Zirconium-88 Neutron Absorption Cross Section at n_TOF EAR2

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$^{88}Zr(n,\gamma)$ - Background

- In 2019, ^{88}Zr was discovered to have a thermal neutron absorption cross section of 861,000 barns (measured) rather than 10 barns (expected).
  - Larger than ^{157}Gd, ^{10}B, ^{6}Li, ^{3}He
  - Smaller than ^{135}Xe
    - Both ^{88}Zr and ^{135}Xe are radioactive
- **n_TOF EAR2 is uniquely positioned to measure a low energy resonance which explains the large cross section**

LETTER

https://doi.org/10.1038/s41586-018-0838-z

The surprisingly large neutron capture cross-section of ^{88}Zr

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^{88}Zr(n,γ) – Current Picture

- 2019 LLNL – thermal neutron capture σ_T= 861,000$\pm$69,000 barns
- 2021 LLNL – thermal neutron capture σ_T = 804,000$\pm$63,000 barns and resonance integral I = 2,530,000$\pm$280,000 barns
- 2023 LANL – thermal neutron capture σ_T = 771,000$\pm$31,000 barns and resonance integral I = 15,210$\pm$670 barns
  - LANL DICER uses neutron ToF with transmission for energy-resolved measurement
  - Controversy with resonance integral
  - **<u>Resonance claimed at 0.171 eV</u>**
  - Nature pre-print, under review

Aqueous harvesting of ^{88}Zr at a radioactive-ion-beam facility for cross-section measurements

Jennifer A. Shusterman, Nicholas D. Scielzo, E. Paige Abel, Hannah K. Clause, Nicolas D. Dronchi, Wesley D. Frey, Narek Gharibyan, Jason A. Hart, C. Shaun Loveless, Sean R. McGuinness, Logan T. Sutherlin, Keenan J. Thomas, Suzanne E. Lapi, J. David Robertson, Mark A. Stoyer, Eric B. Norman, Graham F. Peaslee, Gregory W. Severin, and Dawn A. Shaughnessy

Phys. Rev. C **103**, 024614 – Published 26 February 2021

Research Square

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Discovery of the origin of the enormous 88Zr neutron-capture cross section and quantifying its impact on applications

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OPEN **Production of zirconium-88 via proton irradiation of metallic yttrium and preparation of target for neutron transmission measurements at DICER**

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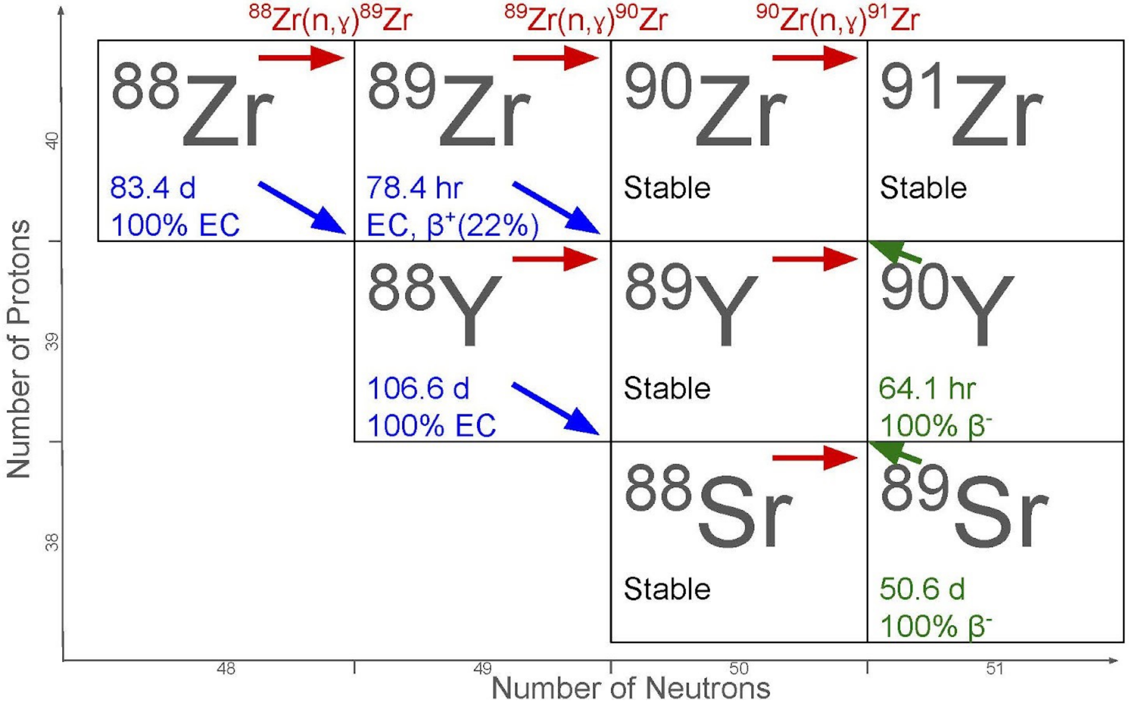
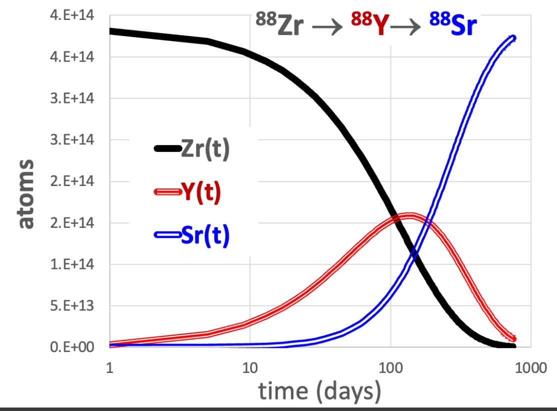
^{88}Zr(EC)^{88}Y and ^{88}Y(EC)^{88}Sr Background

- ^{88}Zr electron capture emits a 393keV gamma ray.
- ^{88}Y electron capture emits 898keV and 1.836MeV gamma rays.

- Two samples:
  - "Test sample" for background studies and setup optimization
    - "Rich" in ^{88}Y, which is the dominant background for n_TOF measurement and main RP concern.
    - By late March: 0.5 mCi of ^{88}Zr and 3.2 mCi of ^{88}Y
  - "Measurement sample":
    - Fresh ^{88}Zr for (n,γ) measurement

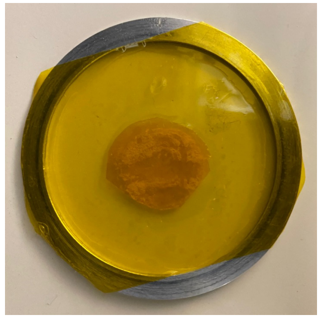
- **Pending INTC approval, the 100mCi ^{88}Zr sample will be made by LANL directly for this measurement to minimize ^{88}Y activity.**



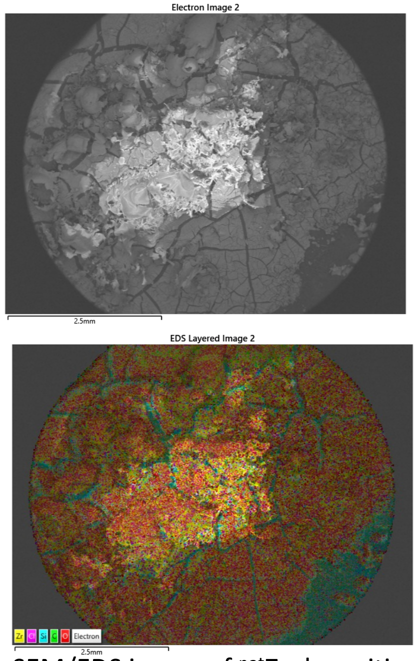


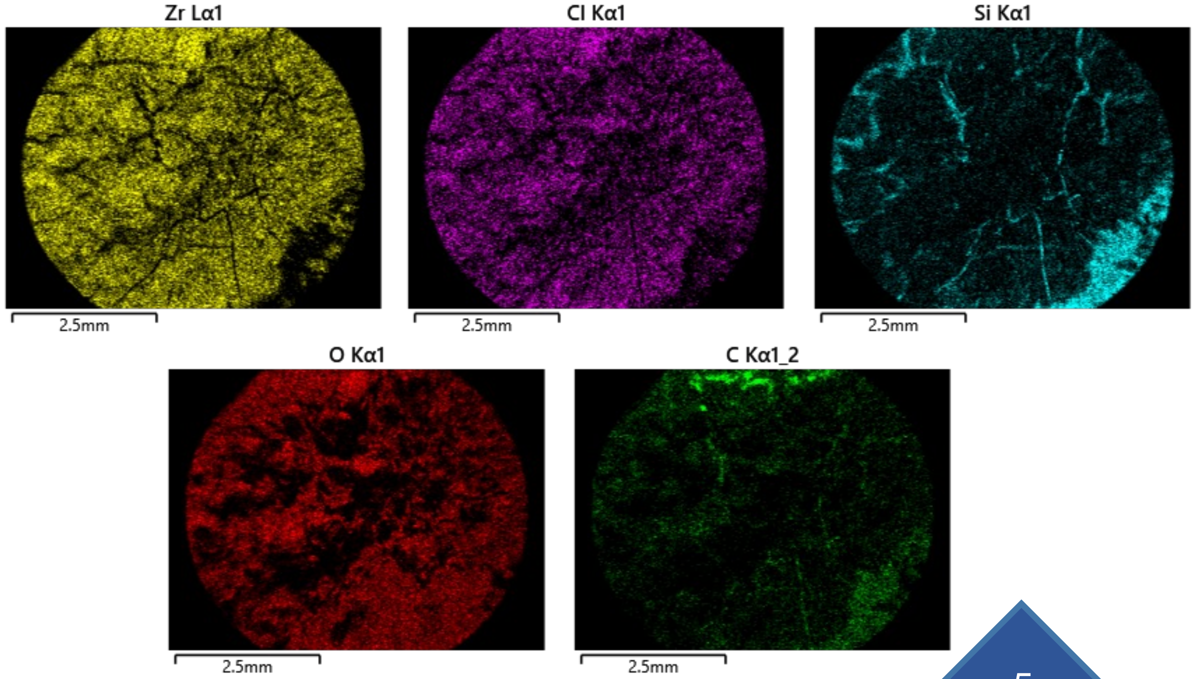
Sample Prep

- The ^{88}Zr sample is provided by US DOE (LANL) in 2N HCl since zirconium is not water soluble.
- Evaporated into 1cm diameter to balance reaction rate and alignment uncertainty.
    - Sample is zirconium chloride and zirconyl chloride.
        - $ZrCl_4 + ZrOCl_2{\cdot}H_20$
    - Demonstration with ^{nat}Zr shown below.
        - Evaporated between two pieces of Kapton tape for conformal containment.
        - Mounted to aluminum ring.



^{nat}Zr within Kapton tape and n_TOF EAR2 aluminum ring

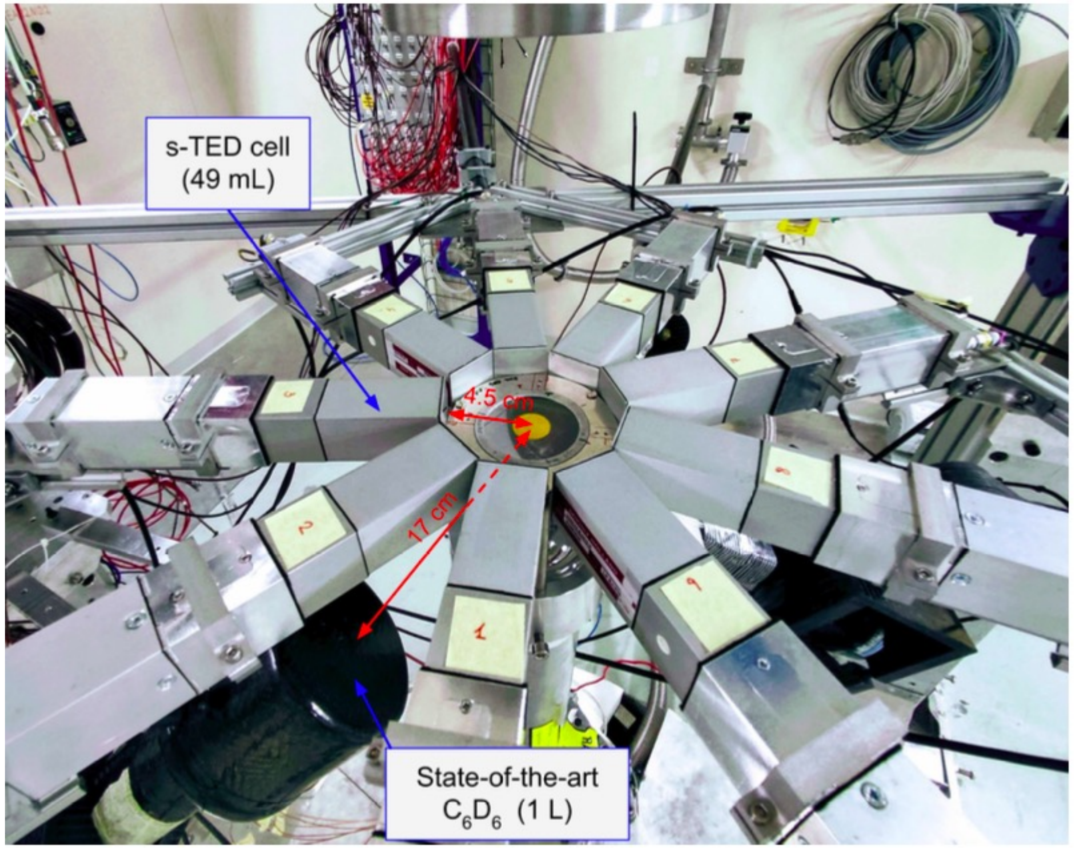




SEM/EDS image of ^{nat}Zr deposition

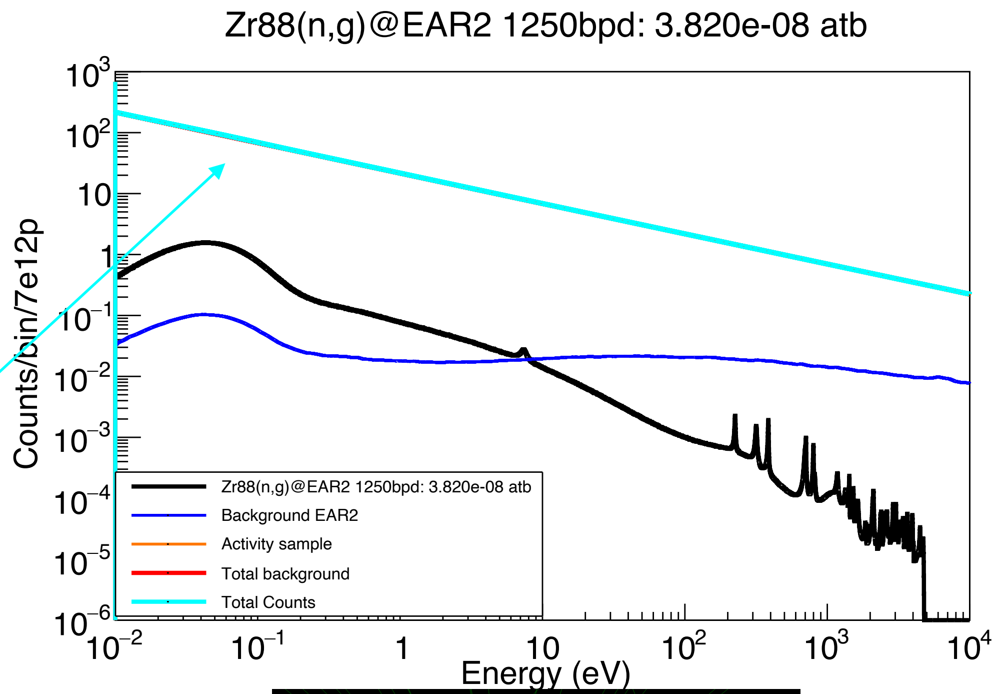
n_TOF EAR2

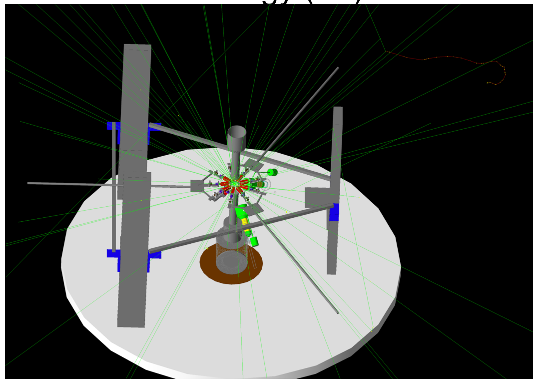
- n_TOF EAR2 is uniquely suited to this measurement
  - Fast data acquisition, sufficient fluence
- We intend to use the segmented total energy detectors (sTEDs) in the circle configuration.
  - Able to sustain the very high rates associated to the activity and the high-flux of EAR2
  - Optimum efficiency, segmentation, and sensitivity.
    - Used before for challenging measurements on radioactive targets (^{79}Se, ^{94}Nb).
  - Able to increase detector stand-off, pending background measurement results.



Measure and Confirm Thermal XS

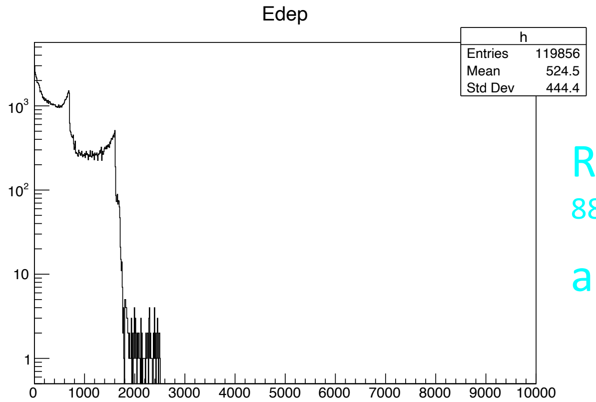
- Counting rate estimates that include the full beamline simulations of n_TOF EAR2
  - This includes the energy resolution function.
- We include the TENDL 2021 database, which is the only library with the updated cross section available at time of the preliminary work
- 1×10^{18} protons for background and 4×10^{18} for signal
- 100mCi ^{88}Zr signal sample
  - Assuming the sample is in the beam from week 3 to week 8 after separation:
    - ^{88}Zr 84 mCi → 63 mCi
    - ^{88}Y 12 mCi → 24 mCi (main background)
- We have simulated the detector efficiency by first generating the cascades using NuDEX and including them into the Geant4 simulations
- Using Geant4 with EAR2 geometry and sTEDs we calculate the detector efficiency >300keV
- 18-20 March test proposed to validate count rate and detector performance.

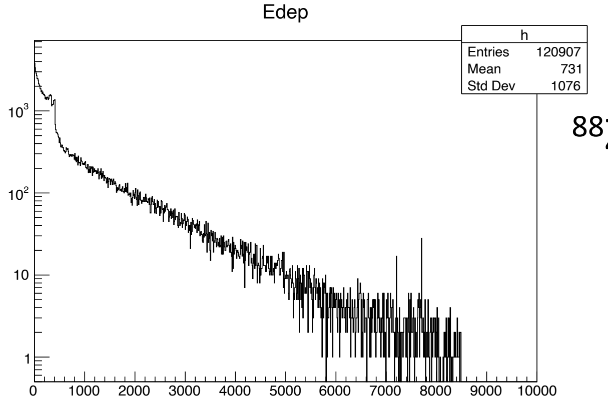




Background Suppression

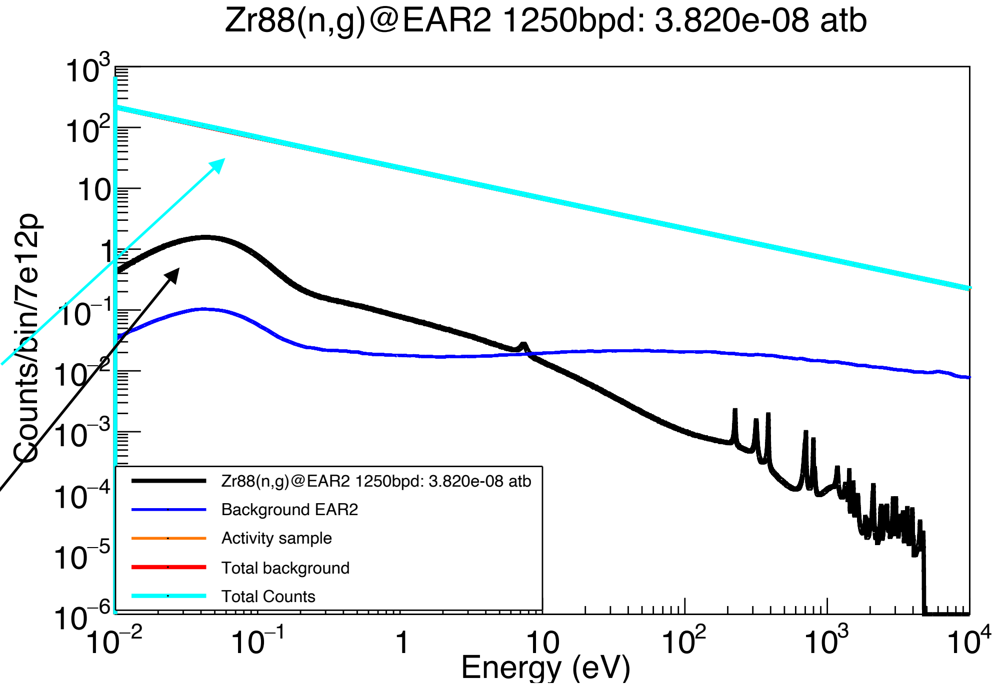
- Based on proposed 18-20 March test sample results, there are options for further background suppression:
  - Can add shielding (Al ring)
  - Gamma thresholds





Radioactivity background: ^{88}Y gammas at 898keV and 1.836MeV

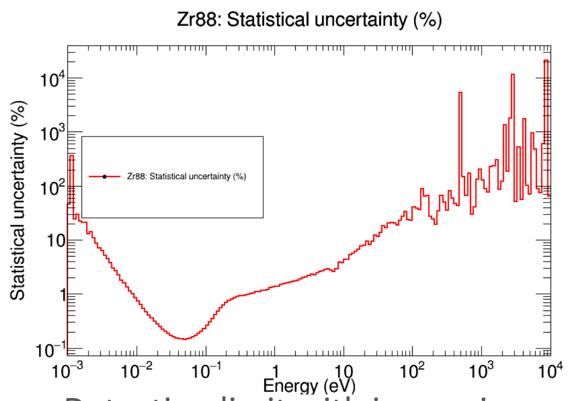
^{88}Zr(n,γ) signal

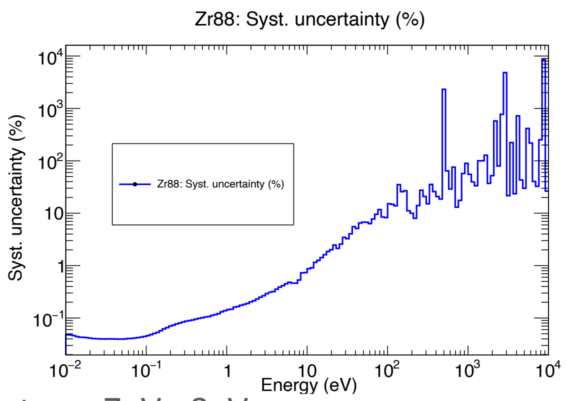


We can use gamma energy thresholds to suppress ^{88}Y background at the cost of statistics…

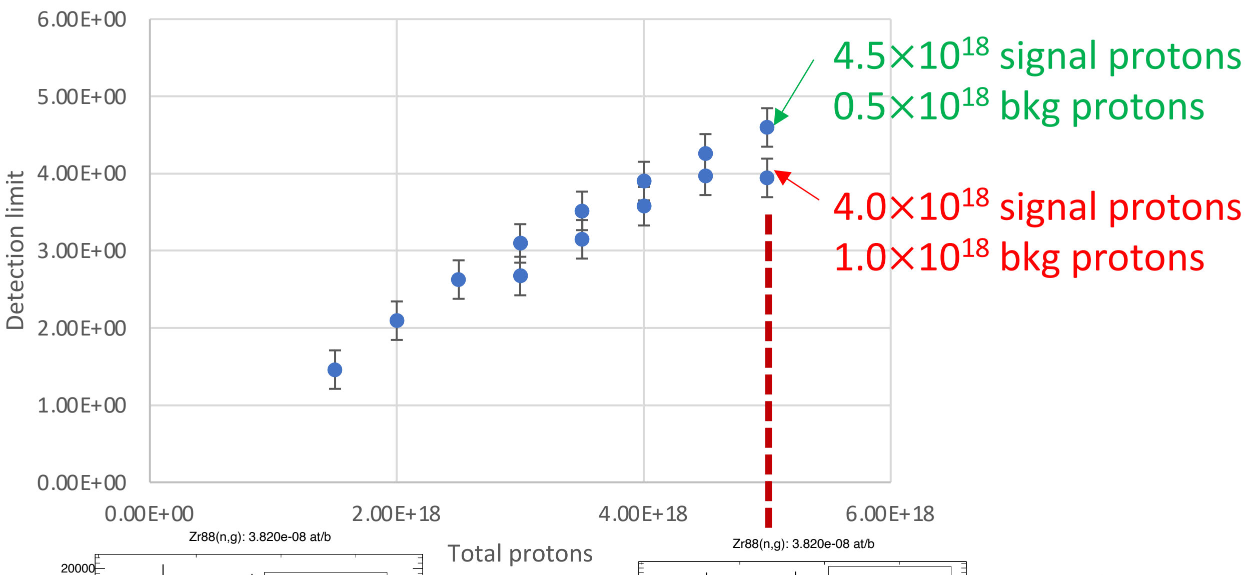
Projected Sensitivity

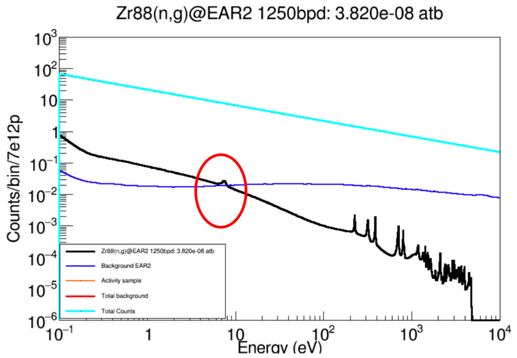
- EAR2 background subtracted with realistic systematic uncertainty of 0.5%. Radioactivity background subtracted with statistical uncertainty only.
  - Able to confirm large thermal neutron cross section
  - Probability of confirming preliminary DICER 0.171 eV resonance. First capture-based confirmation!
  - Able to search for resonances to ~10eV
- TENDL 2021 does not have the 0.171 eV resonance
  - Using the TENDL 2021 resonance at 7 eV as proxy for higher resonance hunting.
  - $D=(C_{Zr}-C_B)/Unc(C_{Zr}-C_B)$
  - Still optimizing mixture of signal to background counts.

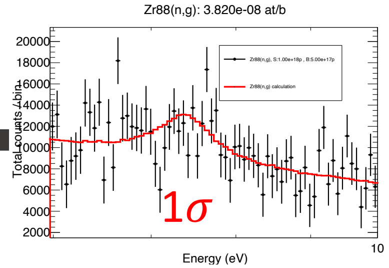


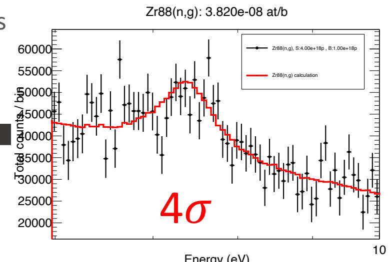


Detection limit with increasing protons, 7eV->8eV





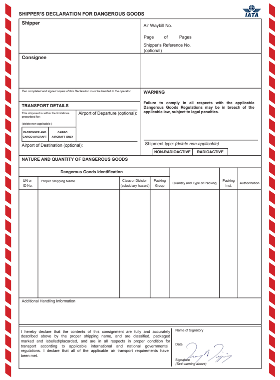


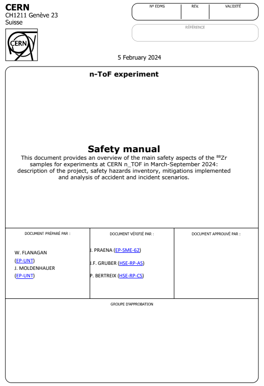


Schedule

Pending INTC:
- Test sample at CERN ~7 MAR.
  - Beam-off tests planned 13-17 MAR.
    - Test DAQ, dead time, etc.
  - Beam-on test planned 18-20 MAR.
    - First 3 days of beam at n_TOF EAR2.

- Shipped by UT NETL Reactor using ALARA Logistics.
- CERN radship request document #830
- Draft DGD and Safety Manual
  - Second draft of Safety Manual to CERN RP.





Week		Notes
8 JAN	▲	Background sample leaves LANL
15 JAN	△	Background sample arrives at UT NETL
22 JAN		
29 JAN		
5 FEB		INTC mtg 7-8 FEB WF at CERN 9 FEB
12 FEB		
19 FEB		
26 FEB	✦	Background sample leaves NETL for CERN
4 MAR	✧	WF at CERN 7-19 MAR
11 MAR		Beam-off tests
18 MAR	❀	Beam start Beam-on tests

	^{nat}Zr tests, chemistry prep, hot cell prep
	Background sample prep
	Beam-off tests (background)
	Beam-on tests (background)
▲	Background sample leaves LANL
△	Background sample arrives at UT NETL
✦	Background sample leaves UT NETL (NLT)
✧	Background sample arrives at CERN (NLT)
❀	Finalize desired ^{88}Zr activity with LANL

18-20 March beam time pending INTC approval.

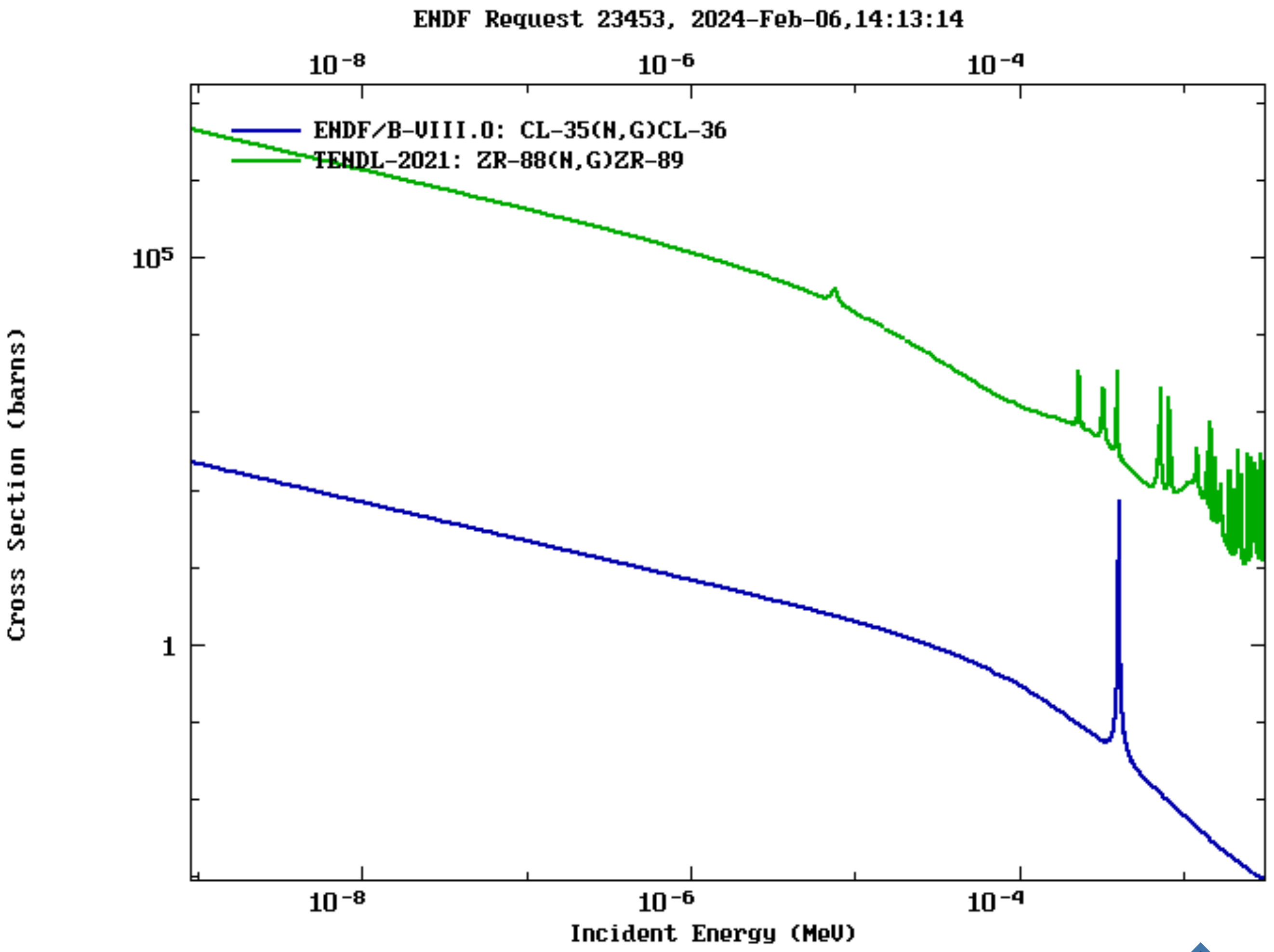
Signal sample collection anticipated *late summer*, pending INTC approval.

…Signal strategy based on background results…

Backup

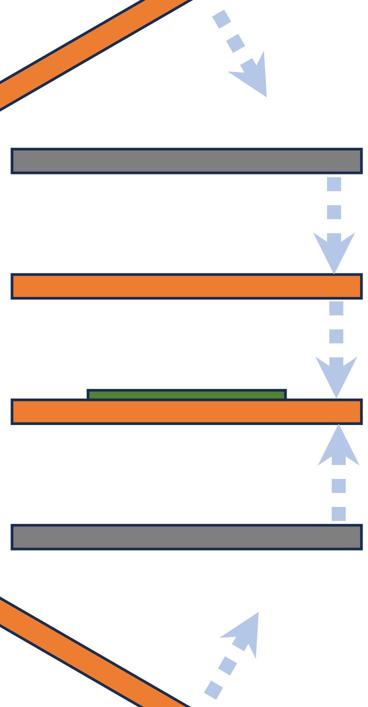
^{35}Cl background

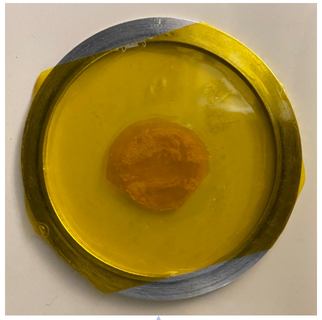
- Sample is $ZrCl_4$ and $ZrOCl_2 \cdot H_20$
- Natural chlorine has a 33 barn cross section
  - ^{nat}Cl is 76% ^{35}Cl with 44 barn cross section
- Though Cl atoms outnumber Zr atoms within sample, the ~800,000 barn cross section of ^{88}Zr mitigates Cl concerns.



Sample Prep (continued)

- $ZrCl_4$/$ZrOCl_2$ evaporated onto Kapton tape
- Additional piece of Kapton tape to conformally trap sample
- Mount to aluminum ring
  - Standard for n_TOF
- Mount to additional aluminum ring on other side to confirm Kapton tape does not separate
- Squeeze aluminum rings together with final piece of Kapton tape.





Initial test with natural zirconium in same initial solution. Note that this sample is not yet conformally wrapped.