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06

Physical foundations of the formation of the silicon-based heterovarizonic structure

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With the formation of binary unit cells based on the A^{II} and B^{VI}, A^{III} and B^{V} elements, a heterovarizonic structure was obtained in the near-surface region of silicon, without destroying the crystal structure, without surface states with a thickness of about $5\,\mu$m. The resulting heterovarizonic structure has special fundamental parameters that ensure the absorption of light in a wide range of the solar spectrum from UV to IR radiation with $\lambda = 0.1-3\,\mu$m, i.e., it covers the entire solar spectrum.

Keywords: silicon, photocell, solar cell, heterovarizonic structure, photovoltaics, $A^{II}B^{VI}$, A^{III}, B^{V} compounds.

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Introduction

Thanks to silicon, modern electronics has achieved very great success. All this is connected with rather large reserves of silicon across the globe, and most importantly, an accessible technology for obtaining silicon crystals with the required purity and electrical parameters and given sizes of single crystals has been mastered. Not for a single semiconductor material will a technology for obtaining crystals of such purity and with such geometric dimensions be developed in the near future.

Photovoltaic energetics can become the main source of energy for humanity not only in the distant future, but also in the near future, as it is the cleanest and most eternal source of energy. At present, silicon has been and remains the main material in photovoltaic energetics, more than 90% of photovoltaic stations operate on the basis of silicon photocells. However, large-scale utilization of silicon photocells in photovoltaic energetics under ground conditions is limited by their low efficiency, which has reached 20% in industry, and 25% [1–3] under laboratory conditions.

Although photocells based on semiconductor compounds $A^{III}B^{V}$ and $A^{II}B^{VI}$ have a fairly high efficiency, especially multi-stage photocells [4–5], but they cannot be used as an energy source under ground conditions, i.e. it is practically impossible to use such photocells on a wider scale. This is due not only to the very high price of such photocells (they are almost 50 times more expensive than silicon-based photocells), but also to the limited supply of elements in semiconductors $A^{III}B^{V}$ and $A^{II}B^{VI}$, their toxicity, and too complicated technology for obtaining such elements [6–8].

Therefore, the only option is to increase the efficiency of photocells based on silicon or to develop a new class of material based on it for the future of photovoltaic energetics under ground conditions.

A deeper analysis shows that low efficiency of silicon solar cells is mainly due to the fundamental parameters of this material, i.e. a small band gap, non-direct-gap structure, and low mobility compared to semiconductor compounds $A^{III}B^{V}$ [9].

Therefore, it can be unequivocally stated that without changing the fundamental parameters of silicon, it is practically impossible to create more efficient photocells [10,11].

As far as we know, at present there are no physical materials and technologies that allow for changing the fundamental parameters of silicon, as well as developing any technology for obtaining new promising materials for the future of photovoltaic energetics.

The purpose of this article was to physically substantiate the possibility of creating a new type of hetero variable-gap structures by forming binary clusters of atoms of III and V groups in the silicon lattice, as well as to reveal new physical and functional capabilities of such a material for optoelectronics and photovoltaic energetics.

1. Physical foundations for the formation of binary elementary cells in a silicon lattice

In the last 10−15 years the scientists of the „Digital Electronics and Microelectronics“ Department of TSTU have been working on the possibility of controlling the fundamental parameters of silicon [12–14]. The scientists of this department managed to physically substantiate the original and accessible technology for obtaining silicon with controlled fundamental parameters. The essence of this technological solution lies in the formation of binary nanoclusters in the silicon lattice, i.e. binary elementary cells with controlled parameters.

As is known, group III and V elements in silicon are mainly located at sublattice points and create small donor and acceptor energy levels in the band gap of silicon. The solubility of these elements in silicon is quite high and reaches $N \geq 10^{21}\,\mathrm{cm}^{-3}$. In the atomic state, they are in the form of singly charged ions A^{III-}, B^{V+} and create an electric potential around themselves, they also create a corresponding additional concentration of charge carriers in both the conduction and valence bands. All this leads to a significant violation of the thermodynamically equilibrium state of the crystal. Therefore, the crystal under such conditions is in a thermodynamically non-equilibrium state [15].

It has been found that it is possible to create such thermodynamic conditions for diffusion doping of silicon, sequentially with III and V groups of elements, as well as optimal thermodynamic conditions for thermal annealing, after diffusion doping, which allows atoms of III and V groups to be nearby in the silicon lattice, i.e. they will occupy places in neighboring lattice points (Fig. 1).

As can be seen, electrically neutral molecules are formed between the A^{III-} and B^{V+} ions. The formation of such molecules completely shields the electric potential of each ion of the atoms of III and V groups. In this case, the atoms of III and V groups in silicon act not as impurity atoms, but as the main atoms of a new lattice cell. Therefore, the atoms of III and V groups do not create energy levels in the band gap, i.e. their presence does not create additional electrons and holes both in the conduction and in the valence bands. The formation of such molecules does not break the tetrahedral bond in the silicon lattice. This means that the formation of such electrically neutral molecules between the ions of elements of III and V groups provides a more favorable thermodynamic equilibrium state of the lattice. This leads to a significant stimulation of the self-organization of the such electrically neutral molecules formation between the ions of the A^{III} and B^{V} elements.

The formation of such electrically neutral molecules is interesting in that a new lattice cell $Si_2A^{III-}B^{V+}$ appears in the silicon lattice (Fig. 1). These lattice cells, in contrast to the lattice cell of silicon, which has a 100% covalent bond, have a partially ionic-covalent bond, i.e., an elementary cell appears that does not exist in nature. The ion fraction in such new binary lattice cells $Si_2A^{III-}B^{V+}$, according to [15,16], is determined by the difference between the electronegativity values of atoms of III and V groups. Calculations show that when the electronegativity difference is within $\Delta x \leq 1$, the fraction of ionic bond in them is about 15−20%. This means that in this case the covalent bond in them will be predominant.

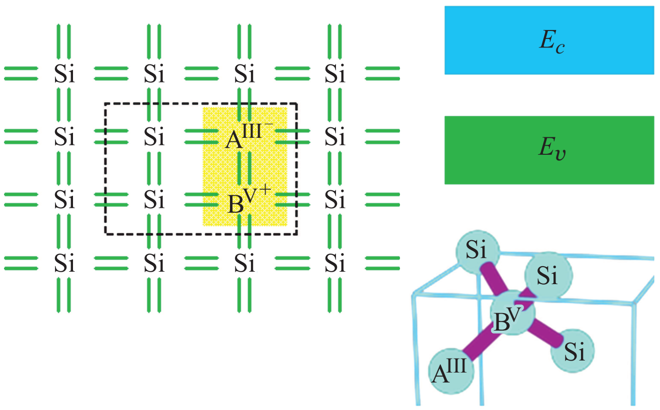

Figure 1. Electrically neutral molecules between A^{III-} and B^{V+} ions and the formation of new elementary cells $Si_2A^{III}B^{V}$ in silicon.

Now the question arises whether all element atoms of III and V groups can form electrically neutral molecules and will they form new binary lattice cell in the silicon lattice Theoretically yes, but for this it is necessary that the following conditions are met [17,18].

1. The sum of the covalent radii of the elements' atoms of III (r_{III}) and V (r_{V}) groups should be sufficiently close to the sum of the covalent radii of two silicon atoms (r_{Si}), i.e. the following relations must be met: $r_{\mathrm{III}} + r_{\mathrm{V}} - 2r_{\mathrm{Si}} \leq \pm 0.2r_{\mathrm{Si}}$, $2r_{\mathrm{Si}} - (r_{\mathrm{III}} + r_{\mathrm{V}}) \leq \pm 0.2(2r_{\mathrm{Si}})$.
2. The difference between the electronegativity values of elements' atoms of III and V groups should not differ significantly from the electronegativity values of the silicon atoms.
3. The solubility of the elements' atoms of III and V groups and their diffusion coefficients should not differ significantly from each other.
4. It is necessary to carry out thermal annealing after diffusion doping of the material. The thermodynamic conditions of thermal annealing (temperature and time) are mainly determined by the chemical and physical parameters of elements of III and V groups.

On the basis of the analysis of the electrical and chemical parameters of elements of groups III and V , as well as their diffusion coefficients and solubility in silicon, the following groups of element pairs of groups III and V have been established. These data show the possibility of forming a fairly wide class of binary lattice cells with the participation of III and V group atoms.

1. The most suitable pairs are BBi, AlP, GaP, BP.
2. Suitable pairs are BSb, AlAs, GaAs.
3. Possible pairs are BAs, GaSb, InP.
4. Non-suitable pairs are AlSb, InAs, InSb.

As is known, the fundamental parameters of each semiconductor material are determined by its structure, composition and chemical bonding of the lattice cells that make up the semiconductor material. Therefore, it is necessary to select the optimal technology conditions of diffusion doping of silicon with elements' atoms of III and V groups, which makes it possible to introduce them with a maximum and identical concentration, and also to distribute these atoms in silicon samples, taking into account their diffusion coefficients, both in the process of simultaneous and sequential diffusion of elements of III and V groups. That is, it is necessary to determine the optimal temperatures and time of additional thermal annealing, which ensures the maximum participation of the introduced impurity atoms in the electrically neutral molecules formation.

With an increase in the concentrations of introduced elements' atoms of III and V groups, the probability of the electrically neutral atoms formation between elements' atoms of III and V groups increases significantly, therefore, the concentration of binary lattice cells increases, and this, in turn, leads to the appearance of some possible combinations of binary lattice cells (Fig. 2).

The energy bonds of atoms in lattice cells and their combinations will differ significantly from each other. This means that the energy required for release of electrons in such lattice cells with their various combinations differs significantly, i.e. there will be significantly different values of the band gap. Therefore, they will not have the same fixed value of the light wavelength for bandgap absorption (λ_b), each of them will have its own values λ_b, i.e. the bandgap absorption spectrum of silicon is significantly expanded. The absorption spectrum will be more noticeable only at a sufficiently high concentration of such lattice cells and their combinations.

When introducing the maximum concentration of impurity atoms of III and V groups $N \geq 10^{21}\,\text{cm}^{-3}$ into silicon, which ensures their identical concentration distribution, in contrast to the above-mentioned different combinations of binary lattice cells, semiconductor compound nanocrystals can be formed based on the introduced impurity elements' atoms of III and V groups (Fig. 3). This means that a local region appears, enriched with nanocrystals of semiconductor compound atoms $\text{A}^{\text{III}}\text{B}^{\text{V}}$, immersed in the silicon lattice. The size of semiconductor compound nanocrystals $\text{A}^{\text{III}}\text{B}^{\text{V}}$ can vary from several nanometers to several tens of nanometers depending on the doping conditions. The energy structure of silicon with high concentration of binary lattice cells and such structures changes significantly, and silicon turns into a new material.

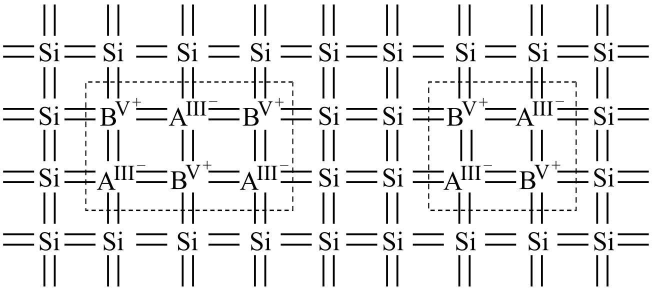

Figure 2. Some possible combinations of binary lattice cells in a silicon lattice.

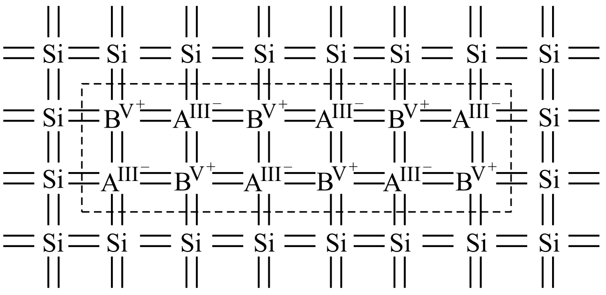

Figure 3. Nanocrystals of a semiconductor compound based on elements of III and V groups in the silicon lattice ($\text{Si}_2\text{A}^{\text{III}-}\text{B}^{\text{V}+}$).

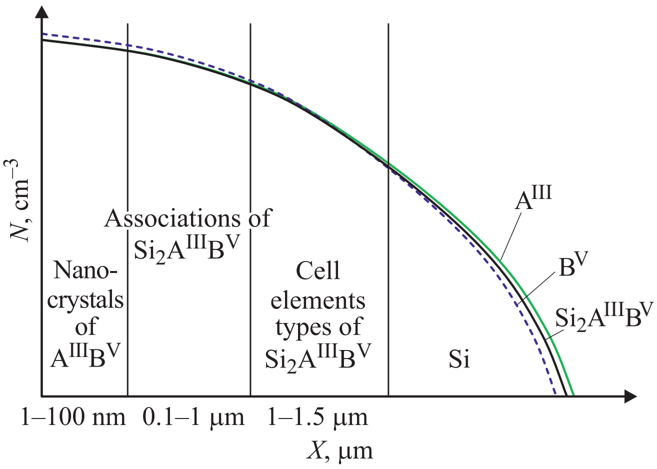

Figure 4. The most probable structures based on lattice cells distribution over the depth of silicon. A^{III} — concentration distribution of elements of III group after diffusion of elements of V group (first stage of diffusion), B^{V} — concentration distribution of elements of V group (second stage of diffusion), $\text{Si}_2\text{A}^{\text{III}}\text{B}^{\text{V}}$ — concentration distribution of binary lattice cells in a silicon lattice.

The most probable structures based on the distribution of lattice cells over the depth of silicon $\text{Si}_2\text{A}^{\text{III}-}\text{B}^{\text{V}+}$ are shown in Fig. 4. As can be seen, a region enriched with nanocrystals of various sizes appears in the near-surface region with thickness of $d = 1{-}100\,\text{nm}$. Further from 0.1 to $1\,\mu$m, a region appears enriched with various combinations of binary lattice cells. After this region, there is a region enriched mainly with individual lattice cells $d = 1{-}1.5\,\mu$m, then up to $d = 1.5{-}3\,\mu$m the silicon region follows with the lowest concentration binary lattice cells.

The thickness of each region and their distribution can be controlled over a wide range depending on the diffusion conditions. Thus, it can be seen from Fig. 5 that in silicon, starting from the surface to the depth X, a region appears in which the structure, composition, and chemical bonds change from semiconductor compounds $\text{A}^{\text{III}}\text{B}^{\text{V}}$ to pure silicon without a significant change in the crystal structure of silicon and the tetrahedral bond, but naturally the properties of each structure are different, and they differ significantly in fundamental parameters from each other.

This structure is not similar to the existing variable-gap structures and is not similar to a heterostructure, i.e. a new type of structure appears — a hetero variable-gap structure without any violations of the structure and surface states. This structure is created using diffusion technology, since the diffusion method of structure formation is used here.

To prove formation of such hetero variable-gap structures, preliminary experiments were carried out with impurity atoms of gallium and phosphorus, which are included in a number of more suitable pairs. Single-crystal silicon KEF-100 ($N_{\text{P}} \sim 10^{13}\,\text{cm}^{-3}$) with an oxygen content $N_{\text{O}_2} \approx (5{-}6) \cdot 10^{17}\,\text{cm}^{-3}$ and dislocation density

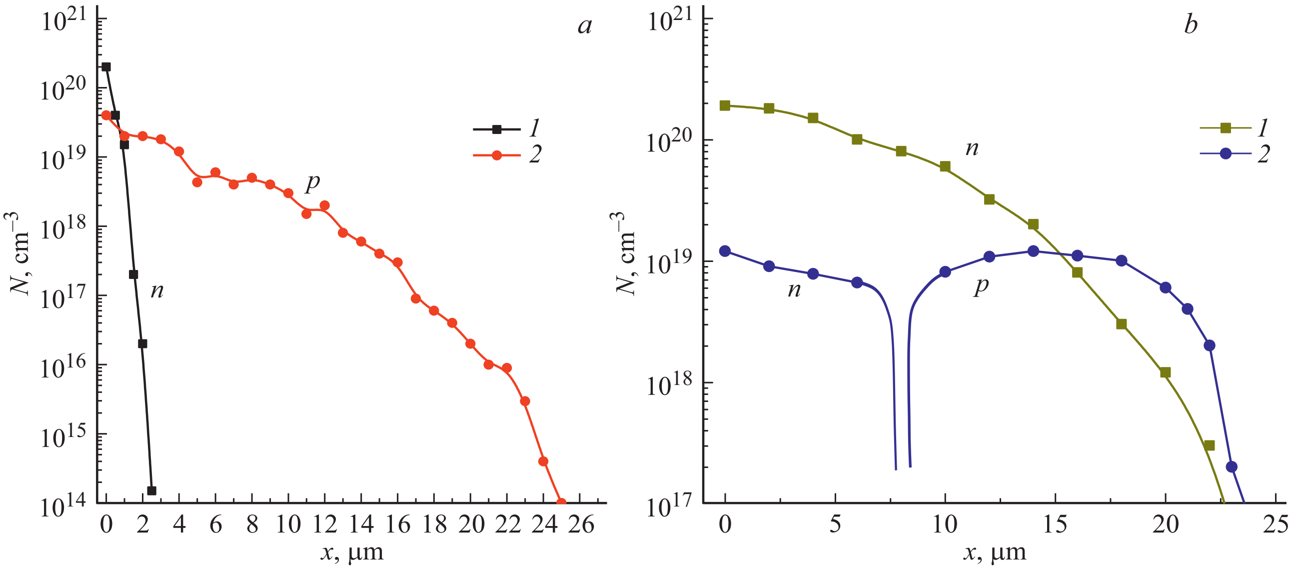

Figure 5. The concentration distribution of electrons (phosphorus atoms) and holes (gallium) in the silicon lattice: *a* — diffused independently, *b* — with successive diffusion. Curve *1* — distribution of electrons (phosphorous). Curve *2* — distribution of holes (gallium).

$N_D \sim 10^3\,\mathrm{cm}^{-2}$ was used as initial material. The sample size was $V \sim 1 \times 4 \times 8$ mm. After the necessary mechanical and chemical treatments of the samples, phosphorus was diffused from the applied ammonium phosphate layer in air at $T = 1000^\circ$C for $t = 2$ h. After diffusion, phosphosilicate glass was removed from the surface in all samples by etching with HF and NH_4F. The concentrations of phosphorus and gallium atoms were studied by the 4-probe method. It was assumed that these atoms are in the lattice in an electroactive state. Along with this, the dependences of charge carriers mobility on the concentration of impurity atoms were taken into account. At the same time, the electron (phosphorus) concentration on the surface of the samples was $N_\mathrm{P} = 2 \cdot 10^{20}\,\mathrm{cm}^{-3}$.

Gallium diffusion in silicon (KEF 100) was carried out from the gas phase at temperature of $T = 1250^\circ$C for 4 h. This choice of gallium diffusion makes it possible to obtain the maximum concentration on the surface and in the volume of silicon.

Then gallium diffusion was carried out in the samples doped with phosphorus at $T = 1250^\circ$C for 4 h. At the same time, samples of phosphorus-doped silicon (without gallium) were simultaneously subjected to thermal annealing, also at $T = 1250^\circ$C, $t = 4$ h, in order to determine the change in the concentration distribution of phosphorus during additional annealings.

Figure 5, *a* shows the concentration distribution of phosphorus atoms in silicon after diffusion $T = 1000^\circ$C, $t = 2$ h. The concentration distribution of gallium after diffusion $T = 1250^\circ$C, $t = 4$ h in KEF-100 samples is also presented.

As can be seen from the experimental results, the phosphorus concentration in the near-surface region is $N_\mathrm{P} = 2 \cdot 10^{20}\,\mathrm{cm}^{-3}$, and at a depth of $x = 2.5\,\mu$m, its concentration is decreased to $\sim 10^{14}\,\mathrm{cm}^{-3}$ (curve *1*), while the samples always remain of *n*-type.

The data obtained correspond to the literature data [12–14]. As can be seen from the figure, the concentration distribution of phosphorus and gallium during their diffusion separately differ significantly from each other.

Fig. 5, *b* shows the concentration distribution of electrons (phosphorus) in the reference samples, which were subjected to additional thermal annealing at $T = 1250^\circ$C, $t = 4$ h (curve *1*), and also the concentration distribution of charge carriers in silicon samples doped with gallium at $T = 1250^\circ$C, $t = 4$ h, which were pre-doped with phosphorus at 1000°C, $t = 2$ h (curve *2*). As can be seen, as a result of additional annealing at $T = 1250^\circ$C, $t = 4$ h the surface concentration of phosphorus slightly decreases, and the penetration depth reaches $x = 25\,\mu$m (curve *1*), in the region $x = 0{-}25\,\mu$m the samples acquire the apparent *n*-type.

Concentration distribution in samples doped with gallium at $T = 1250^\circ$C, $t = 4$ h, after doping with phosphorus at 1000°C, $t = 2$ h (Fig. 5, *b*, curve *2*) shows that in the studied samples up to a depth of $x = 7.5{-}8\,\mu$m there is *n*-conductivity type. In this case, the concentration of electrons (phosphorus) significantly decreases, and at $x > 7.5{-}8\,\mu$m, the samples acquire the *p*-conductivity type. In $x = 7.5{-}10\,\mu$m region the hole concentration slightly increases; and in $x = 8{-}15\,\mu$m region the concentration of holes (gallium) practically remains constant, and at $x > 17\,\mu$m it decreases quite sharply.

These obtained results show that during gallium diffusion (in the presence of a high phosphorus concentration) the phosphorus concentration in $x = 0{-}7.5\,\mu$m region decreases by 1.5−1 times, then the gallium concentration becomes greater than the phosphorus concentration, and the material has the p-conductivity type. Although, as can be seen from Fig. 5, *b* (curve *1*), the concentration of phosphorus up to 15 μm region is greater than the concentration of gallium. These results suggest that the phosphorus presence in silicon leads to increasing of the gallium concentration.

2. Discussion

The obtained experimental results cannot be explained by the mutual compensation of donor (phosphorus) and acceptor (gallium) impurity atoms, since phosphorus and gallium atoms in the silicon lattice are distributed randomly and spatially separated, which, accordingly, should not lead to an increase in the concentration of gallium atoms in the presence of phosphorus atoms. Therefore, it can be assumed that these phenomena are associated with the interaction of phosphorus and gallium atoms. Since the phosphorus atoms in the silicon lattice are located in the crystal lattice sites in the form of a positively charged ions P^+, creating additional electrons in the conduction band, the concentration of which is equal to $N_{P^+} = n$. The presence of a sufficiently high concentration of positively charged phosphorus atoms (P^+) creates a significant electric potential. This potential is distributed from the crystal surface deep into the silicon sample, which stimulates an increase in the gallium atoms concentration during diffusion, which act as an acceptor impurity in silicon in the form of negative ions Ga^- [19,20]. Therefore, it can be assumed that as a result of such interactions the donor-acceptor complexes appear in the silicon lattice, i.e. quasimolecules in the form of $[P^+Ga^-]$. In turn, the $[P^+Ga^-]$ quasimolecules in the silicon lattice and their depth distribution form a hetero variable-gap structure, which changes the energy structure of silicon.

To estimate the contribution to the energy parameters of silicon with hetero variable-gap structures based on $[P^+Ga^-]$, we created $p-n$-junctions in silicon with the participation of gallium and phosphorus impurity atoms separately, and with hetero variable-gap structures. In this case, single-crystal hole-type silicon of the KDB-0.5 brand doped with boron (for doping with phosphorus) and single-crystal silicon of the electronic type of conductivity doped with phosphorus of the KEF-0.3 brand (for doping with gallium) were used as the starting material; the hetero variable-gap structure was created on the basis of silicon KDB-0.5. Diffusion in all samples was carried out under the same conditions $T = 1250^\circ$C, $t = 0.5$ h.

The results of the experiment (Fig. 6) show that in $p-n$-junctions formed by gallium and phosphorus, their spectral dependences of the short-circuit current (I_{sc}) correspond to each other. In the case of the Ga^-P^+ quasimolecule formed in silicon, $I_{sc}(h\nu)$ differs significantly from that doped with phosphorus and gallium separately.

1) The maximum value of Isc in samples with Ga^-P^+ quasimolecules is observed not at $h\nu = 1.1$ eV, but at $h\nu = 1.35$ eV;

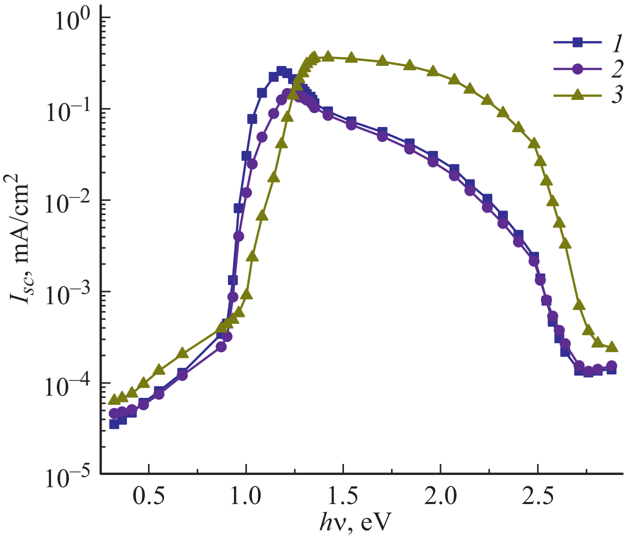

Figure 6. Spectral dependence of the short-circuit current of silicon samples with a $p-n$-junction obtained using: *1* — P, *2* — Ga and *3* — GaP.

2) in this case, significantly increased values of I_{sc} are observed in the region $h\nu = 1.3-2.8$ eV;

3) there is an expansion of the spectral region of sensitivity I_{sc}.

The results obtained suggest that such significant differences in $I_{sc}(h\nu)$ are associated with formation of Si_2GaP binary lattice cells, which result in the formation of a silicon-based hetero variable-gap structure. Since the formation of a [GaP] quasimolecule in the silicon lattice does not create any energy levels in the silicon band gap. This can also serve as evidence for the formation of hetero variable-gap structures taking into account gallium and phosphorus in silicon. More reliable data can be obtained based on the results of X-ray diffraction and mass spectroscopic analyses, which the authors are currently working on.

Let us consider the energy structure of the resulting material depending on the nature of the introduced impurity elements atoms of III and V groups, where nanocrystals of semiconductor compounds $A^{III}B^V$ with large values of band gap energy as in GaP, BP, GaAs, as well as with small values as in GaSb, InSb, etc. are formed. This makes it possible to create various combinations of binary lattice cells, i.e. to obtain a structure at a depth of $d = 0.1-5\,\mu$m in silicon with an energy structure different from the energy structure of semiconductor compounds $A^{III}B^V$. Also, with a varying band gap starting from GaP, GaAs, PB to the silicon band gap $E_g = 1.12$ eV, which ensures maximum absorption of the UV, visible and IR regions of light radiation. Also, the formation of such structures in the near-surface region of the material can form compounds with a band gap smaller than the band gap of silicon (GaSb, InAs, InSb), in which the value of E_g continuously increases from the crystal surface to E_g of silicon at a depth of d. In this case, a material is created that absorbs IR radiation with $\lambda > 1.2\,\mu$m. It should be noted that the band gap of nanocrystals $A^{III}B^V$ and various combinations of binary lattice cells may differ from the band gap of both compounds $A^{III}B^{V}$ and nanocrystals $A^{III}B^{V}$ in the crystal lattice silicon.

Thus, the optimal conditions for doping silicon with various elements' atom pairs of III and V groups make it possible to create a practically new class of materials with a controlled band gap based on the main silicon material.

Formation of a local region in the silicon lattice enriched with $A^{III}B^{V}$ nanoclusters leads to the appearance of a region in the silicon lattice with a direct-gap structure similar to the band structure of the corresponding semiconductor compounds $A^{III}B^{V}$. This means that the absorption coefficient significantly increases in the entire region of the solar spectrum. Thus, with formation of binary lattice cells, it is possible to change the more important fundamental parameter of silicon — the band structure, which is practically impossible to change by any other methods.

As a result of the formation of binary lattice cells with controlled parameters in the near-surface region, it is possible to obtain a sufficiently thick ($5\,\mu$m) silicon layer. With the help of a material with new fundamental parameters that ensure light absorption in a wide range of the solar spectrum from UV to IR radiation $\lambda = 0.1-3\,\mu$m, the entire solar spectrum is covered. None of the existing semiconductor materials has this property. Most importantly, it is obtained on the basis of the basic material of silicon.

Conclusion

It should be especially noted that this diffusion technology for obtaining such materials is sufficiently well-proven, reliable, accessible, and most importantly, it is based on the current planar diffusion technology that does not require expensive equipment, instruments and devices.

Now let us say a few words about the functionality of silicon with binary lattice cells. As shown by preliminary calculations, production of such a material based on silicon with binary lattice cells with the participation of Ga$-$P, P$-$Al and B$-$P can increase the efficiency of photocells by 25$-$30%. Calculations also show that when using the optimal technology, which enables to obtain lattice cells with the participation of GaP, GaAs, BP atoms with the required concentration and distribution, it is possible to manufacture silicon-based photocells with efficiency of more than 50%. This means that in the near future (3$-$5 years) expensive and complex multi-stage photocells based on $A^{III}B^{V}$ will be replaced by photocells with higher efficiency based on silicon with binary lattice cells. Thus, the proposed science-based model for controlling the silicon fundamental parameters ensures the large-scale utilization of photovoltaic energetics under ground conditions. Such materials also enable to create new, reliable and cheap devices for optoelectronics.

Conflict of interest

The authors declare that they have no conflict of interest.

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