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Preparation and Investigation of p-GaAs/n-Cd_{1-x}Zn_xS_{1-y}Te_y Heterojunctions Deposited by Electrochemical Deposition

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Anisotype heterojunctions of p-GaAs/n-Cd_{1-x}Zn_xS_{1-y}Te_y have been fabricated by preparing n-type Cd_{1-x}Zn_xS_{1-y}Te_y thin films onto p-GaAs single crystal wafers using an electrochemical deposition method. The voltammetric behavior of the Cd_{1-x}Zn_xS_{1-y}Te_y thin films on GaAs substrates from aqueous solutions was studied. Electrical and photoelectrical properties of heterojunctions were studied depending on the Cd_{1-x}Zn_xS_{1-y}Te_y films composition ($x = 0.1 \div 0.8$; $y = 0.2$; 0.4 ; 0.9) and heat treatment (HT) regime in argon atmosphere (100–450 °C during 3–16 min). Under AML5 conditions, the open-circuit voltage, short-circuit current, fill factor, and efficiency of our best cell, was $V_{oc} = 584$ mV, $J_{sc} = 14.54$ mA/cm², $FF = 0.6$, and $\eta = 6.7\%$, respectively. [DOI: 10.1115/1.4027694]

Keywords: electrochemical deposition, thin film, heterojunction, heat treatment, solar cell

Introduction

Thin films of II-VI compounds (CdS, CdTe, Cd_{1-x}Zn_xS, Cd_{1-x}Zn_xS_{1-y}Se_y, and Cd_{1-x}Zn_xS_{1-y}Te_y (CZSTE), etc.) have attracted considerable attention from the research community due to their wide uses in the fabrication of semiconductor device technology and solar cells [1–5]. In photovoltaic systems, the replacement of CdS with the higher energy band gap of Cd_{1-x}Zn_xS, Cd_{1-x}Zn_xS_{1-y}Se_y, and CZSTE alloys has led to a decrease in window absorption losses and has resulted in an increase in the short-circuit current. The II-VI quaternary semiconductors seem to be useful materials with photosensitivity in the visible and ultraviolet wavelength regions [6–10]. Since single crystals of GaAs are well-studied materials, their use at manufacturing of heterojunctions p-GaAs/CZSTE will be a good way to deeply study the physical properties of CZSTE films.

There are many techniques used to synthesize thin films of II-VI compounds, such as thermal evaporation, chemical bath deposition, successive ionic layer absorption and reaction, magnetron sputtering, metalorganic vapor phase epitaxy, etc. [11–17]. In photovoltaic applications, where semiconductor films over large areas are required, the electrodeposition technique is specially adequate. In addition, for application in solar cells, electrodeposition allows one to easily alter both the bandgap and lattice constant by composition modulation through the control of growth parameters such as applied potential, pH, and temperature of the bath [11,18–20]. Thus, it is at least in principle possible to easily grow large areas of tandem cells designed for the most efficient conversion of the solar spectrum.

In the present work, anisotype heterojunctions of p-GaAs/n-CZSTE were fabricated by depositing CZSTE thin films as a window using the electrochemical deposition method onto the p-GaAs single crystals.

Experimental

Electrodeposition of the CZSTE films onto the p-GaAs substrates was carried out at a temperature of 80 °C from aqueous solution containing cadmium (CdSO₄), zinc (ZnSO₄), sodium (Na₂S₂O₃), and tellurium (TeO₂ or Na₂Te₂O₃) salts. The thickness and resistivity of the monocrystalline p-GaAs substrates were 0.4 mm and $\rho = 0.2$ – 0.23 Ω cm, respectively. Before the deposition process, the surfaces of the GaAs substrates were etched in an aqueous solution of hydrochloric acid and KOH-KNO₃ (1:3) composition for 3 min. After etching, the GaAs wafers were washed for 2 min in pure alcohol and distilled water maintained at high temperatures (≥ 300 °C).

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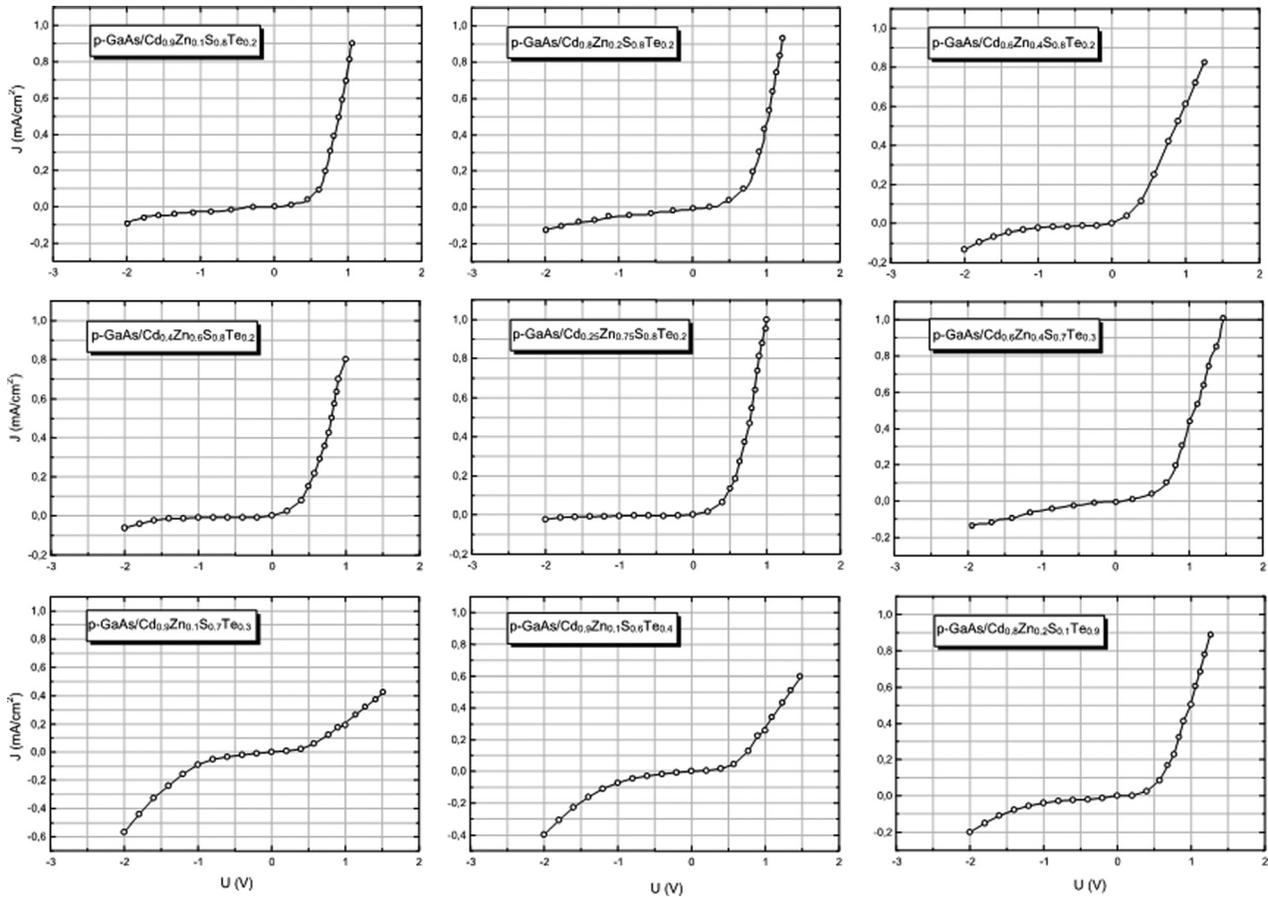


Fig. 1 Dark J - V curves for as-deposited p-GaAs/CZSTE heterojunction

112 Cyclic voltammetry was used to monitor the electrochemical
 113 reactions in separate solutions of CdSO_4 , ZnSO_4 , $\text{Na}_2\text{S}_2\text{O}_3$, and
 114 TeO_2 , then in their combined solution at the same concentration
 115 and $p\text{H}$. The cyclic voltammogram was scanned in the potential
 116 range 1.2 V to -1.2 V versus graphite (or Ag/AgCl) electrodes.
 117 Cyclic voltammogram for mixture of CdCl_2 , ZnCl_2 , $\text{Na}_2\text{S}_2\text{O}_3$, and
 118 $\text{Na}_2\text{Se}_2\text{O}_3$ salts shows that wave $-0.52 \div -0.9$ V corresponded to
 119 the formation of CZSTE layers. The thickness of the CZSTE films
 120 grown by electrodeposition from a solution could be varied in a
 121 wide range from 50 to 1600 nm.

122 In order to fabricate the heterojunctions, an ohmic in electrode,
 123 in reticulate form was evaporated on the CZSTE films with an
 124 area of $\sim 0.82\text{--}1$ cm^2 . An ohmic contact was performed on the
 125 side of GaAs wafers by evaporating an Al electrode.

126 Results and Discussion

127 The dark current-voltage (J - V) curves of the heterojunctions
 128 were measured in the direct and reverse current modes. The experi-
 129 mental J - V curves, measured at 300 K, for as-deposited p-GaAs/
 130 CZSTE heterojunctions, using various values of x and y , are illus-
 131 trated in Fig. 1.

132 These curves definitely proved diode type behavior, with the
 133 forward direction corresponding to the positive potential on
 134 p-GaAs. Thus, according to this figure, the as-deposited junctions
 135 composed of CZSTE films with $x=0.75$ and $y=0.2$ (which is a
 136 good lattice match with GaAs layers) reaches a rectification value
 137 of $k=700$ at voltage $U=1.0$ V (k is the rectification factor), and
 138 decreases when zinc concentration, x , increases. The low rectifica-
 139 tion coefficient is due to the high series resistance within the hetero-
 140 structure. Plotting the natural log of the current density versus
 141 the applied voltage, we are able to identify a characteristic thermally
 142 activated recombination region up to 0.63 V. Usually, such
 143 dependencies are described by the expression

$$J = J_s \left[\exp\left(\frac{eV}{AkT}\right) - 1 \right] \quad (1)$$

Here, J_s is the saturation current density, V is the applied voltage, 144
 e is the electron charge, A is the ideality factor, k is the Boltzmann 145
 constant, and T is the temperature. 146

Increasing the forward bias magnitude ($U > 0.65$ V) resulted in 147
 a less steep dependence of $J(V)$ and its pronounced deviation from 148
 the curve calculated according to the formula (1), which can be a 149
 consequence of the changes of carrier transport mechanism. The 150
 most possible case to be considered is tunneling recombination. In 151
 the as-deposited heterojunctions, the ideality factor was determined 152
 under a forward bias, and it was normally found to range from 1.6 153
 to 2.7 for the different x and y . This established that the value of 154
 ideality factor was minimal for the p-GaAs/n- $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$ 155
 heterojunctions. 156

The mechanism of current passage through the heterojunctions 157
 essentially changes with increasing HT temperature from 0 to 158
 390°C (for 14 min). Notably, tunnel currents sharply decreased 159
 with increasing HT temperature, which testifies to reduction of 160
 defects and decreasing series resistance (Table 1). After the HT in 161
 argon atmosphere at 390°C for 14 min, the ideality factor values 162
 were approximately 1.4 for the heterojunctions with $x=0.75$ and 163
 $y=0.2$. It is significant to note that the best rectification for the 164
 annealed p-GaAs/n- $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$ heterojunctions was 165
 obtained at about $k=3000$, which is attributable to the optimal 166
 HT conditions and lattice mismatch between the solid solution of 167
 $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$ and GaAs. 168

The capacitance versus voltage measurement results ($1/C^2$ - V) 169
 for the heterojunctions p-GaAs/n- $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$ annealed 170
 in argon atmosphere at 390°C for 14 min showed a linear relation- 171
 ship with bias voltage and indicates that the junction is abrupt. 172

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Table 1

HT temperature and duration	Rectification coefficient (k)	Nonideality factor (A)	Series resistance ($R_s, \Omega \text{ cm}^2$)
before HT	200	1.61	260
150 °C; 14 min	540	1.54	200
200 °C; 14 min	970	1.51	176
250 °C; 14 min	1700	1.46	93
300 °C; 14 min	2450	1.44	54
350 °C; 14 min	2600	1.42	30
390 °C; 14 min	3000	1.4	24
430 °C; 14 min	6	2.56	1300

173 Also built-in potential ($V_{bi} = 0.61 \text{ V}$) were calculated by extrapolating ($1/C^2 - V$) plot to ($1/C^2 = 0$).

175 As-deposited (nonheat-treated) p -GaAs/CZSTE heterojunctions were found to possess a photovoltaic effect. As follows from Fig. 2, the efficiency of the heterojunctions depends on the film's composition x and y . Under AM1.5 conditions, the maximal values of open-circuit voltage, short-circuit current, fill factor, and efficiency for cells p -GaAs/ n - $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$, were $V_{oc} = 131 \text{ mV}$, $J_{sc} = 3.4 \text{ mA/cm}^2$, $\text{FF} = 0.43$, and $\eta = 0.2\%$, respectively.

183 To assess the effect of HT on the photoelectric properties of the heterojunctions, the films were annealed in argon atmosphere at 100–450 °C for 3–16 min. Figure 3 shows typical spectral dependences of the photocurrent for p -GaAs/ n - $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$ heterojunctions before and after HT. There occurs a reconstruction of the photosensitivity spectrum after HT, i.e., the spectrum broadens. As the HT temperature increased from 0 to 390 °C for 14 min, photosensitivity in the $\lambda_m = 0.38\text{--}0.8 \mu\text{m}$ wavelength region sharply increased. The near infrared photosensitivity falloff for all heterojunctions indicated GaAs absorber band gaps of 1.42 eV. Figure 3 also shows that after subsequently HT in argon atmosphere for 14 min at $\geq 400 \text{ °C}$ the performance of these cells deteriorated.

196 The observed effect of HT on the heterojunction properties can be understood in terms of electronic–molecular interaction between the surface of CZSTE films and oxygen [3–5]. It is believed that oxygen adsorption, after the removal of CZSTE films from the solution, leads to the formation of deep acceptor states in the surface layer of the films. The oxygen-related acceptors

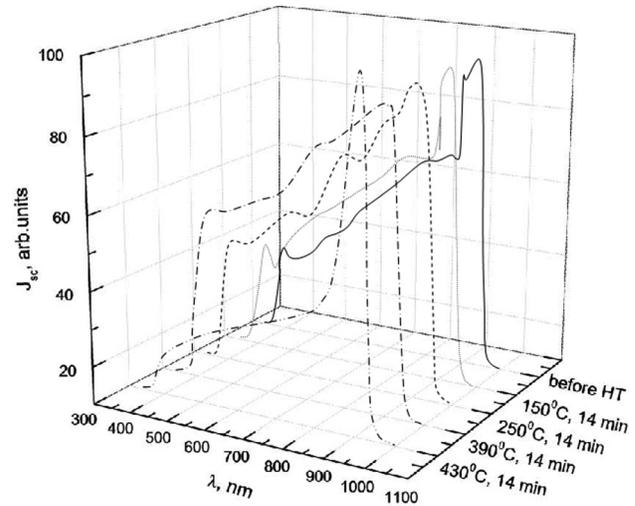


Fig. 3 Spectral dependences of the photocurrent for p -GaAs/ n - $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$ heterojunctions before and after HT

capture electrons from the film bulk, creating a near-surface potential barrier, which is responsible for the low short-wavelength photosensitivity of the nonheat-treated heterojunctions. The small height of the intergranular barriers in polycrystalline films, as compared to the oxygen-related barriers, renders the short-wavelength photoresponse of the p -GaAs/CZSTE heterojunctions to be governed by the density of oxygen-related states. The observed effect of HT on the photoelectric properties of the heterojunctions demonstrates that the donor and acceptor concentrations in the films depend on HT conditions. In particular, it seems likely that, in the initial stages of HT, some of the oxygen desorbs, which enhances the short-wavelength photosensitivity of the heterojunctions. In addition, HT at 390 °C for 14 min results in preferential vaporization of Cd and Zn. The Cd and Zn vacancies forming in the surface layer of the CZSTE films act as r centers. The decrease in the density of surface defects and film recrystallization during subsequent HT shifts the photosensitivity maximum to shorter wavelengths and improves the performance parameters of the films. The sharp decrease in photosensitivity upon heat treatment at 400 °C or higher temperatures indicates that some of

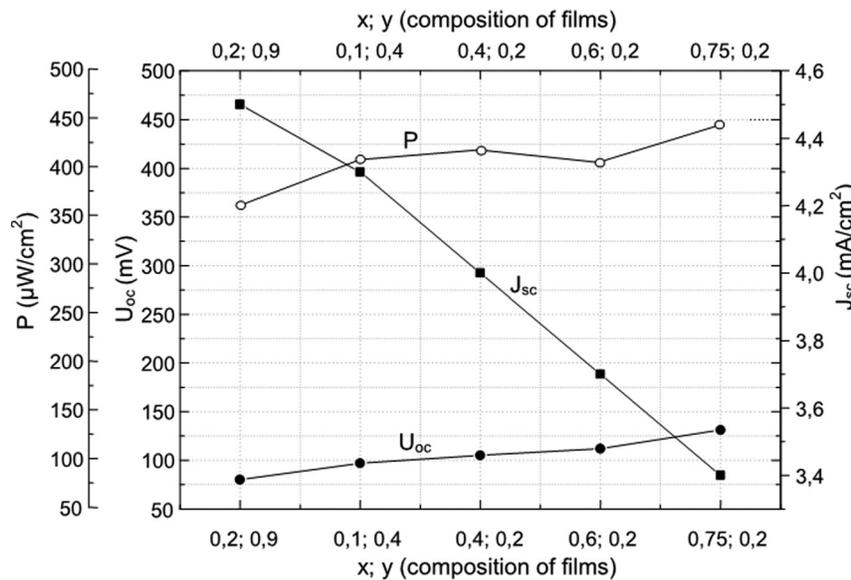


Fig. 2 Dependence of short-circuit current (J_{sc}), open-circuit voltage (U_{oc}), and power output (P) of the as-deposited p -GaAs/ n - $\text{Cd}_{1-x}\text{Zn}_x\text{S}_y\text{Te}_{1-y}$ cells on the films composition

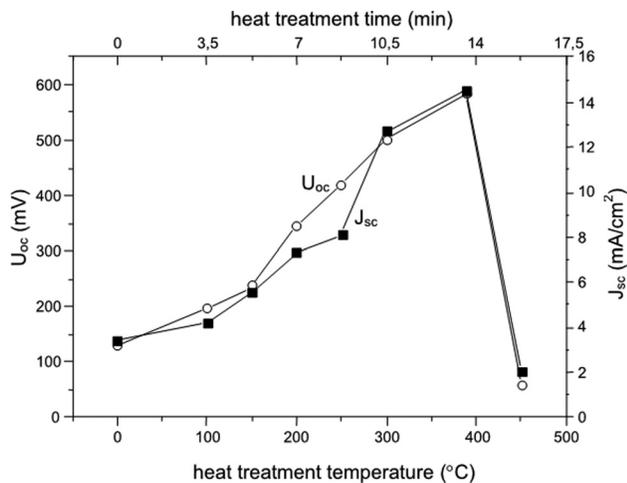


Fig. 4 Dependence of p-GaAs/n-Cd_{0.25}Zn_{0.75}S_{0.8}Te_{0.2} solar cell parameters on the heat treatment time and temperature

the oxygen does not desorb and remains in the surface layer in atomic form. As a result, the r centers begin to play a crucial role in determining the recombination process, and the concentration of holes captured by the r centers increases sharply, reducing the photoresponse of the devices.

Note that, under the conditions of this study, the short-circuit current through the heterojunctions varies nonmonotonically not only with temperature but also with HT time and reaches a maximum after heat treatment at 390 °C for 14 min (Fig. 4). Under AM1.5 conditions the maximal values of open-circuit voltage, short-circuit current, fill factor and efficiency of our best cell, were $V_{oc} = 584$ mV, $J_{sc} = 14.54$ mA/cm², FF = 0.6, and $\eta = 6.7\%$, respectively.

During storage for more than 36 months at room temperature, the parameters of HT p-GaAs/CZSTE heterojunctions experienced no degradation.

Conclusions

p-GaAs/CZSTE heterojunctions prepared by the method of electrochemical deposition are suitable to fabricate high efficiency solar cells. Their electrical and photoelectrical characteristics were studied depending on the composition of CZSTE films and the HT condition. It is established that HT at 390 °C for 14 min in argon atmosphere reduces the concentration of defects, results in formation of heterojunctions and minimum values of nonideality factor ($A = 1.4$) of J - V characteristics and serious resistance ($R_a = 24 \Omega \text{ cm}^2$). The forward current of this junction obeys tunneling-recombination model and (C - V) measurements revealed that heterojunctions are abrupt.

Heterojunctions with $x = 0.75$ and $y = 0.2$ possess a high photosensitivity after the HT in argon at 390 °C for 14 min. Under standard 100 mW/cm² white-light illumination at room temperature, the values of the parameters of our best cell were

$V_{oc} = 584$ mV, $J_{sc} = 14.54$ mA/cm², FF = 0.6, and $\eta = 6.7\%$, respectively.

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