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Gamma Sensitivity of Single-Crystal CVD Diamond Neutron Detectors

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Abstract—We have studied the gamma sensitivity of single-crystal CVD diamond neutron detectors using a ^{252}Cf neutron source placed in a moderator. It has been shown that a major contribution to the count rate of the detectors is made by the gamma rays from the source. We have compared the count rates of a detector with a ^{10}B boron isotope-based slow-neutron converter and without it. With allowance for the theoretically calculated detection efficiency, the difference between the count rates is consistent with the fraction of slow neutrons measured using a scintillation detector.

Keywords: ^{252}Cf source, ^{10}B boron isotope, slow-neutron converter
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INTRODUCTION

The ability to monitor a neutron flux is critical not only for nuclear power generation but also for a variety of scientific and industrial applications. All types of neutron detectors in current use have some drawbacks, which limit their application field. In particular, fission chambers contain fissionable materials, which places special safety requirements and complicates handling of them. Silicon diodes and scintillation detectors have a relatively low radiation resistance and limited temperature range. Gas chambers are cumbersome and sensitive to the gamma-ray background. At the same time, diamond is regarded as an attractive material for the fabrication of particle and radiation detectors intended for operation under harsh conditions [1]. In contrast to silicon detectors, diamond detectors possess high radiation resistance, low noise, and low leakage currents even at elevated temperatures. As a rule, diamond detectors are produced by chemical vapor deposition (CVD). CVD technology makes it possible to synthesize high-quality diamond crystals with an energy resolution of 0.3% [2], which approaches the 0.15% resolution of silicon detectors.

It is worth noting that CVD diamond neutron detectors have been the subject of extensive studies [3–9]. Interest in this material is aroused by the fact that the small nucleus charge $Z = 6$ means that diamond has lower gamma sensitivity, and its high radiation resistance allows it operate at high neutron and gamma fluxes. In the core region of nuclear reactors, the neutron energy ranges from thermal values of 25 meV to fast neutron energies of several megaelectronvolts. Fast neutrons have a small cross section of interaction with matter (no greater than a few barns) and lose little energy when scattered by nuclei in a detector. To detect slow neutrons, use is made of a converter: material that has a large absorption cross section for slow neutrons and decays into charged particles. A converter layer is applied to a CVD crystal. Absorption of a neutron in such a layer produces secondary charged particles, which reach the diamond crystal and create electron–hole pairs. There are reports on studies of diamond neutron detectors with converters based on the ^{6}LiF [4–7], ^{10}B [8–10], and ^{235}U [11] isotopes. In studies where detectors were tested in research nuclear reactors, the count rate of CVD diamond detectors correlated well with the reactor power and data from standard neutron detectors.

Among the above converter materials, the ^{10}B boron isotope possesses the largest absorption cross section for slow neutrons, 3587 b, but the lowest energy of decay products: 2.79 MeV. This means higher detection efficiency and weaker degradation of diamond crystals at high neutron doses. For example, the crystal in ^{6}LiF detectors was observed to suffer irreversible changes starting at neutron fluences above

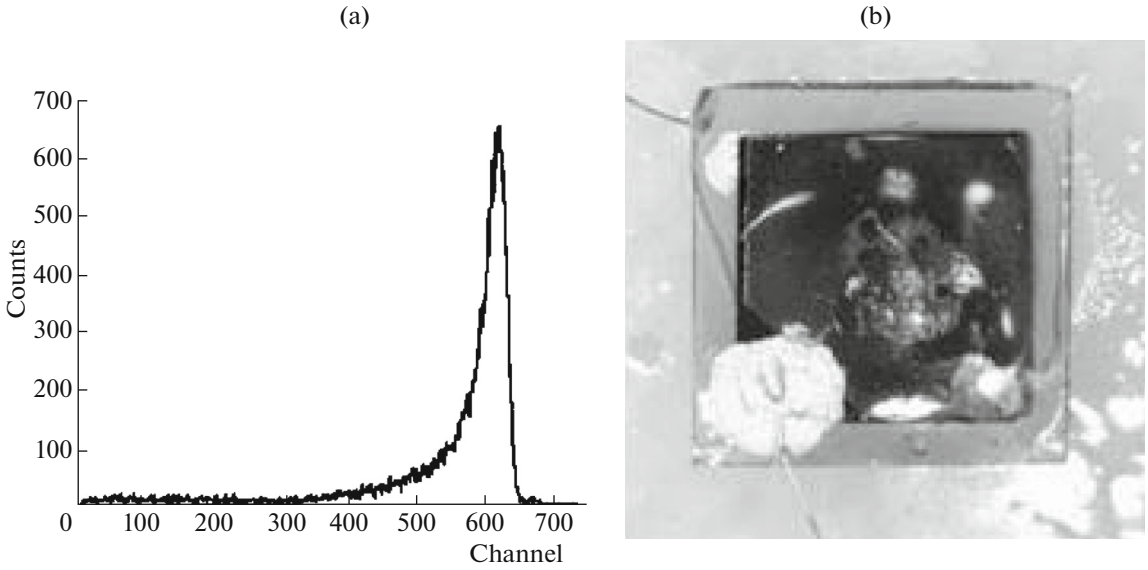

Fig. 1. (a) Characteristic spectrum of the CVD plates under irradiation with a ^{239}Pu alpha source (energy of alpha particles, 5.157 MeV) at a bias of 200 V; (b) detector based on a CVD crystal with a boron oxide layer.

2×10^{16} cm^{-2} [7]. An even faster degradation rate is expected in the case of heavy isotopes. On the other hand, the lower energy release in the case of the ^{10}B(n, α) reaction means a smaller signal amplitude in the detector (the formation of one electron–hole pair in diamond requires about 13 eV), which makes neutron signals difficult to isolate from the gamma-ray background. In this case, it is, therefore, important to understand the response of diamond detectors to the gamma-ray background in order to optimize the detector design.

In this paper, we report a study of single-crystal CVD diamond detectors on a ^{252}Cf neutron source in a moderator. We tested detectors with a ^{10}B-enriched boron oxide converter and without it.

EXPERIMENTAL

We investigated two electronic-grade single-crystal CVD diamond plates (manufactured by Element Six) [13] 3.5 × 3.5 mm^2 in area and 0.5 mm in thickness. The nitrogen and boron concentrations in the plates were no higher than 5 and 1 ppb, respectively. On both sides of the plates, Ti/Al contact layers 50/100 nm in thickness were made by magnetron sputtering in Ar at a pressure of 1333 Pa. The layers were grown through a mask with a 2.5 × 2.5 mm^2 window, which ensured a 0.5-mm separation between the edges of the contact pads and the lateral faces of the plates.

After metallization, the operation of the detectors was tested on a ^{239}Pu alpha source, whose characteristic spectrum is shown in Fig. 1a. The spectra obtained demonstrate that both crystals have high charge collection efficiency. In this case, it is limited by characteristics of the amplifier and a bias of 0.4 V/μm, at which charge collection does not reach a maximum.

Next, boron oxide (B_2O_3) layers about 2 μm in thickness were grown on both sides of one of the plates by vacuum evaporation through a 2.5 × 2.5 mm mask. The starting material used was boric acid 90% enriched in the ^{10}B isotope. The acid was preheated in vacuum until complete dehydration. To protect the plate from atmospheric moisture, an epoxy lacquer layer was applied to the contact pads. Figure 1b shows a detector based on a CVD crystal with a boron oxide layer (The boundary of the layer is seen where a wire is attached to the upper contact pad).

RESULTS AND DISCUSSION

The detectors were tested on a ^{252}Cf neutron source. Given the fraction of ^{252}Cf decayed during the storage of the source, the neutron radiation intensity in our experiments was $(1.2 \pm 0.1) \times 10^6$ s^{-1}. The source was placed in the center of a polyethylene moderator in the form of a cylindrical vessel 14 cm in radius, with a wall thickness of 11 cm. The diamond detectors, placed in shielding metallic boxes, were located about 1 cm from the moderator. The detectors were connected to an amplifier and multichannel analyzer, and spectra were measured for 1 h. To assess the effect of gamma radiation, we performed measurements using bismuth plates with a total thickness of 2 cm, located between the moderator and detectors.

The measured count rate of the detectors is indicated in the table. The multichannel analyzer was calibrated in energy using a ^{239}Pu alpha source. Signals with an energy under 0.1 MeV were left out of account

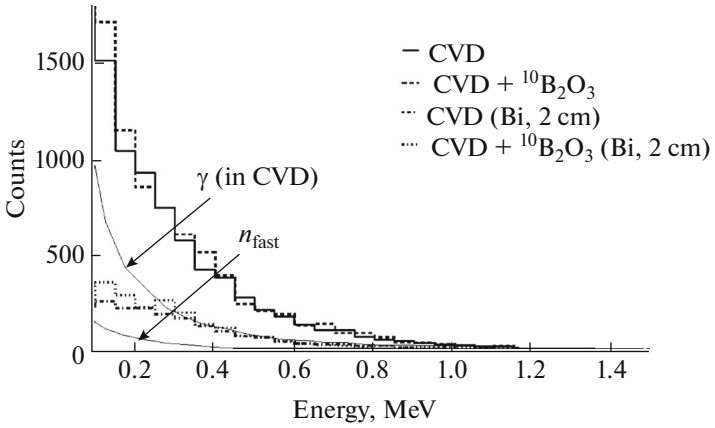

Fig. 2. Energy distribution (0.05 MeV steps) of CVD detector signals obtained using a ^{252}Cf source in a moderator. The lower, smooth curves show a calculated response of the detector to fast neutrons (n_{fast}) and gamma rays (γ) scattered in the CVD crystal (without filters).

because of the possible noise in this range. The table indicates the statistical error in count rate, evaluated as the square root of the number of counts detected over the measurement time.

It is seen from the table that the detectors with and without a $^{10}B_2O_3$ converter have essentially identical count rates over the entire energy range of their response (Fig. 2). On the other hand, the count rate of the radiation that passed through the bismuth layer was a factor of 4 lower. This suggests that a considerable fraction of the diamond detector signals originate from the gamma source. The mean free path of gamma-ray photons in bismuth reaches its maximum level, 2.42 cm, at an energy near 4 MeV [14], so 2 cm of bismuth reduces the gamma intensity by at least a factor of 2.3.

Nevertheless, in both cases we observe a difference of ~0.1 s^{-1} between the count rates, which is near three standard deviations. Such a difference may arise from the difference in properties between the two crystals, metallizations, etc. To exclude this factor, two silicon wafers 10 × 10 mm^2 in area and 0.2 mm in thickness, with $^{10}B_2O_3$ deposited on their surfaces under the same conditions as in the fabrication of the latter detector, were tightly applied to the two surfaces of the detector

Count rate of pulses with an energy >0.1 MeV from a ^{252}Cf source in a moderator and difference of the count rates of the detector with a $^{10}B_2O_3$ layer and without it

Filter	Detector	Count rate, cps	Difference, cps
No	CVD + $^{10}B_2O_3$	2.04 ± 0.02	0.11 ± 0.03
	CVD	1.93 ± 0.02	
Bi, 2 cm	CVD + $^{10}B_2O_3$	0.54 ± 0.01	0.08 ± 0.03
	CVD	0.46 ± 0.01	

with no converter layer. The count rate measured under the same conditions as above was higher by 0.16 ± 0.03 s^{-1}. It is worth noting that boron oxide was applied to the entire surface of the silicon wafers, whereas on the second detector it was only present over the contact pads, 2.5 × 2.5 mm^2 in area. Therefore, the sensitivity of the former detector, with silicon wafers, to slow neutrons may exceed that of the detector with $^{10}B_2O_3$ deposited directly onto the contact pads owing to the larger area of the oxide.

The present results lead us to conclude that the difference in count rate between the detectors arises from the interaction of slow neutrons with ^{10}B in the converter layer, rather than from a difference in characteristics between the detectors. Its low value compared to the background can be explained by the small fraction of thermal neutrons compared to gamma rays in our experiments. To confirm this conclusion, below we analyze the contribution of each kind of radiation to the response of the diamond detectors and describe slow-neutron intensity measurements with a scintillation detector.

Fast neutrons. An 11-cm-thick polyethylene layer is insufficient to reduce the neutron energy to thermal values. After a few collisions, some of the neutrons leave the moderator, and the rest of the neutrons slow down to thermal energies of ~0.025 eV and move along chaotic trajectories until they leave the moderator or become absorbed by hydrogen nuclei. The fraction and spectrum of fast neutrons that leave the moderator and their contribution to the response of the detector were evaluated by the Monte Carlo method using cross sections indicated in the ENDF library [15].

There are on average 3.7 neutrons per spontaneous ^{252}Cf nucleus fission. The neutron spectrum was measured by Meadows [16] and is well represented by a Maxwellian distribution, $N(E) \sim E^{1/2}\exp(-E/T)$, with $T = 1.565$ MeV. Simulation results obtained with this

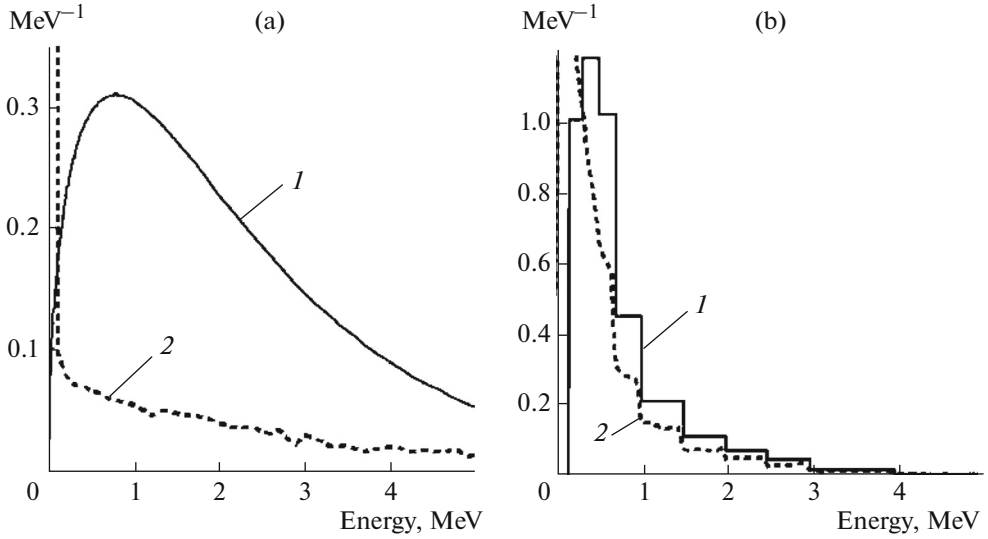

Fig. 3. Simulated energy distributions of (a) neutrons and (b) gamma rays from a ^{252}Cf source: (*1*) input spectra, (*2*) at the moderator output. The distributions are normalized to unit area.

distribution indicate that about 30% of the fast neutrons leave the moderator (Fig. 3a). The rest of the neutrons slow down to thermal energies in the polyethylene. The fraction of neutrons absorbed by hydrogen was not evaluated.

When penetrating a diamond crystal, a neutron with an energy under 4.8 MeV may interact with carbon nuclei only elastically [12]. As a result of scattering, the carbon nucleus involved acquires a kinetic energy $E_C = 0.28E_n(1 - \cos\theta)/2$, which depends on the scattering angle θ in the center-of-mass system. At energies $E_n \sim 1$ MeV, scattering is nearly isotropic, so interaction with monoenergetic neutrons in diamond produces a signal with a uniform energy distribution from zero to $0.28E_n$.

Simulation results indicate that, of all the neutrons emitted by ^{252}Cf, only 0.9% are scattered in diamond. Neglecting events with a released energy under 0.1 MeV reduces the probability to 0.23%, which corresponds to a detector count rate of 0.14 cps under the present experimental conditions.

Gamma radiation. In lightweight materials, the main interaction of gamma radiation in the energy range ~0.1–10 MeV is incoherent (Compton) scattering by electrons of atoms. In heavy elements, the photoeffect prevails up to ~1 MeV. As a result of Compton scattering, an electron acquires an energy from zero to $E_\gamma/(1 + m_e c^2/(2E_\gamma))$ and produces electron–hole pairs when slowing down in the detector crystal. The range of such an electron may reach several millimeters, which leads to considerable uncertainty in calculations of the response of a small detector: one should take into account electrons that result from the interaction of gamma-ray photons with the detector frame and penetrate the CVD crystal.

The gamma spectrum of ^{252}Cf comprises a continuum emitted directly in the course of spontaneous fission and discrete lines due to subsequent decay of fission fragments. The fast gamma spectrum (emitted during the first 10^{-8} s after spontaneous fission) was measured by Verbinski et al. [17] and presented as a table indicating the number of gamma-ray photons at energies in the range from 0.4 to 10 MeV. In recent reports [18, 19], the fast gamma spectrum was presented as plots. There are on average 7.8–8.3 gamma-ray photons per spontaneous ^{252}Cf fission, and the average gamma-ray photon energy is 0.8–0.95 MeV.

The present simulation of the detector response to gamma rays takes into account only the scattering of gamma-ray photons in the diamond crystal of the detector. Because of the small size of the crystal, calculation provides only a qualitative estimate, so it is sufficient to use the steplike distribution in Fig. 3b (constructed using the table from Verbinski et al. [17]) as the input gamma spectrum of ^{252}Cf. In calculating the gamma-ray intensity, we used the ratio $\gamma/n = 8.3/3.7$. Simulation results indicate that 1.9% of the gamma-ray photons are scattered in diamond. Neglecting events with a released energy under 0.1 MeV reduces the probability to 0.6%, which corresponds to a detector count rate of 0.77 cps under the present experimental conditions. Comparison of the simulated detector response to gamma radiation (with no Bi filter) with the measured distribution (Fig. 2) leads us to conclude that a major fraction of detector signals originated from the stopping of electrons knocked out by gamma radiation from the detector frame.

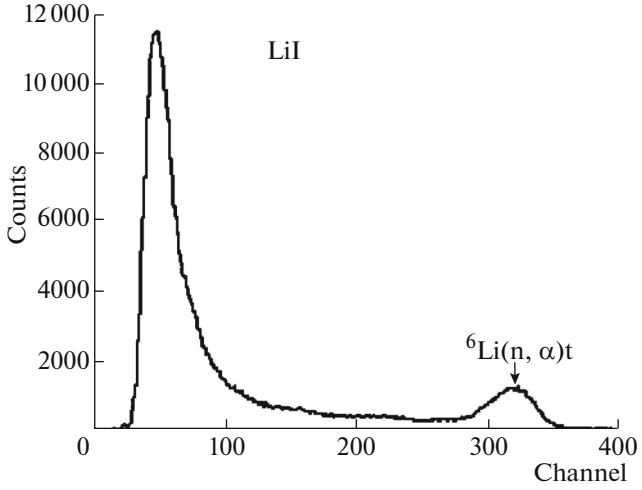

Fig. 4. Spectrum of ^{6}LiI(Eu) scintillation detector signals from a ^{252}Cf source in a moderator. The peak near the 320th channel corresponds to ^{6}Li decay upon absorption of a neutron.

Slow neutrons. The ^{10}B boron isotope has a large absorption cross section for thermal neutrons: σ(2200 m/s) = 3598 b [15]. Absorption leads to decay into ^{7}Li + α, accompanied by energy release of 2.79 MeV. In 94% of cases, the resulting lithium nucleus is in an excited state with an energy 477.6 keV. The kinetic energies of the lithium and helium nuclei are 0.84 and 1.47 MeV, respectively. If absorption in the converter layer occurred near the CVD diamond, one of the nuclei may penetrate the crystal and produce an electric signal in the detector circuit. The probability of detecting a neutron can be evaluated using the following formula:

$$P = \frac{1}{4}\sigma N(^{10}\mathrm{B})(r_\alpha + r_{\mathrm{Li}}),$$

where $N(^{10}\mathrm{B})$ is the concentration of ^{10}B nuclei in the converter material and $r_{\alpha,\mathrm{Li}}$ is the range of the corresponding nucleus in the converter layer. If the thickness of the converter layer, d, is smaller than the ion range r_i (where $i = \alpha$ or Li), $2d - d^2/r_i$ should be used instead of r_i in calculating the probability P. The factor 1/4 takes into account the result of the integration with respect to the solid angles at which one of the charged nuclei penetrates the diamond crystal.

According to calculations with SRIM software [20], the α and Li ranges in $^{10}B_2O_3$ boron oxide (density of 1.85 g/cm^3) are 5.5 and 3.0 μm, respectively. Taking into account the 90% ^{10}B enrichment, the thickness of the layer $d = 2$ μm, and the presence of the layers on the two sides, we find that the probability of detecting a thermal neutron is 3.2%. Neglecting signals with a released energy under 0.1 MeV reduces the detection probability to 2.8%. Thus, a difference between detector count rates of 0.1 ± 0.03 cps corresponds to a slow-neutron flux density at the detector of 60 ± 20 cm^{-2} s^{-1}, or 14 ± 4% of the total neutron flux. Here, we give only a statistical error, which does not take into account other factors, for example, effects on the edges of the detector crystal.

The intensity of slow neutrons from ^{252}Cf in the moderator was measured with a ^{6}LiI(Eu) scintillation detector 30 × 15 mm in dimensions. The neutron detection efficiency was 80%. The detector was placed 30 cm from the source and the spectrum was measured over a period of 10 min (Fig. 4). The beginning of the spectrum is dominated by the gamma-ray background. In the range 280–380 channels, there is a well-defined peak due to the $n + {}^6\mathrm{Li} \rightarrow \alpha + {}^3\mathrm{H} +$ 4.78 MeV reaction. The count rate in the range 280–380 channels gives the slow neutron intensity over the full 4π solid angle, which is 2.6 × 10^5 s^{-1}, or 22% of the total neutron flux.

The count rate of all other pulses gives a gamma intensity of 1.95 × 10^6 s^{-1}, which is a factor of 1.6 higher than the neutron intensity. In this case, the gamma ray detection efficiency was taken to be unity, which is responsible for the distinction from the ratio indicated above: $\gamma/n = 8.3/3.7$ for ^{252}Cf.

CONCLUSIONS

We have studied slow-neutron detectors based on single-crystal CVD diamond 0.5 mm in thickness using a ^{252}Cf neutron source placed in a polyethylene moderator. We have measured the count rates of detectors located near the moderator and after 2-cm-thick bismuth plates. Comparison of the count rates indicates that most signals of the CVD detectors correspond to gamma rays from the source. Comparison of the measured count rate with the count rate evaluated using Monte Carlo simulation leads us to conclude that 40% of the detector signals correspond to gamma-ray photon scattering in the detector crystal and that the other 60% are due to the electrons that result from the interaction of the gamma rays with the materials of the detector frame.

The slight difference in count rate between a CVD diamond detector with a $^{10}B_2O_3$ slow neutron converter and that with no converter suggests that the fraction of slow neutrons is small compared to the overall background. Measurements with a ^{6}LiI(Eu) scintillation detector have shown that only 22% of the neutrons are slowed down and leave the moderator. With allowance for the calculated probability of detecting slow neutrons, this makes it possible to account for the count rate difference within errors.

The present experimental data and calculation results confirm that the slow neutron detection efficiency of the CVD + $^{10}B_2O_3$ structure is on the order of 3%.

Our results lead us to conclude that CVD films with the minimum possible active layer thickness should preferably be used in the fabrication of slow-neutron detectors intended for operation in the presence of a strong gamma-ray background.

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