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Fabrication and characterization of solid-state thermal neutron detectors based on hexagonal boron nitride epilayers



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ABSTRACT

Solid-state thermal neutron detectors with improved detection efficiencies are highly sought after for many applications. Hexagonal boron nitride (hBN) epilayers have been synthesized by metal organic chemical vapor deposition on sapphire substrates. Important material parameters including the mobility-lifetime ($\mu\tau$) product and the thermal neutron absorption length (λ) have been measured. For hBN epilayers with a room temperature resistivity of 5.3 × 10¹⁰ Ω cm, the measured $\mu\tau$ product of electrons is 4.46 × 10⁻⁸ cm²/V and of holes is 7.07 × 10⁻⁹ cm²/V. The measured λ values are 277 μ m and 77 μ m for natural and ¹⁰B enriched hBN epilayers, respectively. Metal–semiconductor–metal detectors incorporating 0.3 μ m thick hBN epilayers were fabricated. The reaction product pulse-height spectra were measured under thermal neutron irradiation produced by a ²⁵²Cf source moderated by high density polyethylene block. The measured pulse-height spectra revealed distinguishable peaks corresponding to the product energies of ¹⁰B and neutron reaction with the 0.84 MeV ⁷Li peak being the most prominent. The detectors exhibited negligible responses to gamma rays produced by ¹³⁷Cs decay. Our results indicate that hBN epilayers are highly promising for realizing highly sensitive solid-state thermal neutron detectors with expected advantages resulting from semiconductor technologies, including compact size, light weight, ability to integrate with other functional devices, and low cost.

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1. Introduction

Hexagonal boron nitride (hBN) is a wide bandgap semiconductor with an energy band gap around 6 eV. It has emerged as an important material for deep ultraviolet photonic device applications [1–5] and for the exploration of new physical properties in two dimensional systems that are complementary to graphene [6–8]. Due to the fact that the boron-10 (10 B) isotope has a capture cross-section of about 3840 barns for thermal neutrons (with 0.025 eV energy) which is orders of magnitude larger than those of most other isotopes [9,10], hBN is also highly promising for the fabrication of solid-state neutron detectors [11,12]. When a 10 B atom captures a neutron, it undergoes the following nuclear reaction:

$$^{10}B + ^{1}_{0}n \rightarrow ^{7}Li(1.015MeV) + \alpha(1.777MeV) 6\%$$
 (1a)

$$^{10}B + ^{1}_{0}n \rightarrow ^{7}Li^{*}(0.840MeV) + \alpha(1.470MeV). 94\%$$
 (1b)

The daughter particles (α particles and 7Li ions) produced by the nuclear reaction have a mean free path (or a range) of $\sim\!5~\mu m$ for

 α particles and $\sim 2 \,\mu m$ for $^7 \text{Li}$ ions [9] and lose their energies by producing a cloud of electron-hole pairs in hBN semiconductor, which serve as the detection signal for thermal neutrons. In hBN neutron detectors, the neutron capture, charge collection, and electrical signal generation occur in the same hBN layer. This is in contrast to boron coated conversion devices, in which the thermal neutron absorption takes place in the boron conversion layer and the generation of electrons and holes occurs in the semiconductor layer [13-15]. Thus, hBN detectors are potentially capable of providing higher detection efficiencies for thermal neutrons than those of the boron coated semiconductor conversion devices. Furthermore, its large energy band gap inherently renders detectors with very low leakage currents [2,11,12,16]. Compared to the commercialized ³He gas filled neutron detectors which have a thermal neutron cross section \sim 5330 barns [9], the density of 3 He gas is significantly lower than the density of ¹⁰B in solid state materials such as in hBN. Thus, the thermal neutron absorption length of ³He is much larger than that of hBN. Therefore, ³He gas detectors are generally large and bulky. Most importantly, there is an urgent issue of ³He gas shortage, which limits its future applications.

Our group is currently exploring wafer scale epitaxial growth of hBN semiconductors by metal organic chemical vapor deposition (MOCVD) technique [2–4]. A continuous irradiation with a thermal

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neutron beam generated an appreciable steady current response in hBN detectors, corresponding to an effective conversion efficiency approaching $\sim\!80\%$ for the absorbed thermal neutrons [11,12]. However, more research is needed to further improve the material quality, device design, and the detection efficiency. In terms of charge carrier collection, neutron detectors operate on the same basic principle as photodetectors. However, the absorption length of thermal neutrons is much larger than the band edge photons in hBN. Therefore, thermal neutron detectors and photodetectors require different design considerations. The typical absorption length of the band edge photons in semiconductors is in the order of 0.1 um and is only around 15 nm in hBN [2]: while the thermal neutron absorption length (λ) in hBN synthesized using natural boron sources is around 230 µm [11,17]. This large absorption length requires the device design considerations to favor the following: (1) the thickness of hBN layer to be large in order to capture a sufficient number of the incoming neutrons; and (2) the carrier mobility-lifetime product ($\mu\tau$) to be large to allow the free electrons and holes to sweeping out and to be collected as a signal in a timely manner. The $\mu\tau$ product is thus one of the most important parameters that characterize the electronic quality of a semiconductor detector material. This parameter determines the charge collection efficiency and hence the suitability of a material for detector applications. This parameter is significantly influenced by crystalline quality and the density of impurities/defects introduced during the crystal growth and device fabrication processes. Since the hBN epilayer and device technologies are just in the development stage, it is crucial to characterize and optimize the $\mu\tau$ product in order to improve the material quality and device performance. To our knowledge, this parameter has not been characterized for hBN in either bulk or thin film form.

We report here on the growth and characterization of the basic material properties of hBN epilayers that are important for the design of neutron detectors, including the $\mu\tau$ product and thermal neutron absorption coefficient and absorption length of hBN epilayers. Thermal neutron detectors based on a metal–semiconductor–metal (MSM) device architecture have been fabricated. The reaction product pulse height spectra of hBN MSM detectors were tested under thermal neutron irradiation produced by a ^{252}Cf source moderated by high density polyethylene block. The results indicated that neutron detectors fabricated from hBN epilayers are capable of resolving the prominent energy peaks as expected from the reaction of Eq. (1) with high spectral resolution.

2. Epilayer growth and basic structural properties

The hBN epilayers employed in this study were grown by MOCVD on c-plane electrically insulating crystalline sapphire substrates. Since hBN materials and devices are in the development stage, the use of the insulating sapphire substrates is more suitable for the thorough characterization of the material properties of hBN for neutron detector applications. Triethylboron (TEB) and ammonia (NH₃) were used as precursors for boron and nitrogen, respectively. Due to the lattice mismatch between hBN and Al₂O₃, a low temperature BN buffer layer of about 10 nm in thickness was deposited on sapphire substrate at 600 °C prior to the growth of hBN epilayer. The hBN epilayers were grown at 1300 °C using hydrogen as a carrier gas. X-ray diffraction (XRD) θ -2 θ scan of the grown hBN [4] revealed a c-lattice constant \sim 6.67 Å, which closely matches the bulk *c*-lattice constant of hBN (c=6.66 Å) [18–20], affirming that our MOCVD grown BN films are of single hexagonal phase. The XRD rocking curve (ω-scans) of the (0002) diffraction peak of hBN exhibited a full width at half maximum (FWHM) of 380 arcsec [4], which is comparable to those of typical GaN epilayers grown on sapphire [21], but is much broader than the typical FWHM (\sim 63 arcsec) of high quality AlN grown on sapphire [22]. The results are indicative of the fact that the development of epitaxial layers of hBN is in its early stage. Secondary ion mass spectrometry (SIMS) results revealed that hBN epilayers have excellent stoichiometry [4]. The grown epilayers also have a good surface morphology as indicated by the scanning electron microscopy (SEM) image of an hBN epilayer (etched into stripes) shown in Fig. 1(a).

3. Device fabrication and mobility and lifetime product characterization

Metal–semiconductor–metal (MSM) detectors with interdigital fingers were fabricated from these hBN epilayers with etched stripes (as illustrated in Fig. 1(a)) based on the processes outlined in Ref. [11,16] to take the advantage of the excellent lateral transport properties of hBN. Fig. 1(b) shows the schematics of the hBN layer structure employed for MSM detector fabrication and the setup for the I–V characteristics measurements. The devices incorporate hBN epilayers with 0.3 μ m in thickness. Since the development of hBN epilayers is in its early stage, the surface morphology of hBN epilayers becomes poorer with an increase in

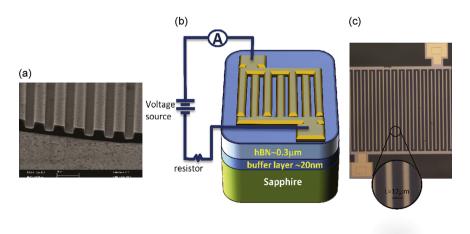


Fig. 1. (a) SEM image of an hBN epilayer etched into stripes. (b) Schematic diagram of the layer structure of hBN epilayer employed for MSM detector fabrication and the setup for the I-V characteristics measurements. (c) Optical microscopy image of a fabricated MSM detector with a device size of $500 \times 500 \, \mu m^2$ and $6 \, \mu m/12 \, \mu m$ finger width/spacing.

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