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INFRARED PHOTODETECTORS BASED ON INASP EPITAXIAL NANOWIRES ON SILICON

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Abstract. In the paper, the upgraded technology of forming the epitaxial arrays of InAsP nanowires (NWs) synthesized on silicon substrates using molecular beam epitaxy has been presented. The optical and electrophysical properties of the NWs were studied. Based on the grown structures, a prototype of a short-wave infrared photodiode for a range from 1.2 μm to 1.9 μm was fabricated. Its band structures were numerically simulated. The temperature dependencies of the current-voltage characteristics and the spectral sensitivity of the photodetector prototype based on the NWs were experimentally studied. The external quantum efficiency of photoconversion of radiation with a wavelength of 1380 nm was found to be about 0.25% at 100 K.

Keywords: InAsP, silicon substrate, molecular beam epitaxy, nanowires, photodetector, quantum efficiency

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Научная статья

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ФОТОДЕТЕКТОРЫ ИНФРАКРАСНОГО ДИАПАЗОНА НА ОСНОВЕ ЭПИТАКСИАЛЬНЫХ НИТЕВИДНЫХ НАНОКРИСТАЛЛОВ INASP НА КРЕМНИИ

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Аннотация. В работе представлена усовершенствованная технология формирования эпитаксиальных массивов нитевидных нанокристаллов (ННК) InAsP, синтезированных на подложках кремния методом молекулярно-пучковой эпитаксии; исследованы оптические и электрофизические свойства этих ННК. На основе выращенных структур создан прототип фотодиода коротковолнового инфракрасного диапазона (1,2 – 1,9 мкм). Промоделированы зонные диаграммы фотодиода. Экспериментально исследованы температурные зависимости вольтамперных характеристик и спектральной чувствительности этого прототипа. Установлено, что внешняя квантовая эффективность фотопреобразования излучения с длиной волны 1380 нм составила примерно 0,25 % при температуре 100 К.

Ключевые слова: InAsP, кремниевая подложка, молекулярно-пучковая эпитаксия, нитевидный нанокристалл, фотодетектор, квантовая эффективность

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Introduction

Heterostructures based on narrow-gap semiconductors (with the band gap less than 1 eV) are widely used in infrared (IR) optoelectronics [1, 2]. The practical significance of research in this field lies in the possibility of fabricating multispectral IR radiometers and thermal imagers for reconnaissance and surveillance systems as well as chemical sensor arrays used for detection and



identification of explosives. The most popular material platform for IR detectors is indium arsenide (InAs). The choice of the narrow-gap (band gap E_g is 0.35 eV at a temperature of 300 K) InAs semiconductor material is due to the high mobility of charge carriers (more than $10^4 \text{ cm}^2/(\text{V}\cdot\text{s})$) and the long lifetime of minority carriers. However, further advances in detector manufacturing are limited by two main factors. First, the small band gap inevitably leads to accelerated thermal generation rates and is accompanied by nonradiative recombination of charge carriers, which in turn leads to high dark currents, increased noise levels, and decreased sensitivity of photodetector arrays [3]. Secondly, lattice mismatch between the layers of planar A^3B^5 heterostructure and substrate significantly limits the possibilities for controlling the band profile of heterostructures [4].

The performance characteristics of photodetector arrays can be improved by transition from planar structures to structures with a well-developed surface, namely, to arrays of nanowires (NWs) [5]. In this case, due to developed lateral surface of the NWs, nanostructures of high crystalline perfection can be grown on lattice-mismatched substrates, in particular, on silicon. Moreover, NW arrays are characterized by stronger light absorption compared with planar layers of the same composition and equivalent thickness [6, 7]. The addition of phosphorus to the InAsP solid solution causes an increase in the band gap with a consequent decrease in dark currents and corresponding noise from photodetectors operating in the short-wavelength IR range (1.0–3.5 μm) [8].

In this paper, we propose an improved technological approach to fabrication of photodetectors in the near and short-wavelength IR ranges (1.3–1.9 μm) using InAsP NW arrays epitaxially grown on silicon substrates as the active area. The effects of localization and resonant amplification of the IR electromagnetic field in the NWs can significantly improve the characteristics of such photodetectors:

- reduce the volume of their active area while maintaining their sensitivity;
- reduce their dark current;
- increase the performance of devices based on them [9].

Materials and methods

The InAsP/Si heterojunction is a type II junction with partial band overlap, which determines the choice of diode polarity. As follows from the band diagram (Fig. 1), only the combination of InAsP with n -type conductivity and Si substrate with p -type conductivity (n -InAsP/ p -Si) is possible for the array to function as a photodiode. The inverse combination (p -InAsP/ n -Si) will lead to the formation of barriers for charge carriers. Thus, the built-in field is generated at the n -InAsP/ p -Si

heterojunction (see Fig. 1,*a*). It is known that the surface states of NWs strongly influence the energy band diagram of nanostructures.

In view of the above, defects were added to the lateral surface of the considered model for InAsP NWs, whose presence in the case of InAsP material with the band gap of about 600–650 MeV leads to forced injection of electrons from the defect levels, band bending near the surface (see Fig. 1,*b*) and additional contribution to separation of charge carriers during photodiode operation.

InAsP NW arrays were epitaxially grown on silicon substrates doped to p -type conductivity (boron-doped p -type Si(111)). The substrates had a diameter of 3 inches, were misoriented by 4° toward (111), and had a resistivity of 0.4–0.6 $\Omega\cdot\text{cm}$.

The Veeco GEN III molecular beam epitaxy (MBE) system used was equipped with an In effusion cell as well as P_2 and As_4 cracker cells with needle valves acting as molecular sources. The intensity of molecular beams of group V and III

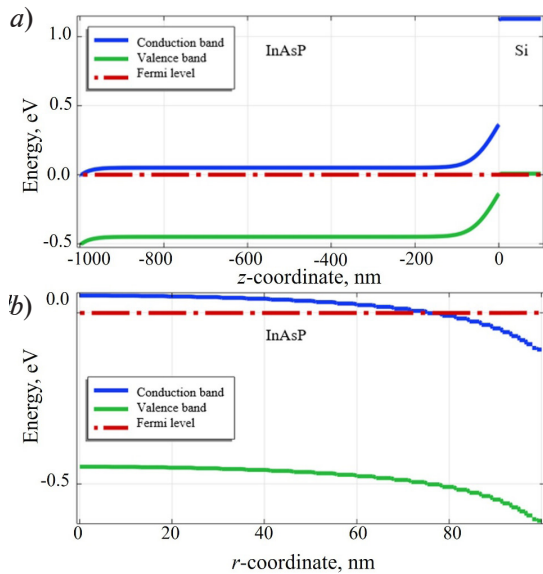


Fig. 1. Band structure of n -InAsP/ p -Si photodiode in longitudinal (*a*) and transverse (*b*) directions

elements was taken proportional to the pressure measured by a Bayard–Alpert ionization gauge. The substrate temperature (growth temperature) was measured by a thermocouple calibrated by the phase transition temperature of Si(111) $7\times 7 \leftrightarrow 1\times 1$ surface reconstruction. Monitoring of the growth process and analysis of the crystal structure of NWs was performed *in situ* by reflection high-energy electron diffraction (RHEED).

The main stages of formation of NW arrays on silicon are shown in Fig. 2. Before the NWs were grown by MBE, the silicon substrates were cleaned by a modified Shiraki method [10]. At the final stage of cleaning, it was carried out by wet chemical oxidation in boiling aqueous solution of ammonia and hydrogen peroxide ($\text{NH}_4\text{OH} : \text{H}_2\text{O}_2 : \text{H}_2\text{O}$) in a ratio of 1 : 1 : 3. As a result of the procedure, a thin (about 1.5 nm) layer of SiO_x oxide was formed on the silicon surface (see Fig. 2,a)

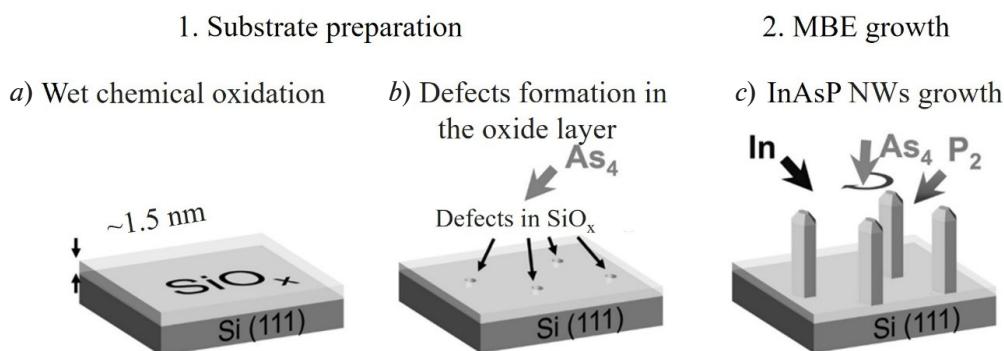


Fig. 2. Main stages of epitaxial growth of InAsP NW arrays on silicon: substrate preparation 1, material growth by molecular beam epitaxy 2; cleaning by wet chemical oxidation (a), formation of defects in SiO_x oxide layer (b); growth of NWs (c)

Immediately before the start of the epitaxial growth stage, silicon substrates were subjected to thermal annealing under ultrahigh vacuum conditions for 30 minutes at a temperature of 780 °C, which led to the formation of defects in the layer of surface silicon oxide serving as NW nucleation centers (Fig. 2,b) [11, 12]. Then, as the substrate cooled to the growth temperature in As_4 flux, the As-substituted silicon surface stabilized in the defect regions of the SiO_x surface layer (Fig. 2,b) [13]. The formation of NWs was initiated by switching group V sources to the necessary fluxes and opening the indium shutter (Fig. 2,c). To ensure uniform distribution of molecular fluxes during the growth process, the substrate was rotated at a speed of 5 rpm. In general, varying the group V growth fluxes makes it possible to regulate the incorporation of phosphorus and arsenic atoms into InAsP NWs, thus controlling the band gap of the NWs to ensure a given spectral sensitivity of photodetectors based on them.

The considered NW arrays were grown at a substrate temperature of 520 °C for 60 minutes and a P/As flux ratio of 1. InAsP NWs were not doped during growth; due to the large number of surface states on the lateral surface of the NWs, the synthesized nanostructures were characterized by *n*-type conductivity. Reference samples of InAs NWs were additionally grown.

Notably, the method applied for self-induced growth of NW arrays allows to achieve an extremely high surface density of NWs ($\geq 10 \mu\text{m}^{-2}$), which significantly reduces light scattering, manifesting as an opaque black surface of the sample. To fabricate photodetector arrays, we used the grown InAsP NWs with a diameter of 110 ± 41 nm, a length of 900 ± 300 nm, and a surface density of $5.5 \mu\text{m}^{-2}$. The morphology of the synthesized arrays was studied by scanning electron microscopy (SEM) using a Zeiss SUPRA 25-30-63 microscope. (see the SEM image in the inset in Fig. 4,a).

Studies of the crystalline structure of NWs by transmission electron microscopy (TEM) show that the axis of NW growth coincides with the [0001] axis. A large number of packing defects are observed in NWs along the (0001) growth plane [14, 15].

The photoluminescent (PL) properties of NW arrays were studied using a Bruker Vertex 80v FT-IR spectrometer equipped with a KBr beam splitter and Ge optical windows. The resolution of the photoluminescence spectra was about 8 MeV. A closed-cycle cryostat (Janis PTCM-4-7,



USA) with zinc selenide windows was used for low-temperature measurements. A continuous-wave diode-pumped solid-state Nd:YAG laser ($\lambda = 1064$ nm) was used to excite the photoluminescent response. The measurements were carried out with p -polarization and a 45° incidence angle of pump radiation. An opto-mechanical modulator with a frequency of 340 Hz was used to modulate the laser radiation. The PL signal was recorded by an InSb photodiode cooled with liquid nitrogen and detected by an SR830 DSP lock-in amplifier.

The virtual crystal approximation (VCA) was used to model the electro-optical properties of solid solutions. The plane -wave basis was used to represent wave functions; the PBEsol exchange-correlation functional was used for geometry optimization [16]; the TB09 meta-GGA potential was used for correct description of electronic properties and for construction of band diagrams [17]. The calculations were carried out taking into account the spin-orbit interaction. According to the calculation results (see Fig. 3,*b*), the observed shift in the position of the PL maximum corresponds to $\text{InAs}_{0.81}\text{P}_{0.19}$.

The electrophysical characteristics and spectral dependence of the photoconversion efficiency of the array were measured by a four-probe system mounted on a cold finger of a nitrogen cryostat. A halogen lamp and a monochromator (SOLAR Laser Systems M266-IV) were used as a tunable source of monochromatic radiation. The photocurrent was measured using a Stanford Research System SR570 current preamplifier and a Stanford Research System SR830 lock-in amplifier. A reference photodiode with known characteristics in the spectral range of 600–2000 nm was used for calibration.

Fabrication of photodetector mesa structures

To study the functional characteristics of the arrays, photodetector mesa structures were fabricated by post-growth technologies based on InAsP NW arrays epitaxially grown on Si. Fig. 3 schematically shows the stages of the formation and a simplified diagram of the NW-based photodetector.

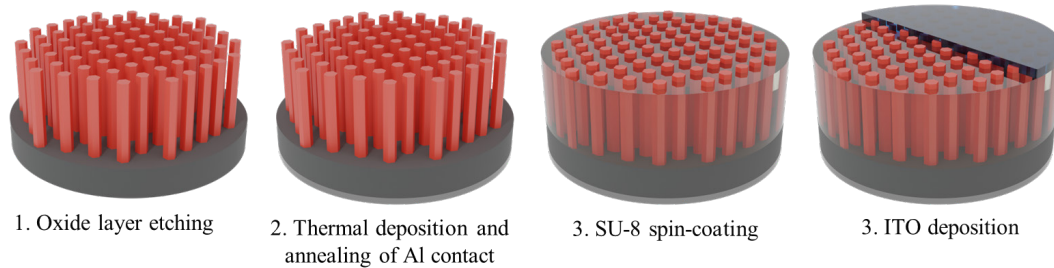


Fig. 3. Fabrication stages and schematic structure of InAsP NW photodetector on Si substrate:

H^+ etching and removal of oxide layer from the back of the substrate (1); thermal deposition and subsequent annealing of Al contact (2); planarization of NW array with SU-8 epoxy resin layer (3); deposition of transparent upper contact made of ITO (4)

At the first stage, H^+ hydrogen plasma was etched at a pressure of 500 mTorr with a discharge power of 20 W for 5 minutes; this treatment is necessary to suppress surface states and recombination centers at the heterojunction between InAs/Si NWs [18]. After the oxide layer is removed, an ohmic back contact (aluminum, 200 nm thick) to the silicon substrate was formed by vacuum thermal evaporation (heating of the target with an electron beam) and rapid thermal annealing (300°C). For partial planarization of the NW array, a layer of epoxy resin (SU-8 photoresist) was applied to the front side of the array by spin-coating, filling the free space between the NWs in the array. The thickness of the applied photoresist layer, controlled by SEM, ensured the formation of front contact to the tips of the NWs (see Fig. 3). To remove the surface oxide from the tips of the NWs, the structures were pretreated in 1 : 3 HCl : H_2O solution. The transparent top contact was a 200 nm thick indium tin oxide (ITO) layer deposited through a shadow mask by high-frequency magnetron sputtering; the Boc Edwards Auto 500 RF 10 Thermal Resistance Evaporator Coater (Great Britain) was used for this purpose.

Results and discussion

The characteristic PL spectra obtained at 5 K from arrays of reference InAs NWs and InAsP solid solution NWs are shown in Fig. 4, *a*. Evidently, the position of the maximum of InAsP NWs is shifted to the region of higher energies by about 120 MeV relative to InAs NWs; this is a consequence of the wider band gap of the InAsP solid solution.

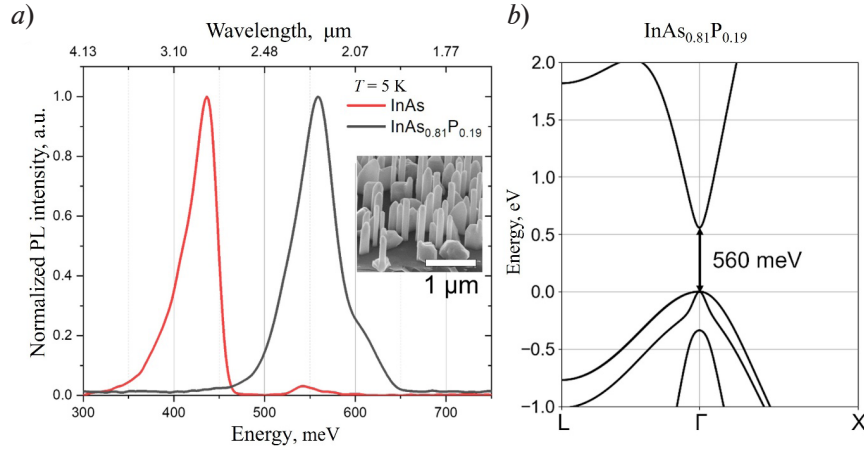


Fig. 4. Low-temperature PL spectra from InAs and InAsP NW arrays epitaxially grown on Si (*a*) and calculated band diagram of $\text{InAs}_{0.81}\text{P}_{0.19}$ with sphalerite crystal structure (*b*)
Inset: SEM image of InAsP NWs

To estimate the phosphorus concentration in the sample under consideration, measurement results of low-temperature PL were compared with the simulation results obtained within the framework of density functional theory (see Fig. 3, *b*). The simulations were carried out in the ABINIT program [19].

Fig. 5, *a* shows the measured and calculated dark I – V characteristics (VAC) at various temperatures. Numerical simulation shows that a constant current value should be observed for a negative bias in the ideal case. This means that the main contribution to the system is made only by thermally generated charge carriers. The direct branch reflects an exponential dependence, which corresponds to the process of injection of holes from silicon substrate into the InAsP array.

The experimentally obtained I – V curve (dependence of the reverse current on the magnitude of reverse-bias voltage) indicates the presence of leakage currents in the array, associated with non-radiative recombination. The latter is caused by the capture of charge carriers into trap states forming near the InAsP/Si interface. A decrease in the operating temperature leads to slowdown

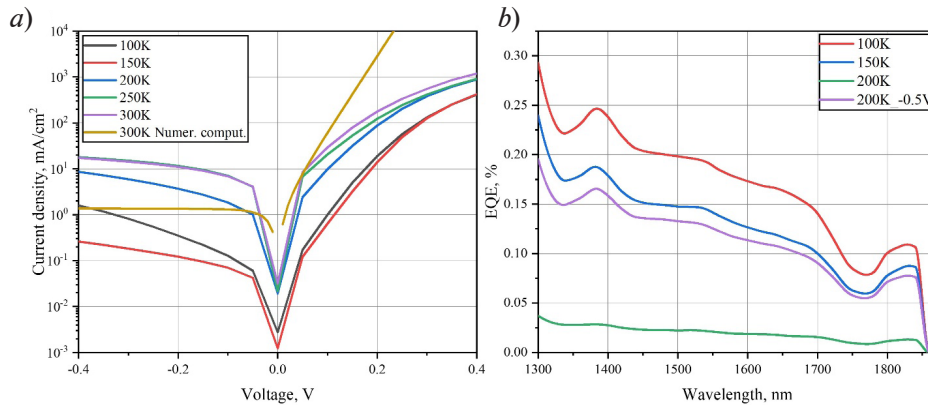


Fig. 5. Electrophysical and spectral temperature-dependent characteristics of photodiode: dependences of current density normalized by the area of the NW array in mesa on voltage (*a*); spectrum of external quantum efficiency (EQE) (*b*)



in both thermal generation of carriers and their recombination through defects, which is manifested as a decrease in reverse current. In addition, there is an increase in the knee voltage of the diode caused by an increase in the band gap of InAsP with a decrease in temperature. The positive branch has a characteristic bend associated with the presence of a series resistance in the measuring circuit, induced, in particular, by the current-collecting top contact.

Spectral dependences of the external quantum efficiency (EQE) of photoconversion were experimentally obtained for the considered photodetector array at zero bias in the wavelength range of 1300–1900 nm. These EQE spectra are shown in Fig. 5, *b*. Notably, the photoresponse for wavelengths less than 1300 nm due to absorption in the silicon substrate was not considered in this study. For example, there is no photoresponse outside the absorption range of silicon at 200 K and above (without the application of an external electric bias), which indicates a high recombination rate of photogenerated charge carriers. As the temperature decreases below 200 K, the photocurrent in the long-wavelength region of the spectrum begins to increase. At 100 K, the EQE value at a wavelength of 1380 nm reaches 0.25%.

We can assume that this behavior of EQE is caused by a change in the carrier transport mechanism in the active area of the NW array with a decrease in temperature. The Debye screening length of the array was estimated to verify this assumption. It was found that at 300 K, the calculated Debye length for InAsP is about 150 nm (particle concentration $n_i = 8 \cdot 10^{14} \text{ cm}^{-3}$), which is significantly shorter than the length of the NWs. Therefore, thermally generated charge carriers effectively screen the built-in electric field, and primarily diffusive carrier transport takes place in the active area of the array. Due to the small lateral size and well-developed surface of the NWs, this type of transport is ineffective, which leads to preferential recombination of photogenerated carriers on the surface states.

As the temperature decreases, the Debye length increases, and the screening area increases. The main transport mechanism in the region of incomplete screening is carrier drift in the built-in electric field between *p*-Si and *n*-InAsP, which occurs due to the difference in the positions of the Fermi levels in the materials.

It can be concluded from the above that the diffusion length of photogenerated carriers in the considered arrays at $T \geq 200 \text{ K}$ is insufficient for effective separation of charge carriers. This is probably due to high density of packaging defects in NWs, reducing the mobility of charge carriers.

Analysis of the observed nonuniform spectral dependence of the array's photoresponse leads us to conclude that the light absorption efficiency strongly depends on the diameter and surface density of the NWs in the array [20]. The calculation data and accounting for the morphology of the NW array (the NW diameter is about 110 nm, the surface density is about $5.5 \text{ } \mu\text{m}^{-2}$) suggest that the absorption coefficient of radiation with a wavelength of 1300 nm in the given array does not exceed 5–7%.

Therefore, to further increase the sensitivity of the InAsP NW-based IR photodetector, it is necessary to optimize the morphological parameters of the NW array by increasing their diameter to 400–500 nm and surface density to $10 \text{ } \mu\text{m}^{-2}$ or more.

Conclusion

We carried out comprehensive studies on the formation of InAsP NW arrays as well as their optical and electrophysical properties. Post-growth technologies were developed to fabricate prototype photodetectors based on InAsP NW arrays on the surface of Si(111) substrate. The temperature dependences of their spectral sensitivity and electrophysical characteristics were studied. We established that the experimentally measured external quantum efficiency of the system under study reaches 0.25% for the IR range.

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