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SEMICONDUCTOR STRUCTURES, LOW-DIMENSIONAL  
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# Main Features of Photostimulated Ion Transport in Heterojunctions Based on Mixed Ion–Electron (Hole) Conductors and the Model of a Thin-Film Ion Accelerator

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**Abstract**—The main features of photostimulated ion transport in heterostructures based on mixed ion–electron (hole) conductors are considered. It is shown that, using correctly chosen physical factors and a heterostructure of special design, one can implement efficient ion acceleration and thus develop a thin-film ion accelerator.

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## 1. INTRODUCTION

Nanoobjects in the form of thin films provide unique possibilities for implementing photostimulated ion transport in heterojunctions composed of a mixed ion–electron (hole) conductor and a conventional electron (hole) semiconductor (or insulator or metal). Generation of photovoltage in this thin-film heterojunction causes photostimulated ion transport, which was observed for the first time in [1].

The space-charge regions in the aforementioned heterojunction are generally of nanoscale size [2]. Therefore, this heterojunction consists of two thin films. One of them is a mixed ion–electron (hole) conductor, while the other is a conventional semiconductor with electron (hole) conductivity (or insulator or metal). The characteristic film thicknesses range from few micrometers to 40 nm.

The question arises of what new information studies of these objects may yield. The processes occurring during photostimulated displacement of ions in artificially formed (mixed ion–electron conductor)—(conventional semiconductor or insulator or metal) heterojunctions are similar to a certain extent to the processes observed in the case of photostimulated diffusion of a metal into a semiconductor (this process is also referred to as photodoping). The investigations of this phenomenon were thoroughly described in monograph [3] and review [4]. Upon exposure of metal layers (Ag, Cu, Zn, Mg, Ni, Pd, Sn) and a chalcogenide glassy semiconductor (or some other appropriate semiconductors, for example, metal halides) successively deposited on a substrate to photoactive light, the metal diffuses into the chalcogenide semiconductor to form an intermediate photodoped layer. Photostimulated penetration of metal into the chalcogenide semiconductor leads to the formation of an

intermediate photodoped metal layer. The physical and chemical properties of such layers differ significantly. In addition the formation of an intermediate doped layer determines also the conditions for the metal photodiffusion (photodoping).

Photodoping is a promising technique for preparing photoresistive layers for nanolithography and microlithography in production of modern large- and very large-scale integrated circuits [5] and for optical data recording [6, 7].

If the thicknesses of the metal and semiconductor layers that form the photosensitive metal–semiconductor structure are chosen correctly, the metal is completely dissolved in the semiconductor as a result of the illumination and the two initial layers are transformed into one semiconductor layer photodoped with metal [3, 4].

Many different models have been proposed to explain the nature of photostimulated diffusion of metal into semiconductor. Let us consider the most typical and well-justified ones.

Even in the early studies of this phenomenon, in some models the light-stimulated diffusion of a metal into a semiconductor (photodoping of semiconductor with a metal) was considered as a solid-state photochemical reaction [8, 9]. Accordingly, this reaction was considered to be based on its energy efficiency. The interaction between As–S films with different arsenic and sulfur contents and an Ag film under illumination of this photosensitive semiconductor–silver structure was investigated using Raman spectroscopy by Firth et al. [8]. They interpreted the photodoping of this structure as a two-stage photochemical reaction. The Ag and S–S bonds, which are initially present in the semiconductor, interact in the initial stage. In the second stage, the reaction continues between Ag<sub>2</sub>S

and the As–S matrix. A model was proposed, according to which the structure of stoichiometric photodoped glass is formed by  $\text{AsS}_3$  pyramids linked through –S–Ag–S– units.

Note that more than ten years before researchers from the United Kingdom published their basic studies in this field [8, 9], a model of photochemical transformations in photosensitive semiconductor–metal structures based on the studies of the Ukrainian researchers was presented in Romanenko's candidate's dissertation [10].

However, these and other similar models could not explain the generation of photovoltage, which has been experimentally recorded in many photosensitive semiconductor–metal structures exposed to light [3, 4]. The model based on photochemical reaction also could not explain the specific features of photocurrent generation during photodoping and some other features of this phenomenon. According to the results obtained by Japanese researchers [11–13], illumination of the Ag–chalcogenide semiconductor–electrode system leads to generation of photovoltage and photocurrent.

Ukrainian researchers [14] have thoroughly investigated the photoelectric processes occurring in a cell of the composition  $\text{In}_2\text{O}_3$  (Sn) layer– $\text{As}_2\text{S}_3$  layer– $\text{As}_2\text{S}_3$  (Ag) layer–Ag layer. Along with the study of the photovoltage generation under cell illumination by white light, the dependence of the photovoltage on the light wavelength was analyzed. For example it was found that the photovoltage generated under cell irradiation in the wavelength range of 450–460 nm can be as high as 220 mV.

These experimental results led to the development of electric-field models of photostimulated diffusion of metals into semiconductors. A great contribution to the justification of the mechanism of this phenomenon was made by Lakshmikummar (United States) [15], who considered the interface between the undoped and photodoped layers as a heterojunction between two materials: undoped and doped semiconductors. This heterojunction model was experimentally proven in [16, 17]. It was established that the generation of photovoltage at the interface between undoped and silver-photodoped chalcogenide glassy semiconductors is accompanied by the separation of electrons and holes in such a way that they form a drawing electric field for silver ions. This electric field makes  $\text{Ag}^+$  ions move from the photodoped layer to the undoped layer. Thus, photodoping leads to the formation of a natural heterojunction, which moves during illumination of the photosensitive semiconductor–metal system.

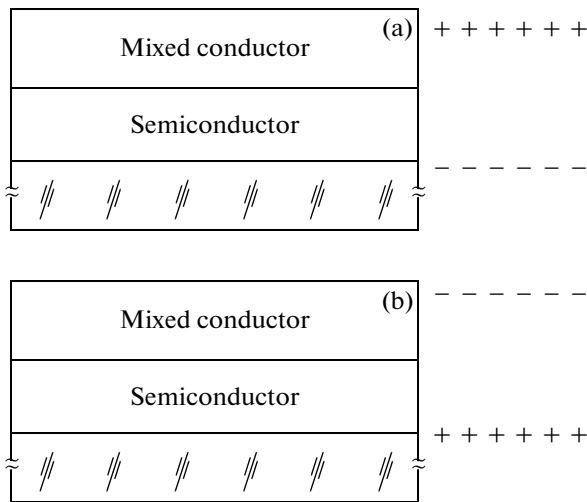
Up-to-date models of photostimulated diffusion of metal into a semiconductor do not reject chemical bonding of metal ions with a chalcogenide glassy semiconductor matrix; however, the importance of the peculiar coordination of metal–chalcogen bonds is indicated. Photodiffusion occurs via sites corresponding to unshared electron pair of chalcogen atoms, and

the conventional covalent metal–chalcogen bonds are formed at the sites of dangling bonds of the semiconductor matrix [18, 19]. The microscopic mechanism of this phenomenon is related to the electric processes at the interfaces between the undoped and doped semiconductors and between the doped semiconductor and metal.

The results of studying the nature and mechanism of photostimulated metal diffusion into a semiconductor (photodoping) gave rise to the concept and problem of studying the photostimulated displacement of ions in artificial heterojunctions between a semiconductor layer photodoped with a metal and a conventional undoped semiconductor [1, 20]. In the most general representation, the case in point is a heterojunction between a mixed ion–electron (hole) conductor and a conventional undoped semiconductor, since the study of semiconductors photodoped with metals showed that, at high metal concentrations (above 15%), these materials are mixed ion–electron (hole) conductors [1, 3, 16]. The essence of this concept is as follows: if photostimulated diffusion of metal into a semiconductor leads to the formation of a natural heterojunction (which provides this diffusion), an artificial heterojunction can be developed in such a way as to make the generation of photovoltage in this heterojunction ensure transport of metal ions through one of the heterojunction layers.

Since natural photodoping is promising for optical data recording, an artificial heterojunction based on a mixed ion–electron (or ion–hole) conductor should also be promising for optical data recording and other applications. The photostimulated displacement of ions in artificial heterojunctions between a mixed ion–electron conductor and a conventional electronic conductor was studied for the first time by one of the authors in [1, 20]. It was shown that these heterojunctions provide ion transport throughout one of the layers forming the heterojunction; i.e., the concept of photostimulated ion transport was experimentally proven for these heterojunctions.

The processes occurring in these heterojunctions can schematically be presented as follows. A thin-film heterojunction, which consists of a solid electrolyte layer with mixed ion–electron (hole) conductivity (this material is also referred to as a mixed ion–electron conductor) and a semiconductor layer with *n*- or *p*-type conduction, is shown in Fig. 1. The separation of photoexcited electrons and holes during photovoltage generation is shown on the right. Depending on the type of the heterojunction band diagram, holes can drift to the quasi-neutral region of the ion–electron (hole) conducting layer, while electrons can drift to the quasi-neutral region of the electronic conductor (Fig. 1a) or vice versa (Fig. 1b). The electric field between the photoexcited electrons and holes sepa-



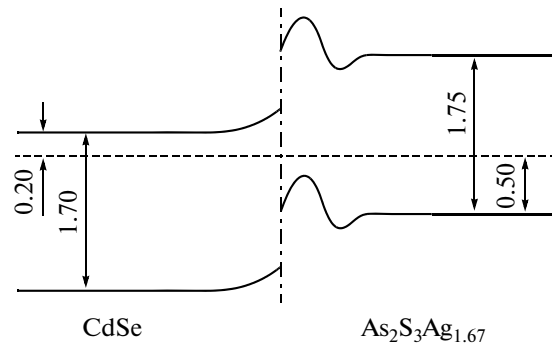
**Fig. 1.** Schematic image of the heterojunction based on a mixed ion–electron (hole) conductor. A layer of a conventional electron (hole) semiconductor are formed on a transparent substrate. It is shown on the right how photoexcited electron–hole pairs are separated under illumination of the thin-film structure. (a) Holes drift to the quasi-neutral region of the mixed conductor, while electrons drift to the quasi-neutral region of the conventional semiconductor. (b) Photoexcited electrons drift to the quasi-neutral region of the mixed conductor, while photoexcited holes drift to the quasi-neutral region of the conventional semiconductor.

rated by the heterojunction field will affect the ions located in the mixed ion–electron conductor layer to make them move in the direction perpendicular to the interface between the layers. If ions are positively charged (Fig. 1a), they move to the interface. In the case presented in Fig. 1b, they will drift to the free surface of the mixed ion–electron (hole) conductor. This schematic representation of the physical processes takes into account the effect of the electric field but neglects the influence of the electron wind [21]; features of its influence will be considered below.

The situation shown in Fig. 1a was implemented when studying CdSe–As<sub>2</sub>S<sub>3</sub>Ag<sub>x</sub> heterojunctions ( $x = 0.9–2.4$ ) [1].

For example, an Auger analysis of a CdSe–As<sub>2</sub>S<sub>3</sub>Ag<sub>1.67</sub> heterojunction illuminated by a KT-220–500 halogen lamp (illuminance  $8 \times 10^4$  lx) revealed an increase in the silver ion concentration (up to 5 at %) at the interface. The formation of silver clusters 0.2–0.5 μm in size and precipitates from 2 to 3 μm was also observed as a result of the silver photodeposition at the interface.

Since the existence of photostimulated ion transport was proven experimentally, this phenomenon can be used to design a thin-film ion accelerator based on heterostructures composed of mixed ion–electron (hole) conductors. This possibility will be demonstrated below.



**Fig. 2.** Energy-band diagram of the CdSe–As<sub>2</sub>S<sub>3</sub>Ag<sub>1.67</sub> heterojunction [1].

2. THEORETICAL ANALYSIS

Photostimulated ion transport is observed when a photovoltage is generated in a heterostructure based on a mixed ion–electron conductor under illumination [1]. It was shown in [20] that there is another necessary condition for photostimulated ion transport: the energy of light photons must exceed not only the energy gaps of the semiconductors forming the heterojunction, but also the barrier in the conduction band (Fig. 2). The energy-band diagram of this heterojunction was considered in [20], where it was shown that the specific features of this diagram are due to the interaction between charged particles of three types: negatively charged electrons, positively charged holes, and positively charged ions. The interaction between holes and electrons, which are the main charge carriers in two different films, is described by the diffusion model [2]. The displacement of ions and the formation of a dense ion layer near the space-charge region (which is formed as a result of the diffusion of holes and electrons) are related to the interaction of positive ions with the electric field of the electron–hole space-charge region. Due to this the energy-band diagram of the heterojunction is described by a wide portion with bent bands and a barrier in the conduction band [1, 20]. This barrier is determined by the spatial arrangement of the space-charge region and the dense ion layer. Photostimulated ion transport was observed when the light reached the interface between the layers forming the heterojunction. These experimental results indicate that the ion motion is caused by the electric field between the photoexcited electrons and holes separated in the space-charge region of the heterojunction. The main mechanism for overcoming the barrier in the conduction band of the heterojunction is the electron injection [20].

At a high light intensity, the field of photoexcited electrons and holes separated in the space-charge region of the heterojunction compensates for the heterojunction field. As a result the band bending is smoothed out. Then the intense ion migration to the

**Table 1.** Ionic conductivity activation energy  $E_a$  for some solid electrolytes

Electrolyte	$E_a$ , eV*
$\alpha$ -AgJ	0.051
KAg <sub>4</sub> J <sub>5</sub>	0.095
NH <sub>4</sub> Ag <sub>4</sub> J <sub>5</sub>	0.095
RbAg <sub>4</sub> J <sub>5</sub>	0.098
Ag <sub>2</sub> J <sub>4</sub> WO <sub>4</sub>	0.16
[(CH <sub>3</sub> ) <sub>4</sub> N] <sub>2</sub> Ag <sub>13</sub> J <sub>15</sub>	0.17
AgCl	0.16
AgBr	0.15
LiCl	0.40
LiBr	0.39
LiJ	0.40
(Ag <sub>2</sub> GeS <sub>3</sub> ) <sub>48</sub> (AgI) <sub>52</sub>	0.28
(Li <sub>4</sub> P <sub>2</sub> S <sub>7</sub> ) <sub>42</sub> (LiJ) <sub>58</sub>	0.42
(Li <sub>2</sub> S) <sub>30</sub> (B <sub>2</sub> S <sub>3</sub> ) <sub>23</sub> (LiJ) <sub>47</sub>	0.33
(Li <sub>2</sub> S) <sub>30</sub> (B <sub>2</sub> S <sub>3</sub> ) <sub>26</sub> (LiJ) <sub>44</sub>	0.3

\* The  $E_a$  values are taken from [22–24].

**Table 2.** Dependence of the ion–electron interaction energy  $U$  on the photoexcited-electron concentration  $C$  in a layer of a mixed ion–electron conductor (the calculation takes into account the interaction of monovalent positive ion with the eight nearest photoexcited electrons)

$C$ , cm <sup>-3</sup>	$U$ , eV
10 <sup>20</sup>	0.77
10 <sup>21</sup>	1.66
10 <sup>22</sup>	3.57

interface, which was observed experimentally [20], can be due to the entrainment of positive ions by the flow of moving photoexcited electrons. Thus, the photostimulated ion displacement is determined to a certain extent by the entrainment of positive ions by the flow of photoexcited electrons.

This conclusion is justified by the comparison of the ionic conductivity activation energy (Table 1) for a number of solid electrolytes and the Coulomb ion–electron interaction energy (Table 2). The latter parameter was calculated for one positive metal ion (Ag<sup>+</sup>) surrounded by eight photoexcited electrons. Comparison of the data of Tables 1 and 2 shows that, at high light intensities, when many photoexcited electrons arise in the mixed ion–electron conductor, they can efficiently entrain ions, because the Coulomb ion–electron interaction energy exceeds significantly the ionic conductivity activation energy for a number of materials. This technique for estimating the influence of the photoexcited electron flow on the ion

transport is in agreement with the modern theoretical models considering the effect of physical factors on the ion transport and with the conventional models [25–27]. In accordance with these models, it is assumed that an ion in a solid is located in a potential well of depth  $\Delta g$ . Then the probability  $W$  for this ion to pass to the neighboring site over unit time is described by the expression

$$W = v \exp\left(-\frac{\Delta g}{kT}\right), \quad (1)$$

where  $k$  is the Boltzmann constant and  $T$  is temperature. If this ion is affected by an external electric field or entrained by the Coulomb potential of the electric charges around it, these factors affect the exponent in the exponential. Due to this the ion mobility  $\mu$  in an electric field  $E$  can be described by the expression [15]

$$\mu = \mu_0 \exp\left[-\frac{(E_a - E\Delta x)}{kT}\right], \quad (2)$$

where  $E_a$  is the ionic conductivity activation energy and  $\Delta x$  is the potential-well width. Based on these considerations, one can draw the following conclusion: since the ion–electron interaction energy either exceeds the ionic conductivity activation energy or is of the same order of magnitude (Tables 1 and 2), ions must be effectively entrained by the photoexcited electron flow.

Thus, the photostimulated ion transport is determined by two physical factors. A positive metal ion is entrained in the following way: when photoexcited electrons move in the heterojunction field, they pass at a sufficiently short distance from the ion to entrain it due to the Coulomb interaction.

In addition photoexcited electrons and holes induce an electric field, which, depending on the type of the energy-band diagram, either compensates for the main (heterojunction) field or provides an accelerating potential for ions. The specific features of the influence of these physical factors are determined by the specific form of the energy-band diagram and the concentration of the electrons, holes, and ions that are involved in the formation of space-charge regions in a heterojunction based on a mixed ion–electron conductor.

Using the two aforementioned physical factors of photostimulated ion transport and heterostructures of special design, one can develop thin-film ion accelerators of different designs.

Provided that there is a high density of electrons after their passage through the space-charge region of the heterojunction, one could implement efficient entrainment of positive ions by these electrons. Due to the strong electron–ion interaction, electrons can capture ions and accelerate them to the electron beam velocity.

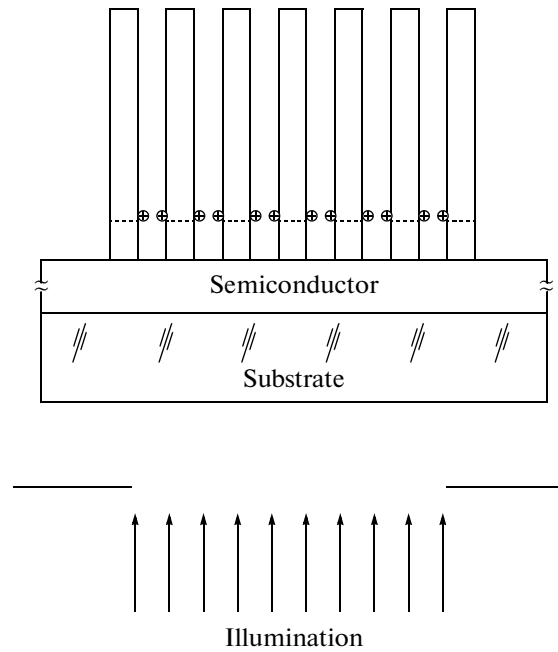
Note that there are two factors that hinder efficient acceleration of ions in heterojunctions based on a mixed ion–electron (hole) conductor. First, when the concentration of ions is high, they compensate for the field between the photoexcited electrons and holes separated in the space-charge region. The second factor is the collisions of ions with the framework of the solid-state matrix of the mixed conductor.

These undesirable factors can be eliminated by forming special narrow channels (oriented perpendicularly to the interface between the layers) in the mixed ion–electron conductor and placing ions on the channel walls (Fig. 3). In this case the concentration of ions will be low and they will not compensate for the field between the photoexcited electrons and holes separated in the space-charge region. The special-design heterostructure in Fig. 3 is in essence a thin-film ion accelerator, because, under the conditions shown in Fig. 3, ions can be efficiently accelerated. In this heterostructure the ions are located behind the space-charge region of the heterojunction. The electron beam emerging from the space-charge region of the heterojunction entrains ions due to the Coulomb interaction and accelerates them. The electrons that leave the space-charge region undergo a quantum-mechanical energy exchange with the electrons located within the space-charge region. The accelerator operates such that both pumping and acceleration of photoexcited electrons lead to acceleration of positive ions.

According to the data of monograph [28], the velocity of photoexcited electrons, which move in the electric field of the space-charge region of a photovoltaic heterojunction or a  $p$ – $n$  junction, is  $10^7$  cm/s. This value corresponds to a de Broglie wavelength of 7.27 nm. Accordingly, the channel width should be equal to the de Broglie wavelength or even smaller to allow for efficient energy exchange between the electron flows from the two different sides of the channel and entrainment of the ions fixed on the channel walls.

If the properties of the materials are chosen such that the energy-band diagram of the heterojunction provides motion of photoexcited electrons to the free surface of the mixed conductor and the drift of holes into the bulk of conventional semiconductor, this separation of photoexcited carriers will lead to the acceleration of positive ions toward the free surface of the mixed conductor. In this case the positive ions will be, on the one hand, entrained by the photoexcited electron flow due to the strong ion–electron interaction and, on the other hand, accelerated by the electric field between the clouds of electrons and holes; i.e., the ions will be accelerated. The possibility of using a heterostructure of special design as an ion accelerator was patented for the first time in [29].

Pulsed illumination of this heterostructure allows one to implement ion escape from a mixed ion–electron conductor. To this end the physical properties of the materials and the illumination conditions must be



**Fig. 3.** Schematic image of a heterostructure based on a mixed ion–electron (hole) conductor with channels. The dashed line shows the boundary of the space-charge region in the mixed ion–electron (hole) conductor. Positive ions are indicated on the channel walls in the mixed conductor beyond the depletion region.

chosen such as to provide ion acceleration primarily due to the electron-beam induced ion entrainment. When ions move through the channel with the electron-beam velocity, their kinetic energy exceeds greatly that of the Coulomb ion–electron interaction and the energy of ion binding with the channel surface in the mixed conductor. As a result ions will escape from the heterostructure when reaching the mixed conductor surface.

When accelerating ions in this way in a heterostructure of special design, one can increase their velocity by providing interaction of them with hot electrons. In accordance with the equation of the internal photoelectric effect,

$$h\nu = E_g + \frac{m_e V_e^2}{2}, \tag{3}$$

where  $h\nu$  is the light photon energy,  $E_g$  is the band gap of the illuminated material,  $m_e$  is the electron mass, and  $V_e$  is the electron velocity. Thus, the photon energy transferred to an electron is spent to overcome the band gap of the illuminated material and acquire a velocity  $V_e$  by the electron. If the light photon energy exceeds greatly the band gap, the conditions corresponding to a high velocity of hot electrons can be formed. Using a special-design heterostructure, one can direct the motion of hot electrons to the mixed-conductor free surface. Then this hot electron flow

can entrain accelerated ions to high velocities. The kinetic energy of ions increases with an increase in their velocity according to the quadratic law.

Another factor that is favorable for accelerating ions is the connection of the special-design heterostructure to an external voltage source. Due to this one can vary the number of electronic carriers that form space-charge regions of the heterojunction and their potentials. Under illumination of this heterojunction, photoexcited electrons will entrain ions more efficiently.

The general estimates of the conditions for accelerating ions in heterostructures of different designs show that two different operation modes can be implemented: either an ion accelerator or an ion laser. The ion-accelerator mode can be implemented most directly when ions are accelerated in a thin-film heterostructure or in the same heterostructure but with narrow channels (Fig. 3), without their escape beyond the heterostructure boundaries. In this case the ion-irradiated material can be either a mixed-conductor matrix or some other material, which can be supplied (in micro- or even nanovolumes) through a channel to the irradiated region. Another interesting possibility is to provide a contact of the free part of the channel with the rest of the material, which must be subjected to ion bombardment.

However, a more important operation mode of ion accelerator implies ion escape from the heterostructure. In this case, if the ion kinetic energy exceeds significantly the ion binding energy with the mixed-conductor matrix in the accelerating channel, the accelerator mode is implemented. However, if the energy of the ion accelerated in the channel exceeds the binding energy with mixed-conductor matrix by a small value comparable with the binding energy, the ion laser mode can be provided. The reason is that the ion-matrix binding energy is a threshold parameter, which synchronizes the ion-flow energy.

### 3. CONCLUSIONS

In this study we analyzed more thoroughly (than in previous studies [1, 20]) the physical factors of photo-stimulated ion transport in heterojunctions based on a mixed ion–electron (hole) conductor and the possibility of applying in practice the effect of these physical factors.

Photostimulated ion transport in heterojunctions based on mixed ion–electron (hole) conductors is determined to a certain extent by the electron-beam induced entrainment of positive ions and the effect of the electric field formed between the photoexcited electrons and holes separated by the heterojunction field. An analysis of the specific features of the effect of these factors shows that a thin-film ion accelerator can be developed based on a special-design heterostructure. A model of this accelerator was patented for the first time in [29] and developed in this study.

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