Detection of Hidden Materials Using Nuclear Resonance Fluorescence Technique: Simulation and Measurements

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***Abstract*–The measured value of the 2.176 MeV ^{238}U Nuclear Resonance Fluorescence (NRF) peak count rate were compared with simulation results. The simulation methods studied include GEANT4, MCNP5 and MCNPX. The simulation results were found out to be consistent but one order of magnitude higher than the measured value. The possible reasons for this discrepancy were discussed.**

I. INTRODUCTION

THE ability to prevent smuggling of clandestine nuclear material has become a problem of contemporary importance and is being actively pursued by researchers globally. The detection and identification of hidden materials can be quite difficult due to the variability of possible environments. Passive and active approaches are being explored, using a wide variety of penetrating particles. One of the most interesting of these involves NRF.

The intelligent Model-Assisted Sensing System (iMASS) includes a real-time Monte Carlo simulation of NRF and analysis combined with the signal data processing for the detection of nuclear materials hidden in cargo containers. This paper presents the Geant4 platform developed for direct application in nuclear material detections. The dynamic flexibility of Geant4 and the ability to be sped up with parallelization is also described. The simulation is benchmarked against the measurement results obtained through a series of experiments using various samples, including aluminum, boron, and depleted uranium [1].

NRF can be considered as the nuclear analog of the atomic X-ray fluorescence process. The resonant energies represent the fingerprints for isotope identification. Resonant photons are highly penetrating and difficult to shield. Bremsstrahlung X-ray has become a widely used excitation photon source. Detectors used to measure the resonant photons ought to have high energy resolution for better signal-to-noise ratio. HPGe detectors provide the best energy resolution among commonly available radiation detectors. Recently developed lanthanum halide scintillation detectors offer good energy resolution and a fast decay constant. System performance with both HPGe detector and lanthanum halide scintillation detector ($LaCl_3$) are presented [2].

The iMASS is envisioned to simulate the cargo scanning in a virtual computational setting copying the real system; the main challenge is to develop a computational platform that will show the detection of nuclear materials in cargo. Geant4 was chosen as the operating “brain” of the iMASS computational platform. The NRF module, developed by Pacific Northwest National Labs (PNNL) [3], is part of the iMASS Geant4 platform. The post processing package, ROOT, is implemented in order to maintain the large structures of data created by Geant4 simulations as described in [1]. Geant4 platform includes simulation of the NRF source, the NRF scanning of the cargo interior, and the creation of data to be mined by the newly developed algorithm for signal analysis [1], [4], [5].

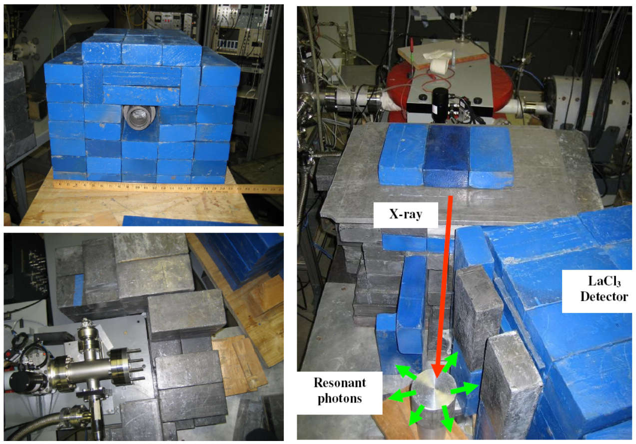

Fig. 1. NRF Experimental Setup

II. EXPERIMENTAL SETUP

The Van de Graff electron accelerator at the Radiation Laboratory of the University of Notre Dame is used as a photon source. The detection method involves exposing materials to a bremsstrahlung X-ray beam and detecting the resonantly-scattered photons. The LINAC can produce bremsstrahlung X-rays with energies of up to 3 MeV. These interrogating photons are highly penetrating and have the potential to investigate the whole body of a small cargo container. Scattered photons are measured using a strictly collimated gamma detector. The experimental setup is shown

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in Figure 1. Customized DAQ system based on FPGA technology was developed to be used together with fast decay lanthanum halide scintillation detector in high rate scenarios [6], [7]. It performs onboard, real-time digital signal processing (DSP). The DAQ board and onboard shaping method are shown in Figure 2.

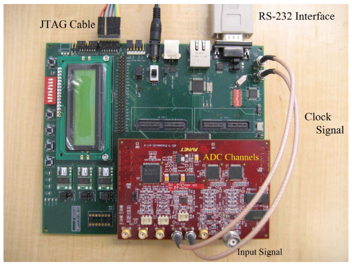

Fig. 2. FPGA based DAQ board and onboard shaping algorithm

Newly-available fast lanthanum halide scintillation detectors have an energy resolution sufficient to observe strong resonances in light materials, as shown in Figure 3. Their fast decay constant makes it possible to perform NRF experiments at a higher beam current, which reduces the detection time. HPGe detectors provide much better energy resolution, thus NRF spectra with good signal-to-noise ratio. Resonance peaks were observed from various samples, including depleted uranium, as shown in Figure 4.

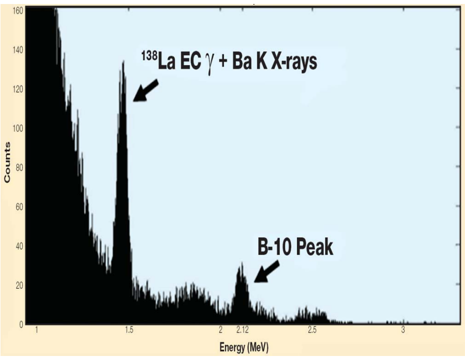

Fig. 3. NRF peak from the boron sample measured by a $LaCl_3$ detector

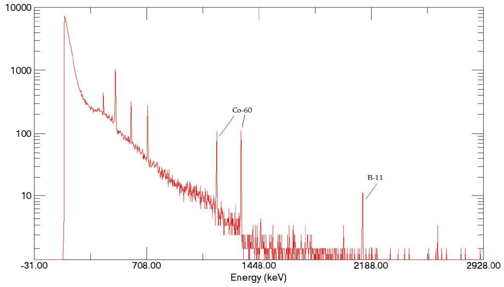

Fig. 4. NRF peak from the boron sample measured by an HPGe detector

III. CALCULATION USING ANALYTICAL METHOD

The shape of the NRF cross section can be best described using the Breit-Wigner formula. For a photon of energy E, the NRF cross section, i.e. the probability to excite a nucleus from a ground state with total angular momentum J_0 to an excited state with energy E_r and total angular momentum J_1, and then de-excite to a final state with total angular momentum J_f can be written as:

$$\sigma_1^0(E) = \pi\bar{\lambda}^2 \frac{2J_1+1}{2(2J_0+1)} \frac{\Gamma_0\Gamma_1}{(E-E_r)^2+\frac{1}{4}\Gamma^2} \tag{1}$$

The energy of an incident photon interacting with a nucleus varies according to the relative movement of the two, i.e. Doppler Effect. If the energy of the photon is E for a nucleus at rest, the apparent energy to a nucleus moving towards the photon source with a velocity is larger than E and can be written as:

$$E' = E(1+v/c)/\sqrt{1-(v/c)^2} \approx E(1+v/c) \tag{2}$$

However, the integrated NRF cross-section is not affected by Doppler broadening. This result is very useful in the estimation of interaction rates, because the width of the NRF effective cross-section is so narrow that in most cases the flux of the incident photon can be approximated by a constant independent of the energy in the calculation. For a thin sample, the NRF interaction rate can be derived as:

$$\begin{aligned} r &= \int N\sigma_D(E)\phi(E)dE = N\phi(E_r)\int \sigma_D(E)dE \\ &= N\frac{\sigma_{\max}^0 \Gamma\pi}{2}\phi(E_r) \end{aligned} \tag{3}$$

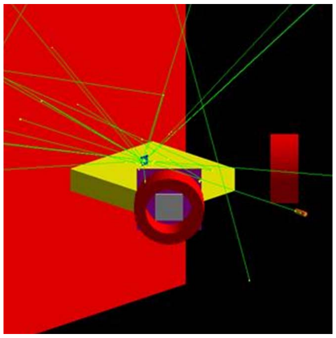

Fig. 5. iMASS rendering of an experimental setup

IV. SIMULATION USING IMASS – GEANT4

The experimental setup described above is modeled and simulated using iMASS. The NRF simulation environment consists of a few modules: a particle simulation program (Geant4), NRF physics module (G4NRF developed by PNNL), large scale data-analysis (ROOT), intelligent algorithms for signal processing and threat identification and C/C++ which glues all of the modules together. Modules are developed in such a way that modifications can easily be made to adapt the simulating environment to any other real world situation. Figure 5 is an artistic rendering of the realistic environment. Various materials have been simulated to qualitatively benchmark the Geant4NRF module that PNNL developed.

V. SIMULATION USING MCNP/MCNPX

NRF is not included in any standard MCNP/MCNPX release up to now. To better understand the feasibility of utilizing this technique in detection of SNM for homeland security application, efforts have been made to include this interaction in MCNP/MCNPX simulations.

Photoatomic interactions were used to imitate NRF in MCNP5 [8]. There are two subroutines in the source code that need to be modified to include NRF in the simulation, ‘photot.F90’ and ‘colidp.F90’. In subroutine ‘photot.F90’, the code obtains photoatomic cross sections (photoelectric, coherent and incoherent scattering, pair production, and total cross section). NRF is approximated using Breit-Wigner, Gaussian function or a hat-top function and added to the total cross section value. In subroutine ‘colidp.F90’, the code samples nuclides (based on total cross section) and type of interaction (based on partial cross section). If NRF is sampled, the photon is re-emitted in all directions isotropically with an energy that equals the incident energy minus recoil energy loss.

The ability to simulate NRF has been planned as one of the new capabilities of the next MCNPX release. Effort has been made to include this feature in currently available MCNPX releases by modifying the ACE files [9]. Due to the limitation of the ENDF format, NRF cross section was approximated using triangular function.

VI. COMPARISON AND CONCLUSION

The experimental setup described above was modeled and simulated using various codes, including modified MCNP5, MCNPX and iMASS (Geant4). The count rate of the resonant peak at 2.176 MeV from our U-238 sample (2 inch by 2 inch by 0.1 inch) was derived from simulation results as well as analytical calculation results. The comparison is shown in Table I below. The measured count rate at 2.176 MeV was only about 0.01 cps. This is an order of magnitude lower than the predicted value. Some possible reasons that can cause this discrepancy include the bad beam condition of the Van de Graaff LINAC, the incompleteness of simulation data library, and the approximation used in simulation approaches.

TABLE I. COMPARISON OF COUNT RATE (UNIT: CPS)

Calculation Methods	Count Rate of the 2.176 MeV Peak
Analytical	0.248
MCNP5	0.267
MCNPX	0.366
iMASS (GEANT4)	0.377

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