Self-powered micro-structured solid state neutron detector with very low leakage current and high efficiency

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We report on the design, fabrication, and performance of solid-state neutron detector based on three-dimensional honeycomb-like silicon micro-structures. The fabricated detectors use boron filled deep holes with aspect ratio of over 12 and showed a very low leakage current density of $\sim 7\times10^{-7}$ A/cm^2 at -1 V for device sizes varying from 2×2 to 5×5 mm^2. A thermal neutron detection efficiency of $4.5\% \pm 0.5\%$ with discrimination setting of 500 keV and gamma to neutron sensitivity of $(1.1\pm0.1)\times10^{-5}$ for single layer was measured without external bias for these devices. Monte-Carlo simulation predicts a maximum efficiency of 45% for such devices filled with 95% enriched ^{10}boron.
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The highly matured existing ^{3}He gas tube based neutron detection technology offers very high efficiency, low noise, and very low gamma sensitivity.[1] However, gas tube detectors have a number of drawbacks such as bulkiness, high bias voltage requirement (>2 keV), and high cost due to the recent high demand but limited supply of ^{3}He gas.[2] Solid state neutron detectors (SSND) can overcome many shortcomings of gas tube detectors by offering fast timing characteristics, lightweight, compactness, low (or no) bias, ability for chip-level integration with other electronic devices and high neutron detection efficiency. Therefore, the SSNDs are highly attractive for a broad range of applications, including fissile materials sensing, neutron therapy, medical imaging, the study of material sciences, probing of protein structures, and oil exploration.[3]

A widely adopted approach for obtaining a SSND is to coat a semiconductor p-n junction or metal-semiconductor junction with high thermal neutron absorption cross-section material, such as ^{10}B or LiF as a neutron to energetic charge particle conversion medium, since semiconductors (e.g., silicon or GaAs) have very low neutron absorption cross-section.[4–7] The energetic charge particles generated in the converter layer as a result of nuclear reaction reach the p-n junction and create electron-hole pairs, which are then separated by the junction built-in field, thus producing the external electrical signal. Since the resulting particle absorption takes place over a shorter distance (the range of alpha particle from ^{10}B (n α) reaction is about $\sim 5\,\mu$m in Si), the p-n junction material thickness can be kept minimum, which helps to reduce the gammas sensitivity. The neutron detection efficiency of such detectors mainly depends on two characteristics: (i) the absorption length/volume of the conversion medium to capture most of the incoming neutron flux and (ii) the range for the energetic particles produced by nuclear reaction (such as alpha particles and lithium ions) to reach the depletion region to create electron-hole pairs without losing much of their energy in the conversion medium. The converter thickness requirements for ^{10}B (density 2.35 g/cm^3) to achieve neutron interaction probability of 90% is 43 μm and the range of Li ions and alpha particles from the reaction in ^{10}B is only around 2–5 μm.[8] These two characteristics, absorption length and the range of daughter particle in converter contradicts each other and limits the efficiency to 2%-5% (Refs. 4–6) of a planer device, in which neutron capture and electrical signal generation occurs in two separate media. In order to overcome the limitation of low efficiency in planer devices, silicon micro pillar or parallel stripe p-i-n diodes were proposed, in which the gap between pillars or stripe is filled with either LiF or ^{10}B.[7,9] Boron is more preferable as a converter due to high absorption cross-section of 3840 barns than Li (940 barns) and it is compatible with silicon processing. A high aspect ratio micro pillar structure that requires for capturing most of the incoming neutron flux (few μm in diameter and about 40-50 μm high) is fragile and also the etched pillar side wall provides a leakage path. The reported leakage current density for such a device was 1.4×10^{-4} A/cm^2, which is three orders of magnitude higher than the planer device structure.[9] The main source of high leakage current in diode having micro-structured surface such as pillars fabricated by dry etching is the surface recombination in the etched surface. The high leakage current not only decreases the device sensitivity to neutron events with increasing noise level but also limits the scaling of device to large area devices necessary for many of the emerging applications.

In this letter, we report the fabrication and characterization of a self-powered solid state thermal neutron detector incorporated with hexagonal holes filled with natural boron (19.8% ^{10}B isotope) arranged in honeycomb fashion in silicon matrix. The devices have continuous p$^+$-n junction over the entire surface of the micro-structure achieved by diffusing boron as a part of boron filling process on the exposed surface of the holes. The fabricated detectors were fully depleted without external bias and showed more than two orders of magnitude improvements in the reverse bias

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leakage current compared to micro pillar silicon p-i-n diode without etched sidewall passivation, which enabled to achieve an extremely low noise level and high thermal neutron detection efficiency. These solid state detectors showed improved overall thermal neutron detection characteristics than that of previous attainments.[9]

The neutron detection efficiency of micro-structured silicon detector incorporated with boron in silicon matrix depends on geometries of the micro-structure such as silicon wall thickness, hole diameter, and depth. We have chosen a honeycomb-like structure based on its physical robustness, and higher possible efficiency compared to other structures,[10] which were simulated using GEANT4 (Ref. 11) to optimize the device geometry for maximum efficiency. The simulation included the hexagonal holes filled with ^{10}B boron and thermal neutron incident normally to the detector surface with an energy deposition discrimination of 0.2 MeV. The simulation result in Fig. 1 shows that the maximum efficiency of about 45% is possible for a Si wall thickness and hole diameter of 1 and 2.8 μm, respectively. The three dimensional cross-sectional view of fabricated device is shown in Fig. 2. The device fabrication process started with the deposition of about 50 μm thick intrinsic silicon layer with resistivity of about 45 Ω-cm on a (100) oriented n + Si substrate and then about 1 μm thick p + layer with a resistivity ∼0.001 Ω-cm. The device mesas with sizes ranging from 2×2 to 5×5 mm^2 was defined by photolithography and reactive ion etching. The mesa etch side wall was passivated by depositing silicon dioxide using plasma enhanced chemical vapor deposition (PECVD). The honeycomb hexagonal holes were patterned on the mesa area using a standard Si Bosch process—deep reactive ion etching (DRIE). The dimensions of the round holes and the thickness of Si side wall were chosen such that the Si wall could be depleted completely without external bias voltage when boron was diffused a few tens of nm (∼100 nm) to make conformal p-layer from two sides of the wall. The hole diameter and Si wall thickness were about 2.8 μm and 1 μm, respectively. The top and the cross-sectional view of the device after DRIE are shown in Figs. 3(a) and 3(b), respectively.

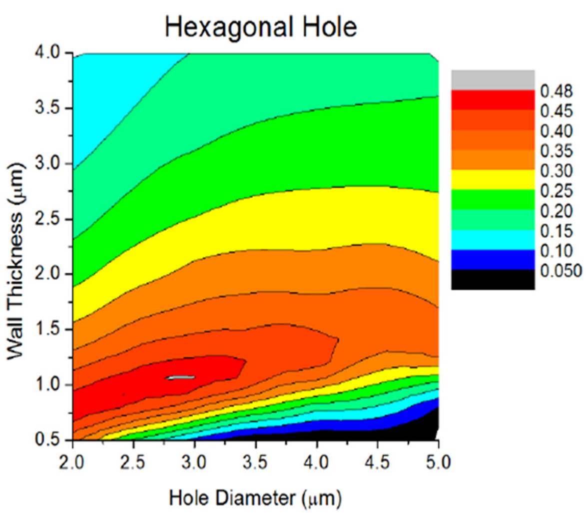

FIG. 1. Results of optimization simulations for hexagonal shaped boron holes, which are 45 μm deep. The maximum efficiency is about 45% for a hole separation of 1 μm and boron hole diameter of 2.8 μm.

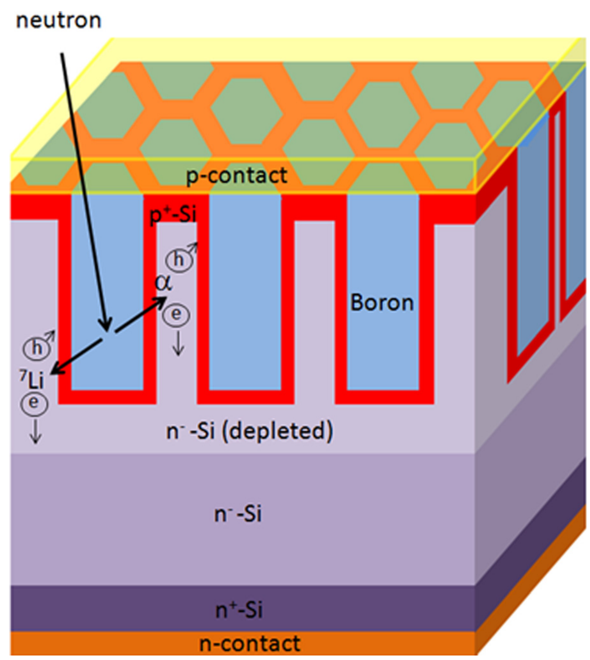

FIG. 2. Schematic diagram of the fabricated neutron detector in p$^+$-n Si substrate (3D cross-sectional view) with deep hexagonal holes filled with boron. Hexagonal holes are patterned in a honeycomb fashion using DRIE technique and the exposed surface inside the hole was boron diffused to make a continuous p$^+$-n junction as a part of boron filling process in LPCVD using diborane precursor. Incoming neutron is absorbed by ^{10}B and emits energetic ^{7}Li and α particles, which lose energy in Si generating electron-hole pairs in depleted region that are ultimately separated by the built-in field.

The holes are about 35 μm deep with aspect ratios up to 12.5. After DRIE, about 10 nm boron was deposited conformally on the Si side wall at 500 °C with natural boron (19.8% ^{10}B isotope) using diborane precursor by low pressure chemical vapor deposition (LPCVD). The wafer was heated to 950 °C for 30 min to diffuse boron in order to make continuous p-n junction on all the exposed area inside holes. The holes were then filled using the same LPCVD process at 500 °C. Complete boron filling of deep holes having a high aspect ratio using LPCVD at a reasonable growth rate is a challenging task. We have used multiple depositions and etch back process to achieve a boron fill factor as high as 90%. Figs. 3(c) and 3(d) show the cross-sectional view after boron deposition (twice) near the top surface and at the bottom, respectively. The boron on the top was then removed using a RIE plasma etching with SF_6 and O_2 as process gases. The etch-stop SiO_2 exposed on the top of p+-Si after etching boron from the top was removed using buffer oxide etchant (BOE). Finally, front and back ohmic contacts were deposited using Ti–Al metallization by sputtering. After completing the wafer fabrication processes, the device wafer was diced into individual dice having devices of different sizes. The devices were wire-bonded to external pins of 24-pin flat pack for electrical and thermal neutron response measurements. The inset in Fig. 4 shows the optical image of packaged chip. Figure 4 shows the leakage current density versus biasing voltage (J-V) characteristics of different size devices. The reverse leakage currents measured at −1 V for 2×2, 3×3, and 5×5 mm^2 devices were 17, 82, and 180 nA, respectively. The corresponding leakage current

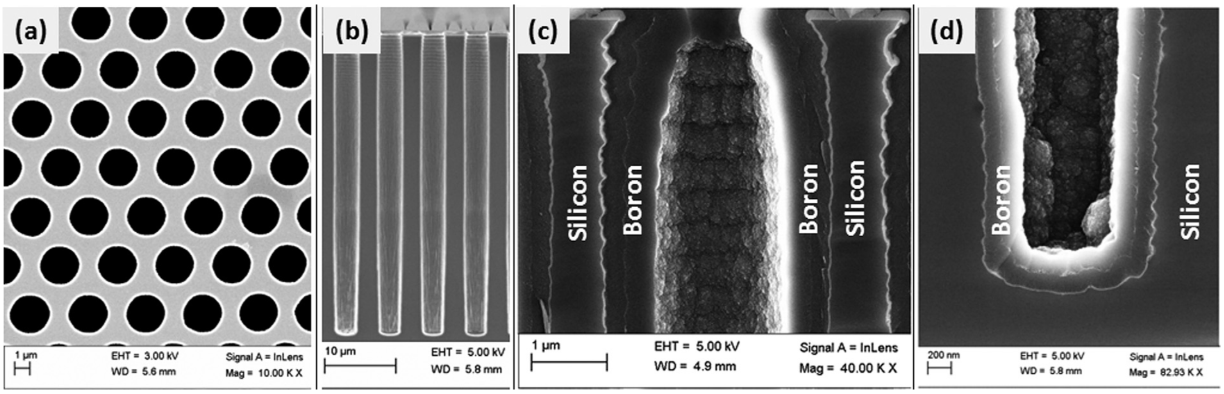

FIG. 3. Scanning electron microscope (SEM) image of (a) top view after patterning deep holes using DRIE (Bosch process). The hole diameter is about 2.8 μm and Si wall thickness is ~1 μm at the top, (b) cross-sectional view after DRIE. The holes are around 40 μm deep with good side wall profile and the aspect ratio around 13, and (c) cross-sectional view after boron deposition (twice) near the top surface (d) and at the bottom of the hole. As the boron thickness approaches around 800 nm, the hole mouth becomes narrow and the deposition rate deep inside the hole decreases significantly. ICP-RIE was used to etch back boron from the mouth before the second deposition. More than 90% boron fill factor was achieved after second deposition.

densities at -1 V for all devices were in the order of $\sim 7\times10^{-7}$ A/cm^2, which is a few orders of magnitude improvement for similar structured devices reported earlier.[9] The extremely low leakage current density in our devices is mainly due to the formation of the continuous p-n junction on the etched surfaces inside the holes, which eliminate the leakage current due to surface recombination. The devices with such a low leakage current density along with low series resistances are extremely important to make the thermal neutron detector having a very low noise level while counting neutrons events.

The intrinsic thermal neutron detection efficiency of the device was measured using a calibrated thermal neutron source consisting of ^{252}Cf source, emitting fission neutrons,

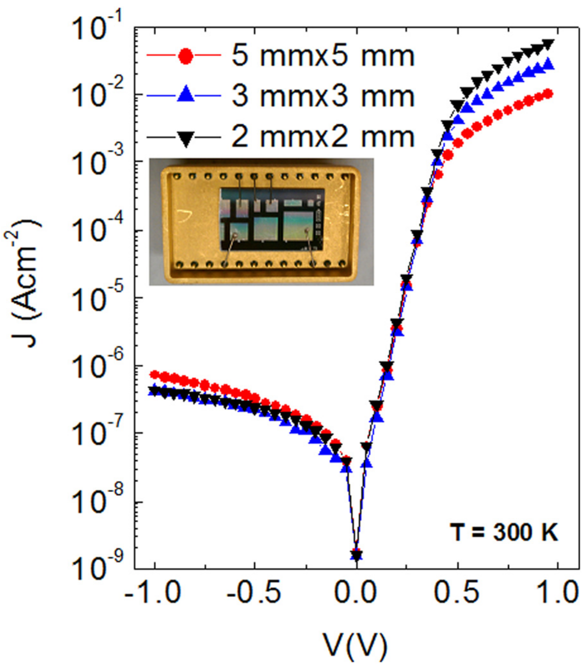

FIG. 4. The current density versus biasing voltage (J-V) characteristics of fabricated devices of different sizes. The current density at -1 V bias is about 7×10^{-7} A/cm^2 for all devices. The very low leakage current density in our devices is attributed to the reduced surface recombination due to the continuous p$^+$-n junction on the entire surface of the diode. The inset shows the optical image of a packaged chip for characterizations.

moderated by high density polyethylene block. The moderator size was $61\times61\times40$ cm^3 with the ^{252}Cf source embedded in the centre of the 61×61 cm^2 face placed 2.5 cm from the face of maximum neutron emission.[12] In order to determine the average thermal neutron flux from the moderated source accurately, the fluxes measured at different distances using three different methods: gold activation foils, a small Li-glass detector, and a BF_3 detector with and without Cd cover, were compared with MCNP (Ref. 13) calculations. At the test distance of 10 cm from the moderator face, the flux was found to be 888 n/cm^{2}s thermal neutrons. The chip with different size devices packaged in flat pack was placed in a cast aluminum box connected to a Ortec[14] preamplifier (model: 142AH) followed by an Ortec shaping amplifier (model 672). The pulse height distribution measured using an Ortec MCA (model 927) under no external bias for 2×2 mm^2 device as an example is shown in Fig. 5; the x-axis was normalized assuming spectrum endpoint is the full charge particle energy deposition (~2.5 MeV). The counts were recorded keeping the detector (i) at 10 cm away from the ^{252}Cf moderator face, red curve-1, (ii) with 2 mm Cd sheet covering the moderator face, black curve-2, (iii) with ^{60}Co gamma source which produces 10 mR/h dose on detector surface, the standard set by Pacific Northwest National Laboratory[15] for gamma insensitivity criteria for neutron detector, blue curve-3, and (iv) removing ^{252}Cf away from the moderator housing, green curve-4. It is clearly seen that the noise level for the device is below 200 keV. The extremely low noise in our devices is the result of the extremely low leakage current achieved due to the continuous p$^+$-n junction on the entire surface of the device. Although the device noise level is below 200 keV, the device also responds to gamma rays which are registered below 500 keV. In order to avoid any contribution from gamma to the intrinsic efficiency, the lower level of detection (LLD) was set at 500 keV. The counts recorded above 500 keV with Cd shield were subtracted from counts recorded without Cd shield and ^{252}Cf in the moderator housing because the Cd shutter blocks neutrons with energy smaller than 0.5 eV but allow gamma ray and high energy neutrons. The net count due to thermal neutron in 13 000 s was 21 489 and the incoming flux was 888 n/cm^{2}s. The estimated intrinsic thermal neutron

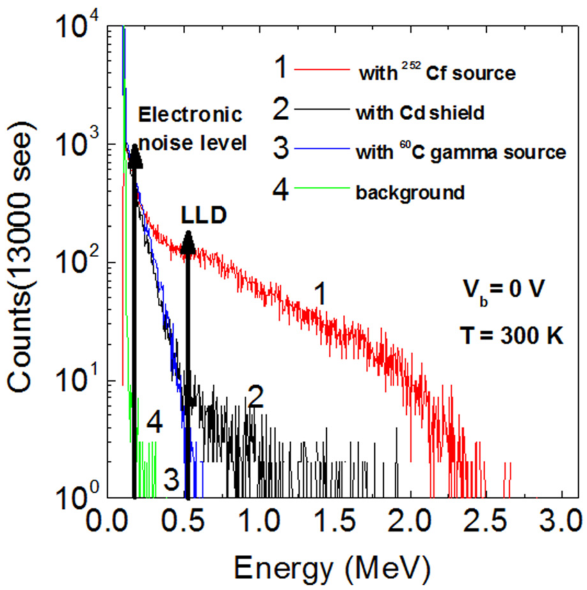

FIG. 5. The measured pulse height distribution of the micro-structure silicon diode with hexagonal holes filled with natural boron as the converter material. The device electronic noise level is below 200 keV (green curve-4), but the device also responds to gamma rays below 500 keV (blue curve-3), so a LLD was set at 500 keV.

detection efficiency and gamma to neutron sensitivity were 4.5% ± 0.5% (and $1.1 \pm 0.1) \times 10^{-5}$ respectively. The efficiency could reach to 6.9% if the gamma sensitivity in the range of 200 to 500 keV could be eliminated with further device fabrication process optimizations, which was not done for the devices. Monte Carlo simulations of the device under test with incomplete boron filling, the moderated ^{252}Cf source and assuming above 200 keV charge particle energy deposition gives an efficiency of 9.9% which is higher than the experimentally measured value of 6.9% for LLD setting of 200 keV, the discrepancy could be related to the variation in Si wall thickness, hole diameter, and boron thickness. However, the measured efficiency was found to be almost the same for all device sizes up to 5×5 mm^2. Further the efficiency also remains unchanged when measured under reverse bias voltages of 0 to 5 V, ensuring that the active area of our devices is completely depleted under zero bias. The efficiency increased to 8% when two layers of devices were stacked facing each other and measured in parallel configuration keeping the LLD at 500 keV. The efficiency could be enhanced by 3 fold when the natural boron is replaced by 95% enriched ^{10}B isotope. The measured efficiency of about 15% for a single layer of device when scaled to 95% ^{10}B enriched boron is about one third of the predicted value of 45% from simulation for the optimum geometry; however, the device having optimum geometry and filled with 95% ^{10}B enriched boron has not been fabricated yet. The difference in observed and simulated results is attributed to: (i) the boron fill factor is only 90%, (ii) the hexagonal holes are only 34 μm deep (vs 45 μm assumed in simulation) and were tapered and the hole diameter decreases from 2.8 μm at the top to 1.7 μm at the bottom, which reduces the volume fraction of converter boron by about 40% than that of assumed in simulation. The efficiency could be further enhanced by optimizing the DRIE process to achieve deep holes with vertical side wall and optimizing LPCVD process to achieve 100% boron fill factor.

In summary, we have designed fabricated and characterized micro-structured silicon diodes with deep hexagonal holes filled with natural boron as a conversion material for thermal neutron detection applications. The fabricated devices of different sizes showed a leakage current density of $\sim 7 \times 10^{-7}$ A/cm^2 at -1 V reverse bias. The extremely low leakage current density observed in our device is attributed to the novel concept of continuous p$^+$-n junction on the entire surface of the microstructure. The thermal neutron detection efficiency measurement showed low electronic noise level below 200 keV, intrinsic thermal neutron detection efficiency of 4.5% ± 0.5%, and gamma to neutron sensitivity of $(1.1 \pm 0.1) \times 10^{-5}$ for single layer device measured without external bias voltage. The efficiency increased to 8% when two layers of devices were stacked together. The outstanding characteristics such as very low leakage current, low noise floor, and the zero bias operation of our devices fabricated by standard Si processing is a step closer to the realization of highly efficient, large area solid state neutron detector at low cost for future applications.

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