

## Film heterophotocell with nanocluster subsystem

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**Abstract.** The effects associated with changes in the properties of a film heterojunction as a result of introducing a nanocluster subsystem into it are considered. Such a modified heterojunction exhibits spectral-inversion photovalve effect. A design of a photocell containing two series-connected opposite  $p$ - $n$  junctions, photoactive in different spectral regions, is proposed.

**Keywords:** heterojunction, film, nanocluster, subsystem, properties, kinetics, irradiation, diffusion, photovalve effect.

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### 1. Introduction

The study of properties of heterojunctions (HJs) deserves attention for solving problems of functional diagnostics of new generation devices. Such studies are relevant for developing effective photovalve elements [1–3].

The task of the work is to consider a possibility of manifestation of new properties of film HJs of the  $p\text{Cu}_2\text{S}-n\text{Si}$  type, into which a cluster raster of subcolloidal dispersion in the form of a nanocluster subsystem (NCS) is introduced, and to determine the optimal operating modes of such a heterophotoelement.

### 2. Experimental

Transition from an individual atom to a macrostate is characterized by formation of structures such as quasi-molecular clusters and subcolloidal and colloidal nanoclusters (NCs), which are extensive in nature, since they consistently change their properties up to the properties of the corresponding macrosubstance [4]. The latter is very important to consider when using quasi-metallic NCs as a doping element for individual materials and heterostructures (HS) [5–10].

Quasi metallic centers (QMCs) in crystals (in particular, dielectric and ionic ones) with nanometer sizes (from 1 to 5 nm) can be formed using special technological methods (irradiation, heat treatment, action of optical radiation). Such QMCs represent an intermediate state between simple point defects (such as aggregates of F-centers) and macroscopic metal colloids.

Influence of the NCs of different sizes on the optical and some other properties of crystals (“red shift” of the color of the matrix material when the centers are enlarged,

*etc.*) has been studied both theoretically and experimentally [11–13]. An analogy with different stages of a photographic process in silver halide materials and on semiconductors would be appropriate. Moreover, the size of NCs in a single-crystal matrix can be varied quite widely [4, 5].

Before depositing the  $\text{Cu}_2\text{S}$  film on the crystalline silicon substrate, a Si cluster pattern with an island structure was formed on the substrate by ion implantation.

By varying the NC formation rate and the substrate temperature, we were able to implant the cluster centers in the form of a nanocluster subsystem (NCS) of silicon into the  $p\text{Cu}_2\text{S}-n\text{Si}$ -HJ. In this way, HJ of the  $p\text{Cu}_2\text{S}-(\text{Si-NCS})-n\text{Si}$  type was formed. For such HJ, “extensive” effects, in particular the spectral inversion effect, were observed.

It should be noted that the NCS in a heterojunction (HJ) can be also created by implanting transition metals, in particular, Fe, Ni, Co, and Cu [5]. Implantation of transition metals into the surface layer of Si is used to create silicides, to modify the electronic structure and to form magnetic nanostructures. Important aspects of this process are high diffusion rates, formation of deep recombination levels and synthesis of ferromagnetic phases. The latter is a good confirmation that the detected effects are to a greater extent due to the presence of NCS, and are not caused by the NCS material.

### 3. Results and discussion

The HJ considered in the work is formed from a Si NC. This material was chosen due to its advantage in terms of purity class over other materials that were at our disposal.

A common property of gate photovoltaic cells based on Si, Ge, Se, CdS and other materials is preservation of

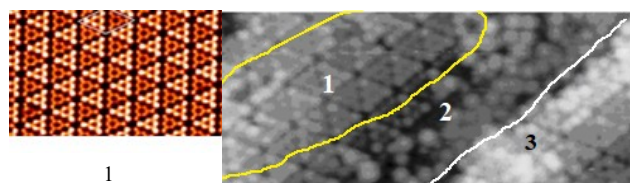
unchanged photovoltage polarity in the entire spectral active region, for which, according to the typical scheme of the gate photoeffect, the  $n$ -region is negatively charged with respect to the  $p$ -region under light illumination. At the same time, a possibility of reversing the polarity of the gate photovoltage by changing the illumination wavelength has been established.

The photoelectric element proposed in this work, which includes HJ with NCS, opens up new opportunities in solving a number of practical issues in optoelectronics related to the control and measurement of color temperatures of radiation sources, recognition of color images, etc.

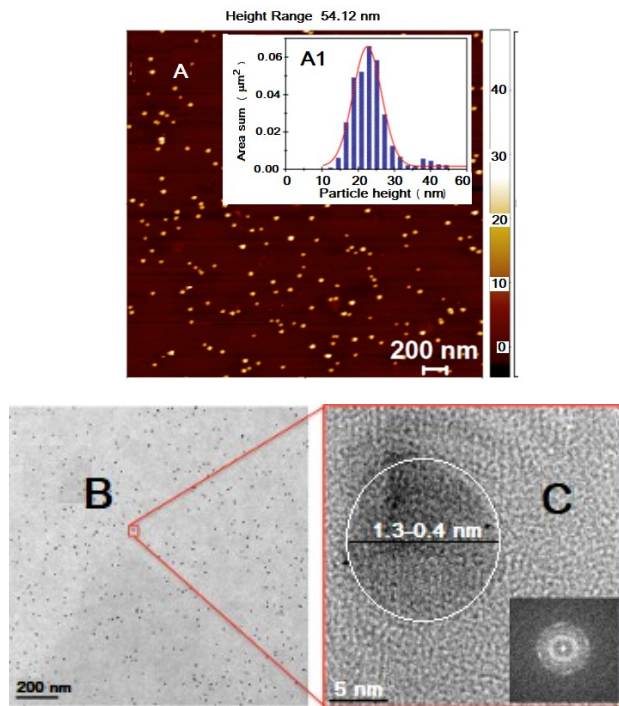
One of the main recommendations is to introduce various impurities (Fe, Ni, Co, etc.) into the  $p$ - $n$  junction region to provide traps and recombination centers of non-equilibrium electron-hole pairs. Another recommendation is to make a photovoltaic cell based on a complex HJ from different materials. This HJ contains two sequentially connected opposite  $p$ - $n$  junctions photoactive in different spectral regions. In both cases, the photosensitivity of the device is low, and the spectral distribution of the photovoltaic effect is strongly asymmetric relative to the point of inversion of the polarity of the photovoltage. In the first variant, this is apparently due to the insufficiently high efficiency of the specified impurities as recombination centers. In the second variant this is due to the complication of optical-photoelectric conditions in a HJ with a sequential chain of transitions.

We have considered a possibility of creating a gate spectral-inversion heterophotocell in a configuration  $p\text{Cu}_2\text{S-Si(NCP)-nSi}$ , which structurally represents a combination of a half-film  $p$ - $n$  heterojunction located at the contact between a  $p$ -type  $\text{Cu}_2\text{S}$  film and an  $n$ -type Si crystal, with an ultrafinely dispersed Si cluster raster (NCS), distributed in the junction plane. The cluster raster forms an "island" structure, which is organically included in the heterojunction structure and, in particular, is embedded in the copper sulfide film.

The technology of obtaining a spectrally inverted  $p\text{Cu}_2\text{S-nSi-HJ}$  photovoltaic cell is generally similar to that described in [4], the only difference being that manufacture of samples begins with implementation of



**Fig. 1.** High-resolution microphotograph (the image size  $270 \times 195 \text{ \AA}^2$ ) of a Si(111) crystal surface: 1 – Si(111)- $7 \times 7$  surface region in the filled state (the STM data show 12 adatoms and 6 rest atoms per a unit cell ( $7 \times 7$ ) [12]); 2 – micrographic NC silicon raster on the c-Si surface. It is a hierarchical two-dimensional large-scale texture combining microcrystalline structural elements and nanoclusters. The nanocluster pattern is created by metal-assisted etching, ion bombardment, and laser micro/nano structuring [13].



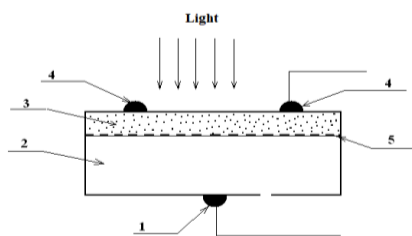
**Fig. 2.** Formation of NCs on a solid surface. Panel A is an AFM micrograph illustrating surface coverage of the Si NCs and their size distribution (inset, A1) [14]. Panels B and C are TEM micrographs of the Si NCs on the substrate [15].

a metal-cluster raster of subcolloidal dispersion with the size of individual particles not exceeding  $20 \text{ \AA}$  on the surface of a Si washer (at a temperature of  $100^\circ\text{C}$ ). The HJ clustering operation was carried out by thermal evaporation in vacuum ( $\sim 10^{-6}$  Torr) or by pyrolytic decomposition of compounds, in a mode that ensures formation of a subcolloidal metal monolayer on the control substrate, which is in the same conditions as the main substrate. This monolayer is clearly distinguishable with an electron microscope (Fig. 1).

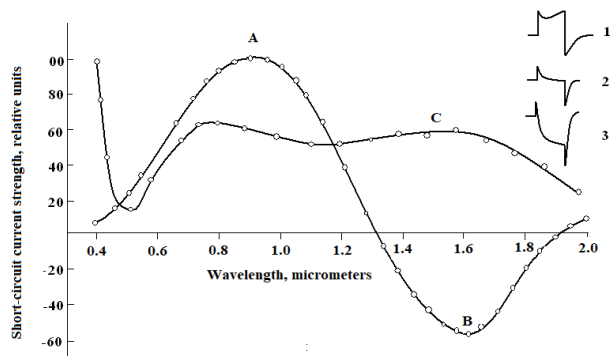
Typically, sputtering of semiconductor materials is more difficult compared to metallic materials. However, the AFM results presented in Fig. 2 (panel A) indicate that Si nanoparticles with sizes of 10 to 20 nm are relatively easily deposited on the c-Si surface [14]. Fig. 2 (panels B and C) also present micrographs obtained using high-pressure transmission electron microscopy [15].

A schematic representation of a spectral inversion heterophotocell is shown in Fig. 3. The most important element of this design is a nanocluster raster. In the nanocluster raster, the NC dispersion degree is of particular importance. The result is a spectral inversion photocell with HJ having a  $(p\text{Cu}_2\text{S-(Si-NCS)-nSi})$  NCS. Such a photocell has a clearly defined sensitivity distribution, which is shown in Fig. 4.

Operation of a spectral inversion photocell of the  $p\text{Cu}_2\text{S-(Si-NCS)-nSi}$  type is ensured by combining two essentially different mechanisms of the gate photoeffect in one HJ, separated by their spectral active regions. The first mechanism implements the well-known scheme of



**Fig. 3.** Schematic diagram of a spectral-inversion heterophotocell  $p\text{Cu}_2\text{S-Si(NCS)-nSi}$ : 1, 4 – thin-layer contacts; 2 – silicon crystal; 3 – copper sulfide film; 5 – subcolloidal nanocluster array (NCS).



**Fig. 4.** Spectral distribution of the inversion photovalve effect of HJ with NCS – ( $p\text{Cu}_2\text{S-Si(NCS)-nSi}$ ): A and B – regions of the normal and anomalous effect, respectively, C – optical absorption spectrum of the  $\text{Cu}_2\text{S}$  film. Plots (top right) of the photo-response kinetics: 1 – in the region of the normal effect, 2 – at the inversion point, 3 – in the region of the anomalous effect.

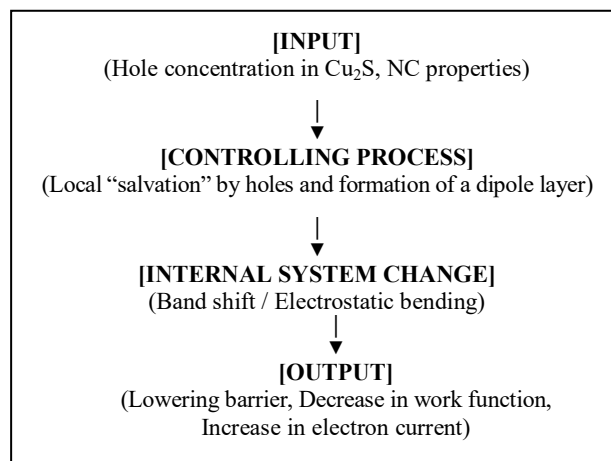
the gate photoeffect in the  $p$ - $n$  junction (Fig. 4A), according to which the  $n$ -region (silicon) is negatively charged under illumination. The spectral distribution of the photovoltaic effect in this case (Fig. 4A, 4B) corresponds to photoactive absorption of light in both materials  $\text{Cu}_2\text{S}$  and Si, which is facilitated by the  $p\text{Cu}_2\text{S-nSi}$  – HJ zone diagram favorable for the photocell. The second mechanism (Fig. 4B) operates in clustered areas of HJ, *i.e.* where NCs are located. Its spectral characteristic corresponds to absorption of light by nanoparticles (Figs. 4B, 4C).

The result of the action of light is generation of photovoltage (photocurrent) in a direction opposite to that caused by an action of the first mechanism, *i.e.* Si is positive under an action of light from this spectral region, and  $\text{Cu}_2\text{S}$  is negative.

The mechanism of photoelectric effect in NCs is much more complex and does not reduce to a simple linear or single-stage “input-output” scheme. Although at the conceptual level, there is an optical signal at the input (photons) and an electric current at the output (electrons). The “input-output” model assumes a direct, instantaneous and proportional dependence of the result on the perturbation. In NCs (quantum dots), the photoeffect is a multistage quantum process with an internal “memory”, dependence on the size, shape and environment of the NC, and nonlinearity of the system [16].

To explain the principle of “input-output” action for the effect of solvation of nanoparticles (in our case, NCs) by holes (Fig. 4B), the Scheme 1 is proposed.

**Scheme 1.** “Input-Output” for the effect of solvation of NCs by holes.



Explanation and detail of the circuit elements.

The circuit parameters are distributed as follows:

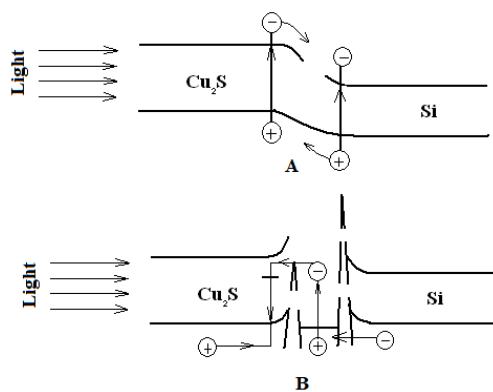
1. Input parameters (Input). Primary input: excess concentration of free holes in the  $\text{Cu}_2\text{S}$   $p$ -film.

2. Transformation process (Black Box/System Operator). Diffusion of holes to the interface boundary and their accumulation around the NC (“salvation”). Formation of a double electric (dipole) sphere at the nanocontact. Change in the macroscopic potential at the interface, which causes a shift of the energy bands (band bending).

3. Output parameters (Output). Energy output: specific reduction in the height of the potential barrier. Functional output: effective reduction in the work function of an electron from NC. Kinetic output: sharp increase in the electron transmission coefficient (emission/transport intensity) into the  $p$ -region of the heterojunction.

The photoelectric effect on NCs changes its electronic structure. Potential wells and energy barriers are formed, and localized electron traps arise, which directly control the transport of current carriers.

At the nanometer scale, the process of electron emission or photocurrent generation depends on the geometric parameters of the NC. When the size of the NC decreases and becomes smaller than 5 nm, movement of electrons is limited in all three dimensions. In this case, the following processes occur: the band gap ( $E_g$ ) of the silicon NC increases (up to 1.5...2.5 eV depending on the NC size.  $E_g = 1.12$  eV for c-Si); the conduction ( $E_c$ ) and valence ( $E_v$ ) bands split into quantum levels; and energy barriers and local energy levels appear. For NCs with sizes up to 3 nm, the quantum energy levels in the conduction band ( $E_c$ ) are located below or above the edges of the bulk substrate bands (within 0.20 eV). This creates specific conditions for charge injection [17].



**Fig. 5.** Band energy diagrams: (A) of HJ free of NCs of the  $p\text{Cu}_2\text{S}-n\text{Si}$  type and (B) HJ with NCs –  $p\text{Cu}_2\text{S}-(\text{Si-NCS})-n\text{Si}$ .

Near the NC, the energy situation also changes significantly. There is a substantial influence of the matrix environment [18]. At the interface between the NC and the matrix environment (e.g.  $\text{SiO}_2$ ), the band diagram becomes complex. Here, the NC functions as a three-dimensional potential well for charge carriers, which is surrounded by high energy barriers of the matrix. This well for the  $\text{SiO}_2$  matrix is equal to 3.1 eV for electrons and 4.7 eV for holes. Due to the small intrinsic capacitance of the NC, the impact of even one electron on the NC creates a powerful energy barrier (a Coulomb repulsion energy arises), which prevents capture of the next electron. A large number of surface states are formed directly near the NC surface. Traps for electrons and holes (unsaturated bonds of silicon or bonds with oxygen) arise creating deep and shallow energy levels within the band gap. These level capture charge carriers and block or enhance current.

Passivation of broken chemical bonds of silicon atoms on the surface of NCs e.g. with hydrogen atoms compensates for localized states from the band gap and equalizes the potential. The accumulated charge in the traps near the NCs creates local electric microfields that distort the energy bands of neighboring matrix regions changing the tunneling conditions (Fowler–Nordheim modes or direct tunneling). Controlled capture by electron traps is used in non-volatile memories. For example, the charge localized in a trap changes the threshold voltage of a transistor. This allows storing a logical bit of information for a long time [19].

According to calculations of the electronic structure of Si NCs by quantum chemistry methods [4], these nanoclusters tend to have a certain excess negative charge in equilibrium. This is a condition for NC stability and further growth up to colloidal sizes. Under such conditions, presence of a high concentration of holes in  $\text{Cu}_2\text{S}$  should lead to a kind of “solvation” of NCs by holes from the  $\text{Cu}_2\text{S}$  film, which results in a decrease in the effective work function of electrons from the NC to the HJ  $p$ -region (Fig. 5).

A photoexcited electron is moved from a NC deep into the  $\text{Cu}_2\text{S}$  film, where, most likely, it is captured on

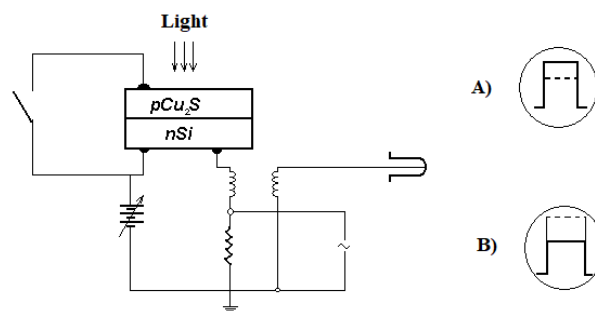
a highly developed system of electron adhesion levels in this quasi-monopolar semiconductor. Simultaneously with the photoexcitation of the first electron in the NC, another one tunnels from the Si valence band *etc.*, i.e., the process that constitutes the essence of the “feed in – feed out” scheme is reproduced.

The spectral activity region of the negative photovoltaic element defines the effective energy depth of the NCs at 0.8 eV. This parameter is of significant importance for spectral inversion of the photoeffect, since it provides tunnel contact with the valence band of silicon. When switching to a cluster raster with large-sized NCs (more than 20 Å), the “feed in – feed out” effect disappears, in other words, the spectral inversion photovoltaic effect is not observed.

This may be explained by extinction of the solvating factors that ensure the necessary “feed in – feed out” mechanism for the operation of the potential barrier profile around the NC, as a result of weakening of the Coulomb field of the large-sized NCs.

Direct confirmation of the mechanism of the spectral-inversion photovoltaic effect discussed above was obtained by parallel measurements of the kinetics of photoconductivity in the base region of the photocell – the Si plate. When the  $p\text{Cu}_2\text{S}-(\text{Si-NCS})-n\text{Si}$  photocell was illuminated with light from the spectral region corresponding to creation of anomalous photovoltaic effect (IRS-3 filter), the photoresponse of the base region of the element was significantly smaller when the photocell was open (idle mode) than in the case of a short circuit (Fig. 6).

For a  $p\text{Cu}_2\text{S}-n\text{Si}$  photocell that does not contain NCs, the behavior in similar situations will be opposite in the entire active region of the spectrum. The results obtained by us are easy to interpret by comparing two mechanisms of the photovoltaic effect – ordinary and “feed in – feed out”. In the latter case, extraction of electrons from silicon on the NCs leads to a decrease in the photocurrent in the base circuit. Naturally, in the short-circuit current mode, such observations are less pronounced due to the continuous restoration of the concentration of non-equilibrium electrons in silicon from the external circuit.



**Fig. 6.** Scheme of observation of the kinetics of photoconductivity (IRS-3 filter) in the low-resistance base layer of the photocell and kinetic diagrams of photocurrent (the dotted line indicates short-circuit mode of the photocell): A)  $p\text{Cu}_2\text{S}-n\text{Si}$  – photocell; B)  $p\text{Cu}_2\text{S}-(\text{Si-NCS})-n\text{Si}$  – photocell.

There are also other considerations that confirm the proposed mechanism of spectral inversion of the photovalve effect in  $p\text{Cu}_2\text{S}-(\text{Si-NCS})-n\text{Si}$  HJ. The fact is that in this situation, it would be possible to assume participation of Schottky microbarriers at the “NC-matrix material” interface in the spectral-inverted effect. These microbarriers create an electric field opposite to the barrier field in the heterojunction and provide an anomalous photovalve effect. However, the latter is unlikely for the following reasons. It is known that significant contact barriers are formed as a result of transition of  $\sim 10^{15}$  electrons from one material to another, which causes a distortion of energy bands. Emergence of Schottky barriers in the place of contact of the NCs with the matrix substance is impossible due to the small NCs size.

Theoretical estimates presented in [4, 5] show that ionization or capture of even one electron in the NCS leads to a sharp change in the position of the Fermi level of the NC by units of eV. On the one hand, this fact significantly complicates further electron exchange between the NC and the matrix material. This is absolutely not enough to distort the energy bands of the matrix crystal in this region due to the small concentration of charge involved in the exchange.

The insignificance of the role of Schottky barriers between the NC and the matrix material also follows from the experimental facts themselves. At NC sizes up to 20 Å, the role of the Schottky barriers should increase up to formation of a mesh structure at the interface. That is, one should expect dominance of the anomalous photovalve effect or even complete transformation of the photovalve effect in the  $p\text{Cu}_2\text{S}-(\text{Si-NCS})-n\text{Si}$  HJ from normal to anomalous one. But this does not actually happen, since the photovalve effect not only does not increase, but, on the contrary, completely disappears with an increase in the NC size to hundreds or more angstroms. For example, the diffusion coefficient of Ni in Si at temperatures of  $\sim 100^\circ\text{C}$  is negligibly small. In other words, atomic doping of Si with nickel during obtaining  $p\text{Cu}_2\text{S}-(\text{Si-NCS})-n\text{Si}$  – photovoltaic cells is effective.

#### 4. Conclusions

A technology for changing the properties of a film HJ of the  $p\text{Cu}_2\text{S}-n\text{Si}$  type by introducing a cluster raster of subcolloidal dispersion in the form of NCS, which provides a smooth band-energy transition profile, is proposed. Such structure is suggested to use as a base for photovalve energy converters. The optimal operation modes of such heterophotocell are determined. The structure of a HJ can be changed by implanting (introducing or depositing) a cluster raster of an island structure at the interface or in one of the HJ layers. This is an effective method of spatial and energetic modification of the HJ properties. This approach is actively used in modern nanoelectronics, photovoltaics, and LED technologies.

Varying the NC formation rate and substrate temperature opens up a possibility of implanting NCs with sizes from 10 to 150 Å. In our opinion, manifestation

of the spectral inversion effect observed in GPs with NCPs is extensive in nature, *i.e.* this effect directly depends on the number of absorbed photons, NCS morphology, defect concentration, and NC size and geometry.

The presented research results deserve attention in solving problems of modern functional diagnostics of devices with NCS as well as in the development of photovalve elements.

The spectral-inversion photodetector described in this work, in addition to its independent application, can be used in designs with nanostructures [3, 4], in particular, when creating optocouplers, such as bichromatic LEDs with controlled emission frequency.

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**Kovalchuk V.V.:** conceptualization, supervision, project administration, writing – review & editing.

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**Dyachok D.O.:** investigation, formal analysis, visualization, writing – original draft.

**Loskutov K.V.:** investigation, resources, writing – original draft.

## Плівковий гетерофотоелемент з нанокластерною підсистемою

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**Анотація.** Розглянуто ефекти, пов'язані зі зміною властивостей плівкового гетеропереходу в результаті введення у нього нанокластерної підсистеми. Такий модифікований гетероперехід виявляє спектрально-інверсійний фотоперехідний ефект. Запропоновано конструкцію фотоелемента, що містить два послідовно з'єднаних *p-n*-переходи зустрічної дії, фотоактивні в різних областях спектра.

**Ключові слова:** гетероперехід, плівка, нанокластер, підсистема, властивості, кінетика, опромінення, дифузія, фотоперехідний ефект.