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Characterization of bismuth tri-iodide single crystals for wide band-gap semiconductor radiation detectors

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ABSTRACT

Bismuth tri-iodide is a wide band-gap semiconductor material that may be able to operate as a radiation detector without any cooling mechanism. This material has a higher effective atomic number than germanium and CdZnTe, and thus should have a higher gamma-ray detection efficiency, particularly for moderate and high energy gamma-rays. Unfortunately, not much is known about bismuth tri-iodide, and the general properties of the material need to be investigated. Bismuth tri-iodide does not suffer from some of the material issues, such as a solid state phase transition and dissociation in air, that mercuric iodide (another high-Z, wide band-gap semiconductor) does. Thus, bismuth tri-iodide is both easier to grow and handle than mercuric iodide. A modified vertical Bridgman growth technique is being used to grow large, single bismuth tri-iodide crystals. Zone refining is being performed to purify the starting material and increase the resistivity of the crystals. The single crystals being grown are typically several hundred mm^3. The larger crystals grown are approximately 2 cm^3. Initial detectors are being fabricated using both gold and palladium electrodes and palladium wire. The electron mobility measured using an alpha source was determined to be 260 ± 50 cm^2/Vs. An alpha spectrum was recorded with one of the devices; however the detector appears to suffer from polarization.

1. Introduction

Wide band-gap semiconductors often possess the material properties necessary for high resolution, room temperature, gamma-ray spectroscopy [1]. Some of the iodine based semiconductors, such as mercuric iodide (HgI_2) and lead iodide (PbI_2), have already demonstrated their ability to be used as radiation detectors. Heavy metal iodide semiconductors tend to have high density, high-Z and large band gaps, all of which are necessary for room temperature gamma-ray spectroscopy [2]. Unfortunately, there are material and growth issues associated with both PbI_2 and HgI_2 that have encouraged the exploration of alternative semiconductors. Bismuth tri-iodide (BiI_3) is a wide band-gap semiconductor that has high density (5.8 g/cm^3) and high atomic number ($Z_{Bi}=83$, $Z_I=53$). Bismuth tri-iodide has material properties similar to those of HgI_2 and PbI_2; however, BiI_3 has only one solid phase (rhombohedral) unlike HgI_2, which suffers from a solid phase transition, and BiI_3 is not as soft as PbI_2 [3]. Thus, BiI_3 is both easier to grow and handle than HgI_2 and PbI_2, making it an attractive option for radiation detection.

2. Crystal growth and characterization

Large, single BiI_3 crystals were grown using a modified vertical Bridgman technique (Fig. 1). The crystals were grown in a custom EDG Sunfire furnace with 24 programmable temperature zones using a previously established temperature profile [4]. The temperature profile was dynamically controlled, which allowed the ampoule to remain stationary throughout the growth process. Optimization of the growth parameters demonstrated which combinations produce single crystals. Rocking curves demonstrated that the highest quality crystal was obtained with a temperature gradient of 10 °C/cm and a growth rate of 0.5 mm/h. Single crystals on the order of 100–200 mm^3 are consistently being grown. The largest single crystal grown thus far, was approximately 2 cm^3. X-ray diffraction spectra were used to verify that the crystals were single crystals and not polycrystalline [5]. The band-gap of the crystals was determined to be 1.68 ± 0.09 eV at room temperature through the use of UV–vis spectra.

Vapor phase transport was utilized to refine the starting powder. Decreasing the impurities in the crystals should increase the resistivity and reduce charge trapping [6]. A 330 °C hot zone and a 250 °C cold zone was used for powder refining. Impurity analysis (ICP-AES) showed that the crystals have impurity levels consistent with vendor reports (MV Laboratories) for the impurities in the starting material. To test for effects of off-gassing

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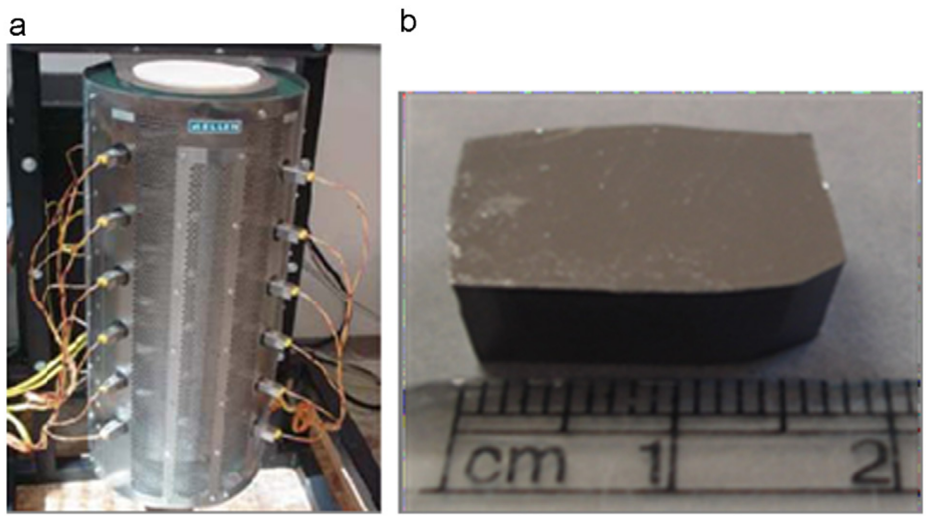

Fig. 1. Modified vertical Bridgman furnace (a) used to grow BiI3 crystals (b).

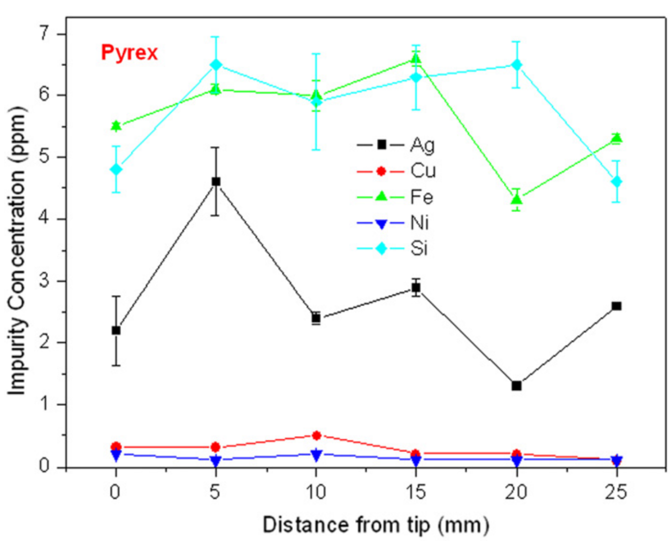

Fig. 2. Impurity analysis performed along the length of the crystal.

by the Pyrex glass ampoules crystals were also grown in quartz ampoules. The crystals grown in the quartz ampoules had statistically insignificant differences in impurity levels from the crystals grown in Pyrex glass ampoules. Measurements of the impurity levels along the length of the crystal indicated that the impurity concentrations were consistent throughout the crystals (Fig. 2).

3. Detector fabrication

The crystals were cut with a diamond wire saw to form detectors. Initial detectors were fabricated with both sputtered gold and palladium electrodes (Fig. 3). Palladium wire was attached to the electrodes with silver epoxy. The electrodes were applied to cut surfaces, surfaces cleaned with nitrogen and surfaces cleaned with argon. *I–V* curves generated by the detectors demonstrated that both types of electrodes resulted in ohmic contacts. Resistivity values with both types of electrodes, obtained from the *I–V* curves, were on the order of 10^8–10^9 Ω cm (Fig. 4). Not all of the detectors fabricated displayed good ohmic behavior; some showed undesirable characteristics that could be due to impurities, poor electrode contacts, improper handling or poor environmental conditions during sputtering (i. e., loss of vacuum).

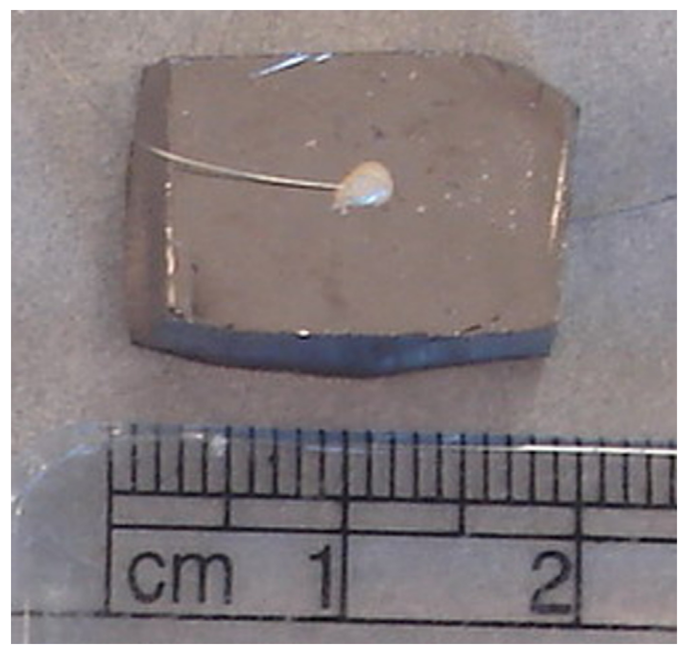

Fig. 3. Detector fabricated with Pd electrodes.

4. Radiation detection

The charge carrier mobilities were measured from the response of a detector to an ^{241}Am alpha source. The electron mobility (Fig. 5) was calculated to be 260 ± 50 cm^2/Vs by measuring the electron rise time. The poor signal from holes prevented a value for hole mobility from being obtained. The poor hole mobility indicates that BiI_3 may be a candidate material for utilizing single polarity charge sensing techniques [7].

An alpha spectrum from the same ^{241}Am source was recorded using a detector with Pd electrodes (Fig. 6). Initial results showed a clear peak; however, after approximately 90 min the spectrum became noisy and the leakage current on the detector overwhelmed any true radiation event signal from being recorded.