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Nuclear Resonance Fluorescence and Isotopic Mapping of Containers

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Abstract. National security programs have expressed interest in developing systems to isotopically map shipping containers, fuel assemblies, and waste barrels for various materials including special nuclear material (SNM). Current radiographic systems offer little more than an ambiguous density silhouette of a container's contents. In this paper we will present a system being developed at LLNL to isotopically map containers using the nuclear resonance fluorescence (NRF) method. Recent experimental measurements on NRF strengths in SNM are discussed.

Keywords: Nuclear Resonance Fluorescence, NRF, FINDER, inelastic gamma ray scattering, transition widths, gamma ray transitions, scanning, national security
PACS: 23.20.Lv, 25.60.Dz, 25.20.Dc, 27.90.+b

INTRODUCTION

Many efforts are underway to develop a system that can identify and characterize illicit materials such as nuclear and radiological materials. Some systems use a passive technique to detect the unique radioactive signatures of materials of interest. Other systems use an active approach with an external source to stimulate materials in containers. Typically these are photon sources or neutron sources. (See Ref. [1-5].)

NRF provides a unique approach to identify materials on an isotopic basis. NRF is a phenomenon in which a photon of an appropriate energy excites a nucleus to a higher level that subsequently decays to a lower lying level by emitting a gamma ray of energy equal to the energy difference between the resonance level and lower-lying level. This process is analogous to atomic fluorescence where photons excite an atomic-bound electron to a higher atomic energy level.

Researchers at Lawrence Livermore National Lab (LLNL) are developing a system that uses a tunable quasi-monoenergetic photon source, also under development. The system called FINDER (Fluorescence Imaging in the Nuclear Domain with Extreme Radiation), uses a transmission technique to identify specific material. In such a setup, the source sits on one side of the container and the detector array sits on the opposite side (See Fig. 1). The detectors are oriented so that they face a witness foil made of a small sample of material of interest. The witness foil is arranged in the beam path at 0-degrees. During operations, the source produces a spectrum of photons around the resonant energies of the material being sought (i.e. witness foil). If the material is present in the container, the photons on resonance will be absorbed and fluoresce into four-pi. This causes a deficit of photons in the source spectrum at the resonant energies. The resultant spectrum then impinges on the witness foil. Since the flux of resonant photons is reduced in the resultant spectrum, the magnitude of NRF scatter from the witness foil is reduced. Conversely, if material matching the witness foil is not in the container, then the resultant spectrum is only attenuated by the material through which it passes through the container. Therefore, either the photon flux will scatter from material within the container or from the witness foil.

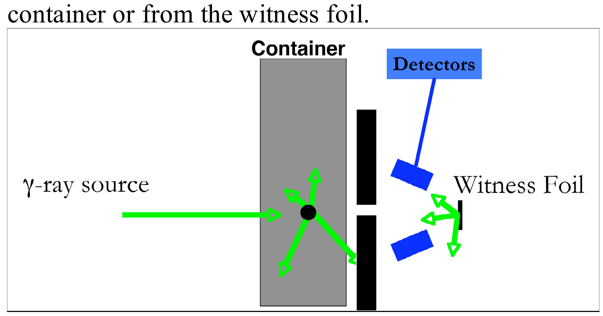

FIGURE 1. Schematic of FINDER. See text for details

In light of developing systems to use NRF to scan containers, searches for NRF states in materials of interest must be performed. At LLNL, we are collaborating with various industrial groups and academic institutions to measure NRF states in isotopes such as ^{239}Pu. NRF measurements of ^{239}Pu will be the focus for the remainder of this paper..

EXPERIMENTAL METHOD

NRF measurements of ^{239}Pu were performed at the High Voltage Research Lab (HVRL) at MIT using a bremsstrahlung beam with endpoint energy of 2.8 MeV. (The setup was designed and constructed by Passport Systems Inc. for their Homeland Security efforts.) Two Pu targets, provided by LLNL, were used where each Pu target was ~93% enriched ^{239}Pu and a mass of 3.5 grams. Each Pu target was encased in 25 grams of Nitronic-40. The two targets were arranged in a side-by-side configuration. The gamma rays were measured with two HPGe detectors situated at 120-degreees with respect to the beam axis and focused on the Pu targets. A thin disk of aluminum was placed in front of each target to determine the NRF strengths in Pu relative to well-known NRF states in Al.

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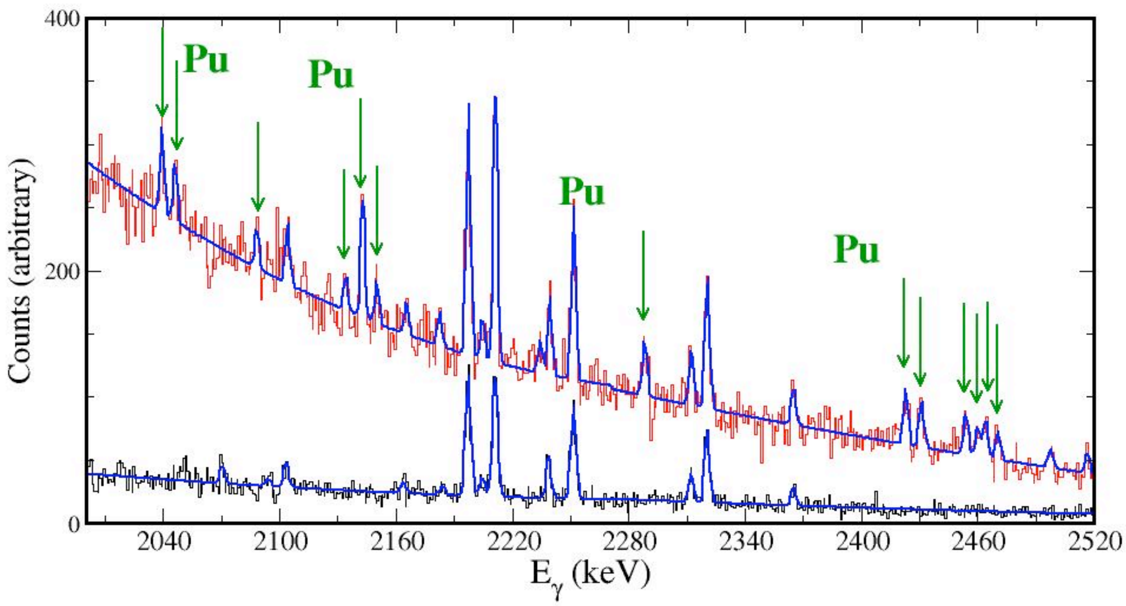

FIGURE 2. Histograms of Pu + Holder data (upper) and Holder only data (lower). Highlighted with arrows are the peaks assigned to Pu transitions from NRF

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RESULTS

The results of the NRF measurements of ^{239}Pu are shown in Figure 2. The upper histogram is data taken from the holder and Pu material and Al disk. The lower histogram is data from the holder and Al disk only. Because of low statistics a smooth curve is overlayed on both histograms to aid the eye.

Because ~93% of the Pu target mass is A=239, it is most likely the case that the transitions are from ^{239}Pu and not from the other constituents. The widths estimated in Table 1 would have to be 16 times larger if they were ^{240}Pu (~80 meV) and even larger for other isotopes (>200 meV). This is unrealistic given that other actinides are ~15-meV(Ref. [6,7]). However, more measurements should be done to verify this.

A proposed level at 2431.7(3)~keV is populated with an incident photon and may appear to decay via two branches: a 2431.7(3)-keV gamma ray directly to the ground state and a gamma ray 2423.5(3)-keV to the 7.861-keV level. This offers further credibility that these transitions are from NRF states in ^{239}Pu. A similar argument could be made for the transitions at 2144 keV and 2135 keV.

See Ref. 8 for a more detailed analysis and discussion.

SUMMARY

Thirteen new transitions have been assigned to ^{239}Pu (see Table 1). These transitions were discovered using photons from a bremsstrahlung source to excite

NRF states. Further effort is required to verify the assignment of the transitions to ^{239}Pu and to determine the polarizations of the states. Efforts to find NRF states in other isotopes of interest are also necessary..

TABLE 1. Results from significant gamma ray transitions associated with 239Pu. Given in the table are transition energies, statistical significance (ratio of peak area to uncertainty), cross sections, and widths (assuming g~2 for Pu239).

Energy (keV)	Statistical Significance	Cross Section (eV barns)	Width (meV)
2040.25(21)	5.8	8(2)	4(1)
2046.89(31)	4.2	5(2)	3(1)
2089.14(35)	3.7	4(1)	3(1)
2135.00(37)	3.5	4(2)	2(1)
2143.56(13)	9.7	13(2)	8(1)
2150.98(31)	4.2	5(2)	3(1)
2289.02(25)	6.2	8(2)	6(1)
2423.48(22)	7.2	10(2)	8(1)
2431.66(25)	6.3	9(3)	7(3)
2454.37(26)	6.2	9(3)	7(3)
2460.46(37)	4.7	6(4)	5(4)
2464.60(30)	5.7	8(4)	7(4)

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