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ISOMERS AND APPS

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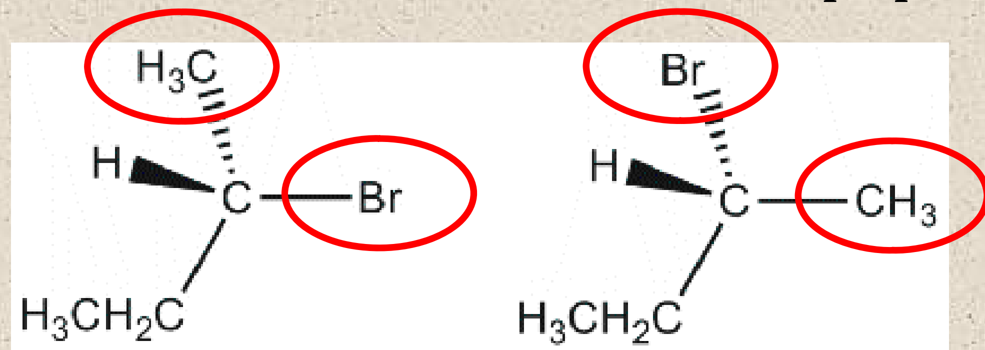
Workshop on Nuclear Isomers: Structure and Applications
University of Surrey, Guildford, UK
May 19 – 21, 2010

CHEMICAL VS. NUCLEAR ISOMERS

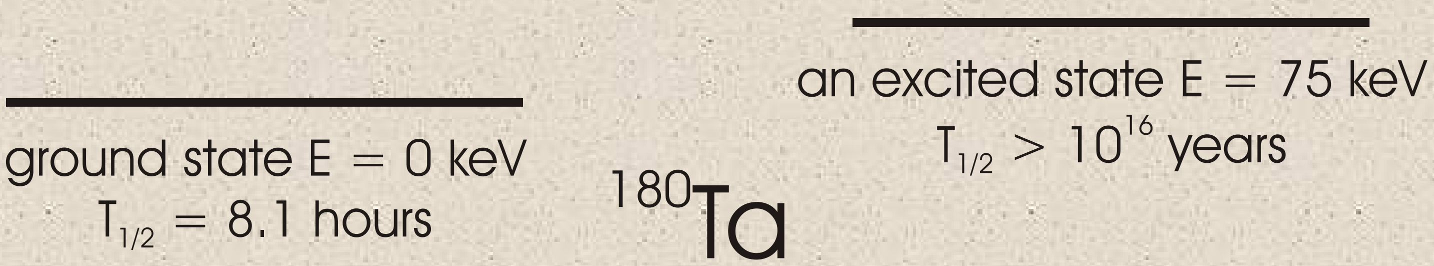
- **Chemical isomers refer to molecules that have the same numbers and types of constituents, but different structure and properties.**



Reflection symmetry in one part of molecule – configurational enantiomer (stereoisomer)

Graphic: wikipedia.org/wiki/enantiomer

- **Nuclear isomers refer to atomic nuclei with the same numbers and types of constituents (thus, the same isotope), yet which can exist within a sample in one of several forms for a significant period of time. One example is ^{180}Ta, which has two long-lived isomeric states:**



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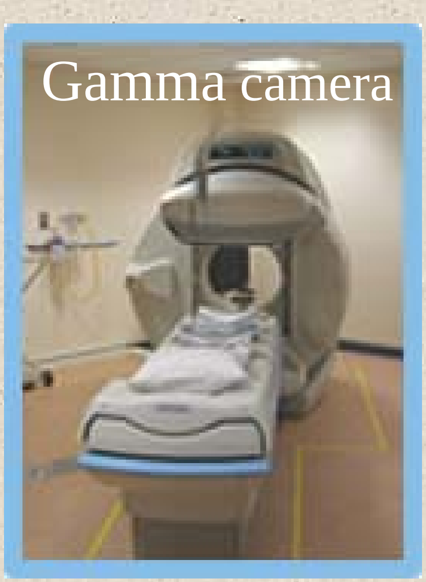
ISOMER EXISTENCE...

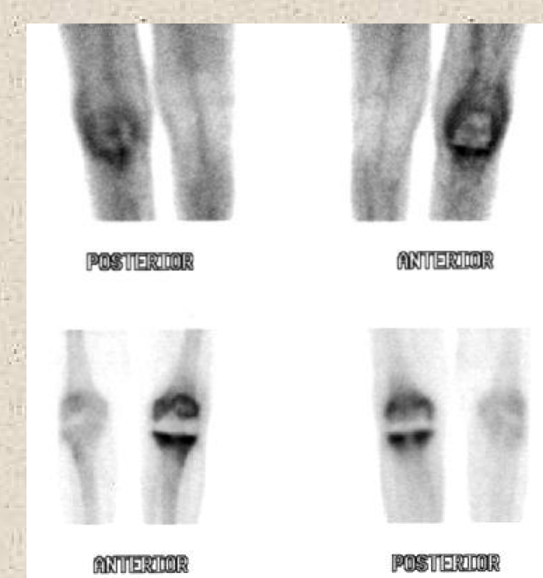
- **Fifteen metastable isomers are known with shelf-lives of ½ year or longer.**
- **Longest-lived isomer is ^{180m}Ta – the only naturally-occurring isomer ($T_{1/2} > 10^{16}$ y), but very rare.**
- **Other metastable isomers must be manufactured and may be costly.**

Enriched to 4% (from 0.012%) –
1.2 mg ^{180m}Ta in 30 mg TaO_2 - $300,000

... AND CURRENT APPLICATION



Royal Infirmary of Edinburgh



University Hospitals of Cleveland

- **^{99m}Tc costs only $60/gram**
- **$T_{1/2}$ ~ 6 hours**
- **Used in medical imaging, “tagged” to seek specific organ/site**
- **100% γ decay at 140 keV**
- **^{99g}Tc $T_{1/2}$ ~ 200,000 y (Q_β = 294 keV)**
- **Inactive and eliminated in a relatively short time after tests**

LASERS

FOUNDATIONS

1917 Einstein describes stimulated emission of electromagnetic radiation

1926 Eddington "Decay of Radium is a spontaneous event if the radium atom is an isolated system. However, this decay can be stimulated by the gamma-radiation field identical in frequency to the gamma-rays emitted in the decay of Radium ..." *The Internal Constitution of the Stars*

1957 Mössbauer discovers recoilless gamma emission in solids

1950s Basov, Prohorov and Townes achieve MASER (Nobel Laureates 1964)

1960 Maiman realizes first optical laser with ruby

CONCEPT FOR A GAMMA-RAY LASER (GRASER)

1961 Rivlin, Soviet Inventor's Certificate, unclassified early 1980s

1963 Baldwin, Proc. IEEE 51; Vali and Vali, Proc. IEEE 51

QUANTUM ELECTRONICS ⇒ QUANTUM NUCLEONICS

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GRASER SCHEME

HIGH PHOTON ENERGIES ⇒ NO REFLECTORS (OR CAVITY)

Single-pass Amplified Spontaneous Emission (ASE) through a filament

GAIN

$\Phi = \Phi_0 e^{\alpha L}$ where $\alpha = (n_u \sigma_{SE} - n_l \sigma_{Abs}) - n_0 \sigma_{atten} = (n_u - n_l)\sigma_{SE} - n_0 \sigma_{atten}$

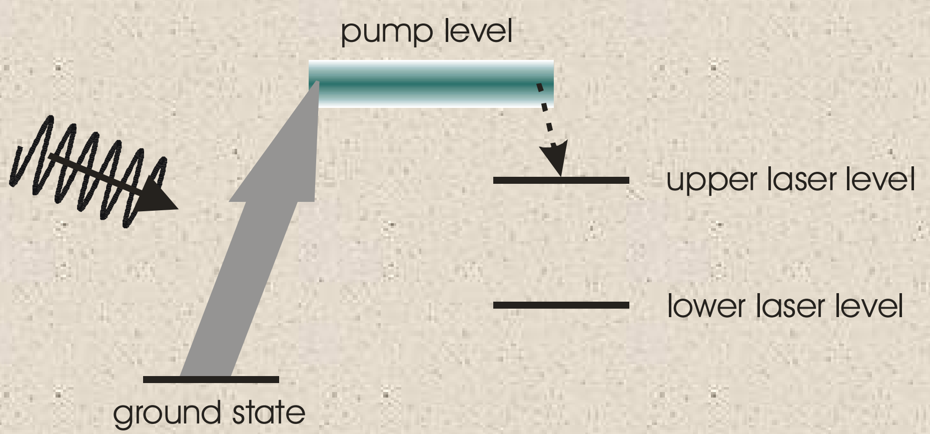
Necessary ⇒ inversion: $n_u - n_l > 0$

Necessary ⇒ photon balance: $(n_u - n_l)/n_0 > \sigma_{atten}/\sigma_{SE} < 10^{-5}$

4-LEVEL LASER SCHEME

$\tau_u \gg \tau_l \Rightarrow n_l \to 0$

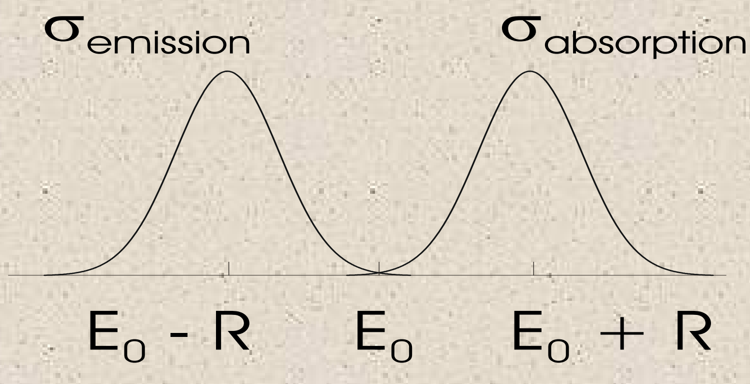
Upper laser level an isomer
(restricted to $T_{1/2} < 10\ \mu s$)



GRASER SCHEME

EMISSION AT LASER TRANSITION

RECOIL of nucleus robs emitted gamma ray of energy $R \sim E^2 / A$

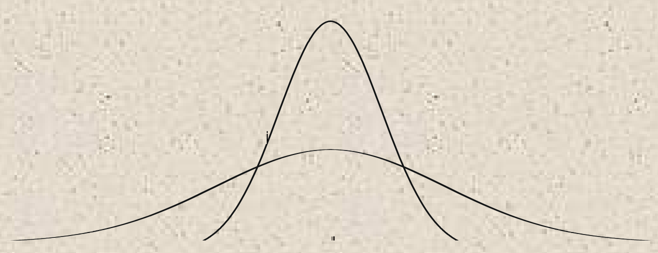


Emitted photon no longer in resonance with additional nuclei to produce stimulated emission

DOPPLER BROADENING of gamma transition robs nuclei of stimulated emission cross section

$$\sigma_{SE} = 2\pi\left(\frac{\hbar c}{E_0}\right)^2 g(J_i, J_f)\frac{\Gamma_{rad}}{\Gamma}$$

'wave' cross section



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COMPARISONS

RUBY LASER	GRASER
$E = 1.79$ eV	$E = 30$ keV
$\tau = 4.33$ ms	$\tau = 1\ \mu s$
$\Gamma_{nat} = 0.15$ peV	$\Gamma_{nat} = 700$ peV
$\sigma_{SE\text{-}Nat} = 7.7 \times 10^8$ Mb	$\sigma_{SE\text{-}Nat} = 2.7$ Mb
$R = 33$ peV	$R = 4.8$ meV ($A = 100$)
$\Gamma_{Doppler} = 5.8$ meV	$\Gamma_{Doppler} = 4.3$ meV (4 K)
$\sigma_{SE\text{-}Doppler} = 20$ kb	$\sigma_{SE\text{-}Doppler} = 0.4$ b
Large enough	**No chance**

RESCUE?

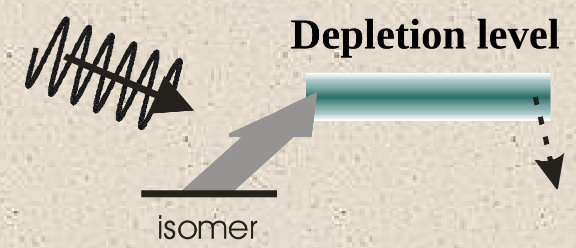
Require Mössbauer emission to prevent both recoil (mismatch with SE resonance) and Doppler broadening (reduction of σ_{SE})

GRASER DILEMMA

- Creating inversion (pumping) likely to destroy Mössbauer effect (sensitive to disruption of crystal environment - see reviews: Baldwin, et al., RMP 1981, 1997; Carroll, et al., HI 2001)

DECADES OF IDEAS …

- Photon pump may be less damaging to crystal, quantum interference (gain without inversion) may reduce pump requirements, hidden inversion in cooled ensembles, etc.
- Use of metastable isomer as "ground state" of 4-level laser scheme may reduce pump requirements



- Even without lasing, process causes induced release of nuclear energy via isomer depletion

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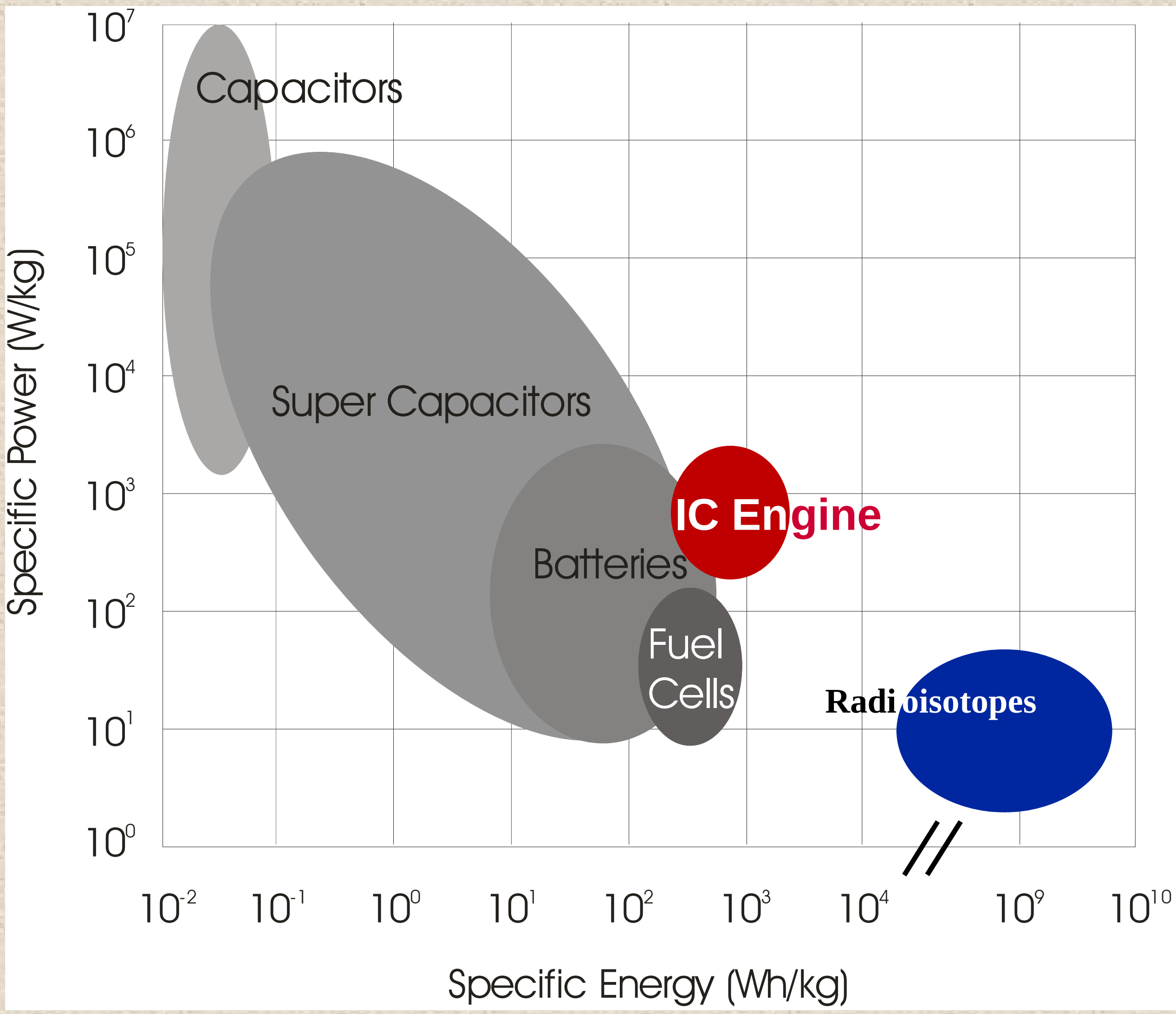
ENERGY STORAGE



EXAMPLE – ^{238}Pu USED BY NASA

Energy storage:

$T_{1/2}$ = 87.74 y

Energy = 6.3×10^8 Wh/kg

Energy use:

$T_{1/2}$ = 87.74 y

Power = 570 W/kg

Inherent limitation of same time scale for storage and release

COULD OUTPUT TIME SCALE BE SHORTENED UPON DEMAND USING NUCLEAR ISOMERS?

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"STORAGE" ISOMERS ($T_{1/2}$ > 1 day)

Isomer properties and production cross sections.

Isomer	$T_{1/2}$	J^{π}	E^{*} [keV]	Production reaction	σ_{th} [b]	I_{γ} [b]
$^{91}Nb^{m}$	61 d	$½^{-}$	105	$^{93}Nb(\gamma, 2n)$		
$^{93}Nb^{m}$	16.1 y	$½^{-}$	31	$^{93}Nb(\gamma, \gamma')$		
$^{97}Tc^{m}$	90 d	$½^{-}$	97	$^{96}Ru(n, \gamma)^{97}Ru \rightarrow$ $^{97}Tc^{m}$	0.29 10^{-4}(eff.)	7.3 $2 \cdot 10^{-3}$(eff.)
$^{102}Rh^{m}$	2.9 y	6^{+}	141	$^{103}Rh(\gamma, n)$		
$^{108}Ag^{m}$	418 y	6^{+}	109	$^{107}Ag(n, \gamma)$	0.33	1.2
$^{110}Ag^{m}$	250 d	6^{+}	118	$^{109}Ag(n, \gamma)$	4.9	72
$^{113}Cd^{m}$	14.1 y	$^{11}/_{2}{}^{-}$	264	$^{112}Cd(n, \gamma)$	< 2	-
$^{114}In^{m1}$	49.5 d	5^{+}	190	$^{113}In(n, \gamma)$	8.1	220
$^{117}Sn^{m}$	13.6 d	$^{11}/_{2}{}^{-}$	315	$^{116}Sn(n, \gamma)$	0.006	0.49
$^{119}Sn^{m}$	293 d	$^{11}/_{2}{}^{-}$	90	$^{118}Sn(n, \gamma)$	0.01	-
$^{121}Sn^{m}$	55 y	$^{11}/_{2}{}^{-}$	6.3	$^{120}Sn(n, \gamma)$	0.001	-
$^{121}Te^{m}$	154 d	$^{11}/_{2}{}^{-}$	294	$^{120}Te(n, \gamma)$	0.34	-
$^{123}Te^{m}$	119.7 d	$^{11}/_{2}{}^{-}$	248	$^{122}Te(n, \gamma)$	0.44	5.1
$^{125}Te^{m}$	57.4 d	$^{11}/_{2}{}^{-}$	145	$^{124}Te(n, \gamma)$ $^{124}Sn(n, \gamma)^{125}Sn \rightarrow$ $^{125}Sb \rightarrow$ $^{125}Te^{m}$	1.1 0.13 10^{-3}(eff.)	1.4 8 0.05 (eff.)

Evaluated Nuclear Structure Data File (ENSDF), Brookhaven National Laboratory

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"STORAGE" ISOMERS ($T_{1/2}$ > 1 day)

Isomer properties and production cross sections.

$^{127}Te^{m}$	109 d	$^{11}/_{2}^{-}$	88	$^{126}Te(n, \gamma)$	0.063	0.64
$^{129}Te^{m}$	33.6 d	$^{11}/_{2}^{-}$	106	$^{128}Te(n, \gamma)$	0.027	0.21
$^{129}Xe^{m}$	8.9 d	$^{11}/_{2}^{-}$	236	$^{128}Xe(n, \gamma)$	0.48	38
$^{131}Xe^{m}$	11.8 d	$^{11}/_{2}^{-}$	164	$^{130}Xe(n, \gamma)$	0.45	16
$^{148}Pm^{m}$	41.3 d	6^{-}	138	$^{148}Nd(d, 2n)$		
$^{166}Ho^{m}$	1200 y	7^{-}	6	$^{165}Ho(n, \gamma)$	3.5	20
$^{174}Lu^{m}$	142 d	6^{-}	171	$^{175}Lu(\gamma, n)$		
$^{177}Lu^{m}$	161 d	$^{23}/_{2}^{-}$	970	$^{176}Lu(n, \gamma)$	2.8	4.7
$^{178}Hf^{m2}$	31 y	16^{+}	2446	$^{181}Ta(p, \alpha)$		
$^{179}Hf^{m2}$	25 d	$^{25}/_{2}^{-}$	1106	$^{178}Hf(n, \gamma)$	-	-
$^{180}Ta^{m}$	> 10^{16} y	9^{-}	75	Stable		
$^{184}Re^{m}$	169 d	8^{+}	188	$^{184}W(d, 2n)$		
$^{186}Re^{m}$	$2 \cdot 10^{5}$ y	8^{+}	149	$^{185}Re(n, \gamma)$	0.3	~ 5
$^{192}Ir^{m}$	241 y	9	155	$^{191}Ir(n, \gamma)$	0.16	~ 0.5
$^{193}Ir^{m}$	10.5 y	$^{11}/_{2}^{-}$	80	$^{192}Os(d, n)$		
$^{193}Pt^{m}$	4.33 d	$^{11}/_{2}^{-}$	150	$^{192}Pt(n, \gamma)$	2.2	~ 20
$^{195}Pt^{m}$	4.02 d	$^{11}/_{2}^{-}$	259	$^{194}Pt(n, \gamma)$	0.1	3.1
$^{242}Am^{m}$	141 y	5^{-}	49	$^{241}Am(n, \gamma)$	54	195

Stores longest – naturally available

Stores most energy (1.3 billion Joules/gram)

Evaluated Nuclear Structure Data File (ENSDF), Brookhaven National Laboratory and other literature

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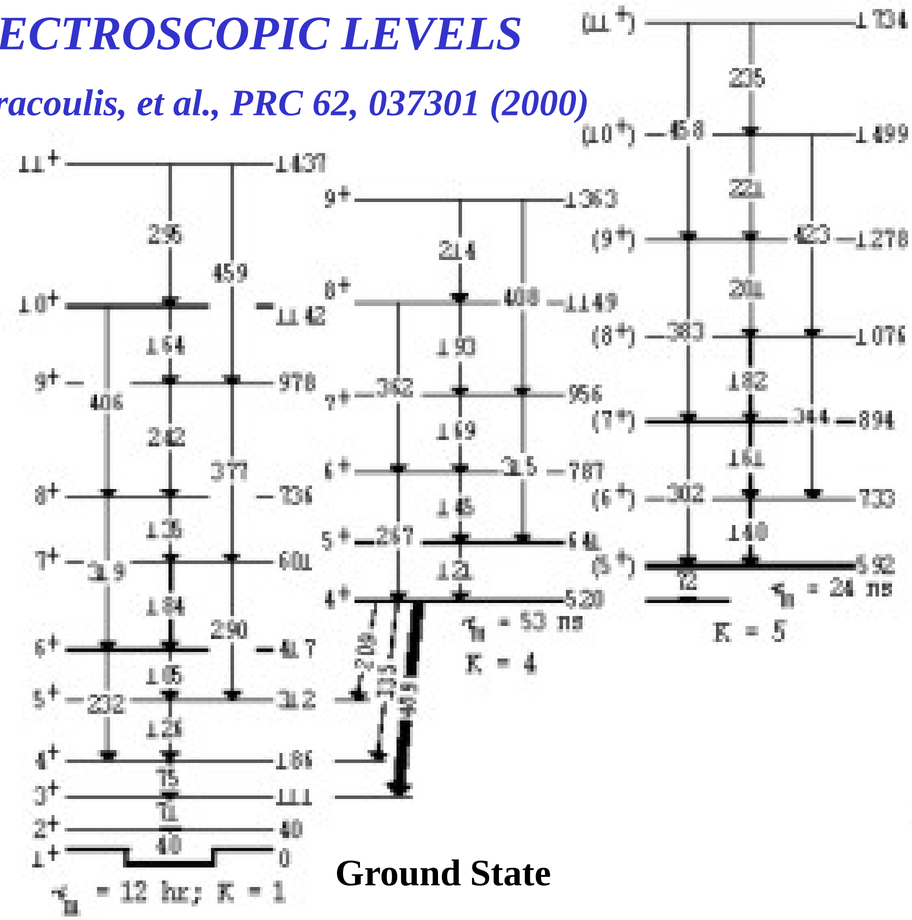
SPECTROSCOPY …

^{180m}Ta

SPECTROSCOPIC LEVELS

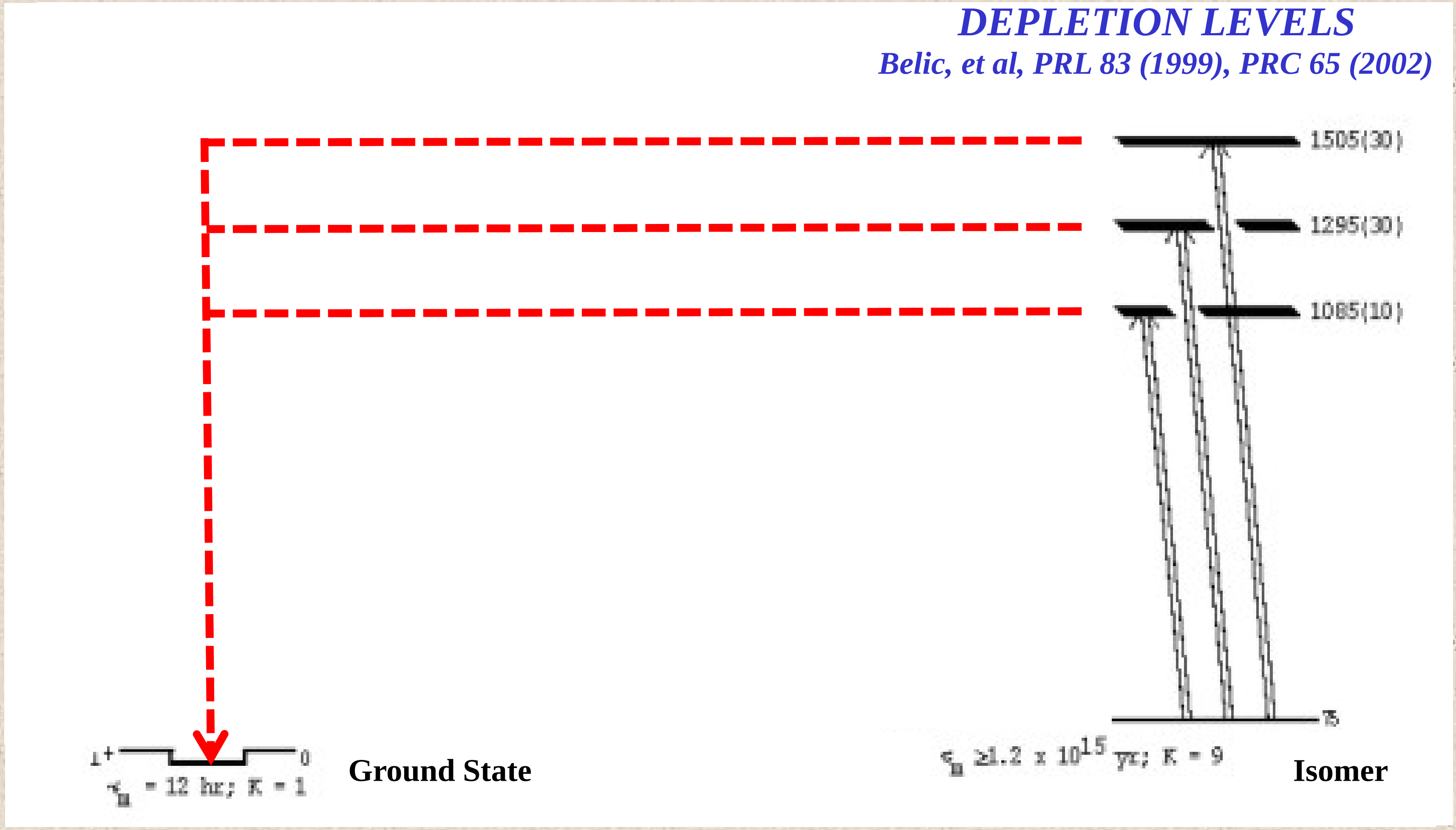
Dracoulis, et al., PRC 62, 037301 (2000)



Ground State

… AND DEPLETION

^{180m}Ta



Depletion first demonstrated Collins, et al., PRC 37, 1988

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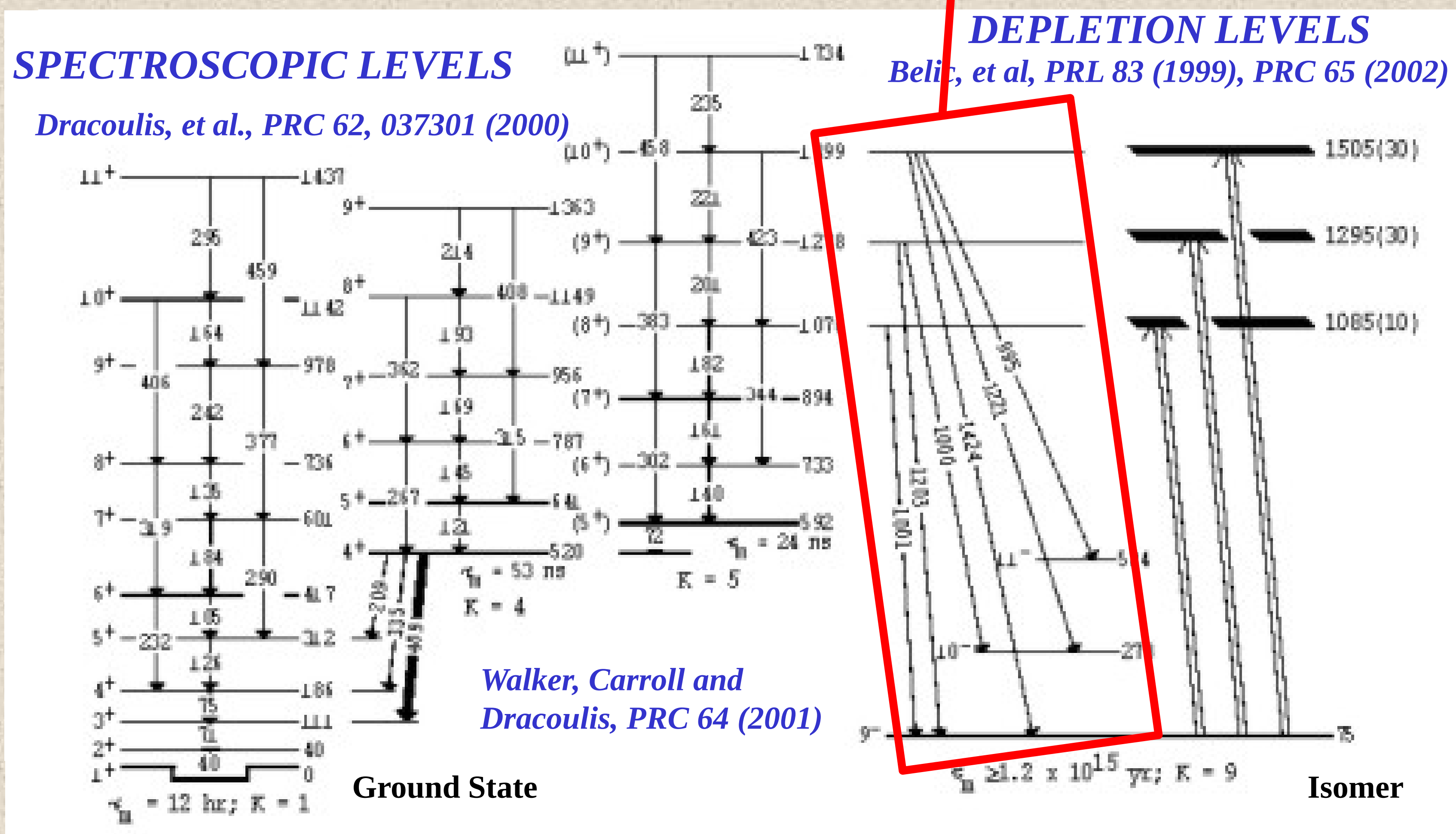
PARTIAL SYNTHESIS

^{180m}Ta

"Back" decays have not been observed



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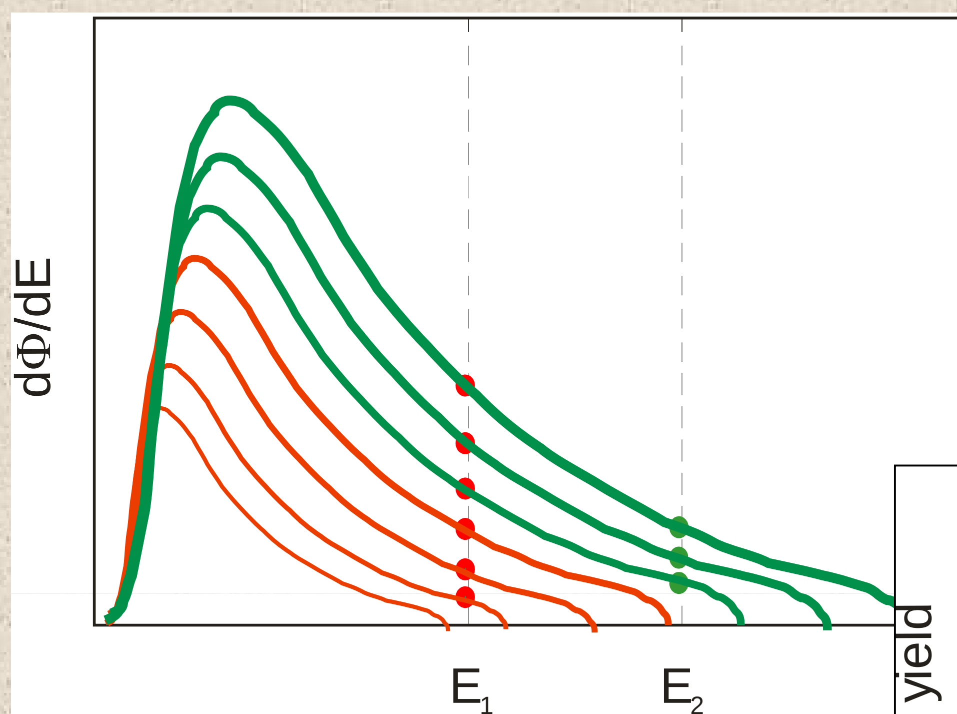
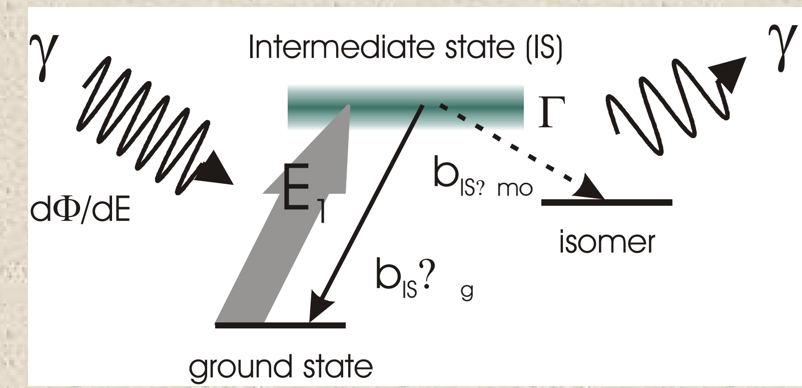
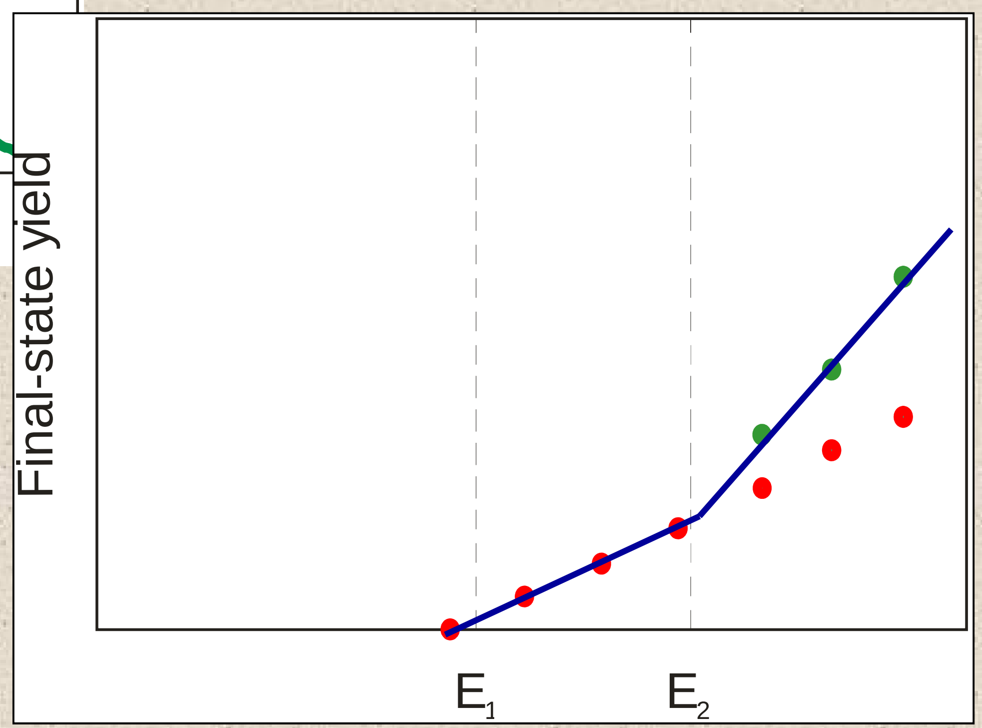
ISOMER PHOTOEXCITATION







Described by an integral cross section (ICS)

$$ICS = 2\pi^2 \left(\frac{\hbar c}{E_{IS_i}}\right)^2 \left(\frac{2I_{IS_i}+1}{2I_g+1}\right) \frac{\Gamma^{\gamma}_{IS\to g}\Gamma_{IS\to m}}{\Gamma}$$

so that

$$Yield = N_m = N_g \sum_{IS} \left(ICS_{IS_i}\right) \left(\left.\frac{d\Phi}{dE}\right|_{IS_i} T\right)$$

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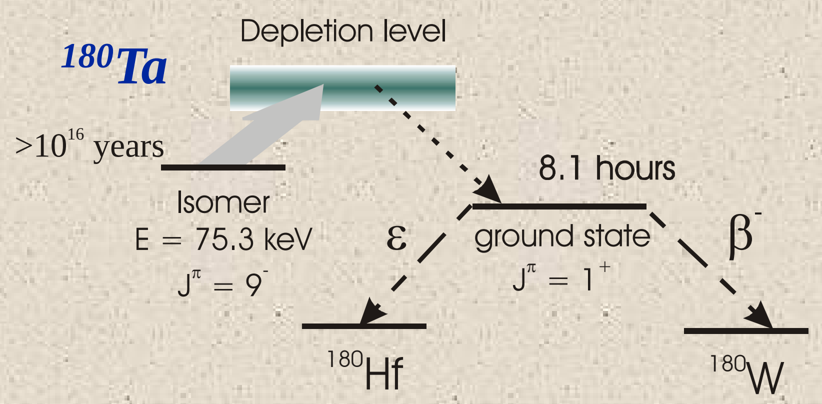
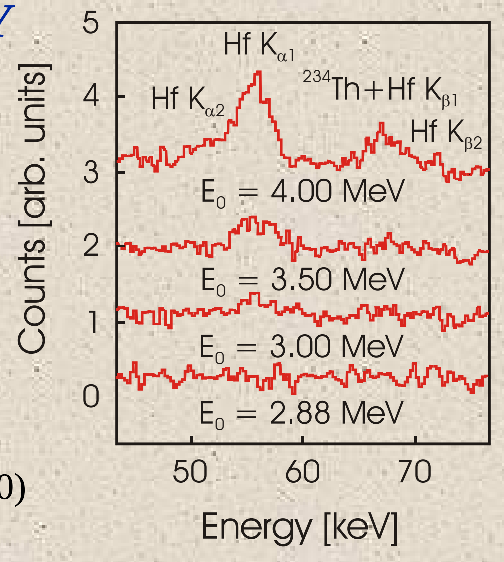
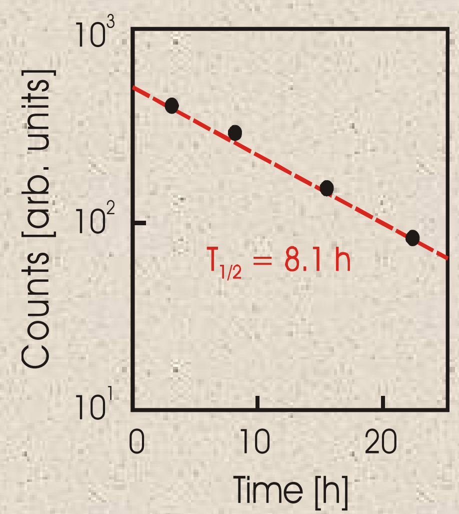
^{180m}Ta DEPLETION

DETECTED BY GROUND-STATE DECAY

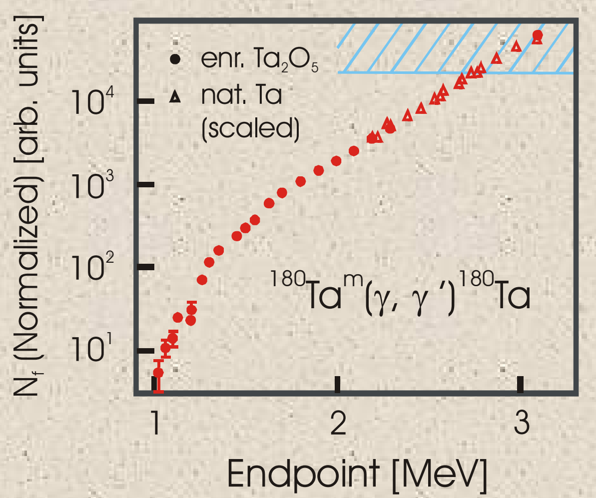






Collins, et al. PRC 37, 2267 (1988), PRC 42, R1813 (1990)
(S-DALINAC at TU Darmstadt)

Belic, et al., PRL 83, 5242 (1999) and PRC 65, 035801 (2002) (natural and enriched targets, U. Stuttgart)



IS Energy [MeV]	(σΓ) [eV b]
1.01 (1)	0.057 (3)
1.22 (2)	0.27(2)
1.43(2)	0.24(4)
1.55(3)	0.70(9)
1.85(5)	1.11(14)

IS Energy [MeV]	(σΓ) [eV b]
2.16(2)	2.8(3)
2.40(6)	3.5(6)
2.64(3)	13(1)
2.8(4)	36(2) - prev. 120(20)

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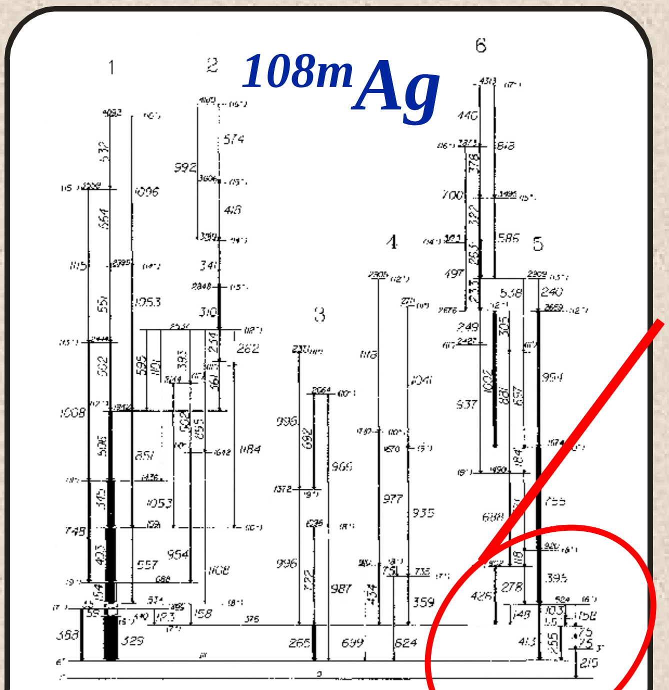
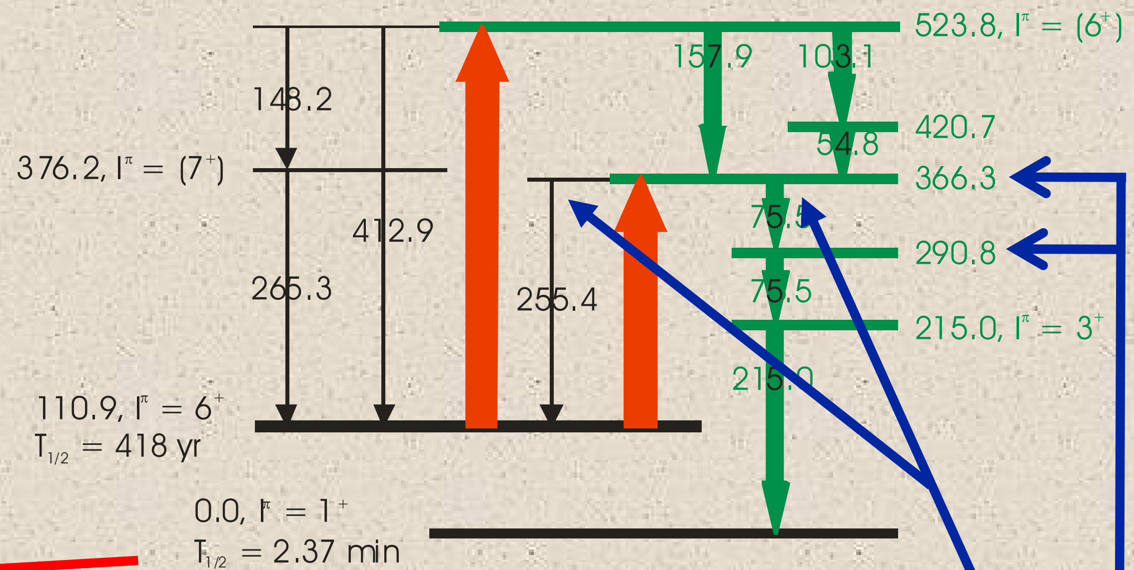
DEPLETION POSSIBILITIES - ^{108m}Ag



F. R. Espinoza-Quinones, et al., PRC 52, 104 (1995)

- Isomer $T_{1/2}$ = 418 years, gs $T_{1/2}$ = 2.37 min
- Two possible depletion paths seen in level scheme



- Depletion by continuous photon spectrum represented by integral cross section (ICS)

$$ICS = 2\pi^2 \left(\frac{\hbar c}{E_{depl}}\right)^2 \left(\frac{2I_{depl}+1}{2I_m+1}\right) \frac{\Gamma^{\gamma}_{depl\rightarrow m}\Gamma_{depl\rightarrow g}}{\Gamma}$$

UNKNOWN:

- Branching ratios from 366 keV level
- Ang. momenta of depletion level & other cascade states

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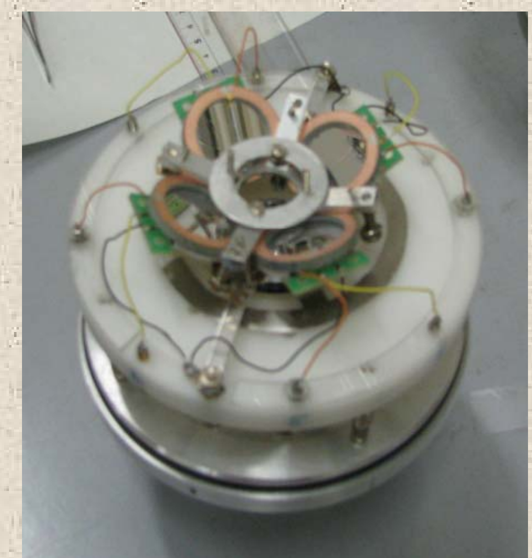
SPECTROSCOPY - ^{108m}Ag

- JAEA Tokai tandem accelerator – ^{18}O beam at 135 MeV
- Free-standing ^{107}Ag target, 4 mg/cm^{2} enriched to 98.22% (ORNL) from natural 51.839%; remaining 1.88% ^{109}Ag
- Detector array with eight HPGe, including four 60% relative efficiency HPGe placed symmetrically around beam axis
- Four ΔE-E Si(Li) telescopes
- 70 hours beamtime
- p-γ^{n} coincidences available



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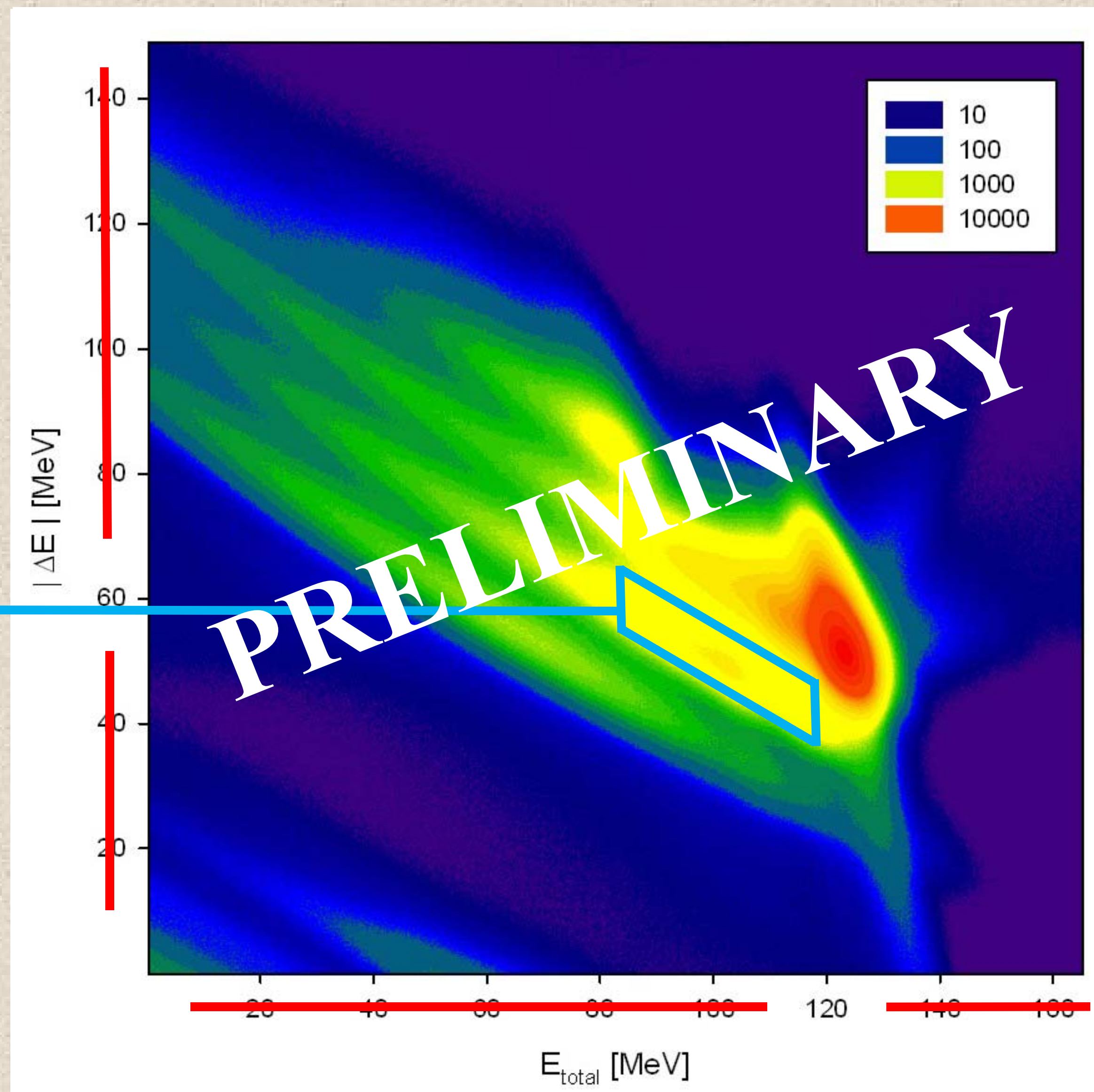
SPECTROSCOPY - ^{108m}Ag

- Many reactions possible – dominant reactions are elastic scattering of ^{18}O and Coulex by ^{18}O
- Primary interest in ^{108}Ag produced by single-neutron transfer – projectile-like ^{17}O ions
- Gate on projectile-like ^{17}O restricted to avoid more than 7.2 MeV excitation into ^{108}Ag (S_n = 7.29 MeV)



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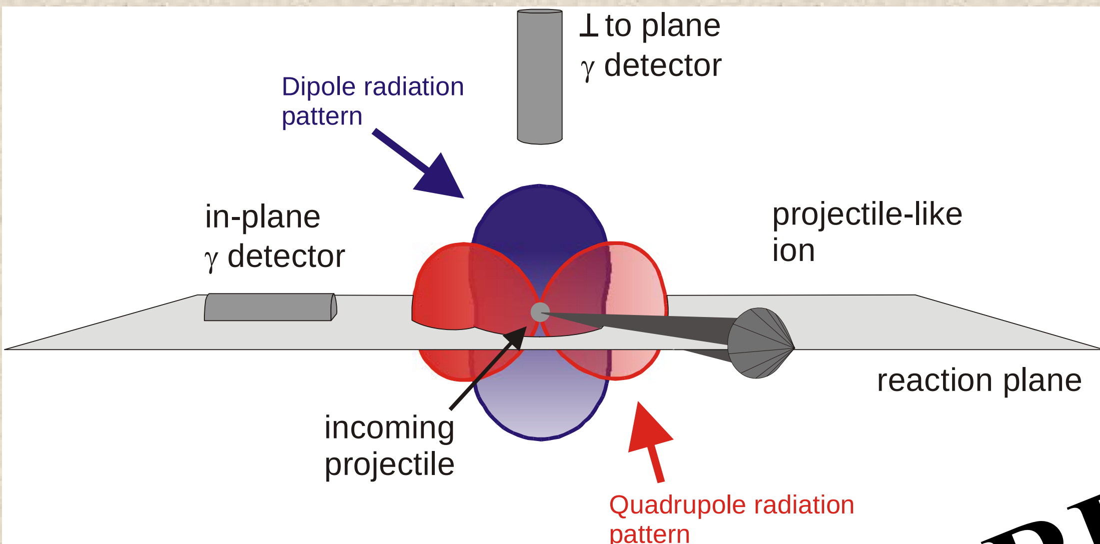
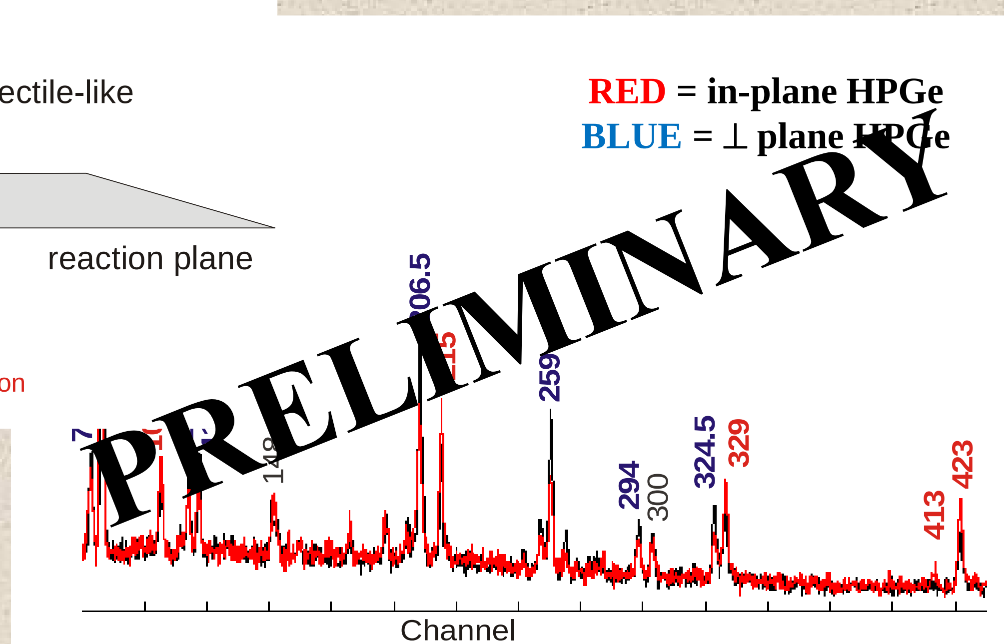
SPECTROSCOPY - ^{108m}Ag

- Gate on ^{17}O projectile-like ions
- Extract singles first (gain matched) from 4 symmetrically-placed HPGe detectors
- Partial alignment of target-like nuclei after reaction leads to gamma angular correlations with respect to reaction plane (here, P_{zz} ~ 0.85)
- Dipole transitions $I_{\perp plane} > I_{in\ plane}$; quadrupole transitions $I_{\perp plane} < I_{in\ plane}$





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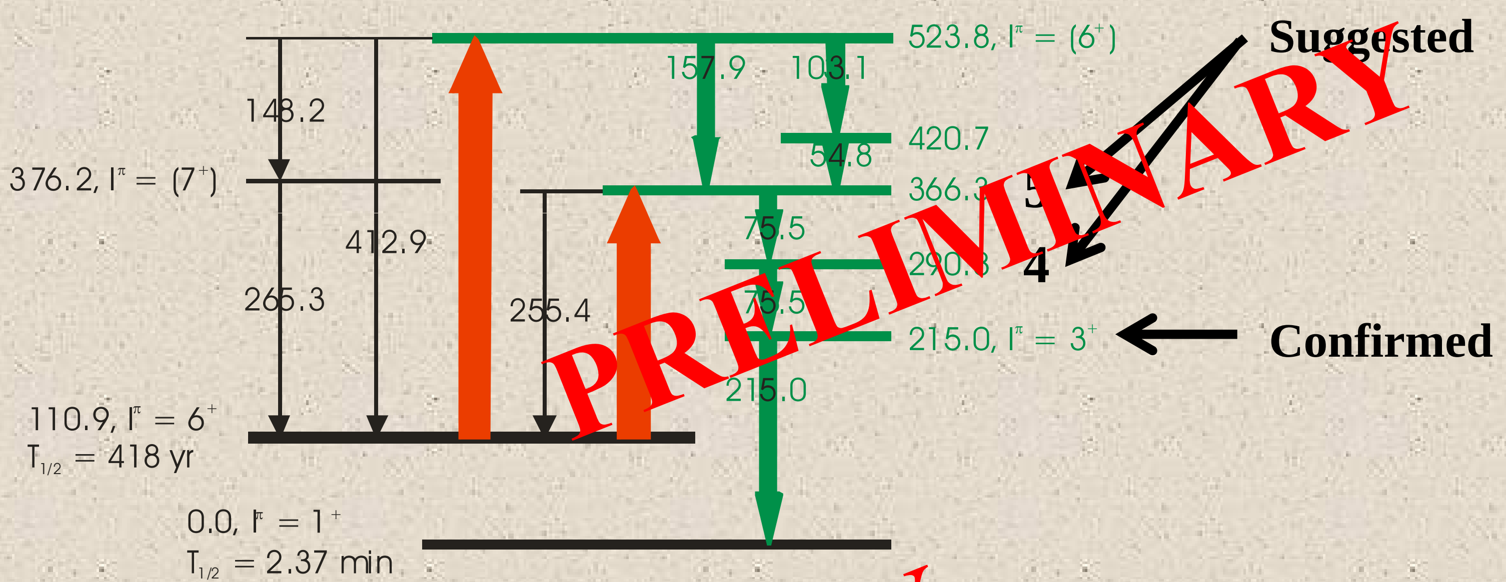
INITIAL ANALYSIS ^{108m}Ag

- Dipole transitions: 75.5 keV, 255.4 keV
- Quadrupole transition: 215.0 keV



- Maximum transition rates if assume electric dipole transitions for 75.5 keV, 255.4 keV
- Use Weisskopf single-particle formulae to estimate unknown branching ratios

$$\Rightarrow \quad ICS \cong 2.3\times10^{-26}\ cm^2\ keV$$

FULL ANALYSIS UNDERWAY

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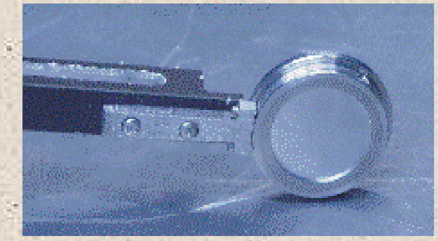
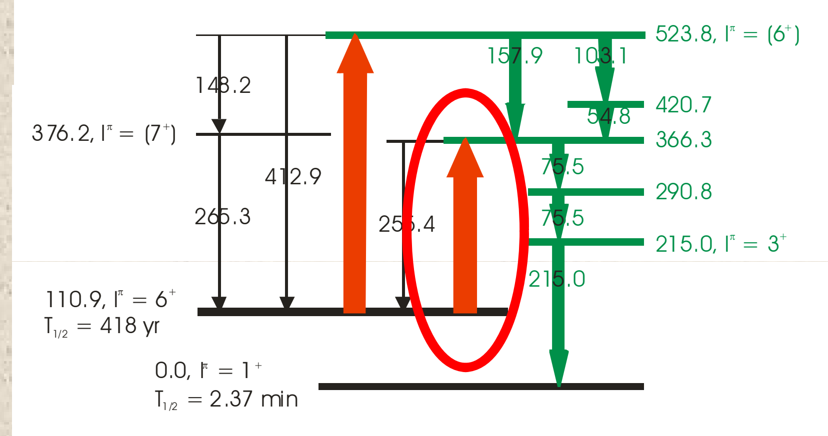
DEPLETION TEST - ^{108m}Ag

- Sample available containing 5.4×10^{16} nuclei of ^{108m}Ag (9.7 μg, 77 μCi) – courtesy of the Naval Research Laboratory

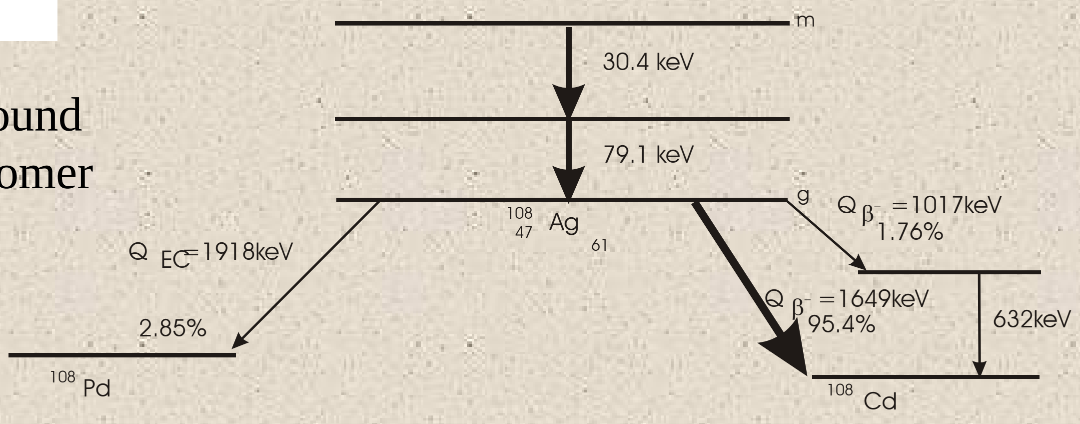


- Encapsulated by Al cover and frame – 50 μm thickness on one side



- Potential depletion transition at 255 keV may be excited by photons – 450 keV (endpoint) industrial bremsstrahlung source at YSU's X-Ray Effects Laboratory, 100% duty cycle (oil-cooled)

- Sample exhibits decay of ^{108}Ag ground state in secular equilibrium with isomer
- Depletion would produce excess ground-state population and decay ($T_{1/2}$ = 2.37 min)



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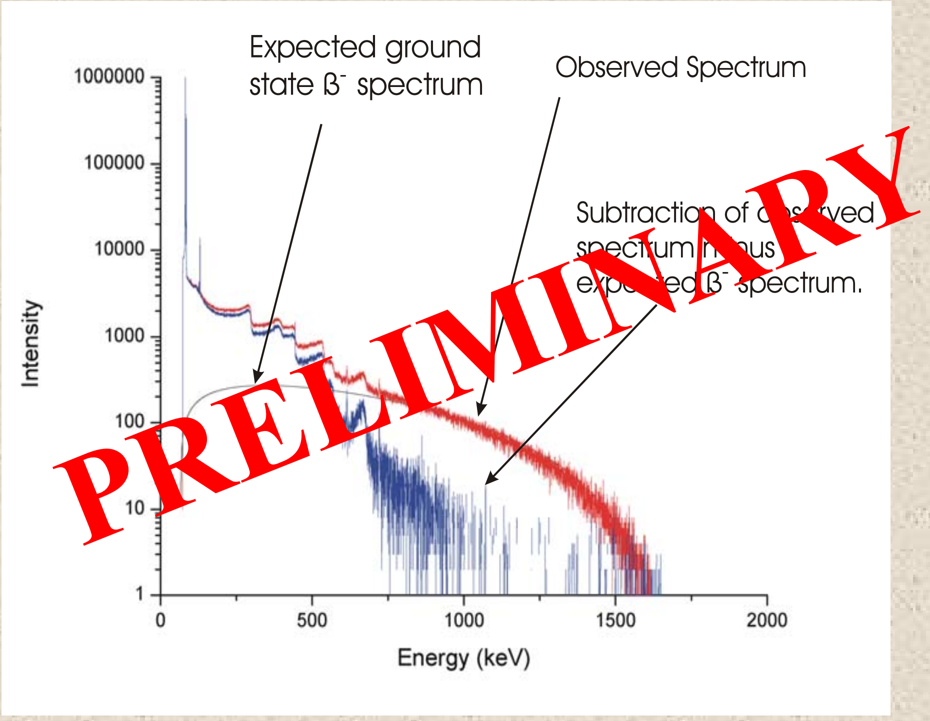
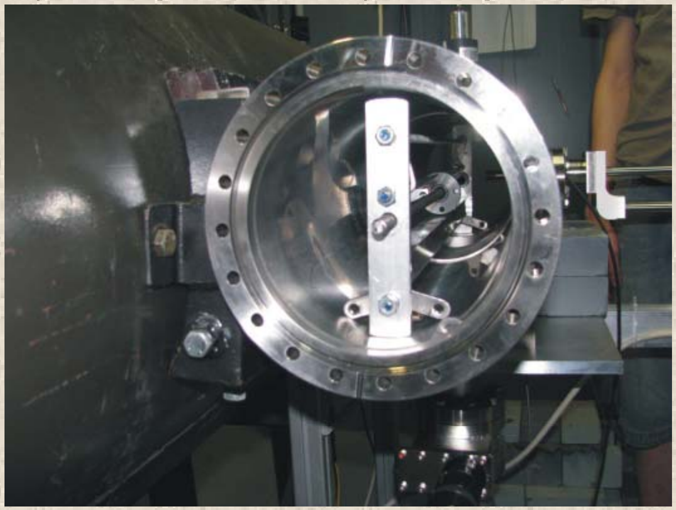
DEPLETION TEST ^{108m}Ag

- Vacuum chamber constructed
  - Labview control system
  - Si(Li) detector on extendable arm
  - Stepper-motor-driven linear transportation for sample





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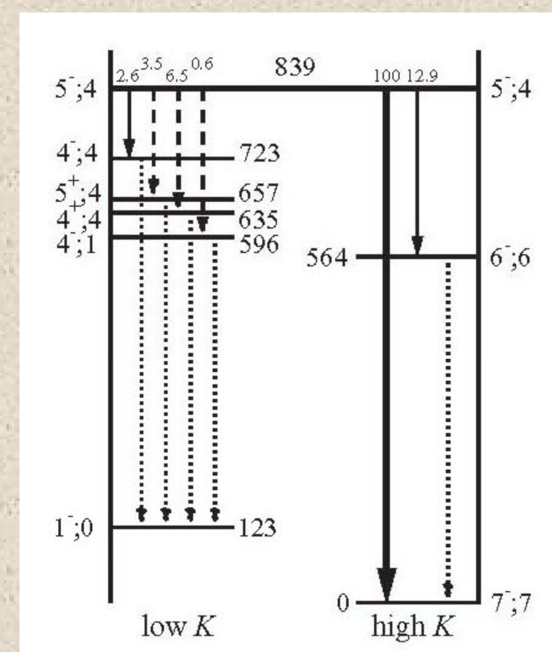
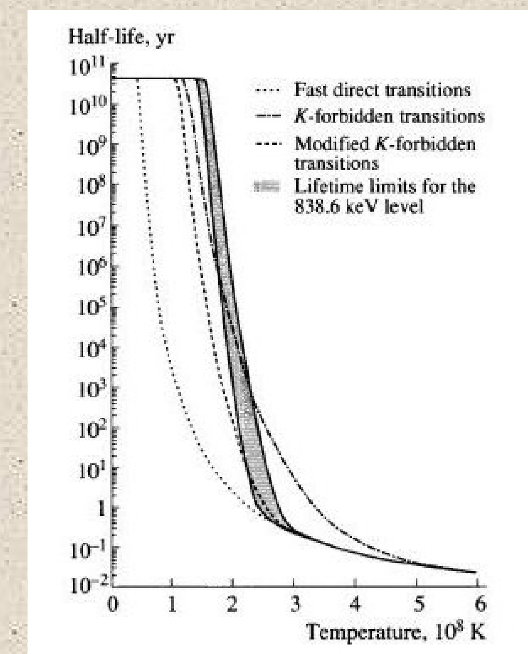
PHOTOACTIVATION - ^{176m}Lu

MOTIVATION

- Proposed as s-process thermometer – natural abundance on Earth dependent on temperature (and photon bath) within stellar environment – lowest-lying intermediate states will be most important
- Photoactivation of ^{176}Lu mimics typical isomer depletion sequence – high-spin level to low-spin level via an intermediate state





- In-beam photon scattering (nuclear resonance fluorescence) could not detect intermediate states below 1.759 MeV

Energy (keV)	I_s (eVb)	$g\Gamma_0^{red}$ (meV/MeV^3)
1759	5.05(122)	0.050(12)
2087	5.66(65)	0.047(5)
2243	4.99(95)	0.039(7)
2249	9.57(147)	0.074(11)
2807	7.08(61)	0.044(4)

Mohr, et al., PRC 79, 045809 (2009)

Stedile, et al., LP 14, 442 (2004)

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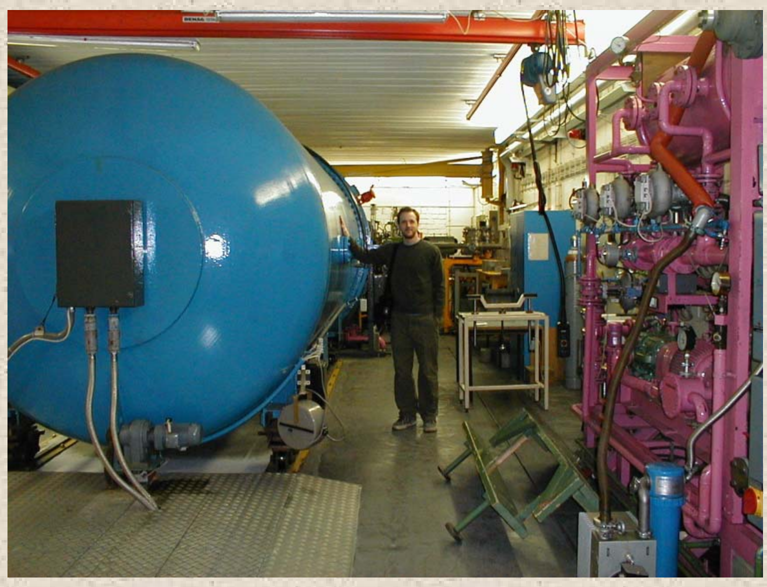
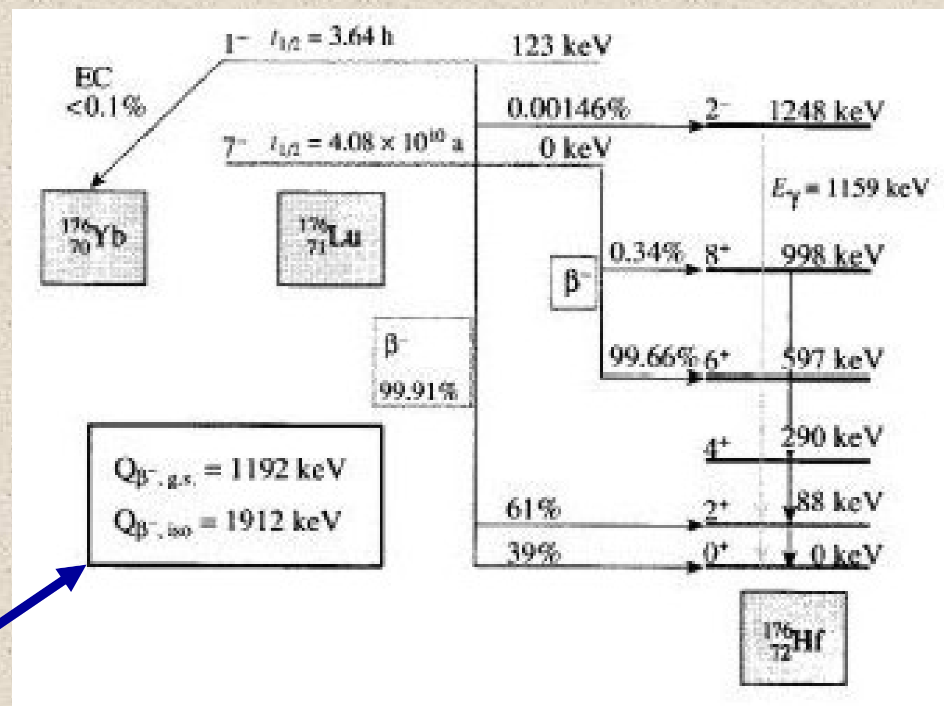
EXPERIMENT

- Irradiation with bremsstrahlung from the (now decommissioned) Stuttgart Dynamitron
- Unique facility: I_{max} ~ 400 μA, E_0 = 0.8 – 4.3 MeV
- Stuttgart/Karlsruhe/Darmstadt/Sofia/YSU collaboration
- Was to have been part of a PhD thesis ~ 2004 – became a forensic project in 2009





Stedile, et al., LP 14, 442 (2004)

- Clear signal from isomer beta decay
- Large-area SSB detector for detection

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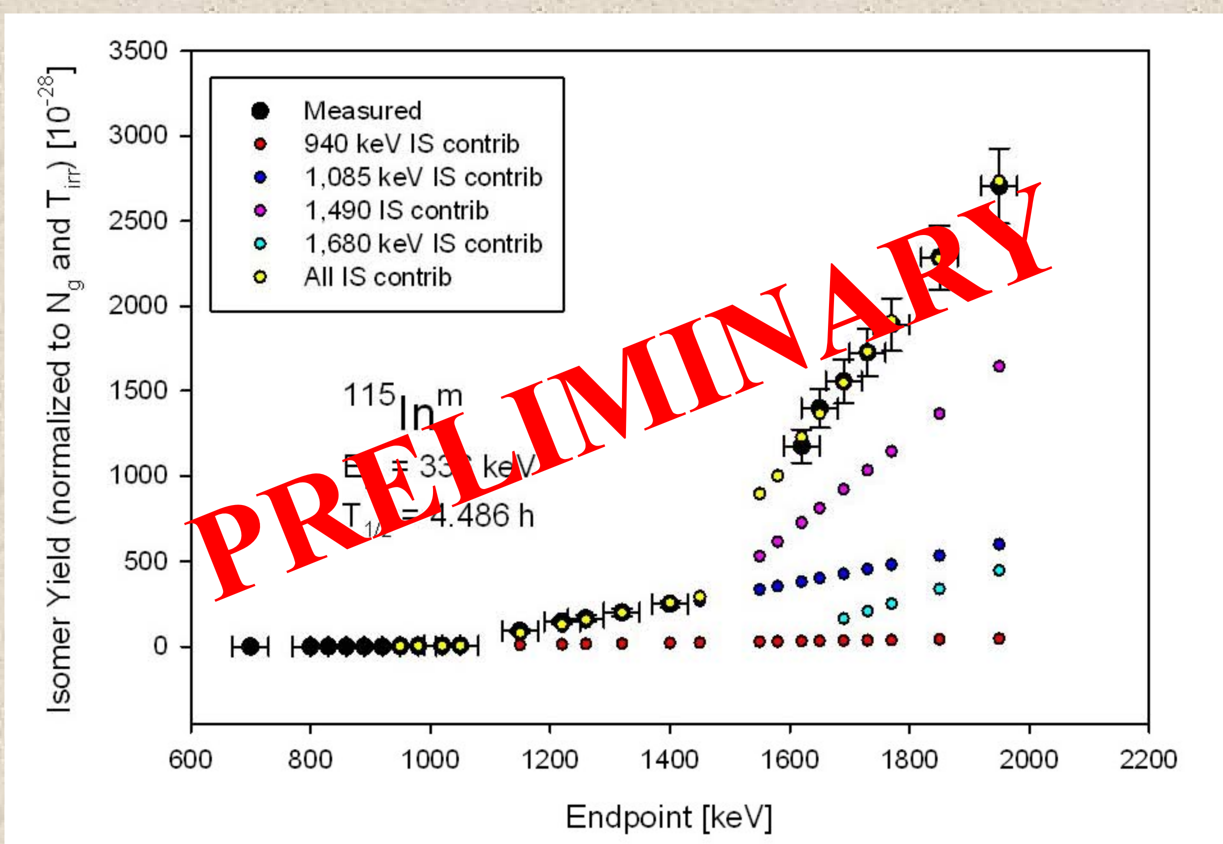
PHOTOACTIVATION - ^{176m}Lu

BREMSSTRAHLUNG SPECTRUM CALIBRATION USING ^{115}In

- Well known intermediate states at 940 keV, 1,085 keV, ,1490 keV, 1,680 keV
- Convenient isomer lifetime and gamma decay signature



- ^{nat}In foil (95% ^{115}In) irradiated in stack with Al-encapsulated enriched $^{176}LuO_2$ sample
- In foil counted with HPGe detector in parallel with beta detection of Lu sample
- Photon spectral intensities at each endpoint by GEANT, normalized to ^{115}In activation

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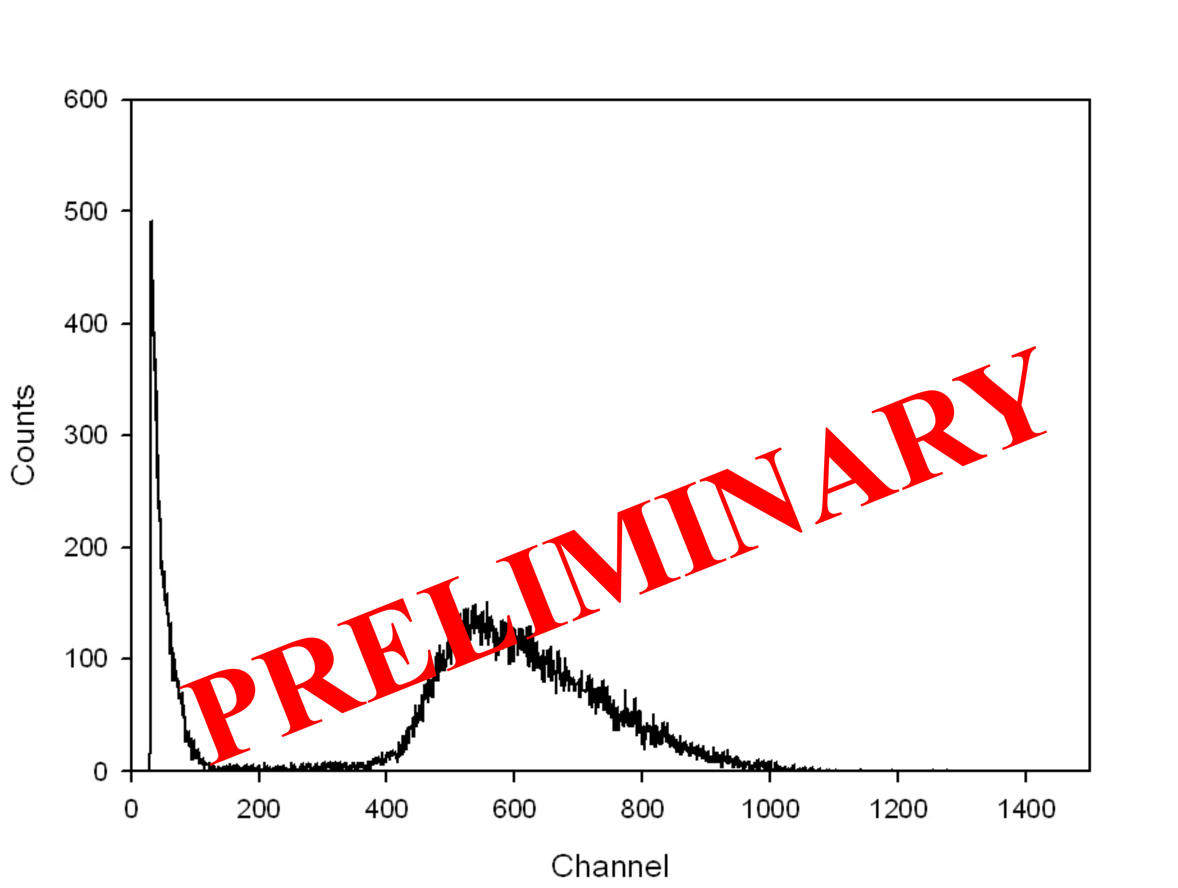
PHOTOACTIVATION - ^{176m}Lu

Lu BETA SPECTRUM

- Strong absorption of lower part of isomer spectrum and ground-state spectrum due to Al filter (in addition to Al encapsulation of sample)
- Detection efficiency for ^{176m}Lu betas by comparison with gamma efficiency (simultaneous measurement) and measured photoactivation at highest endpoint irradiation
- Beta efficiency in agreement with GEANT simulations



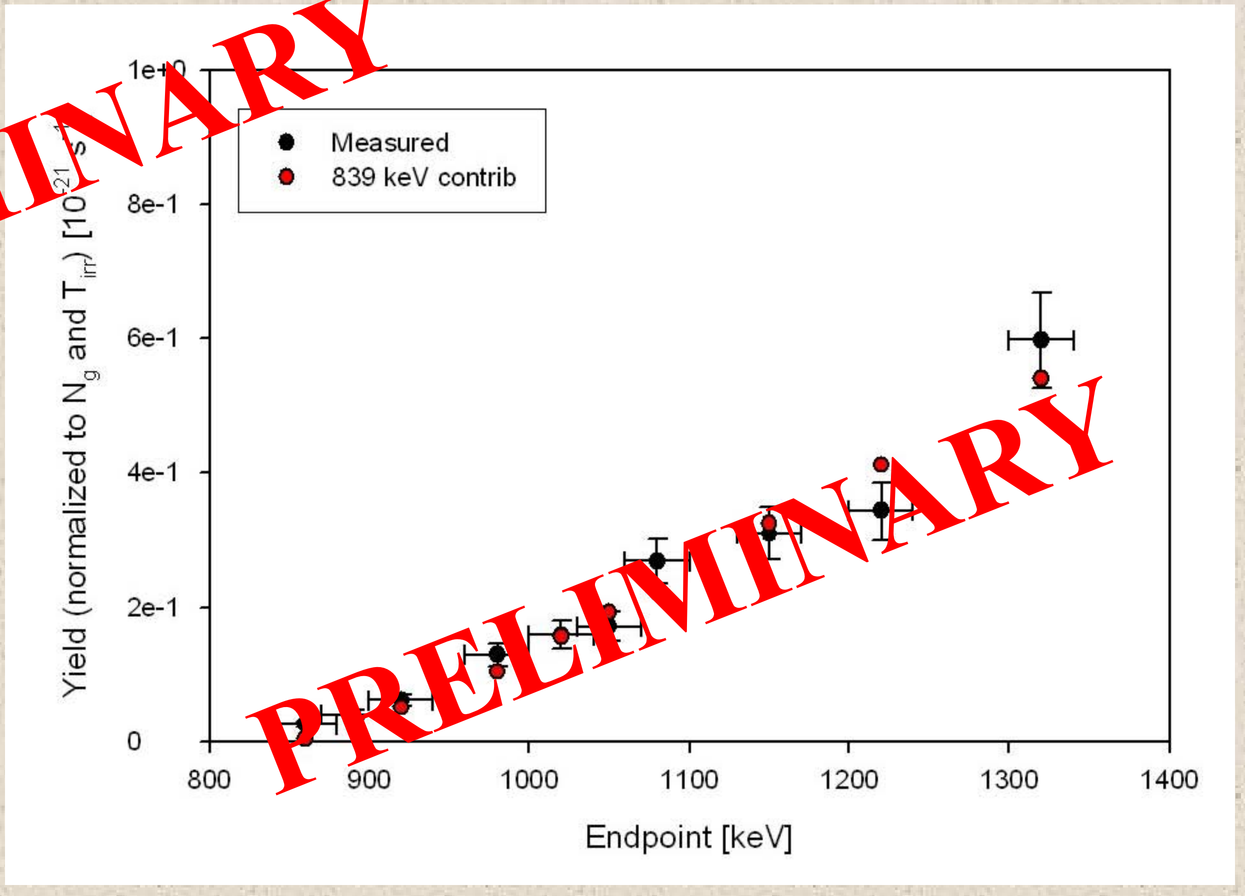
PHOTOACTIVATION - ^{176m}Lu

YIELD CURVE

- Strong absorption of lower part of isomer spectrum and ground-state spectrum due to Al filter (in addition to Al encapsulation of sample)

PRELIMINARY!

- Best fit for intercept ⇒ 839 keV
- Best fit for ICS ⇒ 1.21 × 10^{-29} cm^2 keV
- Mohr, et al. PRC 79 (2009) estimated 1.2 × 10^{-29} cm^2 keV without actual data and based on some assumptions



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SUMMARY/STATUS

APPLICATIONS

- Wait and see – need more data on depletion of metastable isomers to determine feasibility as energetic materials
- Graser remains unrealized (new ideas welcome)

INDUCED DEPLETION DEMONSTRATED

- ^{180m}Ta – 1987 through 1999 photons and Coulex (various sites)
- ^{177m}Lu – 2006 inelastic scattering of slow neutrons (CEA)
- ^{68m}Cu – 2007 Coulex (CERN REX-ISOLDE)
- ^{178m2}Hf – 2009 inelastic scattering of slow neutrons (Flerov Lab, JINR)

ON THE HORIZON

- Depletion tests for ^{108m}Ag (418 y) and ^{166m}Ho (1,200 y)
- Spectroscopy of ^{186}Re (isomer 200,000 y)
- Atomic nuclear interactions (NEET, NEEC) may prove valuable

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