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Title: Introduction to nuclear reactions and the new Device for Indirect Capture Experiments on Radionuclides

Author(s): Stamatopoulos, Athanasios; Koehler, Paul E.; Bond, Evelyn M.; Bredeweg, Todd Allen; Couture, Aaron Joseph; DiGiovine, Brad Joseph; Essenmacher, Scott David; Fassbender, Michael Ernst; Hayes-Sterbenz, Anna Catherine; Keksis, August Lawrence; Matyskin, Artem; Mocko, Veronika; Parsons, Donald Kent; Rusev, Gencho Y.; Ullmann, John Leonard; Vermeulen, Christiaan

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Introduction to nuclear reactions and the new Device for Indirect Capture Experiments on Radionuclides*

A. Stamatopoulos[1], P. Koehler[1], E. Bond[2], T. A. Bredeweg[2], A. Couture[1], B. DiGiovine[1], S. Essenmacher[3,4], M. E. Fassbender[2], A. C. Hayes-Sterbenz[5], G. Keksis[2], A. Matyskin[2], V. Mocko[2], K. Parsons[6], G. Rusev[2], J. Ullmann[1], E. Vermeulen[2]

1. Physics Division, Los Alamos National Laboratory, 87545, NM, USA
2. Chemistry Division, Los Alamos National Laboratory, 87545, NM, USA
3. Department of Chemistry, Michigan State University, East Lansing, MI 48824
4. Facility for Rare Isotope Beams, Michigan State University, East Lansing, MI 48824
5. Theoretical Division, Los Alamos National Laboratory, 87545, NM, USA
6. X-Computational Physics Division, Los Alamos National Laboratory, 87545, NM, USA

LANSCE user group meeting, June 2nd – 3rd , Los Alamos, NM, USA

*Funded by LANL/LDRD-DR

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Table of Contents

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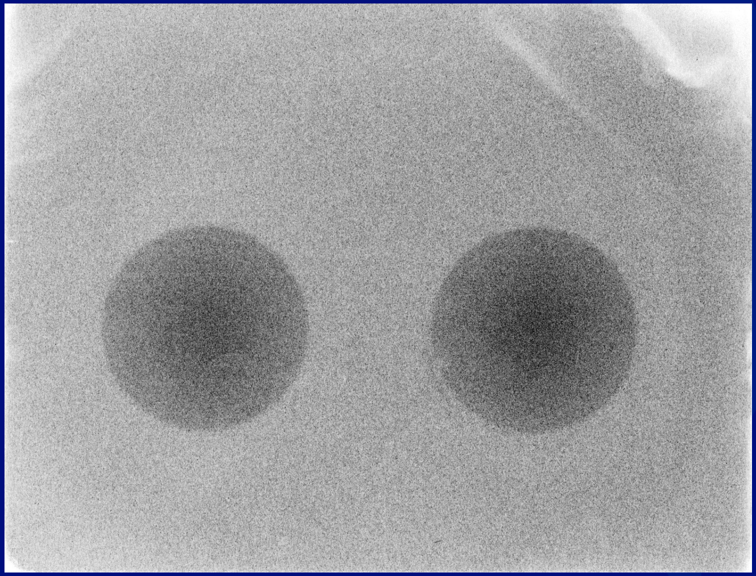
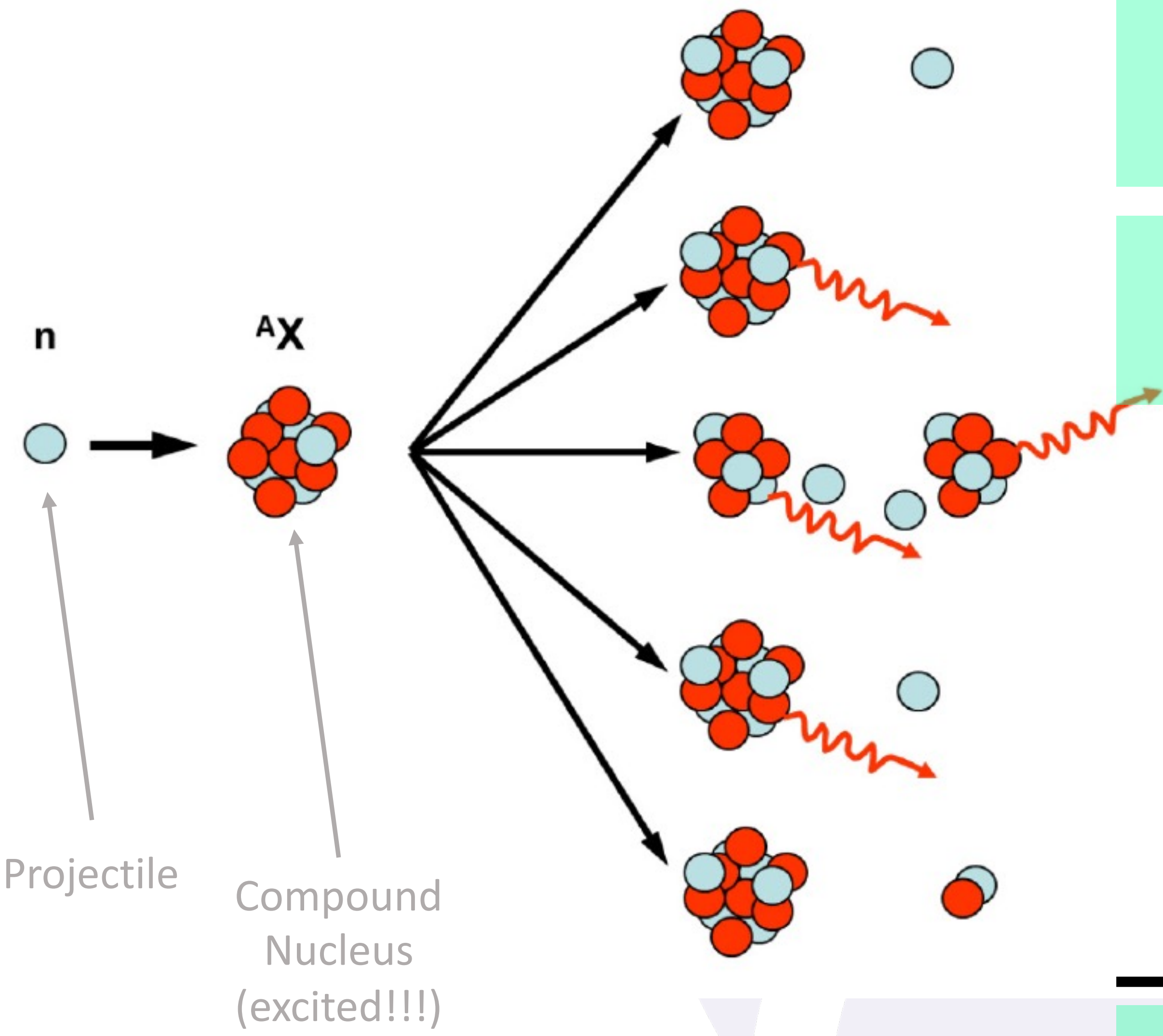
Neutron-induced reaction studies



Neutron-induced reactions

De-excitation channels



elastic scattering
(n,n)

radiative capture
(n,γ)

fission
(n,f)

inelastic scattering
(n,n'γ)

other reactions
(n,p), (n,d), (n,α)

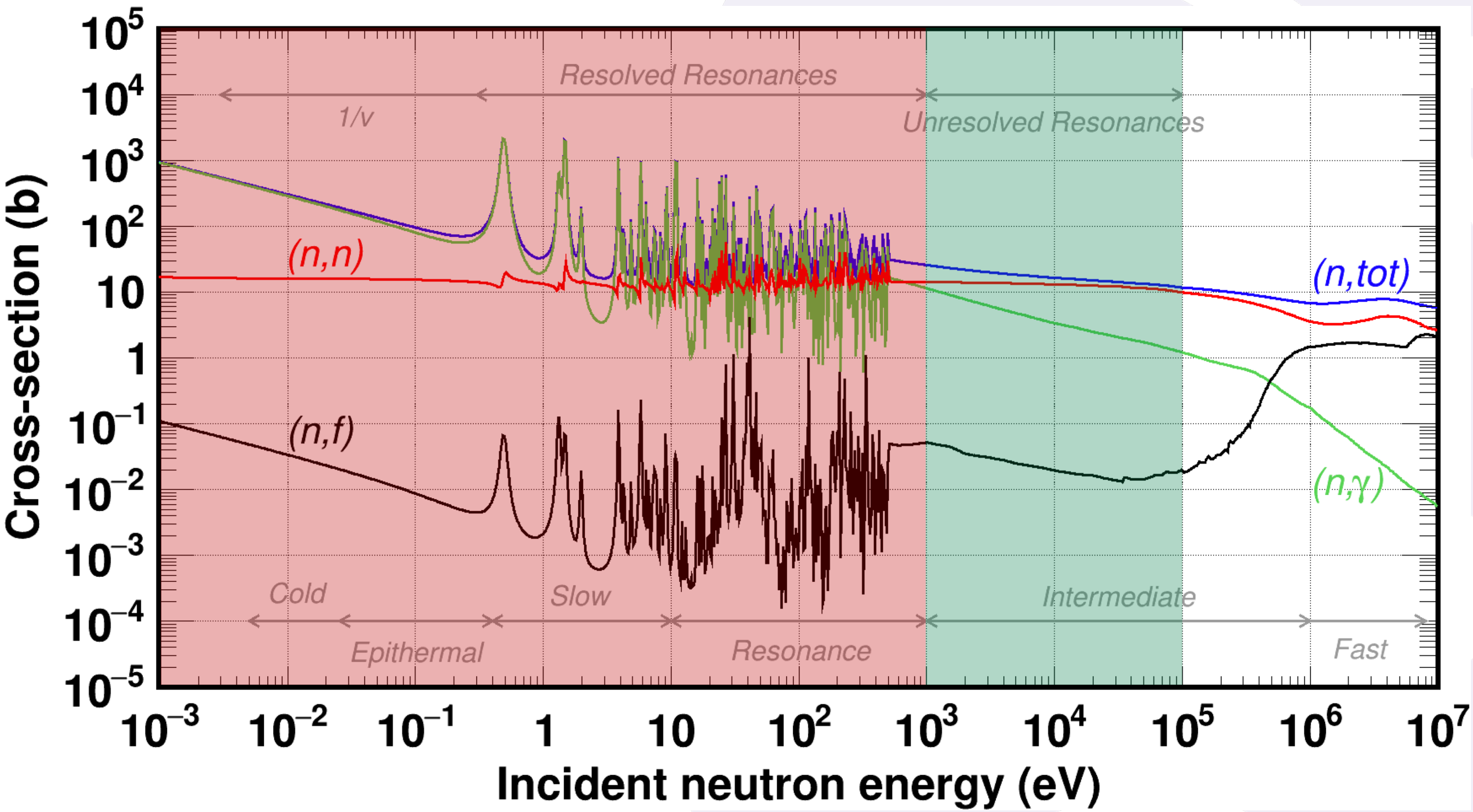
+

Total
(n,tot)

Neutron-induced cross sections

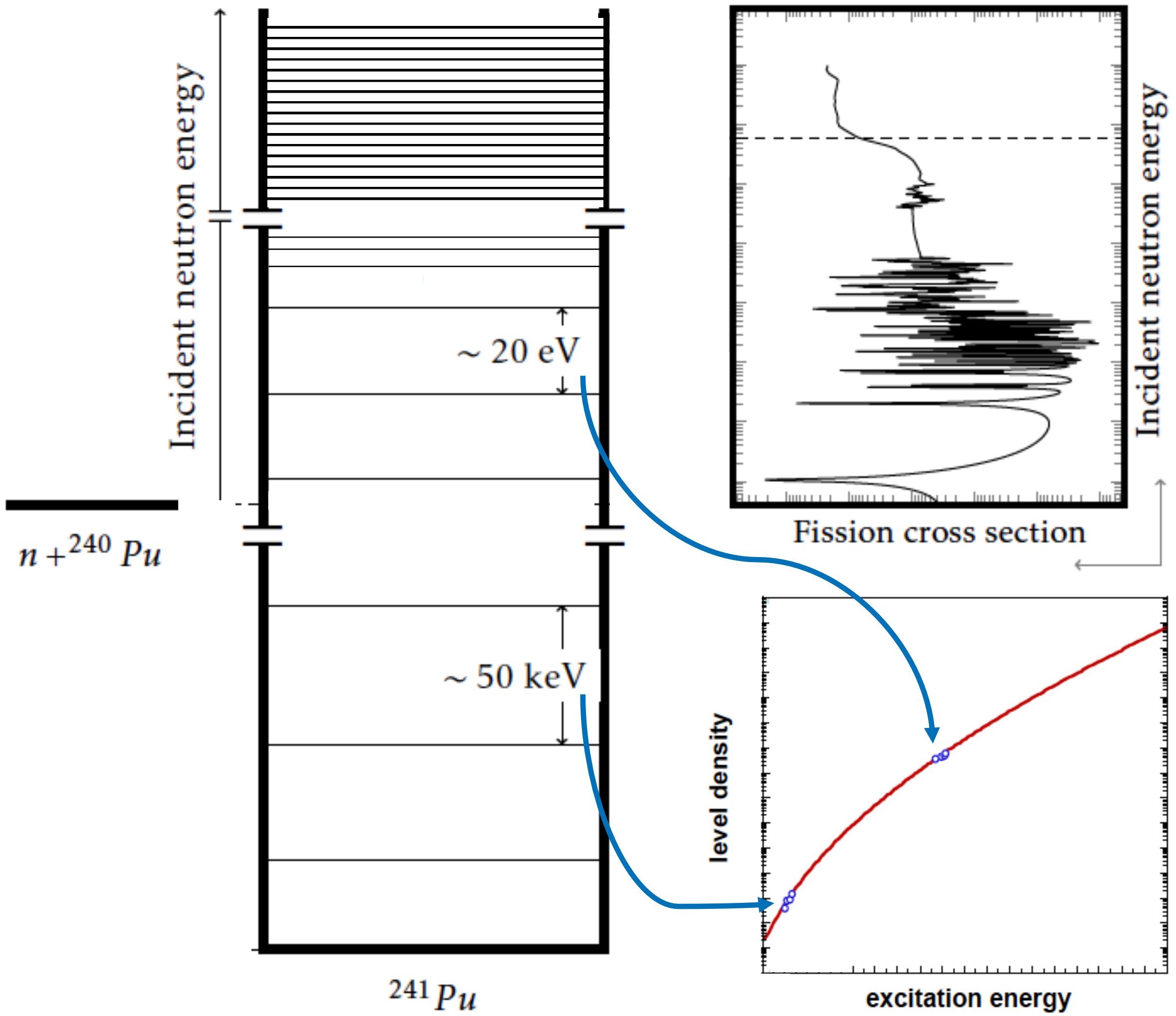
"Size" of a nucleus for a given projectile

- **Cross section (σ_i)** : The probability/area for the reaction *i* to occur (1 barn = 10^{-24} cm^2)



Neutron resonances and nuclear levels

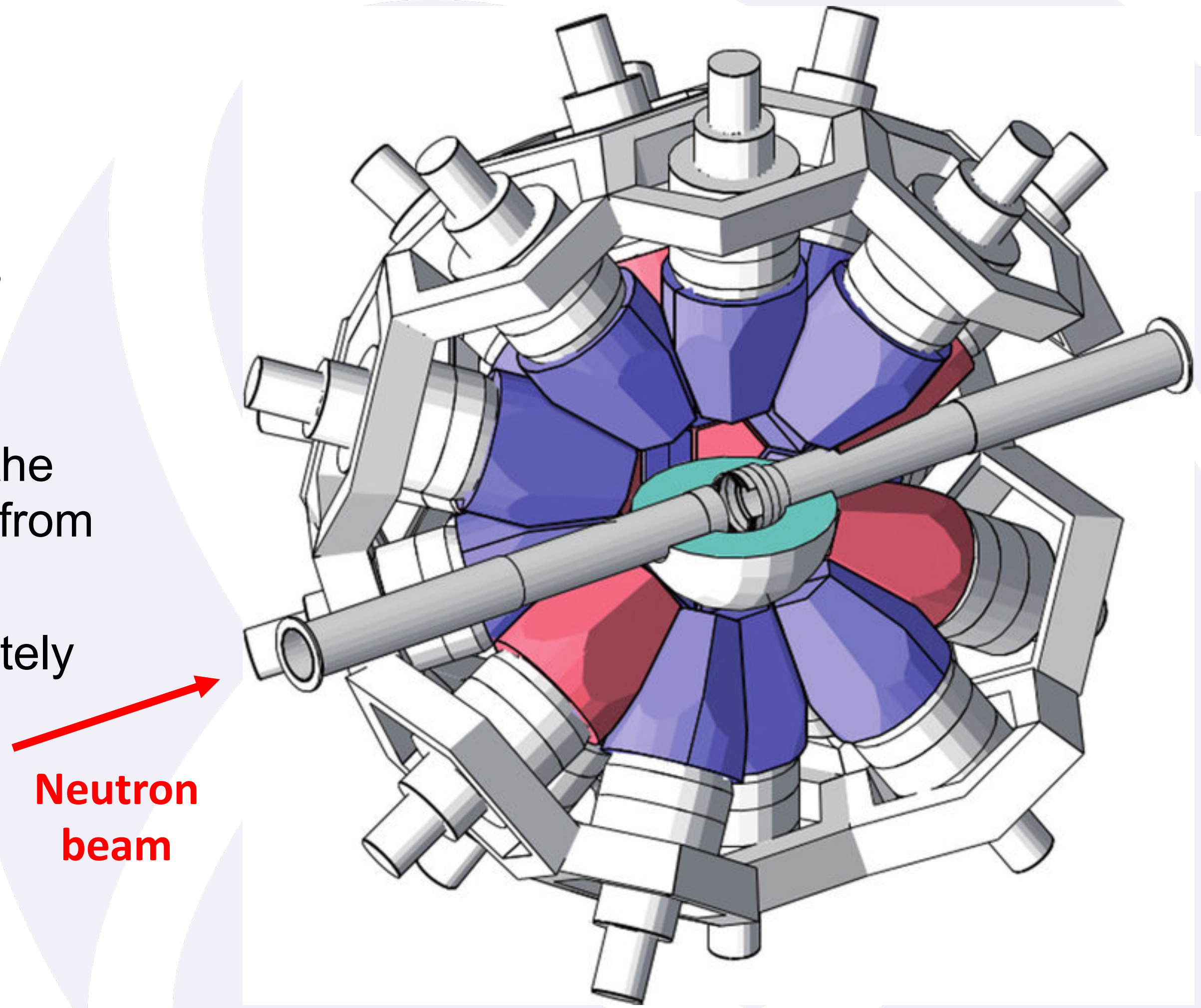
- Nuclear levels show up as spikes (resonances) in the cross section
- The higher the excitation, the closer the levels
- At high excitation energies the levels are so close, that it's hard to resolve them.
- At high excitation energies the (n,γ) cross section drops.



Neutron capture measurements

- **(n,γ) cross sections** are important for:
  - Nuclear physics and astrophysics
  - Nuclear criticality safety, advanced reactors, radiochemical diagnostics

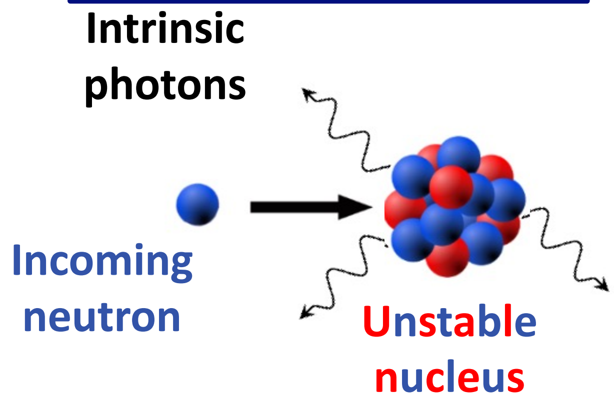
- Neutrons impinge on a sample
- Gamma detectors are close to the sample and catch the gammas from the de-excitation.
- Works fine for stable or moderately radioactive samples.



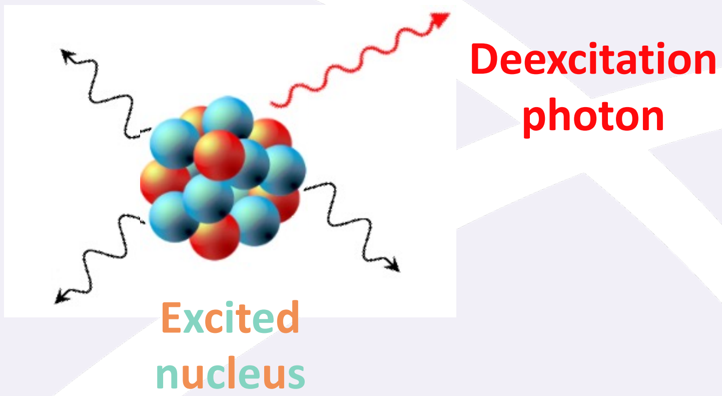
Neutron capture measurements on radionuclides

- Challenging **direct measurements** (especially E_n~keV): **large backgrounds** involved

Before neutron capture



After neutron capture



- **Indirect** measurements and techniques

- A **new technique** is proposed:
  - Neutron **transmission** experiments to measure the **(n,tot)** cross section
  - **R-Matrix fits** of the resonances seen in the transmission spectrum
  - Fit results to **calibrate** the **Nuclear Statistical Model** and tightly constrain (n,γ)

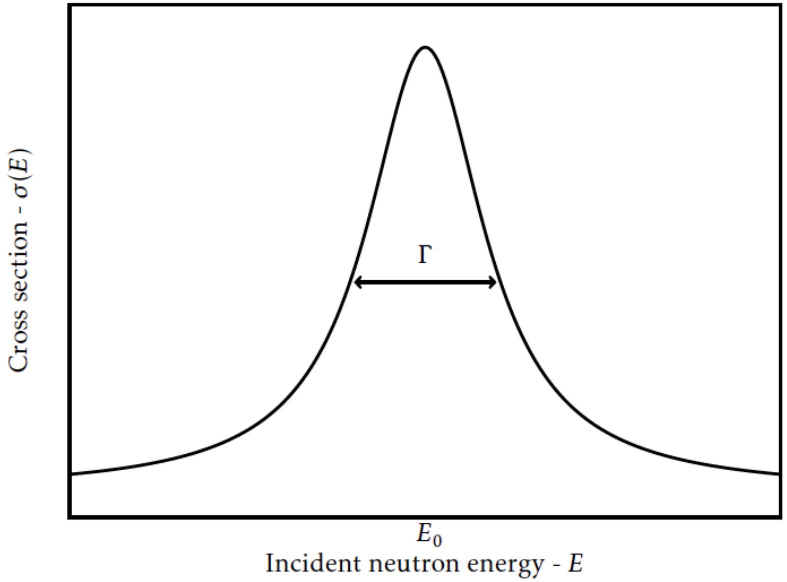
The technique in a nutshell

- Cases where the most probable reactions are capture and scattering

$$\sigma_{tot} \approx \sigma_\gamma + \sigma_n$$

Total — Capture — Scattering

- Each resonance can be analytically described by its **Energy** and **Width**



- Similar to the cross sections, resonance widths are

$$\Gamma_{tot} = \Gamma_\gamma + \Gamma_n$$

Total — Capture — Scattering

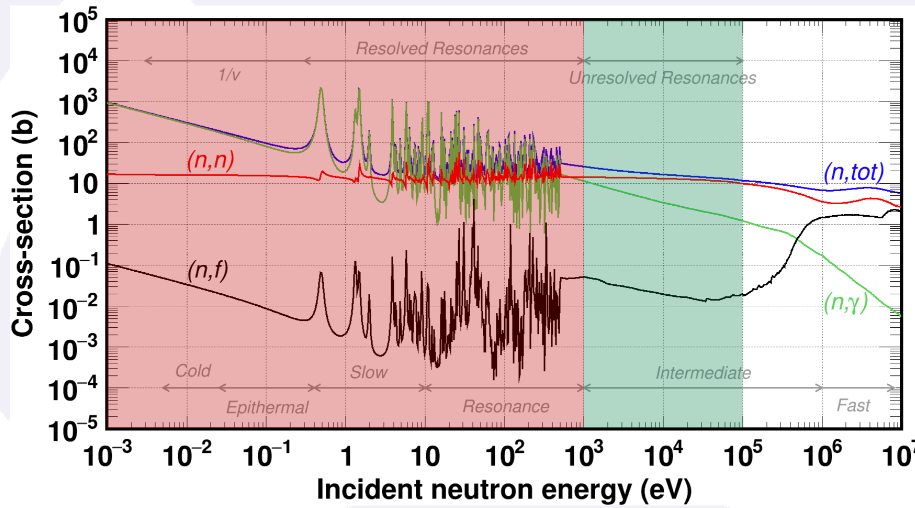
- At resonance energies:

$$\Gamma_\gamma = \Gamma_{tot} - \Gamma_n$$

- At intermediate energies (keV):

Level spacing

$$\sigma_\gamma = f(\Gamma_{tot}, \Gamma_n, D_0)$$



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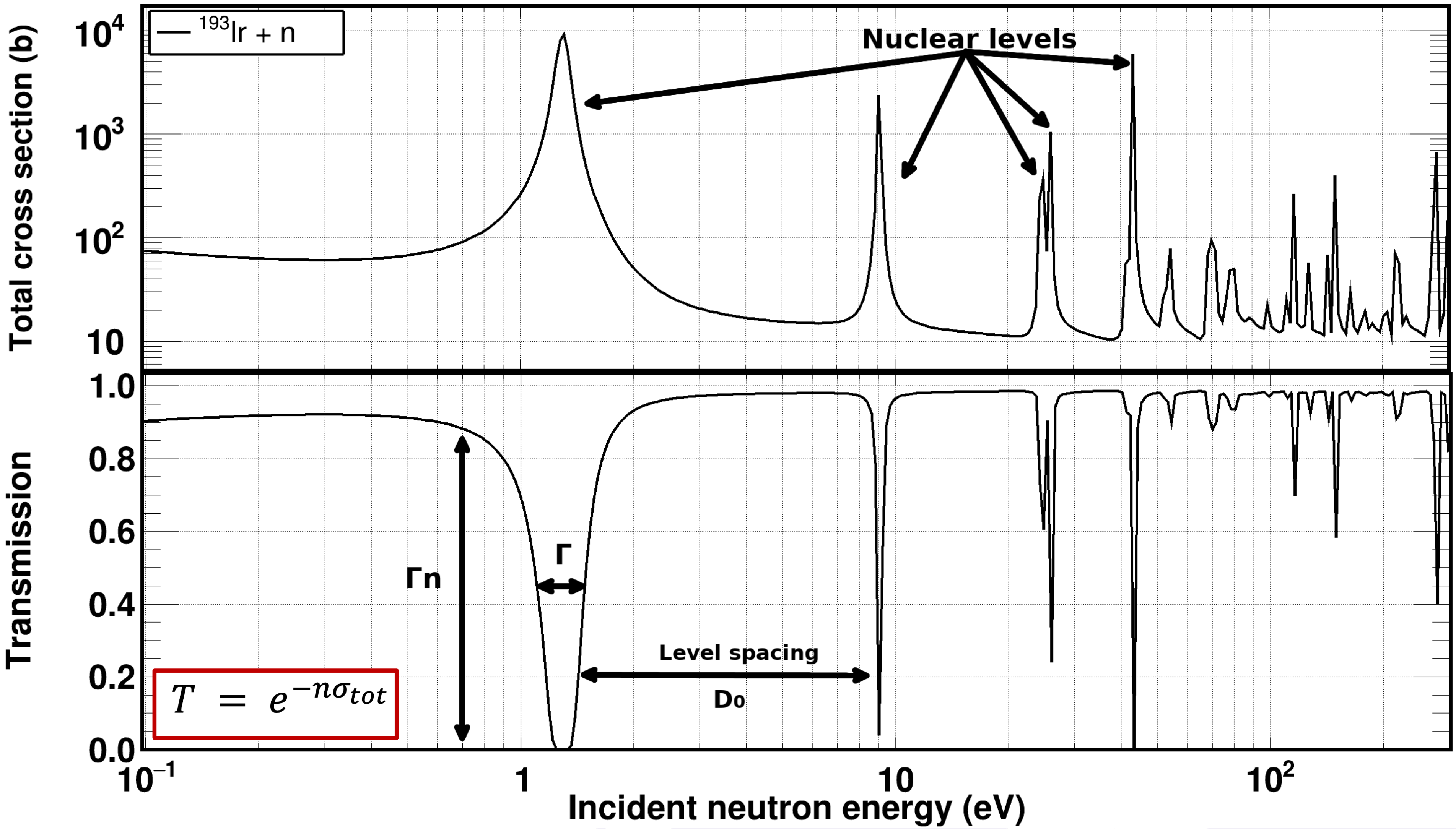
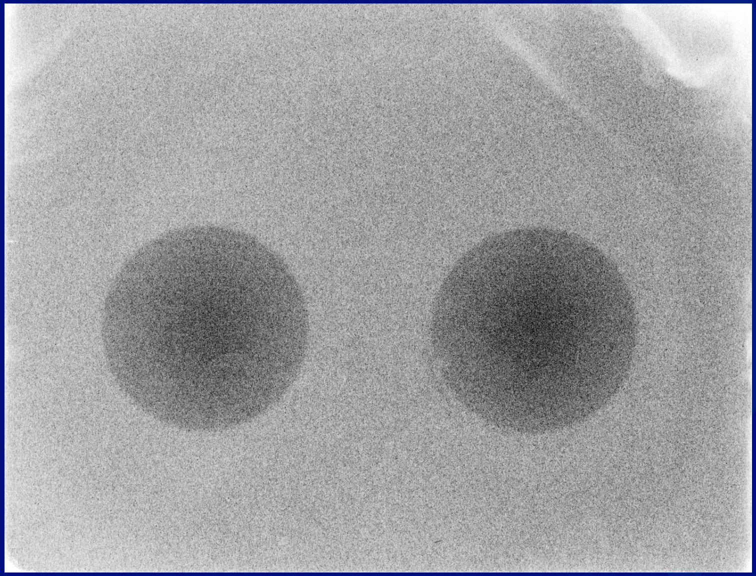
The technique in a nutshell

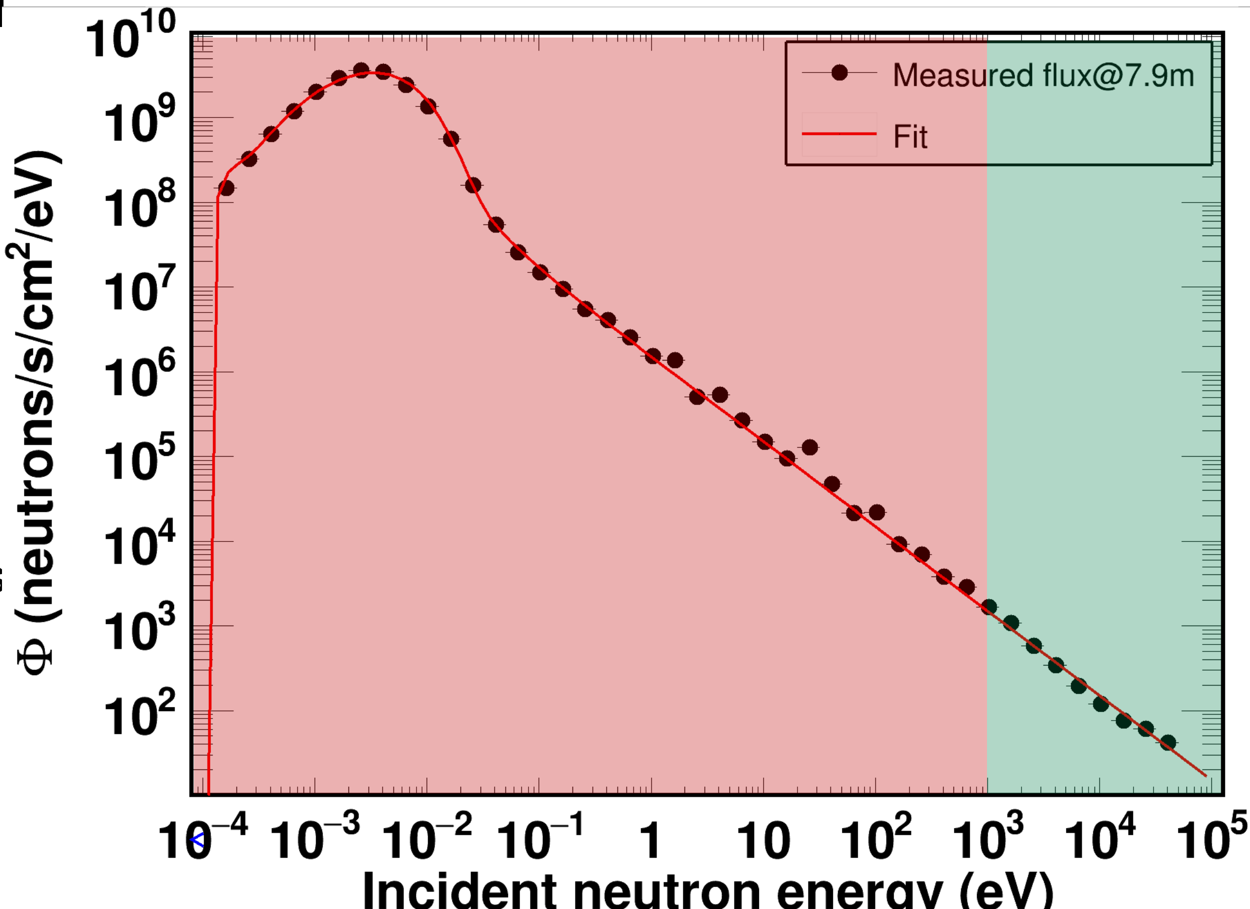


Neutron transmission measurements



Neutron production @ Los Alamos Neutron Science Center

- Neutrons are produced through spallation
- A high energy ion beam impinges on a heavy material
- Neutrons are uncharged and go all over the place
- Collimators: blocks with holes
- Moderation: slow down of neutrons using light materials
- Continuous spectrum (white beam) at **resonance** and **intermediate** energies.



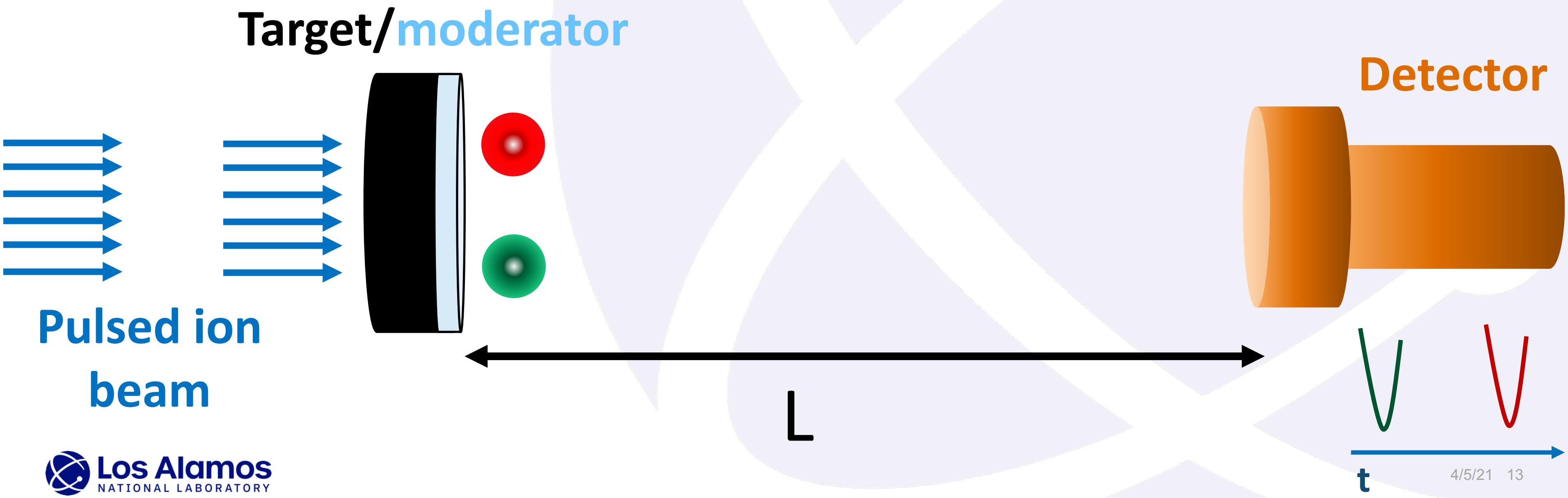
The time of flight (tof) technique

- The neutron velocity *v* is "proportional" to its energy *E*

$$E = \frac{1}{2} m\, v^2 = \frac{m}{2} \left(\frac{L}{t}\right)^2 \cong \left(72.3 \, \frac{L[m]}{t[\mu s]}\right)^2$$

- **Fast** neutrons need less time than **slow** ones, to travel a given distance *L*

- Measuring the travel time or **time of flight** *t*, we reconstruct the incident energy *E*



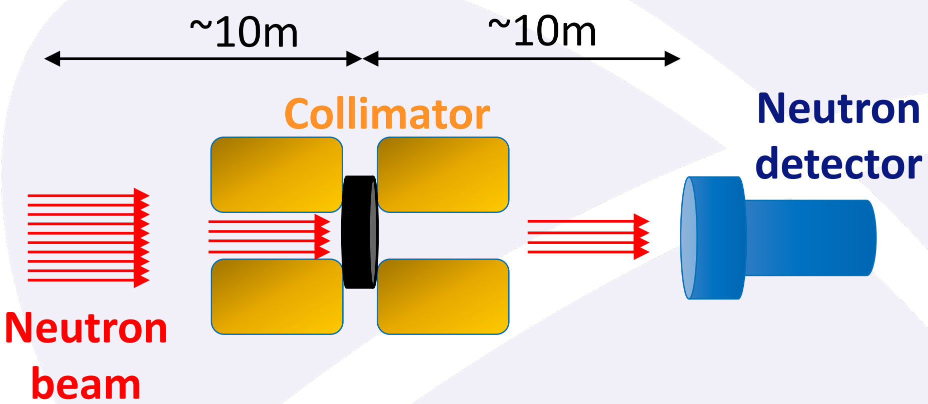
Traditional transmission measurements: How to

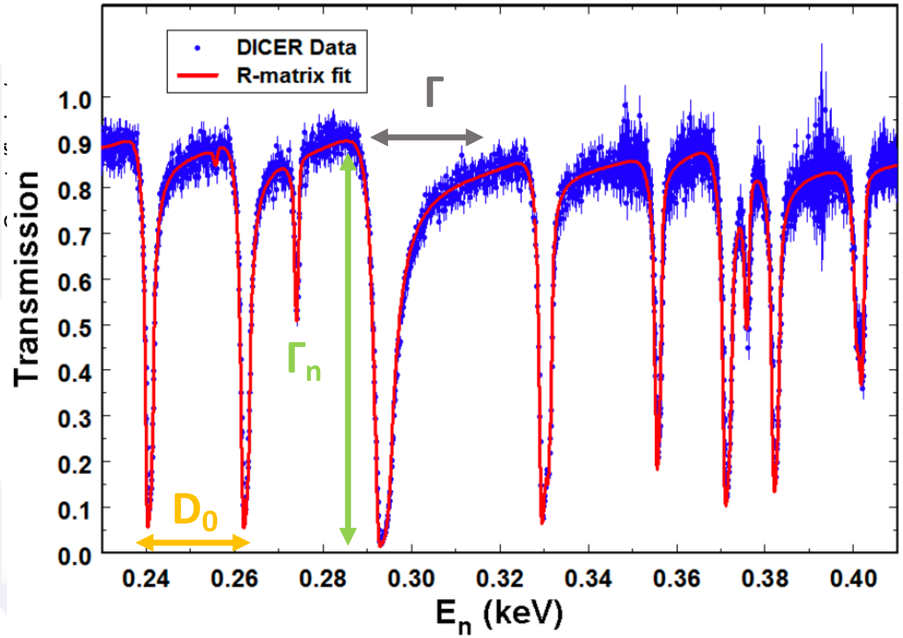
- The neutron spectrum is recorded by a neutron detector (**sample out**)

- A sample, usually big, is installed and absorption dips appear (**sample in**)

- The transmission is the ratio sample in/out

$$T = \frac{Sample\ Out}{Sample\ In}$$





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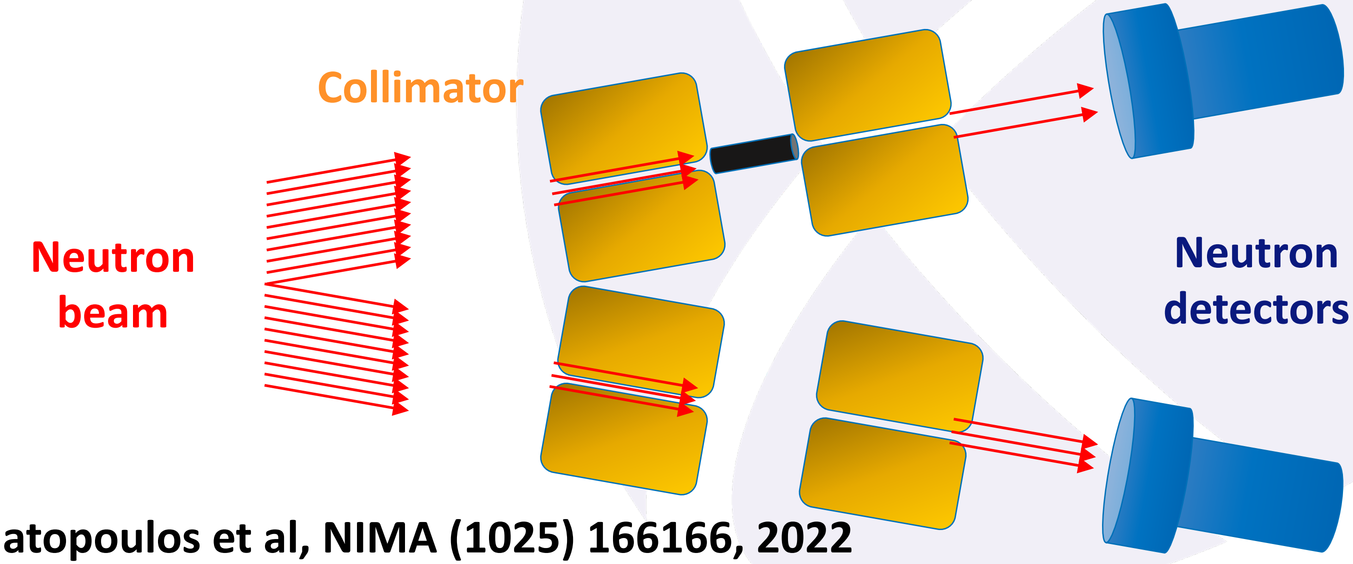
Traditional transmission measurements: Challenges

- Sample cycling is needed therefore a positioning system must be utilized.
- Position systems have a finite accuracy and that's a serious bottleneck, in measuring limited amounts of materials.
- Precise repeatability of the sample's position, relative to the collimation system, is not necessarily ensured.
- Treatment: samples are much larger than the neutron beam diameter which is defined by the collimation system.
- Large samples are sometimes difficult to fabricate when there are dose rates and rarity of the material considerations.
- What if…the sample didn't have to move…?

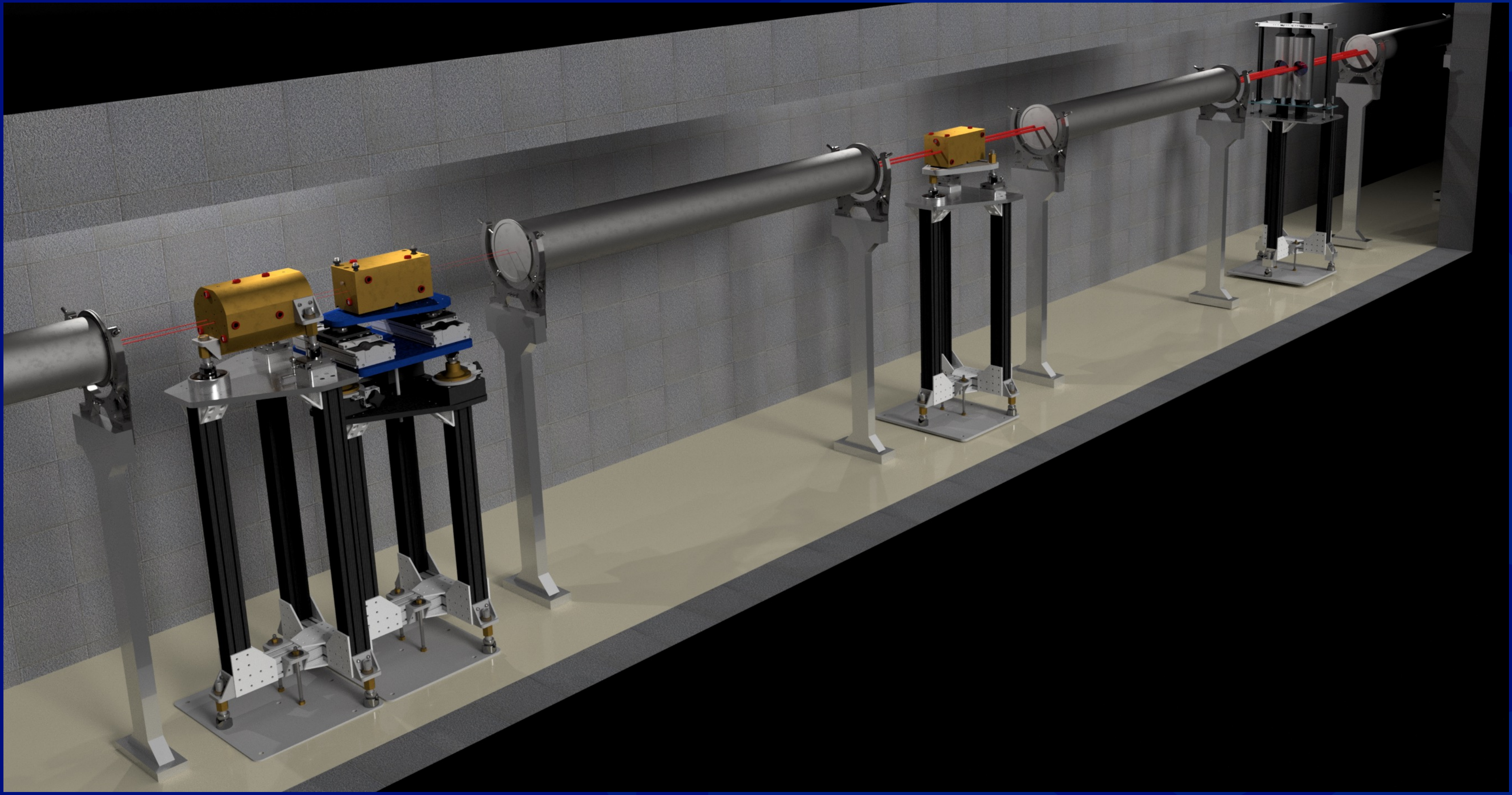
Non-traditional measurements: Binocular approach

- Binocular mode of operation: Simultaneous measurement of sample in/out
- Binocular collimator is a unique concept conceived, designed and executed at DICER.
- No precise repositioning concerns, as long as the sample is precisely positioned beforehand: metrology network ~10μm and ~10 mdeg accuracy
- Added bonus: measurements will be completed 50% faster!
- High flux facility → small samples (~10,000 smaller than typical)
- ~50 radionuclides are within reach ($t_{1/2}$ > 30days, D_0<50eV)

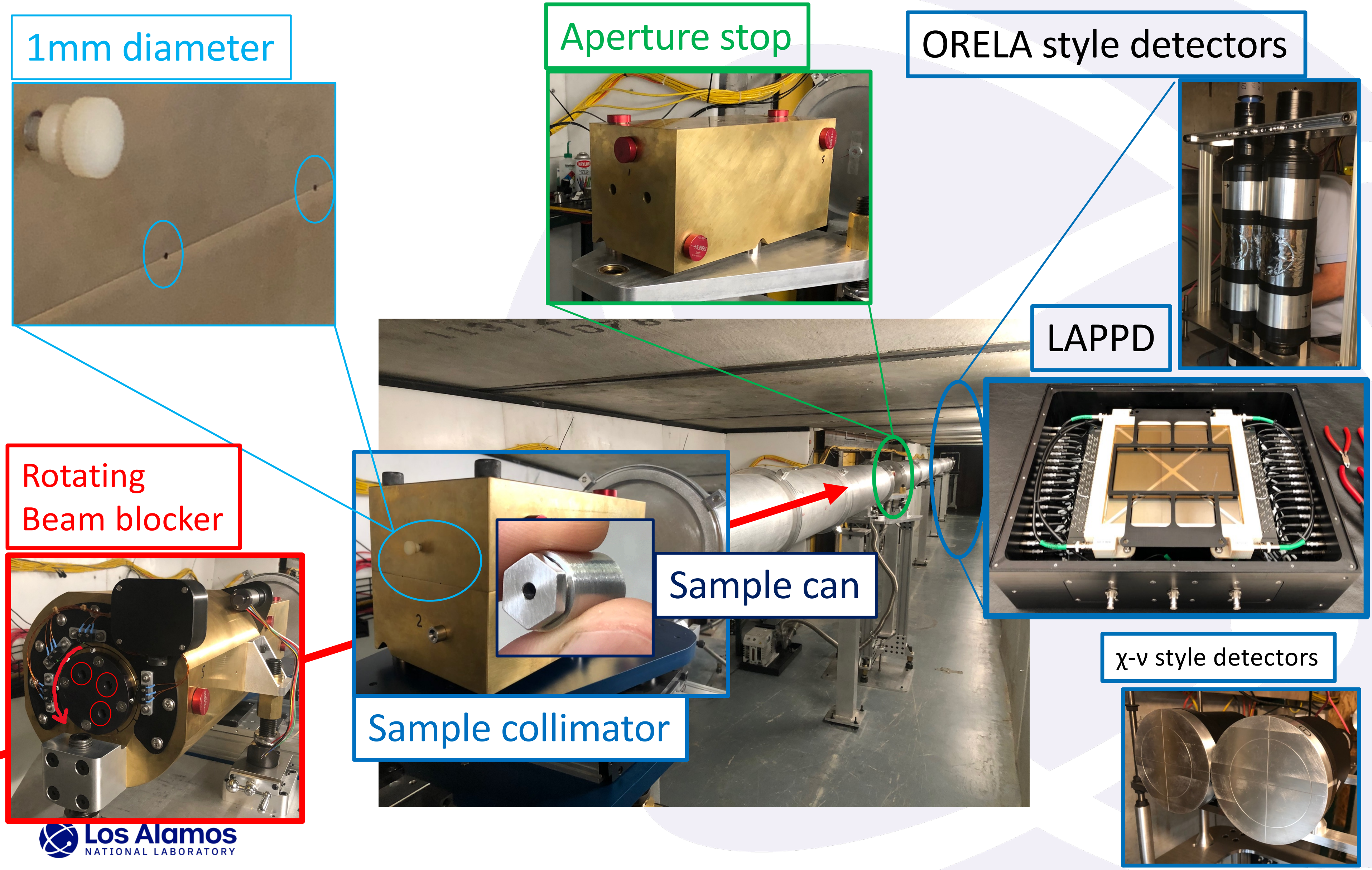


A. Stamatopoulos et al, NIMA (1025) 166166, 2022

Device for Indirect Capture Experiments on Radionuclides



Description of the apparatus:
Device for Indirect Capture Experiments on Radionuclides



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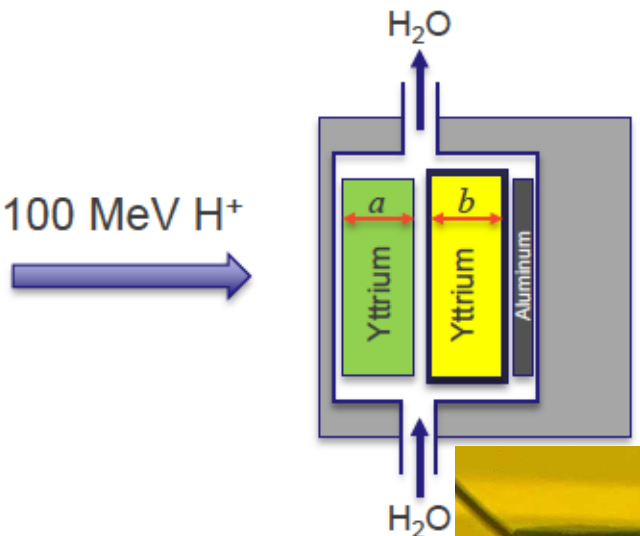
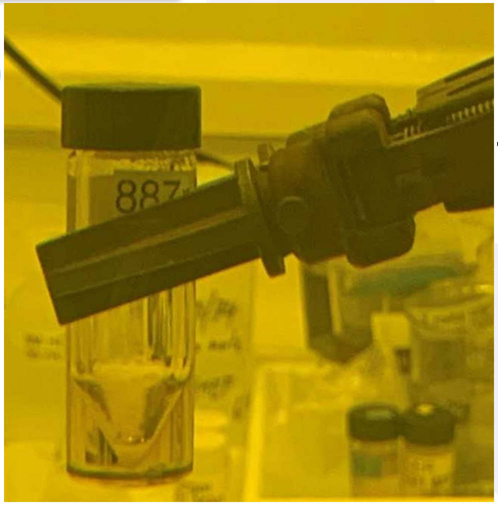
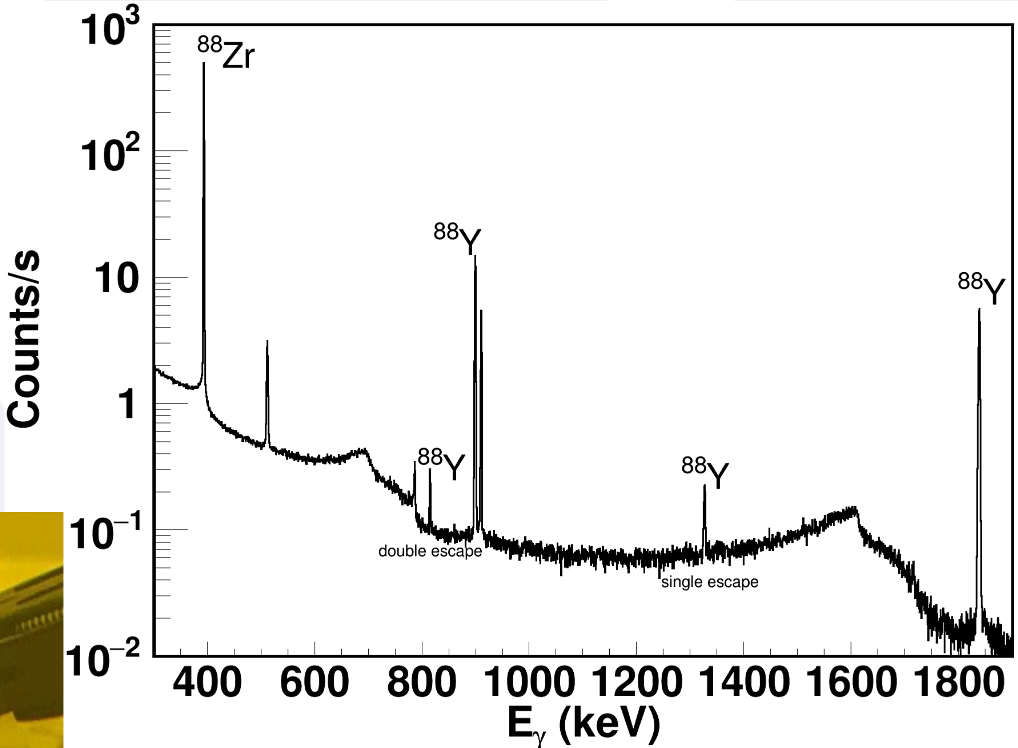
DICER, IPF, C-division synergy: The ^{88}Zr case

- The radioactive sample fabrication relies on the synergy between DICER and IPF
- Proton irradiation of a suitable bulk material
- Chemical separation and purification (600 mCi, 10 mL ^{88}Zr + D_2O•6mol/L DCl)
- Gamma spectroscopy



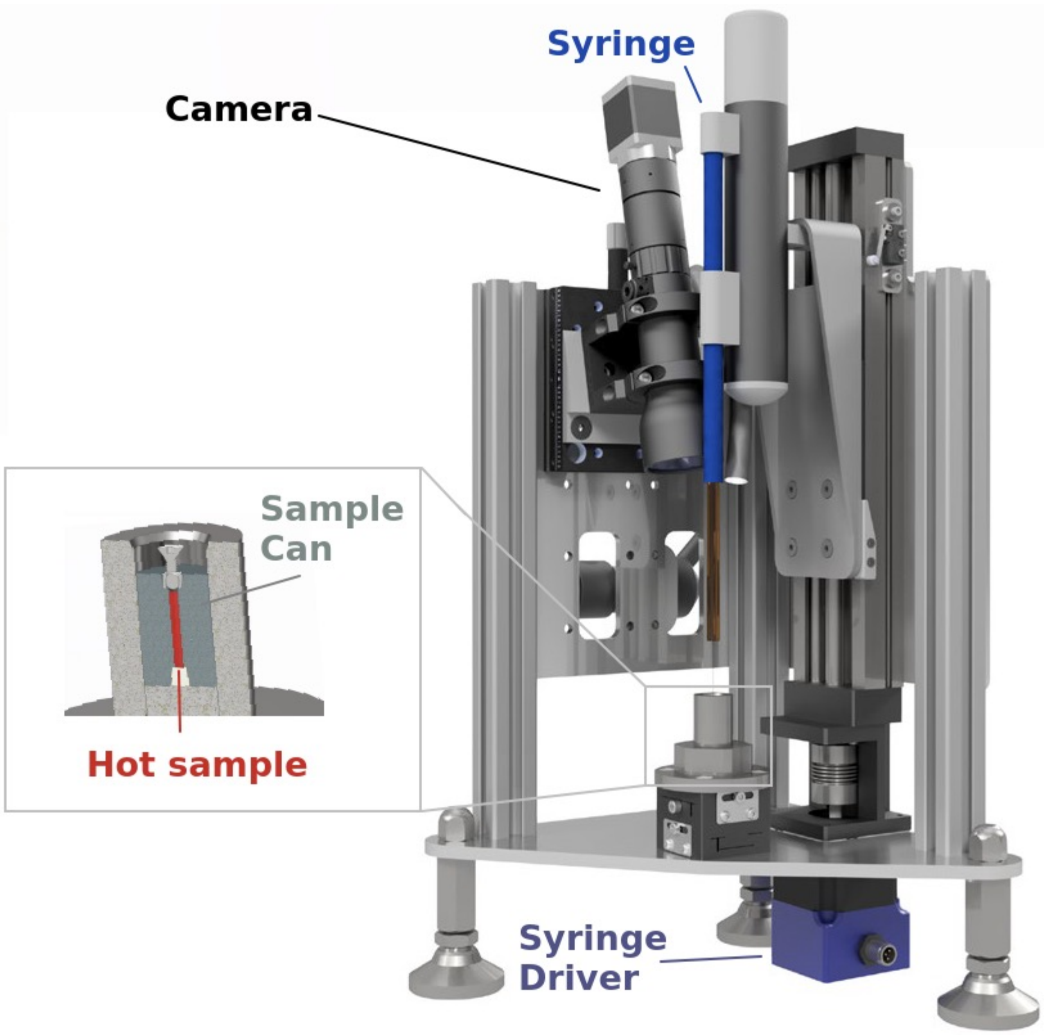


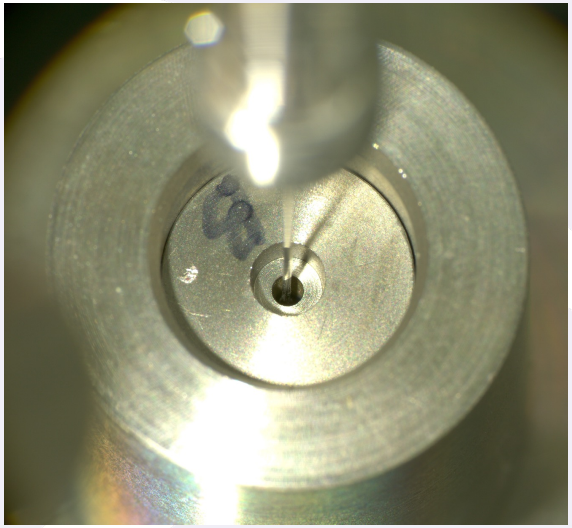


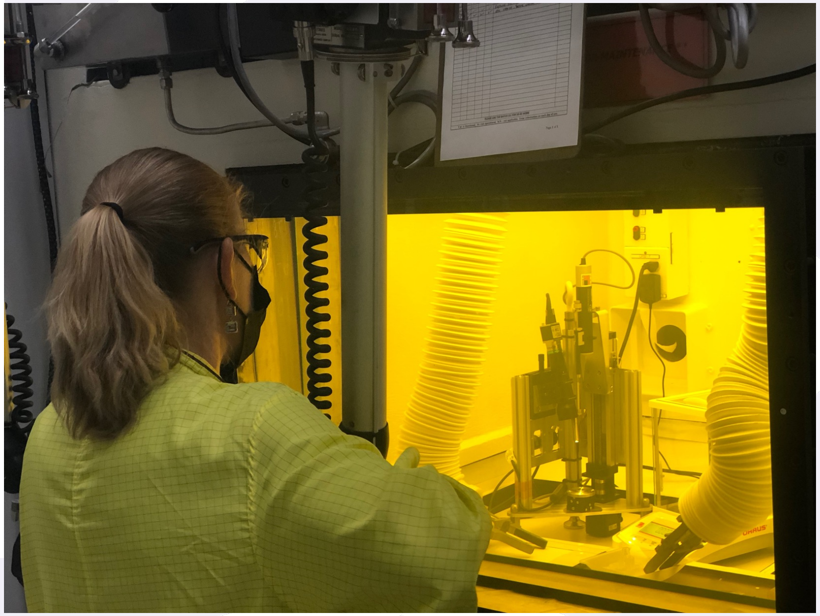
A. Stamatopoulos et al., EPJ., Conf. 260, 03006 (2022)
A. Stamatopoulos et al., J Rad. Nucl. Chem., In press
A. Matyskin et al, Submitted to Scientific Reports

Development of filling station

- Operations have to take place in a hot cell with a remote handling capability

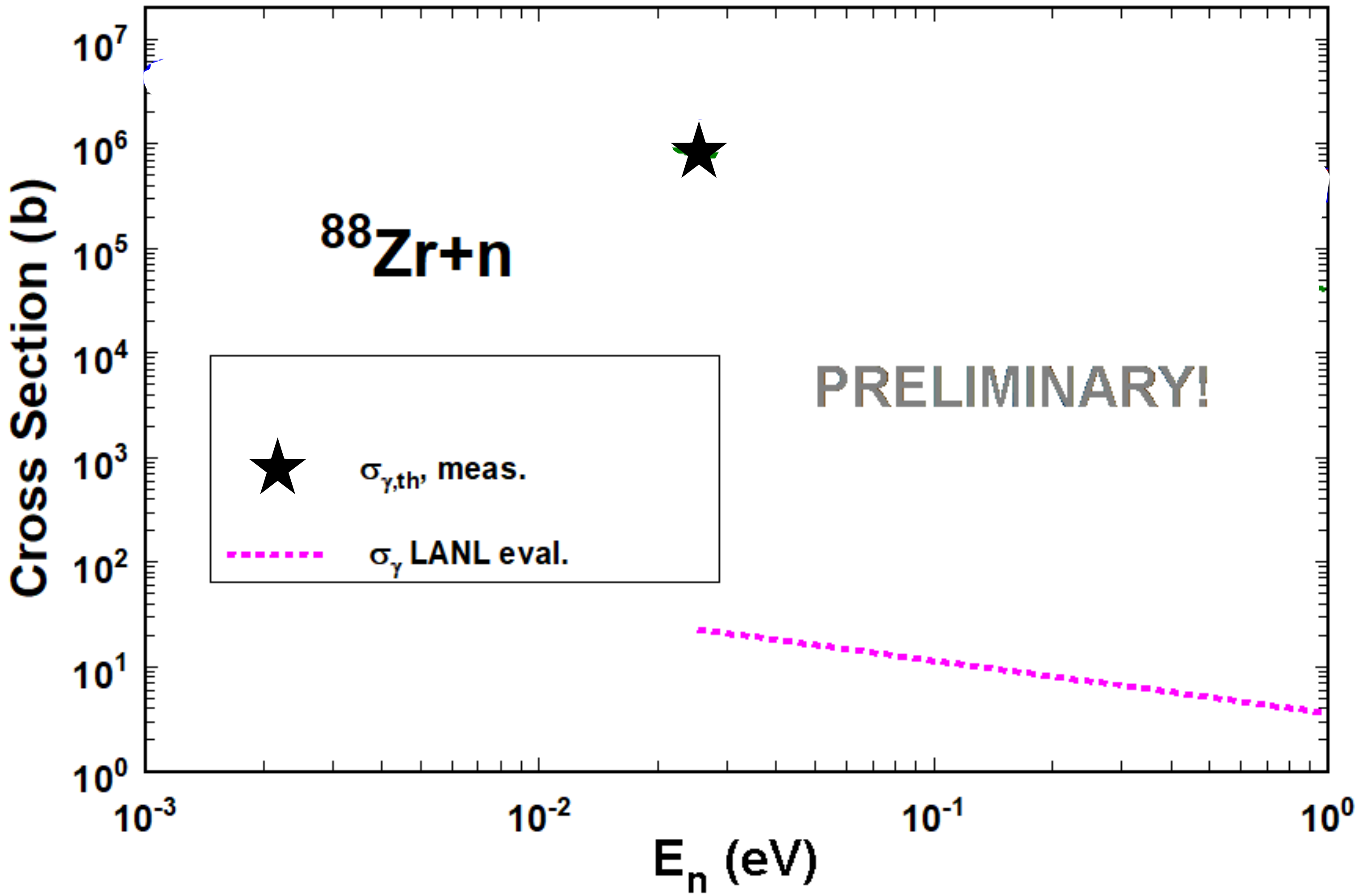






^{88}Zr

- ^{88}Zr was recently reported (Shusterman et al, Nature 565, p. 328 (2019)) to have an enormous capture cross section at thermal: σ_{th} = *8.61 kb*
- We measured a 66 ng ^{88}Zr sample (~8,500 times smaller than the next smallest)

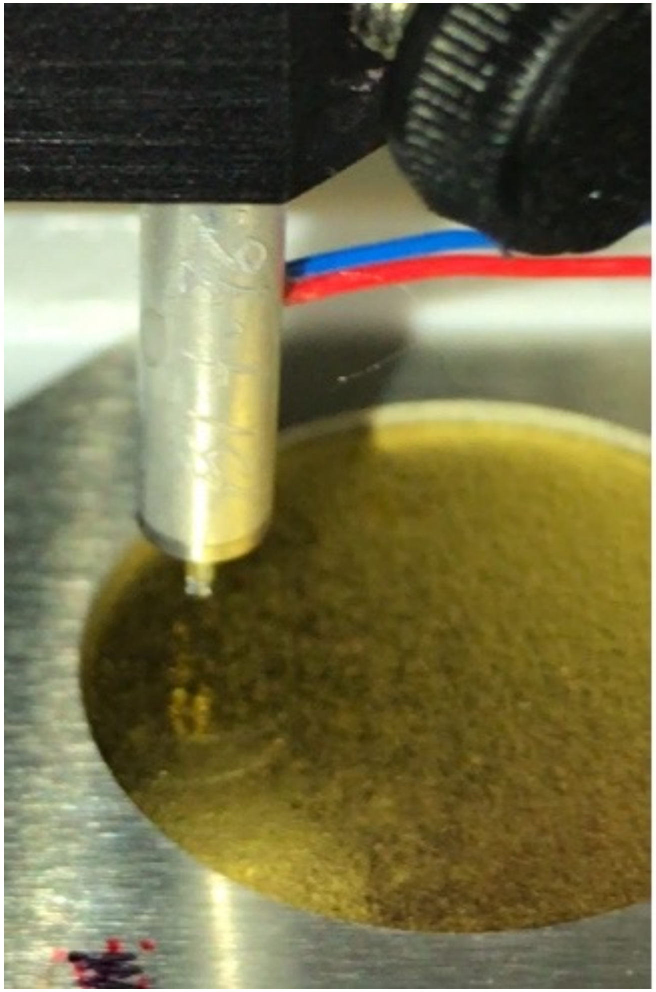


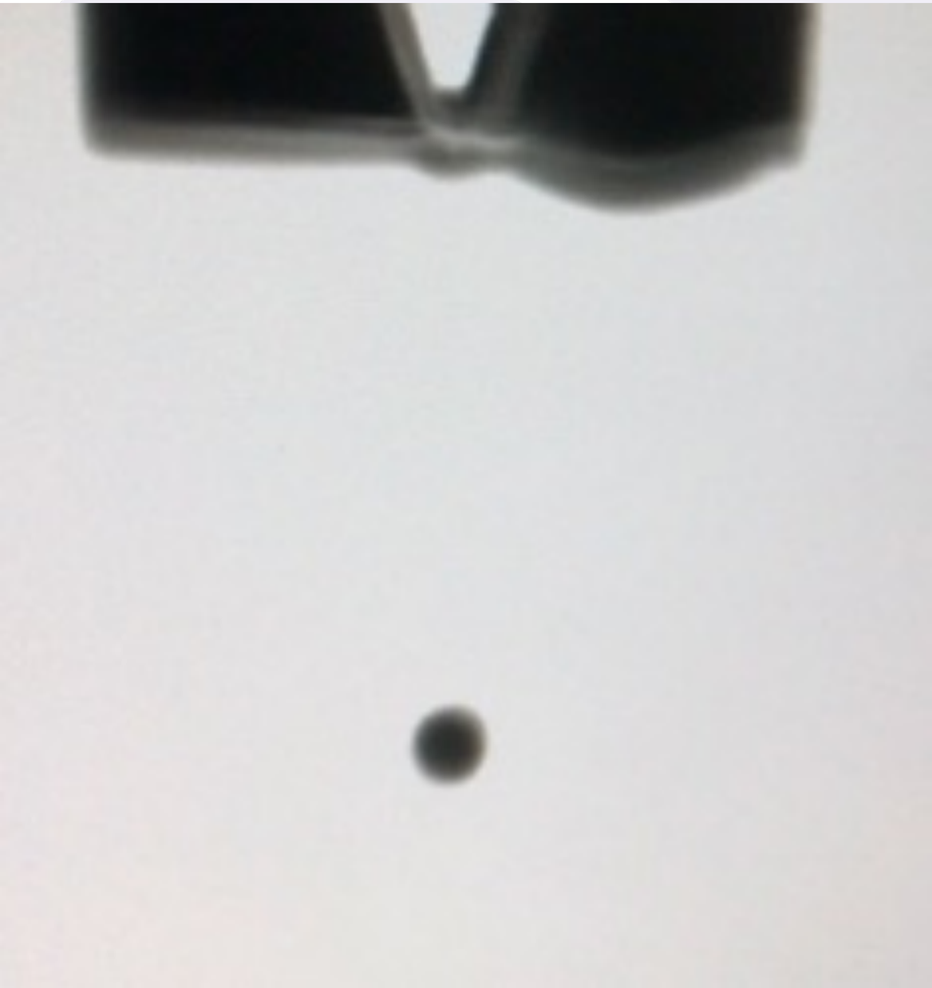

Thank you for your attention!

Back-up slides

Development of inkjet printing

- Inject printing of radioactive samples is a possibility we are currently exploring
- This will allow to print samples with a small diameter (i.e. ^{88}Y)





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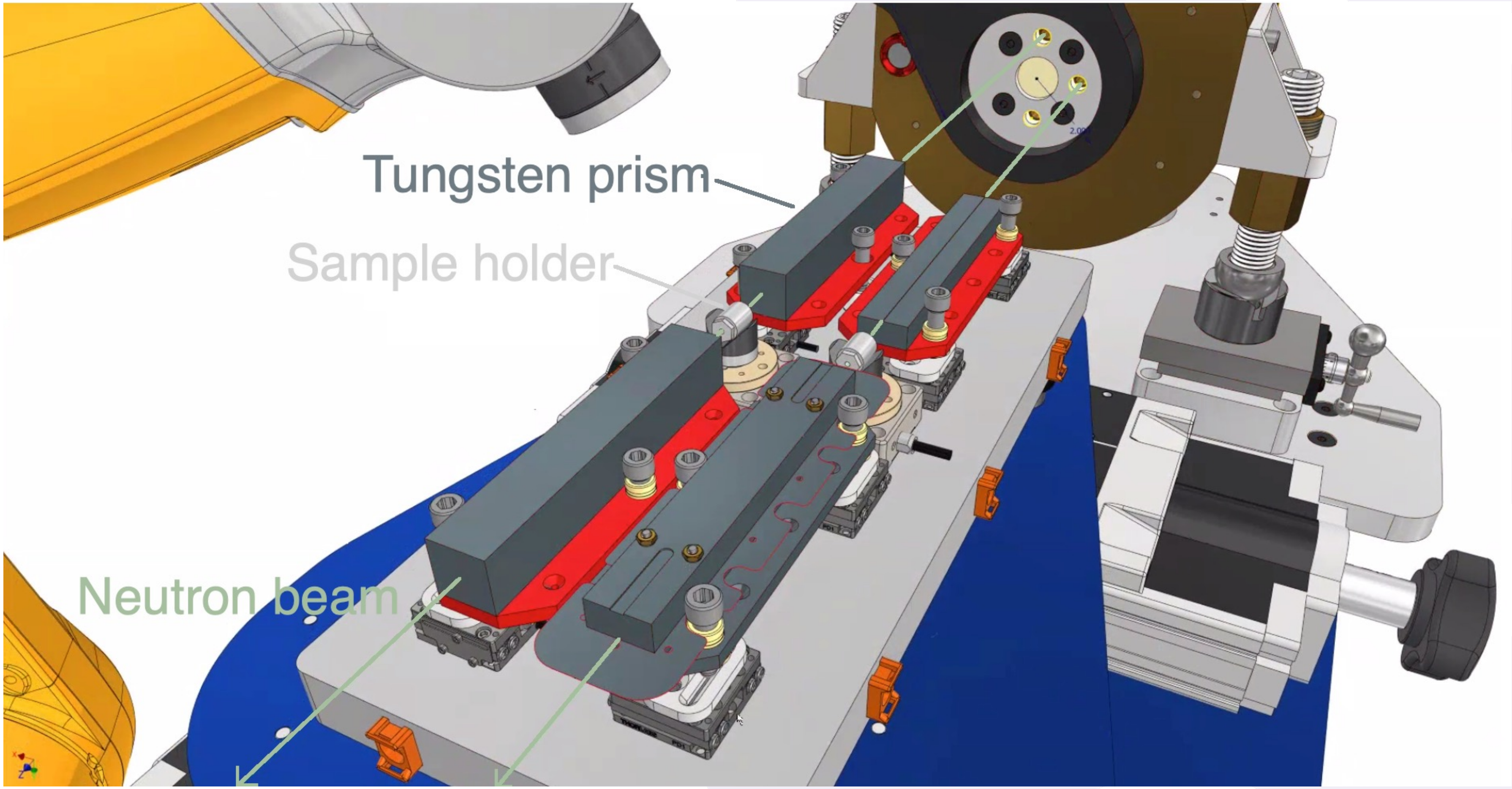
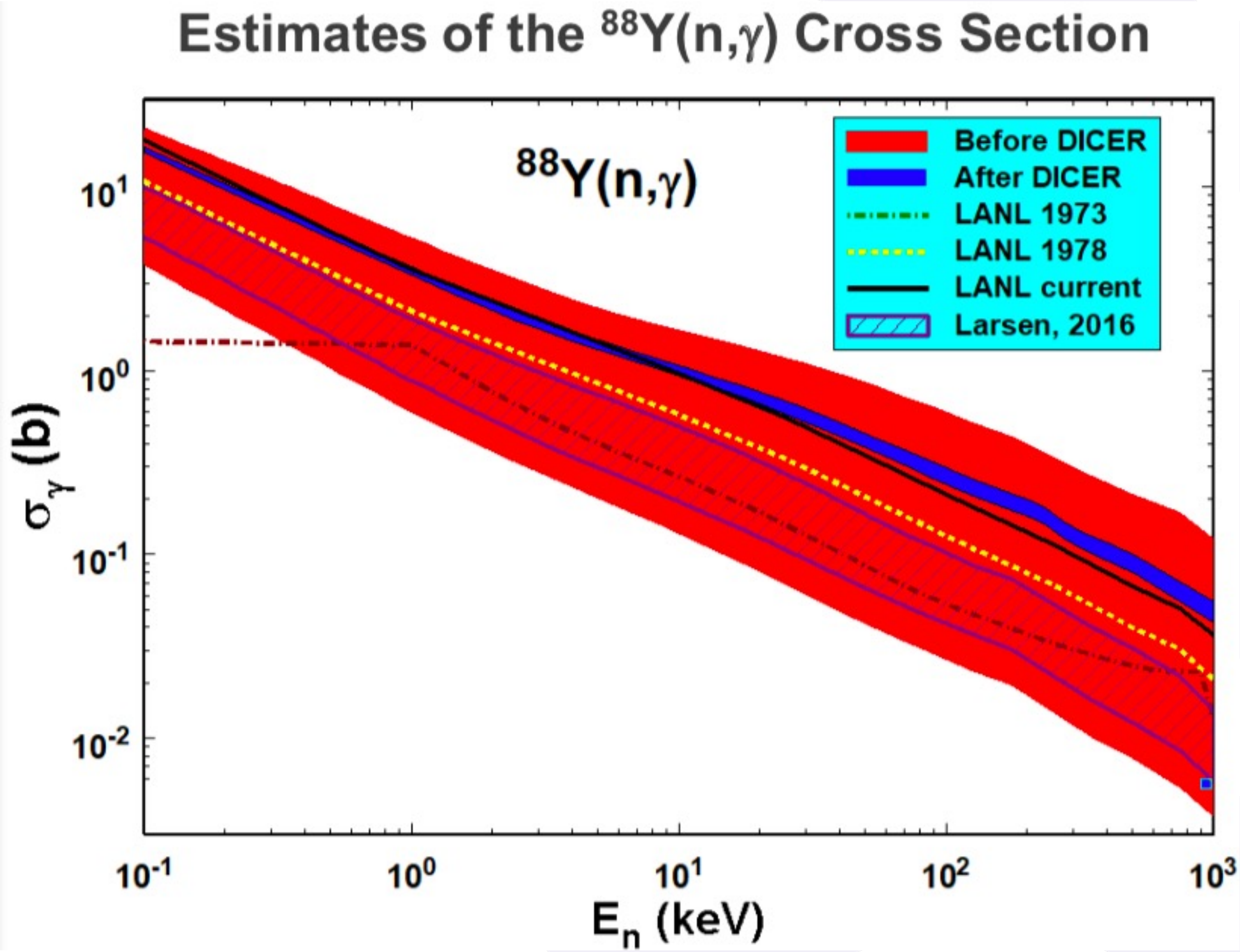
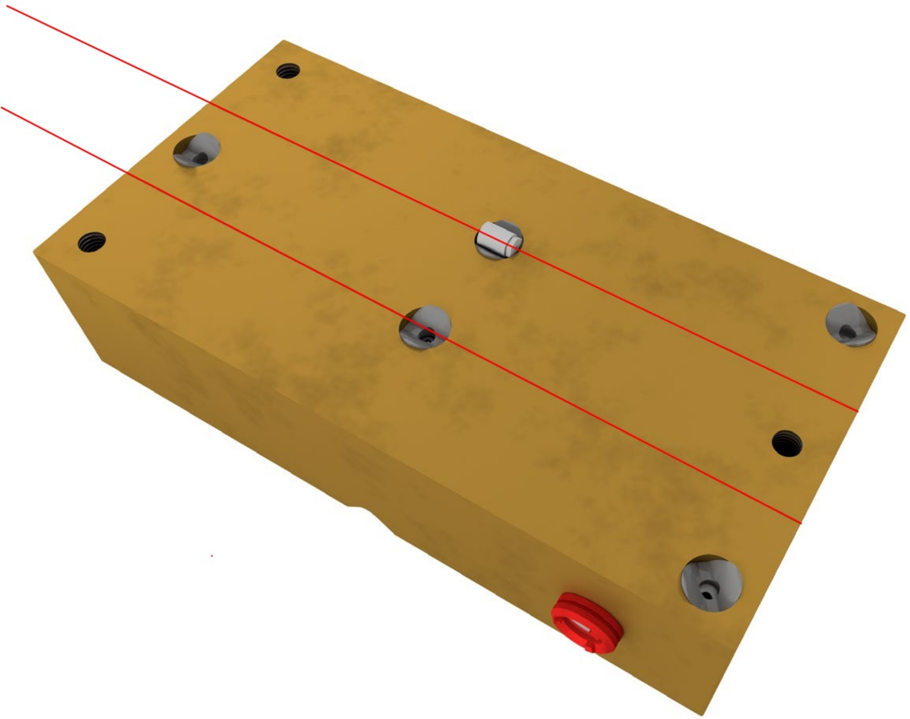
Development of a 0.1 mm collimator

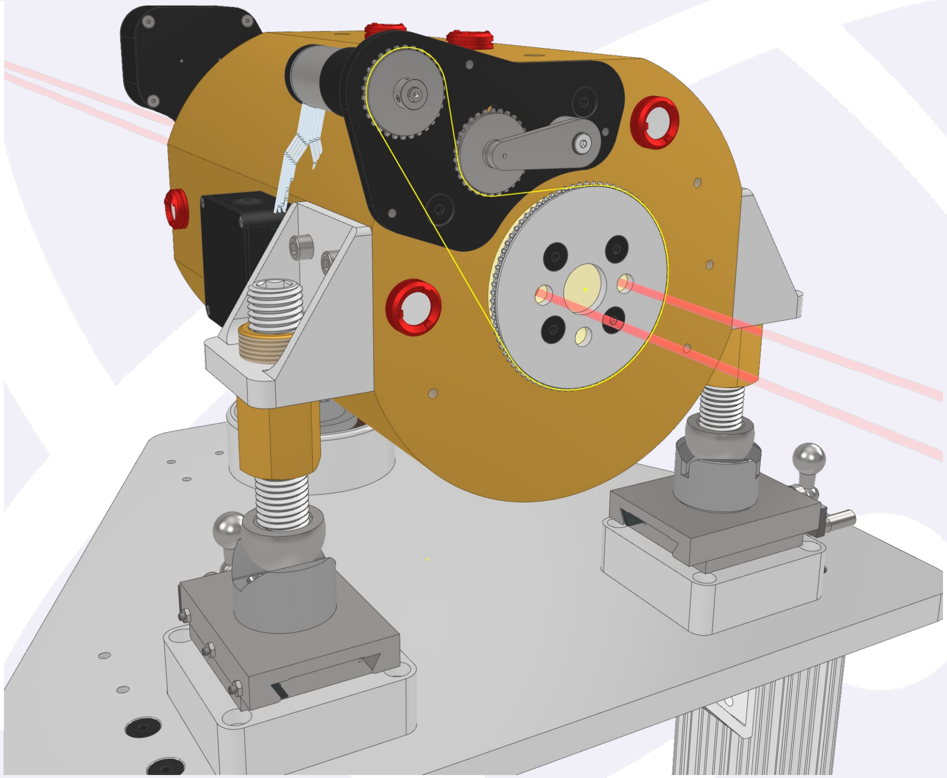
- A smaller collimator will allow measurements on: smaller samples, higher energies, smaller cross sections
- Conceptual design is ready
- 4 tungsten pieces, each with each own precision alignment system

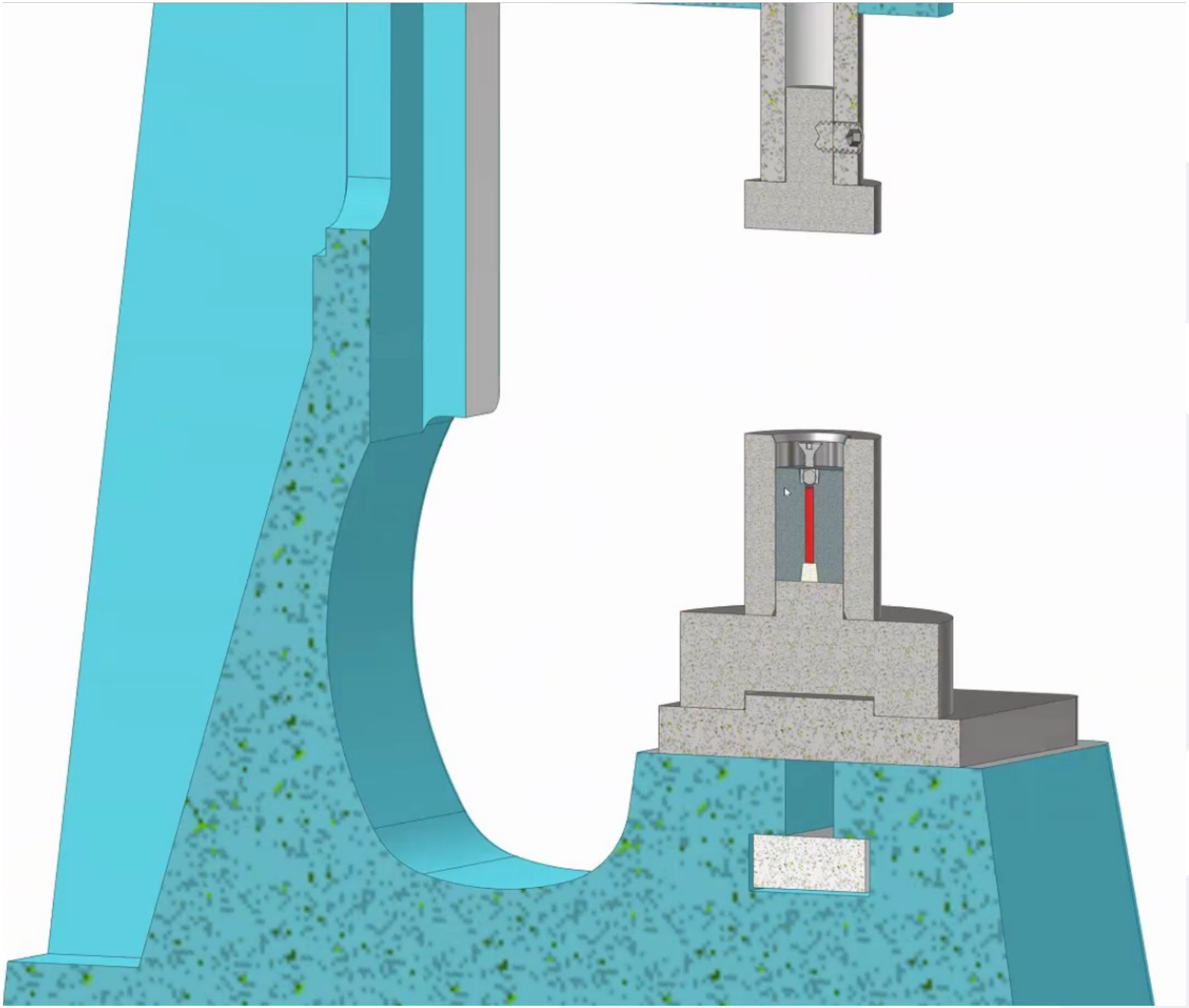


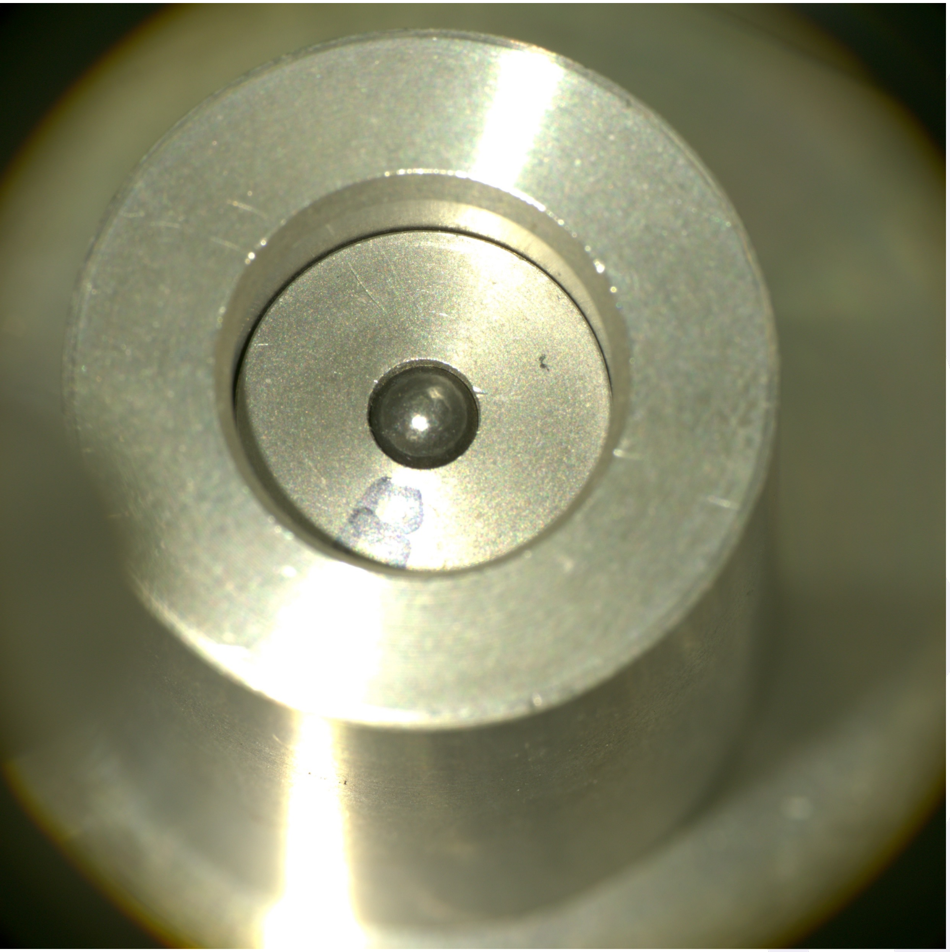
^{88}Y

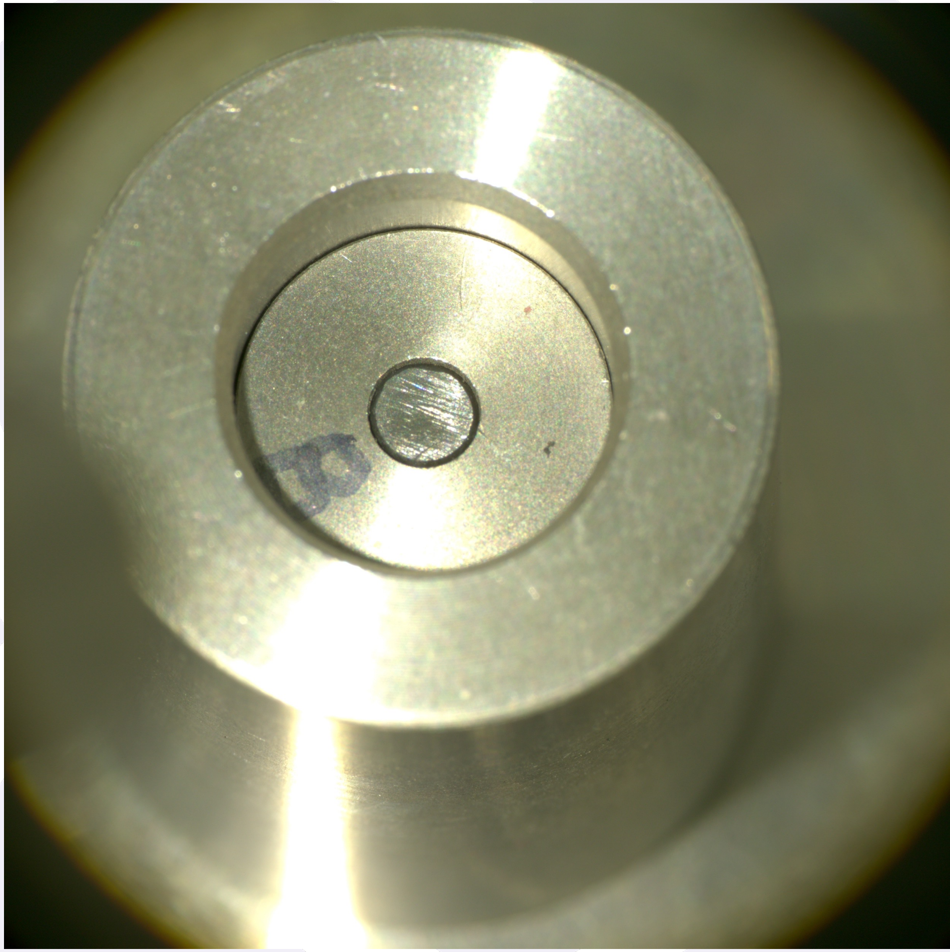

Estimates of the ⁸⁸Y(n,γ) Cross Section
⁸⁸Y(n,γ)
Before DICER
After DICER
LANL 1973
LANL 1978
LANL current
Larsen, 2016
σγ (b)
10¹
10⁰
10⁻¹
10⁻²
10⁻¹
10⁰
10¹
10²
10³
En (keV)





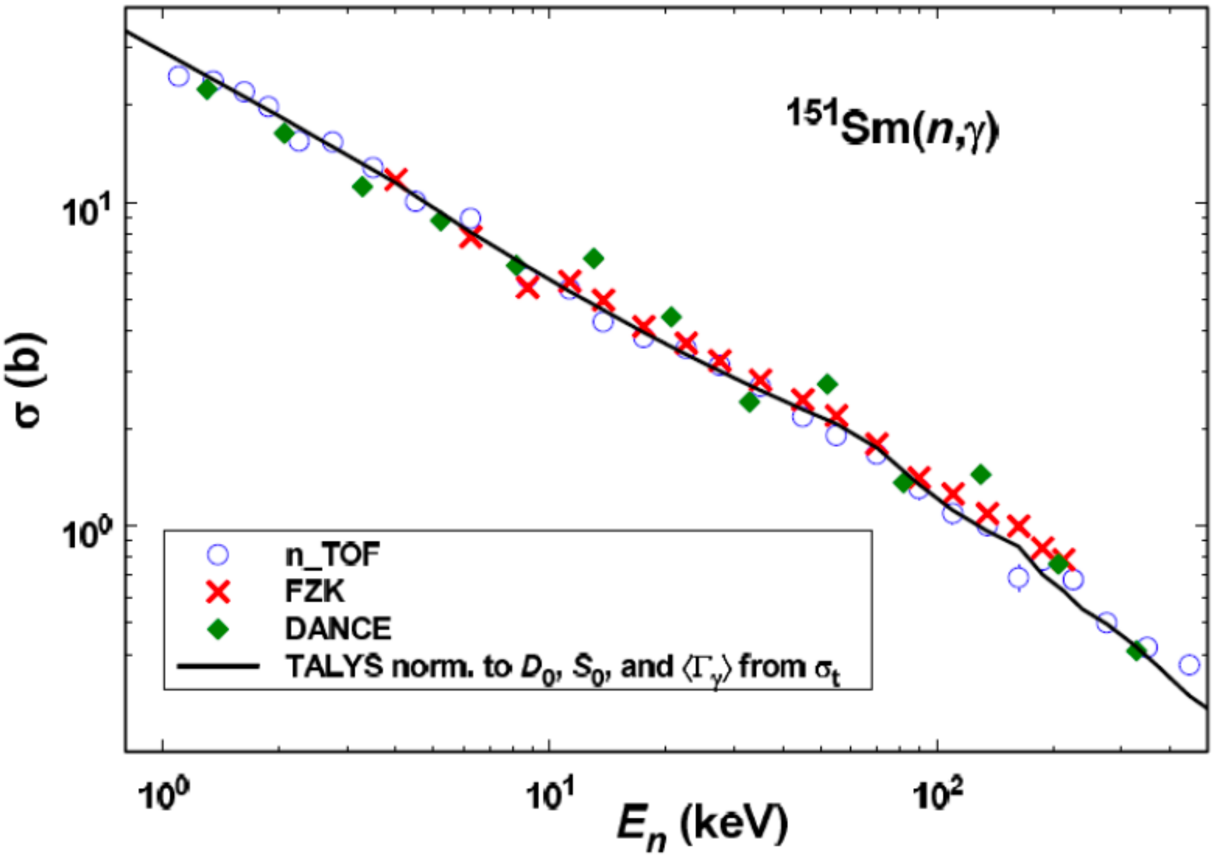






Introduction: Proof of Principle

- Resonance analysis on $^{151}Sm(n,tot)$ data from RPI
- Normalize TALYS calculations and compare to $^{151}Sm(n,\gamma)$ EXFOR data
  P. Koehler, LA-UR-14-21466



Design of sample canisters

- Liquid samples ensure homogeneity
- Hard acidic environment is usually unavoidable
- Need for proper canisters to minimize dose and ensure secure handling