# Heterolayers of Hexagonal *a*-CdTe

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The importance of the direct band structure of CdTe as a necessary condition for the creation of highperformance photoelectronic devices based on it is argued. The possibility of obtaining heterolayers (HLs) of cadmium tellurides by the method of isovalent substitution is shown. Prerequisites for manufacturing are determined and technological modes of isothermal annealing of  $\alpha$ -CdSe and  $\alpha$ -CdS base crystals are developed. For the first time, HLs with a little-used atypical hexagonal modification  $\alpha$ -CdTe crystal lattice with highly stable properties are obtained by isovalent substitution. A technique is developed, and the basic parameters of the band structure and luminescence characteristics are studied by the method of  $\lambda$ -modulation of optical irradiation. The band gap  $E_g = 1.56$  eV is established, and the parameters of the band structure are determined for the first time, namely, the subbands of the valence band split off due to the action of the crystal field  $\Delta_{cr} \approx 0.06$  eV and the spin-orbit interaction  $\Delta_{so} \approx 0.38$  eV. A high quantum efficiency  $\eta \approx 7.9$  % of the dominant radiation in the edge region is established. It is determined by doping with isovalent impurities, which are the atoms of the base substrate. An important prerequisite for obtaining high intensity (compared to CdTe crystals with  $\eta \sim 0.1-0.2$  %) is the formation of a transition layer of the corresponding solid solution between the components of heterostructures, which significantly reduces the concentration of defects and the discrepancy between the crystalline and thermal parameters of the contacting materials. The obtained features of the crystal structure provide the conditions for the formation of effective luminescence, the basic mechanisms of which are interband radiative transitions and the dominant annihilation of excitons bound on isovalent impurities.

Keywords: Cadmium telluride, Hexagonal structure, Isovalent atoms, High quantum radiation intensity.

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#### 1. INTRODUCTION

One of the basic materials for devices of modern functional electronics is cadmium telluride [1, 2]. Based on it, a number of different devices have been obtained, the properties and characteristics of which are determined precisely by the physical and technical parameters of the material used. The direct band structure of cadmium telluride in combination with a sufficiently large band gap  $(E_g = 1.50 \text{ eV})$  is an important condition for obtaining a material with a high radiation efficiency in the near infrared region [2, 3]. It should be noted that, on its basis, highly efficient converters of solar energy into electrical energy have been obtained and such detectors are widely used in various optoelectronic systems [4, 5]. At the same time, the problem of obtaining highly efficient radiation sources, especially on tellurides of the atypical hexagonal modification  $\alpha$ -CdTe, is important and relevant. In this case, an important task of photoelectronics is the combination of emitters and photodetectors in various optoelectronic systems based on  $\alpha$ -CdTe. For them, the possibility of varying the type of electrical conductivity in manufactured devices is important. However, the quantum efficiency of emitters based on it is significantly low due to nonradiative recombination processes due to deep energy levels [6]. They are formed by intrinsic point defects (IPD) of the structure and the process of their formation is poorly controlled [7, 8]. Therefore, to obtain a high efficiency of radiation generation, it is important to find a method for obtaining layers and heterostructures of the base material and simultaneously dope it with an appropriate type of impurities [9].

# 2. EXPERIMENT

## 2.1 Samples Under Study

Heterolayers (HLs) were obtained by isothermal annealing in vapors of isovalent elements [10]. Under the appropriate regimes, atoms of the same valency were replaced in the base substance, which led to the name of the method as isovalent substitution (IVS) [11]. It becomes especially effective in diffusion processes using the closed pipe method. Preliminarily, the base substrates of crystals of II-VI compounds underwent known processes of chemical-mechanical treatment, loaded into quartz ampoules, which were evacuated to no worse than  $10^{-4}$  Torr and sealed off. In them, the substrate of the base material and the canopy of the corresponding diffuser were located at opposite edges. Isothermal annealing was carried out with a suitable location of the ampoule in the furnace, Fig. 1.

Isothermal annealing of the base crystals  $\alpha$ -CdSe was carried out at  $T_a = 800$  °C in a saturated vapor of an isovalent (up to Se) element Te. Equilibrium conditions were obtained, according to which the diffusion of the isovalent impurity (IVI) occurs and the atoms of the base substance of the same valency are replaced. A new chemical compound  $\alpha$ -CdTe is formed on the surface of  $\alpha$ -CdSe according to the reaction [10, 11]

$$\alpha - \mathrm{CdSer}_{\mathrm{s}} + \mathrm{Te}_{\mathrm{G}} \to \alpha - \mathrm{CdTe}_{\mathrm{Ts}} + \mathrm{Se}_{\mathrm{G}}, \qquad (2.1)$$

where indices  $"T_s"$  and "G" correspond to the solid and gaseous states of the reagents, respectively.

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Fig. 1 – Temperature distribution and position of ampoules during the production of diffusion layers: 1 - substrate of the initial material, 2 - sample

In the case of the  $\alpha$ -CdSe base compound, an  $\alpha$ -CdTe/  $\alpha$ -CdSe heterostructure (HS) with time-stable properties is formed. Annealing of hexagonal  $\alpha$ -CdS substrates in a pair of isovalent element Te was also carried out to obtain an  $\alpha$ -CdTe/ $\alpha$ -CdS HS by the reaction

$$\alpha - CdS_{Ts} + Te_G \rightarrow \alpha - CdTe_{Ts} + S_G.$$
(2.2)

The characteristic light-yellow color of the base substance made it possible to detect the boundary between it and the resulting dark HL of cadmium telluride by observing the end face of the structure under a microscope. Based on its position, it is possible to estimate the thickness of HL, which is  $d = 75 \mu$ m upon annealing  $t_a = 1.5$  h and temperature T = 820°C.

The optimality of certain regimes of the annealing process is confirmed by the results of the preparation of a  $\beta$ -CdTe HL of a typical cubic modification. They were obtained by the above technology under certain conditions according to the reaction

$$\alpha$$
-ZnTe<sub>Ts</sub> + Cd<sub>G</sub>  $\rightarrow \alpha$ -CdTe<sub>Ts</sub> + Zn<sub>G</sub>. (2.3)

Obtaining HSs with layers of typical cubic  $\beta$ -CdTe and atypical hexagonal  $\alpha$ -CdTe crystal lattices is confirmed by the results of studies of their band structure. Behind it, the energy parameters of the hexagonal structure were determined: the band gap  $E_g$  and the splitting of the valence band into subbands due to the action of the crystal field  $\Delta_{CR}$  and the spin-orbit splitting  $\Delta_{SO}$ . The last parameter was determined for the cubic lattice of  $\beta$ -CdTe. Its presence and the absence of  $\Delta_{CR}$  confirm the formation of the corresponding type of crystal structure. As the research results showed, hexagonal HLs are characterized by time and temperature stability of properties. This indicates the absence of a phase transition of the wurtzite  $\stackrel{\leftarrow}{\neg}$  sphalerite type structure, which is observed when growing by known classical methods of epitaxy and ion implantation [8, 11].

The stability of  $\alpha$ -CdTe can be explained by the formation of a transition layer of the corresponding solid solution between the constituents of HSs. The transition layer eliminates the mismatch between the crystalline and thermal parameters of the contacting materials [10, 11]. Consequently, the separation limit is characterized by a low concentration of defects. The latter is confirmed, as studies have shown, by a high quantum efficiency of  $\eta \approx 7.9$  % of the radiation of the obtained HLs, whose CdTe crystals are characterized by  $\eta \sim 0.1$ -0.2 %.

The crystal lattice of the base crystal in the IVS method determines and stabilizes the structure of the obtained HLs [7]. In particular, the wurtzite structure of the substrate determines the analogous structure of the crystal lattice of the HL formed on the surface. Similar processes and their results are observed in the case of substances with a sphalerite-type lattice. In this case, the chemical composition of substrates and HLs, as well as their constant crystal lattices, differ. The difference in the crystalline and thermal parameters between them is completely compensated by the transition layers of the corresponding solid solutions [11, 12].

#### 2.2 Research Methods

The properties of the resulting layers were determined by studying their basic optical properties: optical transmission  $T_{\omega}$  and reflection  $R_{\omega}$ . They make it possible to determine the main parameters of the crystal structure, which are the main characteristics of materials. In addition, they are non-destructive research methods, in contrast to electrophysical methods. They were carried out according to well-known classical methods [13]. The optical properties were also studied using the  $\lambda$ -modulation method [14]. Its use made it possible to establish the band structure, determine its parameters and reveal properties that cannot be established by classical methods.

A special ELC/C halogen lamp was used for optical measurements. It is characterized by a monotonic emission spectrum in a wide range  $\Delta \lambda = 0.32$ -1.4 µm and the absence of its features in the intensity distribution. The properties of materials, as well as the establishment of the possibilities of their practical use in functional electronics devices, were carried out by studying their luminescent properties, especially photoluminescence  $N_{\omega}$ . For its excitation, an LGN-21 nitrogen laser with a radiation wavelength  $\lambda = 0.337$  µm was used. To establish a reliable spectral distribution of radiation, the quantum spectral sensitivity of the spectral setup  $S_{\omega}$  was determined. It was determined by the well-known method [14].

These measurements were carried out on a universal optical setup based on the MDR-23 spectral device when radiation was detected by a FEP-79 photomultiplier. The use of  $\lambda$ -modulation made it possible to obtain differential spectra  $T'_{\omega}$ ,  $R'_{\omega}$ ,  $N'_{\omega}$ . They were registered using a synchronous detection system. Based on the obtained differential spectra  $T'_{\omega}$ ,  $R'_{\omega}$  and  $N'_{\omega}$ , the basic parameters of materials, their characteristics and properties were established. Behind them, it was possible to establish the prospects for their practical use in multifunctional electronics devices.

#### 3. RESULTS AND DISCUSSION

Thin HLs of CdTe are the main component of functional devices, including optoelectronics. Their growth by the IVS method made it possible to obtain materials of cubic and hexagonal modifications. The result obtained is important for the manufacture of photosensitive detectors and sources of optical radiation with desired and stable properties on their basis, which is an urgent scientific and practical problem [2, 5].

Note that the various epitaxial methods used make it possible to synthesize  $\alpha$ -CdTe films only at low substrate temperatures ( $T_s \approx 300$  °C) [15]. However, their thickness d does not exceed 0.3 µm. An increase in d(during the manufacture of devices), as well as an increase in the substrate temperature  $T_s$ , causes a partial formation of a cubic structure. In addition, thin ( $d \leq 0.3 \mu$ m)  $\alpha$ -CdTe films eventually transform into  $\beta$ -CdTe even at room temperature. Devices made on their basis do not provide the possibility of using under extreme conditions (high temperatures and radiation) and the temporal and temperature stability of the parameters [5, 11].

The IVS method used makes it possible to obtain thin CdTe HLs with a crystal structure identical to that of the substrate [14]. Their parameters, properties, and characteristics are determined both by the perfection of the structure and by doping with isovalent atoms of the base substrate (they are indicated in the above equations for the growth reactions of HLs). In particular, the isovalently substituted cubic modification  $\beta$ -CdTe layers obtained by the IVS method on  $\beta$ -ZnSe substrates under appropriate technological conditions are automatically doped with residual Zn atoms of IVI [9]. Investigation of the differential transmission  $T'_{\omega}$  and reflection R' of  $\beta$ -CdTe:Zn HL revealed characteristic features on the curves. According to which the type of crystal lattice is determined, and the parameters of the band structure are determined, curve 1 in Fig. 2. The main maximum corresponds to the band gap  $E_g$  of cadmium telluride. For a cubic structure, it is  $E_g = 1.50$  eV, which is consistent with the literature data [16]. The second maximum falls on the quantum energy of 2.40 eV and the value of the spin-orbit splitting of the valence band is determined from it:  $\Delta so = 0.90$  eV. The indicated parameters of the band structure were observed in the study of undoped CdTe crystals and doped with IVI Ca and Mg, namely, CdTe:Ca and CdTe:Mg. Such matching of certain parameters for CdTe doped with the indicated impurities and Zn ( $\beta$ -CdTe:Zn) indicates that IVI does not influence the band structure of the base material.

The results obtained make it possible to explain the results of studies of the optical reflection R' correctly and unambiguously of the HL of the atypical  $\alpha$ -CdTe structure, curve 2 in Fig. 2. Three features are observed behind them at photon energies of 1.56, 0.063 and 0.38 eV. Their presence and quantity indicate the crystal structure of the obtained HLs, namely, the hexagonal structure of the  $\alpha$ -CdTe HL crystal lattice. It is determined by the substrate, which predetermines the identical nature of the structure of the obtained HLs, namely, hexagonal. Accordingly, the main extremum at 1.56 eV corresponds to the band gap  $E_g = 1.56$  eV. The remaining maxima are due to the band structure of the hexagonal lattice. It is determined by the splitting of the valence band into subbands due to the action of the crystal field  $\Delta_{CR}$  and is  $\Delta_{cr} \approx 0.063$  eV. The value of the spin-orbit interaction  $\Delta_{so} \approx 0.38 \text{ eV}$  was established. The specified parameters of the a-CdTe band structure are shown in curve 2 in Fig. 2. The values of these parameters of the

band structure of the obtained  $\alpha$ -CdTe/ $\alpha$ -CdSe HS with a hexagonal lattice were determined experimentally for the first time.



**Fig. 2** – Differential reflectance spectra of CdTe HLs obtained on  $\beta$ -ZnTe (1) and  $\alpha$ -CdS (2) substrates, T = 300 K. The inset shows the band structure of crystals of cubic (*a*) and hexagonal (*b*) modifications

The high perfection of the crystal structure is confirmed by photoluminescence studies. As is known [8], it allows one to establish the role of structural defects and dopants (uncontrolled foreign and specially introduced) in the formation of the properties of obtained materials of various origins. In our case, the use of this method of non-destructive testing of the main properties will allow us to determine the effect of growth modes and substitution by isovalent elements on the perfection of the crystal lattice, the presence of various types of IPD in it, the role of various generation-recombination processes and will give an answer about the possibilities of practical use in the corresponding electronic devices.

The high quantum efficiency of radiation in the edge region is important for the obtained HLs. It is observed in the near IR range and is  $\eta \approx 7.9$  %. Note that it was determined from a reference GaAs sample with  $\eta \approx 100$  % using the well-known technique [10].

The emission spectrum covers the photon energy range  $1.5 \div 1.65 \text{ eV} (\Delta \lambda = 0.751 - 0.826 \ \mu\text{m})$  with a maximum at  $\hbar \omega_m = 1.55 \text{ eV} (\lambda_m = 0.800 \ \mu\text{m})$ , Fig. 3. There is also a second maximum at  $\hbar \omega = 1.62 \text{ eV} (\lambda_m = 0.765 \ \mu\text{m})$ with a slight intensity. The corresponding differential radiation spectrum  $N'_{\omega}$ , measured by  $\lambda$ -modulation, differs significantly in shape from the distribution of photons by the classical method, Fig. 3. Features are observed that testify to the composite nature of the radiation. It is characterized by several bands due to fundamentally different mechanisms of radiative recombination. We will consider their nature.

Note that in the region of photon energies  $\hbar \omega \ge E_g$  (at  $E_g = 1.56$  eV), intense radiation is observed. It is characterized by properties inherent in interband radiative recombination. Such a radiation mechanism is confirmed by good agreement between the experimental curve and the well-known analytical expression [12]

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$$N_{\omega} \sim \left(\hbar\omega\right)^2 \sqrt{\hbar\omega - E_g} \exp\left(-\frac{\hbar\omega - E_g}{kT}\right), \qquad (3.1)$$

where k is the Boltzmann constant, T is the temperature. This component exhibits a maximum at  $\hbar\omega_m = 1.575 \text{ eV} (\lambda_m = 0.787 \text{ µm})$  on the differential spectrum obtained by  $\lambda$ -modulation.

The spectrum does not change position when L varies by 3 orders of magnitude, which is typical for interband recombination of free charge carriers [17, 18].



**Fig. 2** – Spectra of conventional (1) and modulated (2, 3) photoluminescence of  $\alpha$ -CdTe/ $\alpha$ -CdSe HSs of the hexagonal modification, as well as components of the emission bands due to interband radiative transitions of free charge carriers (4, 5) and annihilation of bound excitons (6). The inset shows the dependence of the intensity (7) and the position of the maximum (8) on the excitation level of the *L* component of the exciton emission band, *T* = 300 K

The insignificant contribution of self-absorption processes and the possibility of observing it in the radiation spectrum at energy  $\hbar \omega > E_g$  can be explained both by the perfection of the crystalline structure of the layers and by their small thickness *d*, which is due to the use of the regimes for obtaining layers by the isovalent substitution method. It is these factors that also make it possible to observe the radiation component in the region  $\hbar \omega > E_g$ with a maximum at  $\hbar \omega_m = 1.623$  eV. Analyzing the band structure parameters  $E_g$  +  $\Delta_{cr}$ , we obtain  $\hbar \omega_m = 1.56 + 0.063 \text{ eV} = 1.623 \text{ eV}$ , which corresponds to the experimental result. This suggests that this emission band in the  $\hbar \omega > E_g$  region is due to interband transitions of free charge carriers to the valence subband split off under the action of the crystal field.

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At the same time, the self-absorption effect does not allow one to observe the radiation formed by transitions involving the valence subband split off by the spin-orbit interaction  $\Delta_{so}$ . The value of the radiation photon energies could be 1.56 + 0.38 = 1.84 eV, but the limiting value of the interband radiation range is 1.65 eV.

An important result of the preparation of  $\alpha$ -CdTe HLs by IVS is the formation of a radiation component with a maximum at  $\hbar \omega_m = 1.54 \text{ eV} (\lambda_m = 0.805 \ \mu\text{m})$  in the range  $\hbar \omega \leq E_g$ . It is dominant and unambiguously reveals its nature in studies using the method of modulation spectroscopy [19]. The appropriate differential curve is complex, and its character depends on the excitation level L. The following properties are observed. Firstly, the point E of the intersection of the curve with the abscissa axis (characterizes the maximum  $\hbar \omega_m$  of the ordinary  $N_{\omega}$  curve) shifts with an increase in the excitation level L to the region of lower photon energies, curve 8 in the inset in Fig. 3. Secondly, the intensity I of this low-energy emission band changes with the excitation level according to the law  $I \sim L^{1.5}$ , curve 7 in Fig. 3.

The established features are characteristic of the annihilation of excitons during their inelastic scattering by free charge carriers [20]. They are holes, since *a*-CdTe HLs under study are characterized by hole conductivity due to doping upon substitution of residual Se atoms for the *a*-CdSe substrate. The shift of the maximum (the cross section of the differential curve with the  $\hbar \omega$  axis) towards lower energies is adequately explained by the increase in the concentration of free holes with increasing *L*, which leads to an increase in the probability of exciton scattering. Most likely, the excitons are bound on the isovalent Se impurity.

The presented results of studying the properties of HLs of the atypical  $\alpha$ -CdTe modification indicate the possibility of obtaining highly efficient  $\alpha$ -CdTe/ $\alpha$ -CdSe HS light emitters in the near IR region. Additional temperature studies carried out in the range of 20-200 °C made it possible to establish that some parameters of  $\alpha$ -CdTe HL and  $\alpha$ -CdTe/ $\alpha$ -CdSe HS do not change, as well as during long-term (1-3 years) storage of samples. This indicates the possibility of their use in devices with extended functionality.

## 4. CONCLUSIONS

The use of solid-phase substitution reactions at an IVI concentration of more than 0.1 at. % ensures the formation of  $\alpha$ -CdTe HLs of atypical hexagonal modification of the crystal lattice. For the first time, the parameters of their band structure  $E_g = 1.56 \text{ eV}$ ,  $\Delta_{CR} = 0.06 \text{ eV}$ ,  $\Delta_{SO} = 0.311 \text{ eV}$  have been determined. High intense radiation is obtained, the stability of which is ensured by the formation of transition layers with a low concentration of solid solution defects between the contacting materials. It is formed by the interband recombination of free charge carriers, and the dominant annihilation of excitons bound on isovalent impurities. The possibility of obtaining  $\alpha$ -CdTe/ $\alpha$ -CdSe HSs of hexagonal modification of sources of intense edge radiation with an efficiency of  $\eta \approx 7.9$  % in the near IR region is shown.

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# Гетерошари гексагонального *α*-CdTe

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Аргументовано важливість прямої зонної структури СdTe як необхідної умови при створенні на його основі високоефективних приладів фотоелектроніки. Показано можливість отримання гетерошарів телуридів кадмію методом ізовалентного заміщення. Визначено передумови виготовлення, та розроблено технологічні режими ізотермічного відпалу базових кристалів α-CdSe і α-CdS. Вперше отримано методом ізовалентного заміщення гетерошари з мало використовуваною нетиповою гексагональною модифікацією α-CdTe кристалічної гратки із високо стабільними властивостями. Розроблено методику і досліджено методом λ-модуляції оптичного опромінення базові параметри зонної структури і характеристики люмінесценції. Встановлено ширину забороненої зони Eg = 1,56 eB і вперше визначено параметри зонної структури, а саме, підзон валентної зони, відщеплених внаслідок дії кристалічного поля  $\Delta_{cr} pprox 0,063$  eB і спін-орбітальної взаємодієї  $\Delta_{so} pprox 0,38$  eB. Встановлено високу квантову ефективність  $\eta pprox 7$ -9% домінуючого у крайовій області випромінювання. Воно визначається легуванням ізовалентними домішками, якими є атоми базової підкладки. Важливою передумовою отримання високої інтенсивності (порівняно з кристалами CdTe з  $\eta \sim 0,1-0,2\%$ ) є формування перехідного шару відповідного твердого розчину між складовими гетероструктур, який істотно зменшує концентрацією дефектів та неузгодженість кристалічних і термічних параметрів контактуючих матеріалів. Отримані особливості кристалічної структури забезпечують умови формування ефективної люмінесценції, базовими механізмами якої є міжзонні випромінювальні переходи і домінуюча анігіляція зв'язаних на ізовалентних домішках екситонів.

Ключові слова: Телурид кадмію, Гексагональна структура, Висока квантова інтенсивність випромінювання, Ізовалентні атоми.