

# Effect of substrate temperature and laser wavelength on pulsed laser deposited CdTe thin films

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## Abstract

The effect of substrate temperature and laser wavelength on the laser deposition of CdTe thin films has been studied. CdTe thin films synthesized on glass substrates have been studied using X-ray spectral microanalysis, X-ray diffraction and scanning electron microscopy. It has been shown that the ratio of elements in the CdTe thin films depends on both the substrate temperature and the laser wavelength. Specific features of the crystalline structure, phase composition and structural parameters of the synthesized CdTe films depending on the deposition process conditions (substrate temperature 100–400 °C, laser wavelength 600–1200 nm) have been revealed. Specifically, CdTe thin films deposited using laser radiation of wavelengths of 600 and 1200 nm have a cubic crystalline structure, while CdTe films obtained at a laser wavelength of 1064 nm have either a hexagonal structure or are a mixture of cubic and hexagonal phases, depending on the substrate temperature. At a low substrate temperature (100 °C) the films crystallize to a hexagonal structure, whereas at higher temperatures (200, 300 and 400 °C) the films are a polycrystalline mixture of cubic and hexagonal CdTe phases, growing predominantly along the [111]C direction. It has been shown that CdTe thin films on glass substrates have similar morphologies but different thicknesses, regardless of deposition conditions.

## Keywords

CdTe, crystal structure, microstructure, pulsed laser deposition, thin films

## 1. Introduction

Due to their optical and electrical properties, CdTe thin films attract great attention both as light absorbing material for solar cells and for the fabrication of various purpose infrared photodetectors [1–3], including those

designed to determine coordinates, rotation angles, geometric dimensions and positions of objects [4].

CdTe has been used for the fabrication of ideal *p*-In-Sb/*n*-CdTe type heterojunctions and coordinate-sensitive infrared photodetectors, possessing high resolution, broad-band photosensitivity and high speed [5–7].

The CdTe compound is a direct band gap semiconductor with  $E_g \sim 1.5$  eV, has a high optical absorption coefficient ( $>10^5$  cm<sup>-1</sup>) and can exhibit *p*- or *n*-type conductivity [8, 9].

CdTe thin films are synthesized using various methods: electrochemical deposition [10, 11], vacuum deposition [12–15], atomic layer electrodeposition [12–16], pulsed laser deposition (PLD) [17–23] etc. However, most of the above methods require high synthesis temperatures (450–600 °C) for the formation of polycrystalline CdTe films, which can distort their stoichiometry [24]. For example, Te enrichment of the CdTe film composition can produce *p*-type conductivity, which is unacceptable for photodetector fabrication. However, the PLD technique allows high-precision control of the properties of the deposited material by adjusting the deposition conditions. Furthermore, that technique ensures a high degree of crystallinity of the deposited semiconductor films at relatively low substrate temperatures due to the high mobility of atoms and ions in the plasma, which is provided by the high energy induced by the laser radiation. Low synthesis temperatures also preclude the inter-diffusion of the components and favor the formation of a sharp heterojunction in the InSb/CdTe system.

There are but scarce literary data on the PLD synthesis of CdTe thin films [17–23]. Moreover, CdTe films can have a cubic or hexagonal crystalline structure or represent a polycrystalline mixture of both. The formation and content of the hexagonal CdTe phase in the films depends strongly on a number of factors: substrate material and temperature, gas pressure in the vacuum chamber, evaporation temperatures of the main material (CdTe) and the metal (Cd) etc. [25]. In [25], it was established that in the presence of a large excess of Te, only cubic crystals are formed during the growth of CdTe films, whereas near the stoichiometric composition and, possibly, with an excess of Cd, hexagonal crystallites appear. The authors also found that CdTe films with a hexagonal structure are obtained during vacuum evaporation only at substrate temperature above 200 °C, and with its further increase (up to 350–390 °C), the fraction of the hexagonal phase increases to 95–100%. PLD CdTe films synthesized at a laser wavelength of 355 nm and a substrate temperature of 20 °C had a wurtzite structure, whereas those synthesized at 200–500 °C had a cubic structure [18]. It was also reported [19] that PLD CdTe films synthesized at a laser wavelength of 1064 nm and substrate room temperature had a hexagonal structure, whereas those synthesized at higher temperatures (100–300 °C) had a cubic structure. The cited work also reported that PLD CdTe films deposited at room temperature using a Nd:YAG laser had typically a hexagonal structure, whereas those deposited at a higher temperature had a stable cubic phase. However, in [23], thin CdTe films, also obtained by the PLD method (laser radiation wavelength 248 nm) on a CdS substrate at substrate temperatures of 100–150 °C had a hexagonal structure with a small cubic phase fraction,

the proportion of which increases with a further increase in deposition temperature to 250 °C.

The aim of this work was to study the elemental composition, crystal structure and morphology of CdTe films depending on laser wavelength and substrate temperature during PLD.

## 2. Experimental

CdTe thin films were synthesized by PLD on glass substrates. The power source was a YAG:Nd<sup>3+</sup> laser having the wavelength  $\lambda = 1064$  nm (specimen series Nos. 4–7), Solid State Q-switched Laser AO-K-1202 (China) with a wavelength of 1200 nm (specimen series Nos. 8–10) and 600 nm (second harmonic, specimen series Nos. 1–3). For all the test specimens, the working pressure in the chamber was maintained constant,  $5.3 \cdot 10^{-3}$  Pa. The radiation intensity incident on the CdTe target was about  $2 \cdot 10^8$  W/cm<sup>2</sup>. The length and energy of the laser pulses were 30 ns and 0.35 J, respectively. The laser pulse frequency was 0.16 Hz, their total number during the film deposition process being constant (200). The substrates were ~1 mm thick polished glass wafers placed at a 30 mm distance from the CdTe target. Before deposition, the substrates were heated to 100 °C (specimen series Nos. 1, 4, 8), 200 °C (specimen series Nos. 2, 5, 9), 300 °C (specimen series Nos. 3, 6, 10) and 400 °C (specimen series No. 7). For CdTe films deposited at a laser wavelength of 1064 nm, additional synthesis was carried out at a substrate temperature of 400 °C in accordance with the results of [19], where a phase transition of CdTe from a hexagonal to a stable cubic crystalline structure was observed after the substrate temperature was increased from room temperature to 100–300 °C if a Nd:YAG laser was used. All the synthesized CdTe films had *n*-type conductivity.

The elemental composition of the CdTe thin films was studied using X-ray spectral microanalysis (XSM) under a ZEISSEVO 10 electron microscope with an AZtecLive Advanced attachment and Ultim Max 40 (measurement accuracy 0.1%). Micrographs of the films were obtained using a scanning electron microscope S-4800 (Hitachi). X-ray diffraction (XRD) studies of the CdTe films were carried out on a DRON-3M diffractometer in CuK $\alpha$  radiation ( $\lambda = 0.15406$  nm). The optical components of the diffractometer were as follows: soler, slot 1:8:0.25, graphite mono-chromator and FEU-85 detector. The measurement scheme was Bragg–Brentano. The phase composition of the films was studied by comparing the experimental data against Powder Diffraction File (PDF) standard databases of the International Centre for Diffraction Data (ICDD). The unit cell parameters were calculated using the FullProf software based on the Rietveld multiprofile analysis of X-ray diffraction patterns [26]. Furthermore, the size *D* of coherent scattering regions (CSR) and the mechanical stress in the unit cell ( $\epsilon$ ) were calculated using Eqs. (1) and (2), respectively [27].

$$D = 0.9\lambda/\beta \cos\theta, \quad (1)$$

where  $\beta$  is the full width at half maximum (FWHM, rad) taking into account experimental broadening (0.02 deg);  $\theta$  is the diffraction angle;  $\lambda$  is the diffractometer wavelength (0.15406 nm). The CSR size calculation error was  $\pm 1$  nm.

$$\varepsilon = (a - a_0)/a_0 \times 100\%, \quad (2)$$

where  $a_0$  is the unit cell parameter borrowed from the Powder Diffraction File database for CdTe;  $a$  is the unit cell parameter of the CdTe films synthesized in this work.

### 3. Results and discussion

#### 3.1. Elemental composition of CdTe thin films

Figure 1 shows a typical XSM spectrum of PLD CdTe thin films synthesized at a constant substrate temperature of 300 °C and a laser wavelength of 1200 nm. For the CdTe films deposited at other temperatures (100, 200 and 400 °C) and laser wavelengths (600 and 1064 nm), similar XSM spectra were obtained which did not differ fundamentally from those shown in Fig. 1. All the XSM spectra exhibit peaks of the Cd and Te elements, suggesting the presence of those elements on the surface of the glass substrates. Analysis of all film samples indicates the presence of foreign elements within the sensitivity limits of the method. The most intense peak at 1.7 keV corresponds to Si, due to the use of a glass substrate. The remaining low-intensity peaks also belong to elements contained in the substrate material (Na, Ca, Mg).

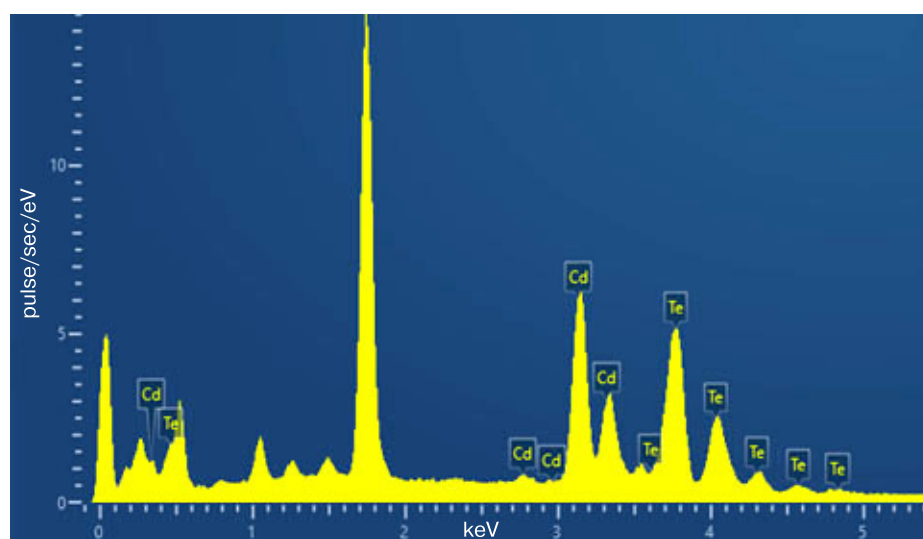
Table 1 summarizes data on the elemental composition of the CdTe films depending on the substrate temperature and laser wavelength. The data suggest that changing the

substrate temperature from 100 to 400 °C increases the Cd/Te elemental ratio in the films, regardless of the laser wavelength used, i.e., 600, 1064 or 1200 nm. For  $\lambda = 1064$  nm, the CdTe films exhibit the most intense increase in the Cd/Te ratio as compared with the films deposited at 600 or 1200 nm. A similar effect of the synthesis temperature on the elemental ratio in the CdTe films was observed earlier [15, 19, 28], where a decrease in the Te content with an increase in the synthesis temperature was attributed to the re-evaporation of Te atoms from the growing film surface.

As can be seen from the data presented in Table 1, an increase in the laser wavelength at a constant substrate temperature of 100 °C does not lead to any significant changes in the Cd/Te ratio of the films (measurement accuracy 0.1%). However, at substrate temperatures of 200 and 300 °C, an increase in the laser wavelength from 600 to 1200 nm initially increased the Cd/Te ratio and then reduced it.

The XSM data show that the composition of the CdTe thin films deposited at  $\lambda = 600$  and 1200 nm is enriched with tellurium ( $\text{Cd/Te} < 1.0$ ) and hence depleted of cadmium. However, for 1064 nm laser wavelength, the composition of the CdTe films deposited at 100 °C was also enriched with tellurium, but at higher temperatures (200, 300 and 400 °C) it was, on the contrary, enriched with cadmium ( $\text{Cd/Te} = 1.03\text{--}1.46$ ). The CdTe thin films deposited at substrate temperatures of 200 and 300 °C at laser wavelengths of 1064 and 1200 nm had the closest to stoichiometry elemental ratio ( $\text{Cd/Te} \approx 1.0$ ).

It is well known that CdTe compound growth exhibits congruent sublimation and condensation [29, 30], allowing a bulk stoichiometric CdTe source (in this work, a crystalline CdTe target) to condense to a film having a close-to-stoichiometry composition. Furthermore, for unbalanced supply of Cd and/or Te to the surface of the growing film, supplied atoms that are not involved in the formation of the CdTe layer re-evaporate, thus precluding



**Figure 1.** Typical XSM spectrum of CdTe thin films, synthesized by pulsed laser deposition on glass substrates (shown for specimen series No. 10, substrate temperature of 300 °C and laser wavelength 1200 nm)

**Table 1.** Elemental composition of PLD CdTe thin films depending on laser wavelength and substrate temperature

| Specimen<br>Series No. | $\lambda$ (nm) | $T$ (°C) | Elemental composition |       |       |       | Cd/Te ratio |        |
|------------------------|----------------|----------|-----------------------|-------|-------|-------|-------------|--------|
|                        |                |          | Cd                    |       | Te    |       |             |        |
|                        |                |          | at. %                 | wt. % | at. % | wt. % | Atomic      | Weight |
| 1                      | 600            | 100      | 47.9                  | 44.8  | 52.1  | 55.2  | 0.92        | 0.81   |
| 2                      |                | 200      | 48.1                  | 45.0  | 51.9  | 55.0  | 0.93        | 0.82   |
| 3                      |                | 300      | 48.7                  | 45.5  | 51.3  | 54.5  | 0.95        | 0.83   |
| 4                      | 1064           | 100      | 48.1                  | 44.9  | 51.9  | 55.1  | 0.93        | 0.81   |
| 5                      |                | 200      | 50.7                  | 47.5  | 49.3  | 52.5  | 1.03        | 0.90   |
| 6                      |                | 300      | 58.4                  | 55.2  | 41.6  | 44.7  | 1.40        | 1.23   |
| 7                      |                | 400      | 59.3                  | 56.3  | 40.6  | 43.7  | 1.46        | 1.29   |
| 8                      | 1200           | 100      | 47.8                  | 44.7  | 52.2  | 55.3  | 0.92        | 0.81   |
| 9                      |                | 200      | 48.7                  | 45.5  | 51.3  | 54.5  | 0.95        | 0.83   |
| 10                     |                | 300      | 49.7                  | 46.5  | 50.3  | 53.5  | 0.97        | 0.86   |

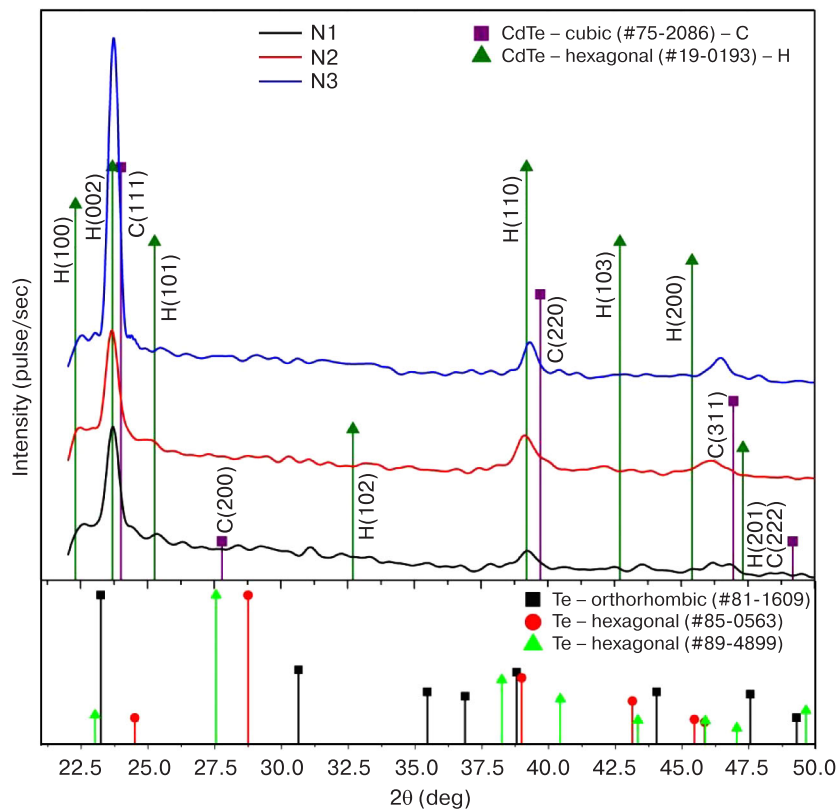
the formation of impurity Cd and Te phases [30], since the Cd (or Te) vapor pressure above the pure Cd (or Te) surface is far higher than the Cd or Te vapor pressure above the CdTe surface [30].

However, at a significantly excess pressure of Cd or Te and/or if the crystal growth rate is higher than the element evaporation rate, elemental inclusions can form [29, 30]. The above factors are in agreement with the observed change in the elemental ratio in the CdTe films

synthesized in this work with an increase in the substrate temperature and a change in the laser wavelength (see Table 1).

### 3.2. X-ray diffraction of CdTe thin films

According to the phase diagram, the single-phase compound CdTe exists with a very close ratio of elements to stoichiometry, but may suffer from a small deficiency of

**Figure 2.** X-ray diffraction patterns of CdTe thin films synthesized at laser wavelength of 600 nm at different substrate temperatures (100, 200 and 300 °C), in comparison with Powder Diffraction File (PDF) ICDD database

Cd or Te [29, 30]. In [30], it was noted that the maximum non-stoichiometry that can be maintained by single-phase CdTe at thermodynamic equilibrium is about 0.01%.

This value is significantly lower than the deviation observed in the composition of CdTe films deposited in this work (see Table 1). Taking this into account, it can be assumed that in films of series Nos. 1–4, 8–10, the coexistence of an impurity phase of Te with the main phase of CdTe is possible, while in films of series Nos. 5–7, the coexistence of a Cd phase is possible. Furthermore, as noted above, the CdTe compound crystallizes in a cubic or hexagonal structure, and the coexistence of these phases or a hexagonal to a cubic phase transition can occur.

Figure 2 shows X-ray diffraction patterns of PLD CdTe thin films synthesized at different substrate temperatures (100, 200 and 300 °C) at a 600 nm laser wavelength. In all the diffraction patterns, the diffraction peaks at  $2\theta \approx 23.7^\circ$ ,  $39.5^\circ$  and  $46.5^\circ$  are typical of the cubic CdTe structure, space group  $F43m$  (PDF#75-2086), and correspond to the (111), (220) and (311) diffraction planes, respectively.

Comparison between the diffraction patterns for the cubic (PDF#75-2086) and hexagonal (PDF#19-0193) CdTe structures, shown in Fig. 2, suggests that the diffraction peaks of the cubic CdTe phase (C) are partially superimposed with the hexagonal CdTe phase peaks (H). The intense peak near  $2\theta \approx 23.7^\circ$  can be attributed to either (111)C or (002)H, since their positions in the XRD patterns differ by only  $0.319^\circ$ . Furthermore, the low-intensity diffraction peaks at  $2\theta \approx 39.5^\circ$  and  $46.5^\circ$  can also be attributed to the (220)C/(110)H and (311)C/(201)H diffraction planes of the cubic or hexagonal CdTe phases, respectively. However, the X-ray diffraction patterns did not contain strong (110)H diffraction peaks at  $2\theta = 39.222^\circ$  typical of the hexagonal CdTe phase, the intensity of which is comparable to that of the (002)H peak as well as the strong (100)H, (101)H, (103)H and (200)H peaks at  $2\theta = 22.319^\circ$ ,  $25.281^\circ$ ,  $42.717^\circ$  and  $45.425^\circ$ , respectively (PDF#19-0193). In particular, based on the broadening of the diffraction peak near  $2\theta \approx 39.5^\circ$  for the films of sample series No. 2, the presence of the hexagonal phase of CdTe along with the cubic phase is difficult to confirm, due to, as noted above, the absence of the characteristic intense peaks (100)H and (101)H. In this case, the peak (111)C in all the obtained XRD patterns undergoes a shift towards the region of smaller angles compared to the data of PDF#75-2086 by no more than  $2\theta \approx 0.37^\circ$  (Table 2). A small  $0.41^\circ$  shift of the CdTe diffraction peaks was reported [8], where the films were synthesized using the closed space sublimation technique. Taking the above into account, one can conclude that the PLD CdTe films synthesized at short laser wavelengths (600 nm) and substrate temperatures of 100, 200 and 300 °C had a cubic crystal structure. Furthermore, the CdTe films deposited under the above conditions were single-phase since the experimental XRD patterns did not contain peaks of an impurity Te phase as per the phase diagram [29, 30]. However, a small Te excess in the CdTe

films could cause the formation of an elemental Te phase in a small amount, in the form of inhomogeneous local inclusions (see Table 1).

The X-ray patterns of the films shown in Fig. 2 suggest that the intensity of the main (111)C peak and its full width at half maximum (FWHM) (Table 2) change but slightly with an increase in the process temperature from 100 to 200 °C, whereas with further increase in temperature to 300 °C the (111)C peak intensity increases, and its FWHM decreases. The coherent scattering region (CSR) size of CdTe increases slightly with an increase in temperature (see Table 2). This indicates an improvement in the degree of crystallinity of CdTe films with increasing temperature when using a laser with  $\lambda = 600$  nm. Furthermore, the improved crystallinity of the obtained CdTe films at 300 °C may be due to their composition being closer to stoichiometry (see Table 1).

Figure 3 shows XRD patterns of CdTe thin films obtained by the PLD method using a laser wavelength of 1064 nm at different substrate temperatures. CdTe films deposited at 100 °C crystallize in a hexagonal structure of the  $P63mc$  space group (PDF#19-0193). The XRD pattern of this sample contains peaks (002)H and (110)H at  $2\theta \approx 23.73^\circ$  and  $39.48^\circ$ , respectively, with close intensities, as well as peaks (100)H, (101)H at  $2\theta \approx 22.7^\circ$ ,  $25.1^\circ$ , respectively. The latter result does not agree with earlier data [18, 25], but agrees in part with other data [19, 23]. With further increase in temperature to 200 °C, the growing CdTe films were cubic, with a small content of the hexagonal CdTe phase, as confirmed by the low-intensity (100)H and (101)H peaks along with the main (111)C, (220)C and (311)C ones (see Table 2). There are literary

**Table 2.** Position and FWHM of main diffraction peak (111)C/(002)H and coherent scattering region size ( $D$ ) of PLD CdTe films synthesized at laser wavelength of 600 nm (Nos. 1–3), 1064 nm (Nos. 4–7) and 1200 nm (Nos. 8–10) and different substrate temperatures: 100 °C (Nos. 1–4), 200 °C (Nos. 2, 5, 9), 300 °C (Nos. 3, 6, 10) and 400 °C (No. 7)

| Specimen Series No. | $2\theta$ (deg) | FWHM (deg) | $D$ (nm) |
|---------------------|-----------------|------------|----------|
| 1                   | 23.70           | 0.37       | 23       |
| 2                   | 23.66           | 0.39       | 22       |
| 3                   | 23.73           | 0.27       | 32       |
| 4                   | 23.73           | 0.49       | 17       |
| 5                   | 23.82/23.89     | 0.23/0.36  | 39/24    |
| 6                   | 24.16/24.67     | 0.21/0.38  | 43/23    |
| 7                   | 23.27/24.49     | 0.53/0.88  | 16/9     |
| 8                   | 23.73           | 0.49       | 17       |
| 9                   | 23.81           | 0.19       | 48       |
| 10                  | 23.71           | 0.25       | 35       |
| PDF#75-2086 - C     | 24.026          | –          | –        |
| PDF#19-0193 - H     | 23.707          | –          | –        |

data on a hexagonal to cubic phase transition in PLD CdTe films synthesized at elevated substrate temperatures (100–250 °C) [21, 31]. It was noted [21] that the hexagonal CdTe phase is metastable, and its presence in deposited CdTe thin films is probably caused by the formation of a nanocrystalline material.

CdTe thin films synthesized at higher temperatures (300 and 400 °C) are polycrystalline mixtures of the cubic and hexagonal CdTe phases, growing predominantly along the [111]C direction. The weak (100)H and (101)H peaks along with the strong one at  $2\theta \approx 24.5^\circ$  corresponding to the (111)C and (002)H planes are present in the XRD patterns of film series Nos. 6 and 7. The diffraction patterns of the CdTe thin films synthesized at a 1064 laser wavelength do not indicate the presence of elemental Cd and Te phases. However, the latter can exist in the form of small local inhomogeneous inclusions.

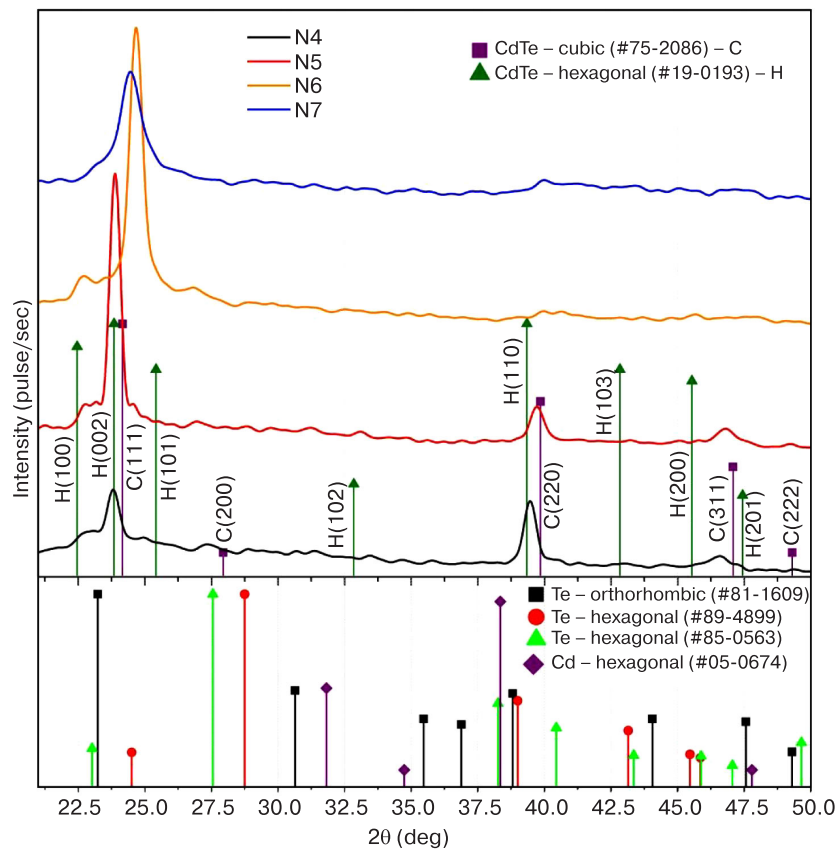
The films synthesized at  $\lambda = 1064$  nm, by analogy with those grown at  $\lambda = 600$  nm, do not show any clear regularity in the position of the main diffraction peak, its FWHM and CSR size with an increase in temperature (see Table 2). With an increase in the substrate temperature from 100 to 300 °C, the position of the main diffraction peak changes noticeably, shifting towards higher angles, and, at 400 °C, towards lower angles. The change in the FWHM of the peaks suggests an improvement of the crystallinity of the CdTe films with an increase in the substrate temperature from 100 to 300 °C and its further

degradation at 400 °C. The greatest CSR size of the deposited CdTe films was observed at a substrate temperature of 300 °C.

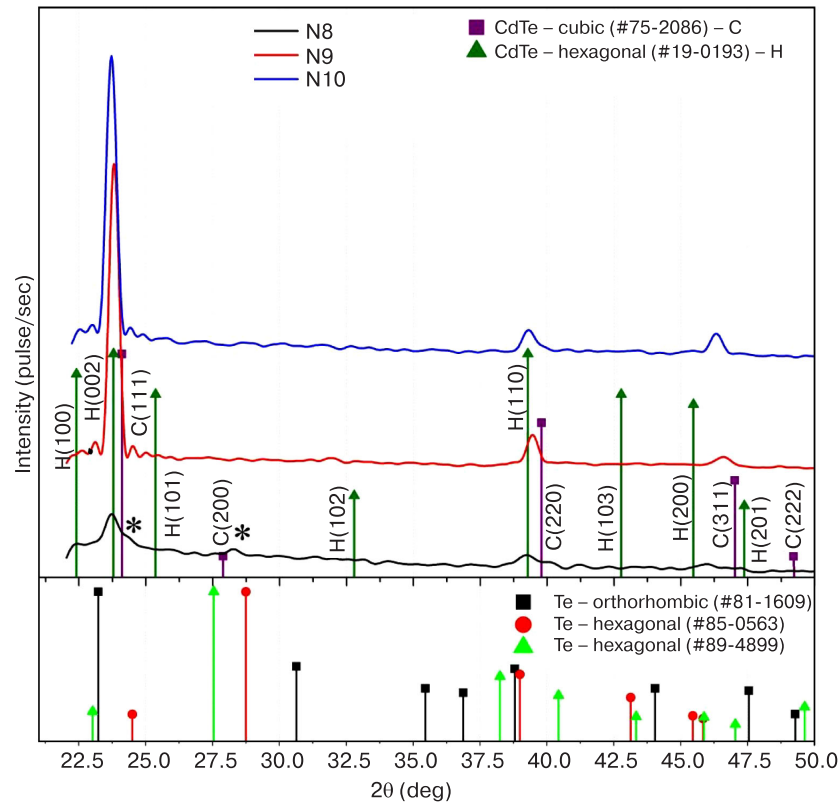
Figure 4 shows the XRD patterns of the CdTe thin films deposited at a laser wavelength of 1200 nm and different substrate temperatures (100, 200 and 300 °C). All those CdTe films, by analogy with those grown at a laser wavelength of 600 nm, had a cubic structure, space group  $F43m$  (PDF#75-2086). All the X-ray diffraction patterns contained intense (111)C and (220)C diffraction peaks, whereas the (311)C peak was only observed for specimen series Nos. 9 and 10.

CdTe films of series No. 8 exhibited a low intensity and large broadening of the main (111)C and (220)C diffraction peaks. Furthermore, the XRD pattern of the specimen series contained peaks at  $2\theta \approx 24.31$  and  $28.33^\circ$ , which, taking into account the broadening of the peak at  $2\theta \approx 39.3^\circ$ , can be attributed to the hexagonal Te phase (PDF#85-0563/89-4899). Noteworthy, the XRD patterns of the CdTe films (Figs. 2 and 3) having close compositions to that of specimen No. 8 (Table 1) did not contain the typical diffraction peaks of elemental Te. The latter fact can suggest, as noted above, the existence of an impurity tellurium phase in the form of inhomogeneous inclusions in Te-rich films [32].

The CdTe films synthesized at a laser wavelength of 1200 nm did not exhibit an increase in the intensity of the main (111)C peak with an increase in the substrate



**Figure 3.** X-ray diffraction patterns of CdTe films synthesized at laser wavelength of 1064 nm and different substrate temperatures (100, 200, 300 and 400 °C), in comparison with Powder Diffraction File (PDF) of ICDD database



**Figure 4.** X-ray diffraction pattern of CdTe films synthesized at laser wavelength of 1200 nm and different substrate temperatures (100, 200 and 300 °C), in comparison with Powder Diffraction File (PDF) of ICDD database

temperature, in contrast to the films deposited at a shorter laser wavelength (600 nm) and by analogy with the film specimens deposited at  $\lambda = 1064$  nm (Figs. 2–4). The main (111)C peak was shifted towards lower angles by  $0.22\text{--}0.32^\circ$ , and its FWHM initially decreased by more than two times and then grew slightly with an increase in the substrate temperature (see Table 2).

The CSR size initially increased from 17 to 48 nm and then decreased slightly to 35 nm. The calculated mean CSR size for all the series of synthesized CdTe films was in agreement with earlier data [32, 33], i.e.,  $24 \pm 1$  nm for vacuum-evaporated films, 21–22 nm for chemically deposited films and 79–125 nm for thermally evaporated films. In earlier work [32], the smaller CSR size as compared with the 79–125 nm was attributed to the CdTe film synthesis temperature used. The CdTe films in [32] were synthesized at an ambient temperature, whereas in another work [33], those were synthesized at  $\sim 397^\circ\text{C}$ . As can be seen from Table 2, larger CSR sizes are typical of higher synthesis temperatures except for the films synthesized at longer wavelengths (1064 and 1200 nm) at 400 and  $300^\circ\text{C}$ , respectively, possibly due to the degradation of the polycrystalline quality of the films or the formation of a polycrystalline mixture of the cubic and hexagonal CdTe phases.

Note that the X-ray diffraction patterns of the CdTe films do not exhibit any dependence on laser wavelength at a constant substrate temperature. However, both the substrate temperature and the laser wavelength exert

impact on the film growth. Therefore, a correct combination of the above process parameters in PLD is beneficial for the improvement of the structural properties of CdTe films.

Table 3 summarizes calculated unit cell parameters and mechanical stresses of the CdTe thin films, for the cubic and hexagonal modifications. It can be seen from the data that the unit cell parameters of the PLD CdTe films are in a good agreement with the Powder Diffraction File (PDF) data. The cubic CdTe films show a decrease in the unit cell parameters  $a$  with an increase in the substrate temperature, but in some cases the decrease is within the experimental error. The unit cell undergoes tensile stress, whose magnitude decreases with an increase in the substrate temperature, and hence the unit cell volume decreases. First of all, this may be due to a change in the Cd/Te elemental ratio in the films, i.e., a decrease in the Te content with an increase in the substrate temperature from 100 to  $300^\circ\text{C}$  (see Table 1). Similar observations were presented earlier in [8, 34]. Tellurium enrichment of the films leads to unit cell expansion due to a greater atomic radius of tellurium than that of cadmium [8, 34]. Therefore, a decrease in the tellurium content in the films leads to a decrease in the unit cell volume. For the CdTe films synthesized at a laser wavelength of 1064 nm, the dependence described above can be different, possibly, due to the formation of a tetragonal phase along with the main cubic one. With an increase in the substrate temperature, the cubic

**Table 3.** Structural parameters of PLD CdTe films synthesized at laser wavelengths of 600 nm (Nos. 1–3), 1064 nm (Nos. 4–7) and 1200 nm (Nos. 8–10) and for different substrate temperatures: 100, 200, 300 and 400 °C.

| Specimen Series No. | Unit cell parameters (nm) |                                                  | $V$ (nm <sup>3</sup> ) | $\varepsilon$ (%) |
|---------------------|---------------------------|--------------------------------------------------|------------------------|-------------------|
|                     | Cubic                     | Hexagonal                                        |                        |                   |
| 1                   | $a = 0.650 \pm 0.002$     | –                                                | 0.27463                | 1.40              |
| 2                   | $a = 0.649 \pm 0.002$     | –                                                | 0.27336                | 1.25              |
| 3                   | $a = 0.647 \pm 0.002$     | –                                                | 0.27084                | 0.94              |
| 4                   | –                         | $a = 0.442 \pm 0.004$ ,<br>$c = 0.752 \pm 0.001$ | 0.12723                | –                 |
| 5                   | $a = 0.644 \pm 0.001$     | $a = 0.453 \pm 0.001$ ,<br>$c = 0.748 \pm 0.001$ | 0.26709/0.13293        | –                 |
| 6                   | $a = 0.645 \pm 0.01$      | $a = 0.474 \pm 0.001$ ,<br>$c = 0.762 \pm 0.001$ | 0.26834/0.14827        | –                 |
| 7                   | $a = 0.649 \pm 0.001$     | $a = 0.458 \pm 0.001$ ,<br>$c = 0.757 \pm 0.002$ | 0.27336/0.13752        | –                 |
| 8                   | $a = 0.652 \pm 0.002$     | –                                                | 0.27717                | 1.70              |
| 9                   | $a = 0.648 \pm 0.002$     | –                                                | 0.27210                | 1.08              |
| 10                  | $a = 0.645 \pm 0.002$     | –                                                | 0.26834                | 0.66              |
| PDF#75-2086 - C     | $a = 0.641$               | –                                                | 0.26337                | –                 |
| PDF#19-0193- H      | –                         | $a = 0.458$ ,<br>$c = 0.750$                     | 0.13625                | –                 |

unit cell volume increases, whereas the volume of the hexagonal unit cell initially increases at below 300 °C and then decreases, with further increase in temperature. However, other origins of the tensile mechanical stress in the CdTe films are possible, such as linear and point defects caused by composition deviation from stoichiometry, grain boundaries (especially for nanocrystalline materials) and substrate material (glass).

### 3.3. Scanning electron microscopy of CdTe thin films

Table 4 shows the thicknesses of the CdTe as measured from SEM images. It can be seen that the thicknesses of the deposited CdTe films vary in a range of 102–401 nm, although the duration of film deposition process was the same in all cases. It is well known that the PLD growth of thin films includes several stages at which supplied particles either impact the substrate surface, or diffuse and start nucleation, or are rejected and desorbed. The above processes are controlled by the process temperature and deposition rate [35]. For example, at low substrate tem-

peratures the mobility of the film-forming complexes and the intensity of the nucleation process are lower, whereas at excessively high substrate temperatures, the deposition efficiency is lower due to the rejection of supplied molecules by evaporation. Moreover, the ablation threshold depends on the light absorption coefficient of the target material and hence the laser photon energy. For low photon flux and/or low proton absorption by the target at a given laser wavelength, laser pulses heat the target, and the outgoing material flux is controlled by the heat-induced evaporation of the target particles. In that case the evaporation fluxes outgoing from a multicomponent target will be controlled by the vapor pressures of the target constituent components.

It was reported [21] that the thickness of PLD CdTe thin films depends on substrate temperature and decreases with an increase in the substrate temperature due to the re-evaporation of particles from the growing film surface. However, the results of this work suggest another regularity of the effect of substrate temperature on the deposited CdTe layer thickness: it depends on laser wavelength.

**Table 4.** Thickness of PLD CdTe thin films as a function of laser wavelength and substrate temperature (specimen numbers are shown in parentheses)

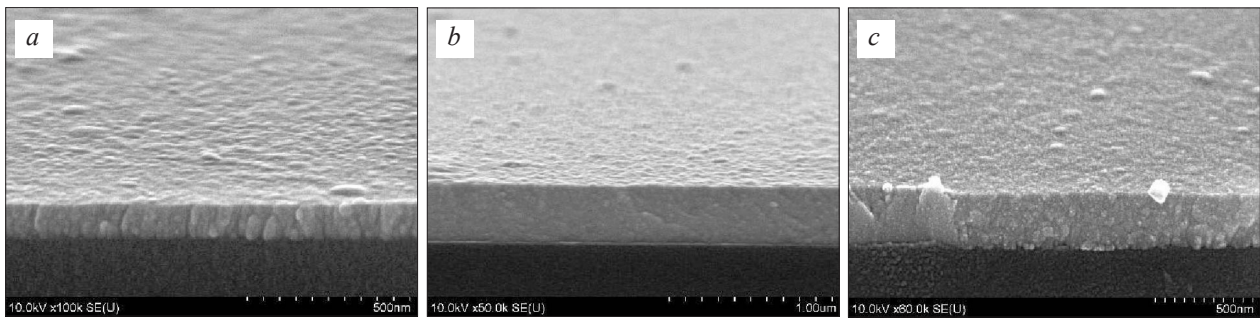
| Wavelength (nm) | Substrate temperature (°C) |                |                 |                |
|-----------------|----------------------------|----------------|-----------------|----------------|
|                 | 100                        | 200            | 300             | 400            |
| 600             | 102 nm (No. 1)             | 321 nm (No. 2) | 223 nm (No. 3)  | –              |
| 1064            | 324 nm (No. 4)             | 341 nm (No. 5) | 401 nm (No. 6)  | 141 nm (No. 7) |
| 1200            | 151 nm (No. 8)             | 173 nm (No. 9) | 204 nm (No. 10) | –              |

For example, at the laser wavelength  $\lambda = 600$  nm, an increase in the substrate temperature from 100 to 300 °C leads to an about 3-fold increase in the CdTe film thickness, followed by its decrease by approx. 1.5 times as the substrate temperature reaches 400 °C. For greater laser wavelengths, i.e.,  $\lambda = 1064$  and 1200 nm, the CdTe film thickness increases with the substrate temperature from 100 to 300 °C and then decreases dramatically at 400 °C. Furthermore, with an increase in the laser wavelength from 600 to 1200 nm at a constant substrate temperature, the thickness of all the CdTe films initially decreased and then increased. The data summarized in Table 4 suggest that the CdTe films deposited at a substrate temperature of 100 °C and at higher temperatures of 300–400 °C have the smallest thickness, regardless of the laser wavelength, except for Specimen No. 6 synthesized at  $\lambda = 1064$  nm and 300 °C. Moreover, the thickness of the deposited CdTe layer is the smallest if the shortest and longest laser wavelengths (600 and 1200 nm) are used. In both cases the smallest CdTe layer thickness is determined, as noted above, by the substrate temperature and the laser photon

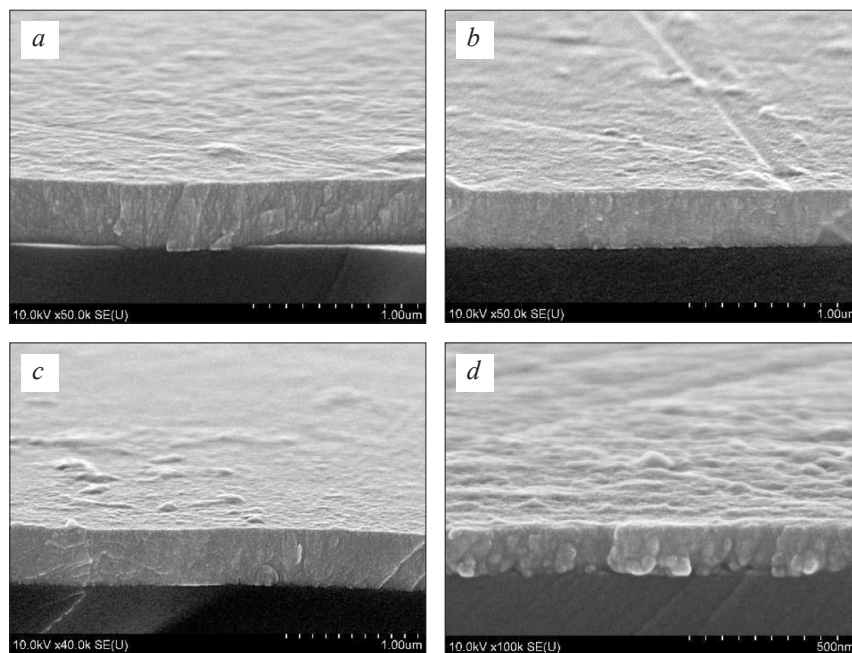
energy: if those are low and/or high, the nucleation intensity and the deposition intensity are lower, due to the rejection of molecules by evaporation and the re-evaporation of particles from the growing CdTe thin film surface [21, 35].

Figures 5–7 show SEM images of CdTe thin films on glass substrates synthesized at different laser wavelengths and substrate temperatures.

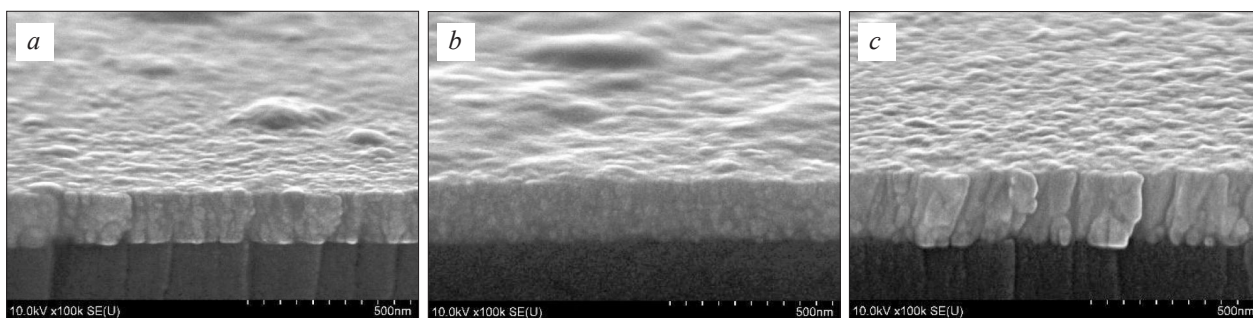
The above SEM images show that all the CdTe thin films deposited on glass substrates are in the form of layers of tightly packed nanometer-sized grains without visible voids, cracks or pores (Figs. 6–7). Noteworthy, the CdTe thin films with a thickness of more than 300 nm, i.e., No. 2 (600 nm, 200 °C), No. 4 (1064 nm, 100 °C), No. 3 (1064 nm, 200 °C) and No. 6 (1064 nm, 300 °C), have denser structures or grain packing than thinner films which typically have a “crumbly” structure with visible grain boundaries. The film specimens Nos. 1, 4, 7, 8, and 10 have columnar structures oriented along the film growth direction with a column width of about 20–70 nm. The difference in the morphology of the CdTe layers most



**Figure 5.** SEM images of CdTe thin films on glass substrates synthesized at laser wavelength of 600 nm and different substrate temperatures: (a) No. 1 (100 °C), (b) No. 2 (200 °C) and (c) No. 3 (300 °C)



**Figure 6.** SEM images of CdTe thin films on glass substrates synthesized at laser wavelength of 1064 nm and different substrate temperatures: (a) No. 4 (100 °C), (b) No. 5 (200 °C), (c) No. 6 (300 °C) and (d) No. 7 (400 °C)



**Figure 7.** SEM images of CdTe thin films on glass substrates synthesized at laser wavelength of 1200 nm and different substrate temperatures: (a) No. 8 (100 °C), (b) No. 9 (200 °C) and (c) No. 10 (300 °C)

likely is originated, as noted above, from the low/high synthesis temperature and a use of long-wave laser radiation. For the CdTe films synthesized at a 600 nm laser wavelength, an increase in temperature from 100 to 200 °C improves the grain packing in the film (Figs. 5a and 5b), but further growth in temperature to 300 °C, on the contrary, leads to structure degradation (Fig. 5c). A similar behavior is observed for the films grown at laser wavelengths of 1064 and 1200 nm (Figs. 6 and 7). If the laser wavelength changes, the films synthesized at substrate temperatures of 100 and 300 °C exhibit a similar regularity, except the series of specimens synthesized at 200 °C.

The surfaces of all the CdTe films, regardless of deposition conditions, are homogeneous and have a clear smooth texture. The inclusions on the surfaces of CdTe film specimens, e.g. Nos. 2 and 3 (see Fig. 3), probably originate from “splashing” which is typical of PLD films [9]. Unlike earlier results [9], this work did not show any clear relationship between the morphology of CdTe thin films and their deposition conditions. All the SEM images taken are in agreement with the mean CSR size calculations, additionally confirming the nanocrystalline nature of the synthesized CdTe thin films.

## Conclusion

CdTe thin films were synthesized using the PLD technique on glass substrates at different substrate temperatures and laser wavelengths. Analysis of their X-ray spectral and X-ray structural data showed the effect of film deposition process parameters on their structural parameters, as well as the elemental and phase composition. It was shown that an increase in the substrate temperature from 100 to 300/400 °C leads to an increase in the cadmium content, regardless of the laser wavelength used. A change in the laser wavelength from 600 to 1200 nm at a constant substrate temperature initially causes cadmium enrichment and then cadmium depletion of the film composition, this effect being more pronounced at higher temperatures (200 and 300 °C).

It was found that CdTe thin films synthesized at laser wavelengths of 600 and 1200 nm have a cubic crystal structure. CdTe films synthesized at a laser wavelength

of 1064 nm have either a hexagonal structure or are mixtures of the cubic and hexagonal phases, depending on the substrate temperature. At a low substrate temperature (100 °C) the films crystallized to a hexagonal structure, whereas at higher temperatures (200, 300 and 400 °C) they were polycrystalline mixtures of the cubic and hexagonal CdTe phases, growing predominantly along the [111]C direction. All the CdTe film specimens were single-phase, except the film synthesized at  $\lambda = 1200$  nm and a substrate temperature of 100 °C. It should be noted, however, that it is difficult to confirm the presence of elemental Cd and Te phases in other CdTe film specimen series due to the possible presence of their inhomogeneous local inclusions. The FWHM derived from the X-ray diffraction patterns allowed evaluating the crystallinity of the CdTe films depending on the substrate temperature and laser wavelength used. The mean CSR sizes calculated using the Scherrer formula depending on the film deposition process parameters were in the 9–48 nm range, suggesting their nanocrystalline nature. For the cubic CdTe films it was found that the unit cell parameter and volume decrease with increasing synthesis temperature. For two-phase CdTe films containing the hexagonal CdTe phase along with the cubic phase, the cubic unit cell volume increases, whereas the hexagonal unit cell volume initially increases and then decreases with an increase in the substrate temperature from 100 to 400 °C. In all cases, the crystal lattices of all the CdTe films undergo tensile mechanical stress, the magnitude of which decreases with an increase in temperature, which is probably caused by a change in the Cd/Te elemental ratio in the film composition (i.e., a decrease in the Te content), and depends on the substrate material.

All the CdTe thin films had uniform surfaces with a clear smooth texture and were in the form of layers of nanosized grains without visible voids, cracks or pores. It was shown that the thickness of the CdTe films deposited for the same duration varies in a 102–401 nm range depending on PLD mode, i.e., substrate temperature and laser photon energy. The structure and grain packing density of the CdTe layers differed depending on film thickness and hence PLD process conditions.

PLD CdTe thin films synthesized at a substrate temperature of 200 °C and a laser wavelength of 1064 nm have close-to-stoichiometry compositions, better

morphology and higher crystallinity in comparison with other specimens. The latter fact suggests that those

CdTe film synthesis conditions are optimum for the fabrication of thin-film solar cells and photodetectors.

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