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Active Interrogation of Nuclear Materials Using $LaBr_3$: Ce Detectors

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Abstract

Active interrogation in ^{235}U was demonstrated with $LaBr_3$:Ce scintillation detectors using the nuclear resonance fluorescence (NRF) technique. An NRF experiment was performed at the High Intensity γ-ray Source (HIγS) facility using quasi-monochromatic circularly or linearly polarized γ-ray beams. Photons scattered at 90^o relative to the incident beam were detected with two different sizes of cylindrical $LaBr_3$:Ce detectors. Clear NRF peaks at 1733 and 1815 keV corresponding to de-excitations to the ground state and/or low-lying levels in ^{235}U were observed within 77 minutes of beam time even under the high background due to the self-activity of $LaBr_3$:Ce and the radioactive decay of ^{214}Bi nuclei existing in the ^{235}U target. The present study shows a possibility of using $LaBr_3$:Ce detector to perform NRF experiments, promoting many options for inspection of special nuclear materials.

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1. Introduction

Nuclear power has provided about 2 millions of GW.d worldwide of clean electricity during the past three decades [1]. Although the prospects have been clouded by the uncertainties concerning the nuclear policies, the nuclear power output grows at the same pace as prior the Fukushima Daiichi accident in early 2011 [2]. The capacity of nuclear materials required as nuclear fuel or generated, as spent fuel, is huge. Nondestructive assay (NDA) of special nuclear materials (SNM) has an increasing requirement in nuclear security and safeguard prospects. NDA for SNM requires not only the elemental composition of an object, but also the isotopic identification for specific isotopes usually being the fuel for nuclear power plants such as ^{235}U and ^{239}Pu. Therefore, a signature of a specific nucleus is essential for the inspection of nuclear material. NRF, used for nuclear structure investigation [3]-[7], is a process of resonant excitation of nuclear levels by absorption of photons and subsequent de-excitation to lower-lying levels by γ-ray emission. Since each nucleus has characteristic resonant energies, the detection of resonant γ-rays provides a unique fingerprint for identification. In addition, high-energy γ-rays used for the resonant excitation are highly penetrable in materials. Thus, the NRF technique can be applied for elemental and isotopic characterization of materials at the inside of heavy shieldings [8].

The main challenge involved in the NRF process comes from the very narrow bandwidth of the nuclear energy level (around eV or meV). Thus, a very small number of incident γ-rays contributes to trigger NRF events. In addition, this small number of NRF events might be hidden by huge background events originating from the atomic scattering and/or radioactive species. Consequently, an NRF measurement may take long time to obtain reasonable statistics [9], which is not suitable for practical use of inspecting materials.

Laser Compton backscattering γ-rays source providing a quasi-monochromatic γ-rays beam provides a very good opportunity to decrease the background events from the atomic scattering at the energy of interest. Furthermore, implementing a detection system based on scintillating detectors with very high counting-rates saves the time for an NRF measurement in comparison with the high purity germanium (HPGe) detectors. Recently, lanthanum halides scintillating detectors are commercially available. $LaBr_3$:Ce detectors which have a nanosecond rise time, relatively high density (5.1 g/cm^3) and a good energy resolution (~ 3% at 662 keV) have good properties for detecting NRF photons.

In the present study, we demonstrate the feasibility of using $LaBr_3$:Ce detector to detect the photon-induced excitations from ^{235}U in a shielding of 1 cm of iron using a quasi-monochromatic photon beam. A simple and efficient analysis technique based on the Statistics-sensitive Nonlinear Peak-clipping (SNIP) algorithm is implemented to improve the signal to noise ratio for NRF peaks [10].

2. Experimental Setup

An NRF experiment was performed at the High Intensity γ-ray Source (HIγS) facility, a joint project between the Triangle Universities Nuclear Laboratory (TUNL) and the Duke University Free Electron Laser Laboratory (DFELL) [9]. Fig. 1 shows the relevant energy scheme of ^{235}U. A circularly polarized γ-ray beam having energy of 1730 or 1850 keV hit a 93.7% enriched ^{235}U target. The target consisted of three square foils (2.54×2.54 cm^2) with a density of 239 mg/cm^2. Cylindrical collimator, with a size of 30.5 cm long and 1.905 cm diameter defined the energy spread of the incident γ-ray beam around 2.4% (FWHM). Alignment of the target was done so that the center of the γ-ray beam coincided with the center of the ^{235}U target and all size of the beam hit the target. Shielding conditions were either bare target or shielded one within 1 cm of iron.

The detection system consisted of two $LaBr_3$:Ce detectors and a 60% (HPGe) detector. Lead and copper absorbers with thicknesses of 3.2 mm and 4.0 mm, respectively, were mounted only on the face of the HPGe to attenuate the intensity of the low-energy background. All detectors were positioned at right angle with respect to both the target and γ-ray beam direction. Along the horizontal axis, the HPGe detector was positioned 10 cm from the center of the target while a 1.5” diameter and 3” length $LaBr_3$:Ce detector was positioned at 5 cm from the center of the target. Along the vertical axis a 3.5” diameter and 4” length $LaBr_3$:Ce detector was positioned at 6.8 cm from the center of the target. The energy resolution of each $LaBr_3$:Ce detector at 662 keV is around 3.5 % (FWHM). The energy distribution and the flux of the incident beam were measured by a 123% HPGe positioned at the downstream of the NRF measurement system. Copper absorbers were used to attenuate the beam before hitting the flux monitor. Multichannel analyzer was used to process signals coming from HPGe detectors while digital signal processing (DSP) unit was used to process signals coming from $LaBr_3$:Ce detectors. Energy calibration was performed using the two natural background peaks of ^{40}K and ^{208}Tl at 1461 keV and 2615 keV respectively. Data analysis was performed by the ROOT analysis toolkit [12]. TSpectrum class in this toolkit provides background estimation and subtraction by SNIP method. The clipping window w used in the SNIP method was fixed at 56 keV for all spectra measured with $LaBr_3$:Ce.

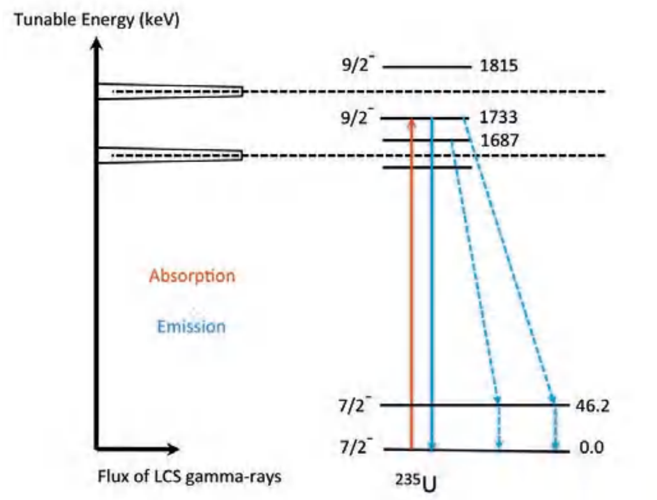

Fig.1. Energy level scheme of ^{235}U nuclide shows the energy levels at 1733 keV and 1815 keV as well as the low lying level at 46.2 keV. The last one is populated via branching transition from the higher levels.

3. Results and Discussion

Background Peaks

Fig. 2 shows an off-beam spectra measured for around 9 hours using $LaBr_3$:Ce and HPGe detectors. Three types of background radiation are observed in these spectra. Natural background resulting from the naturally occurring radioisotopes, ^{40}K and ^{208}Th, at energies of 1461 keV and 2615 keV, respectively. Another type of background is due to some radioactive contaminants existing in the $LaBr_3$:Ce crystal. Of these contaminants, the radioactive isotope of lanthanum (138La with abundance of 0.09%) is the most pronounce. This long-lived isotope ($T_{1/2}$ = 1.02×10^{11} y) emits two photons at 789 keV and 1436 keV. These γ-rays appear in the HPGe detector as well because of the close geometry of the detectors. In addition, a broad-structured peaks from 2000 keV to 2500 keV are attributed to the emission of alpha particles from ^{227}Ac which is thought to exist within the $LaBr_3$:Ce crystal because of its similar chemical properties to La. Peaks due to alpha emitters don’t appear in HPGe spectrum because the very short range of alpha particle. Although, the detector background is harmful, it may be used as a self-energy-calibration [13]. The background peaks due to the target result from the daughters of ^{238}U, basically from ^{214}Bi. The peaks from ^{214}Bi located at 1661, 1729, 1765 and 1847 keV overlap with the NRF energy levels at 1687, 1733, 1769 and 1815 keV respectively. This overlapping is critical in case of $LaBr_3$:Ce detector with energy resolution around 40 keV. Also, even for HPGe the overlapping peaks at 1729 keV make it difficult to separate the NRF peak at 1733 keV.

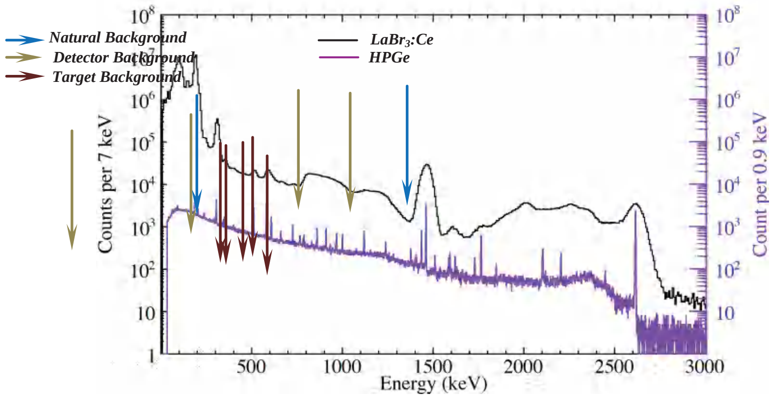

Fig. 2. Background spectra measured with $LaBr_3$:Ce and HPGe detectors for 9 hours.
Arrows indicate peaks due to different types of background

NRF Spectra Measured with $LaBr_3$:Ce Detectors

The response of the 1.5”×3” $LaBr_3$:Ce detector to the incident beam of energy 1730 keV is shown in Fig. 3-a for the case of bare target and Fig. 3-b for the case of shielded target. The beam on target was 77 minutes and the intensity of the incident photon beam was about 1.6×10^7 s^{-1} MeV. The intensity of the NRF peak is reduced by the factor of 25% due to shielding. This reduction is in good agreement with the γ-ray attenuation data. A comparison between the on-beam spectra and the off-beam spectra indicates that the peak at 1733 keV is an NRF with a negligible contribution from the background peak at 1729 keV.

On the other hand, the spectra measured by HPGe detector are shown in Fig. 3-c. with both on-beam and off-beam runs. The NRF peak at 1733 keV is hardly seen as a shoulder of the background peak at 1729 keV even with a collection time of 530 minutes. This is consistent with the data available for ^{235}U [6].

Background Suppressed by gating

As mentioned above, there are many background radiation that accompany the NRF measurement on ^{235}U with $LaBr_3$:Ce. Therefore, a gating signal of 100 ns width, in coincidence with the storage ring RF signal, was applied to the scintillating detector to decrease background events. On the other hand, there was no gating condition applied to the HPGe because the gating frequency is around 5.5 MHz which is too high to coincide with HPGe signals. The NRF spectrum measured with the 3.5”×4” $LaBr_3$:Ce detector under the gating condition when the incident photon beam had an energy of 1850 keV and the intensity of the incident beam was around 6.5×10^7 s^{-1} MeV is shown in Fig. 4-a for the case of bare target.

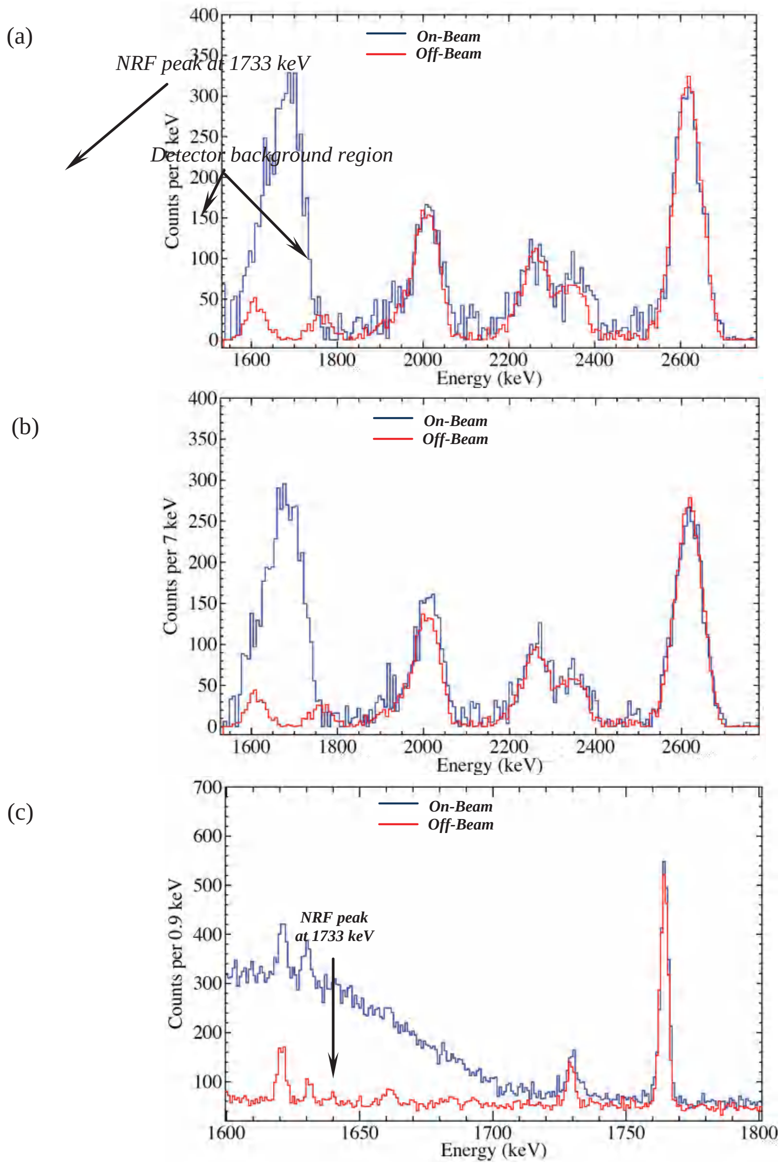

Fig. 3. On-beam spectra at beam energy of 1730 keV with the corresponding off-beam measurements in red. (a) Spectra measured with the 1.5”×3” $LaBr_3$:Ce detectors for 77 minutes while the target was bare. b. Spectra measured with the 1.5”×3” $LaBr_3$:Ce detectors while the target was shielded within 1 cm of iron. The real time is normalized to 77 minutes. In (a) and (b) the off-beam real time is normalized to 77 minutes as well. (c) Spectra summed up from different runs of total real time of 530 minutes measured with HPGe. The off-beam measurement is normalized to 530 minutes. The NRF peak at 1733 keV appears as a shoulder of the background peak at 1729 keV.

The real time is 72 minutes. The off-beam spectrum is normalized by the intensity of the background peak at 2615 keV while the real time normalization is used for the off-beam spectrum measured by HPGe. The signal to noise ratio is greatly enhanced by suppressing the background using the gating technique. The NRF peak measured by $LaBr_3$:Ce detector is a superposition of three levels corresponding to energies of 1815, 1828 and 1862 keV which are clearly observed in the HPGe spectrum. The energy resolution of the scintillating detector is too low to distinguish the three peaks individually.

(a)

(b)

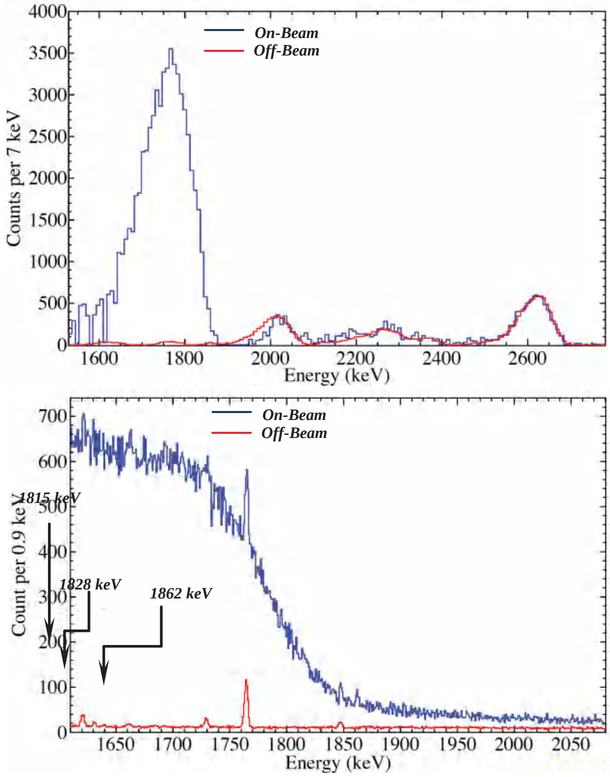

Fig. 4. On-beam spectra at beam energy of 1850 keV with the corresponding off-beam measurements in red. (a) Spectra measured with the 3.5”×4” $LaBr_3$:Ce detectors for 72 minutes while the target was bare under 100 ns gating condition. The off-beam spectrum is normalized using the 2615 keV peak intensity. (b) Spectra summed up from different runs of total real time of 165 minutes measured with HPGe detector. The off-beam measurement was normalized to 165 minutes. NRF peaks at 1815, 1828 and 1862 keV are visible in the spectrum.

Energy Shift in $LaBr_3$:Ce Spectra

As seen from Fig. 3-a, Fig. 3-b, and Fig. 4-a, the NRF peaks in $LaBr_3$:Ce spectra are clearly visible; however, all NRF peaks are down-shifted by an amount of 46 keV. Fig. 5 shows a close view of the NRF peak at 1733 keV. The broad peak seems to be a composition of three NRF peaks corresponding to energy levels of 1733, 1687 and 1641 keV with a difference of 46 keV. This can be explained in terms of branching transition to the low-lying energy level before the de-excitation to the ground level. On contrast, these branching transitions are not clear in the spectra measured by HPGe. On the other hand, the peak at 1687 keV has the highest intensity, even higher than the peak at 1733 keV which has been reported to have the highest cross section among the ^{235}U levels [9], [14]. The fast timing-resolution of $LaBr_3$:Ce detectors (> 300 ps) might enable the detectors to detect photons emitted in different branching transitions separately.

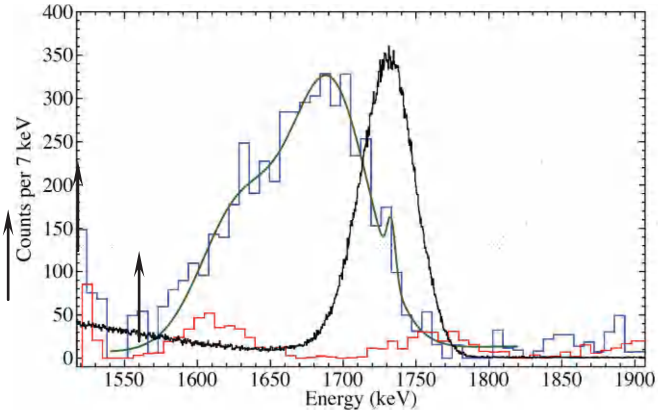

Fig. 5. Energy shift in $LaBr_3$:Ce spectra. Black arrows refer to energies of 1641, 1687 and 1733 keV. Black spectrum is the incident beam profile.

4. Conclusion

NRF measurements were performed on ^{235}U using quasi-mono-energetic photon beams with energies 1730 and 1850 keV. The scattered photons at 90^{o}, with respect to the incident beam, were detected with $LaBr_3$:Ce detectors while the target was either bare or shielded in 1 cm iron box. Broad and clearly visible NRF peaks were obtained from the de-excitations of ^{235}U nuclei at 1733 and 1815 keV by using $LaBr_3$:Ce detectors. Broadness of the NRF peaks is attributed to the branching transitions to the low-lying energy level at 46 keV. Gating technique enhanced the signal to noise ratio in the $LaBr_3$:Ce spectra. The results presented in this report revealed the possibility of using $LaBr_3$:Ce detectors in an NRF measurement on ^{235}U which may have a good impact on the active interrogation of special nuclear materials.

Acknowledgment

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