

FLEXIBLE PHOTOCELLS BASED ON LAYERED $A^{III}B^{VI}$ SEMICONDUCTOR COMPOUNDS

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Abstract- The main characteristics and parameters of flexible thin-layer solar photovoltaic cells, created on the basis of contact of single crystals of layered $A^{III}B^{VI}$ compounds doped with rare-earth elements (erbium and holmium) have been investigated. The possibility of increasing their sensitivity by a purposeful variation of the amount of impurity introduced and the thickness of the contacting layers is shown.

Keywords: Photoelectric Converters, Anti-Isotopic Heterojunction, Layered Single Crystals, Rare Earth Element, Holmium, Erbium, Photosensitivity, Stability, Reproducibility.

I. INTRODUCTION

The problems associated with the field of hydrocarbon power-engineering cause an urgent need to find a viable alternative to non-renewable energy sources. One of the indispensable types of such energy is the energy of solar irradiation. Great potential and inexhaustible energy of the Sun attracts more and more attention.

At present various methods and devices exist to convert the energy of solar irradiation into electrical energy. Among them the most effective both from the energy and ecological points of view are photoelectric methods and semiconductor photoelectric converters based on them, which allow direct and single-step energy transfer.

In various fields of science, technology, production and life, especially in the field of space technology and portable devices, solar batteries are used widely and successfully. Despite the fact that the energy received in this way is much more expensive, in conditions of limited natural resources, environmental problems and in particular, global warming, the use of solar energy by mankind on the Earth's surface and near-Earth space in the near future can make a significant competition of hydrocarbons. In addition, photovoltaic solar energy converters (solar photovoltaics) can be the only and sufficient source of power supply also in geostationary satellites, portable terrestrial electronic devices.

The achievement in this field of power-engineering, in turn is inextricably linked with the development of physics and technology of semiconductor materials, as well as various types of contact structures (metal-semiconductor contact, homo- p-n-junctions, heterojunctions) based on them. The first highly efficient heterostructured solar battery with the use of gallium and arsenic was created in 1970, and in 1995 in experimental and space laboratories the first samples of thin-film solar cells appeared. A few years later, flexible superlight solar panels, photocells were created on copper, indium and gallium selenides, effectively absorbing solar rays. Simultaneously with the use of prototypes, several demonstration devices have been developed, etc.

To improve the efficiency of photovoltaic solar panels, various technological and design techniques are successfully developed and applied at present. These include: the search and use of semiconductors with most suitable width of the band gap for the conversion of solar radiation; purposeful control of the properties of semiconductor materials and structures based on them by the optimal addition of various impurities; transition from homogeneous to heterogeneous semiconductor structures; creation of photocells based on flexible thin-layer contacting pairs, etc.

However, depending on the purpose and conditions of operation of photovoltaic solar converters often come to the fore the features of their design, the spectral region of sensitivity, durability and stability in relation to the physical and chemical impacts of the environment, surface properties than the cost, power and magnitude of the efficiency coefficient. Therefore, the search for semiconductor materials with appropriate physical properties and the study of their photoelectric properties, in addition to modern semiconductor physics and semiconductor electronics, is also one of the topical issues of solar energetics. One such material can be single crystals of $A^{III}B^{VI}$ compounds with a layered structure (indium and gallium monoselenides).

II. FORMULATION OF THE PROBLEM

The appropriate value of the width of the forbidden band, significant photosensitivity in a wide range of the optical spectrum up to a temperature of 350 K [1-3]; the possibility of purposeful control of photoelectric parameters and characteristics by doping them with rare-earth elements (REE), the creation of various contact structures [4-9] and ultra-thin flexible optoelectronic devices [10-12] on their basis by simpler technological methods; easy cleavability to thin layers, high chemical resistance, atomically mirror-like surface make single crystals of A^{III}B^{VI} compounds with a layered structure promising materials for the creation of high-efficiency flexible thin-layer photoelectric converters of solar energy [13, 14]. Elsewhere [15, 16] it was reported on the study of some aspects of the electronic properties of heterostructures created by optical contact of A^{III}B^{VI} semiconductor compounds with a layered crystal structure with various semiconductors, and in [17-21] the possibility of purposeful control of the electrical, photoelectric, luminescent properties of these materials by doping with various rare-earth elements (REE) were demonstrated.

In present paper we attempted experimentally investigate the electronic (electrical and photoelectric) properties of heterostructures, based on thin flexible layers of pure and slightly doped (with the contents of the impurity introduced $N \leq 10^{-1}$ at.%) with certain REEs, in particular, holmium (Ho) and erbium Er) single crystals of indium (n-InSe) and gallium (p-GaSe) monoselenides and identify the possibility of creating on their basis flexible thin-layer solar photovoltaic cells with increased sensitivity

III. OBJECTS AND METHODS OF RESEARCH

The objects of the investigations were anisotropic heterostructures p-GaSe<Ho>/n-InSe<Ho>, p-GaSe<Er>/n-InSe<Er>, p-GaSe<Ho>/n-InSe<Er>, p-GaSe<Er>/n-InSe<Ho>, p-GaSe<Er>/n-InSe and p-GaSe/n-InSe<Er>, p-GaSe<Ho>/n-InSe, p-GaSe/n-InSe<Ho> and p-GaSe/n-InSe created using the technology, described elsewhere [15], for different contents of the introduced impurity ($N \approx 0, 10^{-5}; 10^{-4}; 10^{-3}; 10^{-2}$ and 10^{-1} at.%). GaSe and InSe compounds were synthesized by the joint fusion of composite components (metallic Ga or In and granular Se) of high purity (99.999%) in evacuated quartz ampoules under a specially chosen temperature regime.

Doping was carried out by introducing into the charge mixture in the right quantity of powdered metallic REE of 99.999% purity before the synthesis process. The choice of Ho and Er as impurities was determined with their individual physico-chemical features [22-24]. Single crystals were grown from the synthesized substance by a modified Bridgman method [25-26]. The concentration and mobility of majority charge carriers at 300 K were $p_0 \approx 10^{14} \text{ cm}^{-3}$, $\mu_p \approx 90 \div 100 \text{ cm}^2/\text{V}\cdot\text{s}$ and $n_0 \approx 5 \cdot 10^{14} \text{ cm}^{-3}$, $\mu_n \approx 900 \div 1000 \text{ cm}^2/\text{V}\cdot\text{s}$ for p-GaSe and n-InSe single crystals, respectively.

The structure, phase and elemental composition of the obtained ingots, the state of the surface along the plane of natural layers [27] of the investigated samples were determined by carrying out complex structural analyzes.

For this purpose, modern measuring systems and methods (DSC-910, ADVNCE-8D, SINTECP 21, DRON-4-07 with CuK α - radiation at step 0.05 and 8-135° angle range, Zeiss SEMS with energy dissipation analyzer) were used. As a result of such a complex structural-phase and composite analysis it was established that both groups (p-GaSe and n-InSe) of the obtained samples have a single-crystalline structure. Regardless of the content and chemical nature of the introduced REE impurity the diffraction patterns of the indium monoselenide crystals are indexed in the rhombohedral (γ -polytype) syngony, refer to the space group $R_{zone}(C_{3x}^5)$ with elementary cell periods $a \approx 4.003 \text{ \AA}$, $c \approx 24.955 \text{ \AA}$, and the diffraction patterns of the gallium monoselenide crystals in hexagonal syngony (ε -polytype) and belong to the space group $D_{3h}^1(P\bar{6})$ with elementary cell periods $a \approx 3.744 \text{ \AA}$, $c \approx 15.902 \text{ \AA}$ [28-30]. In the ingots obtained no phases of substitution of selenides, oxides and free lanthanides, as well as heterogeneities and extraneous phases on their surfaces were found. Separate thin layers were cleaved from different parts of the same, as well as from different monocrystalline ingots. The type of conductivity was determined from the signs of the thermo-emf and the Hall coefficient.

The heterostructures studied were created by uniformly compressing to each other thin plates of p-GaSe<REE> and n-InSe<REE>, p-GaSe<REE> and n-InSe, n-InSe<REE> and p-GaSe, as well as p-GaSe and n-InSe in a special holder. The thickness and transverse dimensions of the contacting layers having a shape of the rectangular parallelepiped were $\sim 25 \div 200 \text{ \mu m}$ and $2 \div 6 \text{ mm}$, respectively. The electrical contacts were created by soldering metallic Sn and In without flux on the open air. The geometry of the structures studied was chosen so that when measuring, the current and the light beam are directed along the "C" axis of the crystals (in a direction perpendicular to the natural layers of the single crystals) and perpendicular to the contact plane of the heterostructure. The measurements were carried out in the temperature range $77 \leq T \leq 350 \text{ K}$, at wavelengths (λ) and light intensities (Φ) $0.20 \div 2.00 \text{ \mu m}$ and $\Phi \leq 5 \cdot 10^2 \text{ lx}$, respectively. Experimentally were registered the dark static current-voltage characteristics (CVC) of the investigated heterostructures, the spectral distribution of the photo-emf in them at different temperatures (T), the wavelengths of light (λ), illuminations (Φ), and the impurity content (N).

IV. EXPERIMENTAL RESULTS

It is established that all of the investigated structures under the conditions considered by us have a rectifying character [31], with forward direction, when the positive polarity of the applied external electric voltage is applied to the p-GaSe <REE (or p-GaSe) layer.

The rectification coefficient [31] at voltages $U \approx 4 \div 5 \text{ V}$ is $\sim 4 \div 16$ for different structures, depending on the thickness and doping level of the contacting pairs. At $U \geq 0.5 \text{ V}$ the exponential course of the straight branch of the current-voltage characteristic is replaced by a linear one.

At 300 K, in the first approximation, the cut-off voltage of the considered heterojunctions corresponds to the height of the potential barrier (~0.8÷1.1 V) and the residual resistance is ~10⁵÷3·10⁶ Ω [31]. With the change in the content of the introduced REE impurity, the diode character of the structures under study is preserved, but the numerical value, as well as the stability and reproducibility of its individual parameters (current cut-off voltage, residual resistance, rectification coefficient, etc.) vary. In particular with increasing *N*, at first (at *N*≤10⁻³ at.%) the resistance of these materials increases by almost an order of magnitude relative to the initial one (taking place for the lowest-resistivity pure crystals), and then decreases again (at *N*>10⁻³ at.%), and at *N*≥10⁻¹ at.% becomes even less than the initial one (taking place in structures based on the lowest-resistivity pure p-GaSe and n-InSe crystals).

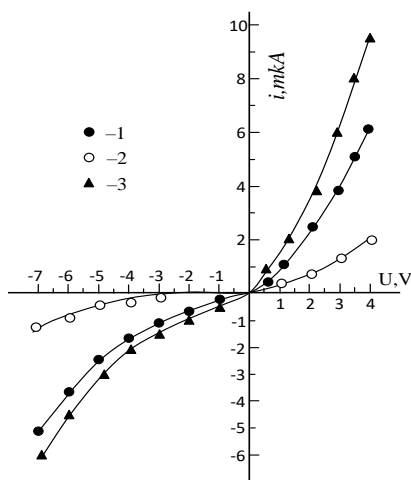


Figure 1. CVC of p-GaSe<Er>/n-InSe<Ho> structures for various *N*, at.%.: 1-0; 2-10⁻⁵; 3-10⁻³; 4-10⁻¹, *T* = 200 K

With the change of *N* non-monotonically change (first worsen, and then improve with respect to the initial value) also the stability and reproducibility of the electric and photoelectric parameters of the structures studied. Capacitive measurements have shown that all investigated heterostructures are sharp ones [31]. The photoelectric properties of the investigated structures were studied in the photovoltaic regime [31]. In all of the cases considered the p-GaSe<REE> (or p-GaSe) component in contact is charged negatively with respect to the second contacting component (n-InSe<REE> or n-InSe). The measurements were carried out in a pulsed mode of illumination. In this case, the pulse frequency varied within 100-150 Hz. It turned out that the maximum value of the photovoltage is observed when the investigated structures are illuminated from the side of p-GaSe<REE> (or p-GaSe) layer.

When illumination is carried out from the side of narrow-band semiconductor - n-InSe<REE> (or n-InSe) layer, the nature of the photosensitivity spectrum is completely determined by the absorption in a narrow-gap semiconductor and is similar to the spectrum of a photoresistor manufactured on its basis.

At that, the difference is only in the fact that in the short-wavelength region of the spectrum (in the wavelength region λ≤0.50 μm) a discontinuity is observed. When illuminated from the side of wide-band component (p-GaSe<REE> or p-GaSe), two regions are observed in photo-emf spectrum. The first (short-wave region) corresponds to light absorption in the layer of wide-band (p-GaSe<REE> or p-GaSe), and the second (long-wave region) corresponds to narrow-band component (n-InSe<REE> or p-InSe) of the contacting pair. Under optimal conditions, the open circuit voltage (*U_{ocv}*) and the short-circuit current (*i_{scc}*) [31] in the structures under study are ~0.34 V and ~42 μA/cm², respectively.

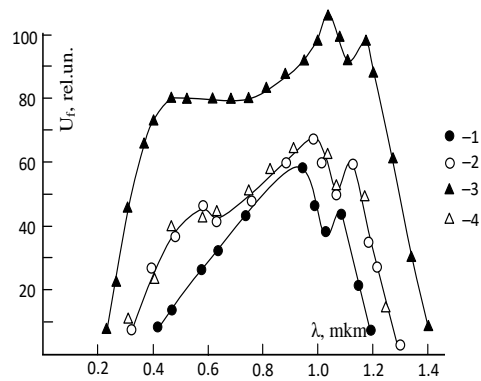


Figure 2. The photo-emf spectrum in p-GaSe<Er>/n-InSe<Ho> structures, *N*, at.%.: 1-0; 2-10⁻⁵; 3-10⁻³; 4-10⁻¹ *T* = 200 K; Φ = Φ_{max}
1- when illuminated from the n-InSe <Ho> side
2-4- when illuminated from the p-GaSe<Er> side

It has been established that in both cases (both under illumination from the side of wide-band layer and from the side of narrow-band semiconductor layer), when doping with REE *N*≈10⁻⁵ at.%, at 77 K the magnitude and width of the spectral distribution increase with respect to those occurring for structures based on pure crystals of these semiconductors. However, at that the stability and reproducibility of the parameters and characteristics of the structures under study are deteriorated. When illuminated from p-GaSe<REE> side in structures with *N*≈10⁻⁴ at.%, the photosensitivity spectrum significantly expands and almost covers the wavelength range from ~0.25 μm to 1.45 μm. But at the same time, the stability and reproducibility of the parameters and characteristics of the structures under investigation are further deteriorating. With further increase in the content of the introduced REE impurity to *N*≈10⁻¹ at.%, the magnitude of the photo-emf and the width of its spectral distribution decrease, approaching to the original one. But, at that, the stability and reproducibility of the parameters and characteristics of the structure under study are improving reaching their maximum.

With a decrease in the thickness of the contacting pairs, in addition to changing (improving) their electrical and photoelectric parameters, also change mechanical properties – increases flexibility and the latter gives them a significant practical advantage.

V. DISCUSSION

The analysis of the obtained experimental results allows us to assume that the tunneling mechanism of current flow dominates in the heterostructures under study. However, at that it is also necessary to take into account the presence of local centers in the forbidden band and partial disorder of the contacting semiconductor crystals. The unequal effect of light from the side of wide-band and narrow-band semiconductor on the current-voltage characteristic and the value U_0 for the heterostructures under study at direct biases are related to the different photosensitivity of the contacting materials. Since p-GaSe<REE> crystals (or p-GaSe) have a relatively small photosensitivity at $T > 300$ K in comparison with n-InSe<REE> (or n-InSe) crystals, illumination of the studied structures from the side of p-GaSe<REE> (or p-GaSe) has little effect on U_0 and C-V characteristic. The broadening of the spectral distribution of the photosensitivity of the investigated heterostructures under illumination from the wide-band contacting component is, to all appearances, due to the "window effect" [31], and the sharp decrease in photosensitivity from the short wavelength side - with a small value of the diffusion length of photocarriers in both contacting semiconductors in the direction perpendicular to the natural layers of the crystal in comparison with the thickness of individual layers.

The dependence of the characteristics and values of the main parameters, as well as their stability and reproducibility on the content of the introduced impurity, is connected with a change in the degree of spatial inhomogeneity of the single crystals of the semiconductors under study upon doping.

Thus, it can be concluded that on the basis of doped with REE (Ho or Er) single crystals of gallium and indium monoselenides it is possible to create flexible thin-layer heterostructural solar photoconverters with increased sensitivity. Although the efficiency, the generated power and the area of the working surface are not large, but they can be useful for space and other portable devices, as well as for the development of the physics of solar photoconverters and layered $A^{III}B^{VI}$ semiconductor compounds.

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