The new Device for Indirect Capture Experiments on Radionuclides at LANSCE: Efforts on measuring the resonance(s) responsible for the extremely large $^{88}$Zr $(n,\gamma)$ cross section

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Abstract. The thermal neutron capture cross section of $^{88}$Zr was recently reported to be the second largest in nature with the largest resonance integral measured. Presumably, these very large values are caused by a resonance or resonances very near thermal energy. Determining their energies and widths, and hence the shape of the cross section away from thermal energies, is useful for applications. The short half-life (83.4 days) and associated large background, renders direct measurements of the neutron capture cross section impossible using current techniques. However, it is possible to measure the total neutron cross section, and hence the resonance properties, using the newly commissioned Device for Indirect Capture Experiments on Radionuclides (DICER) at the Los Alamos Neutron Science Center (LANSCE). Transmission measurements are utilized as a surrogate method to perform capture measurements. The $^{88}$Zr needed for a DICER measurement was produced at the Isotope Production Facility (IPF) and cleanly separated from the production target material. A description of the new instrument, efforts and preliminary results on $^{88}$Zr will be presented.

1 Introduction

The accurate knowledge of neutron capture rates is essential in applications such as radiochemical diagnostics, nuclear forensics and nuclear astrophysics. Several studies have been performed on stable nuclei and a few on long-lived radionuclides [1] using direct techniques, however, studies on $(n,\gamma)$ cross sections on short-lived radionuclides is more challenging.

A number of indirect techniques have been developed and efforts have been devoted to accurately quantify the systematic errors of the results these techniques provide. Frequently used indirect techniques include the Oslo [2] and $\beta$-Oslo methods [3], the $\gamma$-ray strength function method [4, 5] and the surrogate technique [6].

In transmission experiments long sample-detector distances are involved, thus the sample induced backgrounds are appreciably reduced. Neutron transmission measurements, therefore, can be used to tightly constrain $(n,\gamma)$ cross sections [7–9]. The average resonance spacing ($B_n$), average total radiation width ($G_{\gamma0}$), and neutron strength function ($S_0$) can be determined from R-matrix analysis of neutron transmission data and these quantities can be used to tightly constrain the neutron capture cross section using the nuclear statistical model (NSM).

The Device for Indirect Capture Experiments on Radionuclides (DICER) [7, 10–12] was developed at the Los Alamos Neutron Science Center (LANSCE) [13] to perform studies on radionuclides relevant to radiochemical diagnostics, nuclear forensics and nuclear astrophysics [7, 9, 11, 14]. DICER was designed to work best for nuclides having average resonance spacings less than about 20 eV and half lives longer than about 1 month and masses as small as about 1 $\mu$g.

2 The first DICER generation

DICER is located at the Manuel Lujan Jr. Neutron Scattering Center at LANSCE. Currently a source-to-detector distance of 31 m is operational and is shown in Fig. 1. A detailed description of the the DICER instrument is given in Ref. [10].

2.1 Neutron source

Neutrons are produced through spallation from LANSCE’s 800 MeV proton beam, pulsed at 20 Hz [13].
The neutron flux at DICER used to perform the $^{88}$Zr study, is shown in Fig. 2 and spans from 0.1 meV - 100 keV in incident neutron energy. The shown flux was measured with the ORELA-style detectors at 31 m from the spallation source. For more details on the neutron production and neutron spectrum, see Ref. [10]. Currently the neutron source viewed by DICER is being upgraded to have higher flux at keV energies and above and better resolution [15].

![Figure 1. The DICER instrument at LANSCE. Figure not to scale.](image)

2.2 Collimation system

The collimation system allows the simultaneous sample in and sample out measurements which is important when studying small-scale samples. Briefly, DICER utilises two non-parallel neutron beam lines, which converge at the same spot on the liquid hydrogen moderator.

The first collimation element, the rotating beam blocker, is a brass cylinder (30.5 cm in length, 21.5 cm in diameter), is installed at 14.35 m from the neutron source. An inner cylinder (10 cm diameter) is inserted in the cylindrical shell and is equipped with three 0.8 cm diameter holes. These holes provide four beam configurations, used for background measurements: (i.) two beam lines, (ii.) no beam lines, (iii.) a beam line to the left and (iv.) a beam line to the right. The inner rotating cylinder is “stepped” with two different diameters, to eliminate streaming paths between the inner and outer parts.

Further downstream, at a 14.85 m distance from the spallation target, a brass right rectangular prism, the binocular collimator (30 × 15 cm), is installed. This collimation element serves both as a collimator and sample holder and provides two tiny beam lines, 1 mm in diameter. The binocular collimator is designed to house a cylindrical sample enclosure of 1.5 cm in length and 1 cm in diameter.

Lastly, 18.5 m from the neutron source, a rectangle brass collimator, the aperture stop (30 × 15 cm), is installed to clean up the beam penumbra. The collimation system of DICER, is shown in Fig. 1).

2.3 Detectors

The default detection system of DICER consists of two $^6$Li-glass disks, 10 cm in diameter and 4 mm in thickness. Each glass is viewed by two photomultipliers which are perpendicular to the neutron beam. The detectors are installed roughly 31 m from the neutron source and are shown in Fig. 1.

3 The $^{88}$Zr experiment

Shusterman et al. ([Refs. [16, 17]]) measured the $(n, \gamma)$ cross section of $^{88}$Zr at the thermal point and reported an enormous 0.861(69) Mb value. Most probably, a resonance near thermal energies is responsible for this large cross section, and DICER can provide point-wise data over a large neutron energy range to confirm or reject that assumption.

3.1 Production of $^{88}$Zr

The DICER approach on producing radionuclides and fabricating radioactive samples, relies on the collaboration with the Isotope Production Facility (IPF)[18, 19]. An example that demonstrates the successful synergy between DICER and the IPF, is the production of the $^{88}$Zr radionuclide and the first exploratory experiment of two 66 ng (∼1 mCi) and a 33 ng (∼0.5 mCi) liquid sample. The liquid format ensures that $^{88}$Zr is uniformly distributed. At those small scales, fabricating a uniform solid sample would have been extremely difficult. It has to be noted that only one of the 66 ng samples will be discussed.

29 g of an yttrium metallic target, (2.90 mm in length, 46 mm in diameter), encapsulated in an aluminum enclosure, was irradiated at the IPF for a total of ∼9 hours, at an average proton current of 96.1 µA. Roughly 8 GBq 88Zr were isolated and then converted to 1.9 mol/L DCl. More details on the $^{88}$Zr preparation process are given in Refs. [9, 20].
3.2 The $^{88}$Zr samples

The liquid $^{88}$Zr sample was enclosed in a cylindrical tungsten (W) canister, with an inner diameter of 1.2 mm and was sealed with two spheres made out of lead (Pb), which is shown in Fig. 3. The challenging task of precisely dispensing the right volume of the liquid sample (66 ngr $^{88}$Zr dissolved in 8 µL DCl) through the small opening in the canister, was made possible through the design of a filling station, shown in Fig. 4. The station was remotely operated in a hot cell environment and a camera ensured a visual on-the-fly inspection of the filling operation.

The two Pb spheres, shown in Fig. 3, acted as seals and ensured that the liquid radioactive sample will not leak out of the canister. A hand press, shown in Fig. 5, operated remotely inside the hot cell, was used to swage the Pb spheres into the ends of the W canister, forming a hermetically tight seal.

3.3 Preliminary results

The $^{88}$Zr sample was irradiated at DICER for a total of three days. For one of the three days, special beam conditions were requested: the duty cycle of the accelerator was reduced from 20 Hz to 1 Hz to avoid wraparound effects. To check the 1-Hz measurement and analysis systems, a short (∼20 min.) run with a metallic $^{95}$Mo sample was made. As shown in Fig. 6, the preliminary DICER data can be reproduced using ENDF/B-VIII.0 resonance parameters through the R-Matrix code SAMMY.

In addition, the “sample out” had the same volume of DCl used to dissolve the $^{88}$Zr sample. A transmission measurement using this sample out as a “sample in” was performed and a blank W can with Pb windows was used as the sample out. The results are in excellent agreement with ENDF/B-VIII.0.

The DICER data on $^{88}$Zr confirm the enormous $(n, \text{tot})$ cross section, as shown on the transmission plot, in Fig. 7. In fact, the $(n, \text{tot})$ cross section at the thermal point is of the order of a few Mb and appears to be appreciably larger than the $(n, \gamma)$ 1 cross section. The blue triangle in Fig. 7 corresponds to the transmission expected at thermal energy for a cross section equal to that reported by Shusterman et al. The red curve in Fig. 7 corresponds to a single-resonance fit to the DICER and Shusterman data using the R-Matrix code SAMMY [21]. That is, the resonance parameters
fit both the DICER transmission and the Shusterman capture.

Finally, additional DICER measurements with the same $^{88}$Zr sample, after most of the $^{88}$Zr has decayed away, are planned for the 2022 LANSCE run cycle. This will check if contaminants could have contributed to the transmission measured during the 2021 run cycle. We also plan new measurements with fresh, larger samples during the 2022 run cycle.

4 Conclusion

DICER is a new neutron transmission instrument, that was recently commissioned at LANSCE to study neutron capture on radionuclides. The first exploratory measurement of a radioactive sample took place recently with the production of a 66 ng, liquid $^{88}$Zr sample. The preliminary results indicate that the $(n, \text{tot})$ cross section is of the order of a few Mb at thermal energy. A sub-thermal resonance, appears to be responsible for this large cross section. Additional measurements are planned to be performed at the end of 2022.

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