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SEMICONDUCTOR THALLIUM CHALCOINDIANATES AND ALLOYS ON THEIR BASIS WITH RARE-EARTH ELEMENTS

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Abstract: In the presented work, the specific features of the crystal lattice of the TISe type and the principle of producing semiconducting thallium chalcoindianates and alloys based on them with rare-earth elements are described in detail. Correlations between the physical properties and the content of rare-earth elements in the compositions of the studied materials are given. The results of experimental studies on the radiation effects arising under the influence of gamma radiation are presented.

Keywords: semiconductor chalcoindiantes, electrical conductivity coefficient, Hall, thermopower, thermal efficiency, gamma irradiation

It is known that the special and very interesting properties of rare-earth elements area primarily associated with the structural features of the electron shell of the atoms of these substances, namely, the presence in the shell of an expanding 14-local deep 4f layer [1].

The outermost 6s 6p 5d electrons form a band (zone) of conductivity, leaving rare earths in a solid trivalent. The properties of this band determine their electrical, optical properties and transport phenomena.

In TISe lattices, thallium atoms exhibit variable valence. Monovalent thallium and indium ions in octahedral are responsible for the formation of crystal lattices of this type, while the semiconductor features of compounds with this structure are mainly determined by trivalent ions (TI^{3+}, In^{3+}) in tetrahedra [2,3,4].

For the same reasons, in the case of partial substitution of In atoms in $TIInX_2$ by lanthanide atoms, we obtained quadruple compounds and solid solutions according to the following scheme [5]:

$$TI^{+}TI^{3+}X_2^{VI} \rightarrow TI^{+}In^{3+}X_2^{VI} \rightarrow Tl^{+}In_{1-x}^{3+}Ln_x^{3+}X_2^{VI}$$
$$\rightarrow TI_2^{+}In^{3+}Ln^{3+}X_4^{VI} \rightarrow TI^{+}Ln^{3+}X_2^{VI}$$

In these compounds, in our opinion, the external electron shells of two pairs of chalcogen atoms are extended to neutral argon, krypton and xenon by $6p^1$ - electrons TI, $5s^2$ $5p^1$ - indium electrons $5d^1$ $6s^2$ - lanthanides.

In all triple $TIInX_2^{VI}$, $TILnX_2^{VI}$, $TI_2InLnX_4^{VI}$ trials that we studied, thallium atoms exhibit a monovalent and indium and lanthanide atoms trivalent state [6-8].

The physico-chemical, X-ray phase, microstructural analyzes, the study of microhardness and density were used to plot the state diagrams $TIInS_2(Se_2, Te_2)$- $TILnS_2(Se_2, Te_2)$, in which areas are observed solubilities based on $TIInS_2(Se_2, Te_2)$and new quadruple compounds with the general formula $\llbracket TI_2InLnS_4(Se_2, Te_2)$. Solid solutions in $TIInX_2$- $TILnX_2$systems and new phases TI_2InLnX_4, crystallize in the same tetragonal syngony, the lattice parameters of solid

solutions based on ternary compounds of the type $TIInX_2$ with increasing content lanthanides increases additively law [7.8].

A characteristic feature of the $TIIn_{1-x}Ln_xTe_2$ solid solutions crystals is the presence of high concentration stoichiometric vacancies in their crystal lattice $(10^{18} - 10^{19} см^{-3})$ and associated localized states in the band gap [6, 7], and this value increases with an increase in the relative amount of lanthanides in the composition of solid solutions. The formation of solid solutions leads to changes in the electron and lattice subsystems of the crystal, causing static lattice deformation and disturbance of the electronic spectrum. With an increase in the concentration of vacancies in a solid solution, the average distance between the atoms of the solute $TILnTe_2$ reaches the values at which the interimpurity interaction begins to make a significant contribution to the energy of the solvent crystal $TILnTe_2$, and this leads to the appearance of fundamentally new properties not characteristic of the material [9].

At high impurity concentrations, the band splits, the impurity levels approach the bottom of the conduction band, merge with it, and all observed features disappear. In these crystals, the band gap is sufficiently large, the concentration of charge carriers for the initial crystals is small, they are easily doped, and small deviations from stoichiometry also create acceptor levels. To confirm the presence of an acceptor zone in them, the curves $R_x(T)$,$\alpha(T)$ and σ (T) were analyzed, the results of which agree well with the values obtained on the basis of the electrical properties in strong electric fields [8]. The nature of the dependence of electrical conductivity for all samples, observed in a wide range of temperatures and fields, indicates an exponential increase in electrical conductivity depending on the electric field strength. Experimental data on the change in the electrical conductivity of the samples studied in strong electric fields is qualitatively explained by the Frenkel theory, according to which upon thermionic ionization, the activation energy of charge carriers decreases linearly with increasing field strength:

$$\Delta E = \Delta E_g - 2e\sqrt{e\,E/\varepsilon} = \Delta E_g - \Delta E_n, \qquad (1)$$

where ΔE_g is the activation energy in the absence of an electric field,

ΔE– change in activation energy under the influence of an electric field.

Using the extrapolation method, the activation energy of charge carriers in the absence of an electric field for various compositions of solid solutions $TIIn_{1-x}Ln_xS_2(Se_2, Te_2)$ and $InS_2(Se_2, Te_2) and TI_2InLnS_4(Se_4, Te_4)$.

The study of the electrical conductivity of single crystals of ternary compounds $TIInS_2(Se_2, Te_2)$ and solid solutions based on them showed that the conductivity in the direction parallel to the layers ($\sigma_\parallel$) is much greater than in the direction perpendicular to the layers of tetrahedra ($\sigma_\perp$). It was established that the ratio $\sigma_\parallel/\sigma_\perp$ in $TIInS_2(Se_2, Te_2)$ and solid solutions $TIIn_{1-x}Ln_xS_2(Se_2, Te_2)$ with increasing electric field strength and with decreasing temperature increases. This means that $\sigma_\parallel$ is less dependent on temperature than $\sigma_\perp$, which confirms the arguments about the structure and chemical bonds in the studied samples.

The obtained results can be interpreted from the standpoint of the Frenkel-Poole theory, which suggests that the considered dependences of electrical conductivity σ, activation energy ΔE, and coefficient β on the electric field strength and temperature in strong electric fields are due to an increase in the concentration of charge carriers in solid solutions $TIInX_2$- $TILnX_2$due to the action of the mechanism of thermionic ionization [8].

One of the unsolved problems in microelectronics is the development of solid-state inductive elements, since there is still no proposed method for localizing the magnetic field in a semiconductor. Therefore it is necessary to use indirect methods for creating inductances, or

rather the so-called. analogues of inductances. For this, it is necessary that the inductance analog has inertia to a change in current and provides a phase shift between current and voltage by an angle $\varphi = \pi/2$. Such phenomena take place in devices with negative resistance (NR) in the presence of positive feedback (PFB) between the exit and the entrance. Feedback can be of two types - current and voltage. PFB current should lead to an increase in the current through the device, and PFB voltage should increase the voltage. In the first case, the element exhibits inductive properties, and in the second, capacitive [10, 11].

With positive current feedback (S-type IVC), the current is delayed relative to the voltage, therefore, in devices with S-type NR, the reactive component of the impedance is inductive. Similarly, in an N-type NR device with a voltage across the voltage, the voltage change is delayed with respect to the current, so they have a reactive component of the impedance of a capacitive nature [10].

The study of the current-voltage characteristics of the $TIInS_2(Se_2, Te_2)$-$TILnS_2(Se_2, Te_2)$ showed that all of them have switching properties with memory. The parameters that characterize the switching effect are investigated depending on the temperature and thickness of the active region. The following linear and quadratic regions, the region of sharp current growth, the region of negative differential resistance (NDR) and the regions of small positive differential resistance are distinguished on the NDR [12].

On the basis of experimental data, it was obtained that the threshold voltage is weakly dependent on the temperature and thickness of the sample. Such a weak dependence of the threshold voltage on temperature is due to the fact that, at temperatures of 77-200 K, an electronic switching mechanism takes place, i.e. as a result of a decrease in the dissipated power, thermal breakdown at low temperatures becomes electronic. And at higher temperatures (T-200-350K), the locally advantageous thermal switching mechanism is located.

The promise of the investigated compositions is obvious, since on their basis, using microelectronic methods, it is possible to make memory elements, semiconductor analogues of inductance, light-driven oscillators, without complicating the technology and design of the corresponding devices.

In [7, 8], it was shown that $TlIn_{1-x}Ln_xTe_2$ is characterized by defects (in the opinion of the authors, these defects are point-like), In (or Ln) vacancies, interstitial sites that appear due to statically located In atoms in the sublattice. Therefore, it is necessary to take into account the mobility contribution μd, calculated using the relaxation time with the scattering mechanism on point defects for the standard zone

$$\tau_d(T) = \frac{\pi\hbar^4}{(2m^*kT)^{1/2} m^* V_0^2 N_d^{|}} \left(\frac{E}{kT}\right)^{-1/2}, \quad (2)$$

where V_0 is a constant characterizing the amplitude δ of the potential, is the concentration of point defects, which was taken as ≈Na. Taking into account the values of V_0, and m * in the corresponding expressions [9], we determine τ_d (T), after which we can determine μ_d (T). Then we define the effective mobility as follows:

$$\mu_3 = \left(\frac{1}{\mu_{i,ac}} + \frac{1}{\mu_{op}} + \frac{1}{\mu_d}\right)^{-1}, \quad (3)$$

where from σ (T) and α (T) were calculated [9].

A quantitative comparison of the data on μ_p (T) showed that the calculated values of hole mobility are greater than experimental ones. This may be due to two reasons: 1) in $TlIn_{1-x}Yb$, the value of the screening radius differs from the value of the lattice constant, 2) in this material the hole gas is strongly degenerate. The second reason is more likely. To confirm this assumption, the reduced chemical potentials () were determined and it was found that for all samples at T = 100K ≥10. With an increase in temperature, degeneracies are lifted, with the result that the spread between calculated and experimental data increases. According to the experimental data of μ_p (T), it is quantitatively difficult to distinguish scattering by acoustic and optical phonons in strongly degenerate cases. The prevailing scattering mechanism is easily distinguished from the concentration dependence μ (n). As noted in [17], μ_{ac}~n^{-1}, μ_{op}~$n^{1/3}$, μ~$n^{2/3}$. From the data obtained it can be seen that in the dependence p ~ the exponent α_0≈0.7 and α_0 is almost independent of the hole concentration. This means that up to T≤700K, the hole scattering mechanism is mixed. Compared to other narrow-gap semiconductors [9], the mobility of electrons and holes in $TlIn_{1-x}Yb_xTe_2$is low, the likely cause of which is the large effective mass of charge carriers in them.

So, in the temperature range studied, the $TlIn_{1-x}Yb_xTe_2(0 \le x \le 0{,}10)$ solid solution has hole conductivity in a strongly degenerate state of hole gas. At T≥700K, a domain of intrinsic conductivity is observed and the nature of the temperature dependences of σ and α is explained by the presence of two types of charge carriers. Starting from x = 0.05, when going from $TlInTe_2$ to $TlIn_{1-x}Yb_xTe_2$, the temperature dependence σ (T) differs qualitatively and quantitatively. It is assumed that this transition is accompanied by transitions from a semiconductor to a narrow-gap semiconductor. The proposed model with one (holes) and two (electrons and holes) types of charge carriers, taking into account the scattering of charge carriers at ion centers, acoustic and optical phonons, as well as defects, describes well the electrical and thermoelectric properties of $TlIn_{1-x}Yb_xTe_2$ solid solutions at 0.05≤ x≤0.10 [13].

The temperature dependence of the thermal conductivity of the compounds $TIInS_2(Se_2, Te_2)$, $TI_2InLnS_4(Se_4, Te_4)$, $TILnS_2(Se_2, Te_2)$ and solid solutions $TIIn_{1-x}Ln_xS_2(Se_2, Te_2)$, in the temperature range of 80÷450K. The calculation of the electronic part of the thermal conductivity showed that it is much less than the total in all the studied samples and can be neglected [7, 8, 14, 15].

In the single crystals $TIInS_2(Se_2, Te_2)$, the pronounced anisotropy of the lattice thermal conductivity takes place in the entire temperature range studied. In the series La → Yb, the thermal conductivities of the compounds $TIInX_2$ and TI_2InLnX_4 both decrease for sulphides and selenides and tellurides, which can be explained by a partial change in the rigidity of the crystal lattice of sulphides when the metal atoms are replaced with analogs with large charge numbers due to which weakens the strength of ionic bonds between metal and chalcogen atoms. In a similar way, one can explain the change in thermal conductivity for the same compounds in the series S → Se → Te. The thermal conductivity of three-component compounds of the type $TITITe_2$ with a variation of the cation composition in the direction of an increase in the atomic weights of substitution cations naturally decreases. However, with the transition from compounds with an ordered arrangement of atoms to solid solutions of substitution on their basis, there is a deviation from the usual pattern of changes in thermal conductivity from atomic weight. This circumstance is probably due to the fact that for mixed crystals of variable composition $TIIn_{1-x}Ln_xX_2$ phonon scattering from local point defects prevails, and the main factor affecting the thermal resistance due to point defects is local change in the density and elastic properties of the medium. The local change in the density is mainly dependent on the average atomic weights, the local change in the elastic properties of the difference between the atomic radii of the solvent and the solute.

Crystals of variable composition of the type $TIIn_{1-x}Ln_xX_2$ according to crystal structure considerations seem to be extremely convenient for testing the theory of phonon scattering from local distortions of crystal lattices developed in recent years. Crystals of $TIIn_{1-x}Ln_xX_2$ solid solutions are presented to us as a result of the partial replacement of dissimilar elements of cations based on the TISe-type lattice, which is also supported by the results of structural and electrophysical studies. When testing the theory of the interaction of phonons with point defects, the problem is simplified by the fact that as a result of the partial substitution of indium atoms by lanthanide atoms in $TIInX_2$ compounds, the nature of interatomic bonds remains completely unchanged, since the substitution atoms are isovalent and differ in mass. Thus, this model is closer in that idealized model, where local changes in density in the lattice are not accompanied by a corresponding (local) change in elastic properties. The results were interpreted within the framework of a theory that takes into account the roles of three phonon transfer processes (U-processes) and normal processes (N-processes), as well as point defects in phonon scattering, the results of the study of the thermoelectric properties of these compounds and solid solutions are built$TIInS_2(Se_2, Te_2)$- $TILnS_2(Se_2, Te_2)$.

It has been established that in the temperature range 80–450 K in the scattering of phonons in these compounds, the main role is played by three-phonon transfer processes (U-processes), and in solid solutions simultaneously with

U – processes, active processes are played by normal processes (N – processes) and scattering on point defects.

In all materials, the temperature and concentration dependences of the elastic properties were investigated and the results of calculations showed that the Debye temperature decreases with increasing lanthanide content in solid solutions; the propagation of longitudinal ultrasonic waves is increasing. This is due to the fact that the formation of ternary compounds $TIInX_2$ in a chemical bond from each atom of indium involves three electrons. At the same time, some of these electrons, which are in the collectivized state, are shifted to the residuals of the chalcogen atoms, and some of them remain in a localized form in the vicinity of the indium atoms and participate in the formation of a covalent bond.

Compounds of groups A^3B^6, $A^3B^3C_2^6$ and mixed crystals based on them are a wide class of semiconductor materials of interest to which their extremely anisotropic (chain and layered) crystalline structure is caused. The study of such compounds is associated with the solution of a number of fundamental problems of solid state physics. In addition, experimental studies have shown the promise of using this class of compounds in laser technology, nonlinear optics, in electroluminescent switching devices, as modulators and frequency converters of receivers of the visible and infrared range, strain gauges and other purposes.

Many potential practical applications of these crystals turned out to be unrealized due to the lack of detailed information on the dynamics of the crystal lattice. The monograph [17] presents the results of studies of the vibrational spectra of crystals from the group of A^3B^6 compounds with a chain structure, and from the group $A^3B^3C_2^6$ compounds which crystallize in both the chain and layered structure. It examines the effect of anisotropy, disorder, and temperature instability of the crystal lattice on the phonon spectra of these crystals, and employs experimental methods: inelastic coherent and incoherent neutron scattering, far-infrared spectroscopy, Raman and Mandelstam-Brüllian scattering.

In [18], the vibrational spectra of $TlInS_2$ crystals in the terahertz (0.1–3 Hz) region were studied and analyzed. The observed low-frequency band is associated with vibrational oscillations of nanolayers (“boson peak"). The relationship between coherent low-frequency oscillations and the disordered nature of the crystal structure is shown. It is concluded that there is a topological disorder, which is manifested at the level of the layered subsystem.

In [19, 20], the temperature dependences of the dielectric conductivity ε (T), electrical conductivity σ (T), and current-voltage characteristics (VAC) of A^3B^6 and $A^3B^3C_2^6$ crystals were studied, which allowed us to establish the mechanisms of: conductivity in a wide temperature range; observable giant dielectric relaxation; and "S" -shaped switching and memory phenomena in these systems. Since defects make a significant contribution to the formation of the observed features, the authors also studied the effect of defects formed under the influence of gamma radiation with a dose of up to 250 Mrad on the above features. It is shown that the dependences of σ (T) in $TlGaTe_2$ crystals subjected to doses (0–250 Mrad) of γ-irradiation have a hopping character and are described in the Mott approximation.

It was found that, when γ-irradiated, in the band gap of a $TlGaTe_2$ crystal, impurity energy levels arise due to radiation defects. Thermal filling of these levels occurs at a lower temperature than in the case of an unirradiated crystal. With an increase in the dose of irradiation to 100 Mrad, the conductivity decreases (T = 200 K), in the dose range 100-200 Mrad remains constant, a further increase in the dose of radiation leads to an increase in conductivity. It is shown that in these crystals the maximum value of the dielectric conductivity irradiated with a dose of 100 Mrad is ~ 6 times higher than the initial value. It was established that in $TlGaTe_2$ crystals the maximum value of specific conductivity at a dose of 100 Mrad is an order of magnitude higher than the initial one, and the transition temperature to the superionic state shifts towards low temperatures. The critical values of the radiation dose at which an abrupt change in conductivity is observed.

The monograph [21] summarized and presented the results of studies on the nature of effects arising in layered semiconductors of the A^3B^6 and $A^3B^3C_2^6$ type and in structures based on them, under the influence of ionizing radiation. The influence of radiation effects on the photoelectric, photoluminescent properties of these crystals is considered. Radiation effects in diode structures based on layered single crystals of A^3B^6 and $A^3B^3C_2^6$ types are analyzed and recommendations on the radiation resistance of these photosensitive structures are given.

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ПОЛУПРОВОДНИКОВЫЕ ХАЛЬКОИНДИАНАТЫ ТАЛЛИЯ И СПЛАВЫ НА ИХ ОСНОВЕ С РЕДКОЗЕМЕЛЬНЫМИ ЭЛЕМЕНТАМИ

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Резюме: В предъявленной работе подробно изложен специфические особенности кристаллической решетки типа TISe и принцип получения полупроводниковых халькоиндианатов таллия и сплавов на их основе с редкоземельными элементами, приводятся корреляции между физическими свойствами и содержанием редкоземельных элементов в составах исследуемых материалов. Представлены результаты экспериментальных исследований по изучению радиационных эффектов, возникающих под воздействием гамма-излучения.

***Ключевые слова*:** полупроводниковые халькоиндианаты, коэффициент электропроводности, Холла, термоэдс, термоэффективность, гамма-облучение

TALLİUM HALKOİNDİANAT YARIMKEÇİRİCİLƏR VƏ ONLAR ƏSASINDA NADİR TORPAQ ELEMENTLİ ƏRİNTİLƏR

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Xülasə: Təqdim edilmiş işdə TlS tipli kristallik qəfəsin məxsusi xüsusiyyətləri və tallium halkoindianat yarımkeçiricilərin və onlar əsasında nadir torpaq elementli ərintilərin alınma prinsipləri ətraflı verilmişdir. Tədqiq edilən materialların tərkibində olan nadir torpaq elementliərinin miqdarı və onların fiziki xassələri arasında asılılıqlar araşdırılmışdır.Qamma şüalanmanın təsiri nəticəsində müşahidə olunan radiasiya effektlərinin təcrübi tədqiqıt nəticələri araşdırılmışdır.

Açar sözlər: yarımkeçirici halkoindianatlar, elektrik keçirmə əmsalı, Hool, termoehq, termoeffektivlik, qamma şüalanma**.**