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Structural and Optical Properties of Vacuum Evaporated $Znse_{0.5}te_{0.5}$/si Thin Films: Effect of Thickness

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Abstract: Semiconducting $ZnSe_{0.5}Te_{0.5}$ thin films were prepared with different thicknesses on Si <100> and glass substrates at a constant substrate temperature of 473K using vacuum evaporation technique. XRD study show that the vacuum evaporated $ZnSe_{0.5}Te_{0.5}$ thin films are polycrystalline cubic structure. Structural parameters such as lattice parameter and inter-planar spacing were obtained from Bragg's angle of predominant orientation <111>. Grain size, dislocation density and microstrain were calculated from FWHM of <111> diffraction line. Extinction coefficient was calculated from optical transmittance and optical absorption values using spectrophotometer technique. The direct optical band gap of the $ZnSe_{0.5}Te_{0.5}$ thin films deposited on glass substrates with different thicknesses (171-386nm) were lying in the range 2.457 – 2.184eV. The band tail energy was estimated from optical absorption curves. These structural and optical parameters are found to be very sensitive to the kind of substrate and to the thin film thickness.

Key words: ZnSeTe, vacuum evaporation, silicon substrates, lattice parameter, grain size, dislocation density, microstrain, band tail energy, energy band gap, extinction coefficient.

INTRODUCTION

Group II-VI semiconducting thin films have attracted considerable interest for investigators because of their optical, electrical and photoinduced properties. Zinc semiconducting chalcogenides thin films have been extensively studied recently due to their technologically important applications in optoelectronic devices, solar cells, IR detectors and lasers. Ternary ZnSeTe thin films; have been successfully used in light emitting devices (Shinozaki *et al.,* 1999 and Tamargo *et al.,* 2000 and Eason *et al.,* 1995), full-color display elements (Tamargo *et al.,* 2000), scintillating material in radiation detectors (Ryzhikov *et al.,* 2003), solar cells (Vakkalanka *et al.,* 2007) and semiconducting lasers (Heonsu, 1994).

Thin films of ZnSeTe were prepared using many deposition techniques such as vacuum evaporation (El-Nahass *et al.,* 1996 and Dutta *et al.,* 1994), electrodeposition (Riveros *et al.,* 2002), physical vapour transport (Su *et al.,* 2000), metal organic vapor deposition MOCVD (Kamata *et al.,* 1997 and Naumov *et al.,* 1993), molecular beam epitaxy MBE (Peiris *et al.,* 2001 and Yang *et al.,* 1998 and Brasil *et al.,* 1991), and pulsed laser deposition PLA (Chen *et al.,* 1991 and Aydinli *et al.,* 1991).

There are several reports available on the deposition of binary and ternary Zn(Se,Te) thin films onto different kind of substrates such as glass (El-Nahass *et al.,* 1996 and Dutta *et al.,* 1994), quartz (El-Nahass *et al.,* 1996), stainless steel, GaAs (Kamata *et al.,* 1997 and Chang *et al.,* 1999 and Naumov *et al.,* 1993) and InP (Kontos *et al.,* 2003 and Strassburg *et al.,* 2003) substrates. The structural (El-Nahass *et al.*, 1996 and Dutta *et al.,* 1994), optical (El-Nahass *et al.,* 1996 and Dutta *et al.,* 1994 and Su *et al.,* 2000 and Yang *et al.,* 1998), photoluminescence PL (Mochizuki *et al.,* 1996 and Su *et al.,* 2000 and Chang *et al.,* 1999) and cathodoluminescence (Chang *et al.,* 1999) properties of ternary ZnSeTe thin films have been studied. The efficiency of ZnSeTe thin film devices is strongly affected by the structural, optical and electrical characteristics of these films. Ternary ZnSeTe chalcogenides has a cubic crystal structures with <111> planes preferred orientation with a lattice constant ranging from 5.68Å to 6.165Å (El-Nahass *et al.,* 1996 and Chang *et al.,* 1999).

Zn (Se, Te) thin films have a wide transparency range from the visible to beyond 10μm (Amutha *et al.,* 2006). Ternary materials, such as $ZnSe_xTe_{1-x}$ are technologically useful because the band-gap at room temperature can be tuned between the end-member values 2.26eV for ZnTe and 2.7 eV for ZnSe as the chemical composition parameter, is varied (Amutha *et al.,* 2006 and El-Nahass *et al.,* 1996 and Kumaresan *et al.,* 2002).

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In this paper $ZnSe_{0.5}Te_{0.5}$ thin films have been deposited on glass and Si <100> substrates at a constant substrate temperature of 473K using vacuum evaporation technique. The structural and optical properties of the prepared thin films were obtained using XRD and optical absorption techniques. This study attempted to determine the variations in structural and optical parameters caused by substrate kind and thin film thickness.

MATERIALS AND METHODS

$ZnSe_{0.5}Te_{0.5}$ thin films were prepared using vacuum evaporated technique. Appropriate solid mixture containing pure Zn, Se and Te (99.99%) elements was prepared and fused in evacuated silica tubes. The charged tubes were gradually heated to 1373K in rotating furnace for 36h, after which the tubes were slowly cooled to room temperature at a rate of 30K/h. Then the solid solution ingot of $ZnSe_{0.5}Te_{0.5}$ was taken out from the tube and made into powder form. The $ZnSe_{0.5}Te_{0.5}$ thin films were deposited onto Si and Corning glass slides in evacuated chamber (better than 10^{-5} Torr achieved using oil diffusion pump) using a Tantalum boat charged with the source material. The deposition substrate temperature for all investigated thin films is 473K. The distance between the rotating substrate holder and the Tantalum boat is about 25cm. Thickness of prepared thin films was obtained by a micro gravimetrical method using the relation $t = m/(\rho A)$ where m and A are the mass and the surface area, respectively, and ρ is the density of the bulk material (Pejova *et al.,* 2002 and Nascu *et al.,* 2004).

The structural properties of $ZnSe_{0.5}Te_{0.5}$ thin films were studied by a Philips PW 1710 X-ray diffractometer using CuKα radiation of wavelength λ = 1.540562Å in the range of 2θ between 20˚ and 80˚. The optical properteis of the deposited thin films were measured in the wavelength visible spectrum region by an UV spectrophotometer Shimadzu-3101.

RESULTS AND DISCUSSIONS

The vacuum evaporated thin films of $ZnSe_{0.5}Te_{0.5}$ were deposited with different thicknesses on glass and Si <100> substrates at a constant substrate temperature of 473K. The X-ray diffraction study based on precise measurements upon the position broadening and shape of X-ray profiles on the polycrystalline thin film gives information about the microstructural parameters such as lattice parameter, inter-planar spacing, grain size, dislocation density and microstrain which are characterizes the microstructural variations in the film. It is well known that pure Zn (Se,Te) materials exist in two crystalline phases, i. e., a cubic form with sphalerite structure and a hexagonal form with wurtzite structure. The XRD patterns for ternary $ZnSe_{0.5}Te_{0.5}$ thin films deposited with different thicknesses on glass and Si <100> substrates are presented in figure (1). Their X-ray diffraction pattern indicated the polycrystalline nature with zinc blend structure. The X-ray diffractogram of $ZnSe_{0.5}Te_{0.5}$ thin films showed the <111>, <220> and <311> reflections, of which the intensity of the <111> orientation is predominant. This study reveals that the vacuum evaporated $ZnSe_{0.5}Te_{0.5}$ thin films using a solid mixture containing pure Zn, Se and Te elements with different thicknesses on glass and Si <100> substrates are polycrystalline cubic structure. The similar crystalline structures were generally observed for ZnSeTe thin films (El-Nahass *et al.,* 1996). The intensity of the <111> peak for all prepared thin films decreases as thin film thickness increases.

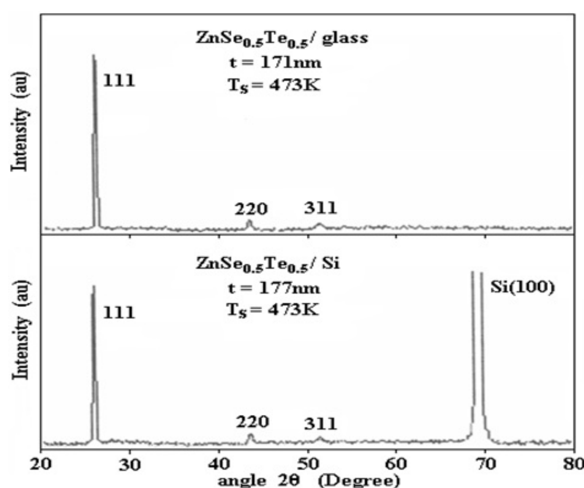

Fig. 1: – X-ray diffractogramms of vacuum evaporated $ZnSe_{0.5}Te_{0.5}$ thin films at substrate temperature 473K on glass and Si <100> substrates.

From XRD patterns the angular full width at half maximum FWHM of the cubic <111> peak decreases as the thin film thickness increases, see table (1). The relative small shift in XRD peaks of the films is clearly observed in table (1). The inter-planar spacing d between successive atomic planes in the crystal was calculated from the position of the <111> peak using the Bragg's relation (Cullity, 1978):

$$d = \frac{\lambda}{2\sin\theta}$$

Where, λ is the X-ray wavelength and θ is the scattering angle and it is called the Bragg angle. The calculated inter-planar spacing d for cubic $ZnSe_{0.5}Te_{0.5}$ thin films prepared with different thicknesses on glass and Si <100> substrates are presented in table (1). The peaks in an X-ray diffraction pattern are related to the cubic unit cell dimensions. The cubic structure is characterized by one lattice parameter a, the lattice parameter can be determined for <111> planes by using the plane-spacing equation (Cullity, 1978):

$$a_{hkl} = d\sqrt{h^2 + k^2 + l^2}$$

Where, hkl are the Miller indices. The calculated lattice parameter a_{111} for deposited thin films with different thicknesses on glass and Si <100> substrates are shown in figure (2).

Table 1: XRD patterns, inter-planar spacing and angular full width at half maximum for cubic $ZnSe_{0.5}Te_{0.5}$ thin films evaporated at substrate temperature 473K with different thicknesses on glass and Si <100> substrates.

Substrate	Thickness (nm)	hkl	2θ (Degree)	d (Å)	FWHM X10^{-3} (rad)
		111	26.148	3.405	
	171	220	43.387	2.084	7.23
		311	51.476	1.774	
		111	26.161	3.403	
	232	220	43.449	2.081	6.56
Glass		311	51.693	1.767	
		111	26.375	3.376	
	318	220	43.676	2.071	4.96
		311	51.076	1.787	
		111	26.243	3.393	
	386	220	43.707	2.069	3.71
		311	51.476	1.774	
		111	26.170	3.402	
	177	220	43.659	2.071	5.23
		311	51.354	1.778	
		111	26.270	3.390	
	228	220	43.558	2.076	5.05
Si(100)		311	51.542	1.772	
		111	26.238	3.394	
	316	220	43.488	2.079	4.44
		311	51.636	1.769	
		111	26.256	3.391	
	394	220	43.543	2.077	3.54
		311	51.731	1.766	

The lattice constant data presented in figure (2) for films evaporated with different thin film thickness shows a curvilinear behaviour. El-Nahass *et al.,* (1996) reported that the calculated unit cell lattice constant a_{111} for $ZnSe_xTe_{1-x}$ in powder form and molecular fraction x shows a linear dependence following Vegard's law. The lattice parameter for all prepared thin films was in the range 5.898 to 6.848Å for glass substrates and in the range 5.893-5.871Å for Si substrates. Also, these results are in a good agreement with the results published in previous papers (El-Nahass *et al.,* 1996 and Chang *et al.,* 1999).

The crystallite size D of the films was calculated from the Debye Scherer's formula from the full-width of half-maximum intensity β (FWHM) expressed in radians (Cullity, 1978 and Warren, 1969):

$$D = \frac{0.94\lambda}{\beta\cos\theta}$$

Figure (3) represents the grain size D of the $ZnSe_{0.5}Te_{0.5}$ thin films. In the investigated polycrystalline thin films, FWHM and the grain size are strongly influenced by the used substrate and thin film thickness. The grain size of the films deposited on Si substrates with different thin film thickness are found to be higher than the values for the thin films deposited on glass substrates, see figure (3). For the grain size, values ranging from 206-401Å for glass substrates and 284-420Å for Si substrates were found, when the deposition thickness of $ZnSe_{0.5}Te_{0.5}$ films varied between 171 and 394nm. The increase of grain size with the thickness was due to the improvement in the degree of crystallinity of the thin films.

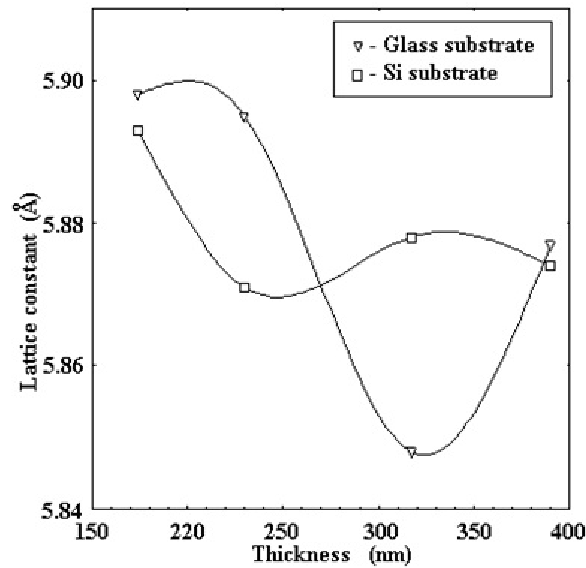

Fig. 2: Lattice constant for cubic $ZnSe_{0.5}Te_{0.5}$ thin films evaporated at substrate temperature 473K with different thicknesses on glass and Si <100> substrates.

The dislocation density δ defined as the length of dislocation lines per unit volume of the crystal and can be evaluated from the particle size D by the relation (Gopal *et al.,* 2005):

$$\delta = \frac{n}{D^2}$$

Where n is a factor, when equal unity giving minimum dislocation density. The origin of the microstrain is related to the lattice misfit, which in turn depends upon the deposition conditions. The microstrain ε is calculated using the relation (Gopal *et al.,* 2005):

$$\varepsilon = \left[\frac{\lambda}{D\cos\theta} - \beta\right]\frac{1}{\tan\theta}$$

Figures (4a) and (4b) represent the dislocation density δ and the microstrain ε of prepared $ZnSe_{0.5}Te_{0.5}$ thin films evaporated with different thicknesses on glass and Si <100> substrates. It was observed that the dislocation density and microstrain exhibit a decreasing trend with the deposition thin film thickness.

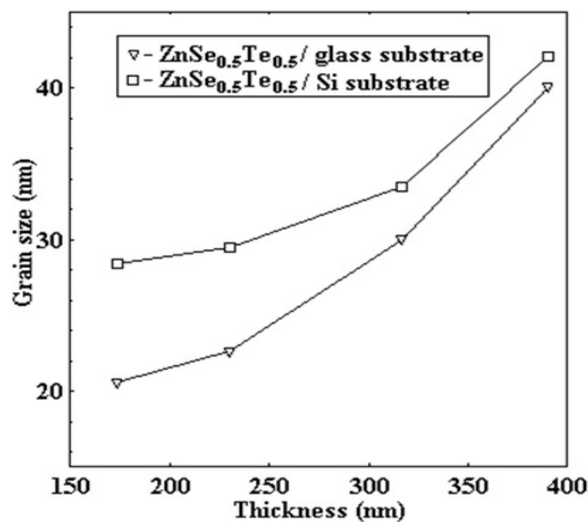

Fig. 3: Grain size for $ZnSe_{0.5}Te_{0.5}$ thin films evaporated at substrate temperature 473K with different thicknesses on glass and Si <100> substrates.

Dislocation density and microstrain of the films deposited on glass substrates with different thin film thicknesses are found to be higher than the values for thin films deposited on Si substrates, see figures (4a) and (4b).

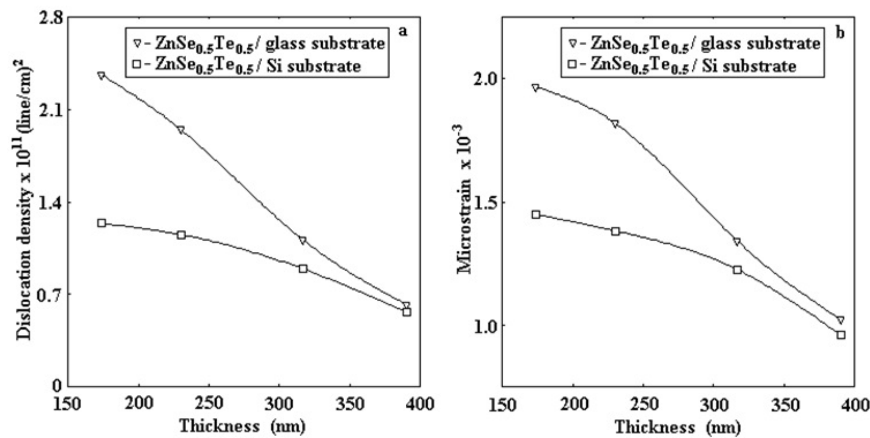

Fig. 4: Calculated (a) dislocation density and (b) microstrain for $ZnSe_{0.5}Te_{0.5}$ thin films evaporated at substrate temperature 473K with different thicknesses on glass and Si <100> substrates.

The absorption coefficient α, which is a function of the incident photon energy hυ, was determined from the transmission data in a high absorption spectrum region (when the reflectance is neglected) according to the expression (Tarey *et al.,* 1985):

$$\alpha = \frac{1}{t} \ln \left[\frac{1}{T} \right]$$

Where, T is the transmittance and t is the thickness of the thin films. Absorption coefficient α can be used to determine the band gap of the material. For the incident photon energy hυ greater than the optical band gap and above the exponential tail, the optical absorption follows a power law (Moss, 1973 and Pankove, 1971):

$$\alpha h\upsilon = A(h\upsilon - E_g)^n$$

Where A is a constant nearly independent on photon energy and known as the disorder parameter, E_g is the band gap of the material, the exponent n depends on the type of optical transition process. The exponent n may have value ½ corresponding to the direct allowed optical transition. The values of the band gap for the direct optical transition can be determined by extrapolating the straight line portion of the $(\alpha h\upsilon)^2$ vs. hυ graphs to the hυ – axis (Moss, 1973 and Pankove, 1971):

$$(\alpha h\upsilon)^2 = B(h\upsilon - E_g^{dir})$$

The $(\alpha h\upsilon)^2$ vs. hυ plots for $ZnSe_{0.5}Te_{0.5}$ thin films deposited with different thicknesses on glass substrates are shown in figure (5).

Photons with energy greater than band gap of the material will be absorbed and the photons of longer wavelength will be transmitted. The increase of absorption coefficient in the photon energy range 2-2.55eV can be related to the decrease of optical transmission spectrum. The shift in the fundamental absorption edge towards lower wavelength side is clearly observed in figure (5) with decreasing thin film thickness from 386nm to 171nm.

From table (2) it can be noticed that the direct band gap energy decreases as the thin film thickness increases. Direct band gap energy of $ZnSe_{0.5}Te_{0.5}$ thin films decreases from 2.457 to 2.184eV as the thin film thickness increases from 171 to 386nm. These values of direct energy gap are in good agreement with the values reported by El-Nahass *et al.,* (1996) using vacuum evaporation technique.

The optical absorption coefficient α depends exponentially on incident photon energy hυ. The absorption edge in many materials follows the Urbach rule (Pankove, 1971 and Abu El-Fadl *et al.,* 2004):

$$\alpha(h\upsilon) = \alpha_0 \exp \left[\frac{h\upsilon}{\Delta E} \right]$$

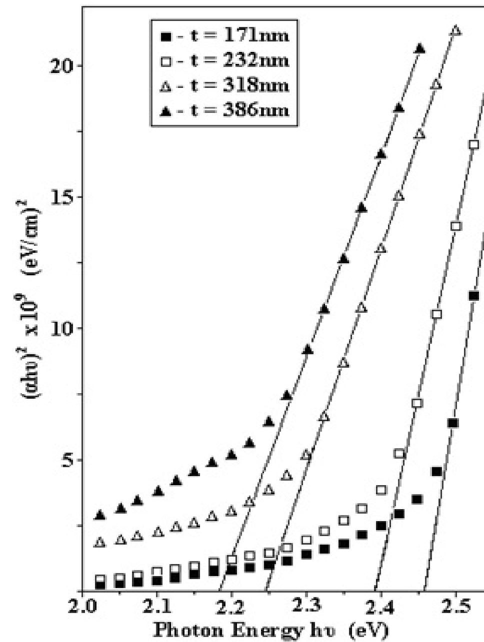

Fig. 5; The $(\alpha h\upsilon)^2$ vs. photon energy curves for $ZnSe_{0.5}Te_{0.5}$ thin films evaporated at substrate temperature 473K with different thicknesses on glass substrates.

Where α_0 is a constant of exponential dependence and ΔE is the energy width of the tail of localized state in the normally forbidden band gap. The width of localized states (band tail energy or Urbach energy) ΔE is estimated from the slopes of $\ln(\alpha)$ vs. $h\upsilon$ plots. The energy width of the tail ΔE could be obtained by extrapolating the linear portions of these curves. The estimated energy width of the tail ΔE for $ZnSe_{0.5}Te_{0.5}$ thin films prepared with different thicknesses on glass substrates are listed in table (2). From table (2) it can be noticed that the band tail energy ΔE decreases as the thin film thickness increases.

Table 2: Energy band gap E_g and energy width of the tail ΔE for $ZnSe_{0.5}Te_{0.5}$ thin films prepared with different thicknesses on glass substrates.

Thickness (nm)	E_g (eV)	α_0 (cm^{-1})	ΔE (eV)
171	2.457	5.9582	1.9175
232	2.393	7.0922	1.8557
381	2.246	12.8936	1.8402
386	2.184	11.0330	1.5155

The extinction coefficient k as a function of absorption coefficient $\alpha(h\upsilon)$ is given by (Pankove, 1971 and Gittleman *et al.,* 1979):

$$k(h\upsilon) = \frac{\lambda}{4\pi}\alpha(h\upsilon)$$

The calculated values of extinction coefficient k for $ZnSe_xTe_{1-x}$ thin films were plotted as a function of photon energy $h\upsilon$ and are shown in figure (6). The extinction coefficient of the films is high at higher incident photon energy. It is observed from figure (6) that the extinction coefficient in the wavelength ranges 445-765nm decreases from 0.3443 to 0.0086 with increasing thin film thickness. The increase of the extinction coefficient with the increase of thickness can be related to the change in the transmittance of the films. The structural and optical parameters studied in this work are found to be very sensitive to the kind of used substrate and to the thin film thickness.

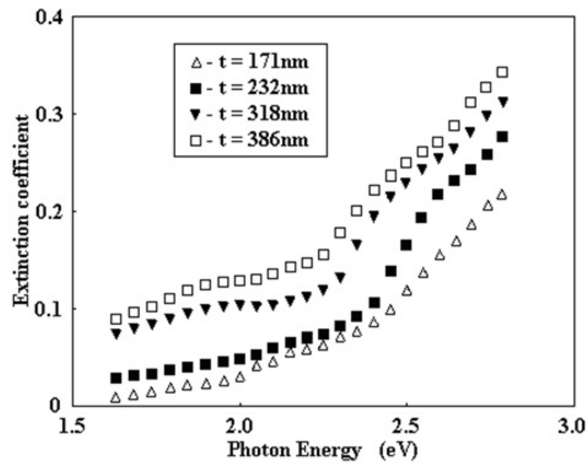

Fig. 6: Extinction coefficient vs. photon energy for $ZnSe_{0.5}Te_{0.5}$ thin films evaporated at substrate temperature 473K with different thicknesses on glass substrates.

Conclusions:

Semiconducting $ZnSe_{0.5}Te_{0.5}$ thin films were prepared with different thicknesses on Si (100) and glass substrates at a constant substrate temperature of 473K using vacuum evaporation technique. XRD study show that the vacuum evaporated $ZnSe_{0.5}Te_{0.5}$ thin films are polycrystalline cubic structure. Structural parameters such as lattice parameter and inter-planar spacing were obtained from Bragg's angle of predominant orientation <111>. Grain size, dislocation density and microstrain were calculated from FWHM of <111> diffraction line. These structural parameters are found to be very sensitive to the kind of substrate and to the thin film thickness. Extinction coefficient was calculated from optical transmittance and optical absorption values using spectrophotometer technique. The band tail energy was estimated from optical absorption curves. It is found that the band tail energy ΔE decreases as the thin film thickness increases. In conclusion we say that the direct band gap for $ZnSe_{0.5}Te_{0.5}$/glass can be tuned from 2.457 – 2.184eV by varying the thin film thickness (171-386nm).

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