Isotopes at LERF:

Photonuclear Reaction Cross Sections
(and Why it Matters)

From 0 – 140 MeV: Isotope Production, Physics and Applications

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What are the basic science questions?

Photonuclear reactions below the pion production threshold, with photons in either the entrance channel or the exit channel (or both), have been studied since the late 1950s.

- The competitive advantage of real photons in exploring E1 excitation strength and decay channels and, to a lesser extent, E2 and M1 excitations, led to the systematic measurement and establishment of photonuclear sum rules for these low multipole excitations. Moreover, tagged-photon scattering provides the "best" means for measuring absolute total photo-nuclear cross sections.

- Quantities such as integrated photo-absorption cross sections up to pion photo-production threshold, and the distribution of E1, E2 and M1 strength, are well established for many nuclei, but, in many cases for either, for "natural" targets (i.e. – ^{nat}Z targets) or for "selectively interesting" isotopes.

  - What about isotopic-specific data?
  - What about the data of individual reaction channels, and the models that predict them?
  - What about measurements over most of the sub-pion threshold energy range? Note that a large portion of the data were only measured over a small energy range.
  - What about the systematics of these data, as functions of A, Z, exit channel, and E_γ, and how do these constrain nuclear models?
  - What are the astrophysical origins of proton-rich nuclei (they cannot be made in the standard astrophysical r-process and s-process)?

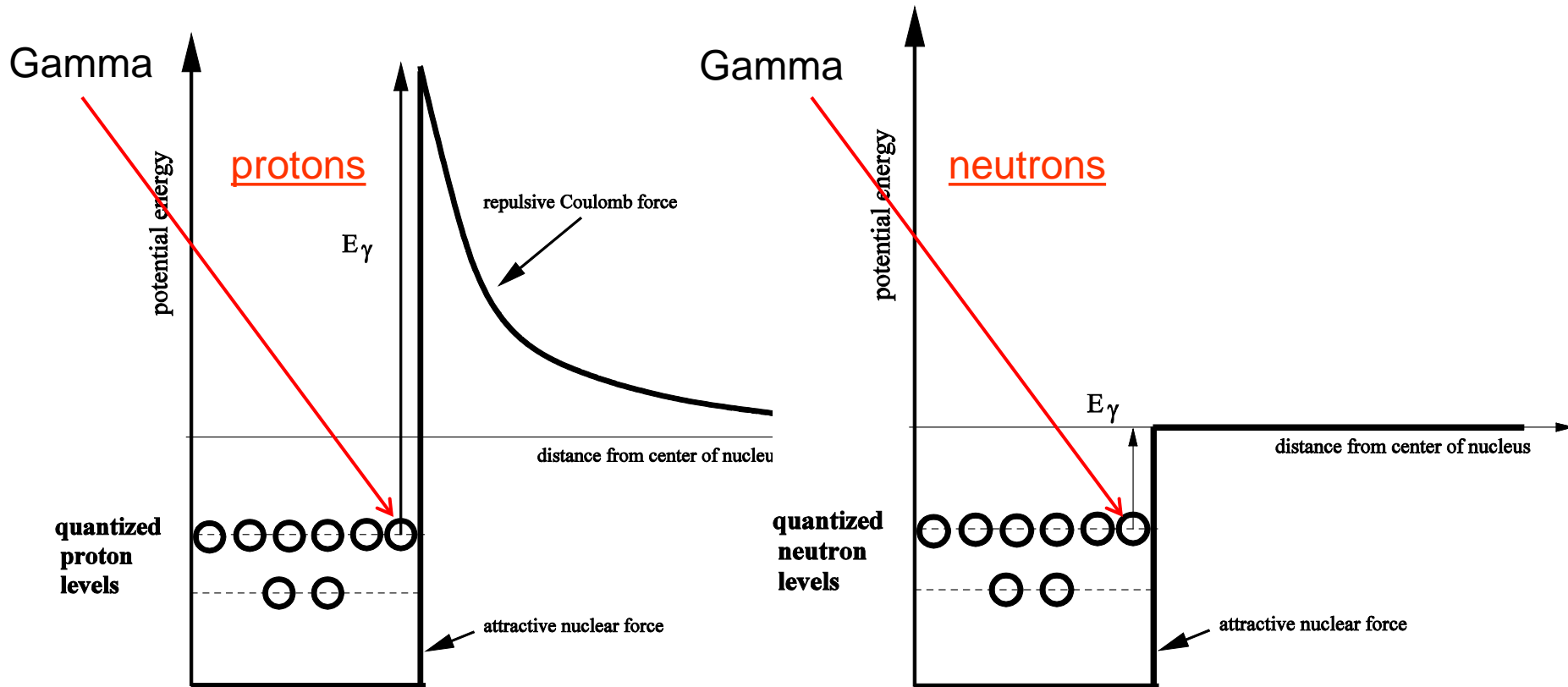   - Why do measurements of these processes matter?

2

Why does this matter for practical applications?

There are many proposed and important applications of photo-nuclear physics that require or would greatly benefit from well-measured cross sections of specific isotopes and individual reaction channels. These include:

- radiation shielding, dosimetry and transport codes,
- radio-isotope production for medicine, industry and research,
- photon activation analysis (PAA), forensics, attribution and illegal trade,
- nuclear non-proliferation, homeland security and nuclear forensics,
- photo-neutron sources for accelerator-driven subcritical systems (ADSS)

The need for these photo-nuclear data can best be filled by a bright, CW photon source capable of high fluxes in the energy range from approximately 10 to 140 MeV.

What are Photonuclear Processes? 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)
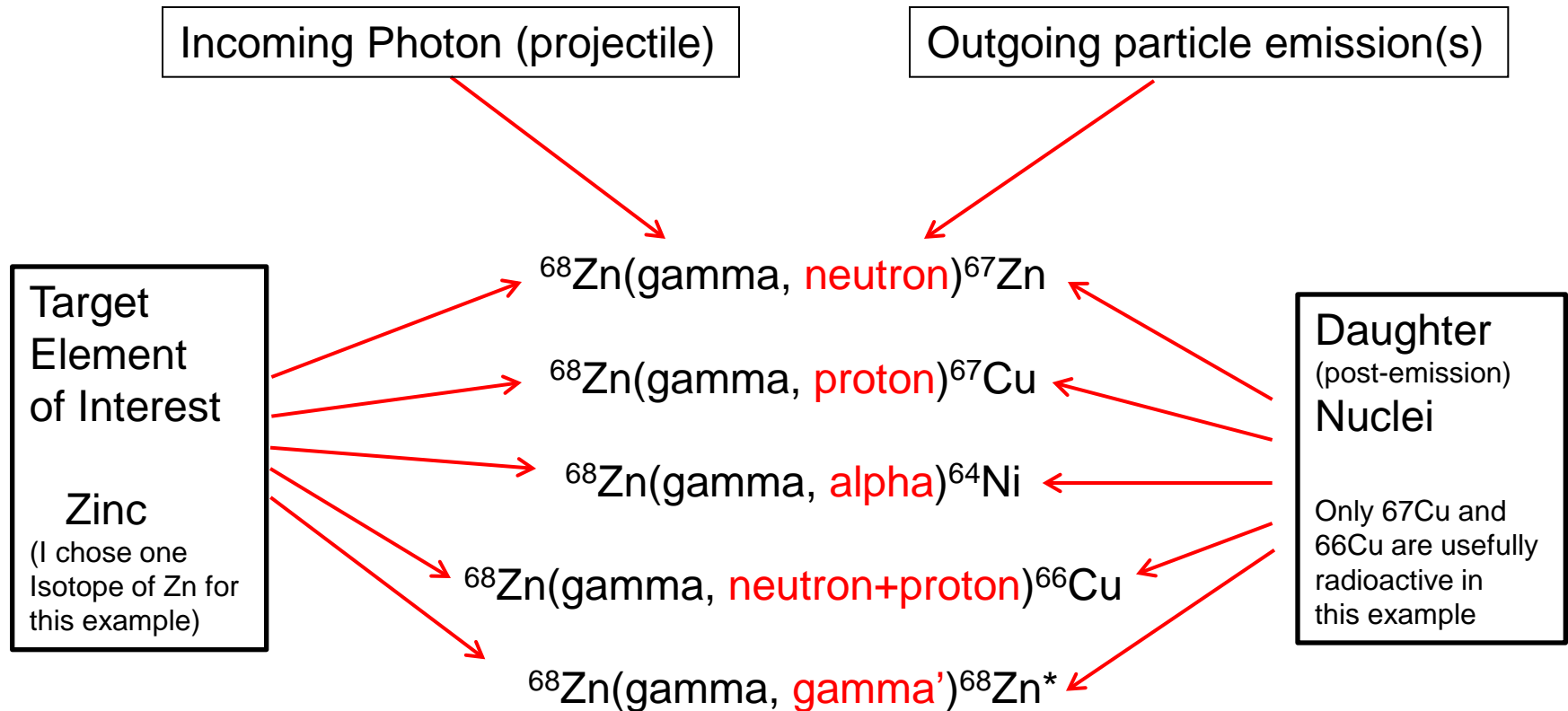
Gamma

potential energy

protons

E_γ

repulsive Coulomb force

distance from center of nucleus

quantized proton levels

attractive nuclear force

Gamma

potential energy

neutrons

E_γ

distance from center of nucleus

quantized neutron levels

attractive nuclear force

Coulomb energy barrier for protons …   **but not for neutrons.**

What are common photo-production mechanisms?

Photon (gamma) absorption "heats up" a nucleus of a target element of interest, followed by emissions:

Incoming Photon (projectile)

Outgoing particle emission(s)

Target Element of Interest

Zinc
(I chose one Isotope of Zn for this example)

^{68}Zn(gamma, neutron)^{67}Zn

^{68}Zn(gamma, proton)^{67}Cu

^{68}Zn(gamma, alpha)^{64}Ni

^{68}Zn(gamma, neutron+proton)^{66}Cu

^{68}Zn(gamma, gamma')^{68}Zn*

Daughter
(post-emission)
Nuclei

Only 67Cu and 66Cu are usefully radioactive in this example

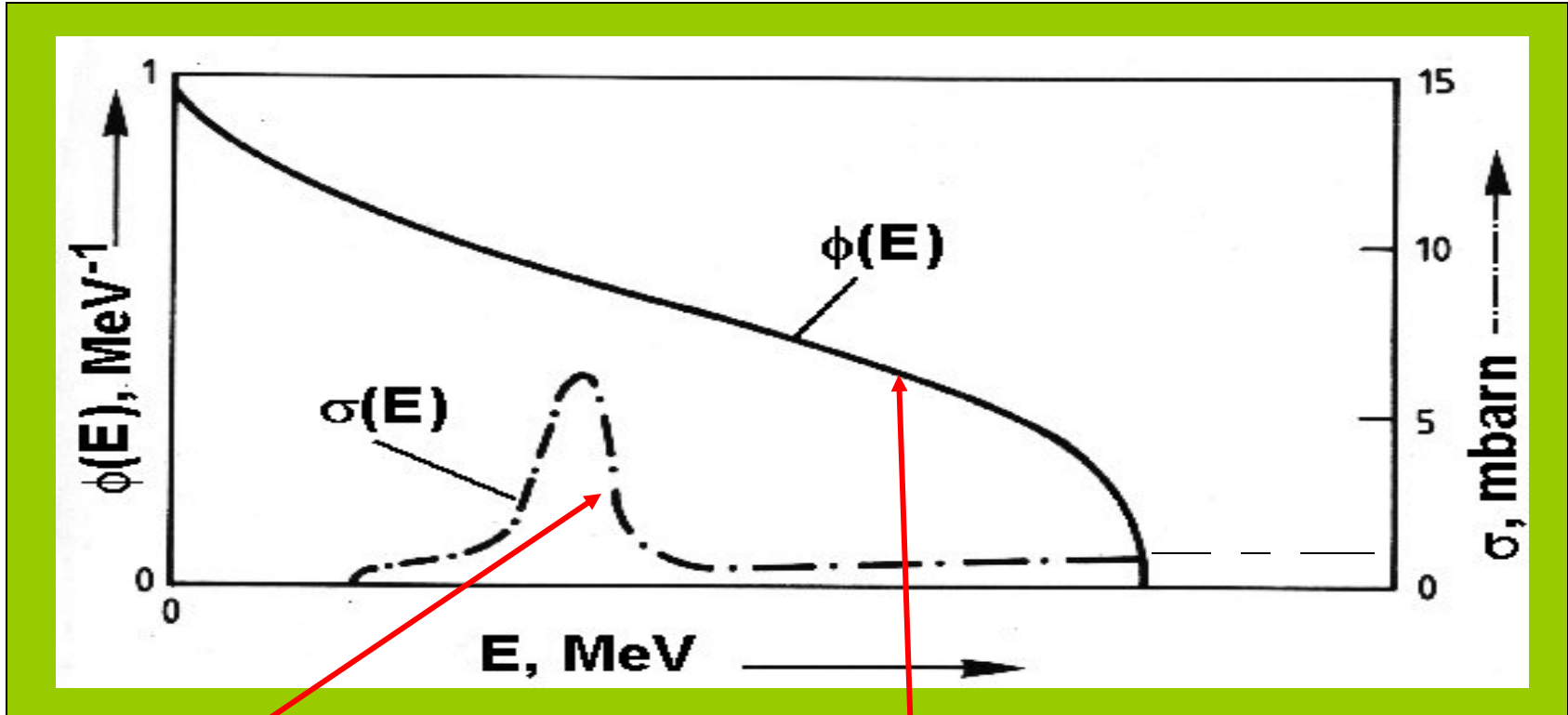etc.

* - note that beta/positron emission can sometimes be usefully employed, too.

Photo-nuclear reactions enable fuller exploitation of the moderately Proton-Rich side of the Valley of Stability ...

The valley of stability

nuclei with excess nucleons move down the valley towards stability

the neutron side of the valley is poorly understood - scientists aren't sure where the dripline lies

dripline

superheavy elements?

Lead

Gold

Hafnium

Xenon

Tin

more protons

more neutrons

stable elements

accelerators   reactors
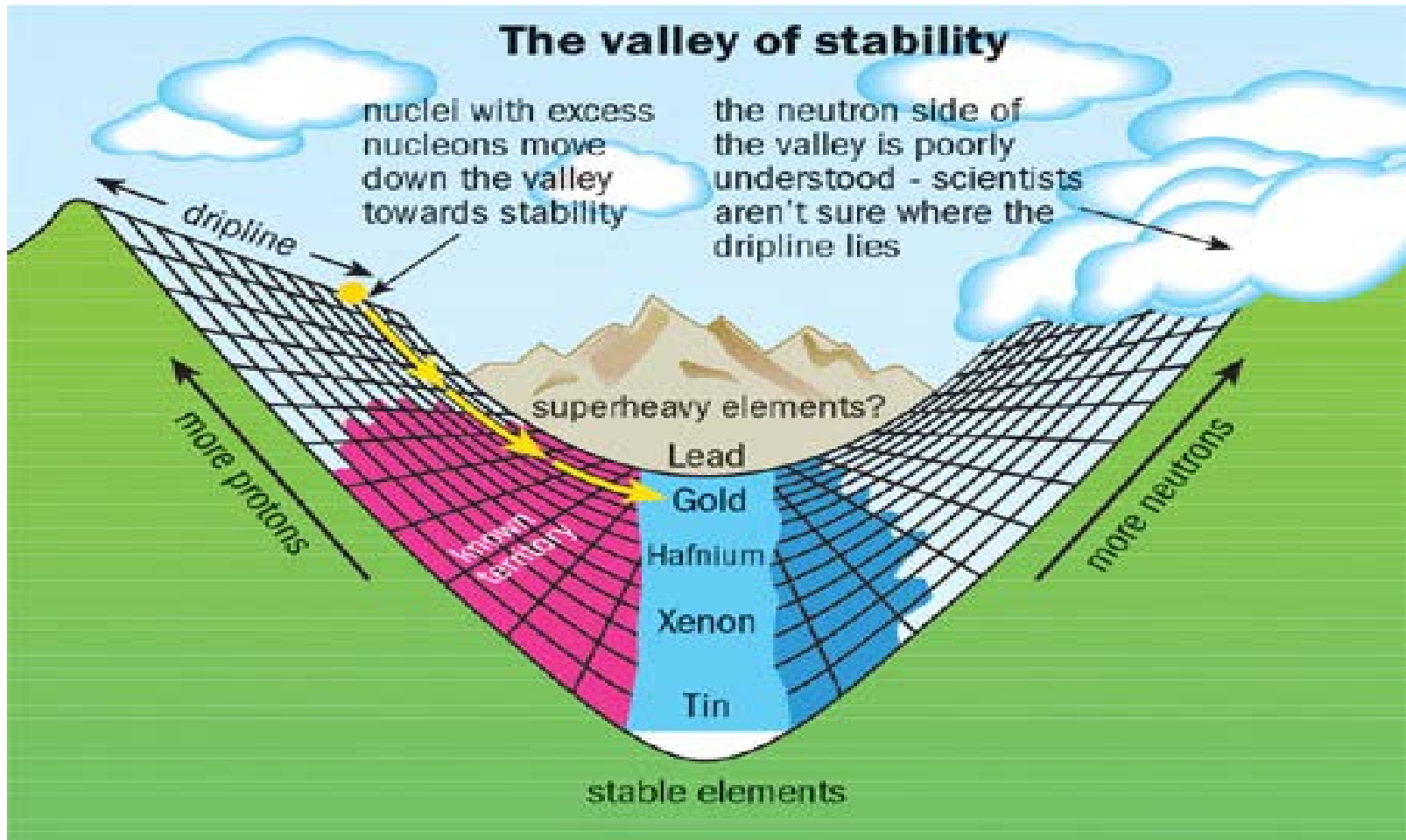
Yields are high!!!:

a fact not often appreciated:

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

σ(E): Cross-section of the nuclear reaction under study, from 10s mb ranging up to ≈ 1 b, with a width of approximately 5 MeV.

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

Isotope	Crude yield per 100 kW per week from 100g targets at 50 MeV electron beam energy
F-18*	9 kCi/wk
Cu-64*	10 kCi/wk
Ba-131*	15 kCi/wk
Mo-99*	2 kCi/wk (approx. 10% national need)
In-111*	2 kCi/wk
Y-88*	6 Ci/wk
Cu-67*	12 Ci/wk

These rates of production are <u>very significant</u> when compared to the national demand. But do not forget about SA!

* - Taken from <u>measured yield at 2 kW</u> using IAC linac and scaled up by power, mass, beam energy, isotopic enrichment, geometric corrections, etc. Note that these rates are consistent with published rates of Segebade et al. and the Mo-99 estimates of TRIUMF.

"Photo-induced reaction cross section data are of importance for a variety of current or emerging applications. Among them are:

• Radiation shielding design and radiation transport analyses,

• Calculations of absorbed dose in the human body during radiotherapy,

• Physics and technology of fission reactors (influence of photoreactions on neutron balance) and fusion reactors (plasma diagnostics and shielding),

• Activation analyses, safeguards and inspection technologies,

• Nuclear waste transmutation, and

• Astrophysical nucleosynthesis."

Note that they missed isotope production (more about this later)
– although isotope production is implicit in the last three applications.

A word about the data IAEA considered, and the data that they did not:

- Did include traditional bremsstrahlung experiments, photon-tagging data (except photon scattering), annihilation in-flight data.

- Did Not include (i) photon scattering data, or (ii) activation data (mostly done by Segebade et al.), or (iii) capture reaction data.

Why is the world's data sparse?

- Many of these partial cross sections are small (≈µb),

- Incident beam (photons) and outgoing products (neutrons) are electrically neutral,

- Outgoing projectiles (neutrons, protons, etc.) emitted with ≈ 1MeV or less KE. For protons, this means it is difficult to get the protons out of the target,

- Photons offer the advantage of a well-understood reaction, but the disadvantage of only exciting a few modes of excitation. (i.e. – "less interesting")

One potential application of isotope production as a forensic tool (PAA) which may be of some importance:

"Counterfeiting is one of the fastest growing economic crimes of modern times...."[1]

The International Chamber of Commerce estimated that the global value of counterfeit goods rose to a staggering **$1.8 trillion in 2015**, up nearly three-fold from the 2008 estimate of $650 billion.[2] Counterfeits account for roughly: 1% of the medicine in the U.S. and 10%-30% in developing countries, sometimes with fatal consequences. Fake automotive parts surged by 83% in the U.S. in 2014. And the U.S. Department of Commerce reports that annual incidents of major counterfeit microelectronics for DoD systems number in the tens of thousands.

1 - International Chamber of Commerce, "Countering Intelligence Bureau,"
http://www.iccwbo.org/products-and-services/fighting-commercial-crime/counterfeiting-intelligence-bureau/

2 - International Chamber of Commerce, "Counterfeiting Statistics,"
http://www.iacc.org/resources/about/statistics

What is the state of the world's data?

- On the positive side:
  - Reasonably good integrated, total (and absolute) cross section up to pion threshold for low-Z and high-Z targets.

- On the negative side:
  - Very little quality data on angular distributions, exclusive reactions, full energy range, most isotopes, absolute normalization, multipolarity, and energy distributions of emissions.

What's at stake in terms of physics?

- Absolute normalization of photo nuclear cross sections,
- Sum rules and nuclear models,
- Partial cross sections (emission products) and nuclear models.
- Energy and angular dependence of emission products.
- Anomalous abundancies of elements and questions of the astrophysical origins of proton-rich nuclei (see T. Rauscher et al., Rep. Prog. Phys. 76 (2013) 066201)

Photo-Nuclear Cross Sections have strong energy dependence.

10s to 100s of mb – up to approximately 1300 mb

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

Giant Resonance

Quasi-Deuteron-Disintegration

Photo-Meson Production

σ_{tot}
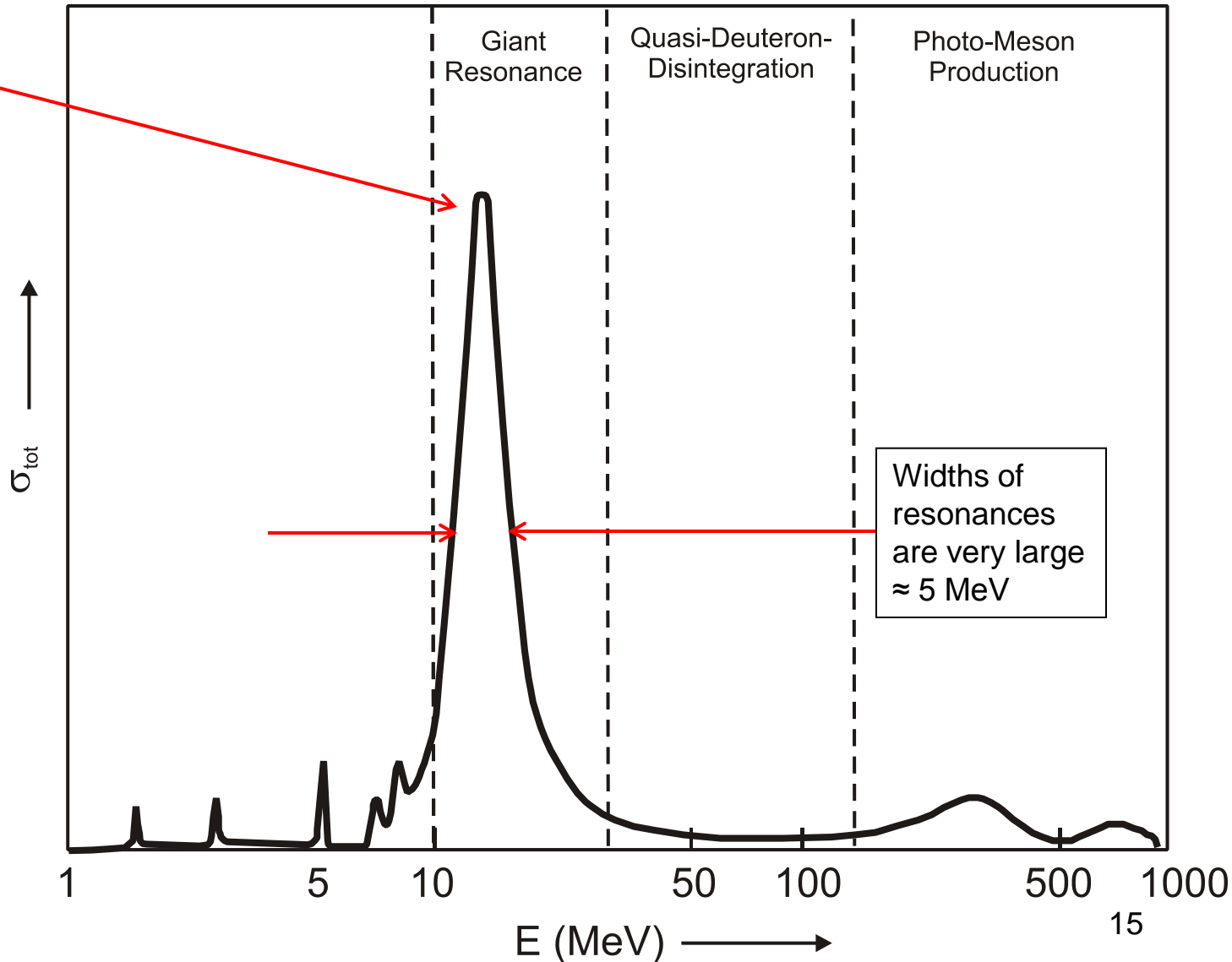
Widths of resonances are very large ≈ 5 MeV

1   5   10   50   100   500   1000

E (MeV)

# How are these cross sections measured? Four dominant techniques:

- Bremsstrahlung irradiation:
  - Deconvolution of cross section from yield curves.

- Tagged bremsstrahlung:

- Positron annihilation in-flight:

- Attenuation measurements:

- Inverse reactions (capture data…)

Photon Tagging, quasi-monochromaticity, and absolute normalization, Illinois was the most important player in this energy region.

BREMSSTRAHLUNG CALIBRATION
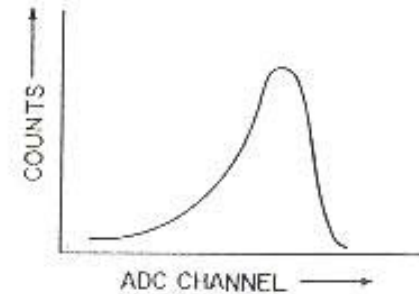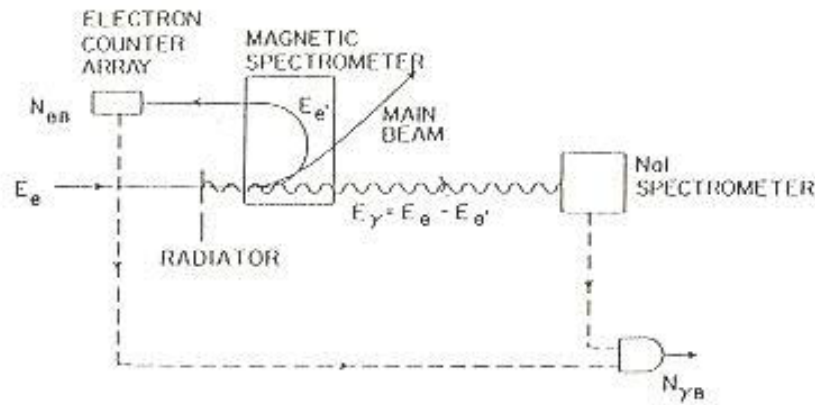
SCATTERING
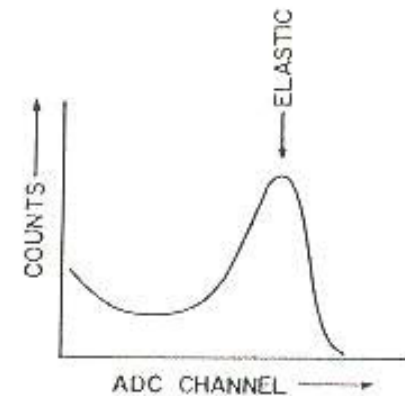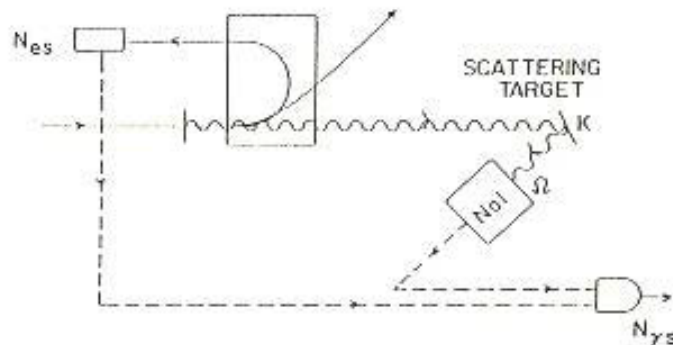
IF the same detector is used to measure flux and yield
➔ detector efficiency cancels in experimental normalization.

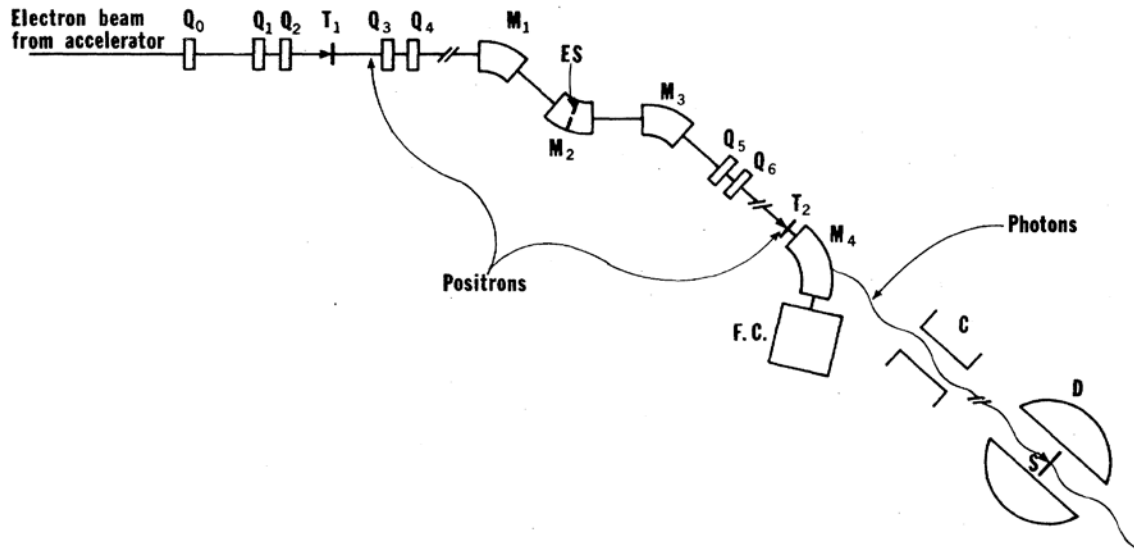# Positron annihilation in-flight (Saclay and LLNL):

FIG. 2. Schematic diagram of the annihilation-photon beam facility at Saclay: M_i—bending magnets; Q_i—quadrupole magnets; ES—energy-analyzing slit; T_1—positron converter target; T_2—annihilation target; FC—Faraday cup; C—collimator; S—nuclear sample; D—neutron detector. (from Bayart, 1969)

And Attenuation Measurements, completed by Ahrens et al. at Mainz, measures bremsstrahlung flux via thin foil pair-production (KE of e^+/e^- pair measured) before and after a nuclear target.   Note that this requires a large and theoretically-based subtraction of atomic attenuation processes.

The IAEA protocol, greatly (over)simplified, was:

- Put all of the bremsstrahung, annihilation in-flight and tagged data on the "same footing",

- Compared the consensus data (if there is a consensus) with statistical-model calculations,

- If agreement was found between calculations and data, then recommending cross sections is easy.

- If there was little or no consensus with the experimental data, the theoretical calculations "tipped" the scale in favor of the experimental data that mostly closely matched the theory.

Quality data, setting aside normalization, typically looks like this (note that the data generally only spans a small part of the 0-140 MeV range):
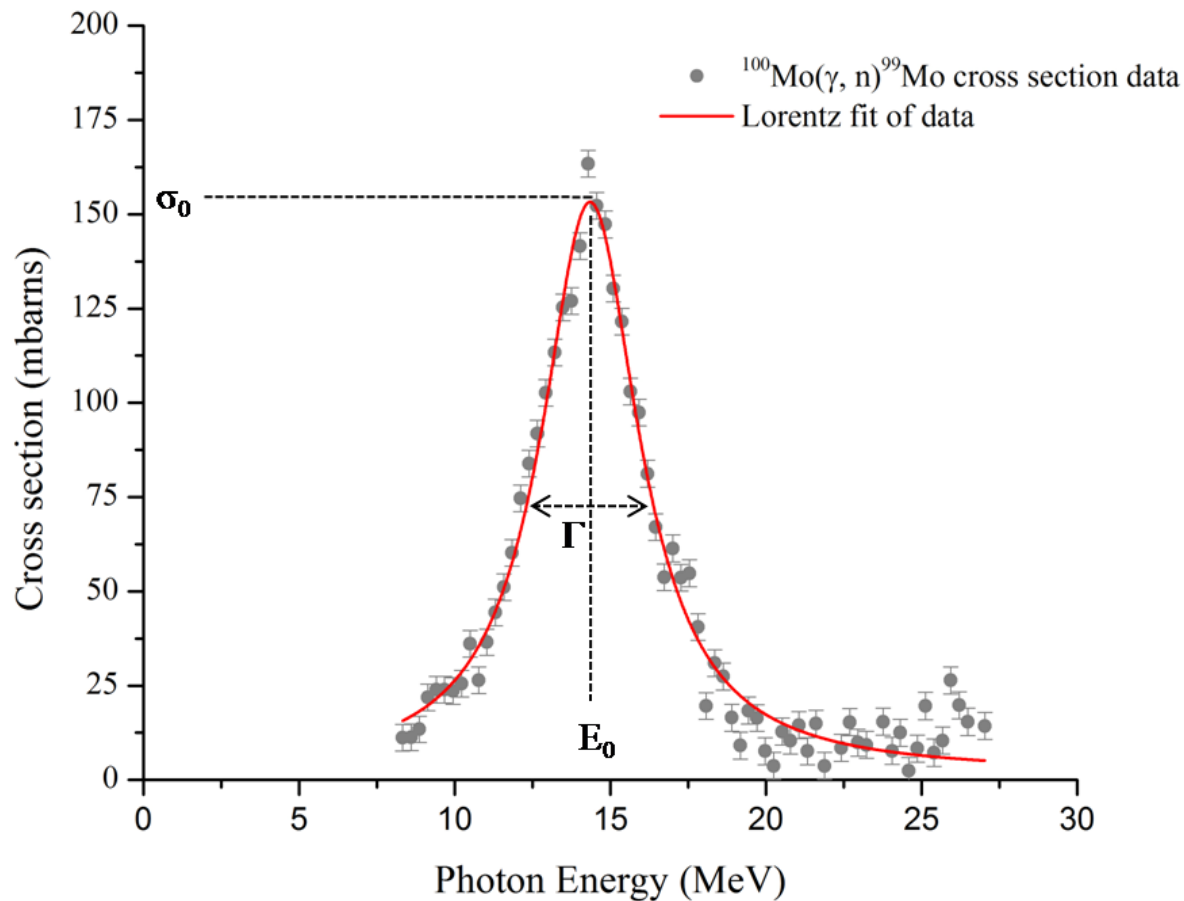
What difference would photon-scattering data make?
Qualitatively, photon scattering can be thought of as
"photo-absorption, squared."
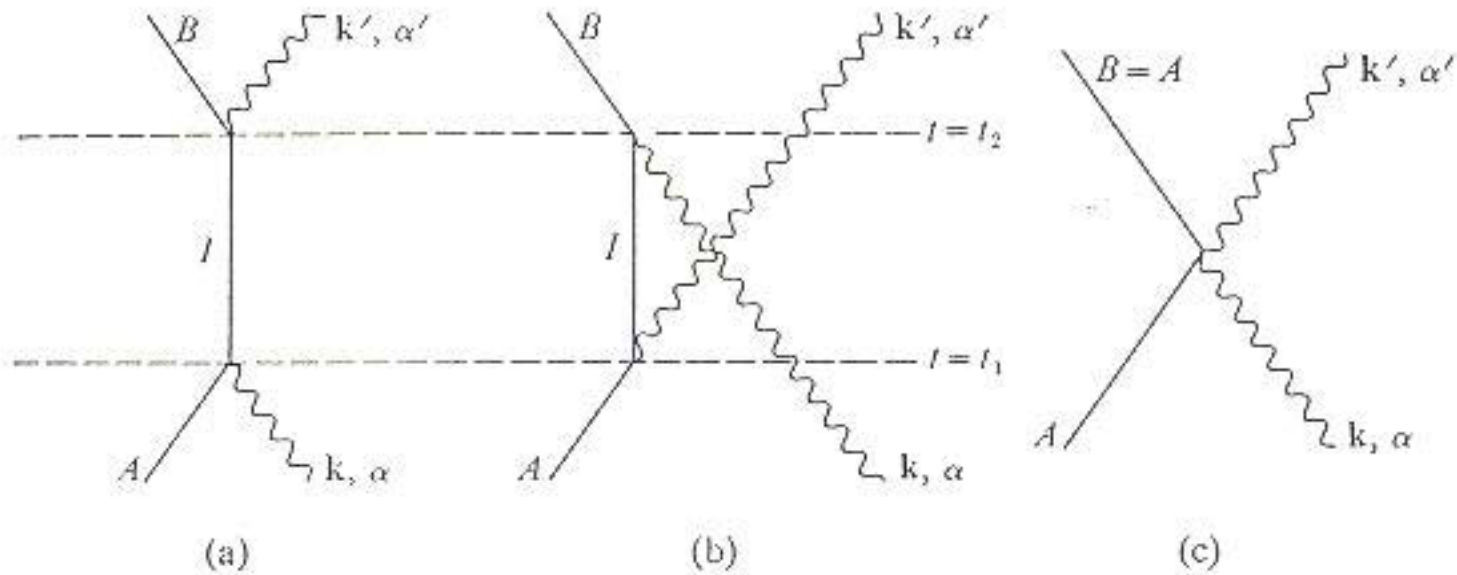
But photon scattering is related to photo-absorption in a model-independent fashion:

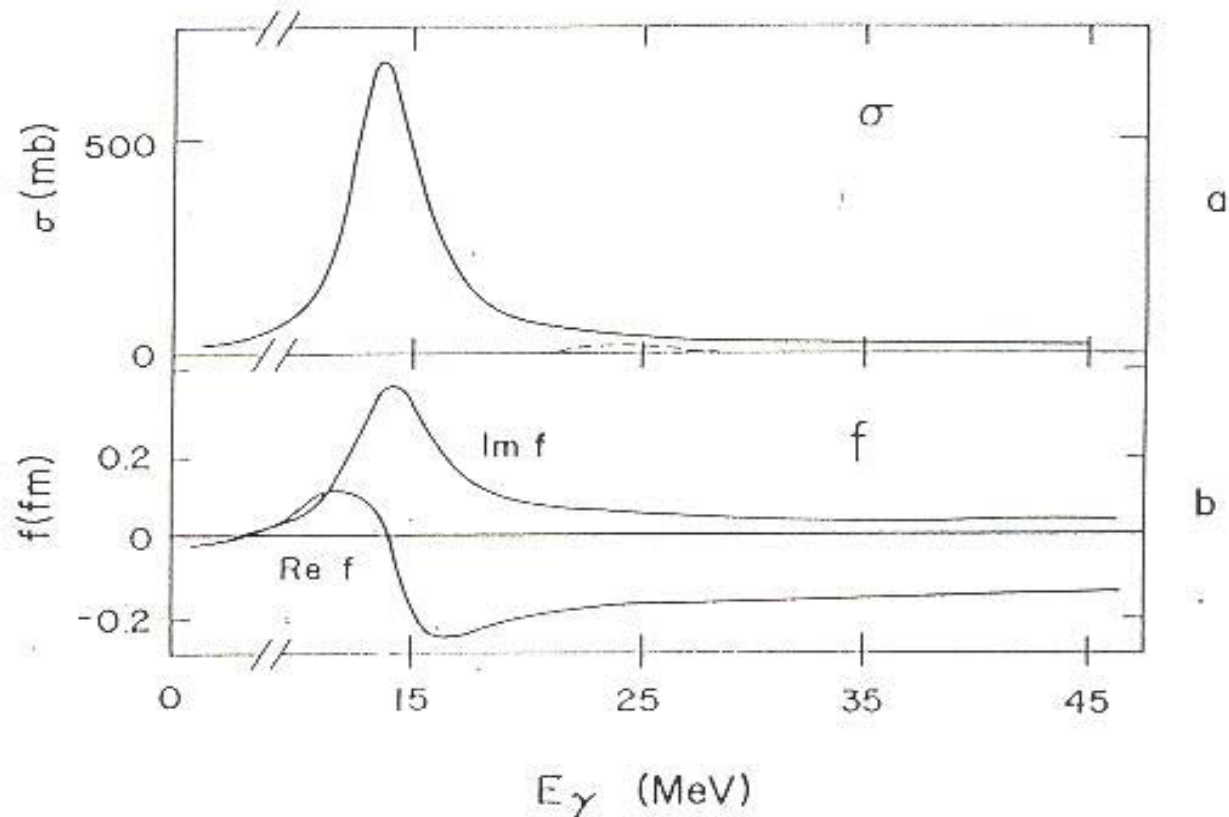$$\frac{d\sigma}{d\Omega} = |R(E, \theta)|^2.$$

$$R(E, \theta = 0) = f(E) + D_0(\theta = 0), \qquad D_0 = -\frac{Z^2}{A}\frac{e^2}{Mc^2}\vec{\epsilon}\cdot\vec{\epsilon}\,',$$

$$\mathrm{Im}(f(E)) = \frac{E}{4\pi\hbar c}\sigma_\gamma(E), \qquad \mathrm{Re}(f(E)) = \frac{E^2}{2\pi^2\hbar c}\int_0^\infty \frac{\sigma_\gamma(E')dE'}{(E'^2 - E^2)},$$

The ingredients to these relations are causality and unitarity.

$$\text{Im}(f(E)) = \frac{E}{4\pi\hbar c}\sigma_\gamma(E), \qquad \text{Re}(f(E)) = \frac{E^2}{2\pi^2\hbar c}\int_0^\infty \frac{\sigma_\gamma(E')dE'}{(E'^2 - E^2)},$$

Note that especially at the peak of the resonance, the vanishing of the imaginary part of the scattering amplitude ➔ direct proportionality to absorption.

Sum Rules:
1 TRK =

$$\int_0^\infty \sigma(E)\,dE = \frac{2\pi^2 e^2 \hbar}{Mc}\frac{NZ}{A} = 60\left(\frac{NZ}{A}\right)\text{MeV·mb},$$
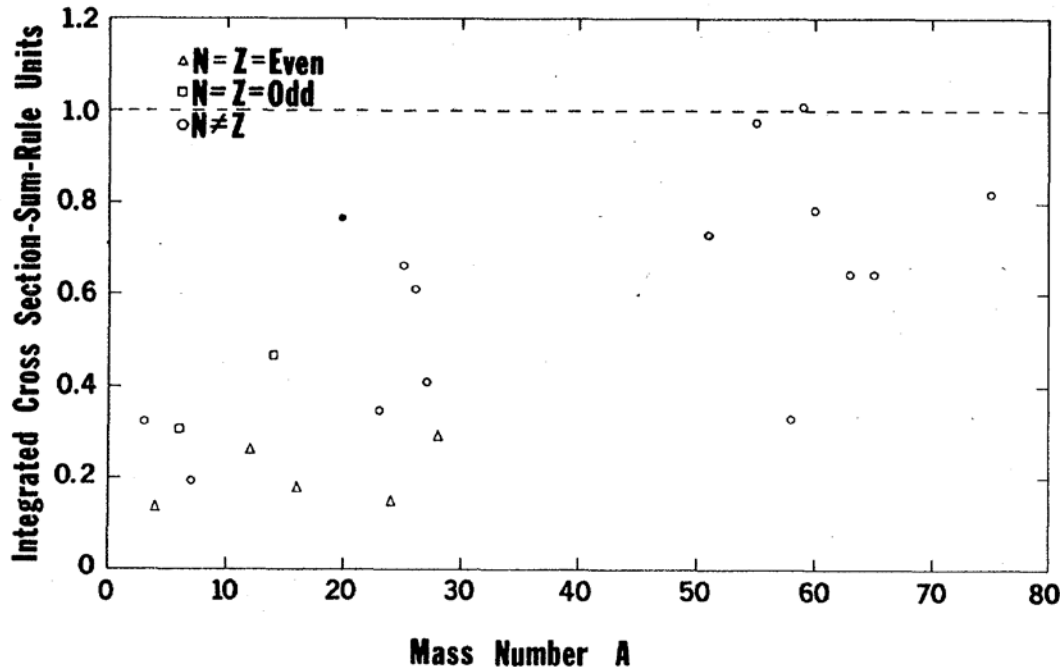

FIG. 40. Measured integrated total photoneutron cross sections $\sigma_{\text{int}}(\gamma,\text{total})$ for nuclei having $A < 80$ in units of $60NZ/A$ MeV-mb, where

$$\sigma_{\text{int}} = \int_{E_{\text{thr}}(\gamma,\,n)}^{E_{\gamma\max}} \sigma(E)\,dE.$$

24

1 TRK =

$$\int_0^\infty \sigma(E)\,dE = \frac{2\pi^2 e^2 \hbar}{Mc}\frac{NZ}{A} = 60\left(\frac{NZ}{A}\right)\text{MeV-mb},$$
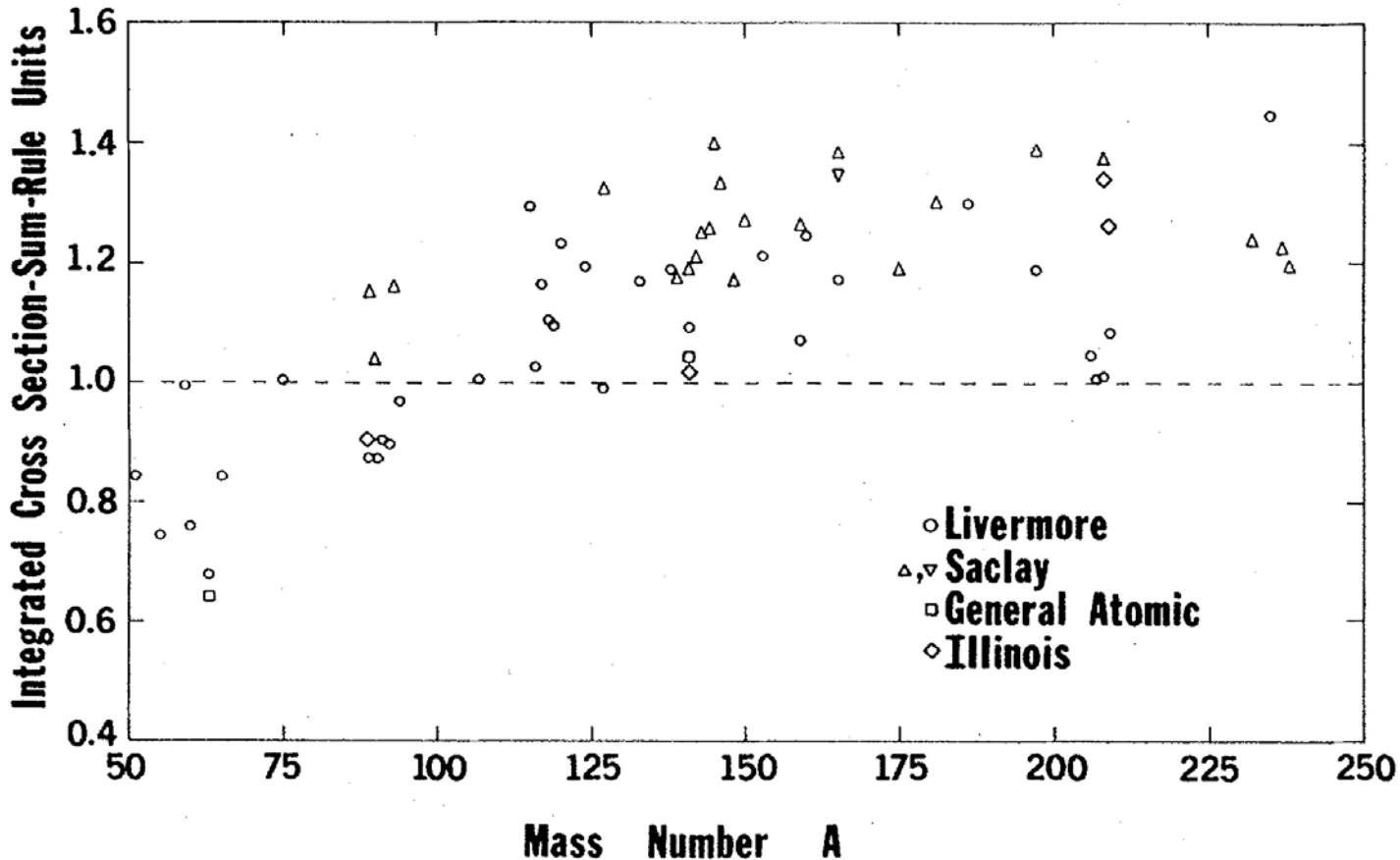

FIG. 47. Extrapolated integrated cross sections derived from the Lorentz parameters in units of 60NZ/A MeV-mb.

What's at stake in these discrepancies?

- For example, if one adopts the Berman et al. conclusions, nuclei have substantial and variable deviation of integrated absorption from the TRK sum rule.

- If one adopts the IAEA protocol, one comes to a similar conclusion, albeit with a higher normalization.

- If one combines tagged photon scattering data with that of Ahrens total absorption and Saclay's annihilation in-flight [this is my favorite option, essentially ALL nuclei obey 2 TRK sum rules from 0-140 MeV (+/- 5-10%)].

Who's Right?

So why hasn't photo-nuclear isotope production been exploited?

In two words: Specific Activity (or lack thereof)

SA is the Achilles Heel of photo-nuclear methods, especially the predominant cross sections of (γ, n).

So why hasn't PAA been exploited?

In three words: Atoms for Peace,
sometimes referred to as "cost."

So why hasn't Photo-Production been exploited? Chemical separations of (γ, charged-particle) reactions….

Example: ^{67}Cu – photo-production via (gamma, p):

The maximum Specific Activity of ^{67}Cu in a post-gamma-irradiated ^{68}Zn target is dominated by the impurities in the target and the impurities transmitted in the separation processes.

Need 1-10 kCi/g SA (reliably) at roughly 100 mCi/dose for clinical trials … eventual routine use in medicine will probably require approx. 10 kCi/g SA.

(In the U.S., ANL, IAC, RPI and others have led this effort, in parallel with efforts by BNL, LANL, Trace Life Sciences, etc. to use (out-going) light-ion reactions)

What about photo-nuclear (γ, n) reactions, which are the largest cross sections?
Can Kinematic Recoil Methods work?

Example: ^{99}Mo – photo-production via (gamma, n):

The Specific Activity of ^{99}Mo in a post-gamma-irradiated pure ^{100}Mo target is "typically" of the 1-10 Ci/g range (see Bennett, 1998 or TRIUMF, 2008).

But if one separates the ^{99}Mo from a post-irradiated pure ^{100}Mo target, the answer is very different. Nano-materials may enable one to produce (maybe):

better than 10 kCi/g specific activity of ^{99}Mo on a laboratory scale (tens of uCi) – can it be scaled up?

Separation via Photo-nuclear <u>Kinematic Recoil</u>: significant improvement in the specific activity of accelerator-based photon production?

- A mechanical separation technique can, in principle, use the kinematic recoil (≈ 10 - 20 keV) of the residual (post neutron emission) molybdenum nucleus to eject the material from the small diameter nanoparticle

nanoparticles of catcher material
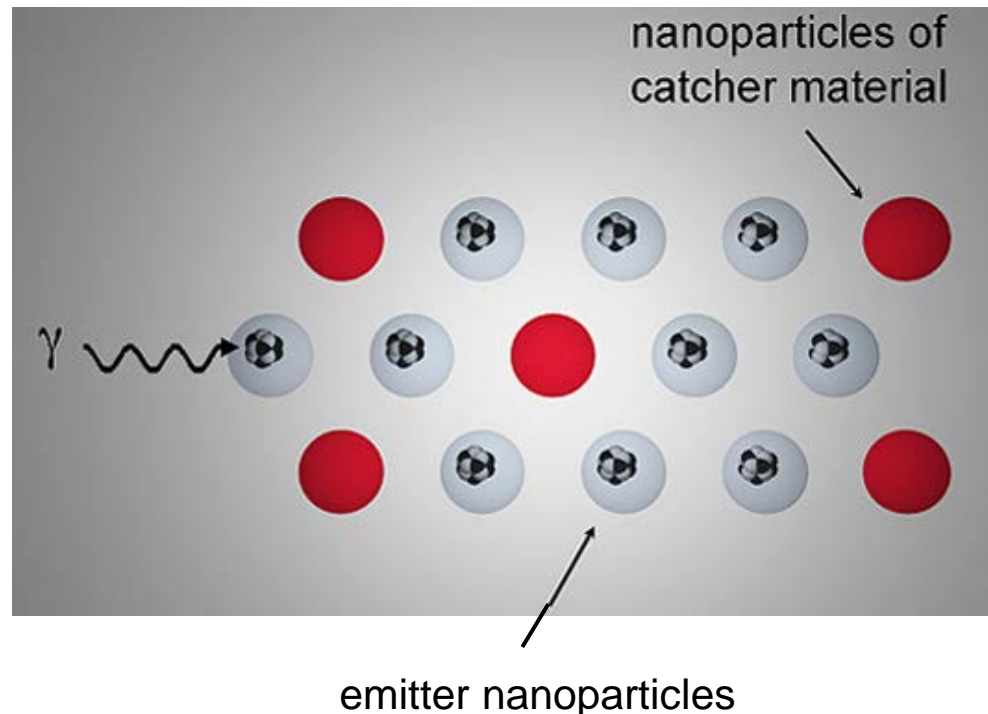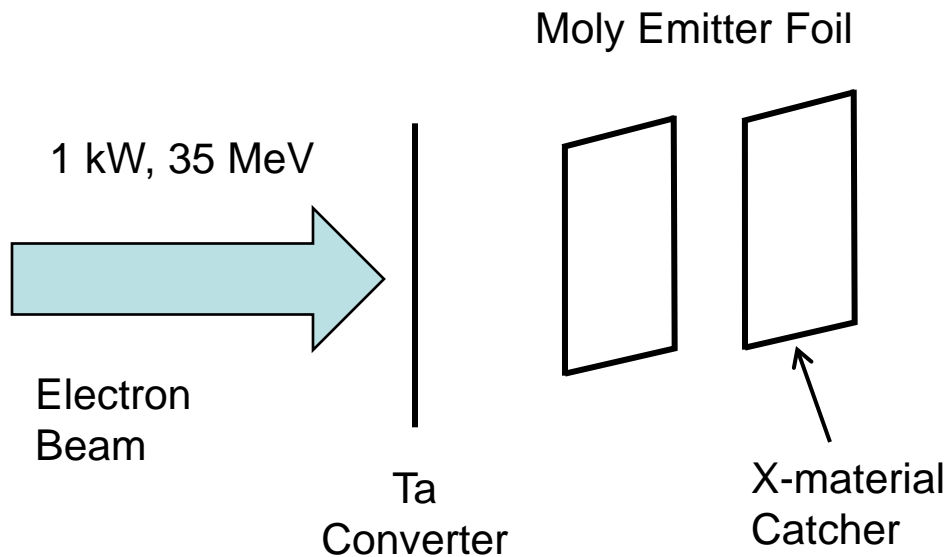
emitter nanoparticles

Illustration of one kinematic recoil emitter/catcher scheme. Here nanoparticles of the isotope production target (the emitter) are co-mingled with nanoparticles of catcher material.

Specific Activity in 'Catcher' Material
is controlled by non-^{99}Mo mass transfer:

Moly Emitter Foil

1 kW, 35 MeV

Electron
Beam

Ta
Converter

X-material
Catcher

- Non-^{99}Mo transfer by:
- Radiolytic Processes:
  - Measured to be "zero" up to ≈ 100 kGy with peak dose rates at approx. 33 kGy/sec.
- Knock-ons:
  - Calculated with SRIM to be approx. 50:1 ➔ 15 kCi/g ^{99}Mo
  - Calculations with nano-coatings on the Mo emitter ➔ 3:1 is possible ➔ ≈ 170 kCi/g ^{99}Mo

Conclusions

1) Photo-production of isotopes (and applications thereof) is an under-developed technology

2) Photo-nuclear cross sections are of fundamental interest to nuclear models and nuclear astrophysics

3) There are large gaps in photo-nuclear data that LERF could fill – probably better than anyone else.

Questions?