RADIATIVE RECOMBINATION MECHANISMS in CdTe THIN FILMS DEPOSITED BY ELEMENTAL VAPOR TRANSPORT

**Shamara Collins¹, Sergiu Vatavu^{1,2}, Vamsi Evani¹, Md Khan¹, Sara Bakhshi¹,
Vasilios Palekis¹, Corneliu Rotaru², and C. Ferekides¹**

*¹Department of Electrical Engineering, University of South Florida, 4202 East Fowler
Ave, Tampa, FL 33620, USA;*

*²Faculty of Physics and Engineering, Moldova State University, 60 A. Mateevici str.,
MD-2009 Chisinau, Republic of Moldova*

Abstract

A photoluminescence (PL) study of the radiative recombination mechanisms for CdTe films deposited under different Cd and Te overpressure by elemental vapor transport is presented. The experiment and analysis have been carried out in the temperature range of 12-130K. The intensity of the PL laser excitation beam was varied by two orders of magnitude. It has been established that the bands in the 1.47-1.50 eV are determined by transitions involving shallow D and A states and the 1.36x-1.37x eV band is due to band to level transitions. Deep transitions at 1.042 eV and 1.129 eV are due to radiative transitions to levels determined by CdTe native defects.

Keywords

CdTe, Photoluminescence, elemental vapor transport, defects

1. Introduction

Cadmium telluride is a material constantly acquiring attention from the world PV community. After about ten years of stagnation in efficiency increase for CdTe based solar cells, recent developments have lead small area cell efficiencies above 20% [1,2,3]. Understanding the material properties responsible for achieving such high performances and being able to advance them even further is a critical factor for CdTe PV. In this context, it is necessary to evaluate the properties of thin films under different deposition conditions. An important aspect of all II-VI semiconductors like CdTe is the role of intrinsic defects. Understanding and controlling their formation s a key issue in improving the performance of CdTe based thin film solar cells. Most deposition processes typically utilized for thin films of CdTe do not offer any means of controlling or varying film stoichiometry. The Elemental Vapor Transport process can be used for the deposition of CdTe films under excess Cd or Te partial pressures therefore offering an opportunity to influence the film stoichiometry and therefore the formation of native defects. This research study aims at elucidating the influence of this deposition technology on the energetic levels in the bandgap due to variations in the film stoichiometry. Low-temperature photoluminescence (PL) is one of the methods used to reveal the radiative levels' energetic diagram relative to different deposition conditions (i.e. different Cd/Te pressure ratios will force the creation of a different distribution of the radiative levels in the band gap). It has been proven that PL is effective in understanding the transitions in the bandgap [4, 5] and their possible correlation to photovoltaic efficiency. These results will allow further correlation of gap states in CdTe films to native defects.

2. Experimental details

The Elemental Vapor Transport (EVT) deposition method has been used for the deposition of CdTe thin films onto alumina substrates. Additional information on the EVT process and films can be found elsewhere [6, 7].

As-deposited CdTe thin films deposited under a wide range of Cd/Te vapor ratios (0.35 to 2.0) have been investigated by PL over the 0.8 – 1.6 eV energy range.

The measurements of the photoluminescence spectra have been carried out by use of an in-house assembled system based on SPEX 500M monochromator with a 600 mm^{-1} grating. The energetic resolution was 1 meV or better. The PL signal was detected by using a synchronous detection system using a lock-in amplifier (EG&G model 5209), chopper (EG&G Park model 197), a Standard Research Systems model SR560 Low noise Preamplifier, and an InGaAs photodiode. The PL has been excited by 514.5 nm line of a Coherent Innova 70 laser (10 – 15 mW output power). The incident laser beam power was varied by two orders of magnitude using neutral density filters. The radiative transitions have been investigated in the temperature range of 12K to 130K using a closed loop He cryosystem, consisting of: model 22 CTI-Cryogenics (Helix Technology Corporation) refrigerator (cryo head) with a custom built optical head (fused silica windows) and CTI-Cryogenics model SC compressor. A Lakeshore 805 controller with a silicon diode for temperature sensing was used to control the sample temperature.

3. Results & discussion

The analysis of the photoluminescence spectra is presented for three spectral regions: (a) excitonic (1.570-1.590 eV), (b) 1.330-1.560 eV region and (c) the 0.820-1.250 eV spectral region (deep transitions). Due to the complexity of the PL bands under analysis, the spectra have been deconvoluted, using the natural line-shape determined by a Lorentzian function [8]. The dependence of each elemental component

vs. temperature and incident excitation power has been investigated. The deconvolution results show several main bands present in the initial spectra, namely: 1.36x-1.37x eV (the third digit after comma depends on Cd/Te vapor pressure ratio), 1.497 eV, 1.486 eV, 1.47x (x=5-7) eV, 1.547 eV, 1.598 eV (Fig. 1). The contribution to the radiative transitions is also given by 1.44x eV, 1.505 eV, 1.515 eV, 1.52x (1.53x) eV bands – their weight decreases steeply as the temperature increases or excitation intensity decreases.

3.1 Excitonic photoluminescence in the CdTe EVT films Analysis (using deconvoluted data) of the spectral region (1.575-1.598 eV, 15K), characteristic for CdTe excitons radiative annihilation, reveal that the band with a width of approx. 25 meV is present in all samples and consists of 4 subbands of Lorentzian shape (the precision is determined by the mathematical deconvolution, the physical meaning will have only 3 digits after comma). The deconvoluted data (15K) is given in Table 1. Considering the exciton Rydberg in CdTe equal to 9.1 meV and the band gap 1.606 eV (0K) [9], the excitonic band mentioned above, consists of a free exciton (FE) line at 1.598 eV, and the Lorentzian with peak energy at 1.592 eV is determined to be a bound exciton (to a Donor) [10] with a 6 meV binding energy. The elementary band at 1.585 eV is determined by annihilation of electrons bounded to an Acceptor (13 meV binding energy) [11]. The PL component at 1.577 eV might be related to the 1st phonon replica of the FE, but its intensity (Poisson distribution does not fit) and width are inconsistent with that assignment.

The estimation of donor and acceptor position (and ionization state) has been done according to ref. 9, chapter 8. The results show that neutral donors (D) and acceptors (A) might be positioned between 18-109 meV and 39-236 meV respectively; the ionized D and A might be positioned at 30 meV and 65 meV respectively.

The intensity of the exciton bands are comparable to the intensity of the PL defect bands detected in these films. All the bands exhibit a linear dependence on the PL excitation power, which is given by [12]:

$$I_{PL} = AP^n \quad (1)$$

where P is laser beam excitation power, A – is a constant. The values of n are given in Table 2 as a function of the Cd/Te vapor ratio. It should be noted that the value of the power exponent lies in the normal range for CdTe [13] only for the P_{Cd}/P_{Te} ratio of 1.00. The high values of n observed for the other Cd/Te ratios might be the result of an increase in the number of recombination paths. One of the models that explain a square dependence on the excitation power ($I_{PL} \sim P^2$) considers that the concentration of neutral donors (N_D^0) and acceptors (N_A^0) are constant [14] and do not depend on excitation power; the intensity of FE recombination is given by formula (2), [14]:

$$I_{FE} = \frac{B}{\tau_{FE} [eN_{A^0} + g(N_D - N_{D^0})]^2} P^2 \quad (2)$$

and in the case of bound exciton annihilation:

$$I_{D^0X} = \frac{CN_{D^0}}{(1 + \frac{\tau_{DX}}{\tau_{DX}^{nr}}) [eN_{A^0} + g(N_D - N_{D^0})]^2} P^2 \quad (3)$$

where τ_{FE} , τ_{DX} , τ_{DX}^{nr} is the lifetime of FE, DX and non-radiative DX lifetimes; g is the transition rate of the generation process; B, C are constant coefficients; e is the electron charge. A similar formula describes the AX recombination. The case where $n > 2$, the PL intensity in this region also increases due to an increase of N_D in comparison to N_D^0 and increase of n (concentration of free electrons) as the Cd/Te overpressure ratio increases. A similar analysis explains the behavior of PL intensity in the band edge region when the Cd/Te vapor pressure ratio decreases.

The free exciton line intensity tends to increase as the P_{Cd}/P_{Te} ratio decreases from 1.00 to 0.35, surprisingly reaching its maximum value for the 0.45 ratio and not 1.00 as one might expect. The PL plots for P_{Cd}/P_{Te} ratio=2.00 shows the lack of FE transitions in films due to probably high concentration of Cd atoms screening the exciton formation. A similar trend is observed for the DX and AX bands. The P_{Cd}/P_{Te} ratios of 0.45 and 1.25 indicate a higher radiative recombination rate (Fig. 2).

3.2 PL in the 1.330-1.560 eV region The spectra obtained in this range vary extensively as the Cd/Te ratio varies as shown in Fig. 3 (spectra measured at 15K). The deconvolution analysis shows that the plots consist of at least 8 to 10 elementary bands, having different nature (Table 2). The dependence of the peaks' intensity at different temperatures shows an activation dependence [4]. The temperature quenching of the bands given in Fig. 3 reveal one or two slopes, and the activation energies for selected are given in Table 3. The typical PL quenching for the selected bands is presented for the ratio 0.60 Fig. 4a and the dependence on the laser excitation – in Fig. 4b for the same P_{Cd}/P_{Te} ratio.

3.2.1 1.497 eV, 1.486 eV, 1.474 eV bands This set of transitions were detected in nearly all CdTe films. They exhibit a linear dependence with photoluminescence excitation, which suggests that these are due to transitions to levels (presumably acceptors, because the peaks mentioned above disappear in Cd-rich samples). On the other hand the value of the power exponent varies from the 1.33-1.41 (usually seen in CdTe samples) to 1.91-3.15 – a quite high value for this material. One cannot exclude the formation of impurity bands, or at least the presence of several acceptor levels quite close (kT or more) one to each other. As result from the above analysis (1.00 ratio), 1.474 eV band is determined by D (67 meV from CB, transitions to this states are non-radiative probably due to a high number of D states) to ionized acceptor transition (0.065 eV). The origin of this band might find another interpretation, which is not

strictly correlated to the value of the activation energies (40-60 meV). Considering its power dependence on excitation intensity and the value of the bandgap at 0K – 1.606 eV [15] e-A transitions can determine the formation of this band. This radiative transition is persistent in spectra recorded for all P_{Cd}/P_{Te} ratios and has the maximum value for the 0.45 ratio. It is assumed the acceptor is the V_{Cd}^0 [16].

The 1.486 eV band is determined by $E_c - 0.021$ meV (CB to D transitions by means of a large number of shallow defect levels) to ionized acceptor (the same as for the 1.474 band); to be underlined that the donor position determined above is close energetically to the value of neutral donor to which excitons are bounded ($E_c - 0.018$ meV). The 1.486 eV band – has similar features to the 1.474 eV band and might be determined by e-A transitions. The nature of the acceptor is not clear.

The 1.497 band (same ratio 1.00) might result from direct CB to Te_i [17] transitions (Te_i – probably ionized). The 1.497 eV band disappears as the P_{Cd}/P_{Te} ratio goes over 1.00 and 1.474 band is restructured, and 1.470 band appears for P_{Cd}/P_{Te} ratio of 2.00. It should be noted that the 1.486 eV and 1.497 eV bands are completely extinguished in samples deposited at Cd/Te ratios >1 , suggesting that they are determined by transitions to A.

3.2.2 The origin of the **1.40x, 1.42x 1.44x eV**, bands (15K) remains unclear for the moment due to the difficulties with the deconvolution process at different temperatures, excitations and ratios (ambiguous treatment on the number of bands is possible). It is noted that the 1.44x band coincides energetically with the well known Cd-vacancy related defect band and the 1.42x – with its 1st phonon replica.

3.2.3 1.36x-1.37x eV band 1.36x-1.37x eV (depending on ratio of Cd and Te vapour pressure), (also known as Z-band in CdTe) has its peak position varying as one can see in Table 3 depending on the Cd/Te pressure ratio and is completely extinguished for Cd/Te pressure ratio of 0.35; it has the highest intensity for a ratio of 1.25 (see Fig.

5). With an activation energy decreasing from 52 meV to 22 meV (Table 4) as the Cd/Te vapor pressure ratio decreases, and a power law dependence on the excitation (the exponent exceeding 1.88), the nature of this band according to [18] is determined by band to acceptor transitions, the last one being present on the crystallite boundary and caused probably by dislocations. The activation energies for this band is consistent to the one obtained in [18]. One cannot exclude that the Cd_i^{2+} to VB transitions occur (Cd_i^{2+} is positioned at $E_c - 0.23$ eV [17]).

3.2.4 Based on the behavior of the 1.505 eV, 1.515 eV, 1.52x (1.53x) eV elementary bands as a function of temperature and excitation, one can conclude that all of these are of plasmonic origin.

3.2.5 1.547 eV band The 1.547 eV band does not shift or disappear as the Cd/Te vapor pressure ratio varies to its extremities. The intensity of radiative transitions for this band reaches its maximum value for the 0.45 Cd/Te vapor pressure ratio and the photoluminescent transitions involving defect levels at 23 meV occur. This band is most likely due to a D (it might be the same donor as the one which assists excitons binding, one can expect a large number of shallow states in-between CB and this level because one notices only a power function on the excitation intensity or, it might be a Donor impurity band) and acceptor ($E_v + 0.036$ eV). A definitive conclusion on its origin cannot be reached at this time, but the possibility of existence of a level caused by dislocation and assisting this transition cannot be excluded.

3.3 PL in 0.8-1.25 eV region As one can see in Fig. 6. deep radiative transitions in CdTe are seen in the 0.82-1.20 eV region. The preliminary analysis of this band reveals that the wide band and its intensity strongly depend on Cd/Te overpressure ratio and at least two elemental component exists at 1.042 eV and 1.126 eV. These results are consistent with the ones presented in [19]. It is expected that the bands are determined by CB to Te_i^{2-} transitions and Cd_i^{2+} to Te_i^{2-} or V_{Te}^+ to Te_i^- .

4. Conclusions

A large number of bands have been detected from analysis of the as-deposited CdTe on Alumina thin films measured at different temperatures and excitation rates. It has been determined that high intensity excitonic peaks are among the main radiative paths in the material and the trend persists for all ratios. An unusually high value of the power exponent (more than 3) has been determined for the bands in the 1.47-1.5 eV region. It is believed that the radiative transitions in the 1.36x-1.37x eV are assisted by levels caused by dislocations. The deep transitions in CdTe are determined by native defects.

An important observation – shift on the Cd/Te overpressure ratio to its limits enhances the radiative annihilation of excitons in CdTe. The change in deposition conditions of the films also influences the other (1.497 eV, 1.486 eV, 1.474 eV) bands as well, and for the 0.45 and 1.25 P_{Cd}/P_{Te} ratios, the intensity of the PL reaches its max value. It can be concluded that for these ratios the concentration of created intrinsic defects reaches its highest value. The PL intensity decreases for a Cd/Te pressure ratio of 0.35, and points at the fact the Te related defect concentration has reached its maximum value, and intensity decrease is related to other processes. A similar explanation for the PL of the samples obtained at large Cd/Te pressure ratios.

The 1.577 eV band which has not been discussed in the main discussion is present in all samples, its temperature quenching has a typical activation character, the activation energies varying between 15 meV (2.00 ratio) and 31 meV (0.35 ratio). With a power dependence (formula 1) on PL excitation, band to level transition determines this PL spectral component although one might also consider that it's a LO phonon replica (21.3 meV) of the FE line.

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Figure captions

Figure 1 The photoluminescence spectra at 15K for CdTe as-deposited samples having different Cd/Te overpressure ratios (for comparison, the plots are given in linear and semilog scales, excitation – 10 mW).

Figure 2 The edge radiative recombination in CdTe: intensity of FE, DX, and AX bands vs Cd/Te vapor pressure ratio at 15K.

Figure 3 The PL spectra at 15K for the 1.3-1.53 eV region deconvolution vs P_{Cd}/P_{Te} ratio of as-deposited samples (excitation – 10 mW).

Figure 4 a) The temperature dependence of the selected elementary bands (after deconvolution) for CdTe/Alumina as-deposited samples, P_{Cd}/P_{Te} ratio 0.60 (excitation – 10 mW).

b) The PL intensity for selected bands vs laser excitation power at 15K, CdTe/Alumina as-deposited samples, P_{Cd}/P_{Te} ratio 0.60.

Figure 5 The PL intensity for selected bands vs Cd/Te pressure ratio at 15K (top) and thermal quenching of the 1.36x band (bottom).

Figure 6 The PL spectra (15K) in the 0.8-1.25 eV spectral region for samples obtained in different (P_{Cd}/P_{Te}) conditions.

Table captions

Table 1

Exciton radiative annihilation: peaks and ratios (mathematical model) at 15K

Table 2

The power exponent for the dependence of the PL intensity vs PL excitation power, 15K.

Table 3

The deconvoluted peaks' energetic positions at different ratios present in the photoluminescence spectra at 15K for CdTe as-deposited samples having different P_{Cd}/P_{Te} ratios, 15K.

Table 4 The dependence of the intensity for selected bands vs temperature and their activation energies for different P_{Cd}/P_{Te} ratios.

Table 1
Exciton radiative annihilation: peaks and ratios (mathematical model) at 15K

Cd/Te vapor pressure ratio	Peak position (eV)	FWHM (eV)	Intensity (arb. units)
2.00	1.59195	0.01141	9.04452
	1.58515	0.01049	16.7177
	1.57612	0.01298	18.88657
	1.56675	0.01452	11.97628
1.25	1.59772	0.00679	411.99553
	1.59172	0.00752	439.79255
	1.58556	0.00929	465.6471
	1.57684	0.01172	439.20216
1.00	1.59863	0.00647	78.53111
	1.59209	0.00786	78.22671
	1.58505	0.01071	97.71827
	1.57621	0.01750	83.98095
0.75	1.59902	0.00757	93.60356
	1.59211	0.00791	105.11827
	1.58520	0.00997	147.9624
	1.57702	0.01253	99.15457
0.60	1.59865	0.00706	250.7919
	1.59176	0.00792	247.35655
	1.58571	0.01016	319.26807
	1.57674	0.01478	242.76749
0.45	1.59836	0.00717	1192.78244
	1.59156	0.00834	1381.51929
	1.58531	0.00941	1513.39446
	1.57718	0.00777	900.06437
0.35	1.59789	0.00601	83.94339
	1.59198	0.00795	131.44915
	1.58526	0.00968	177.54792
	1.57778	0.01376	118.81508

Table 2

Table 2
The power exponent for the dependence of the PL intensity vs PL excitation power, 15K.

Cd/Te vapor pressure ratio	n _{1.598 eV}	n _{1.592 eV}	n _{1.585 eV}	n _{1.577 eV}	n _{1.547 eV}	n _{1.497 eV}	n _{1.486 eV}	n _{1.474 eV}	n _{1.470 eV}	n _{1.36x- 1.37x eV}
2.00	-	-	-	-	-	-	-	-	3.15	2.47
1.25	3.07	3.03	3.24	3.35	2.20	1.96	1.91	1.75	-	2.31
1.00	1.86	1.60	1.68	1.62	1.61	1.51	1.48	1.31	-	1.83
0.75	1.73	2.06	2.07	2.08	1.55	1.41	1.6	1.36	-	1.71
0.60	2.02	1.99	2.00	1.85	-	1.44	1.31	1.33	-	1.79
0.45	2.23	2.15	2.24	2.08	1.83	1.59	1.74	1.49	-	1.88
0.35	2.26	2.18	2.13	2.40	-	1.94	1.77	1.88	-	-

Table 3
The deconvoluted peaks' energetic positions at different ratios present in the photoluminescence spectra at 15K for CdTe as-deposited samples having different P_{Cd}/P_{Te} ratios, 15K.

P_{Cd}/P_{Te} ratio	2.00	1.25	1.00	0.75	0.60	0.45	0.35
I	1.566						
	1.550						
		1.545	1.547	1.546	1.547	1.548	1.546
					1.532	1.533	
			1.528				
		1.520		1.522			1.519
			1.518		1.518	1.519	
	1.499						
	II	1.505	1.506	1.506	1.506	1.506	1.506
	III	1.496	1.497	1.496	1.498	1.497	1.497
IV	1.486	1.486	1.487	1.487	1.487	1.487	1.487
V		1.474	1.474	1.474	1.474	1.475	1.476
	1.470						
						1.456	
							1.460
	1.450						
		1.448		1.446			
					1.441		
			1.439				
	1.431						
				1.423		1.425	
	1.407						
VI	1.370	1.367	1.368	1.367	1.372	1.377	

Note
The bands are grouped from I to VI, the precision is determined by math accuracy of deconvolution, so the bands differing by 1-2 meV in position are believed to have the same origin

Table 4

Table 4 The dependence of the intensity for selected bands vs temperature and their activation energies for different P_{Cd}/P_{Te} ratios.

P_{Cd}/P_{Te}	1.36x– 1.37x eV		1.470 eV	1.474 eV		1.486 eV		1.497 eV		1.547 eV	
	ΔE_1 (meV)	ΔE_2 (meV)	ΔE_2 (meV)	ΔE_1 (meV)	ΔE_2 (meV)	ΔE_1 (meV)	ΔE_2 (meV)	ΔE_1 (meV)	ΔE_2 (meV)	ΔE_1 (meV)	ΔE_2 (meV)
0.35	22	2	-	-	2	18	3	22	3	-	4
0.45	25	<1	-	42	-	39	<1	44	<1	23	<1
0.60	30	2	-	27	<1	34	1	43	1	-	-
0.75	39	1	-	27	<1	45	1	21	2	15	2
1.00	52	1	-	50	<1	64	<1	66	<1	49	2
1.25	43	-	-	-	5	44	8	-	4	48	6
2.00	-	5	7	-	-	-	-	-	-	-	-

Figures

Figure 1

Figure 2

Figure 3

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Figure 3

a

b

Figure 4

Figure 5
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Figure 5

Figure 6