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The sensitivity of $LaBr_3$:Ce scintillation detectors to low energy neutrons: Measurement and Monte Carlo simulation

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ABSTRACT

The neutron sensitivity of a cylindrical ⌀1.5 in. × 1.5 in. $LaBr_3$:Ce scintillation detector was measured using quasi-monoenergetic neutron beams in the energy range from 40 keV to 2.5 MeV. In this energy range the detector is sensitive to γ-rays generated in neutron inelastic and capture processes. The experimental energy response was compared with Monte Carlo simulations performed with the Geant4 simulation toolkit using the so-called High Precision Neutron Models. These models rely on relevant information stored in evaluated nuclear data libraries. The performance of the Geant4 Neutron Data Library as well as several standard nuclear data libraries was investigated. In the latter case this was made possible by the use of a conversion tool that allowed the direct use of the data from other libraries in Geant4. Overall it was found that there was good agreement with experiment for some of the neutron data bases like ENDF/B-VII.0 or JENDL-3.3 but not with the others such as ENDF/B-VI.8 or JEFF-3.1.

1. Introduction

Inorganic scintillation detectors find many applications in γ-ray spectroscopy mainly because of the very large detection efficiencies which can be attained. In comparison with high-purity germanium (HPGe) detectors larger intrinsic efficiencies can be obtained because of the larger density and/or effective atomic number. Also in comparison with HPGe detectors, larger solid angle coverage can be reached (close to 4π), because large crystal volumes can be grown and the dead material in multidetector arrangements is more easily minimized. However HPGe detectors offer a much better energy resolution ($\sim 0.2\%$ at 1 MeV) than scintillation detectors ($\sim 5\%$ in NaI(Tl), for comparison). The advent [1] of $LaBr_3$:Ce scintillation material, which has about a factor of two better energy resolution compared to NaI(Tl), about 30% larger intrinsic efficiency and much faster scintillation time response, has triggered many new applications. Several properties of $LaBr_3$:Ce used in scintillation detectors have been the subject of thorough investigations as revealed in a search of bibliographic databases. In the present publication we investigate the question of $LaBr_3$:Ce neutron sensitivity to low energy neutrons. Although the results of this study may have other applications we are particularly concerned with the characterization of neutron induced background in the context of nuclear γ-ray spectroscopy applications.

In nuclear research it is often found that the emission of γ-rays, being the primary object of the study, is accompanied (either simultaneously or alternatively) by the emission of neutrons. An example of this situation is the production of neutrons in the reactions used to populate nuclear exited levels in in-beam γ-ray spectroscopy. Another example is the emission of β-delayed neutrons in γ-ray spectroscopy studies of β-decay. Neutrons produced in both cases have in common a relatively low energy (up to a few MeV) and the possibility of inducing background signals through interaction with the γ-ray detector. Therefore a careful investigation of the sensitivity of inorganic scintillation detectors to low energy neutrons is of relevance in these fields.

As an example we take the study of β-decay using the total absorption γ-ray spectroscopy (TAGS) technique [2]. The technique aims to determine accurately the β-intensity distribution for complex decays using a high efficiency 4π scintillation detector.

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The intensity is obtained from the total absorption spectrum by deconvolution with the appropriate response. In the study of β-decay of neutron-rich nuclei one encounters the phenomenon of β-delayed neutron emission in nuclei away from the valley of β-stability. If the decay proceeds to levels above the neutron separation energy in the daughter nucleus the emission of neutrons competes efficiently with γ-ray de-excitation. The emitted neutron can interact with the detector introducing effects in the spectra which must be subtracted if we are to obtain the correct information from the analysis of the data. The quantification of this contamination has been the primary motivation for a research programme looking at several inorganic scintillation crystals, using both Monte Carlo (MC) simulations and measurements. In the present publication we report on the studies performed for the case of $LaBr_3$:Ce material that has been considered for the construction of a new spectrometer in the DEcay SPECtroscopy (DESPEC) experiment [3] at the Facility for Antiproton and Ion Research (FAIR).

Neutrons interact with matter in a complex way through a series of processes whose probability varies strongly depending on isotope and neutron energy. The neutron sensitivity of a scintillation detector, or probability of detecting an incoming neutron, and the associated energy response are influenced by all intervening materials and their geometrical disposition. The experimental determination of the neutron sensitivity requires measurements with the actual detector at the neutron energies of relevance, which is difficult to do. The use of MC simulations for the estimation of this quantity is very appealing because of its generality. However, it is not obvious that MC codes available on the market can provide the detector response with enough accuracy to be useful. In this work we investigate experimentally the response of a $LaBr_3$:Ce detector to neutrons at several energies below 2.5 MeV and compare them with MC simulations. This study is performed with well characterized pure neutron beams as a necessary step towards the application to more complex mixed neutron-gamma fields encountered in spectroscopy experiments.

2. Neutron interactions

Low energy neutrons interact with matter through a series of processes whose importance, as quantified by the cross-section, depends strongly on the neutron energy and the isotopic composition. The main processes contributing to the total neutron cross-section below few MeV for non-fissile isotopes are elastic and inelastic scattering and radiative capture. The questions considered below, exemplified by the case of $LaBr_3$:Ce, apply to inorganic scintillation materials in general.

Fig. 1 shows the macroscopic group cross-sections for $LaBr_3$:Ce(5%) for the three processes that exhaust the total cross-section in the energy range from 1 keV to 10 MeV. The group cross-sections are obtained with the code PREPRO [4] from the ENDF/B-VII.0 [5] evaluated nuclear data file. Group cross-sections represent averages over neutron energy intervals (shown in the figure). They take into account the neutron cross-sections for all the stable isotopes of the three relevant elements Br, La, and Ce. As observed the elastic process dominates in the whole energy range, the inelastic channel grows rapidly above the threshold related to the lowest excited state, and the capture channel decreases strongly with energy.

The elastic scattering process generates signals in the scintillation crystal through the energy loss of recoiling nuclei. Given the large masses of the target nuclei, the recoil energies are small (a maximum of 0.25 MeV for the collision of a 5 MeV neutron with ^{79}Br). In addition, due to the large ionization density produced by the low energy heavy recoil, the light output of the scintillation crystal will be strongly quenched with respect to the light output produced by electrons (γ-rays). This is a well known phenomenon in scintillation materials. In the case of $LaBr_3$:Ce a quenching factor larger than 2 has been observed [6] for α particles and even stronger quenching can be expected for heavier particles [7]. As a consequence the recoiling nuclei will give a negligible signal, below practical detection thresholds, for neutrons up to several MeV and are of no concern for our purposes.

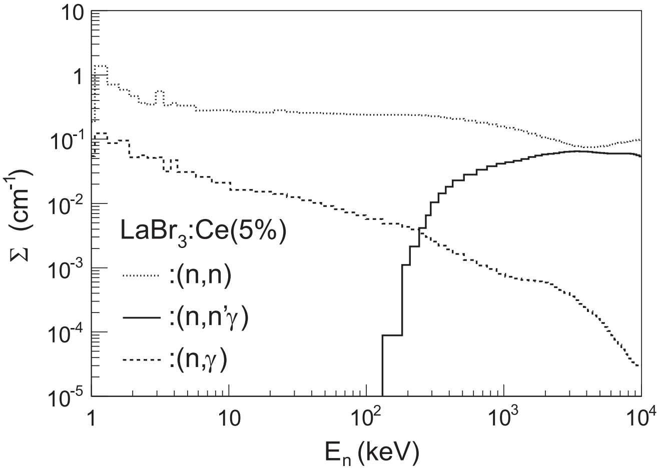

Fig. 1. $LaBr_3$:Ce(5%) group cross-sections for elastic, inelastic and radiative capture neutron reaction channels.

In the case of inelastic and radiative capture reactions γ-rays are generated as secondary particles. They can easily produce a detectable signal indistinguishable from the primary radiation of interest, making these channels the biggest concern in γ-ray spectroscopy. Considering the cross-section dependence with neutron energy (see Fig. 1), one expects that the inelastic channel dominates except below the threshold (few hundred keV). The energy release is limited by the neutron energy in the case of inelastic reactions, while in the case of capture reactions it is equal to the neutron separation energy (typically ranging from 5 to 10 MeV) plus the neutron energy. Because of this, the contamination coming from inelastic scattering concentrates at low energies in the spectrum while that coming from capture shows up at high energies. In addition, due to the nuclear level density variation with excitation energy, the inelastic channel is associated with the emission of a few γ-rays mostly of well known energy, while the capture channel is associated with an electromagnetic cascade of average multiplicity $m = 4–5$ and unknown energy distribution, including high energy primary γ-rays. How this radiation deposits energy in a given detector depends on its geometry. Therefore the measured neutron sensitivity and spectral distribution is specific to a given set-up. For example, the capture process will produce a continuum distribution in small detectors while high energy peaks corresponding to the full absorption of the cascade will appear for large volume detectors [8].

Modern general purpose MC simulation codes for the transport and interaction of radiation in matter have reached a high degree of predictive power in the case of electromagnetic interactions. In order to be applied to the calculation of the sensitivity of γ-ray spectrometers to low energy neutrons, a MC code must include a proper description of the neutron transport and reaction final state. This description should be based on the information contained in evaluated databases for reaction cross-sections, angular distributions, etc., as well as on the information available in nuclear structure databases for excited level schemes and de-excitation patterns.

3. Experiment

In order to investigate the neutron sensitivity of $LaBr_3$:Ce material a small detector was irradiated with neutron beams of well defined energy spanning the energy region from 40 keV (pure radiative capture) to 2.5 MeV (dominated by inelastic scattering).