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Electrical Properties and Photosensitivity of n-Mn_2O_3/p-InSe Heterojunctions Produced by the Spray Pyrolysis Method

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The conditions of application of thin semiconductor Mn_2O_3 films on p-InSe crystalline layered semiconductor substrates at a temperature of 623 K by the spray-pyrolysis method to create anisotypic heterojunctions n-Mn_2O_3/p-InSe were investigated. InSe is a promising material for photoelectronics. The use of the Mn_2O_3 film, which is transparent in the region of maximum photosensitivity of InSe, makes it possible to effectively exploit the optical properties of InSe in the fabrication of various semiconductor devices. The advantage of using layered semiconductors in the production of heterojunctions is that high-quality interfaces are obtained even with a significant discrepancy in the parameters of the crystal lattices of the starting materials. This significantly expands the choice of heterojunction materials. Electrical and photoelectric parameters of n-Mn_2O_3/p-InSe heterojunctions were measured and theoretical models describing them were proposed. The graphical dependencies of *I-V* characteristics, series resistance, height of the potential barrier and photosensitivity are constructed. It was established that these heterojunctions are photosensitive and have rectifying properties. Using the energy parameters of the starting materials, an energy diagram of the heterojunction was constructed, which allows for the analysis of physical processes in the obtained heterojunctions. Based on the temperature dependence of both direct and reverse *I-V* characteristics, the dynamics of changes in the energy parameters of the heterojunction with temperature, as well as the mechanisms of current flow through the heterojunction, are established. The spectral photosensitivity of the heterojunction was analyzed.

Keywords: Indium Selenide, Mn_2O_3, Heterojunction, Spray Pyrolysis, *I-V* Characteristics, Photosensitivity.

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1. INTRODUCTION

The value of the band gap $E_g = 1.26$ eV of indium monoselenide InSe makes this material attractive for the creation of photodetectors of the visible spectrum and photoconverters of solar energy. InSe crystals are characterized by a structure based on layers that are connected by van der Waals bonds. The production of InSe substrates for the creation of heterojunctions is carried out by peeling off the material from the ingot and eliminates the operations of mechanical and chemical treatment of the surface of the plates in order to clean them. Indium selenide is used to create diodes and photosensitive structures based on the Schottky barrier and heterojunctions [1-4].

Among the various crystalline modifications of manganese oxide Mn_2O_3 [5], the cubic phase α-Mn_2O_3 is one of the most stable. α-Mn_2O_3 is used in chemical analysis, devices for energy conversion and storage [6]. This material is non-toxic and cost-effective.

As a manganese oxide semiconductor, heterojunctions with g-C_3N_4 [7], CuOx [8], Fe_2O_3/Mn_2O_3 [9], n-Mn_2O_3/n-CdZnTe [10] are used. Thin films of Mn_2O_3 with a band gap $E_g \approx 2.01 \div 2.4$ eV [11, 12] are promising as a frontal material for heterojunctions. α-Mn_2O_3 films are produced by spray-pyrolysis [12, 13], electrodeposition [14], sol-gel method [15] etc. The spray-pyrolysis method is characterized by simple hardware implementation and mobility of mode correction in the production of α-Mn_2O_3 films with given physical properties. Taking into account the successful application of the method for creating heterojunctions based on indium selenide and heterostructures using manganese oxide [4, 10, 16], the purpose of the study was to manufacture and study the photovoltaic properties of n-Mn_2O_3/p-InSe heterojunctions. We have already produced photosensitive heterojunctions based on the contact of Mn_2O_3 with n-InSe, which show good rectifying properties [4]. Therefore, the idea of using p-InSe is quite obvious. This paper analyzes the results of studies of the electrical properties and spectral characteristics of the quantum efficiency of n-Mn_2O_3/p-InSe heterojunctions, which were produced by the method of spray pyrolysis of an aqueous solution of manganese chloride salt on p-InSe substrates.

2. EXPERIMENTAL

For the production of n-Mn_2O_3/p-InSe anisotypic heterojunctions, substrates up to 500 μm thick were used, which were exfoliated from indium monoselenide p-InSe crystals grown by the Bridgman method. Hole electrical conductivity was ensured by doping with cadmium (0.1% by mass). The concentration of charge carriers determined on the basis of Hall studies was $p \sim 10^{14}$ cm^{-3} at room temperature. The mobility of holes in the perpendicular direction relative to the symmetry axis $\boldsymbol{c}$ was equal $\mu_{pH} \approx 50$ cm^2/(V·s). n-Mn_2O_3/p-InSe heterojunctions were produced by applying thin Mn_2O_3 films with a thickness of $w \approx 0.5$ μm on the surface of p-InSe substrates by the spray pyrolysis method. Spray pyrolysis was carried out under atmospheric conditions (with access to the substrate air). A 0.1 M aqueous solution of manganese crystal hydrate salt $MnCl_2 \cdot 4H_2O$ was used to

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generate an aerosol over the surface of the substrates.

The temperature of the substrates during the pyrolysis of manganese dichloride was maintained at a stable $T_S = 623$ K. During the pyrolysis of salt, atoms of elemental manganese are formed on the substrate, which, when interacting with atmospheric oxygen, form a film of the α-Mn_2O_3 binary compound, which has an n-type conductivity, a high resistivity at $\rho \approx 10^7$ Ohm·cm (at $T = 300$ K) and an optical band gap $E_g \approx 2.12$ eV. For n-Mn_2O_3 films, a high resistivity is characteristic together with a low electron diffusion coefficient $D_n = 5 \cdot 10^{-3}$ cm²/s [17] and a low concentration $n \approx 1.1 \cdot 10^{12}$ cm^{-3}. Contacts to p-InSe substrates and n-Mn_2O_3 films were made using a conductive paste containing finely dispersed silver particles.

The *I-V* characteristics of the n-Mn_2O_3/p-InSe heterojunctions were measured on the SOLARTRON SI 1286, SI 1255 hardware complex in the temperature range $265 < T < 321$ K. The spectral photosensitivity of the heterojunctions was studied at room temperature using the MDR–3 monochromator, which had a resolution of 2.6 nm/mm. The photosensitivity spectra were normalized with respect to the photon flux.

3. RESULTS AND DISCUSSION

The analysis of electronic processes at the n-Mn_2O_3/p-InSe heterojunction was carried out using the energy diagram (see Fig. 1), which is built on the basis of the energy parameters of n-Mn_2O_3 and p-InSe semiconductors. The electron affinity energy of Mn_2O_3 $\chi = 4.45$ eV determined by the Kelvin method [13] was used, which is characteristic of Mn_2O_3 films produced by the spray pyrolysis method. The used value of the band gap $E_g(Mn_2O_3) = 2.12$ eV, which is obtained from optical studies of Mn_2O_3 films, was tested when constructing the energy diagram of n-Mn_2O_3/n-CdZnTe heterojunctions [10]. The depth of the Fermi level $\delta_1 = E_C - E_F \approx 0.41$ eV in the band gap of n-Mn_2O_3 was calculated using the expression for non-degenerate semiconductors [18]:

$$E_C - E_F = \delta = kT \cdot \ln\left(2 \cdot \left(\frac{2\pi m_n kT}{h^2}\right)^{3/2} \cdot \frac{1}{n}\right), \qquad (1)$$

where m_n is the effective mass of electrons $m_n = 0.48 \cdot m_0$ [19] for n-Mn_2O_3.

The concentration of charge carriers in n-Mn_2O_3 films $n \approx 1.1 \cdot 10^{12}$ cm^{-3} was determined based on the results of the research of the specific electrical resistance $\rho \approx 10^7$ Ohm·cm by the formula $n = (q\rho\mu_n)^{-1}$). The mobility of electrons is $\mu_n = D_n/kT$, where the diffusion coefficient for electrons $D_n = 5 \cdot 10^{-3}$ cm²/s [20], k is the Boltzmann constant.

The electron affinity energy χ(InSe) = 4.6 eV and the band gap E_g(InSe) = 1.2 eV for the used indium monoselenide crystals with the depth of the Fermi level $\delta_2 = E_F - E_V = 0.3$ eV is used to determine the energy profile of InSe-based heterojunctions [16].

The magnitude of the contact potential difference $q\Phi$ of the n-Mn_2O_3/p-InSe heterojunction is significantly affected by the difference in electron affinities, which determines the gap in the conduction band. The absolute value of $\Delta E_C = |\chi_1 - \chi_2| = 0.15$ eV. The value of $q\Phi$ can be found by next formula:

$$q\Phi = E_{g2} - \delta_1 - \delta_2 + \Delta E_C, \qquad (2)$$

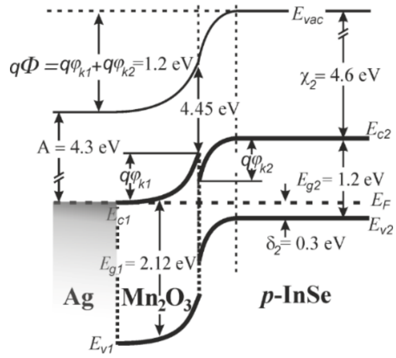

Fig. 1 – Energy diagram of n-Mn_2O_3/p-InSe heterojunction

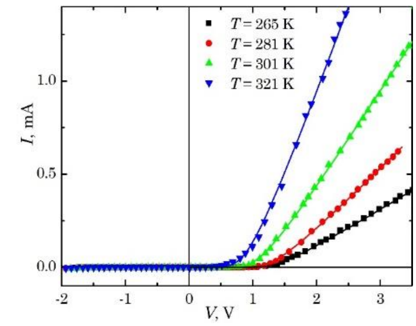

Fig. 2 – *I-V* characteristics of n-Mn_2O_3/p-InSe heterojunction at different temperatures (points are experimental data, curves are approximation by formula (3))

ΔE_C has a positive effect on the value of $q\Phi$, which is equal to 1.2 eV, but is able to create obstacles for the movement of electrons in the conduction zone at the heterojunction. It should be noted that despite the high resistance of n-Mn_2O_3 films, when using a silver paste for forming of contact to the heterojunction, the n-Mn_2O_3 film is enriched with electrons from Ag and the Ag/n-Mn_2O_3 contact has ohmic properties. This is facilitated by the difference in affinity energies between the metal and the film. On the *I-V* characteristics of anisotypic n-Mn_2O_3/p-InSe heterojunctions in the temperature range of $T = 265 \div 321$ K (see Fig. 2), diode properties are revealed. An increase in the direct current of the heterojunction was observed when a positive potential was applied to the p-InSe region. With the reverse bias (positive potential on the n-Mn_2O_3 film), the reverse current in the entire range of investigated voltages did not exceed 2 μA. The rectification coefficient at $T = 301$ K was $\sim 10^2$ (at voltages $|V| = 2$ V).

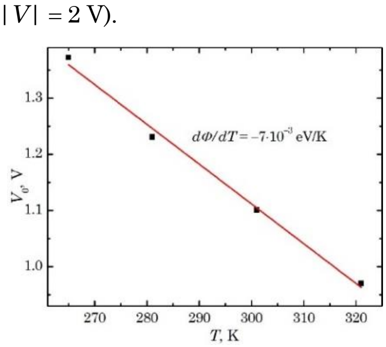

Fig. 3 – Temperature dependence of voltage V_0 to determine the temperature coefficient of change in barrier height $d\Phi/dT$

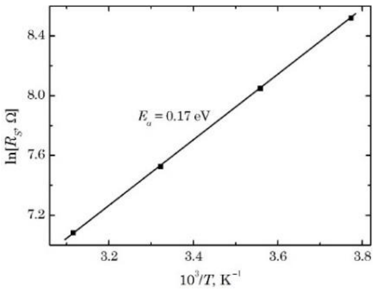

Fig. 4 – Temperature dependence of series resistance R_S to determine the activation energy of electrical conductivity E_a

Extrapolation to the voltage axis of the *I-V* characteristics during direct displacement (from straight-line sections) allows to find the cut-off voltage V_0 and estimate the height of the energy barrier $qV_0 \approx q\Phi$ of the n-Mn_2O_3/p-InSe heterojunction. When the temperature increases from $T \approx 265$ K to $T = 321$ K, the value of V_0 decreases linearly from 1.37 V to 0.97 V (see Fig. 3). The temperature coefficient $d(q\Phi)/dT$ in the temperature range of the studies is $7 \cdot 10^{-3}$ eV/K. The order of magnitude of $d(q\Phi)/dT$ agrees well with the temperature coefficient of change in the height of the energy barrier at indium selenide heterojunctions [16] due to the increase with increasing temperature in the concentration of intrinsic charge carriers and the effective density of N_C and N_V states in the conduction band and in the valence band, respectively.

In the n-Mn_2O_3/p-InSe structure, the high-resistance materials are both the Mn_2O_3 film and, due to the anisotropy of the properties, the p-InSe substrate. With the current direction perpendicular to the plane of the layers of the p-InSe crystal, a significant series resistance R_S may occur in the heterojunctions. The change in the angle of inclination of the straight sections of the straight branches of the *I-V* characteristics is a consequence of the decrease with increasing temperature of the series resistance R_S of the heterojunction. Determined from the graphical dependence $R_S = f(10^3/T)$ (see Fig. 4), the activation energy of conduction is $E_a \approx 0.17$ eV. The obtained value does not correspond to the depth of the band gap of the main energy levels for InSe [21] and best agrees with the activation energy of electrical conductivity in n-Mn_2O_3 thin films ($E_a \approx 0.11$ eV) [20]. In n-Mn_2O_3/p-InSe heterojunctions, the n-Mn_2O_3 film makes the main contribution to the series resistance R_S. The evaluation of the resistance of the n-Mn_2O_3 film in the structure, taking into account the geometry of the samples and the current flow lines, confirms the data of the temperature dependence of R_S.

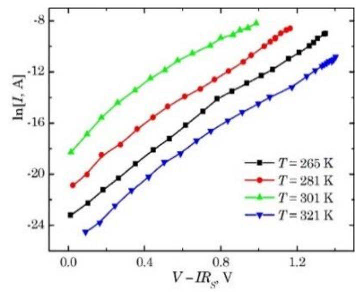

Fig. 5 – Dependencies of $\ln I = f(V - I \cdot R_S)$ at direct bias of n-Mn_2O_3/p-InSe heterojunction

For a correct analysis of the mechanisms of current formation on heterojunctions, which are made using high-resistance semiconductors, it is necessary to take into account that part of the external applied voltage falls on the series resistance $I \cdot R_S$. To determine the mechanisms of current flow through the energy barrier at direct bias of the n-Mn_2O_3/p-InSe heterojunction, the *I-V* characteristics were analyzed in the coordinates $\ln I = f(V - I \cdot R_S)$ (see Fig. 5).

The rectilinearity of the dependences $\ln I = f(V - I \cdot R_S)$ with an angle of inclination to the voltage axis that does not change with temperature is characteristic for the implementation of the tunneling mechanism of direct current flow, which is described by the exponential dependence $I = B \cdot \exp(a(V - I \cdot R_S))$. The tunnel current is described by the expression [22]:

$$I = B \cdot N_t \exp[-\frac{4(2m^*)^{1/2} q^{1/2} (\varphi_{k1} - (V - IR_S))}{3\hbar H}], \qquad (3)$$

where B is a constant, N_t is the concentration of states in the conduction band of p-InSe to which tunneling is carried out, m^* is the effective mass of electrons in the conduction band of the Mn_2O_3 film, φ_{k1} is the height of the energy barrier from the side of the Mn_2O_3 film through which electron tunneling takes place, H is the thickness of the energy barrier in the Mn_2O_3 film.

The result of approximation of experimental data using formula (3) are shown in Fig. 1 by solid curves.

The reverse *I-V* characteristics of the n-Mn_2O_3/p-InSe heterojunction in the temperature range $T = 265 \div 321$ K at reverse voltages correspond well to the expression for the tunnel current:

$$I = a_0 \exp(-b_0(\Phi - (V - IR_S)^{-1/2})), \qquad (4)$$

where a_0 is a parameter that depends on the probable electron filling of the energy levels in the conduction band of p-InSe from which tunneling occurs, b_0 is determined by the dynamics of changes in current relative to voltage.

According to the formula (4), the *I-V* characteristic of the n-Mn_2O_3/p-InSe heterojunction in coordinates $\ln I = f(\Phi - (V - I \cdot R_S)^{-1/2})$ is linear (see Fig. 6) at voltages $-2\text{ V} < V < 0.1$ V The reverse current in the n-Mn_2O_3/p-InSe heterojunction is formed by the tunneling of electrons from the conduction band of p-InSe to the conduction band of n-Mn_2O_3 through the barrier caused by ΔE_C.

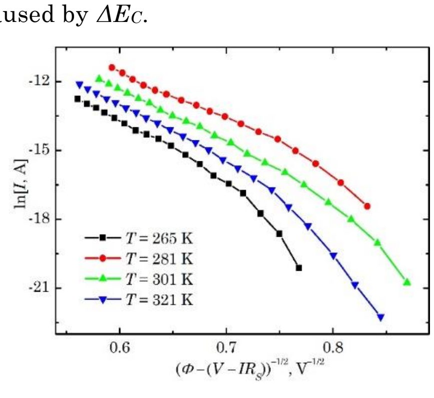

Fig. 6 – Dependence of $\ln I = f(\Phi - (V - I \cdot R_S)^{-1/2})$ at reverse bias of n-Mn_2O_3/p-InSe heterojunction

Upon irradiation of the n-Mn_2O_3/p-InSe heterojunction from the side of the Mn_2O_3 thin film, the spectral sensitivity for the quantum efficiency covers the range of photon energies from 1.2 eV to 3.2 eV. It is characterized by two maxima at energies of 1.25 eV and 2.1 eV (see Fig. 7). The fundamental absorption of radiation in the region of *p*-InSe determines the long-wavelength edge for photosensitivity at the energy of light $h\nu = 1.2$ eV. For non-main charge carriers (electrons) generated by light in *p*-InSe in the conduction zone, there is an obstacle in the form of an energy barrier for their transition to the conduction zone of *n*-Mn_2O_3, which arises due to the difference in the electron affinities of the contacting materials. However, the barrier thickness in the *n*-Mn_2O_3 film decreases when the heterojunction is illuminated due to the displacement of the electrical transition in the forward direction. The probability of tunneling through it of photogenerated electrons in *p*-InSe increases.

At energies of light commensurate with the band gap of Mn_2O_3 films, absorption occurs in the Mn_2O_3 frontal layer of the heterojunction. At the same time, an increase in photosensitivity at $h\nu \approx 2.1$ eV is observed on the spectral dependence of the quantum efficiency. Due to the polycrystalline nature of *n*-Mn_2O_3 thin films, the photosensitivity peak of the *n*-Mn_2O_3/*p*-InSe heterojunction at $h\nu \approx 2.1$ eV is less sharp compared to the peak at $h\nu \approx 1.25$ eV. In *n*-Mn_2O_3 films, unlike single-crystal materials, radiation with energy $h\nu < E_g \approx 2.12$ eV is partially absorbed at the grain boundaries. For holes generated by light in the *n*-Mn_2O_3 film, there are no obstacles to their transition (separation) to the *p*-InSe region.

The full width for the spectrum of the relative quantum efficiency of the *n*-Mn_2O_3/*p*-InSe heterojunction at half-height $\delta_{1/2}$ is ~ 1.4.

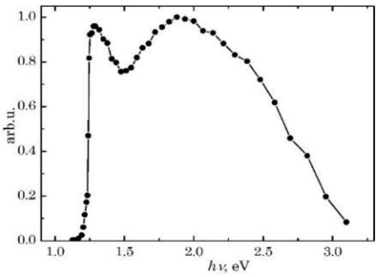

Fig. 7 – Spectral characteristics of the quantum efficiency of *n*-Mn_2O_3/*p*-InSe heterojunction

4. CONCLUSION

Photosensitive *n*-Mn_2O_3/*p*-InSe heterojunctions in the visible region of the spectrum were produced using the spray pyrolysis method of thin Mn_2O_3 films on *p*-InSe substrates. The rectification of the current at the resulting heterojunctions is due to the energy barrier that arises at the contact potential difference $q\Phi \approx 1.2$ V. When forward and reverse currents flow through the *n*-Mn_2O_3/*p*-InSe heterojunction, the main role is played by the processes of electron tunneling through the energy barrier in the conduction band of *n*-Mn_2O_3, which is formed due to the difference in electron affinities $\Delta\chi = 0.15$ eV between Mn_2O_3 and InSe. At forward bias, the probability of tunneling increases due to a decrease in the thickness of the barrier in the *n*-Mn_2O_3. At reverse bias, the tunneling current is caused by an increase in the concentration of electrons in the *p*-InSe.

The photosensitivity spectrum in the range of energies of 1.2÷3.2 eV makes it possible to effectively use *n*-Mn_2O_3/*p*-InSe heterojunctions as radiation photoreceptors.

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Електричні властивості і фоточутливість гетеропереходів *n*-Mn_2O_3/*p*-InSe, виготовлених методом спрей-піролізу

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Досліджені умови нанесення методом спрей-піролізу при температурі 350 °C тонких напівпровідникових плівок Mn_2O_3 на підкладки з кристалічного шаруватого напівпровідника *p*-InSe для створення анізотипних гетеропереходів *n*-Mn_2O_3/*p*-InSe. InSe є перспективним матеріалом для фотоелектроніки. Використання плівки Mn_2O_3, яка є прозорою в області максимальної фоточутливості InSe, дозволяє ефективно експлуатувати оптичні властивості InSe при створенні різних напівпровідникових пристроїв. Перевагою використання шаруватих напівпровідників при виготовленні гетеропереходів є те, що якісні інтерфейси отримуються навіть при значному неспівпаданні параметрів кристалічних граток вихідних матеріалів. Це значно розширює вибір матеріалів гетеропереходів. Проведені виміри електричних та фотоелектричних параметрів гетеропереходів *n*-Mn_2O_3/*p*-InSe та запропоновано теоретичні моделі, що їх описують. Побудовано графічні залежності *I-V* характеристик, послідовного опору, висоти потенційного бар'єру та фоточутливості. Встановлено, що дані гетеропереходи є фоточутливі та володіють випрямляючими властивостями. Використовуючи енергетичні параметри вихідних матеріалів, побудовано енергетичну діаграму гетеропереходу, яка дозволяє провести аналіз фізичних процесів в отриманих гетеропереходах. За температурною залежністю як прямих, так і зворотних вольт-амперних характеристик встановлена динаміка зміни з температурою енергетичних параметрів гетеропереходу, а також механізми протікання струму крізь гетероперехід. Проаналізована спектральна фоточутливість гетеропереходу.

Ключові слова: Селенід Індію, Mn_2O_3, Гетероструктури, Спрей-піроліз, Вольт-амперні характеристики, Фоточутливість.