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Charge Trapping Effects in THM and VGF Grown CdZnTeSe Radiation Detectors

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Abstract—The performance of state-of-the-art $\text{Cd}_{1-x}\text{Zn}_x\text{Te}_{1-y}\text{Se}_y$ (CZTS) detectors, an emerging wide bandgap high-resolution room temperature semiconductor radiation detector, is limited by the presence of various charge trapping centers including impurities and other intrinsic defects. In this paper we report the growth of CZTS single crystals using two different methods viz., travelling heater method (THM) and vertical gradient freeze (VGF) method. Detectors in planar configuration have been fabricated from the THM- and VGF-grown crystals. Both types of detectors exhibited very high bulk resistivity $>10^{10} \Omega\text{-cm}$ and excellent electron mobility-lifetime ($\mu\tau$) product $>3 \times 10^{-3} \text{ cm}^2/\text{V}$. Although the THM-grown crystal detector showed a $\mu\tau$ product almost twice that of the VGF grown crystal detector, an electron drift mobility of $\approx 1000 \text{ cm}^2/\text{V/s}$ has been observed in the VGF detector, which is much higher than that usually observed in THM detectors. The defects involved in trapping have been studied using photoinduced current transient spectroscopy (PICTS) to understand the observed difference in the charge transport parameters between the two types of the CZTS crystals. Biparametric correlation studies have been carried out through digital gamma spectroscopy to study the effect of charge trapping on pulse-height and risetime of the detector output pulses and to recover the gamma spectra from the effects of charge trapping.

Index Terms— Semiconductor detectors, CdZnTeSe (CZTS), wide bandgap semiconductors, Biparametric correlation, charge trapping, deep level defects, and gamma ray detection.

I. INTRODUCTION

WIDE bandgap semiconductor $\text{Cd}_{1-x}\text{Zn}_x\text{Te}_{1-y}\text{Se}_y$ (CZTS) is an emerging quaternary material for room-

temperature detection of x-rays and gamma-rays for medical imaging, nuclear material safeguards, homeland security, and high-energy physics [1]–[3]. CZTS is grown by adding selenium to the $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ (CZT) matrix during growth wherein selenium (Se) replaces some of the tellurium (Te) atoms from its position in the CZT zinc-blend crystal lattice [3]. CZTS has reportedly demonstrated unprecedented high crystal growth yield due to excellent spatial homogeneity and reduction in crystal defects compared to that obtained for CZT [4]. Crystal growth yield above 90% compared to that of 33% in CZT ensure a lowering of the cost of detector production. CZTS single crystals exhibit very high energy resolution for a wide range of gamma-photon energy because of the excellent electron transport properties [1]. The present state-of-the-art CZTS detectors grown by travelling heater method (THM) exhibit very high energy resolution of $\approx 0.7\%$ for 662-keV gamma photons [1]. The performance of CZTS detectors, irrespective of the growth method, is limited by the charge trapping/recombination in various intrinsic and extrinsic defects [3]–[8]. In the present work, we demonstrate the fabrication of CZTS detectors through a vertical gradient freeze (VGF) method. VGF is a low-temperature growth method and avoids relative motion between the heater and the ampoule containing the precursor materials, thus avoiding thermal drift or temperature fluctuations [9]–[10]. The VGF grown detectors have been characterized in terms of radiation detection performance and compared with that of standard THM-grown detectors. The defects responsible for charge trapping have been studied through photoinduced current transient spectroscopy (PICTS). The effects of charge trapping on high energy gamma-ray detection and subsequent recovery scheme have been investigated through digital biparametric (BP) correlation studies.

II. EXPERIMENTAL METHOD

The THM CZTS crystals in the stoichiometry $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}_{0.98}\text{Se}_{0.02}$ were synthesized from 6N purity $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ and CdSe precursor (pre-synthesized) materials at Brookhaven National Laboratory (BNL). The VGF CZTS crystals were grown from 7N purity elemental precursors with a target stoichiometry $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}_{0.97}\text{Se}_{0.03}$ at the University of South Carolina (UofSC). For both the methods, the precursors were sealed in conically tipped quartz ampoules under high

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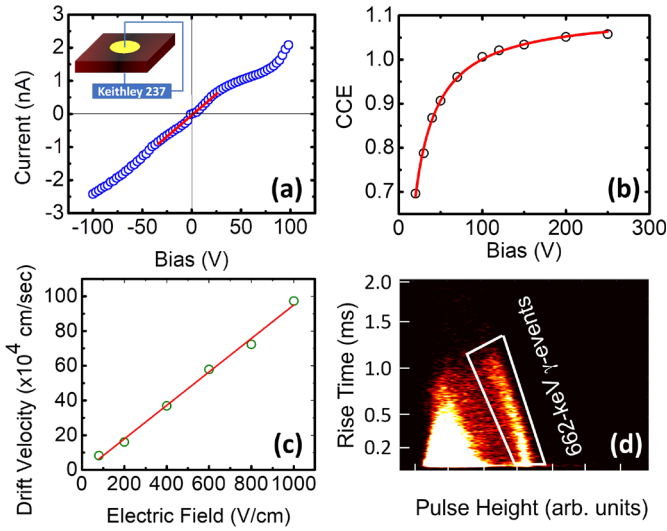


Fig. 1. (a) Current-voltage (I - V) characteristics measured at room temperature. (b) Variation of charge collection efficiency as a function of bias voltage. The solid line shows a single-polarity Hecht equation fit. (c) Variation of electronic drift velocity as a function of electric field. The solid line shows a linear fit. (d) A biparametric (BP) correlation plot obtained by exposing the detector to a ^{137}Cs radiation source. The enclosed region shows the 662-keV γ -events affected by charge trapping for a VGF grown CZTS planar detector.

vacuum ($\sim 10^{-7}$ torr), and all the growths were unseeded. Three-zone vertical furnaces have been used for the growth runs. For the THM growth the temperatures of the three zones were set to achieve the required temperature gradient near the growth interface, while only two heating zones out of three have been used for the VGF growth to obtain the desired temperature gradient.

Radiation detectors in planar geometry have been fabricated after cutting wafers from the grown ingots followed by polishing, cleaning, and gold (Au) deposition. Current-voltage (I - V) characteristics measurements were carried out to determine the leakage currents and the electrical bulk resistivities of the CZTS detectors. Analog alpha spectroscopic methods have been used to determine the radiation response and the electron mobility-lifetime ($\mu\tau$) product measurements. A digital alpha particle spectrometer was used to obtain the electron drift mobilities and the BP correlation plots. Defect studies have been carried out through PICTS where current transients induced by a 635-nm pulsed laser were recorded as a function of temperature in the range 85-400 K.

III. RESULTS AND DISCUSSIONS

Figure 1(a) shows the I - V characteristic of a planar CZTS detector grown using the VGF method. A resistivity of $3 \times 10^{10} \Omega\text{-cm}$ has been measured from the I - V plot which is similar to that obtained for standard THM-grown CZTS crystals [3]. A leakage current of ≈ 2 nA has been measured at bias voltage of ± 100 V. Figure 1(b) shows the variation of charge collection efficiency (CCE) measured as a function of the bias voltage applied to the VGF grown CZTS crystal, which was fit to a single polarity Hecht equation to obtain a $\mu\tau$ product of 3×10^{-3}

cm^2/V . The obtained $\mu\tau$ product was lower by a factor of 2 than that reported for THM-grown CZTS crystals. The variation of electron drift velocity with applied electric field obtained for the VGF detector is shown in Fig. 1(c). An electron drift mobility of $\approx 1000 \text{ cm}^2/\text{V}\cdot\text{s}$ was calculated from the plot. Figure 1(d) presents a BP correlation plot showing the effect of charge trapping on gamma-photon induced detector pulses obtained by exposing the VGF detector to a ^{137}Cs source emitting 662-keV gamma photons.

IV. CONCLUSION

$\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}_{1-y}\text{Se}_y$ ($y = 0.02$ and 0.03) single crystals have been grown through THM and VGF crystal growth methods. Radiation detectors in planar configuration were fabricated and characterized through electrical and radiation detection measurements. Defect studies have been carried out through PICTS measurements and will be presented to correlate the detection performance of the CZTS detectors with the charge trapping effects. The results of the behavioral difference of the charge trapping observed between the detectors fabricated from two different growth methods will be presented.

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