

Influence of Annealing Temperatures on Nonlinear Optical, Dielectric, Semiconducting Results, and Fermi Level Position for CdP_{0.03}Te_{0.97} Thin Film

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Abstract: CdP_{0.03}Te_{0.97} thin films were deposited at room temperature using thermal evaporation and annealed at 100 and 200°C. The effect of annealing temperature T_{ann} on both dispersion energy E_d and oscillating energy E_o were studied. The lattice dielectric constant ϵ_L and free carrier concentration/effective mass N/m^* were calculated for these samples. The values of the first order of moment M_{-1} , the third order of moment M_{-3} , and static refractive index n_o were determined. Both of dielectric loss ϵ'' and dielectric tangent loss ϵ'' for these films increased with photon energy ($h\nu$) and had the highest value higher than the energy gap E_g . All of the optical parameters such as real part of optical conductivity σ_1 , the imaginary part of optical conductivity σ_2 and the relation between Volume Energy Loss/ Surface Energy Loss (VEL/SEL) were determined. The linear optical susceptibility $\chi^{(1)}$ increased with T_{ann} . The influence of annealing temperatures on all of the non-linear refractive index n_2 , the third-order non-linear optical susceptibility $\chi^{(3)}$, and non-linear absorption coefficient β_c were studied. Both of the electrical susceptibility χ_e and relative permittivity ϵ_r increased with T_{ann} and had the highest value higher than E_g . The dependence of density valence band, conduction band, and position of Fermi level E_f were studied.

Keywords: CdP_{0.03}Te_{0.97} thin film; thermal annealing; dielectric results; linear optical susceptibility; non-linear optical parameters and semiconducting results.

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1. Introduction

CdTe is II-VI crystalline compound with a zinc blende crystal structure and has a direct bandgap of 1.44 eV [1] which is suitable for electronic applications such as photovoltaic devices [2] light-emitting diodes [3,4] solar cells [5,6] X-ray and gamma detectors [7] Field Effect Transistors (FETS) [8] Lasers [9] and non-linear integrated optical devices [10]. The structure of CdTe thin films was carried out by many authors [11-15]; it was found that CdTe thin films had a polycrystalline structure [11-12], and the annealing process increased the crystallinity of CdTe films [15,16]. The optical properties of CdTe thin films were studied [17-23] it was found that, the energy gap values were (1.44-1.60 eV) [17] (1.45-1.52 eV) [18] 1.534 eV [19] (1.43-1.50 eV) [21] and also CdTe had a high absorption coefficient (10^4 cm^{-1})

[22,23]. The annealing effect on optical properties was investigated [24-28]; it was found that the annealing process decreased the energy gap of CdTe [27] and reduced both transmission and reflection for CdTe [28]. On the other hand, the doping effect on CdTe thin films' optical properties was studied [29-35]. It was found that Sn dopant increased absorption coefficient [29] Al, Sb dopant ratio decreased energy gap [31] band gap increased refractive index and extinction coefficient decreased with increasing Ni ratio in Cd_{1-x}Ni_xTe films [34]. The transmittance spectra decreased by the addition of Cu [35]. The increase in bandgap energy and the effect of annealing temperature on structure and optical properties of CdPTe thin films were studied [36]; it was found that the optical energy gap increased with annealing temperature.

This work investigates the annealing temperatures' effect on dielectric semiconducting results, Fermi level position, first-order optical susceptibility, and non-linear optical results for CdTe thin-film doped with phosphorus.

2. Materials and Methods

CdTe doped with phosphorus compound was synthesized by the direct fusion method. Mixtures of high purity elements (Cd, P, and Te) (99.999 %) in stoichiometric portions (obtained from Aldrich Chemical Co.) were used. The composition mixture was taken in a graphitized silica tube evacuated at a pressure of 10⁻⁵ kPa. The evacuated tube was then placed into a furnace whose temperature was raised gradually to 1000 °C. During the synthesis, the molten material was shaken every 2 h to ensure homogeneity. Then the temperature of the furnace was kept at this temperature for 144hs and then cooled to room temperature. Thin cadmium telluride doped with phosphorus films was prepared by vacuum thermal evaporation on glass substrates using an Edward coating unit under pressure 10⁻⁵ kPa. Glass substrates were cleaned by a cleaning solution rinsed in distilled water than pure alcohol in an ultrasonic cleaner. Transmittance (*T*) and reflectance (*R*) of the as-deposited, annealed thin films at 100 and 200°C were measured at normal incidence using a Jasco (V-570) spectrophotometer from 500 to 2500 nm. The thickness of the evaporated films was determined by a quartz thickness monitor and confirmed by multiple-beam interferometers.

3. Results and Discussion

3.1. Dielectric, optical conductivity, and linear optical susceptibility results.

The structure of these films with different annealing temperatures is illustrated in previous work [36]. The optical transmittance *T* and reflectance *R* were measured and discussed in previous work [36]. The single oscillator theory was expressed by Wemple–DiDomenico relationship [37]:

$$n^2(E) - 1 = \frac{E_o \cdot E_d}{E_o^2 - E^2}$$

Where *n* is the refractive index values of these samples which are determined in previous work [36] *E* is the photon energy, *E_o* is the oscillator energy, and *E_d* is the dispersion energy. The values of *E_o* and *E_d* for all samples are shown in Table 1.

Table 1. The determined values of CdP_{0.03}Te_{0.97} film with different annealing temperatures such as lattice dielectric constant (ϵ_L), Oscillation energy (E_o), Dispersion energy (E_d), the first order of moment (M_{-1}), third order of moment (M_{-3}), Field strength (f), static refractive index (n_o), (N/m^*), the density of conduction band N_c (cm^{-3}), the density of valence band N_v (cm^{-3}) and Fermi level position.

Sample	Lattice dielectric constant ϵ_L	Oscillation energy E_o (eV)	Dispersion energy E_d (eV)	M_{-1} (eV)	M_{-3} (eV)	Field strength (f) (eV) ²	n_o	N/m^*	N_c	N_v	Fermi Level Position (eV)
As-deposited	16.00	3.40	6.50	4.70	2.55	22.10	1.70	2.1E+49	7.18E+22	3.1E+21	0.11
Annealed at 100 °C	18.00	3.60	6.80	4.95	2.61	24.48	1.69	2.8E+49	1.15E+23	5.0E+21	0.15
Annealed at 200 °C	21.50	3.75	7.10	5.16	2.66	26.63	1.71	1.5E+50	1.64E+23	7.2E+21	0.19

Figure 1 shows the relation of n^2 and λ^2 to determine the ratio of effective mass to carrier concentration using the following equation [38]:

$$n^2 = \epsilon_L - \left(\frac{eN}{4\pi c^2 \epsilon_o m^*} \right) \lambda^2$$

Where ϵ_L is the lattice dielectric constant, ϵ_o is the permittivity of free space, e is the charge of the electron, n , k are the linear refractive index and the absorption index of these films, respectively, which was determined in previous work [36] N is the free carrier concentration for CdP_{0.03}Te_{0.97} film with different annealing temperatures and c is the speed of light so the values of N/m^* are shown in Table 1. From this table, the annealing temperatures affected the ratio of N/m^* the high annealing temperature induces a higher number of free electrons. The values of the first order of moment M_{-1} and the third order of moment M_{-3} derived from the relations [38]:

$$E_o^2 = \frac{M_{-1}}{M_{-3}}$$

Table 1 shows the values of M_{-1} and M_{-3} for these thin films. The oscillator strength (f) was calculated as following [39]:

$$f = E_o \cdot E_d$$

The values of f are shown in Table 1.

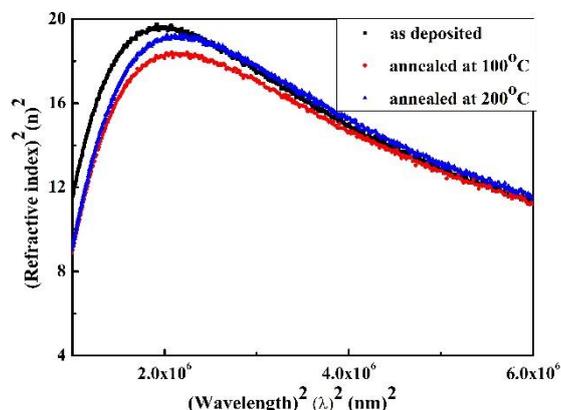


Figure 1. Relation of n^2 and λ^2 for CdP_{0.03}Te_{0.97} thin film with different annealing temperatures.

Another important parameter depending on both E_o and E_d is that static refractive index n_o , which was determined using the following equation [40]:

$$n_o = \left[\left(\frac{E_d}{E_o} \right) + 1 \right]^{0.5}$$

The values of n_0 for all these samples are shown in Table 1. Figure 2 represents the dependence of $(n^2-1)^{-1}$ on $h\nu$ for these thin films. From this figure, it was seen that all these samples had the same behavior, and the values of $(n^2-1)^{-1}$ increased with annealing temperatures.

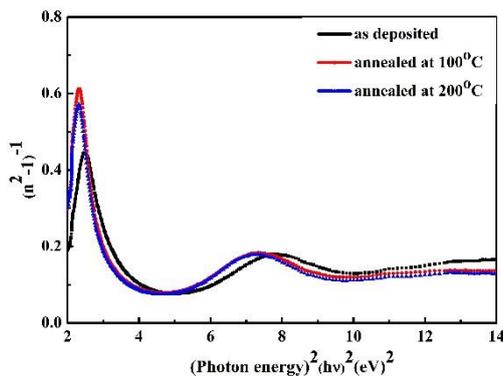


Figure 2. $(n-1)^{-1}$ vs. $h\nu$ for CdP_{0.03}Te_{0.97} thin film with different annealing temperatures.

The dielectric loss (ϵ'') and dielectric tangent loss (ϵ''') for these films were calculated as following [41]:

$$\epsilon'' = (n^2 + k^2)$$

$$\epsilon''' = [(n^2 + k^2)^2 - (n^2 - k^2)^{0.5}]$$

Figure 3(a,b) shows the relation between both ϵ'' and ϵ''' and $h\nu$ for CdP_{0.03}Te_{0.97} film with different annealing temperatures. From this figure, it was seen that, at energy less than energy gap, the values of both of ϵ'' and ϵ''' decreased with annealing temperatures for all studied samples, and the peak maximum values position decreased with increasing annealing temperatures, this is due to the increasing of electron motilities with annealing temperatures.

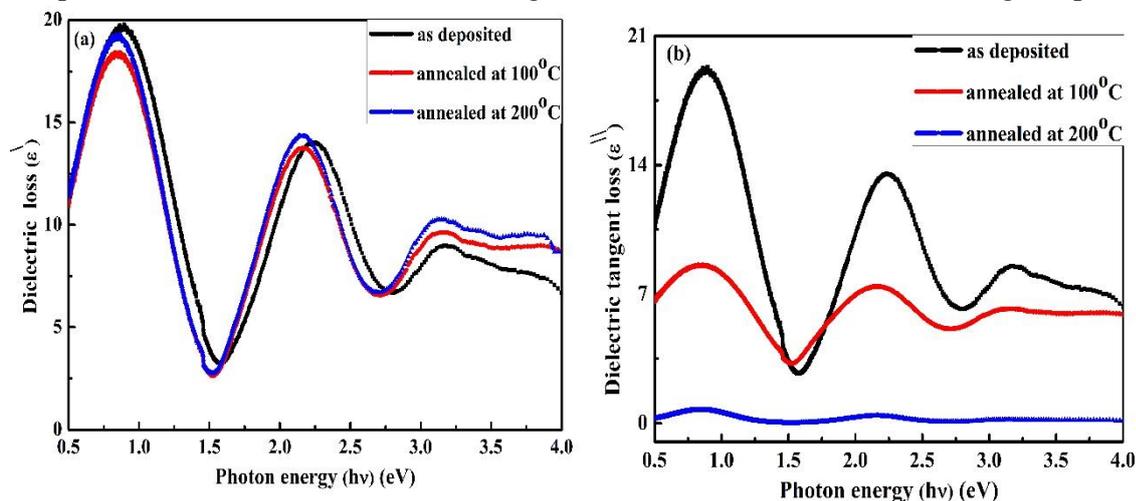


Figure 3. (a) The dependence of ϵ'' ; (b) ϵ''' on $h\nu$ for CdP_{0.03}Te_{0.97} film with different annealing temperatures.

The optical conductivity was calculated from the following equations [42]:

$$\sigma_1 = \left(\frac{\epsilon''' \cdot c}{2\lambda} \right)$$

$$\sigma_2 = \frac{(1 - \epsilon'') \cdot c}{4\lambda}$$

The dependence of σ_1 and σ_2 on $h\nu$ for these films is shown in Figure 4(a,b). The behavior of both σ_1 and σ_2 for all these studied films is the same with $h\nu$ and increase with $h\nu$ for all these samples.

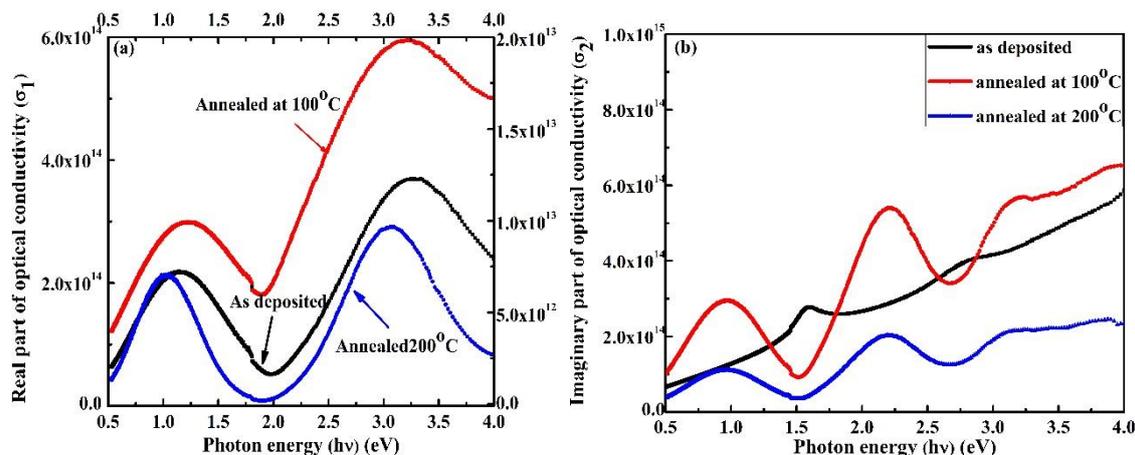


Figure 4. (a) The dependence of σ_1 ; (b) σ_2 on $h\nu$ for CdP_{0.03}Te_{0.97} film with different annealing temperatures.

The values of Volume Energy Loss Function (VELF) and Surface Energy Loss Function (SELF) for these films were determined optically as following [38]:

$$VELF = \frac{\epsilon''}{\epsilon'^2 + \epsilon''^2}$$

$$SELF = \frac{\epsilon''}{(\epsilon' + 1)^2 + \epsilon''^2}$$

The dependence of VELF/SELF on annealing temperatures is shown in Figure 5(a). $\chi^{(1)}$, which describes the effect of wave length on films optical response, was calculated as [43]:

$$\chi^{(1)} = \frac{(n^2 - 1)}{4\pi}$$

The relation between $\chi^{(1)}$ and $h\nu$ for CdP_{0.03}Te_{0.97} thin film with different annealing temperatures is shown in Figure 5(b). From this figure, it was seen that the linear optical susceptibility $\chi^{(1)}$ increased with annealing temperatures, and the values of $\chi^{(1)}$ had maximum values higher than E_g . This means that there is a possibility of a wide change in optical properties by thermal annealing.

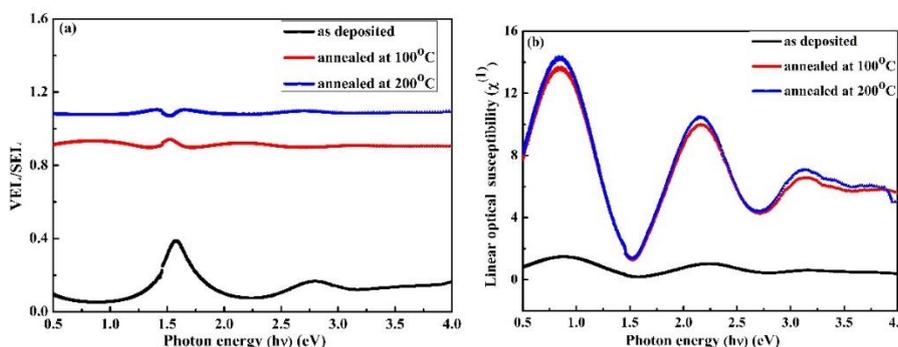


Figure 5. (a) Dependence of VEL/SEL ; (b) $\chi^{(1)}$ on $h\nu$ for CdP_{0.03}Te_{0.97} film with different annealing temperatures.

3.2. Non-linear optical properties.

The non-linear refractive index n_2 , which can be explained as when intense light go through media the refractive index change, which is non-linear phenomena[44] n_2 was determined from the following simple equation [45-46]:

$$n_2 = \frac{(12 \pi \chi^{(3)})}{n_o}$$

The dependence of n_2 on wavelength for CdP_{0.03}Te_{0.97} thin film with different annealing temperatures is shown in figure 6(a). The values of n_2 decrease with wavelength and annealing temperature for all these studied samples; this is due to increased transmittance with annealing temperature [36, leading to a decrease of propagated light and may be attributed to the degree increase of homogeneity of the thin film. An important parameter to assess the degree of nonlinearities in the third-order non-linear optical susceptibility $\chi^{(3)}$ which was determined using the following equation [47]:

$$\chi^{(3)} = A \left[\frac{E_o \cdot E_d}{4\pi(E_o^2 - (h\nu)^2)} \right]^4$$

Where $A = 1.7 \times 10^{-10}$ e.s.u [45]. The dependence of $\chi^{(3)}$ on $h\nu$ for CdP_{0.03}Te_{0.97} film with different annealing temperatures is shown in Figure 6(b). It was noticed that the behavior of $\chi^{(3)}$ is the same for all the studied sample the values of $\chi^{(3)}$ increase with $h\nu$ this is due to when $h\nu$ increases, the deflection of the incident light beam increases and the values of $\chi^{(3)}$ decrease with annealing temperature this could attribute to the variation of free carrier concentration which leads to the increase of electrons mobility with annealing temperatures. On the other hand, another important non-linear parameter such as the non-linear absorption coefficient β_c , which determined as following [48]:

$$\beta_c = \frac{48 \cdot \pi^3 \cdot \chi^{(3)}}{n^2 \cdot c \cdot \lambda}$$

The values of β_c increase with $h\nu$ for all these samples, as shown in Figure 6(c), because of the higher values of $h\nu$ a large number of excited electrons overcoming the bandgap.

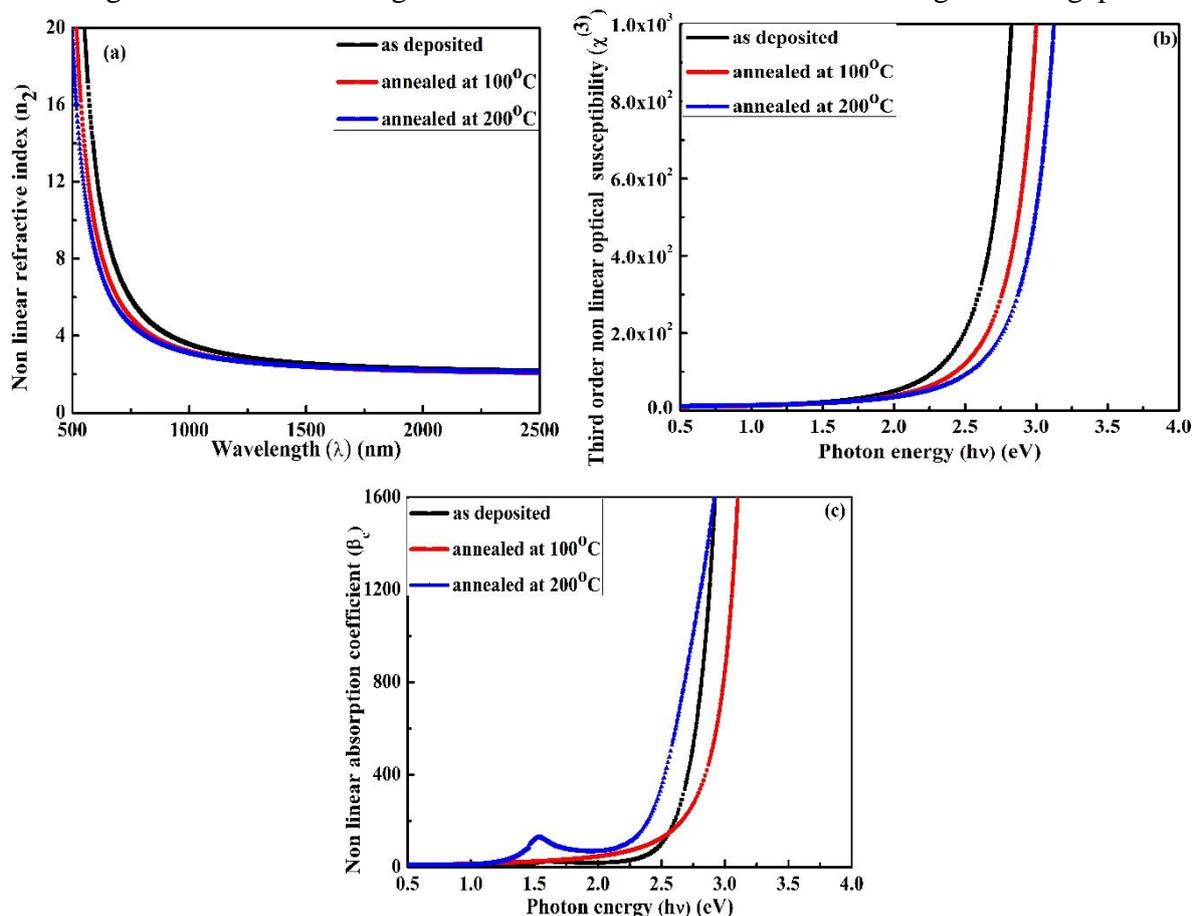


Figure 6. (a) Relation between n_2 and λ , the influence of $h\nu$ on both of; (b) $\chi^{(3)}$; c β_c for CdP_{0.03}Te_{0.97} film with different annealing temperatures.

3.3. Electrical results.

Electrical susceptibility $\chi_{(e)}$ was determined using the following relation [49]:

$$\chi_{(e)} = \frac{(n^2 - k^2 - \epsilon_o)}{4\pi}$$

Figure 7(a) shows χ_e vs. $h\nu$ of these investigated samples. From this figure, it is clear that the values of χ_e increase with annealing temperatures. This is due to the increasing electron mobility. The relative permittivity ϵ_r was calculated using the following relation [50]:

$$\epsilon_r = (\chi_e + 1)$$

The relation between relative permittivity ϵ_r and wavelength for CdP_{0.03}Te_{0.97} thin film with different annealing temperatures is shown in Figure 7(b). It was found that ϵ_r had strongly affected by annealing temperatures; this could be attributed to the electron mobility changes with annealing temperature.

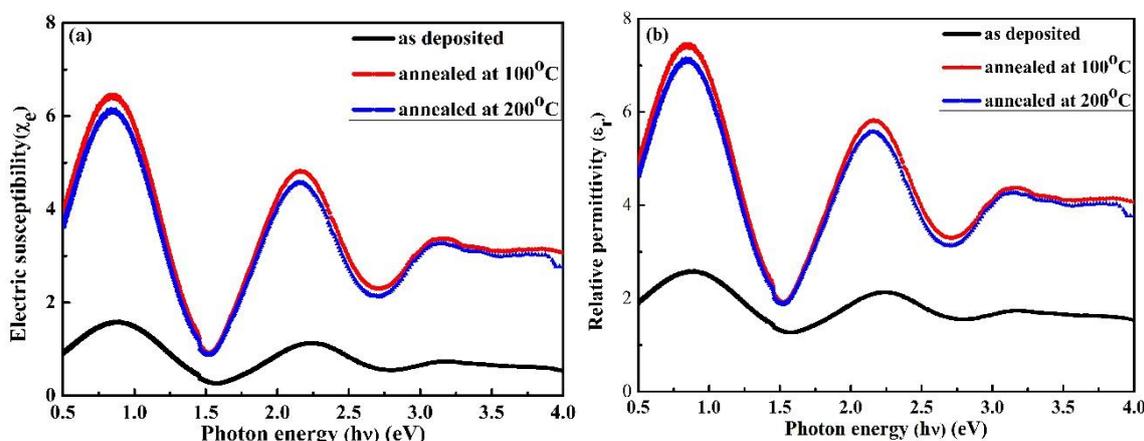


Figure 7. (a) The dependence of electrical susceptibility $\chi_{(e)}$; (b) relative permittivity ϵ_r on $h\nu$ for CdP_{0.03}Te_{0.97} film with different annealing temperatures.

3.4. Semiconducting and electronic results.

Both N_v and N_c play very important rules examination of the linear optical transition and non-linear optical properties. N_v and N_c are calculated as following [51]:

$$N_v = 2 \left[\frac{(2\pi m^*_h KT)}{h^2} \right]^{3/2}$$

$$N_c = 2 \left[\frac{(2\pi m^*_e KT)}{h^2} \right]^{3/2}$$

where N_v and N_c are the density of states for both valence and conduction bands respectively, effective mass of electrons in (CdTe) $m^*_e = 0.11$ [52] effective mass of holes in (CdTe) $m^*_h = 0.18$ [52] effective mass of holes in (CdP) $m^*_h = 0.4$ [53] effective mass of electrons in (CdP) $m^*_e = 0.05$ [53] and K is a Boltzmann constant. The determined values for both N_v , N_c are shown in table 1. Another important factor that was determined theoretically is the position of Fermi level [54]:

$$E_f = \left(\frac{KT}{q} \right) \cdot \ln \left(\frac{N_c}{N_v} \right)$$

The values of Fermi level position for these investigated thin films are shown in Table 1.

4. Conclusions

The values of E_d and E_o increased with annealing temperatures for $CdP_{0.03}Te_{0.97}$ (E_d from 6.50 to 7.10 eV), and also E_o had the values from (3.4 to 3.75 eV). The values of N/m^* increased with annealing temperatures T_{ann} , which increased free carrier. The values of M_{-1} and M_{-3} also increased with increasing T_{ann} . Both of ϵ^{\parallel} and ϵ^{\perp} increased with $h\nu$, and the maximum values decreased with increasing T_{ann} due to increased electron mobility with increasing T_{ann} . Both σ_1 and σ_2 decreased with T_{ann} . $\chi^{(1)}$ increased with T_{ann} due to increasing carrier concentration. The values of n_2 increased with λ for all these samples, while χ^3 increased with $h\nu$. This means that these samples could change their optical properties by changing wavelength and applied field. The non-linear absorption coefficient (β_c) decreased with T_{ann} , while both of the χ_e and ϵ_r increases with T_{ann} and had the highest value higher than the energy gap. The annealing temperatures affected the values of both N_v and N_c , while E_f affected slightly with T_{ann} .

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Conflicts of Interest

The authors declare no conflict of interest.

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