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ОПТИЧЕСКИЕ СВОЙСТВА ТОНКОЙ ПЛЕНКИ CdTe, ПОЛУЧЕННОЙ МЕТОДОМ ВЫСОКОЧАСТОТНОГО МАГНЕТРОННОГО РАСПЫЛЕНИЯ

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Тонкие пленки теллурида кадмия (CdTe) относятся к соединениям $A^{II}B^{VI}$, демонстрируют полупроводниковые свойства и представляют собой важную область исследований из-за их широкого применения в различных оптоэлектронных устройствах. Солнечные элементы на основе CdTe привлекают внимание, поскольку CdTe характеризуется прямой энергетической запрещенной зоной E_g и высоким коэффициентом поглощения, что делает CdTe отличным светопоглощающим слоем солнечных элементов. Испарение материала в вакууме методом высокочастотного магнетронного распыления является одним из наиболее эффективных методов получения однородных пленок. Настоящая работа посвящена исследованию оптических свойств тонкой пленки CdTe, полученной на кварцевой подложке методом высокочастотного магнетронного распыления. Определены спектры оптического пропускания, отражения и μ-комбинационного рассеяния тонкой пленки CdTe. Линейность спектральной зависимости коэффициента оптического поглощения α тонкой пленки CdTe в координатах $(\alpha h\nu)^2$ *vs* $h\nu$ свидетельствует о прямом характере оптических переходов, соответствующих длинноволновому краю фундаментального поглощения. Оптическая ширина запрещенной зоны исследуемой тонкой пленки CdTe составляет E_g = 1,53 эВ. Пики экспериментальных спектров комбинационного рассеяния света при 121; 139; 142; 167 и 331 см$^{-1}$ приписываются фононам в кристаллических CdTe и Te.

Ключевые слова: пропускание; коэффициент отражения; спектры комбинационного рассеяния; запрещенная зона; тонкая пленка.

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OPTICAL PROPERTIES OF CdTe THIN FILM OBTAINED BY HIGH-FREQUENCY MAGNETRON SPUTTERING METHOD

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Cadmium telluride (CdTe) thin films relate to $A^{II}B^{VI}$ compounds and show semiconductor behaviour. They present an important research field because of their wide application in various optoelectronic devices. CdTe-based solar cells attract attention since CdTe is characterised by the direct energy bandgap E_g and high absorbance, which makes it an excellent light-absorbing layer of solar cells. Material evaporation in vacuum by using the high-frequency magnetron sputtering method is one of the most advantageous methods for obtaining uniform films. The present work is dedicated to the investigation of the optical properties of CdTe thin film, which is produced on quarts substrate by the high-frequency magnetro sputtering method. The optical transmission, reflectivity, and μ-Raman spectra of the CdTe thin film have been determined. Linearity of the spectral dependence of the coefficient of optical absorption α of CdTe thin film in the coordinates $(\alpha h\nu)^2$ *vs* $h\nu$ indicates for the direct character of optical transitions corresponding to the long-wavelength edge of fundamental absorption. The optical bandgap of the studied CdTe thin film is found to be $E_g = 1.53$ eV. The peaks of the experimental μ-Raman spectra at 121; 139; 142; 167 and 331 cm^{-1} are attributed to the phonons in crystalline CdTe and Te.

Keywords: transmission; reflectivity; Raman spectra; bandgap; thin film.

Introduction

Cadmium telluride (CdTe) based solar cells is the one of leading technology of solar energy production because of its significant photovoltaic (PV) conversion efficiency, performance stability, low costs of fabrication and short payback time [1]. The last one was obtained by *First Solar* (USA)[1]. In recent years a significant increase of the PV conversion efficiencies up to 18.6 and 22.1 % of the PV modules and corresponding small area devices based on CdTe solar cells was taken place [2].

The short-circuit current density of the modern CdTe solar cells has already approached its theoretical limit [3]. According to the thermodynamic limit of Shockley – Queisser, the optimal band gap of the materials required for reach the maximum of theoretically calculated efficiency of solar cell for the air mass coefficient AM1.5 is equal to 1.34 eV [4; 5]. However, the value of band gap of CdTe is slightly larger (1.5 eV in [3]).

The aim of the present work is the investigation of the optical properties of CdTe thin film, which is produced on the quarts substrate by the high-frequency magnetron sputtering method. In this paper, we present results of investigation the transmission, reflection and Raman spectra. The bandgap, refractive index, high-frequency dielectric constant, static dielectric constant, electron effective mass and thickness of thin filmare determined from the measurements of transmittance spectra.

Experimental details

CdTe films were deposited on quarts substrates with a size of 16.0 × 8.0 × 1.1 mm by the method of high-frequency magnetron sputtering (13.6 MHz) using a VUP-5M vacuum station (*Selmi*, Ukraine) [6]. A single crystal disc of 99.999 % purity with a thickness of 2 mm and a diameter of 40 mm was used as a target. The target – substrate distance was 75 mm. The deposition time was 1200 s. The start and end of the process were controlled by means of a movable shutter.

Before the sputtering process, the chamber was evacuated. The gas pressure inside the chamber was $4 \cdot 10^{-4}$ Pa. This pressure is achievable with using the Polyphenyl ether (5Ф4Е) diffusion fluid in the vapor oil vacuum pump, which provides a low partial vapor pressure ($9 \cdot 10^{-7}$ Pa).

The sputtering was carried out at a pressure of argon (Ar) in the range of 1.0–1.3 Pa. The power of the HF magnetron was maintained at the level of 50 W and the temperature of the substrate at 489 K. For heating the substrates, a high-temperature tungsten heater with a power of 300 W was used. The temperature was controlled by means of a proportional – integral – derivative (PID) controller for controlling heating and cooling rates, as well as for ensuring the temperature conditions of deposition.

[1]First Solar achieves yet another cell conversion efficiency world record [Electronic resource]. 2016. URL: https://investor.firstsolar.com/news/press-release-details/2016/First-Solar-Achieves-Yet-Another-Cell-Conversion-Efficiency-World-Record/default.aspx (date of access: 10.01.2021).

The spectral dependence of the optical transmittance and reflectivity (Shimadzu UV-3600 (Japan)) of the obtained samples in the visible and near infrared regions (300–1500 nm) was studied at room temperature [6; 7].

Raman spectra were recorded by the micro-Raman (μ-Raman) method. The Raman spectra were performed with using a T64000 Jobin Yvon spectrometer (France) at the room temperature. The He – Ne laser ($\lambda = 633$ nm) and Ar laser ($\lambda = 514.532$ nm) were used as light excitation. In the measurements performed, the laser power waschosen between 1 and 250 mW, the exposure time wasregulated between 5 and 120 s, and the light spot diameter was ~1 μm.

Experimental results

Figures 1 and 2 show the transmission and reflection spectra of CdTe thin films – substrate combinations, respectively. The transmission and reflection coefficients strongly depend on the film structure, which is determined by the preparation methods, film thickness and deposition conditions.

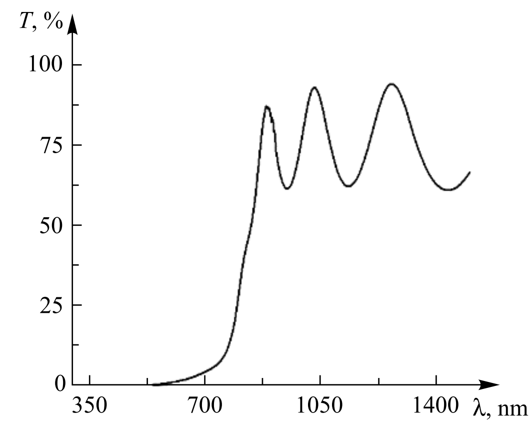

Fig. 1. Transmission spectrum of CdTe thin film

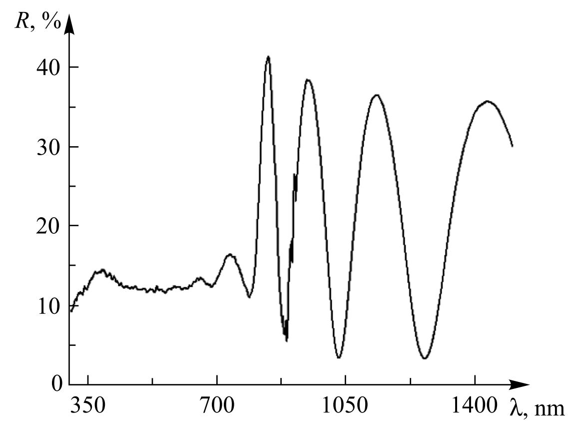

Fig. 2. Reflection spectrum of CdTe thin film

The transmission and reflection spectra of the thin film exhibit periodic peaks and minima associated with interference effects, indicating high structural perfection of the film. The surface roughness destroys the observed interference extremadue to the multiple reflections.

For each film studied the full transmittance and reflectivity spectra were calculated and the corresponding averaged values were evaluated using the relations (1)

$$\tilde{T} = \frac{1}{b-a}\int_a^b T d\lambda,\ \tilde{R} = \frac{1}{b-a}\int_a^b R d\lambda, \quad (1)$$

where $\tilde{T}$ and $\tilde{R}$ are the averaged transmittance and reflectivity in the wavelength range $a - b$ (see fig. 1 and 2). The obtained values $\tilde{T}$ and $\tilde{R}$ of the film in the wavelength range of 300–1500 nm are equal to $\tilde{T} = 43.14$ % and $\tilde{R} = 19.27$ %.

Clear interference maxima and minima in the wavelength range from 300 to 1500 nm are presented in fig. 1 and 2, the positions of which are depended by the film thickness.

To calculate the thickness d of the studied film, the following equation has been used,

$$d = \frac{M\lambda_1\lambda_2}{2\left(n(\lambda_1)\lambda_2 - n(\lambda_2)\lambda_1\right)}, \quad (2)$$

where λ_1 and λ_2 are the wavelengths corresponding to the neighbouring extremum points in the transmittance spectrum. Here, $M = 1$ for two neighbouring extrema of the one type (max – max, min – min) and $M = 0.5$ for two neighbouring extrema of the opposite types (max – min, min – max) [6]. The thickness of CdTe thin film calculated by equation (2) is equal to $d = 717.7$ nm.

The bandgap energy E_g was determined using different methods. In the first one, the bandgap energy corresponds to the wavelength position of the last maximum of the transmittance derivative $\frac{dT}{d\lambda}$ (fig. 3). This method gives satisfactory results, although it tends to underestimate slightly E_g value [8]. From the wavelength spectrum of $\frac{dT}{d\lambda}$ presented in fig. 3, it can be directly seen that the bandgap energy of CdTe thin films is 1.55 eV.

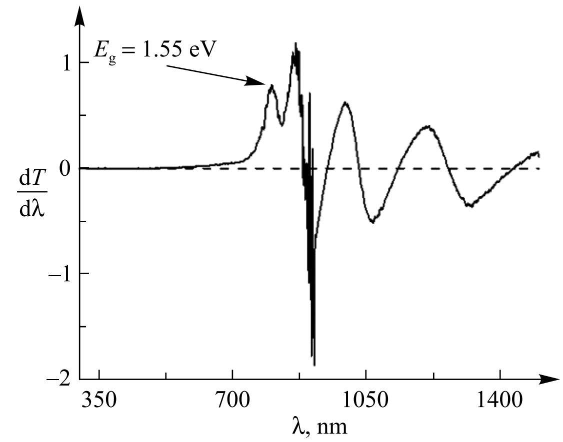

Fig. 3. Wavelength spectrum of the transmittance derivative $\frac{dT}{d\lambda}$ of CdTe film. The arrow indicates the maximum of $\frac{dT}{d\lambda}$, position of which determines the optical bandgap E_g

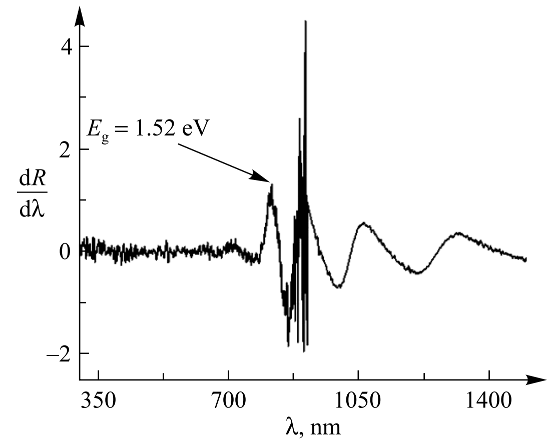

Fig. 4. Wavelength spectrum of the reflectance derivative $\frac{dR}{d\lambda}$ of CdTe film. The arrow indicates the maximum of $\frac{dR}{d\lambda}$, position of which determines the optical bandgap E_g

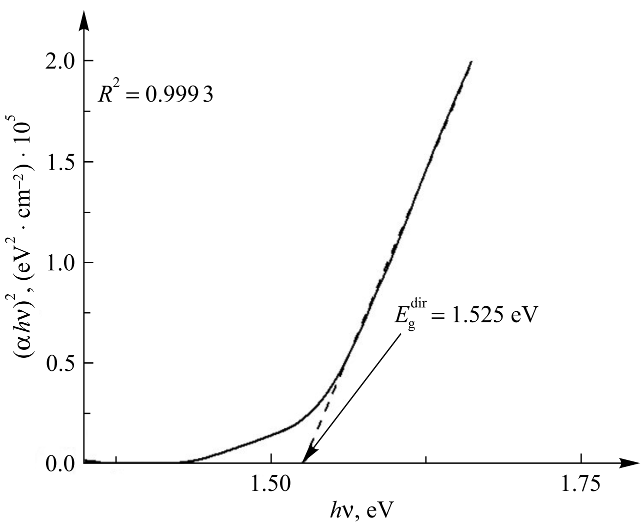

Fig. 5. Absorption coefficient α in presentation of $(\alpha h\nu)^2$ as function of photon energy $h\nu$: R^2 is the coefficient of determination; E_g^{dir} is the direct bandgap

In the second method, the bandgap E_g is taken as a position of the last maximum of the reflectivity derivative $\frac{dR}{d\lambda}$ spectrum (see fig. 4). The obtained values E_g = 1.52 eV is a bit smaller than that obtained on the basis of the transmittance spectrum E_g = 1.55 eV (see fig. 3).

One more method of the bandgap determination is based on the analysis of the absorption spectrum of a film at the long-wavelength edge of fundamental absorption [7]. Here, the spectral dependence of the optical absorption of CdTe thin film in the $(\alpha h\nu)^2$ *vs* $h\nu$ (α is the absorption coefficient and $h\nu$ is the photon energy) coordinates (see fig. 5) is approximated satisfactorily by the straight line. Extrapolation of the linear segment of the $(\alpha h\nu)^2$ curve until intersection with the axis of photon energyhν permits to obtain the optical bandgap of CdTe film as E_g = 1.53 eV, which is close to the values E_g = 1.55 eV and E_g = 1.52 eV obtained by using the previous methods.

The observed linear behaviour of the dependence presented in fig. 5 in the range of 1.55–1.67 eV indicates that here, the direct interband optical transitions form the long-wavelength absorption edge of CdTe film.

Now we proceed to the refractive index, which is a fundamental optical parameter related to the band structure of optoelectronic materials. A number of empirical relations between therefractive index n and the bandgap E_g are known from the literature. The most often used are the relations suggested by Moss [9], Ravindra [10], Herve and Vandamme [11], and Tripathy [12]. According to these models, the refractive index n of a semiconductor can be calculated correspondingly as

$$n_M^4 E_g = C,\ n_R = a - bE_g,\ n_{H-V}^2 = 1 + \left[\frac{A}{E_g + B}\right]^2,\ n_T = n_0\left[1 + \alpha e^{-\beta E_g}\right], \tag{3}$$

where the normal conditions are assumed, the bandgap units are eV, and the n's subscripts abbreviate the authors mentioned above. In equation (3), we have C = 95 eV, a = 4.084 eV, b = 0.62 eV^{-1}, A = 13.6 eV, B = 3.47 eV, n_0 = 1.73, α = 1.901 7 and β = 0.539 eV^{-1} [10–13]. Depending on the material studied, these different empirical relations (3) may be applied to the experimental values n and E_g more or less successfully [13].

Using different models given by equation (3), we have calculated the refractive indices n of CdTe thin film, which correspond to the determined bandgaps E_g (table 1).The refractive index n = 2.682 0 (λ = 600 nm, T = 300 K) was obtained in [14]. Comparing the obtained results (see table 1) with the literature data (n = 2.682 0, λ = 600 nm, T = 300 K) [14], one may assume that the Moss relation between E_g and n is the most appropriate for CdTe film studied.

The high-frequency dielectric constant ε_∞ in table 1 has been calculated on the basis of the refractive index n, as $\varepsilon_\infty = n^2$.

Table 1

Optical parameters obtained from results of experimental studies of transmission and reflection spectra

Parameters			Methods of determination		
			$dT/d\lambda$	$dR/d\lambda$	$(\alpha h\nu)^2$ vs $h\nu$
Value of bandgap, eV			1.55	1.52	1.53
ε_0			9.78	9.85	9.85
$\frac{m^*}{m_e}$			0.18	0.19	0.19
Parameters	Methods of bandgap determination	Moss relation	Ravindra relation	Herve and Vandamme relation	Tripathy relation
n	$\frac{dT}{d\lambda}$	2.8	3.13	2.89	3.16
	$\frac{dR}{d\lambda}$	2.81	3.14	2.90	3.18
	$(\alpha h\nu)^2$ vs $h\nu$	2.81	3.14	2.90	3.17
ε_∞	$\frac{dT}{d\lambda}$	7.83	9.76	8.35	9.98
	$\frac{dR}{d\lambda}$	7.90	9.86	8.42	10.1
	$(\alpha h\nu)^2$ vs $h\nu$	7.89	9.85	8.41	10.09

The static dielectric constant (ε_0) of the CdTe thin film has been calculated using thepolynomial expansion of ε_0 by the bandgap E_g, valid for semiconductors (4) [15].

$$\varepsilon_0 = -33.268\,76 + 78.618\,05E_g - 45.707\,95E_g^2 + 8.324\,49E_g^3. \tag{4}$$

Comparing the obtained results of ε_∞ and ε_0 (see table 1) with the reference data for CdTe [16] (ε_∞ = 7.15 and ε_0 = 10.3), one may assume that the Moss relation (3) and the relation (4) reproduce satisfactorily the dielectric constants ε_∞ and ε_0 on the basis of the measured bandgap E_g.

The relation (5), which is similar to (4), may be used for expansion of the normalised electron effective mass $\frac{m^*}{m_e}$ by the bandgap E_g powers [15],

$$\frac{m^*}{m_e} = 5.170\,04 - 7.466\,99E_g + 3.632\,86E_g^2 - 0.575\,25E_g^3. \tag{5}$$

The values $\frac{m^*}{m_e}$ for CdTe thin film calculatedby using the relation (5) (see table 1) areapproximately twice larger than the corresponding reference value $\frac{m^*}{m_e} = 0.1$ [15].

The experimental Raman spectra of CdTe thin film are presented in fig. 6 and 7. Here, four phonon modes may be selected. Laser excitation with the wavelength of 633 nm, which is close to the bandgap of CdTe thin film, leads to the appearence of TO (Cd – Te), LO_1 (Cd – Te) and LO_2 (Cd – Te) phonon modes at 139; 167 and 331 cm^{-1}, respectively (table 2). Tellurium modes A (Te – Te) and E (Te – Te) are most clearly observed on the background of Cd – Te modes when the green laser excitation is realised, which heats the sample studied (see fig. 7, curve *2*). It is known that the crystalline Te shows Raman peaks at the phonon modes 90 cm^{-1} (E mode), 121 cm^{-1} (A mode) and 142 cm^{-1} (E mode), and that the cross section of Raman scattering is about 75 times larger than that in CdTe [16] at the same excitation conditions. The presence of mode A (Te – Te) is an indicator of Te inclusions in the CdTe polycrystalline film. The form of these inclusions (in the clusters or thin films) and their possible location (on the surface or in the volume of CdTe thin films) remain unclear.

Table 2

Characterisation of Raman spectra

Peak position, cm^{-1}	121	139	142	167	331
Phonon mode	A	TO	E	LO_1	LO_2
Compound	Te	CdTe	Te	CdTe	CdTe

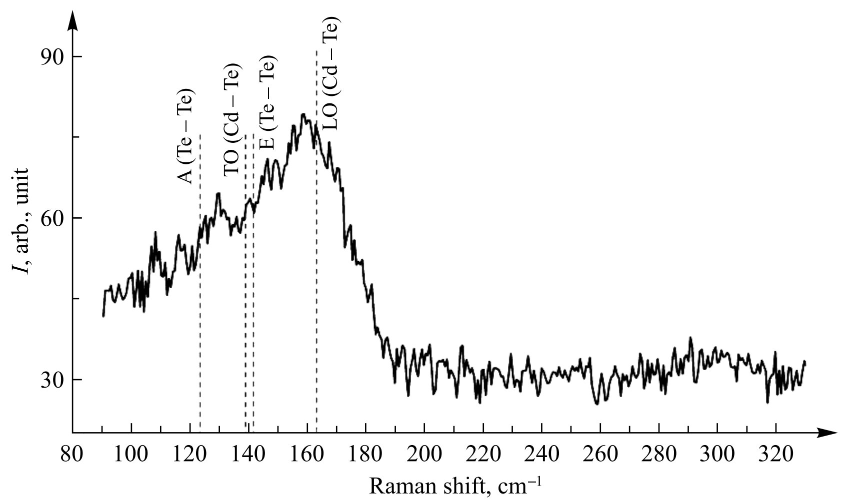

Fig. 6. Raman spectra of CdTe thin film: laser excitation wavelength is $\lambda = 514.532$ nm; laser power is 1 mW; the exposure time is 60 s

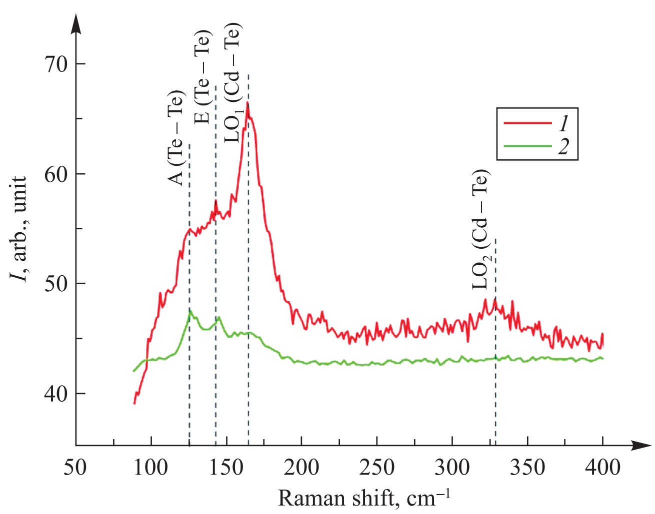

Fig. 7. Raman spectra of CdTe thin film: laser excitation wavelengths are $\lambda = 633$ nm (curve *1*) and 514.532 nm (curve *2*); laser power is 3 mW; the exposure time is 30 s

Conclusion

CdTe thin films were deposited onto quarts substrate by the HF magnetron sputtering method. The fundamental absorption edge position in the transmittance spectrum of studied thin films corresponds to the values that are typical for CdTe compound. The film thickness of $d = 717.7$ nm was obtained by optical interference analysis of the transmittance spectrum $T(\lambda)$ of CdTe films. The value of optical bandgap E_g obtained for CdTe by different methods was identified to be between 1.52 and 1.55 eV.

Based on the obtained experimental data of the bandgap of CdTe, the refractive index, high-frequency dielectric constant, static dielectric constant and electron effective mass were calculated. It is established that in the case of CdTe film the Moss relation allows determining the values of refractive index and high-frequency dielectric constant most adequately.

Raman spectra show phonon modes at 139; 167 and 331 cm^{-1}, which confirm the formation of CdTe phase. The phonon peaks observed at 90 and 121 cm^{-1} correspond to the crystalline tellurium.

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