Analysis of the Nuclear Structure of ^{186}Re Using Neutron-Induced Reactions

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Committee and Sponsors

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Outline

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Thesis Statement

Measurements of γ-ray spectra from $^{187}\text{Re}(n, xn\gamma)^{188-x}\text{Re}$ reactions would provide valuable input to nuclear structure databases such as the Experimental Unevaluated Nuclear Data List (XUNDL) and the Evaluated Nuclear Structure Data File (ENSDF).

New levels and transitions discovered in ^{186}Re, and in particular levels that directly populate the ^{186m}Re isomer, would lead to a greater understanding of the isomer and a potential application in a radioisotope power source.

Research Objectives

- Examine nuclear structure of ^{186}Re using γ-ray spectra obtained from $(n, 2n\gamma)$ reactions on an enriched ^{187}Re target
  - Of particular interest: transitions that feed the 149(7) keV isomer ^{186m}Re
- Generate γ-ray excitation functions and partial cross sections for comparison with theoretical models

Background

- Rhenium ($Z = 75$) is a rare earth metal, the last stable element to be discovered
- Two stable isotopes
  - $A = 185$ and 187
- ^{186}Re decays with $t_{1/2} = 3.7$ d
  - 92.5% β^- to ^{186}Os
  - 7.5% EC to ^{186}W
- ^{186}Re possesses an isomer with $t_{1/2} = 2 \times 10^5$ y
  - Isomer excitation energy (adopted value) is 149 ± 7 keV
  - Decays only by γ-ray cascade to the ground state

Motivation

- Contribution to nuclear structure databases
  - Level scheme of ^{186}Re includes many tentative assignments
- Astrophysical implications
  - Neutron-induced reaction cross sections used in calculations related to ^{187}Re/^{187}Os as a cosmochronometer
  - No consideration of ^{186m}Re in current model
- High-energy density applications
  - Energy density of nuclear isomers is second only to fission
  - Long-lived isomers may be induced to release excess energy
  - β^- decay of ground state ^{186}Re could be used in a switchable power storage device

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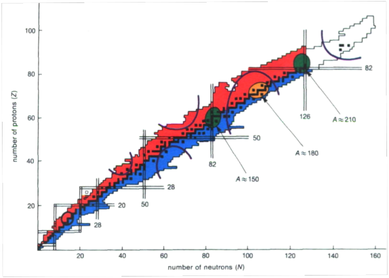
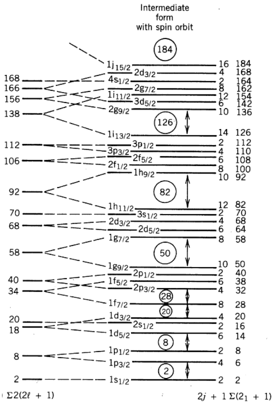
Nuclear Landscape

- Chart of the nuclides: plot of neutron number N versus Z
  - Black dots: stable nuclides
  - Blue: β^- decay
  - Red: β^+ decay or e^- capture
- Nuclear properties explained by shell model
  - Shell closures result in magic numbers (double lines)
  - Green regions: spin trap isomers
- Nilsson model for deformed nuclei
  - Purple arcs: deformation
  - Orange region: K-trap isomers



Shell Model

- Most successful nuclear structure model to date
- Neutrons and protons fill successive orbits per Pauli principle
- Groupings in energy levels define distinct shells and magic numbers
- Nuclear spin and parity (J^{π}) determined from unpaired nucleons



Transition Selection Rules

- Quantum-mechanical selection rules for γ-ray transitions between nuclear levels

$$|\ell_i - \ell_f| \leq L \leq \ell_i + \ell_f$$

$$\Delta\pi = \text{no} \rightarrow \text{even electric, odd magnetic}$$

$$\Delta\pi = \text{yes} \rightarrow \text{odd electric, even magnetic}$$

  - $\ell_{i,f}$: spin quantum number of initial, final levels
  - π: level parity
  - L: multipole order of transition
- Lowest permitted multipole order is most likely
- Electric more likely than magnetic

Weisskopf Estimates

- Shell model used to derive estimates of γ decay rates λ $[s^{-1}]$
  - E is transition energy [MeV] and A is atomic number

Electric	Magnetic
$\lambda(E1) = 1.0 \cdot 10^{14} A^{2/3} E^3$	$\lambda(M1) = 5.6 \cdot 10^{13} E^3$
$\lambda(E2) = 7.3 \cdot 10^{7} A^{4/3} E^5$	$\lambda(M2) = 3.5 \cdot 10^{7} A^{2/3} E^5$
$\lambda(E3) = 34 A^{2} E^7$	$\lambda(M3) = 16 A^{4/3} E^7$
$\lambda(E4) = 1.1 \cdot 10^{-5} A^{8/3} E^9$	$\lambda(M4) = 4.5 \cdot 10^{-6} A^{2} E^9$

- $\sim$ 5 order of magnitude decrease in transition rate for incremental increase in multipole order

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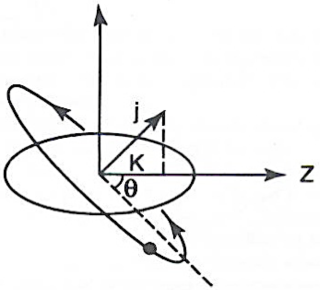
Nilsson Model

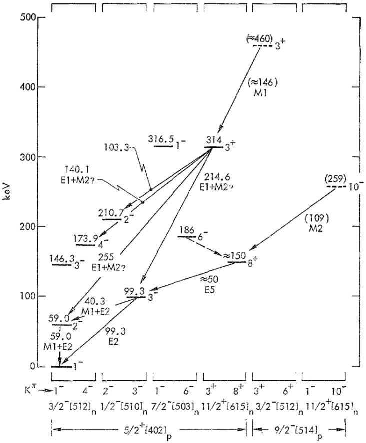
- Away from magic numbers, deformation is important
  - J ceases to be a good quantum number
  - Quantum number K, the projection of angular momentum on the nuclear axis of symmetry
- Prolate (football-shaped) and oblate (frisbee-shaped) nuclei
- Degree of deformation modifies energy levels in shell model



Nucleon of spin j orbiting a prolate nucleus

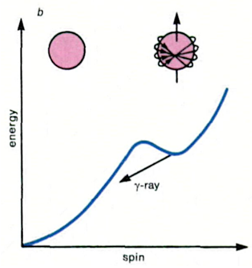
Structure of ^{186}Re

- Configurations of energy levels in Nilsson model
  - Convention $K^\pi\ [N\ell m_\ell]$
  - Arises from spherical state spin-parity J^π
- Energies are approximate for $J^\pi = 8^+$ isomer and γ transition to 3^- level at 99.3 keV



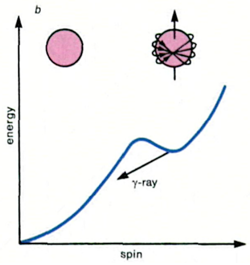
Spin Traps

- Local minimum in excitation energy plotted against angular momentum $\sqrt{J(J+1)}\hbar$
- Spin difference between states affects γ-ray transition rate (Weisskopf estimates)
- For ^{186}Re, $J = 8$ for isomer and $J = 1$ for ground state
    - γ-decay via cascade involving $J = 3$ state at 99.3 keV



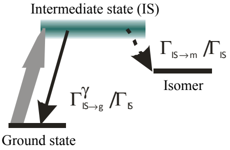
K-Traps

- Occur in non-spherical nuclei, as in midshell region $150 \leq A \leq 190$
- Local minimum in excitation energy plotted against K
- Difference in K between states inhibits transition ($L < \Delta K$ forbidden)
- For ^{186}Re (prolate), $\Delta K = 4$ for direct transition to ground state

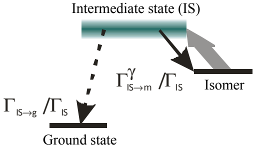


Isomer Photoexcitation and Depletion

- Goal is to achieve stimulated isomer energy release (depletion)
- Excitation and depletion are analogous processes
  - Resonant (γ, γ') reaction using bremsstrahlung source
  - Involve transitions to/from IS, width Γ
  - Smaller ΔJ or ΔK between initial state and IS than for direct transition
- Photoexcitation reaction $^{115}\mathrm{In}(\gamma, \gamma')^{115m}\mathrm{In}$ demonstrated using WSU Dynamitron



(a) Photoexcitation



(b) Depletion

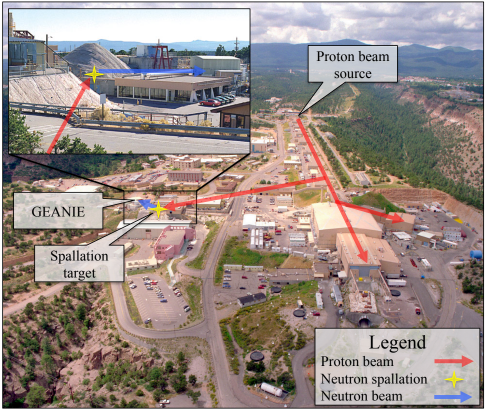
Outline

Summary

- Experiment took place 10 November – 9 December 2014 at Los Alamos Neutron Science Center (LANSCE)
- Examined γ-rays produced from inelastic neutron scattering reactions:
  - $^{187}\text{Re}(n, n'\gamma)^{187}\text{Re}$
  - $^{187}\text{Re}(n, 2n\gamma)^{186}\text{Re}$
- Spectrometer: GErmanium Array for Neutron-Induced Reactions (GEANIE)
- Target:
  - Rhenium metal powder, 981.7 mg
  - Enriched to 99.52% ^{187}Re at ORNL
  - Pressed and encapsulated in polycarbonate holder

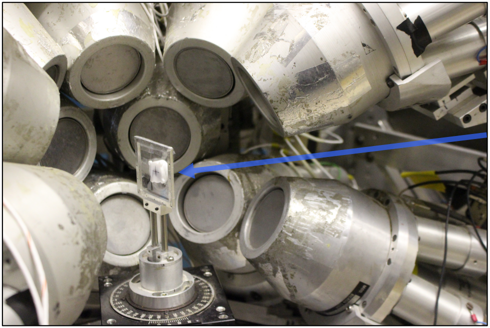
Los Alamos Neutron Science Center (LANSCE)

- 800 MeV proton LINAC
- Pulsed beam operated at 60 or 120 Hz
- Spallation neutrons from tungsten target
- 0.6 – 200 MeV neutrons delivered to GEANIE (60° right off beam path)



GErmanium Array for Neutron-Induced Reactions (GEANIE)

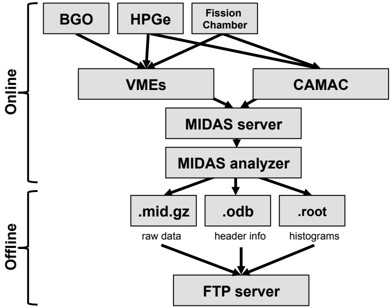
- 18 HPGe detectors
  - Mix of planar and coaxial geometries
  - E_γ and neutron time-of-flight signals sent to hardware
- Compton-suppressed using BGO detectors
- Neutron flux measured by fission chamber
  - ^{235}U and ^{238}U foils



Outline

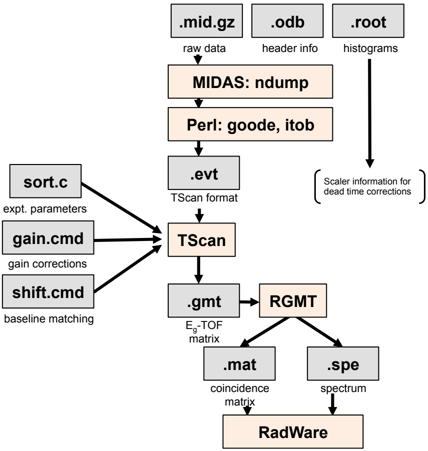
Data Acquisition

- 14 detectors (7 coaxial and 7 planar) produced usable data
- Over 62 GB of data collected, comprising 441 runs
- Online and offline data analysis involved both open-source and proprietary LANSCE software
- Each γ-ray event was tagged with analog-to-digital converter (ADC) and time-to-digital converter (TDC) channels



Offline Analysis

- Good events were selected in software with knowledge of known faults
- ADCs had to be gain-matched such that γ-ray peaks were aligned prior to adding spectra
  - Gain drift corrections
  - Coaxial and planar detectors analyzed separately
- Energy calibration using data from ^{152}Eu source

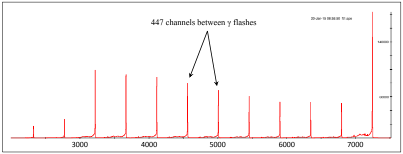


Neutron Energy Determination

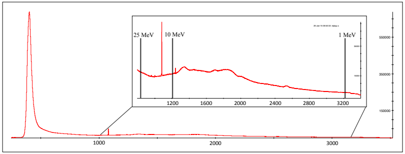
- TDC spectra collapsed to time-of-flight (ToF) spectra
- Neutron energies were calculated from time-of-flight in sort routine:

$$E_n = \frac{E_0}{\sqrt{1 - v^2/c^2}}$$

  - $E_0 = 939.57$ MeV is the neutron rest energy
  - Neutron velocity v determined from the ToF and known distance (20.34 m) from spallation source to target



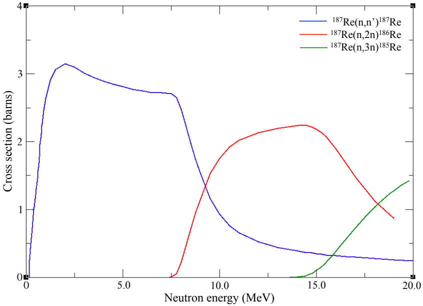
(a) TDC spectrum



(b) TOF spectrum

Reaction Product Selection

- Known neutron energies permitted selection of reaction products according to reaction channel
- ENDF cross sections for $^{187}\mathrm{Re}(n, xn)^{188-x}\mathrm{Re}$ reactions define neutron energy gates



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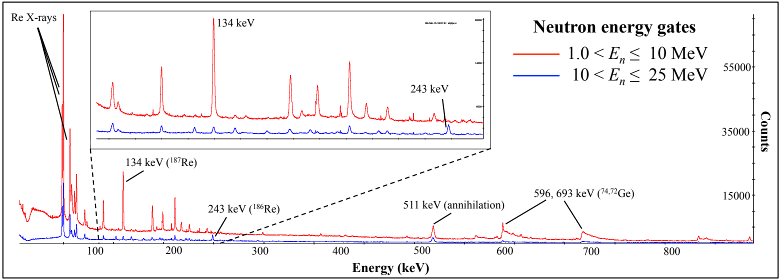
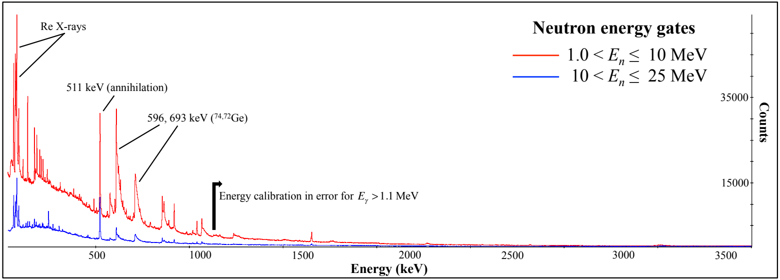
Planar Detector γ-ray Spectra



Summed planar detector γ-ray spectra ($10 \leq E_\gamma \leq 900$ keV)

Coaxial Detector γ-ray Spectra



Summed coaxial detector γ-ray spectra ($50 \leq E_\gamma \leq 3750$ keV)

Identification of New $\gamma-$ray Transitions

- Peaks in γ-ray spectra compared with transitions in ENSDF and XUNDL
- Known sources of background excluded
    - X-rays and γ-rays from ^{209}Bi in BGOs
    - γ-rays from ^{19}F in target capsule
    - Neutron scattering on ^{72,74}Ge (long tails)
    - $e^- e^+$ annihilation (Doppler broadened)
- Remaining peaks compared with list of energy level differences for product nucleus
- Assignment based on lowest multipole order allowed per selection rules

Newly-observed $\gamma-$ray Transitions in ^{186}Re

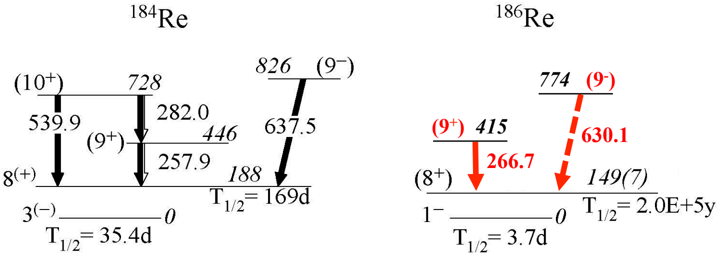
Energy (keV)	Error	Levels (initial and final)	Transition
266.7386	0.0168	414.9(5) keV to 149(7) keV J^π unassigned to $J^\pi = 8^+$	$E1$?
354.2677	0.0867	500.722(16) keV 146.274(4) keV $J^\pi = (4)^+$ to $J^\pi = (3)^-$	$E1$
381.2162	0.0568	559.976(9) keV to 180.1(5) keV $J^\pi = (5)^+$ to $J^\pi = (6)^-$	$E1$
492.6613	0.1167	1069.8 keV to 577.732(16) keV $J^\pi = (2^-, 3^-)$ to $J^\pi = (2^-)$	$M1$
630.1362	0.1277	689.3 keV to 59.010 keV $J^\pi = (1^-)$ to $J^\pi = (2)^-$ **OR** 774.2(15) keV to 149(7) keV J^π unassigned to $J^\pi = 8^+$	$M1$ $M1$?

Newly-observed $\gamma-$ray Transitions in ^{187}Re

Energy (keV)	Error	Levels (initial and final)	Transition
660.3193	0.0749	1661(6) keV to 1000.93(12) keV $J^\pi = (1/2^-, 3/2^+, 5/2^-)$ to $J^\pi = (5/2^-, 7/2^+)$	$E1$
719.4028	0.0764	230.10(4) keV to 511.768(7) keV $J^\pi = (3/2^+, 5/2^+)$ to $J^\pi = 1/2^+$	$M1$ or $E2$
992.5207	0.0703	1200(3) keV to 206.252(7) keV $J^\pi = (9/2^-)$ to $J^\pi = 9/2^-$	
1014.5449	0.0193	1220.80(25) keV to 206.252(7) keV J^π unassigned to $J^\pi = 9/2^-$	

Isomer Feeding Mechanism in ^{186}Re

- Similarities between ^{184}Re and ^{186}Re level schemes motivated transition assignments in ^{186}Re
- Known level at 414.9(5) keV and new transition with energy 277.74(2) keV implies isomer energy is 148.2(5) keV



Outline

Excitation Functions

- Determine γ-ray excitation functions for new transitions

$$X_\gamma(E_n) = \frac{I_\gamma(E_n)}{\Phi(E_n)}$$

  - I_γ is γ-ray intensity in photons/s
  - Φ is the neutron flux in neutrons/MeV·s
- Excitation function peak defines reaction channel
  - $1 \leq E_n \leq 10$ MeV for $(n, n'\gamma)$ reaction channel
  - $10 \leq E_n \leq 20$ MeV for $(n, 2n'\gamma)$ reaction channel
- Shape provides information about spin of level populated

Transition Cross Sections

- Transition cross sections $\sigma(E_n)$ proportional to γ-ray excitation functions

$$\sigma(E_n) = X_\gamma(E_n) \cdot \frac{\mathrm{DeadTime}_\gamma \cdot (1+\alpha) \cdot C_\gamma(E_n)}{\mathrm{DeadTime}_\Phi \cdot \epsilon(E_\gamma) \cdot t} \cdot N$$

  - $\mathrm{DeadTime}_{\gamma,\Phi}$ is the dead time correction for the HPGe and fission chamber detectors
  - α is the internal conversion coefficient for the transition
  - $C_\gamma(E_n)$ is an angular correction factor (accounts for self-absorption and angular distribution)
  - $\epsilon(E_\gamma)$ is the total efficiency of the detector array at the photon energy E_γ
  - t is the areal density of the target, in atoms/barn
  - N is a normalization factor from ^{56}Fe measurement

Theoretical Models

- Nuclear reaction codes output partial γ-ray cross sections, equivalent to transition cross sections up to $(1 + \alpha_\gamma)$ factor
- Statistical codes
  - CoH_3 developed at LANL
  - TALYS 1.6 code developed by European team
  - Both used previously to validate experimental cross sections from GEANIE data

Data from ^{nat}Re Target

- 24 hours irradiation of ^{nat}Re (37.4% ^{185}Re and 62.6%^{187}Re) target
- Limited data from reactions on ^{185}Re:
  - $^{185}\mathrm{Re}(n, n'\gamma)^{185}\mathrm{Re}$
  - $^{185}\mathrm{Re}(n, 2n\gamma)^{184}\mathrm{Re}$
- Excitation function for transitions feeding the 188 keV isomer in ^{185}Re could be compared with those claimed to feed the isomer in ^{186}Re

Outline

Significant Findings

- Level schemes of both ^{186}Re and ^{187}Re enriched
  - 5 new transitions in ^{186}Re
  - 4 new transitions in ^{187}Re
- First claimed discovery of transition that feeds the $J^{\pi} = 8^{+}$ isomer in ^{186}Re
- Improved estimate of isomer energy
  - Adopted value: 149 ± 7 keV
  - Proposed: 148.2 ± 0.5 keV
- Abstract submitted for presentation at APS April Meeting

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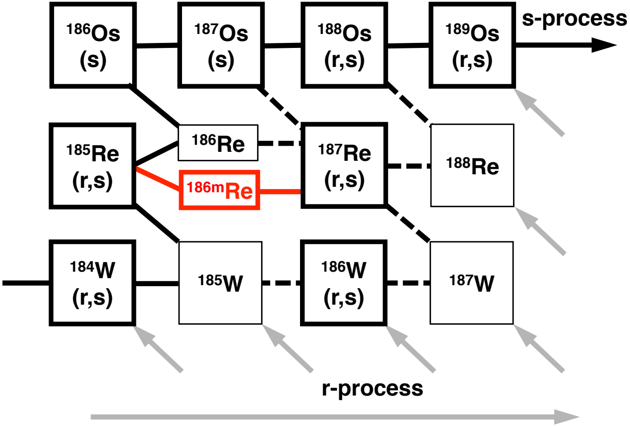
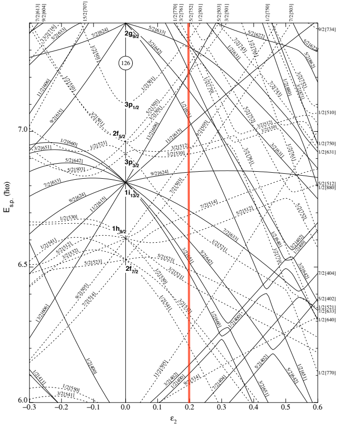
Questions?

Outline

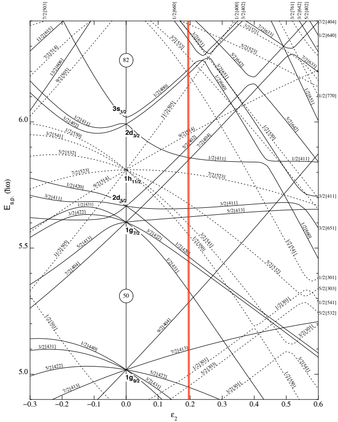
Nucleosynthesis of ^{187}Re and ^{187}Os



Nilsson Diagrams



(a) Neutron energy levels



(b) Proton energy levels

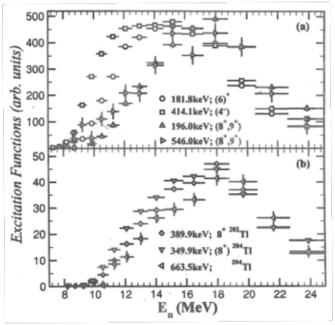
Vertical red line shows ^{186}Re deformation parameter $\epsilon = 0.195$

Significance of Excitation Functions

- Excitation function peak defines reaction channel
- Shape provides information about spin of level populated
    - Greater neutron energy required to populate high-spin state
    - Classically, impact parameter corresponds to $\vec{r}$

$$\vec{L} = \vec{r} \times \vec{p}$$

- Can compare excitation functions from similar isotopes



Excitation functions from (a) $^{205}\mathrm{Tl}(n, 2n\gamma)^{204}\mathrm{Tl}$ and (b) together with those from $^{203}\mathrm{Tl}(n, 2n\gamma)^{202}\mathrm{Tl}$