NATIONAL NUCLEAR SECURITY AND OTHER APPLICATIONS OF RARE ISOTOPES

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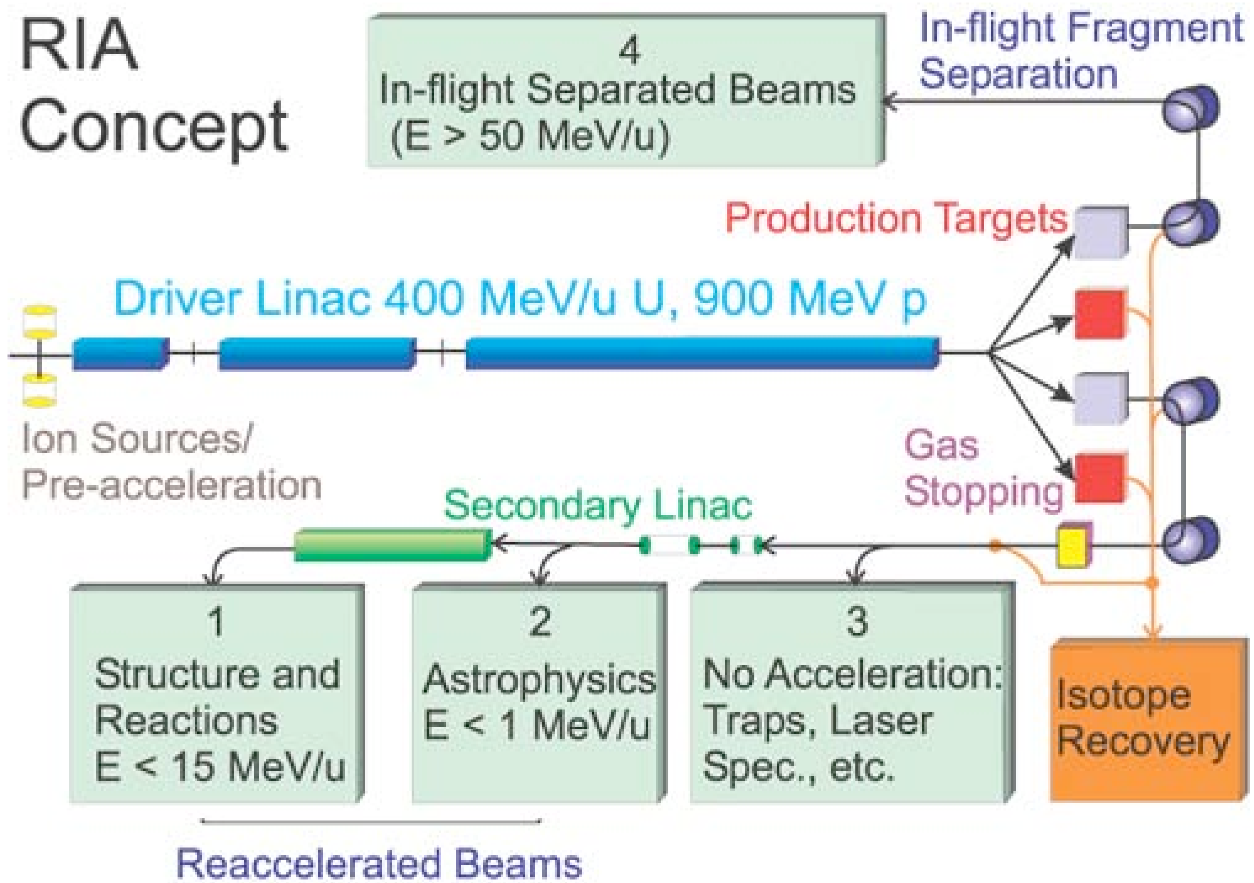
What is a Rare Isotope Beam?

- A Rare (Radioactive) Isotope Beam (RIB) is an energetic beam of multiply ionized non-stable nuclei.
- Nuclei with half-lives greater than one second can be produced easily in spallation, multi-fragmentation or fission.
- RIB's are necessary because:
  - Reactions with stable targets and beams allow the study of less than 10% of all bound nuclei
  - Radioactive targets have many problems including:
    - High backgrounds (β,γ,α)
    - Short half-lives
    - High Cost, Safety considerations, etc.

The Rare Isotope Accelerator (RIA)

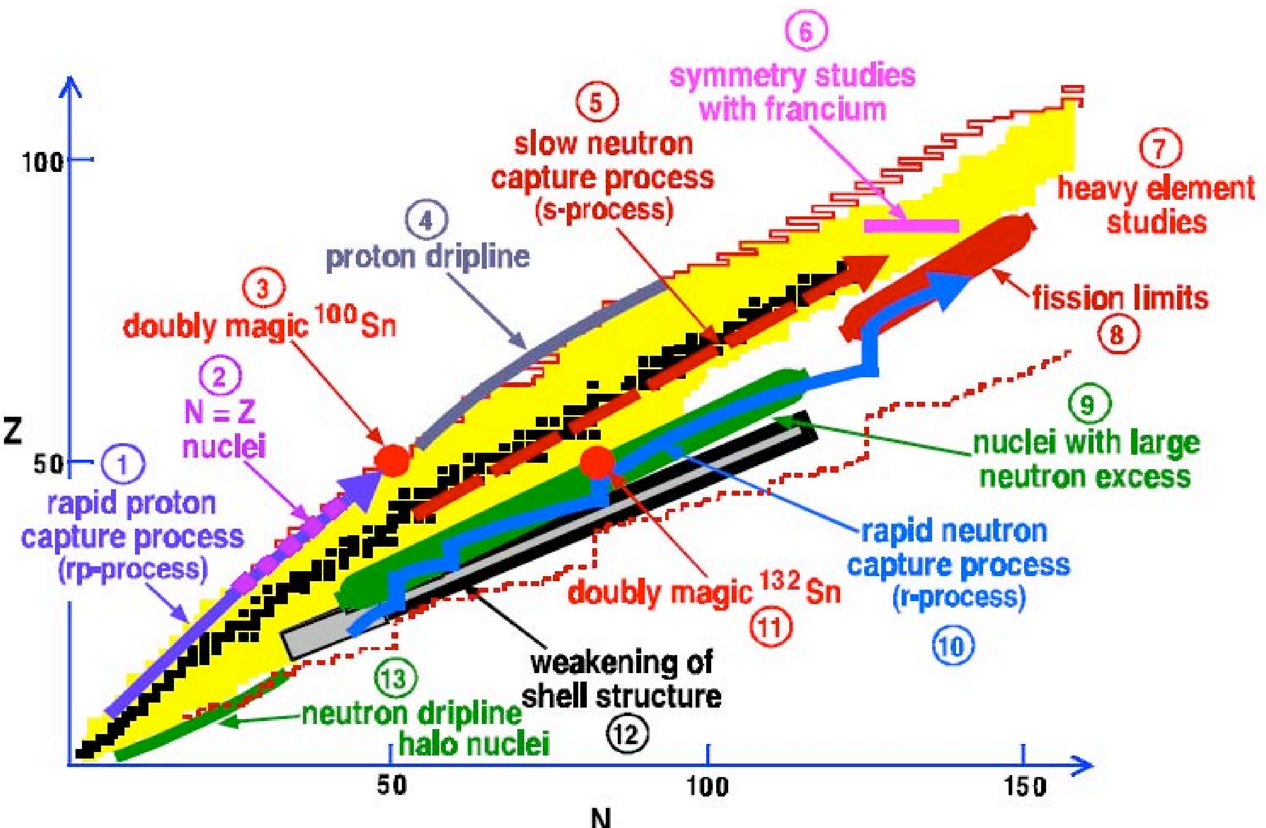
RIA will produce radioactive isotopes via:

- **proton beams on low Z targets (ISOL)**
- **heavy ion beams on low Z targets (Fragmentation)**



RIA is a proposed nuclear physics accelerator facility which will dramatically increase the number of different radioactive isotopes available for experimental measurements.

The Justification for RIA is the Wealth of Nuclear Science



Exploring the nuclear force in systems far from stability

- **Changing of "Magic Numbers" – closed shell in single particle level scheme**
- **Location of Neutron Drip Line – limit of nuclei existence**
- **Nucleosynthesis of elements heavier than iron – NRC grand challenge**
- **Test of fundamental symmetries – Parity and time reversal**

There are important applications of RIA technology

- *National Security Mission*
  - Stewardship of the Nuclear Stockpile
  - Homeland Security
- *Nuclear Energy*
  - Global Nuclear Energy Partnership
- *Oncology Research*

  New Medical Isotopes

Stockpile Stewardship

•Premise

World-class science, with detailed engineering investigations and high-fidelity three-dimensional simulations, can maintain a reliable nuclear weapons stockpile

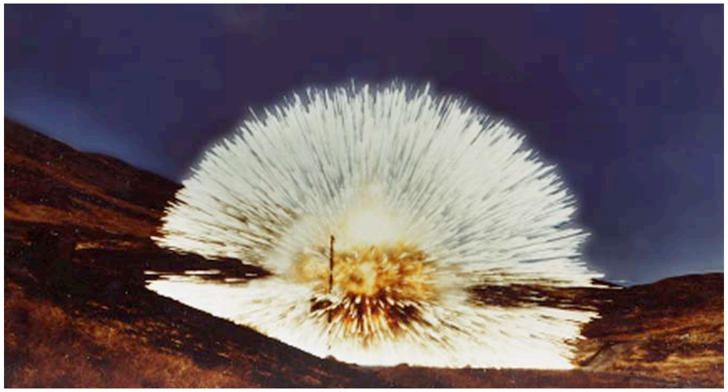
•Requirement

Dynamic, vital and broad physical science community that engages the specific issues that matter in nuclear weapons

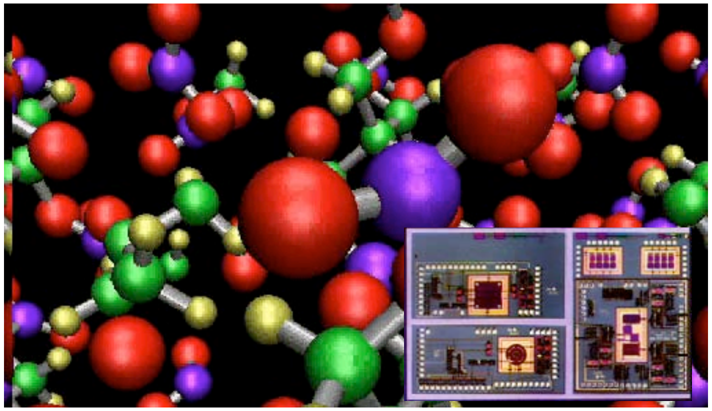
Computing & Simulation

High-Energy-Density Physics

Hydrodynamics



Materials, Physical Data, and Microsystems



Stewardship Goals and RIA

- ✓ **Measure cross sections and reaction rates on unstable nuclei**
  - **Allows neutron flux measurements in environments with *very high* instantaneous fluxes**
- **Conduct detailed studies of fission processes (mass distributions, lifetimes etc.)**
- ✓ **Fill major holes in nuclear data bases**
- ✓ **Guarantee a source of low energy nuclear physicists and nuclear chemists for the NNSA labs.**

Example of a key SBSS problem

- **Determine the neutron energy spectrum, flux, and angular dependence in environments with extremely high instantaneous neutron flux.**
- **Such fluxes exist in only a few places:**
  - **Inside stars**
  - **Near an ignited capsule at the National Ignition Facility**
  - **Archival nuclear test data**
- **Interpreting experimental observations is difficult.**

Stockpile Stewardship

Key challenge in past nuclear weapon tests is measuring neutron flux during test.

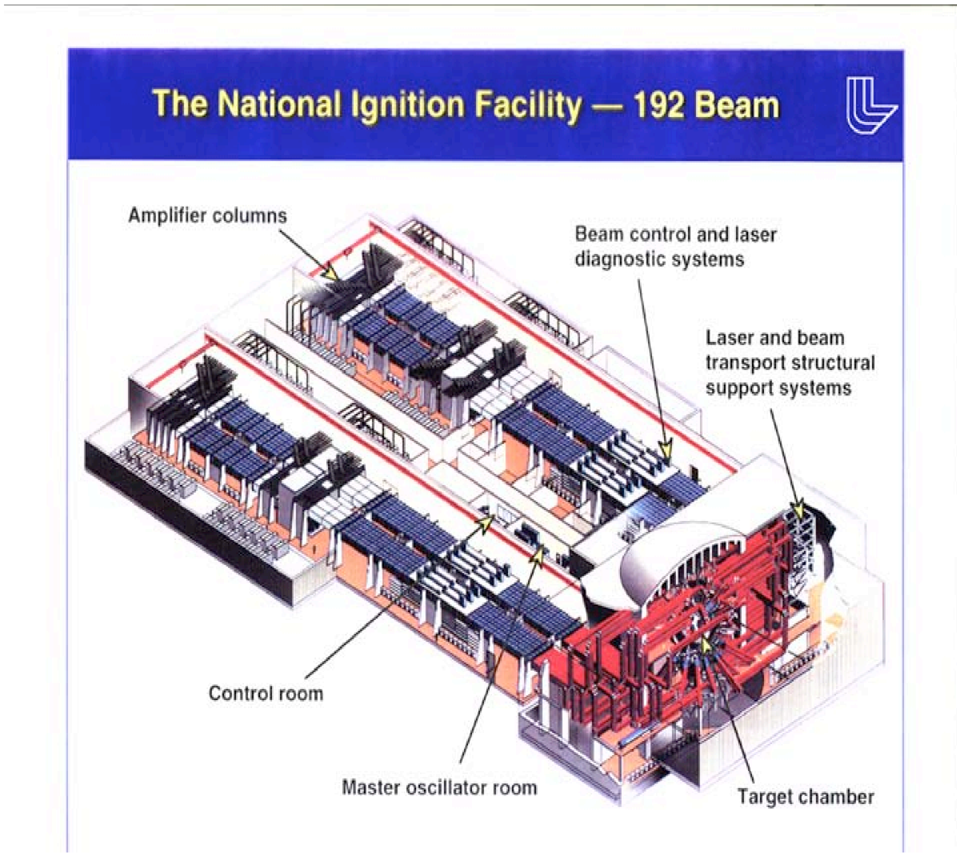
Answer: Use certain isotope as neutron flux monitors.

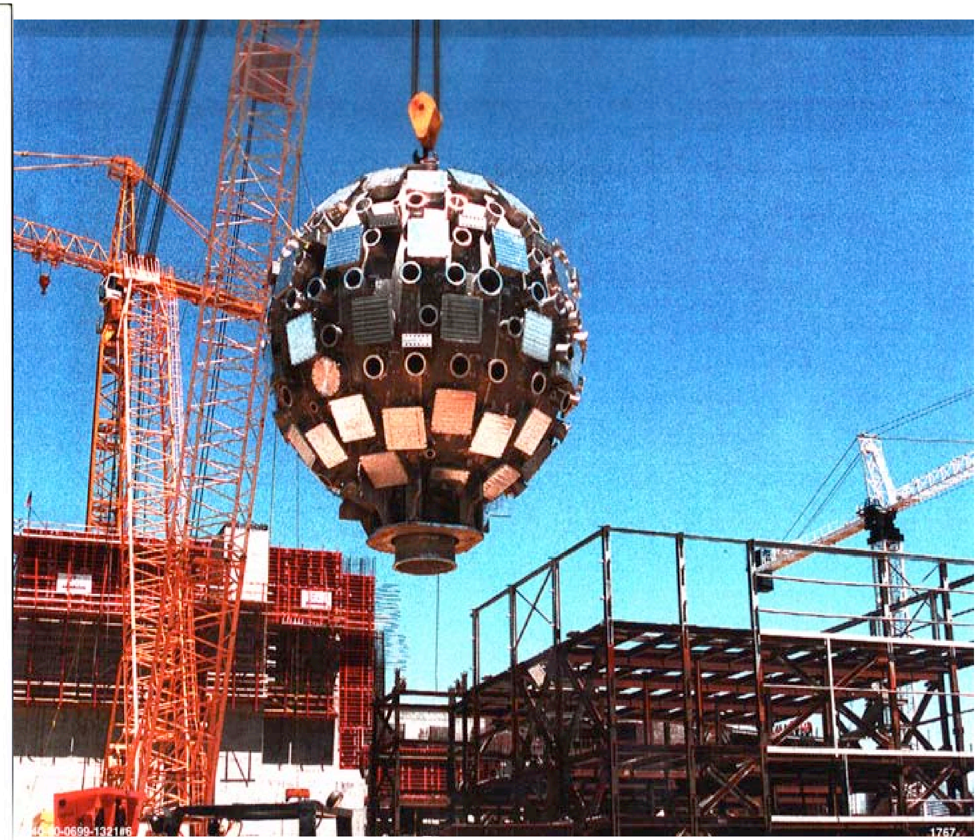
1. **Load isotope into device.**
2. **Extract core sample after test and perform radiochemical processing.**
3. **Interpret measured isotope production to infer information about neutron flux (neutron cross-sections needed).**

Key challenge at present is to reduce uncertainty of interpretation.

Answer: Improve quality of neutron cross-section data.

Stockpile Stewardship is DOE program to improve modeling capability of nuclear explosions.


The National Ignition Facility — 192 Beam
Amplifier columns
Beam control and laser diagnostic systems
Laser and beam transport structural support systems
Control room
Master oscillator room
Target chamber

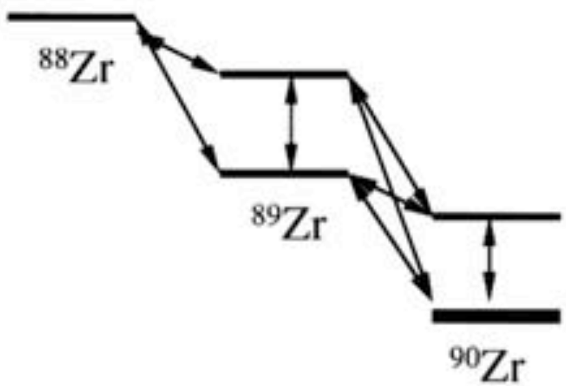


- **To measure the neutrons produced in a mm-sized hohlraum at the center of a 10 m target chamber:**
  - **Install foils of various isotopes**
  - **Measure daughter products after the implosion**
  - **(Often the neutron flux is highly non-symmetric)**
  - **Examples:**
  - **Foils of ^{90}Zr**
  - **Measure $^{89}Zr/^{90}Zr$ and $^{88}Zr/^{89}Zr$**

Most of the intermediate cross sections are not measured – RIA is NEEDED!

Toy Model -- limited reaction network
-- simple neutron spectrum

The Reaction Network



Impact of change in a Zr Cross Section*

	Reaction	50% change	
		^{89}Zr/^{90}Zr	^{88}Zr/^{89}Zr
S	^{90}Zr(n,2n)^{89g}Zr	36%	20%
S	^{90}Zr(n,2n)^{89m3}Zr	6.5%	10%
U	^{89}Zr(n,2n)^{88}Zr	11.5%	42%
U	^{89}Zr(n,γ)^{90m4}Zr	21%	5.5%
U	^{89}Zr(n,γ)^{90}Zr	16%	4.5%
U	^{88}Zr(n,γ)^{89m3}Zr	1%	15.5%
U	^{89m3}Zr(n,2n)^{88}Zr	0.5%	8%
U	^{90m4}Zr(n,2n)^{89}Zr	5%	2.5%

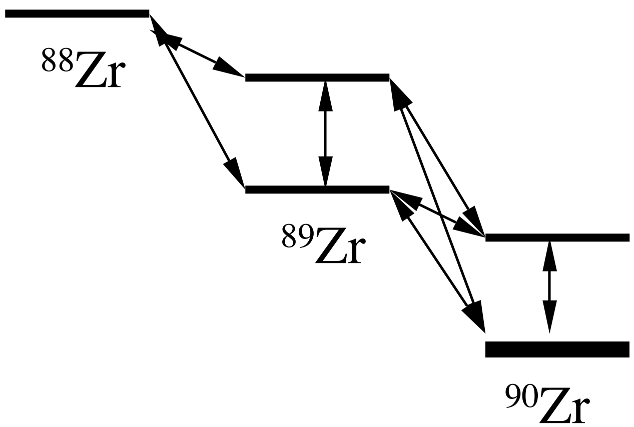
To use this technique we need to measure cross sections well at RIA

Cross sections are not well known

Reaction	Knowledge
$^{90}Zr(n,2n)^{89g}Zr$	Sum (g.s.+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
$^{89}Zr(n,\gamma)^{90m4}Zr$	Calculation only
$^{89}Zr(n,\gamma)^{90}Zr$	Calculation only
$^{88}Zr(n,\gamma)^{89m3}Zr$	Calculation only
$^{89m3}Zr(n,2n)^{88}Zr$	Calculation only
$^{90m4}Zr(n,2n)^{89}Zr$	Calculation only

Sensitivity Study for Simple Reaction Network

The Reaction Network



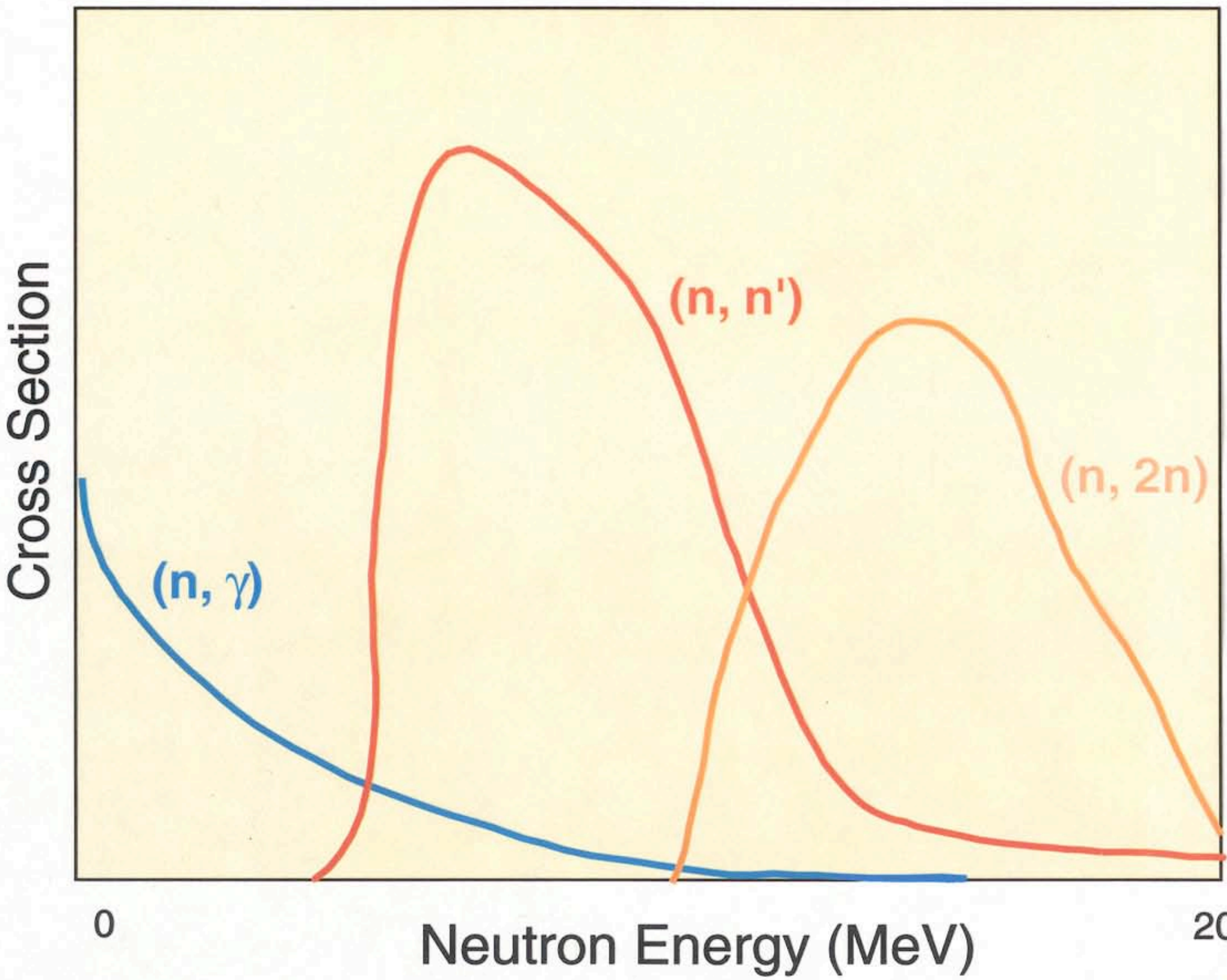
Reaction	**Change in Isotope Ratio**			
	For 10% change in cross section		**For 50% change in cross section**	
	$^{89}Zr/^{90}Z_L$	**$^{88}Zr/^{89}Zr$**	**$^{89}Zr/^{90}Zr_L$**	**$^{88}Zr/^{89}Zr$**
$^{90}Zr(n,2n)^{89g}Zr$	7.2%	4.0%	36%	20%
$^{90}Zr(n,2n)^{89m3}Zr$	1.3%	2.0%	6.5%	10%
$^{89}Zr(n,2n)^{88}Zr$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

Green: Cross section for sum (g.s + 0.94m) known to 2% for E_n = 13-15 MeV.
Red: Cross section determined by calculation only.

It is important to measure these cross sections accurately from threshold to 20 MeV.

ah.j.33a

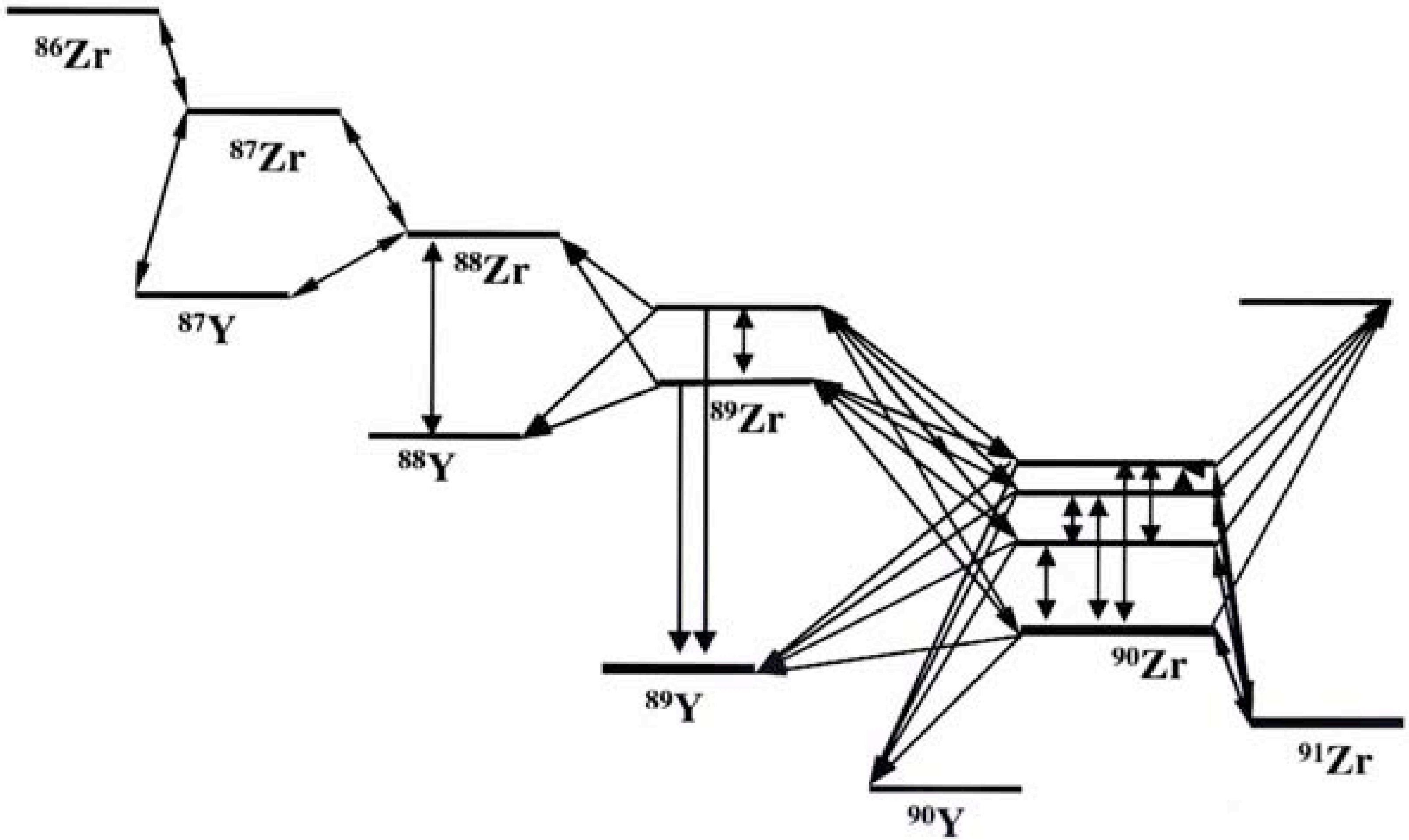
Three Nuclear Reactions to Monitor the Neutron Flux



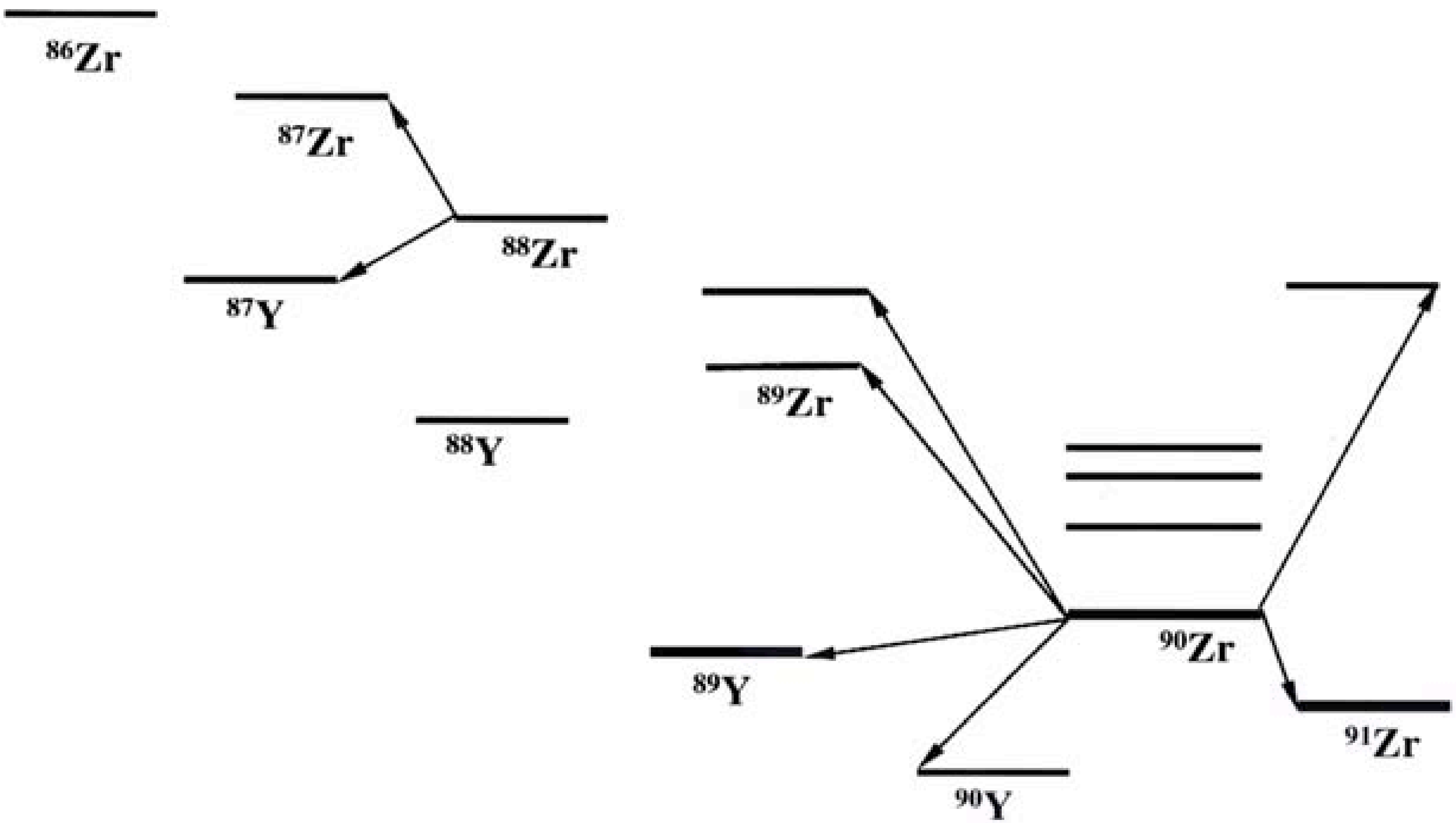
NNSA National Nuclear Security Administration

Los Alamos NATIONAL LABORATORY

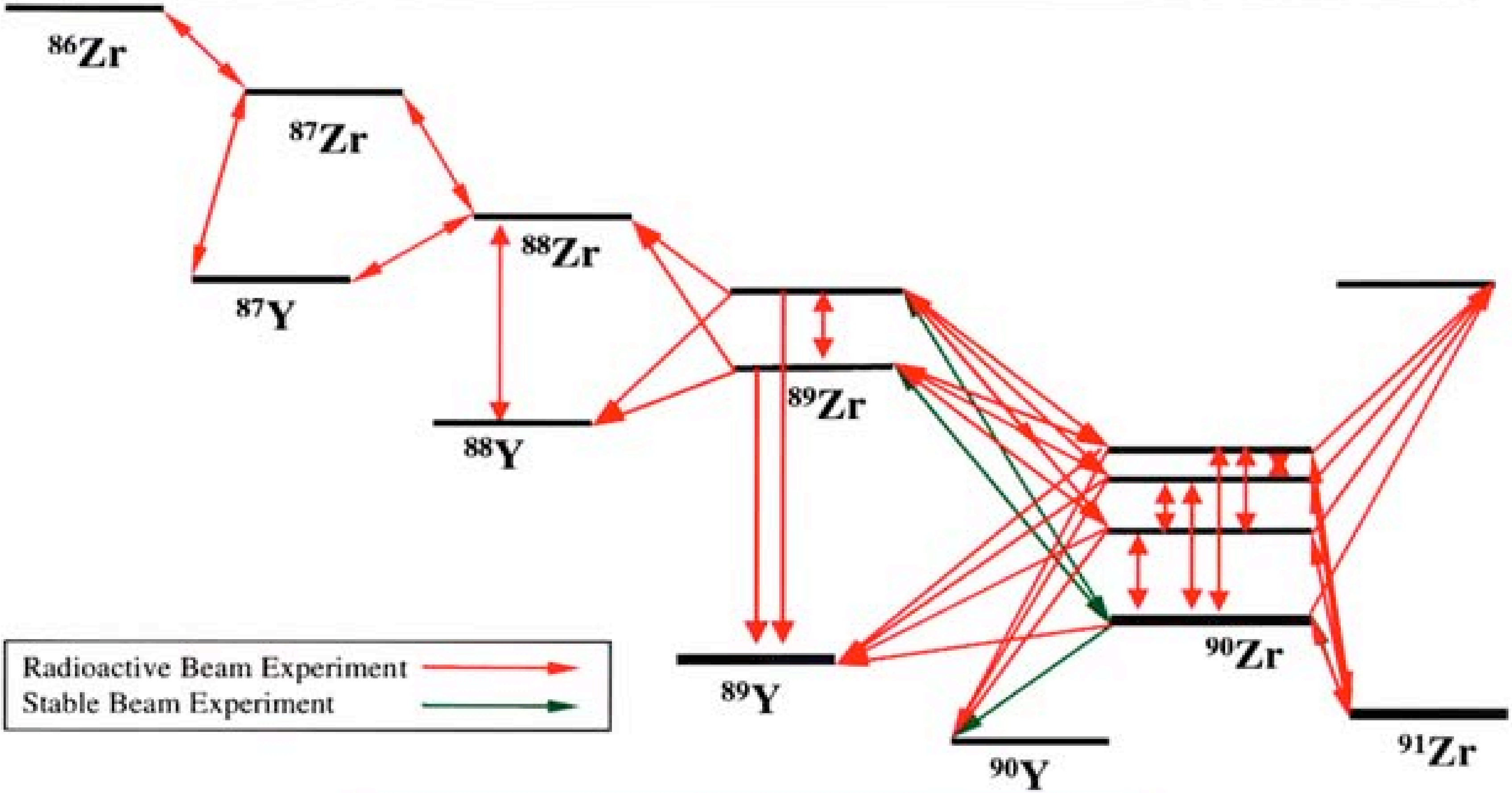
Moving closer to the real world, the need for good measurements is even more apparent



Only a few of these cross sections have been measured

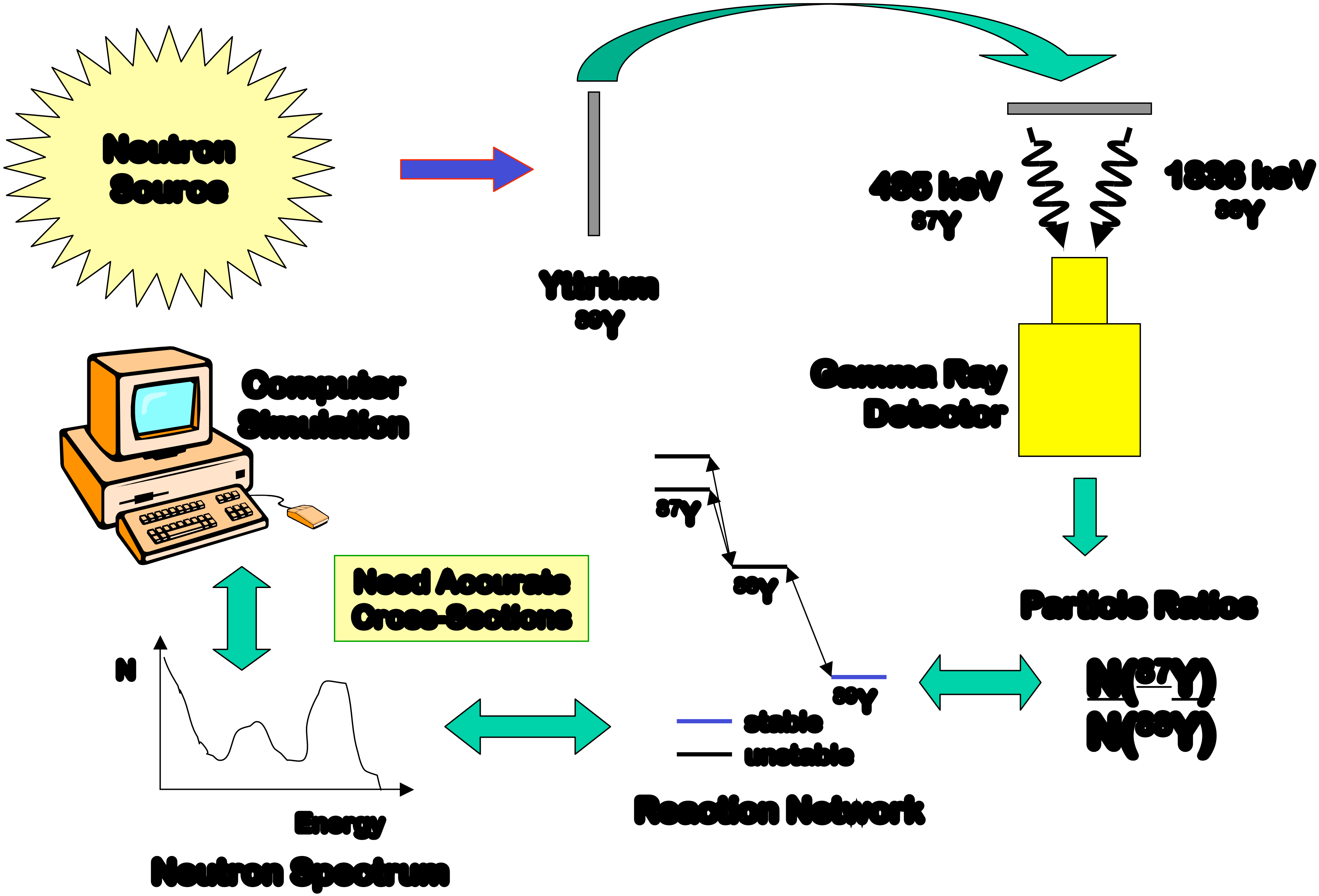


What needs to be measured

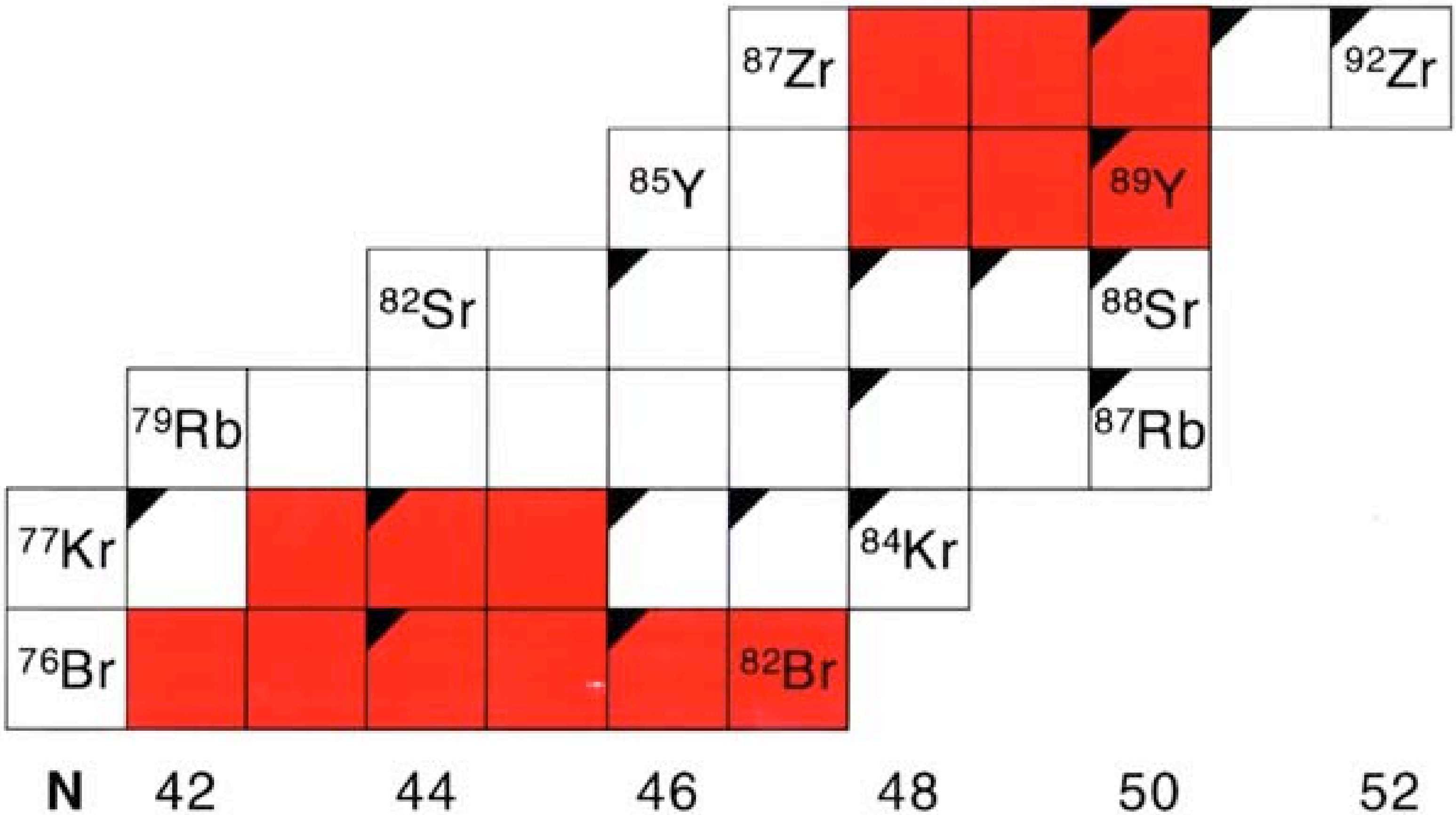


RIA is the place to do this physics

One Possible Measurement Scheme



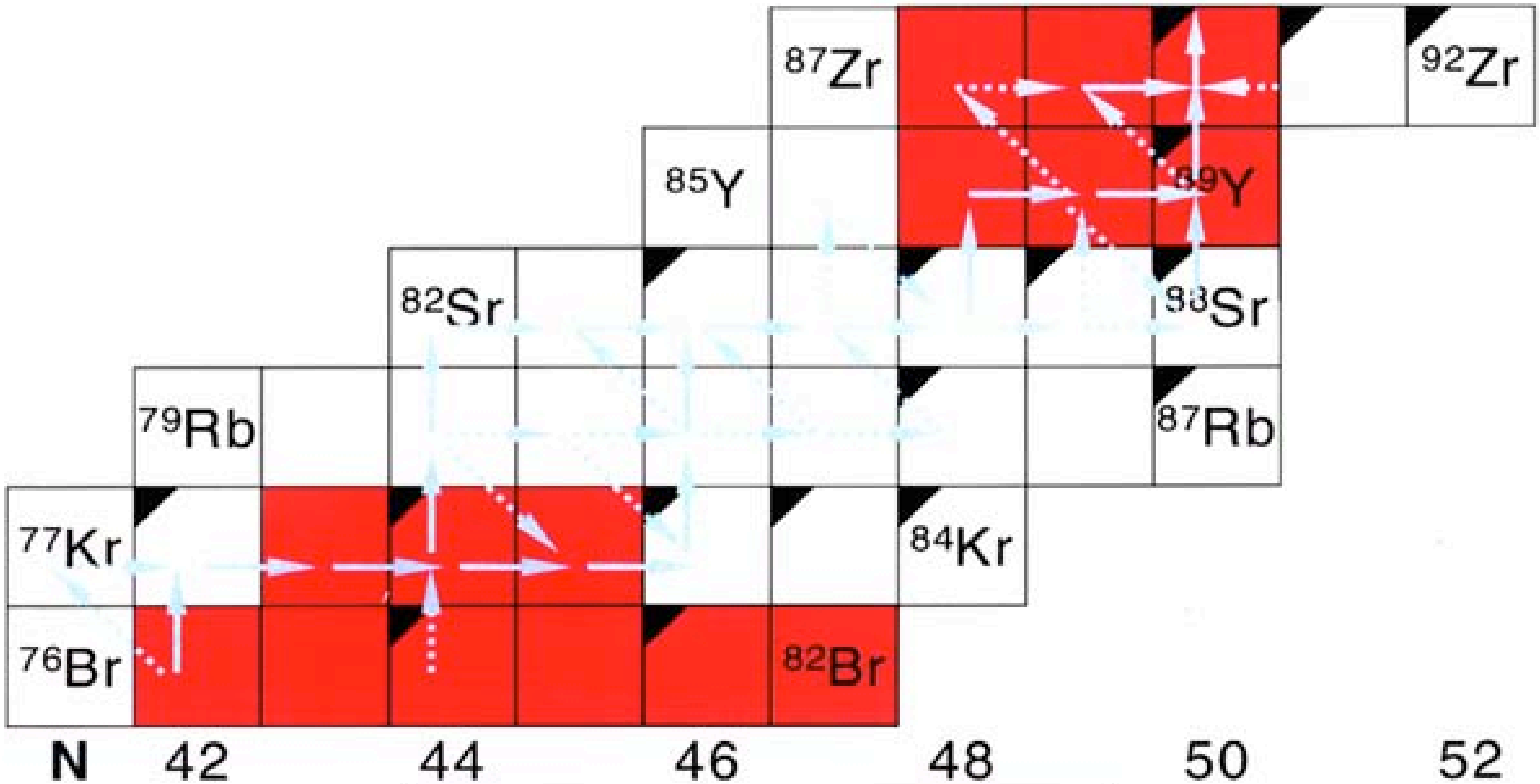
Important Radiochemical Detectors near Y and Zr*



*(D.R. Nethaway & M.G. Mustafa, UCRL-ID-133269)

P01300-lab-u-008 6

Nucleosynthesis of "light-p" nuclei*



Two motivations per experiment

*(R.D. Hoffman *et al.*,1996 ApJ 460: 478)

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NNSA and RIA

- NNSA will support groups working at RIA
  - Stewardship Science Academic Alliances program
  - NNSA Laboratories will fund groups doing relevant RIA experiments
- Expect NNSA scientists to help design the best experimental program
- Would like to help show a broad interest in RIA from other parts of DOE

Department of Energy
National Nuclear Security Administration
Washington, DC 20585

January 10, 2003

MEMORANDUM FOR: Dr. Ray Orbach
Director, Office of Science

FROM: Everet H. Beckner
Deputy Administrator for Defense Programs

SUBJECT: Rare Isotope Accelerator (RIA)

DISCUSSION: As discussed at our meeting on December 9, we believe that a future Rare Isotope Accelerator (RIA) will be important to science-based stockpile stewardship and therefore to the national security mission of the NNSA. There is significant interest at the NNSA laboratories in conducting experiments at an RIA to measure cross sections and reaction rates involving unstable, short-lived nuclei that would be extremely difficult to measure elsewhere. These data will provide the scientific underpinnings to reevaluate results from the radiochemical diagnostics used in the underground nuclear test program and to conduct precise determinations of neutron fluxes at new facilities such as the National Ignition Facility. Perhaps equally important, we also expect RIA to train many of the next generation of NNSA laboratory nuclear physicists.

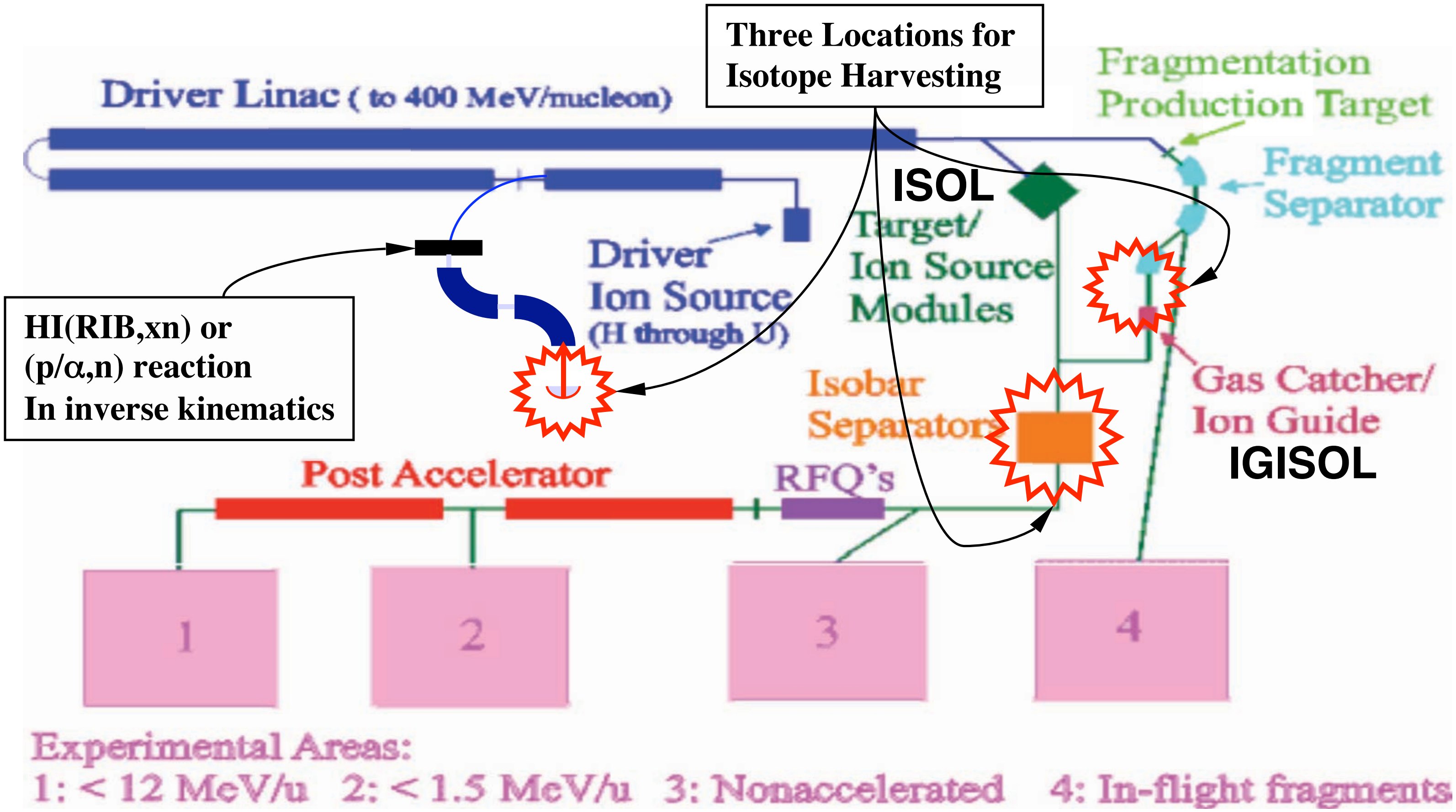
While the NNSA could not build such a facility to fulfill the needs we have for nuclear data, we will be users with interest in nuclear science as well as in specific data. Scientists at the NNSA laboratories are already collaborating with scientists in the broader nuclear physics community to help design the best accelerator complex and experimental facility. If and when the Office of Science decides to proceed on the construction of RIA, the staff at the NNSA laboratories in general and at Livermore in particular are ready to help.

Experimental Challenges

- Harvesting Isotopes of Interest
- Separation and Manufacturing of Targets
- Transportation to a Co-Located Neutron Source
- The Co-Located Neutron Source
- Detection technique backgrounds
  - For example, intense radiation environments
  - Purity of targets

Ingenious Solutions Proposed for All of These

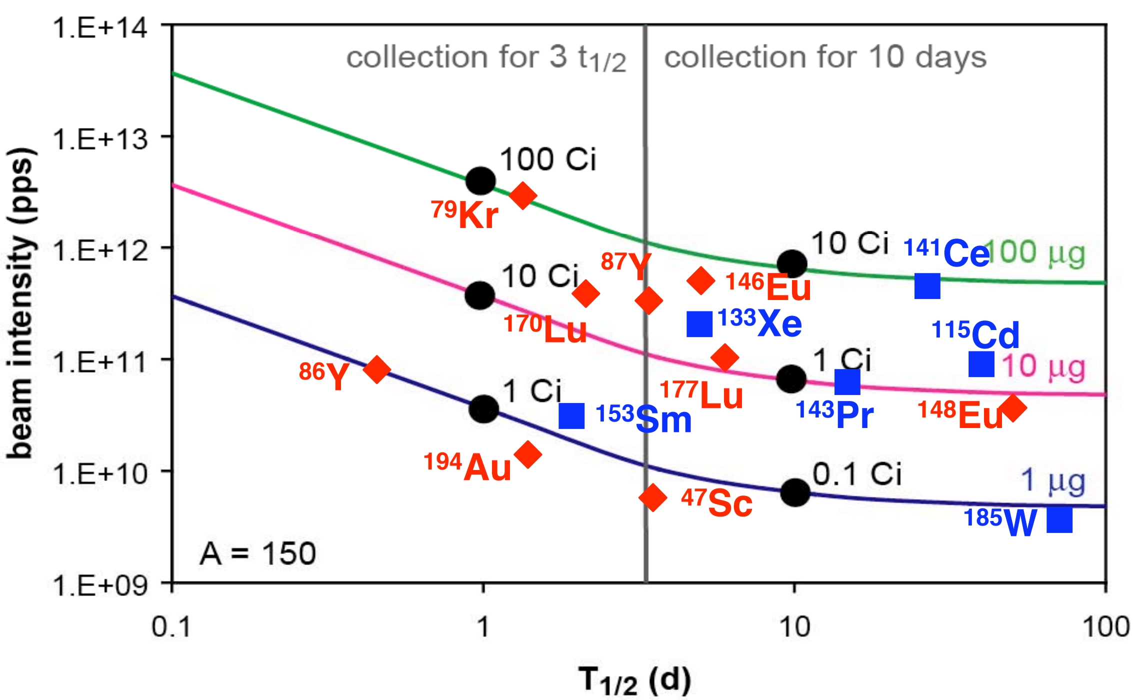
Harvesting Isotopes at RIA



1. **Production at first stripper – Direct Reactions**
2. **ISOL with Mass Separator**
3. **Fragmentation with IGISOL system**

How Much Can Be Harvested?

$$dN(t)/dt = P - \lambda N(t) \rightarrow N(t) = P/\lambda(1 - e^{-\lambda t})$$



Assume collection period of 3 half lives or 10 days, whichever is shorter.

Blue squares are several s-process branch points and red diamonds are several important isotopes for Stockpile Stewardship

For 1 day half-life isotope with production rate of 10^{11} pps, 1×10^{16} atoms (~ 2 Curies) can be collected in 3 days.

For 1 year half-life isotopes with production rate of 10^{11} pps, 8×10^{16} (~ 50 mCuries) can be collected in 10 days.

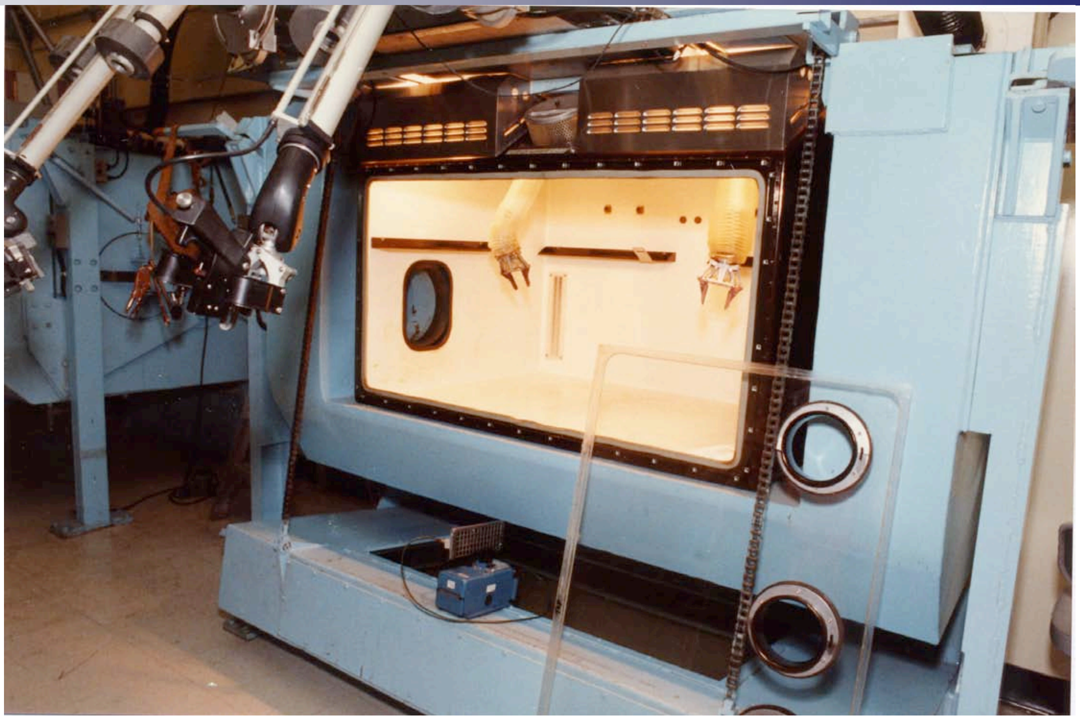
Radiochemistry Facility and Transportation

- **Production limits imply shortest half-life of produced species ~ 1 hour**
- **Will probably wait on order of one hour before trying to handle production products.**
- **10 μg of 1 hour half-life isotope implies ~150 C one hour after production run.**
- **Depending on efficiency of separation and decay products, may be significant contribution from other isotopes.**

- **Need radiochemistry lab to process material in targets**
  - **Handle up to 1 kC of activity**
  - **Z separation of production products**
- **Need to transport from production area to lab to neutron facility**
- **Also may need to do chemistry on target after neutron irradiation**

Radiochemistry

- Harvested isotope will be 10 Curies of activity.
- Other radioactive isotopes will be present.
- Gamma and beta rays will be dominant form of radiation.



Hot cell capable of handling 1 kCi of gamma ray activity.

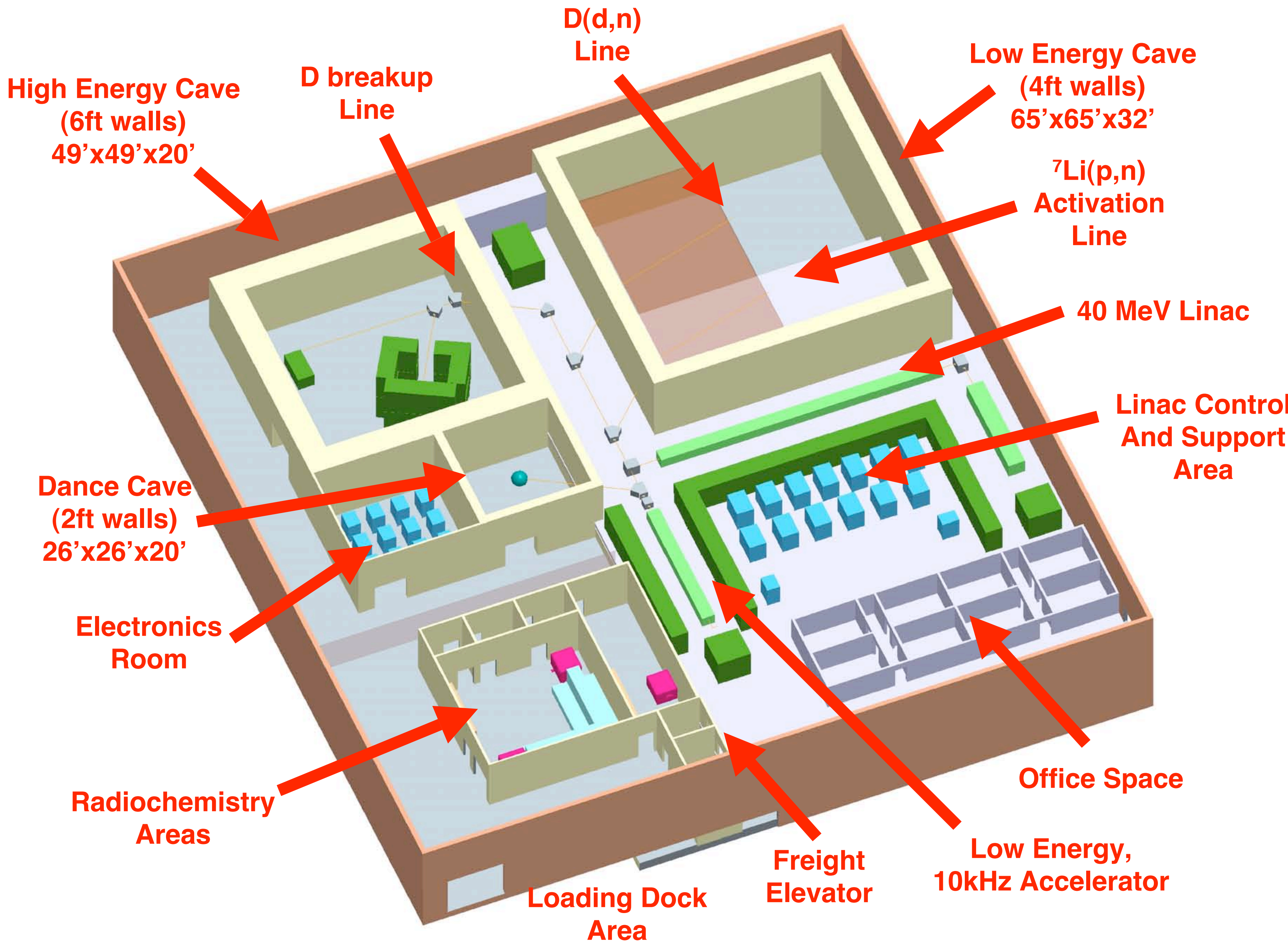
Hot cell capable of handling 100 Curies of activity required

Neutron Source Facility at RIA

1. **Co-located but separate facility.**
2. **Mono-energetic neutrons from ~10 keV to 20 MeV.**
3. **High neutron fluxes, up to 10^{11} neutrons/sec on target.**
4. **Radiochemistry facility for processing targets.**

Different production mechanism are appropriate for different energies.

- **$^{3}H(^{2}H,n)^{4}He$: 14+ MeV**
- **Deuteron Breakup: 7+ MeV**
- **$^{2}H(^{2}H,n)^{3}He$: 2-9 MeV**
- **$^{3}H(p,n)^{3}He$ or $^{7}Li(p,n)^{7}Be$: 0.1-2 MeV**
- **Moderated reactions: Below 100 keV**

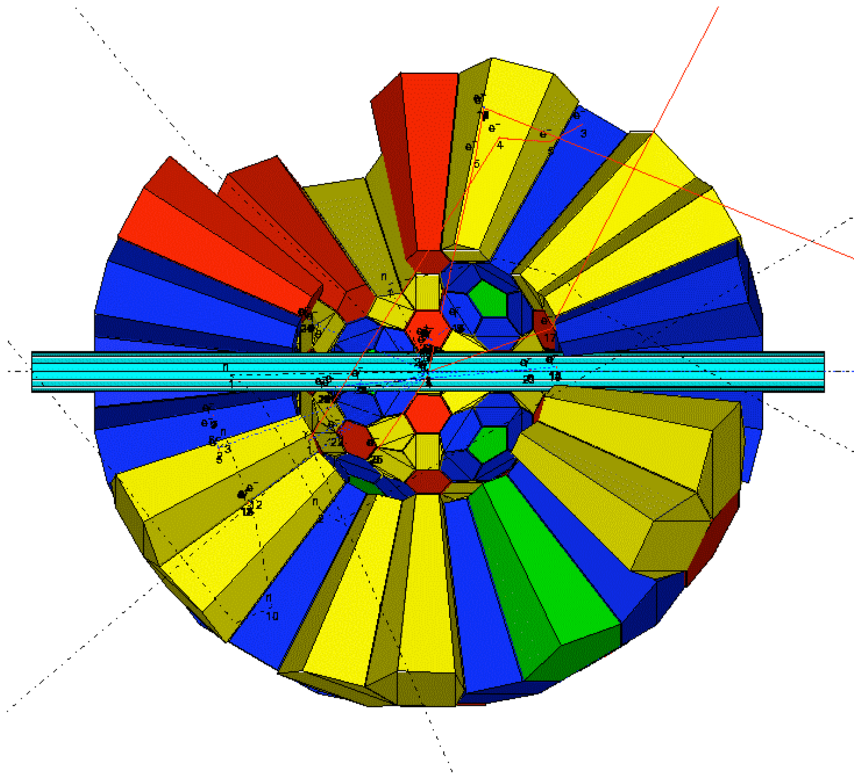


High Energy Cave
(6ft walls)
49'x49'x20'
D breakup
Line
D(d,n)
Line
Low Energy Cave
(4ft walls)
65'x65'x32'
^{7}Li(p,n)
Activation
Line
40 MeV Linac
Linac Control
And Support
Area
Dance Cave
(2ft walls)
26'x26'x20'
Electronics
Room
Radiochemistry
Areas
Loading Dock
Area
Freight
Elevator
Low Energy,
10kHz Accelerator
Office Space

Detector Techniques

- Depending on details of a particular isotope, various excellent ideas have been discussed. Measurements could be made directly or indirectly.
- *DANCE*
- *GEANIE*
- *GRETINA*
- *Surrogate Approach*
- *Some new brilliant idea*

The DANCE barium fluoride array



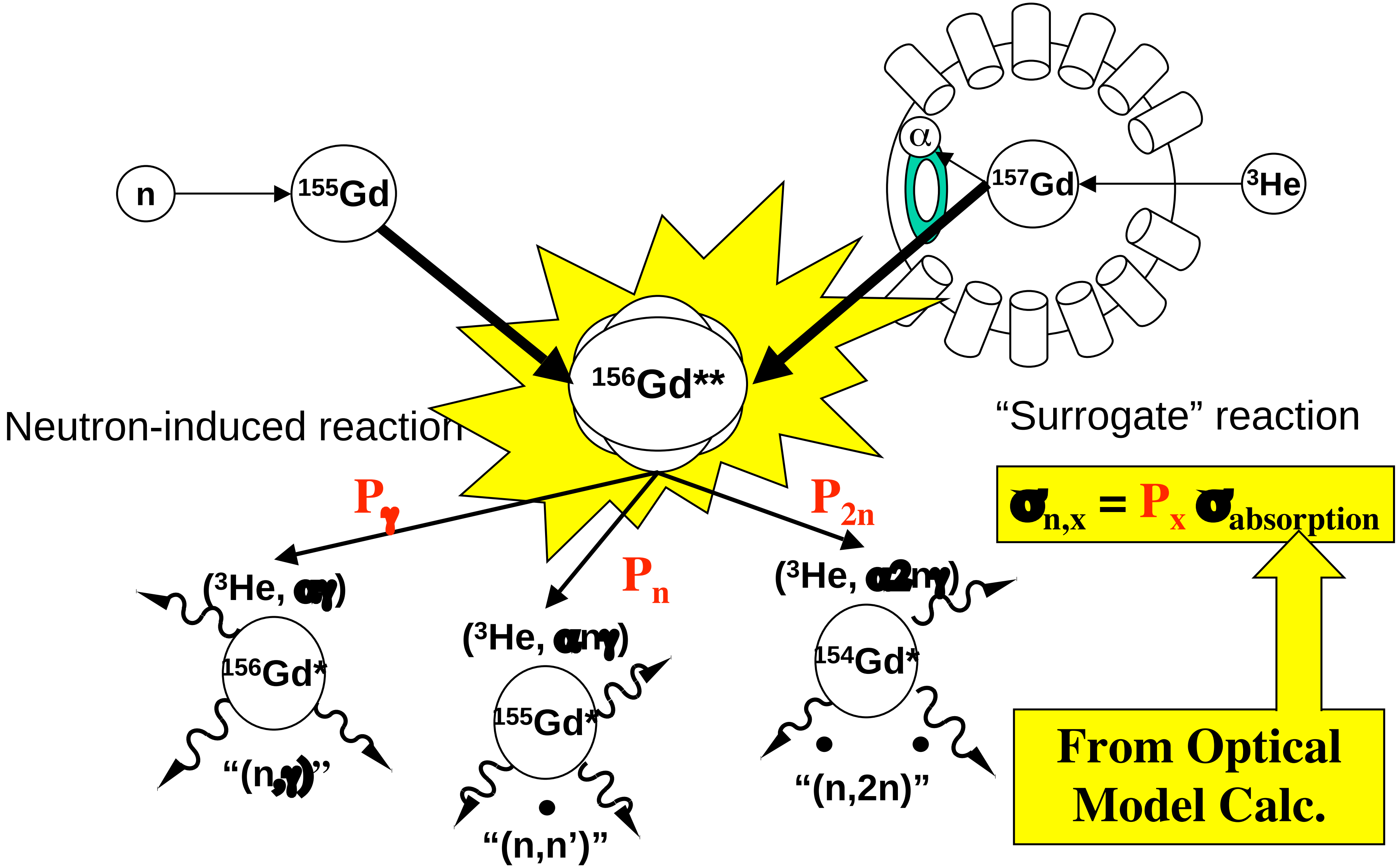
- 162 segments with 4 different shape crystals (159 segments with crystals)
- High efficiency will allow measurements on milligram samples
- Highly segmented to allow detection of radioactive targets
- Hit pattern analysis and reaction calorimetry to minimize backgrounds
- Inner radius = 17 cm
- Crystal depth = 15 cm
- Extensive Monte Carlo simulations to design detector
- All crystals will be delivered in FY2002
- State-of-the-art fast digitizers for data acquisition
- Array will be completed in 2002, but some data may be obtained with partial array.
- M. Heil, et al., Nucl. Instr. Meth. A459, 229-246 (2001)

Monte Carlo (GEANT) Simulations
- M. Heil
- R. Reifarth
- F. Kaeppeler

Forschungzentrum Karlsruhe

LANSCE
Los Alamos Neutron Science Center

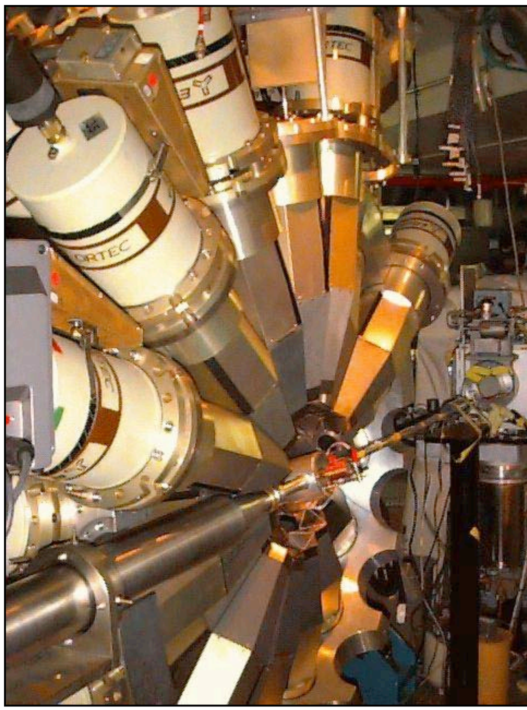
Surrogate neutron-induced reactions using charged particle beams

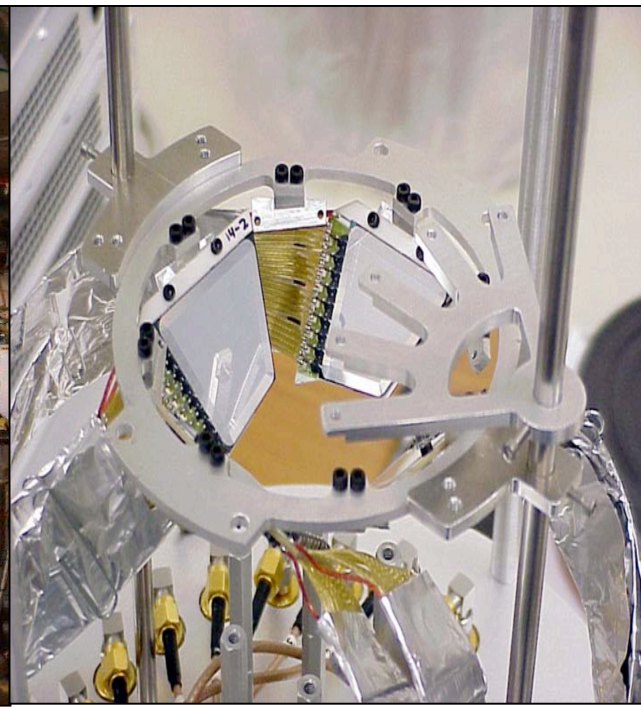


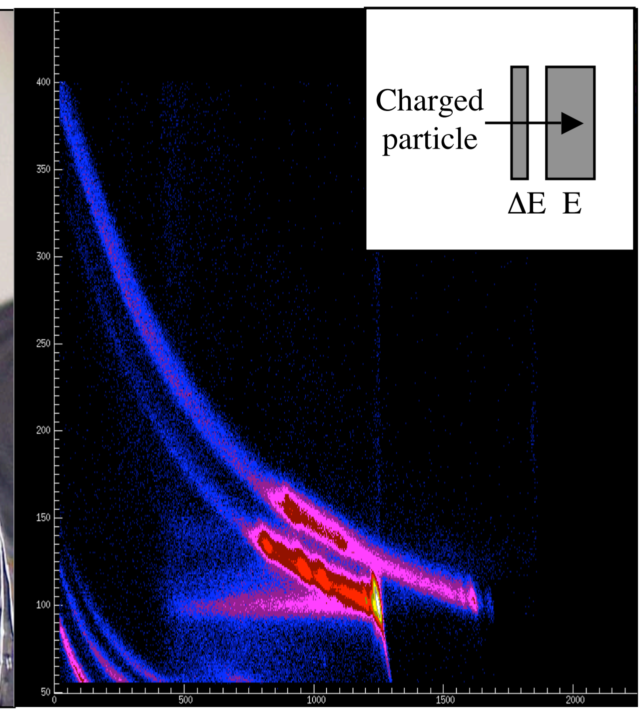
Silicon Telescope Array for Reaction Studies coupled to GAMMASPHERE

- ^{157}Gd(^{3}He,α /^{3}He)$^{156.157}$Gd at E(^{3}He) = 45 MeV
- 3 day run, Average Current = 0.2-0.3 pnA

GAMMASPHERE	STARS	ΔE-E Particle ID







First experiment completed: 4/02

Other Applications

- ***Test Readiness Program***
  - Congress has mandated that the nation must be prepared to resume underground testing should the President so order.
  - The challenges at a Rare Isotope Facility are identical (separation techniques, hot radiochemistry etc.) to those that would be encountered in analyzing tests.
  - The scientific staff needed for this program most likely would come from RIA.

Homeland Security

The Problem: A Nuclear Bomb has been detonated. The President needs to know

What kind of nuclear device was it?

Where did it come from?

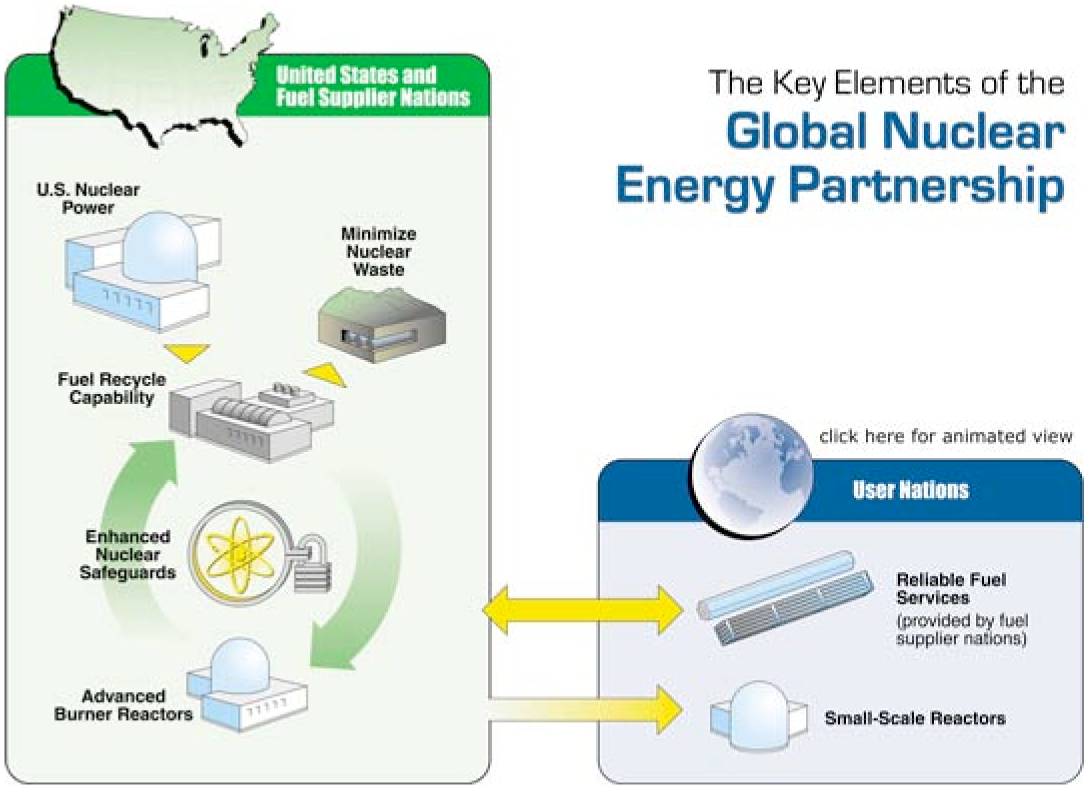
95	235Am 10.3 M ε	236Am 3.6 M α	237Am 73.0 M ε	238Am 98 M ε	239Am 11.9 H ε	240Am 50.8 H ε	241Am 432.2 Y α	242Am 16.02 H β-	243Am 7370 Y α
	234Pu 8.8 H ε	235Pu 25.3 M ε	236Pu 2.858 Y α	237Pu 45.2 D ε	238Pu 87.7 Y α	239Pu 24110 Y α	240Pu 6561 Y α	241Pu 14.290 Y β-	242Pu 3.75E+5 Y α
93	233Np 36.2 M ε	234Np 4.4 D ε	235Np 396.1 D ε	236Np 154E+3 Y ε	237Np 2.144E+6 Y α	238Np 2.117 D β-	239Np 2.356 D β-	240Np 61.9 M β-	241Np 13.9 M β-
	232U 68.9 Y α	233U 1.592E+5 Y α	234U 2.455E+5 Y 0.0054% α	235U 7.04E+8 Y 0.7204% α	236U 2.342E7 Y α	237U 6.75 D β-	238U 4.468E9 Y 99.2742% α	239U 23.45 M β-	240U 14.1 H β-
91	231Pa 3.276E+4 Y α	232Pa 1.31 D β-	233Pa 26.975 D β-	234Pa 6.70 H β-	235Pa 24.44 M β-	236Pa 9.1 M β-	237Pa 8.7 M β-	238Pa 2.27 M β-	239Pa 1.8 H β-
	140		142		144		146		148

Reducing the uncertainty in answering these questions is essential. Improved accuracy of many actinide cross sections is needed.

Measurements at a Rare Isotope Facility will be very important.

Nuclear Energy

The Global Nuclear Energy Partnership (GNEP) is part of the AFCI (Advanced Fuel Cycle Initiative) studying technology options for burner reactors to transmute long-lived radioisotopes into shorter lived ones.



Improved nuclear cross sections are essential.

GNEP Needs at RIA

1. Improved Nuclear Data

--to design better reactors

2. Personnel

-- If GNEP is successful, there are not enough trained radiophysicists or radiochemists to handle the challenges such as the degradation of reactor materials.

Nuclear Reactor Community estimate of uncertainties needed and estimate of present uncertainties on cross sections.

SFR

Isotope	Cross Section	Energy Range	Uncertainty %	
			Initial	Required
Pu239	σ_{capt}	498 KeV-183 KeV	15	9.4
		183 KeV-67.4 KeV	15	8.1
		67.4 KeV-24.8 KeV	10	9
		24.8 KeV-9.12 KeV	10	7.7
	σ_{fiss}	6.07 MeV-2.23 MeV	5	3.9
		2.23 MeV-1.35 MeV	5	3.6
		1.35 MeV-498 KeV	5	2.1
		498 KeV-183 KeV	5	1.8
		183 KeV-67.4 KeV	5	2
		67.4 KeV-24.8 KeV	5	2.8
		24.8 KeV-9.12 KeV	5	3.1

SFR

Isotope	Cross Section	Energy Range	Uncertainty %	
			Initial	Required
Pu241	σ_{fiss}	6.07 MeV-2.23 MeV	20	8.8
		2.23 MeV-1.35 MeV	10	7.8
		1.35 MeV-498 KeV	10	4.6
		498 KeV-183 KeV	10	3.6
		183 KeV-67.4 KeV	10	3.5
		67.4 KeV-24.8 KeV	10	4.5
		24.8 KeV-9.12 KeV	10	4.7
		9.12 KeV-2.03 KeV	10	7.3
		2.03 KeV-454 eV	10	6

Tables from M. Salvatores, Nuclear Physics and Related Computational Science R&D for Advanced Fuel Cycle Workshop, August 10-12, 2006 Bethesda, Maryland

RIA and Medical Applications

- RIA will be a great place to make *research* quantities of tailor-made medical isotopes
- Could explore technologies to make specific isotopes
- Systemic Therapy R&D is an interesting possibility
  - The goal: localize sources in tumor cells with radiation penetrating only cellular dimensions
- ***Competition for beam time etc. makes RIA an unlikely factory for material for therapy.***

Candidate radionuclides for radioimmunotherapy

^{47}Sc	^{64}Cu	^{67}Cu
^{90}Y	^{105}Rh	^{103}Pd
^{111}Ag	^{124}I	^{142}Pr
^{149}Pm	^{153}Sm	^{159}Gd
^{166}Ho	^{177}Lu	$^{186/188}Re$
^{194}Ir	$^{193m,195m}Pt$	^{211}At

Conclusions

- There are compelling reasons why the U.S. should invest in a new facility to produce Rare Isotopes ***NOW***
- World-class science
- Important applications
  - National Security
    - Stockpile Stewardship
    - Homeland Security
  - Nuclear Energy
  - Medical Research

Let's Get It Started!!!