The Energy Spectra of Alpha Particles from the Decay Products of Uraninite Emanations

The silicon detector assigned to you has probably been exposed recently to alpha emitters of which not all have decayed to negligible levels of activity. You can use the residual activity to test the detector and spectrum analysis equipment, and to adjust the gains of the system to achieve a convenient spread of the spectral lines (0–10 MeV) with good linearity.

Connect the detector and other components as shown in Figure 3, **except for do not turn on or connect the detector bias voltage supply**. Place the detector in its rubber stopper face down on the table *so that room light does not strike the surface when the ~ 45 -VDC bias voltage is applied*. Set the MCA lower level discriminator (in software) to ~ 100 channels to eliminate detector noise.

Turn on the Ortec 428 bias supply, set it to +45 VDC and measure the voltage at the SHV connector before connecting to the preamplifier’s bias voltage input (the Ortec 428 dial is a rather coarse control and you want to be sure not to exceed the manufacturer’s maximum voltage recommendation at the risk of destroying the (expensive) detector.

After setting up the scope trigger and gain, adjust the spectroscopy amplifier so that the highest-energy pulses occur at ~ 8 VDC (MCA channel ~ 1600). Beware, you may be plagued by 80MHz rf pickup, probably from the MIT FM radio station broadcasting over the MIT 110V power line!) Record all the settings of all the component in your setup so that you can reestablish the same measurement conditions in your next lab session.

1. Record a 300 s spectrum with the MCA memory with the detector face down on the table.
2. Seal your can with your stopper/detector. Record a second 300 s spectrum with no high voltage applied to the can. Compare this spectrum with the one you obtained with the detector face down on the table.
3. Connect the high voltage (~ 425 V) power supply in black plastic box between the can and lab ground and record a third 300 s spectrum .

4. Reverse the voltage and record a fourth 300 s spectrum.

Try to understand everything you see in these four spectra. Ask for advice if there are mysteries you can’t solve. Record a one hour spectrum with ~ 425 V applied to the can with the polarity that produces the most action. While you are waiting, get started on the long-term measurement of the mean life of Rn^{222} . You will need to get data over several days, so it’s wise to start early on this measurement.

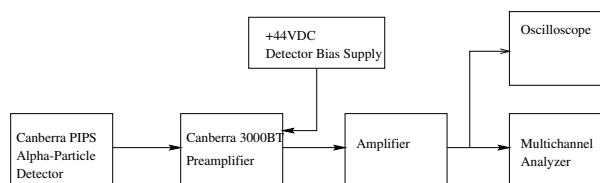


FIG. 3: Arrangement of components for measuring the energy spectra of alpha particles from nuclides deposited on the surface of the detector.

Measuring Mean Life of Rn^{222} and Po^{218}

Radon is a closed shell atom, the heaviest in the sequence of noble gasses which begins with helium. Formed in the earth’s crust by the decay of radium, radon diffuses into the atmosphere to become part of the air we breathe. Radon in the environment is now recognized as the major source of ionizing radiation to the general population. There is a concerted effort by federal and local agencies to mitigate “high” radon levels found in some homes. We place the quotes around “high” to emphasize that the adjective is appropriate only for very long term exposures to the general population. The radon intensities of concern in the environment are measured in picocuries, which is a millionth the source strength of the radioactive sources used in smoke detectors or the calibration sources we use in the laboratory. The radioactivity of the radon sources used in the present experiments are very weak and pose no health hazard.

Radon is adsorbed by charcoal. This property can be exploited to prepare a solution of radon and its decay products in a liquid scintillation cocktail in which the beta and alpha decay events can be detected as scintillations by the Beckman counter described below. Hydrocarbons in the scintillation cocktail elutes radon from charcoal. **While the commercial liquid scintillator (Beckman’s Formula-989) used in Junior Lab is not particularly dangerous, please wear gloves and note the safety precautions listed on the Material Safety Data Sheet (MSDS) posted by the hood.** In a lead box in the chemical hood there is a tightly capped jar containing packets of charcoal and

uraninite ($\sim 325\text{g}$ of nearly pure UO_2). After several days (comparable to the mean life of radon so that it is in equilibrium with its precursors) of exposure to the emanations of the mineral, the charcoal carries a load of adsorbed radon which can be transferred to liquid scintillator. The jar is split into two halves, labeled **ripe** and **unripe**, please use the former!

The maximum activity of radon atoms in equilibrium with one mole of uranium is $\frac{N}{\tau_{\text{U}^{238}}}$ disintegrations s^{-1} , where $N = 6.03 \times 10^{23}$ is Avogadro's number, and $\tau_{\text{U}^{238}} = 6.5 \times 10^9 \text{ yr}$ is the mean life of uranium. Thus we find that the maximum activity we can expect to find in the charcoal to be $2.9 \times 10^6 \text{ s}^{-1}$ ($\sim 70 \mu\text{Ci}$). If this is divided among 10 packets of charcoal, then the maximum activity of each is $2.9 \times 10^5 \text{ s}^{-1}$, which is more than enough for these experiments.

You will determine the mean life of Po^{218} by measuring the rapid growth (minutes) of its radioactivity in a freshly prepared sample of Rn^{222} , and the mean life of Rn^{222} from the slow decay products. You will have to work rapidly at the beginning of this measurement in order not to miss the early stages in the build up of the Po^{218} activity. Study the following procedure before you begin. **Make up a data table in your lab book to record the times (starts and durations), the number of counts, and your comments.** Practice using the Trump-PCI MCA so that you can record successive energy spectra for various "preset" time intervals. You must also learn how to use the ROI (region of interest) feature to display the total number of counts recorded between two different selected amplitudes in a spectrum, as well as the number with background subtracted.

The "sum" signals from the Beckman scintillation counter (ignore for now the "coin" signals) should be fed directly to the signal input of the MCA located on the back of the computer. The amplifier should be set for a negative input with fast pulse shaping and a gain that spreads the full spectrum over about 2/3 of the range of the display. You may need to set the lower level discriminator (LLD) at 2 or 3% to suppress the counting of very small pulses (which may be rf interference).

Detailed Procedure for Rn^{222} and Po^{218}

Make up a solution of radon in liquid scintillator hermetically sealed in a glass vial in which the alpha-decay activity of Rn and its shorter-lived decay products can be assessed from time to time over the next several days with the Beckman scintillation spectrometer. Place in a glass vial a radon-loaded charcoal packet extracted from the "radon farm" jar in the lead box in the hood. Pump in 10 cc of liquid scintillator from the reservoir. Cap the vial and rock it gently for \sim ten seconds. Remove the packet and discard it in the waste jar. To prevent false results due to radon leaking from the vials, it is essential

that they be hermetically sealed. You can improve the seal by applying vacuum grease to the rim of the vial and capping it tightly so the radon does not leak out during the several days required for the measurements. Lower the vial into the counting chamber and immediately start a sequence of measurements, appropriately spaced in time so as to define accurately the build up of the Po^{218} activity. The following is a suggested sequence:

1. Record three 30-second spectra with the shortest possible intervals between, making note of the starting times of each measurement.
2. After the third spectrum, wait one minute and take another 30 second spectrum. Repeat with intervals of 2, 4, and 8 minutes between the starts of successive accumulations, each time setting the preset duration of the measurement to be half the time since the start of the last measurement (e.g. to 60 sec after the 2-minute wait, 120 sec after the 4-minute wait, etc.) These increased integration times will give you better statistical accuracy as you approach the asymptotic rate.

The eight spectra should reveal the growth of the ~ 6 MeV alpha particle peak from Po^{218} as a shoulder on the peak due to the alpha particles from Rn^{222} . You should also begin to see the beginnings of a peak at 7.6 MeV due to alpha particles from the decay of the ultra short-lived Po^{214} .

Measure on the MCA and plot, as a function of time, the intensity of the combined (and unresolvable) 6 MeV and the 5.5 MeV alpha-particle groups. To do this you will have to set up identical ROI's in each spectrum with the lower level set at the lower boundary of the alpha particle peak to as to exclude as much as possible, the counts due to the beta decays, and the upper limit set in the valley between the 6 and 7.6 MeV peaks so as to exclude as efficiently as possible the alpha decays of Po^{214} whose rate will continue to grow and confuse the result. The MCA will give you both the "gross area" (total number of counts) and the "net area" (the number of counts above a straight-line drawn from just to the left boundary of the ROI to the right boundary of the ROI (see the Trump-PCI manual for details). The area under the straight line is attributable mostly to the beta-decay events which continue to grow in rate due to the accumulation of Pb^{214} and Bi^{214} long after the rate of alpha decays from Po^{218} has reached its maximum rate.

Think about your preliminary results and how you might obtain improved data on your second run. Make a practice run to familiarize yourself with the procedure and to make any needed adjustments of the electronics. Make a second run for a good set of data. Each team should attempt to get at least two good sets of data.

After you have completed the comparatively rapid series of measurements required for a determination of the

Po²¹⁸ mean life, and extracted with the ROI feature of the MCA a good measure of the unresolved activities of the Rn²²² and Po²¹⁸, you can turn your attention to the much more gradual process of the decay of the parent nuclide itself, i.e. Rn²²². Since the mean life of radon is several days, you will have to follow the decay of your sample for several days or a week to get accurate results. Here you must overcome three problems. The first is that the beta activities of Pb²¹⁰ and Bi²¹⁰ and the alpha activity of Po²¹⁴ will continue to grow for several hours before they reach the equilibrium condition of being equal to the activities of Rn²²² and Po²¹⁸. Since you cannot isolate the counts due to Rn²²², any rate you measure will be influenced by these other activities. The best thing to do, therefore, is to wait until all the activities are nearly equal (a couple of hours). Then any reproducible ROI will give you a rate proportional to the Rn²²² activity from which you can determine the mean life. The second problem is to define the ROI in terms of features of the spectrum that you can identify from day to day as you return to the setup after it has been altered by other users. The upper limit can simply be set somewhat above the obvious upper limit of the entire spectrum. The lower limit can be set just below the lower limit of the distribution of alpha pulses, i.e. where the alpha peak just starts to blend into the plateau of beta-ray counts. The third problem is the possible leakage of the radon gas from the vial.

Detailed Analysis for Rn²²² and Po²¹⁸

Given the following facts:

1. The mean life of Po²¹⁸ is much shorter than that of Rn²²²,
2. The mean lives of the Po²¹⁸ decay products that lead to the production of alpha-active Po²¹⁴ are long compared to the mean life of Po²¹⁸,
3. Some Po²¹⁸ may be dissolved in the liquid scintillator along with Rn²²²,

one can show that the measured counting rate $r(t)$ is represented to a good approximation by a function of the form

$$r(t) = a - be^{-t/\tau} \quad (13)$$

where a, b and τ are constants. Thus

$$\ln[a - r(t)] = \ln b - t/\tau \quad (14)$$

The constant a is the total counting rate that is approached asymptotically by the two alpha activities at times much longer than the mean life of Po²¹⁸, but still much less than the mean life of Rn²²². It can be estimated from an examination of a plot of r vs. t . A plot

of $\ln[a - r(t)]$ vs. t during the first few minutes should be nearly a straight line with a slope of $-1/\tau$ where τ is the mean life of Po²¹⁸. (Note: as r approaches a , statistical fluctuations will cause wild gyrations or worse in the values of $\ln a - r$, so the most useful information about τ must be derived from the early part of the rising curve of activity.

Additional questions:

1. Describe how the pulse height spectrum changes with time?
2. Identify the part of the pulse height spectrum due to the beta decays of Pb²¹⁴ and Bi²¹⁴?
3. What is the effect of the accumulating radionuclide Po²¹⁰ on your results?

After initiating the Rn²²² measurements, return to the silicon detector experiment and measure the channel numbers of all the peaks of the one-hour spectrum. With the aid of the chart of the nuclides (located on the wall above the experiment), your preparatory question answers and [9], identify the peaks and explain their shapes. Do you see evidence of the famous high-energy alphas from Po²¹²? If so, estimate their energies and the fraction of the decays that give rise to them.

- Try to explain any discrepancies between the spectrum of pulse heights you have observed and their expected energies (e.g. consider the effect of a thin coating of protective material or dust over the source or the detector.)
- In light of the likely age of the uranite source and the opportunity it has had to achieve equilibrium among the quantities of its radioactive decay products, try to explain the inequalities you may see in the intensities of the various alpha particle groups.

At the end of the first lab session, seal your can with the stopper/detector and connect your high-voltage box between the can and the shield of the BNC connector with the appropriate polarity to draw the polonium ions to the detector. This will build up a supply of radioactive nuclides in the gas in the can and on the surface of the detector for analysis at your next session. Note the time in your lab notebook.

DAYS 2 AND 3: DETERMINATION OF THE MEAN LIVES OF SEVERAL NUCLIDES

In preparation for your second session devise a plan for the determination of the mean lives of as many nuclides as possible with data that you will be able to obtain from the setup used in the first session. Base your plans on the possibilities of measurement implied by equations 11

and 12 above. There are two alpha decays and two beta decays whose mean lives can be measured by appropriate manipulations with the silicon detector. Consider:

1. Disconnecting the high voltage and measuring the decay of the nuclides stuck on the detector surface
2. Reconnecting the high voltage and measuring the build up of activities
3. Flushing the accumulated radon out of the can with nitrogen, sealing the can, and then measuring the build up of activities as radon accumulates in the gas in the can.

You may want to consult your instructor about your strategies.

Do not break the hermetic seal of your can and let the accumulated radon escape before you have extracted all the useful information from the state of things that exists after a long accumulation of radon in the can.

Restore the settings of the measurement chain you established in the first session. Use the MCA to record the spectra and, for your convenience in analysis, make whatever fine adjustments of the amplification may be necessary to bring the highest energy peak into the same channel as before.

Record a one-hour spectrum for comparison with the one you recorded in the first session.

Now proceed with the strategy you have devised to measure the mean lives of as many of the activities as you can.

Sometime during your second session, measure the activity of your liquid scintillator with the Beckman spectrometer and record the date and time.

MEASURING THE MEAN LIFE OF Po^{212}

The mean-life of Po^{212} is very short. However, it can be measured by taking advantage of the fact that each of birth of a Po^{212} atom is signaled by emission of an energetic electron in the beta-decay of its parent Bi nucleus. The setup employs a scintillation detector with the scintillator covered by a thin aluminum foil that can be penetrated by most of the decay electrons. The silicon detector must first be plated with sufficient precursors of Po^{212} by exposure to radon gas in a can with the uraninite with the plating voltage source connected ($> \sim 1$ day). Then it is placed in the well of the scintillation counter so that electrons emitted downward in the decay of the precursor Bi nucleus will excite the scintillator. If the subsequent decay of the daughter Po nucleus produces an alpha particle going upward, the silicon detector will produce a pulse of the characteristic energy

of the alpha decay. The scintillation pulse is used to start the timing sequence of the time-to-amplitude converter (TAC); the silicon pulse is used to stop it. And the MCA records the exponential distribution of lifetimes from which the mean life can be determined.

The rate of accidentals during the short lifetime of Po^{212} is so small that no pulse-height selection of the alpha pulses is necessary. Thus the setup shown in Figure should work.

Recommended settings:

Canberra 2021:	coarse gain	100
	fine gain	0.75
	input pol	+
	shape time	1.0 μs
	rate	high
	mode	assymmetric
	threshold	auto
Ortec 459:	coarse	+1000 v
Canberra 814:	coarse gain	64
	fine gain	9
	amp	pos
	preamp out	
TAC:	range	100 ns
	mult	100
	TAC inhibitout	
Scope:	1 $\mu\text{s}/\text{div}$	
	$\pm 2\text{-}3$ v pulses	on chan 1 & 2

Caution: Cover the scintillation detector with several layers of black cloth to stop light leaks.

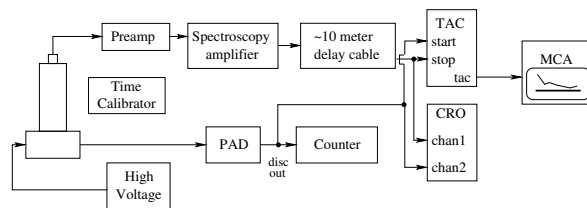


FIG. 4: Setup for measuring the decay of Po^{212} . The time calibrator puts out negative pulses so the input switch on the spectroscopy amplifier must be changed to negative for the calibration. The TAC is set to accept positive signals which should have amplitudes greater than 2 volts.

DAY 4: REVIEW AND REFINE YOUR MEASUREMENTS.

Measure the activity of the large radon in liquid scintillator sample, and derive the mean life of the radon isotope that has accumulated in the can.

Perfect the other measurements in light of your experience.

ANALYSIS

- Identify the parent nuclides in the uraninite.
- Identify all the nuclides whose alpha decays you have observed.
- Explain the shapes of the alpha peaks, especially the Po^{212} peak.
- Plot all the decay data and fitted decay curves, and estimate the mean or half-lives with errors.

DETERMINATION OF THE ENERGY-MEAN LIFE RELATION

The bottom line of this experiment is in the title: The Quantum Mechanics of Alpha Decay. With the data in hand on the energies and lifetimes of four nuclides of the uranium family, you can display the energy-mean life relation and contemplate the extraordinary dependence of the decay rate on the barrier height and its explanation in terms of the quantum theory of barrier penetration.

The analysis of your data on energies and mean lives should be carried out with regard for the statistical uncertainties. You should be able to determine the relative energies of the alpha particle groups from the silicon barrier detector data with uncertainties of less than 5% and the corresponding mean lives to within 10%.

Plot the log of the mean lives versus the alpha particle energies. Derive from your data a formula that expresses approximately the Geiger-Nuttall relation between mean life and energy.

Compare your experimental results with the expected relation from barrier penetration theory.

SOME POSSIBLE THEORETICAL TOPICS FOR THE ORAL EXAM

1. Barrier penetration and alpha decay.
2. The Bateman equations.
3. The relation between A and Z for nuclei stable against beta decay.
4. The energy loss of charged particles in matter.

EQUIPMENT

The following is an introduction to the measuring equipment. Additional information about some of the devices can be found in Melissinos [1966] and in the equipment manuals. In addition to the major pieces of

equipment described below, there are three power supplies used in this experiment. Costly damage can occur if they are inadvertently interchanged. The Ortec Model 428 Detector Bias Supply provides the low (~ 45) voltage to the Silicon Barrier Solid State Detector. This supply should be connected to the 'HV' input on the Canberra Model 3002BT preamplifier. The second supply is housed in a black plastic box and provides ~ 425 VDC for biasing the can containing the uraninite with respect to the solid state detector, causing polonium ions to be attracted to the surface of the detector. The third supply provides high voltage to the photomultiplier of the scintillation counter used to detect beta-decay electrons emanating from the nuclides deposited on the surface of the Silicon Detector.

The Beckman Liquid Scintillation Spectrometer

The Beckman scintillation detector consists two photomultiplier tubes (PMT's) that view the flashes of light in the liquid scintillation cocktail contained in a sample vial placed between them. Particle energy dissipated in the scintillator produces a flash of blue light at a rate of about one photon per 30 eV. Some of the light is absorbed by the metallic coating on the inside of the glass window of the PMT causing emission of a burst of photoelectrons. The photoelectrons enter a series of dynode stages (electron multiplier) from which an amplified burst of electrons emerges to be measured as a pulse of charge proportional to the amount of light in the scintillation flash.

Liquid scintillation cocktails are generally xylene or other organic liquid with a small quantity of dissolved complex aromatic hydrocarbon. A liquid scintillator converts into visible light about 1% of the energy lost by an energetic electron traversing the liquid. Thus a 1 MeV electron that stops in a liquid scintillator by virtue of energy loss due to ionizing collisions with carbon and hydrogen atoms yields a flash of several thousand photons (~ 1 visible photon per 100 eV of energy loss) which is readily detected by a PMT. The liquid scintillator is also an efficient detector of alpha particles. However, the rate of energy loss along the path of a stopping alpha particle is so great that the conversion capacity of the aromatic molecules is saturated and only about 0.1% of the ionization energy is converted into optical photons.

Figure is a schematic diagram of the Beckman liquid scintillation spectrometer. It has a light-tight, radiation-shielded counting chamber and associated electronics to supply power and process signals. In the chamber are two PMT's that view a platform on which a sample of liquid scintillator can be lowered for counting. The platform is raised and lowered through a light-tight shutter by an electromechanical elevator. The outputs from the PMT's are amplified and fed to an addition circuit and

a coincidence circuit contained in a chassis mounted by the side of the chamber. The output of the addition circuit is a negative 0–1V pulse proportional to the sum of the amplitudes of the signals from the two PMT's produced by a single scintillation event. The output of the coincidence circuit is a positive 5V logical pulse produced whenever two pulses above a certain minimum size arrive nearly simultaneously from the two PMT's. Dark current pulses from the PMT's are random and uncorrelated, so the chances of their occurring in two PMT's within the resolving time of the circuit is small. On the other hand, a faint scintillation event that produces enough light to cause one or more photoelectrons to be ejected from each of the two photocathodes will yield pulses that are simultaneous within the decay time of the phosphor, which is on the order of 10^{-8} s. Thus the coincidence logic signals are a very sensitive indicator of the occurrence of faint scintillation events, though they do not carry amplitude information.

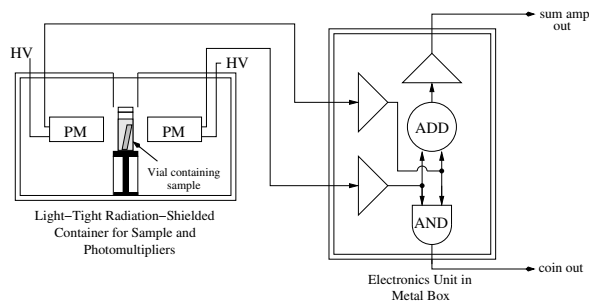


FIG. 5: Schematic diagram of the Beckman instrument for measuring the scintillations from alpha decays in the liquid scintillator.

Silicon Barrier Solid State Detector

When a charged particle dissipates an amount of kinetic energy, K , in the sensitive volume of a solid state detector, an average of K/ε valence electrons are raised to the conduction band and swept out by the bias field to form a pulse of negative charge. In silicon $\varepsilon \approx 3$ eV, so an alpha particle with an energy of several MeV produces a charge pulse containing so many electrons that the fluctuations are a negligible source of width of a line in a typical alpha energy spectrum — the actual line width ΔE depends on the quality of the detector, and in a good detector it should not exceed $\Delta E \approx 0.01E$. The solid state detector is connected to a preamplifier through which the bias voltage ($< \sim 50V!$) is supplied to the detector, and in which the voltage induced by the charge pulse at the gate of an input field-effect transistor is amplified before passing to a second amplifier and on to a multichannel analyzer for pulse-height analysis (the

physics of solid state detectors is described in Melissinos, 1986.)

Although silicon barrier detectors are supposed to work fine at room temperature, you will reduce thermal line-width broadening if the detector is cooled. One possible setup involves placing an upside-down metal beaker into a small, filled liquid nitrogen dewar. The bottom surface of the metal beaker should not be submerged. The barrier detector is then placed on the bottom surface of the beaker and cooled. The whole setup is covered by black cloth and can also be insulated.

Plastic Scintillation Counter

Beta-decay electrons emitted by the atoms plastered onto the silicon detector surface will be detected by a scintillation detector with a plastic scintillator in an arrangement shown schematically in Figure . The plastic scintillator is covered with a thin aluminum foil which is readily traversed by the beta-decay electrons. Information about the wiring and operation of a photomultiplier can be found in Melissinos.

Multi-Channel Analyzer

The distribution in amplitude (pulse-height spectrum) of pulses from a detector or the TAC (see below) is measured with a 2048 channel Perkin-Elmer multi-channel analyzer (MCA) card mounted in the computer. Details of the MCA operation can be found on the web at <http://web.mit.edu/8.13/labguides/mca/mca.pdf>. These cards allow full software control over upper and lower level discrimination voltages, have a coincidence (gated) detection mode, and offer a simple set of tools for establishing energy calibrations and determining peak statistics.

Time-to-Amplitude Converter/Single Channel Analyzer (TAC/SCA)

The Ortec Model 567 TAC/SCA time to amplitude converter (with its internal jumper set for positive input pulses) is used to convert the time intervals between pulses at the “start” and “stop” inputs into the amplitudes of output pulses. Thus, if a start pulse is generated in the plastic scintillator by a beta decay event heralding the birth of the radioactive nuclide Po^{212} , and the stop pulse is generated in the silicon detector by its decay, then the distribution in amplitude of the TAC output pulses will be proportional to the distribution in duration of the lifetimes of the Po^{214} atoms.

The SCA feature of the Ortec 567 is enabled by flipping the toggle switch adjacent to the output jacks. It

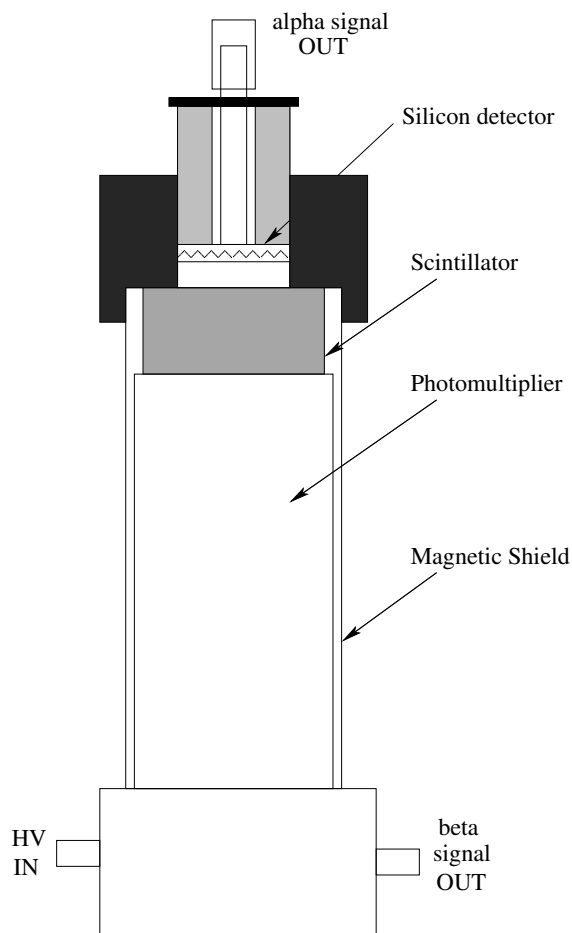


FIG. 6: Schematic diagram of the scintillation counter with the silicon detector in place.

will inhibit the TAC output except for pulses with amplitudes that fall within the “window” set by the lower and upper discriminator dials. Within that window the TAC output remains proportional to the start-stop time intervals. For each pulse in the window, a +5 VDC logic pulse is generated at the SCA output. It part of the experiment, it may be useful to use the SCA logic pulse to gate the MCA in an analysis of the amplitudes of pulses from the ‘sum’ output of the Beckman scintillation detector in order to identify which of the alpha particle groups is associated with the very short lived ($\sim 100\mu\text{s}$ nuclide.

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BATEMAN EQUATIONS MATRIX SOLUTION

The Bateman equations can be solved using matrices. The Bateman equations are

$$\frac{dN}{dt} = N, \quad \Lambda = \begin{bmatrix} -\lambda_1 & & & \\ \lambda_1 & -\lambda_2 & & \\ & \lambda_2 & -\lambda_3 & \\ & & & \ddots & \ddots \end{bmatrix} \quad (15)$$

N is the nuclide number row vector, with the parent nuclide the first entry. Λ is the decay constant matrix ($\lambda_i = 1/\tau_i$). The solution to this matrix differential equation, for initial conditions N_0 , is given by the matrix exponential,

$$N(t) = e^{\Lambda t} N_0 \quad (16)$$

The matrix exponential is defined as

$$e^{\Lambda t} = 1 + \Lambda t + \frac{(\Lambda t)^2}{2!} + \frac{(\Lambda t)^3}{3!} + \dots \quad (17)$$

and we can check that it solves Eq. (??) by substitution. Although we now have an elegant solution that is similar in form to the single-nuclide solution, calculating the matrix exponential numerically is computationally intensive. Using matrix algebra, Eq. (16) can be simplified for computational purposes. Since Λ is triangular, its eigenvalues are the main diagonal elements, the negatives of the decay constants. Let $D = \text{diag}\{-\lambda_i\}$ be the matrix of eigenvalues. Then Λ is diagonalized by the matrix of eigenvectors, V , as follows,

$$\Lambda V = DV \implies \Lambda = VDV^{-1} \quad (18)$$

Next, we express the matrix exponential $e^{\Lambda t}$ in terms of D ,

$$e^{\Lambda t} = e^{VDV^{-1}t} = Ve^{Dt}V^{-1} \quad (19)$$

This is a great computational improvement because the matrix exponential of a diagonal matrix is easy to calculate,

$$e^{Dt} = \begin{bmatrix} e^{-\lambda_1 t} & & & \\ & e^{-\lambda_2 t} & & \\ & & e^{-\lambda_3 t} & \\ & & & \ddots & \ddots \end{bmatrix} \quad (20)$$

The new equation for $N(t)$, substituting from Eq. (19), is

$$N(t) = Ve^{Dt}V^{-1}N_0 \quad (21)$$

and can easily be implemented in a program such as MATLAB.

MATLAB IMPLEMENTATION

```
function R=bate(t,lambda,No)
% R=bate(t,lambda,No)
%
% Calculates Bateman Equations
%
% t: time vector
% lambda: decay constants vector
% No: initial conditions vector (number of each nuclide)
%
% R: size(length(No),length(t)) matrix containing either decay
% counts (see comment below) for each nuclide in No
No = No(:);
Lambda = -diag(lambda)+diag(lambda(1:(end-1)),-1);
[P,D] = eig(Lambda);
O = P*diag(inv(P)*No);
E = D*repmat(t,length(No),1);
% Uncomment the first line for decay rates,
% the second line for decay counts
% R = diag(lambda)*O*exp(E);
R = O*exp(E);
```

FIG. 7: The Three Naturally Occurring Radioactive Decay Chains. From "Nuclides and Isotopes, 15th Edition", see <http://www2.bnl.gov/ton>