

Determination of trap levels in CZT:In by thermally stimulated current spectroscopy

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Abstract: Many defects in semi-insulating (SI) cadmium zinc telluride ($\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ or CZT) ingots grown by the melt methods act as trapping centers to introduce deep levels in the band gap, which has strong effects on CZT detection properties. The thermally stimulated current (TSC) spectroscopy was used to measure these traps, and the initial rise method and the simultaneous multiple peaks analysis (SIMPA) method were introduced to characterize trap levels in SI-CZT:In. The results show that there is a larger error in the determination for the trap peaks with the initial rise method due to the interference of overlapping peaks, while the SIMPA method demonstrates a better performance in resolving these overlapping peaks simultaneously for a full characterization of trap levels. On this basis, a theoretical SIMPA fitting, which is composed of ten trap levels and a deep donor level E_{DD} dominating the dark current in SI-CZT:In, is achieved. Furthermore, the reason of high resistivity in CZT:In was explained by the relationship between E_{DD} level and Fermi level.

Key words: $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$; trapping; deep levels; thermally stimulated current spectroscopy

1 Introduction

Semi-insulating (SI) cadmium zinc telluride ($\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ or CZT) is an advanced material for fabricating X-ray and gamma-ray detectors operating at room temperature because of its high resistivity and good mobility-lifetime products [1–4]. The high resistivity is obtained by electrical compensation processes: usually, group III (Al, Ga, In) or group VII (Cl) elements are doped to compensate the native defects effectively in the material [5,6]. However, there are different defects in CZT ingots grown by the melt methods, including native defects, residual impurities and complexes, which act as trapping centers to introduce deep levels in the band gap [7,8]. These deep levels exert strong effects on the electrical properties and charge transport behaviors, determining the final detector performance properties [9,10]. Therefore, the identification and characterization

of the electrically active defects are of considerable importance for controlling and improving CZT detection properties.

Defects in SI-CZT crystals can be characterized by thermally stimulated current (TSC) spectroscopy. In TSC measurement, the energy level of a particular trap is related to the temperature at which it is emptied, while the number of traps contributing to the stimulated peak is obtained from the amount of charge release. Actually, the precise determination of trap levels in CZT material by TSC measurement is difficult due to the mutual interference of overlapping peaks and their poor resolutions [11,12]. In previous studies, PAVLOVIC and DESNICA [13] have proved an assumption that the measured TSC spectra in CdZnTe material were composed of many overlapping peaks. The initial rise and the SIMPA methods were introduced to comparatively resolve TSC spectra for a full characterization of trap levels which affect charge

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transport behaviors of indium-doped $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ ($x=0.1$) detector. As a consequence, an effective analysis method was determined for an accurate evaluation of trap levels in SI-CZT material.

2 Experimental

SI- $\text{Cd}_{1-x}\text{Zn}_x\text{Te:In}$ ($x=0.1$) crystal with the indium content of 1×10^{-5} was grown by the modified vertical Bridgman (MVB) method in our laboratory, which was described in detail [14,15]. Wafer in the size of $10 \text{ mm} \times 5 \text{ mm} \times 1 \text{ mm}$ cut from the ingot was etched in 2% bromine methanol solution to remove the damaged surface layer, and then electroless Au contacts were deposited with AuCl_3 solution. The electrical resistivity of the crystal was measured to be about $100 \text{ G}\Omega\cdot\text{cm}$ at room temperature by an Agilent 4155C semiconductor parameter analyzer.

In TSC measurement, the wafer was placed in a closed-cycle cryostat with liquid helium (Sumitomo DE-202 closed-cycle refrigerator) and cooled in darkness to 25 K, where the deep traps were filled using a halogen lamp. Then, as the traps were emptied by thermal emission in darkness until 310 K at heating rate of 0.25 K/s with bias voltage of 10 V, TSC currents as a function of temperature were obtained by a Keithley 6514 electrometer. During the experiments, the wafer was kept in darkness to prevent the excitation of charge carriers by light, avoiding unintentional deep trap filling. In the meantime, the temperature dependence of dark current (I_{DC}) was determined by performing the above described procedures without illumination at low temperature.

In addition, the temperature-dependent resistivity was investigated by current-voltage measurement with planar contacts in the range from 271 K to 321 K with bias voltages from -10 V to 10 V .

3 Results and discussion

3.1 Initial rise method

The initial rise method, a kind of “thermal cleaning” procedure, is based on the assumption that the traps begin to empty as the temperature increases [16]. A whole TSC curve from 25 K to 310 K in a SI-CZT:In sample is obtained where the heating rate was 0.25 K/s with bias voltage of 10 V in Fig. 1 (the dashed curve), the sample was subsequently re-cooled down to 25 K and then irradiated again. When it was measured through the same cycle once more, the heating was stopped at a temperature T between T_1 and T_2 . In this way, the traps responsible for the current peaks for $T < T_2$ were substantially emptied and thus the resulting TSC currents were obtained. After subtracting the impact of the dark current, the isolated TSC peak at the temperature of T_1

was obtained. Then, the sample was re-cooled and re-heated in darkness in the temperature range from T_2 to T_3 so as to obtain the isolated T_2 peak. Finally, the results of this procedure are illustrated in Fig. 1, where the solid curves are the isolated TSC currents for the corresponding temperature intervals defined above. By isolating the whole TSC curve with the initial rise method, five main trap peaks are observed, namely T_1 , T_2 , T_3 , T_4 and T_5 at the energy level of 0.091, 0.151, 0.202, 0.337 and 0.436 eV, respectively. It is clearly shown that only parts of the isolated trap peaks with the initial rise method are similar to the results of the whole measured TSC curve, such as the traps of T_1 and T_4 , while others (like the traps of T_2 , T_3 and T_5) are apart from the whole measured TSC result or even isolated multi-peaks. This indicates that the TSC curve from CZT:In is not a classical single spectrum but complex. During the thermal emission procedure of TSC measurement, the simultaneous release of charge carriers from different traps may lead to the neighboring traps overlapping and thus cause the additional shifts of maximum peak in the whole measured TSC curve. The existence of overlapping peaks with poor resolutions and inevitable interference may bring a larger error to the isolation of trap peaks with the initial rise method. As a result, the simultaneous isolation of overlapping peaks in a complex TSC spectrum is very important for characterizing trap levels to the full.

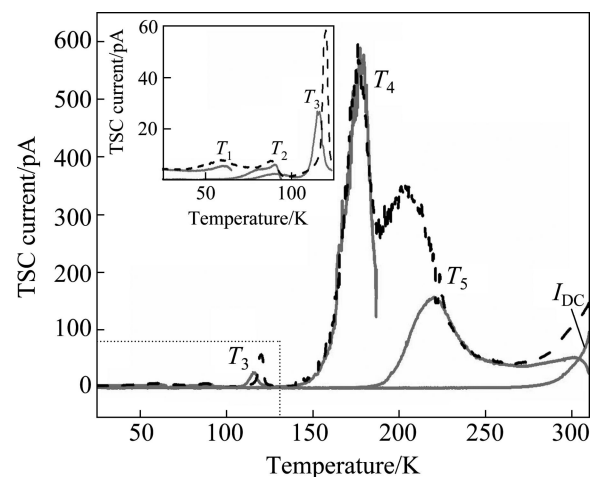


Fig. 1 TSC spectra from SI-CZT:In sample and isolation of trap peaks with initial rise method (Dashed curve presents whole TSC curve from 25 to 310 K and solid plots are isolated TSC currents with initial rise method)

3.2 Simultaneous multiple peak analysis (SIMPA) method

In order to determine all trap levels simultaneously and decrease the sum of offset errors to a minimum from the TSC spectrum with overlapping peaks in SI-CZT:In, the SIMPA method reported by PAVLOVIC and

DESNICA [13] is introduced.

The temperature dependence of fitting function, I_{SIMPA} , comprising the sum of TSC peaks belonging to the specific deep levels I_{TSC} and the dark current I_{DC} , is defined by

$$I_{\text{SIMPA}}(T) = \sum_{i=1}^m I_{\text{TSC}}^i(T) + I_{\text{DC}}(T) \quad (1)$$

where $I_{\text{TSC}}^i(T)$ is the i th individual TSC peak; m is the number of deep traps involved in calculation; $I_{\text{DC}}(T) = C \cdot \exp[-E_{\text{DD}}/(kT)]$, C is a constant, k is the Boltzmann's constant, and E_{DD} is the deep donor activation energy which dominates the dark current.

In the first order kinetics approximation, a single TSC peak resulting from an electron trap can be described as

$$I_{\text{TSC}}^i(T) = N_{T_i} e \mu A E \tau \times 3.0 \times 10^{21} \frac{m}{m_0} \sigma_i T^2 \times \exp \left\{ -\frac{E_{a,i}}{kT} - \frac{3.0 \times 10^{21} \frac{m}{m_0} \sigma_i k}{\beta E_{a,i}} \times T^4 e^{-E_{a,i}/kT} \left[1 - 4 \frac{kT}{E_{a,i}} + 20 \left(\frac{kT}{E_{a,i}} \right)^2 \right] \right\} \quad (2)$$

where e is the electron charge, μ is the carrier mobility, τ is the carrier lifetime, A is the area of electrode, E is the applied electric field, T is the absolute temperature, β is the heating rate, m_0 is the rest mass, m is the effective mass, $E_{a,i}$ is the thermal activation energy of the i th trap which is related to the TSC peak position, and σ_i is the capture cross section of the i th trap.

In addition, N_{T_i} represents the carrier concentration of the i -th trap at the beginning of the temperature ramp, which can be calculated from

$$N_{T_i} = \frac{Q_{T_i}}{2\mu_n(T_i)\tau_n(T_i)eAE} \quad (3)$$

where Q_{T_i} denotes the temporal integral of I_{TSC}^i , that is $Q_{T_i} = \int I_{\text{TSC}}^i dt = \frac{1}{\beta} \int_{T_0}^T I_{\text{TSC}}^i dT$. Therefore, the trap signatures concentration (N_T), thermal activation energy (E_a) and capture cross section (σ) of deep levels in SI-CZT:In can be obtained by above calculations.

This method clearly indicates where TSC peaks are “missing” and/or overlapping. Figure 2 presents the SIMPA fit of the TSC curve obtained from the SI-CZT:In. The dashed curve denotes the whole TSC spectra as mentioned above and the solid curves correspond to each individual TSC peak defined by Eqs. (1) and (2) which belongs to a specific trap. As a result, a better fit is achieved by a theoretical function, which is composed of ten trap levels and the E_{DD} level dominating the dark

current. By calculation with the SIMPA method, the trap signatures of observed traps are characterized simultaneously and the results are presented in Table 1. In comparison with the initial rise method, the SIMPA fit demonstrates very good performance in resolving the composition of overlapping peaks simultaneously to the individual trap peaks and therefore results in the evaluation of real trap values.

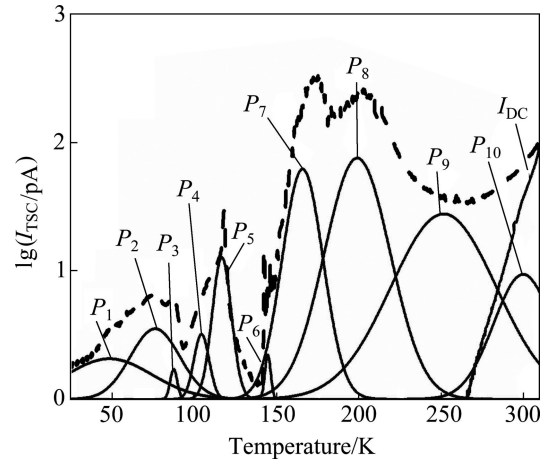


Fig. 2 TSC spectra on SI-CZT:In with SIMPA method (Dashed curve is experimental whole TSC curve, and solid curves are individual SIMPA fitting peaks which belong to specific traps)

Table 1 Trap signatures of observed trap levels in SI-CZT:In determined with SIMPA method

Trap level	Peak maximum/K	Activation energy/eV	Concentration/cm ⁻³	Capture cross section/cm ²
P_1	48	0.07	2.1×10^{14}	1.00×10^{-21}
P_2	76	0.123	2.5×10^{14}	3.61×10^{-19}
P_3	87	0.145	1.5×10^{13}	1.54×10^{-18}
P_4	104	0.18	7.2×10^{13}	4.77×10^{-18}
P_5	116	0.205	2.0×10^{14}	4.83×10^{-18}
P_6	145	0.265	1.9×10^{13}	2.97×10^{-18}
P_7	166	0.312	6.8×10^{14}	2.46×10^{-18}
P_8	199	0.387	1.1×10^{15}	6.66×10^{-18}
P_9	252	0.509	1.4×10^{15}	2.53×10^{-18}
P_{10}	300	0.626	3.8×10^{14}	8.80×10^{-18}
E_{DD}	—	0.704	—	—

Among these traps, trap P_1 , with an activation energy of 0.07 eV, a capture cross section of $1.00 \times 10^{-21} \text{ cm}^2$ and a concentration of $2.1 \times 10^{14} \text{ cm}^{-3}$, is deduced as an acceptor level. The defect associated with this level is probably correlated with the impurity (Au, Ag, etc) [17]. Traps with similar activation energies are also often identified as the acceptor level of Cd vacancy [18].

Trap P_2 with E_a , σ and N_T of 0.123 eV, $3.61 \times 10^{-19} \text{ cm}^2$, $2.5 \times 10^{14} \text{ cm}^{-3}$, respectively, and trap P_3 (0.145 eV,

$1.54 \times 10^{-18} \text{ cm}^2$, $1.5 \times 10^{13} \text{ cm}^{-3}$) may correspond to the shallow acceptor level (or band) of the so-called A center, which is reported in the range of 0.12–0.15 eV [19]. The microscopic origin of this level has been attributed to ($\text{V}_{\text{Cd}}\text{-In}_{\text{Cd}}$) complex.

Trap P_4 (0.18 eV, $4.77 \times 10^{-18} \text{ cm}^2$, $7.2 \times 10^{13} \text{ cm}^{-3}$) and trap P_5 (0.205 eV, $4.83 \times 10^{-18} \text{ cm}^2$, $2.0 \times 10^{14} \text{ cm}^{-3}$) may be assigned to the electron levels of Te_{Cd^+} or the Te_{Cd} related defects [10,20], which correspond to the level at 0.20 eV in Ref. [10] and the TEES defect P_0 or P_1 in Ref. [20].

Trap P_6 (0.265 eV, $2.97 \times 10^{-18} \text{ cm}^2$, $1.9 \times 10^{13} \text{ cm}^{-3}$) is at the same position as the peak of level C found in $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ by CAVALLINI et al [21]. The origin of this level is probably attributed to the structural defects introduced by compositional segregation, precipitation, inclusion and/or constitutional super-cooling.

Trap P_7 (0.312 eV, $2.46 \times 10^{-18} \text{ cm}^2$, $6.8 \times 10^{14} \text{ cm}^{-3}$) corresponding to the trap T_3 at 0.321 eV confirmed by an inductively coupled plasma mass spectroscopy (ICP-MS) analysis in Ref. [12] may be ascribed to the acceptor level formed by impurity Cu.

Trap P_8 (0.387 eV, $6.66 \times 10^{-18} \text{ cm}^2$, $1.1 \times 10^{15} \text{ cm}^{-3}$) is similar to the level at 0.4 eV reported by BERDING [22], which is assigned to an electron trap from the doubly ionized tellurium antisite ($\text{Te}_{\text{Cd}}^{2+}$) related defects or complexes.

Trap P_9 with E_a of 0.509 eV may be attributed to Cd vacancy (V_{Cd}) above the valence band. EMANUELSSON et al [23] also observed this level by photo-electron paramagnetic resonance (Photo-EPR) measurement (0.47 eV), which corresponds to the acceptor level of double ionized Cd vacancy.

Trap P_{10} (0.626 eV, $8.80 \times 10^{-18} \text{ cm}^2$, $3.8 \times 10^{14} \text{ cm}^{-3}$) may be claimed to have an acceptor character and Zn-related origin. The defects at 0.55–0.65 eV associated with Zn cation vacancy have been found in PICTS measurements of $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ and $\text{Cd}_{0.8}\text{Zn}_{0.2}\text{Te}$ [19, 24].

Furthermore, the deep donor level E_{DD} dominates the dark current above room temperature in SI-CZT:In, which is similar to the ‘EL2’ mid-gap level in SI-GaAs [25]. By calculation, the E_{DD} level at 0.704 eV near the mid-gap of CZT:In at room temperature ($E_g=1.57$ eV) is determined. It is generally accepted that high resistivity CZT materials are achieved through the pinning of the Fermi level E_F near mid-gap by compensation. The E_{DD} level plays an important role in compensation processes, and it is responsible for the Fermi level pinning near the mid-gap [26]. To further confirm the hypothesis, the Fermi level of the SI-CZT:In by the linear fit of temperature dependence of resistivity was calculated, $\ln \rho \propto \Delta E/(kT)$ [27], as shown in Fig. 3. By fitting the plots of $\ln \rho - 1/(kT)$, the Fermi level in SI-CZT:In is determined to be (0.740 ± 0.047) eV.

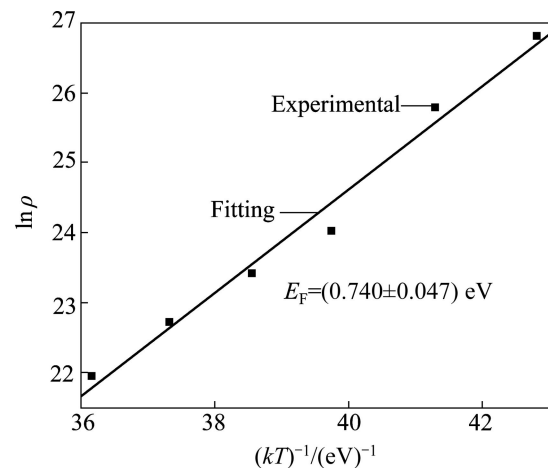


Fig. 3 Fermi level by fitting plots of $\ln \rho$ versus $(kT)^{-1}$

4 Conclusions

1) There is a larger error in evaluating the values of deep traps with the initial rise method, which is caused by the interference of overlapping peaks.

2) The SIMPA method demonstrates a better performance in isolating the composition of overlapping peaks simultaneously to the individual trap peaks and thus to obtain more reliable values of trap levels.

3) Based on the SIMPA fit, ten trap levels and a deep donor level E_{DD} dominating the dark current in SI-CZT:In and their origins are identified. At the same time, the high resistivity of CZT:In is explained to be responsible for the Fermi level pinning near the mid-gap.

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热激电流谱确定 CZT:In 中的陷阱能级

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摘 要: 熔体法生长的半绝缘碲锌镉($\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ 或者 CZT)晶体中存在着很多缺陷, 这些缺陷作为陷阱中心在带隙中引入了深能级, 从而严重影响 CZT 的探测性能。分别采用初始上升法和同步多峰分析法(SIMPA)分析热激电流谱(TSC), 从而获得了半绝缘的镉掺杂的 $\text{Cd}_{0.9}\text{Zn}_{0.1}\text{Te}$ 晶体中的陷阱能级分布。结果表明: 由于重叠峰的干扰, 初始上升法在确定陷阱峰的最大值时会产生较大的误差; 而 SIMPA 法被证实适用于分离重叠峰, 可同步获得较全面的陷阱能级分布。基于此, 获得了半绝缘 CZT:In 晶体的缺陷能级分布结果, 即包含十个陷阱能级和一个影响暗电流分布的深施主能级 E_{DD} 。此外, 通过 E_{DD} 能级与费米能级的关系, 解释了 CZT:In 晶体获得高阻特性的原因。

关键词: $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$; 陷阱; 深能级; 热激电流谱

(Edited by LI Yan-hong)