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APPARATUS

Study of the Separation of Neutrons and Gamma Quanta by $LiInSe_2$ Crystals Using Highly Enriched ^{6}Li Isotope

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Abstract—The results of studying two $^6LiInSe_2$ single crystals using highly enriched ^{6}Li isotope are presented. The interaction of neutrons with crystals has been calculated with the aid of the Geant4 toolkit, theoretical values of the detection efficiency were obtained, and a comparison with a ^{3}He detector was carried out. The possibility of separating neutrons and gamma quanta by the pulse-shape discrimination method is considered.

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INTRODUCTION

$^6LiInSe_2$ single crystals are a promising material for thermal-neutron detection due to their unique physical properties. Unlike other converters, $^6LiInSe_2$ crystals have a number of advantages, such as a high theoretical efficiency due to the large neutron capture cross section of ^{6}Li nuclei, compactness, low level of noise [1], as well as a wide temperature range of stable operation.

Neutron detection using a $^6LiInSe_2$ crystal is based on the nuclear reaction $^6Li(n,\alpha)^3H$. The high energy, released in this reaction (about 4.78 MeV), allows one to efficiently detect signals from neutrons and gamma radiation [1]. A high neutron detection efficiency (more than 50%) is achieved when using thin single crystals due to the mean free path of thermal neutrons (920 μm) [2] in $^6LiInSe_2$. The neutron signal is generated in a $^6LiInSe_2$ crystal as a result of the ionization by alpha particles and tritons, whereas gamma quanta interact with the material much weaker, generating signals of smaller amplitude and another shape. This circumstance allows for the use of pulse-shape discrimination (PSD) methods to efficiently select neutron events against the gamma radiation background.

Optimization of the process of $^6LiInSe_2$ crystal growth is currently a key line of research. In order to reduce the number of lattice defects, crystals are subjected to a high-temperature annealing. This procedure increases the neutron capture and detection efficiency and improves the separation of neutrons and background signals, as was shown in [3, 4]. The use of the highly enriched ^{6}Li isotope in $^6LiInSe_2$ single crystals significantly increases the thermal neutron detection efficiency.

In this work, we performed a comparative analysis of the $^6LiInSe_2$ single crystal, unannealed and annealed in a selenium atmosphere, for its future application as a semiconducting thermal neutron detector.

The purpose of the study was to perform computation of the interaction of neutrons with crystals using the Geant4 toolkit in order to obtain theoretical values of the detection efficiency and compare them with the ^{3}He detector efficiency, to determine the detection efficiency using ^{252}Cf neutron source, and to evaluate the possibility of separating neutrons and gamma quanta by the pulse-shape discrimination method.

SIMULATION

The interaction of neutrons with a $^6LiInSe_2$ crystal was simulated in the Geant4 (version 4.10.p02) toolkit using the QGSP_BERT_HP physical sheet (a set of physical models) [5]. In the simulation, we considered

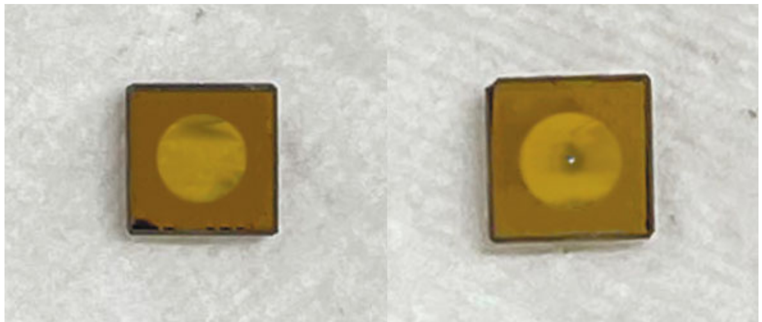

Fig. 1. $^6LiInSe_2$ crystals: (a) raw and (b) annealed in a selenium atmosphere.

a simple geometry, in which, a neutron beam with a cross-section of 5 × 5 mm is incident on a detector ($LiInSe_2$ crystal 5 × 5 × 1.5 mm in size or a ^3He counter with a diameter of 18, 140 mm at a gas pressure of 2.8 atm) at right angle to the detector surface. The conversion efficiency was simulated for two neutron wavelengths: 1.8 and 4.0 Å. The calculation results showed that the neutron conversion efficiency (the $^6Li(n,\alpha)^3H$ capture process) for a crystal with a natural ^6Li content (7.5%) is 6.9 and 12.8% for neutrons with wavelengths of 1.8 and 4.0 Å, respectively. A crystal with a 100% level of enrichment with the ^6Li isotope was used in the calculations, which made it possible to determine the maximal theoretical conversion efficiency: 59.5 and 77.1%. This result allows for evaluation of the maximum neutron detection efficiency of the detector. The efficiency of a ^3He counter is 73.1 and 93.8% at the aforementioned wavelengths.

Similar values of the theoretical detection efficiency of neutrons with a wavelength of 4 Å were reported in [6]; it was evaluated to be up to 95% when using the ^6Li isotope in $^6LiInSe_2$ single crystals.

EXPERIMENTAL

In the previous study devoted to $^6LiInSe_2$ single crystals [4], the application of natural lithium (and, as a consequence, low content of the ^6Li isotope) was one of possible reasons for the low efficiency of thermal-neutron detection. In this study, $^6LiInSe_2$ single crystals were applied to increase the neutron detection efficiency; the raw material for growing these crystals was highly enriched (95%) with the ^6Li isotope. $^6LiInSe_2$ single crystals were grown by the Bridgman–Stockbarger vertical method [7].

Two 1.5-mm-thick plates 5 × 5 mm in size were cut from the grown crystal. One of the crystals was not subjected to additional treatment. The second was annealed in a selenium atmosphere at a temperature close to 800°C. Both crystals were optically transparent in visible light and similar to the crystal studied in [8]. Their photographs are shown in Fig. 1.

A 1000-Å-thick gold coating was deposited on the crystals as contact pads, to which gold electrical contacts with a diameter of 40 μm and a length of 4 mm were soldered. Since the length of contacts didn't allow for their connection to the preamplifier, a new connection technique was used. It consists in fixing a crystal between two textolite panels with contact pads, which provides a reliable electrical contact between the planar surfaces of single crystals and feeding electrodes. A layer of highly conductive glue was deposited as a conductor between the crystal and textolite for relaibility (Fig. 2b).

The measurement procedure consisted in irradiation of the measurement cell of a $^6LiInSe_2$ single crystal by a thermal neutron flux (^{252}Cf source (10^5 n/(cm^2 s)) + 100-mm polyethylene) and registration of signals from the detector anode (Fig. 2a). The distance from the source to the crystal was 200 mm. To measure the sensitivity to gamma radiation, the single crystals were irradiated by a flux of gamma quanta from a ^{60}Co source. The source was placed on the measurement-cell surface. To isolate the necessary irradiation area, shield blocks and diaphragms made of cadmium and lead were used. The measurements were performed at room temperature for 7200 s.

A Digitizer CAENN6730 with the PSD firmware [9], as well as a fast amplifier with an integration constant *RC* of 20 ns and a differentiation constant *CR* of 200 ns were used in the work. The block-diagram of measurements is shown in Fig. 2.

PULSE-SHAPE DISCRIMINATION METHOD

The pulse-shape discrimination method consists in integration of a digitized input pulse in two time windows relative to the discriminator start and establishment of a relationship between them, which allows one to separate the contributions of the neutron and gamma radiation signals [10, 11].

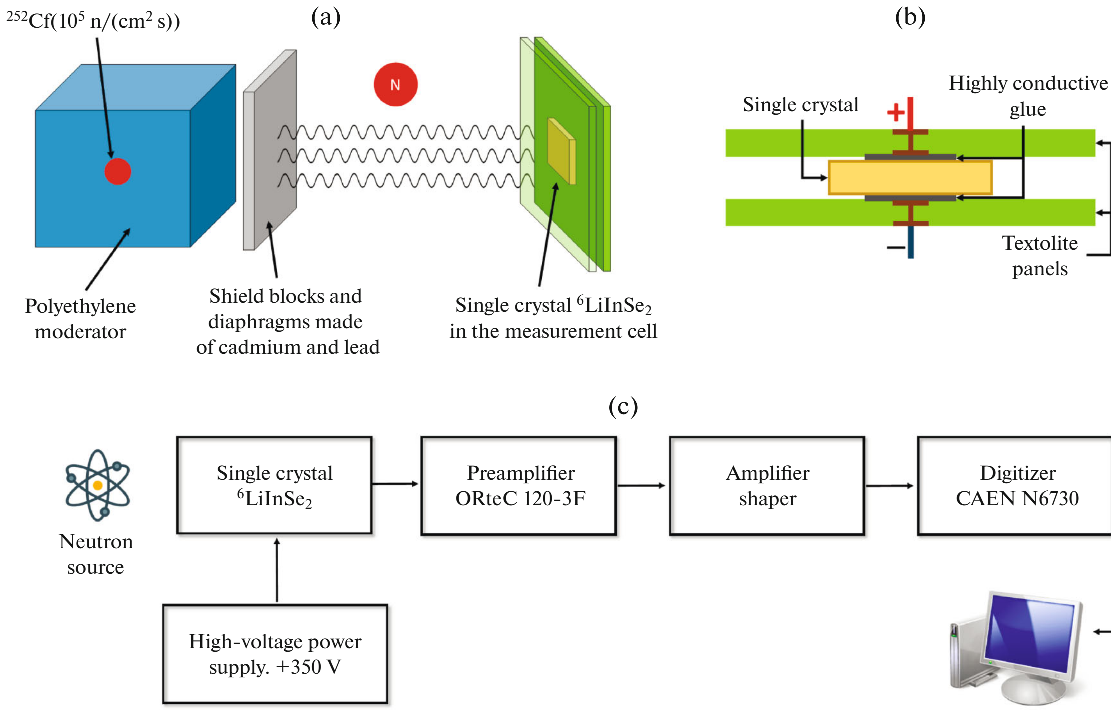

Fig. 2. (a, c) Measurement layout and (b) measurement cell based on ^{6}LiInSe$_2$ crystals.

The application of two time windows is depicted in Fig. 3, where the shape of a neutron signal is presented as an example. It was found that a short 100-ns time window and a long 500-ns window provide an optimal quality of separation, characterized by the PSD_p parameter. The charge integrated in the long window is used to form energy spectra. The PSD_p parameter is given by the formula

$$PSD_p = (Q_{\text{long}} - Q_{\text{short}})/Q_{\text{long}}, \quad (1)$$

where Q_{short} and Q_{long} are the charges integrated in the short and long windows, respectively.

RESULTS

Figures 4 and 5 show the energy spectra and the pulse-shape discrimination spectra of the ^{6}LiInSe$_2$ crystal; Figs. 6 and 7 present similar spectra for the annealed crystal. The spectra were obtained using a ^{252}Cf neutron source and a cadmium filter.

It follows from the analysis of the energy spectra of the crystals (Figs. 4, 6) that the energy discrimination doesn't allow one to separate overlapping spectra of neutrons and gamma quanta. Two peaks are observed in the pulse-shape discrimination spectra (Figs. 5, 7), each corresponding to a certain type of radiation.

A rough estimate of the measurement results showed that the average charge formed when detecting one neutron in the crystal is on the order of 6300 electrons. A charge per neutron was calculated using the average amplitude at the Digitizer input and the integration coefficient (pK/V) per channel. It was found that 720 eV is required to form a single pair of carriers at energy of 4.7 MeV released in the crystal as a result of the reaction. The noise in the electronic devices is on the order of 300 electrons (mean-square devia-

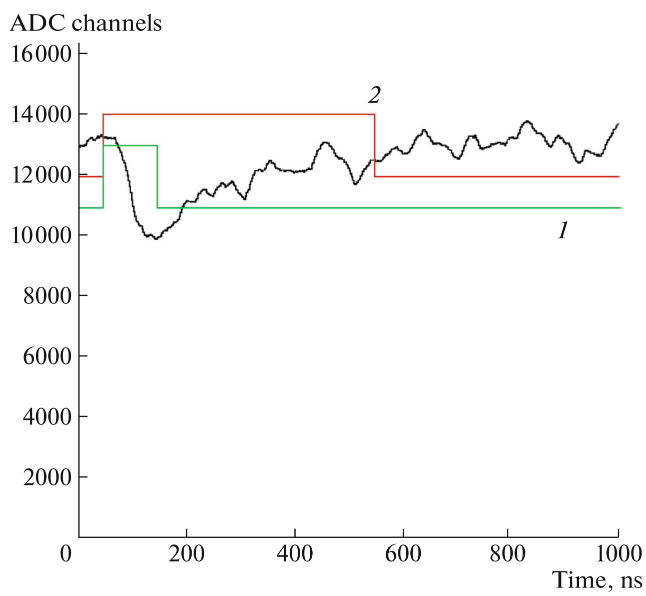

Fig. 3. Shape of the signal obtained by applying (*1*) short (100 ns) and (*2*) long (500 ns) time windows; 1 channel = 0.12 mV.

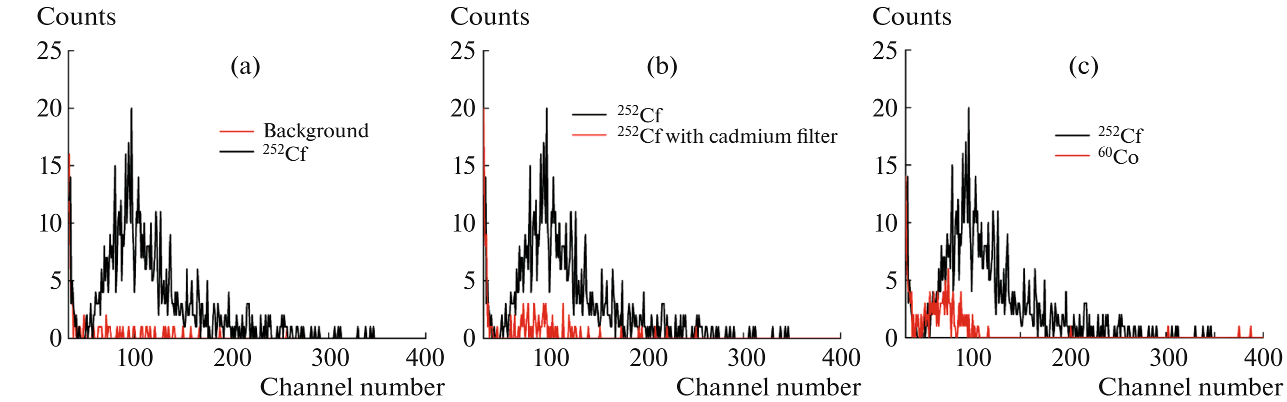

Fig. 4. Energy spectra of the unannealed crystal, measured using (a) a ^{252}Cf neutron source, (b) a Cd filter, and (c) a ^{60}Co source.

tion), which suggests a very small level of measured signal.

COMPARATIVE MEASUREMENT OF THE NEUTRON DETECTION EFFICIENCY

As previously in [4], a Gelii-18/190-8.0/OTs gas-filled helium neutron detector [12] was applied to estimate the neutron detection efficiency with the aid of crystals. The helium detector was mounted at the same position as the crystals; the distance from the source to the detector was 200 mm. A 2-mm-thick cadmium filter with a cut window 5 × 5 mm in size, which corresponded to the crystal area, was mounted before the helium detector. Therefore, the neutron detection efficiency was successfully estimated using crystals relative to the helium neutron counter; it was found to be 1.2% for the crystal without annealing and 4% for the crystal annealed in a Se medium.

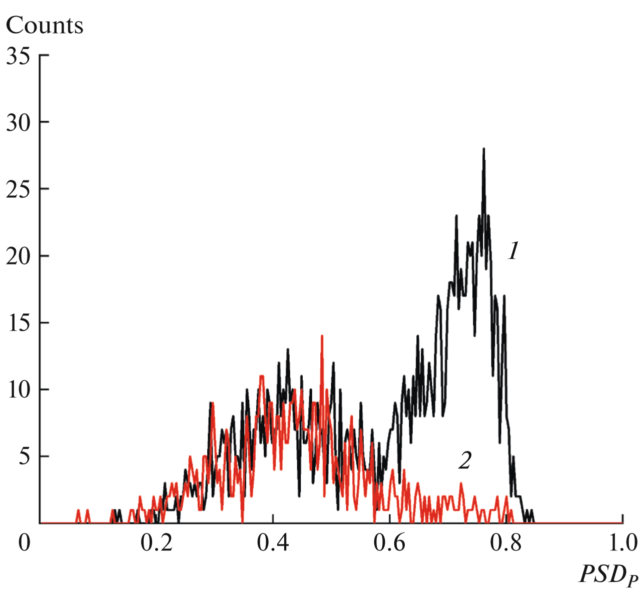

Fig. 5. Pulse-shape discrimination spectra of the unannealed crystal, recorded using a ^{252}Cf neutron source (*1*) without a filter and (*2*) with a cadmium filter.

The detector efficiency was determined by comparing the numbers of neutrons detected by the helium neutron counter and using $^6LiInSe_2$ crystals.

CONCLUSIONS

$^6LiInSe_2$ single crystals with a highly enriched ^{6}Li isotope are a promising material for designing efficient and stable detectors of thermal neutrons. The application of these crystals in combination with the modern detection technologies makes it possible to design devices with improved sensitivity and efficiency.

Two crystals, grown by the Bridgman–Stockbarger vertical method using highly enriched ^{6}Li isotope, were studied. A simulation of the interaction of neutrons with the crystal was performed, which showed neutron absorption at a level of 60%. Energy spectra and pulse-shape discrimination spectra of the crystals were recorded, which indicate that the crystal annealed in a selenium atmosphere has better characteristics as compared to the unannealed crystal, which is related to the crystal processing conditions. A possibility of separating signals of neutrons and gamma radiation from a ^{252}Cf source was also demonstrated. The results of studying the detection efficiency showed that the efficiencies for the annealed and unannealed crystals are, respectively, 4 and 1.2%.

One of the possible reasons for the low efficiency of neutron detection using crystals is the small signal magnitude, which is due to the high energy necessary to form a single pair of carriers in the crystal. The experimental magnitude on the order of 720 eV, which is very high, hinders neutron detection and negatively affects efficiency.

The quality of grown $LiInSe_2$ crystals is of great importance for the efficiency and reliability of neutron detectors. A detector based on a crystal 5 × 5 ×

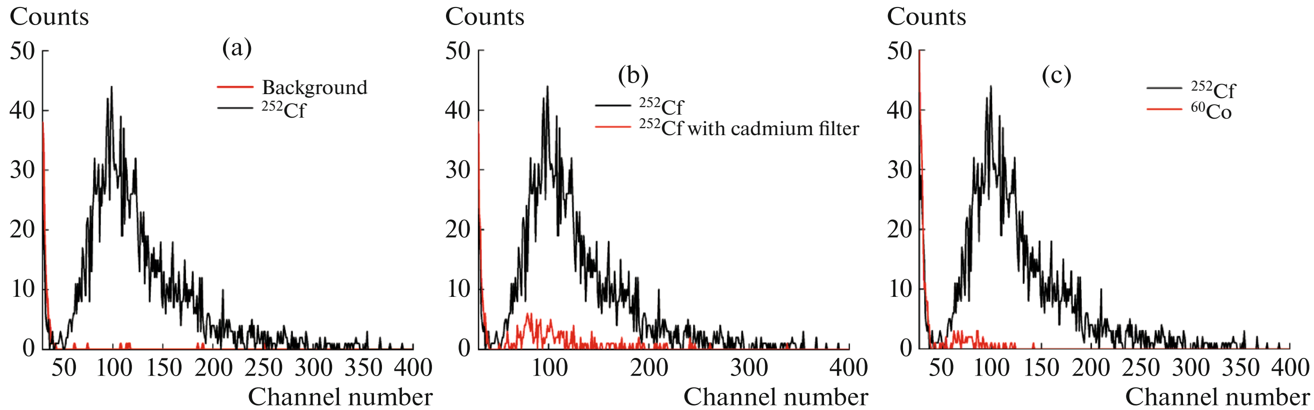

Fig. 6. Energy spectra of the annealed crystal, measured using (a) a ^{252}Cf neutron source, (b) a Cd filter, and (c) a ^{60}Co source.

0.56 mm in size, enriched with ^{6}Li isotope up to 95%, was demonstrated in [2]; it showed a neutron detection efficiency in individual pixels from 5 to 65%, whereas the average efficiency was 38%. It was noted that, improving the quality of crystal growth, one can minimize the significant difference in the area distribution of crystal efficiency, which was explained by the presence of intrinsic defects and inhomogeneities in the crystal. It should be also pointed out that a 0.56-mm-thick crystal was studied in [2], whereas 1.5-mm-thick crystals were applied in this work; the difference in thickness may also affect the detection efficiency.

It is worth noting the following. To use such crystals in a detection system, high-quality contact pads and electrodes must be formed, as was done in [13]. In this work, the quality of the contact pads on $LiInSe_2$ crystals may be insufficient, which reduces the detector operating efficiency. A high resistance at the metal–semiconductor interface leads to signal loss, which reduces the detector sensitivity. Optimization of contacts and synthesis conditions is a key factor for operation of the neutron detectors based on $LiInSe_2$.

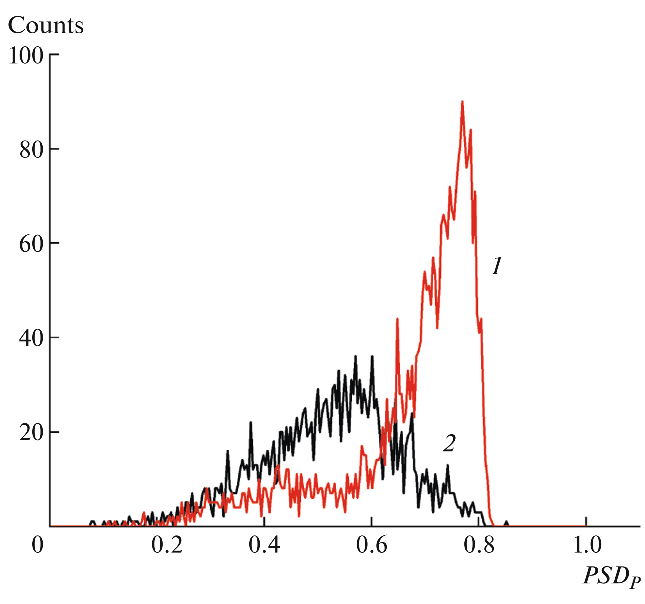

Fig. 7. Pulse-shape discrimination spectra of the annealed crystal, obtained using a ^{252}Cf neutron source (*1*) without a filter and (*2*) with a cadmium filter.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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