

Charge transport performance of high resistivity CdZnTe crystals doped with In/Al

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Abstract: To evaluate the charge transport properties of as-grown high resistivity CdZnTe crystals doped with In/Al, the α particle spectroscopic response was measured using an un-collimated ²⁴¹Am (5.48 MeV) radioactive source at room temperature. The electron mobility lifetime products $(\mu\tau)_e$ of the CdZnTe crystals were predicted by fitting plots of photo-peak position versus electrical field strength using the single carrier Hecht equation. A TOF technique was employed to evaluate the electron mobility for CdZnTe crystals. The mobility was obtained by fitting the electron drift velocities as a function of the electrical field strengths, where the drift velocities were achieved by analyzing the rise-time distributions of the voltage pulses formed by a preamplifier. A fabricated CdZnTe planar detector based on a low In concentration doped CdZnTe crystal with $(\mu\tau)_e = 2.3 \times 10^{-3} \text{ cm}^2/\text{V}$ and $\mu_e = 1000 \text{ cm}^2/(\text{V}\cdot\text{s})$, respectively, exhibits an excellent γ -ray spectral resolution of 6.4% (FWHM = 3.8 keV) for an un-collimated ²⁴¹Am @ 59.54 keV isotope.

Key words: CdZnTe crystals; α particle pulse height spectra; charge transport performance; In/Al doping

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1. Introduction

CdZnTe (CZT) crystals have been gradually attracting more attention over the last decade, and are considered as a promising material for the fabrication of room temperature X/ γ -ray detectors with excellent energy resolution^[1,2]. A high sensitivity and response X/ γ -ray spectrometer and imaging system have been produced by using a CZT pixelated array or Frisch grid detector combining with standard integrated reading circuits, without any additional cooling system. CZT detectors have been widely used in environmental monitoring, nuclear medicine, astrophysics, high-energy physics, safety defense, inspection of nuclear wastes and many other fields, and have been considered as substitutes for scintillator detectors such as NaI (Tl) and CsI (Tl)^[3].

To obtain high resistivity ($\rho > 10^{10} \text{ }\Omega\cdot\text{cm}$) CZT single crystal is essential and significant for detector fabrication and operation under relatively high bias voltages. However, the resistivity of CZT material grown by the melt growth method is usually degraded due to the high Cd partial pressure during CZT crystal growth, resulting in a non-stoichiometric melt, and therefore producing abundant Cd vacancies (V_{Cd}) acting as acceptors. Intentionally introducing donor impurity (such as Ge, In, Sn, Cl) into the CZT crystals has been considered a promising compensation method to approach high resistivity, and plenty of literature has been published on this^[4-6]. The charge carrier transport behavior is a significant parameter which can reflect both charge collection efficiency and energy resolution. Therefore, it is meaningful to characterize the charge transport properties in order to select high-quality detector-grade CZT crystals. In this work, as-grown high re-

sistivity CZT crystals doped with In/Al were evaluated by α particle pulse height spectra. The electron mobility lifetime product $(\mu\tau)_e$ was obtained, and the electron mobility μ_e was calculated by analyzing the rise time distribution of the pulse shapes.

2. Experiments

Three Cd_{0.9}Zn_{0.1}Te ingots doped in different situations were grown using a modified vertical Bridgman method, as seen in Table 1. As-grown wafers CZT1, CZT2 and CZT3 were directionally sliced from the middle part of the corresponding ingot, with a surface orientation of {111} and dimensions as shown in Table 1. The wafers were consecutively mechanically polished and chemically etched prior to the evaporation. The Au/CZT/Au devices were fabricated by thermally evaporating Au contacts on each face of the wafers with a thickness of 70 nm; the electrode sizes are shown in Table 1. Subsequently, the bared CZT surface was passivated by using 30% H₂O₂ solution for 2 min in order to reduce the leakage current. Finally, one face of the Au/CZT/Au was bonded by graphite paste to a ceramic chip carrier, while the opposite face was connected using Ag wire with Au paste for further measurements.

Current-voltage (I - V) measurements were performed on the planar devices by using a Keithley 487 picoammeter/voltage supply, and the resulting resistivity is shown in Table 1. The charge transport properties of the CZT crystals were evaluated using α particle spectroscopy. The measurements were carried out by placing the devices inside a temperature-controlled vacuum chamber at pressure below 2×10^{-2} Torr.

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Table 1. Properties of the samples CZT1, CZT2 and CZT3.

Sample	Dimension (mm ³)	Resistivity (Ω·cm)	Electrode areas (mm ²)	Dopant condition
CZT1	8 × 8 × 3.5	3.9 × 10 ¹⁰	5 × 5	Heavily doped with In
CZT2	10 × 10 × 2	3.4 × 10 ¹⁰	6.5 × 6.5	Lightly doped with In
CZT3	10 × 10 × 1.2	7.5 × 10 ¹⁰	6.5 × 6.5	Al

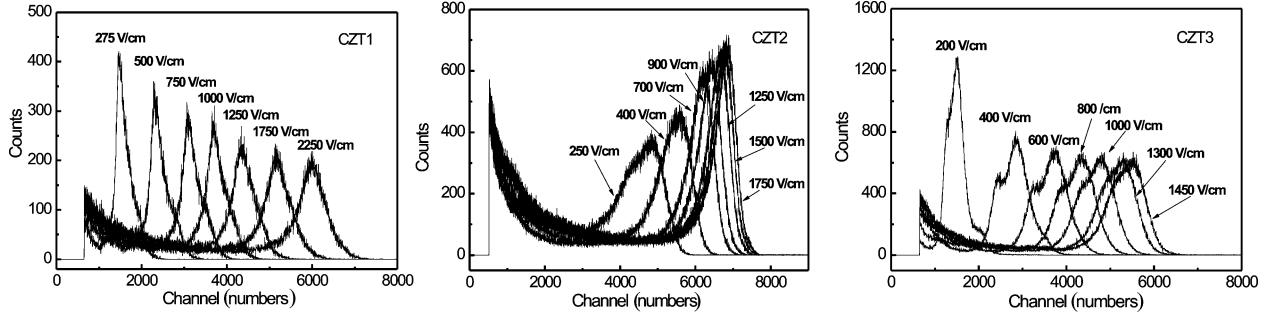


Fig. 1. α particle pulse height spectra as a function of electrical field strength for CZT1, CZT2 and CZT3, respectively.

Pulse height spectra as a function of bias voltages were obtained, illuminated by an un-collimated ²⁴¹Am 5.48 MeV α particle source. Pulse shape information was simultaneously recorded by connecting the pre-amplifier output to a high-speed waveform digitizer card. The rise time distribution from the CZT detector was analyzed by using Scopeview soft. In addition, an ORTEC 710 bias supply, an ORTEC 142 charge sensitive pre-amplifier and an ORTEC 570 shaping amplifier with an optimized shaping time of 2 μ s were used during the measurement. Finally, a standard multi channel analyzer (CAN-DERRA Multiport II) was used for the pulse height spectra acquisition.

3. Results and discussion

3.1. α particle induced pulse height spectra

Generally, the charge collection efficiency (CCE) is dependent on the drift length (λ) of the charge carrier and the thickness (d) of the sample. For the non-saturated situation, drift length is related to mobility (μ), lifetime (τ) and applied electrical field strength (E) in the form of

$$\lambda = \mu\tau E, \tag{1}$$

$$E = \frac{V}{d}, \tag{2}$$

where V is the bias voltage. According to the Hecht equation^[7]:

$$CCE(x) = \frac{\lambda_h}{d} \left(1 - \exp \frac{-x}{\lambda_h} \right) + \frac{\lambda_e}{d} \left(1 - \exp \frac{x-d}{\lambda_e} \right). \tag{3}$$

x is the distance from the position of electron-hole pair generation to the illuminated electrode, and λ_e and λ_h are the drift lengths for electron and hole, respectively. Due to the short range of 5.48 MeV α particles in CZT crystal (approximately 18 μ m^[8]), the electron-hole pairs are generated in a shallow region, which is very close to the irradiated contact, and the resulting induced charge signal is predominantly due to the

drift of a single carrier polarity, so one item in Eq. (3) can be ignored. Taking Eq. (1) into Eq. (3), the single carrier Hecht equation is given by the following equation:

$$CCE \approx \frac{\mu\tau V}{d^2} \left(1 - \exp \frac{-d^2}{\mu\tau V} \right). \tag{4}$$

Figure 1 shows typical ²⁴¹Am (5.48 MeV) α particle pulse height spectra as a function of bias voltages for CZT1, CZT2 and CZT3, respectively, under cathode irradiation at room temperature. It is revealed that the photo-peak centroid shifts to the high channel direction as the applied bias increases. Because the photo-peak position is sensitive to CCE, the resulting $(\mu\tau)_e$ values for CZT1, CZT2 and CZT3 are $(0.62 \pm 0.02) \times 10^{-3}$, $(2.34 \pm 0.02) \times 10^{-3}$ and $(0.46 \pm 0.01) \times 10^{-3}$ cm²/V respectively, by fitting the photo-peak centroid versus the electrical field strength using Eq. (4), as illustrated in Fig. 2. Similar data were obtained for holes $(\mu\tau)_h$, with the bias polarity reversed so that the irradiated surface acted as the anode. Since $(\mu\tau)_h$ is normally approximately 2 orders of magnitude lower than $(\mu\tau)_e$, and the applications of CZT detectors are mainly based on the electron performance, hole properties are neglected in this paper.

3.2. TOF method for evaluation of charge mobility

The use of a time of flight (TOF) technique to evaluate the charge mobility of CZT crystals was reported in Ref. [9]. Firstly, the voltage pulses output from the pre-amplifier were collected by a high-speed waveform digitizer card. Then the charge drift time was acquired by analyzing the pulse height rise time distribution (normally, a 10%–90% amplitude rise time $\tau_{10\%-90\%}$ was selected). Figure 3 shows the typical 10%–90% rise time distribution for CZT2, and the inset is a representative voltage pulse from the corresponding situation. According to the expression given as follows:

$$V_{dr} = \frac{d}{t_{dr}}, \tag{5}$$

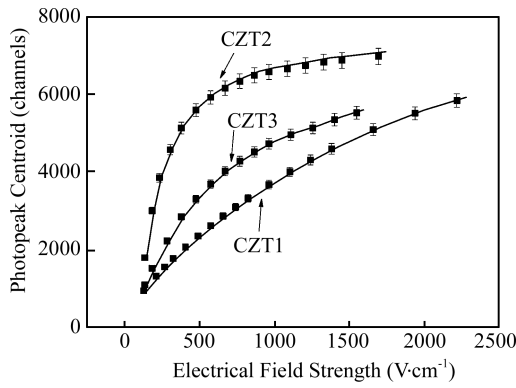


Fig. 2. Comparison of the electron mobility lifetime products of CZT1, CZT2 and CZT3, fitted using the single carrier Hecht equation.

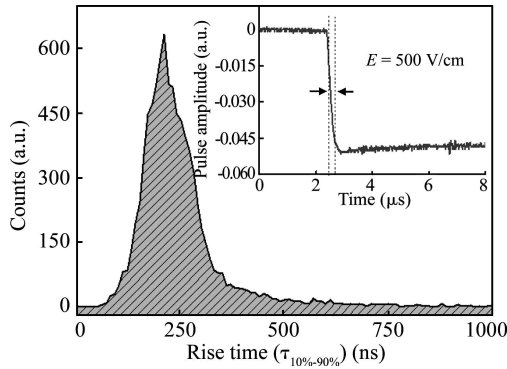


Fig. 3. Resulting distribution of the pulse shape rise time (10%–90%) for CZT2 under an electrical field strength of 500 V/cm. Inset is a typical pulse shape.

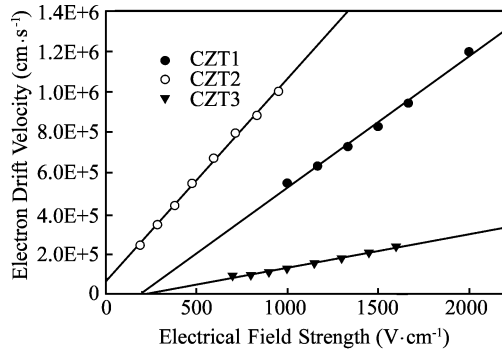


Fig. 4. Electron drift velocity as a function of electrical field strength for CZT1, CZT2 and CZT3, respectively.

the charge drift velocity (V_{dr}) can be achieved. Due to the correlation of

$$V_{dr} = \mu E, \tag{6}$$

the charge mobility can be examined by fitting the plot of V_{dr} – E . The resulting electron mobility μ_e for CZT1, CZT2 and CZT3 was (690 ± 40) , (1000 ± 19) and (200 ± 12) $\text{cm}^2/(\text{V}\cdot\text{s})$, respectively, as illustrated in Fig. 4. Similar data were obtained for holes μ_h .

Taking into account the data above, the deterioration of $(\mu\tau)_e$ for CZT1 was mainly attributed to the short electron lifetime τ_e , which suggested abundant electron trapping centers in the material. Generally, deep level defects in the crystals were recognized as charge carrier trapping centers. Currently,

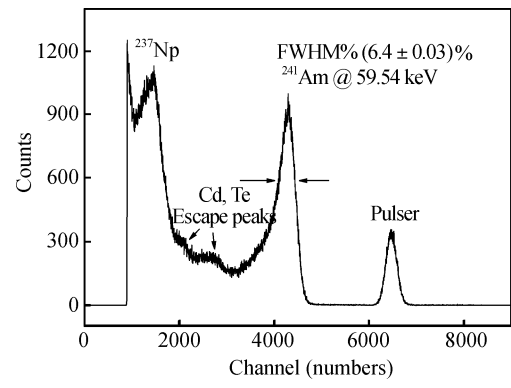


Fig. 5. Room temperature γ -ray spectroscopic response for CZT2 planar detector irradiated by an un-collimated ^{241}Am (59.54 keV) source.

however, there is still controversy about the origin of the different deep level defects or complexes in CZT materials^[10, 11]. In our recent study^[12], Cd_i^{2+} acting as an electron trap center was proposed as a possible deep level defect in heavy In doped samples CZT1, which in turn degraded the τ_e . In contrast to CZT3, the main contribution to the deterioration of $(\mu\tau)_e$ was mostly the low μ_e . In general, mobility was influenced by ionized impurity scattering and lattice vibration scattering. In terms of heavy Al doped CZT3, surplus Al atoms could introduce plenty of interstitial Al_i into the material caused by the low doping efficiency, and therefore produced the majority of the ionized impurity scattering centers, which are responsible for the low μ_e . In the case of lattice vibration scattering, it can be divided into acoustics scattering and optics scattering. The former is dependent on the temperature and the variation of the conduction band caused by deformation of the material, while the latter is usually determined by the temperature. They were both ignored as the measurement was carried out at room temperature. Besides, the Cd vacancy, Te vacancy and other point defects were also recognized as scattering centers. The dislocations and neutral impurities scattering likely degrade the μ_e to a certain extent. Nevertheless, the influence is quite weak according to the adopted high purity (7N) raw materials. The $(\mu\tau)_e$ and μ_e for CZT2 are high, suggesting that the deep level traps and charge scattering centers in the material are very low, which indicates the excellent crystal quality.

3.3. ^{241}Am (59.54 keV) γ -ray spectroscopic response

A typical γ -ray spectroscopic response for the fabricated planar detector based on sample CZT2 was acquired using 59.5 keV γ -rays from an un-collimated ^{241}Am isotope at room temperature, as shown in Fig. 5. Generally, the full-width at half-maximum (FWHM) of the full-energy peaks was assumed to be a measure of the energy resolution of the detector. The resulting energy resolution was $(6.4 \pm 0.03)\%$ (FWHM = (3.81 ± 0.02) keV) without any additional signal processing. The ^{237}Np peak can be observed on the low energy side, and the Cd and Te escape peaks are clearly resolved. The electronic noise of the preamplifier system is simultaneously indicated by the pulser peak, which indicates a remarkable deterioration

in the charge collection. Therefore, the energy resolution of the detector can be improved by decreasing the adverse effect above.

The γ -ray spectroscopic responses for detectors CZT1 and CZT3 irradiated by ^{41}Am (59.54 keV) were also acquired in the same measuring situation, in which the photo-peaks are clearly seen; the energy resolution was 13.1% (FWHM = 7.8 keV) and 15.3% (FWHM = 9.1 keV), respectively. High resistivity crystals can ensure detector operation on high bias voltages. However, the charge collection efficiency and energy resolution of the detector were mainly determined by the charge transport behavior.

4. Conclusions

In this work, electron mobility lifetime products $(\mu\tau)_e$ for as-grown high resistivity CZT crystals doped in different situations were obtained, illuminated by an un-collimated ^{241}Am 5.48 MeV α particle source. In parallel, the electron mobility μ_e was calculated by using the TOF technique. The results demonstrate the following:

(1) Cd_i^{2+} acting as an electron trap center was proposed as possibly the main deep level defect in heavy In doped CZT crystals, which in turns degraded the τ_e value.

(2) The resulting $(\mu\tau)_e$ and μ_e for the low In doped CZT crystal was $2.3 \times 10^{-3} \text{ cm}^2/\text{V}$ and $1000 \text{ cm}^2/(\text{V}\cdot\text{s})$, respectively. The energy resolution was approximately 6.4% (FWHM = 3.8 keV) for the fabricated CZT planar detector, irradiated by 59.5 keV γ -rays from an un-collimated ^{241}Am isotope at room temperature.

(3) In the case of the Al doped CZT crystal, surplus Al atoms could introduce plenty of interstitial Al_i into the material, and therefore produced sufficient ionized impurity scat-

tering centers, which were mostly responsible for the low μ_e .

References

- [1] Li G, Jie W, Hua H, et al. $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$: growth and characterization of crystals for X-ray and gamma-ray detectors. *Prog Crystal Growth and Charact*, 2003, 46: 85
- [2] Triboulet R, Scheel H J, Fukuda T. *Crystal growth technology*. New York: Wiley, 2003
- [3] Szeles C. CdZnTe and CdTe materials for X-ray and gamma ray radiation detector applications. *Phys Status Solidi B*, 2004, 241(3): 783
- [4] Fiederle M, Babentsova V, Franca J, et al. Defect structure of Ge-doped CdTe. *J Cryst Growth*, 2002, 243: 77
- [5] Yang G, Jie W, Li Q, et al. Effects of In doping on the properties of CdZnTe single crystals. *J Cryst Growth*, 2005, 283: 431
- [6] Fiederle M, Fauler A, Konrath J, et al. Comparison of undoped and doped high resistivity CdTe and (Cd, Zn)Te detector crystals. *IEEE Trans Nucl Sci*, 2004, 51: 1864
- [7] Knoll G F. *Radiation detection and measurement*. 3rd ed. New York: Wiley, 2000
- [8] Ruzin A, Nemirovsky Y. Methodology for evaluation of mobility-lifetime product by spectroscopy measurements in CdZnTe spectrometers. *J Appl Phys*, 1997, 82: 4166
- [9] Sellin P J, Davies A W, Gkoumas S, et al. Ion beam induced charge imaging of charge transport in CdTe and CdZnTe. *Nucl Instrum Methods B*, 2008, 266: 1300
- [10] Du M, Takenaka H, Singh D J. Carrier compensation in semi-insulating CdTe: first-principles calculations. *Phys Rev B*, 2008, 77: 094122
- [11] Saucedo E, Franc J, Elhadidy H, et al. Investigation of the origin of deep levels in CdTe doped with Bi. *J Appl Phys*, 2008, 103: 094901
- [12] Xu Y, Jie W, Sellin P, et al. Study on temperature dependent resistivity of indium-doped cadmium zinc telluride. *J Phys D*, 2009, 42: 03505