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Fabrication and testing of antimony doped bismuth tri-iodide semiconductor gamma-ray detectors

Sasmit S. Gokhale [a, *], HyukSu Han [b], Oswaldo Pelaez [a], James E. Baciak [a], Juan C. Nino [b], Kelly A. Jordan [a]

[a] *Nuclear Engineering Program at the Department of Materials Science and Engineering, 100 Rhines Hall, University of Florida, Gainesville, FL 32611, USA*
[b] *Department of Materials Science and Engineering, 100 Rhines Hall, University of Florida, Gainesville, FL 32611, USA*

HIGHLIGHTS

- Antimony (Sb) doped BiI_3 crystals were grown by the modified vertical Bridgman technique.
- Effect of surface treatment on BiI_3 radiation detectors was investigated.
- Radiation response was measured by recording gamma-ray spectra at room temperature.
- Maiden response of BiI_3 to gamma-rays was reported.
- Electron mobility-lifetime product was estimated.

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ABSTRACT

Antimony (Sb) doped bismuth tri-iodide (BiI_3) radiation detectors were fabricated from large single crystals that were grown using the modified vertical Bridgman technique. Detectors were prepared by subjecting the crystal surfaces to different mechanical and chemical treatments. Surface quality of the detectors was evaluated using optical microscopy. The influence of surface quality on detector performance was analyzed by measuring the leakage current for each of the detectors. The radiation response of the detectors was measured using an Americium (^{241}Am) gamma-ray source at room temperature. The first successful use of BiI_3 detectors for gamma-ray spectroscopy is reported here with energy resolution of 7.5% at 59.5 keV. The mobility-lifetime product for electrons was also estimated to be about 5.2×10^{-4} cm^2/V.

1. Introduction

Bismuth tri-iodide demonstrates a number of properties that are apt for room temperature semiconductor radiation detection, and especially gamma ray spectroscopy. The high atomic number ($Z_{Bi} = 83$ and $Z_I = 53$) and the relatively high density (5.78 g/cm^3) causes the material to have good photon stopping power (Matsumoto et al., 2002), while the large band-gap (1.67 eV (Qiu et al., 2013)) allows it to function as a room temperature radiation detector without cooling.

There are several factors that affect the detector response to radiation. Some of these factors such as the material atomic number and charge carrier mobility and lifetime are inherent to the detector crystal, while extrinsic factors such as impurities and detector surface quality also affect the performance of radiation detectors (Goorsky et al., 1996). Generally detector performance can be improved by controlling these factors. BiI_3 has a layered crystal structure with weak van der Waals bonding between the layers, and therefore the crystals are mechanically soft (Fornaro et al., 2006) and precautions have to be taken to prevent damage during detector fabrication.

Detector fabrication is carried out in a number of steps, which involves cutting the crystal boule, polishing the surfaces, and subsequently etching the polished surfaces. Each of these steps is aimed at eliminating the surface damage resulting from the previous steps. The surface damage that results from cutting the detector crystal may enhance the surface leakage current by providing

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* Corresponding author.
E-mail address: sasmitg@ufl.edu (S.S. Gokhale).

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more conductive paths and altered electric field distribution, which results in an increase in noise. It can also enhance carrier recombination by providing more carrier trapping sites, which reduces the charge collected at the electrodes (Cui et al., 2004).

Mechanical polishing of the surfaces followed by chemical etching are two important steps in the fabrication of detectors. Mechanical polishing removes the large surface distortions arising during cutting; however it does not leave the surface defect free. Chemical etching is carried out to remove the residual damage after mechanical polishing. Other investigations have reported the effect of surface treatment on the surface quality of semiconductor detectors (Cui et al., 2004), (Fornaro et al., 2010), (Oliveira et al., 2005), and (Zheng et al., 2011). These studies have shown that the leakage current is correlated to the surface morphology. Since leakage current is a major component of detector noise, it is important to optimize the detector fabrication process to produce detectors with good response.

Recently, Han et al. have demonstrated that Sb doping can effectively improve the electrical characteristics of BiI_3 single crystals (Han et al., 2014). It was observed that the resistivity of the doped BiI_3 crystals was on the order of 10^{10} Ω-cm. This value of resistivity is comparable to the resistivity of the BiI_3 platelets grown by physical vapor deposition (PVD) (Fornaro et al., 2004), (Cuna et al., 2004), and (Aguiar et al., 2009) and is orders of magnitude higher than the value of resistivity of undoped BiI_3 detectors reported in our previous work (Lintereur et al., 2011). Resistivity values in the range of 10^{10} Ω-cm were also reported for BiI_3 crystals grown by the Bridgman method, as well as for crystals grown by vapor transport (Dmitriev et al., 1999), (Nason and Keller, 1995), and (Gokhale et al., 2015). Additionally it is theorized that the Sb doping may delay the onset of detector polarization and as a result the detectors could be operated for longer duration.

In this work Sb doped BiI_3 detectors were subjected to the various surface treatment steps and their surface quality was evaluated. The electrical characteristics and the radiation response of the treated detectors were measured. A number of the detectors showed response to gamma-ray irradiation. This was the first instance where BiI_3 detectors were used to record gamma-ray spectra which could be spectrally resolved. The radiation response measurement was performed at room temperature using a^{241}Am sealed gamma-ray source.

2. Experimental techniques

2.1. Crystal growth

Sb doped BiI_3 single crystals were grown by the modified vertical Bridgman method. Pyrex glass ampoule was selected as the growth chamber due to relatively low annealing (=560 °C) and softening point (=815 °C), however those temperatures are still higher than the maximum temperature (=440 °C) for the BiI_3 single crystal growth process. Customized Pyrex glass ampoules (inner diameter = 1.905 cm, tip length ≅5 cm, and tip angle ≅70°) were purchased from Southern Scientific, Inc. The Pyrex ampoule was cleaned with glassware cleaning solution (Decon Contrad® 70) to remove residual organics. The ampoule was soaked in the solution overnight under a fume food, and it was then rinsed with DI water (ρ ≅ 17.0 MΩ-cm) followed by baking in a drying oven (120 °C) overnight.

Typical loads of 20 g of BiI_3 powder (99.999%, MV laboratory Inc.) with appropriate amount of SbI_3 (99.9%, Strem Chemicals Inc.) based on the doping concentration were weighed and loaded in the ampoule. SbI_3 was added to the ampoule first followed by the BiI_3 powder. The ampoule was then evacuated to a pressure of 0.03 torr by using mechanical roughing pump, and then molecular turbo pump to finally evacuate the ampoule to 7.5×10^{-5} torr. The vacuumed ampoule was then sealed by using a propane hand torch. The sealed ampoule was vertically mounted on a standing steel frame using galvanized steel wires, and placed in programmable 24 multi heating zone furnaces (EDG-13, Mellen Company). The previously optimized growth condition, such as temperature gradient of 10 °C/cm and growth rate of 0.5 mm/h, were utilized for the single crystal growth process (Lintereur et al., 2011).

In order to determine optimal doping concentration, a series of BiI_3 single crystals with 0.5 at%, 1.0 at%, and 5.0 at% Sb were grown. Detectors were then prepared from each crystal and electrical properties, such as bulk resistivity and leakage current were measured and compared. The gold electrodes sputtered onto the detectors exhibited ohmic behavior. As can be seen from Fig. 1(a), the resistivity of the detectors doped with 0.5 at% and 1.0 at% Sb was of the order of 10^{10} Ω-cm (3.48×10^{10} Ω-cm for 0.5 at% and 1.7×10^{10} Ω-cm for 1 at%), while the 5 at% doped material had lower resistivity at around 3.75×10^{9} Ω-cm.

In addition, Fig. 1(b) shows the comparison of the leakage current density in the various Sb doped BiI_3 single crystal detectors. The detectors were biased such that the electric field was held constant at 100 V/cm and the leakage current was measured over a period of time. It can be seen that the leakage current density appears to change over time. This change may be attributed to polarization effects. Polarization may be caused due to a change in the internal electric field over time due to defects, traps, or impurities. Similar polarization phenomenon has been observed in other detector materials such as HgI_2 and TlBr (Ponpon et al., 2000) – (Hitomi et al., 2009). The effect of polarization on the radiation response of the detectors is discussed later on in the manuscript. The lowest leakage current was observed in the 0.5 at% doped BiI_3 detector. A detector with the higher resistivity and low leakage current should have a better response to radiation since a lower dark current and a higher signal to noise ratio can be achieved.

The measurements were repeated on more than eight detectors harvested from the different crystal boules to confirm the reproducibility of the results. The trends seen in Fig. 1a and b were consistently observed in all the detectors fabricated from the different doped crystals i.e. the 0.5 at% Sb doped detectors had higher resistivity and lower leakage current that the detectors with other doping concentrations. Therefore, Sb (0.5 at%) doped BiI_3 was selected as it was the most promising compound for growing single crystals and fabricating radiation detectors, and the following results and discussion are only focused on this compound.

2.2. Detector fabrication

Detectors were fabricated by cutting the as grown crystal into parallelepipeds using a diamond wire saw. Any mechanical stress caused due to cutting and handling the crystals can increase the concentration of intrinsic structural defects by local deformation. This limits the performance of the detectors (Owens et al., 2003). Therefore the crystals were cut slowly, using mineral oil for lubrication to minimize damage. The wire saw cuts were made in a direction perpendicular to the [001] plane. The [001] plane is the cleavage plane, and since BiI_3 has a layered structure with weak van der Waals bonding between the layers, the crystals cleave easily along the cleavage plane.

After cutting, the crystals were then polished with abrasive slurries (Fig. 2). Polishing was carried out in a number of steps with slurries of progressively smaller grain size from 3 to 0.05 μm. The slurries were made by suspending alumina powders in mineral oil. Each mechanical polishing step causes some surface damage as well, which extends into the crystal bulk to a depth of approximately three times the size of the powder being used (Kargar et al.,