Radioactive decay and the Bateman equation

Introduction to Nuclear Science

Simon Fraser University
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Outline

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Natural radioactivity

- Natural radioactivity has been discovered by Henri Becquerel and Marie Skłodowska-Curie in late 1890s.
- Radioactive polonium and radium elements were isolated by Marie and Pierre Curie in 1898.
- The process involves spontaneous disintegration of the parent element and a formation of a daughter element.
- A number of long-lifetime processes were identified since then:
  - α-decay : emission of ^{4}He
  - β^--decay: emission of an electron and electron anti neutrino
  - electron capture: capture of an electron from an atomic orbit by a proton
  - β^+-decay: emission of a positron and electron neutrino
  - heavy fragment emission: for example ^{12}C or ^{16}O
  - fission: split of a nucleus into two fragments of comparable mass and charge

Natural radioactivity

- To quantify the decay process several measures can be introduced:
  - Activity A: number of disintegration per second
  - half-life $T_{\frac{1}{2}}$: time after which the number of radioactive nuclei in a sample is reduced to half of its initial value
  - lifetime τ : time after which the number of radioactive nuclei in a sample is reduced by a factor of $e \approx 2.718$ of its initial value.

Units

- Lifetimes and half-lives are measured in units of time.
- Nuclear lifetimes span broad range from 10^{-20} s up to infinity (for stable nuclei).
- The SI unit for activity is 1 Becquerel, abbreviated as [Bq],

$$1\ [\text{Bq}] = 1\ [\text{dps}]\ (\text{decay/disintegration per second}). \tag{1}$$

- An often used non-SI unit is 1 Curie, abbreviated [Ci].

$$1\ [\text{Ci}] = 3.7 \times 10^{10}\ [\text{Bq}] \tag{2}$$

- 1 [Ci] corresponds to activity of 1 g of Radium and is a sizable unit.
- Typical environmental levels of radioactivity are pico-nano Curie (0.01-10 [Bq]), research calibration sources are typically of micro Curie (10 [kBq]) activity, a reactor upon a shutdown have activity in the range of giga Curie (10^9 Ci or 10^{19} [Bq]).

Natural radioactivity

Some naturally occurring long-lived radioactive isotopes

Nuclide	Half-life [years]	Natural abundance
$^{40}_{19}$K	1.28×10^{9}	0.01%
$^{87}_{37}$Rb	4.8×10^{10}	27.8%
$^{113}_{48}$Cd	9×10^{15}	12.2%
$^{115}_{49}$In	5.5×10^{14}	95.7%
$^{128}_{52}$Te	7.7×10^{24}	31.7%
$^{130}_{52}$Te	2.7×10^{21}	33.8%
$^{138}_{57}$La	1.1×10^{11}	0.09%
$^{144}_{60}$Nd	2.3×10^{15}	23.8%
$^{147}_{62}$Sm	1.1×10^{11}	15%
$^{148}_{62}$Sm	7×10^{15}	11.3%

Nuclear decay

- One can show experimentally that the sample activity A is proportional to the number N of nuclei in the sample (decay is the first-order reaction).
- Denoting the proportionality constant by λ and calling it the decay rate one obtains

$$A = \lambda N$$

- Activity is the number of disintegration per second,

$$A\Delta t = N(t) - N(t + \Delta t) = -(N(t + \Delta t) - N(t))$$
$$A = -\frac{N(t + \Delta t) - N(t)}{\Delta t} = -\frac{dN}{dt}$$

- Above equations when combined give

$$-\frac{dN}{dt} = \lambda N$$
$$N(t) = N(0)\exp(-\lambda t)$$

Nuclear decay

- Defining lifetime as

$$\tau = \frac{1}{\lambda}$$

the nuclear decay law can be written as

$$N(t) = N(0) \exp\left(-\frac{t}{\tau}\right)$$

- It is easy to note that after time $t = \tau$ the number of radioactive nuclei in the samples is reduced by the factor of e

$$N(\tau) = N(0) \exp\left(-\frac{\tau}{\tau}\right) = N(0) \exp^{-1} = \frac{1}{e} N(0)$$

Nuclear decay

- The half-life is

$$N(t = T_{\frac{1}{2}}) = N(0)\exp\left(-\lambda T_{\frac{1}{2}}\right) = \frac{1}{2}N(0)$$

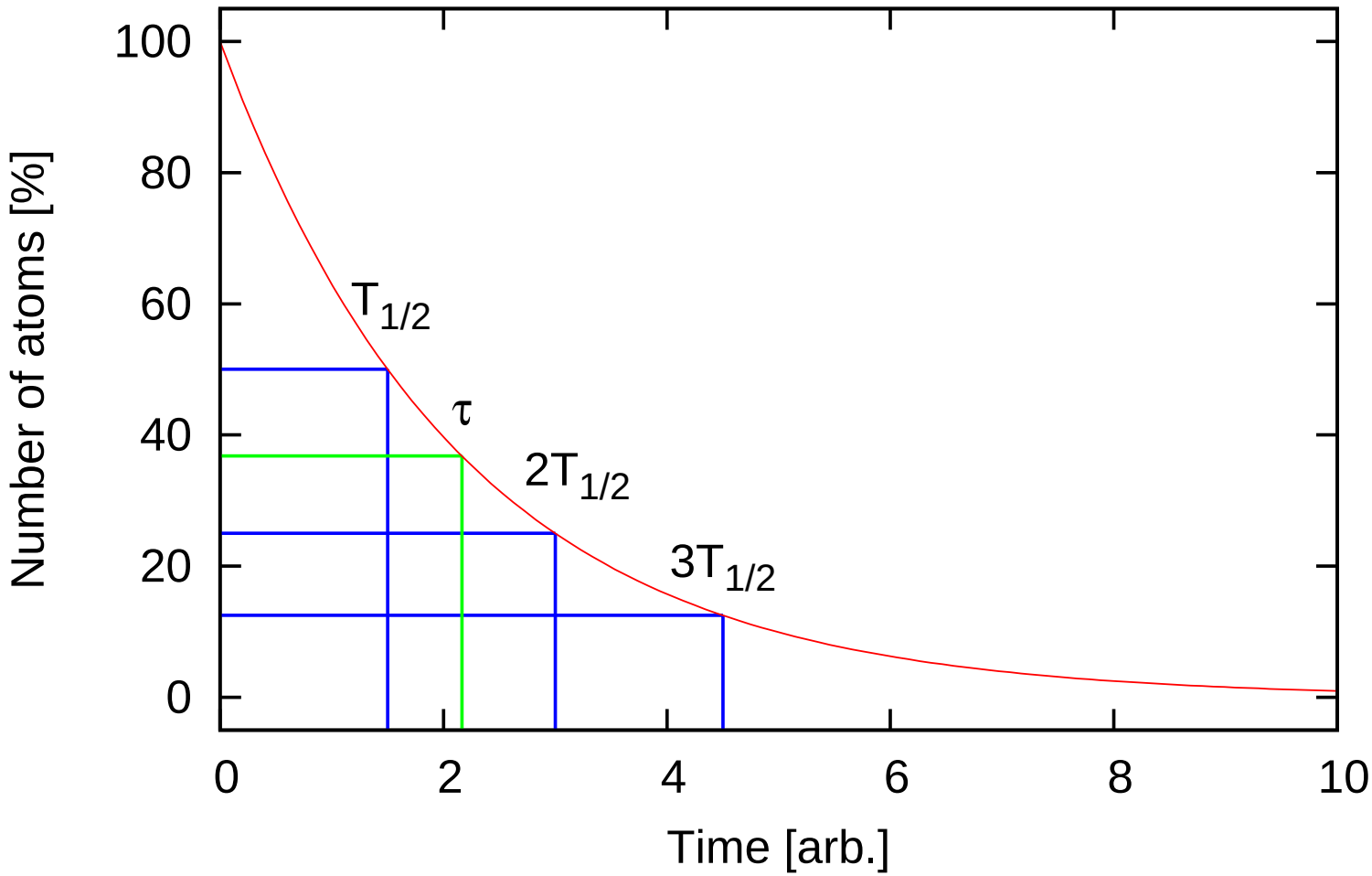
$$\exp\left(-\lambda T_{\frac{1}{2}}\right) = \frac{1}{2}$$

$$-\lambda T_{\frac{1}{2}} = \ln\left(\frac{1}{2}\right) = -\ln(2)$$

$$T_{\frac{1}{2}} = \frac{\ln(2)}{\lambda} = \ln(2)\tau = 0.693\tau$$

Nuclear decay

$$T_{1/2} = 1.5,\ \tau = T_{1/2}/\ln(2) = 2.16$$



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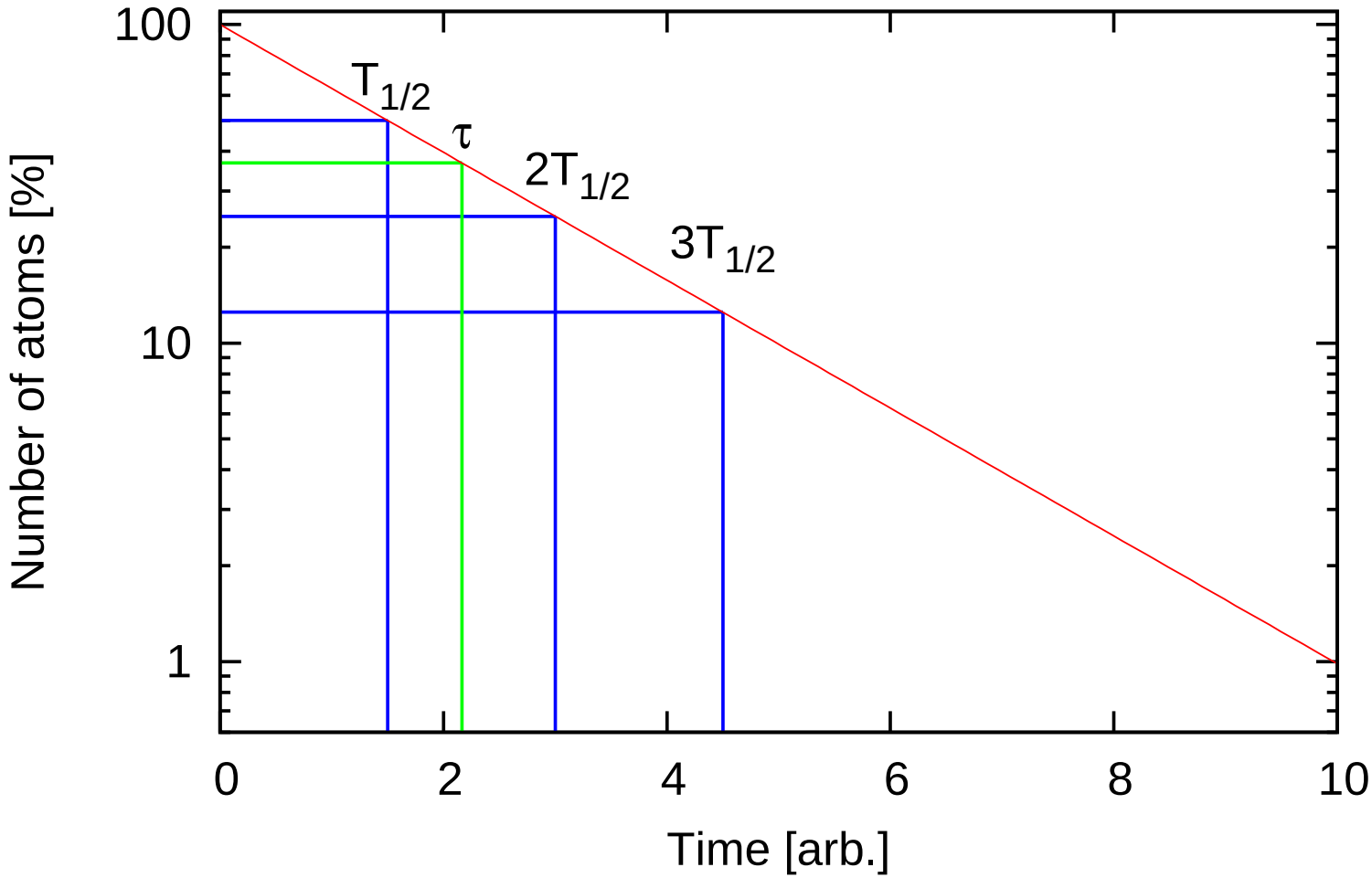

Decay rate measurements

- For large range of lifetimes measurements of the decay curves shown on the graphs above can be carried out and lifetimes/decay rates can be fitted.
- However, for lifetimes comparable or longer than the span of a human life there are no measurable changes in the activity of a sample which prohibits direct decay curve measurements.
- In these cases the decay rates are deduce from the ratio of observed activity A to the absolute number of radioactive atoms N in a sample.

$$A = \lambda N \implies \tau = \frac{1}{\lambda} = \frac{N}{A} \tag{3}$$

- The absolute number of atoms can be established based on the total mass of the sample and its isotopic composition.
- Isotopic composition can be established using mass spectroscopy.

Activity

- It should be stressed that the activity of a sample depends on its mass m (number of radioactive atoms) and the decay rate. Denoting the molar mass by μ and the Avogadro number by N_A one gets

$$A = \lambda N = \lambda \frac{m}{\mu} N_A = \frac{1}{\tau} \frac{m}{\mu} N_A \tag{4}$$

- This implies that small mass of short-lived isotopes may have the same activity as a large mass of long-lived isotopes.
- For example 1 MBq of tritium ($T_{1/2}$=12.33 y) corresponds to 5.59×10^{14} or 1.1 [nmole] of atoms or 2.78 [ng] of mass
- 1 MBq of ^{14}C ($T_{1/2}$=5730 [y]) corresponds to 2.6×10^{17} or 0.43 [μmole] of atoms or mass of 6 [μg].

Detection efficiency

- Radiation detectors are built to detect decay products.
- As such detectors respond to activity.
- In a typical experiments number of counts N_C corresponding to detection of radiation of interest in a detector is recorded per unit of time. The units of N_C are counts per second.
- This number of counts is related to the activity by the response function of a detector ϵ called efficiency

$$N_C = \epsilon A \tag{5}$$

- Efficiency depends on the type and geometry of the detector, as well as type and energy of detected radiation.

Activation analysis

- Tritium is produced in atmosphere by reaction of fast neutrons generated by cosmic rays with ^{14}N

$$n + {}^{14}N \rightarrow {}^{12}C + {}^{3}H \tag{6}$$

- Tritium is then incorporated into water and remove from atmosphere as rain and snow.
- A 50 ml sample of water typically show 1 [dpm] (disintegration per minute) associated with the β^- decay of tritium to ^{3}He. Based on that let us estimate number ratio of tritium to hydrogen in water.
- The number of tritium atoms in the sample is

$$N_{^3H} = \frac{1}{\lambda}A = \tau A = 12.33\ [\text{y}] * 1\ [1/\text{min}] = 6.5 \times 10^6 = 1.08 \times 10^{-17}\ [\text{mole}] \tag{7}$$

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- The mass of the 50 ml sample is 50 g.
- The number of water molecules in the sample is

$$N_{H_2O} = \frac{m}{\mu} = \frac{50}{18.02} = 2.77 \text{ [mole]} = 16.7 \times 10^{23} \tag{8}$$

- There are two hydrogen atoms per molecule, thus the tritium to hydrogen number ratio is

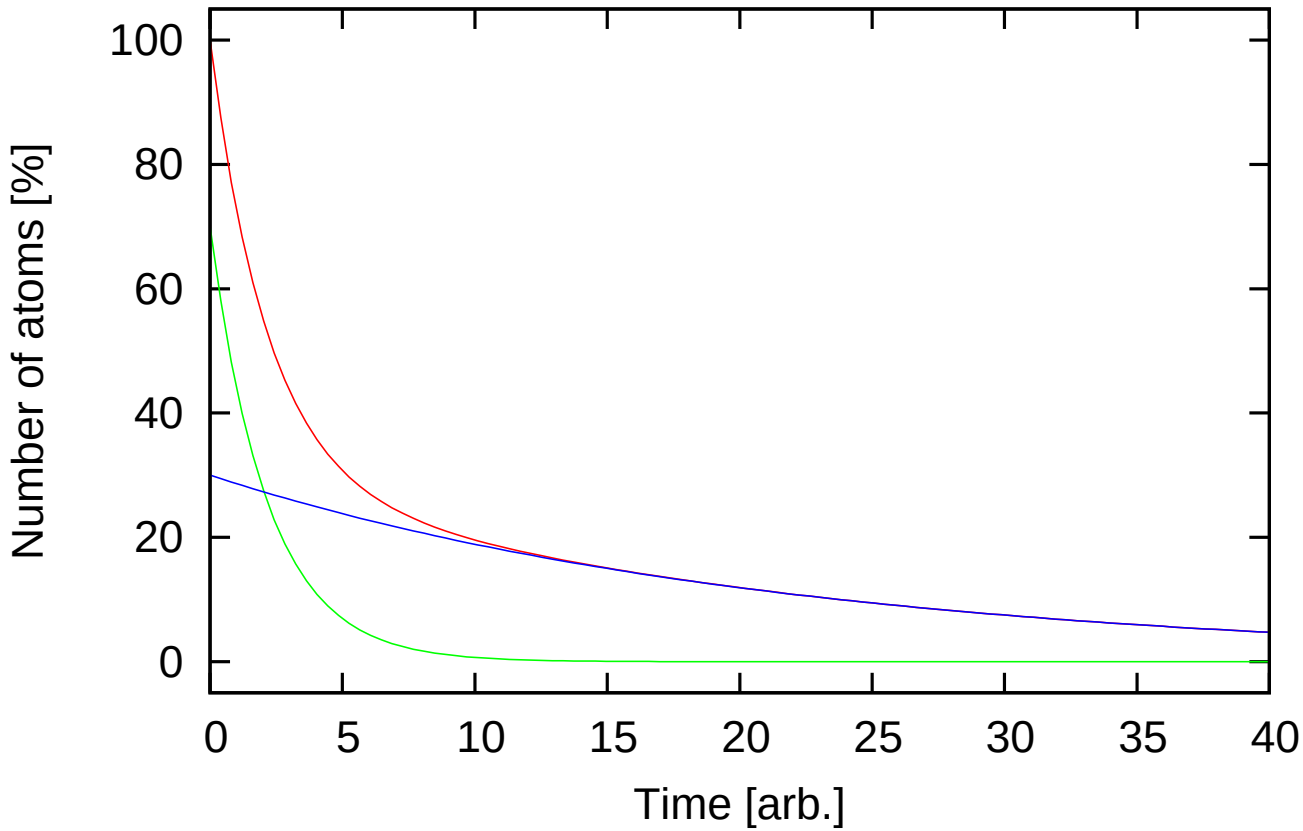
$$\frac{N_{^3H}}{N_{^1H}} = \frac{1.08 \times 10^{-17}}{5.54} = 2 \times 10^{-18} \tag{9}$$

Independent decay of radioactive mixture

- Quite often radioactive samples are mixtures of radioactivities decaying at different rates.
- If the decay products from both samples are the same (for example electrons from β^- decay) a detector will see the combined decay of the mixture.
- In such cases a special care has to be taken if lifetimes are extracted. A common procedure is to extract parameters for the longest-lived activity first, subtract it from the data, analyze the next longest lived, etc.
- Currently this is done using computer fits.
- An example for a two-component mixture is analyzed below.

Independent decay of two radioactivities

$$N(t) = 70 \times \exp\left(-\frac{t}{\ln(2)1.5}\right) + 30 \times \exp\left(-\frac{t}{\ln(2)15}\right)$$



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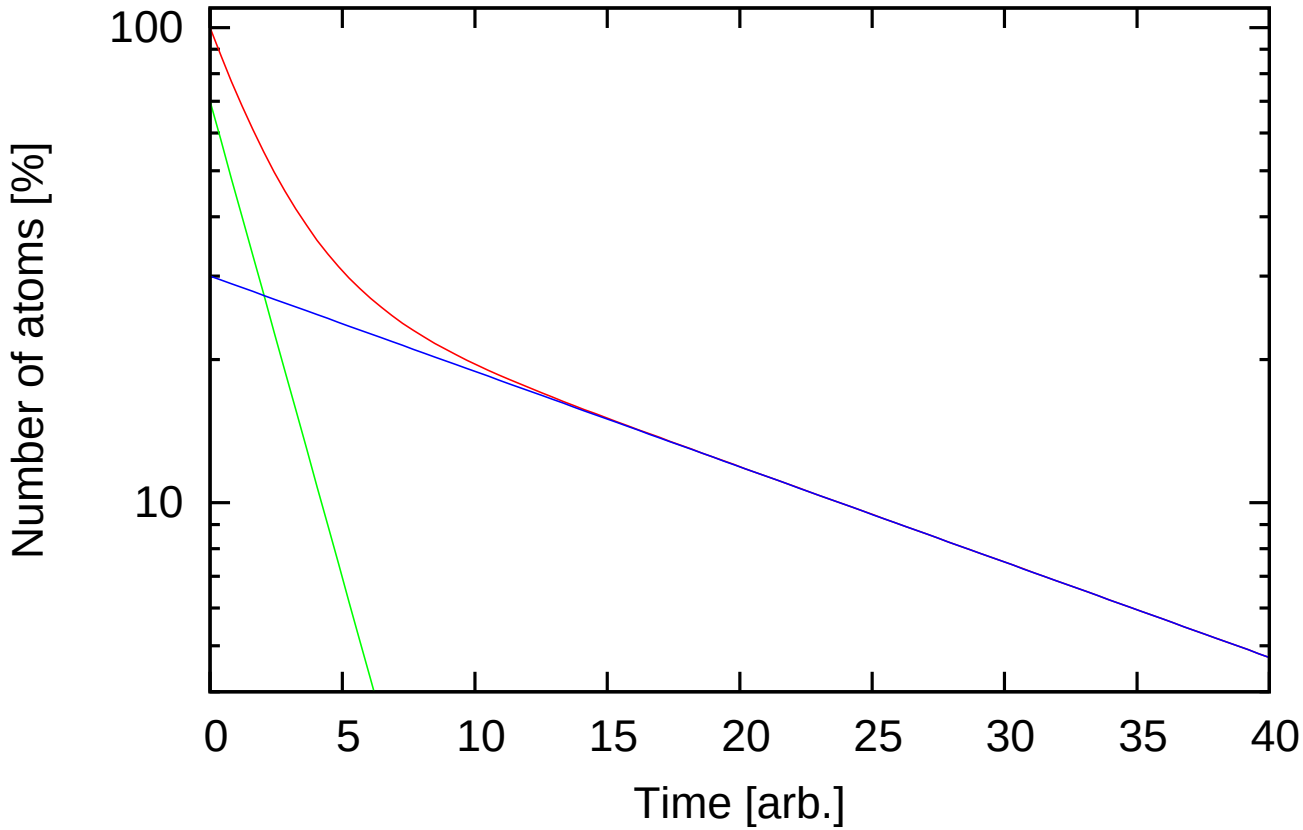
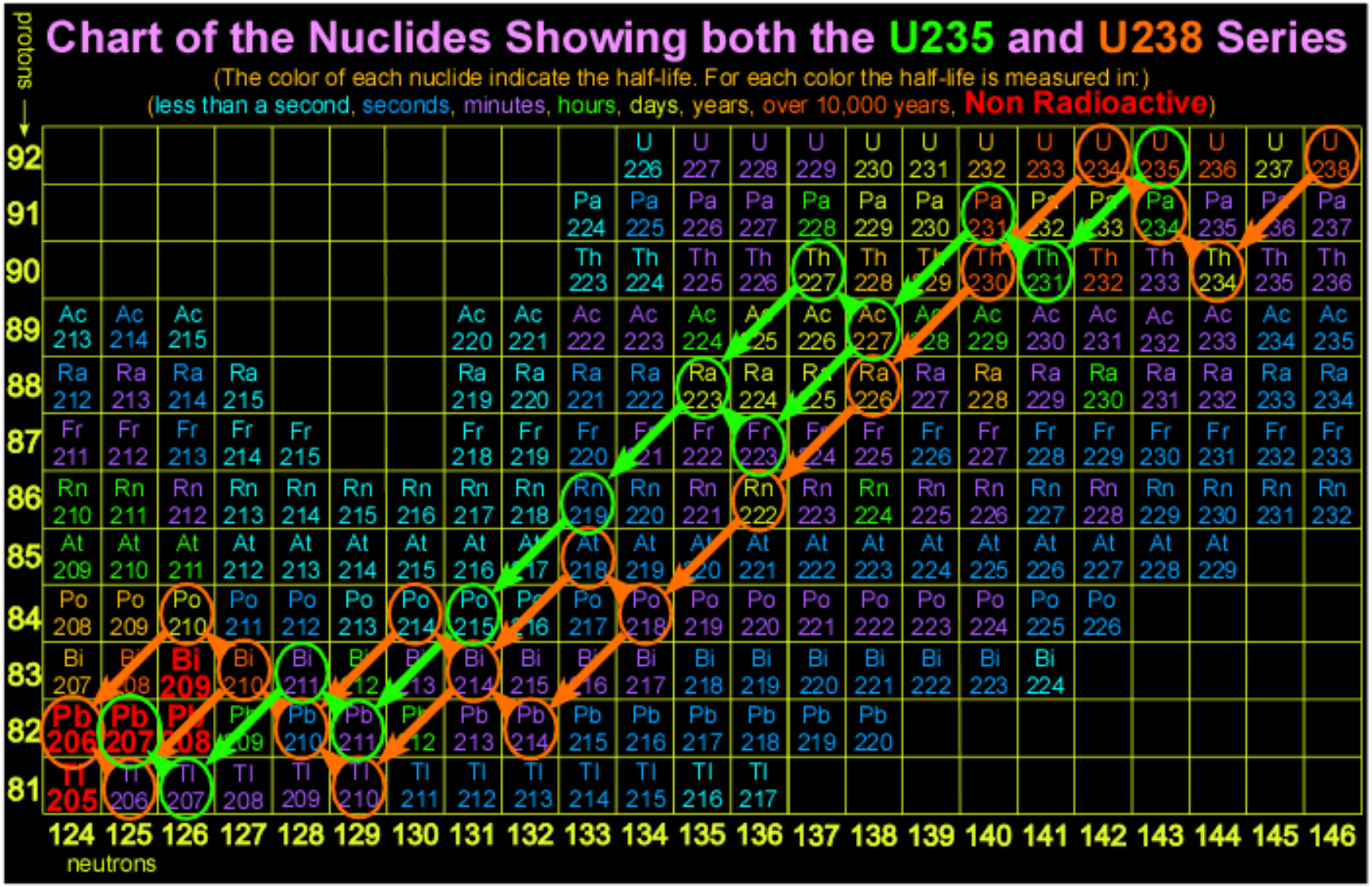


Decay chains

- Decay chains in which radioactive decay of an unstable isotope feeds radioactive decay of another unstable isotope are commonly encountered in nature and experimental nuclear science.
- For example, there are three naturally occurring, long-lived chains of α and β decays originating in the long-lived isotopes of ^{232}Th ($T_{1/2}$=14.1 Gy), ^{235}U ($T_{1/2}$=0.7 Gy) and ^{238}U ($T_{1/2}$=4.5 Gy).
- Another example is a sequence of β decay of unstable isotopes along the mass parabolas for a fixed mass number until the most stable isotope is reached.
- In case of the decay chain activities and abundances of radioactive isotopes are not independent. Rather, they are determined by the history of the decay: the decay rates and abundances in the preceding part of the chain.
- Thus the chain decay is different then independent decay of a mixture considered so far.

^{235}U and ^{238}U chains



The Bateman equation

- The Bateman equation is a mathematical model describing abundances and activities in a decay chain as a function of time, based on the decay rates and initial abundances.
- The Bateman equation is not a single equation, rather it is a method of setting up differential equations describing the chain of interest based on its known properties.
- We are going to consider the simplest case with a parent feeding single daughter.
- Then by varying the parameters such as the decay rates and relative initial abundances we will investigate the chain evolution as a function of time.

Two-decay chain

- Let us denote
  - Initial number of parent and daughter atoms as $N_1(0)$ and $N_2(0)$
  - Number of parent and daughter atoms in time as $N_1(t)$ and $N_2(t)$
  - Parent and daughter activities in time as $A_1(t)$ and $A_2(t)$
  - Parent and daughter decay rates by λ_1 and λ_2
- The equation for the time evolution of the parent is the same as for a single step decay

$$\frac{dN_1(t)}{dt} = -\lambda_1 N_1(t) \tag{10}$$

- The equation for the time evolution of the daughter, however, includes a term describing daughter decay but also daughter feeding by the parent

$$\frac{dN_2(t)}{dt} = -\lambda_2 N_2(t) + \lambda_1 N_1(t) \tag{11}$$

Two-decay chain

- The solution of Eq. 10 is

$$N_1(t) = N_1(0)\exp\left(-\lambda_1 t\right) \tag{12}$$

- Taking this into account Eq. 11 become

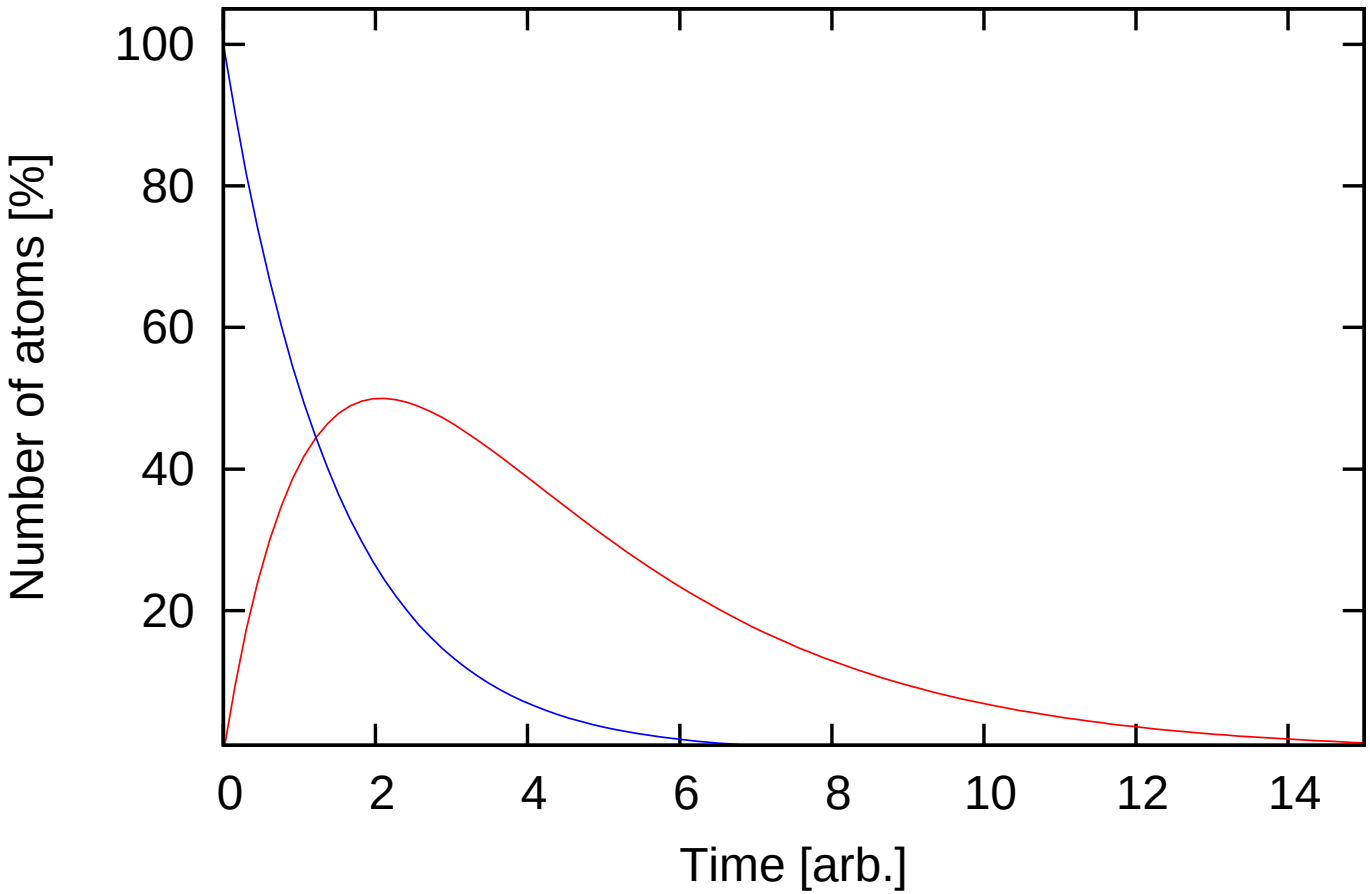
$$\frac{dN_2(t)}{dt} = -\lambda_2 N_2(t) + N_1(0)\exp\left(-\lambda_1 t\right) \tag{13}$$

- The solution for $N_2(t)$ is

$$\begin{aligned} N_2(t) &= N_2(0)\exp\left(-\lambda_2 t\right) + \\ &- N_1(0)\frac{\lambda_1}{\lambda_2 - \lambda_1}\left(\exp\left(-\lambda_2 t\right) - \exp\left(-\lambda_1 t\right)\right) \end{aligned} \tag{14}$$

Two-decay chain: abundance for a special case

Blue: parent $T_{1/2}$=1.5, $N_1(0) = 100\%$
Red: daughter $T_{1/2}$=3, $N_2(0) = 0\%$



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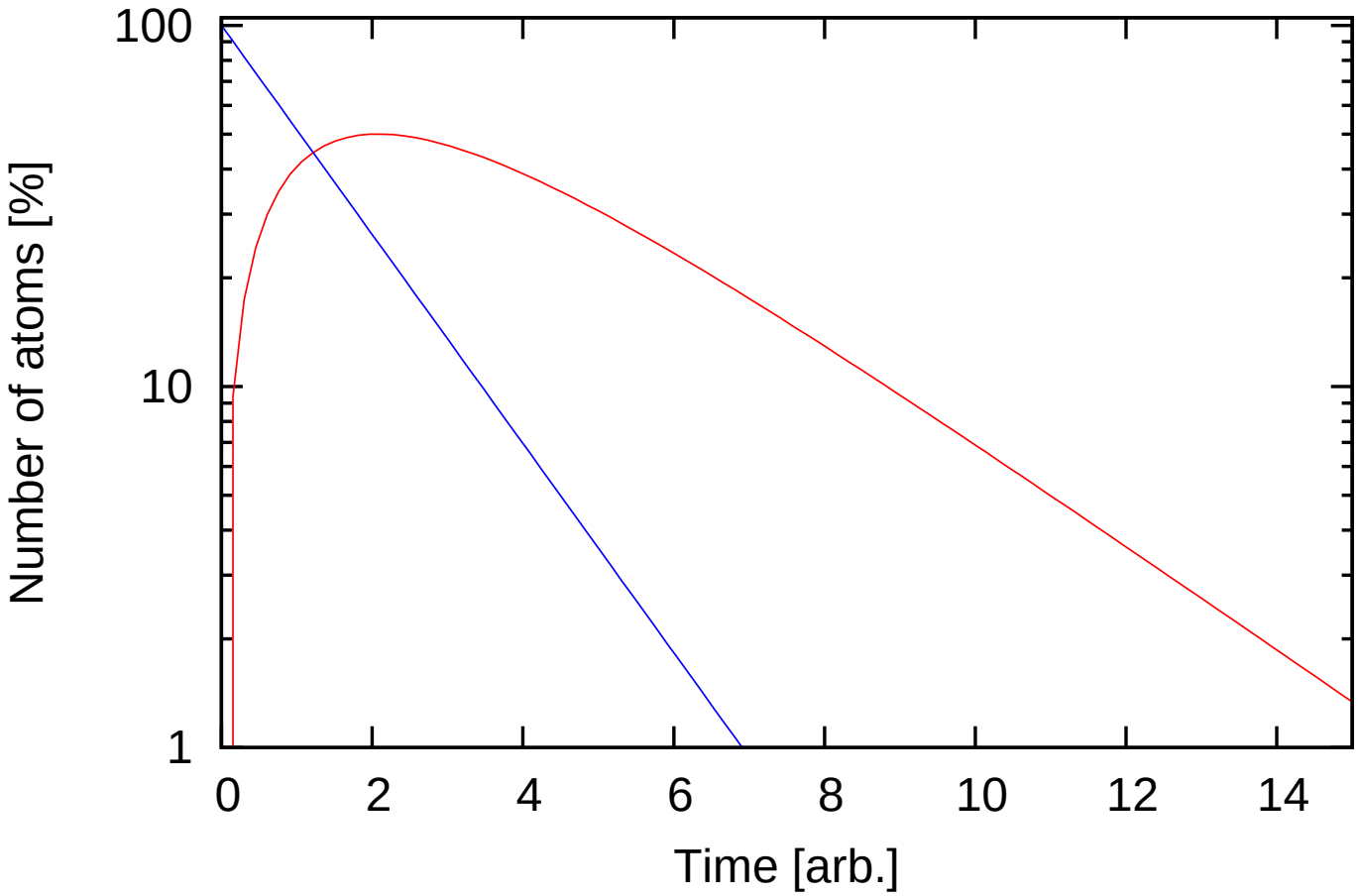

Two-decay chain: activities

- Eqs 12 and 14 define abundances
- Activities of the parent and the daughter can be calculated from

$$\begin{aligned} A_1(t) = \lambda_1 N_1(t) &= \lambda_1 N_1(0) \exp\left(-\lambda_1 t\right) \\ A_2(t) = \lambda_2 N_2(t) &= \lambda_2 N_2(0) \exp\left(-\lambda_2 t\right) + \\ &- N_1(0) \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} \left(\exp\left(-\lambda_2 t\right) - \exp\left(-\lambda_1 t\right)\right) \end{aligned} \tag{15}$$

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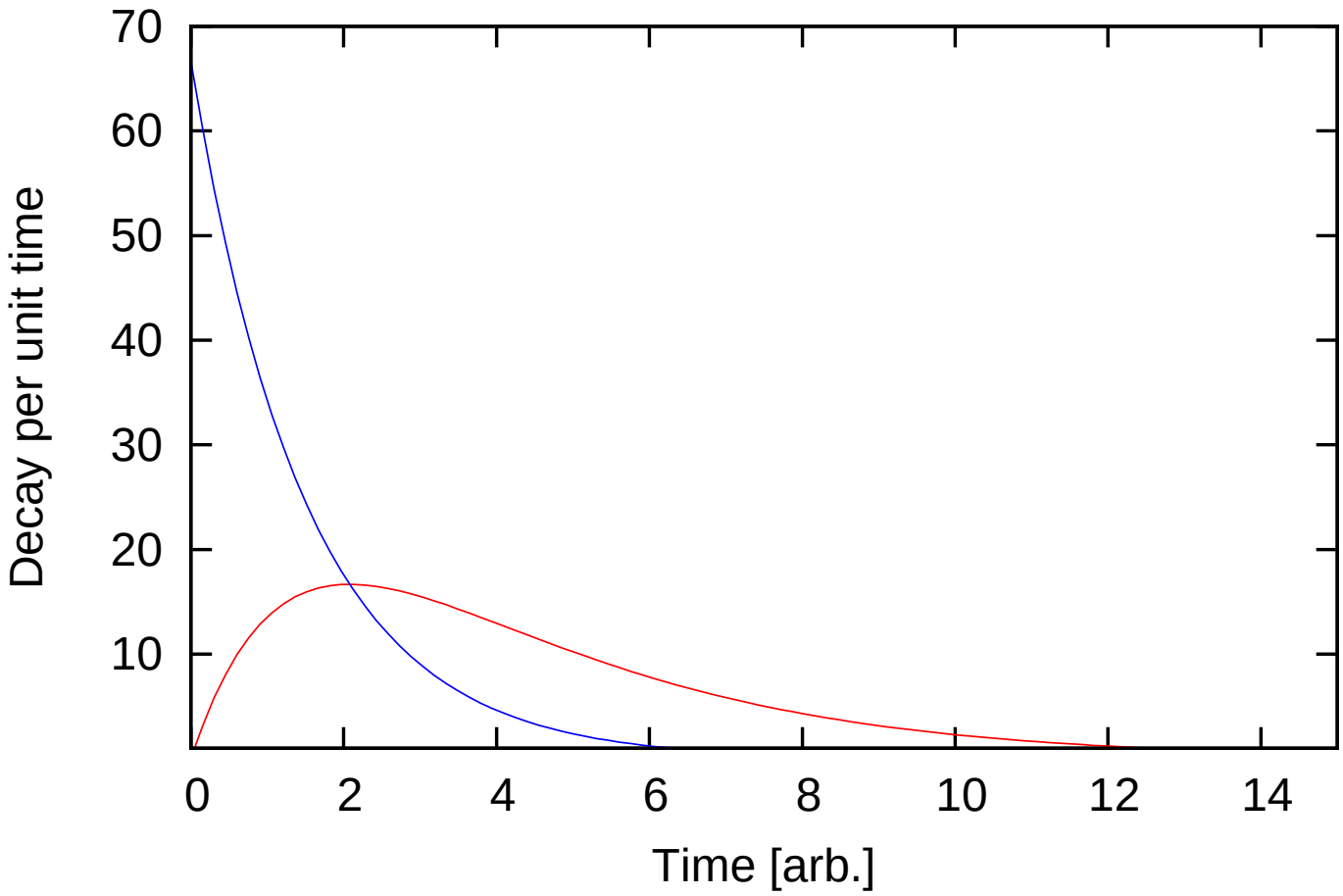

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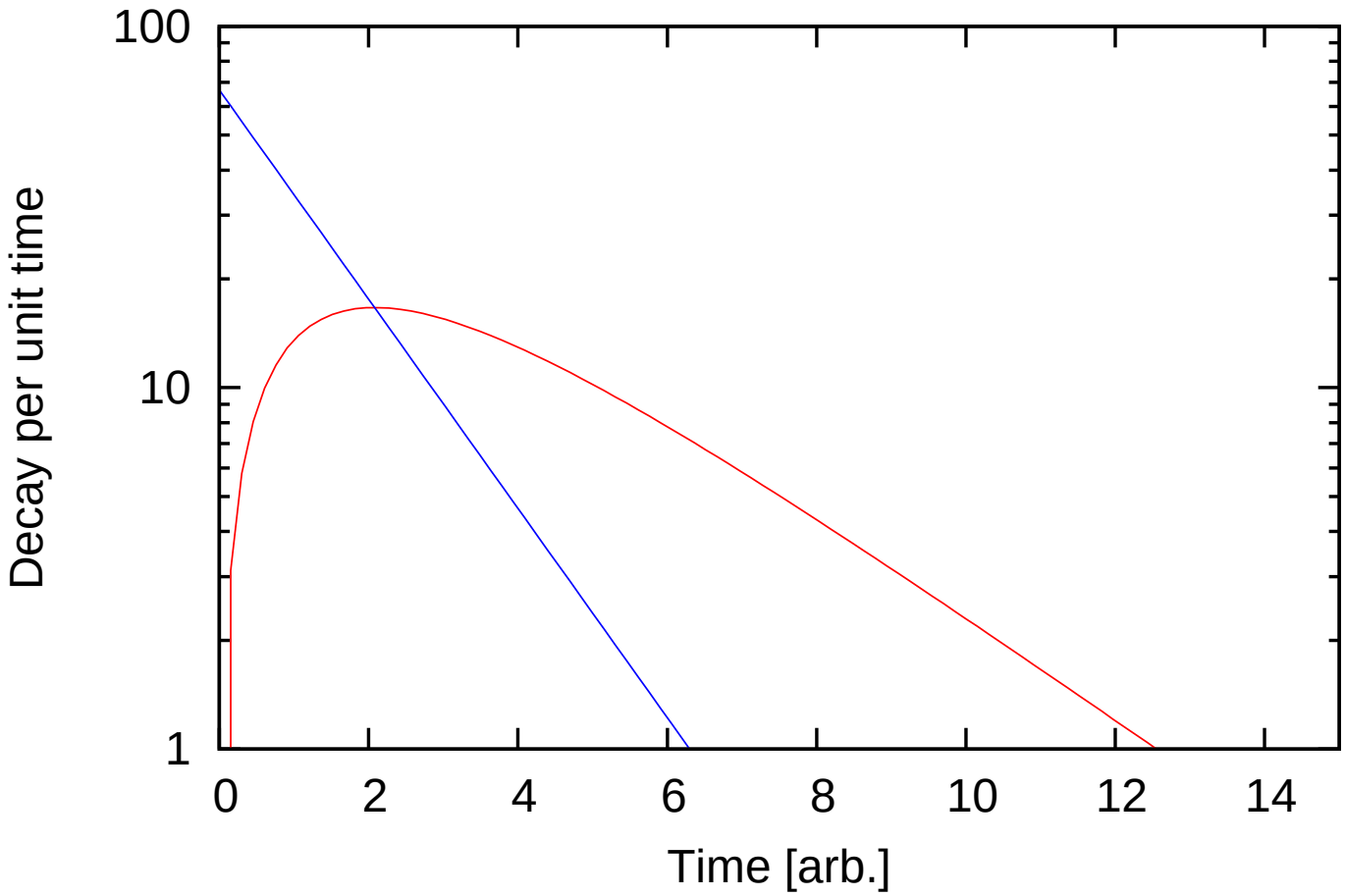
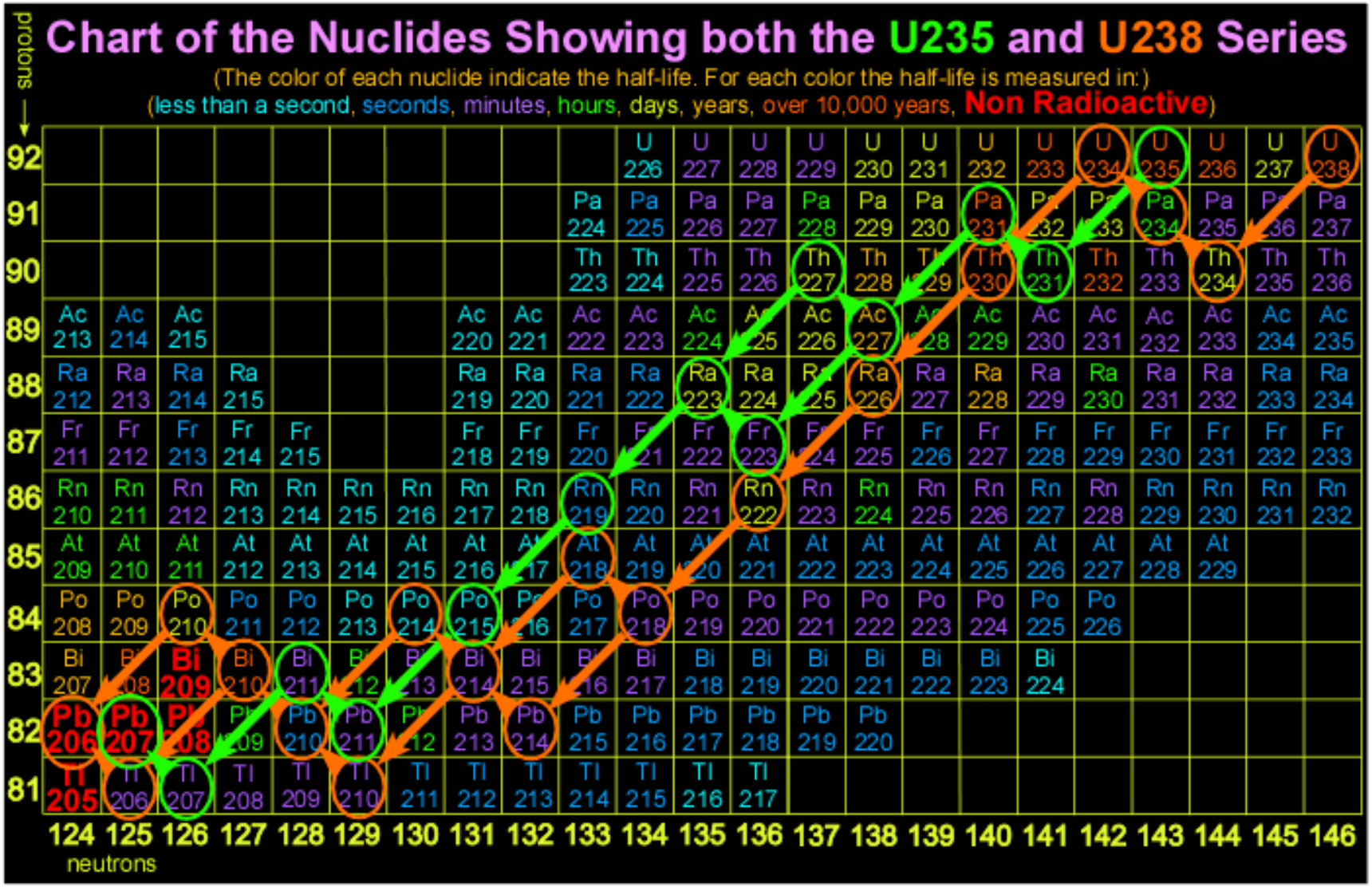


Decay branches

- Decay branches are observed when there is more than a single process for disintegration of the parent nucleus.
- For example, in the decay chain of ^{238}U ^{218}Po can α-decay to ^{214}Pb or β^- decay to ^{218}At.
- Another example is in the decay chain of ^{235}U with ^{227}Ac having an α branch to ^{223}Fr and a β^- decay branch to ^{227}Th.
- Yet another example are decays of ^{235}U and ^{238}U by spontaneous fission which is a tiny, however, existing branch as compared to the dominating α decay.

^{235}U and ^{238}U chains



Total decay rate and branching ratios

- For clarity, let us consider two competing branches in the decaying parent: an α and a β branch.
- The decay rates λ_α and λ_β define probability per unit time for disintegration by the respective process. The total probability for disintegration is

$$\lambda = \lambda_\alpha + \lambda_\beta \tag{16}$$

- Relative probability for each branch decay, called the branching ratio, is the ratio of the respective decay rate to the total decay rate

$$\begin{aligned} \text{br}_\alpha &= \frac{\lambda_\alpha}{\lambda} = \frac{\lambda_\alpha}{\lambda_\alpha + \lambda_\beta} \\ \text{br}_\beta &= \frac{\lambda_\beta}{\lambda} = \frac{\lambda_\beta}{\lambda_\alpha + \lambda_\beta} \end{aligned} \tag{17}$$

Lifetime and partial lifetimes

- The decay (without feeding) is defined by the total rate

$$\begin{aligned} \frac{dN(t)}{dt} &= -\lambda N(t) = -(\lambda_\alpha + \lambda_\beta)N(t) \implies \\ N(t) &= N(0)\exp(-\lambda t)) = N(0)\exp(-(\lambda_\alpha + \lambda_\beta)t) = \\ & N(0)\exp(-\lambda_\alpha t)\exp(-\lambda_\beta t) \end{aligned} \tag{18}$$

- Lifetime of the parent is defined by the total rate

$$\tau = \frac{1}{\lambda} \tag{19}$$

- Partial lifetimes for the decays are defined as

$$\tau_\alpha = \frac{1}{\lambda_\alpha}, \quad \tau_\beta = \frac{1}{\lambda_\beta} \tag{20}$$

Lifetime and partial lifetimes

- Note that while

$$\lambda = \lambda_\alpha + \lambda_\beta \tag{21}$$

and the total rate is dominated by the larger of λ_α, λ_β partial rates.

- For the lifetime and partial lifetimes this implies

$$\frac{1}{\tau} = \frac{1}{\tau_\alpha} + \frac{1}{\tau_\beta} \tag{22}$$

and the lifetime of the parent is dominated by the shorter partial lifetime.

- It should be stressed that

$$\tau \neq \tau_\alpha + \tau_\beta$$

and that there is only one lifetime τ defining decay of the parent. Partial lifetimes can be extracted from measured branching ratios.