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Influence the number of laser pulses and annealing temperature and the Structure and optical properties of In_2O_3: CdO films prepared laser induce plasma

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Abstract
In this Research, (In_2O_3: CdO) films were prepared using pulsed laser deposition (PLD) method on glass substrate at room temperature deposited at laser influence 500mJ/cm^{2}with different shoots N= (200,300,400,500and600). the structural, and the optical properties and the films are studied with different annealing temperatures (523and 623) K. Optical measurements and the films were analyzed by UV-VIS absorption spectra. The structural properties of samples were investigated by x-ray diffraction patterns of the films and show that the films and polycrystalline Structure with all shoots. Transmittance spectrum found is equal to 93.17%, refractive index range is 1.635 and energy gap range is 2.75-3.15ev.

Keywords: In_2O_3 thin films, X-ray diffraction, Structural, Optical properties, Pulsed laser deposition: In doped CdO nanostructures

تاثير عدد نبضات الليزر ودرجة حرارة التلدين على الخصائص البصرية والهيكلية لمركب المحضر كغشاء بطريقة الحث بالبلازمIn_2O_3: CdO

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الخلاصة
في هذا البحث ، تم تحضير أفلام (In2O3: CdO) باستخدام طريقة الترسيب بالليزر النابضي (PLD) على ركائز زجاجية في درجة حرارة الغرفة بتأثير طاقة ليزر 500 mj / cm^2 مع نبضات الليزر المختلفة N=(200،300،400،500and600). تمت دراسة البنية الهيكلية، والخصائص البصرية للاغشية مع درجات حرارة التلدين المختلفة (523 و 623) K. تم تحليل ودراسة القياسات البصرية للاغشية بواسطة أطياف الامتصاص UV-VIS. تم فحص الخواص التركيبية للعينات من خلال أنماط حيود الأشعة السينية وتبين أن الأغشية متعددة التبلور لجميع نبضات الليزر. وجد ان طيف النفاذية يساوي 93.17٪ ، معامل الانكسار هو 1.635 و فجوة الطاقة هو2.75-3.15 الكترون فولت .

Introduction

The extensive and intensive investigation of indium oxide (In_2O_3) during the last two decades can be directly linked to its remarkable combination of electronic and optical properties. Specifically, higher values for both electrical conductivity and transmission in the visible and near infrared make feasible the exploitation of In_2O_3 thin films in numerous optoelectronics applications, from solar cells to liquid crystal panel displays and switching devices. recent years much regard has been focused on

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reactive Pulsed Laser deposited In_2O_3 films because of their characteristic such as deposited from metallic indium target in the presence of reactive gas of oxygen, high deposition rates, film uniformity on small area and precise control over the composition of the deposited film [1]. The physical properties of In_2O_3 films prepared by this technique mainly depend on the depositing parameters like oxygen partial pressure, substrate temperature, laser energy, number of laser pulses, the angle of deposition. In order to achieve optical transparency, the substrate should be kept at high temperatures during deposition of the Thin films this investigation, the influence of substrate bias on the crystallographic structure and optical properties of the films prepared by reactive pulsed laser deposition CdO is transparent conducting oxides (TCOs) materials that possess both high electrical conductivity and high optical transparency (>80%) in the visible light region of the electromagnetic spectrum [2]. CdO is a n-type semiconductor with nearly metallic conductivity [3]. It has a direct energy band gap (Eg) of ~2.3 eV and two indirect transitions at lower energies [4]. Mixed oxides such as CdO: In_2O_3 have at a recent time received large attention, since they combine many beneficial characteristics of both In_2O_3 and CdO. Un doped and doped In_2O_3 thin films have been obtained by different techniques [5], spray pyrolysis [6], and pulsed laser deposition [7]. The aim of this research is to optimize the Structure and optical properties of In_2O_3-doped cadmium oxide thin films deposited by pulsed laser deposition (PLD).

Experimental

In_2O_3: CdO films were prepared by pulsed laser deposition and the experiment was accomplished in a vacuum chamber in generally (10^{-3} mbar) vacuum conditions, at a low pressure of the gas background for specified cases of oxides and nitrides, which shows the arrangement of the target and substrate holders inside the chamber with regard to the laser beam. The focused Nd: YAG Q-switching laser beam was coming through a window to be incident on the target surface at an angle 45°. The substrate was placed in front of the target and parallel and. Fixed about (3cm) the real distance was kept between the target and the substrate so that the substrate holder did not champer the incident laser beam. Modification of the deposition technique is done by Many investigators to obtain the better-quality films by this method. These included the rotation of the target, and the positioning of the substrate with respect to target. In_2O_3, CdO, and doped In_2O_3: CdO nanoparticles ratio concentration of as (9) wt. % high purity of 99.9 % is pressed under pressure of 5 ton to form a target of 1.5 cm diameter and 0.2 cm thickness. It should be as dense and homogeneous as much as possible to ensure a good quality of the deposit. the prepare thin films of with In_2O_3 with doping ratios the concentration of added oxidized of as (9) wt. %. of CdO and are deposited on glass substrates, using laser energy (500) mJ as laser influence and different shoots = (200,300,400,500,600). The experiment was done in a vacuumed chamber to about (10^{-3} mbar), type of substrates glass, pulse laser deposition using Nd: YAG laser with 1064 nm wavelength, Pulse duration of laser beam 6ns and annealed film at 250 ,350 °C for two hours.

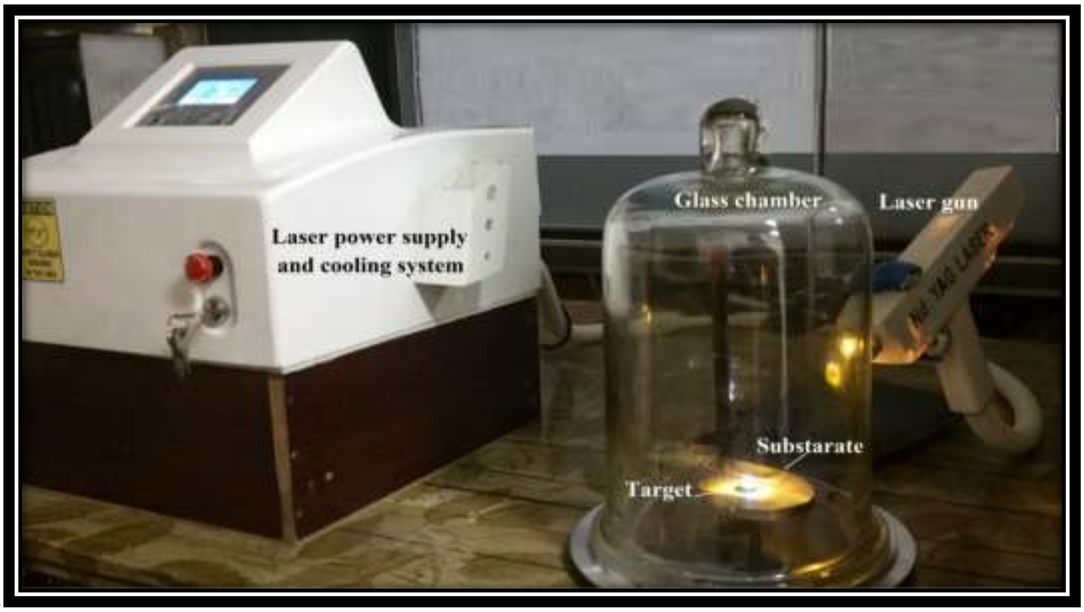


Figure 1-Pulse laser deposition experimental set up

Results and discussions

1- X-Ray diffraction for Thin films1

The patterns of the In_2O_3 thin films as shown Figure- 2. The x-ray diffraction patterns for undoped In_2O_3 film grown on glass substrates. Deposit and annealing temperature 523,623 K. Can be take notice from an XRD pattern in the case annealing at 523K the peaks at (30.6607°, 35.5400°, 51.0928°, 60.6989°) referred to (222), (400), (440) and (622) direction, respectively. In the case annealing at 623 can be noticed from that the peaks at (21.5121°, 30.5845°, 35.4638°, 51.0928°) and (60.9276°) referred to (211), (222), (400), (440) and (622). In the case (RT) did not peak because appears amorphous. Which perfectly matches with the In_2O_3 reference of rhombohedral according to (card No 96-101-0589). These results are the same with the other researches [8]. From Figure-2 we also see that the intensities of thin films the peaks increase with the increase of the heat treatment of thin film. The (FWHMs) of 222 peaks in 523 and 623 K were 0.5337° and 0.4574°, respectively. The average grain size evaluated from 222 peaks increased from 15.44 to 18.01 nm with the increase of substrate temperature from 523 to 623 K, which was ascribed to the improvement in the crystallinity of the films. The same with the other researches [9]. Figure-3 shows the X ray diffraction (XRD) patterns of doped the prepared 9% In_2O_3: CdO film. The pattern shows polycrystalline of cubic CdO structure films are composed of crystallites of CdO and In_2O_3. XRD shows nor mixed phases. Seen that the film is orientated along (222) crystallographic directions, and this is in agreement with the result obtained by others on film prepared by pyrolysis method for, vacuum evaporation [10] and spray pyrolysis [11]. The (FWHM) method that is often calculated by Scherrer's relation [12],

$$D = \frac{k\lambda}{\beta\, cos(\theta)} \quad \ldots\ldots (1)$$

Where λ is the wavelength of X-ray used (1.54 A), calculated crystalline size (D) and The FWHM and the grain size of the samples are shown in the Table-1, In the case (RT) did not peak because appears amorphous. Which perfectly matches with the In_2O_3 reference of rhombohedral according to (card No 96-101-0589) and also see that the intensities of thin films the peaks increase with the increase of the heat treatment of thin film. The (FWHMs) of 222 peaks in 523 and 623 K were 0.5337° and 0.4574°, respectively. The average grain size evaluated from 222 peaks increased from 15.44 to 18.01 nm with the increase of substrate temperature from 523 to 623 K. Shown in the Table-2, The FWHM decrease with annealing indicate on the increase in crystalline size, all pattern was polycrystalline cubic structure (rock salt) identical with standard card (No. 96-101-0589). XRD shows peaks corresponding to CdO and In_2O_3 phases, and not presence of ternary compound. The preferred orientation was along (222) for In_2O_3 structure.

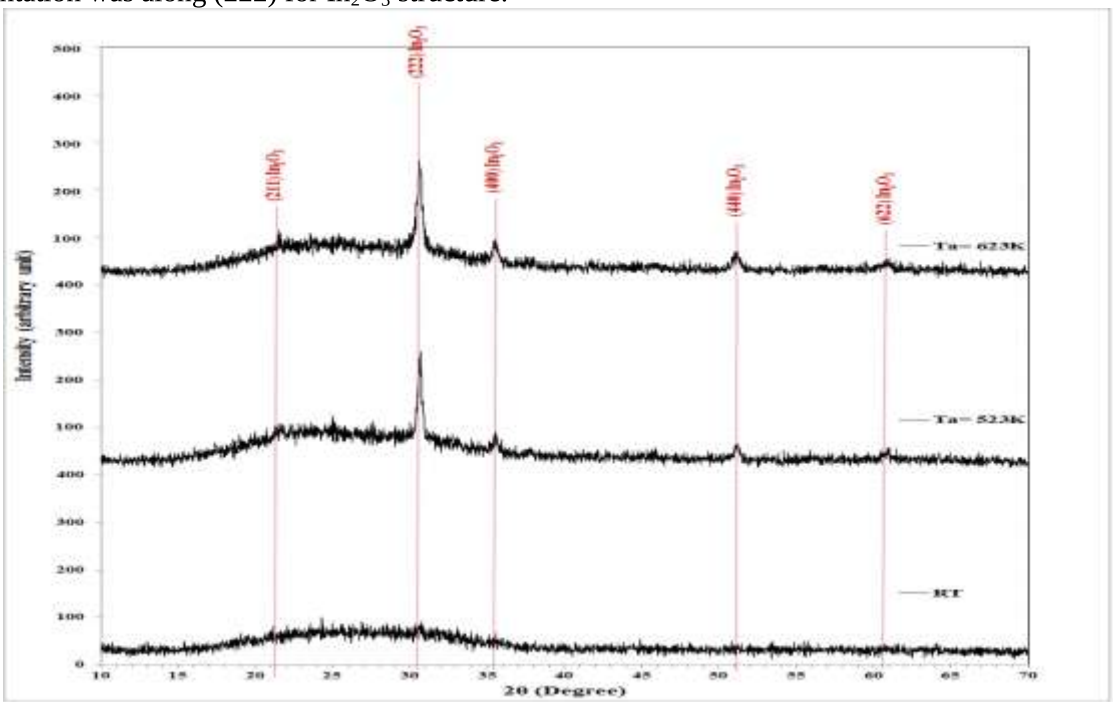


Figure 2-XRD patterns of In_2O_3 in three cases of temperatures RT, 523and623 K, thickness of this sample at 159nm.

Table 1-Illustrates the parameters: 2θ, d_{hkl}, (hkl), FWHM and G.S of In_2O_3 films at three cases of temperatures RT, 523,623 K

Tea (K)	2θ (Deg.)	FWHM (Deg.)	d_{hkl} Exp. (Å)	Crystalline size (nm)	hkl	d_{hkl} Std. (Å)	Phase	Card No.
RT	Amorphous							
523	30.6607	0.5337	2.9136	15.44	(222)	2.9214	In_2O_3	96-101-0589
	35.5400	0.6099	2.5239	13.68	(400)	2.5300	In_2O_3	96-101-0589
	51.0928	0.7624	1.7862	11.55	(440)	1.7890	In_2O_3	96-101-0589
	60.6989	0.7624	1.5245	12.08	(622)	1.5256	In_2O_3	96-101-0589
623	21.5121	0.4574	4.1275	17.69	(211)	4.1315	In_2O_3	96-101-0589
	30.5845	0.4574	2.9207	18.01	(222)	2.9214	In_2O_3	96-101-0589
	35.4638	0.4574	2.5292	18.24	(400)	2.5300	In_2O_3	96-101-0589
	51.0928	0.8386	1.7862	10.50	(440)	1.7890	In_2O_3	96-101-0589
	60.9276	0.8387	1.5193	10.99	(622)	1.5256	In_2O_3	96-101-0589

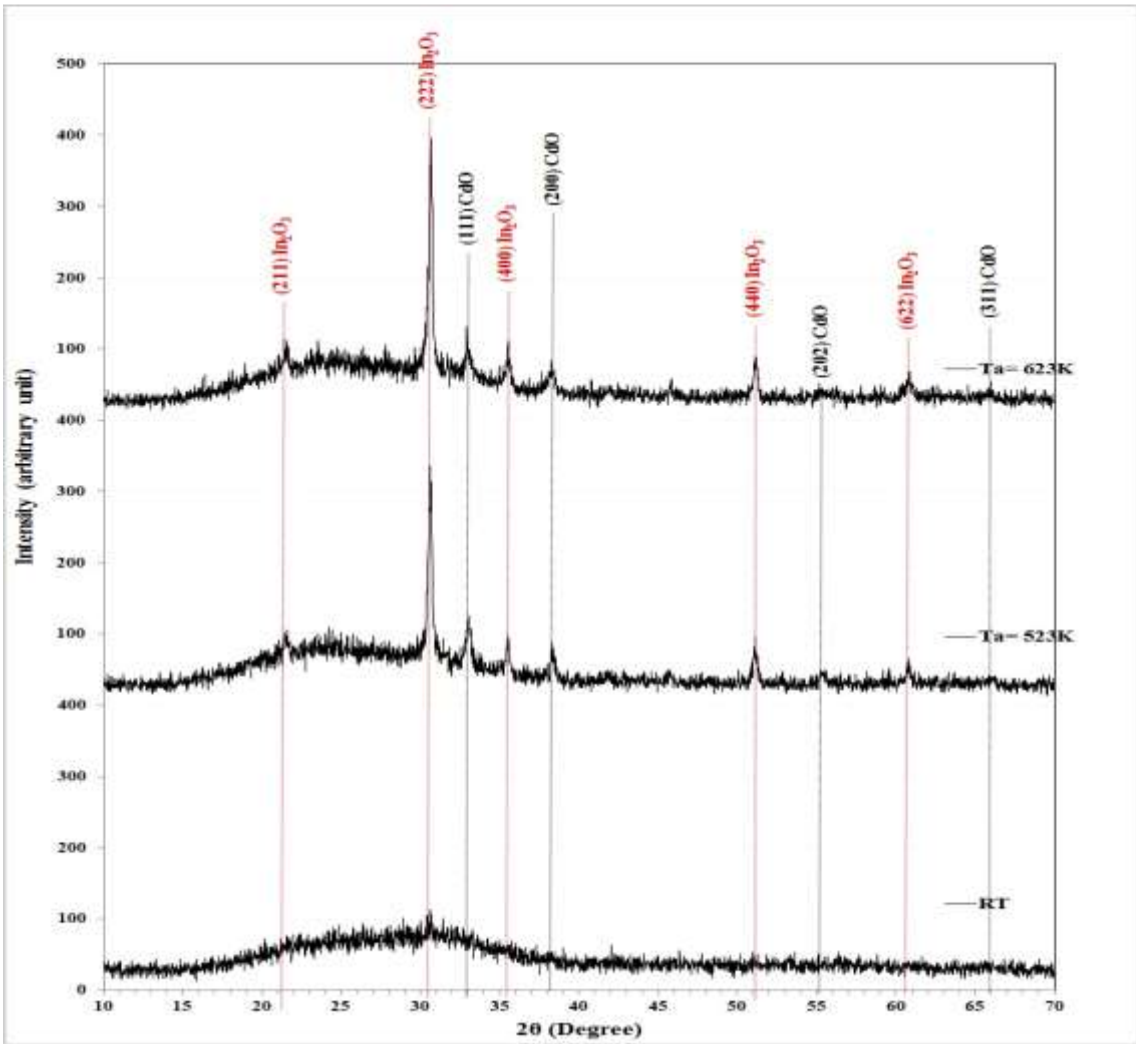


Figure 3-X-ray diffraction patterns of doped In_2O_3: CdO in three cases of temperatures RT, 523 and 623 K, thickness of this sample at 221nm.

Table 2- Illustrates the parameters: 2θ, d_{hkl}, (hkl), FWHM ad G.S of compound In_2O_3: CdO films at three cases of temperatures RT, 523and 623 K

Tea (K)	2θ (Deg.)	FWHM (Deg.)	d_{hkl} Exp.(Å)	Crystalline size (nm)	hkl	d_{hkl} Std.(Å)	Phase	Card No.
RT	30.5845	0.7624	2.9207	10.81	(222)	2.9214	In_2O_3	96-101-0589
523	21.4358	0.4574	4.1420	17.69	(211)	4.1315	In_2O_3	96-101-0589
	30.5845	0.3050	2.9207	27.02	(222)	2.9214	In_2O_3	96-101-0589
	33.0241	0.4575	2.7103	18.12	(111)	2.6848	CdO	96-900-6690
	35.3875	0.4574	2.5345	18.24	(400)	2.5300	In_2O_3	96-101-0589
	38.2846	0.5337	2.3491	15.76	(200)	2.3251	CdO	96-900-6690
	51.0928	0.4575	1.7862	19.25	(440)	1.7890	In_2O_3	96-101-0589
	55.3621	0.5337	1.6582	16.82	(202)	1.6441	CdO	96-900-6690
	60.7751	0.4575	1.5228	20.14	(622)	1.5256	In_2O_3	96-101-0589
	65.8831	0.7624	1.4166	12.42	(311)	1.4021	CdO	96-900-6690
623	21.5121	0.4574	4.1275	17.69	(211)	4.1315	In_2O_3	96-101-0589
	30.6607	0.2287	2.9136	36.04	(222)	2.9214	In_2O_3	96-101-0589
	32.8717	0.4575	2.7225	18.11	(111)	2.6848	CdO	96-900-6690
	35.5400	0.3812	2.5239	21.89	(400)	2.5300	In_2O_3	96-101-0589
	38.2084	0.5336	2.3536	15.76	(200)	2.3251	CdO	96-900-6690
	51.0928	0.3812	1.7862	23.11	(440)	1.7890	In_2O_3	96-101-0589
	55.1334	0.4575	1.6645	19.60	(202)	1.6441	CdO	96-900-6690
	60.8513	0.6100	1.5211	15.11	(622)	1.5256	In_2O_3	96-101-0589
	65.8831	0.6862	1.4166	13.80	(311)	1.4021	CdO	96-900-6690

2- Optical measurements

The transmission spectrum of In_2O_3: CdO films for different annealing temperatures (273, 523, and 623) K has been determined by UV-Visible transmission spectrum in the pulse laser (300-1100) nm on glass substrate. Fig (4) (A, b and c) Shows The transmission spectrum shifts to longer wavelengths with increasing of temperature for all different pulse laser times. It is Obvious that the transmission increases with increasing annealing temperature and this may be due to improving the crystalline size, or due to decrease in the reflection that occurs due to the variation in particle size. This is in agreement with the result [13,14]. The transmission decreases, but not systemically with the high number of pulses with small area of target that mean overlap between pulses because in the case of more atoms are present in the film so more states will be available for the photons to be absorbed. The behavior of the transmission spectra is opposite completely to that of the absorption spectra. Shoots pules 200, 300, 400,500and 600 and constant energy at 500mj. This figure shows that the transmittance decreases with increasing no. of shoot pules due to increasing of thicknesses.

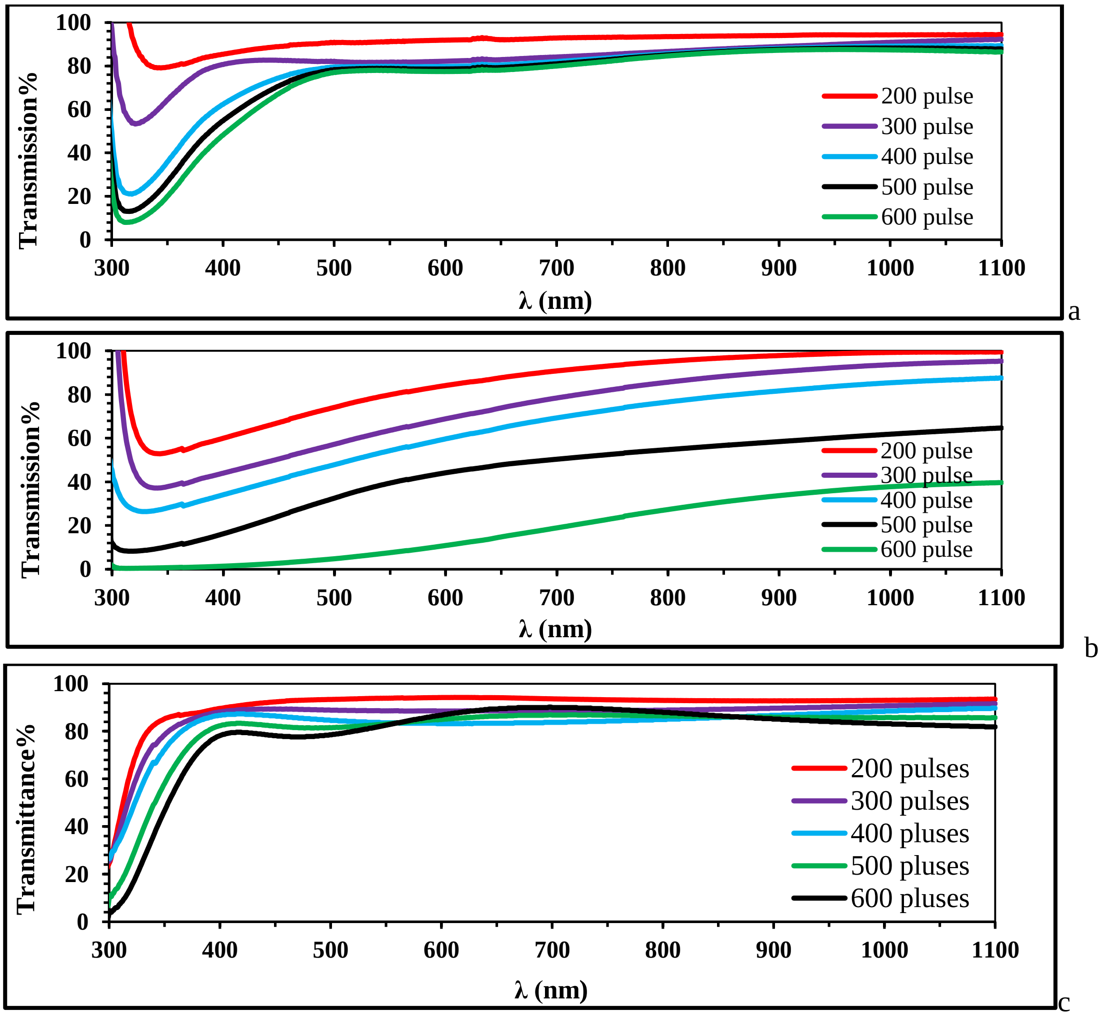
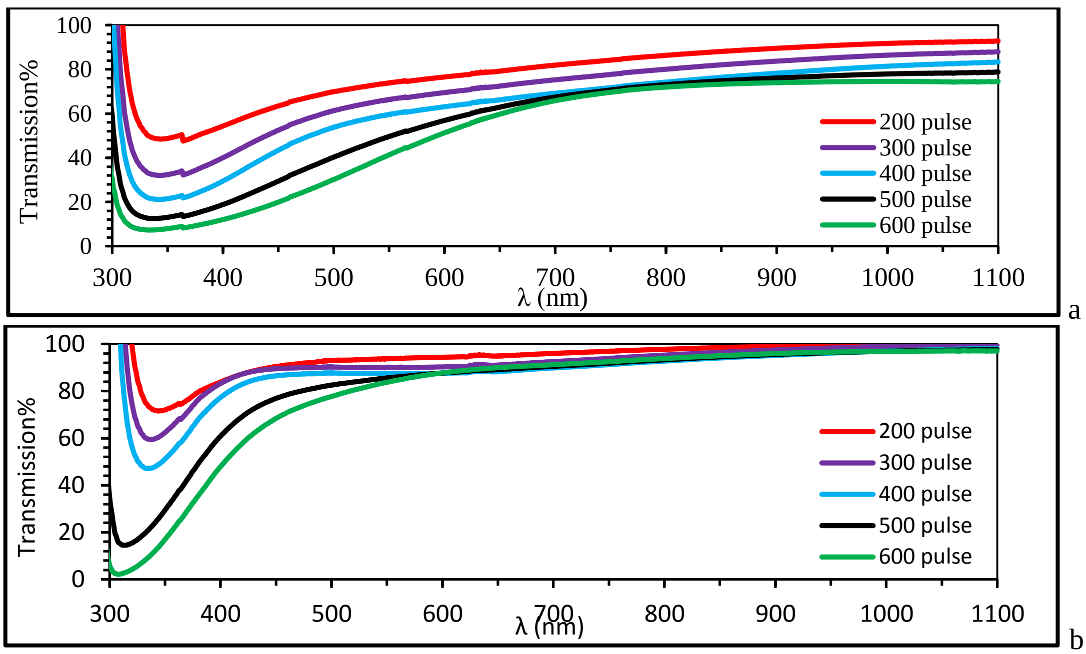


Figure -4 (a, b and c): Transmission spectrum as a function of wavelength for In_2O_3 of pulse laser. for undoped In_2O_3 films, at. (a) For RT, (b) annealing523 K, (C) annealing 623 K.



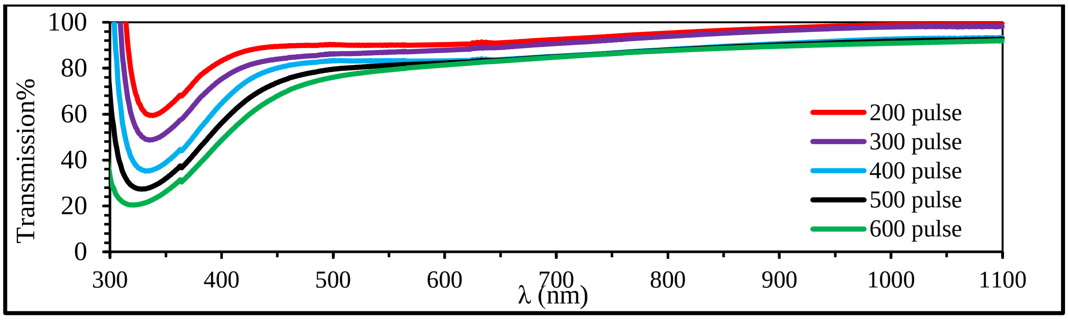


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Figure 5-(a, b and c): Transmission spectrum as a function of wavelength for In_2O_3 of pulse laser. for doped ratios as (9) wt. %. Of CdO film, at. (a) For RT, (b) annealing523 K, (C) annealing 623 K.

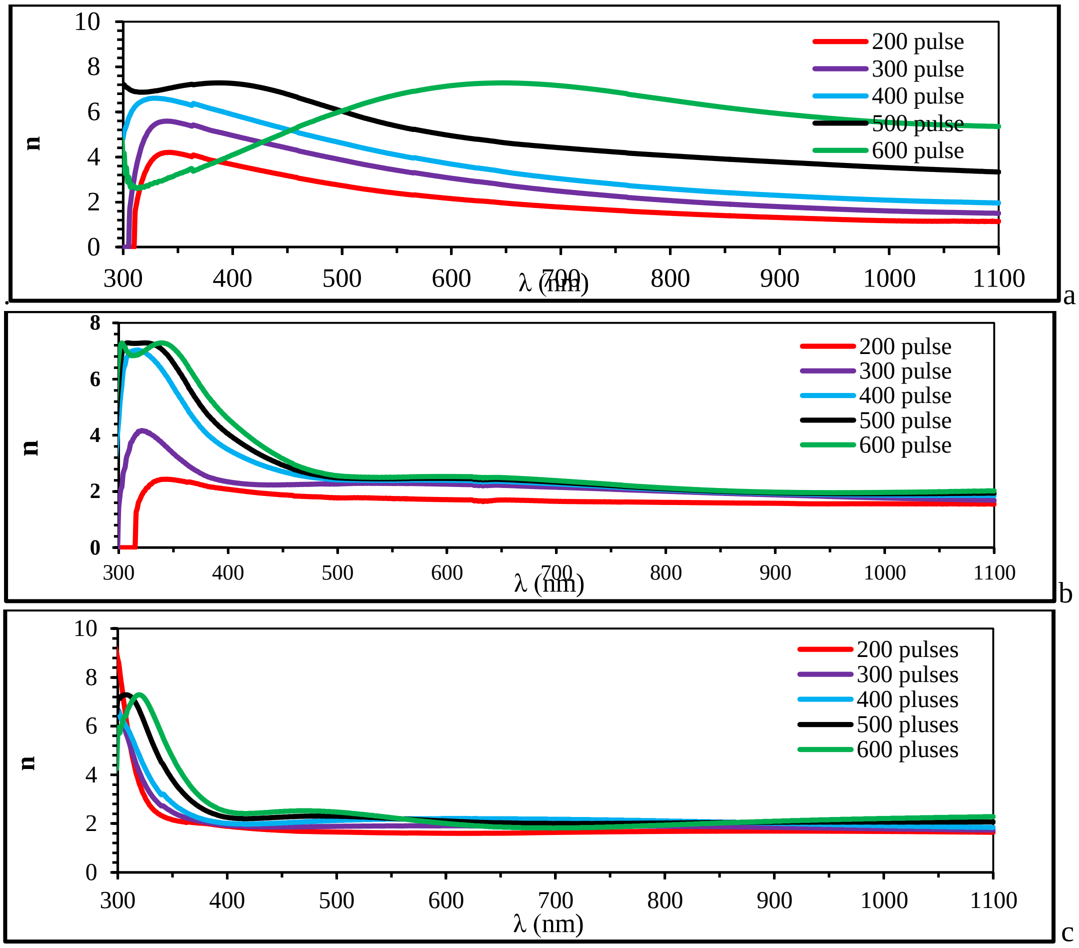


Figure 6-(a, b and c): Variation of, refractive Index as a function of wavelength of pulse laser. for undoped In_2O_3 films, at. (a) For RT, (b) annealing523 K, (C) annealing 623 K.

Shown in Figure -6(a, b and c), which indicate that the refractive index figures that the refractive index, in general decreases slightly with annealing temperature (Ta=623K. This conductance is due to the increase in energy gap which is probably due to the increased grain size and decrease of the defect density which means decreasing of the reflection. Also, we can see from this fig that the refractive index decreases with the increasing of the wavelength of the incident to photon. The results are in close agreement with [15].

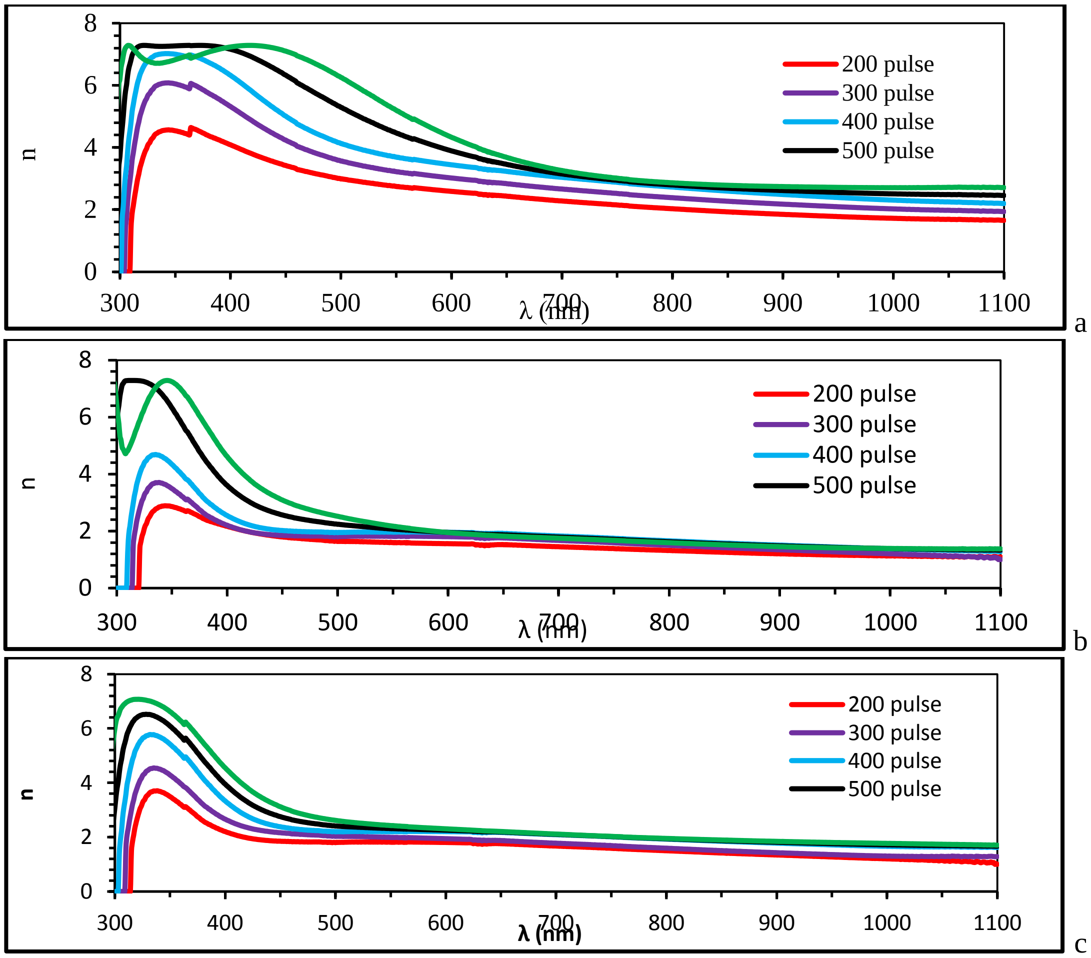
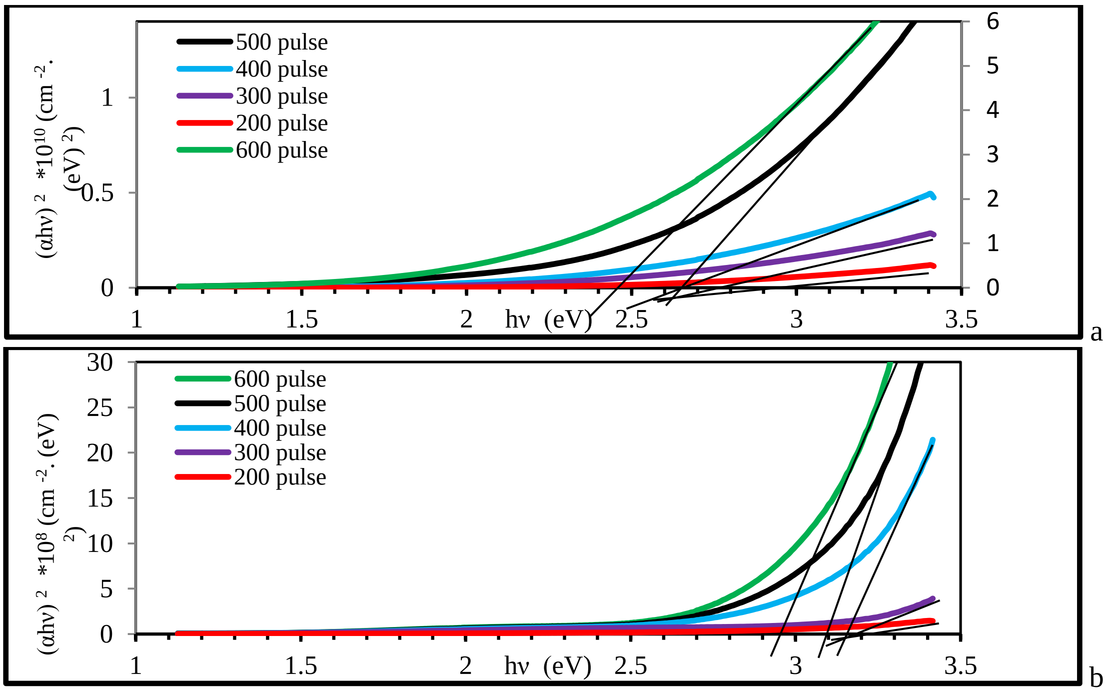


Figure 7-(a, b and c): Variation of refractive Index as a function of wavelength of pulse laser for doped In_2O_3: CdO films, at. (a) For RT, (b) annealing523 K, (C) annealing 623 K.



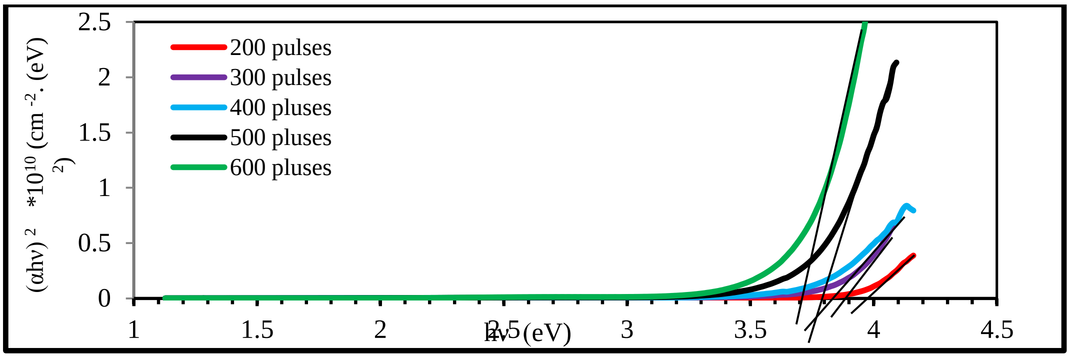


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Figure 8-(a, b and c): Variation of $(\alpha h\nu)^2$ as a function of hν for pulse laser for undoped In_2O_3 films at. (a) For RT, (b) annealing523 K, (C) annealing 623 K

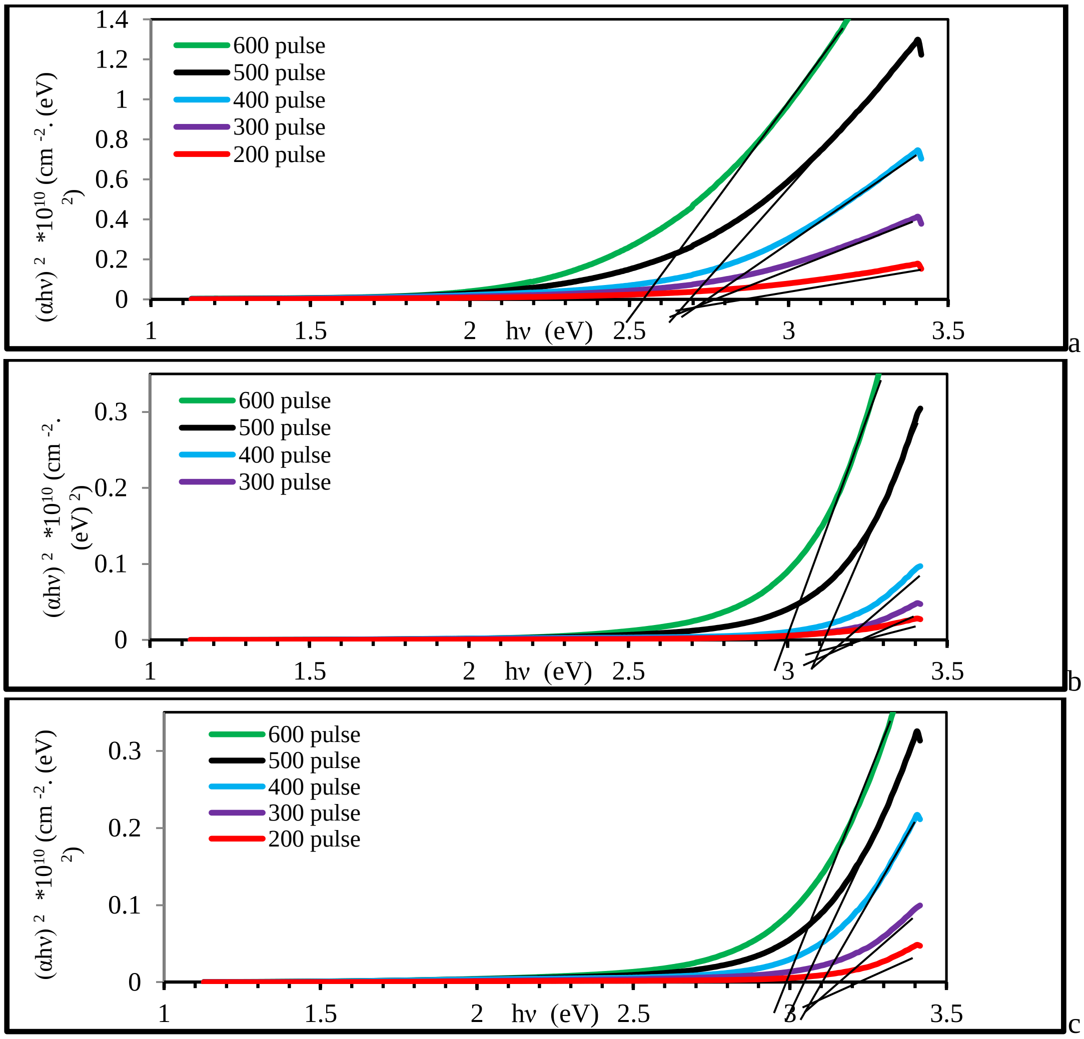

Figure 9-(a, b and c): Variation of $(\alpha h\nu)^2$ as a function of hν for pulse laser for doped In_2O_3: CdO films at. (a) For RT, (b) annealing523 K, (C) annealing 623 K

(α) is calculated using the equation, [16]

$\alpha = \ln (1/T) / d$ …………………………………………………………………………….. (2)

Where T is transmittance and d are film thickness. The absorption coefficient (α), and the incident photon energy (hν), are related by the following equation [17]:

$(\alpha h\nu)^2 = A (h\nu - E_g)$ ………………………………………………………………….. (3)

The scheme model of (αhν) 2 versus hν for In_2O_3 thin films with (200, 300,400,500 and 600) nm. Shoot deposited on glass substrate is shown in Figures- (7,8,9). It is observed that increase in no. shoot of laser lead to increase in the optical band gap from undoped 2.70eV to 3.95eV and doped 2.75eV to 3. 0eV.The results are in close agreement with [18].

Conclusions

In this work We have study the influence of no. Shoot of pulse laser in the characteristics of undoped In_2O_3, and doped In_2O_3: CdO thin films. It was found that pure In_2O_3 films pattern, in the case (RT), has amorphous structure. When the films annealed at 523 and 623 K temperatures the films crystallinity was improved and became polycrystalline structure with rhombohedral type and has preferred orientation along (222) direction. In_2O_3: CdO thin films mixed samples with 9 wt.% were polycrystalline with cubic structure with preferred orientation along (222) for In_2O_3 structure. The energy gabs All prepared films with increasing numbers of shoots, and increasing after annealing temperature

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