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Sensitive neutron detection method using delayed coincidence transitions in existing iodine-containing detectors

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ABSTRACT

This work explains a new, highly sensitive method for the detection of neutrons, which uses the $T_{1/2} = 845$ ns delay in the decay of ^{128}I at the 137.8 keV energy level, resulting from the capture of thermal neutrons by iodine nuclei in NaI and CsI scintillation detectors. The use of delayed coincidence techniques with a several μs delay time window for delayed events allows for the highly effective discrimination of neutron events from any existing background signals. A comparison of ambient neutron measurements between those identified through the suggested method from a cylindrical, ø 63 mm × 63 mm NaI(Tl) scintillator and those from a low-background proportional ^{3}He counter experimentally demonstrates the efficacy of this neutron detection method. For an isotropic, 4π, thermal neutron flux of 1 n cm^{-2} s^{-1}, the absolute sensitivity of the NaI detector was found to be 6.5 ± 1 counts s^{-1} with an accidental coincidence background of 0.8 events day^{-1} for any delay time window of $\Delta t = 1$ μs. The proposed method can provide low-background experiments, using NaI or CsI, with measurements of the rate and stability of incoming neutron flux to a greater accuracy than 10^{-8} n cm^{-2} s^{-1}.

1. Introduction

The search for alternative neutron detectors to replace those that use ^{3}He presents a pressing issue in modern physics [1,2]. The fairly low cost, widespread availability, and ease of use of NaI(Tl), CsI(Tl), CsI(Na), and CsI(pure) scintillation detectors makes their implementation for the detection of neutrons an incredible opportunity. Neutrons have previously been detected by these scintillation devices through various approaches (see Table 1).

It is important to note that, in [4], when a NaI spectrometer was compared to a ^{3}He-based portal monitor with a comparable active volume, the detection efficiencies and minimum detectable activities of the devices were found to be similar.

In general, neutron measurements with NaI and CsI detectors require accurately accounting for background signals in the neutron detection region of interest because of the high sensitivity of these detectors to cosmic and γ-rays. To efficiently discriminate neutron events from background signals, the neutron detection method proposed in this work uses delayed coincidence techniques with a short delay time window.

2. Description of the method

Iodine has only one stable isotope: ^{127}I. This isotope has a cross section for thermal neutron capture of $\sigma_\gamma^0 = 6.2 \pm 0.2$ b [10], which is 860 times lower than ^{3}He's cross section of 5333 ± 7 b. However, it is important to note that NaI (solid) has 109 times as many moles as an equal volume of ^{3}He (gas, 500 kPa), and CsI (solid) has 77 times as many moles. Thus, despite its relatively low cross section, the fairly small ø 63 mm × 63 mm NaI(Tl) detector (used for the experiment described in this work, and henceforth referred to with $^{63\times63}$NaI) has an ~50% neutron capture (on ^{127}I) effectiveness, as obtained from Geant4 [11] modeling based on assumptions about the uniform thermal neutron flux (the Maxwell-Boltzmann distribution at room temperature). ^{128}I in its excited state (6.8 MeV) is produced as a result of neutron capture, and its decay to the ground state proceeds through a series of low-energy levels (see Table 2). This decay process often includes the energy level 137.8 keV with $T_{1/2} = 845 \pm 20$ ns. To identify neutron capture by iodide in a detector, thereby measuring neutron flux, measurements of the following delayed coincidences by the same detector can be used: the decay transition from 6.8 to 137.8 keV (the neutron prompt signal) and the transition from 137.8 keV to the

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Table 1
Prior approaches for neutron detection with NaI and CsI.

Reference	Detector	Neutrons	Short description
[3]	NaI	thermal	boron lining with available NaI detectors
[4]	NaI	thermal	high-energy photons following (n,γ) reactions in the NaI
[5,6]	NaI(Tl)	thermal	triple $\beta-\gamma-\gamma$ coincidences in two detectors following (n,γ) reactions on ^{23}Na
[7]	NaI	thermal	activated NaI detector (^{128}I $\beta-$ decay, $T_{1/2}$ = 25 min and ^{24}Na $\beta-$ decay, $T_{1/2}$ = 15 h)
[8]	CsI(Na)	fast	57.6 keV signal from ^{127}I(n,n') inelastic scattering
[9]	NaI(Tl), CsI(Tl)	fast	1–200 MeV neutrons, (n,p) and (n,α) reactions, pulse-shape discrimination

Table 2
^{128}I excited energy levels below 200 keV and their decay transitions (from [10,12]).

Energy level in ^{128}I (keV)	$T_{1/2}$ (ns)	Energy levels following decay (keV)
180		160.8, 85.5, 27.4
167.4	175 ± 15	137.8
160.8		27.4, 0
151.6		85.5, 27.4
144.0		133.6
137.8	**845 ± 20**	**133.6, 85.5**
133.6	12.3 ± 0.5	85.5, 27.4, 0
128.2		85.5
85.5		27.4
27.4		0
0		

ground state (the neutron delayed signal).

According to [12], following neutron capture by iodide, 40 ± 10% of all de-excitations pass through the 137.8 keV energy level. This energy level is filled and emptied via a series of low-energy, highly converted transitions, including the 4.2 keV transition, which has never been experimentally investigated on its own. These transitions raise uncertainty about the probability of decay via this energy level, causing uncertainty when estimating the detector's overall sensitivity. The lack of direct measurements of low-energy γ-transition intensities and of internal conversion coefficients also results in significant uncertainty in the detector's effectiveness at recording neutron prompt and delayed events. Nevertheless, Geant4 MC's estimations obtain a >75% absorption probability for low-energy γ-rays and electrons released in the $^{63\times63}$NaI detector, following neutron capture. Thus, the overall effectiveness of this NaI detector at detecting neutrons is roughly estimated with performed MC to be ~10% and has significant uncertainties, as described above.

3. The experiment

For experimental verification of the suggested method, a simple spectrometer was created, as shown in Fig. 1. A 3″ PMT R6091 (Hamamatsu) was attached to a cylindrical, ø63 mm × 63 mm NaI(Tl) scintillator via optical grease, and this device was placed in a 5-cm thick lead brick well. The top part of this lead brick shield was left open. A cardboard box, covered along the inside in black paper, shielded the detector from light. A CAEN DT5780 Dual Digital Multi Channel Analyzer collected data using an event by event list mode. Consecutive events within a delay time smaller than 1.8 μs could not be properly registered due to the NaI scintillator's ~250 ns decay time, after-pulses in the PMT, and impulse shapes with tails extending up to approximately 1.5 μs after the initial peak. Therefore, 1.8 μs was chosen as the trigger holdoff. With an energy threshold slightly below 100 keV, the total count rate of the detector was ~7.3 Hz.

Fig. 1. On left: NaI spectrometer inside lead shield (front side of shield removed). 1) ø 63 × 63 mm NaI(Tl) scintillator, 2) R6091 PMT, 3) lead bricks. On right: NaI and ^{3}He neutron spectrometers during data acquisition. 4) Black box with NaI detector inside, 5) CHM-57 ^{3}He filled proportional counter, 6) ^{3}He neutron detector's preamplifier, 7) DT5780 Dual Digital Multi Channel Analyzer used to acquire data from both detectors.

The energy response in the 137 keV expected region for delayed signals was calibrated with measurements collected in the presence of a ^{139}Ce radioactive source (165.9 keV γ-line). In the first successful test following the calibration measurements, a highly active (>10^4 n s^{-1}) PuBe neutron source was used to see if the number of neutron events could be measured using a NaI scintillator via the suggested method (Fig. 2). This test did not involve any special moderators: fast neutrons from the source were thermalized by surrounding materials.

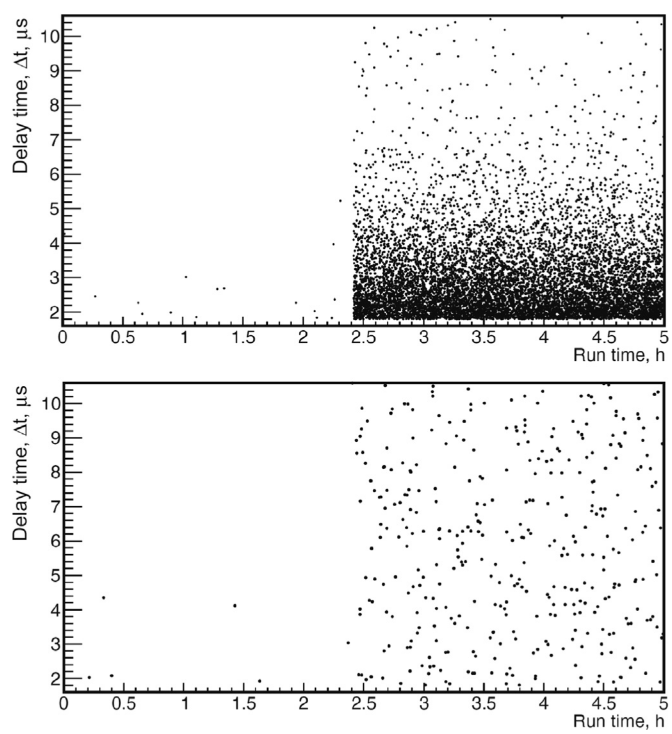

Fig. 2. Delayed coincidences registered without (left half of each graph) and in the presence of (right half of each graph) the PuBe neutron source. Each dot represents one coincidence event. The upper graph shows coincidences with delayed events in the 137 keV region, i.e. ADC channels from 150 to 350 (see Fig. 3). During the run with the PuBe source, the number of coincidence events in the upper graph is evidently greater for a delay time of Δt < 7 μs than for a larger delay time. The lower graph shows delayed coincidence events registered from ADC channel 1000–8000 (well above the expected signal for a neutron event). In this graph, the noticeable increase of random coincidences for the run with the PuBe source, as compared to that without it, does not depend on the delay time.