



REGULAR ARTICLE

Optical and Magnetic Properties of Silicon with Binary $\text{Ge}_x\text{Si}_{1-x}$ Nanocompounds

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The paper presents the results of a study of the optical and magnetic properties of silicon with binary compounds obtained by diffusion technology from the gas phase. Diffusion of impurity atoms of germanium and manganese was carried out by a two-stage low-temperature diffusion technology, in which, taking into account the vapor pressure of germanium and manganese diffusants during diffusion, samples were obtained without erosion of the surface of the original material.

The optical properties of the obtained silicon samples with binary compounds $\text{Ge}_x\text{Si}_{1-x}$ were studied on a Lambda 950 UV/Vis/NIR spectrometer. The results of the studies showed that in silicon samples with binary compounds $\text{Ge}_x\text{Si}_{1-x}$, one of the main fundamental parameters of the original silicon changes, i.e. the energy of chemical bonds. A change in the energy of the chemical bond of silicon-germanium atoms will lead to a change in the lattice constant of the original silicon. Analysis of the study results showed a change in the optical properties of the original silicon. The magnetic properties of silicon doped with impurity atoms of germanium were studied after additional doping with impurity atoms of manganese. The selection of the impurity atom of manganese for additional doping is justified, because manganese atoms in silicon lead to an increase in the magnetic property of the silicon sample doped with impurity atoms of germanium. The results of the study showed that due to the unfilled d-shell of manganese atoms, the effect of the magnetic field magnitude leads to a strong change in the spin directions. The obtained results of the study allow creating new semiconductor devices based on silicon with binary $\text{Ge}_x\text{Si}_{1-x}$ and three-component $\text{MnGe}_x\text{Si}_{1-x}$ compounds.

Keywords: Silicon, Compounds, Diffusion, Germanium, Mobility, Concentration, Impurity, Physical mechanism.

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1. INTRODUCTION

Studies of electrophysical, photoelectric and optical properties of binary compounds and heterostructures obtained on the basis of silicon substrates allow to create new modern high-speed semiconductor devices with extended functional capabilities. In most cases, to obtain such structures it is necessary to use complex technologies with the use of expensive devices and installations. At present, studies are conducted on obtaining nanosized structures for the production of modern nanoelectronic devices, with the formation of nanoclusters of impurity atoms [1-4]. In this work, the possibilities of obtaining binary nanocompounds $\text{Ge}_x\text{Si}_{1-x}$ on the surface and near-surface layer of silicon by the method of diffusion technology are shown [5]. The developed diffusion technology from the gas phase allowed to create binary nanocompounds $\text{Ge}_x\text{Si}_{1-x}$ in silicon, which led to a change in the optical and magnetic properties of the original material.

2. EXPERIMENTAL DETAILS

2.1 Material and Methods

To obtain binary compounds $\text{Ge}_x\text{Si}_{1-x}$, the initial monocrystal silicons of the KDB-10 and KEF-100 brands grown by the Czochralsky method (Cz-Si) were used, in which the concentration of the initial boron and phosphorus atoms was $\sim 3 \cdot 10^{15} \text{ cm}^{-3}$ and $\sim 10^{13} \text{ cm}^{-3}$, respectively. Diffusion of impurity germanium atoms was carried out using the developed low-temperature two-stage diffusion technology. The essence of the developed low-temperature diffusion technology is as follows: the studied initial silicon samples and germanium diffusant with a certain mass (this depends on the volume of the quartz ampoule) are placed in quartz ampoules and the vacuum is pumped out in the limit of $P \sim 10^{-6} \text{ mm Hg}$. After that, the quartz ampoules were installed in an MG17-60/300 diffusion electric furnace at a temperature of $T = 30^\circ \text{C}$. The temperature of the electric furnace with quartz ampoules was gradually raised at a rate of 5 degrees/min.

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In this case, the silicon samples were heated to the temperature of the first stage of diffusion, which is in the temperature range of $T = 600 \div 650^\circ\text{C}$ and were maintained at this temperature for $t = 20 \div 30$ minutes. Then the temperature of the electric furnace was relatively quickly raised ($t = 20 \div 30$ degrees/min) to the temperature of maximum solubility of impurity germanium atoms in silicon ($T = 1150 \div 1250^\circ\text{C}$) and the samples were maintained at this temperature for $t = 5$ hours, after which the quartz ampoules were removed from the electric furnace and cooled at a rate of 200 degrees/sec and dropped into cold technical oil [6, 7]. The absorption spectra of the obtained silicon samples doped with impurity germanium atoms were studied on a Lambda 950 UV/Vis/NIR spectrometer at $T = 300$ K (Fig. 1).

From the results of the study and the solution of the Kubelka-Munk function, the width of the forbidden zone of the formed binary compounds $\text{Ge}_x\text{Si}_{1-x}$ in silicon was determined based on the spectra of reflected light (Eq. 1). The calculation results show that the width of the forbidden zone of the original samples of monocrystal silicon was equal to $E_{\text{Si}} = 1.12$ eV, the width of the forbidden zone with binary compounds $\text{Ge}_x\text{Si}_{1-x}$ was $E = 0.76$ eV (Fig. 1a).

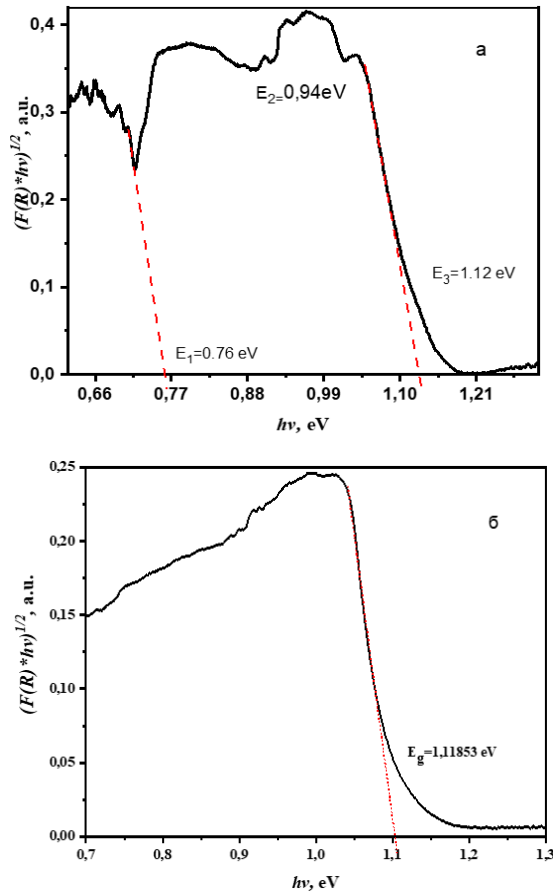


Fig. 1 – a) Absorption spectrum of silicon doped with germanium impurity atoms, b) Absorption spectrum of the original monocrystal silicon of the KDB = 10 brand, at $T = 300$ K

$$F_{KM} = \frac{(1-R)^2}{2R} \quad (1)$$

$$h\nu \cdot F_{KM}^{1/n} = A(h\nu - E_g)$$

where: R is the reflection coefficient, A is the proportionality coefficient, $h\nu$ is the energy determined from the slope of the absorption spectrum, E_g is the band gap.

From the slope angle θ and the solution of the Kubelka-Munk function, the band gap of the formed $\text{Ge}_x\text{Si}_{1-x}$ compounds in silicon was determined based on the spectra obtained on a Lambda 950 UV/Vis/NIR spectrometer (Eq. 1).

From the analysis of the results of measuring the absorption spectra, it was established that the absorption spectrum of silicon doped with impurity germanium atoms differs from the original monocrystal silicon [8]. Using the reverse calculation according to Vegard's law (Eq. 2), the ratio of the lattice constant of the original silicon and silicon with binary compounds $\text{Ge}_x\text{Si}_{1-x}$ was determined, as well as the value of the forbidden band (E_g) of the obtained materials, which was $E_{\text{SiGe}} = 0.79$ eV at $x = 0.25$.

$$E_{g,(\text{Ge})_x\text{Si}_{(1-x)}} = (1-x) \cdot E_{g,\text{Si}} + x \cdot E_{g,\text{Ge}} \quad (2)$$

$$0.79 = (1-x) \cdot 1.12 + x \cdot 0.68$$

Thus, it can be said that the calculated energy $E_{\text{SiGe}} = 0.79$ eV corresponds to the energy of the chemical bond of the newly formed compound $\text{Ge}_{0.25}\text{Si}_{0.75}$ in silicon.

The value of the band gap of the formed $\text{Ge}_x\text{Si}_{1-x}$ compounds, which is equal to $E_g = 0.79$ eV (at $x = 0.25$), is much smaller than the band gap of the original silicon and larger than that of the original germanium. This indicates the formation of microheterovarizional structures of the $\text{Si}_2\text{Ge}_x\text{Si}_{1-x}$ type in silicon. In [4, 9], the authors obtained binary solid compounds $\text{Ge}_x\text{Si}_{1-x}\text{-Si}_2$ on the surface of silicon, in which heterostructures were formed. In our case, we obtained microheterovarizional structures $\text{Ge}_x\text{Si}_{1-x}\text{-Si}_2$ using the conventional diffusion technology of doping impurity atoms of germanium into silicon. The composition and optical properties of the obtained binary compounds $\text{Ge}_x\text{Si}_{1-x}$ in silicon were studied using an infrared Fourier spectrometer FSM-1202. Before studying the optical properties of the obtained silicon samples doped with impurity atoms of germanium, two opposite surfaces were polished with diamond paste on a polishing machine, and the remaining four sides of the silicon samples were ground to $\sim 50 \mu\text{m}$ in order to remove the near-surface enriched layer of impurity atoms of germanium. These studies made it possible to accurately measure the elemental composition of the formed binary compounds $\text{Ge}_x\text{Si}_{1-x}$ on the surface and near-surface layer of silicon (Fig. 2).

The analysis of the obtained results showed that the formation of binary compounds of impurity germanium atoms with atoms of the initial silicon leads to the appearance of a peak associated with the absorption of the frequency of 1066 cm^{-1} . From the comparative analysis it was found that the peaks observed at 885 cm^{-1} and 850 cm^{-1} correspond to germanium oxides (GeO_2), the peak

at 1104 cm^{-1} is associated with the formation of silicon oxygen compound (Si-O). The intensity of other peaks at 463 cm^{-1} , 814 cm^{-1} , and 1104 cm^{-1} showed the presence of silicon oxide (SiO_2). The transmission coefficient at the peak of 1066 cm^{-1} showed the presence of binary compounds of the $\text{Ge}_x\text{Si}_{1-x}$ type in silicon. For the study, silicon samples doped with germanium impurity atoms subjected to additional heat treatment in the temperature range $T = 800 \div 1000^\circ\text{C}$ for $t = 2$ hours were used.

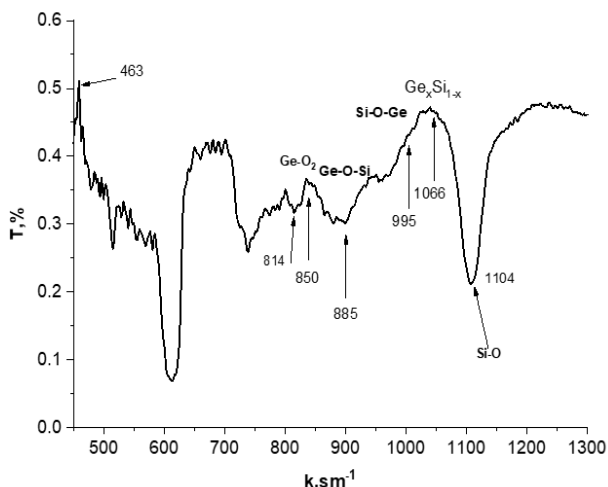


Fig. 2 – Elemental composition of silicon surface doped with germanium impurity atoms. The study was conducted on an FSM 1202 spectrometer, at $T = 300\text{ K}$

To study the magnetic properties of silicon with binary compounds $\text{Si}_{1-x}\text{Ge}_x$, the obtained samples were additionally doped with manganese impurity atoms. The choice of the manganese impurity atom was justified by the analysis of literature data. In works [10-13], the authors studied the magnetic properties of silicon samples with germanium impurity atoms by additionally introducing manganese impurity atoms into the crystal lattice with the formation of ferromagnetic compounds of the GeMn type in silicon [14]. Also, in work [15], the authors obtained magnetic nanowires with binary compounds $\text{Ge}_{0.99}\text{Mn}_{0.01}$ taking into account the high reactivity of germanium atoms with manganese atoms. An analysis of literature has shown that although the electrical, photoelectric and optical properties of silicon doped separately with manganese impurity atoms have been studied in sufficient detail by many authors [16-17], silicon containing a $\text{Ge}_x\text{Si}_{1-x}\text{Mn}$ type compound has not been studied to date.

Magnetic properties were studied in silicon samples with binary $\text{Ge}_x\text{Si}_{1-x}$ compounds additionally doped with manganese impurity atoms. Magnetic properties were measured using an FM-Nanoview 1000 AFM magneto-atomic force microscope. The results of the study showed that the presence of nanoclusters of $\text{MnGe}_x\text{Si}_{1-x}$ ternary compounds leads to the appearance of a ferromagnetic state at a temperature of $T = 300\text{ K}$ (room temperature). It should be noted that for the first time, ferromagnetic properties of

silicon with compounds of germanium and manganese impurity atoms were obtained using diffusion technology.

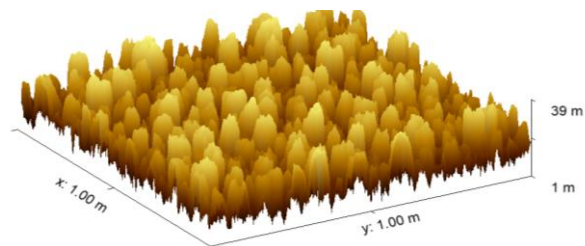


Fig. 3 – Topology of magnetic domains in silicon doped with germanium and manganese impurity atoms, obtained in an FM-Nanoview 1000 AFM magnetic-atomic force microscope at $T = 300\text{ K}$

Fig. 3 shows the topology of magnetic domains in silicon samples doped with impurity atoms of germanium and manganese. As can be seen from the figure, the silicon surface is completely covered with magnetic domains consisting of impurity atoms of germanium and manganese, which not only enhances the ferromagnetic properties of silicon, but also leads to a shift in the Curie temperature to higher values, in which silicon samples with $\text{MnGe}_x\text{Si}_{1-x}$ compounds pass from the paramagnetic state to the ferromagnetic state (Fig. 4). It was found that in silicon samples with impurity atoms of germanium and manganese, ferromagnetic properties were first observed at a temperature of $T = 300\text{ K}$ and higher.

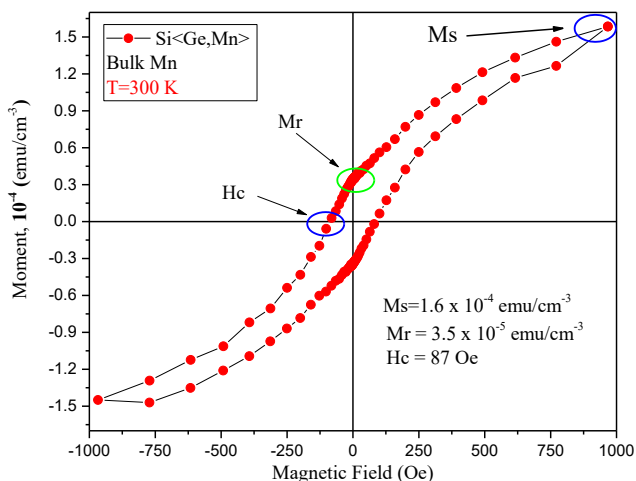


Fig. 4 – Dependences of the magnetic moment of silicon doped with impurity atoms of germanium and manganese on the magnetic field, at $T = 300\text{ K}$, obtained in a Quantum Design MPMS-3 superconducting quantum interference magnetometer

3. RESULT AND DISCUSSION

From the analysis of the obtained results, it was established that at the additional annealing temperature $T = 800^\circ\text{C}$ the maximum formation of binary compounds $\text{Ge}_x\text{Si}_{1-x}$ occurs, and at this temperature the minimum thickness of oxidation of the Si substrate is formed. Two dips in the transmittance (absorption peaks) at 850 cm^{-1} and 885 cm^{-1} are shown in Fig. 2, indicating the formation

of a germanium oxide layer O-Ge-O (GeO_2) with a significant thickness on the silicon surface [8]. These data were confirmed by the results obtained by other authors in [18, 19]. These results of the study confirm that impurity atoms of manganese in silicon lead to the formation of $\text{MnGe}_x\text{Si}_{1-x}$ nanoclusters with magnetic properties [20-22]. Analysis of the obtained research results confirmed that in silicon samples containing ternary compounds $\text{MnGe}_x\text{Si}_{1-x}$ the magnetic properties are enhanced due to the unfilled d -shell of impurity manganese atoms in which the spin directions change greatly. This leads to a strong manifestation of the ferromagnetic properties of silicon and a shift in the Curie temperature to higher values.

4. CONCLUSION

The analysis of the obtained results showed that impurity germanium atoms form binary compounds of the $\text{Ge}_x\text{Si}_{1-x}$ type on the surface and near-surface layer of silicon. The formation of such compounds with a high concentration leads to the formation of microheterostructures based on binary $\text{Ge}_x\text{Si}_{1-x}\text{Si}$

compounds, which in turn lead to a significant increase in the photoconductivity value, and also expand the spectral sensitivity range relative to the original silicon. These research results indicate that silicon with binary $\text{Ge}_x\text{Si}_{1-x}$ compounds can be used as a promising material for the development and manufacture of photodetectors with extended spectral sensitivity and efficient photocells. Optical and magnetic properties of silicon with ternary $\text{MnGe}_x\text{Si}_{1-x}$ compounds obtained by diffusion of impurity germanium and manganese atoms from the gas phase showed that these samples are a new interesting material in magnetoelectronics and spintronics. The observed physical phenomena are of particular interest to specialists, since silicon is the main material in the manufacture of semiconductor products using planar technology. The results of the study were carried out with the support of the international Uzbek-Turkish project "AL-2021022215, Integrated microfluidic system for capturing circulating cancer cells with ferromagnetic nanoclusters in Si".

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Оптичні та магнітні властивості кремнію з бінарними наносполуками $\text{Ge}_x\text{Si}_{1-x}$

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У статті представлені результати дослідження оптичних та магнітних властивостей кремнію з бінарними сполуками, отриманими за допомогою дифузійної технології з газової фази. Дифузію домішкових атомів германію та марганцю проводили за двоступеневою низькотемпературною дифузійною технологією, в якій, враховуючи тиск пари дифузантів германію та марганцю під час дифузії, отримували зразки без ерозії поверхні вихідного матеріалу.

Оптичні властивості отриманих зразків кремнію з бінарними сполуками $\text{Ge}_x\text{Si}_{1-x}$ досліджували на UV/Vis/NIR спектрометрі Lambda 950. Результати досліджень показали, що у зразках кремнію з бінарними сполуками $\text{Ge}_x\text{Si}_{1-x}$ змінюється один з основних фундаментальних параметрів вихідного кремнію, тобто енергія хімічних зв'язків. Зміна енергії хімічного зв'язку атомів кремній-германій призведе до зміни постійної кристалічної решітки вихідного кремнію. Аналіз результатів дослідження показав зміну оптичних властивостей вихідного кремнію. Магнітні властивості кремнію, легованого домішковими атомами германію, досліджували після додаткового легування домішковими атомами марганцю. Вибір домішкового атома марганцю для додаткового легування є виправданим, оскільки атоми марганцю в кремнії призводять до збільшення магнітних властивостей зразка кремнію, легованого домішковими атомами германію. Результати дослідження показали, що через незаповнену d -оболонку атомів марганцю, вплив величини магнітного поля призводить до сильної зміни напрямків спінів. Отримані результати дослідження дозволяють створювати нові напівпровідникові прилади на основі кремнію з бінарними сполуками $\text{Ge}_x\text{Si}_{1-x}$ та трикомпонентними $\text{MnGe}_x\text{Si}_{1-x}$.

Ключові слова: Кремній, Дифузія, Германій, Рухливість, Концентрація, Домішка, Фізичний механізм.