EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of the zirconium-88 neutron absorption cross section at EAR2

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Abstract:

This document proposes to measure the neutron capture cross section of ^{88}Zr. This isotope was discovered in 2019 to have the second largest thermal neutron cross section, second only to ^{135}Xe. A pre-print by the DICER collaboration measured this cross section from 0.0253eV to 500eV, confirming the massive thermal cross section but finding a factor of 200 discrepancy in the reported resonance integral. DICER further suggests a resonance at 0.171eV. n_TOF can confirm this resonance and possibly extend the reach of these measurements to determine the contribution of any additional resonances. 5×10^{18} protons on target will allow very high-precision below 5 eV and to maintain the statistical uncertainty below 10% up to 100 eV. This energy range is important to determine the resonance integral. ^{88}Zr has the difficulty of being a radioactive isotope with 83.4 day half-life and a radioactive daughter (^{88}Y), leading to careful consideration of sample preparation and evolution. The Segmented Total Energy Detector (sTED) array within EAR2 has unique speed and acquisition characteristics to minimize dead-time and pile-up effects due to the instrinsic radioactivity of the target sample.

Requested protons: 5×10^{18} protons on target
Experimental Area: EAR2

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Introduction

The thermal neutron capture cross section of ^{88}Zr was recently measured to be 861,000±69,000 barns when a 10 barn cross section was expected [1]. A subsequent study by the same group refined their result to 804,000±63,000 barns with a resonance integral of 2,530,000±280,000 barns. If correct, this would be the largest resonance integral by two orders of magnitude [2]. These measurements state only a thermal cross section as they utilize reactor facilities. The DICER collaboration at LANL has published a pre-print with the first energy-resolved neutron capture measurement on ^{88}Zr. This measurement essentially confirms the thermal neutron absorption cross section (771,000±31,000 barns) while disputing the resonance integral (15,210±670 barns). This measurement further suggests that the large thermal cross section is due almost entirely to a resonance at 0.171eV [4].

Reference Database Tabulations

Due to the recent nature of the ^{88}Zr(n,γ) measurements, the ENDF general purpose libraries have yet to reflect the LLNL and LANL DICER results [5]. The TENDL-2021 database reflects the five order of magnitude increase in total cross section but does not attribute it to a resonance [6]. The JEFF-3.3 database retains the pre-2019 expected cross section and does not claim a resonance in the region reported by recent DICER measurements [7]. As ^{88}Zr resonances have never been measured before DICER, what is seen in these evaluated files are based on statistical assumptions and knowledge of neighboring nuclides. The proposed n_TOF measurement will aim to clarify these reference values as various ^{88}Zr applications begin to be explored.

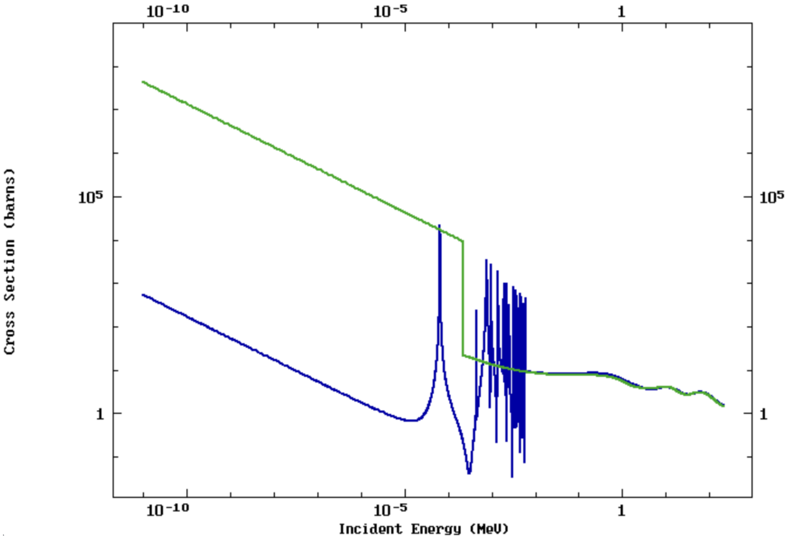

Figure 1: The current ^{88}Zr(n,γ) cross sections from the ENDF database as of this proposal submission. The TENDL-2021 cross section is shown in green while the JEFF-3.3 cross section is given in blue.

^{88}Zr Sample Evolution

^{88}Zr has an 83.4 day half life. It decays through electron capture to ^{88}Y and emits a 393keV gamma ray. ^{88}Y is also radioactive with a 106.6 day half life and decays through electron capture to stable ^{88}Sr, emitting 898keV and 1.836MeV gamma rays with 93% and 99% branching ratios, respectively. As ^{88}Y is an irreducible component within the sample, this activity represents the dominant background. The abundance of ^{88}Y can be easily obtained with the Bateman equation and will realistically represent a sizeable fraction of the ^{88}Zr activity, see Figure 3.

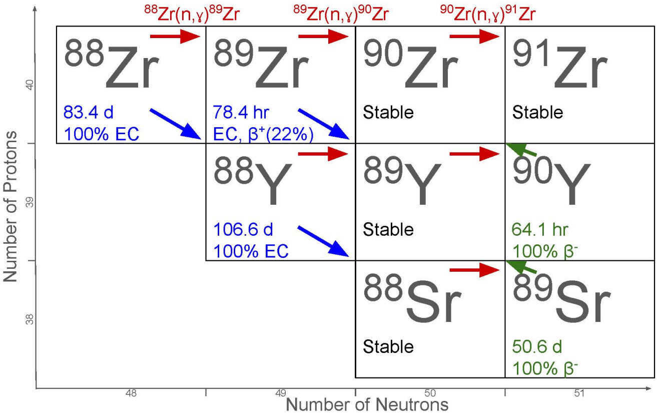

Figure 2: Chart of relevant nuclides in the neighborhood of ^{88}Zr. The blue arrows indicate electron capture and positron emission while the green arrows indicate beta decay and the red arrows indicate neutron absorption.

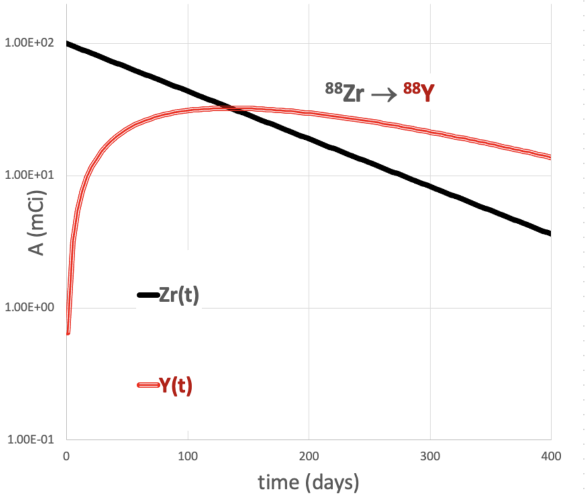

Figure 3: Composition of a sample which is initially 100mCi of pure ^{88}Zr. If it is assumed that n_TOF begins taking data 3 weeks after ^{88}Zr production/separation and collects 5 weeks of data, the ^{88}Zr activity will decrease from 84mCi to 63mCi during beam while the ^{88}Y activity will increase from 12mCi to 24mCi.

Sample Preparation

Due to the large thermal cross section of ^{88}Zr, significant care needs to be taken to avoid self-shielding. Utilizing the most recently reported thermal capture cross section of 771,000 barns [4], the neutron path length is 0.3μm ($\mu = \frac{1}{n\sigma}$) assuming a solid zirconium number density of 4.3×$10^{10}\mu$m^{-3}. The desired thickness is 3nm which would maintain a constant flux throughout the sample to within 1% utilizing the standard absorption probability (P=1-$e^{-x/\mu}$).

The ^{88}Zr sample is provided through the U.S. Department of Energy National Isotope Development Center. In particular, the sample is made by the Los Alamos Isotope Production Facility (IPF) with proton irradiation of a yttrium target (^{88}Y$(p,n)^{88}$Zr). As zirconium is not water soluble, the sample is provided in a 2N HCl solution (pH=-0.3). The sample will be deposited within single-walled carbon nanotube paper [8] at the University of Texas Nuclear Engineering Teaching Laboratory (NETL). The samples will further be encased in mylar foils to maintain sample integrity. After HPGe characterization, the samples will be sent to CERN. NETL frequently sends radioactive samples to Europe and has experience using courier services to avoid operational delays.

Measuring sample uniformity from carbon nanotube filter paper with NIST zirconium standard samples at NETL is planned for the weeks of 8 January and 15 January. After depositing the zirconium acid solution on the filter paper, the zirconium concentration of multiple annuli will be measured with standard neutron activation analysis techniques. A 1mCi ^{88}Zr initial test sample is also arriving to NETL the week of 8 January. This will allow for tests of handling procedures as well as neutron radiography and radioactivity mapping of the final proposed sample configuration.

In the case that the proposed filter paper method proves unsatisfactory, the zirconium can be evaporated on the bottom of a flat quartz vial as was done for the LLNL measurements [1][2] or remain in a strong acid solution as was done by DICER [3][4].

Dead-Time and Pile-Up

n_TOF has previously measured neutron absorption cross sections of radioactive isotopes such as ^{79}Se[9], ^{94}Nb[10], and ^{204}Tl [11]. With 40 times more fluence than in EAR1 arriving in a smaller time interval, leading to an instantaneous flux which is a few hundred times higher than in EAR1, EAR2 is ideal for radioactive samples. The ^{88}Y gamma ray energies nonetheless present unique difficulties. The collaboration has recently published the detection efficiency of individual sTED detectors with ^{137}Cs and ^{88}Y to be 0.20% and 0.46%, respectively, assuming the closest detector (compact ring) configuration [12]. As mentioned previously, the activity of ^{88}Zr and ^{88}Y may realistically evolve from 84mCi to 63mCi and 12mCi to 24mCi, respectively, as the ^{88}Zr decays and the ^{88}Y activity initally builds in accordance with Bateman evolution. Conservatively applying the recently published ^{137}Cs efficiency to ^{88}Zr and the ^{88}Y efficiency to ^{88}Y activity, this gives initial and final count rates in individual sTED of 8.3 c/μs and 8.7 c/μs, respectively. This is a challenging scenario, but is within the 10-16 c/μs maximum count rate to which dead-time and pile-up corrections can be applied [13]. Based on results from a preliminary exposure, detailed in the Run Plan section, the detectors may be placed further away or utilize shielding if necessary.

Sensitivity Study

A preliminary study suggests that a 80mCi ^{88}Zr sample (3.0×10^{16} target atoms) with 5×10^{18} protons on target can maintain a signal to background ratio above unity and statistical uncertainty under 1% up to an energy of 5eV and to maintain the statistical uncertainty below 10% up to 100eV in order to confirm the thermal resonance and extend the DICER results. Of the 5×10^{18} requested protons, 3×10^{18} are for the signal sample and 2×10^{18} are for ancillary (background and normalization) measurements. Though 100mCi of ^{88}Zr is proposed to be produced, 80mCi is utilized to allow for realistic sample preparation and transport times. The signal and background rates are given in Figure 4. The ^{88}Zr(n,γ) rates utilized TENDL-2021 libraries, which include the increased thermal neutron cross section but not the proposed DICER resonance, see Figure 1. The expected background subtracted counts and statistical uncertainty as a function of energy are shown in Figure 5.

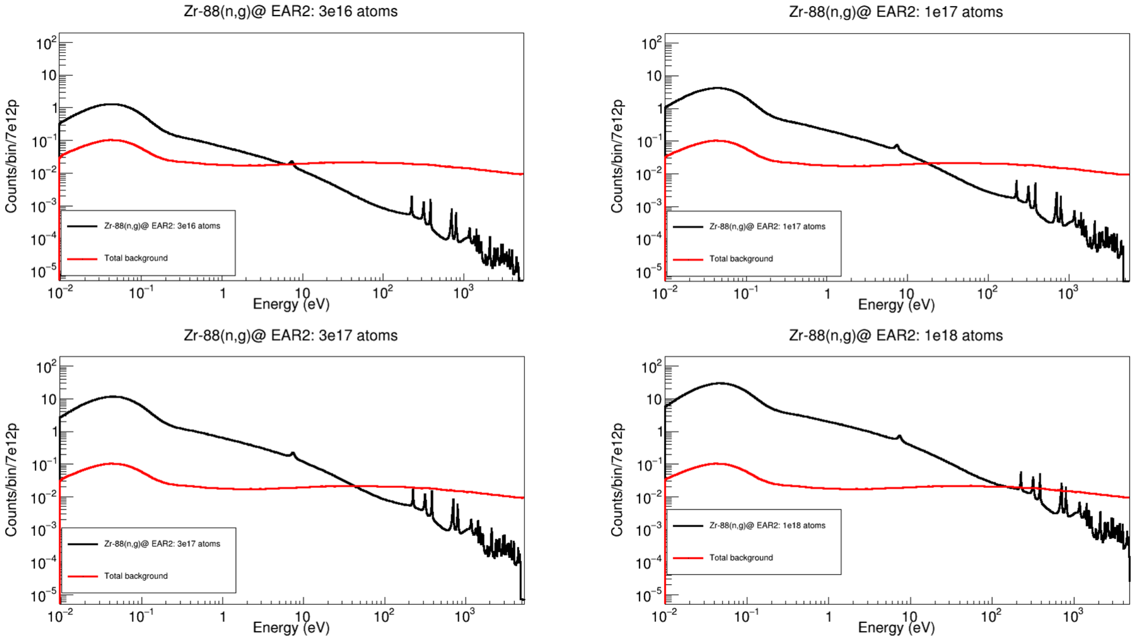

Figure 4: Signal and background rate estimates at EAR2. The 80mCi sample proposed here corresponds to 3×10^{16} atoms, top left. The background due to ^{88}Y is not included. The results shown correspond to 1250 bins per decade.

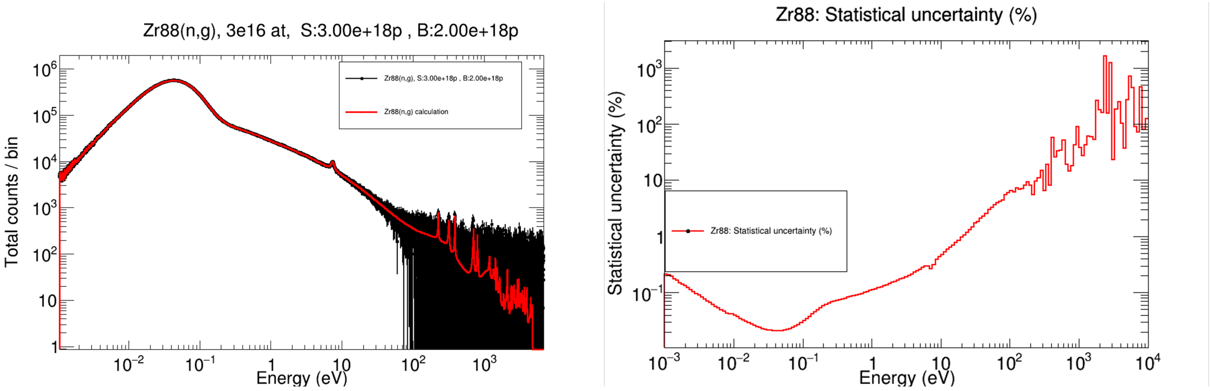

Figure 5: Background subtracted counts (left) and statistical uncertainty (right) for 80mCi ^{88}Zr(n,γ) with 3×10^{18} protons on target for the sample and 2×10^{18} protons on target for ancillary measurements. The background due to ^{88}Y is not included. The results shown correspond to 1250 bins per decade.

Run Plan Strategy

The initial 1mCi ^{88}Zr test sample which arrives to NETL the week of 8 January has a large ^{88}Y activity (4.82mCi) by virtue of being over a year since production. After this initial sample is used to demonstrate sample preparation procedures, it is proposed to arrive to CERN by 1 April. By this point, the ^{88}Zr and ^{88}Y activities will be 0.5mCi and 3.2mCi, respectively. Despite having two orders of magnitude less ^{88}Zr than the signal sample, it will be within an order of magnitude of the maximum expected ^{88}Y activity. Given the higher detection efficiency of ^{88}Y and its role as the leading background, a short run is envisioned in April to diagnose and correct any dead-time or pile-up issues. Additionally, with the help of Geant4 simulations, we will aim to minimize the dead-time and pile-up corrections while keeping the maximum signal-to-background ratio. Furthermore, ^{197}Au will be used for the absolute normalization and carbon and lead samples for the neutron and gamma ray scattering in the sample [12]. The larger ^{88}Zr signal sample will be made explicitly by Los Alamos IPF for this measurement and will be scheduled to minimize ^{88}Y buildup.

Summary

5×10^{18} protons on target are requested for measurement of the ^{88}Zr(n,γ) cross section. With a signal to background rate above unity and sub-percent statistical uncertainty in background subtracted counts through 5eV, there is a high likelihood that this campaign will be able to confirm the preliminary DICER resonance and further expand our knowledge of this 800,000 barn thermal neutron capture cross section.

Summary of requested protons: 5×10^{18} protons.

References

[1] J. A Schusterman et al., Nature 565 (2019), 328-330

[2] J. A Schusterman et al., Phys. Rev. C 103 (2021), 024614

[3] A. V. Matyskin et al., Scientific Reports 13 (2023), 1736

[4] A. Stamatopoulos et al., Pre-print (2023)

[5] D. A. Brown et al., Nuclear Data Sheets 148 (2018), Pages 1-142

[6] O. Iwamoto et al., J. Nucl. Sci. Technol. 60 (2023), 1

[7] A. J. M. Plompen et al., Eur. Phys. J. A56 (2020), 181

[8] ACS Material, Single-Walled Carbon Nanotube Paper Technical Data Sheet

[9] J. Lerendegui-Marco et al., EPJ Web Conf. 279 (2023), 13001

[10] J. Balibrea-Correa et al., arXiv 2301.11199 (2023)

[11] A. Casanovas, Thesis (2020)

[12] V. Alcayne et al., Radiation Physics and Chemistry 111525 (2024)

[13] J. Balibrea Correa, Submitted to NIM A (2023)

Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
EAR2 with sTED detector array	☒ To be used without any modification ☐ To be modified
University of Dallas and University of Texas NETL: • 1mCi ^{88}Zr sample with significant ^{88}Y activity within carbon nanotube filter paper, encapsulated in mylar foils. • 100mCi ^{88}Zr sample within carbon nanotube filter paper, encapsulated in mylar foils.	Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards	Radioactive Sample	☒	See comments below

^{88}Y has a specific activity of 19.8 mR/hr per 1 mCi at 30 cm due to 898keV and 1.836MeV gamma rays with 93% and 99% branching ratios. ^{88}Zr has a specific activity of 1.0 mR/hr per 1 mCi at 30 cm. As such, the ^{88}Y dominates the equivalent dose of both samples.

The first sample will have ^{88}Zr and ^{88}Y activities of approximately 0.5mCi and 3.2mCi, respectively. These abundances will be monitored with HPGe counting at NETL before shipping to CERN and Bateman evolution calculations will be used to project varying sample compostion while at CERN.

Within the second sample, the ^{88}Zr activity will decrease from 84mCi to 63mCi during beam while the ^{88}Y activity will increase from 12mCi to 24mCi. Similar to the first sample, HPGe counting and Bateman evolution will be continually employed to project source activity.