Chapter IV:
Radioactive decay
Summary

1. Law of radioactive decay

2. Decay chain/Radioactive filiation

3. Quantum description

4. Types of radioactive decay
History

• Radioactivity was discovered in 1896 by Henri Becquerel while working on uranium salts.
• On Earth, some elements show «natural» radioactivity due to their lifetime comparable to the age of the Earth (principally uranium and thorium).
• Artificial radioactivity (artificial production of radioactive elements) is possible with the use of particle accelerators or nuclear reactors → first time (Irène and Frédéric Jolliot-Curie):

\[
\begin{align*}
\frac{27}{13}\text{Al} + \frac{4}{2}\alpha &\rightarrow +\frac{30}{15}\text{P} + n \\
\frac{30}{15}\text{P} &\rightarrow +\frac{30}{14}\text{Si} + \beta^+\nu_e
\end{align*}
\]
Law of radioactive decay (1)

• Radioactive decay has a statistical character → impossible to precisely predict when a disintegration will happen → only probabilities may be given

• If 4 conditions are fulfilled:
  1. atoms are identical
  2. they are independent
  3. their mean life is long
  4. their number is large

→ the Poisson statistic can be applied:

\[ P_x(T) = \frac{(\lambda T)^x}{x!} e^{-\lambda T} \]

• \( P_x \) is the probability for \( x \) disintegrations in a time interval \( T \) and \( \lambda \) is the probability of disintegration per unit time = decay (desintegration) constant (independent of the nucleus age)
Law of radioactive decay (2)

- $\lambda dt$ is the disintegration probability of a nucleus in the time interval $dt$
- The survival probability of a nucleus at time $t$ (if existing in $t = 0$) →

\[ P_0(t) = e^{-\lambda t} \]

- If $N_0$ is the initial number (at $t = 0$) of nuclei → the number of survival nuclei $N(t)$ at time $t$ is:

\[ N(t) = N_0 e^{-\lambda t} \]
Half-life and activity (1)

• The half-life $T_{1/2}$ is time taken for half the radionuclide's atoms to decay $\rightarrow$

$$N(T_{1/2}) = \frac{N_0}{2} = N_0 e^{-\frac{T_{1/2}}{\lambda}}$$

$\Rightarrow$

$$T_{1/2} = \frac{\ln 2}{\lambda}$$

• The mean lifetime for decaying atoms $\tau$ is defined as the arithmetic mean of all the atoms' lifetimes $\rightarrow$

$$\tau = \frac{\int_0^\infty t \left| \frac{dN}{dt} \right| dt}{\int_0^\infty \left| \frac{dN}{dt} \right| dt} = \frac{\int_0^\infty \lambda t e^{-\lambda t} dt}{\int_0^\infty \lambda e^{-\lambda t} dt} = \frac{1}{\lambda}$$
Half-life and activity (2)

- Activity \( A(t) \) at time \( t \) is defined as the mean number of disintegrations per time unit →

\[
A(t) \equiv \lambda N(t) = \left| \frac{dN}{dt} \right|
\]

- The activity unit is Becquerel (Bq) → 1 Bq = 1 disintegration per second (old unit → Curie (Ci) corresponding to the activity of 1 g of \(^{226}\)Ra → 1 Ci = \(3.7 \times 10^{10}\) Bq)
Radioactive filiation (1)

• Simple case: Radioactive nucleus 1 ($N_0$ at time $t = 0$) decays with decay constant $\lambda_1$ to stable nucleus 2 $\rightarrow$

$$N_1(t) = N_0 e^{-\lambda_1 t}$$
$$N_2(t) = N_0 \left(1 - e^{-\lambda_1 t}\right)$$
Radioactive filiation (2)

- Two decay modes are sometimes possible → $\lambda_a$ and $\lambda_b$
- Total decay rate:
  
  $$ - \left( \frac{dN}{dt} \right) = - \left( \frac{dN}{dt} \right)_a - \left( \frac{dN}{dt} \right)_b = (\lambda_a + \lambda_b)N = \lambda_tN $$

- The total decay constant is $\lambda_a + \lambda_b = \lambda_t$
- Practically $\lambda_t$ is observed while $\lambda_a$ and $\lambda_b$ are determined by the final number of isotopes of each mode:
  
  $$ N_1(t) = N_0 e^{-\lambda_t t} $$

  $$ N_{2,a}(t) = \frac{\lambda_a}{\lambda_t} N_0 \left( 1 - e^{-\lambda_t t} \right) $$

  $$ N_{2,b}(t) = \frac{\lambda_b}{\lambda_t} N_0 \left( 1 - e^{-\lambda_t t} \right) $$
Radioactive filiation (3)

Measurement of the 2 $\gamma$ rays of $^{56}$Mn
Radioactive filiation (4)

• We suppose now $X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} X_3$

• The number of $X_1$ (« parent ») decreases following an exponential equation →

$$\frac{dN_1}{dt} = -\lambda_1 N_1 \rightarrow N_1(t) = N_1(0)e^{-\lambda_1 t}$$

• The number of $X_2$ (« daughter ») increases due to disintegration of $X_1$ and disintegrates with the disintegration constant $\lambda_2$ →

$$\frac{dN_2}{dt} = -\lambda_2 N_2 + \lambda_1 N_1 = -\lambda_2 N_2 + \lambda_1 N_1(0)e^{-\lambda_1 t}$$

• The solution is→

$$N_2(t) = N_2(0)e^{-\lambda_2 t} + \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(0) \left( e^{-\lambda_1 t} - e^{-\lambda_2 t} \right)$$
Radioactive filiation (5)

- The number of $X_3$ changes as

$$\frac{dN_3}{dt} = \lambda_2 N_2$$

$$N_3(t) = N_3(0) + N_2(0) (1 - e^{-\lambda_2 t}) + N_1(0) \left( 1 - \frac{\lambda_2 e^{-\lambda_1 t} - \lambda_1 e^{-\lambda_2 t}}{\lambda_1 - \lambda_2} \right)$$

- Practically $\rightarrow$ measures of activities $A_1 = \lambda_1 N_1$ and $A_2 = \lambda_2 N_2$ $\rightarrow$

assuming $N_2(0) = N_3(0) = 0$ $\rightarrow$

$$A_1(t) = A_1(0)e^{-\lambda_1 t} \quad \text{and} \quad A_2(t) = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1(0) \left( e^{-\lambda_1 t} - e^{-\lambda_2 t} \right)$$
Equilibria (1)

• We note that $A_1(t)$ is maximum at $t = 0$ and zero at $t = \infty$ and that $A_2(t)$ is zero at $t = 0$ and $t = \infty \rightarrow A_2(t)$ has a maximum for $dA_2(t)/dt = 0 \rightarrow$

$$
\frac{d(A_2)}{dt} = 0 = -\lambda_1 e^{-\lambda_1 t_m} + \lambda_2 e^{\lambda_2 t_m}
$$

$$
t_m = \frac{\ln \frac{\lambda_2}{\lambda_1}}{\lambda_2 - \lambda_1}
$$

• This maximum happens when the activities of parent and daughter are equal $\rightarrow A_1(t_m) = A_2(t_m)$

$$
e^{-\lambda_1 t_m} = \frac{\lambda_2}{\lambda_2 - \lambda_1} \left( e^{-\lambda_1 t_m} - e^{\lambda_2 t_m} \right)
$$

$$
t_m = \frac{\ln \frac{\lambda_2}{\lambda_1}}{\lambda_2 - \lambda_1}
$$
Equilibria (2)

- At $t_m \rightarrow$ we have «ideal equilibrium»
- The ratio of activities of $X_2$ and $X_1$ is $\Rightarrow$

\[
\frac{A_2(t)}{A_1(t)} = \frac{\lambda_2}{\lambda_2 - \lambda_1} \left(1 - e^{-(\lambda_2-\lambda_1)t}\right)
\]

- For $t < t_m \rightarrow$ always $A_1 > A_2$
- For $t > t_m \rightarrow$ always $A_1 < A_2$
- The specific relation between parent and daughter depends on the relative values of their disintegration constants $\Rightarrow$ 3 cases $\Rightarrow$
  1. $\lambda_2 < \lambda_1$
  2. $\lambda_2 > \lambda_1$
  3. $\lambda_2 \gg \lambda_1$
Non-equilibrium: $\lambda_2 < \lambda_1$

- $X_1$ isotopes disintegrate faster than filiation products $X_2 \rightarrow$ the ratio of activities increases without limit

![Graph showing the ratio $A_2(t)/A_1(t)$ over time $t/T_{1/2}^{(1)}$ with $\lambda_2 = 0.1\lambda_1$]
Example with $\lambda_2 < \lambda_1$

- Disintegration of metastable tellurium $\rightarrow$

  \[
  ^{131m}\text{Te} \quad T_{1/2} = 30h \quad \rightarrow \quad ^{131}\text{I} \quad T_{1/2} = 193h \quad \rightarrow \quad ^{131}\text{Xe}
  \]

- We have thus $\rightarrow \lambda_1 = 2.31 \times 10^{-2} \text{ h}^{-1}$ and $\lambda_2 = 3.59 \times 10^{-3} \text{ h}^{-1}$
Transient equilibrium: $\lambda_2 > \lambda_1$

- The activities ratio increases as a function of time and reaches a constant value $\rightarrow$ for $t \rightarrow \infty$:

$$\frac{A_2(t)}{A_1(t)} \sim \frac{\lambda_2}{\lambda_2 - \lambda_1}$$

- The daughter activity decreases at the same rate as that of the parent $\rightarrow$ this equilibrium is called transient equilibrium
Secular equilibrium: $\lambda_2 \gg \lambda_1$

- The activities ratio increases as a function of the time and reaches 1 pour $t \to \infty$:
  \[ \frac{A_2(t)}{A_1(t)} \approx 1 \]

- The parent and daughter activities become equal → secular equilibrium

- Example → disintegration of radium →
  
  \[
  ^{226}_{88}\text{Ra} \xrightarrow{T_{1/2} = 1602 \text{ an}} \hspace{1cm} ^{222}_{86}\text{Rn} \xrightarrow{T_{1/2} = 3.8 \text{ j}} ^{218}_{84}\text{Po}
  \]

- We have → $\lambda_1 = 1.18 \times 10^{-6} \text{ j}^{-1}$ and $\lambda_2 = 1.81 \times 10^{-1} \text{ j}^{-1}$
Bateman equations

- We consider \( X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} X_3 \rightarrow \ldots \)
- Generalization of previous equations \( \rightarrow \)

\[
dN_i = \lambda_{i-1} N_{i-1} \, dt - \lambda_i N_i \, dt
\]

- General solution for \( N_0 \) nuclei of type 1 and none for other types is given by the Bateman equations \( \rightarrow \)

\[
A_n = N_0 \sum_{i=1}^{n} c_i e^{-\lambda_i t}
\]

where \( c_m = \frac{\prod_{i=1}^{n} \lambda_i}{\prod_{i=1}^{m} (\lambda_i - \lambda_m)} \)

- ' means that term with \( i = m \) is omitted
- Secular equilibrium is possible \( (A_1 = A_2 = \ldots) \)
ORIGEN

- Bateman formula can be implemented easily in computer code → but if $\lambda_p \approx \lambda_j$ for some isotope pair → cancellation can lead to computational errors → other methods such as numerical integration or the matrix exponential method are in use
- ORIGEN (Oak Ridge Isotope GENeration in SCALE) code calculates the decay chains by the matrix exponential method
- ORIGEN was developed for the Nuclear Regulatory Commission and the Department of Energy (USA) → easy-to-use standardized method of isotope depletion/decay analysis for spent fuel, fissile material, and radioactive material
- It can be used to solve for spent fuel characterization, isotopic inventory, radiation source terms, and decay heat
Application: Production of radioelements (1)

• Stable element placed into a reactor or an accelerator (such a cyclotron) → nucleus captures a neutron or a charged particle → possible production of a radioelement

• The production rate $R$ (unit: $m^{-3}s^{-1}$) depends on the target atom density $N_0$ (unit: $m^{-3}$), on the density of current $J$ of the beam (unit: $m^{-2}s^{-1}$) and on the reaction cross section $\sigma$ unit: $m^2$) → $R = N_0\sigma J$

• As $\sigma \approx 10^{-24}$ cm$^2$ and $J \approx 10^{14}$ cm$^{-2}s^{-1}$ → the probability to convert a stable particle is $\approx 10^{-10}$ s$^{-1}$ → the number of converted particle is small → the number of target nuclei is constant → $R$ is constant
Application: Production of radioelements (2)

- We consider: \( N_0 \overset{R}{\rightarrow} N_1 \overset{\lambda_1}{\rightarrow} N_2 \)
- We have thus:

\[
dN_1 = R dt - \lambda_1 N_1 dt \rightarrow N_1(t) = \frac{R}{\lambda_1} (1 - e^{-\lambda_1 t}) \rightarrow A_1 = R (1 - e^{-\lambda_1 t})
\]

- If the irradiation time \( T \ll T_{1/2} \rightarrow A_1 = R \lambda_1 T \)
- If the irradiation time \( T \gg T_{1/2} \rightarrow A_1 = R \) (secular equilibrium)

Production \(^{61}\text{Cu} \, (T_{1/2} = 3.4 \text{ h})\) due to bombardment of \(^{61}\text{Ni}\) by deuteron → use for positron emission tomography (PET)
Application: Carbon-14 dating

Radiocarbon (14-C) is constantly being created in the atmosphere by the interaction of cosmic rays with atmosphere:

\[
\frac{14}{7}N + \frac{1}{0}n \rightarrow \frac{14}{6}C + \frac{1}{1}H
\]

The resulting radiocarbon is incorporated into plants by photosynthesis → into animals by eating the plants

During its life → a plant or animal is exchanging carbon with its surroundings → same proportion of 14-C as the biosphere

When dying → no more 14-C acquiring and decay of 14-C in the organic sample

The measurement of the ratio \( \frac{^{14}C}{C_{\text{total}}} \) gives the sample age

As \( T_{\frac{1}{2}} = 5730 \pm 40 \) years → dating is possible for age between a few hundred years and about 50,000 years
Quantum description of radioactive decays (1)

- Solving the Schrödinger equation for various time-independent potentials $\rightarrow$ energy levels are *stationary* states
- A system in a particular stationary state will remain in that state for all times $\rightarrow$ no transition $\rightarrow$ no decay
- If we assume one state being the mixture of two (or more) states $\rightarrow \psi = c_1\psi_1 + c_2\psi_2$ $\rightarrow$ probability $|c_1|^2$ to be found in 1 and $|c_2|^2$ to be found in 2 $\rightarrow$ for time-independent potential $\rightarrow c_1$ and $c_2$ are independent on time $\rightarrow \neq$ with observation
- We are forced to abandon the notion of pure states with well-defined wave-functions $\rightarrow$ difficult interpretation of nuclear structure
Quantum description of radioactive decays (2)

• We assume a potential of the form $V + V'$ where $V$ is the nuclear potential that gives stationary states and $V'$ is a very weak additional potential that causes the transition between the states.

• Neglecting $V'$ we obtain the static nuclear wave functions.

• These wave functions are used to calculate the transition probability between the « stationary states » under the influence of $V'$ this transition probability is $\lambda$.

• Fermi Golden Rule →

$$
\lambda = \frac{2\pi}{\hbar} |V'_{fi}|^2 \rho(E_f) \text{ where } V'_{fi} = \int \psi^*_f V' \psi_i d\mathbf{r}
$$
Quantum description of radioactive decays (3)

- The potential $V'$ depends on the particular type of transition which is considered.

- The transition probability is thus influenced by the density of final states $\rho(E_f)$ within an energy interval $dE_f$ the number of states accessible to the system is $dn_f = \rho(E_f)dE_f$ the transition probability is larger if the number of final states accessible for the decay is large.
Width of the states (1)

- Solving the Schrödinger equation for time-independent potential $V \rightarrow$ stationary states of the nucleus $\psi_i(r) \rightarrow$ the time-dependent wave function $\Psi_i(r,t)$ is

$$\Psi_i(r, t) = \psi_i(r) e^{-iE_i t/\hbar}$$

where $E_i$ is the energy of the state

- The probability of finding the system in the state is $|\Psi_i(r,t)|^2 \rightarrow$ independent on time for stationary state

- To be consistent with the radioactive decay law we have to introduce the decrease with time $\exp(-t/\tau_i)$ with $\tau_i = 1/\lambda_i \rightarrow$

$$|\Psi_i(t)|^2 = |\Psi_i(t = 0)|^2 e^{-t/\tau_i}$$

- The expression of $\Psi_i(r,t)$ becomes $\rightarrow$

$$\Psi_i(r, t) = \psi_i(r) e^{-iE_i t/\hbar} e^{-t/2\tau_i}$$
Width of the states (2)

• The resonant state (non-stationary state) can be written →

\[ \Psi_i(r, t) = \psi_i(r) \exp \left( \frac{-it}{\hbar} \left( E_i - \frac{i\lambda \hbar}{2} \right) \right) \]

• Complex energy: \( E_i - i\lambda \hbar/2 \)

• Alternatively the state has no a definite energy → the wave function is a superposition of components having different energies (with \( A(E) \) the probability amplitude to find the state at energy \( E \)) →

\[ \exp \left( \frac{-it}{\hbar} \left( E_i - \frac{i\lambda \hbar}{2} \right) \right) = \int A(E) \exp \left( \frac{itE}{\hbar} \right) \]
Width of the states (3)

• The probability for finding the state at energy $E_i$ is given by the absolute square of the amplitude $\rightarrow$

$$|A(E)|^2 = \frac{1}{4\pi^2} \frac{1}{(E - E_i)^2 + \Gamma_i^2/4}$$

• The shape of such a distribution is Lorentzian and $\Gamma_i = \hbar/\tau_i$ is the width of the state $i \rightarrow$ full width at half maximum (FWHM) of such a distribution

• The width is the measure of our inability to determine precisely the energy of the state $\rightarrow$ it is not a question of instrumental uncertainty
Width of the states (4)

• Another way to understand it $\Delta E \Delta t \geq \hbar/2 \rightarrow$ if $\Delta t \rightarrow 0$ we can precisely determine the energy of the state because $\Delta E = 0$

• If the state lives on an average for a time $\tau \rightarrow$ we cannot determine its energy except to within an uncertainty of $\Delta E \sim \hbar/\tau$

• It is always possible to speak of transitions between distinct levels because the widths $\Gamma$ of nuclear levels (typically $\Gamma < 10^{-10}$ MeV) is small compared with their energy spacing ($\sim 10^{-3}$ MeV)
Types of radioactive decay

• There are 3 principal types of decay: $\alpha$-, $\beta$- and $\gamma$-decay processes

• In $\alpha$ and $\beta$ processes $\rightarrow$ an unstable nucleus emits an $\alpha$ or a $\beta$ particle as it tries to become a more stable nucleus

• In $\gamma$-decay process $\rightarrow$ an excited state decays toward the ground state without changing the nuclear species
\( \alpha \)-decay

- The nucleus emit an \( \alpha \) particle i.e. a nucleus of helium: \( ^4_2\text{He}_2 \)
- The \( ^4\text{He} \) nucleus is a tightly bound system → the kinetic energy released is maximized
- The decay process is

\[
\frac{A}{Z}X_N \rightarrow \frac{A-4}{Z-2}X'_{N-2} + \frac{4}{2}\text{He}_2
\]

- The number of protons and neutrons are separately conserved
- Example (with \( T_{\frac{1}{2}} = 1600 \) years and \( E_{\text{kin}}(\alpha) = 4.8 \) MeV):

\[
^\text{226}_{88}\text{Ra}_{138} \rightarrow ^\text{222}_{86}\text{Rn}_{136} + \frac{4}{2}\text{He}_2
\]
β-decay (1)

- The nucleus can correct a proton or a neutron excess by directly converting a proton into a neutron or a neutron into a proton
- Three possible ways for this process → each of them involves another charged particle to conserve electric charge and a (anti-)neutrino to conserve the electronic lepton number
  - $\beta^-$ decay: $n \rightarrow p + e^- + \bar{\nu}$
  - $\beta^+$ decay: $p \rightarrow n + e^+ + \nu$
  - electron capture ($\epsilon$): $p + e^- \rightarrow n + \nu$
- For the electron capture → an atomic electron too close to the nucleus is swallowed
- In all cases where $\beta^+$-decay is allowed energetically → electron capture is allowed (competing process) but not the opposite
In $\beta^-$- and $\beta^+$-decays → a particle is created (electron - « negatron » - and positron, respectively) → they did not exist inside the nucleus before the decay → in contrast with $\alpha$-decay in which the emitted nucleon were inside the nucleus before the decay

We also note that the emitted $\beta^-$ and $\beta^+$ show an energy spectrum → the total energy is shared between the 3 bodies

In electron capture → the neutrino energy is fixed

Examples:

$$\frac{131}{53}I_{78} \rightarrow \frac{131}{54}Xe_{77} + \beta^- + \bar{\nu}$$

$$\frac{25}{13}Al_{12} \rightarrow \frac{25}{12}Mg_{13} + \beta^+ + \nu$$

$$\frac{54}{25}Mn_{29} + e^- \rightarrow \frac{54}{24}Cr_{30} + \nu$$

In these processes → $Z$ and $N$ each change by one unit but $Z + N = \text{constant}$
\( \gamma \)-decay

- An excited state decays to a lower excited state or possibly the ground state by emission of a photon of \( \gamma \) radiation with energy equal to the difference between the nuclear states (less a usually negligible correction due to the recoil)
- \( \gamma \)-decay is observed in all nuclei that have excited bound states \((A > 5)\) and generally follows an \( \alpha \)- or \( \beta \)-decay (daughter nucleus in an excited state)
- \( T_{1/2} \) is generally small \(< 10^{-9} \text{ s}\) but sometimes can be \( \gg \) (isomeric or metastable states) \( \rightarrow \) no clear distinction between states which are isomeric or not \( \rightarrow 10^{-6} \text{ s is isomeric and } 10^{-12} \text{ s is not } \rightarrow \) in between: fuzzy
- Competing process \( \rightarrow \) internal conversion: the energy is transferred to an atomic electron (no \( Z \) and \( N \) changes for the nucleus but the atom becomes ionized)
Other processes

• **Spontaneous fission**: some nuclei spontaneously fission → a heavy nucleus with an excess of neutrons splits roughly in half into two lighter nuclei → the final nuclei are not rigidly determined but are statistically distributed over the entire range of medium-weight nuclei + neutrons + γ-rays +... (examples: $^{256}\text{Fm}$ with $T_{1/2} = 2.6$ h or $^{254}\text{Cf}$ with $T_{1/2} = 60.5$ days)

• **Nucleon emission**: As we move further and further from the stability valley → the energy differences between neighboring isobars ↑ can be larger than the nucleon binding energy ($\approx 8$ MeV) → radioactive decay by nucleon emission → occurs most frequently in fission products having a large neutron excess → delayed neutrons → very important in the control of nuclear power plants (ex: $^{138}\text{Xe} \rightarrow ^{137}\text{Xe} + n$ or $^{73}\text{Br} \rightarrow ^{72}\text{Se} + p$)
Branching ratios (1)

• Often it exists several possible decay processes in competing mode → the decay schemes may be very complicated
• We specify the relative intensities of the competing modes by their branching ratios → example:

$^{226}$Ac decays by $\alpha$ emission (0.006%), $\beta^-$ emission (83%) and $\epsilon$ (17%) → then ...
Branching ratios (2)

- Frequently → branching ratio is specified by giving the partial decay constant of partial half-life.

- For $^{226}$Ac →

  \[
  \lambda_t = \frac{0.693}{29h} = 0.024 h^{-1} = 6.6 \times 10^{-6} s^{-1}
  \]

  \[
  \lambda_{\beta} = 0.83\lambda_t = 5.5 \times 10^{-6} s^{-1}
  \]

  \[
  \lambda_{\epsilon} = 0.17\lambda_t = 1.1 \times 10^{-6} s^{-1}
  \]

  \[
  \lambda_{\alpha} = 6 \times 10^{-5}\lambda_t = 4 \times 10^{-10} s^{-1}
  \]

- Partial half-life ($T_{\frac{1}{2},i} = 0.693/\lambda_i$) are convenient to represent branching ratio → but only total half-life has a sense.
interactive chart of nuclides

• All information → interactive chart of nuclides: http://www.nndc.bnl.gov/chart
Summary of various decay processes