



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, $\phi 63$ mm \times 63 mm NaI(Tl) scintillator and those from a low-background proportional ^3He counter experimentally demonstrates the efficacy of this neutron detection method. For an isotropic, 4π , thermal neutron flux of $1 \text{ n cm}^{-2} \text{ s}^{-1}$, 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 $\Delta t = 1 \mu\text{s}$. 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 $10^{-8} \text{ n cm}^{-2} \text{ s}^{-1}$.

1. Introduction

The search for alternative neutron detectors to replace those that use ^3He 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 ^3He -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 ^3He '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 ^3He (gas, 500 kPa), and CsI (solid) has 77 times as many moles. Thus, despite its relatively low cross section, the fairly small $\phi 63$ mm \times 63 mm NaI(Tl) detector (used for the experiment described in this work, and henceforth referred to with $^{63\times 63}\text{NaI}$) has an $\sim 50\%$ 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

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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 $\beta - \gamma$ decay, $T_{1/2} = 25$ min and ^{24}Na $\beta - \gamma$ decay, $T_{1/2} = 15$ h)
[8]	CsI(Na)	fast	57.6 keV signal from $^{127}\text{I}(n,n')$ 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	175 \pm 15	160.8, 85.5, 27.4	
167.4		137.8	
160.8	845 \pm 20	27.4, 0	
151.6		85.5, 27.4	
144.0		133.6	
137.8		133.6, 85.5	
133.6		85.5, 27.4, 0	
128.2		85.5	
85.5		27.4	
27.4		0	
0			

neutron prompt signal) and the transition from 137.8 keV to the ground state (the neutron delayed signal).

According to [12], following neutron capture by iodide, 40 \pm 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 $^{63\times 63}\text{NaI}$ detector, following neutron capture. Thus, the overall effectiveness of this NaI detector at detecting neutrons is roughly estimated with performed MC to be \sim 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, $\phi 63$ mm \times 63 mm NaI(Tl)



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

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 \sim 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 \sim 7.3 Hz.

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.

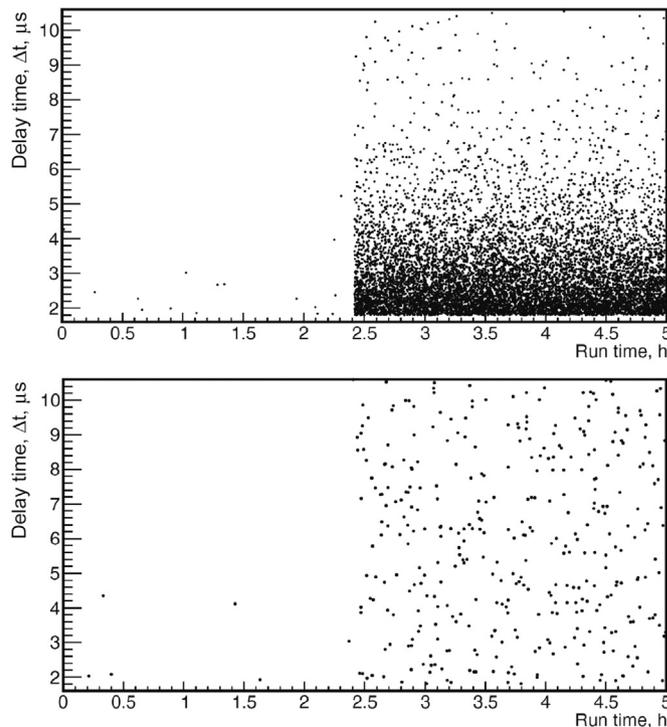


Fig. 2. Delayed coincidences registered without (left half of each graph) and in the presence of (right half of each graph) the PuBe neutron source. Each dot represents one coincidence event. The upper graph shows coincidences with delayed events in the 137 keV region, i.e. ADC channels from 150 to 350 (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 $\Delta t < 7$ 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.

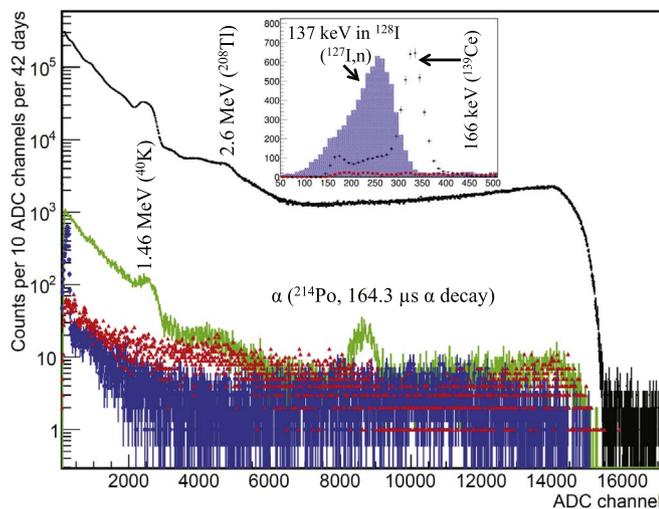


Fig. 3. The experimental energy spectra measured during 42 days in the LNP, JINR with a $^{63}\times^{63}\text{NaI}$ spectrometer. Black dots - energy spectrum without any cuts; blue dots with error bars and red triangles - neutron prompt and neutron delayed spectra, respectively (delay time window is 1.8 – 10 μs , no ADC window cuts were applied, neutron events in the delayed spectrum are in a peak corresponding to ADC channels 150–350); green dots - delayed events with delay time window from 10 to 500 μs . Insert: blue filled area - delayed event spectrum with delay time window from 1.8 to 10 μs (i.e. neutron events), asymmetrical peak shape caused by prompt signal influence on delayed signals; red squares - delayed event spectrum with shifted delay time window (11.8 to 20 μs); dots with error bars - calibration with ^{139}Ce (experimental calibration spectrum scaled down to match amplitude of delayed event spectrum). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Ambient neutron measurements using the aforementioned spectrometer were conducted in one of the buildings of JINR's Laboratory of Nuclear Problems (Dubna, Russia, ~ 120 m above sea level). Simultaneously, a low-background ^3He neutron detector (CHM-57 [13–15]) also performed neutron flux measurements, as shown in Fig. 1. This ^3He detector had a neutron sensitivity, which refers here and hereafter to the isotropic, 4π , $1 \text{ n cm}^{-2} \text{ s}^{-1}$ thermal neutron flux in the detector's absence, of $243 \pm 24 \text{ counts s}^{-1}$.

Fig. 3 displays the experimental energy spectra obtained by the NaI(Tl) detector over the course of 42 days of measurements from May to June 2016. As evident from these spectra, it is impossible to discern neutron events from background signals without using coincidence detection techniques to distinguish the delayed event line in the 137 keV energy region. The $T_{1/2}$ of $825 \pm 12 \text{ ns}$ (Fig. 4), obtained from measurements for this energy level, agrees with the tabulated value of $845 \pm 20 \text{ ns}$ [10]. Since systematic effects on the determination of $T_{1/2}$, such as event time precision, were not studied in this work, the acquired $T_{1/2}$ value should not be considered an improvement of the tabulated one. The ^{214}Bi - ^{214}Po peak in coincidence events in data from measurements with a delay time window of 10–500 μs helps to indirectly verify the delay time window approach to identifying coincidence events. The experimentally acquired $T_{1/2} = 168.3 \pm 10.8 \mu\text{s}$ for ^{214}Po agrees with the tabulated value of $164.3 \pm 2.0 \mu\text{s}$ [10].

Fig. 5 compares the neutron flux measured by the $^{63}\times^{63}\text{NaI}$ detector with that measured simultaneously by the low-background proportional ^3He counter, thereby demonstrating the sensitivity of the NaI-based neutron detection approach. Both spectrometers detected the small changes in ambient neutron flux, on the level of $10^{-3} \text{ n cm}^{-2} \text{ s}^{-1}$, caused by the operation of one of JINR's acceleration facilities. Based on analyses of the simultaneous measurements from the two detectors, the absolute sensitivity of the $^{63}\times^{63}\text{NaI}$ spectrometer was found to be $6.5 \pm 1 \text{ counts s}^{-1}$ for a neutron flux of $1 \text{ n cm}^{-2} \text{ s}^{-1}$ (without accounting for loss of effectiveness due to the 1.8 μs trigger holdoff). Meanwhile, the number of coincidence events occurring within a delay time window of 1 μs was determined to be 0.8 day^{-1} based on the constant background signal for a delay time greater than 10 μs (see Fig. 4), as

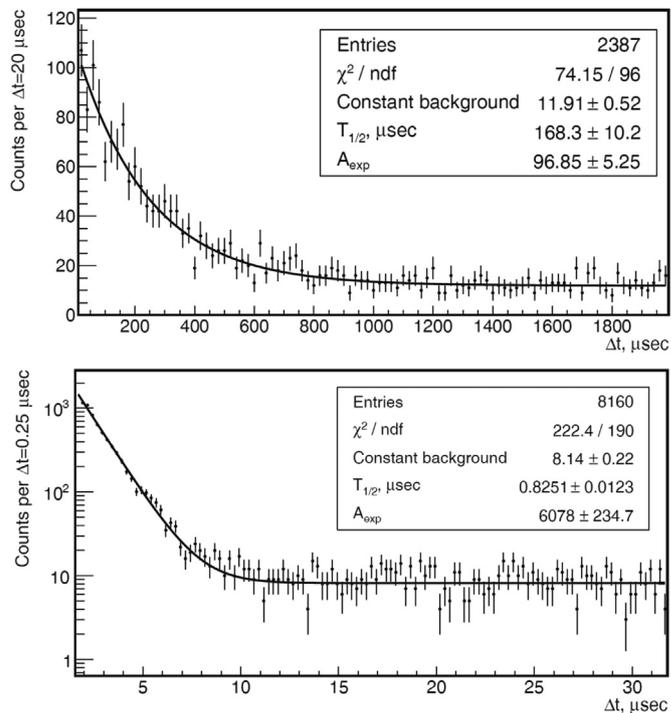


Fig. 4. Delayed coincidences. Upper graph shows ^{214}Po 's α -decay. Lower decay curve illustrates 137 keV level's $T_{1/2}$. On both plots, dots with error bars correspond to observed coincidences with particular delay times. Solid lines show the fit of the function: $f = \text{constantbackground} + A_{\text{exp}}e^{ln(2)t/T_{1/2}}$, from which the half-lives of the data sets can be determined.

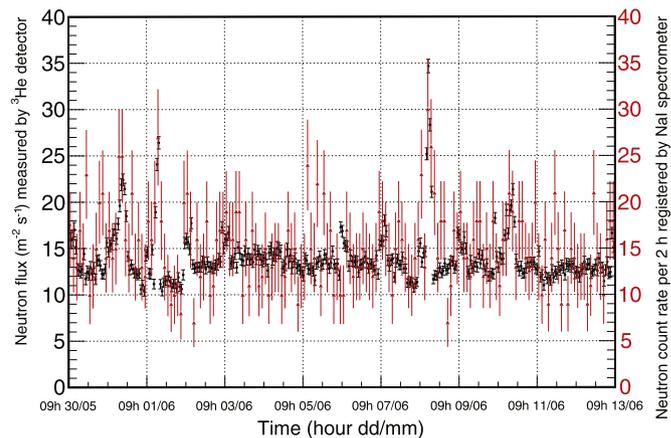


Fig. 5. Comparison of neutron flux measured by ^3He detector and number of neutron events registered by NaI spectrometer (from measurements taken in May–June 2016). Black dots with error bars - neutron flux from ^3He detector; red triangles with error bars - neutron count rate from $^{63}\times^{63}\text{NaI}$ spectrometer.

well as on matching calculations with known event count rates above the energy threshold and in the 137 keV energy region.

4. Discussion

Multiplying the event count rates in the prompt and delayed energy regions yields the frequency of coincidence events comprising the background signal for the suggested method of neutron measurement, which is almost quadratically proportional to the overall background signal. Thus, the proposed method is especially sensitive in low-background experiments that use iodine-containing detectors with masses of 10 s or 100 s of kilograms as active veto systems or as the main detectors. Some examples of these types of experiments include the DAMA/LIBRA dark matter search experiment, which uses a 250 kg

NaI(Tl) detector [6]; the KIMS dark matter search experiment, which implements a 103 kg CsI(Tl) detector [16]; the ν GeN reactor neutrino nuclear coherent scattering experiment, which uses an approximately 400 kg NaI(Tl) anti-Compton veto system [17]; and the COHERENT experiment, which uses a 14 kg CsI(Na) detector to search for coherent elastic neutrino scattering from the Spallation Neutron Source [18].

In the aforementioned experiments, neutrons comprise one of the most uncertain sources of background events. Considering the sensitivity calculated for the $^{63}\times^{63}\text{NaI}$ scintillator described in this work, it is also possible to estimate the ability to detect the level and stability of thermal neutron flux in low-background experiments. For a thermal neutron flux on the level of $10^{-8} \text{ n cm}^{-2} \text{ s}^{-1}$, approximately 1 neutron per day can be detected with a 100 kg NaI detector (the precise number can only be estimated after accounting for the influence of the detector's geometry on neutron capture efficiency and the detection of prompt and delayed signals). For many low background experiments, only the detection of fast neutrons, which is a difficult and often even impossible task, is of interest. However, since no thermal neutron sources exist in nature, the thermal neutron flux is always in equilibrium with the fast neutron flux. Thus, despite the typically unknown ratio of fast to thermal neutrons, the measurement of thermal neutrons provides useful information about the total neutron flux and stability. For example, in [6], the upper limit on the thermal neutron flux passing through the multicomponent DAMA/LIBRA shield was determined to be: $<1.2 \times 10^{-7} \text{ cm}^{-2} \text{ s}^{-1}$ (90% C.L.).

The majority of existing neutron detectors are used for nuclear safeguards, security, and reactor instrumentation. The “golden standard” among such detectors is the ^3He -filled proportional counter with its high neutron detection efficiency and simultaneously low sensitivity to γ -radiation. A thorough comparison of NaI detectors with ^3He detectors in the particular case of neutron portal monitors already exists in [4]. Along with all other iodine-containing detectors, NaI spectrometers with comparable active volumes to ^3He -based portal monitors have similar detection efficiencies and minimum detectable activities. The strong suppression of γ -background radiation marks the main advantage of the delayed coincidence approach to the detection of neutrons via an NaI (CsI) spectrometer. At the same time, the $1.8 \mu\text{s}$ trigger holdoff and the fact that not all of the transitions pass through the delayed 137 keV level reduce the efficiency by a factor of ~ 10 , which can be prevented through improved data acquisition and/or a different light detector. To implement the proposed approach in a high intensity neutron field, the important, degrading effect of high-dose fast neutron bombardment on crystals must be taken into account.

For large detectors, such as that used in [4] or larger, a subsequent measurement of the total, high-energy transition cascade (above the natural radioactivity threshold) coupled with measurements from the delayed 137.8 keV energy level can effectively suppress background signals, even with a high γ -count rate. This differs from the NaI detector used in this experiment and other relatively small detectors, where only a fraction of events in the high-energy cascade from 6.8 to 137.8 keV are registered.

Another important point of discussion is the detection of neutron events with a high multiplicity (neutrons from spontaneous fission, those produced by high energy muons in surrounding detector materials, etc). The proposed method can measure such events only after careful consideration and changes in the experiment: conducting the experiment with a short delay time window for prompt-delayed events of $<3 \mu\text{s}$ would likely still not exceed the average time between neutron production and capture in the detector.

The decay process of ^{128}I has another notable excited state at 167.4 keV, which has a $T_{1/2}$ of $175 \pm 15 \text{ ns}$ (Table 2). This energy level is emptied exclusively via a 29.6 keV transition to the aforementioned 137.8 keV energy level, providing a potential way to radically suppress background signals through triple coincidence measurements. However, this goal necessitates the detection of delayed events in a time window of 100 s of ns. This is a difficult task when using NaI, and

especially CsI, scintillators because they have a decay time of ~ 200 –500 ns. Triple coincidence measurements were not investigated in this research.

A similar neutron detection method can be implemented for Ge-detectors by using ^{73}Ge 's delayed transitions from the 13.3 keV ($T_{1/2} = 2.95 \mu\text{s}$) and 66.7 keV ($T_{1/2} = 0.499 \text{ s}$) energy levels, where the latter is only helpful in ultra-low-background measurements. However, it is important to note several experimental differences: ^{72}Ge 's thermal neutron capture cross section of 0.98 b is 6 times smaller than that of ^{127}I , and the ^{72}Ge isotope only comprises 27.7% of the atoms in $^{\text{nat}}\text{Ge}$. Furthermore, unlike ^{128}I , ^{73}Ge is a stable isotope, so it can be excited to the aforementioned energy levels by fast neutron inelastic scattering, causing ambiguous results. For low-background neutron measurements, the cosmogenic isotope ^{73}As 's decay needs to be accounted for in the analysis. Also, in a Ge-detector it is necessary to consider the influence of the cold cryostat, the amount of nitrogen in the Dewar vessel, and the vessel itself on the incoming neutron energy distribution (i.e. the neutron capture cross section).

5. Conclusion

In summary, this work proposes a fundamentally new neutron detection technique involving spectrometers with existing, iodine-containing (NaI or CsI) scintillation detectors. The additional use of delayed coincidence techniques allows for the highly effective discrimination of neutron events from background signals. The precise measurements from a simple, inexpensive NaI(Tl) spectrometer of the ambient neutron flux at sea-level demonstrate the sensitivity of this technique.

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