DICER: A new instrument at LANSCE to constrain neutron capture rates on radionuclides

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Abstract. With very few exceptions, direct measurements of neutron capture rates on radionuclides have not been possible. A number of indirect methods have been pursued such as the surrogate method [1], the γ-ray strength function method [2, 3], the Oslo method [4–7] and the β-Oslo method [8]. Substantial effort has been devoted to quantify the usually large systematic errors that accompany the results from these techniques. A new instrument has been developed at the Los Alamos Neutron Science Center (LANSCE) to provide more accurate data on several radionuclides relevant to nuclear criticality safety, radiochemical diagnostics, astrophysics, nuclear forensics and nuclear security, by measuring the transmission of neutrons through radioactive samples and studying resonance properties. The Device for Indirect Capture on Radionuclides (DICER) [9–13] and associated radionuclide production at the Isotope Production Facility (IPF), both at LANSCE, as well radioactive sample fabrication, have been under development the last few years. A description of the new apparatus, data on a few mid-weight stable isotopes and efforts on radionuclide measurements will be presented.

1 Introduction

The Device for Indirect Capture Experiments on Radionuclides (DICER) is an instrument developed at the Los Alamos Neutron Science Center (LANSCE) with the aim of addressing the challenging task of measuring neutron capture cross sections, particularly for short-lived and rare radionuclides. Accurate neutron capture data are crucial for numerous fields including nuclear security, astrophysics, radiochemical diagnostics, and nuclear forensics, yet direct measurements on many radionuclides are often impossible due to their inherent radioactivity.

DICER employs an indirect technique based on neutron transmission measurements through small radioactive samples. This method overcomes the limitations of traditional gamma-detection techniques by utilizing a high-precision dual-beam collimation system, which significantly reduces sample size requirements and measurement times. By focusing on neutron transmissions and resonance interactions, DICER enables determinations of neutron capture cross sections, even for very small samples on the order of μg to mg, and for neutron energies up to a few keV.

The development and deployment of DICER mark a significant advancement in neutron physics, providing the ability to measure the neutron transmission and extract capture cross sections for approximately 50 radionuclides. Initial tests have demonstrated the instrument's capability, including successful measurements on radionuclides such as ^{88}Zr, and ongoing efforts are focused on expanding its use to other isotopes. As the production of radioactive samples remains a bottleneck, further development is directed towards enhancing sample fabrication techniques and refining DICER's measurement precision to support a wider array of radionuclide studies.

2 Description of the apparatus

DICER [9–12] is located on flight path 13 (FP13) at the Manuel Lujan Jr. Neutron Scattering Center at LANSCE and is shown in Fig. 1. The apparatus has two measurement stations, one at a source-to-detector distance of 31 m and another at 64 m. The former is the main measuring station whereas the latter is currently under development.

2.1 The neutron source

Neutrons are generated by spallation reactions, when an 800 MeV proton beam from LANSCE's linear accelerator impinges on a tungsten-water assembly. The proton beam is nominally pulsed at 20 Hz and has a nominal width of 125 ns, however different duty cycles (i.e. 1 Hz) and proton widths (i.e. 10 ns) are possible upon request.

The target-moderator assembly, shown in Fig. 2 consists of three separate tungsten pieces that serve separate types of science. The upper tier target is used for nuclear physics experiments and consists of three tungsten disks, 10 cm in diameter, 3.8 mm in thickness that are separated by 1 mm, as shown in Fig. 3.

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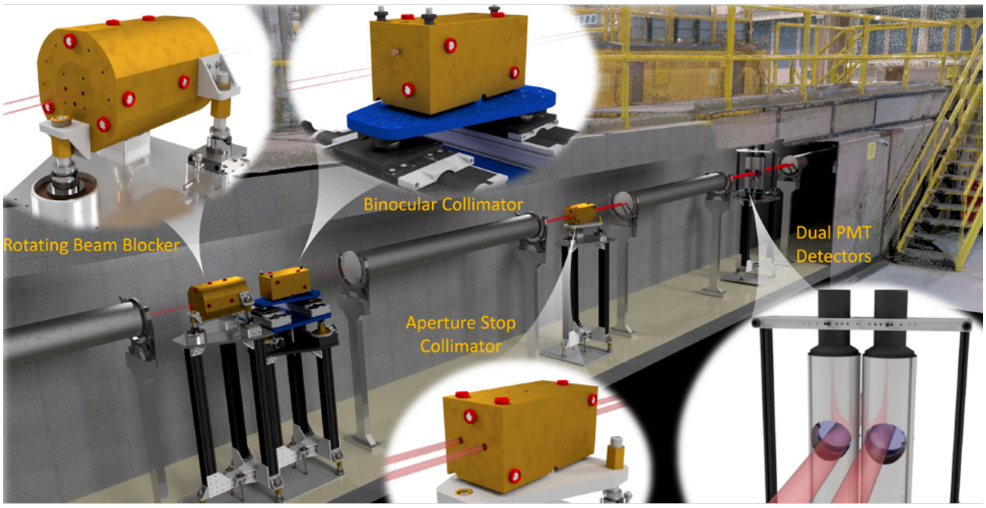

Figure 1. The Device for Indirect Capture Experiments on Radionuclides (DICER) employs a unique binocular collimation system. See text for details.

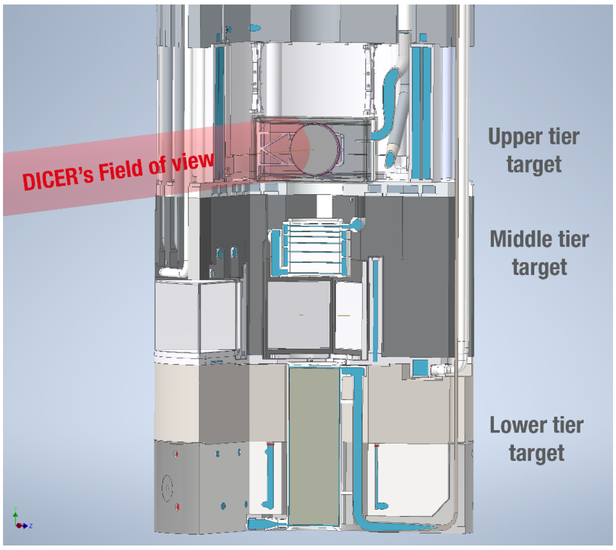

Figure 2. The new moderated neutron source at LANSCE, termed Mark-IV [14]. See text for details.

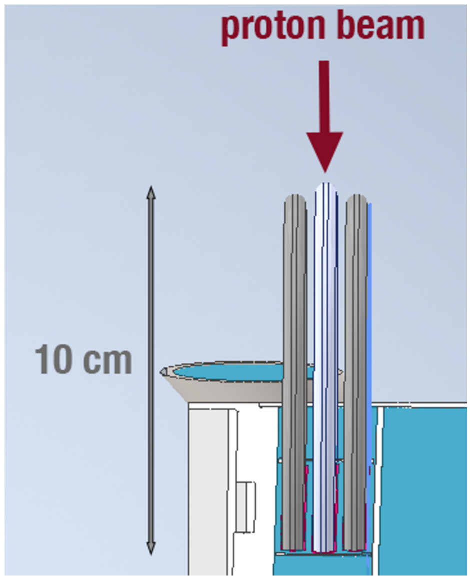

Figure 3. Sideview of the upper tier portion of the Mark-IV target assembly. The three tungsten disks, are emerged in water and have a diameter of 10 cm, a thickness of 3.8 mm and are separated by 1 mm.

Mark-IV replaced the previous generation neutron spallation target, termed Mark-III, which resulted in a shifted towards higher neutron energies neutron spectrum, as shown in Fig. 4.

2.2 Filter box

A filter box is positioned 8 m from the spallation target. This box contains five remotely controlled paddles made of various materials: cadmium (to absorb neutrons bellow $\sim$ 0.5 eV), lead (to reduce gamma ray background), bismuth (for black resonance measurements), and polyethylene (to scatter neutrons bellow $\sim$ 200 keV as shown in Fig. 5). These paddles filter the beam and reduce unwanted background radiation during measurements. The thicknesses of the paddles are shown on table 1.

Table 1. List with DICER paddles and thicknesses. The top rows correspond to upstream positions while bottom rows correspond to downstream paddles.

Position	Material	Thickness (cm)
1	Cadmium	0.08
2	Lead	0.32
3	Bismuth	2.54
4	Polyethylene	2.54
5	Polyethylene	2.54

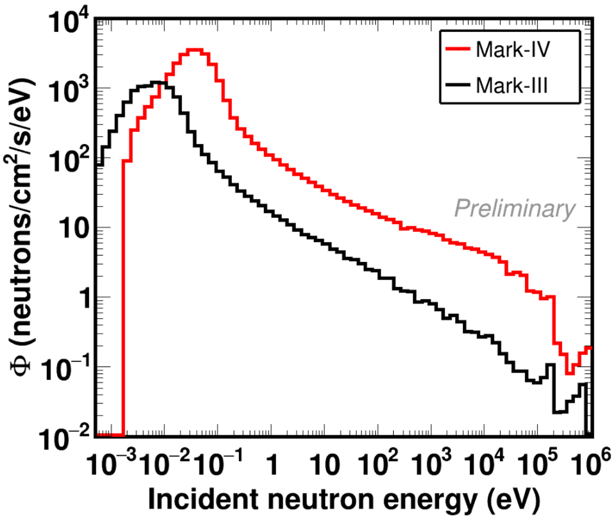

Figure 4. Comparison between the Mark-III and Mark-IV neutron flux. The Mark-IV flux is preliminary.

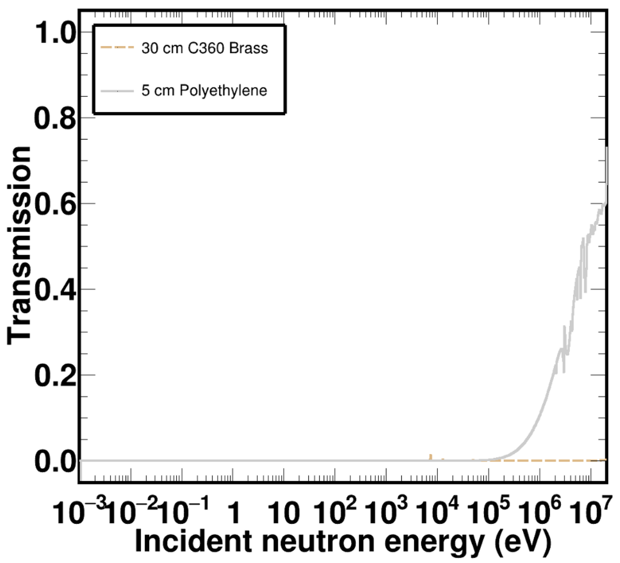

Figure 5. Transmission through 30 cm of brass and 5 cm of polyethylene, which corresponds to one of the three collimator and paddles, respectively.

2.3 Collimation system

DICER's collimation system is one of its most important components, allowing it to perform simultaneous sample-in and sample-out measurements without moving the radioactive sample during the experiment. This innovative binocular mode of operation minimizes the required sample size and shortens measurement times by half compared to traditional methods. The collimation system includes three key components: the rotating beam blocker, the binocular collimator, and the aperture stop.

The rotating beam blocker, located 14.35 m from the neutron source, is a brass cylinder equipped with three 0.8 cm diameter holes arranged in an L-shape pattern. By rotating the inner cylinder, different beam configurations can be selected, allowing for two beam channels, one beam channel (left or right), or complete blockage of the neutron beam. This system is remotely controlled through a programmable logic controller (PLC).

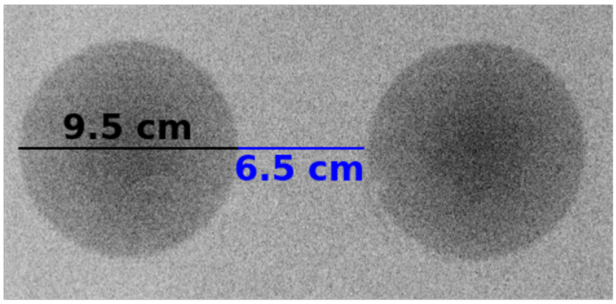

Figure 6. The collimation system of DICER, provides two sharp beam spots at 32 m from the spallation source, 9 cm in diameter which are separated by 6.5 cm.

The binocular collimator, located 14.85 meters from the neutron source, serves both as a collimator and sample holder. It is split into two halves, allowing for precise positioning of the sample, which is critical for accurate measurements. The grooves through which the neutrons pass are only 1 mm in diameter and are not parallel, but converge to the same area on the neutron source.

Finally, the aperture stop, located 18.5 m from the spallation target, refines the beam by removing any unwanted halo effects. This ensures that the beam spots are well-defined when they reach the detectors, shown in a neutron image plate in Fig. 6.

2.4 Detectors

DICER uses three types of detectors to measure neutron transmission: ORELA-type, $\chi - \nu$-type and the Large Area Picosecond Photo Detector (LAPPD) detectors, all based on ^{6}Li-doped glass scintillators. In the ORELA-type detectors, 10 cm diameter glass disks are positioned perpendicular to the neutron beam, and the scintillation light generated by neutron interactions is collected by two photomultiplier tubes (PMTs) on either side of the disk. This setup minimizes neutron interactions with the PMT thus reducing beam-induced background noise, though the light collection efficiency is not optimal. To mitigate this, the interior of the detector is lined with highly reflective material to maximize light collection.

The $\chi - \nu$-type detectors place the glass scintillator in direct contact with the PMT window, improving light collection efficiency. However, this design exposes the PMT to direct neutron interactions, which increases background noise.

The PMTs in both detectors are equipped with anti-magnetic shielding to prevent gain shifts due to changes in the magnetic field around the experimental area.

Finally, an imaging array, the LAPPD detector [13] is currently under development. The LAPPD is a multichannel plate (MCP)-based detector equipped with picosecond-level timing and millimeter-scale spatial reso-

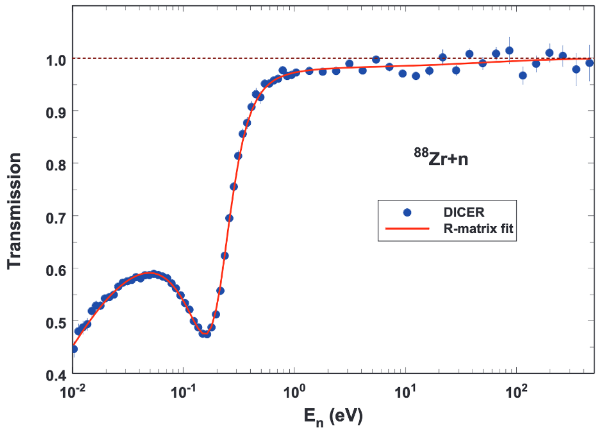

Figure 7. A resonance at 0.17 eV was observed for the first time and it is responsible for the large neutron capture cross section of ^{88}Zr.

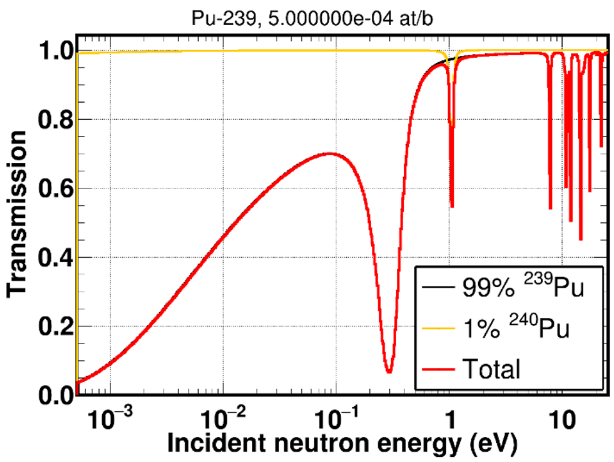

Figure 8. Neutron transmission of a fictional ^{239}Pu sample that contains 1% ^{240}Pu. The percentile refers to atoms. The contribution of ^{240}Pu to the total transmission around 1 eV is significant despite the small atomic fraction.

lution capabilities. It is coupled with a ^{6}Li-glass scintillator, which converts neutron interactions into detectable light. The LAPPD has an 8*x*8 resistive anode array, providing an active area of 20*x*20 cm^2, and can capture beam images with mm spatial resolution. This detector can also perform time-of-flight measurements by capturing neutron arrival times, making it a powerful tool for imaging and detecting neutron transmissions at DICER.

3 Some experimental results

Although DICER has been operational since 2022 several nuclei have been studied such as ^{nat,191,193}Ir, ^{88}Zr, ^{147,149}Sm, ^{239}Pu, ^{95}Mo, ^{nat}Cu, ^{197}Au, ^{nat}Cd, ^{nat}Gd, ^{209}Bi, ^{nat}Y and ^{88}Y. Furthermore, the following measurements are foreseen to be executed in the near future: ^{63}Cu, ^{239}Pu, ^{133}Cs, ^{88}Y, ^{151}Sm, ^{73,74}As and ^{171}Tm.

3.1 ^{88}Zr+n

^{88}Zr ($t_{1/2}$ = 83.4 days) was recently reported to have the second largest neutron-capture cross section at the thermal point (804(63) kb) [15] and the largest resonance integral (2.53(28) Mb). [16].

In collaboration with the Isotope Production Facility (IPF) within LANSCE, we fabricate a liquid ^{88}Zr sample of about 1 μ g and used DICER to discover a resonance at 0.17 meV, shown in Fig. 7 which is responsible for the large neutron capture cross section.

3.2 ^{239}Pu+n

Although DICER is mainly developed to perform studies on radionuclides, there are cases where measurements are needed in rare-to-find, material. Such a measurement is the ^{239}Pu+n measurement, where high accuracy data are needed below 20 eV for nuclear criticality purposes. The main reason is the usual ^{240}Pu contamination in ^{239}Pu sample. As shown in Fig.8, even 1% of ^{240}Pu is enough to dominate the transmission around 1 eV.

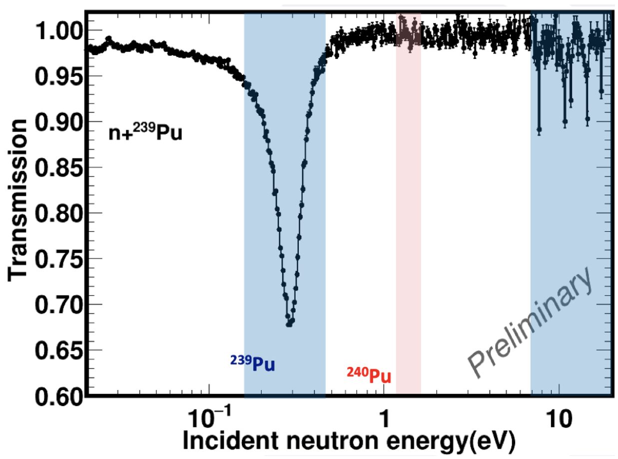

Figure 9. Preliminary DICER transmission of a 99.94% ^{239}Pu sample. The red shaded area around 1 eV, highlights the sample does not contain any significant amount of ^{240}Pu.

To perform such a sensitive measurement the ^{240}Pu atomic fraction had be smaller than 0.1%. Such a material was identified with the Los Alamos National Laboratory, however, only 100 mg were available. This amount of material is more than enough for a DICER measurement, therefore a liquid $^{239}PuCl_4$ sample in 6 mol/L DCl in D_2O was fabricated and irradiated at DICER. The preliminary transmission is shown in Fig. 9 where there is no significant signature of ^{240}Pu. The data are currently under analysis.

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