• Readings: Modern Nuclear Chemistry, Chap. 9; Nuclear and Radiochemistry, Chapter 3
• Energetics
• Decay Types
• Transition Probabilities
• Internal Conversion
• Angular Correlations
• Moessbauer spectroscopy

• Emission of photon during deexcitation of the nucleus
  ▪ Wide range of energies
  ▪ Different % yields

• Isomers
  ▪ Two different nuclear configurations for same isotope
  ▪ Different total angular momenta and energy differences
  → long-lived nuclear states are called isomeric states
  * gamma ray decay is called isomeric transition (IT)

• Gamma decay energy range from few keV to many MeV
Gamma decay example: $^{152}$Eu

- Many gamma transitions from decay of $^{152}$Eu
  - Different decay modes of isotope
    - EC and $\beta^-$
- What gamma data provides % yield
  - From chart of the nuclides, gamma energies at 121.8 keV, 1408 keV, and 344.3 keV
Gamma Data

- Table of the isotope data
  - % yields and transitions
  - 121.8 keV, 1408 keV, and 344.3 keV
Gamma Data

$\gamma^{(152}\text{Sm})$ from $^{152}\text{Eu}$ (13.542 y) EC+$\beta^+$ decay

< for $l\gamma$% multiply by 0.020879>

121.8 keV, 1408 keV, and 344.3 keV
### Gamma Data

121.8 keV, 1408 keV, and 344.3 keV

<table>
<thead>
<tr>
<th>$E_γ$ (keV)</th>
<th>$I_γ$ (%)</th>
<th>Decay mode</th>
<th>Half life</th>
<th>Parent</th>
</tr>
</thead>
<tbody>
<tr>
<td>12.598 15</td>
<td>0.29 3</td>
<td>γ</td>
<td>96 m l</td>
<td>152m² Eu</td>
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<tr>
<td>18.265 7</td>
<td>0.26 21</td>
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<td>96 m l</td>
<td>152m² Eu</td>
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<tr>
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<tr>
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<td>152m² Eu</td>
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<td>344.285 12</td>
<td>26.5 4</td>
<td>β⁻</td>
<td>13.537 y 6</td>
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<tr>
<td>1389.00 1</td>
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<td>9.3116 h 13</td>
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<tr>
<td>1390.36 16</td>
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<tr>
<td>1406.85</td>
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<td>152m² Eu</td>
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<td>1408.006 3</td>
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</tr>
<tr>
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<td>0.0006 4</td>
<td>e+β⁺</td>
<td>9.3116 h 13</td>
<td>152m² Eu</td>
</tr>
<tr>
<td>1457.643 11</td>
<td>0.502 5</td>
<td>e+β⁺</td>
<td>13.537 y 6</td>
<td>152m² Eu</td>
</tr>
<tr>
<td>1460.64 13</td>
<td>0.0016 4</td>
<td>β⁻</td>
<td>9.3116 h 13</td>
<td>152m² Eu</td>
</tr>
</tbody>
</table>

- **Search for % yield for specific isotope**
  - [http://nucleardata.nuclear.lu.se/toi/](http://nucleardata.nuclear.lu.se/toi/)
    → Enter element and isotope

- **Isotope browser for android**
Nuclear Excited State Transitions

• De-excitation of excited states
  ▪ $\alpha$- and $\beta$-decay processes leave product nucleus in either ground state or excited state

• De-excitation can include
  ▪ Emission of electromagnetic radiation ($\gamma$ radiation)
  ▪ newly created electron and positron (higher energy)
    $\rightarrow$ Excited stated greater than 1.02 MeV
  ▪ Internal conversion from interaction between nucleus and extranuclear electrons leading to emission of atomic electron
    $\rightarrow$ kinetic energy equal to difference between energy of nuclear transition involved and binding energy of electron
γ Transitions

- **Pair production**
  - Exceeds 1.02 MeV
  - Emitted with kinetic energies that total excitation energy minus 1.02 MeV
  - Uncommon mode

- **Gamma decay characterized by a change in energy without change in Z and A**
  - $E = h\nu$
  - Majority of γ transitions have very short lifetimes, 1E-12 seconds
    - Table of the Isotopes provide data
    - Longer lived states are metastable

- γ transitions used for determining nuclear energy levels and decay schemes
Energetics

• Recoil from gamma decay
  ▪ Energy of excited state must equal
  → Photon energy, and recoil
    * $M\cdot c^2 = M c^2 + E_\gamma + T_r$
  ▪ Momentum same for recoil and photon

• If $E_\gamma = 2$ MeV, and $A=50$
  ▪ recoil energy is about 40 eV
→ Use 931.5 MeV/AMU

• Important for Moessbauer spectroscopy (page 19)
• Find recoil from 15.1 MeV photon from $^{12}C$

$$T_r = \frac{E_\gamma^2}{2M} = \frac{15.1^2}{2 \times 12 \times 931.5} = 1.02E - 2\text{MeV} = 10.2\text{keV}$$
Multipole Radiation & Selection Rules

- Since $\gamma$ radiation arises from electromagnetic effects, it can be thought of as changes in the charge and current distributions in nuclei
  - Charge distributions resulting electric moments
  - Current distributions yield magnetic moments
- Gamma decay can be classified as magnetic (M) or electric (E)
  - E and M multipole radiations differ in parity properties
- Transition probabilities decrease rapidly with increasing angular-momentum changes
  - as in $\beta$-decay
Angular momentum from decay
- \( l=1,2,3,4 \)
- \( 2^l \)-pole (dipole, quadrupole, octupole...)

Shorthand notation for electric (or magnetic) \( 2^l \)-pole radiation
- \( E_l \) or \( M_l \)
  \( \rightarrow \) \( E_2 \) is electric quadrupole

Determine multipole of decay
- \( I_i^+ I_f \geq 1 \geq |I_i-I_f| \), where \( I_i \) is initial spin state and \( I_f \) is final spin state
  - Initial and final state have same parity
    - allowed transitions are:
      - electric multipoles of even \( l \)
      - magnetic multipoles of odd \( l \)
  - If initial and final state different parity
    - electric multipoles of odd \( l \)
    - magnetic multipoles of even \( l \)

Example:
- Transition is between a 4+ and a 2+ state
  - \( l \) between 6 and 2
    - \( 4^+2 \) to \( 4^-2 \)
  - Same parity, both +
    - \( E \) even, \( M \) odd
      - \( E_2, M_3, E_4, M_5, E_6 \) transitions are allowed
      - Generally lowest multipole observed
      - Expect \( E_2 \) as the main transition

- 137Cs example
  - \( 11/2^- \) to \( 3/2^+ \)
  - \( 11/2+3/2 = 7 \)
  - \( 11/2-3/2 = 4 \)
  - Different parity between states
    - \( E \) odd, \( M \) even
    - \( M_4, E_5, M_6, E_7 \)
Isomeric Transitions

- Isomeric transition (IT) is a $\gamma$ decay from an isomeric state

- Transition probability or partial decay constant for $\gamma$ emission
  \[ \lambda_\gamma \propto E^{2l}A^{2l/3} \quad (l \text{ not } 1) \]

- For given spin change, half lives decrease rapidly with increasing $A$ and more rapidly with increasing $E$

- Weisskopf single particle model
  \- Model predicts low-lying states of widely differing spins in certain regions of neutron and proton numbers
  \- Numbers preceding shell closures at $N$ or $Z$ values of 50, 82, 126
  \- Coincide with “islands of isomerism”

  \* Large number of isomeric states near magic numbers

- Predictions strong for M4 isomers
  - E2 isomers 100 faster than predicted
  \[ \rightarrow \text{Variations in nuclear shape} \]

---

**Table 3-4 Partial Half Lives for Gamma Transitions Calculated on the Single-Particle Model\textsuperscript{a}**

<table>
<thead>
<tr>
<th>Transition Type</th>
<th>Partial Half Life $t_\gamma$ (s)</th>
<th>Illustrative $t_\gamma$ Values (s) for $A = 125$, $E = 1$ MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E1$</td>
<td>$5.7 \times 10^{-15}$ $E^{-3}A^{-2/3}$</td>
<td>$2 \times 10^{-15}$</td>
</tr>
<tr>
<td>$E2$</td>
<td>$6.7 \times 10^{-9}$ $E^{-5}A^{-4/3}$</td>
<td>$1 \times 10^{-6}$</td>
</tr>
<tr>
<td>$E3$</td>
<td>$1.2 \times 10^{-2}$ $E^{-7}A^{-2}$</td>
<td>$8$</td>
</tr>
<tr>
<td>$E4$</td>
<td>$3.4 \times 10^{-6}$ $E^{-9}A^{-8/3}$</td>
<td>$9 \times 10^{7}$</td>
</tr>
<tr>
<td>$E5$</td>
<td>$1.3 \times 10^{11}$ $E^{-11}A^{-10/3}$</td>
<td>$1 \times 10^{15}$</td>
</tr>
<tr>
<td>$M1$</td>
<td>$2.2 \times 10^{-14}$ $E^{-3}$</td>
<td>$2 \times 10^{-11}$</td>
</tr>
<tr>
<td>$M2$</td>
<td>$2.6 \times 10^{-8}$ $E^{-5}A^{-2/3}$</td>
<td>$1 \times 10^{-4}$</td>
</tr>
<tr>
<td>$M3$</td>
<td>$4.9 \times 10^{-2}$ $E^{-7}A^{-4/3}$</td>
<td>$8 \times 10^{2}$</td>
</tr>
<tr>
<td>$M4$</td>
<td>$1.3 \times 10^{4}$ $E^{-9}A^{-2}$</td>
<td>$8 \times 10^{9}$</td>
</tr>
<tr>
<td>$M5$</td>
<td>$5.0 \times 10^{11}$ $E^{-11}A^{-8/3}$</td>
<td>$1 \times 10^{17}$</td>
</tr>
</tbody>
</table>

\textsuperscript{a} The energies $E$ are expressed in MeV. The nuclear radius parameter $r_0$ has been taken as 1.3 fm. Note that $t_\gamma$ is the partial half life for $\gamma$ emission only; the occurrence of internal conversion will always shorten the measured half life.
Non-photon emission for de-excitation

- 0 → 0 transitions cannot take place by photon emission
  - Photon has spin and therefore must remove at least one unit of angular momentum

- If no change in parity in 0 → 0 transition de-excitation occurs by other means
  - emission of an internal-conversion electron
    \[ \rightarrow ^{72}\text{Ge}, ^{214}\text{Po} \]
  - simultaneous emission of an electron-positron pair (\(\Delta E > 1.02\) MeV)
    \[ \rightarrow ^{16}\text{O}, ^{42}\text{Ca} \]

- Transitions between two I=0 states of opposite parity cannot take place by any first-order process
  - requires simultaneous emission of two \(\gamma\) quanta or two conversion electrons
Internal Conversion Coefficients

• Excited nucleus ejects atomic electron
  ▪ Discrete energy emission, only one particle
  ▪ Generally k shell electrons
• Interaction between nucleus and extranuclear electrons
  ▪ emission of electron with kinetic energy equal to difference between energy of nuclear transition and electron binding energy
• Internal conversion favored when:
  ▪ energy gap between nuclear levels is small
  ▪ \(0^+ \rightarrow 0^+\) transitions
• Internal conversion coefficient $\alpha$
  - ratio of rate of internal conversion process to rate of $\gamma$ emission
    * ranges from zero to infinity
    * coefficients for any shell generally increase with decreasing energy, increasing $\Delta I$, and increasing $Z$

• Internal conversion electrons show a line spectrum
  - correspond to $\gamma$-transition energy minus binding energies of electron shells in which conversion occurs
  - difference in energy between successive lines are used to determine $Z$
Internal conversion spectrum

- $\alpha_K / \alpha_L$ ratios can be used to characterize multipole order
  - Determine $\Delta I$ and $\Delta \Pi$
  - Compare to table on previous page
- If $Z$ of x-ray-emitting species known, it can be determined whether it decays by EC or IT
  - X-rays generated from daughter isotope
    - For EC, x-rays will be of $Z-1$
    - IT x-rays from $Z$
- Specific lines generated from nuclear transition
  - Overlaid on beta spectrum
  - Can determine specific peaks and electron binding energies

**Binding energies for $^{203}$Tl (keV)**
- $K$ 85.529
- $L_I$ 15.347
- $L_{II}$ 14.698
- $L_{III}$ 12.657
- $M$ 3.704
Angular Correlations of Gamma Decay

• Assumes $\gamma$ rays have no track of multipole interaction from production
  ▪ In some cases multipole fields give rise to angular distributions of emitted radiation with respect to nuclear-spin direction of emitting nucleus

• Generally not observed during gamma decay
  ▪ ordinarily samples contain randomly oriented nuclei
  ▪ Observed angular distribution of $\gamma$ rays is isotropic due to random orientation
    → Would be remove if nuclei aligned
Angular correlation

- If nuclear spins can be aligned in one direction, angular distribution of emitted $\gamma$-ray intensity would depend on initial nuclear spin and multipole character of radiation
  - Align nuclei in magnetic or electric field at near 0 K
  - Observe a $\gamma$ ray in coincidence with a preceding radiation
    - $\rightarrow$ Alpha, beta, or gamma

- Coincidence experiment
  - Angle $\theta$ between two sample-detector axes is varied, coincidence rate will vary as a function of $\theta$

Correlation function:

$$W(\theta) = 1 + a_2 \cos^2 \theta + a_4 \cos^4 \theta$$

$$A = \frac{W(180^\circ) - W(90^\circ)}{W(90^\circ)}$$

Where $A = a_2 + a_4$ (fits)
Angular Correlations

- Correlate gamma emission with preceding radiation
  - Need very short gamma lifetime
  - Measure coincidence as function of $\theta$
- Schematic diagram of angular correlations
  - $\gamma_1\gamma_2$ cascade, Z axis defined by $\gamma_1$
  - Requires time and spatial correlated detectors
CHEM 312: Lecture 6 Gamma Decay

- Readings: Modern Nuclear Chemistry, Chap. 9; Nuclear and Radiochemistry, Chapter 3
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- Internal Conversion
- Angular Correlations
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Mössbauer Spectroscopy

- Uses of gamma emission and absorption to determine chemical information

- Principles
- Conditions
- Spectra

**Principles**

- Nuclear transitions
  - emission and absorption of gamma rays
    → sometimes called nuclear gamma resonance spectroscopy
- Only suitable source are isotopes
  - Emission from isotope is essentially monochromatic
  - Energy tuning performed by Doppler effect
    → Vibration of source and absorber
  
  * spectra recorded in mm/s (1E-12 of emission)
Recoil

- Recoil converted to vibrational energy
- Associated recoil energy of emitter
  - With gamma decay $E$ is large enough to have a measurable effect
- Molecules in gas or liquid cannot reabsorb photon
- In practice lattice vibrational modes may be excited during absorption
  - Recoil can be observed in solid
  - Entire solid recoils (recoil free)
- Emitting nuclei in chemical system
  - Thermal equilibrium, moving source
  - Doppler shift of emitted photon
    $\rightarrow$ Slight variation of photon energy with vibration

$$E_r = \frac{\rho^2}{2M} = \frac{E^2}{2M}$$

$$E_r(eV) = \frac{537E^2}{2M}$$

$E$ in Mev

$$\Delta E = \frac{V}{c} E \cos \vartheta$$

$\vartheta$ is angle between direction of motion of nucleus and emitted photon, $v$ is nucleus velocity
Recoil Free Fraction

- $\vartheta$ can vary from -1 to 1, so distribution is $E_T - E_R$
  - $E_T$ is gamma transition energy from excited to ground state
  - $E_R$ is recoil energy
  - Distribution around 0.1 eV at room temp

- Some chemical energy goes into photon, and some recoil energy goes into lattice phonon
- Heisenberg uncertainty implies distribution of energy from finite half-life
  - $\Gamma$ (in eV) = $4.55E-16/t_{1/2}$ (sec)
    - $\Gamma$ level width, which is finite due uncertainty principle

- What Mössbauer did
  - Total recoil in two parts, kinetic and vibrational
  - If emitter and absorber are part of lattice, vibrations are quantized
    - Based on phonon
  - Recoil energy transfer only in correct quanta
Recoil Free Fraction

- If recoil energy is smaller than quantized vibration of lattice whole lattice vibrates
- Mass is now mass of lattice
  - $v$ is small as is recoil kinetic energy
- $E$, $E_T$ and recoil energy goes into lattice phonon system
  - Lattice system is quantized, so it is possible to find a state of system unchanged after emission
Recoil free fraction

- Energy goes into lattice phonons
- For $E > 150$ keV nearly all events vibrate lattice
  - Gives rise to Mössbauer spectra
  - $\rightarrow$ recoil-free fraction
    - Portion of radiation which is recoil free
- Vibration of lattice reduced with reduced temperature
- Recoil-free fraction increases with decreasing temperature
- Temperature range from 100 to 1000 K
- For gamma level half-lives greater than $1E-11$ seconds, natural width around $1E-5$ eV
  - For gamma decay of 100 keV
    - $\rightarrow$ Doppler shift of $1E-5$ eV is at a velocity of 3 cm/s
Isomeric or Chemical Shift

- Volume of nucleus in excited state is different from ground state
  - Probability of electron orbitals found in nucleus is different
    → Can be used to evaluate chemical state
- Difference appears as a difference in total electron binding state and contributes to transition energy
  - $E_T = \Delta E(nucl) + \Delta E(elect)$ [binding energies]
  - Consider an emitting nucleus (excited) and absorber (ground) in different chemical states
  - Difference in $\Delta E(elect)$ and therefore $E_T$
  - Change is chemical shift

$$\Delta E(elect) = \frac{2}{5} \pi Z e^2 (\bar{r}_{ex}^2 - \bar{r}_{gr}^2) \left[ |\psi_{ex}(0)|^2 - |\psi_{gr}(0)|^2 \right]$$
Magnetic Dipole Splitting

- magnetic moment will add to transition energy
  - \[ E_T = \Delta E(\text{nucl}) + \Delta E(\text{elect}) + \Delta E(\text{mag}) \]
- Change in magnetic moment will effect shift
- Split also occurs \((2I+1)\) values
- around 1cm/s

Electric Quadrupole Splitting

- inhomogeneous magnetic field
  - \[ E_T = \Delta E(\text{nucl}) + \Delta E(\text{elect}) + \Delta E(\text{mag}) + \Delta E(\text{quad}) \]
Technique

- Intensity of photon from emitter is detected
- Velocity of emitter and absorber recorded
  - important to know these values
- May be cooled and place in magnetic field
- Used in
  - amorphous materials
  - catalysts
  - soil
  - coal
  - sediments
  - electron exchange
Mössbauer Devise

First Mössbauer Spectrum Recorded on Martian Surface
Gusev Crater, January 17, 2004 (3h25min)

Fe^{2+}/Fe_{total} \sim 0.6
$^{237}$Np Moessbauer spectroscopy

- 68 ns excited state lifetime
- Isomer shift suitable for analysis of chemical bonds
- Can record radiation spectrum from absorber
  - 60 keV from $^{241}$Am
- Shift correlated with oxidation state and number of 5f electrons present

Fig. 3. $^{237}$Np Mössbauer spectra of NpFeGa$_5$ at 10 K.
Topic Review

• Trends in gamma decay
  ▪ How does it come about, how is it different from alpha and beta
• Energetics of gamma decay
• Decay Types
  ▪ Photon emission, IC, pair production
• E and M transitions
  ▪ Probabilities, modes, and how to define
• Angular Correlations
  ▪ How are they measured and what do they inform about nucleus
• Moessbauer spectroscopy
Questions

- $^{195}$Pt has a ground state spin and parity of $\frac{1}{2}^-$, with excited states at 99 keV (3/2-) and 130 keV (5/2-). Does the 5/2 level decay primarily to the 3/2- level or to the $\frac{1}{2}$- level? Why? What is the transition multipolarity?
- What is the spin of a photon?
- What type of gamma decay is expected from a 0+ to 0+ transition?
- Classify the most likely multipolarity for the $\gamma$-ray decay of $^{60m}$Co.
- Describe Mössbauer spectroscopy
- Why do angular correlations arise in the nucleus? How are they measured

- emission of an internal-conversion electron
- simultaneous emission of an electron-positron pair ($\Delta E > 1.02$ MeV)

$^{195}$Ir $^{195}$Pt

$Q_{\beta^-} = 1120.1$ keV

2.5 h half life

$129.777$ 0.67 ns

$98.882$ 0.170 ns

13% 7.0 $1/2^-$

57% 6.2 $5/2^-$

26% 6.6 $3/2^-$

1% 4.0 $7/2^-$

$^{60m}$Co

$Q_{\beta^-} = 52.1$ keV

$Q_{\gamma} = 20.7$ keV

$2^+$ to $5^+$: 3 to 7, Same parity E even $M3, E4, M5, E6, M7$

$^{60m}$Co

$Q_{\gamma} = 20.7$ keV

$2^+$ to $5^+$: 3 to 7, Same parity E even $M3, E4, M5, E6, M7$

2+ to 5+: 3 to 7, Same parity E even $M3, E4, M5, E6, M7$

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$Q_{\beta^-} = 52.1$ keV

$Q_{\gamma} = 20.7$ keV

$2^+$ to $5^+$: 3 to 7, Same parity E even $M3, E4, M5, E6, M7$
Questions

- Determine gamma decay yields
  - $^{95}$Zr
  - $^{241}$Am
  - $^{60}$Co

http://nucleardata.nuclear.lu.se/toi/
Lund LBNL data site, nuclide search
Questions

• What are metastable isotopes?

• Provide the half-life of $^{99m}$Tc

• Why do isomeric states exist?

• Where do isomers exist?

Long-lived nuclear states
Gamma ray decay isomeric transition (IT)

Large spin changes

Large number of isomeric states near magic numbers
• Comment on blog
• Provide response to PDF quiz 6