High efficiency hexagonal boron nitride neutron detectors with 1 cm$^2$ detection areas

Cite as: Appl. Phys. Lett. 116, 142102 (2020); doi: 10.1063/1.5143808
Submitted: 6 January 2020 · Accepted: 18 March 2020 · Published Online: 6 April 2020

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ABSTRACT

We report the realization of 1 cm$^2$ hexagonal boron nitride (h-BN) thermal neutron detectors with an unprecedented detection efficiency of 59%. This was achieved through improvements in material quality, as reflected in a sixfold enhancement in the electron mobility and lifetime product and a threefold reduction in the surface recombination field, which resulted in a higher detection efficiency at a lower applied electric field over that of a previous state-of-the-art lateral detector with a detection area of 30 mm$^2$. The attainment of 1 cm$^2$ h-BN neutron detectors capable of retaining a high detection efficiency represents a significant milestone toward the practical applications of h-BN detectors.

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Due to the large thermal neutron absorption cross section of the $^{10}$B isotope ($\sigma \sim 3840$ barns = $3.84 \times 10^{-21}$ cm$^2$) and the high atomic density of $^{10}$B ($\sim 5.5 \times 10^{23}$/cm$^3$) in 100% $^{10}$B-enriched h-BN (h-10BN), the thermal neutron absorption length ($\lambda$) in h-10BN is as short as $\sim 47.3$ μm [\(\lambda = \frac{\sigma}{\rho} = \frac{(\text{N}_0\alpha)}{M} = (5.5 \times 10^{23} \times 3.84 \times 10^{-21})^{-1} \text{cm}

Therefore, h-10BN neutron detectors are inherently lightweight and compact with excellent form factors. The high electrical resistivity (>10$^{15}$ Ω-cm) and high temperature stability of h-BN make these detectors suitable for high temperature operation. Furthermore, h-10BN neutron detectors require a lower operating voltage than 3He gas detectors because h-10BN is a semiconductor and have a demonstrated low sensitivity to gamma radiation due to the low atomic numbers of constituent B and N atoms.

Due to the nature of low neutron flux from a primitive nuclear device ($\sim 3 \times 10^7$ neutrons/s) and low reflectance of neutrons in porosity measurements, large size detectors capable of providing high sensitivities are desirable for practical uses. The sensitivity of a detector or the count rate ($C_D$) detected by a detector is proportional to its detection efficiency ($\eta$) and detection area ($A_D$), i.e., $C_D = \varphi \eta A_D$, where $\varphi$ is the neutron flux. However, there are technical challenges to scale up the detector size, which include increased dark current, capacitance, and surface recombination field ($E_s$), all of which tend to decrease the detection efficiency. It was shown recently that compared...
to vertical detectors consisting of planar contacts on top and bottom, the adoption of a lateral detector configuration can alleviate these issues to some extent, which led to the demonstration of h\textsuperscript{10}BN detectors with a detection area of 0.3 cm\textsuperscript{2} and a detection efficiency of \(~50\%\).\textsuperscript{12} In this work, we report the realization of 1 cm\textsuperscript{2} h-BN thermal neutron detectors with an unprecedented detection efficiency of 59\%. Improved material quality plays a key role in enabling the scale up to this detection area, as reflected in the increased charge carrier mobility-lifetime (\(\mu t\)) product and reduced surface recombination field (\(E_s\)), which are two important parameters greatly influencing the charge collection efficiency and hence the overall detection efficiency of h\textsuperscript{10}BN neutron detectors.\textsuperscript{12}

Epilayers of h\textsuperscript{10}BN of \(~100 \mu m\) in thickness were deposited on c-plane sapphire substrates of 4-in. in diameter, using metal-organic chemical vapor deposition (MOCVD). Based on the insights of previous studies,\textsuperscript{9–13} a low temperature BN buffer layer of about 20 nm in thickness was deposited on the c-plane sapphire substrate at about 800 °C prior to the growth of the h\textsuperscript{10}BN epilayer, whereas the growth temperature of the subsequent thick h\textsuperscript{10}BN epilayer was \(\sim 1400 \pm 40 \pm C\). The primary purpose of using a reduced growth temperature of 1400 °C in the present work as compared to that of \(> 1400 \pm C\) in previous studies\textsuperscript{10,12} was to reduce the oxygen diffusion from the sapphire substrate,\textsuperscript{13} whereas oxygen impurities in h-BN are known to occupy nitrogen sites (ON), acting as donors.\textsuperscript{14} Moreover, the presence of ON donor impurities tends to decrease the formation energies of boron-related deep level defects in h-BN,\textsuperscript{14} which are expected to be detrimental to the charge collection efficiency of h-BN detectors. Layer structured h-BN has a different expansion coefficient than the sapphire substrate, enabling natural separation of thick h\textsuperscript{10}BN layers from sapphire substrates during cooling down after epi-growth. An optical image of a portion of the diced h\textsuperscript{10}BN wafer used in this study is shown in Fig. 1(a). This freestanding h\textsuperscript{10}BN wafer was diced into strips of \(~1.3 \pm mm\) in width. Adopting the same and proven lateral device architecture used in the 0.3 cm\textsuperscript{2} detector,\textsuperscript{11} we combined 5 strips with lengths varying from 13.5 to 17 mm to form a detector with a total detection area of 1 cm\textsuperscript{2}. A highly resistive adhesive material was used to mount these strips on a sub-mount (sapphire). E-beam evaporation was used to deposit a metal bi-layer of Ni (100 nm) and Au (40 nm) on the clipped edges of the h\textsuperscript{10}BN strips using a shadow mask of 1.1 mm width leaving around \(~100 \mu m\) metal covering on the edges, as schematically depicted in Fig. 1(b). The finished 1 cm\textsuperscript{2} h\textsuperscript{10}BN detector is shown in Fig. 1(c).

The dark current–voltage (I–V) measurements were performed for the 1 cm\textsuperscript{2} detector from 0 to 500 V and the result is shown in Fig. 2(a). A linear fit of the I–V characteristic provides values of resistance, \(R = 1.77 \times 10^{12} \Omega\) and resistivity, \(\rho = 1.05 \times 10^{12} \Omega \cdot cm\). A deuterium UV lamp (DS421, Acton Research Corporation, spectral range of 190–350 nm) was used as an excitation light source for characterizing the I–V characteristics under illumination. A metal mask (hole diameter of 190–350 nm) was used as an excitation light source for characterizing the I–V characteristics under illumination. An optical image of a portion of the diced h\textsuperscript{10}BN sample used in this work is shown in Fig. 1(a). This freestanding h\textsuperscript{10}BN wafer was diced into strips of \(~1.3 \pm mm\) in width. Adopting the same and proven lateral device architecture used in the 0.3 cm\textsuperscript{2} detector,\textsuperscript{11} we combined 5 strips with lengths varying from 13.5 to 17 mm to form a detector with a total detection area of 1 cm\textsuperscript{2}. A highly resistive adhesive material was used to mount these strips on a sub-mount (sapphire). E-beam evaporation was used to deposit a metal bi-layer of Ni (100 nm) and Au (40 nm) on the clipped edges of the h\textsuperscript{10}BN strips using a shadow mask of 1.1 mm width leaving around \(~100 \mu m\) metal covering on the edges, as schematically depicted in Fig. 1(b). The finished 1 cm\textsuperscript{2} h\textsuperscript{10}BN detector is shown in Fig. 1(c).

FIG. 1. (a) Optical image of a portion of the diced h\textsuperscript{10}BN sample used in this work. (b) Schematic diagram of detector strips mounted on a sapphire sub-mount with UV light illuminated through a metal mask, allowing the measurement of electron and hole transport separately. (c) Optical image of a 1 cm\textsuperscript{2} neutron detector fabricated from a 100 \mu m thick h\textsuperscript{10}BN freestanding wafer by combining five detector strips.

The measured I–V characteristics of electron transport under UV illumination are shown in Fig. 2(b). To determine the charge carrier \(\mu t\) product and surface recombination field \(E_s = \frac{1}{\mu t}\), the measured photocurrent in Fig. 2(b) was fitted by Many’s equation,\textsuperscript{15}

\[
I_i(V) = I_0 \left[ \frac{1 - e^{-\frac{u_i}{\mu t V_b}}}{V_i W} \right] \left[ 1 + \frac{s_i W}{\mu t V_b} \right]
\]

(2)

Here, the saturation current is denoted as \(I_0\), and \(\mu t V_b\) (\(\mu t\)) and \(s_i\) (\(s_i\)) are the mobility-lifetime product and surface recombination velocity for electrons (holes), respectively. \(V_b\) is the applied voltage between two electrodes of distance \(W\), providing an applied electric field of \(E_b = \frac{V_b}{W}\). The numerator and the denominator term of Eq. (2) describe the charge collection efficiency limited by the bulk trapping effect (or...
μτ product) and “surface recombination field” or the ratio of surface recombination velocity to mobility (Ea = s/μ), respectively,9–11 whereas both μτ and s/μ are directly correlated with the overall material quality. From Eq. (2), conditions for a high charge collection efficiency are (a) \( \frac{W_{i}}{s/\mu} \ll 1 \), which means that the charge carrier drift length (\( l_{e} = \mu V/\tau_{e} \)) must be greater than the transit length (spacing between the electrodes, W) and (b) \( \frac{W_{l}}{s/\mu} = \frac{W}{l_{e}} \ll 1 \) (where i = e, h), which means that the applied electric field must be greater than the surface recombination field.9–11 The measured values of \( \mu_{e} \tau_{e} \) and \( s/\mu_{e} \) for this detector were 5.0 \( \times 10^{-3} \) cm\(^2\)/V and 5.5 \( \times 10^{2} \) V/cm, respectively, which provide values of \( \frac{W_{i}}{s/\mu} \approx 0.007 \) and \( \frac{W_{l}}{s/\mu} \approx 0.14 \) at a bias voltage of 500 V, and a theoretical charge collection efficiency of 87.4% under UV irradiation. It is important to recognize that the measured \( \mu_{e} \tau_{e} \) product of this detector has been enhanced by a factor of 6, while \( E_{e} \) has been reduced by a factor of 3 in comparison to those of a previous reported state-of-the-art 0.3 cm\(^2\) h-10BN lateral detector.12 These improvements were achieved primarily through MOCVD epitaxial growth optimization discussed above.

Figure 3 shows the pulse height spectra of this 1 cm\(^2\) h-10BN detector. The detector was placed 1 m away from the \(^{252}\)Cf source moderated by a HDPE cube. The blue curve is the response to thermal neutrons which was recorded by placing the detector 1 m away from the \(^{252}\)Cf source moderated by a HDPE cube. The green curve is the response to gamma photons emitted from a \(^{137}\)Cs source measured at the same bias voltage. The green curve is the response to gamma photons emitted from a \(^{137}\)Cs source measured at the same bias voltage.

FIG. 2. Current–voltage characteristics measured for the device configuration shown in Fig. 1(b): (a) in the dark and (b) under UV radiation for electron transport.
velocities reported from other III–V semiconductors suggested that the surface recombination velocity of h-BN can be as low as 10^2 cm/s with further improvements in material quality and device processing. The realization of 1 cm^2 h-BN neutron detectors capable of retaining a high detection efficiency represents a critical step toward the commercial adoption of high sensitivity h-BN neutron detectors for practical applications.

This research was supported by DOE ARPA-E (No. DE-AR0000964) and monitored by Dr. Isik Kizilyalli. Jiang and Lin are grateful to the AT&T Foundation for the support of Ed Whitacre and Linda Whitacre endowed chairs.

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FIG. 4. Surface recombination velocities of representative semiconductors vs the energy bandgap $E_g$, with published data from Ref. 23 for GaAs, Ref. 18 for InP, Ref. 24 for Al$_{x_1}$In$_{x_0}$As, Ref. 20 for InGaAs, Ref. 19 for InGap, Refs. 21 and 22 for GaN, Ref. 25 for Si, Ref. 26 for 4H-SiC, and Ref. 16 for h-BN. The black lines serve as a guide to the eye revealing the trend of decreasing surface velocity with an increase in the energy bandgap of semiconductors.