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A Systematic Study of Photonuclear Reactions for Isotope Production

March 2024

Changing the World's Energy Future

Douglas P. Wells, Van Romero, Edna S Cardenas, Michael A Reichenberger, Tony Forest

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A Systematic Study of Photonuclear Reactions for Isotope Production

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Idaho National Lab: **Edna Cárdenas and Michael Reichenberger**

Idaho State University: **Daniel Dale and Tony Forest**

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NEW MEXICO TECH SCIENCE • ENGINEERING • RESEARCH UNIVERSITY

Idaho State University

INL Idaho National Laboratory

Norfolk, Virginia AccelApp'24

Who? Team of Three Institutions (and includes research at multiple nuclear labs)

The Team of PIs

New Mexico Tech (NMT):
Faculty: D. Wells, V. Romero,

Idaho State University (ISU):
Faculty: D. Dale, T. Forest,

Idaho National Lab (INL):
INL Staff Scientists:
E. Cárdenas, M. Reichenberger,

Nuclear Labs Involved

- Idaho National Lab
- John's Hopkins University's Applied Physics Lab – "possible"
- ISU's Idaho Accelerator Center
- Argonne National Lab "LEAF" facility – "probable"
- Rensselaer Polytechnic Institute – "probable"
- NMT's developing lab

What are the fundamental challenges that are motivating this work?

And why are they important?

The U.S. (and the rest of the world) lacks:

1. Facilities to produce radio-isotopes, primarily for medicine, but also for research and many other applications,
2. People with the requisite expertise, especially young people,
3. Methods to produce next-gen isotopes for research, medicine and industry,
4. Nuclear Data to make informed decisions about next-gen methods and facilities.

Why does isotope production matter?

- What are the needs, challenges and opportunities? National use in isotopes, especially medical isotopes, is absolutely critical for the nation's medical care.
  - In the U.S., roughly 1.5 MILLION Americans per month receive a diagnostic or therapeutic procedure that uses radio-isotopes. This is the primary diagnostic tool to detect and track the progress of cancer and cancer therapy.
- Yet the U.S. does not have a domestic supply for many of the most heavily used or most promising isotopes.
  - Electron linacs and photonuclear reactions from bremsstrahlung beams can be a part of the solution for the national supply, which leads to many challenges and related opportunities.
- DOE/NSF Nuclear Science Advisory Committee (NSAC) recommendations:
  - "Meeting Isotope Needs and Capturing Opportunities for the Future: The 2015 Long Range Plan for the DOE-NP Isotope Program", www.osti.gov/biblio/1298983

Why photonuclear?

- There is a remarkable dearth of photo-nuclear data, facilities and people.
- Of roughly 300 stable nuclear species, only ~10% of (γ,n), (γ,p), (γ,np), and (γ,α) reaction cross sections have been experimentally measured over the (isovector) Giant Dipole Resonance range, and less if one goes to energies above the GDR.
- And photonuclear reactions offers the potential for an unexploited and (relatively) inexpensive and low nuclear-waste approach to isotope production.

What are the tasks?

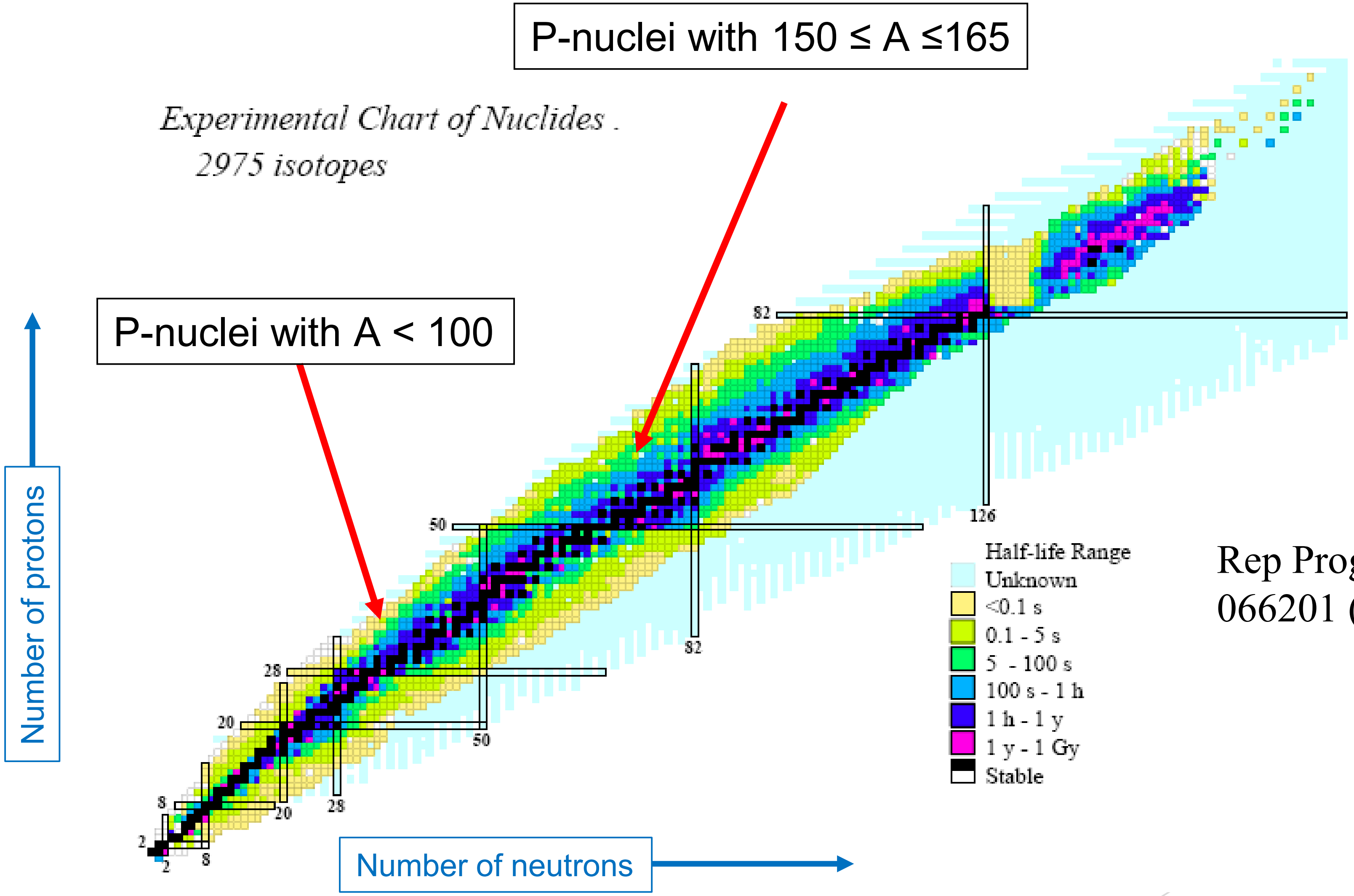
This team's (NMT + Idaho State University + Idaho National Lab) Four Major Tasks are:

1. Measure bremsstrahlung-weighted excitation functions and infer cross sections to fill in some of the many gaps in the world's photonuclear data, including (γ, α), (γ, p), (γ, n), (γ, np), and possibly other reaction channels.
2. Measure Figures-of-Merit (FOM) for isotope production for selected reactions versus bremsstrahlung end-point energy, both for enriched and natural targets.
3. Investigate kinematic recoil separations of radioisotopes to address the isotope separation challenge of using (γ, n) reactions for isotope production.
4. And, most important of all, educate new talent who go on to contribute to the nation's isotope programs.

What are the important questions and applications of photonuclear physics, and why do they matter?

- What are some of the fundamental and applied nuclear physics questions?
  - What do we know about the resonant "Normal Modes" of excited nuclei, sum rules and violations thereof, cross sections for specific "exclusive" nuclear reactions, and nuclear structure implications?
  - What are the contributions of photonuclear reactions to astrophysical origins of natural "proton-rich" nuclei?
  - Isotope Production: primarily for medical applications
  - Activation Analysis: forensics, nuclear non-proliferation, trace element analysis, and related applications
  - Next-Gen reactor materials
  - Radiation effects, biological or materials/devices, especially space systems
  - Accelerator-Driven Subcritical Systems (i.e. – not quite a nuclear reactor) and nuclear waste "burn-up"

Why else does this research matter? We are mostly investigating p-rich isotope production via (γ, n) or other reactions. Beyond isotope production, other implications include the astrophysical origin of proton-rich isotopes…



Rep Prog Phys **76**, 066201 (2013)

What are photonuclear reactions?

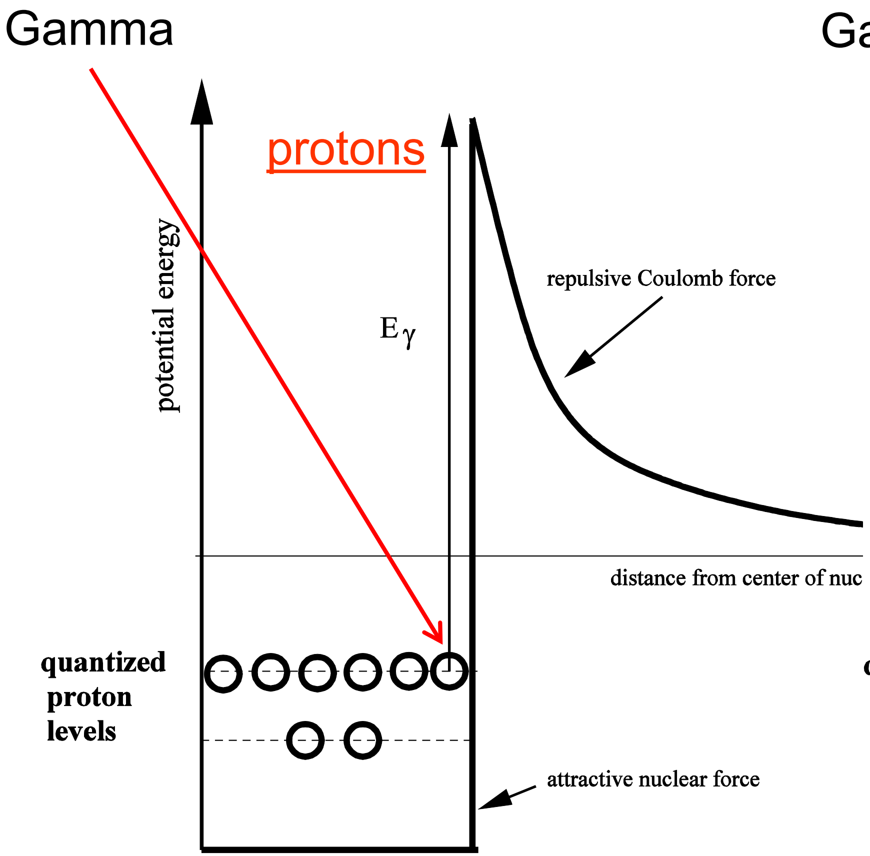
- The semi-classical picture is that:
  - photons (gammas) are absorbed by the nucleus which, in turn,
  - Typically statically "equilibrates" (multi-nucleon excitation) before emitting particle(s) to de-excite:
  - this process heavily favors neutron emission for metals (medium to high atomic number)

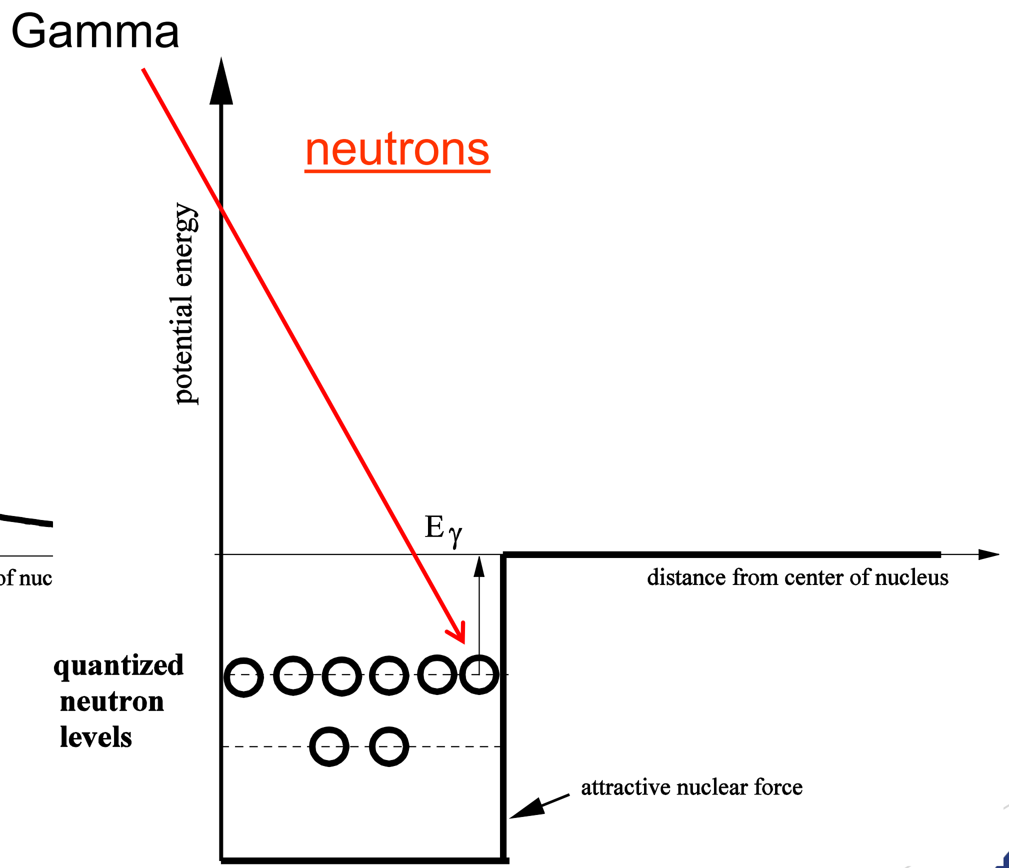
In general, nuclear reactions include the interactions of three* of the four known fundamental interactions:

- (Residual) Strong Nuclear Force (mediated by π's, ρ's, etc.),
- Electromagnetic Force (mediated by photons),
- Weak Nuclear Force (mediated by W's and Z's).

* - one can plausibly also include the gravitational force, when one notes that a neutron star is also a nucleus.

What are Photonuclear Reactions? Photons (gammas) are absorbed by the nucleus which, in turn, typically "equilibrates" (multi-nucleon excitation) before emitting particle(s) to de-excite: favors neutron emission for metals (medium to high atomic number)





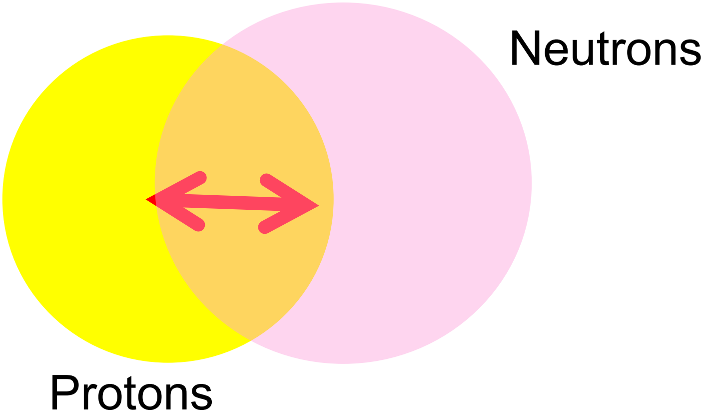
Coulomb energy barrier for protons … but not for neutrons.

Photonuclear reactions in the ≈ 10 - 50 MeV range are dominated by statistical "compound nucleus" processes that trigger the "Normal Modes" of a nucleus.

The "normal modes" of a nucleus are the "natural" resonances - a superposition of many excited states - which, from a time-scale perspective, requires an equilibration process that is called the "compound nucleus" (developed in the 1930s to explain statistical properties of nuclear fission and the neutron resonance widths (~ eV, or less) that induce nuclear fission):

Uncertainty Principle:

$$\Delta t = \tau \geq h/\Gamma = h/(\Delta E)$$



Compound nuclear lifetimes range from 10^{-21} s (or so) to 10^{-14} s, which are very large when compared to the 10^{-23} – 10^{-22} s transit times for nucleons across a nucleus. Shown is an exaggerated picture of the Isovector Giant Dipole Resonance (GDR), which comprises approximately 80% or more of the integrated photonuclear cross section below 50 MeV.

Photonuclear cross sections are *strongly* energy dependent

Semi-classical Picture:

1) Gamma excites proton (1p1h state).

2) Excited proton exchanges energy with neutron (2p2h state),

3) Excited proton and neutron exchange energies with other protons and neutrons,

4) Etc ….

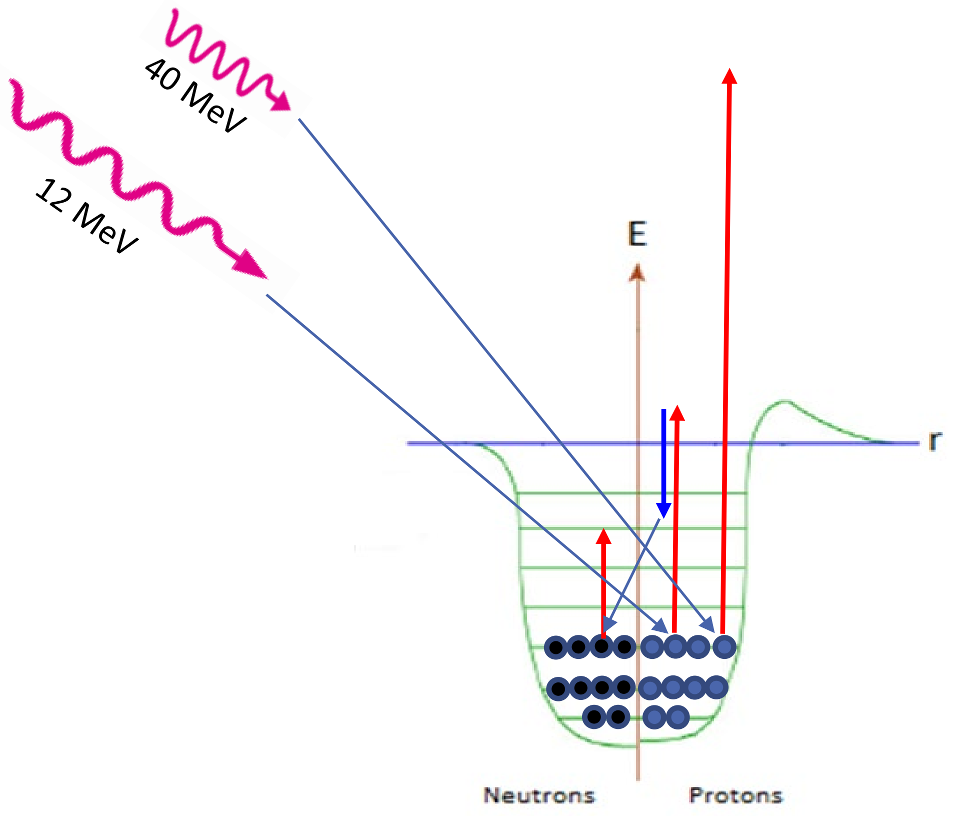


Figure courtesy of Geno Santistevan

Why do these states live a long time?

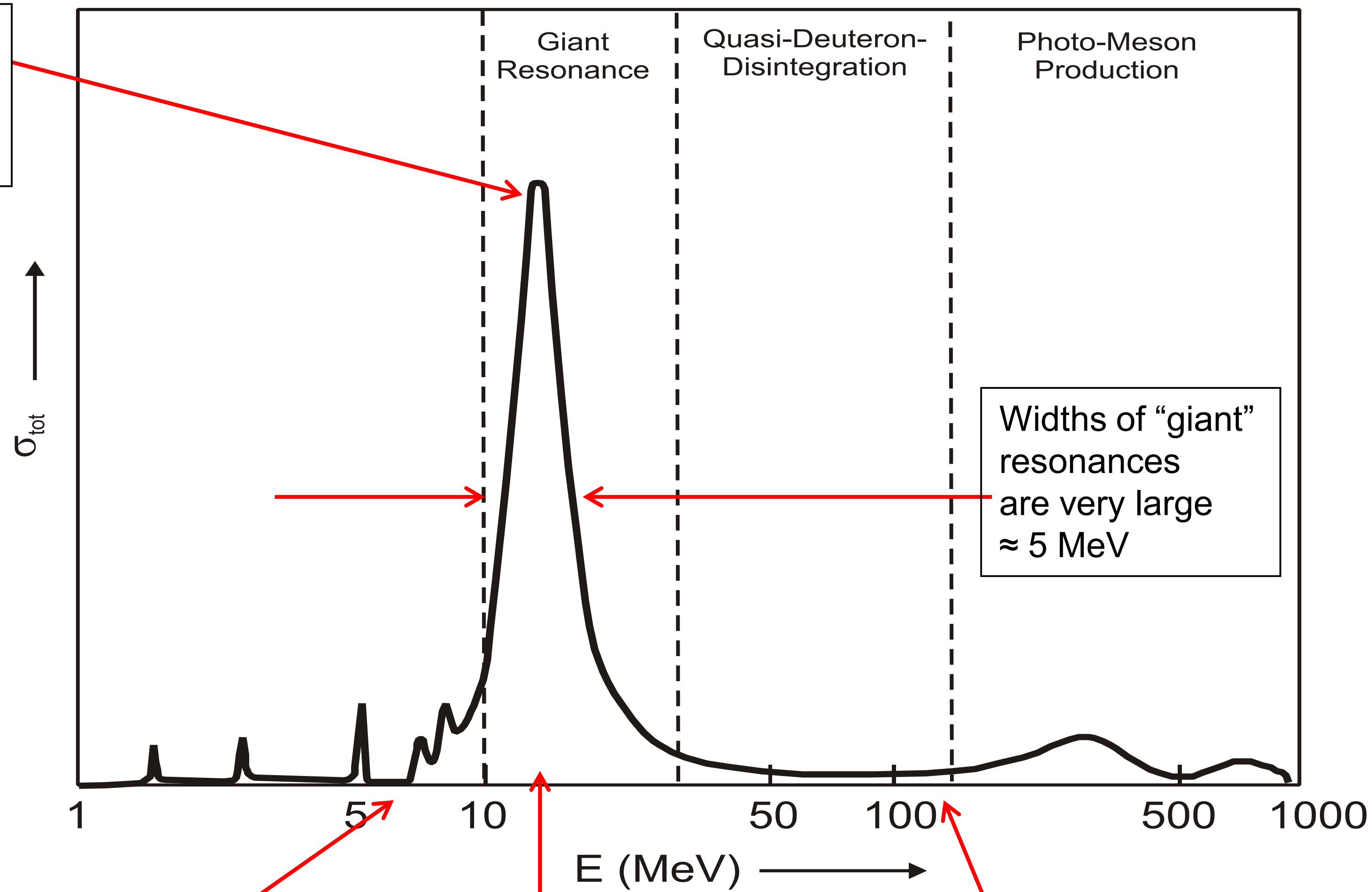
Because while the nuclear state is in the energy continuum, all of the individual nucleons are in bound states.

The physics of photonuclear reactions varies with photon energy…

10s to 100s of mb – up to approximately 1300 mb
(1 b = 10^{-28} m^2 = 100 fm^2)

Note:

Energy dependence of reaction channels makes optimization dependent on what you are trying to make (or not make).



Widths of "giant" resonances are very large ≈ 5 MeV

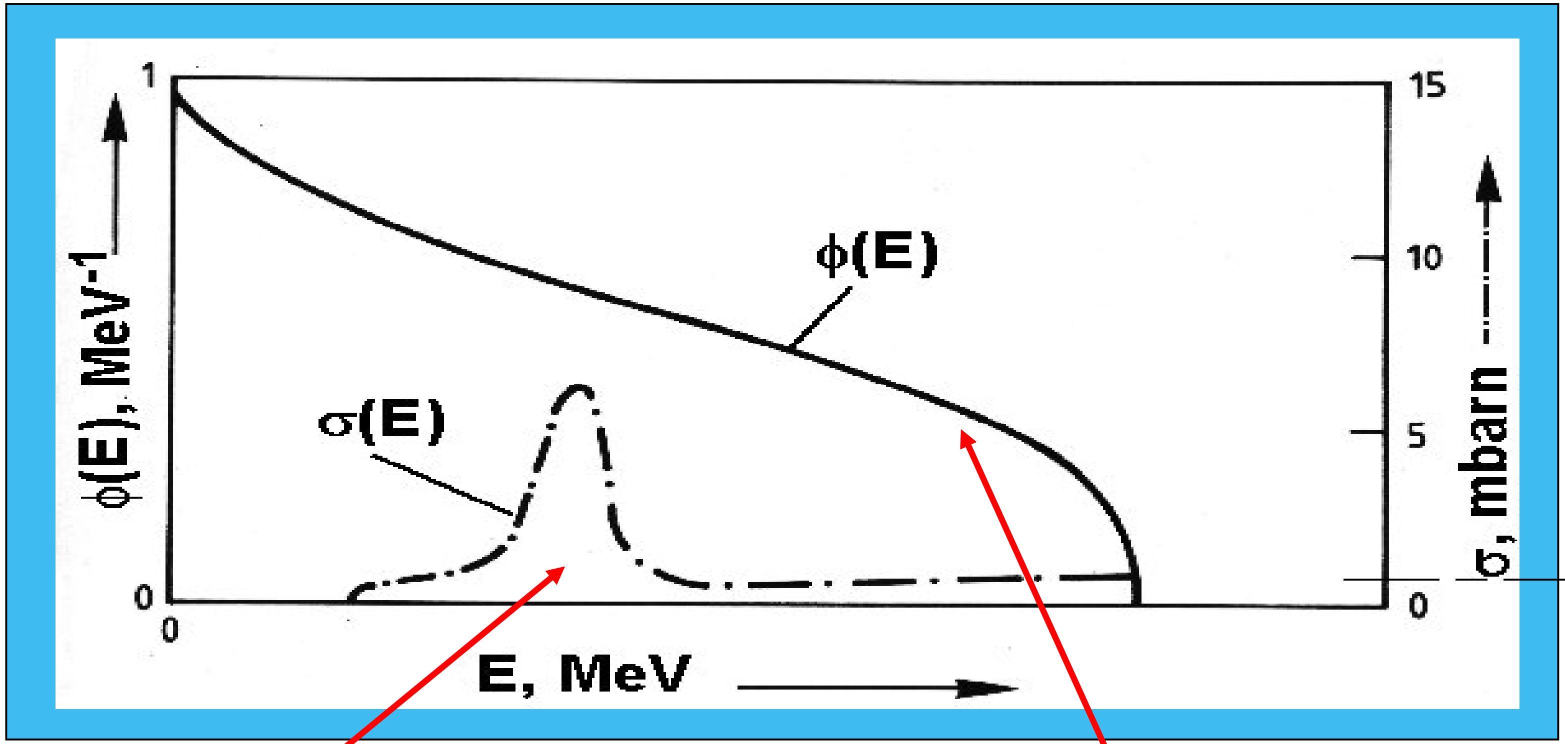
Astrophysical Nucleosynthesis

Isotope Production

Quasi-deuteron regime

But are photonuclear yields adequate? In some cases, Yes.

$$Y \propto M \int_{E_S}^{E_{max}} \varphi(E_\gamma) \cdot \sigma(E_\gamma)\, dE_\gamma$$



σ(E): Cross-section of the nuclear reaction under study, i.e.: the quantified 'probability' that the reaction occurs

ϕ(E): Flux density of the activating particles, e.g. bremsstrahlung photons produced by the accelerator.

Smaller photo-nuclear peak cross sections are compensated by thick targets and integration over a broad energy range:

$$Y \propto M \int_{E_S}^{E_{\max}} \varphi(E_\gamma) \cdot \sigma(E_\gamma)\, dE_\gamma$$

General Rule-of-thumb – for particle beam energies of order 30-100 MeV, maximum neutron production yields (a good measure of isotope production rate) are of order 10^{12} per kW, regardless of the projectile (light ions, heavy ions, electrons (bremsstrahlung)).

But, if photonuclear production is so wonderful, why hasn't photonuclear production been exploited?

Specific Activity: Two (potentially) viable paths are available:

1) Chemical Separation: which requires either (γ, charged-particle) reactions OR the (γ, n) daughter to β-decay to another element,

2) Kinematic Recoil: In which the post (γ, n) daughter recoils ($\approx$ 10 keV kinetic energy) out of the target material into another material for post-irradiation collection.

Time Scales for Experiments: Rough Guidelines

10^{-23} s – 10^{-22} s ≈ transit time across nucleus, of particle moving at c.

10^{-21} – 10^{-19} s ≈ equilibration and decay time scale for statistical nuclear reactions

10^{-19} s ≤ t ≤10^{-8} s ≈ "prompt" delayed decays amenable to coincidence experiments.

10^{-8} s ≤ t ≤10^{0} s ≈ delayed decays amenable to measurements between accelerator pulses.

10^{0} s ≤ t ≤10^{7} s ≈ delayed decays of traditional activation experiments.

10^{7} s ≤ t ≈ delayed decays amenable to ultra-low background measurements.

Isotope of Interest	Half-Life	Probable Photonuclear Reaction Production Mechanism	Alternative Photonuclear Reaction Production Mechanism	Notes:
^{44}Sc	3.97 h	^{45}Sc(γ, n)^{44}Sc		Possible candidate for kinematic recoil
^{47}Sc	3.35 d	^{51}V(γ, α)^{47}Sc		
^{64}Cu	12.7 h	^{65}Cu(γ, n)^{64}Cu		Possible candidate for kinematic recoil
^{67}Cu	3.79 d	^{71}Ga(γ, α)^{67}Cu		
^{67}Ga	3.62 d	^{69}Ga(γ, 2n)^{67}Ga	^{70}Ge(γ, 2np)^{67}Ga	Commercially available. Could also produce ^{67}Ge via ^{70}Ge(γ,3n)^{67}Ge then let it decay to ^{67}Ga.
^{68}Ga	1.13 h	^{69}Ga(γ, n)^{68}Ga	^{70}Ge(γ, np)^{68}Ga	
^{68}Ge	271 d	^{70}Ge(γ, 2n)^{68}Ge		
^{72}As	26.0 h	^{75}As(γ, 3n)^{72}As		
^{77}As	38.8 h	^{81}Br(γ, α)^{77}As	^{79}Br(γ, 2p)^{77}As	
^{76}Br	16.2 h	^{79}Br(γ, 3n)^{76}Br		
^{77}Br	72.0 h	^{79}Br(γ, 2n)^{77}Br		
^{89}Zr	3.27 d	^{90}Zr(γ, n)^{89}Zr	^{91}Zr(γ, 2n)^{89}Zr	Possible candidate for kinematic recoil
^{86}Y	14.7 h	^{89}Y(γ, 3n)^{86}Y	^{88}Zr(γ, np)^{86}Y	
^{90}Y	2.67 d	^{91}Zr(γ, p)^{90}Y	^{92}Zr(γ, np)^{90}Y	Commercially available
^{117m}Sn	14 d	^{118}Sn(γ, n)^{117m}Sn	^{119}Sn(γ, 2n)^{117m}Sn	
^{124}I	4.18 d	^{127}I(γ, 3n)^{124}I		
^{161}Tb	6.91 d	^{162}Dy(γ ,p)^{161}Tb	^{163}Dy(γ, np)^{161}Tb	
^{195m}Pt	4.0 d	^{197}Au(γ, np)^{195m}Pt		
^{198}Au	2.69 d	^{200}Hg(γ, np)^{198}Au	^{199}Hg(γ, p)^{198}Au	
^{225}Ac	10.0 d	^{226}Ra(γ ,n)^{225}Rn ==> ^{225}Ac	^{232}Th(γ, 7n)^{225}Th ==> ^{225}Ac	Both mechanisms require beta-decay to "feed" ^{225}Ac

Outline of the Program:

A partial list of isotopes of interest and potential photonuclear production mechanisms.

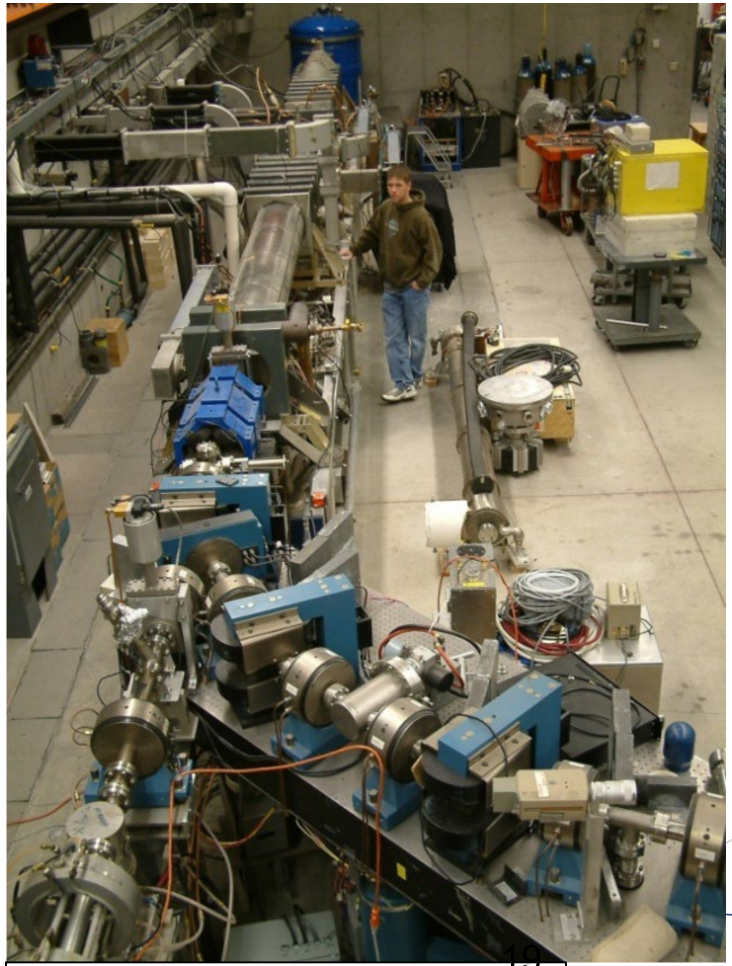
Where will student-led experiments be done?

Several labs, including INL, ISU, NMT and probably ANL, RPI, possibly JH-APL.
Shown is one accelerator at ISU's Idaho Accelerator Center (IAC).

One of the Labs: Idaho Accelerator Center:

- **Electrons (and gammas from bremsstrahlung):**
  - (2) 40 MeV electron linacs (~10 kW)
  - 25 MeV electron linac (~ 1 kW)
  - 25 MeV, 1 kW high-resolution LINAC (shown).
- **Infrastructure to support researchers:**
  - Radiochemistry Lab,
  - Machine Shop,
  - Electronics Shop,
  - Detector Lab,
  - etc.

Idaho Accelerator Center



L-Band Traveling Wave Linac

Part of the team: Shown are Dr. Edna Cárdenas of INL, along with Kean Martinic of ISU, and Robert Bentley and Geno Santistevan of NMT

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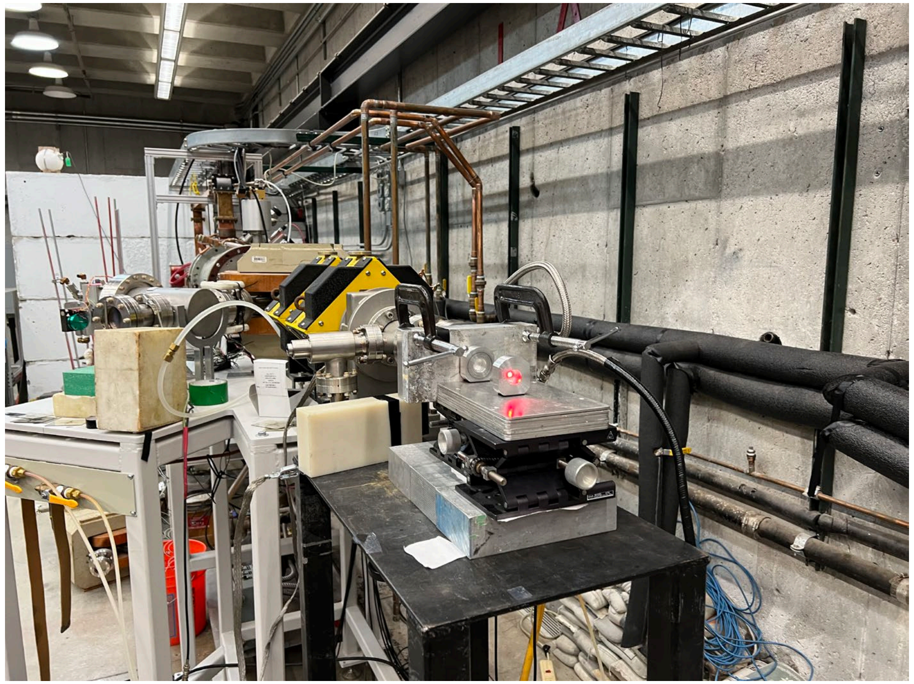
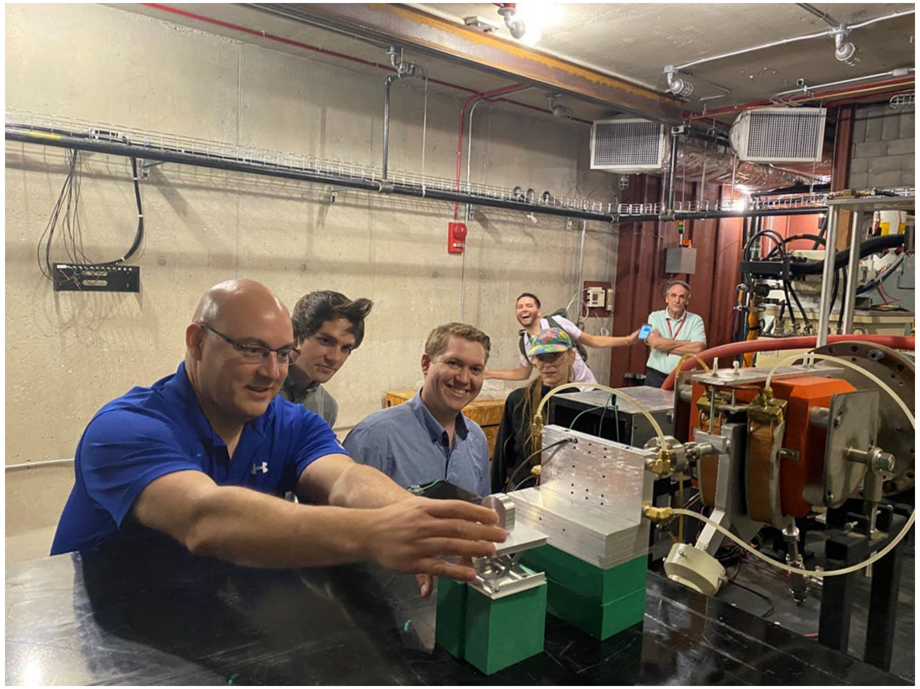
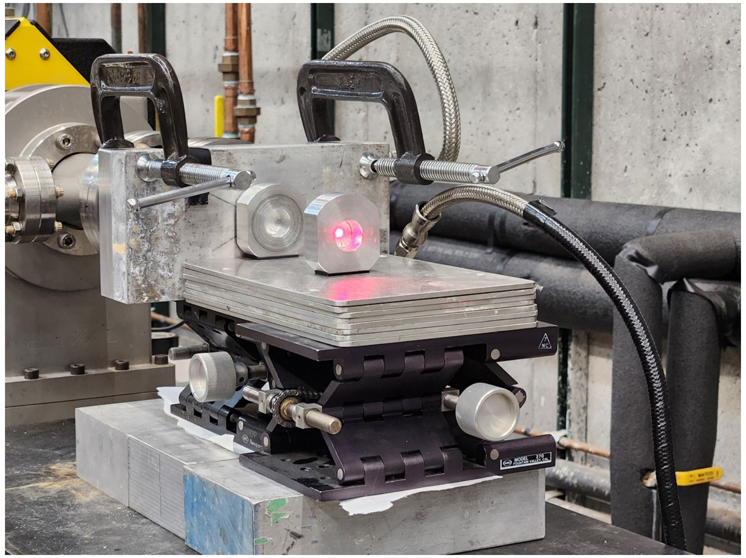




Lining up the targets….



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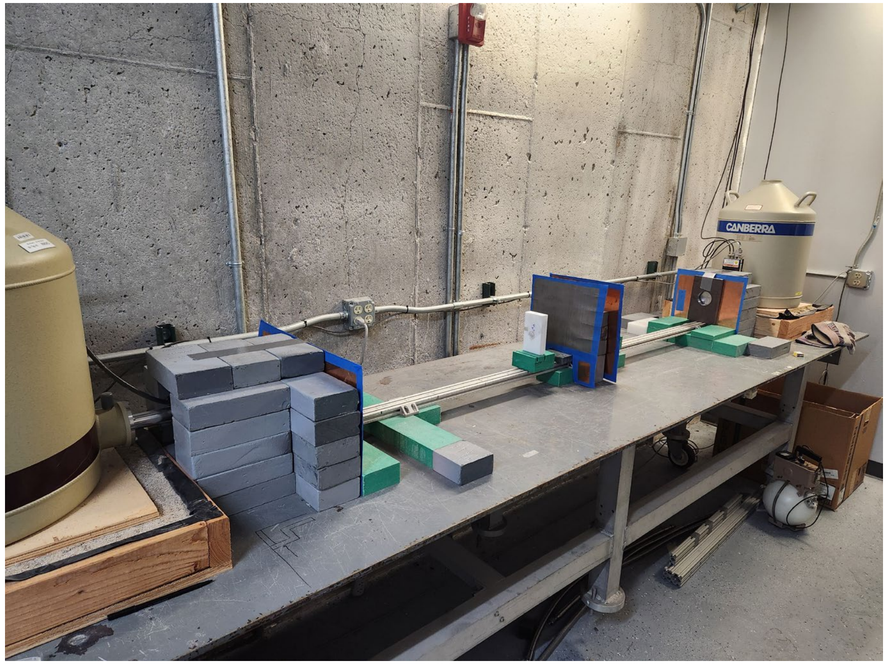
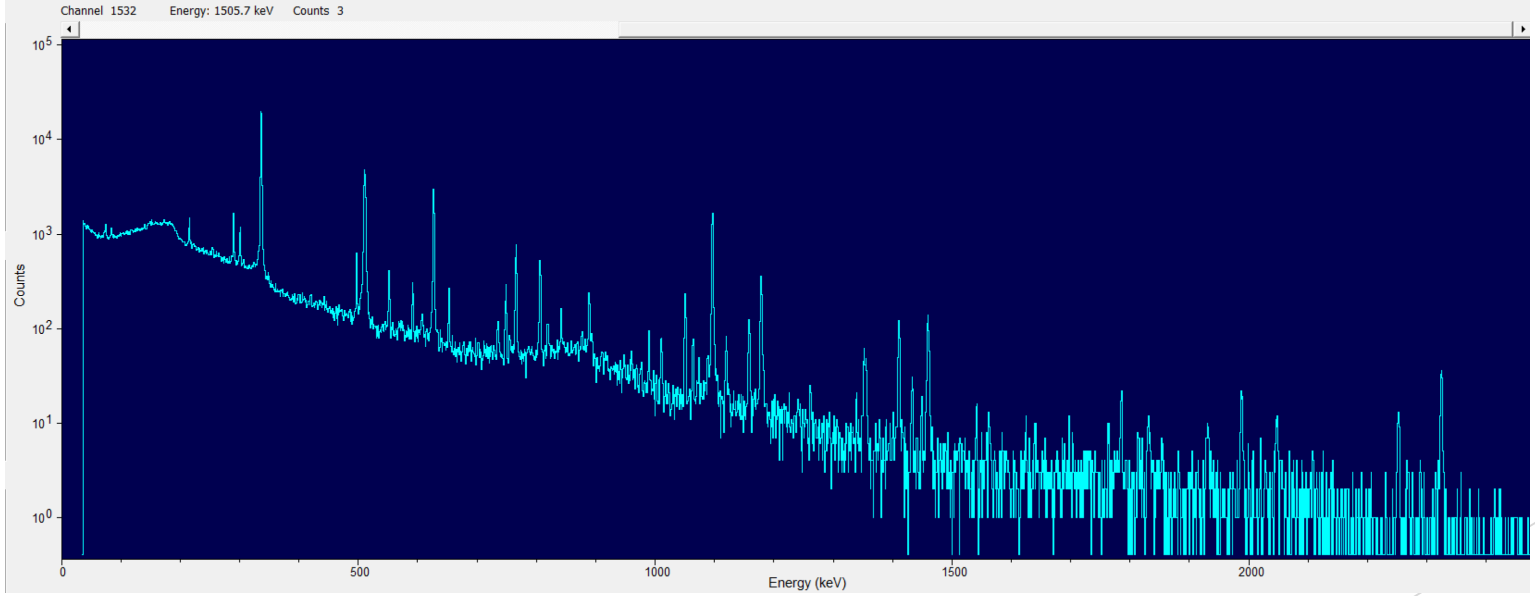


Counting the samples at the IAC



Focus on Task 4: And, most important of all, educate new talent who go on to contribute to the nation's isotope programs.

This is a 5-year research and education program that started in January, 2023 with, currently:

- Six masters or doctoral students,
- Ten baccalaureate students
- Summer internships at Idaho National Lab

Summary:

A three-institution collaboration team has been formed (NMT + Idaho State University + Idaho National Lab), which will pursue:

- Measure bremsstrahlung-weighted excitation functions and infer cross sections to fill in some of the many gaps in the world's photonuclear data, including (γ, α), (γ, p), (γ, n), (γ, np), and possibly other reaction channels.
- Measure Figures-of-Merit (FOM) for isotope production for selected reactions versus bremsstrahlung end-point energy, both for enriched and natural targets.
- Investigate kinematic recoil separations of radioisotopes to address the isotope separation challenge of using (γ, n) reactions for isotope production.
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Thank You!