Cross section Measurements for Radiochemistry using RIA

L. A. Bernstein
M.A. Stoyer
LLNL

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Outline

- **What are Radiochemical Detectors and how are the used?**
- **What is the current status of radchem databases?**
- **Which Cross Sections do we need to measure?**
  - **5-level Toy Model for Zr network.**
- **How can RIA help?**
  - **Direct Measurements - Neutron-induced Cross Sections**
    - **Radioactive Target Assembly Measurements**
  - **Indirect Measurements**
    - **Charged Particle in Inverse Kinematics (Astro connection).**
      - (d,p) as a surrogate for (n,γ).
    - **Level Densities**
    - **Total Cross Sections**

What are radchem detectors and how are they used?

- **“Detector” elements are pre-loaded.**
  - **Neutron Monitors: Y,Zr,Lu,Bi are “most reliable”***
  - **Experiments to measure cross sections on stable Y,Zr,Lu,Bi were run in 1998-2000. Analysis is in progress**
  - **Ti, Cr etc. are used as charged-particle monitors**
    - **Neutron-induced reactions form $s_{destruction}$,etc.**
- **Isotope ratios are determined from post-test counting**
  - **Examples: $^{89}Zr/^{90}Zr_L$, $^{88}Zr/^{89}Zr$, etc.**
- **Vast majority of cross sections are calculated NOT measured**
  - **Models are qualitative at best.**

*Nethaway & Mustafa UCRL-ID-133269

Example: The Zr Cross Section Network

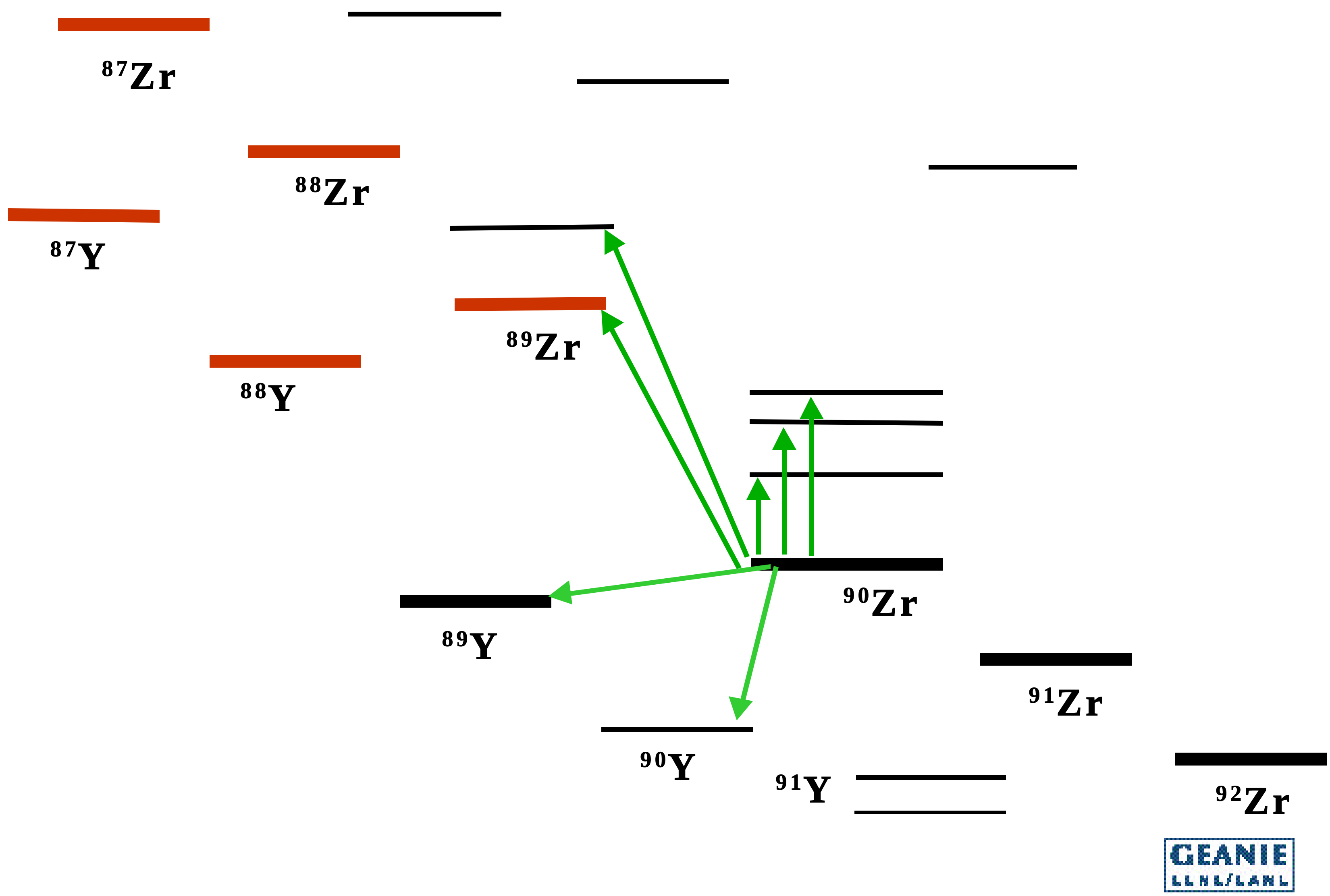
Reaction	Knowledge
$^{90}Zr(n,2n)^{89g}Zr$	Sum (g+0.94m) known to 2% for E_n = 13-15 MeV
$^{90}Zr(n,2n)^{89m3}Zr$	
$^{89}Zr(n,2n)^{88}Zr$	Calculation only
$^{89}Zr(n,g)^{90m4}Zr$	Calculation only
$^{89}Zr(n,g)^{90}Zr$	Calculation only
$^{88}Zr(n,g)^{89m3}Zr$	Calculation only
$^{89m3}Zr(n,2n)^{88}Zr$	Calculation only
$^{90m4}Zr(n,2n)^{89}Zr$	Calculation only

Guessing a 10% uncertainty is unrealistic
(30% uncertainty is closer to reality)

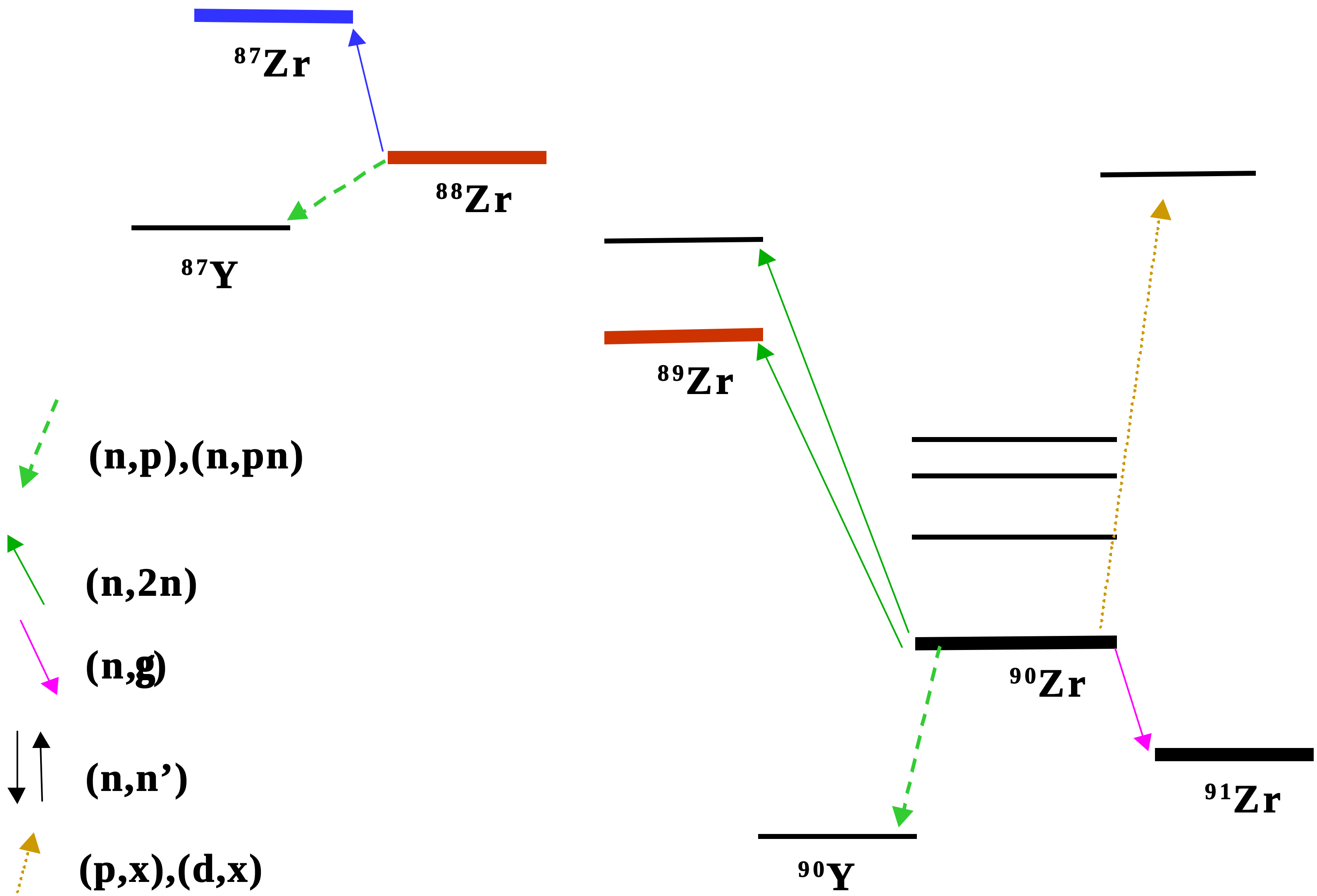
Current Status of the Nuclear Data
(from Nethaway & Mustafa UCRL-ID-133269)

- **^{50}Cr (Cr0386):**
  - **2 measured values for $^{50}Cr(n,t)^{48}V$ @ 14.6 MeV differ by 65%**
  - **$^{50}Cr(n,np)^{49}V$ is based on a single measurement @ 14.7 MeV. Excitation function is ACTL X 1.7 to match @ 14.7 MeV.**
  - **29 other neutron reactions are solely based on ACTL values.**
- **^{90}Zr (Zr0892):**
  - **All were calculated by M. Gardner & D. Gardner.**
  - **Calculations were normalized to measurements :**
    - **$^{90}Zr(n,2n)^{89g+0.94m}Zr$ agrees within 2% from 13-15 MeV.**
- **^{89}Y (YT0585 & YT0488):**
  - **All were calculated by M. Mustafa, M. Gardner & D. Gardner**

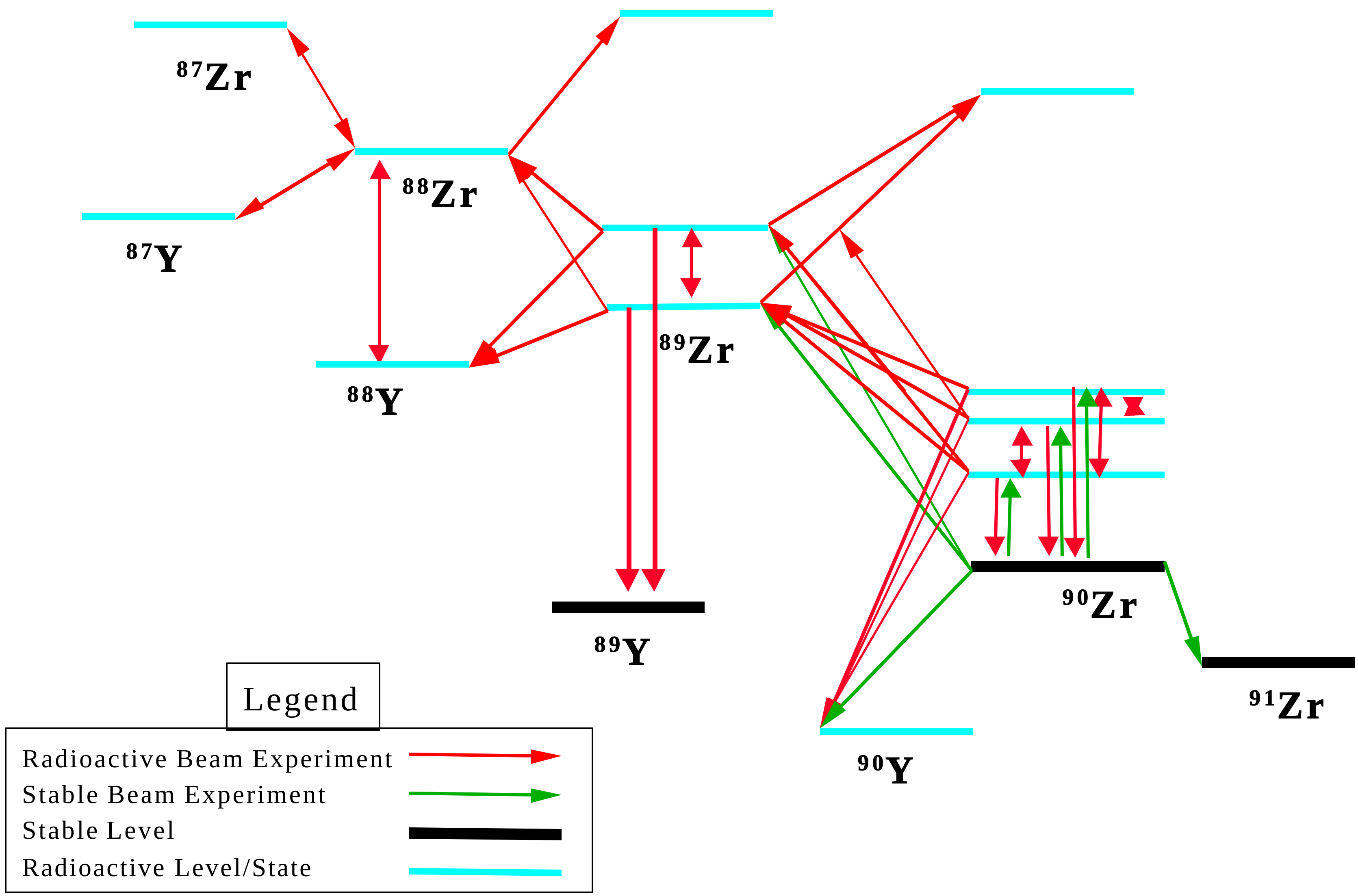
What we measured using GEANIE: $^{90}Zr(n,x)$



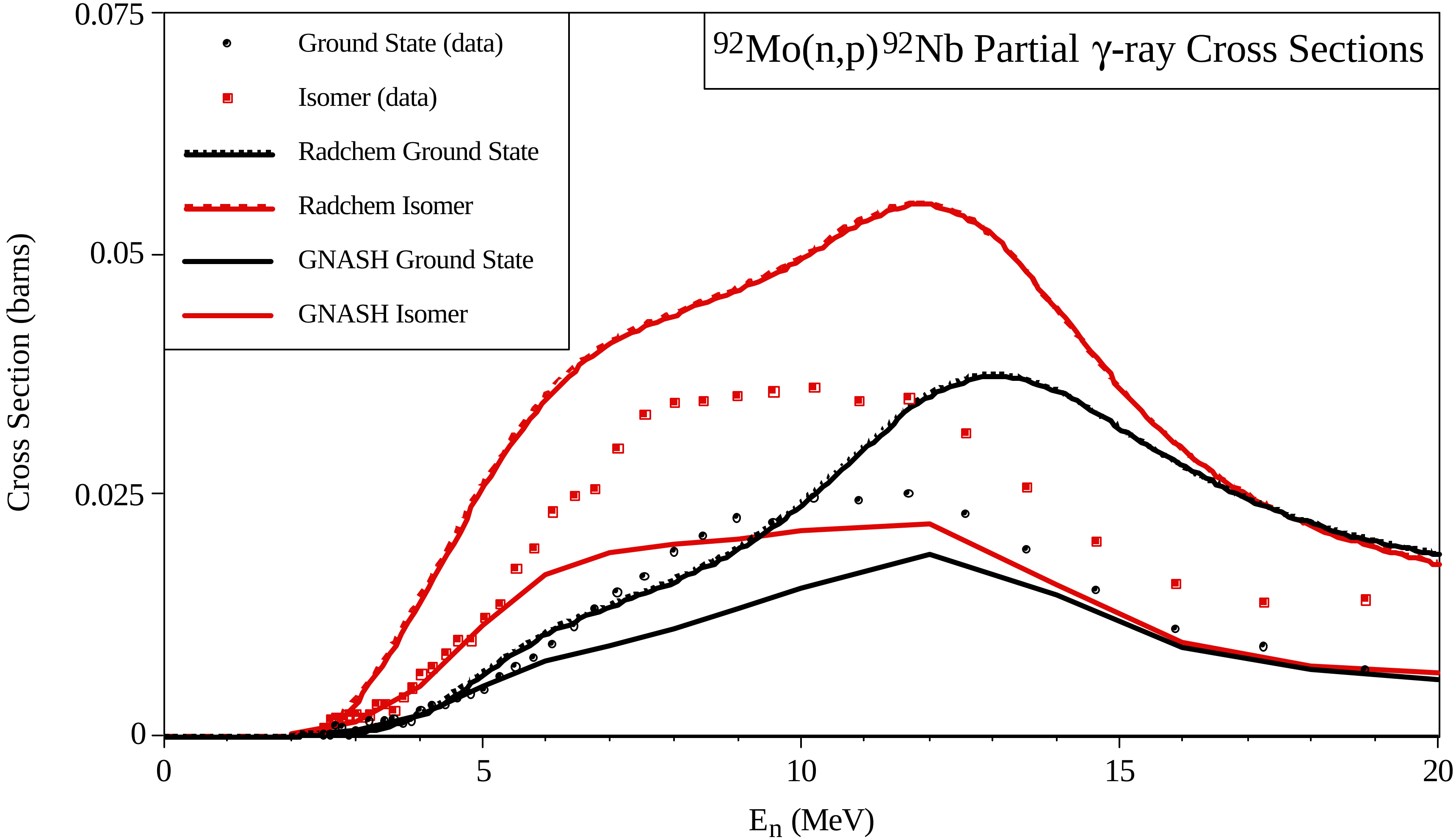
Zr cross sections for which some data exist



Most of the cross sections in a radiochemical network are measurable only using a radioactive target or beam



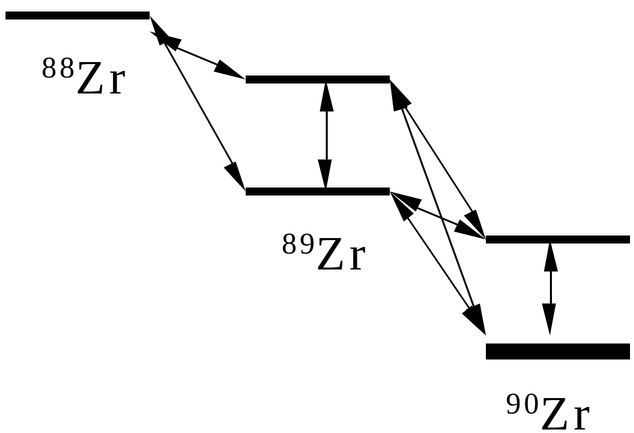
How reliable are the codes & libraries?



Models alone will not fill the need

Which of these Zr Cross Sections matter?
- Preliminary Sensitivity Studies have been done (E.A. Henry)

The Reaction Network



Impact of a change in the Cross Section

Reaction	10% change		50% change	
	$^{89}Zr/^{90}Zr_L$	$^{88}Zr/^{89}Zr$	$^{89}Zr/^{90}Zr_L$	$^{88}Zr/^{89}Zr$
$^{90}Zr(n,2n)^{89g}Zr$	7.2%	4.0%	36%	20%
$^{90}Zr(n,2n)^{89\ m3}Zr$	1.3%	2.0%	6.5%	10%
$^{89}Zr(n,2n)^{88}Zr$	**2.3%**	**8.4%**	**11.5%**	**42%**
$^{89}Zr(n,g)^{90\ m4}Zr$	**4.2%**	**1.1%**	**21%**	**5.5%**
$^{89}Zr(n,g)^{90}Zr$	**3.2%**	**0.9%**	**16%**	**4.5%**
$^{88}Zr(n,g)^{89\ m3}Zr$	0.2%	3.1%	1%	15.5%
$^{89\ m3}Zr(n,2n)^{88}Zr$	0.1%	1.6%	0.5%	8%
$^{90\ m4}Zr(n,2n)^{89}Zr$	1.0%	0.5%	5%	2.5%

Green: Stable (can be done now)

Red: Unstable (needs RIA/RIB)

Some of these cross sections need to be measured well.

How does this fit into the Campaign Structure?
- FY2001 DOE/Defense Programs Budget Request

- **Secondary Certification and Nuclear-Systems Margins (Campaign 4).**
  - ***Determine and document the minimum primary factors necessary to produce a militarily effective weapon.***

FY99-01 Performance Measure:

Complete the reevaluation of primary-yield determination (radiochemistry and prompt diagnostics analysis.)

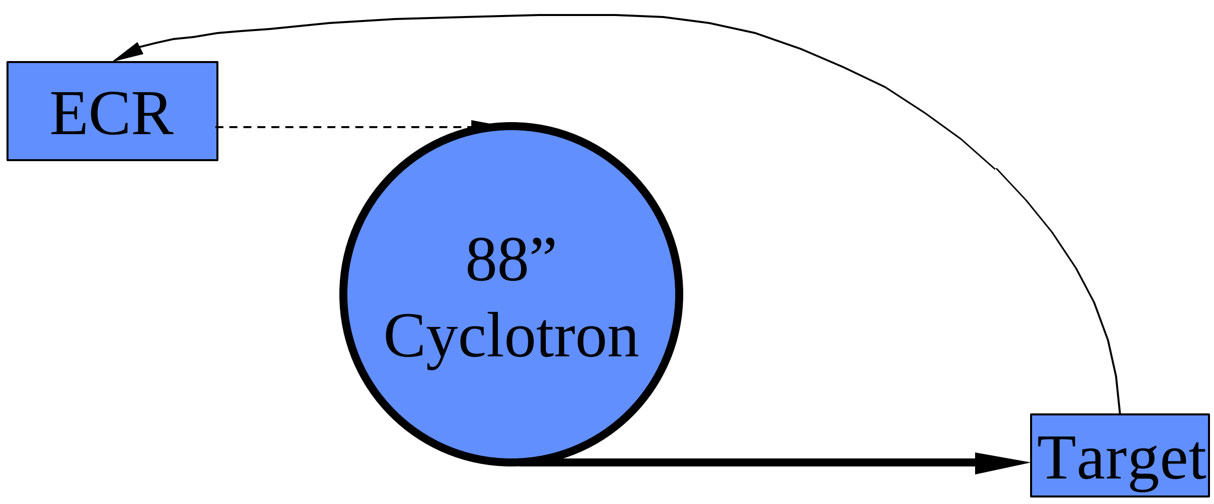
- **Primary Certification (Campaign 1)**
  - ***Supports experimental abilities to develop and implement the ability to certify, without nuclear testing, rebuilt and aged primaries to within a stated yield level.***

FY99-01 Performance Measure:

Evaluate historical test data for archiving.

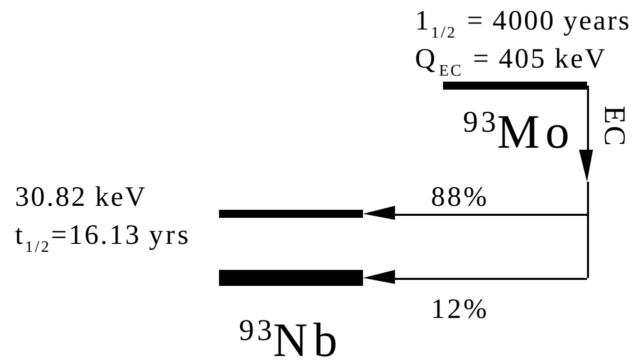
Re-cyclotron Technique

- **Use proton- or α-induced reaction to produce radioactive nuclei (e.g. $^{77}Se(p,n)^{77}Br$) with $t_{1/2}$>16 hours.**
- **Trap radioactive product and transfer to cyclotron ion source (Electron Cyclotron Resonance) Source.**
- **Re-accelerate radioactive beam**

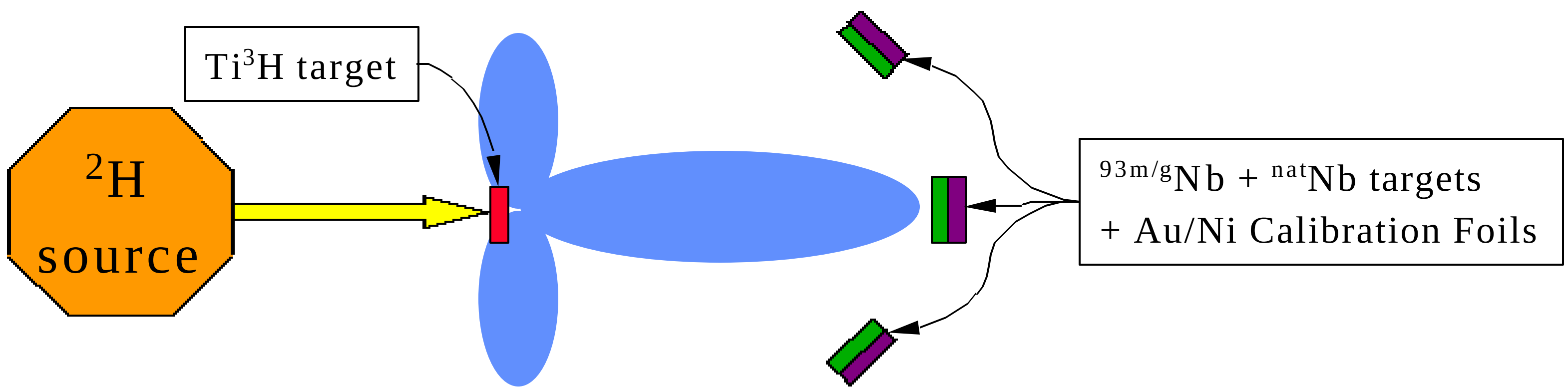


Isomers in Radchem Detectors done today:

^{93m}Nb(n,2n)^{92m}Nb/^{92g}Nb



- **To get a 1 mg target from 93 g "cow" requires 24 days.**
  - **This example can be done now. ^{93}Mo is commercially accessible.**



First Measurement of (n,2n) cross section in A=90 region

Example using Existing Facilities:

^{89}Zr(n,2n)^{88}Zr ($t_{1/2}$=78 hrs.) Count Rate Estimate

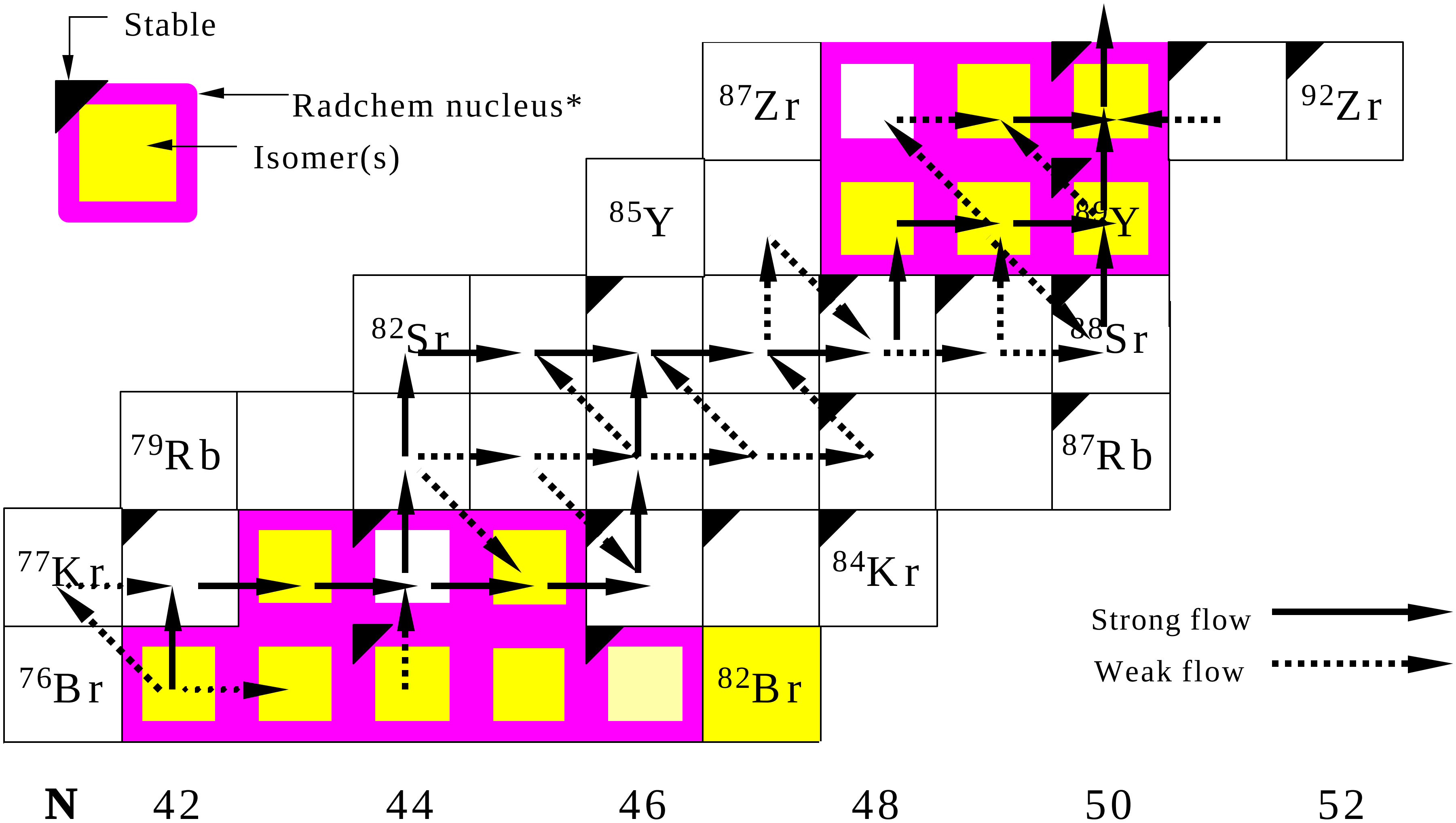
- **10 day Production Run at the 88”/UC Davis cyclotron:**
  - **Average s " 0.5 barn for ^{89}Y(p,n)^{89}Zr reaction @ E_p"12 MeV**
  - **I_{beam} " 10 pμa = 1 x 10^{14} protons/sec**
  - **Max t_{target}" 100 mg/cm^2 ® 6 x 10^{21} atoms (P_{target}= 40 W).**

N(^{89}Zr) = $\sigma_{(p,n)}$ x I_{beam} x N(^{89}Y) x t_{run} = 2.6 X 10^{16} atoms of ^{89}Zr

- **RTNS-II irradiation run (Dt"3 days)**
  - **e_{chem} = chemical separation efficiency " 50%, $s_{(n,2n)}$ " 500 mb**
  - **F_n = neutron fluence on target = 10^{11} neutrons/sec (challenge)**
  - **$t_{1/2}$(^{88}Zr) = 83.4 days**
  - **R_{branch}(^{88}Zr ® ^{88}Y + 392 keV g-ray) = 100%**

Peak: N(^{88}Zr→^{88}Y+ 392 keV γ–ray)=ε $\sigma_{(n,2n)}$ Φ_n N_{target} Δt R_{branch}=1.7 x 10^8
No Background after 30 days (10 $t_{1/2}$)
0.01% measurement possible

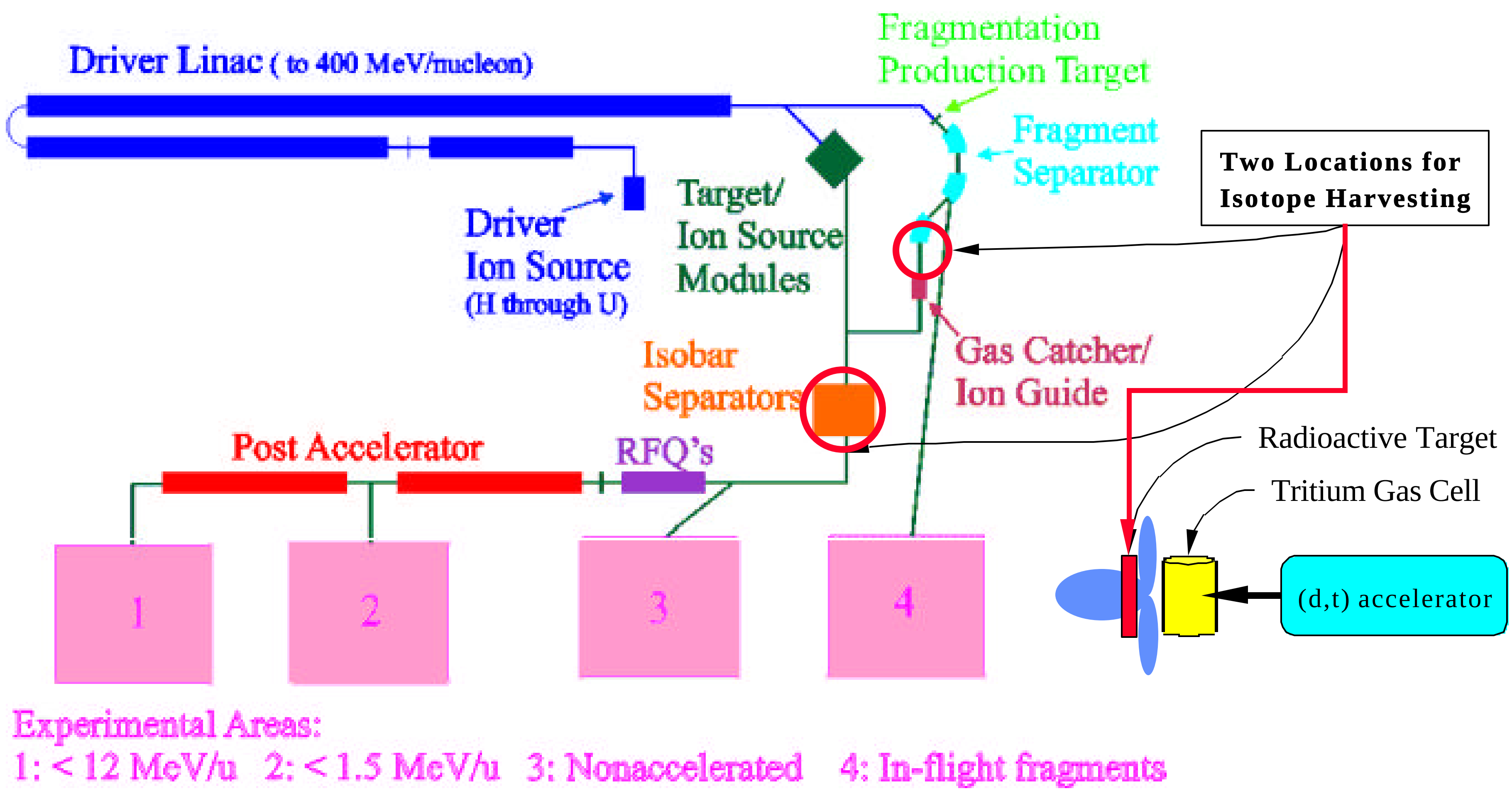
Radiochemistry: How elements are formed in devices[1]
Astrophysics: How elements are formed in stars[2]



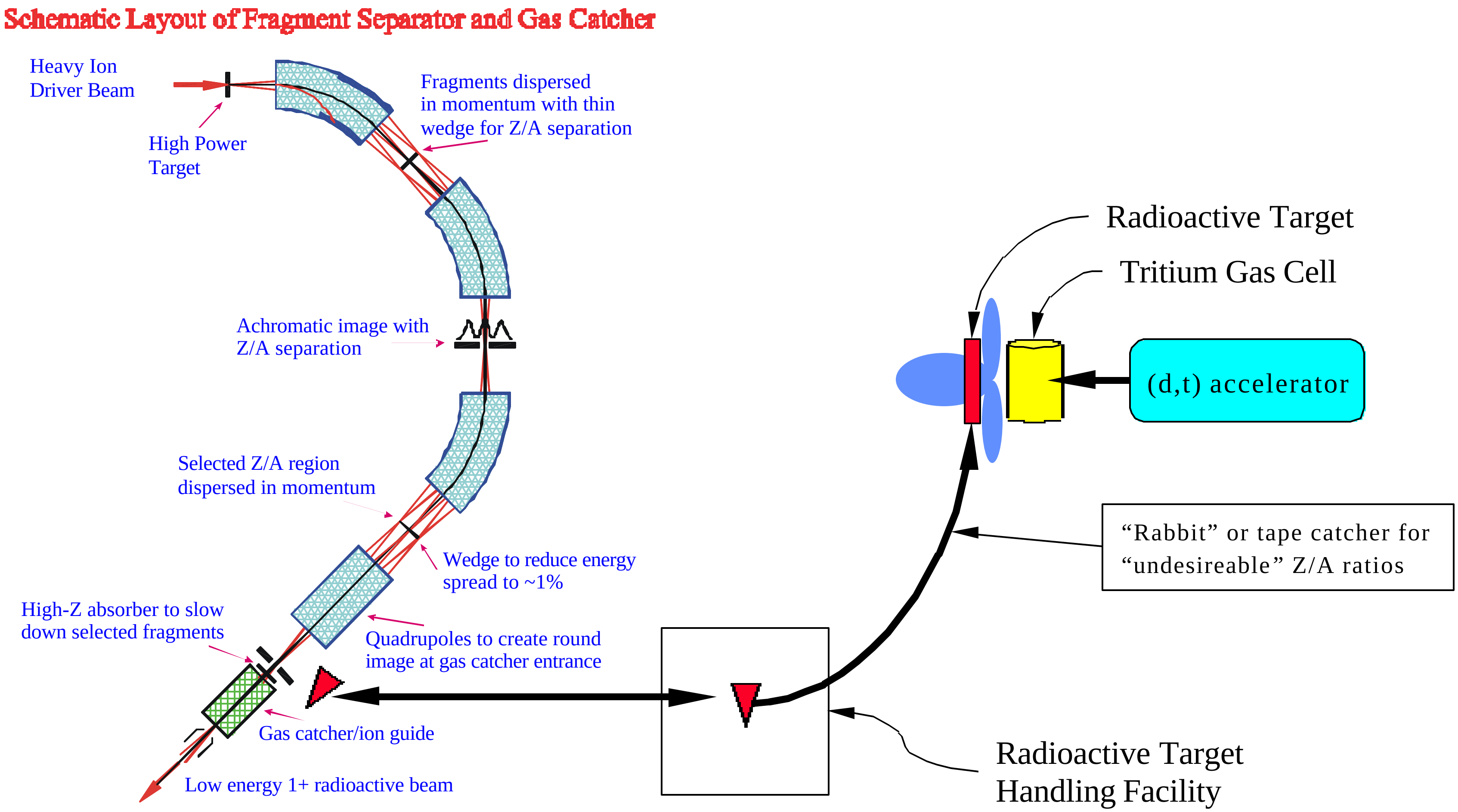
[1]Nethaway & Mustafa UCRL-ID-133269

[2]Hoffman *et al.*,1996 ApJ 460: 478

How can RIA help?
Harvesting isotopes for radioactive target assembly.



The IGISOL target offers an opportunity to allow RIA to run in a multi-user mode



Uses include Radioactive Target & Medical Isotope Production

Two possible neutron sources

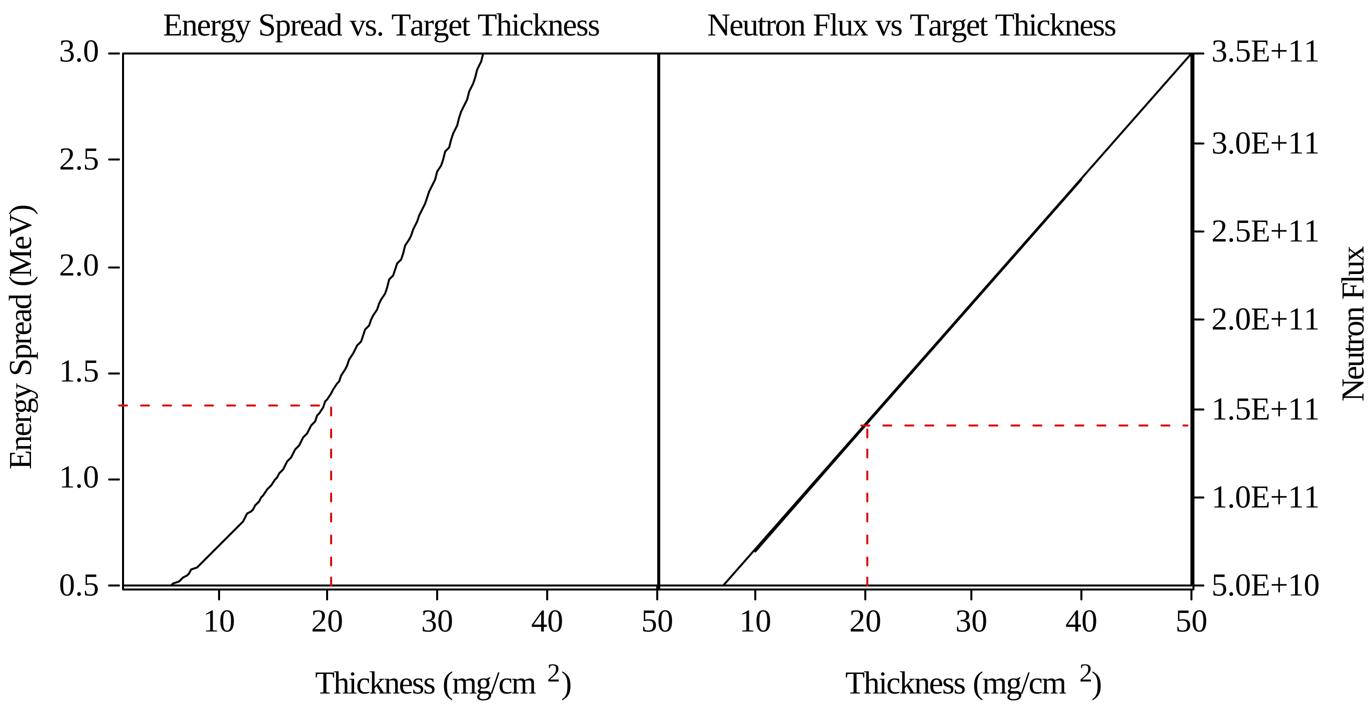
- The ICT/RTNS (Built at LLNL. Currently U.C. Berkeley)
  - Deuterium Linear accelerator + Ti^3H target.
  - $I_{max} = 6 \times 10^{12}$ neutrons/sec into 4π. (At 14 MeV only)
- The neutron-radiography source (Jim Hall & Brian Rusnak)
  - Prototype to be built at LLNL, duplicated at Y-12 by E.S.P.
  - RFQ + DTL "after-burner" + Window-less plasma arc D-gas cell.
  - 10^8 neutrons/sec with $1 < E_n$ (MeV)< 20 well focused into 1.0 cm^2.
- The machine would also be useful to for non-neutron physics:
  - 4-20 MeV ^1H or ^2H and 10-40 MeV α's for transfer reactions
    - Doesn't matter that the target is radioactive
  - $^7Li(p,n)$ low-energy neutrons for s- and r-process studies.

Both Sources Represent Modest Expenditures for DOE/DP

The Neutron Radiography Source:

- **Energy Spread Limits Target Thickness ® Neutron Flux**

$^{7}Li(p,n)^{8}Be$ at E_p = 5 MeV



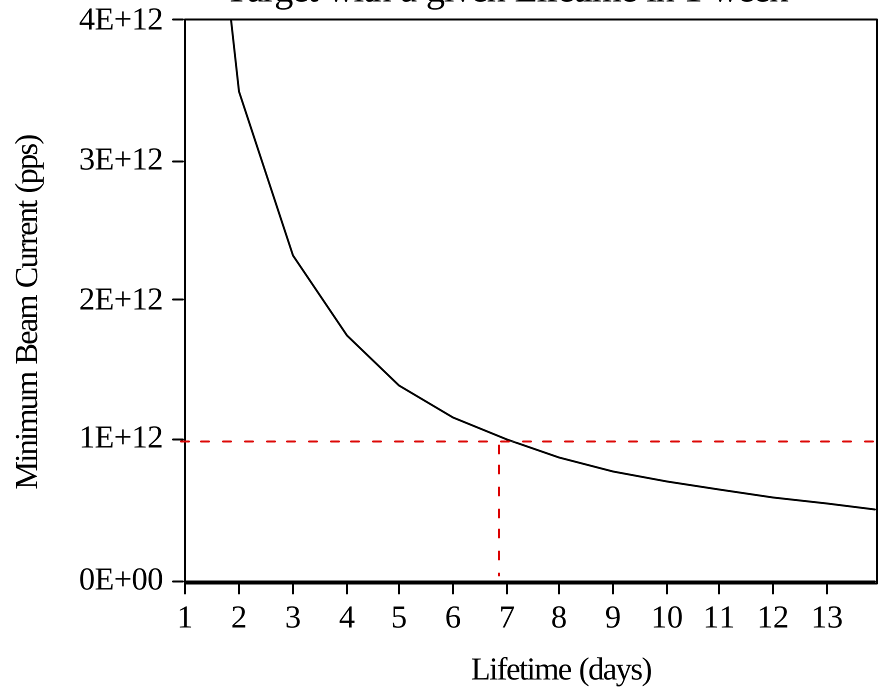
Practical Limit is 10^{11} neutrons/second/sr for Li, d etc.

What are the shortest lived states we can make targets of using RIA?

The equation governing the target material build-up:

$$dN(t)/dt = I - \lambda N(t) \rightarrow N(t) = I/\lambda + e^{-\lambda t}$$

Current Required to Assemble a 100 µg
Target with a given Lifetime in 1 week



Radioactive target experiments require full beam intensity

An Example of a direct (n,2n) measurement that requires RIA: $^{87}Y(n,2n)^{86}Y$ ($t_{1/2}$= 80 hours)

- **Neutron irradiation run (3 days)**
  - **t = decay of target during run = 0.6**
  - **$s_{(n,2n)}$ = 500 mb**
  - **F_n = neutron fluence on target = 10^8 neutrons/sec on target**
  - **Measure neutrons *vis-a-vis* Frehaut *et al.*,**
- **Peak = $t\ s\ F_n N_{target} Dt$ =4.8 x 10^7**
- **Statistical errors on the order of 0.01% (Systematics will dominate).**

Even less material would work, but target manufacture would be tough

An Example of a direct (n,2n) measurement that we can do today: $^{48}V(n,2n)^{47}V$ ($t_{1/2}$=16.0 days)

- **Neutron irradiation run (10 days)**
  - **6 x 10^{17} = atoms of ^{48}V collected. 1/2 decay during during.**
  - **$s_{(n,2n)}$ = 500 mb**
  - **F_n = neutron fluence on target = 10^8 neutrons/sec**
  - **Neutron Efficiency = 75%**
  - **Measure neutrons *vis-a-vis* Frehaut *et al.*,**
- **Peak = $s\ F_n N_{target} Dt$ =9.8 x 10^6**
- **Statistical errors on the order of 0.1% (Systematics will dominate).**

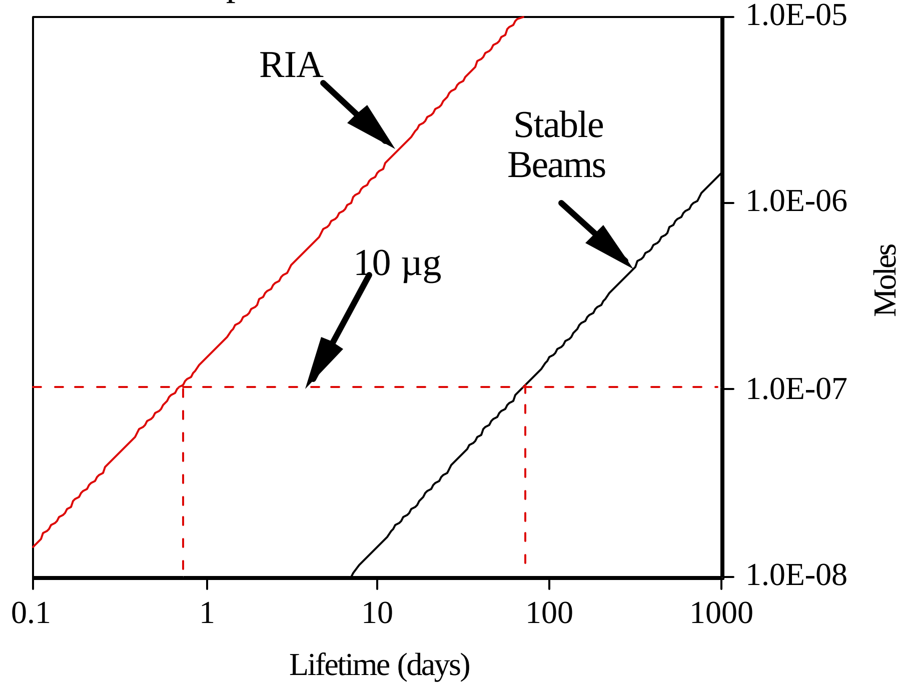
Even less material would work, but target manufacture would be tough

RIA compared to a Compound Nucleus Reaction etc. done today:

- **s=0.5 barn for $^{89}Y(p,n)^{89}Zr$ reaction @ E_p=12 MeV, I_p=10 μa**
- **100 mg/cm^2 target ® 10^{20} atoms (P_{target}= 40 W), 10 day run**

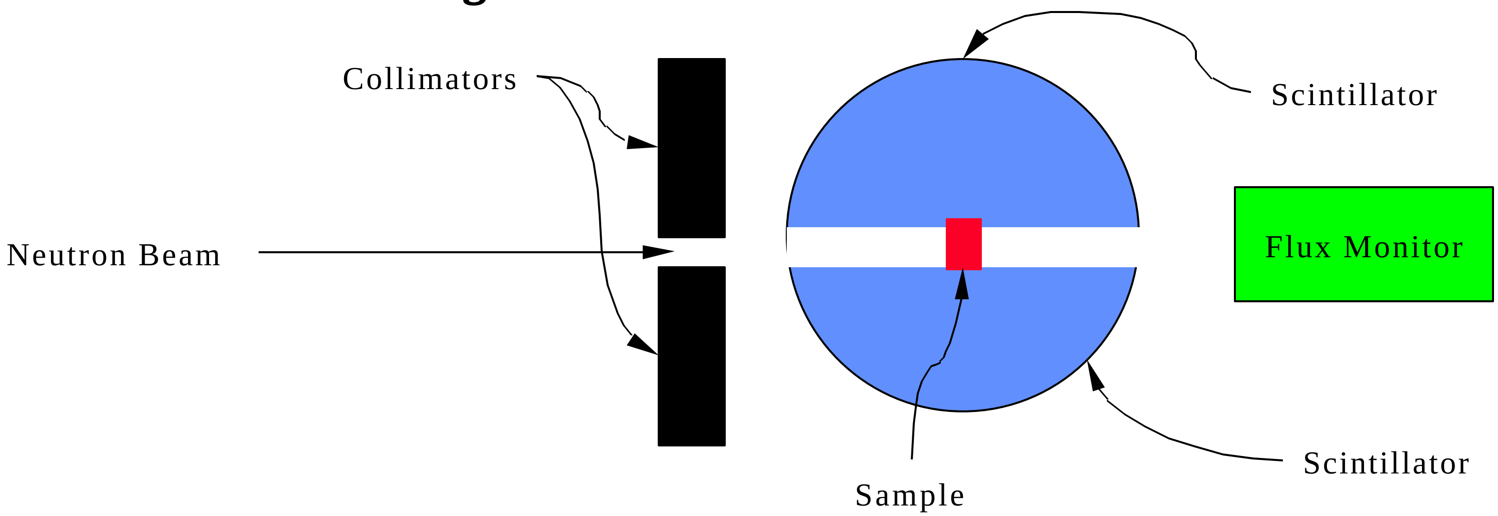
$\sigma_{(p,n)}$ x I_p x N(^{89}Y) x t_{run} = **10^{10} s^{-1}** of ^{89}Zr
RIA Production run - **10^{12-13} s^{-1}**

Time required to make a target using Stable Beams/Compound Nucleus Reactions vs RIA.



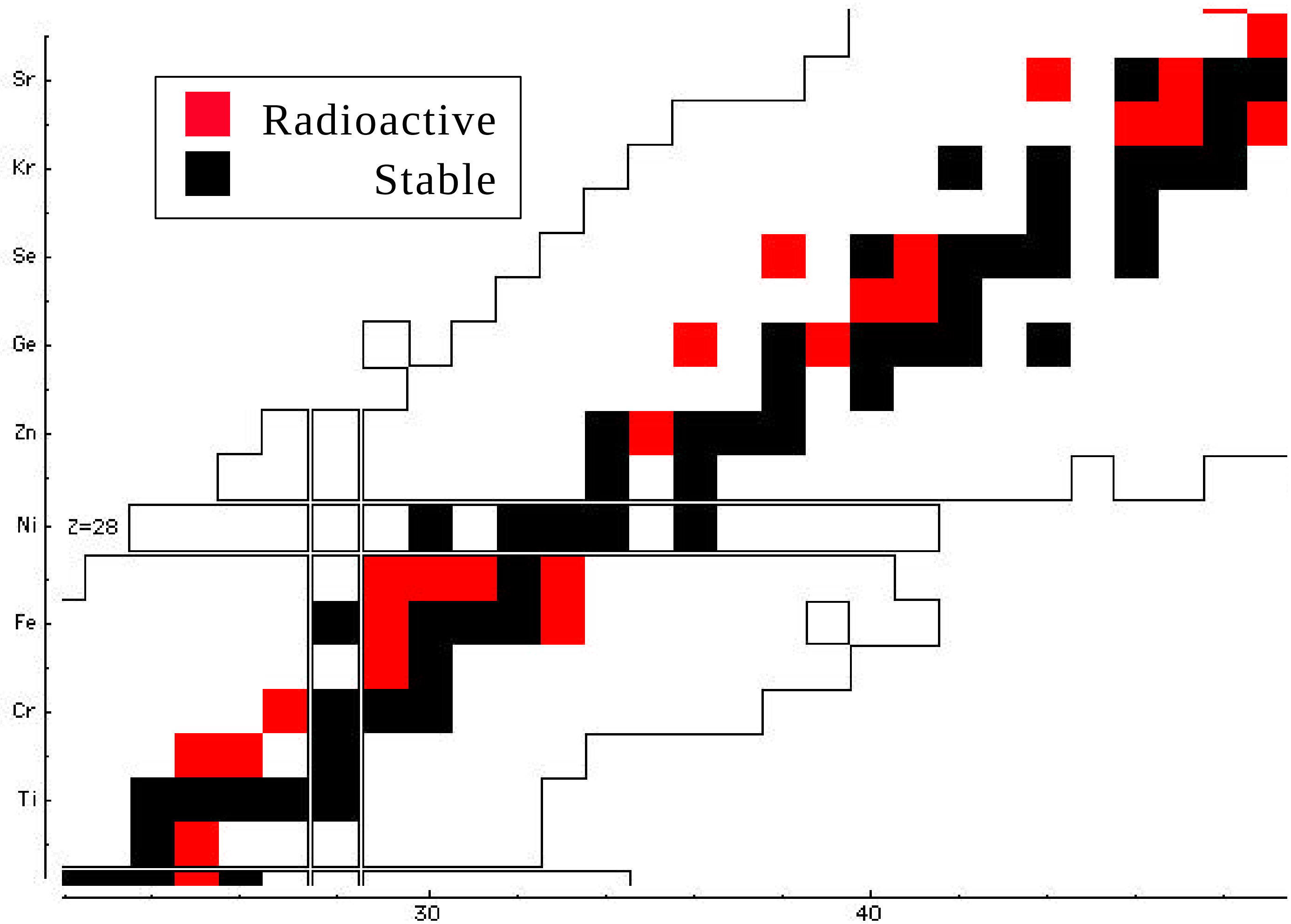
The Approach of Choice: Measure Evaporated Neutrons (J. Frehaut *et al.*,)

- **Use Collimated Neutron Beam and Gd-imbued Scintillator**
  - **Excellent efficiency (75%)**
  - **No neutron background for nuclei w/o fission.**

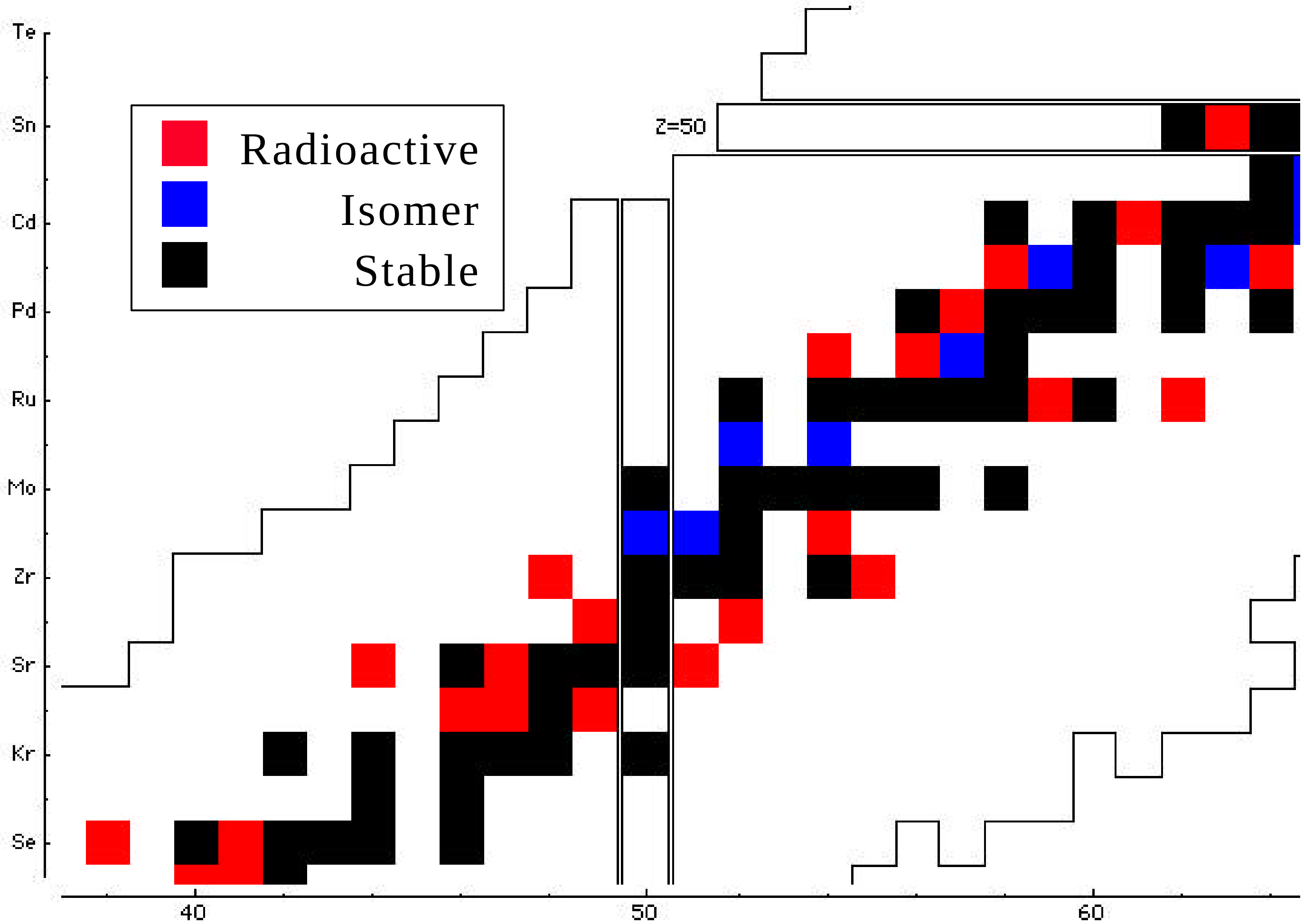


The new neutron source is nearly 4 orders of magnitude more intense than the one used in the earlier measurements

Potential Radchem Targets from RIA near A=50



Potential Radchem Targets from RIA near A=90

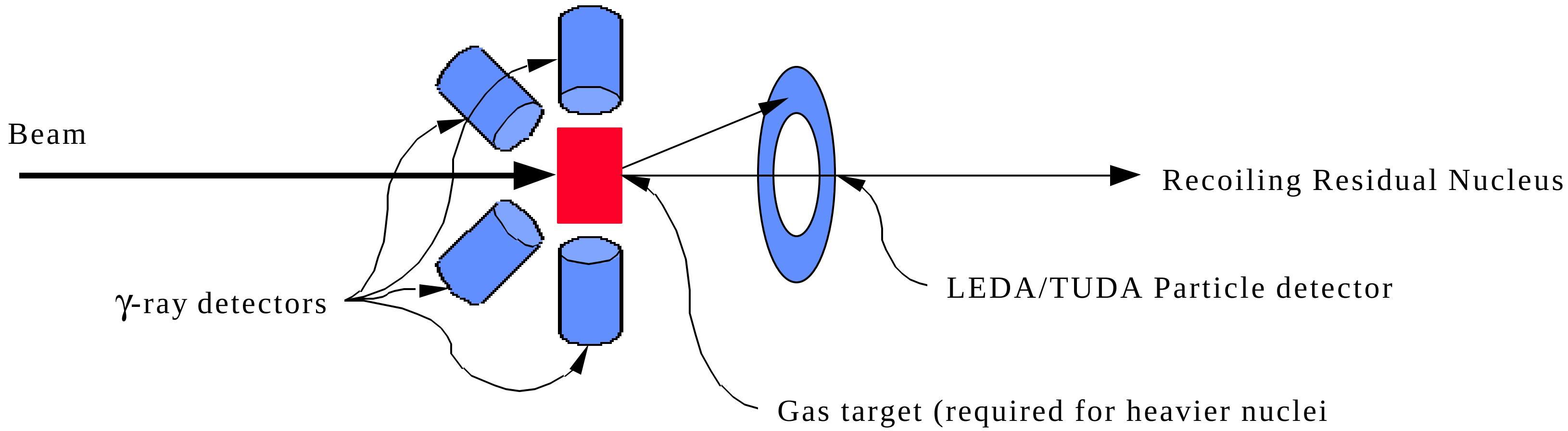


Indirect Measurements -
Improving the Reaction Model Calculations

- **Direct Component of (n,g) using transfer reactions**
  - **Radioactive Target with d-beam or Inverse Kinematics w/RIB.**
  - **Requires less material than neutron target assembly (10-100 µg ® 10^{16-17} atoms).**
  - **Radioactivity of the target not an issue**
- **Level Density Measurements**
  - **Statistical g-ray measurements need RIBS to access near stability nuclei over a wide spin range.**
- **Total Cross Section Measurements (Optical Model)**
  - **Transmission experiments using RIBS**

Indirect Technique #1:
Transfer Reaction using RIBs to get $\sigma_{direct}(n,\gamma)$

- **Accelerate beam onto a p/d/t gas target (Inverse Kinematics)**
- **Measure the reaction cross section using γ-ray/particle tag.**
- **Complementary to Astrophysics measurements**

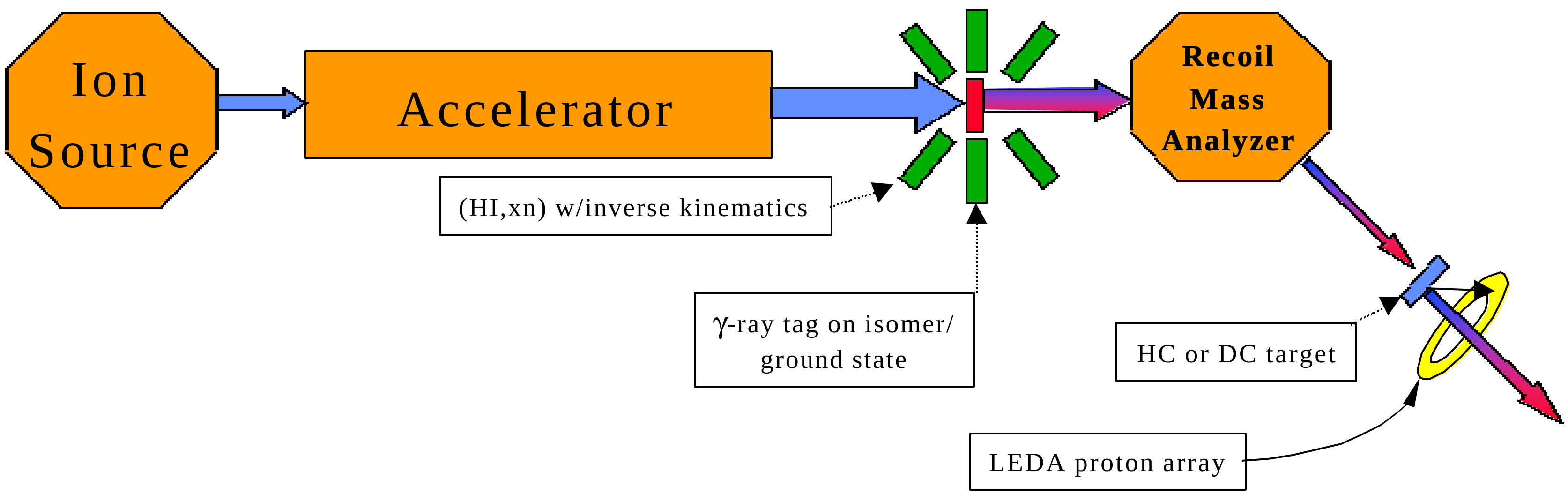


Removes a significant source of uncertainty in the reaction model

Program: Cross Sections. Facility: ANL

Cross Sections on Isomers w/secondary beams

- **Ground/isomer beam formed via multi-fragmentation.**
  - **(1) g-ray tagging at production target identifies whether isomer or ground state is populated.**
  - **(2) Measure s_{isomer} & $s_{ground\ state}$ at back-plane.**
  - **(3) Post-doc to be hired pioneered the effort (J.Schwartz)**



Maintains our involvement with ANL Personnel

Secondary Beams Cross Section Measurements: Count Rate Estimates I

- **Secondary Beam Production at the FMA entrance:**
  - $s_{(HI,xn)}$ " 200 mb (e.g., ^{24}Mg(^{58}Ni,3pn) etc.)
  - I_{beam} = 1 pμa
  - t_{target} = 1 mg/cm^2 ® 1.6 X 10^{17} atoms

$$I_{secondary} = \sigma_{(HI,xn)} \text{ X } I_{beam} \text{ x } N_{target} = 1.6 \text{ X } 10^{6} \text{ particles/second}$$

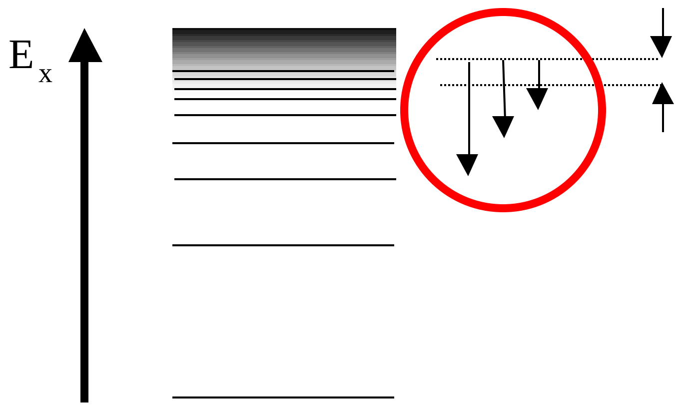
- **Measurement at the back-plane of the FMA:**
  - e_{FMA} " 20%
  - t_{target} = 5 mg/cm^2 (Gas Target Required) ® 4 X 10^{18} atoms
  - $s_{(p,p')}$ " 1 barn

$$R_{proton\text{-}array} = \varepsilon_{FMA} \text{ X } \sigma_{(p,p')} \text{ X } I_{secondary} \text{ x } N_{target} = 6.4 \text{ events/second}$$

8 hours for 1% statistics into each of 10 proton angles

Indirect Measurements Technique #2:
Level Densities & g-ray Strength functions from g-rays.

- **Step I: Use neutron-rich RIB to access near stability nucleus**
- **Step II: Measure "First Generation" of g-rays, G(E_x,J^p)**



- **Step III: Assume G(E_x,J^p) = r(E_x,J^p) X F(E_x, E_g) and iteratively adjust r and F to minimize c^2.**
- **Proof-of-principle experiment run 5/99 at LBNL - $^{24}Mg(^{3}He,p)^{26}Al$**
- **4 additional experiments to be run in Oslo - December 2000.**

Important for Hauser-Feshbach calculations including γ-decay

Conclusions

- Direct Measurements of Cross Section on Radioactive Nuclei and Isomeric states near stability would remove uncertainties in radchem network calculations.
- Indirect Measurements using transfer reactions, γ-rays, etc. could significantly reduce reaction model uncertainties.

What do we need to do now?

- Research needs to be done to allow for parallel "isotope harvesting" with RIA to make radioactive targets.
- Develop the high-intensity neutron source sited at RIA for Stockpile Stewardship, Astrophysics and Nuclear Structure (already happening).