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A novel method to assay special nuclear materials by measuring prompt neutrons from polarized photofission

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ABSTRACT

A novel method of measuring the enrichment of special nuclear material is presented. Recent photofission measurements using a linearly polarized γ-ray beam were performed on samples of ^{232}Th, ^{233,235,238}U, ^{237}Np, and ^{239,240}Pu. Prompt neutron polarization asymmetries, defined to be the difference in the prompt neutron yields parallel and perpendicular to the plane of beam polarization divided by their sum, were measured. It was discovered that the prompt neutron polarization asymmetries differed significantly depending on the sample. Prompt neutrons from photofission of even–even (non-fissile) targets had significant polarization asymmetries ($\sim$0.2 to 0.5), while those from odd-A (generally fissile) targets had polarization asymmetries close to zero. This difference in the polarization asymmetries could be exploited to measure the fissile versus non-fissile content of special nuclear materials, and potentially to detect the presence of fissile material during active interrogation. The proposed technique, its expected performance, and its potential applicability are discussed.

1. Introduction

The general problem of non-destructively measuring the enrichment of Special Nuclear Material (SNM) or attempting to identify the presence of shielded SNM is sufficiently complex that many potential solutions have been developed. Generally, these solutions are divided into passive or active measurements. In many cases, solutions which work for one subset of SNM do not work for other types of SNM.

In passive measurements, natural emissions from a sample are used to identify the presence of SNM or measure the enrichment of SNM. These natural emissions could be neutrons or γ rays from spontaneous fission, or α particles and γ rays from α-decay. ^{240}Pu has a relatively short spontaneous fission half-life, and therefore the passive measurement of prompt neutrons is sufficient to discover samples of ^{240}Pu, but the spontaneous fission half-lives of ^{235,238}U are sufficiently long such that this technique is less practical [1]. A variety of passive techniques have been developed to use γ rays to measure the enrichment of ^{235,238}U [2–5] and ^{239,240}Pu [6], but it may be difficult to use these techniques to measure the enrichment of samples composed of ^{233}U and ^{232}Th because of the long half-life of ^{232}Th and the lack of γ rays emitted from its decay. Finally, passive measurements of α-particles could be used to assay samples of ^{233,235,238}U [7] and ^{232}Th, but this technique is more difficult to use with ^{239,240}Pu since their α-particle decay energies are very close, though there is progress on unfolding measured spectra to assay plutonium samples [7,8]. The use of passive measurements of α particles is also complicated by the fact that the α particles will straggle in the sample itself and in any casing or shielding, so sample preparation is generally required.

In active measurements, a beam of particles, generally neutrons or high-energy γ rays [9], is used to induce nuclear reactions in a sample. Active interrogation with thermal neutrons, either moderated at the neutron generator or within the sample, can be particularly sensitive to the presence of fissile nuclear material. In this technique, one would detect prompt neutrons (such as with the differential die-away technique [10]), delayed γ rays [11], or delayed neutrons [12]. However, active interrogation with thermal neutrons is not sensitive to the presence of non-fissile material such as ^{238}U or ^{232}Th, and therefore cannot be easily used to assay the enrichment of SNM. Fast neutron beams are sensitive to the presence of fissile and non-fissile materials, and by detecting delayed γ rays, the enrichment of a sample may be deduced due to small differences in the mass distribution of the daughters following fission, though this technique is still under development [13].

Active measurements with γ-ray beams can also be used to measure the enrichment of a sample. Delayed γ rays from fission

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could be used to measure the enrichment of samples [14] in a technique similar to that of Ref. [13]. Nuclear Resonance Fluorescence (NRF) could be used to quantify the amount of each nuclide present in a sample of SNM [15,16]. The NRF technique seems to be the most versatile to distinguish different isotopes in SNM, though it is still under development.

A new technique is presented in this paper that involves detecting prompt neutrons from photofission induced by a linearly polarized γ-ray beam. This technique is sensitive to both fissile and non-fissile fissionable material, and is capable of measuring the enrichment of SNM. In principle, this technique could measure the enrichment of any two-component subset of fissile+non-fissile SNM. Because it relies on interrogation with a γ-ray beam, which can penetrate low-density shielding, and detection of fast prompt neutrons, which can penetrate high-density shielding, it is expected that this technique could in principle be used to detect and identify shielded SNM.

2. Description of the experiment

Details of the experiment and the analysis are described elsewhere [17,18], therefore only a short description is provided here. A nearly 100% linearly polarized, high-intensity ($\sim 5\times 10^6 \gamma/\mathrm{s}$), pulsed γ-ray beam was generated using the High Intensity γ-Ray Source (HIγS) located at Duke University [19–21]. The γ-ray beam was created by colliding a high-power Free Electron Laser (FEL) beam with an electron beam in a storage ring. γ-ray beams of energies between 5.3 and 7.6 MeV were generated. The Full-Width at Half Maximum (FWHM) at each γ-ray beam energy was approximately 3%. The γ-ray beam was polarized in the horizontal plane.

The γ-ray beams were incident on one of seven actinide targets: ^{232}Th, ^{233,235,238}U, ^{237}Np, or ^{239,240}Pu. Prompt fission neutron yields were measured using an array of 12–18 liquid scintillator detectors filled with BC-501A. These detectors were placed in the horizontal plane (azimuthal angles of 0° and 180°) and the vertical plane (azimuthal angles of 90° and 270°). When using all 18 detectors, four were placed at each scattering angle of 55°, 90°, and 125° and two were placed at each scattering angle of 72°, 107°, and 142°. Pulse shape discrimination was used to eliminate γ-ray backgrounds. The energies of detected neutrons were determined based on their time-of-flight, and neutron energy thresholds were set at $E_n = 1.5$ MeV to ensure that all detected neutrons came from fission and not the (γ, n) reaction. Possible neutron backgrounds, due to either delayed fission neutrons from earlier events or to ^{27}Al(α,n) neutrons from α-particles in the actinide sample interacting with the casing of the sample, were subtracted from the prompt neutrons by measuring the out-of-time yield with respect to the beam pulse. The resulting prompt fission neutron yield was integrated over all neutron energies above the energy threshold, and the polarization asymmetry Σ:

$$\Sigma(\theta) = \frac{1}{P}\frac{Y(\theta,\phi=0^\circ)+Y(\theta,\phi=180^\circ)-Y(\theta,\phi=90^\circ)-Y(\theta,\phi=270^\circ)}{Y(\theta,\phi=0^\circ)+Y(\theta,\phi=180^\circ)+Y(\theta,\phi=90^\circ)+Y(\theta,\phi=270^\circ)} \quad (1)$$

was measured as a function of scattering angle θ and beam energy. The fractional beam polarization P was assumed to be 1.0. Experimentally, it was determined that the polarization asymmetry was maximal at a scattering angle of 90°, which is in agreement with theory [18] and with previous measurements of unpolarized prompt neutron angular distributions [22]. The polarization asymmetries were corrected for instrumental asymmetries, finite-size effects, and the contamination of each sample with other isotopes.

3. Experimental results

Fig. 1 shows the experimental results for the polarization asymmetry at a scattering angle of 90°. The error bars indicate the statistical uncertainties, and the systematic uncertainties are typically smaller than the size of the data points and are not shown. The polarization asymmetries are very different for the even–even and the odd-A actinides. The even–even actinides (^{232}Th, ^{238}U, and ^{240}Pu) generally show large polarization asymmetries, particularly at the lower beam energies studied, where the asymmetries are as large as 0.45. However, the odd-A actinides generally show polarization asymmetries much closer to zero. Above beam energies of 6 MeV, the odd-A actinides show polarization asymmetries nearly consistent with zero.

The physical origin of these polarization asymmetries in the prompt neutrons can be traced back to underlying fragment polarization asymmetries. It has been well established that the angular distributions of the fission fragments are highly anisotropic for even–even actinides [23] but mostly isotropic for odd-A actinides [24–26]. This difference in the fragment angular distributions is expected to be due to the difference in target spins of even-even (spin 0) isotopes and odd-A (spin > 0) isotopes, but

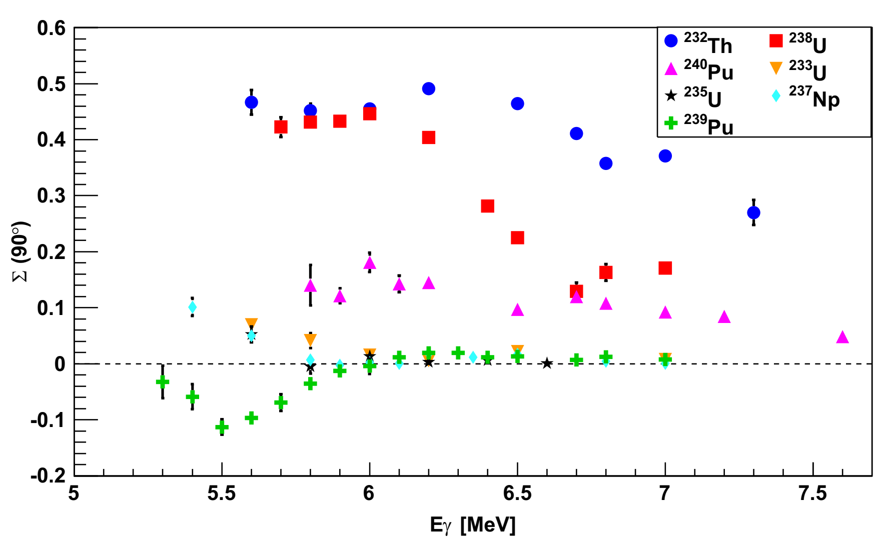

Fig. 1. The measured polarization asymmetry Σ at a scattering angle of 90° is shown as a function of beam energy E_γ. The error bars indicate the statistical uncertainties. Uncertainties not shown are smaller than the size of the data points.