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MAGNETIC PROPERTIES OF SILICON WITH PARAMAGNETIC IMPURITY ATOMS[†]

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One of the possible ways to obtain silicon with magnetic properties is the introduction of paramagnetic impurities into silicon: Cr, Mn, Fe, Ni, and Co. In our opinion, silicon materials containing magnetic nanosized clusters are most suitable for spintronic devices. The possibility of obtaining silicon with magnetic properties by diffusion doping was studied in this work. To obtain silicon doped with Cr, Mn, Fe and Ni impurity atoms, *p*-type single-crystal silicon with a specific resistance of ρ = 5 Ohm·cm and ρ = 0.5 Ohm·cm was used, and for doping with Co atoms, *n*-type silicon with resistivity ρ=10 Ohm·cm was used. The diffusion temperature and time were chosen such that, after diffusion annealing, the samples with impurity Cr, Fe, and Mn atoms remained highly compensated *p*-type, and when doped with impurity Co atoms, they remained high-resistance *n*-type. The results of the study showed that with decreasing temperature, the value of the negative magnetoresistance Δρ/ρ in the Si<Mn> samples increases and reaches its maximum value (about 800%) at *T* = 240 K, a further decrease in temperature leads to a decrease in the magnetoresistance, and at a temperature *T* = 170 K, the sign of the magnetoresistance is inverted. In Si <Cr> samples, with decreasing temperature, the positive magnetoresistance turns into a negative one, the value of which increases with decreasing temperature, and is achieved at *T*=100 K Δρ/ρ = 45–50%. In Si<Fe> samples, with decreasing temperature, the value of negative magnetoresistance increases monotonically and at *T*=100 K its value is Δρ/ρ = (100÷120) %. The study in Si<Co> samples showed that with decreasing temperature the value of positive magnetoresistance increases and at *T*=100 K it reaches Δρ/ρ = (17÷20) %. The study of magnetoresistance in samples - Si<Ni> showed that with decreasing temperature the value of positive magnetoresistance increases and at *T*=100 K it reaches Δρ/ρ = (10÷15) %. When studying the magnetic properties of p-Si <B, Mn> samples at low temperatures (below *T*=30 K), a ferromagnetic state was found, i.e. succeeded in obtaining a magnetic semiconductor material by the method of diffusion of a paramagnetic impurity. In the overcompensated Si <B, Mn> (*n*-type) samples, no magnetic hysteresis was found. This shows a significant effect on the magnetic properties of the manganese impurity in silicon of its charge and, accordingly, spin state. Based on the results obtained, it can be argued that diffusion doping of silicon with manganese can be used to obtain silicon with magnetic properties.
Keywords: *Silicon; Manganese; Nickel; Nanocluster; Magnetoresistance; Ferromagnetic; Hysteresis*
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INTRODUCTION

Obtaining magnetic semiconductor materials for modern spintronics [1-3] and studying their magnetic properties is of great scientific and practical interest. One of the possible ways to obtain silicon with magnetic properties is the introduction of paramagnetic impurities into silicon: Cr, Mn, Fe, Ni, and Co. The electronic structure and parameters of these impurity atoms are presented in Table 1.

As can be seen from Table 1, these elements in the silicon lattice act as paramagnetic impurities with fairly high spin values. The main disadvantage is the low limiting solubility in the electroactive state. It was established [5] that chromium and manganese atoms are mainly located in interstitial positions in the silicon crystal lattice and act as donor impurities. Ni, Fe, and Co atoms can be located both in interstices and in the nodes of the silicon crystal lattice [5]. The concentration of electroactive atoms of these impurities, except for Ni and Co atoms [5], depends significantly on the diffusion conditions and the cooling rate. At the maximum cooling rate of silicon samples, it is possible to obtain a material in which all introduced impurity atoms (Cr, Mn, and Fe) are in an electroactive state [6].

In addition, the nonelectroactive part of the dissolved impurity in the silicon lattice in the form of interstitial atoms, dimers, nanoclusters, microclusters, precipitates, and magnetic silicides can also have magnetic properties.

The magnetic properties due to microclusters, precipitates and silicides are unsuitable for spintronic devices due to the strong inhomogeneity of the magnetic properties over the volume. An example of such an impurity is nickel, which easily forms microclusters, precipitates, and silicides, due to which the nonelectroactive nickel concentration near the surface reaches 10^{20}–10^{21} cm^{-3} [8, 9]. At the same time, the electroactive solubility of nickel does not exceed 10^{16} cm^{-3}; therefore, doping with nickel hardly changes the resistivity of the samples [10].

In our opinion, silicon materials containing magnetic nanosized clusters (including those containing atoms of other impurity elements – impurity complexes) are most suitable for spintronic devices. Such formations were found for

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manganese impurities in the form of BMn_4 nanoclusters in the silicon lattice [11]. In addition, ferromagnetic properties have been found at room temperature in silicon containing boron and about 1% manganese atoms [2,12].

Table 1. Electronic structure and parameters of Cr, Mn, Fe, Ni, and Co impurity atoms in silicon [7].

Element	Electronic structure	Spin	Total limiting solubility, cm^{-3}	The electroactive part of the limiting solubility, cm^{-3}	Energy levels in silicon
Cr	$3d^54S^1$	$\frac{5}{2}$	10^{16}	10^{14}	Ec – 0.41
Mn	$3d^54S^2$	$\frac{5}{2}$	$2\cdot10^{16}$	10^{16}	Ec – 0.43 Ec – 0.53 Ev + 0.45
Fe	$3d^64S^2$	2	$4\cdot10^{16}$	$2\cdot10^{14}$	Ec – 0.14 Ec – 0.51 Ec – 0.40
Co	$3d^74S^2$	$\frac{3}{2}$	10^{16}	$6\cdot10^{15}$	Ev + 0.53 Ev + 0.35
Ni	$3d^84S^2$	1	$7\cdot10^{17}$	$7\cdot10^{14}$	Ev + 0.35 Ev + 0.23

The formation of magnetic structures in semiconductors can be carried out by various methods: chemical vapor deposition, molecular beam epitaxy, or ion implantation. Thus, the implantation of crystalline silicon with transition metal ions (Co, Ni, and Fe) is used to create magnetic nanoclusters, as well as metal silicides [13–16].

In this work, the possibility of obtaining silicon with magnetic properties due to diffusion doping with Cr, Mn, Fe, Ni, Co impurities was studied.

Project AL-202102215 is developing an integrated microfluidic system that captures circulating cancer cells with ferromagnetic clusters in silicon.

TECHNOLOGY AND RESEARCH METHODS

To obtain silicon doped with Cr, Mn and Fe impurity atoms, we used *p*-type single-crystal silicon with a resistivity ρ=5 Ohm·cm, and for doping with Co atoms, we used *n*-type silicon with a resistivity ρ=10 Ohm·cm. To obtain silicon doped with Ni impurity atoms, *p*-type silicon with a resistivity ρ=0.5 Ohm·cm was used. The diffusion temperature and time were chosen such that, after diffusion annealing, the samples with impurity Cr, Fe, and Mn atoms remained highly compensated *p*-type, and when doped with impurity Co atoms, they remained high-resistance *n*-type.

Before and after diffusion, the samples were subjected to mechanical and chemical treatment (cleaning in an ammonium peroxide solution and etching for 1 minute in a HF + HNO_3 1:3 mixture) to remove impurities and remove mechanical damage to the surface. The magnetoresistance of the obtained samples was measured on a setup that allows you to adjust the magnitude of the magnetic field from 0.1 to 2 Tl, as well as the electric field strength applied to the sample from 0.1 to 1000 V/cm, in the temperature range T=100÷300 K [7].

Measurements of the magnetization of the samples at low temperatures were carried out on a SQUID magnetometer.

THE RESULTS OBTAINED AND THEIR DISCUSSION

The electrical parameters of the obtained samples are shown in Table 2.

Table 2. Electrical parameters of silicon doped with Cr, Mn, Fe, Co, and Ni atoms

Samples	*ρ*, Ohm·cm	Conductivity type	Charge carrier concentration, cm^{-3}	The mobility of charge carriers, $cm^2/V\cdot c$
Si <Cr>	$(5÷6)\cdot10^3$	*p*	$3.8\cdot10^{12}$	250÷270
Si <Mn>	$(5÷8)\cdot10^3$	*p*	$5.2\cdot10^{12}$	160÷200
Si <Fe>	$(5÷7)\cdot10^3$	*p*	$4.2\cdot10^{12}$	200÷250
Si <Co>	$10^3÷10^4$	*n*	$6.2\cdot10^{11}÷6.9\cdot10^{12}$	900÷1000
Si <Ni>	0,5	*p*	$4\cdot10^{16}$	250÷350

Table 3 shows the values of the magnetoresistance (Δρ/ρ) of the samples at T=300 K obtained with the same values of the electric (E=200 V/sm) and magnetic fields (near 2 Tl).

Table 3. Values of resistivity and magnetoresistance of samples at T=300 K

Samples	*ρ* (Ohm·cm)	Maximum value Δρ/ρ, %	Type of magnetoresistance in the range of magnetic fields, (0÷2 Tl)
Si<Cr>	$5\cdot10^3$	from -7 to +8	Weak negative and positive
Si<Mn>	$5.3\cdot10^3$	-97	Big negative
Si<Fe>	$5.5\cdot10^3$	-7	Weak negative
Si<Co>	$5.1\cdot10^3$	5	Weak positive
Si<Ni>	0.5	6	Weak positive

It has been established that in samples doped with impurity Ni, Cr, and Co atoms, predominantly weak (less than 6%) positive magnetoresistance (PMS) takes place. At the same time, high (more than 100%) negative magnetoresistance (NMR) is observed in Si<Mn> samples. In Si<Fe> samples, NMR is also observed, but the value of NMR is much less than Δρ/ρ = 5–7 %. It was found that the NMR value in the Si<Mn> samples increase significantly with an increase in the applied electric (E) and magnetic (B) fields.

Interesting results were obtained in the study of the temperature dependence of the magnetic resistance Δρ/ρ in the temperature range *T*=100÷380 K.

Previously, it was found that in silicon samples doped with impurity manganese atoms (Si<Mn>), with decreasing temperature, the value of the NMR increases and reaches its maximum value (Δρ/ρ = 300%) at a temperature of *T*=235÷240 K [17,18].

We have studied the magnetoresistance depending on temperature in the samples Si<Mn>, Si<Cr>, Si<Fe>, Si<Co>, Si<Ni> in the field E=200 V/cm and B=2 Tl (Fig. 1).

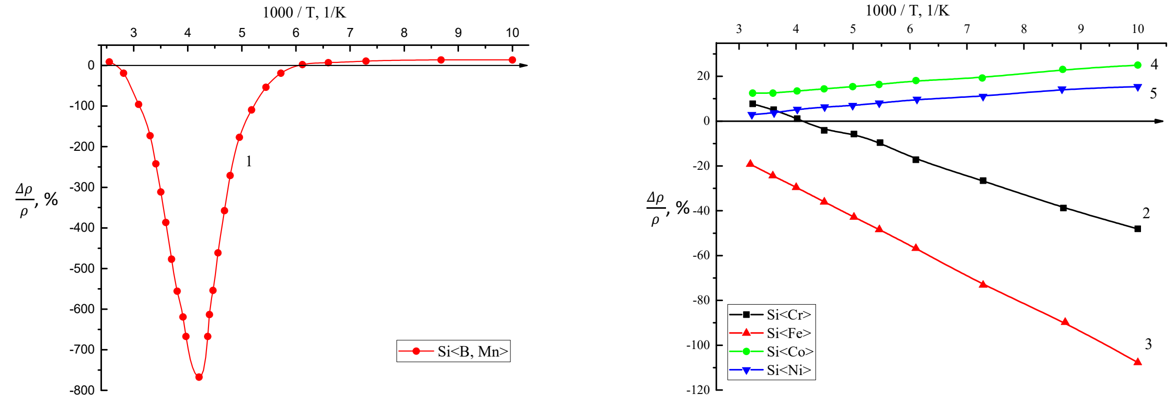

Figure 1. Temperature dependence of magnetoresistance for samples: 1- Si<Mn>, 2- Si<Cr>, 3- Si<Fe>, 4- Si<Co>, 5- Si<Ni> at E=200 V/cm, B =2 Tl.

The results of the study showed that with decreasing temperature, the NMR value in Si<Mn> samples increase and reaches its maximum value (about 800%) at *T*=240 K, further temperature decrease leads to a fairly rapid decrease in NMR and at temperatures *T*=160÷170 K there is an inversion of the sign of the magnetoresistance i.e. The NMR passes to the PMR, the value of the PMR in this temperature range weakly depends on temperature. At a temperature of *T* > 380 K, a slight PMR is also observed.

In Si<Cr> samples, with decreasing temperature, the PMR transforms into NMR, the value of which grows quite rapidly with decreasing temperature, and is reached at *T*=100 K Δρ/ρ = 45–50%. In this case, the NMR value increases linearly with an increase in both the electric and magnetic field strengths.

In Si<Fe> samples, in contrast to Si<Mn> and Si<Cr>, with decreasing temperature, the value of the NMR increases monotonically and at *T*=100 K its value is Δρ/ρ = (100÷120) %. In these samples, the magnetoresistance sign inversion is not observed in the studied temperature range; always observed only NMR.

The study of the magnetoresistance in Si<Co> samples showed that with decreasing temperature, the value of the PMR increases and at *T*=100 K it reaches Δρ/ρ = (17÷20) %. This quantity depends very weakly on the applied electric and magnetic fields.

The study of the magnetoresistance in Si<Ni> samples showed that with decreasing temperature, the value of the PMR increases and at *T*=100 K it reaches Δρ/ρ = (10÷15) %.

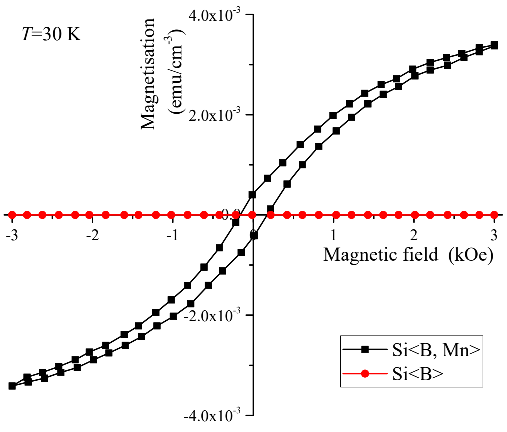

Figure 2. The dependence of the magnetization of the samples *p*-Si <B, Mn> and *p*-Si <B> on the magnetic field (hysteresis) at *T*=30 K

When studying the magnetic properties of *p*-Si <B, Mn> samples in the low temperature region, we found a ferromagnetic state at *T*=30 K (Fig. 2), i.e. succeeded in obtaining a new magnetic semiconductor material by impurity diffusion. In the overcompensated Si <B, Mn> (*n*-type) samples, no magnetic hysteresis was found. This shows a significant effect on the magnetic properties of the manganese impurity in silicon of its charge and, accordingly, spin state.

Previous studies using the EPR method showed that only in *p*-Si<B, Mn> samples, nanoclusters of the BMn_4 type with a high magnetic moment are formed [17-19]. Under optimal doping conditions, the concentration of such clusters reaches $N=10^{15}$ cm^{-3} [20, 21].

In the Si <Ni> samples, a ferromagnetic state was also found at *T* = 300 K, which is probably due to the presence of nickel precipitates [22-26] on the surface and in the bulk of the samples. This assumption is confirmed by the fact that when the samples are etched to a depth of 100 μm, the magnetization of the samples is greatly reduced.

On the basis of the obtained results, it can be argued that diffusion doping of silicon with manganese with the formation of BMn_4 nanoclusters can be used to obtain silicon with magnetic properties.

CONCLUSION

The high spatial homogeneity of the p-Si <B, Mn> samples with BMn_4 nanoclusters and the ferromagnetic state observed in them at T=30 K makes it possible to use this material in devices operating on the principle of spin polarization. A significant NMR value, reaching 50% at room temperature, makes it possible to create sensitive magnetoresistive sensors for magnetic measurements.

Conflict of Interest

The authors declare that they have no conflict of interest

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МАГНІТНІ ВЛАСТИВОСТІ КРЕМНІЮ З ДОМІШКАМИ ПАРАМАГНІТНИХ АТОМІВ

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Одним із можливих шляхів отримання кремнію з магнітними властивостями є введення в кремній парамагнітних домішок: Cr, Mn, Fe, Ni та Co. На нашу думку, кремнієві матеріали, що містять магнітні нанорозмірні кластери, найбільш придатні для пристроїв спінтроніки. У роботі досліджено можливість отримання кремнію з магнітними властивостями шляхом дифузійного легування. Для отримання кремнію, легованого домішковими атомами Cr, Mn, Fe та Ni, використовували монокристалічний кремній p-типу з питомим опором ρ = 5 Ом·см і ρ = 0,5 Ом·см, а для легування атомами Co використовувався кремній n-типу з питомим опором ρ=10 Ом·см. Температуру та час дифузії вибирали так, щоб після дифузійного відпалу зразки з домішковими атомами Cr, Fe та Mn залишалися висококомпенсованими p-типу, а при легуванні домішковими атомами Co залишалися високоомними n-типу. Результати дослідження показали, що зі зниженням температури величина від'ємного магнітоопору Δρ/ρ у зразках Si<Mn> зростає і досягає максимального значення (близько 800%) при T = 240 K, подальше зниження температури призводить до до зменшення магнітоопору, а при температурі T = 170 K знак магнітоопору змінюється. У зразках Si <Cr> зі зниженням температури позитивний магнітоопір переходить у від'ємний, величина якого зростає зі зниженням температури, і досягається при T=100 K Δρ/ρ = 45–50%. У зразках Si<Fe> зі зниженням температури величина від'ємного магнітоопору монотонно зростає і при T=100 K його значення становить Δρ/ρ = (100÷120) %. Дослідження на зразках Si<Co> показало, що зі зниженням температури величина позитивного магнітоопору зростає і при T=100 K досягає Δρ/ρ = (17÷20) %. Дослідження магнітоопору в зразках - Si<Ni> показало, що зі зниженням температури величина позитивного магнітоопору зростає і при T=100 K досягає Δρ/ρ = (10÷15) %. При дослідженні магнітних властивостей зразків p-Si <B, Mn> при низьких температурах (нижче T=30 K) виявлено феромагнітний стан, тобто вдалося отримати магнітний напівпровідниковий матеріал методом дифузії парамагнітної домішки. У надкомпенсованих зразках Si <B, Mn> (тип n) магнітного гістерезису не виявлено. Це свідчить про істотний вплив на магнітні властивості домішки марганцю в кремнії його зарядового і, відповідно, спінового стану. На підставі отриманих результатів можна стверджувати, що дифузійне легування кремнію марганцем може бути використане для отримання кремнію з магнітними властивостями.

Ключові слова: *кремній; марганець; нікель; нанокластер; магнітоопір; феромагнетик; гістерезис*